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APPENDICES TO
COMMENTS OF THE GENERAL ELECTRIC COMPANY
ON THE
AUGUST 1991 REVIEW COPY
OF THE
PHASE 1 REPORT - INTERIM CHARACTERIZATION AND EVALUATION
FOR THE
HUDSON RIVER PCB REASSESSMENT RI/FS

October 24, 1991

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1. Institute for Evaluating Health Risks. 1991. Reassessment of Liver Findings in Five PCB Studies in Rats.

2. Comments of the General Electric Company on the Advanced Notice of Proposed Rulemaking of the United States Environmental Protection Agency Concerning Disposal of Polychlorinated Biphenyls, 55 F.R. 26738. 1990.

3. Chase, K.H., J. Doull, S. Friess, J.V. Rodricks and S. Safe. 1989. Evaluation of the Toxicology of PCBs.

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2. Xu, Y.J. 1991. Transport Properties of Fine-Grained Sediments, Ph.D. dissertation Abstract, UCSB.

3. Gailini, J., C.K. Ziegler, W. Lick. 1991a. The Transport of Suspended Solids in the Lower Fox River, J. of Great Lakes Research, in press.

4. Ziegler, C.K., J. Lick and W. Lick. 1990. SEDZL: A User-Friendly Numerical Model for Determining the Transport and Fate of Fine-Grained, Cohesive Sediments, UCSB report.

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2. Palermo, Autumn 1991, Equipment Choices for Dredging Contaminated Sediments, Remediation Journal.

3. Rice and White, 1987, PCB Availability Assessment of River Dredging Using Caged Clams and Fish, 6 Env. Tox. 259-274 (1987).

4. Swan, Analysis of Dredge Safety Hazards, United States Dept. of the Interior, Bureau of Mines.

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3. Abramowicz, D.A. (1990) Aerobic and Anaerobic Biodegradation of PCBS: A Review. V. 10 I.3, pp. 241-251 Critical Reviews in Biotechnology.

4. Bedard, D.L. (1990) Bacterial Transformation of Polychlorinated Biphenyls. V. 4 Biotechnology and Biodegradation.

5. General Electric Company Corporate Research and Development (1990) Research and Development Program for the Destruction of PCBS: Ninth Progress Report

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A. SPECIFIC PAGE BY PAGE COMMENTS

APPENDIX A
SPECIFIC PAGE BY PAGE COMMENTS

Executive Summary

Pg. E-1, par. 1: EPA cites an upper bound of GE discharges of 1.3 million pounds. To GE knowledge there is not any reliable information to support the use of this number in this or any other document. EPA should carefully review all data presented to it.

Pg. E-1, par. 2: The discussion of the Fort Edward Dam implies that the dam was a GE dam and the removal was done by GE. The report should clarify that this was not the case.

Pg E-1, par. 3: The discussion of the commercial fishing ban on the taking of striped bass fails to mention the conservation reason for the ban as well as the fact the majority of the PCBs in the striped bass do not originate from within the Hudson River.

Pg. E-5, par. 3: EPA's conclusion that the historical loading to the Lower Hudson was dominated by those from the Upper Hudson is an assumption proposed by others and accepted by EPA without critical evaluation. As discussed in these comments, the data does not in fact support such a conclusion.

Pg. E-5, par. 4: EPA states that the PCB deposition in the Lower Hudson occurred around 1973, implying that this coincides with the removal of the Fort Edward Dam. The actual data from the lower river does not support this and, in fact, show that the maxima in PCB concentration may have, in fact, occurred in 1971, two years prior to the dam removal. The PCB maxima in the sediments is not inconsistent with the national use patterns and data obtained from other rivers. This and other significant information point directly to other dischargers as being important.

Pg. E-10, par. 3: The EPA's assessment of the carcinogenic potency of PCBs is based on out dated information. This information has been supplied to EPA and must be considered as part of this reassessment.

E-10, par. 3: EPA Region II points to emerging evidence on neurological or developmental effects of PCBs. GE is unaware of any thorough evaluation of the published data on these topics being done by EPA. It is premature for EPA to conclude such effects occur due to PCB exposure.

Pg. E-13, par. 3: GE urges EPA not to move forward, at this time, with the data collection proposed in the Phase 2A sampling plan. GE's comments on that document are included here by reference. GE, also, would like to express the concern that EPA has not allowed public comment on the Phase 2A sampling plan.

Section I - Introduction

Pg. I-1, par. 2: Many months ago EPA asked for comment on the January 1991 review copy of the Phase 1 work plan for the Reassessment Remedial Investigation and Feasibility Study (RRI/FS). To date, EPA has not made available to GE a response to comments or a final work plan. GE formally requests that EPA make all comments received available to GE and that EPA issue a final work plan.

Pg. I-2, par. 3: EPA states that the reassessment is being performed for the river bottom sediments from Hudson Falls to the Federal Dam in Troy. In a number of places in the Phase 1 Report, EPA seems to ignore this definition and to expand the scope. Specifically, EPA baseline risk assessment includes the risks posed by PCBs from the sources above Rogers Island. Additionally, EPA is including excavation technologies that may have application to removing the remnant deposits in the initial screening of technologies. EPA needs to stay within the scope of the defined project. GE believes the EPA should allow GE to determine the impacts of the EPA mandated remedy of the remnant deposits on the rest of the river. GE believes monies expended by EPA to study the remnants area in a way that duplicates work GE is required to perform will not be recoverable by EPA from GE.

Pg. I-3, par. 1: Both GE plants did not begin using PCBs in 1946. The source of information relied upon for making the statements about the history should be identified.

Pg. I-3, par. 2: EPA states that particularly large quantities of PCB were released downstream during spring floods of 1976 and 1983. Records prepared by the NYDEC and reported in the main body of GE comments show that the majority of sediment moved into the Thompson Island Pool in 1973. Lesser amounts moved during the spring of 1974 and lesser amounts during the spring of 1976. The USGS monitoring data indicates some increase above the prior year occurred during the spring of 1984 but nothing like the magnitude of sediment movement that occurred right after the dam was removed. It is also important to note that the vast majority of sediment were trapped behind the dams a short distance downstream of the dam that was removed (limited movement).

Section A - Lower Hudson Characterization

Synopsis, par. 3: The statement made that PCB loads form the lower river sources are of the same magnitude as those form the Upper Hudson River is misleading. The most recent analysis prepared by Thomann, et al. (1989) shows that the vast majority (78%) of PCBs are coming from lower river sources. EPA states (pg. 4-28) that Thomann's estimate of upper river loading may be over-estimated by 90%. This would indicate that Thomann's estimate that 78% of the PCBs in the lower river are from the lower river is probably much greater. EPA appears to have contradictory

information in the report. The statement needs to be revised.

Pg. A. 1-3, par. 2: The presence of a cyclic trend is not obvious in the precipitation data presented in figure A.1.1. A more interesting and relevant observation from this data is that the period of the mid-1970's, experienced the highest sustained precipitation in the entire record and at that period of time the precipitation was not typical.

Pg. A.1-12: The reference to NYDOT in December of 1984 is not in the references section of the report.

Pg. A.1-12 to A.1-1-28, Section A.1.4 (Aquatic Resources of the Lower Hudson): Salinity and dissolved oxygen (DO) are well documented water quality parameters that influence the distribution of aquatic biota. Salinity effects are acknowledged. DO is not discussed. Additionally, the abundance of the fish populations are presented as being principally dependent on abundance of food. Although food availability is important, abiotic factors (temperature, salinity and DO) are also important. EPA should review data on thermal and DO impacts on the distribution and abundance of aquatic biota to present a balanced treatment of the topic.

Pg. A.1-24, par. 2: 3 parts per thousand salinity is equal to 3 ppt, not 0.3 ppt.

Pg. A.2-2, par 2: The section on the physical and chemical properties of PCBs needs to be greatly expanded. EPA needs to compile on a homolog (and possibly on a congener basis) the octonal-water portion coefficients and Henry's Law constants. In the literature there is contradictory information on these types of parameters. A critical analysis of this information for this project, to help understand the processes affecting PCBs in the Hudson, is necessary.

Pg. A.2.2, par. 2: EPA's estimation of PCB load to the lower river from the upper river is greatly flawed. As discussed in Section 6.0 of GE's comments, the rise of PCBs levels in the sediments over time follow a national trend in PCB use. Additionally, the sediment record, as analyzed at Foundry Cove in the lower river, shows a peak in PCB concentration that corresponds to the peak in cesium level which is in 1971. This is 2 years before the dam at Fort Edward was removed.

Additionally, GE believes the core sample analyzed by Dr. Bopp from mile point 188.5 is misinterpreted. The core from mile point 188.5, shows low levels of PCBs in the sediment at depth. In shallower sections of the core there is an abrupt increase in PCB and cesium level. This has been interpreted by Dr. Bopp and Dr. Simpson as showing a build up to a peak (corresponding to the 1973 dam release). This interpretation assumes that the sedimentation

rate in the core was constant with time. This is clearly not in keeping with the facts. After the dam was removed, significant volumes of sediment were transported into the Thompson Island Pool. A more plausible interpretation of the core is that prior to 1973, there were very low levels of PCBs moving past the dam that were removed. This is shown by the low levels of PCBs found in the core at depth. At this same depth, the cesium 137 level is detectable indicating that the deposits occurred after 1954. The abrupt increase in PCB and cesium 137 level occurred in the fall of 1973 (dam removal) and continued through the spring of 1976. The important finding is that prior to the dam removal the transport of particle bound PCB was probably very small. Therefore. contribution to the lower river by the upper river prior to 1973 was probably very small.

EPA's suggestion that the sediment cores confirm that peaks of PCB contamination in the lower river occurred in 1973 is not confirmed by the data. Additionally, prior to 1973 the data do not show that significant PCBs were being transported down stream. The NYDEC sediment data collected in 1977 also shows that the deposit wedge of sediment released during the breaching of the dam in 1973 extended only 20 or so miles down stream of the breached dam. EPA must review all data being relied upon and also must begin looking for the other obvious sources of PCBs in the Hudson River system.

By EPA's own estimate, from 1977 to 1989 approximately 15,000 kilograms of PCBs were transported from the Upper Hudson River to the Lower Hudson River. Even if this estimate is greatly increased to account for the period of time between 1973 to 1977, the numbers still do not show that the Upper Hudson was the major source implied by EPA and others. The discrepancy between this estimate and the estimates of others that show greater amounts of PCBs in the Lower Hudson River sediments is another significant indication that the Upper Hudson River is not the major PCB source to the lower river. It is also significant that by EPA's own admission, the input from the upper river is known with more certainty than the other inputs. The information presented indicates the other sources were and continue to be significant.

Pg. A.2-4, apr. 2: 0.2 lbs/day equals approximately 0.1 kg/day, not 1.0 kg/day.

Pg. A.2-5, Section A.2.6 (Atmospheric Deposition): The discussion on atmospheric deposition of PCB in the Phase 1 Report does not adequately characterize the total atmospheric flux of PCB to the Lower Hudson River. Atmospheric transport and deposition is a complex process which is difficult to quantify. Therefore, extreme caution must be used when applying empirical data from remote sources or limited data from local sources to estimate atmospheric PCB loading. The Phase 1 Report fails to account for the elevated atmospheric concentrations of PCB near urban centers which has been measured to be 5 to 10 times higher than in rural areas (Eisenreich

et al., 1981; Doskey and Andren, 1981). It also relies on estimates from Thomannn et al. (1989) which do not appear to include PCB flux due to dry deposition. Recent data suggests that PCB associated with coarse particles contribute significantly to the total PCB dry deposition flux and that this flux is much greater in urban areas than in nonurban areas (Holsen et al., 1991).

There are no reports of direct measurement of atmospheric deposition of PCB to the Lower Hudson River. Direct measurement would provide the best means of determining the fraction of the total downstream load represented by atmospheric deposition. In the absence of direct measurements, models which incorporate the most recent findings regarding the different forms of atmospheric PCB flux should be utilized.

Considering the lack of direct measurements of Lower Hudson River deposition and recent data underscoring the significance of particulate PCB flux in urban areas, it is evident that EPA estimates of PCB loading based on empirical models of atmospheric PCB deposition are inadequate. Atmospheric flux to the Lower Hudson River is not characterized well enough to estimate the importance of this source.

Pg, A.2-6, par. 2: 1.0 ppb equals 1.0 ug/l, not 1.0 mg/l.

Pg. A.3.1, par. 2: EPA relies heavily on the work of the researchers of Lamont-Doherty Geological Observatory and has proposed funding for them to continue past research efforts for this project (see EPA Phase 2A work plan). In theory the use of radioisotope markers would be useful in establishing absolute dates and then to correlate the dates with events and PCB levels. However, as the published work by the Lamont-Doherty Observatory shows, the vast number of cores do not yield information that is readily interpretable. For those that do, great care in interpreting the results needs to be exercised. As an example is the core obtained from Foundry Cove in the Lower Hudson River. This core shows two peaks in cesium 137 content. The first peak is attributed to the maxima in atmospheric fallout that occurred worldwide in 1963. The second peak is attributed to releases from the nuclear reactor at Indian Point. While GE is still researching the date the second peak occurred, we do have serious concerns with the date of the first peak. While the peak in atmospheric discharge may have occurred in 1963, the peak in river sediments should actually lag behind this by a couple of years while the particles deposited by atmospheric deposition, in the basin are translocated downstream. This shift in the peak to years after 1963 has been documented in Great Lake sediments (Oliver, et al., 1989). Until this is understood the only absolute dates that can reasonably be assigned to the cesium data would be the 1971 data (source is localized and basin lag would not apply) and the 1954 date in which cesium 137 first appeared, and of course the date

which the core was obtained.

EPA also needs to carefully review those interpretations presented that make arbitrary assignments to time based on the PCB level. The 1977 NYSDEC survey indicates that there is a deposition wedge of PCB that extends approximately 20 miles downstream of the dam. This indicates there was not mass movement of PCBs to the lower river on sediment particles in 1973. However, it is assumed by the Lamont-Doherty researches that this mass movement did occur and they arbitrary assigned the peak in PCB in the core from Foundry Cove as occurring in 1973. The actual data form the cesium information demonstrates that the peak actually occurred in 1971.

The Lamont-Doherty work also has limited application to extrapolating any estimates of PCB values to any location other than the cores examined. As is seen in the data supplied in section 6.0 of GE's comments, the point measurements vary by over an order of magnitude in a small distance. This is not unusual for a river system for a contaminant like PCBs, particularly given the following:

1. PCB preferentially bind to fine grained particle

2. The shear stresses vary on a small scale in the river due to local variations in flow velocity

3. There are probably numerous local sources causing great spatial variation.

Therefore, any attempt to take a measurement of sediment must account for all these factors. While a particle of sediment suspended in the water is related (possibly in a very complex way) to the PCB level in the water column, particles are sorted to some extent by size during the sedimentation process so an area of extremely low shear stress will accumulate finer material with higher PCB content, while depositional areas subject to higher stresses will preferentially deposit larger particles that probably have lower amounts of PCBs. Therefore, attempts to back calculate PCB levels in the water column based on sediment measurement are potentially very inaccurate.

EPA must carefully evaluate the techniques that have been applied by others and to objectively evaluate the interpretations that can be made. In fact, EPA should convene a peer review group to evaluate the data generated by Lamont-Doherty Geological Observatory prior to collecting additional core samples.

Pg. A.3-2: Par 2: On Figure A.3-1, EPA presents an interpretation without any apparent attempt to review the underlying data. A person not familiar with what is going on could be mislead into concluding that the figure presents raw data. As will be seen, the assigned dates used as substitutes for the actual raw data

measurement of depth are subject to interpretation.

As an example, when the data were first published in 1985, the graphic display showed a significant PCB increase in 1963 and the peak in 1974. The results are also presented in a 1989 report by Bopp and Simpson (Contaminated Marine Sediments -- Assessment and Remediation: National Academy of Sciences). In the 1989 report, the same data are present on a different time scale. The first increase in PCB level is shown occurring in 1960 and then the peak in PCB concentration is shown in 1973. The fixing of this date appears completely arbitrary and is based on the assumption that the peak corresponds to the dam removal. This change in data presentation and the lack of correspondence to other data calls in serious questions the validity of assumptions derived from this core. GE is very concerned that EPA is not performing a detailed, critical evaluation of this information. EPA has in its possession the information showing these discrepancies. This is even illustrated in the EPA Phase 1 Report. In Figure A.3-1 data on one time scale from the core taken at mile point 188.5 is given. In Figure B.3-6, the same core is shown with a different time scale. GE again requests that EPA not perform additional work along these lines until a detailed critique and review is performed by the EPA.

1. Core from mile point 188.5 - The results of this core were presented in a report dated June 30, 1985 submitted by Bopp et al. to the NYSDEC (Report Number NYS-C00708). The cesium data from Table IV of the report shows that the bottom portion of the core contains detectable levels of cesium in the bottom with low levels of PCB (less than 2 parts per million). There is than an abrupt increase in cesium level and PCB. This abrupt rise in PCB is attributed to the removal of the dam in 1973. The deposition rate during this period was undoubtedly significantly greater than prior to the upstream dam being removed. The most plausible explanation of circumstances from this core is that low levels of PCB transport occurred in the 1960's. After the dam was taken out, a significant amount of sediment was transported into the Thompson Island Pool. This fits with all available data.

2. Core from mile point 53.8: This location is from Foundry Cove, and has been published in a number of articles. This location is within the saltwedge and would be greatly influenced by down stream sources. In an article published by Bopp et al. (1982) Figure 6 clearly shows that the peak in PCB concentration is in approximately 1970 to 1971. This date was set by the 1971 peak in radionuclides related to a known release from a local nuclear power plant. The most obvious conclusion is the peak in PCB content occurred before the dam in Fort

Edward was removed.

3. Core from mile point 88.6: The data from this core is only available to GE in report by Bopp and Simpson (1989). The data presents PCB by year instead of by depth. The cesium data is not presented. Due to the problems noted with other interpretations, the actual raw data should be obtained and reviewed. GE requests EPA make this date available to GE.

4. Core from mile point 91.8: In this core the cesium 137 values in the lower part of the core are not reported (54-40 centimeters). The PCB values are very low in this section of core. An abrupt peak in cesium occurs at 24-28 centimeters and than shows a fairly steady level until the core was collected in 1977. The peak in cesium is reported as being 1963. The PCB levels increase to a peak of 28.8 parts per million. This peak is arbitrarily selected as occurring in 1973 to coincide with the removal of the dam. There is no independent data that can be used to show that this approach (i.e. PCB peak equals 1973) is valid in this part of the river. It is not known if this area had ever been dredged, had constant sedimentation rates (or nearly constant), etc. This arbitrary use of assigning the PCB peak to 1973 also does not fit what was believed to be a 3 year period (until the 1976 flood) of significant sediment movement within the Upper Hudson River.

5. Cored from mile points -1.65 and -1.7: The data from these core is only presented in the paper by Bopp and Simpson (1989) and the raw uninterpreted cesium data is not present. EPA should obtain and review this data and make it available to GE and others for review. The data as presented in the EPA figure shows a significant peak in 1970, well before the removal of the Fort Edward Dam.

From the data, and the fact that out of the cores obtained (in excess of 35), only a small number (4?) have any utility arises a serious concern about the viability of the techniques and also any reliance placed upon it by EPA. EPA should convene a peer review group to evaluate the data generated by the Lamont-Doherty Geological Observatory prior to collecting additional core samples.

Pg. A.3-2, par. 2: EPA states, with out any supporting information, that the increase in PCB content of the sediments from what they believe is 1954 to 1970 as being due to GE. GE requests specific data relied upon to come to such a conclusion. The increase in PCB in the sediment, as well as the apparent decrease in chlorination level, as well as the nation peak usage patterns shows that the other numerous, significant PCB sources that EPA continues to refuse to investigate, are just as likely a cause.

Pg. A.3-2, par 2: EPA states that dam removal is seen in all the cores of the lower Hudson as a peak in PCB values corresponding to 1973. As discussed above, the peak in cores in the lower Hudson occurred before 1973. The peak in PCB value has been assumed by many to correlate to the removal of the dam at Fort Edward. There is no independent data that shows this to be the cause in the lower river. At best, the work by the Lamont-Doherty Geological Observatory gives evidence showing the peak actually occurred before the dam was removed. This, combined with the data from the upper river showing that prior to the dam removal in the Thompson Island Pool the PCB concentrations in the sediments immediately below the Fort Edward Dam were very low, shows that the theory that the PCBs in the lower river are mainly from the upper river is unsupportable.

Pg. A.3-2, par 2: EPA attributes the decline in PCBs since 1977 to the reworking of the sediments released from behind the dam at Fort Edward and also to the discontinued use by GE. While this may be a theory that could be tested, the data presented does not prove this to be the case. The data also shows nationwide declines that have occurred due to the limitations on use placed upon PCBs.

Pg. A.3-2, par 3: GE agrees that the data indicates the presence of PCB sources other than those from the upper river.

Pg. A.3-2, par. 3: EPA concludes from the data presented in Figure A.3-1, that not only did higher chlorinated congeners in the cores come from the upper river but they also come from the GE plant and also match discharge records. This is pure speculation and not supported by the evidence. This conclusion is based on data from two cores in a 200 mile stretch of a river that has been industrialized for over 100 years. GE requests that EPA remove this statement from the document.

Pg. A.3-6, par. 2: EPA appears to agree that one could estimate the water column concentration of PCBs by simply looking at the concentration of PCBs in the sediments. This technique employs numerous assumptions, not the least of which is knowing the time history of deposition. If EPA accepts this approach as valid, GE believes a more through discussion and analysis is warranted.

Pg. A.3-7, par. 2: EPA seemed to accept without question the opinion that the reason the striped bass fishery is closed is due to the presence of PCBs. EPA needs to perform a thorough analysis of this issue and will find that fish conservation motives have long been an important consideration in setting fishing restrictions.

Pg. A.3-8, par 1: It is indicated that the EPA has the results of PCB analysis of fish from the NYDEC in a data base. GE requests that this data be made available to ensure the data base supplied to GE by the NYDEC is the same data base being employed by EPA.

Pg. A.3-8, par. 1: The citation to the Nadea and Davis report of 1974 is interesting since it concludes that there were other sources of Aroclor 1016 upstream of Fort Edward and that GE was not the sole source of PCB contamination. EPA should consider this information in the areas where GE is alone singled out by EPA as being responsible for the PCBs in the Upper (and Lower) Hudson River.

Pg. A.3.8, par. 3: The conclusions given in this paragraph are not attributed to anyone. Where did this information come from and can GE get access to it for our review?

Pg. A.3-9, par. 2: GE is concerned with the assessment that the PCBs in the striped bass caught in the river during spring migration bear little resemblance to PCBs found in the location the fish were caught (i.e. sediment, water, or food). EPA needs to evaluate the information presented by GE in the main body of these comments that show the importance of PCB sources outside of the river where these migratory fish obtain the majority of there PCB body burden.

Pg. A.3-9, par. 3: The Sloan et al. (1988) report mentions that not only was the detection limit for Aroclor 1221 changed but also that for all the measured Aroclors. The Phase 1 Report should be modified to reflect this.

Pg. A.3.9, par. 4: In the next to the last sentence of the paragraph the date should refer to 1987 instead of 1988. It is our understanding the results were reported in 1988.

Figure A.3.4: The decline in PCB in the striped bass is very apparent in this figure. It also shows that the increase in PCB in the fish in the early 1980's was due to primarily Aroclor 1254. This rise of PCB has been reported in the early 1980's in a number of places and appear to be due to a source of PCBs with a chlorination levels higher than those present in the sediments in the upper river.

Pg. A.3-10, par. 1: The fourth sentence states that the levels of Aroclor 1016 were relatively constant between 1983 and 1987. However, the April 1987 report from Sloan et al. reflects that the levels of Aroclor 1016 were also declining. The report should be modified to reflect this fact.

<u>Section B - Upper Hudson Characterization</u>

Pg. B.1-3 to B.1-4, Section B.1.2.1 (Water Quality): The EPA Phase 1 Report discusses the results of the 1987-1988 Rotating Intensive Basin Studies (RIBS) Report (NYSDEC, 1990, Phase 1 Report, pages B.1-3 and B.1.4). Portions of the RIBS Report summarize the results of surface water monitoring of the Hudson River. The Phase 1 Report's discussion of the "parameters of concern" identified in

the RIBS Report lists copper and iron as the only constituents "detected at elevated concentrations with sufficient frequency to be considered parameters of concern." (Page B.1-4). However, the RIBS Report identifies cadmium, copper, iron, lead, phenol, total coliform and fecal coliform as "parameters of concern" at concentrations exceeding criteria.

The identification of these additional "parameters of concern" in the Hudson River indicates that PCBs are not the only compounds impacting water quality in the Hudson River. Should a large scale program focusing on the removal of PCBs from the Hudson River be implemented, the presence of these non-PCB parameters in the Upper Hudson River as residuals could represent a condition where the overall water quality of the Upper Hudson River has not improved significantly.

A summary and discussion of the RIBS document, including the results of our review of the RIBS data, is presented below.

The RIBS program incorporates water column, bottom sediment, macroinvertebrate and fish monitoring to produce an integrated approach to the assessment of ambient water quality in New York State. The 17 drainage basins in New York State were divided into three basin groupings which were each intensively monitored for two year. As such, during a six year period, all basins are monitored and the cycle is repeated every seventh year. Water quality is evaluated based on a comparison of detected contaminant concentrations with assessment criteria for 20 parameters presented in the document. These 20 parameters were selected because they have standards and/or guidance values and have been consistently detected above their analytical reporting levels during previous studies. Due to analytical variability, different sampling/analytical methods, and differences in criteria specifications and analytical reports, water quality is evaluated based on a relative degree of adherence to standards, rather than a precise quantification. The following table presents the assessment criteria used in the RIBS program (NYSDEC, 1990).

RIBS ASSESSMENT CRITERIA

PARAMETER	CRITERIA				
Water Temperature	> 25° C				
pH	< 6.5 or > 8.5				
Dissolved Oxygen	< 5.0 mg/l				
Phenol	< 1.0 ug/l				
Dissolved Solids	> 500 mg/l				
Ammonia	> 0.164 mg/l*				
Total Coliform	median > 2400/100ml 20% > 5000/100ml				
Fecal Coliform	Geometric Mean > 200/100ml				
Trichlorethylene	> 1.0 ug/l				
Chloroform	> 1.0 ug/l				
1,1,1-Trichloroethane	> 1.0 ug/l				
Aluminum	> 100 ug/l				
Mercury	> 0.2 ug/l				
Zinc	> 30 ug/l				
Iron	> 300 ug/l				
Manganese	>300 ug/l				
Hardness as $CaCO_3$	10-40	50-77	80-102	120	185
Cadmium**	≥ 1	≥ 1	> 1.3	> 1.3	> 1.5
Copper**	> 1	> 7	> 11	> 13	> 20
Lead**	≥ 5	≥ 5	≥ 5	> 3.7	> 7.0
Nickel**	> 40	> 68	> 90	> 108	> 150

* This value represents worst case conditions for pH and temperature. Any reported ammonia value which exceeded this screening criteria was compared to a computed criteria for the actual reported pH and temperature on that date.

** Assessment criteria based on standards that are hardness dependent.

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Under the RIBS program, a parameter which exceeds its assessment criteria more than 15 percent of the time is considered a "parameter of concern." The identification of "parameters of concern" is used to focus attention on these parameters in other media sampled to evaluate specific impacts. "Parameters of concern" were identified in each of the five locations on the Upper Hudson River including Corinth, Fort Edward, Schuylerville, and Waterford. Results from each of these monitoring locations are discussed below.

Hudson River at Waterford - The RIBS document identified cadmium, copper, lead, phenol, total coliform, and fecal coliform as "parameters of concern" in the water column at Waterford. The Phase 1 Report does not discuss this location or the identified contaminants in its discussion of water quality and the RIBS document (Phase 1 Report, pages B.1.2-B.1.5). A review of the analytical data generated during the RIBS program indicates that aluminum and iron also qualify as "parameters of concern" as they were detected above the assessment criteria 89 percent and 44 percent of the time, respectively. It should also be noted that the assessment criteria for manganese at 300 ug/1 is well above the federal MCL of 50 ug/1. Using 50 ug/1 as an assessment criteria could include manganese as a "parameter of concern" for Waterford due to its detection at or above 50 ug/1 38 per cent of the time monitored.

Hudson River at Schuylerville - The RIBS document identified copper and iron as "parameters of concern" in the water column in Schuylerville. This fact is accurately presented in the Phase 1 Report. However, a review of the analytical data generated during the RIBS program indicates that phenols (22% excedance), total coliform (79% excedance), aluminum (56%), cadmium (25%), and lead (19%) would also qualify as "parameters of concern" at Schuylerville.

Hudson River at Fort Edward - The Phase 1 Report correctly summarized the RIBS document which identified copper as a "parameter of concern" at Fort Edward. However, the RIBS data indicate that total coliform (100%), fecal coliform (100%), aluminum (38%), cadmium (29%), and lead (29%) would also qualify as "parameters of concern" at Fort Edward.

Hudson River at Corinth - The Phase 1 Report acknowledges that copper was found in water column samples from the Hudson River at Corinth. The RIBS document identifies copper and lead as "parameters of concern". The RIBS data also indicate that aluminum (22%) and cadmium (21%) qualify as "parameters of concern" at Corinth.

Hudson River at North Creek - The Phase 1 Report acknowledges that copper was found in water column samples from the Hudson River at North Creek. However, the RIBS document identifies cadmium, copper

and lead as "parameters of concern". Moreover, the RIBS data indicate that aluminum (56%) qualified as a "parameter of concern" at North Creek.

Conclusions - The RIBS Program (NYSDEC, 1990) has provided data that indicates the presence of cadmium, copper, lead, total coliform, fecal coliform, aluminum, manganese, iron, and phenols at concentrations and frequencies which qualify them as "parameters of concern". Several of these parameters were also present in background water column samples collected from the Upper Hudson River at Corinth and North Creek. It is recommended that the data generated by the RIBS Program on the parameters, other than PCB, which were detected at concentrations exceeding regulatory criteria be reevaluated to determine their significance to Upper Hudson River water quality and that this be factored into the RRI/FS.

Pg. B.1-5, Section B.1.2.2: The Phase 1 Report discusses the use of Hudson River water as a raw source for public drinking water. The report states (Phase 1 Report, page B.1-5):

> The Hudson River is used as a source for public water supplies (municipal and institutional drinking water) in sections of the river classified as Class AA or A. Along the Upper Hudson, three communities draw directly Hudson River water.

Although this statement is accurate, it should be noted that: 1) two of these three communities are located upstream of the regions of the river considered as the site for EPA's Reassessment RI/FS, and 2) raw water from the Hudson River is treated in municipal water treatment plants before it is distributed to the public. Monitoring data from Hudson River water treatment plants indicates that the treatments are very efficient (up to 98 percent removal) at removing PCB from raw Hudson River water. In addition, although the raw water is treated, the average raw water PCB concentrations have been below the federal MCL of 0.5 ppb and the NYSDOH action level of 0.1 ppb since September 1983 (Metcalf and Eddy, 1990). Both of these factors should be discussed and referenced in order to provide a balanced perspective on the implications of the uses of Hudson River water as a drinking water source by these three communities.

Drinking Water Use

Queensbury and Winebrook Hills - The three communities which draw Hudson River water for public water supply are the Town of Waterford in Saratoga County, the Town of Queensbury in Warren County, and the Winebrook Hills Water District in Essex County. The intakes for Queensbury and Winebrook Hills are located upstream of Glens Falls. According to the Phase 1 Report, the USGS monitoring station of Glens Falls provided upstream background levels of PCB in the Hudson River from 1977 - 1983. Of 45 observations for total PCB, only two had detectable levels of PCB.

These observations occurred on December 5, 1978 and September 28, 1990 and were both reported at 0.1 ug/l. Referencing the fact that these two communities' water intakes are upstream of Glens Falls would clarify that users of water from these facilities do not have the potential to ingest sediment-borne PCB which may have originated downstream of Glens Falls. However, it is significant to note that the water treatment plants for Queensbury and Winebrook Hills, although upstream of PCB contamination, also utilize coagulation/filtration treatments, which have been demonstrated to effectively remove waterborne PCB (O'Brien & Gere, 1981).

Waterford - The Waterford water treatment plant (Waterford WTP) is the closest downstream public water source to Glens Falls. The Waterford WTP utilizes a coagulation/filtration treatment process which is typically used to treat surface water for drinking water supplies. The coagulation/filtration treatment works via the following steps (AWWA, 1971):

1. a coagulant is mixed with raw water;
2. the coagulant de-stabilizes the particulate material and also forms a precipitate (floc);
3. the floc is mixed to increase the size of the floc particles;
4. the floc is settled and the resulting sludge is withdrawn; and
5. the supernatant is filtered to remove any floc that did not settle.

Since PCB absorb strongly to solids such as sediments (log K^{OC} values from 4.40 to 7.64 [USEPA, 1983]), effective removal of particulates results in removal of a majority of the PCB. The efficiency of coagulation/filtration treatment technologies in removing PCB from Hudson River water was studied by O'Brien and Gere in 1981 as part of a report prepared for NYSDEC entitled "Hudson River Water Treatability Study" (O'Brien & Gere,1981). The study included a series of bench scale jar tests to evaluate optimum conditions for coagulation of Hudson River water. Coagulant, additive, and pH were varied in the tests to evaluate the removal of particulates and turbidity. Results of the bench tests indicated that removal efficiencies could reach 99 percent by regulating the process variables discussed above, but that 90 percent removals by the coagulation/filtration method represent the highest reduction consistently achievable in existing water treatment plants due to the degree of control required to meet changing raw water conditions.

The conditions that gave the best removals of turbidity and particulates in the study (alum coagulant at a dose of 30 to 40 mg/l at a pH of 6.9), similar to the conditions found at the Waterford WTP, appeared to be providing sufficient treatment with the coagulant type and dosages in use at the time of the study.

The results of the 1981 bench scale study are supported by monitoring results at the Waterford WTP. Based on raw water and finished water PCB monitoring at the Waterford WTP between 1975 and 1983, the Waterford WTP averaged an 86 percent removal rate for PCB (USEPA), 1987). These data verify the ability of the coagulation/filtration treatment to efficiently remove PCB in practice.

Action Levels and Standards - Irrespective of the ability of current water treatment practices to effectively remove PCB, monitoring of Hudson River surface water has shown that it meets the NYSDOH established action level for PCB of 0.1 ug/l. PCB concentrations in the Hudson River have been regularly monitored since as early as 1975 by various programs under the USGS, the Waterford Water Works, and NYSDOH. As given in the Phase 1 Report, the highest PCB concentrations in the water column since 1985 were detected at Fort Edward, where the upper 95th percent confidence interval on the adjusted mean is 0.06 ppb. Based solely on PCB concentrations, this water would be an acceptable drinking water source, even without treatment, according to the NYSDOH established action level of 0.1 ppb. NYSDOH has used the National Academy of Science value of 0.16 ug/l PCB to set the action level of 0.1 up/l PCB.

The overall NYSDOH guideline for PCB drinking water is 1 ug/l (NYSDEC, 1986). At or above a PCB concentration of 0.1 ug/l, additional monitoring is triggered and steps are taken to reduce the PCB concentration to below 0.1 ug/l. Drinking water with a PCB concentration at or above 1 ug/l is considered by NYSDOH unfit for human consumption.

There are currently no federal or state drinking water standards for PCB. However, on July 30, 1992 a federal Maximum Contaminant Level (MCL) of 0.5 ug/l will be established for PCB drinking water (40 CFR 141.61). An MCL is defined as the maximum permissible level of a contaminant in water which is delivered to any user of a public water system. Therefore, detected concentrations of PCB in the Upper Hudson River have been below the established enforceable standards and criteria since 1983. It is, therefore, suggested that reference to this fact be added into section B.1.2.2, to provide a frame of reference to the statement that the Hudson River is used as a source of drinking water.

The Phase 1 Report states (Section B.1.2.2, page B.1-5):

Hudson River water is also used for domestic (watering lawns and gardens) and agricultural purposes (irrigating crops).

The source or basis for the statement that there are domestic and agricultural uses of the Hudson River was not provided. Personal communications with County and State Health Departments and NYSDEC indicated that there are few residences or agricultural lands adjacent to the Hudson River which would rely on the Hudson River for water supply. It should also be noted that Upper Hudson River water has consistently been below regulatory limits for PCB since 1983 (Metcalf and Eddy, 1990). Therefore, the use of Hudson River water for residential gardens or agricultural irrigation would not impact crop or garden food quality. This statement should be made as a frame of reference for evaluating the implications of the irrigation and agricultural uses of the Hudson River.

Although Upper Hudson River water is used as a drinking water source, the water is treated at municipal water treatment plants by methodologies which have demonstrated efficiency in the removal of PCB. Irrespective of the treatment, recent Upper Hudson River water monitoring data for PCB indicates the levels of PCB have consistently been below the federal MCL of 0.5 ug/l and at or below the NYSDOH action level of 0.1 ppb with an estimated mean PCB concentration of 0.6 ug/l. Furthermore, this data indicates that the use of Hudson River water for domestic uses other than drinking does not pose a significant route for human exposure.

It is recommended that, when discussing the uses of the Hudson River as a source of drinking water or irrigation, the Phase 1 Report reference the following facts:

- Municipal water uses of the Hudson River include treatment by coagulation/precipitation which is effective at removing PCB.
- Raw surface water from the Hudson River meets NYSDOH drinking water guidelines of 0.1 ug/l PCB.

Pg. B.1-7, par 1: EPA states that the barge canal has experienced a decline in recreational use. This is factually incorrect. The attached figure, derived from NY Department of Transportation records (NY DOT, 1989), clearly show an increasing use of the barge canal for recreation use.

Pg. B.2-1: The estimates of PCB discharges by GE as given by EPA and others are very speculative. This is particularly true of those that assume all PCBs in the Upper and Lower Hudson River came from the GE Plants in Fort Edward and Hudson Falls. Additionally, the use of only production records of PCB purchase records do not, by themselves provide useful information on actual discharges. One also has to consider the significant changes in production and waste generation processes that occurred over time. EPA should not

employ the 1.3 million pound figure since it is not based on a reliable estimating technique.

Pg. B.3-1, Section B.3.1 (Overview of Sources and Data Base): The explanation of the electronic data base developed by EPA to manage the extensive data available on the Hudson River System is useful. A further explanation of EPA's review of the data input to ensure that transcription errors did not occur would also be useful. Additionally, GE requests that EPA make available to GE and others a copy of data files in electronic format as well as make available copies of all the original hard copy files of the data (i.e. reports , laboratory data sheets, etc.).

Pg. B.3-1, par. 2: EPA did not consider the following publically available data (as presented in Table B.3-1):

1. 1975 report by the Division of Pure Waters (NY) on the PCB content of sediment, water and effluent. This report contains data on approximately 15 sediment samples from the Upper Hudson River and 15 sediment samples from tributaries. Additionally, PCB measurements are reported for a number of potential PCB dischargers (Sprague Electric, Glens Falls Landfill, Glens Falls Portland Cement, Saratoga Board Mill, Galente Company, and the Jard Company.)

2. The 1983 Gahagen & Bryant Probing Report. Although this report does not contain PCB analysis of sediments, it does provide information on sediment textures and bed elevations.

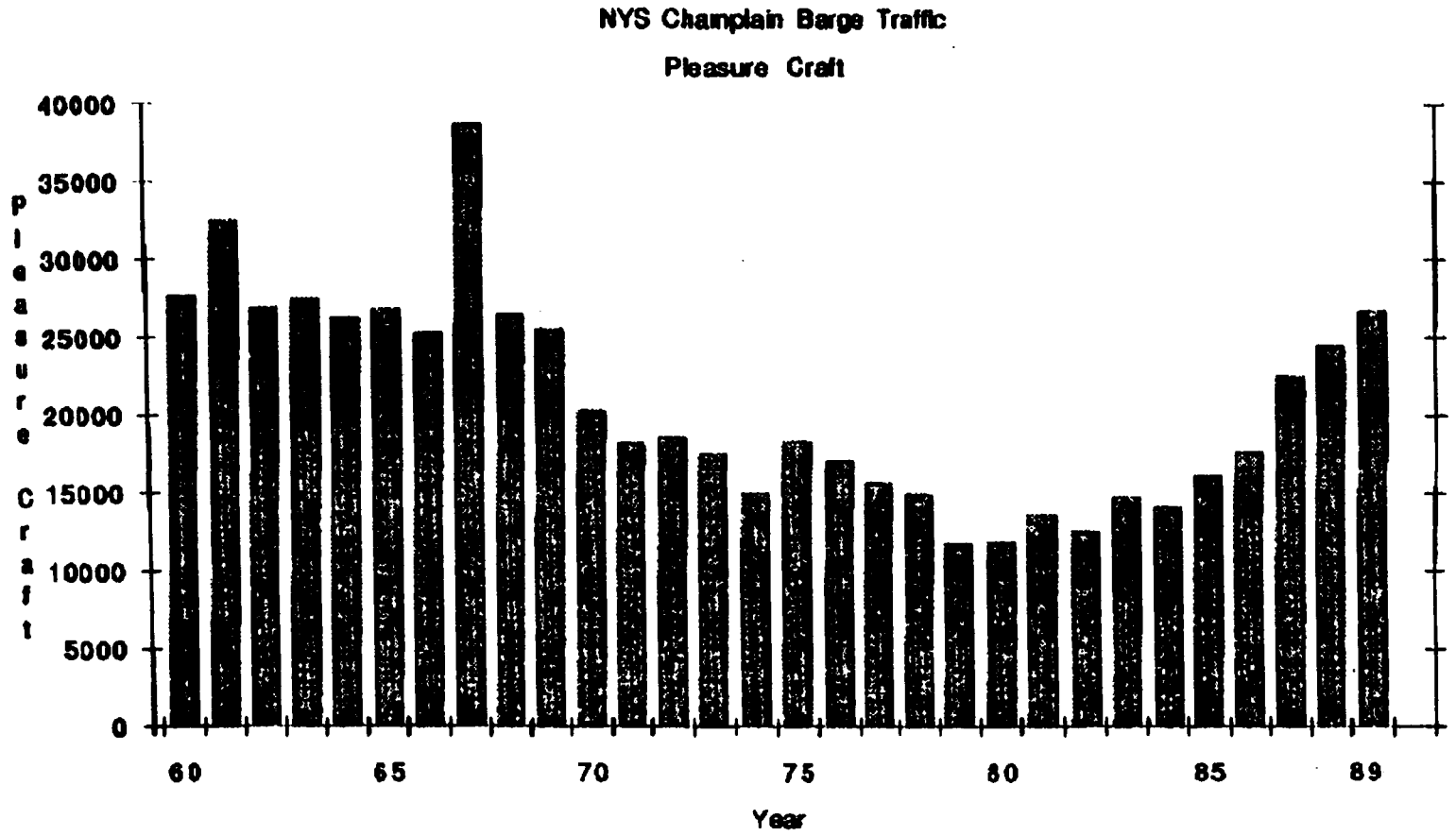
3. A 1989 report by NYDOT by Long Lake Energy Corporation that found that the maximum concentration at "Hot Spot" 34 (Northumberland Dam) to be 10 parts per million instead of the 500 parts per million reported in 1977.

4. As part of the RCRA Closure and Corrective Action permit, Ciba Geigy has generated a significant amount of information on sediment and fish contamination above Fort Edward.

Pg. B.3-5, par. 4: EPA reports they do not have detection limits for the 1976-1978 sediment samples. GE has this data and would be glad to share it with EPA if it is so desired.

Pg. B.3-6, par. 3: The statement concerning the NYDEC designation of 100 polygons containing PCBs greater than 50 parts per million of PCBs is not correct. The NYDEC defined 138 polygons in the Thompson Island Pool. Of this number, only 14 had an average PCB concentration (as defind by the NYDEC) greater than 50 parts per million. The text should be corrected.

Pg. B.3-9, par. 2: EPA's approach for calculating average PCB concentations in the sediment from the 1984 data greatly overestimated the PCB levels. In the 1984 data, all samples were


NYS Champlain Barge Traffic
Pleasure Craft
40000
35000
30000
25000
20000
15000
10000
5000
0
Pleasure Craft
60
65
70
75
80
85
89
Year

screened by mass spectrometer, a semi-quantitative technique used to classify sediments into four ranges, less than 10 parts per million (ppm), 10 - 50 ppm, 50 - 100 ppm, and greater than 100 ppm. Many of the samples that were "cold" (less than 10 ppm), were never analyzed by the more precise gas chromatograhic techniques. DEC's screening technique precludes direct comparison of the two data sets. However, it is clear that not including these "cold" samples in the analysis of average concentrations will elevate the calculated averages. The EPA ignored the results from over 400 grab samples and 600 cores. Even if the data points screened as being less than 10 ppm are conservatively assigned a value of 5 ppm (one half the detection limit) the calculated average will be cut nearly in half (approximately 25 ppm).

Pg. B.3-12, par 1: As discussed earlier, the use of radionuclied dating of sediments form the Upper Hudson River have not yielded conclusive results. Furthermore, it is clear that EPA must perform a through review of the data presented by the Lamont-Doherty Geological Observatory researchers. The interpretation of the data presented in Figure B.3.6 is far from unambiguous. When the raw data is reviewed in context of other data it can clearly show that prior to the dam being removed (early to late 1960's when cesium 137 was present) the amount of PCB in the sediment from above the Fort Edward Dam was fairly small indicting PCB discharges where being effectively contained behind the dam. After the dam was removed, a large amount of sediment moved very quickly (non-uniform sedimentation rate) into the Thompson Island Pool. The best marker for the 1973 date is not the peak PCB level, but rather the point of sharp increase from low to high PCB content. This conclusion is much different from that offered by EPA, NYDEC, or Lamont-Doherty.

Pg. B.3-12, par. 2: GE concurs with the presence of highly altered PCB due to biological degradation in the Upper River. In fact, it was not Bopp who first reported this, but rather John Brown from GE's Corporate Research and Development Center. Additionally, what is probably more significant, is that the vast majority of sediments in the Upper Hudson River have had significant amounts of chlorine removed and this alone may account for the mass reduction seen between the 1977 and 1984 sediment surveys. This will require much further analysis to confirm. The extent of anaerobic dechlorination is described in the main portion of GE comments.

Pg. B.3-14, par. 2: The interpretation of the data provided by GE as presented in Table B.3-8 is incorrect. EPA admitted this deficiency at a recent public meeting. A revised table needs to be made available to GE and others. Additionally, EPA needs to revise the text that derived erroneous conclusions from the misinterpreted data in the table.

Pg. B.3-15, par. 1: While a limited number of tests have been performed on metals leachability, the presence of the metals in the sediments may make any proposed treatment and disposal more

complicated. Additionally, while it is thought the metals may not be readily leachable, if the reason for this is the presence of highly organic matrix under reducing conditions, than if treatment to destroy PCBs also changes these conditions, these metals may become more readily available. It is also intersting to note that the "parameters of concern" defined in the RIB's program includes a number of the metals seen in elevated levels in the sediment.

Pg. B.3-17, par. 2: With respect to discrepancies in the data base, GE agrees that this does present some limitations to interpretation and use. To better understand why the data base employed by EPA does not match that discussed in the literature, GE requests access to the data base being utilized by EPA.

Pg. B.3-19, par. 1: EPA states that several major flood events were associated with the mass erosion of the remnant deposits. What information does EPA have to show there was mass erosion and that this coincided with flood events?

Pg. B.3-20, par. 4: In Table B.3-10, EPA presents summary statistics on the suspended sediments measurements made in the Upper Hudson River by the U.S. Geological Survey. There is a difference between the number of measurements of the suspended sediment concentration and the number of sediment load data points. An explanation as to why this is would be helpful.

Pg. B.3-20, par. 3: Why is the data prepared by the U.S. Geological Survey, a sister federal government agency, available to the NYDEC yet not the EPA?

Pg. B.3-21, par. 3: The detection limit of the U.S. Geological Survey's PCB analytical method is reported as changing from 0.1 milligrams per liter to 0.01 milligrams per liter in 1984. Is this a result due to a change in the sample preparation, analytical techniques or quantitation method or some combination of the above? Does EPA have a written description of any of the analytical or sampling methods employed by the U.S. Geological Survey? Have these been evaluated to see if they are "acceptable"?

Pg. B.3-21, par. 5: In Table B.3-11, the average PCB values for the water column monitoring stations are calculated. Is this an arithmetic average? In environmental samples, many parameters are found to be log-normally distributed, in which case the geometric mean is a better measure of central tendency. Did EPA determine the distribution of the PCB values to see if they were normally distributed? Is the reported standard deviation based on the nondetects reported at the detection limit or as being one half the detection limit?

Pg. B.3-22, par. 1: The observation that the water column data suggests that there may be little loss (or addition) of PCB during transit in the Upper Hudson River is important. If this is

generally the case, then the importance of the source of PCBs between the Fort Edward monitoring station and the Glens Falls monitoring station becomes the focus point of the investigation. EPA should make every attempt to obtain the most recent monitoring data from the U.S. Geological Survey as this is the only information available on the water column monitoring since the EPA mandated remnants remedy occurred. If EPA is not going to do this, is EPA going to collect water column samples on a routine basis for a period sufficient to monitor the variation over a complete cycle of high to low flows (one year?)? The importance of this PCB source is apparent in the data shown in Table B.3-12.

Pg. B.3-23, par. 1: EPA poses a question on whether there has been any genuine trend in PCB loading to water over time, or whether the apparent year to year trends are due to actual variability in the hydrolog. All the data presented by EPA demonstrate that there has been a significant reduction in PCB level and load in the river over time. Why EPA would think this is not genuine is not known. There is a downward trend over time in the PCB water column values.

Pg. B.3-23, par. 4: Are the reported means geometric means (i.e. based on an assumption of a log-normal distribution)?

Pg. B.3-23, par. 4: In Table B.3-12, it appears as if the mean values were calculated using data from 1986 to 1989. According to EPA, one purpose of calculating the current year mean is as an input parameter to the risk assessment. GE believes this method greatly overestimates the PCB values that will be present over the period of potential exposure (the next thirty years). A more appropriate method is to note the obvious decline in PCB levels in the water column and to project the values out over the next thirty years, using something like an exponential decay function.

Pg. B.3-26, par. 4: EPA is attributing the higher PCB levels seen in the summer to the presence of boat traffic and the increased use of locks. EPA believes that this causes an increase in suspended sediment load and therefore PCB level. Does EPA really believe this to be the cause and has the suspended sediment measurements from low flow periods been evaluated to see if this theory is supported by data?

Pg. B.3-27, par. 1: The theory presented that little or no PCBs are being release from the sediment anaerobic zone is based upon the reconstruction of PCB homologs from packed column analysis. While this is an interesting theory, it is also possible that PCBs, if released from the anaerobic zone into the aerobic zone, may be undergoing complete biological destruction since they will contain predominately mono and di PCBs. Additionally, GE believes the old data being relied upon for reaching such important conclusions should be replaced with data from capillary column GC-ECD that is collected using appropriate sample handling and preservation techniques.

Pg. B.3-27, par. 3: GE request that EPA make available to it the referenced data that was given to EPA from the NYDOH on PCB levels in the water column samples.

Pg. B.3-29, par. 1: It is claimed that the spring flows during the 1980's were abnormally low. What data is the basis for this conclusion? Is this a qualitative statement that should rather indicate that the peak flows in the 1980,s are lower than those seen during the 1970's. By all indications the peak flows in the 1970's were abnormally high (see Figure B.3-7). Does this indicate that the flows in the 1980's are actually normal?

Pg. B.3-19, par. 4: GE requests that EPA provide access to the fish data base being employed (electronic form). GE believes it has the same information (supplied by Ron Sloan of the NYDEC) yet GE is not able to match some of the results presented by EPA.

Pg. B.3-33, par. 2: EPA reports the mean level of PCB from all fish in the Upper Hudson River from the years 1986-1988. Did EPA employ a mean assuming that the population (as weighted by population) is normally distributed? Is a geometric mean a better measure of central tendency in this case? Is it also reasonable to average all the fish in the data base (1986-1988) which may include a large number of fish species that will not be preferentially consumed if caught or that may not be representative of the actual distribution (as weighted by population) of the fish species in the river?

Pg. B.3-33, par. 2: EPA's use of a constant PCB value for fish in a risk assessment that assumes that exposure will occur over a thirty year period is outrageous. All the data presented by EPA and an analysis of the physical, chemical and biological processes taking place in the river clearly shows that a significant downward trend in PCB concentration should be employed for such an assessment.

Pg. B.3-39, par. 3: It appears that EPA may have misstated the explanation for the absence of lower chlorinated congeners in the chironomid and the absence of the higher chlorinated congeners in the water. The statement on the last sentence of the paragraph should read that the higher chlorinated congener were present in concentrations below detection limits.

Pg. B.3-41, par. 3 and 4: Reference is made to air data generated jointly by USEPA and NYDEC in 1987. Reference is also made to other air monitoring events conducted by the NYDEC in 1987, yet specific data is not supplied. Specific reference is not included in the references cited section to these investigations so GE is unable to review the cited information. GE requests that this data be made available to GE and others.

Pg. B.3-42, par. 2: EPA cites data from a state wide PCB air monitoring program and compares it to the local data generated over time in the Fort Edward area and concludes that the maximum concentrations are higher in the Fort Edward area. Are the data sets comparable in technique of collection? Did the state wide samples focus on potential local sources or on general ambient air quality? Were samples from Fort Edward biased to attempt to maximize the PCB concentration levels detected?

Pg. B.3-42 to B.3-43: The Phase 1 Report discusses the potential for volatilization of PCB to the air in the Lower Hudson River by citing a study on PCB transport performed by Bopp (1983). The Phase 1 Report, EPA states (Phase 1 Report, page B.3-43):

> For low chlorinated homologues (di- and trichlorobiphenyls), Bopp (1983) estimates that about 40 percent is lost, because of gas exchange, as a given parcel or water travels from Troy to Poughkeepsie. For the tetra- and pentachlorobiphenyls 10 to 20 percent is lost as a result of gas exchange.

Volatilization in the Hudson River is controlled by a number of factors, many of which were not taken into account by Bopp (1983) in verifying his model-driven estimate. Because of competing sinks and sources, it is likely that volatilization of PCB from the Hudson does not occur as stated in the Phase 1 Report. A qualifying statement regarding the referenced estimate should be provided in the Phase 1 Report for the following reasons:

- The values for the physiochemical constants utilized in the formula for volatilization by Bopp (1983) were derived solely on a theoretical basis and could not be verified by measurement of those constants.

- Transport of water and suspended sediment in the Hudson River was assumed by Bopp (1983) to take place according to "plug flow" and did not consider interactions between the water column and deposited sediments.

- Seasonal variations in temperature, wind and flow rates, sediment type, PCB levels and sediment load, as well as physical, chemical or biological degradative processes were not considered in the generation of the estimate.

- The model was verified using water column data only, collected at two locations at one point in time, and did not consider verifying assumptions through measurement of PCB in deposited sediments at the two locations.

- Bopp states in the paper that he does not intend for his proposed model to be used as a quantitative measure of mass flux.

Bopp adopted the gas exchange model developed by Liss and Slater (1974) to describe PCB flux associated with the Hudson River. He rearranged and simplified the equation and utilized predictive equations to determine input variables in order to develop an equation for a homolog-specific gas exchange constant computed as a function of Henry's Law constants, film thickness, and diffusion coefficients. The procedures used to generate these parameters are described below.

Henry's Law constants were computed by grouping PCB congeners according to degree of chlorination as estimated by the elution order of major chromatogram peaks. The activity coefficients were then estimated for each peak by means of graphical interpolation based on the number of chlorine atoms and the estimated activity of the Aroclor mixture. Vapor pressure was calculated using a theorized linear extrapolation of vapor pressure over the number of chlorine molecules.

Diffusion coefficients were computed theoretically using a Wilke Chang (1955) equation, modified for aqueous solutions by Hayduk and Laudie (1974). A boundard layer thickness of 1.8 x 10^{-2}cm for water in the Hudson River was assumed as per Emerson (1975), while a gaseous boundard layer thickness of 1 cm was assumed based on an evaporation model proposed by Sverdrup (Defant, 1961).

Utilizing the above described theoretical equations and estimates and the Liss and Slater (1974) equation, Bopp generated a half-life equation for homolog-specific PCB volatilization taking in to consideration: 1) an estimate of the mass of suspended matter, 2) an estimate of the average depth of the river, 3) a sediment/water distribution coefficient calculated by analyzing PCB in two samples of filtered and unfiltered Hudson River water, and , 4) the gas exchange constant for each chromatogram peak as calculated via the previously estimated physicochemical constants.

The resultant values generated by the equation were "verified" over the stretch of the Hudson from Poughkeepsie to Troy, New York. First, an estimate of residence time was developed based on calculated water transit times between Poughkeepsie and Troy. Predicted values for each homolog at Poughkeepsie were then generated for each homolog based on the concentrations at Troy and the half-life equation. The predicted water column concentrations at Poughkeepsie based on the equation for volatilization half-life were then compared against unfiltered water column samples collected at Poughkeepsie. Reduction in PCB concentrations from Troy to Poughkeepsie were assumed to be losses strictly due to volatilization after adjusting for particle settling.

Bopp reported that the model produced good agreement between predicted losses of lower chlorinated PCB and the observed difference between upstream and downstream samples in lower

chlorinated PCB. The model predicted lower losses among higher chlorinated homologs due to the greater affinity of higher congeners for suspended matter, thus making lower congeners available for volatilization. Sampling results indicated that the magnitude of losses in PCB concentrations by homolog, as predicted by the model, were inversely proportional to the degree of chlorination.

The model described and utilized by Bopp (1983) to generate the PCB volatilization estimated has a number of possible shortcomings which could lead to inaccurate predications. They are primarily related to a high reliance on theoretically derived input variables (versus measure verified data), the reliance on only two points of surface water data and the disregard of factors which influence volatilization. These factors are described in the following text.

In verifying his model, Bopp treats the water between Troy and Poughkeepsie as a "plug" of water which receives no dilution from tributaries, runoff or recharge, and does not gain, lose or recycle sediment. The model does not consider the existence of additional sources of flow and sediment between sampling locations. If inputs to the Hudson River of water and suspended matter between Troy and Poughkeepsie contained PCB, estimates of volatilization of PCB from the River would be biased low. Conversely, if water and suspended matter contributed to the River between Troy and Poughkeepsie contained lower concentrations of PCB, estimates of volatilization from the River would be biased high.

One of the assumptions inherent in the model is that the residence time of suspended matter equals hydraulic residence time. This is not appropriate because suspended matter is continually deposited and scoured to and from the river bottom. The residence time for suspended matter in the water column is, therefore, much greater than the time required for a given volume of water to pass from one point to another. Although water sampled at Poughkeepsie may represent the water column which flowed past Troy approximately 20 days earlier, the suspended matter included in the sample at Poughkeepsie includes resuspended matter that existed in the water column which flowed past Troy at a much earlier date. It could have been scoured from an area which was deposited any weeks or months prior or included runoff from the stream banks or storm drains. Consequently, suspended matter included in water samples analyzed from Poughkeepsie is likely to have undergone physical, biological and chemical transformation for a period of time longer than that used to approximate water transit time. It is, therefore, inappropriate to assume that changes in the sediment load had not taken place and is, therefore, invalid to provide a direct comparison between the two points and ascribe observed differences to volatilization alone. Further investigation of the assumption would have included a number of water column samples from intermediate points. Also, if the relationship were valid, then a comparison of upstream versus downstream sediments should

also reflect a reduction in the lower homologs. This was apparently not considered.

A total of two samples were collected for PCB analysis, one each at Troy and Poughkeepsie. In order to determine or confirm trends with respect to PCB concentrations in the River, additional samples are necessary. Additional samples collected at the same locations as well as at locations between Troy and Poughkeepsie would provide a larger sample size and could be used to evaluate spatial trends in PCB concentrations with a greater degree of confidence.

The effects of biodegradation on PCB in the Hudson River have not been considered in the analysis. Aerobic and anaerobic biodegradation of PCB have been demonstrated to occur in the sediments of the Hudson River (Rhee et al., 1989). Aerobic biodegradation is widespread and primarily degrades lower chlorinated PCB (Abramowicz, 1990). Anaerobic dechlorination of higher chlorinated PCB results in lower congeners subsequently available for aerobic degradation (Brown et al., 1987). Because aerobic organisms that degrade PCB could be more widespread than anaerobes that do the same, and because aerobic processes progress more rapidly than anaerobic reactions, it follows that lower PCB congeners could undergo a reduction in water column concentrations more quickly than higher chlorinated PCB. The differences between aerobic and anaerobic biodegradation could be the basis for, in part, higher degree of reduction in lower congeners measured by Bopp (1983).

Non-Filterable Adsorbed PCB - In the calculation of the distribution coefficients for the various homologs, PCB adsorbed to organic colloids, plankton, and other small biomass would have been measured as dissolved PCB, or these materials would have passed through the filter. Based on their work on PCB flux in Lake Superior, Baker and Eisenreich (1990) found that filtered samples used for PCB analysis contained not only dissolved PCB but PCB adsorbed to non-filterable biomass. Consequently, distribution coefficients developed by Bopp (1983) are likely to be biased low because the "dissolved" PCB bound to colloids and other filterable material are not available for volatilization. This bias would result in a calculated volatilization half-life which is higher than the volatilization half-life expected in the presence of hydrophobic colloidal materials.

Seasonal and Temporal Variations - The impact of variable environmental conditions such as wind, temperature, ice cover, and turbulence were not considered in the model developed by Bopp (1983). Paris et al., (1978) found that the rate of volatilization from water is strongly influenced by the turbulence of the water. Baker and Eisenreich (1990) state that the largest uncertainty in their flux calculations "results from the strong dependence of the equilibrium and mass-transport parameters on environmental conditions. Specifically wind speed and temperature." They state

further that ice cover must also be considered in northern waters. Therefore, even if the 40 day estimate of Bopp (1983) was accurate, it would vary significantly based on seasonal and temporal variability in temperature and wind speed. It is, therefore, misleading to consider, or cite, the 40 day estimate as applicable to the Hudson River.

Uncertainties in Model - Bopp (1983) provides a conceptual approach to modeling the behavior at the air water interface. It was not the intention of the model to provide a quantitative evaluation of PCB flux to the atmosphere from the Hudson River. Bopp acknowledged the limitations of his model when he stated "the main point of the exercise does not lie in detailed consideration of transit time, boundary layer thickness, or mass fluxes of PCB components." (Emphasis added.) Furthermore, Baker and Eisenreich (1990) state that "further efforts are required to develop and validate methodologies that directly measure [hydrophobic organic compound] fluxes across the air-water interface."

Conclusion and Recommendation re Volatization - Based on the limited amount of data utilized in the derivation and calculation of the predicted volatization half-life for the PCB homologs, the failure to consider processes affecting volatilization and the reliance on only two water column data points to verify the predicted values, the volatization estimates provided in Bopp (1983) should not be considered an accurate basis for predicting the behavior of the PCB in the Hudson River.

Pg. B.3-43, par 1: GE concurs that the volitization process is one that needs to be carefully evaluated. While EPA believes this process has "important ramifications for highly chlorinated homologs in the lower Hudson," GE believes the real importance is with the lighter chlorinated homologs that now reside in the highly degraded PCBs of the Upper Hudson River sediments.

Pg. B.3-43 to B.3-47: The Phase I report discusses PCB uptake by plants to consider whether significant uptake and accumulation of PCB in air by plants could result in elevated PCB concentrations in food crops (Phase 1 Report Section B.3.5.2). The discussion of this topic centers on a study performed by Bush et al., (1986). The Phase 1 Report states the conclusions of this study as follows (page B.3-45):

- the main route of PCB uptake in purple loosestrife is via the root system
- soil, as opposed to air, served as the major pathway of PCB uptake
- PCB uptake at the air-leaf interface also occurred.

A review of the study indicates that PCB plant uptake from soils

and air demonstrated inconsistent results and that additional studies would be required to verify and support the conclusion that root translocation is a significant pathway of uptake of PCB into plants. It should also be noted that although uptake of PCB plants does occur, it occurs to a limited extent, as compared to bioaccumulation in fatty tissues of animals.

The paper presented by Bush, et al., (1986), which was the central point of the Phase 1 Report's discussion of plant uptake of PCB, evaluated PCB uptake into purple loosestrife plants as a function of PCB levels in soil and air. PCB concentrations in plants that were transplanted and translocated between a control site with low soil concentrations of PCB (30.2 ng/g) and a Hudson River site with elevated (118.9 ng/g) PCB soil levels were compared. PCB concentrations in the air at the control and Hudson River sites were measured to be <40 ng/m^3, respectively. Three sets of conditions were evaluated in the study: Hudson River site plants translocated with Hudson River site soils to the control site, control site plants translocated with control site soils to the Hudson River site, and control plants transplanted to Hudson River site soils at the Hudson River site. Existing plants at the control site and Hudson River site were monitored for PCB as controls. Monitoring was performed by analyzing plant leaves for PCB. Within six weeks of the test start, the three scenarios tested all indicated increases in PCB concentrations which exceeded concentrations exhibited by the Hudson River control plants. This observation confounds interpretation of the results because the Hudson River control, which should have indicated worst case conditions and thus maximum PCB concentrations, contained lower PCB levels than the plants grown in clean soils and translocated to the Hudson River site.

The fact that the Hudson River site plants in Hudson River site soils which were translocated to the control site also exhibited higher PCB concentrations than the Hudson River site control demonstrates that the uptake of PCB by the plants is inconsistent, and that some factor operating in the study, other than those monitored and controlled, caused the inconsistent results.

The inconsistencies in the plant PCB uptake demonstrated in the study performed by Bush et al., (1986) indicate that uptake of PCB from contaminated soils and air has not been characterized to the extent required to justify the conclusions drawn. It is, therefore, recommended that the conclusions of the Bush et al., (1986) study be qualified within the text of the Phase 1 Report to reflect these inconsistencies.

Pg. B.3-50, chart: The presented chart should be expanded to include all the important data sets including the water column data generated by the U.S. Geological Survey.

Pg. B.3-48, Section B.3.7 (Adequacy of PCB and Aroclor Measurement): EPA must perform a detailed review of every piece of data employed by EPA, particularly the data used in the risk assessment. The assessment of the data presented in the Phase I report is not adequate to meet the EPA requirements given in EPA's own guidance documents. In particular GE would like to bring to EPA's attention the following:

1. Guidance for Data Useability in Risk Assessment, EPA/540/G-90/008, OSWER Directive : 9285.7-05, October 1990. On Page 26 of the document the following statement is made:

"The quality of historical data must be determined prior to its use in the RI. The difficulty in using historical analytical data in the RI is that the methods and the detection limits may not be documented. Also whether data review was performed may not be known. This information is required if analytical data are used in the quantitative components of risk assessment."

"Historical data of unknown quality may be used in developing the conceptual model or as a basis for scoping, but not in the determination of exposure concentration. Analytical data from PA/SI that meet minimum data useability requirements can be combined with data from the RI to estimate exposure concentrations. Similarly, historical data of lower quality may be used if the concentrations are confirmed by subsequent RI analysis."

2. Risk Assessment Guidance for Superfund, Volume I Human Health Evaluation Manual, EPA/540/1-89/002, December 1989

Both of these guidance documents specify the types of evaluations EPA should perform on data used for Superfund risk assessment. GE requests that EPA evaluate all the data in a methodical fashion and make available the result of the evaluation to GE and others. Of particular concern is the results from the fish data and EPA's conclusion on Page B.3-62 where it is stated that "the Aroclor measurements were performed by one laboratory, giving what should be a consistent set of results. Aroclor results appear reliable." Did EPA perform a review of any or all of the laboratory to see if laboratory problems occurred or were the summarized results as presented by the NYDEC relied upon completely without any independent verification of data quality? Would EPA make available to GE and others a copy of the laboratory protocols followed by the laboratory employed by the NYDEC for the analysis of the fish? Would EPA make available copies of chromatograms from the PCB anlaysis of fish (NYDEC) and water (U.S. Geological Survey)? Would EPA make available a copy of the analytical method employed by the U.S. Geological Survey for the analysis of PCBs in the water column?

Pg. B.4.1, par. 2: EPA presents three pathways for PCBs to interact with biota. EPA should consider the effects of the accumulation of PCBs through the food web.

Pg. B.4-3: Estimates of the daily average flood flow rates for the Hudson River below Sacandaga are presented and equivalent estimates at Fort Edward are presented in Table B.4-1. A comparison of the two shows a discrepancy with the average daily flow estimates at Fort Edward. It is indicated that the daily average flows at Fort Edward are lower that than those at the Hudson just below Sacandaga. Due to the difference in drainage area, one would expect a larger flow at Fort Edward. This potential discrepancy should be investigated.

Pg. B.4-7, par. 2: GE supports the reassessment of the magnitude of the 100-year flood and concurs with EPA that it is much less than previously estimated.

Pg. B.4.7, par. 3: EPA presents a statement concerning the lower relative suspended sediment concentrations in the Hudson River compared to other similar rivers and attributes this to lower sediment input and the presence of dams. What information does EPA use to come to this conclusion?

Pg. B.4-8, par. 2: EPA states that only after the dam was removed was it determined that the sediments contained large amounts of PCBs. What information does EPA have to support this conclusion?

Pg. B.4-10, par. 2: EPA states that a reduction in suspended sediment levels occurred after the sediments reached equilibrium and the remnant deposits remediation occurred. The remnant deposits remediation has just been completed and data on suspended sediment after this have not been presented by EPA in the Phase 1 Report. How can EPA attribute this decline to the remnant deposit remediation without data?

Pg. B.4-12, par. 3: The concern on whether dredging may have made the transport of PCB much greater than would have occurred is a legitimate concern. Care must be taken than when making statements about sediment stability based on the historical records due to the actions of New York that caused destabilization.

Pg. B.4-17, par. 3: EPA concludes that major portions of the yearly load may be transported during a few brief flood events. This seems to be somewhat contradictory to the statement that the relationship to flow and PCB transport is weak particularly in the most recent data. Is this a theory or a conclusion? If it is a theory it should be so stated and EPA should test it by data collection in later phases of the project.

Pg. B.4-24, par. 3: The statement made that PCBs mobilized in the Upper Hudson River are transported through to the Lower Hudson

River is misleading. What the data shows is that the concentration of PCBs in the water column at various points in the Upper River do not change as one moves downstream. Why this is so is unknown. Numerous reasons could be suggested, and the notion that PCB transport on scoured sediments occurs to the Lower River is by no means the only explanation.

Pg. B.4-24, par. 3: The conclusion reached that the PCB load in the Upper Hudson River originates above the Thompson Island Pool is of major significance. This shows that removing the sediment will have little or no effect on the PCB load passing through the Upper Hudson River in any given year. This clearly shows the importance of obtaining monitoring data from the remnant deposits remediation project to see what effect the remedation has had on the water column PCB values. This monitoring is mandated to be done by GE as part of the agreement entered into by the Federal government and GE for the remediation of the remnant deposits.

Pg. B.4-25, par. 2: GE does not agree with the approach being advocated for estimating annual PCB flux in the river. The uncertainty of all the methods employed to date is very great. The basic limitation is a lack of data to accurately determine the actual PCB flux to the Lower River and the lack of a physically based model for determining PCB transport. As discussed in Section 2.0 of GE comments, the appropriate way to understand the transport of PCB is to develop a quantatative framework (i.e. model) that can handle the complexities of the processes being modeled. The statistical model employed by EPA is an oversimplification of a very complex system. EPA needs to interpret the data in a framework that realistically models the river conditions.

Pg. B.4-26, par. 3: The EPA assessment of PCB loading to the Lower River from the Upper River presented in this section is very speculative and the purpose for presenting this is unclear. The estimates of PCBs discharged are speculative and GE would like EPA to more critically evaluate this information if the numbers will continue to be used. The EPA estimates for loading for PCBs from 1977 to 1988, while limited in quality, are the best estimates of PCB transport to the Lower River. The only reliable estimate that can be made is that it appears that greater than 15,000 kg of PCB, were transported into the Lower River. Conclusions based on the rest of the information presented by EPA is nothing but speculation.

Pg. B.4-28, par 1: As discussed elsewhere in these comments, the use of the dated cores does not allow one to conclude that a peak in PCB concentration occurs in the sediment that corresponds to the removal of the dam in the Upper River in 1973.

Pg. B.4-30, par. 2: As discussed elsewhere, the use of dated cores in the Lower River has been accepted by EPA without a critical review of the information. EPA has not presented any information

to suggest that data from such cores could be used to determine, in a reliable way, the pre-1976 loading over the Federal Dam of PCBs.

Pg. B.4-30, par. 3: It is stated that the data on the decline in PCB levels in fish will not be sufficient to reduce the PCB burdens in many fish species to acceptable levels in a reasonable period of time. What does EPA consider to be an acceptable level? What is a reasonable time? If a remediation occurs, will this reduce in a significant way the amount of time for the PCB levels to reach the "acceptable level"? Has EPA prejudged the need for a remedy or the effectiveness of a remedy?

Pg. B.4-31, par. 2: Does the peak in fish concentration occur at the same time as maximum dredging activity?

Pg. B.3-35, par. 2: GE agrees that the risk assessment should include the reduction of PCBs over time. However, the analysis presented by EPA also need to remove the background PCB concentration in fish. This will become significant in future years.

Pg. B.3-37, par 1: It appears that EPA utilizes a simple arithmetic mean for calculating PCB levels in the fish. This method is most likely inappropriate and overestimates the PCB levels. EPA should determine whether the data are log-normally distributed (like many other environmental data) and employ the geometric mean as opposed to the arithmetic mean. This is a more appropriate measure of the central tendency of the data.

Pg. B.4-37 to B.4-40, Section B.4.4.3 (Relation Between PCB Concentration in Fish and Water): EPA attempts to define a direct linear relationship between the PCB levels measured in the water column and the PCB level measured in the fish. This relationship is generally referred to as the Bioaccumulation Factor (BAF). Based on the data presented in Figure B.4-25, the relationship was apparent with the data from 1979-1981. However, an obvious relationship in the later years (when PCB water column values dropped dramatically) does not occur. This lack of a simple empirical relationship demonstrates the need to more fully analyze the dynamics of PCBs within the food chain of the river biota. This type of analysis needs to consider variations due to PCB homologs that might help explain the difference over time in the presence of Aroclor 1016 and Aroclor 1254. The conclusion presented on page B.4-42 (second bullet) is not supported by the data for the most recent years and the presented BAF's do not appear to have any use in the RRI/FS. As discussed in the main body of GE comments, EPA must analyze all the data in a framework that allows the best understanding of PCB dynamic in the river system to be understood. A simple empirical model (i.e. BAF) does not explain the data nor allow reasonable predictions to be made.

Pg. B.4-42, par. 2: EPA claims that the water and fish data are extensive and reliable and the sediment data is somewhat less reliable. This is a very subjective statement. To address data uses and limitations, EPA will need to carefully and methodically develop and document data quality objectives as required by EPA guidance. After the data quality objectives are defined, then the adequacy of the historical data can be assessed in a more objective way. Additionally, it does not appear that EPA actually reviewed the quality of the data from the fish or water column and, therefore, the reliability of the data should be qualified until such time that EPA reviews the quality of the data. The lack of clearly defined and articulated data quality objectives is of great concern since the Phase 2A data collection work plan prepared by EPA was nearly devoid of them.

Pg. B.4-42, bullet 3: The characterization of the projected PCB level based on a decline with time as the "best-case" estimate is incorrect. The projection does not assume a more reasonable rate of decline nor does it employ simple averages. EPA should present the range of PCB values for the worse case scenarios that it has evaluated (12 to 1.5 parts per million).

Pg. B.5-6, par. 3: GE concurs that the current sediment scour estimates made by the NYDEC greatly overestimate the amount of sediment resuspension that would occur in the Thompson Island Pool during a 100 year flood. If EPA is going to utilize scour estimates that might occur during a flood scenario, GE believes that the use of a noncohesive sediment transport model based on realistic flow projections will be necessary.

Pg. B.6-2, par. 2: The scope of the risk assessment and the scope of the project (as presented on page I-1) may not be consistent. It is GE's understanding that the EPA is attempting to assess the risk associated with the PCBs in the sediments of the Upper Hudson River. This particular focus is on the bulk of the contaminated sediments which are below Rogers Island. This does not include potential PCB sources within the remnant deposit area or above Bakers Falls. Therefore, the risk assessment must be based on information related to the sediments of concern and contributions to risk from these "background" sources must be removed so that a real "baseline" risk assessment can be made. EPA needs to carefully define the project scope.

Pg. B.6-4, par. 1: As noted in an earlier comment, the limitations of the existing data are not fully understood and there is a concern that inappropriate conclusions from the data may be made. GE believes that EPA must completely evaluate the data being used to understand all of it's limitations. Until this is done, any risk calculations should at best be considered qualitative in nature.

Pg. B.6.4, par. 3: Care must be taken in using the results of the fishing study done by NYDEC on the Hudson River. The study does not distinguish among the major sections of the river, particularly above and below Fort Edward. These two segments of the river are vastly different and, due to the fishing ban, it would appear that the information presented on rates of fishing clearly do not apply to the section of river between Fort Edward and Troy.

Pg. B.6.6, par. 2: It is stated that it is EPA's policy not to assume that fishing bans or other similar types of institutional controls have any significant long-term effect in reducing the intake of contaminated fish. Please provide a copy of this policy with an explanation of how it applies. GE has to believe that the statement is not meant as an evaluation of the effectiveness of the remedy (i.e. fishing restrictions) but rather a statement meant to convey EPA's policy that for the purposes of the "baseline" risk assessment, EPA will assume the no-action alternative is occurring. This allows EPA to evaluate all remedies (including institutional controls) against a common baseline. If this is not the case, GE believes EPA must provide all information that has been relied upon by EPA to conclude that all types of fishing restrictions are ineffective in preventing or limiting consumption. This would be a puzzling conclusion for a risk assessment and is more appropriate for the feasibility study. The Phase I feasibility study does not even mention fishing restrictions. Has EPA Region 2 determined that fishing restrictions are not effective and therefore will not be evaluated in this feasibility study?

Pg. B.6-8, par. 4: When the final risk assessment is prepared in Phase 2 EPA will need to reevaluate the rate of decline and average fish concentration based on the anticipated NYDEC fish data. Additionally, since realistically a ROD will not be issued until 1993, EPA will need to perform the projections from 1994-2023.

Pg. B.6-9, par. 1: As mentioned previously, the use of the arithmetic average is probably not the most appropriate indicator of central tendency. The use of geometric mean is probably the best estimator.

Pg. B.6-13, par. 2: EPA should request the information from the NYDOH to substantiate the claims made in the Phase 1 Report concerning PCB levels in breast milk.

Pg. B.6-13, par. 3: EPA needs to clearly differentiate the subpopulation being considered in Messena, New York and that along the Hudson below Fort Edward. There is no indication that subsistence fishing is of concern on the Hudson River.

Pg. B.6-17, par. 3: EPA attempts to utilize skin-adherence values based on soils. Since the exposure assumed is to contaminated sediments, which are generally submerged, the adherence values should be very much less than those reported for soils. Based on

the data reported by EPA in the Phase I report, a value very much less than 1.0 mg/sq. cm. is reasonable.

Pg. B.6-18, par. 2: As discussed earlier, the EPA estimate of average PCB content of the Thompson Island Pool is greatly overestimated since a large amount of the original screening data was ignored. Additionally, the assumption that all contact will occur in the Thompson island pool is not reasonable. EPA should consider the average (a better measure would probably be the geometric mean) PCB concentration in the entire Upper Hudson River. The 1977 data would indicate a value less than 25 parts per million would still be a conservative estimate (see Figure B.3-1).

Pg. B.6-19, par. 3: The assumption that children, 1 to 6 years of age, eat 200 grams of sediment per day is absurd. Why would very young children even be near the water, let alone in the water, where ingestion could reasonably occur. Secondly, the sediments being below the water would tend to be washed off hands and items children may place in their mouths. Therefore, the rate of ingestion would be much less than for soil consumption. The average PCB level in the sediment also needs to be adjusted as described in the previous comment.

Pg. B.6-22, par. 1: Absorption of PCBs from river water through the skin should be a function of the dissolved PCB level verses the amount absorbed on particles. Since it is thought the dissolved load is a small fraction of the total (including that transported as colloidal material or as macromolecules) EPA should reduce the average PCB level dramatically. Additionally, since the exposure is assumed to occur over a long time period, the average PCB level in the water column over a thirty year period should be used.

Pg. B.6-30, Section B.6.3.4 (Toxicity of Specific PCB Congeners): The TEF approach does not currently have a sound technical basis for application to PCB toxicity. Since the approach is described in the Phase 1 Report, EPA should make available the source of the information provided (transcript of the December 1990 meeting?).

Pg. B.6-34, par. 2: While EPA's attempt to explain why the use of the FDA advisory level for PCBs in fish may not apply to a Superfund risk assessment, due to specific fish consumption assumptions, it is not clear why EPA and FDA utilize different toxicity factors for PCBs. In particular, why, substantively, does FDA utilize different carcinogenic potency factors (Aroclor 1260 verses Aroclor 1254) for different PCBs and EPA does not?

Pg. B.6-44, par. 1: EPA indirectly references the reevaluation of the pathology slides from the relevant PCB feeding studies. As EPA knows, the evaluation is complete. The results with the interpretation were submitted to EPA prior to the completion of the risk assessment. GE has transmitted the report to EPA-Washington and EPA-New York and has asked that the results be evaluated.

Additionally, GE has requested that this important new information, which is directly applicable to the Hudson River RRI/FS, be placed into the Administrative Record. To date GE has not had a formal response related to this new information and EPA has yet to place this date in the Administrative Record.

Pg. B.6-45, par. 1: In the uncertainty analysis EPA mentions a number of obtuse issues relate to why the risks might be underestimated and fail to recognize that due to the way EPA manages uncertainty in the Superfund process that the risks estimated by EPA are grossly overestimated and the real risk is most likely much lower.

Pg. B.6-45, par. 2: The data as presented by EPA is insufficient to calculate the risk in the Lower River due to PCBs in the Upper River sediments. The attempt to qualitatively estimate the risk is no better than speculation.

Pg. B.7-2, par. 4: The description of the terrestrial ecosystem is inconsistent with that shown in Plate B.1-4.

Pg. B.7.3, par. 2: The presence of pockets of wetlands are mentioned, yet the importance of these along with emergent wetland areas (fish habitat) is not discussed.

Pg. B.7-16 to B.7-17: There may be an error in the trophic partitioning. The bluegill's dietary preference would be more appropriately described as epibenthic invertebrates and yellow perch as fish/macroinvertebrate. The list provided in the text appears to be based on two literature citations. Confusion over the dietary preferences for these fish may be due to age (i.e. juvenile fish sometimes have different preferences than adults).

Pg. B.7-18, par. 2: EPA states that the improving conditions as measured by improving biological measures should not necessarily reflect the lack of influence of PCBs. EPA should be more open minded and recognize that generally there does not appear to be a negative effect on the aquatic system due to the presence of PCBs.

Pg. B.7-22, Table B.7-1: Table B.7-1 indicates a low level of confidence in data for both the herring gull and mink. In spite of insufficient data, the Phase I report provides, in Table B.7-3, proposed ecological guidleines for limits to PCB concentrations in birds and mammals. Although the footnote indicates that the values are not enforceable standards, presentaion in this table implies more knowledge than is currently available regarding allowable concnetrations of PCBs in wildlife.

Pg. B.7-23, Table B.7-1: The assumed sediment PCB level is not reasonable and should be reduced to less than 25 parts per million. See the earlier discussion in the Human Health risk assessment.

Pg. B.7-34, par. 4: It is indicated that while few reports of the effects of PCBs on aquatic insects exist, many of the species seem to exhibit sensitivities similar to those of Daphnia magna. EPA should make this data available to GE and others.

Pg. C1-1: The requirements for a feasibility study (FS) from the National Contingency Plan (NCP) are correctly repeated. However, EPA does not explain what the purpose of performing the Phase 1 FS was since we are very early in the RI/FS process. Therefore, it is difficult to know if the objectives were met or if there will be additional opportunities for parties to comment on all aspects of the FS. If the purpose is to identify data needed to complete the field investigation, the result is incomplete. The only data identified as being needed is bench testing of four technologies. GE believes the following data are also needed:

1. Sufficient data on the physical/chemical and biological nature of the river so that the fate and transport of PCBs can be determined under various remedial scenarios. This will necessitate a complex model that will allow projections of the PCB concentration (for biota as the receptor) into the future (for the risk assessment) for various remedial actions including the no- action alternative (comparative risk reduction).

2. Evaluation of the feasibility of dredging in the unique environment presented by the near shoreline locations of the sediment deposits in the Upper Hudson River.

Pg. C.1-2: EPA mentions that during Phase 2 data on the nature and extent of contamination will be gathered. To date, EPA has not defined what the objectives of such an evaluation would be or, overall, what the data needs might be for the risk assessment or the feasibility study. EPA not only needs to define to what degree the nature and extent of contamination must be determined, but also must determine the time dependent interactions between the various media. Understanding the nature and extent is not sufficient to understand the complex chemical/physical/biological system where the PCBs interact over time. EPA must perform a methodical analysis of data quality objectives as dictated by EPA guidance. As an example, if EPA is going to consider dredging as a remedy, and furthermore, EPA targets areas of high concentration in an effort to reduce volume, EPA will need to be able to determine volume to see if the type of equipment to handle the volume of material is available. Additionally, there are doubts whether EPA can define "Hot Spots" and remove them. Therefore, to determine feasibility of dredging, EPA will need to understand how the PCBs are distributed on a scale that is meaningful for the type of dredge and project being considered.

Pg. C.2-1, par. 3: EPA claims it is apparent that the main problem are impacts upon aquatic life and consumers of aquatic life. The

only hypothetical impacts documented by EPA, using EPA default assumptions on toxicity, fish consumption, and other exposure parameters is human consumption of fish (see earlier comments on the risk assessment). It is imperative that EPA carefully define the preliminary remedial action objectives as dictated by the RI/FS guidance, based on appropriate exposure pathways and receptors.

Pg. C.2-1: Under the no-action scenario, EPA lists the use of institutional controls. Institutional controls are considered to be an action. EPA should define another category of action referred to as institutional controls which may include such items as monitoring, fishing restrictions (various types), and land use controls. Otherwise, the use of the term is misleading. EPA should also consider another category that is any combination of the above actions. It is possible that there could be a combination of activities.

Table C.2-1: 1. The table of institutional controls should include as process options fishing ban, managed fishery (i.e. catch and release), monitoring, educational programs, land use restriction. 2. Excavation as a remedy is given as is dredging. This would imply that EPA is evaluating something other than the sediments within the river. What is the scope of the RRI/FS? 3. A very good compilation of sediment removal options and technologies is given in the document entitled: Review of Removal, Contaminant and Treatment of Contaminated Sediment in the Great Lakes (U.S Army Corps of Engineers - December 119, Miscellaneous Paper EL 90-25). EPA should consider this relevant and timely information. This document notes the importance of dredged material transport options. It is suggested that EPA create a subcategory under dredging for material transport.

Pg. C.3-1: EPA seems to indicate that nonpromulgated advisories or guidance documents could substitute for ARARs where there are no specific ARARs applicable to a situation or where ARARs are not protective. GE strongly disagrees that any guidance or nonpromulgated standard can ever be treated as an ARAR for the purposes of Superfund/CERCLA.

Table C.3-2: Wetland impacts caused by dredging must be considered and weighed against the alternative. GE agrees that this requirement is applicable.

Pg. C.4-1, par. 3: It is stated that various issues related to capping need to be investigated, such as ability to withstand scour, leaching, etc. GE agrees such investigations are needed and believes that EPA should begin the investigation of these technologies at this time. This is a defined data need.

Pg. C.4-7, par. 3: EPA is relying on published data for determining the fugitive sediment releases from dredging. This is acceptable as long as the same type of dredge(s) is evaluated in

the same type of condition (small channel, high velocity riverine system). If the data is not comparable, EPA will need to perform tests to determine if this will be a significant issue.

Pg. C.4-6: An important component of any dredging program assessment is the evaluation of transportation of the dredged material. EPA will need to include an evaluation of this in the future work.

Pg. C.4-8, par. 2: Mention is made that, historically, the NYDOT has dredged in river. The occurrence of dredging historically cannot be used alone to determine that dredging is feasible. NYDOT dredging is in the navigational channel in the center of the River, not in the near bank areas and in backwater eddies where PCB containing sediments accumulate.

Pg.C.4-8, par. 2: Is the EPA specifically evaluating the option considered either historically or currently by the NYDEC?

Pg. C.4-8, Section C.4.4 (Treatment Technologies): This section is difficult to follow and is overly simplistic. GE believes EPA needs to prepare preliminary remedial action objectives (RAOs) prior to evaluating individual technologies. It is not clear what level of PCB treatment is needed (i.e. What residual level is acceptable and why?). Also, a major confounding problem with the treatment of the Hudson River sediment may very well be other constituents present in the sediments such as metals. This important consideration is not even discussed. GE believes EPA should first prepare general site-specific preliminary-RAOs and then compile and evaluate candidate technologies based on the RAOs. EPA also relies heavily on information that is either not referenced, or if it is referenced, is not readily available. GE again requests that EPA prepare a complete administrative record so GE can evaluate conclusions reached based on the information that is otherwise not available to GE.

GE also would like to caution EPA against utilizing vendor supplied information without independent corroborating information. Additionally, GE strongly believes that if EPA chooses a treatment option in the ROD (GE does not advocate this) and that if a vendor-specific technology is selected that the costs will be uncontrollable. GE believes EPA should allow selection of the specific technology as part of remedial design. EPA would instead set performance standards for the technology to meet.

Pg. C.4-8, par. 3: EPA states that the incineration of PCB contaminated sediments is the most widely practiced and permitted method for the management. GE is unaware of any significant amounts of sediment (contaminated with anything) that have been incinerated and then landfilled. Additionally, GE is unaware of any significant upland sediment disposal sites that have been used for the disposal of significant quantities of PCB (or other

contaminants) contaminated sediments. EPA should provide the data that indicates the assertion is correct. This information would be helpful in focusing future work.

Pg. C.4-8, Section C.4.4: If EPA selects a type of technology, GE strongly recommends that EPA not be overly prescriptive in its selection, but rather leave latitude for design, where cost and schedule issues can be optimized. EPA should instead provide realistic treatment levels (i.e. performance standards). Specifying a specific technology that may only come from a single vendor will result in enormous costs. As an example, EPA may find that a number of chemical treatment technologies have been shown to be feasible. GE should be allowed some latitude in deciding which one is most appropriate after detailed design occurs. In the record of decision, EPA could specify a class of treatment (e.g. chemical treatment) and a performance standard. GE would then, based on detailed design, select the most efficient method. This same comment would apply to removal technologies.

Pg. C.4-9, par. 1: EPA should consider the work being done as part of the Federal ARCS program (see comment on Table C.2.1). The listing of potentially applicable technologies need to be further evaluated with such contemporary literature. Additionally, the more recent evaluations performed at Massena, New York and New Bedford Harbor need to be considered.

Pg.C.4-9, par. 4: EPA refers to a study conducted by Research Triangle Institute (RTI) yet does not supply a reference to a published report. Since this information seems to be the basis of EPA's views on the PCB treatment technologies in general, GE believes EPA must make this report available for review in the administrative record.

Pg. c.4-14, par. 1: Reference is made to a recent study by RCC for processing liquid sludge or contaminated sediment. The source of the information is not divulged. This information should be made available to GE.

Pg. C.4-14, Table C.4-4: In discussions on LEEP, EPA references Steimer (1991). An examination of the reference section of the report indicates this was some sort of seminar material. Since GE is unaware of the seminar or how to obtain the materials, GE requests that the information be made available to GE by EPA.

Pg. C.4-13: For the B.E.S.T. process, EPA should provide cost information if it is available.

Pg. C.4-13, par. 2: EPA indicates that the B.E.S.T. process was able to achieve a PCB reduction of 99 percent. This result was from sediments that contain much higher levels of PCBs than generally found in the Hudson River (5,800 and 420 ppm). Would the same extraction efficiency occur for sediments with lower amounts

of PCB? It is also indicated that the process yielded sediments having a PCB level of 16 ppm after six extraction stages. Is this level acceptable? What is the remedial action objective being used as a measuring stick for evaluating the effectiveness for these and other technologies?

Pg. C.4-15, par. 2: It is indicated that LEEP, has feed stream particle size limitations. What are these limitations and are they an issue with Hudson River sediment? Does particle size information on Hudson River sediments need to be gathered?

Pg. C.4-15, Propane Extraction: The only reference to information on the propane extraction process is a reference cited in Table C.4-5. In the reference section, all that is given is a personal communication with the vendor. GE requests that EPA make available relevant information and not to rely on vendor supplied information.

Pg. C.4-16, par. 2: A number of problems with the propane extraction system are listed. These problems were found at New Bedford and the material handling and low extraction efficiencies seem to indicate that the use on sediments would be limited. Is EPA planning to gathering the field data necessary to characterize the Hudson River sediments sufficiently to evaluate these problems? EPA indicated the process was rejected for the New Bedford sediments project due to these problems. Why has EPA not rejected this technology for the Hudson River sediments? What additional information does EPA have?

Pg. C.4-16, par. 2: What cost estimates are available for the propane extraction technology?

Pg. C.4-16, Thermal Treatment Technologies: EPA must consider the fact that significant amounts of metals are present in the sediments and not just consider the PCB destruction potential but also the effect the metals might have on the system as well as the need for extensive emission controls and the fact that after incineration the metal may be more mobile since organic materials have been destroyed.

Pg. C.4-17, par. 1: EPA indicates that the Toxic Substances Control Act (TSCA) requires that PCBs be incinerated at a temperature of 2,200 degrees fahrenheit. While this is true for PCBs of certain concentration or those generated under certain conditions, it does not appear that those sediments in the Hudson River would fall under the given requirement. EPA also indicates that PCBs in the residue material would have to be treated to a level of 2 parts per million or less. Please provide a reference for this specific requirement and an explanation of why the Hudson River sediments would have to meet this requirement.

Pg. C.4-17, par. 3: EPA states that due to the anticipated load for this project that more than one thermal system will be required. What is the load (yards of sediment?) being assumed by EPA?

Pg. C.4-17, par. 3: EPA needs to consider early on in the feasibility study the likely public opposition to incineration. This is particularly true given the statement that multiple units will be necessary.

Pg. C.4-17: EPA does not mention methods for handling materials that may include separation into differing size fractions or for separating the water. Both of these need to be considered for pretreatment of sediments. These may be very important as a pretreatment technology for the thermal technologies since they reduce the volume of material treated as well as significantly change the required energy inputs.

Pg. C.4-19, par. 3: EPA again mentions the requirement for destroying PCBs at a temperature of 2200 degrees fahrenheit. EPA needs to explain its rationale for arriving at the conclusion that the sediments, regardless of PCB concentration or time an method of origin are regulated by the specified requirement.

Pg. C.4-20, par. 2: The fluidized bed incinerator is criticized for not being transportable. Is ease of transport an important consideration for the type of project EPA is envisioning?

Pg. C.4-21, par. 3: EPA states that the circulating fluidized bed incinerator has been shown to destroy PCBs at a temperature below 2200 degrees fahrenheit. Please make available to GE the information being relied upon by EPA to make this statement. Does this also indicate that EPA will accept this as equivalent to a TSCA incinerator?

Pg. C.4-21, par. 3: Does this technology require that dewatering of sediments occur?

Pg. C.4-21: On page C.4-20, paragraph 2, EPA rejects the fluidized bed incinerator due to the fact it does not destroy PCBs at sufficiently high temperatures and the technology is not transportable. These same limitations appear to apply to the circulating bed incinerator, yet EPA has not rejected it as a technology.

Pg. C.4-21, Conveyor Furnace: EPA does not provide any information on the use of this technology for PCB contaminated sediments. Is this technology capable of handling high moisture content material and given the low temperatures cited, (operates at less than 2200 degrees fahrenheit) is this material applicable, in EPA's opinion, to the PCB contaminated materials?

Pg. C.4-29, par. 3: EPA states that a compositing operation would require a RCRA liner and leachate collection system meeting the RCRA minimum technology requirements. Has EPA decided that RCRA is an ARAR for the Hudson River sediments?

Pg. C.4-31, par. 3: EPA identifies additional evaluation and data it needs to more fully evaluate the applicability of bioremediation of Hudson River sediments. As pointed out in the other GE comments, some of this information is available and needs to be considered by EPA. Based on this statement, it is not clear if EPA will perform the needed work or if they are identifying this for GE to perform. GE is willing to work closely with EPA and others to gather the information if EPA will consider the information during the Hudson RRI/FS.

Pg. C.4-32, par. 3: EPA states that subaqueous confinement will not be considered further since EPA believes that relatively steep (hydraulic) gradients exist. This is not totally correct and EPA needs to more thoroughly evaluate the use of either subaqueous disposal or a technology that will be used to further stabilize existing deposits of contaminated sediment. While the topographic gradient in the Upper Hudson is relatively steep compared to that in the Lower Hudson River, the hydraulic gradients within the pools behind the dams are relatively shallow. The majority of the Upper River is composed of pools of relatively small hydraulic gradients. EPA needs to make a more meaningful comparison of water velocities that the materials in the Upper River will be exposed to and compare those to tidal areas where disposal has occurred in other areas of the country. The continued stability of the sediments present in the Upper Hudson attest to the viability of in river containment. GE suggests these potential actions be retained for further analysis.

Pg. C.4-32, par. 4: EPA states that a landfill will need to meet regulatory requirements for the disposal of PCB contaminated material. EPA needs to perform a more careful analysis of this point. It is possible that if dredging were to occur, a vast majority of the material would contain less than 50 parts per million of PCB. This material should not have to be put in a landfill meeting the TSCA requirements. This also points to the possibility that only a small amount of material dredged may need to be treated as a TSCA material. Additionally, if the material is first processed into size fractions, then the vast majority of PCBs would be transferred to the very fine grained fraction of the sediment. The majority of the sediment volume may be relatively clean and will not need to be managed within the regulatory framework.

Pg. C.4-32, par.4: EPA states the technology for a landfill is readily available. This may true in the sense that the type of materials can be controlled in lined landfills. However, the application is extremely limited due to potential constraint

presented by the site hydrogeology and the inability to site the landfill. Based on past failed attempts to site a landfill by the State of New York for the purpose of disposing of contaminated sediments, EPA should view this technology as having limited potential for application for the project.

Pg. C.6-1, par. 1: EPA states that the data upon which the initial screening of technologies was based is from various sources. GE requests that EPA make the data or information available to GE. GE also request that EPA specifically identify the data and information relied upon.

Pg. C.6-1, par. 1: The statement made by EPA that it is possible to render some judgements considering remedial options is somewhat confusing. The analysis presented by EPA was focused on technologies without much consideration of how they might be combined to form a remedial option. Therefore, it is not clear how EPA will reach a decision. GE believes, as stated in EPA's own guidance, that before any technologies are screened preliminary remedial action objectives must first be identified.

Pg. C.6-1, par. 2: EPA states that technologies related to excavation of remnant deposits have not been evaluated nor screened for further evaluation. Additionally, Figure C.6.1 shows that excavations as a removal technology has been retained for further analysis. GE is very concerned that EPA is attempting to increase the scope of the RRI/FS without directly acknowledging it. Page I-1 of the EPA Phase 2 report clearly states that the scope for the project is limited to the sediments in the Upper River and not to the remnant deposits of other areas outside the river. EPA has also stated in public meetings that the focus is only on the sediments within the Upper River and not outside of the banks. GE has implemented an agreement for remediation of the remnant deposits. The "ink" has barely dried on the agreement and EPA is suggesting that the 6 millon dollars spent by GE to implement the EPA chosen remedy was for nothing. Why is EPA considering excavation of the remnants? EPA must remove all considerations of remnant excavation from within the confines of this RRI/FS.

Pg. C.6-1, par. 3: EPA states that it is waiting to evaluate the three types of dredging pending availability of additional information on material characteristics and quantities. What specific information is required and when is EPA planning to obtain this information?

Pg. C.6-2, par. 2: On Table C.6-1, EPA eliminates from consideration the possibility of subaquesous disposal. As stated in an earlier comment, EPA should retain this option for further analysis.

Pg. C.7-1, par. 2: EPA should either perform biological testing as part of its RI/FS efforts, or work with GE so GE efforts can be

better directed to meeting EPA program needs for the Hudson River RRI/FS.

Pg. C.7-1, par. 3: EPA states that they will test a number of technologies on the bench scale and that they will work with the developers. GE has extensive experience in such endeavors and needs to voice a few words of caution. The first is that EPA must carefully oversee vendors since they have a strong vested interest in showing their technologies have application. The second problem is the possibility that if the sediment to be tested is TSCA regulated (we are not suggesting it is, however, EPA may think it is), the vendors will have to obtain TSCA research and development permits prior to testing. This is a time consuming cumbersome process. GE also requests that EPA develop specific work plans from each test and specific objectives for measuring success. These work plans should be made available for public comment prior to implementation.

References (pp. R-1 et seq.)

Many references that are by the same author or organization in the same year are not differentiated making it difficult to determine what the source of information was. As an example see USEPA 1990.

Numerous citations in the text are not included in the references making it nearly impossible to evaluate conclusions reached by EPA.

The materials referenced by EPA are a diverse range of information, most of which is difficult to obtain. For some material refernced by EPA to a source is not clear and GE does not know exactly what the information being referred to is. GE believes, as part of EPA's community involvement project, that a central library of information be established that will house all data or information for the project. This would include copies of all published reports (including peer review and journal articles) that have relevance to the study.

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B. COMMENT ON THE CHARACTERIZATION OF PCBS FOUND IN ENVIRONMENTAL SAMPLES

APPENDIX B

COMMENT ON THE CHARACTERIZATION OF PCBs FOUND IN ENVIRONMENTAL SAMPLES

Section B.3.7 of the Phase 1 Report contains a discussion of PCB nomenclature and methods of analyzing environmental samples for PCB content. It correctly points out that PCBs were manufactured and sold as mixtures of congeners and that the "Aroclor" designations were applied to these mixtures. It also correctly points out that as soon as any given Aroclor (a mixture of known composition) is subject to environmental influences, the composition is altered in a way that cannot be known without congener-specific analysis. It then notes that the convention of characterizing PCBs found in environmental samples by Aroclor is both inaccurate and misleading, since:

> "Aroclor designations are no longer descriptive of the congeners present and their relative amounts" (p. B.3-51).

In Subsection B.3.7.3, the Phase 1 Report summarizes the various problems that arise in using the existing data because of both the inaccuracy of the Aroclor nomenclature when applied to environmental samples as well as the inability of commonly used laboratory techniques to accurately quantitate the PCBs in such samples.

GE agrees with EPA's frank criticism of the historic database. Unfortunately, after saying all of the correct things, throughout the Phase 1 Report EPA uses the data as if it were (a) fully accurate and (b) uniformly collected and analyzed. In the

absence of congener-or homolog-specific data, EPA repeatedly treats all PCBs the same, jumps to conclusions as to PCB source and fate based on crude Aroclor designations, and makes assumptions about the presence, composition, and masses of PCBs in various environmental media.

Since, as EPA elsewhere in the Phase 1 Report acknowledges, the toxological and other properties of individual members of the PCB family vary greatly, glossing over the nomenclature and analytic ambiguities of the existing data base can lead to erroneous conclusions. GE believes that EPA's conclusion on page B.3-63 is correct. GE regrets that EPA has not acted consistently with that conclusion in the rest of the Phase 1 Report. For the future phases of the RI/FS, and in the ultimate EPA decision-making process, GE urges that EPA not just pay lip service to its own conclusion, but act as if that conclusion were valid and highly significant.

In addition, the Phase 1 Report presents an inappropriate comparison of sediment PCB concentrations collected from the same regions of the river by GE in 1990, EPA in 1983, and Malcolm Pirnie in 1978 (p. B.3-14; Table B.3-8). The report states:

> "In four of the six locations, the GE samples indicate average PCB levels above both the 1976-78 and 1983 values; the remaining two locations show 1990 PCB levels lower than the 1976-78 and/or 1983 results" (p.B.3-14).

This statement is inaccurate and was made without regard to the differences in analytical techniques employed for the different surveys.

The 1978 and 1983 data used in this comparison were generated by packed column techniques. Packed column separations of sediment PCB extracts typically cannot resolve Webb and McCall peak numbers 11 and 14, which contain the more volatile monochlorinated biphenyls, because they elute with the solvent in the extract (Webb and McCall, 1973). In contrast, the capillary column techniques employed to analyze the 1990 GE data provide sufficient separation to quantify monochlorinated biphenyls.

In samples which have undergone reductive dechlorination, monochlorinated biphenyls can constitute as much as 50 percent of the total PCBs (Brown et. al., 1987). Therefore, for dechlorinated samples, capillary column techniques produce a greater total PCB concentration than packed column methods. For this reason, the comparisons in total PCB concentrations made in Section B.3.2.4 are inappropriate and lead the reader to the erroneous conclusion that PCB levels in the sediments are elevated relative to historical data. Indeed, comparing the results from the capillary column technique to the packed column technique is akin to comparing apples to oranges.

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C. COMMENT ON "TBC" ITEMS

APPENDIX C

COMMENT ON "TBC" ITEMS

The National Contingency Plan (NCP) permits EPA, as part of its duty to identify applicable or relevant and appropriate requirements to identify "other advisories, criteria, or guidance to be considered for a particular release" (TBCs) (NCP § 300.400(g)(3)). The Phase 1 Report briefly refers to these TBCs in its Section C.3.1.3, and several TBCs are preliminarily identified in Tables C.3-1 and C.3-2.

One of the items preliminarily identified as a TBC in Table C.3-1 is a New York State document, identified as "Sediment Criteria" and dated December 1989 Sediment Criteria. The Phase 1 Report improperly classifies this document as a TBC, however, because (1) it does not meet the NCP's definition of a TBC; (2) it was never intended to be used to establish a clean-up level or other remedial goal and thus is not appropriate for use in an RI/FS; and (3) it is scientifically invalid and unreliable.

The document identified in Table C.3-1 apparently refers to a paper dated December 1989 written by Dr. Arthur J. Newell of the New York State Department of Environmental Conservation entitled "Sediment Criteria - December 1989." The cover of this paper states:

> "Note: This document is used as a guidance by the Division of Fish and Wildlife. It is neither a standard nor a policy of the Department."

Because this document has not been expressly adopted by the State as an official standard or policy, it cannot reasonably be said to have been "developed" by the State. It is thus quite clear that the paper was not "developed by EPA, other federal agencies, or states" (NCP § 300.400(g)(3)). Therefore, it fails to meet the NCP's express definition of a TBC.

Furthermore, the paper itself is clearly not intended to be used in establishing sediment clean-up goals. Its sole purpose is to propose a methodology for mathematically simulating the fate and transport of a contaminant found in sediments to the evaluate the effects on human health and the environment of such contaminants. Thus, if it is relevant all to the RI/FS process, it is relevant to the site characterization, human health risk, and ecological assessment elements. EPA has not used the paper for such purposes, and GE's comments on those elements of the Phase 1 Report are contained in Sections 2 and 3 of these comments. In any event, it should be clear that the unpromulgated, unofficial NYSDEC paper it is not relevant to any of the NCP's evaluation criteria and therefore cannot be "appropriate" for use in setting remedial goals or in comparing alternatives (NCP § 300.400(e)(9)).

Lastly, if by some stretch of the NCP standard the paper is found to be a "state" guidance that is "appropriate," the preamble to the NCP (see 55 Fed. Reg. 8745) makes it clear that TBCs must be evaluated by EPA for "reliability and validity." As far as GE is aware, EPA has not performed any

evaluation of this non-peer reviewed paper and must do so if it is to be used as a TBC. In fact, EPA has rejected sediment criteria for compounds such as PCBs, due to significant technical and scientific concerns regarding the validity of the approach (i.e., equilibrium partitioning) employed in the paper.

Even a cursory review of the paper, however, reveals a fundamental flaw. The approach described in the paper relies on the bioaccumulation factor (BAF) approach to relate PCB levels in fish and PCB levels in water. As discussed in Section 2.0 of these comments, the BAF approach (which assumes a linear relationship between PCB levels in fish and PCB levels in water) has no physical, chemical or biological basis and is invalid for the entire range of PCB concentrations. Moreover, the paper's reliance on the BAF approach is additionally flawed because the paper fails to account for the fact that different PCB congeners behave differently in different environmental media and have significantly different toxicities and physical-chemical properties.

GE therefore believes that the sediment criteria paper is improperly identified in the Phase 1 Report as a potential TBC. It is not a guidance adopted or used by any State agency, it is not applicable to the setting of remedial goals, and it is scientifically unreliable and invalid.

D. RECENT SCIENCE ON PCB TOXICOLOGY

1. Institute for Evaluating Health Risks. 1991. Reassessment of Liver Findings in Five PCB Studies in Rats.

2. Comments of the General Electric Company on the Advanced Notice of Proposed Rulemaking of the United States Environmental Protection Agency Concerning Disposal of Polychlorinated Biphenyls, 55 F.R. 26738. 1990.

3. Chase, K.H., J. Doull, S. Friess, J.V. Rodricks and S. Safe. 1989. Evaluation of the Toxicology of PCBs.

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REASSESSMENT OF LIVER FINDINGS in FIVE PCB STUDIES IN RATS

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REASSESSMENT OF LIVER FINDINGS IN PCB STUDIES IN RATS

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Pathology of Chronic Polychlorinated Biphenyl (PCB) Feeding in Rats. Schaeffer, E., Greim, H., & Goessner, W. Tox and Applied Pharm. (1984) 75:, 272-288.

IEHR

Institute for Evaluating Health Risks

President John A. Moore

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NAS-Beckman Center
Irvine, California

July 1, 1991

The Honorable Hank Habicht
Deputy Administrator
Environmental Protection Agency
401 M Street S.W.
Washington, DC 20460

Dear Hank:

The Institute for Evaluating Health Risks has just completed a project, funded by General Electric, in which the pathological diagnoses in five key rat PCB studies were reassessed. This reassessment is the consensus diagnoses of eminent pathologists who are particularly experienced in rodent liver tumors. Using these diagnoses the studies were then analyzed for consistency of result and to determine whether the differences in tumor incidence and type were due to the differing levels of chlorination in the tested PCB mixtures.

The analysis clearly indicates that a reconsideration of the Agency's traditional PCB cancer risk policy is warranted. The policy positions to which I refer are: 1) an assumption that all PCB formulations are probable human carcinogens; 2) the assumption that all PCB formulations have the same quantitative potency to cause cancer.

Both of these positions were initially established years ago when our knowledge base from which to determine the cancer potential of PCBs was meager. They represent the use of conservative default assumptions. However, since then new data and knowledge have accrued that have not been effectively incorporated into the PCB risk assessment. Data, when available, should have priority over default assumptions.

A revised Agency PCB cancer risk assessment should reflect the following:

<u>Develop separate risk assessments for each of the major PCB formulations.</u>

The reassessed data underscore that there are major differences in carcinogenic potential based on the degree of chlorination of the PCB mixture. While the results from studies of mixtures with 60% chlorination consistently report a high incidence of liver tumors, studies in rats which were fed mixtures with 54% or 42% chlorination did not detect statistically significant elevations of liver tumors.

It is not proper to continue a policy which does not consider data, developed subsequent to the initial judgement, that demonstrates other formulations are either not carcinogens or at best, weak carcinogens. There is precedent for such action; several years ago the Science Advisory Panel, which advises the State of California on cancer designations under Proposition 65, voted to recommend Aroclor 1260 as a carcinogen rather than all PCBs.

Utilize all available data when calculating cancer potency for PCB mixtures that have 60% chlorination

When one compares the consensus pathology diagnoses across four studies, in three different laboratories, there appears to be no scientific basis for continuing the practice of selecting only part of the available data for deriving potency estimates. Using this approach, the current EPA value of 7.7 would be replaced with a value of approximately 1.9.

I am not asking you to focus on an issue that is only of arcane scientific interest. The current cancer policy is clearly overstating the cancer risks associated with many exposures to PCBs in the environment. In a number of instances it is driving regulatory decisions that, by any standard are a major economic impact for, at best, trivial public health gain. As an illustration, mixtures with 60% or greater chlorination comprised about 12% of total PCB sales in this country; yet current policy calculates all PCB exposure as if it were equivalent to Aroclor 1260. While PCBs in the environment undergo changes in composition none develops into the chemical "fingerprint" that identifies Aroclor 1260. Therefore, 88% of the PCB that was used is being treated as if it were a potent carcinogen when the data indicate that these lower chlorinated mixtures are either of markedly diminished potency or not carcinogenic at all!

A request to develop a risk assessment utilizing all pertinent data, I believe, is consistent with the Agency's stated goals of focusing on risks which represent true public health or environmental concern and of reducing the uncertainties in risk assessment by applying sound scientific knowledge.

I would be pleased to work with the Agency in explaining the results of this project and discussing alternative approaches to estimating PCB risks. A copy of the pathology reassessment and a letter sent to Agency colleagues which provides greater details on risk related issues is enclosed.

Sincerely,

/s/

John A. Moore

IEHR

Institute for Evaluating Health Risks

President John A. Moore

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NAS-Beckman Center
Irvine, California

July 1, 1991

The Honorable Erich Bretthauer
Assistant Administrator
Office of Research and Development
Environmental Protection Agency
401 M Street SW
Washington, DC 20460

Dear Erich:

The Institute for Evaluating Health Risks has just completed a project in which the pathological diagnoses in five key rat PCB studies were reassessed.[1] Based on the results of this reassessment, a copy of which is enclosed, these studies could then be analyzed for consistency of result and it could be determined whether the differences in tumor incidence and type were due to the differing levels of chlorination in the tested PCB mixtures. The analysis clearly indicates that a reconsideration of the Agency's traditional cancer risk policy is warranted.

In the studies that were reviewed in this project rats were chronically exposed to commercial PCB formulations with three different levels of chlorination. The results of the pathology reassessment are briefly summarized as follows:

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[1] The project, which was funded by General Electric, was managed by the Institute for Evaluating Health Risks; coordination of the pathology reassessment was performed by Experimental Pathology Laboratories Inc.

These specific studies were selected because they were utilized or discussed in previous EPA risk assessments and they represent the best studies for evaluating the cancer potential of these mixtures of chemicals.

reaffirmed that chronic dietary exposure of rats, in three different studies, to 60% chlorinated PCB formulations resulted in the development of benign and malignant liver tumors;

reaffirmed that chronic exposure of rats to a PCB formulation that was 54% chlorinated did not yield a statistically significant increase of either benign or malignant tumors;

revealed that rats chronically exposed to a PCB formulation that was 42% chlorinated did not develop any increase in malignant tumors or a statistically significant increase in benign tumors.

These reassessment results indicate that the following two traditional EPA PCB policy positions be reconsidered: 1) an assumption that all PCB formulations are probable human carcinogens; 2) the assumption that all PCB formulations have the same quantitative potency to cause cancer.

Both of these positions were initially established years ago when our knowledge base from which to determine the cancer potential of PCBs was meager. They represent the use of conservative default assumptions. However, since then new data and knowledge have accrued that have not been effectively incorporated into the PCB risk assessment.[2]

I believe that a revised PCB cancer risk assessment should reflect the following:

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[2] Because of insufficient data default assumptions commonly are a necessary component of a risk assessment. However, there is another policy position which should guide the decision that determines the use of defaults; that overarching policy should establish a clear bias for the use of data whenever it is available. In other words the operant policy position is to use data, the burden should lie on the risk assessor to clearly establish why available data should not be used before it can be replaced by a default assumption.

Develop separate risk assessments for each of the major PCB formulations.

The reassessed data underscore that there are major differences in carcinogenic potential based on the degree of chlorination of the PCB mixture. While the results from studies of mixtures with 60% chlorination consistently report a high incidence of liver tumors studies in rats which were fed mixtures with 54% or 42% chlorination did not detect statistically significant elevations of liver tumors. It is not proper to continue a policy which does not consider data, developed subsequent to the initial judgement, that demonstrates other formulations are either not carcinogens or at best, weak carcinogens. There is precedent for such action; several years ago the Science Advisory Panel, which advises the State of California on cancer designations under Proposition 65, voted to recommend Aroclor 1260 as a carcinogen rather that list all PCBs.

The tissue diagnoses of the expert group of pathologists should be used for risk assessment.

There are three factors that support the use of these consensus diagnoses:

1) it reflects the use of current pathology conventions that are endorsed by the National Toxicology Program and the Environmental Protection Agency;

2) it represents the consensus opinion of pathologists that are experienced in the evaluation of rodent bioassays; specifically liver tumors.

3) the results of the present review permits greater confidence that observed differences in tumor incidence and type in each study are due to differences in the test substances.

Utilize all available data when calculating cancer potency for PCB mixtures that have 60% chlorination.

There is no logical basis to continue the current practice of only using the results obtained in female Sprague Dawley rats. A comparison of the results of each of these studies[3] shows a striking similarity in the nature of the tumor response. It should be noted that three separate strains of rats were used and that the similarity of response is apparent when one compares female Sherman rats, male Wistar rats, and female Sprague Dawley rats. Male Sprague Dawley rats, while developing the same type of liver tumors, did so at a lower incidence. To assume that this reduced response reflects a generic tendency of male rats not to develop tumors is not supported by the data. The greatest incidence of liver tumors (91.2%) was observed in male Wistar rats. The results in male Wistar rats also do not support continuing the practice of censoring the male Sprague Dawley results from the calculation of a cancer slope factor.

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[3] Induction of Liver Tumors in Sherman Strain Rats By Polychlorinated Biphenyl Aroclor 1260. Kimbrough, R.D. et al, JNCI (1975) 55:6, 1453-1459.

Polychlorinated Biphenyl Induction of Hepatocellular Carcinoma in the Sprague Dawley Rat. Norback, D.H. & Weltman, R.H., Env Hlth Perspect (1985) 60:, 97-105.

Pathology of Chronic Polychlorinated Biphenyl (PCB) Feeding in Rats. Schaeffer, E., Greim, H., & Goessner, W., Tox & Applied Pharm. (1984) 75:, 272-288.

When using the results from each of these studies one should apply a consistent decision rule to the censoring of animals from studies; each author used a different convention in their publications. Observing the convention employed by the National Toxicology Program may be more appropriate and consistent for all studies.[4] The group size in several of these studies would increase if this recommendation were adopted.

Employing the geometric mean of the cancer potency factors of the four study groups, female Sherman, male Wistar, male Sprague Dawley, and female Sprague Dawley rats would reflect a less arbitrary use of all existing data. There is ample precedent for this approach in a number of Agency decisions. The geometric mean, using the re-evaluation results, would yield a cancer potency factor of approximately 1.9. The current value calculated by EPA is 7.7 using only the female Sprague Dawley rat.

The reassessment of the NCI study[5] clarifies the significance of "nodular hyperplasia"

This study which evaluated a PCB mixture with 54% chlorination, essentially reaffirmed the original findings that the bioassay did not show a carcinogenic response in either male or female F344/N rats. The group size at each treatment level was 24 rats.

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[4] Censor all rats that died during the first year of the study or censor rats that died prior to the diagnosis of the first tumor in a target organ; whichever date is earlier.

[5] Bioassay of Aroclor 1254 for Possible Carcinogenicity. NCI Carcinogenisis Technical Report Series, Number 38, 1978.

Utilizing the current pathology nomenclature the consensus diagnoses by the expert panel classified "nodular hyperplasia" lesions, a designation used in the original report, as nonneoplastic. Therefore, continuing to incorporate the incidence of nodular hyperplasia in a cancer potency calculation, as was done in the most recent Water Quality Criteria Document[6] would fail to have a supportable scientific basis.

Rather than exclusively focus on how to estimate a cancer potency factor for the 54% chlorination PCB mixture I would urge consideration of a more fundamental question; namely, the estimation of cancer potency from any negative study.

<u>The reassessment of the pathology diagnoses of lesions in the liver of rats fed a PCB mixture containing 42% chlorination reveals that there is no statistically significant increase in tumors.</u>[7]

This study, which was performed in parallel with a PCB mixture with 60% chlorination, has not been accorded the attention that it deserves from a risk assessment perspective.[8]

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[6] Drinking Water Criteria Document for Polychlorinated Biphenyls, April 1988, (PB89-192256) pp VIII-32 to VIII-35.

[7] Liver tumor incidence in controls 8/120 (hepatocellular adenoma 6/120, hepatocellular carcinoma 2/120). Liver tumor incidence in treated group 16/128 (hepatocellular adenoma 14/128, hepatocellular carcinoma 2/128). Fisher exact test, one tailed, p =.098). It is arguable that a two tailed test should be used given that a decrease in pituitary tumors and endocrine tumors was reported in several of these studies. A two tailed test would further erode the p value.

[8] Pathology of Chronic Polychlorinated Biphenyl Feeding in Rats. Schaeffer, E., Greim, H., & Goessner, W., Tox. & Applied Pharm. (1984) 75:, 272-288

Factors which underscore the value of this study include:

1) it is the only major study of a PCB mixture with this level of chlorination.

2) it has far better statistical power to detect an effect than do most bioassays, e.g., the number of animals studied were about two and a half times greater than required by EPA or used by the National Toxicology Program.

3) the selection of male rats as the test subject would not appear to be a limitation. A parallel group of male rats, fed a PCB mixture containing 60% chlorine, yielded a liver tumor incidence of 91%, the highest incidence reported in any of the studies that were reassessed.

4) the study duration was approximately 118 weeks, this is three months longer than the protocol requirements of either EPA or the National Toxicology Program. It is generally held that studies of longer duration favor the detection of tumors, particularly with these types of chemicals.

I am not asking you to focus on an issue that is only of arcane scientific interest. The current cancer policy is clearly overstating the cancer risks associated with many exposures to PCBs in the environment. In a number of instances it is driving regulatory decisions that, by any standard are a major economic impact for, at best, trivial public health gain. As an illustration, mixtures with 60% or greater chlorination were about 12% of total PCB sales in this country; current policy calculates <u>all</u> PCB exposure as if it were equivalent to Aroclor 1260.

While PCBs in the environment undergo changes in composition they do not develop into the chemical "fingerprint" that identifies Aroclor 1260. Therefore, 88% of the PCB that was used is being treated as if it were a potent carcinogen when the data indicate that these lower chlorinated mixtures are either of markedly diminished potency or not carcinogenic at all!

A request to develop a risk assessment utilizing all pertinent data, I believe, is consistent with the Agency's stated goals of focusing on risks which represent true public health or environmental concern and of reducing the uncertainties in risk assessment by applying sound scientific knowledge.

I would be pleased to work with the Agency in explaining the results of this project and discussing alternative approaches to estimating PCB risks.

Sincerely,

/s/

John A. Moore

EPL

EXPERIMENTAL PATHOLOGY LABORATORIES, INC.

REASSESSMENT OF LIVER FINDINGS IN PCB STUDIES IN RATS

PATHOLOGY WORKING GROUP REVIEW

Submitted to:

Institute for Evaluating Health Risks
Washington, DC 20005

Submitted by:

Experimental Pathology Laboratories, Inc.
Research Triangle Park, NC 27709

June 27, 1991

PWG PARTICIPANTS:

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Experimental Pathology
Laboratories, Inc. (Chairperson)

Dr. W. Ray Brown
Research Pathology Services, Inc.

Dr. Ernest E. McConnell
Consultant

Dr. James A. Popp
Chemical Industry Institute
of Toxicology

Dr. Robert A. Squire
Johns Hopkins University

Dr. Jerrold M. Ward
Consultant

Dr. Deborah A. Banas
Experimental Pathology
Laboratories, Inc.
(Reviewing Pathologist)

REASSESSMENT OF LIVER FINDINGS IN PCB STUDIES IN RATS

PATHOLOGY WORKING GROUP REVIEW

NARRATIVE SUMMARY

INTRODUCTION

Polychlorinated biphenyls (PCBs) are compounds whose physical and chemical properties led to their widespread use for a number of commercial applications. Because of their extensive use in the past and their resistance to chemical and biological breakdown, PCBs are now widely distributed in the environment.

PCBs were manufactured commercially by the chlorination of biphenyl. The number and placement of chlorine atoms introduced into the biphenyl molecule determines each PCBs' structure. There are 209 possible PCB congeners or homologs. Commercial PCB formulations are composed of complex mixtures of individual PCBs rather than a single congener. The percent chlorine by weight increases as the average number of chlorine atoms per biphenyl is increased. The chlorine content of various commercial formulations differs according to the desired physical characteristics for specific applications (Siberhorn, et al., 1990).

A number of studies have been undertaken to assess the potential toxicity and carcinogenicity of commercial PCB preparations. The main target organ of PCBs is the liver. A number of investigators have studied the potential carcinogenic effect of various PCBs in the

liver of rats and mice. These studies have indicated that benign or malignant hepatocellular tumors and various nonneoplastic hepatic changes are observed in the liver of rodents when given at appropriate doses for extended periods of time. These studies also indicated that PCB mixtures with a high chlorine content are more potent in inducing neoplastic nodules and hepatocellular carcinomas than mixtures with less chlorination. In most of these studies, the criteria for diagnosis and nomenclature designations according to Squire and Levitt were used by the investigators to classify the hepatocellular changes (Siberhorn, et al., 1990).

In the recent past, there have been changes in the criteria and nomenclature for hepatoproliferative lesions of rats (Maronpot, et al., 1986). These changes resulted from the increased accumulation of data, as well as a better understanding of the mechanisms of toxicity and carcinogenesis. In light of these changes, it was considered reasonable to reexamine the livers from several earlier PCB studies to assess the risk posed by these compounds based on the current understanding of hepatic changes. At the request of the Institute for Evaluating Health Risks (IEHR), Experimental Pathology Laboratories, Inc. (EPL) assembled all liver slides from five (5) key chronic PCB studies for the purpose of reassessment of the liver findings following current diagnostic criteria and nomenclature. These studies included the following: Aroclor 1260 in female Sherman rats (Kimbrough, et al.,

1975); Aroclor 1260 in male and female Sprague-Dawley rats (Norback and Weltman, 1985); National Cancer Institute (NCI) Aroclor 1254 study in male and female Fischer 344 rats (NCI, 1977); Clophen A 60 in male Wistar rats (Schaeffer, et al., 1984); and Clophen A 30 in male Wistar rats (Schaeffer, et al., 1984). Additionally, the liver slides from male and female rats used in a reproduction study in male and female Sherman rats with Aroclor 1260, in which the exposure was limited to nine months, were reexamined to characterize the degree and extent of hepatic lesions resulting from subchronic exposure to Aroclor 1260 (Linder, et al., 1974). References for each of these studies are included in Appendix F. The concurrent review of these studies provide a unique opportunity to compare the incidence, type and severity of hepatic lesions observed in each.

A summary of the experimental design for each of the studies included in this current review is presented on the following table.

SUMMARY OF EXPERIMENTAL DESIGN
PCB STUDIES IN RATS

Study	Chemical	% Cl by Wt.	Strain	Sex	Dosage levels	Duration of Exposure*
Kimbrough, et al., 1975	Aroclor 1260	60%	Sherman	F	100 ppm	23 Months
Norback and Weltman, 1985	Aroclor 1260	60%	Sprague-Dawley	M/F	100 ppm for 16 months followed by 50 ppm for 8 months then control diet for 5 months	29 Months
Schaeffer, et al., 1984	Chlopen A 60	60%	Wistar	M	100 ppm	801-832 Days
NCI, 1977	Aroclor 1254	52-54%	F344	M/F	25 ppm, 50 ppm 100 ppm	104-105 Weeks
Schaeffer, et al., 1984	Clophen A 30	40-42%	Wistar	M	100 ppm	801-832 Days
Linder, et al., 1974 (Reproduction Study)	Aroclor 1260	60%	Sherman	M/F	100 ppm	9 Months

*Duration of exposure published by original investigator.

METHODS

After assembling the hematoxylin and eosin stained microscope slides and data from each of the studies to be reviewed, the procedures developed by the National Toxicology Program which utilizes a Pathology Working Group (PWG) were generally followed to conduct the review. The PWG members consisted of Veterinary Pathologists with extensive experience in the microscopic evaluation and interpretation of hepatic changes observed in bioassay studies conducted in rodents. The PWG's task was to confirm the incidence of hepatocellular neoplasms, resolve diagnostic discrepancies, validate treatment-related effects and discuss the biological significance of the potential effects present.

Prior to the PWG review, all slides within each individual study were randomized by animal number and then coded in ascending numerical order. The randomized slides were examined without knowledge of treatment group by the Reviewing Pathologist, Dr. Deborah Banas, Experimental Pathology Laboratories, Inc. The reviewing pathologist recorded all changes present in the liver sections including both neoplastic and nonneoplastic lesions. After microscopic examination, the data was decoded and presented by treatment group for interpretation of the results and preparation for the Pathology Working Group review.

The Pathology Working Group was chaired by Dr. Jerry F. Hardisty, Experimental Pathology Laboratories, Inc., who organized and presented the material to a panel of five board certified Veterinary

Pathologists. The PWG review was performed on May 29-31, 1991 in the Research Triangle Park, North Carolina. Individuals attending or participating in the PWG review are listed as follows:

Dr. W. Ray Brown	(PWG Participant)
Dr. E.E. McConnell	(PWG Participant)
Dr. James A. Popp	(PWG Participant)
Dr. Robert A. Squire	(PWG Participant)
Dr. Jerrold M. Ward	(PWG Participant)
Dr. Deborah A. Banas	(Reviewing Pathologist)
Dr. Jerry F. Hardisty	(PWG Chairperson)
Dr. Ronald Moch	(Observer, FDA)
Dr. D. Singh	(Observer, EPA)
Dr. Jack Moore	(Observer, IEHR)
Dr. Renate Kimbrough	(Observer, IEHR)
Dr. W. Goessner	(Observer, GSF)
Dr. Diane Norback	(Observer, University of Wisconsin)
Dr. William M. Busey	(Observer, EPL)

Each participant recorded his diagnoses and comments on worksheets which were prepared by the PWG Chairman. The worksheets for each participant are on file at EPL. Each lesion was discussed by the group, reexamined if necessary and the final opinions were recorded on the Chairperson's Worksheets also maintained on file at EPL, Inc. The consensus diagnoses of the PWG was reached when at least three of five PWG participants were in agreement.

After the PWG completed the slide review for each study and the diagnoses recorded by the PWG Chairperson, the slides were decoded by treatment group. The consensus diagnoses was entered into a

computerized pathology reporting system at EPL, and Histopathology Incidence Tables for individual animals and Summary Incidence Tables were prepared for evaluation and interpretation of the PWG findings. The Histopathology Incidence Tables and Summary Incidence Tables are presented in Appendices A - E for each of the studies reviewed.

In addition to providing specific diagnoses for neoplastic and hyperplastic hepatocellular lesions present on the slides examined, the PWG participants also provided comments and opinions on the presence of a variety of nonneoplastic changes including foci of cellular alteration, and other lesions which may be indicative of potential hepatotoxicity. The PWG members were consistently in agreement with the reviewing pathologist concerning the nonneoplastic changes present. Since the reviewing pathologist had examined all liver slides in both control and treated animals in these studies using consistent criteria for diagnosis and severity grading, there was agreement by the PWG members to accept the reviewing pathologist's findings for nonneoplastic changes as the "consensus diagnosis". Therefore the consensus diagnoses presented in the Histopathology Incidence Tables and Summary Incidence Tables represent the majority opinion of the PWG for all neoplastic and hyperplastic changes and the reviewing pathologist's diagnosis for other nonneoplastic changes.

The diagnostic criteria used by the PWG participants and the reviewing pathologist for diagnosis of foci of cellular alteration,

hepatocellular hyperplasia and hepatocellular neoplasms is summarized below:

Foci of Cellular Alteration

1. Localized lesions recognized by virtue of tinctorial staining variation from surrounding hepatic parenchyma.
2. Usually do not compress but merge imperceptibly with surrounding parenchyma.
3. Minimal disruption of hepatic lobular architecture.
4. Hepatocytes within focus may have clear, eosinophilic, or basophilic cytoplasm or a mixture of these and may be larger or smaller than surrounding hepatocytes.

Focal Hepatocellular Hyperplasia

1. Usually a multifocal nodular lesion found associated with previous or concurrent hepatic damage. In this context, may be considered as multifocal regenerative hyperplasia.
2. Lesion consists of spherical proliferation of hepatocytes without nuclear atypia. May contain cytologic alterations similar to those seen in foci of cellular alteration.
3. An increased number of mitoses may be evident. Hyperplastic cells may be hypertrophic or contain intracytoplasmic vacuoles. The cells within a hyperplastic focus are usually uniform and have a homogeneous growth pattern.

4. Hepatic lobular architecture is evident but may be distorted. Portal triads can often be found within hyperplastic foci.

Hepatocellular Adenoma

1. Nodular proliferations of hepatocytes which are sharply demarcated by definite compression of surrounding hepatic parenchyma and usually by virtue of tinctorial staining differences.
2. Hepatic plates of an adenoma are usually not continuous with surrounding liver plates but impinge with them at a sharp angle.
3. Loss of normal lobular architecture.
4. Often have an increased mitotic index, may contain areas of cellular atypia, and have an irregular growth pattern.
5. Cells within an adenoma may be degenerative, hypertrophic, and/or contain intracytoplasmic vacuoles.

Hepatocellular Carcinoma

1. Usually considerably larger and more irregular than hepatocellular adenoma.
2. Compress or extend into surrounding hepatic parenchyma.
3. Characterized by one or more of the following: cellular atypia, local invasiveness, haphazardly arranged cells, broad sheets of cells, "trabecular" patterns, gland-like formations.

RESULTS

A. Aroclor 1260 in Female Sherman Rats (Kimbrough, et al., 1975)

Only one control female rat had a hepatocellular neoplasm. Control female 8231 had a hepatocellular carcinoma. In treated female rats, 21 rats had hepatocellular carcinomas and 135 rats had hepatocellular adenomas. A few of the treated female rats had both hepatocellular adenomas and hepatocellular carcinomas. The hepatocellular carcinomas were well-differentiated trabecular types. The hepatocytes varied from a normal appearance to enlarged, acidophilic or diffusely basophilic cells. The cytoplasm often contained eosinophilic inclusions within vacuoles. Hepatocellular adenomas were generally spherical and well demarcated. The cells in the adenomas were generally enlarged with nuclear and cytoplasmic features similar to the carcinomas. In three of the treated rats the liver tumors had glandular, papillary patterns giving the appearance of both hepatocytic and biliary epithelium (8326, 8377 and 8406). These tumors were considered to represent a subtype of hepatocellular adenoma or carcinoma and were not given a unique diagnosis. A summary of the incidence of rats with only hepatocellular adenoma and those with at least one hepatocellular carcinoma is presented as follows:

	Control	Aroclor 1260
No. Examined	187	189
No. Animals with only Hepatocellular Adenoma	0 (0%)	117 (61.9%)
No. Animals with at Least one Hepatocellular Carcinoma	1 (0.5%)	21 (11.1%)
Total Animals with Hepatocellular Neoplasms	1 (0.5%)	138 (73.0%)

Eosinophilic foci were present in 173 of the treated rats and only seven of the control rats. Other foci frequently occurred jn addition to the eosinophilic foci in treated rats. The incidence of focus/foci of cellular alteration in the liver of control and treated female rats is presented as follows:

	Control	Aroclor 1260
No. Examined	187	189
Focus/Foci, Eosinophilic	7 (3.7%)	173 (91.5%)
Focus/Foci, Basophilic	4 (2.1%)	67 (35.4%)
Focus/Foci, Clear Cell	14 (7.5%)	67 (35.4%)
Focus/Foci, Mixed Cell	1 (0.5%)	38 (20.1%)
No. Animals with Any Type of Focus/Foci	25 (13.4%)	177 (93.7%)

Nonneoplastic lesions which appeared to be increased with treatment included centrilobular hepatocytomegaly, centrilobular fatty change,

bile duct hyperplasia, and pigment deposition. Oval cell proliferation, angiectasis, and dilated bile ducts were also increased in treated rats. Focal necrosis was often noted in treated rats in association with hepatocellular carcinomas. Cholangiofibrosis characterized by atypical bile ducts surrounded by abundant dense collagenous connective tissue was present in three treated rats. The incidence of nonneoplastic lesions which were increased in treated rats as compared to control rats is presented below:

	Control	Aroclor 1260
No. Examined	187	189
Centrilobular Hepatocytomegaly	1 (0.5%)	108 (57.1%)
Centrilobular Fatty Change	1 (0.5%)	32 (16.9%)
Bile Duct Hyperplasia	4 (2.1%)	29 (15.3%)
Pigment Deposition	9 (4.8%)	66 (34.9%)
Oval Cell Proliferation	2 (1.1%)	17 (9.0%)
Angiectasis	1 (0.5%)	17 (9.0%)
Dilated Bile Ducts, Focal	0 (0%)	14 (7.4%)
Focal Necrosis	0 (0%)	13 (6.9%)
Cholangiofibrosis	0 (0%)	3 (1.6%)

B. Reproduction Study With Aroclor 1260 in Male and Female Sherman Rats, F$_0$ Generation (Linder, et al., 1974)

Slight to severe centrilobular hepatocytomegaly was noted in all ten treated males, and minimal to moderately severe centrilobular hepatocytomegaly was present in seven of ten treated females. This lesion was characterized by enlarged hepatocytes, occasionally with atypical nuclei surrounding the central veins and, in the case of moderately severe or severe lesions, extending over a fair portion of the liver lobule. Cytoplasmic inclusions were prominent in these enlarged hepatocytes in two of the males. Minimal to slight centrilobular fatty change was noted in two of the treated females and focal fatty change occurred in one. Single eosinophilic foci were noted in two treated females. These lesions were characterized by foci of enlarged, eosinophilic hepatocytes having a ground-glass appearance to the cytoplasm and having a distinctly different tinctorial appearance compared to the surrounding parenchyma. Multiple, small, clear cell foci were noted in one treated male and were characterized by enlarged hepatocytes having a clear or slightly granular pale cytoplasm. Brown pigment deposition was noted in four of the treated females.

Other lesions noted in the liver sections of both control and treated rats were microgranulomas. These lesions consisted of focal accumulations of macrophages and mononuclear inflammatory cells, and

were unrelated to treatment with Aroclor 1260. The incidence of these changes are summarized as follows:

	Male Rats		Female Rats	
	Control	Aroclor 1260	Control	Aroclor 1260
No. Examined	10	10	10	10
Centrilobular Hepatocytomegaly	0 (0%)	10 (100%)	0 (0%)	7 (70%)
Focus/Foci, Eosinophilic	0 (0%)	0 (0%)	0 (0%)	2 (20%)
Focus/Foci, Clear Cell	0 (0%)	1 (10%)	0 (0%)	0 (0%)
Fatty Change, Centrilobular	0 (0%)	0 (0%)	0 (0%)	2 (20%)
Fatty Change, Focal	0 (0%)	0 (0%)	0 (0%)	1 (10%)
Cytoplasmic Inclusions	0 (0%)	2 (20%)	0 (0%)	0 (0%)
Microgranuloma(s)	3 (30%)	2 (20%)	5 (50%)	3 (30%)
Pigment Deposition	0 (0%)	0 (0%)	0 (0%)	4 (40%)

C. Aroclor 1260 in Male and Female Sprague-Dawley Rats (Norback and Weltman, 1985)

Hematoxylin and eosin stained slides were examined from 31 male control, 40 male treated, 45 female control and the 46 female treated Sprague-Dawley rats. Due to the fact that partial hepatectomies were performed on two control females (30 and 31) and two treated males (189 and 212), these animals are represented twice in the Histopathology Incidence Tables. The animal number followed by "A" represents the liver section examined following the partial hepatectomy and followed by "B" for the liver section examined at termination.

A marked sex difference in the number of hepatocellular neoplasms was present in this study. No hepatocellular neoplasms were present in any of the control male rats. In treated male rats four had

hepatocellular adenomas and one had a hepatocellular carcinoma. Only one control female rat had a hepatocellular adenoma. Hepatocellular adenomas were present in 29 treated female rats and hepatocellular carcinomas were present in 19 treated female rats. Additionally, three treated female rats had cholangiocarcinomas. Most of the hepatocellular carcinomas were well-differentiated trabecular neoplasms. Hepatocellular neoplasms in treated female rats 192, 212, 213 and 214 had glandular, papillary patterns similar to that described above for the Kimbrough study in female Sherman rats. Hepatocellular adenomas were generally spherical, well-demarcated tumors composed of well-differentiated cells which produced mild to moderate compression of the surrounding hepatic parenchyma. A few of the treated female rats had both hepatocellular adenoma and carcinoma. A summary of the incidence of male and female rats with only hepatocellular adenoma and rats with at least one hepatocellular carcinoma is presented as follows:

	Male Rats		Female Rats	
	Control	Aroclor 1260	Control	Aroclor 1260
No. Examined	31	40	45	46
No. Animals with only Hepatocellular Adenoma	0 (0%)	4 (10%)	1 (2.2%)	22* (47.8%)
No. Animals with at Least one Hepatocellular Carcinoma	0 (0%)	1 (2.5%)	0 (0%)	19* (41.3%)
Total Animals with Hepatocellular Neoplasms	0 (0%)	5 (12.5%)	1 (2.2%)	41 (89.1%)

*Two female rats with hepatocellular adenoma and one with hepatocellular carcinoma also had a cholangiocarcinoma.

An increased incidence of eosinophilic cell foci was present in treated male and female rats as compared to control rats. Although a

few basophilic, clear cell and mixed cell foci were also observed they did not occur in a treatment-related manner. A summary of the incidence of foci of cellular alteration in the liver in Sprague-Dawley Rats is presented below:

	Male Rats		Female Rats	
	Control	Aroclor 1260	Control	Aroclor 1260
No. Examined	31	40	45	46
Focus/Foci, Eosinophilic	1 (3.2%)	16 (40%)	5 (11.1%)	36 (78.3%)
Focus/Foci, Basophilic	1 (3.2%)	0 (0%)	2 (4.4%)	1 (2.2%)
Focus/Foci, Clear Cell	4 (12.9%)	0 (0%)	1 (2.2%)	0 (0%)
Focus/Foci, Mixed Cell	0 (0%)	2 (5%)	0 (0%)	0 (0%)
No. Animals with Any Type of Focus/Foci	5 (16.1%)	16 (40%)	7 (15.6%)	36 (78.3%)

Other hepatic lesions which appeared to be increased in treated groups as compared to control male and female rats included centrilobular hepatocytomegaly, centrilobular fatty change and centrilobular necrosis. In treated female rats the incidence of bile duct hyperplasia, cholangiofibrosis, cystic bile ducts, periportal fibrosis and pigment deposition was also increased in incidence as compared to control female rats. The incidence of selected hepatocellular lesions which were increased in incidence in treated male and/or female rats as compared to controls are presented as follows:

	Male Rats		Female Rats	
	Control	Aroclor 1260	Control	Aroclor 1260
No. Examined	31	40	45	46
Centrilobular Hepatocytomegaly	0 (0%)	15 (37.5%)	0 (0%)	5 (10.9%)
Centrilobular Fatty Change	0 (0%)	17 (42.5%)	0 (0%)	2 (4.3%)
Centrilobular Necrosis	0 (0%)	7 (17.5%)	2 (4.4%)	4 (8.7%)
Bile Duct Hyperplasia	6 (19.4%)	8 (20%)	2 (4.4%)	19 (41.3%)
Cholangiofibrosis	0 (0%)	0 (0%)	0 (0%)	4 (8.7%)
Cystic Bile Ducts	0 (0%)	1 (2.5%)	1 (2.2%)	5 (10.9%)
Periportal Fibrosis	2 (6.5%)	3 (7.5%)	0 (0%)	12 (26.1%)
Pigment Deposition	1 (3.2%)	2 (5.0%)	2 (4.4%)	13 (28.3%)

D. Clophen A 60 in Male Wistar Rats (Schaeffer, et al., 1984)

Hepatocellular adenomas were present in six control males and in 85 treated males. Hepatocellular carcinomas were present in two control males and in 67 treated males. Additionally one of the control males with a hepatocellular carcinoma also had a cholangiocarcinoma in the liver. Although most of the hepatocellular neoplasms consisted of well-differentiated adenomas and carcinomas, nine of the tumors present in the treated group had glandular, papillary patterns suggestive of a mixture of hepatocellular and biliary epithelium. Although this pattern was observed only in treated rats, it was considered to most likely represent a subclassification of hepatocellular adenoma or carcinoma and was not given a unique morphologic diagnosis. A few of the treated male rats had both hepatocellular adenomas and hepatocellular carcinomas. A summary of the incidence of male Wistar rats with only hepatocellular

adenoma and rats with at least one hepatocellular carcinoma is presented as follows:

	Control	Clophen A 60
No. Examined	120	125
No. Animals with only Hepatocellular Adenoma	6 (5.0%)	47 (37.6%)
No. Animals with at Least one Hepatocellular Carcinoma	2*(1.7%)	67 (53.6%)
Total Animals with Hepatocellular Neoplasms	8 (6.7%)	114 (91.2%)

*One control male also had cholangiocarcinoma.

Eosinophilic foci were present in the liver of 51 control and 101 treated rats. Additionally, the number of male rats with clear cell and mixed cell foci were greater in the treated group than in the control group. The incidence of focus/foci of cellular alteration in the liver of control and treated rats is presented as follows:

	Control	Clophen A 60
No. Examined	120	125
Focus/Foci, Eosinophilic	51 (42.5%)	101 (80.8%)
Focus/Foci, Basophilic	2 (1.7%)	4 (3.2%)
Focus/Foci, Clear Cell	7 (5.8%)	28 (22.4%)
Focus/Foci, Mixed Cell	7 (5.8%)	28 (22.4%)
No. Animals with Any Type of Focus/Foci	55 (45.8%)	108 (86.4%)

With the exception of an increased incidence of focal necrosis which was associated with hepatocellular carcinomas in rats receiving Clophen a 60, other nonneoplastic lesions appeared to be comparable or decreased in treated rats.

E. Clophen A 30 in Male Wistar Rats (Schaeffer, et al., 1984)

The Clophen A 30 study in Wistar rats was conducted at the same laboratory at the same time as the Clophen A 60 study discussed above and therefore shared the same control group. In the Clophen A 30 treated rats 14 hepatocellular adenomas and two hepatocellular carcinomas were present as compared to six adenomas and two carcinomas in the control group. All of the hepatocellular tumors in the control and treated rats occurred as singular tumors and none of the rats had both a hepatocellular adenoma and a hepatocellular carcinoma. One control male had a cholangiocarcinoma in addition to a hepatocellular carcinoma. A summary of the incidence of male rats with hepatocellular neoplasms is presented below:

	Control	Clophen A 30
No. Examined	120	128
Hepatocellular Adenoma	6 (5.0%)	14 (10.9%)
Hepatocellular Carcinoma	2 (1.7%)	2 (1.6%)
Total Hepatocellular Neoplasms	8 (6.7%)	16 (12.5%)

The incidences of focus/foci of cellular alteration of all types (basophilic, eosinophilic, clear cell and mixed cell) were greater in treated male rats than in control male rats. The incidence of focus/foci of cellular alterations present in control and treated groups is summarized as follows:

	Control	Clophen A 30
No. Examined	120	128
Focus/Foci, Eosinophilic	51 (42.5%)	98 (76.6%)
Focus/Foci, Basophilic	2 (1.7%)	15 (11.7%)
Focus/Foci, Clear Cell	7 (5.8%)	39 (30.5%)
Focus/Foci, Mixed Cell	7 (5.8%)	49 (38.3%)
No. Animals with Any Type of Focus/Foci	55 (45.8%)	106 (82.8%)

The incidence of other nonneoplastic lesions were either decreased or unchanged in the treated group as compared to the control group.

F. Aroclor 1254 in Male and Female Fischer 344 Rats (NCI, 1977)

The results of the PWG review generally confirmed the conclusion of the NCI Technical Report that "under the conditions of the bioassay Aroclor 1254 was not carcinogenic in Fischer 344 rats at the doses tested; however, a high incidence of hepatocellular proliferative lesions in both male and female rats was related to administration of the chemical".

During the PWG review, the PWG panel examined all sections of liver in which a diagnosis of a hepatocellular neoplasm or nodular hyperplasia had been either diagnosed by the reviewing pathologist or reported in the NCI Technical Report.

No hepatocellular neoplasms were observed in control male or female rats. Hepatocellular adenomas and/or carcinomas were present in low numbers in all treated groups. When present, they occurred as singular tumors and none of the affected rats had both a hepatocellular adenoma and a hepatocellular carcinoma. The incidences of hepatocellular neoplasms present in male and female F344 rats are summarized as follows:

Male F344 Rats

	Control	Low	Mid	High
No. Examined	24	24	24	23
Hepatocellular Adenoma	0 (0%)	1 (4.2%)	1 (4.2%)	1 (4.3%)
Hepatocellular Carcinoma	0 (0%)	0 (0%)	0 (0%)	2 (8.7%)
Total Animals with Hepatocellular Neoplasms	0 (0%)	1 (4.2%)	1 (4.2%)	3 (13%)

Female F344 Rats

	Control	Low	Mid	High
No. Examined	23	24	24	24
Hepatocellular Adenoma	0 (0%)	1 (4.2%)	2 (8.3%)	1 (4.2%)
Hepatocellular Carcinoma	0 (0%)	0 (0%)	0 (0%)	0 (0%)
Total Animals with Hepatocellular Neoplasms	0 (0%)	1 (4.2%)	2 (8.3%)	1 (4.2%)

Most of the hepatocellular lesions which were reported as hyperplasia, nodular in the NCI Technical Report were considered to be foci of cellular alteration when reexamined by the PWG. An increased number of foci of cellular alteration were present in treated groups in this study and consisted primarily of basophilic, eosinophilic and mixed cell foci. The incidences of foci of cellular alteration in the liver are summarized as follows:

Male F344 Rats

	Control	Low	Mid	High
No. Examined	24	24	24	23
Focus/Foci, Eosinophilic	0 (0%)	4 (16.7%)	5 (20.8%)	4 (17.4%)
Focus/Foci, Basophilic	0 (0%)	1 (4.2%)	0 (0%)	7 (30.4%)
Focus/Foci, Mixed Cell	0 (0%)	2 (8.3%)	4 (16.7%)	8 (34.8%)
Focus/Foci, Clear Cell	0 (0%)	0 (0%)	0 (0%)	0 (0%)
Total Animals with Any Type of Focus/Foci	0 (0%)	7 (29.2%)	9 (37.5%)	16 (69.6%)

Female F344 Rats

	Control	Low	Mid	High
No. Examined	23	24	24	24
Focus/Foci, Eosinophilic	0 (0%)	10 (41.7%)	13 (54.2%)	6 (25%)
Focus/Foci, Basophilic	2 (8.7%)	2 (8.3%)	0 (0%)	4 (16.7%)
Focus/Foci, Mixed Cell	0 (0%)	1 (4.2%)	0 (0%)	5 (20.8%)
Focus/Foci, Clear Cell	0 (0%)	0 (0%)	1 (4.2%)	2 (8.3%)
Total Animals with Any Type of Focus/Foci	2 (8.7%)	12 (50%)	14 (58.3%)	15 (62.5%)

Other lesions which occurred more frequently in Aroclor 1254 treated rats included centrilobular hepatocytomegaly which occurred in a few treated rats of each sex in each group and pigment deposition which occurred frequently in treated females, often in conjunction with focal/multifocal granulomatous hepatitis. The incidence of these nonneoplastic changes are presented as follows:

Male F344 Rats

	Control	Low	Mid	High
No. Examined	24	24	24	23
Centrilobular Hepatocytomegaly	0 (0%)	3 (12.5%)	3 (12.5%)	3 (13%)
Pigment Deposition	0 (0%)	0 (0%)	2 (8.3%)	2 (8.7%)
Focal/Multifocal Granulomatous Hepatitis	0 (0%)	0 (0%)	1 (4.2%)	3 (13%)

	Female F344 Rats			
	Control	Low	Mid	High
No. Examined	23	24	24	24
Centrilobular Hepatocytomegaly	0 (0%)	3 (12.5%)	1 (4.2%)	3 (12.5%)
Pigment Deposition	3 (13%)	16 (66.7%)	16 (66.7%)	18 (75%)
Focal/Multifocal Granulomatous Hepatitis	3 (13%)	4 (16.7%)	11 (45.8%)	14 (58.3%)

SUMMARY AND CONCLUSIONS

Of the six studies reviewed by the PWG, four studies used the PCB Aroclor 1260 or its equivalent, Clophen A 60. Three of these studies were long-term studies to examine the potential chronic toxicity and carcinogenic activity of Aroclor 1260 or Clophen A 60. The fourth study was a reproduction study in which the F_0 generation male and female rats were examined following nine months of exposure to 100 ppm Aroclor 1260. The other two studies examined by the PWG included long-term studies of Clophen A 30 and Aroclor 1254. These chemicals differ from Aroclor 1260 and Clophen A 60, in that they both have a lower chlorine content. Clophen A 30 has the lowest chlorine content of the chemicals reviewed.

The studies reviewed varied in the strain of rat used, the sex of rat utilized, and the age of sacrifice for tissue examination. The results of the PWG review revealed that the most prominent

difference determining the carcinogenic potential was the specific chemical tested. Additionally, strain and sex differences were noted in the spectrum of liver lesions present in the various studies reviewed.

In F_0 generation male and female Sherman rats that received 100 ppm of Aroclor 1260 for nine months in a reproduction study, centrilobular hepatocytomegaly was the principle effect and appeared to be slightly more severe in male rats than in female rats at this age. However, the early appearance of small eosinophilic foci occurred in female Sherman rats as did the deposition of brown pigment in Kupffer cells. The overall degree of toxicity in the liver of male and female rats following nine months of treatment was judged to be minimal and the centrilobular hepatocytomegaly represents an adaptive hepatocellular response to Aroclor 1260 by the liver.

One of the objectives of the PWG review was to compare the results of the long-term studies which were conducted in various strains of rats with different polychlorinated biphenyls. The incidence of hepatic neoplasms, foci of cellular alteration and centrilobular hepatocytomegaly are presented for each of the long-term studies on the following page:

Comparison of PMG Results for Chronic Studies Conducted in
Four Strains of Rats with Aroclor 1260, Clophen a 60, Clophen a 30 and Aroclor 1254*

Chemical:	Aroclor 1260		Aroclor 1260				Clophen			Aroclor 1254			
Strain:	Sherman		Sprague-Dawley				Wistar			Fischer 344			
Dose:	100 ppm		100 ppm				100 ppm			100 ppm			
Sex:	Female		Male		Female		Male			Male		Female	
	Control	Test	Control	Test	Control	Test	Control	A 60	A 30	Control	Test	Control	Test
No. Examined	187	189	31	40	45	46	120	125	128	24	23	23	24
Hepatocellular Adenoma	0	135	0	4	1	29	6	85	14	0	1	0	1
Hepatocellular Carcinoma	1	21	0	1	0	19	2	67	2	0	2	0	0
Animals with Hepatocellular Adenoma and/or Carcinoma	1	138	0	5	1	41	8	114	16	0	3	0	1
Cholangiocarcinoma	0	0	0	0	0	3	1	0	0	0	0	0	0
Centrilobular Hepatocytomegaly	1	108	0	15	0	5	1	2	2	0	3	0	3
Focus/Foci, Eosinophilic	7	173	1	16	5	36	51	101	98	0	4	0	6
Focus/Foci, Basophilic	4	67	1	0	2	1	2	4	15	0	7	2	4
Focus/Foci, Clear Cell	14	67	4	0	1	0	7	28	39	0	0	0	2
Focus/Foci, Mixed Cell	1	38	0	2	0	0	7	28	49	0	8	0	5
Animals with Focus/Foci of Any Type	25	177	5	16	7	36	55	108	106	0	16	2	15

*Aroclor 1254 given at dosages of 25, 50 and 100 ppm. Only data for 100 ppm groups presented for comparative purposes.

In female Sherman rats that received 100 ppm of Aroclor 1260 for up to 23 months, 73% of the treated rats examined were diagnosed with hepatocellular neoplasms. The overwhelming majority of these neoplasms were benign (62% of the treated rats examined had only hepatocellular adenomas; approximately 11% of the rats had hepatocellular carcinomas or both benign and malignant tumors). Three rats had unusual tumors with glandular, papillary patterns giving the appearance of both hepatocellular epithelium and biliary epithelium simultaneously within a single tumor. Multiple eosinophilic foci occurred in nearly all of the treated rats examined, frequently in rats already diagnosed with hepatocellular neoplasms. The distinction between large eosinophilic foci with enlarged hepatocytes exhibiting compression and hepatocellular adenomas was sometimes very difficult. Other types of foci were also increased in treated rats. Centrilobular hepatocytomegaly was also a very frequent finding occurring in over 50% of the treated rats.

In male and female Sprague-Dawley rats that received Aroclor 1260 at 100 ppm for 16 months, followed by eight months at 50 ppm and up to an additional five months on laboratory chow, 12.5% of the males and approximately 90% of the females had hepatocellular neoplasms. In males, four of the five neoplasms were diagnosed as hepatocellular adenomas. In females, approximately equal numbers of benign and malignant neoplasms were diagnosed with approximately 48% having only

benign neoplasms and approximately 41% of the female rats having malignant or both benign and malignant neoplasms. However, it was noted that a few of the rats in this study were subjected to surgical procedures (partial hepatectomy). The time on study for the rats examined varied as much as 12 months with tissues not examined from rats prior to 18 months, then examined at monthly intervals thereafter up to 29 months. These factors could affect the incidence of various neoplasms.

Hepatocellular tumors appearing to have mixed hepatocellular and biliary elements occurred in four female rats and biliary neoplasms (cholangiocarcinoma) occurred in three treated female rats. These neoplasms usually occurred in livers in which purely hepatocellular neoplasms had already been diagnosed. Eosinophilic foci occurred in slightly less than half of the treated males and in more than three-fourths of the treated females. These lesions were similar to those noted in the female Sherman rats and occasionally the distinction between large eosinophilic foci with enlarged hepatocytes exhibiting compression and hepatocellular adenomas was sometimes very difficult. Centrilobular hepatocytomegaly was a common diagnosis in treated males similar to that noted in male Sherman rats sacrificed at nine months. In females, only a few rats were diagnosed with this lesion, but this may be due to the limited amount of nonneoplastic tissue available for examination.

In male Wistar rats that received 100 ppm of Clophen A60 for up to 832 days, approximately 91% of the rats examined were diagnosed with hepatocellular neoplasms. Approximately 38% of the rats had benign neoplasms only, while 54% of the rats had hepatocellular carcinomas or both benign and malignant neoplasms. This is a reversal of the proportions noted in the study utilizing female Sherman rats in which the majority of the rats had only benign neoplasms. However, it should be noted that the length of the time the rats received the test material was also increased. Tumors appearing to have mixed hepatocellular and biliary elements occurred in nine of the treated rats, frequently in livers in which purely hepatocellular tumors also had been diagnosed. Eosinophilic foci occurred in approximately 80% of the treated rats examined. However, the eosinophilic foci in this study were smaller and blended better with adjacent parenchyma compared to the large compressive foci in the study with female Sherman rats. Centrilobular hepatocytomegaly was not a frequent finding in the treated male Wistar rats but the diagnosis of this lesion may have been hampered by the limited amount of hepatocellular tissue available for examination from each rat. Frequently, only single sections of liver lobes were available and the entire section was neoplastic.

The principle toxic change noted in rats receiving Aroclor 1260 or Clophen A 60 appeared to be proliferative in nature. Beginning with centrilobular hepatocytomegaly, probably due to enzyme induction

resulting in proliferation of smooth endoplasmic reticulum and cell hypertrophy, the affected livers appear to progress to foci of cellular alteration (principally eosinophilic) and ultimately to neoplasia. Hepatocellular hyperplasia was only infrequently diagnosed in the rats treated with Aroclor 1260 due to the overall absence of significant areas of degeneration or necrosis which would have triggered attempted regeneration of hepatic parenchyma. Although bile duct proliferation, oval cell proliferation and/or periportal fibrosis were present as treatment-related lesions in some of the studies, these lesions were not usually very severe, and there did not appear to be any significant loss of hepatic parenchyma in these rats.

In male Wistar rats that received 100 ppm of Clophen A 30 for up to 832 days the incidence of hepatocellular carcinomas was identical in the control and test group. A mild increase in the incidence of hepatocellular adenoma was present in the treated group as compared to the control group (5% in the control and 11% in the treated group). Increased incidences of foci of cellular alteration of hepatocytes was present in male Wistar rats given 100 ppm Clophen A 30. As noted for Wistar rats given Clophen A 60, centrilobular hepatocytomegaly was not a frequent finding but this observation may have been affected by the tissue sampling limiting the amount of hepatic parenchyma available for microscopic examination.

EPL

EXPERIMENTAL PATHOLOGY LABORATORIES, INC.

Administration of Aroclor 1254 to male and female Fischer 344 rats at dosages of 25, 50 and 100 ppm for 104-105 weeks resulted in an increase in the incidence of foci of cellular alteration in the liver. A few hepatocellular neoplasms were diagnosed in treated rats, however; the total number of affected rats was small and within the expected range for rats of this age and strain. Centrilobular hepatocytomegaly and pigment deposition, often occurring with multifocal granulomatous hepatitis, also occurred more frequently in Aroclor 1254 treated rats.

Jerry F. Hardisty D.V.M.

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PWG Chairman

June 27, 1991
Date

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APPENDICES AND REFERENCES OMITTED;

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ADVANCE NOTICE OF PROPOSED RULEMAKING

OF THE

UNITED STATES ENVIRONMENTAL PROTECTION AGENCY

CONCERNING

DISPOSAL OF POLYCHLORINATED BIPHENYLS

(OPTS-66009; FRL 3845-4)

56 FEDERAL REGISTER 26738-26744 (JUNE 10, 1991)

REGULATORY DOCKET REFERENCE NO. OPTS-66009

AUGUST 9, 1990

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EXECUTIVE SUMMARY

GE has participated with EPA for over a decade in developing rules and practical guidance for the proper use and disposal of PCBs. GE has also performed and supported much research in order to understand better the potential risks represented by PCBs to human health and the environment. We fully support the Agency's effort in this proposed rulemaking regarding various issues such as large volume items, mixed waste and other issues.

However, we believe that there are more fundamental issues that EPA should address as part of the rulemaking and that these issues should take precedence over the practical issues that EPA plans to address. The crux of the issue is that EPA continues today to regulate all PCBs as if they represented equivalent risk to Aroclor 1260 (60% chlorinated mixture) in the face of scientific information that only the 60% chlorinated mixtures are carcinogenic in rats. Lesser chlorinated mixtures, which made up about 90% of PCBs sold in this country, are shown to be non-carcinogenic in rats and are less persistent in the environment.

We are submitting a new analysis of 5 earlier long-term cancer bio-assay studies of rats fed various PCB mixtures conducted by the Institute for Evaluating Health Risks (IEHR). These studies show that 60% chlorinated PCB mixtures are less potent than originally determined and that 54% and 42% chlorinated mixtures are non-carcinogenic.

Another concern regarding PCBs has been the persistence of PCBs released into the environment and the uptake of environmental PCBs by

people. We are providing information that PCB levels are decreasing in many environmental compartments, including the Hudson River and the Great Lakes. Furthermore, substantive studies of population groups involved in activities that were expected to result in significant exposures to PCBs did not show PCB blood levels elevated over that of national background levels. These studies included two groups presumed to consume PCB contaminated fish. This lack of elevation of PCB levels is believed to be due to the reduction in environmental contamination levels, as well as the fact that exposure to contaminated soils, air and water bodies results in far less real exposure to PCBs than generally believed by risk assessors.

Studies of workers, both clinical and retrospective mortality, having average levels of exposure over an order of magnitude greater than background exposure levels has not demonstrated adverse clinical effects or significantly increased causes of death, including cancer.

The suggestion that environmental PCB exposure is associated with minor decreases in neurological development of children has not been validated. It is not clear that these decreases are real or significant, and if they are, that they are associated with PCBs, rather than other environmental contaminants, such as heavy metals, or lifestyle factors, such as smoking and alcohol consumption.

We are recommending that EPA immediately begin a re-evaluation of all the scientific information in order to arrive at a new basis for regulating PCBs. On an interim basis, the new information available from the IEHR

study should be used to establish risk for PCB mixtures. This approach can be taken without raising risks to human health or the environment.

. Recognition of the reduced health risks posed by PCB exposure supports a reconsideration of the present regulatory framework for PCB management and disposal. Under the current scheme, incineration and TSCA landfilling are the nearly exclusive options for PCB management, without regard to human health or environmental risk posed by the materials. Alternative disposal technologies are rarely considered, even when they could provide equal (or nearly equal) protection in a more cost-effective manner. Furthermore, the application of stringent regulatory criteria to all PCB materials, consumes limited incineration and landfill capacity better preserved for more hazardous materials.

The greatest consumption of landfill and incineration capacity results from the stringent application of the Agency's anti-dilution rule. The rule is now applied to historic contamination which, while never actively diluted, is now of sufficiently low concentrations that alternative management scenarios would be appropriate. Application of the anti-dilution role, however, often requires that these materials be excavated and incinerated at enormous and unnecessary cost. The Agency should consider revisiting this approach, and at the same time continue to investigate the utility of alternative disposal technologies.

I. INTRODUCTION

General Electric apppreciates the opportunity to respond to EPA's June 10, 1991 Advance Notice of Proposed Rulemaking ("ANPR") regarding revision of the Agency's PCB regulations. See 56 Fed. Reg. 26738. EPA has stated that it "inten[ds] to reconsider portions of its PCB regulations based upon information and experience acquired over the past 12 years . . . [and] to modify the current PCB regulations to allow for maximum flexibility in controlling PCBs and PCB Items based on their risk to human health and the environment . . . . Id.

General Electric strongly supports U.S. EPA's initiative and has prepared the following comments to provide EPA with scientific and technical information GE believes EPA should consider as part of this rulemaking process. Contained in Section II is a summary of recent scientific developments regarding PCBs which provides strong support for the conclusion that PCB regulation should be re-evaluated to comport with new scientific information. Section III discusses the approach GE believes EPA should take in performing such a reevaluation and addresses the problems inherent in the current regulatory structure as it relates to PCB cleanup and disposal issues.

Constrained by the August 9, 1991 deadline for submittal of comments, GE has not been able to include all the relevant scientific, technical and cost information it has begun to compile in response to the ANPR. It is GE's understanding, however, that EPA has agreed to accept comments during the sixty-day period following the formal August 9 deadline. Thus, GE reserves the right to supplement this submittal no later than October 8, 1991 with additional comments concerning both issues raised in this submittal and other matters addressed in the ANPR.

II. RECENT SCIENTIFIC DEVELOPMENTS CONCERNING THE TOXICITY AND PERSISTENCE OF PCBS

A. Introduction

The re-evaluation of EPA's approach to the regulation of PCBs is a natural aspect of the modern, evolutionary process of science-based regulation. That is, as new scientific information becomes established, regulation must respond to and incorporate the new understanding so that the regulation properly reflects scientific reality.

In particular, where a class of chemicals has previously been assumed to possess similar characteristics and is thus regulated without regard to individual differences within the class, the emergence of new information that casts doubt on the initial assumption requires the regulatory agency to re-assess its regulatory approach accordingly. Indeed, EPA has adopted this approach on numerous occasions. For example, EPA's rule on incidental generation of PCBs in manufacturing operations recognizes the difference

between very lightly chlorinated PCBs and other PCBs by applying discounting factors of 50 and 5, respectively, for mono- and di-chlorobiphenyls.[1] Thus, for purposes of determining if a chemical mixture containing incidentally generated PCBs reaches the regulated level of 50 ppm, the concentration of monochlorinated biphenyl is divided by 50, and the concentration of dichlorinated biphenyl by 5.

Similarly, regulations and health standards for many other chemicals recognize that individual members of chemical families are not equally hazardous. For example, in the PCDD (dioxin) and PCDF (furan) families, which EPA has determined are more toxic than PCBs, only about ten percent of the individual PCDD and PCDF congeners are considered toxic enough to be measured for risk assessment purposes. This ten percent consists of congeners that have chlorines in the 2,3,7,8-positions, with additional chlorines in other positions. Based on toxicity tests, "toxic equivalency factors" have been calculated relative to the most toxic congener, 2,3,7,8-TCDD. These equivalency factors are used to calculate the sum of "TCDD equivalents" for any mixture of dioxins and furans. Other examples of differential regulation within a chemical family include FDA and EPA standards that measure and regulate methyl mercury instead of total mercury and chromium +6 (hexavalent chromium) instead of total chromium.

GE therefore urges EPA to consider the new scientific information seriously and carefully, so that EPA's regulation of PCBs will be based on "sound science"[2] rather than outdated speculation.

B. Discussion

The regulation of PCBs in the 1970s was based on the information that was available and the risks that were perceived at the time. The primary scientific bases for regulation at that time included:

> The 1968 "Yusho" human poisoning incident in Japan, which produced chloracne and other symptoms.
>
> The 1975 finding, by Dr. Renate Kimbrough of the Centers for Disease Control, that high dosage feeding with Aroclor 1260, a mixture containing highly chlorinated PCBs, caused liver cancer in rats.[3]
>
> The belief that PCBs are generally persistent in the environment and resistant to biodegradation, which is nature's usual way of dealing with environmental organic chemical contaminants.

Since EPA's initial regulation of PCBs in 1979, substantial "information and experience acquired over the past 12 years in dealing with PCBs"[4] has been developed. In particular, recent scientific studies have shown that:

- The adverse health effects observed in the "Yusho" and "Yucheng" incidents are not attributable to PCBs.

- Although Kimbrough's finding regarding Aroclor 1260 has been supported by subsequent studies, subsequent feeding studies have shown that lower chlorinated mixtures do not cause liver cancer. Moreover, the potential of Aroclor 1260 to cause liver cancer has been shown to be less than originally determined. Finally, other data from these studies suggest that these results are not relevant to human carcinogenicity.

- The threat of PCBs in the environment is less than originally feared because PCBs have been found to biodegrade, and levels are decreasing in many environmental media.

In addition, new scientific information pertaining to the human health effects of PCBs has been discovered. These findings indicate that:

- Exposure to PCBs in the environment today does not result in elevated PCB blood levels.

- Clinical studies of workers exposed to PCBs show no association between adverse health effects and high levels of exposure.

- Epidemiology (mortality) studies of PCB-exposed workers do not indicate that PCB exposure leads to elevated causes of death, whether based on overall cancer mortality or deaths due to individual cancer types.

- The perceived relationship between PCB exposure and chloracne is most likely spurious. Any observed linkage likely arises from contamination of PCB and from uses of PCBs in conjunction with active agents.

- Any suggestion that reproductive or neurodevelopmental effects in humans is related to low-level exposure to PCBs has not been validated.

These findings strongly suggest that human health risks from PCB exposure have been significantly overestimated in current regulations and that EPA should undertake a thorough re-evaluation of the actual risks posed by PCB exposures.

1. Recent Scientific Information Indicates That Prior Speculation Regarding PCB Risks Was Erroneous

a. The "Yusho" Incident Was Not Attributable To PCBs

The "Yusho" incident, in which about 1500 persons in Japan in 1968 became ill after consuming rice oil that was accidentally contaminated with a PCB mixture known as "Kanechlor 400," became known as the "PCB Poisoning Incident" in Japan. A similar incident, known as "Yucheng," occurred in Taiwan in 1979. Typical symptoms were chloracne, swelling of eyelids and eye discharges, brown pigmentation of the nails and skin, and curling of fingernails and toenails. Signs of the disease were also observed in some offspring of affected mothers. Although the major symptoms disappeared over the next sixteen to twenty years, subsequent studies suggested a possible increase of cancer and adverse developmental and behavioral effects in offspring.

The cause of the incident was extensively studied and the rice oil was found to contain high levels of polychlorinated dibenzofurans ("PCDFs"), a chemical that is 100 to 1,000 times more toxic than PCBs. After finding that workers exposed to much higher levels of PCBs showed minimal adverse health effects, and after performing dose-response studies on the rice oil mixture, Japanese and Taiwanese scientists concluded that PCDFs were the prime causal factor in the Yusho and Yucheng incidents.[5]

In 1985, Dr. Kimbrough and Dr. Goyer of the National Institutes of Health unequivocally concluded that:

> The scientific community assumes now that most of the effects observed in these two outbreaks were caused by the ingestion of the polychlorinated dibenzofurans.[6]

Likewise, the Halogenated Organics Subcommittee of EPA's Science Advisory Board reviewed a PCB health advisory from EPA and concluded that:

> The health effects section suggests that the short-term human exposure to Yusho poisoning is representative of polychlorinated biphenyl toxicosis. Recent studies indicate that the major etiologic agents in Yusho were polychlorinated dibenzofurans rather than polychlorinated biphenyls. . . . Thus, a discussion of the human health effects of polychlorinated biphenyls should not use 'Yusho' as an example. Industrial exposure data more accurately reflect human health effects.[7]

Significantly, this scientific re-interpretation of the Yusho and Yucheng incidents is consistent with data from animal studies that show a relatively

low level of acute toxicity – e.g., LD-50s ranging from about 1 to 11 mg/kg-body-weight in rats, depending on the Aroclor mixture.[8] Moreover, this explanation is consistent with the numerous studies (discussed below) that show no significant adverse health effects in workers who had been exposed to average levels of PCBs higher than the Yusho patients were.

b. Re-evaluation of Prior Animal Studies Has Resulted In A Reassessment Of Their Conclusions

After the 1975 Kimbrough study reported that high doses of Aroclor 1260 caused liver cancer in rats, additional animal studies have been carried out with Aroclor 1260[9] and Clophen A60 (both of which are 60-percent-chlorinated mixtures)[10], Aroclor 1254 (54-percent-chlorinated),[11] and Clophen A30 (42-percent-chlorinated)[10]. Although the studies performed with Aroclor 1260 and Clophen A-60 have supported Kimbrough's original finding, the studies with Aroclor 1254 and Clophen A30 suggest that only the 60-percent-chlorinated mixtures are carcinogenic in rats.

Recently, the liver tissue slides from each of the five original studies were screened by a panel of expert pathologists using current guidelines for interpreting liver lesions, as developed by the National Toxicology Program and endorsed by EPA. The panel's proceedings were observed by representatives from EPA, FDA, the original studies, Experimental-Pathology Laboratories, Inc., and the Institutes for Evaluating Health Risks ("IEHR")[12].

Although this review confirmed that 60-percent- chlorinated mixtures are carcinogenic to rats, the expert panel found that the number of animals with benign or malignant liver tumors was less than originally reported. The panel also confirmed that the study of Aroclor 1254 performed by the National Cancer Institute and the study of Clophen A30 yielded negative results. These findings demonstrate that different PCB mixtures have significantly different carcinogenic effects, at least in rats, and suggests that appropriate regulation of PCBs requires distinguishing between 60-percent-chlorinated PCBs and those that are less than 60-percent-chlorinated.

Thus, according to Dr. John Moore, president of IEHR, the panel's clarification of the results of the original studies presents EPA with the opportunity of modernizing PCB cancer risk assessments by (1) developing separate risk assessments for each of the major PCB formulations, and (2) utilizing all the available data when calculating cancer potency for PCB mixtures below 60 percent chlorination. It is, of course, significant that almost all (88 percent) of the PCBs sold in this country by the sole domestic supplier from 1957 to the end of production were mixtures that were below the 60 percent chlorination level.[12] (See Exhibit 1)

In addition, the panel's review of the animal studies revealed further information that should be taken into consideration in estimating the risks of PCBs:

- The PCB-exposed rats, including those with liver tumors, lived significantly longer than the controls (unexposed rats).[10]

- The PCB-exposed rats had significantly fewer cancers of all types, i.e., sum of all cancers, than did the controls (unexposed rats).[10]

- The liver tumors, although formally classified as cancers, did not metastasize to other organs or invade blood vessels.[9]

In other words, PCB exposure in rats appears to produce benign (non-invasive, non-life-threatening) rat liver cancers and indeed may well produce beneficial effects (significant life extension and reduction in number of other cancers relative to the controls). These conclusions seriously call into question the relevance of the rat liver tumors to human risk. Rather, they strongly suggest that, at a minimum, the declassification of PCB mixtures having less than 60-percent chlorination as animal carcinogens can be made without endangering human health.

2. New Information On Human Health Effects Necessitates New Determination Of PCB Risk Assessment

a. Exposures To PCBs In Today's Environment Do Not Result in Elevated Blood Levels

The Centers for Disease Control has conducted a study of PCB blood levels of persons thought to have the highest risk of exposure to PCBs at 12 waste sites where PCBs had been disposed.[13] Site contamination levels ranged from 3,436 to 330,000 ppm on-site to 3 to 133,000 ppm off-site. Persons who were chosen for inclusion in the study reported participating in activities involving one or more contaminated environmental pathways (e.g., swimming in contaminated waters, eating contaminated fish, direct contact with contaminated soils, etc.).

The blood-level screening study resulted in a finding that:

> [I]n 10 of the 12 site-specific investigations conducted under this protocol, no excess proportion of potentially exposed persons was found to have serum PCB levels greater than 20 ppb attributable to nonoccupational exposures from the sites in spite of high PCB levels in soil or leachate on the sites. As a result, we concluded that there was no need for further studies.

Moreover, in the two settings where elevated blood levels were found, one was thought to be attributable to "historical prevalence of occupational exposures," and the other (in New Bedford, Massachusetts) was believed to be a result of substantial consumption of PCB-contaminated fish. As a result of the latter finding, the Centers for Disease Control embarked on a broad

population survey of PCB blood levels in New Bedford.[14] That study found that levels in 1985-86 were within national population background levels.

In addition, the Agency for Toxic Substances and Disease Registry surveyed a population of residents of Paoli, Pennsylvania, who lived in a neighborhood where PCB levels in soil ranged from 1 to 6,400 ppm.[15] The study found that "[T]he geometric mean and distribution of serum PCB concentrations in this group did not differ from the means and distribution of a large sample of persons from across the United States having no known environmental exposure." Accordingly, the report concluded that "the population near the site in Paoli did not show exposure different from other U.S. populations having no known unusual source of exposure." (See Exhibit 2)

These findings -- that exposure to PCBs in the environment do not result in elevated blood levels -- coupled with the evidence (discussed below) that environmental PCB levels are declining, strongly indicate that the public health risks of PCB sites are much lower than originally assumed.

b. Clinical Studies of PCB-Exposed Workers Do Not Indicate That Adverse Health Effects Are Associated With High PCB Exposures

The most extensive long-term exposure of humans to PCBs has occurred in capacitor plants; 17 capacitor plants used PCBs in the United States. Many employees in these plants had daily PCB skin contact for years and inhaled PCBs (primarily Aroclor 1242 or 1016) at levels in the 100 to 1000 µg/m range.[16]

In capacitor plants, the most frequent PCB-related health effect observed was transient skin rashes that affected a small percentage of exposed employees. In contrast to chloracne, this condition responded to simple topical treatment and employee reassignment to other work areas. The medical records of these worker populations have shown no obvious incidence of systemic disease attributable to PCBs.

Medical surveillance by GE of the group of 174 heavily exposed capacitor workers has consisted of multiple examinations over the last fifteen years. [17] The length-of- service of this group averages over 20 years and ranges from 1 to 40 years. PCB blood tests of 174 heavily exposed GE capacitor workers have indicated a mean serum level of about 500 ppb. Ten percent of the individuals' analyses were above 1000 ppb. By comparison, mean serum PCB background levels for people exposed to environmental background levels in the United States range from 2 to 24 ppb, with a mean of approximately 6 ppb. The only clinical parameter found to be statistically correlated with serum PCB level is that of serum triglycerides. The interpretation of this correlation is confounded by the fact that PCBs distribute equally among all lipid pools in the body, including those in the blood; hence, for any given PCB body burden the serum PCB level must vary directly with

the level of serum lipids. Thus, for any given PCB body burden, increased serum PCB levels are the result, not the cause, of increased serum lipids.[18]

Medical examinations of these workers have not revealed serious health problems related to PCB exposure. The pulmonary function tests of the non-smokers in this heavily exposed population were in the normal range.

A number of other worker clinical studies have been carried out in this country and abroad.[19] Observations of high PCB blood levels, some dermal conditions, and isolated cases of chloracne have been reported in the industrial hygiene literature of Japan, Finland, Australia and Italy. Although the reported biochemical examinations identify scattered individual abnormalities in serum enzymes, the various liver function tests were generally considered normal. The Italian chloracne cases were observed in men who had worked where the air levels of Aroclor 1254 were 5200 to 6800 ppb (i.e., 10 to 14 times the U.S. permissible exposure level). The Finnish capacitor workers were also found to be in good health, despite PCB concentrations in their blood approximately 50 times greater than that of a control group. The researchers were unable to detect any biological effect caused by PCB in these workers.

The findings from these studies were summarized by A.B. Smith, M.D., of NIOSH, as follows:

> None of the published occupational or epidemiological studies (including ours) have shown that occupational exposures to PCBs is associated with any adverse health outcome . . . except for the occurrence of chloracne.[20]

Notably, chloracne was not observed by the NIOSH researchers. The relationship of PCB exposure to occurrence of chloracne is discussed below.

c. Epidemiological Studies Do Not Link PCB Exposure to Excess Mortality

Seven cohort mortality studies involving PCB-exposed workers have been performed.[21] Most of these studies involve capacitor manufacturing workers who were exposed to lightly chlorinated PCBs, such as Aroclors 1242 or 1016 (predominant exposure) and Aroclor 1254 (minor exposure). One case-control study involved transformer manufacturing workers who were exposed to Aroclors 1260 (major exposure) and 1254 (minor exposure).[22] (See Exhibit 3) In none of the seven studies did the authors conclude that PCB exposure was associated with cancer or any other cause of death.

With a few exceptions, the seven studies suffered from the fact that the absolute number of deaths was small. As a result, the ratios of the observed to the expected numbers of deaths due to individual cancer types ("odds ratios") were usually based on small numbers, and results were therefore generally

not statistically significant. This effect can be overcome, however, by combining various cohort studies into a single cohort and calculating combined odds ratios. This approach, called "meta-analysis," tends to average out findings that are not consistent from study to study and to highlight those that are.

Such a meta-analytic approach was used by Nicholson in 1987 in which he combined data from five such studies into a single cohort.[23] Although Nicholson's study may not be a valid basis for reaching conclusions about causality because of substantial dissimilarities among the studies and because of methodological flaws in performing the meta-analysis, it remains useful as an hypothesis-generating tool.

Nicholson's analysis indicates a significant increase in deaths resulting from a grouping of liver, biliary and gall bladder cancers.[24] (See Exhibit 4) However, when the data from the Taylor (Harvard: See Exhibit 5) and Sinks (NIOSH) studies were added to the Nicholson combined cohort, with appropriate adjustments because of overlap of Taylor's cohort with two of the previous cohorts in the Nicholson study, the results showed a statistically insignificant relationship.[25]

Other considerations further reduce the likelihood of a PCB-related increase. For example, five of the persons had worked a year or less in PCB exposed jobs; five of the eleven cancers were not confirmed pathologically; and only one of the cancers was confirmed as intrahepatic (the animal studies indicate that only hepatocellular carcinomas occur). Furthermore, the epidemiology of liver, gall bladder, and biliary tract cancer indicates that the etiologies of these cancer types are quite different and suggest that they should be studied separately when attempting to determine causality. Although this evidence does not support a finding that PCB exposure causes liver, biliary and gall bladder in humans, it is prudent to continue to observe results in this area for further clarification.

GE's analysis of the epidemiological data is consistent with that of Chase, Doull, Friess, Rodricks and Safe, who concluded:

> There is insufficient evidence to show a causal relationship between PCB exposure and the subsequent development of any form of cancer. In light of the long-term and widespread usage of PCBs in the workplace and, in some cases, the extensive exposures of workers, it is likely that evidence of carcinogenicity in humans would have been observed in the various epidemiological studies discussed above if PCBs were in fact potent carcinogens.[26] (See Exhibit 6)

d. Any Linkage Between Chloracne And PCB Exposure Is Probably Spurious

Because PCBs are sometimes contaminants in, or are contaminated by, other halogenated aromatic compounds, the interpretation of both animal toxicity and human health effects studies has been difficult. The first commercial use of PCBs was as a low-level additive in chlorinated napthalenes, which are known to be chloracnegenic and to cause liver toxicity. These mixtures were used as solid electrical insulating compounds called "Halowax" or "Chlorowax." Exposure to these mixtures during their manufacture and use resulted in reports of chloracne and liver disease.

Following one such occurrence, Bennett, Drinker, and Warren (1938) conducted studies of rats given doses of individual components of the Halowax compound and reported that "chlorinated diphenyl gave evidence of being the most toxic."[27] A year later, Drinker reported that this compound had been erroneously labeled as chlorinated diphenyl. An authentic sample of 68-percent-chlorinated biphenyl proved to be "almost non-toxic."[28] As noted by NIOSH in 1977, "[t]hese animal experiments reported by Drinker and by Bennett have continued to be erroneously cited."[29]

The first incident involving chloracne and PCB exposure in the absence of chlorinated napthalenes was reported in 1936.[30] After performing skin patch tests with suspect chemicals, including PCBs, on PCB-exposed workers, the authors of this report concluded that the cause was an impurity in the benzene used to make the biphenyl and that "the chlorinated diphenyl can absolutely be absolved as the irritating agent."

The second episode involving PCBs and chloracne occurred in 1950 and 1951, when 14 people were exposed to PCB vapors (reported at 100 $\mu g/m^3$) from a leaky heat exchanger, and seven of the 14 developed chloracne.[31] A third episode was noted in the early 1960s when 13 of 16 people exposed to vapors from an oven in which PCB-plasticized enamels were being baked were similarly affected.[32] Other occurrences of chloracne have involved PCB usage abroad, where data on conditions of use or contaminant concentrations do not permit one to draw reliable conclusions about the cause of the health effect.

In light of the circumstances surrounding these PCB exposures, i.e., impurities in the materials and the heating of PCBs under oxidative conditions, it seems reasonable to attribute the chloracne to contamination by polychlorinated dibenzofurans ("PCDFs"). As demonstrated by the Yusho/Yucheng incidents, and confirmed in the laboratory, PCDFs can be formed by partial oxidation of PCBs at elevated temperature. PCDFs also occur in varying concentrations in commercial PCB mixtures, with higher concentrations in Japanese and European products than in aroclors. As pointed out by NIOSH (1977), "[c]hloracne has frequently been associated with processes where the PCBs were heated."[28]

Perhaps most revealing, however, is the fact that in the three largest and most recent studies of capacitor manufacturing and transformer repair

workers, not one case of chloracne was identified.[17, 20, 32] This result is particularly significant because the mean PCB serum levels in one of the studies were two orders of magnitude greater than national population mean levels and because one of the researchers, Dr. E. Emmett of Johns Hopkins University, was a dermatologist and made a special search for signs of chloracne.[33]

In short, much like the initial hypotheses that surrounded the Yusho incident, subsequent study has shown that any relationship between PCB exposure and chloracne is likely spurious. No reliable study has shown that, absent confounding factors, PCB exposure causes chloracne.

e. Any Relationship Between Reproductive Or Neuro-developmental Effects In Humans And Low-level PCB Exposure Has Not Been Validated

The suggestion that neurodevelopmental effects in humans is associated with low-level exposure to PCBs in the environment is based on two sets of studies, i.e., Fein, Jacobsen, et al.,[34] and Gladen, et al.[35] While it is generally acknowledged that these studies are not sufficient to establish a causal relationship of PCB exposure to the reported deficits, it is important to recognize that there are shortcomings in the studies that may make it impossible to reach conclusions with these cohorts.

Fein, et al. reported that newborns born to mothers who ate fish containing PCBs had significantly decreased birth weight, head circumference, and gestational age. However, other statistically significant differences between the control and exposed populations were observed including the fact that the incidence of mothers ingesting alcohol during pregnancy was three-fold higher in the exposed populations. Alcohol is well known to produce a number of birth defects. As the changes in birth weight and head circumference described by Fein, et al. in the newborns of "PCB-exposed mothers" were small, the increased alcohol consumption by these women could easily explain the observed changes. In addition, the use of cough and cold preparations and caffeine was also higher in the group listed as the PCB-exposed population. Fein, et al. failed to account for the possible effects of other persistent environmental contaminants (such as dioxins and furans) which may be present in Great Lakes fish. Therefore, because significant differences between the exposed and control populations in addition to alcohol were not controlled for in the Fein, et al. study, the effects described in this paper cannot be attributed to PCBs.

The Gladen, et al. study involves 802 infants who were assessed for exposure to PCBs and DDE transplacentally and from mothers' milk. These subjects were followed up frequently with numerous observations of social, medical and developmental status, in particular, observations were made at about six and twelve months of age with regard to a Mental Development Index (MDI) and a Psychomotor Development Index (PDI). Of sixteen associations that were examined, three achieved statistical significance. These

associations indicated that transplacental PCB exposure was inversely related to PDI (more so at twelve than six months of age) and that DDE exposure was positively related to MDI only at six months of age. The authors make no explicit interpretive statement of the relationship between transplacental PCB exposure and PDI. Rather, they state, "Clinically, the relatively small differences we observed . . . are undetectable, [and] . . .can easily be seen when a single child is retested." They acknowledge that the association found may be ". . . related to some factor that we did not measure, or to residual uncontrolled confounding." There is a strong likelihood that the association reported between PCBs and PDI is attributable to chance, bias or to residual confounding.

The dose-response relationship is based almost entirely on cases at the lowest and highest levels of exposure. Intermediate exposure levels show no suggestion of dose response, but instead, a suggestion that higher PCBs are associated with higher PDI. Other factors, not completely accounted for, such as education and income, could also strongly confound the results.

In summary, GE believes that these findings are of interest and that further studies should be planned. However, prior to carrying out additional studies, a standard protocol should be developed in order to maximize the opportunity to obtain reliable information. Among other things, this protocol should contain detailed procedures to identify, quantitate and evaluate, statistically, the role of all environmental contaminants, including heavy metals as well as organics, in affecting neurological development in children.

3. Current Science Does Not Support The Application of Dioxin TEFs To PCBs

In December 1990 the U.S. EPA sponsored a workshop in Washington, DC, to discuss the concept of Toxicity Equivalency Factors for PCBs.

The scheme, as proposed by Dr. Stephen Safe, of Texas A&M University, and discussed at the workshop, would assign TEFs for three so-called 'coplanar' PCBs, for 8 congeners with a single chlorine in an ortho position, and for certain di-ortho congeners. These proposed TEFs are based primarily on in vitro studies done by Dr. Safe.[36]

The attempt to develop a common basis for risk assessment and regulations of several families of chlorinated hydrocarbons is laudable since it would in theory be simple to implement. However, there are certain criteria that must be met before such an approach is feasible. At a minimum, these are:

- The expression of toxicity for endpoints of concern for each of the families of chemicals must proceed through a common mechanism - in this case proceeding through binding with the Ah receptor.

- Since the TEF approach is based on adding TCDD toxic equivalents to determine risk, the sum of TEQs should, with reasonable accuracy, predict the toxicity of mixtures..

- TEFs should be based on data that most closely represent toxic endpoints of concern, i.e., those endpoints that are used in risk assessment.

- Analytical capabilities should be readily available to measure congeners having TEFs.

Our analysis of the situation indicates that the proposed TEFs for PCB congeners do not meet any of these criteria. Comments follow:

The toxicity endpoint of greatest concern for PCBs, i.e., the endpoint on which risk assessment is based is carcinogenicity. More specifically, EPA uses data from a rat feeding study with Aroclor 1260 of Norback and Weltman to calculate the carcinogenic potency factor (CPF) that is used in risk assessments for all PCBs. We have used specific congener analytical data and the TEFs proposed by Safe to calculate toxic equivalents for Aroclor 1260, 1254 and Clophen A30 and CPFs calculated using the rat slide reread data of Moore.

We determine from these data that cancer potency of PCB mixtures bears no relationship to either total TEQs, coplanar congener TEQs, or mono ortho coplanar TEQs. (The trends for CPFs and diortho coplanar TEQs are in the proper direction, but we are not suggesting a causal relationship).

We also considered an alternative explanation, i.e., that PCB congener TEQs do predict carcinogenicity, but that levels in the feeding experiments were too low to elicit a response. This requires that the positive Aroclor 1260 result was caused by an alternative mechanism. Therefore, we calculated daily TEQ feedings for the three PCB mixture bioassays and compared them to the Kociba feeding of TCDD. We determined that the feeding of TCDD equivalents in each of the PCB feeding experiments exceeded the Kociba feeding of TCDD by factors of 2-6.

Thus, each experiment should have resulted in a strong positive response if PCB TEQs were "dioxin like" in the carcinogenic bioassay. In fact, only the experiment with 60% Cl mixtures, with the lowest daily TEQ feeding was positive.

In addition, it was acknowledged by Dr. Safe at the December workshop that the sum of PCB TEQs overestimates the AHH induction potency measured for Aroclor 1254.

There are a number of possible explanations for the failure of PCB mixture TEQs to predict mixture toxicity results. All may be correct, as discussed below:

- TEFs as proposed by Safe are wrong. The range of relative potency data in which the TEFs are selected is quite large, up to three orders of magnitude, and Safe has generally selected TEFs at the high end of the potency range; for example, he describes the value for mono-ortho congeners as "highly conservative".

- The analysis of "DL" congeners is inaccurate. This is almost certainly the case but is not likely to change the conclusions discussed above. The workshop concluded that much work needs to be done on analysis.

- TEFs based on in vitro and sub-chronic in vivo tests do not predict carcinogenicity in rats. This is clearly the case for PCBs, and may be the case for dioxins and furans.

- Other PCB congeners interact with the Ah receptor and diminish the activity of DL congeners. Numerous studies have demonstrated this type of effect, while others have shown the opposite. These results argue against the additivity of TEQs.

At the December workshop, it was suggested that PCB TEQs be added to TEQs for PCDDs and PCDFs to estimate the total "dioxin-like" risk for environmental mixtures. Since PCB TEQs do not predict either carcinogenic potency or AHH induction in PCB mixtures, it is clear that current science does not support this approach.

4. Widespread Natural Biodegradation Is Occurring

New evidence shows that PCBs, which were once thought to be virtually indestructible, have been naturally biodegrading throughout the world. PCB measurements have shown significant decreases, for example, in a variety of environmental media in Connecticut,[37] Pennsylvania,[38] Michigan,[39] New York,[40] Wisconsin,[41] Massachusetts.[42] Canada,[43] and Yugoslavia.[44]

In particular, these decreases may be attributed to two different, yet complementary, microbial remediation processes: aerobic PCB degradation and anaerobic PCB dechlorination. Aerobic microorganisms are capable of oxidatively attacking lightly-chlorinated PCBs, resulting in their ultimate conversion to carbon dioxide and water. Anaerobic microorganisms, on the other hand, reductively attack PCBs, resulting in a stepwise dechlorination to less chlorinated PCBs. These different natural activities are complementary, with the anaerobic process first dechlorinating even highly chlorinated PCB congeners into products that are readily degraded by aerobic organisms.[45] (See Exhibit 7)

a. Aerobic PCB Biodegradation

Organisms capable of aerobically degrading PCBs are well known[46] and are known to be widespread in the environment.[47] In general, these organisms are able to degrade only lightly chlorinated PCB congeners (i.e., PCBs containing less than four chlorines). However, organisms capable of degrading most of the congeners in Aroclor 1242 and even Aroclor 1254 are also known.[48] (See Exhibit 8) These organisms are effective in the destruction of PCBs by reducing the toxicity, volume, and mobility of the contaminated material. For example, over 90 percent removal of Aroclor 1242 can be accomplished by naturally occurring bacteria within 24 hours.[49] In addition, the mobility of the residual contamination is generally reduced, because the more available PCBs in environmental samples are preferentially degraded.[45]

b. Anaerobic Dechlorination

Natural anaerobic PCB dechlorination, which results in the preferential loss of meta and para chlorines from even highly chlorinated PCBs, occurs in many different aquatic systems.[47] For instance, in the upper Hudson River, the most studied system to date, natural anaerobic dechlorination is widespread and nearly ubiquitous.[47] Indeed, anaerobic microorganisms have been shown to be capable of degrading even Aroclor 1260,[48] a highly chlorinated mixture of PCBs that cannot be adequately degraded by aerobic microorganisms.

Anaerobic PCB dechlorination is particularly effective in reducing the toxicity of PCB-contaminated materials, because it removes the meta and para chlorines, thereby significantly reducing the theoretical health risks associated with the PCB residues. Specifically, the detoxification afforded by meta and para dechlorination results in an 80 percent reduction in the "dioxin-like" PCB congeners.[45]

In addition, recent studies have suggested that anaerobic dechlorination may remove ortho-chlorines as well.[50] Anaerobic microbial dechlorination alone can therefore reduce the volume of contaminated material by decreasing PCB concentrations via ortho, meta, and para removal.

C. Recommendations For Re-evaluation Of PCB Regulation

As noted in the Toxicological Profile for Selected PCBs prepared by the Agency for Toxic Substances and Disease Registry ("ATSDR") (ATSDR/TP-88/21; June 1989), EPA's position that PCBs, as an undifferentiated group, are probable human carcinogens is based primarily on studies conducted with Aroclor 1260, and specifically on the results of a single animal study by Norback and Weltman.[9] The risk assessment based on this study superseded an earlier EPA assessment based on a 1975 study by Kimbrough, et al.[3]

As detailed above, however, recent studies have changed the conclusions of these two studies, and indicates that PCB mixtures having less than 60% chlorination are not carcinogenic. EPA should now re-evaluate its approach toward the regulation of PCBs. Two approaches can be considered: 1) the toxic equivalency factor ("TEF") approach; and 2) the "closest Aroclor" approach. As will be discussed, the TEF approach has significant shortcomings that render it indefensible at this time. Therefore, the approach advocated by GE is the "closest Aroclor" approach.

1. The Toxic Equivalency Factor Approach

One possible approach toward recognizing the different health risk posed by different PCB congeners is the toxic-equivalency-factor ("TEF") approach. As discussed above, such an approach would require EPA to assign a TEF to each PCB congener that exhibits "dioxin-like" effects and to determine total TCDD equivalents for any mixture of PCBs.

GE believes, however, that such an approach does not appear appropriate for the regulation of PCBs at this time. The cancer potencies of PCB mixtures do not correlate with the sum of the TEFs derived from the toxicological data. Although it might be possible to design a more rigorous set of experiments to estimate more accurately the TEFs for the various Aroclors, the current data do not permit reasonable reliance on the TEF approach for the regulation of PCBs. To the extent EPA is interested in pursuing such research, GE would, of course, be happy to cooperate with EPA in conducting the research necessary to support this regulatory approach.

2. The "Closest Aroclor" Approach

Another approach, and one that GE believes EPA should adopt, is to regulate PCBs according to their degree of chlorination, rather than as a group. EPA's risk assessment policy currently treats all PCBs as having the same cancer potency as Aroclor 1260, even if no Aroclor 1260 was present at the site in question.

On the basis of the recent scientific studies described above, and in particular the IEHR study, GE submits that a clear and sufficient scientific basis is now available to warrant regulation of PCBs by their degree of chlorination ("closest Aroclor" approach). To accomplish this, GE believes both a short-term and long-term plan is needed.

For the short term, the re-read results allows EPA to treat the risk assessment of Aroclors 1260, 1254 and 1242 differently. With respect to Aroclor 1260, EPA should utilize a cancer potency of 1.9. There is no logical basis to continue the current practice of only using the results obtained in female Sprague-Dawley rats. A comparison of the results of each of the cancer bioassays with 60% chlorination shows a striking similarity in the nature of

the tumor response. It should be noted that three separate strains of rats were used and that the similarity of response is apparent when one compares female Sherman rats, male Wistar rats, and female Sprague-Dawley rats. Male Sprague-Dawley rats, while developing the same type of liver tumors, did so at a lower incidence. To assume that this reduced response reflects a generic tendency of male rats not to develop tumors is not supported by the data. The greatest incidence of liver tumors (91.2%) was observed in male Wistar rats. The results in male Wistar rats also do not support continuing the practice of censoring the male Sprague-Dawley results from the calculation of a cancer slope factor.

Employing the geometric mean of the cancer potency factors of the four rat study groups--female Sherman, male Wistar, male Sprague-Dawley, and female Sprague-Dawley--would reflect a less arbitrary use of all existing data. There is ample precedent for this approach in a number of Agency decisions. The geometric mean, using the re-evaluation results, would yield a cancer potency factor of approximately 1.9. The current value calculated by EPA is 7.7 using only the female Sprague-Dawley rat.

With respect to the studies on lower chlorinated PCB mixtures, the results do not show a statistically significant increase in tumor incidence over control groups. Therefore, EPA should not treat these as carcinogens at all! However, as an interim measure, EPA could calculate apparent cancer potency factors of 0.3 and 0.2 for Aroclor 1254 and Aroclor 1242, respectively, as a basis for regulation until sufficient data on non-carcinogenic endpoints is available and has been assessed for estimating risk. While GE does not subscribe to utilizing this data, i.e., negative studies, to define cancer potency factors, if EPA chose to do so, it would clearly be protective of human health and the environment.

In the long term, GE is beginning to plan basic feeding studies of laboratory animals of all three Aroclors for the purpose of putting all studies on an equal footing. GE believes this will not only reconfirm that Aroclor 1242 and 1254 are not carcinogenic in laboratory animals, but also that the potency factor of 1260 is less than currently utilized by EPA. GE believes that EPA involvement in designing, reviewing and overseeing the results will allow the science of PCB toxicity to advance to a significant degree.

GE requests EPA to begin immediately to re-examine the PCB scientific information and literature and to reconsider the way in which PCBs are regulated . . . specifically to reconsider the regulation of all PCBs as if they are equivalent to Aroclor 1260. We offer to participate with EPA in developing a forum that is appropriate for the discussion and debate of these issues.

GE believes that recent science allows EPA to better manage the potential threats and better balance the risks and benefits of PCBs. The combined short and long term approach will also allow EPA to act expeditiously since the re-read results provide the basis for allowing a short term change.

III. THE RESULTS OF RECENT SCIENTIFIC STUDIES WARRANT A RE-EVALUATION OF THE PRESENT REGULATORY FRAMEWORK FOR PCB CLEANUP AND DISPOSAL

A. PCB Cleanup and Disposal Requirements Must Be Revised To Reflect The Rational Prioritization Of PCB Regulation Within Our National Waste Management Program

s the preceding discussion demonstrates, the scientific evidence since Congress' decision in 1976 to regulate PCBs strongly indicates ubstances do not pose the extreme risk to human health and the t that we once believed them to present. Over the last fifteen PA and the regulated community have learned much about the isk of PCBs and how to more accurately assess that risk as it n health and environmental protection. It is time, therefore, evise the approach taken to date regarding PCB regulation to vely reflect the reduced risk that scientific evidence by the presence of PCBs in the environment.

known speech last year before the National Press Club, eviewed the strengths and weaknesses of this country's ironmental policy over the last two decades and has come to start taking aim before we open fire" on facing our society. "Aiming Before We Shoot; The nmental Policy," September 26, 1990. As part of ator called for an Agency-wide "strategic ocusing our attention and resources on areas of ntial for risk reduction." Id. In view of this PA use this rulemaking as an opportunity to has taken to PCB regulation over the past gram based on a realistic assessment of this sts of PCB management and disposal, in ve on human health and environmental pacity, development of alternative materials recycling and conveyance of

EPA to conduct a comprehensive lity, effectiveness, and cost of all idered for inclusion in a revised ill likely receive some data in the e available for submittal of VPR request will undoubtedly n. Thus, EPA should invest hensive study of the status ering information from ed community. Much

valuable information concerning the effectiveness and feasibility of various PCB disposal options has been generated by EPA and private industry which the Office of Toxic Substances should have in re-evaluating the PCB regulatory program.

A comprehensive information-gathering effort would be most successful if the Agency had full participation from the regulated community. Given the need for such data and the number of issues that have arisen since the PCB regulations were promulgated, GE suggests that the Agency make this proposed rulemaking a negotiated process to facilitate a cooperative effort between EPA, the regulated community and other interested parties. GE believes such an approach would be a much more effective means of addressing the PCB issues presently being considered than would the traditional rulemaking process.

To evaluate the appropriateness of various disposal options without adequate information, as is presently the case, is not an efficient use of the rulemaking process and will likely result in a superficial treatment of the scientific, technical, and regulatory issues involved in PCB waste disposal. EPA needs to develop full information concerning the technical, economic and social aspects of all available disposal options so that it can evaluate the appropriateness of using each option in a given cleanup situation.

To illustrate, there exist a number of disposal mechanisms that are or could be provided for under a revised PCB regulatory disposal scheme. These can be generally categorized as follows:

- Incineration;
- Disposal in a TSCA-permitted landfill;
- Alternative disposal technologies, such as thermal destruction, physical separation, stabilization and biological or chemical dechlorination; and
- Disposal in place or in landfills meeting requirements less stringent than TSCA units.

Incineration and TSCA landfilling are the principal options provided for under the current regulatory scheme and were made the favored means of disposal at a time when EPA knew much less than is now known about the disposal characteristics of PCBs and their relative lack of potential to persist and migrate in the environment. While the stringency of these disposal options may maximize human health and environmental protection, serious questions arise as to the practical feasibility of requiring their use for disposal of most PCB waste, much of which contains low-level contamination. EPA needs to reexamine its emphasis on use of incineration and TSCA landfilling, factoring in such issues as the growing scarcity of available landfill and incineration capacity, the strong public opposition to new facilities siting, and

the risks attendant in the long distance transport required to get PCB waste to a permitted facility. This requires collection of a body of information EPA does not presently have but should obtain to aid it in the policymaking process.

Alternative disposal technologies are beginning to be developed but are still in varying degrees of nascence, in large part because of impediments posed by the present regulatory framework, a problem discussed in more detail below. Notwithstanding the preliminary nature of these technologies, data do exist concerning their potential feasibility, effectiveness and cost. EPA needs to compile all available information concerning these alternative disposal methods and evaluate which of the technologies that have been or are being developed may provide long-term solutions to PCB waste disposal. EPA appears to be moving in this direction through issuance of draft guidance on certain non-destructive disposal technologies. See "Draft Interim Guidance on Non-Liquid PCB Disposal Methods To Be Used As Alternatives to a 40 C.F.R. 761.75 Chemical Waste Landfill" (July 3, 1990) ("EPA Alternative Disposal Guidance"). GE strongly supports EPA's initiative and urges the Agency to undertake an even more systematic, in-depth look at the priority alternative disposal technologies which should be considered among other available PCB disposal options.

Finally, EPA needs to consider the appropriateness of allowing in-place disposal or disposal in landfill units that are not subject to the same requirements as TSCA landfills. EPA made the determination that PCB-contaminated soil and debris generated during cleanup operations could be TSCA-landfilled rather than incinerated without compromising concerns for environmental protection. In light of the scientific evidence discussed above, the Agency now needs to examine how it should approach disposal of low-level PCB-contaminated soil, industrial demolition debris and machinery. If considerations of realistic risk, limited landfill capacity and cost effectiveness are kept in mind, EPA will likely find that in-place and alternative landfill options would be appropriate for a number of PCB waste types.

B. A National PCB Disposal Policy Is Needed To Give Certainty To The Regulatory System

Re-evaluating the present regulatory structure is the first step required to render the system more resource-efficient. However, EPA will not achieve its goal of increasing flexibility in the safe, effective management of PCBs unless the Agency adopts a national policy that articulates a federal preemptive interest in the uniform application of PCB disposal requirements. If a regulatory program is to achieve optimum compliance and encourage voluntary cleanup efforts among the regulated community, the elements of the program must be clear and unambiguous. In the world of PCBs, inconsistency is one of the biggest obstacles to understanding the dictates of the regulatory program, in large part because of the myriad of standards and requirements imposed at the EPA region, state and local levels in the area of

PCB cleanup and disposal. As discussed in more detail below, the regulated community is faced with extreme inconsistency in the enforcement of PCB regulations, and as a result, must spend inordinate amounts of time and money negotiating with multiple regulatory authorities over cleanup standards and procedures that do not necessarily bear any relation to human health or environmental risk. To restore certainty and predictability to the regulatory process, GE urges EPA to establish a national PCB disposal policy, consolidate management of this area at the federal level, and set clear, concise standards that can be uniformly applied by the EPA regions across the country.

C. Issues Involved In Addressing PCB Soil Contamination

1. Present Regulatory Framework Allows Limited Disposal Options

The problems associated with addressing historical PCB contamination in soil provide a perfect illustration of how the present regulatory system is ill-designed to facilitate environmentally protective, cost-effective prioritization of available disposal options. Under the present regulatory system, excavation is the principal means available for addressing PCB-contaminated soil, and EPA's application of the anti-dilution rule to PCB-contaminated soil further expands the scope of materials that must be excavated. As a result, facilities across the country are being required to excavate and TSCA landfill or incinerate thousands of cubic yards of material--much of which is contaminated at concentrations that, in the case of "new" spills, are allowed to be left in place under the PCB Spill Cleanup Policy. Furthermore, limited TSCA landfill space is being used for disposal of material that is not even subject to regulation, because there are few non-TSCA landfills willing to accept low-level, unregulated PCB-contaminated soil. Considering the average cost of $500 per cubic yard for analysis, transportation and disposal of such waste--not to mention the road miles involved in moving the material to its destination--requiring TSCA-landfilling of low-level contaminated waste does not make good economic or environmental sense.

2. Development of Alternative Disposal Technology Is Impeded

The PCB regulations do provide mechanisms for employing alternative methods of disposing of PCB-contaminated soil. For example, 40 C.F.R. § 761.60(e) provides for the permitting of alternative destruction technologies, and EPA has applied this section to permit the use of non-destructive disposal methods. Also, 40 C.F.R. § 761.75(c)(4) allows the permitting of landfills with less stringent requirements. However, the process involved in obtaining approval of alternative technologies and the restrictions and inconsistencies associated with implementation create such impediments that it is not often feasible for the regulated community to take advantage of alternative disposal mechanisms. Part of the problem has

to do with the fact that the PCB regulations create a preference for destruction technologies which are much more difficult to develop than other methods that reduce the mobility and toxicity of the PCB material. See, e.g., Memorandum from John Smith to Bob Murphy, EPA Region VI, attached as Exhibit 9. Moreover, the Agency's equivalency standard based on destruction of PCBs to 2 ppm can not be practically achieved by most alternative methods. Maintaining a bias in favor of destruction technologies will ensure inhibition of research and development into innovative PCB waste disposal methods. EPA's Draft Alternative Disposal Guidance is a step toward correcting that bias and should be further developed to allow advancement of non-destructive PCB disposal options.

A larger problem involving application of these alternative disposal technologies involves the lack of uniform standards to which the regulated community can look to determine the extent of cleanup required. Case-by-case determinations are made by regional EPA offices which set cleanup levels varying from background concentration to 25 or 50 ppm. This inconsistency occurs not only from region to region but even within a particular region with respect to similar sites. To aggravate matters, state and local regulatory agencies often become involved in site cleanup determinations and set standards even lower than those prescribed by the region. Faced with such inconsistent and often unattainable cleanup levels, very few companies in private industry can afford--and even fewer lending institutions are willing to invest in--uncertain technology that may not be approved because it cannot achieve stated cleanup levels.

Equally as troublesome are the burdensome permit requirements with which industry must comply to perform research and development on promising technologies that have potential commercial use. The process of applying for and obtaining a permit for each step in the R&D process--from bench scale test to pilot study to demonstration--is time consuming and expensive. In GE's experience, an average of six to twelve months has been required for permit preparation and approval at a cost of $50,000 to $75,000 per permit. In many cases, parties simply forgo conducting site-specific studies on alternative disposal technologies because the time and cost involved in developing and negotiating an alternative disposal permit with the Agency outweighs the cost of excavation and redisposal in a TSCA landfill. Further, the pressures of other regulatory deadlines, such as remediation timeframes established in Superfund orders, do not allow for the significant amount of time needed to obtain a TSCA permit. This is especially a problem in cases where both headquarters and regional approval are required for technology to be used in more than one region.

These restraints on research and development also impede this country from taking advantage of potentially useful PCB waste treatment technologies being developed around the world. Under existing regulation, access to these technologies requires that foreign personnel and their equipment be brought to the United States so that feasibility studies of their technology can be conducted on soil from domestic facilities. This has been

necessary on two occasions in connection with R&D permits granted to GE in the last year. It is an extremely burdensome and expensive undertaking to arrange short term lab projects here in the U.S. just for the purpose of making an initial evaluation of the applicability of a treatment technique. Yet, given the present structure of the regulations, cost-effective alternatives are not available.

3. EPA Must Revamp The System Using A More Resource-Efficient Approach To PCB Soil Disposal

In short, the regulated community is faced with a regulatory structure that stymies development of innovative technology and limits available PCB disposal options to excavation and use of scarce landfill space with no appreciable increase in environmental protection. In light of the scientific evidence discussed above, such an approach is no longer appropriate and needs to be re-evaluated to take into account the true risk, cost, and effectiveness of both conventional and emerging methods of PCB soil disposal.

The first step in EPA's re-evaluation must involve eliminating application of the anti-dilution rule to cleanup of historical soil contamination. GE agrees that the anti-dilution rule serves an important function as applied to recent spills of PCBs and maintenance of active PCB electrical equipment. However, rote application of the rule to pre-existing contamination makes no sense and results in the inappropriate use of limited chemical waste landfill capacity and an undue financial burden on the regulated community. The Superfund program has recognized this logic and eliminated application of the rule to CERCLA cleanups; there is no principled reason why the rule should apply to cleanup of old PCB contamination. In fact, taking PCB contamination levels in soil and other materials "as found" would be consistent with EPA's regulation of electrical equipment, because in reality, the PCB concentration that triggers regulation of a transformer or capacitor is actually an "as found" concentration of material that was originally 100% PCBs and has become diluted over time. Furthermore, eliminating application of the rule to the form and concentration of old PCB contamination in soil would not mean parties would have incentive or be free to further dilute contaminated materials to avoid disposal requirements, because the rule would still hold for such unauthorized activities. However, it is a wasteful misuse of resources and does not serve any tangible goal of environmental protection to require excavation and TSCA landfilling of low-level contamination merely because the contamination originated years before from a high-level PCB concentration source.

In re-examining the scope of materials that are inappropriately subject to regulation, GE also urges EPA to clarify the issue concerning retroactive application of the PCB regulations raised in In Re Standard Scrap Metal Company, TSCA V-C-288, Appeal No. 87-4, August 2, 1990. In enacting the

Toxic Substances Control Act, Congress did not evince a legislative intent to regulate PCBs disposed or otherwise placed in the environment prior to enactment of the statute. To resolve the uncertainty that the Standard Scrap decision has created, EPA is urged to clarify that the regulatory obligation to clean up applies only to PCB contamination that occurred after February 17, 1978.

Another issue GE believes EPA should consider is the appropriateness of allowing disposal in place and use of non-TSCA landfills as protective, cost-effective disposal options for certain contaminated soils. EPA has recognized that the major potential exposure pathways associated with the disposal of non-liquid PCBs are inhalation and dermal contact. EPA Alternative Disposal Guidance at 2. In-situ remediation techniques that contain PCBs can effectively address potential risks posed by these pathways. For example, capping of a site as an interim measure until a use change occurred--with provision for re-evaluation of remedial options at the time the use changed--would be a more workable solution at an ongoing facility than extensive excavation. EPA could also consider establishing fixation requirements for low level contaminated soil, and allow disposal in an industrial or special waste landfill.

With respect to alternative disposal technologies, GE has experience with a number of promising methods that have potential field application, and information concerning such technologies is provided below. However, if EPA is truly serious about investigating and developing alternative disposal methods, it must revamp the PCB regulatory structure to encourage rather than inhibit research and development. First, a reasonable, uniform standard or set of standards for addressing historical contamination must be developed and implemented on a national basis so that the regulated community has certainty as to the level of cleanup required. While different standards may be required depending on the location and nature of the contamination, generic standards applicable to various types of situations can and should be developed based on realistic assessments of risk, cost, constructability and achievable treatment levels. Based on the scientific evidence discussed above, there is strong support in favor of soil cleanup levels in the range of or higher than those contained in the PCB Spill Cleanup Policy. Additional support is provided in a recent study conducted by Terra, Inc. which concludes that the soil cleanup levels prescribed under the Spill Cleanup Policy provide much more protection than the Agency's traditional excess cancer risk factor of 10^{-4} to 10^{-7}. A copy of the report is attached as Exhibit 10. EPA needs to review these data, revise its risk assessments, and develop uniform cleanup standards that the regulated community can reasonably be expected to meet.

Second, EPA must revise the manner in which it regulates research and development activities to allow greater ease and flexibility in conducting bench scale and pilot tests of new technologies. GE believes the best way to accomplish this is through adoption of provisions analogous to the exemptions set forth in 40 C.F.R. §261.4 (d), (e) and (f) for hazardous waste samples, treatability samples and samples undergoing treatability

studies at laboratories and testing facilities. The requirements contained in these provisions concerning agency notification, sample identification, recordkeeping, storage and transportation would provide an ample measure of protection, but would greatly ease the administrative burden laboratories presently face under the current regulatory system. GE also suggests that EPA modify the RCRA provisions as follows to ensure that the exemptions are workable in the context of PCB research and development. Many of the suggestions below have been incorporated in current GE R&D permits and could be adopted on a general basis:

1) Accumulation, transport and study of samples at least as large as twelve cubic yards (10,000-15,000 kg) should be allowed. It is often necessary, for example, to perform parametric studies on a pilot plant scale to determine optimum operating conditions and reliably estimate the practicality and benefit effectiveness of a process. (Parametric pilot scale treatability studies should not be confused with demonstration tests of an optimized process leading to the granting of an operating permit.) Extrapolation of the results of bench scale feasibility studies performed at a level of pounds per day to operation of a process at a level of tons per day can lead to inaccurate conclusions. Pilot plant studies fill this gap and can require quantities of sample substantially larger than 1000 kg, the upper limit contained in the RCRA regulations.

2) The duration for exemption from disposal of PCB waste samples at labs and test facilities should be independent of the date of collection or shipment, and should be defined as one year following completion of treatability testing. It is not reasonable to mandate that R&D studies leading to a successful treatment process for a PCB waste material be completed within one year of initiation. Such a limitation actually deters investment of time and energy in R&D to develop superior technology. Further, because of the demand for disposal facilities, it is often necessary to schedule actual disposal more than ninety days in advance. The one-year period for disposal following completion of treatability studies allows the studies to reach their natural conclusion rather than a premature ending imposed by disposal scheduling.

3) Provision should be made for allowing transfer of small samples (1 kg or less) from research or test facilities to third-party laboratories for the purposes of PCB waste analysis and other characterization.

4) Exemption should be made for archival retention of small samples taken before, during and after treatment.

5) The exemptions should be extended to import and export of small R&D samples (for example, five gallons or twenty-five kilograms) to facilitate the introduction of innovative foreign technologies for the treatment and disposal of PCBs.

6) The RCRA exemption provisions provide for Regional Administrator discretion in making certain additions to the exemption. When labs in more than one region are involved in sample collection, transport, and performance of treatability studies, provision should be made to centralize discretionary decisions through the region in which the studies are being performed.

Another means EPA could consider to ease present restrictions on research and development would be through issuance of permits by rule. EPA has substantial experience in reviewing and issuing TSCA alternative disposal permits. This wealth of experience could be used to establish generic standards to be met in order to conduct studies. This approach would also eliminate the burdensome and time-consuming procedural hurdles of the permit process, but allow EPA to retain substantial control over the conditions under which studies are conducted.

4. Alternative Treatment Technologies Show Great Promise And Should Be Encouraged

In EPA's Alternative Disposal Guidance, the Agency identifies five groups of treatment technologies that may be appropriate for use in the disposal of non-liquid PCBs. GE has experience with a number of the technologies and treatment groups identified. This experience includes laboratory research, pilot testing, and full-scale demonstration of technologies conducted by GE individually or in collaboration with technology vendors. Based on this experience, GE offers the following comments.

a. Bioremediation

Bioremediation of PCBs is an area of research at the forefront of technology, and it offers the realistic prospect of an effective, economic, and environmentally sound remedial or disposal alternative for PCBs. As EPA itself has noted:

> EPA recognizes that new, innovative technologies are needed to effectively prevent, treat, and remediate the growing environmental problems facing the nation. The Agency believes that biotechnology offers significant potential for the prevention, reduction and treatment of pollution.

Because aerobic and anaerobic organisms are capable of surviving under very different environmental conditions, bioremediation applies in a wide range of environmental media. Moreover, although the cost of PCB bioremediation is difficult to estimate with precision, given the importance of site-specific factors, one researcher has estimated that aerobic biological treatment may cost from $50 to $100 per ton of contaminated material.

Perhaps the most significant aspect of bioremediation is that it has minimal, if any, negative environmental impacts. Naturally-occurring microorganisms, nutrients, and oxygen may be all that is necessary to apply the technology successfully. In fact, the widespread abundance of natural PCB-degrading microorganisms, may make inoculation with bacteria unnecessary for most sites.

In sum, both aerobic and anaerobic PCB biodegradation offer reasonable alternatives to traditional, high-energy PCB destruction processes. An important advantage of these new methods is a greater compatibility with the environment. Changes in current PCB regulations will, however, be required for these technologies to achieve their potential, because these low-energy processes are slower and accomplish less complete PCB destruction than traditional methods. EPA's recognition of the promise of bioremediation of PCBs as a remedial and disposal alternative, combined with the demonstrated effectiveness of bioremediation, provides clear and sufficient reason to adapt to this new technology.

b. Solidification/Stabilization

Chemical Fixation - In Situ Inorganic Polymer. Since 1985, GE has worked with International Waste Technologies ("IWT"), a vendor of solidification/stabilization ("S/S") technology on the remediation of a nonregulated, PCB-contaminated site located in Hialeah, Florida. GE allowed IWT and Geo Con, the installer, to use the site in EPA's Superfund Innovative Technology Evaluation ("SITE") Program.

GE believes that the results from the SITE demonstration conducted in 1988 show that S/S effectively isolates PCBs from the environment. The results also convinced local regulators to accept chemical fixation as an acceptable alternative for site remediation. The results of this demonstration project, which was conducted on soils containing total PCB concentrations ranging from less than 1 ppm to 950 ppm PCB, are reported in the following two reports published by EPA:

- "International Waste Technologies/Geo-Con In Situ Stabilization/Solidification Applications Analysis Report" (EPA/540/A5-89/004 August 1990)
- "Technology Demonstration Summary International Waste Technologies/Geo-Con In Situ Stabilization/Solidification Update Report" (EPA/540/55-89/004a January 1991)

Copies of these reports are attached as Exhibits 11 and 12.

In order to measure the effectiveness of the S/S process, EPA conducted a number of tests on the treated material, including TCLP testing, permeability, and the unconfined compressive strength. GE believes that each of these tests showed significantly positive results. TCLP testing of

unweathered samples conducted twelve months post-treatment showed less than 0.2 ug/l PCB, with many results below 0.1 ug/l PCB in the leachate. Permeability testing showed that S/S achieved a reduction in permeability to 0.6×10^{-8} to 5.6×10^{-7} cm/s in samples analyzed one year following treatment. Similarly, unconfined compressive strength testing demonstrated increases by over a factor of two in samples analyzed one year following treatment.

Future tests of the effectiveness of S/S technology should be aimed at measuring S/S efficiency by evaluating the leaching potential of the treated media. An appropriate test of leaching potential should simulate the conditions that exist at the treatment site rather than using a standard TCLP test, which requires grinding a sample and leaching with an acidic solution. Grinding a sample totally defeats the purpose of S/S. Rather than using TCLP, GE suggests that the EPA adopt testing procedures similar to ANS 16.1 and MCC-1 so the fixed mass can be tested as a monolith. Descriptions of the ANS 16.1 and MCC-1 tests are attached as Exhibits 13 and 14.

In determining the acceptability of S/S, EPA should also look at the relationship between the permeability of the fixed mass and the permeability of the surrounding soil. Depending on where the fixed mass sits in relationship to the water table and the gradient of the water table, one or two orders of magnitude difference in permeability could be sufficient to isolate contamination. EPA should not arbitrarily set permeability requirements at 10^{-7} cm/sec.

Experience with S/S in the SITE Program and during full scale remediation has given GE many ideas for process improvement. The process as demonstrated in the Site Program and operated during remediation is described in Exhibits 11 and 12, discussed above. Operation to depths of 25 to 30 feet was uneventful. The installer was able to treat contaminated soil with 15% by weight additive when the soil had a void space of about 17 - 19%. TCLP leach tests run on treated soil during installation had PCB leach values less than 0.2 ppb on most samples which is similar to the results achieved in the SITE Program demonstration. Below 30 feet , the contractor had difficulty penetrating the soil and controlling injection. Excess fluid was injected to ease penetrations. GE believes this problem can be minimized by changing the design of the cutting head and by relocating the fluid injection points to the outer edge of the cutting head.

GE believes S/S as applied at Hialeah effectively isolates PCBs from the environment. Although GE has only used this process on a sandy soil, it believes the process will be effective for reducing PCB mobility in most soils, although clay and cobble soil types may be difficult to treat. Additionally, S/S is likely to be an economical alternative to incineration or a TSCA landfill provided organic contaminant concentration is not too high. ("High" cannot be defined at this time.) This process also appears to be effective in the treatment of heavy metals, low-level volatile organic contaminants ("VOCs"), and in situations where treatment below the water table is required. Attached as Exhibit 15 is a report entitled "Treatability Study Report

for the Effects of Fixation on VOC's" which describes the results of a test conducted by GE on low-level VOC contamination.

GE has recently seen indications that the S/S process may also act to dechlorinate PCBs. It appears that S/S additives containing iron act to dechlorinate halogenated compounds. It is likely that this reaction will only take place in the presence of water. Because this process is so new, it is not possible at this time to tell whether sufficient water will be present in the fixed mass to permit the dechlorination reaction to take place or whether the iron will shift valence states reducing the opportunity for the reaction to occur. This or a similar reaction has been verified by the University of Waterloo and a patent application has been filed for its use in a reactive cut-off wall for treatment of groundwater plumes contaminated with chlorinated solvents. If this or a similar reaction is taking place in the fixed mass, it is conceivable that the PCBs will be destroyed.

The incremental cost of implementing S/S at the Hialeah site is approximately $\$160/yd^3$. In comparison, the cost of TSCA landfill disposal is about $\$500/yd^3$. Using S/S at the Hialeah site to treat approximately 11,000 yd^3 of material will result in a cost savings of over $3 million when compared to landfill cost. The cost effectiveness is even greater if one considers the cost of shoring and water treatment that would be required with excavation.

Vitrification. GE has worked with Battelle and its spin-off, Geosafe, on use of this process at its Spokane facility. A Bechtel report entitled "Evaluation of In-Situ Vitrification for Stabilization of PCB Soil Contamination" evaluates this technology and is enclosed as Exhibit 16. Vitrification of PCB-contaminated soil showed PCB destruction efficiency of 99.9999%. Process costs are favorable when compared to TSCA landfill costs. The Bechtel report shows vitrification costs of $220/ton in comparison to TSCA landfill costs of $290/ton at a hypothetical site.

c. Thermal Destruction

In GE's experience, on-site thermal destruction is not economical in comparison to disposal in a TSCA landfill for small volumes of contaminated soils less than 7,000-10,000 yd^3. On-site thermal destruction is also limited in its application due to the extensive space requirements for implementing this technology.

d. Physical Separation

GE and Mosmans Mineraaltechniek of Oss, the Netherlands, collaborated to perform physical separation bench-scale testing using water and additives. Separation results were promising and achieved results below 25 ppm PCB for the soil types tested. A report entitled "Indicative Testing on Soil Contaminated with PCBs" describes this process and is attached as Exhibit 17. GE is investigating this technology further to determine its potential.

e. Chemical Dechlorination

GE developed a KOH/PEG (potassium hydroxide/polyethylene glycol) process for treatment of PCB contaminated oil and soil and has worked with Galson Technical Services to further develop the process. Work by GE shows the process to be sensitive to water (10% moisture content greatly lowers efficiency) and less effective on dechlorination of lower chlorinated Aroclors than higher chlorinated Aroclors.

As the foregoing illustrates, certain alternative disposal technologies are further along in their development than others. Companies such as GE are eager to continue research efforts to determine which of these and other methods are best suited as long-term commercially workable PCB treatment options. The rate of progress toward that goal will increase once EPA removes the impediments to research and development by adopting the reforms suggested in these comments.

D. Issues Involved In Addressing Industrial Building Contamination

For businesses which have been in operation for the better part of this century, dealing with historical PCB contamination in industrial buildings is a two-fold issue: 1) addressing decontamination of buildings that have continued industrial or other uses; and 2) disposal of materials from building demolition. Because EPA has no uniform cleanup standards that prescribe acceptable cleanup levels, the regulated community is faced with the same inconsistency among regions and state agencies that plagues the area of soil cleanup. In most cases, overly conservative cleanup levels are imposed that cost industry hundreds of thousands of dollars per year in environmentally unnecessary expenditures.

1. Non-Impervious Surfaces

The extent of the problem is most serious with respect to cleanup of porous surfaces such as concrete, brick, wood, cinder block and sheet rock, and stems essentially from application of the wipe test to these porous surfaces. EPA notes in its preamble to the PCB Spill Cleanup Policy that old PCB contamination is generally more difficult to clean up than more recent spills, "particularly on porous surfaces such as concrete . . . ." 52 Fed. Reg. 10688, 10689 (April 2, 1987). Notwithstanding its inappropriateness, in most cases, the surface cleanup requirements contained in the PCB Spill Cleanup Policy are either directly applied or used as guidance in setting cleanup levels for historical building contamination. As a result, standards of 10 ug/100 cm^2 or lower are prescribed. Experience shows that attaining these levels of decontamination is an extremely time-consuming and costly undertaking.

To illustrate, after one cleaning of a concrete surface, approximately 75% of a remediated area is often below 100 ug/100 cm^2, and most of the area is below 300 ug/100 cm^2. Depending on how the site was used, only small areas--approximately 10% of the total space--are above 1000 ug/100 cm^2. Three and sometimes four recleanings are required to achieve 10 ug/100 cm^2 with several localized areas between 10-100 ug/100 cm^2 remaining. These areas must be scarified to remove a quarter inch or more of the surface. One-time detergent cleanings cost approximately $.50-1.00 per square foot. By contrast, scarification costs in the range of $8-10 per square foot with no guarantee that wipe test results below 10 ug/100 cm^2 will be achieved. Thus, in an average building of 50,000 square feet, the cost to achieve 100 ug/100 cm^2 would be in the range of $150,000 - $250,000, whereas the cost to achieve 10 ug/100 cm^2 would be $350,000 - $450,000.

The costs involved in attaining cleanup levels at or below those prescribed in the PCB Spill Cleanup Policy are not justified in view of the risk analysis used to arrive at those levels. EPA acknowledges, again in its preamble to the Spill Cleanup Policy, that PCB transfer rates are likely much lower with respect to rough, porous surfaces than they are for impervious surfaces such as metal or glass. 52 Fed. Reg. at 10696. Nonetheless, the same transfer rate is used for both impervious and non-impervious surfaces in the risk analysis used to determine acceptable surface cleanup standards. Id.

EPA's assumptions about other factors such as volatilization, lifetime average contact fraction, fraction area contaminated, absorption rate and potency factor put into further question the reliability of EPA's basis for surface cleanup standards. These are discussed in more detail in a study entitled "A Critique of EPA's Risk Analysis of PCB-Contaminated Surfaces" conducted by Everest Consulting Associates in 1986, a copy of which is attached as Exhibit 18. While some of the assumptions EPA ultimately relied on in developing the PCB Spill Cleanup Policy may differ slightly from those reviewed by Everest, the report highlights the general ultra-conservatism of EPA's approach. In light of the most recently generated scientific data concerning PCBs, it is time to revisit the exposure and risk analyses relied upon in determining surface cleanup levels, develop more realistic analytic inputs based on the characteristics of porous materials, and establish reasonable cleanup levels for historical PCB contamination of these surfaces.

As part of this re-evaluation, EPA should also revise the regulations to standardize the use of encapsulation of low-contact industrial surfaces for both old and new PCB contamination. Encapsulation is presently considered an acceptable discretionary remedial method for new spills onto low-contact non-impervious surfaces. See 40 C.F.R. § 761.125(c)(3)(iii) and (c)(4)(iv). To ensure consistent implementation, EPA could easily establish standards that would facilitate monitoring of the encapsulated area. For example, EPA could require that a uniform warning color, such as the yellow and black used on PCB stickers, be applied directly on top of the

contact surface with the encapsulating material then applied on top of the warning color. This way, it would be clear as to when the encapsulation was wearing off and needed to be reapplied. Whatever criteria EPA might choose, given the technical and cost effectiveness of encapsulation, it should be considered a viable cleanup technique for minimizing exposure from low-contact porous surfaces.

EPA also needs to develop a new test for sampling of these surfaces. Given the nature of porous materials and the tendency of old PCB contamination to settle into the pores of the material, the wipe test yields extremely inconsistent results that depend in large part on the pressure applied during the wipe. Aside from the inconsistency that results, such a method is inherently unrepresentative of the actual transfer that takes place when clothing or skin contacts a non-impervious surface, and thus, cannot accurately reflect the true human exposure potential of the PCBs.

A similar problem arises in testing of these materials for disposal. Given that the primary concern in a disposal situation is the potential of the PCBs to migrate out of the waste and into the environment, a leach test of the material in the form in which it is disposed would be much more representative of the effect of the waste on the environment than a wipe test or a measurement of total PCBs in a core or chip sample, which in GE's experience, have been the test methods required. With respect to core samples, for example, the distribution of PCBs throughout the core varies depending on the orientation of the core to the exposed surface, the permeability, and the physical and chemical composition of the material. Thus, analysis of core samples will yield different results depending on whether the total core is analyzed or whether the core is sectioned and the various strata are analyzed individually. For these reasons, GE believes that EPA should adopt test procedures similar to the ANS 16.1 and MCC-1 methods discussed above which provide for testing of material in its fixed form. If a stated cleanup objective is to be completely reliable, the sampling method used to determine attainment of the objective must be representative. The sample methods presently used do not reflect the real potential human or environmental risk posed by PCB-contaminated non-impervious surfaces and, therefore, should be replaced.

2. Impervious Surfaces

In the area of cleanup of historically-contaminated impervious industrial building surfaces, there are three main issues GE urges EPA to address: 1) the difficulty involved in cleaning isolated low-contact surfaces, such as wall-to-ceiling joints, rafters and hard to reach building recesses; 2) disposal of building materials in connection with demolition; and 3) standards for cleanup of incidentally-contaminated machinery. The problem with respect to all of these issues lies, again, in the inconsistent, ultra-conservative requirements that regions and states apply in the absence of reasonable, uniform cleanup and disposal standards for these materials. Isolated upper surfaces and recesses of a building pose a minimal dermal

exposure potential because of the fact that they are so inaccessible. Yet cleanup standards applied to these areas are often in the range of 10 ug/100 cm^2 or lower, based on the PCB Spill Cleanup Policy. The time and effort required to reach these levels multiplies the cost of a cleanup by several factors with no discernible increase in environmental protection. It would be much more reasonable and cost effective to apply to high level recesses and surfaces the 100 ug/100 cm^2 standard the Spill Cleanup Policy applies to low contact outdoor surfaces in industrial facilities, see 40 C.F.R. § 761.125(c)(3), because of the similarity of these areas regarding potential for dermal exposure.

Regulatory certainty as to the acceptable level required for disposal of impervious building materials such as steel and metal in connection with demolition is also needed. Given the lack of uniform standards, regulatory agencies often set standards that are impractical to meet, and landfills and scrap facilities around the country either refuse to take the material or set varying, arbitrary levels for acceptance. Confusion and uncertainty would be minimized if EPA would establish national criteria for cleaning impervious building materials to specified levels based on the planned disposition of the material as scrap, as reuseable machinery, for smelting, or for disposal in a TSCA or special/industrial waste landfill, depending on the concentration of PCBs remaining on the material.

National standards of the type described above should also be applied to cleanup of incidentally-contaminated machinery to resolve the inconsistency and uncertainty that results from EPA's lack of uniform standards for addressing this category of PCB-contaminated materials. Of equal importance with respect to cleanup of industrial machinery is the need to develop a more rational sampling approach than is presently provided by the standard wipe test. Given the number of different surfaces on a piece of equipment, use of the standard size templates of ten by ten centimeters provided for under 40 C.F.R. § 761.123 is not always possible and, in any event, to sample the entire machine, requires analysis of hundreds of templates at an exhorbitant cost. A re-evaluation of the most economically and technically feasible method of sampling PCB-contaminated machinery for proper disposal is needed to address this problem.

One final issue with respect to cleanup of incidentally-contaminated machinery that also warrants EPA's attention concerns the disposal of wastewater generated during cleaning of this equipment. Characteristically, this wastewater contains low-level PCB contamination up to 30 ppb. Yet, by operation of EPA's application of the anti-dilution rule, this water would be considered to contain the PCB concentration of the original source and, thus, would have to be incinerated in accordance with the PCB disposal regulations. This is an absurd result that requires needless expenditure of huge sums of money that can and should be put to more environmentally beneficial uses. In the interests of wise resource allocation, EPA is again strongly urged to reinterpret the anti-dilution rule not to apply to historical

PCB contamination or wastes generated during the cleanup of such contamination.

IV. CONCLUSION

GE appreciates the complexity of the process involved in translating scientific data into regulatory standards that are protective of human health and the environment. Nonetheless, as knowledge in the scientific area becomes more refined, it is incumbent upon the Agency to make adjustments in both the assumptions that underlie the framework of regulation and the regulatory framework itself. These comments are directed at both of these issues and are intended to provide information the Agency should have before it in reassessing the scientific and regulatory status of PCBs. The issues GE has raised herein are of serious concern to it and other members of the regulated community. Thus, the Agency is urged to consider them carefully.

As noted above, time constraints have prevented GE from providing comments and technical data on all of the issues raised in the ANPR. Therefore, in accordance with the Agency's willingness to consider information provided subsequent to the ANPR's August 9, 1991 close of the comment period, GE plans to supplement its submittal within the next several weeks concerning both the issues raised in these comments and other matters discussed in EPA's Advance Notice.

If there are questions about the comments and suggestions submitted above, please contact Stephen B. Hamilton, Ph.D. at 203/373-2316.

END NOTES

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EVALUATION OF THE TOXICOLOGY OF PCBs

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CONTENTS

C O N T E N T S
(continued)

I. EXECUTIVE SUMMARY

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Polychlorinated biphenyls (PCBs) are a class of chemical compounds that were widely used as lubricants and fluids in electrical equipment, among many other uses, until concerns about their potential hazards from environmental exposures arose in the 1970s. This report sets out an assessment of the available scientific literature concerning the hazards, particularly cancer, resulting from human exposure to PCBs. It was prepared under the guidance of a panel of internationally recognized experts in medicine, toxicology, and epidemiology and has been subjected to scientific peer review. (The curricula vitae of the panel are set out in Appendix A.) The panelists have unanimously endorsed the findings presented in the study.

The study focuses on whether low-level environmental exposure to PCB is likely to cause cancer in humans. The panel observes that other possible human impacts from low-level environmental exposures to PCBs are of far lower likelihood than any possible carcinogenic effects. Health effects, other than carcinogenicity, that have been observed following exposure of laboratory animals to PCB mixtures include liver and stomach lesions and reproductive effects. The panel notes that these effects, as well as skin lesions (i.e., chloracne) that have been reported in humans following exposure to very high levels of PCBs, occur at higher doses than those associated with risk levels of concern for potential carcinogenic effects. Thus, exposure levels that pose no significant risk of cancer should not be of concern with regard to other health effects.

A variety of epidemiologic studies have examined whether PCB-exposed workers have incurred a greater likelihood of cancer mortality than the general population. The report summarizes the literature in the field. The panel concludes that the body of epidemiological evidence does not demonstrate a causal relationship between PCB exposure and any form of cancer. This conclusion is confirmed by reviews of several other expert groups, including the EPA, FDA, and the World Health Organization. In light of the long-term and widespread usage of PCBs in numerous industrial settings and the extensive exposure of workers in some cases, it is likely that evidence of carcinogenicity would have already been revealed by the studies if PCBs were in fact a potent human carcinogen.

Experiments have also been conducted with laboratory animals to estimate the potential impacts of PCBs on humans. In such studies the subject animals are chronically exposed over a long time period to high does of PCBs through, for example, the addition of PCBs to the animals' food. The studies show that, of the PCB mixtures that have been adequately tested, only certain mixtures -- commercial mixtures of PCBs containing approximately 60 percent chlorine by weight (Aroclor 1260 and Clophen A60) -- produce a statistically significant increase in the incidence of malignant liver tumors in rodents. In a study conducted by the National Cancer Institute, Aroclor 1254 (approximately 54% chlorine by weight) was not found to be carcinogenic. Other Aroclors have not been tested in acceptable animal bioassays for carcinogenicity. Clophen A30, which closely matches Aroclor 1242 in composition (approximately 42 percent chlorine by weight), has been tested; it causes an increase in benign (non-cancerous) liver tumors and is less potent than Clophen A60 when both mixtures were tested in bioassays of identical design.

Based on the fact that no statistically significant increase in malignant tumors is seen in animal studies with lower chlorinated PCB mixtures, the panel concludes that, if the lower chlorinated PCB mixtures are carcinogenic at all, their potencies are far less than those of mixtures with 60 percent chlorine content.

The panel points out that animal studies have limitations as a reliable indicator of effects in humans. Because of interspecies differences in factors such as absorption, metabolism, and elimination of a test substance, effects that are observed in animals may not occur in human populations, or may occur with different frequencies. Moreover, in order to apply the results from animal studies to humans, it is necessary to make adjustments to account for the fact that environmental exposures of humans are many orders of magnitude less than those to which the rodents were exposed in the animal studies. There are substantial experimental data from which it can be inferred that the carcinogenic effects, if any, from PCB exposure arise only after a certain threshold exposure has been exceeded. Thus, at the low doses that are typical from human environmental exposures to PCBs, the panel concludes that no cancer risk may exist.

Despite the limitations of the animal data, it is common for regulatory agencies to estimate potential risks to humans by extrapolating observations in highly exposed animals to humans. The typical extrapolation procedure also assumes the absence of a threshold and a direct proportion between risk and exposure at low doses. Such an application of the animal data reflects a policy decision to adopt a conservative estimate of potential human risk from exposure, and it is the approach taken in this report. Even if this approach were applied to the data on PCBs, however, the panel concludes that it is necessary, at the least, to recognize the differences in potency among the various types of PCB mixtures. The report defines the appropriate adjustments for the various mixtures.

In summary, while the data show that some carcinogenic effects are observed in rodents that are exposed over a long term to high doses of 60 percent chlorinated PCB mixtures, these animal data have uncertain implications for human exposure. Moreover, no statistically significant increase in malignancies are seen in animals exposed to high levels of lower chlorinated PCB mixtures. Thus, even if the animal data were deemed relevant to the environmental exposure of humans, at the least, adjustments must be made for lower potency of the lower chlorinated PCB mixtures. In any event, the panel concludes that the evidence does not demonstrate a causal relationship between exposure to PCBs of any type and any form of human cancer.

II. INTRODUCTION

II. INTRODUCTION

Polychlorinated biphenyls (PCBs) are a class of chemical compounds that were widely used as lubricants and as fluids in electrical equipment, among many other uses, until concerns about their potential hazards arose in the late 1970s.

This report examines the available scientific data regarding the potential carcinogenicity of commercial PCB mixtures. It has been prepared by a group of independent experts who have been engaged to assist Texas Eastern and subjected to scientific peer review. The curricula vitae of the members of the group are set out as Appendix A.

This analysis is confined to an examination of the data regarding the potential tumorigenicity of PCBs because this is the toxicity endpoint of primary concern with regard to low-level, environmental exposures to PCBs. Other health effects demonstrated in animal models occur at higher doses than the doses associated with risk levels of concern for potential tumorigenic effects. Thus, the exposure levels that pose no significant risk for tumorgenicity from chronic exposure should not be of concern for other health effects.

III. SCIENTIFIC EVIDENCE RELATED TO THE TUMORIGENIC POTENTIAL OF PCBs

III. SCIENTIFIC EVIDENCE RELATED TO THE TUMORIGENIC POTENTIAL OF PCBs

A. Summary and Conclusions

This report provides a summary of epidemiological and toxicological data related to the potential tumorigenic effects of PCBs. The data reveal that there is insufficient evidence to classify PCBs as human carcinogens, i.e., the available epidemiological data do not show a causal relationship between PCB exposures and human cancer. This conclusion is supported by reviews conducted by several expert panels (e.g., EPA 1988, ATSDR 1987).

The data also show that certain mixtures of PCBs, those associated with Aroclor 1260,[1/] produce excess tumors[2/] of the liver in rodents in long-term feeding studies at very high doses. Because there is some equivocal evidence that other mixtures of PCBs may produce excess tumors in experimental animals, we cannot reject the hypothesis that other mixtures of PCBs (those associated with other Aroclors) are animal tumorigens. Nevertheless, it will be shown that, if these other mixtures of PCBs are tumorigens, they are certainly of lower tumorigenic potency (tumorigenic risk per amount of exposure) than those associated with Aroclor 1260.

1/ As will be discussed herein, there are a variety of different types of PCBs. Aroclor 1260 was a tradename for a particular PCB mixture containing 60 percent chlorine by weight. Lower chlorinated Aroclor mixtures (for example, Aroclors 1248 and 1242, 48 and 42 percent chlorine, respectively) were also widely used.

2/ The excess tumors may be either benign (non-cancerous) or malignant (cancerous). It is conservatively assumed by regulatory agencies that both types should initially be counted equally in estimating carcinogenic potency. PCBs are therefore referred to as tumorigens, i.e., capable of producing either type of tumor.

In the concluding part of this report, we shall present estimates of the tumorigenic potencies of the mixtures of PCBs associated with Aroclors 1260, 1254, 1248, 1242, and 1232. These potency values are included in this report because of their use in estimating the potential carcinogenic risk that might result from human exposure to these mixtures.

Before presenting a review of the data related to carcinogenicity, a discussion of the chemical compositions of various sets of PCBs is presented.

B. Chemical Composition of Aroclors

Polychlorinated biphenyls (PCBs) are mixtures of chemically related compounds. The various PCBs all share the same basic biphenyl (12-carbon) structure (figure 1) with a varying number of chlorine atoms. Up to ten of the carbon atoms of the biphenyl molecule can chemically bond (attach) to chlorine atoms. If only one chlorine atom is bonded to the biphenyl molecule, the product is referred to as monochlorobiphenyl. It is possible for the single chlorine atom to bond to carbon atoms in different positions in the biphenyl structure, and each such bonding creates a new chemical (figure 1). There are thus several different forms of monochlorobiphenyls that may have different properties. These different structures are referred to as monochlorobiphenyl isomers. Similarly, different isomers may be created when two chlorines are present (dichlorobiphenyls), when three chlorines are present (trichlorobiphenyls), etc., depending on the location of the chlorine atoms with reference to the carbon atoms in the biphenyl structure. PCBs that have different numbers of chlorines, e.g., 5 chlorines (pentachlorobiphenyls) and 6 chlorines (hexachlorobiphenyls), are called congeners. The various PCB mixtures are referred to as sets of congeners, even though some of these sets contain PCBs related to each other as isomers.

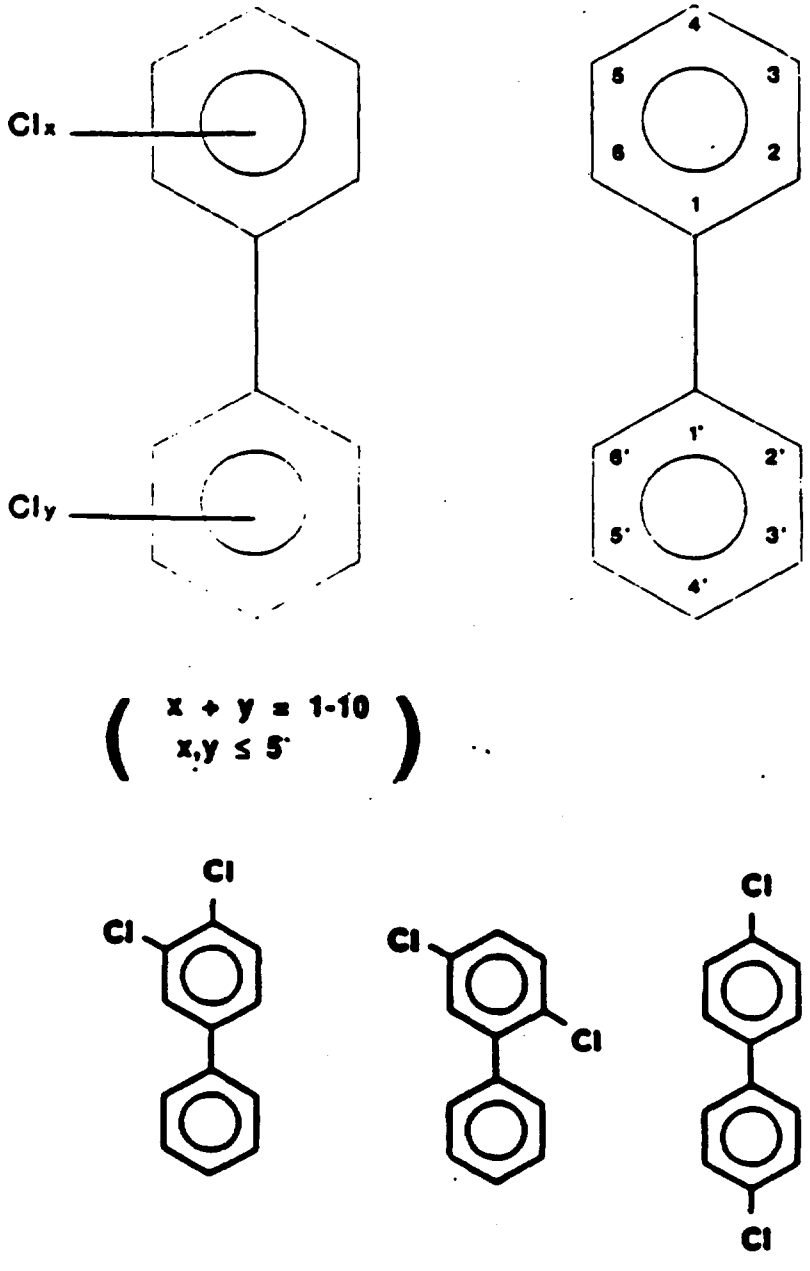

Figure 1. Structure and nomenclature of PCBs. Top left: Biphenyl structure with notation of variable number of chlorines. Top right: Biphenyl structure with numbering of carbon atoms for designating isomers. Bottom: Three isomers of dichlorobiphenyl: 3,4-dichlorobiphenyl, 2,5-dichlorobiphenyl, and 4,4'dichlorobiphenyl.

Individual chlorinated biphenyls can be separated from a mixture by a technique called gas chromatography, and their structures can be confirmed using mass spectroscopy. Figure 2 is a schematic that was produced from the results of such a procedure for Aroclors 1260, 1254, and 1242. When the results of such a separation are represented in a graph (i.e., a chromatograph) as in figure 2, the horizontal location of the bar (labelled "Peak Number") is an indication of the specific PCB congener, or congeners if the separation is not complete. (The relation between these Peak Numbers and PCB congeners by chemical designation is described in Appendix B.) The height of the bar is a measure of the amount (i.e., mass fraction) of each of the various chlorinated biphenyls that are present. Such chromatographs are sometimes called fingerprints, because they can be used to determine the degree of similarity between two mixtures of chemicals even when all of the components of the mixture have not been identified.

Although the correlation is imperfect, as a general rule the number of chlorine atoms per biphenyl molecule increases with Peak Number. Most of the congeners in Aroclor 1260 have more than 4 chlorines, and the average is about 6 chlorines. The chlorine content of Aroclor 1254 is lower than for Aroclor 1260, averaging about five chlorines, and that of Aroclor 1242 lower still, averaging about three chlorines. Thus, although all three are mixtures of PCBs, the actual chemical composition of the mixtures (i.e., the fingerprint) is quite different. Even so, it is readily apparent from figure 2 that the spectra of congeners (i.e., the chemicals present) in the three mixtures overlap (i.e., have some chemicals in common for the mixtures), especially for Aroclor 1260 and Aroclor 1254. Aroclor 1260 has relatively little overlap with Aroclor 1242. Because the chemicals that comprise these mixtures are different, the chemical, physical, and toxicological properties of each mixture would also be expected to differ.

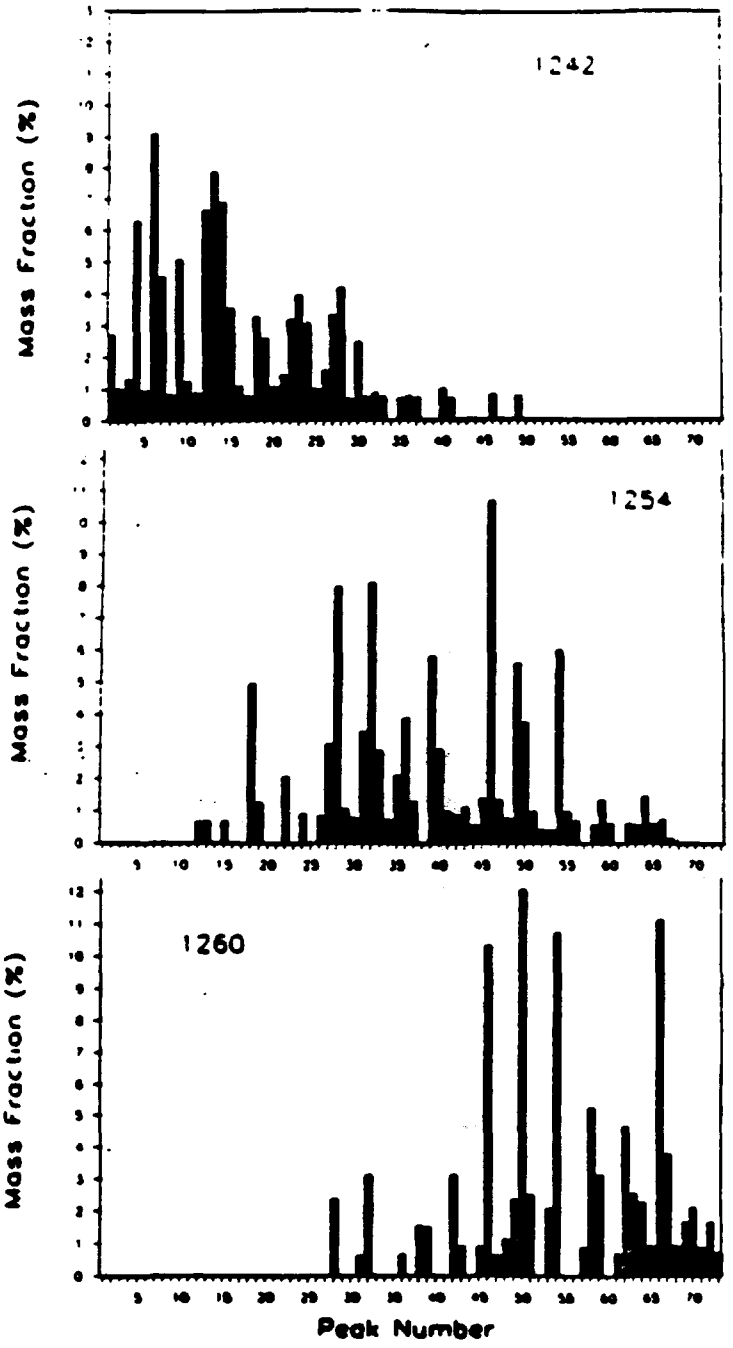

Figure 2. Distribution of mass fractions of PCB congeners in Aroclors 1242, 1254, and 1260.

Two other commercially produced PCBs -- Clophen A30 and Clophen A60 -- are relevant to this discussion. These substances were produced in West Germany and have compositions similar to the Aroclors. Specifically, Clophen A60 is approximately identical to Aroclor 1260, and Clophen A30 is similar to Aroclor 1242. The importance of these similarities will be discussed later.

C. Epidemiological Data

1. General Considerations

The study design that is most commonly used to examine the occurrence of cancers in PCB-exposed populations is a type of cohort study (mortality study) in which death from cancer (all cancers and site-specific[1/] cancers) is the endpoint of interest. In the majority of these studies, the cancer mortality of the population exposed to PCBs is compared to cancer mortality rates of the general population. Standardized Mortality Ratios (SMRs) are then determined by dividing the number of cancer deaths (either total or site specific) in the exposed group by the number of deaths that would be expected by applying rates developed from a reference population. The choice of the reference population (e.g., U.S. average, regional, or state) is critical for the analysis because the normal occurrence of cancer or a particular type of cancer may vary with the population selected. It is often preferable to use a local population (i.e., from the same region or locality as the study population) as the reference population to account for possible unknown confounding variables that could

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[1/] Refers to the anatomical site (e.g., liver) at which the cancer was identified.

influence the mortality experience of the study. For example, if drinking water contaminants increased the cancer mortality of the study (and local) populations, the use of cancer mortality rates based on the U.S. population would not account for this, whereas the use of local rates would.

Statistical analyses are generally conducted on the findings to determine whether any observed increase in the general or site-specific mortality rates in the exposed cohort is statistically significant.[1/] It should be noted that the probability that one or more comparisons will be found to be statistically significant by chance alone increases with the number of comparisons that are made (e.g., number of site-specific cancer mortality rates in the study cohort vs. the number expected based on reference population rates) (Daniel 1983).

If a statistically significant increase in the overall or site-specific rate of cancer mortality is found in the study cohort, one must assess whether the observed difference might have resulted from bias in the manner of data collection. This involves evaluating the ability of the investigators to determine who was exposed (and to what extent) and the methods used for identifying cases of cancer mortality. One must also determine whether the finding might have resulted from the effects of uncontrolled or "confounding variables." This is done by assessing the degree to which investigators accounted for other risk factors (e.g., smoking) in the study design and analysis.

1/The level of statistical significance, or p-value, is generally used to determine this. Traditionally, if a p-value is less than 0.05, chance (although always a possibility) is considered to be an improbable explanation of the results. Conversely, if the p-value is greater than 0.05, chance is considered to be a likely explanation for the observed effect, e.g., cancer.

In the particular case of examination of epidemiological data related to cancer endpoints, there are a number of specific methodological issues that should be considered. These include:

Latency Period: For many cancer endpoints, a long interval may occur between exposure and detectable tumor development. Thus, any cohort study should have a sufficiently long period of follow-up to account for this factor.

Misclassification: It is necessary to classify accurately the exposure status of humans to the substance of interest over time. Important differences in interpretation of epidemiological findings may be obscured if persons are misclassified with regard to exposure.

Pathological Verification of Disease: It is important that investigators clearly state the methods used for identifying disease. There is considerable uncertainty associated with the diagnoses based on death certificates. These should be confirmed using pathology records whenever possible. This is especially important in identifying primary cancer sites because of the tendency of malignancies to metastasize, i.e., to spread to organs other than the site affected by the substance of interest.

Confounding Variables: Exposure to an agent, such as PCBs, may be associated with other possible determinants of cancer risk. For example, cancer rates differ on both a regional and community basis. These differences may in some cases be attributable to qualitative and quantitative variations in the

spectrum of environmental agents (e.g., chemicals in air and water) to which local populations are exposed. Exposure associated with personal habits (e.g., smoking) and past occupations may also account for an observed increase in cancer mortality in a study population. If information is available on the potential confounding variables, it may be possible to adjust for their effects in the analysis. In many cases, however, the relevant confounding variables may be unknown or difficult to measure, complicating the determination of conclusions regarding causation.

After assessing the possible effects of the above factors in an individual epidemiological study, it is generally necessary to apply certain additional criteria in making a judgment as to causality -- i.e., whether exposure to the agent was the cause of the observed health effect. These criteria are frequently presented and discussed in general textbooks of epidemiology (e.g., Rothman 1982, Mausner and Kramer 1985). Most authors base their discussions of the subject on the nine criteria that were noted by Hill (1965) as especially important for consideration when reaching a judgment on the causal nature of an association. The current practice among epidemiologists is to adopt a set of criteria that represent a modification of those originally presented by Hill. Mausner and Kramer (1985) identified the following criteria to evaluate the likelihood that an association is causal:

Strength of the Association: This criterion refers to the degree to which the incidence of the disease is elevated in the exposed population as compared to the control population. In mortality studies, the strength of the association is indicated by the magnitude of the Standardized Mortality Ratio (SMR).

Statistical analysis of the data is necessary to determine the likelihood that an observed association between exposure to an agent and the subsequent development of disease (e.g., as indicated by an elevated SMR) is a chance outcome or is indicative of a true association.

Dose-Response Relationship: This refers to the criterion that the risk of developing the disease usually increases as the exposure increases. The demonstration of such a dose-response relationship increases the likelihood of a causal association.

Consistency of the Association: This criterion refers to the repeated observation of an association in different populations under different circumstances (e.g., under different patterns of exposure).

Temporally Correct Association: Exposure to the suspected causative agent must precede the effect in time. Also, with respect to cancer, a sufficient latency period (i.e., period between exposure and the development of the disease) is necessary for the association between exposure to the agent and the development of disease to be biologically plausible.

Specificity of the Association: This criterion requires that exposure to a causative agent should lead to a unique effect. While the observation of specificity is strong evidence for causal association, its absence is of less significance. Some agents (e.g., smoking) have been strongly linked to multiple effects.

Coherence With Existing Information: This criterion usually refers to the extent to which a causal interpretation is biologically plausible given the current state of scientific knowledge. The likelihood of an association is stronger if it is supported by experimental evidence.

As noted by Rothman (1982), there is no rigid rule to specify when a causal relationship has been established. Any conclusion regarding the likelihood of a causative association is ultimately based on individual, expert judgment. The more criteria that are met for an exposure in question, the greater the likelihood of a causal association. The observation of a statistical association in one or more epidemiological studies usually is insufficient, by itself, to establish causation.

2. Review of Studies

The potential biological effects of human exposure to PCBs have been examined in several populations, primarily workers exposed through occupational activities. As noted previously, EPA (1988) and the Agency for Toxic Substances and Disease Registry (ATSDR 1987) have concluded that the available data are not sufficient to demonstrate that PCBs cause cancer in humans. Nevertheless, the available data, and the strengths and weaknesses of the studies, are reviewed in this section.

The majority of these studies involve occupationally exposed cohorts, and most include only rough measures of exposure (e.g., duration of employment, employment category). The studies differ in the extent to which investigators were able to account for possible confounding variables. As noted above, in some cases such variables could account for any observed increases in cancer mortality in the study population.

a) Brown and Jones (1981); Brown (1987)

Brown and Jones (1981) reported the results of a retrospective mortality study of 2,567 electrical capacitor workers in two plants located in the United States. An update of this study was subsequently published by Brown in 1987. The type of PCB mixture used in the plants varied over the years and included Aroclors 1254, 1242, and 1016. (The latter is a purified version of Aroclor 1242.)

In the initial study (Brown and Jones 1981), the cohort was followed until January 1, 1976, and included all workers with at least 3 months of employment (after 1940) in areas where there was potential PCB exposure. The expected number of deaths in the cohort was determined using age-adjusted U.S. mortality rates (white males and white females) for the appropriate time periods. The total mortality in the study cohort was lower than expected (163 observed vs. 182 expected), as were the total number of cancer deaths (39 observed vs. 44 expected). The Standardized Mortality Ratio (SMR = [observed deaths/expected deaths] x 100) for cancer deaths in the study cohort was 89. Thus, there were fewer cancer deaths among those exposed to PCBs than would be expected in a general, non-exposed population.

With regard to site-specific cancer mortality, Brown and Jones (1981) reported a greater than expected number of deaths due to rectal cancer (4 observed vs. 1.19 expected) and cancer of the liver, gallbladder, and biliary passages (3 observed vs. 1.07 expected). These findings, however, were not statistically significant which strongly suggests that the observed increases are chance occurrences.

In the update of the original study, Brown (1987) followed the mortality experience of the

original cohort through 1982. Once again, the total mortality in the study cohort was lower than expected (295 observed vs. 317.6 expected), with an SMR of 78.

During the additional observation period no additional deaths from cancer of the rectum were observed, resulting in a lowering of the SMR from 336 to 211 (4 observed vs. 1.9 expected). Two more deaths from the disease categories that include cancers of the liver, gallbladder, or biliary passage were reported; these sites were not analyzed individually for statistical significance. This resulted in a statistically significant excess in mortality when the observed number of deaths from these different categories were combined (5 observed vs. 1.9 expected). But the grouping of the 5 cases of liver, gallbladder, and biliary tract cancer into one category (and thus treating them as a single disease) is questionable.[1/] The etiology of the cancers also suggests that they should be considered separately. The evidence for an association between exposure to some environmental agents (e.g., mycotoxins, hepatitis B virus) and an increased risk of developing hepatocellular (liver cell) carcinoma is relatively strong, whereas that for cancer of the hepatobiliary tract (bile ducts found within the liver) is much less compelling (for example, see Zimmerman 1978). Moreover, none of the 5 cases that were grouped by Brown (1987) were identified as a primary carcinoma of the liver, suggesting that liver

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1/The International Classification of Diseases (ICD) code (Eighth Revision) for deaths from cancer at each of these sites is different, indicating that ICD considers them different diseases.

might merely be the common site of metastasis for cancers from sites of unrelated origin.

An analysis of the data did not show an increase in risk with an increase in latency (time since first employment) or any indication of a dose-response relationship (as measured by length of employment) among the deaths from cancers of the liver, gallbladder, or biliary tract. In discussing the study findings, Brown noted that, due to the small number of deaths and the variability of specific cause of death (i.e., within the category including mortality from malignancies of the liver, gallbladder, and biliary tract), it is difficult to interpret the significance of the findings with regard to PCB exposure.

b) Bertazzi et al. (1982, 1987)

Bertazzi and coworkers (1982) reported preliminary results of a retrospective mortality study of production workers in a capacitor manufacturing facility who were employed for at least 6 months between 1946 and 1970. During the early years of production, workers were primarily exposed to PCB mixtures containing 54% chlorine (Aroclor 1254 and Pyralene 1476) that were later replaced by mixtures containing 42% chlorine (Pyralene 3010 and 3011). Mortality was observed between 1954 and 1978 and compared to local rates. The authors reported a statistically significant increase in cancer mortality among males. The observed excesses in cancer deaths in males were primarily attributed to malignancies of the lymphatic and hematopoietic (blood forming) tissues and the digestive system.

In the update of this study (Bertazzi et al. 1987), the cohort was expanded to include non-production workers. The investigators also

decreased the minimum period of employment (after 1946) needed for inclusion in the cohort from 6 months to 1 week. The mortality experience of the cohort was followed from 1946 to 1982.

The short minimum exposure period is a major flaw in the study design. By defining the cohort in this manner, the authors could attribute cases of cancer mortality to PCB exposure that were likely due to other causes or factors. For example, of the 12 cases of mortality that were identified in females, only four were fully characterized with respect to parameters such as length of exposure, and one of these had an exposure period of only three months. It is possible that the remaining eight cases include women who were exposed for very limited periods, making it much less likely that the cancers were associated with work-related exposures.

The total number of deaths in the cohort by 1982 was 64 (30 men and 34 women). Bertazzi et al. used both national and local mortality rates (adjusted by age, sex, and year) to determine the expected number of deaths in the study cohort. Total mortality (i.e., from all causes) was not elevated for males, but there was a statistically significant increase in overall cancer deaths (as indicated by the Standardized Mortality Ratio [SMR]) and in cancers of the gastrointestinal tract, based on either national or local mortality rates.

In females, statistically significantly increased SMRs were observed only when local mortality rates were used to determine expected numbers of cause-specific deaths. Significant excesses were observed for the categories of deaths due to malignant tumors (cancers) (SMR = 226) and deaths due to hematologic neoplasms (cancers of the blood system) (SMR = 377). The local mortality rates

for females in the age group of concern (generally less than 45 years old), however, are associated with a high degree of uncertainty because of the relatively few deaths that occurred among women of this age group in the town (population 150,000).

Bertazzi et al. (1987) reported that when the data were analyzed by duration of exposure, latency, and the year of first exposure, no pattern or trend in mortality was observed for any category of cancer mortality in males or females. They also noted that, in some cases, an examination of the employment history of cancer victims tends to reduce the probability of an association with PCB exposure, in particular with regard to the males with the excess of digestive system cancer (6 observed vs. 1.7 [national] or 2.2 [local] expected). Upon closer analysis of these cases, the authors state that one individual with stomach cancer had been hired at an advanced age and received a very short exposure. Furthermore, two of the individuals (one with stomach cancer, one with pancreatic cancer) had been security guards with no history of direct PCB exposure. This suggests that only 3 of the observed cases may be in people who had any significant exposure to PCBs, and only one individual (with pancreatic cancer) was exposed for more than one year.

The findings of the epidemiological study conducted by Bertazzi et al. (1987) are not indicative of a causative link between exposure to PCBs and the subsequent development of cancer in humans. In the cancer mortality cases that were identified, no dose-response relationship was observed and no pattern was observed with regard to latency and disease. As noted by the authors, some of the male cancer mortality cases had little or no opportunity for direct PCB exposure. Finally,

interpretation of the study results is constrained by the small number of deaths that had occurred in the study cohort, the short minimum exposure period required for inclusion in the study cohort, and the use of relatively unstable local mortality rates as a standard of reference.

c) Gustavsson et al. (1986)

Gustavsson et al. (1986) reported the results of a study of the mortality and cancer incidence among a cohort of 142 male Swedish capacitor manufacturing workers during the period of 1965 to 1982 (with cancer incidence followed through 1980). The workers had been employed for a period of at least six months between 1965 and 1978 and had been exposed to Aroclor 1242 (or equivalent). Airborne PCB levels were measured at 0.1 mg/m^3 in 1973, with possibly higher levels in the 1960's.

A total of seven cancer deaths were identified in the cohort, which was not significantly different from the expected number (5.4), calculated using national statistics. There was also no tendency towards an increase in the mortality or cancer incidence in the most highly exposed subgroup of 19 workers. Although the results indicate no increase in cancer mortality in the study cohort during the study period, the results are not conclusive because of the small cohort size and brief follow-up period.

d) Bahn et al. 1976

Bahn et al. (1976) reported a statistically significant ($p < 0.001$) increase in deaths due to malignant melanoma (2 observed vs. 0.04 expected) in a small group (31) of research and development employees believed to have been heavily exposed to PCBs. The major pathways of exposure were not

identified by the authors. The workers were exposed to Aroclor 1254, among other chemicals, during various periods between 1949 and 1957. Although the authors suggest that PCB exposure may account for the observed excess of malignant melanoma, the small size of the study cohort and the fact that individuals were exposed to other toxic and potentially carcinogenic compounds during their employment makes it impossible to attribute the excess cancer cases to any specific agent.

Bahn et al. reported their findings in the form of a letter. They have never been presented in the form of an epidemiological study with data that can be independently evaluated and published in a journal for peer review. Therefore, these findings are difficult to evaluate as part of the "weight of evidence" regarding the carcinogenicity of PCBs in humans.

A letter by Lawrence (1977) questioned whether the study demonstrated any adverse effects from exposure to PCBs due to concommitant exposure of workers to other, possibly carcinogenic, chemicals. In response, Bahn et al. (1977) maintained the assertion of a "possible association" between PCBs and malignant melanoma, but agreed that the data were not conclusive.

e) Studies in Populations Following the Accidental Ingestion of PCBs, PCDFs, and Other Contaminants

Kuratsune et al. (1986) reported on the results of mortality studies of Japanese "Yusho" patients who had ingested contaminated rice oil in 1968. The oil was contaminated with Kanechlor 400 (similar in PCB composition to Aroclor 1248) as well as polychlorinated dibenzofurans (PCDFs) and polychlorinated quaterphenyls (PCQs). The

composition of the Kanechlor 400 involved in this incident had been altered (i.e., there was a much higher concentration of contaminants than the commercial grade mixture) as a result of its use in a heat exchanger. It is also probable that additional contaminants were generated during the use of rice oil in cooking.

In the 887 males who were included in the cohort, statistically significant increases in mortality from all malignancies (33 observed vs. 15.5 expected), liver cancer (9 observed vs. 1.6 expected), and lung cancer (8 observed vs. 2.5 expected) were reported, based on national rates. The use of local rates decreased the SMR for liver cancer from 560 to 390, which was still statistically significant. No SMR based on local mortality rates was calculated for lung cancer. No significant excesses in cancer mortality were observed for the 874 female patients included in the cohort.

There is evidence that confounding factors could have influenced the findings of Kuratsune et al. (1986). It has been reported that 70% of the identified Yusho patients are from two prefectures that have reported the highest incidence of liver cancers in Japan (Kuratsune 1986), suggesting the possible existence of local factors that have not been identified. For example, as reported in Ikeda et al. (1986), the rate of mortality from liver cancer was substantially different for the Yusho patients in Fukuoka prefecture than in Nagasaki prefecture. This led the authors to conclude that "Such a remarkably uneven geographic distribution of livear [sic] cancer deaths makes it hard to consider the observed increased risk of liver cancer as simply due to the poisoning." Kuratsune et al. (1986) were also unable to control for possible confounding

factors such as smoking habits (especially important with regard to the observed excess of lung cancers), drinking habits, and occupational exposures.

The liver cancer diagnoses on which Kuratsune et al. rely were obtained from death certificates, without any confirmation of pathology through tissue examination. The cases were thus not restricted to primary liver cancers, but also would have included cases in which the liver was a site of metastasis for cancers originating at other sites. The significance of the elevated incidence of liver cancer is thus subject to question.

Many investigators also believe that exposure to PCDF congeners is the primary cause of the symptom pattern observed in Yusho (Miyata et al. 1985, Kashimoto et al. 1985, Masuda 1985). They support this contention by reference to the high degree of toxicity (primarily to the liver) of certain of the PCDF congeners in laboratory animals. These toxic PCDF congeners (including 2,3,7,8-tetrachlorinated; 2,3,4,7,8-pentachlorinated; and 1,2,3,4,7,8-hexachlorinated dibenzofuran isomers) were identified in Yusho oil and in the tissues of Yusho victims (Miyata et al. 1985). As further evidence that PCDFs were the agents most likely responsible for the severity of Yusho symptoms, Kashimoto et al. (1985) and Hara (1985) refer to the relatively mild symptoms observed in PCB-exposed workers who had serum PCB levels similar to those observed in Yusho victims, but without detectable levels of PCDFs.

A second major outbreak of disease caused by ingestion of contaminated rice oil (called Yu-Cheng in Chinese) occurred in central western Taiwan in 1979. The oil that was responsible for this incident contained PCBs, PCDFs, and PCQs that were comprised of congeners similar to those identified in Yusho

specimens (Miyata et al. 1985). No data were located regarding the incidence of cancer mortality in Yu-Cheng victims. It is possible that there has been an insufficient number of deaths in this group for any meaningful analysis of mortality data.

3. Conclusions Regarding Epidemiology Data

Although several studies have investigated the possibility of an association between PCB exposure and human cancer, the results do not support a causal relationship. The primary reasons for this conclusion are:

(i) Strength of the Association: When the excess cancer cases observed by Bertazzi et al. (1987) and Brown (1987) are examined closely, the relationship of the excess cancers to PCB exposure appears doubtful. For example, 2 of the 6 cases of digestive system cancer that were identified in the male subcohort by Bertazzi et al. were in individuals whose jobs involved little or no direct PCB exposure, and a third case was in a worker who began employment at an advanced age. Furthermore, any perceived linkage between any chronic effect and employment is dubious because the cohort includes individuals with only one week of employment. None of the 5 excess liver, gallbladder, or biliary tract cancers observed by Brown (1987) was identified as primary liver cancer, thus rendering suspect the identification of the liver as the target organ. The findings of statistically significant increases in liver cancer among male Yusho victims (Kuratsune et al. 1986) cannot be attributed to PCBs because of concurrent

exposure to high concentrations of other toxic contaminants (e.g., PCDFs).

(ii) Dose-Response Relationship: In studies in which excess cancers were observed, there is no relationship between degree of PCB exposure and cancer risk. For example, in the follow-up study by Brown (1987), 4 of the 5 excess liver or biliary tract cancer cases were observed in the lowest exposure group, with none in the highest exposure category. Bertazzi et al. (1987) were also unable to identify a dose-response relationship between PCB exposure and increased cancer risk.

(iii) Consistency and Specificity of the Association: There is no consistent pattern of associations among the various studies, either with respect to the type of human cancers observed or the nature and extent of PCB exposures.

(iv) Temporally Correct Association: For some of the cases identified by Bertazzi et al. (1987), it appeared that there was little or no opportunity for exposure before development of disease. Also, no pattern of increased risk with an increase in latency was reported by Brown or Bertazzi et al.

(v) Coherence With Existing Information: Experimental data do not suggest that PCBs are a causative agent for cancer in mammals at sites other than the liver. The evidence that PCBs are causative agents for liver cancer in humans is inadequate. A statistically significant increase in mortality from cancer

of the liver, gallbladder, or biliary tract (combined) was observed in one occupationally exposed cohort (Brown 1987); however, none of these cases were identified as primary liver cancer. There was no confirmation, by tissue analysis, of the Yusho liver cancer victims identified by Kuratsune et al. (1986). These cases were also not restricted to primary liver cancers.

There is insufficient evidence to show a causal relationship between PCB exposure and the subsequent development of any form of cancer. In light of the long-term and widespread usage of PCBs in the workplace and, in some cases, the extensive exposures of workers, it is likely that evidence of carcinogenicity in humans would have been observed in the various epidemiological studies discussed above if PCBs were in fact potent carcinogens.

D. Animal Studies

1. Introduction

The numerous human studies are insufficient to show that PCBs cause cancer in humans. When data from human exposure are inadequate to assess the potential hazards from a substance, experiments with laboratory animals are often performed to identify potential adverse effects that might occur in humans. While animal studies have been accepted as a general indicator of possible effects in humans, not all effects observed in all animals will occur in humans. A chemical-specific evaluation may indicate the data from animals is inappropriate, especially when the effects are observed in only one species of animal and cannot be duplicated in other species. For this reason, consistent results from studies in several species are required to justify convincingly that it is proper to

extrapolate results of studies of laboratory animals to humans.

Although laboratory animal testing is, in general, a useful tool for predicting the impact of an agent on humans, the limitations of these tests must be acknowledged. For some chemicals or chemical-specific effects (e.g., tumor forming potential), there can be considerable uncertainty with regard to the applicability of test results in predicting human response. The most obvious and important reason for this is the fact that such animals are physiologically different than the human species. No matter how convincing the results from animal studies, a question always remains about their relevance to human populations because of interspecies differences in factors such as absorption, metabolism, and elimination of a test substance.[1/] In addition, some types of tumor responses (e.g., the rodent liver tumors that are the only clear animal response produced by any of the PCBs) are much less certain predictors of human cancer than are other types of tumor responses.[2/] Finally, all studies must be critically evaluated with respect to the quality of test designs and conduct.

Although there are limitations associated with animal tests, such studies are frequently used for regulating

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1/For example, metabolic differences may undermine the validity of extrapolating from animals to man if the carcinogen is a metabolite of the original chemical and the animals used in the bioassay differ substantially from humans in their production of that metabolite.

2/There is a high and variable incidence of liver tumors in various strains of commonly used laboratory mice, as well as a high spontaneous incidence in the livers of rats of preneoplastic cells (i.e., cells in an altered state that may have carcinogenic potential) that can be stimulated by promoting agents to produce tumors (Nutrition Foundation 1983, Schulte-Herman et al. 1983).

environmental carcinogens. The use of animal test data in this fashion, even when available human data do not suggest a problem (as in the case of PCBs), is based on the policy goal of regulators of providing maximum assurance of public health protection in the absence of complete scientific certainty.

For purposes of this report, the animal data on PCBs will be used to determine potential health risk because data from human exposures show no demonstrable health effects other than chloracne. Therefore, the animal data are presented on the most sensitive endpoint of concern with respect to PCBs: rodent tumorigenicity. In choosing this endpoint as the most sensitive, the conservative assumption is made (as it is by regulatory agencies) that all of the different commercial PCB mixtures are tumorigenic, even though this has not been demonstrated in laboratory or epidemiological studies.

There are effects other than tumorigenicity that have been observed in animals exposed to PCBs at relatively low exposure levels. These include reproductive effects such as reduced birth weight and hyperactivity in the offspring of exposed monkeys (Barsotti and Van Miller 1984, Bowman et al. 1981) and altered menstrual cycles in exposed monkeys (Allen et al. 1979), as well as induction of hepatic microsomal enzymes (enzymes produced by liver cells) in rats (Litterst et al. 1972). Tumorigenicity, however, is the most sensitive endpoint for low-level, environmental exposures to PCBs. Therefore, protection of public health based on tumorigenic risk is protective of adverse effects for other sensitive potential endpoints, such as reproductive effects. Other PCB-related effects would have to be considered if we were concerned with short term, high-level exposure to PCBs.

2. Animal Studies Regarding the Tumorigenicity of Commercial PCBs

The PCB mixtures that have thus far been tested in acceptable animal chronic bioassays for tumorigenicity

include Aroclor 1260, Clophen A60, Aroclor 1254, and Clophen A30. Each of these bioassays is discussed below.

There are no acceptable bioassays concerning the carcinogenicity of Aroclors 1248 or 1242. While there are some animal studies on Aroclor 1242; Aroclor 1248; and Kanechlors 300, 400, and 500 (Japanese commercial mixtures), which qualitatively add to the body of knowledge concerning the potential tumorigenicity of PCBs, these studies are not conclusive and cannot be relied upon for quantitative determinations. This is primarily because of inadequacies in the design (e.g., insufficiencies in study length, numbers of test animals, dose levels tested) of the studies that have been conducted to date. Cancer bioassays conducted by Industrial Bio-Test Laboratories (IBT) have generally been considered invalid by regulatory agencies (cf. Garmon 1981); therefore, this series of chronic animal studies on Aroclors 1242, 1254, and 1260 will not be used for this quantitative analysis.[1/]

In light of the limited number of studies, the cancer potency factors for the various Aroclor mixtures must be derived from the sets of animal data for Aroclor 1260, Clophen A60, Aroclor 1254, and Clophen A30. The importance of the difference in tumorigenic potency among the congeners, as well as the procedure used for adjusting these data for use in risk assessments, is discussed later.

1/Both the FDA and EPA consider these studies to be invalid because of severe procedural and record-keeping deficiencies. Also, the results of a re-evaluation of the original data by Calandra (1976) used terminology that does not conform to current practice for diagnosing hepatocellular proliferative lesions (tissue in which liver cells are dividing at an abnormally fast rate).

The analysis presented in this report follows the current regulatory practice of treating PCBs as complete carcinogens[1/]. Several factors, however, demonstrate that some of the carcinogenic effects of PCBs are due to promotion rather than initiation:

1. A substantial number of experiments have shown that PCBs do not cause direct genetic effects in several assay systems (e.g., as reviewed by ATSDR 1988).

2. PCBs have been shown to cause the promotion of liver tumors in rodents initiated by other compounds (e.g., Kimura et al. 1976; Nishizumi 1976, 1979; Tatematsu et al. 1979; Preston et al. 1981).

Other compounds that act as promoters have threshold doses (e.g., a dose below which no effect is observed) that have been demonstrated experimentally (e.g., as reviewed in Butterworth and Slaga 1987 and Schulte-Hermann 1985). It is believed that a threshold exists for all chemicals that act solely as promoters.

These points have important implications for carcinogenic risks from exposure to PCBs at very low dosages, such as might arise from environmental

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[1/]The process of carcinogenesis is generally regarded as a multistage process. It is considered to consist of, at a minimum, an initial stage in which the genetic material of a cell is permanently altered (initiation) followed by later stages (that may occur many years later) in which the initiated cell undergoes changes which are not fully understood, but which include cell division (promotion). A complete carcinogen is a substance which acts as both an initiator and a promoter in that it can, by itself, cause an increase in tumor formation.

exposures. If the tumorigenic effects of PCBs in laboratory animals are solely or primarily due to promotion, the potential tumorigenic risk will be greatly overstated at very low dosages. Studies with other promoters indicate that the carcinogenic effects of promoters are, at least to some degree, reversible and that a threshold exposure level must be exceeded to produce any effect on carcinogenesis. Thus, the no-threshold, linearized multistage model, which assumes that any level of exposure has some risk, will overstate the risk, especially at low doses (exposures). If PCBs were solely promoters, no tumorigenic risk whatsoever would be expected from doses (exposures) that are below the threshold. It is thus very possible, even if the animal data are reliable indicators of effects in humans at high doses, that no risk would result from low-dose environmental exposures to humans.

a) Aroclor 1260

There are two cancer bioassays of Aroclor 1260: Norback and Weltman (1985) and Kimbrough et al. (1975).

Norback and Weltman initially exposed 70 male and 70 female Sprague-Dawley rats to dietary concentrations of 100 ppm Aroclor 1260 for 16 months, 50 ppm for 8 subsequent months, and control diets for 5 months. The control group consisted of 63 male and 63 female control rats. At months 1, 3, 6, 9, 12, 15, and 18, four controls and six PCB-treated rats had partial hepatectomies (removal of the liver) in order to observe sequential morphological changes and progression to neoplasms[1/]. One set of rats was

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[1/]The term neoplasm refers to a new and abnormal formation of tissue, which can be in the form of a tumor. A neoplasm may be benign (not spreading into surrounding tissues) or malignant (i.e., cancerous).

sacrificed at 24 months; at 29 months, terminal sacrifices on all remaining rats were completed. Sequential observations showed that an increase in the size of some liver cells (centrilobular cell hypertrophy) was present at 1 month; small organized regions of changes in liver cells (foci of hepatocyte alterations) were seen at 3 months; larger areas of liver cell (hepatocyte) alterations were observed after 6 months; benign, i.e., non-cancerous, tumors (neoplastic nodules) appeared at 12 months and malignant tumors (trabecular carcinoma and adenocarcinoma)[1/] were apparent later (after 15 and 24 months, respectively).

The total incidence in Norback and Weltman of trabecular carcinoma was 23% (21/93) with 2/46 and 19/47 in males and females, respectively. Adenocarcinoma appeared at an incidence of 26% (24/93) of which 24/47 occurred in females and 0/46 in males. Neoplastic nodules were observed in 8% (7/93) of the Aroclor 1260 animals (5 males and 2 females). Neoplastic nodules were observed in one female control animal, resulting in a total incidence of 1% (1/81) for neoplastic nodules in controls. No other hepatocellular neoplasms (liver tumors) occurred in the control group. Bile duct hyperplasia (excessive proliferation of normal cells), cysts, and adenofibrosis (benign tumor containing connective tissue) were seen in 38%, 8%, and 9% of the treated animals, and 5%, 1%, and 4% of the control animals, respectively. Although hepatocellular neoplasms were present in 96% of the treated females and 15% of the

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1/Adenocarcinoma refers to a malignant tumor arising from glandular tissue (in this case the liver); trabecular carcinoma refers to a specific type of liver cancer.

treated males, the neoplasms did not metastasize or cause increased mortality relative to controls.

An analysis of the data using Fisher's Exact Test shows the incidence of carcinoma in the females was statistically significantly greater than control females; however, this was not so for males. The incidence of total liver tumors (carcinomas and neoplastic nodules) in males and females was statistically significantly greater than their respective control groups.

Kimbrough et al. (1975) also performed a rodent bioassay for Aroclor 1260. Initially, 200 female Sherman strain rats were fed 100 ppm of Aroclor 1260 for 21 months. Dietary exposure was discontinued for six weeks before all exposed animals were sacrificed. The initial control group consisted of 200 female rats. Malignant tumors (hepatocellular carcinomas) were observed in 26 of the 184 surviving PCB-exposed rats, and benign (non-cancerous) tumors (neoplastic nodules) of the liver were observed in an additional 144 of the exposed rats. Only 1 of the 173 surviving control animals developed hepatocellular carcinoma while none of the control rats developed neoplastic nodules. Analysis of this data using Fisher's Exact Test shows the incidence of carcinoma in exposed rats was statistically significantly greater than in controls.

Kimbrough et al. (1975) also reported the incidence of tumors in organs other than the liver. A number of organ sites showed lower tumor incidence in PCB-treated animals than in the controls. If the total number of tumors at all sites is summed, however, the lower incidence of certain tumor types in the PCB-treated animals as compared to controls was more than counterbalanced by the increase in liver and other tumors compared to control animals.

b) Clophen A60

Clophen A60, the German commercial equivalent of Aroclor 1260, was tested in a cancer bioassay in Wistar rats by Schaeffer et al. (1984). Male Wistar rats received dietary concentrations of 100 ppm Clophen A60 over a period of 832 days. Malignant tumor (hepatocellular carcinoma) incidence in the treated group at 48% (61/126) was statistically significant compared to 0.76% (1/131) in controls. Benign tumors (neoplastic nodules) of the liver were also statistically significant at 49% (62/126) in the Clophen A60 group compared to 3.8% (5/131) in the control groups.

It is important to note, however, that there was a statistically significant lower survival in control animals compared to the Clophen A60 group, i.e., the animals that were exposed to PCBs tended to live longer than animals that were not exposed (controls). This lower survival of the control animals may have led to a lower tumor incidence in controls than might have been seen if survival among the controls had been equivalent to that of the exposed animals, because tumor incidence generally increases with age. When comparing animals with the same length of survival, however, there is still a statistically significant increase in liver tumors in the Clophen-exposed animals versus controls.

c) Aroclor 1254

The National Cancer Institute (NCI 1978) and Kimbrough and Linder (1974) conducted bioassays on the carcinogenicity of Aroclor 1254 in rats and mice, respectively.

The NCI study protocol consisted of 24 male and 24 female Fischer 344 rats that received diets containing either 0, 25, 50, or 100 ppm Aroclor 1254

for 104 to 105 weeks. There was a small, dose-related increase in the incidence of combined benign (adenoma) and malignant (carcinoma) tumors. There was a larger increase in the incidence of nodular hyperplasia.[1/] Although the occurrence of the liver lesions in these rats was not statistically significant, none of the benign or malignant changes in the liver (including hyperplastic nodules, adenomas, or carcinomas) were observed in control animals. Additionally, four adenocarcinomas and one carcinoma of the gastrointestinal tract observed in treated rats may have been treatment related, according to NCI, because the historical incidence of these tumors in this laboratory is only 6/600 in males and 2/600 in females. In this bioassay, however, few sections of the stomach had been evaluated. NCI (1978) concluded that the high incidence of hepatocellular proliferative lesions in male and female rats were related to treatment, but that Aroclor 1254 was not carcinogenic in this bioassay.

A re-evaluation of the NCI data by Morgan et al. (1981), which focused only on the tumors of the gastrointestinal tract, revealed greater numbers of stomach tumors than originally reported. These tumors were not statistically significantly greater than controls and did not appear to be dose-related. When compared with the incidence of historical controls (includes all control animals of this strain from past studies), the total incidence of adenocarcinomas of the stomach in all dose groups

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[1/] Hyperplasia is the condition in which normal appearing cells are proliferating at an excessive rate. A nodule is a small aggregation of these cells.

combined (6/144) was significant. It is important to note, however, that the historical controls may not have been examined in a manner as sensitive to detecting tumors as that used by Morgan et al.

In a subsequent paper, Ward (1985) reported the results of the same re-evaluation by Morgan et al. including data concerning proliferative lesions of the liver, as well as the glandular stomach. This re-evaluation showed a statistically significant increase in benign (i.e., non-cancerous) tumors (hepatocellular adenomas) in male rats exposed to 100 ppm Aroclor 1254 compared to controls. The original NCI bioassay had only reported one hepatocellular adenoma in high-dose males; Ward reported seven adenomas. Ward also showed a dose-related trend in hepatocellular adenomas. This difference may be due to a disagreement in pathological evaluations of tissues.

In a study by Kimbrough and Linder (1974), 9/22 (41%) male BALB/cJ mice fed 300 ppm Aroclor 1254 for 11 months developed tumors of the liver (hepatomas). A similar group receiving the treated diet for only 6 months, followed by control diet for 5 months only had a 4% (1/24) incidence of hepatomas. No hepatomas were observed in 58 control mice. Additionally, all PCB-treated mice had enlarged livers and adenofibrosis of the liver. A major limitation of the study was the high early mortality with subsequent autolysis (tissue degeneration following the death of an animal), thereby eliminating over 50% of the original mice from the final results.

In sum, Aroclor 1254 has not been shown to be carcinogenic in animal studies. There is some evidence that there was a treatment-related increase in non-cancerous changes (hepatocellular proliferative lesions) in rats in the NCI (1978)

study; however, the response was weak (and not statistically significant). No conclusions can be drawn from the re-evaluation of these data by Ward (1985) and Morgan et al. (1981) because of uncertainties associated with their analyses. The findings of Kimbrough and Linder (1974) suggest a treatment-related increase in benign liver tumors in mice treated with Aroclor 1254. The interpretation of these results are limited by previously noted study inadequacies.

d) Aroclors 1248, 1242, and 1232

At present, there are no studies concerning the tumorigenicity of Aroclors 1248, 1242, or 1232 from which reliable carcinogenic potency factors could be derived. Mammalian carcinogenicity bioassays of acceptable quality (e.g., sufficient duration, number of test animals, and test doses) have not been conducted on these Aroclors. Moreover, these PCB mixtures may not be of sufficient tumorigenic potency to cause an observable increase in tumor incidence when tested in a standard rodent bioassay. There are, however, two primate studies (one on Aroclor 1248 and the other on Aroclor 1242) that describe modifications and lesions of the gastric mucosa. Although these studies do not show tumorigenicity as an endpoint, they may be qualitatively significant in light of the stomach adenocarcinomas observed in Aroclor 1254-exposed rats (NCI 1978, Morgan et al. 1981, Ward 1985).

Two studies, Allen et al. (1973) and Becker et al. (1979), reported PCB-induced changes in the stomach, but no increase in stomach tumors. The severity of the effect was correlated with the duration and level of exposure and was observed at relatively low concentrations (0.12 mg/kg/day in the

Becker et al. study). Neither of these studies was designed to examine carcinogenesis nor can they be used for cancer potency estimation of Aroclor 1248 or Aroclor 1242. When considered along with the results of the NCI bioassay, these results suggest that the stomach cannot be discounted as a potential target organ for PCB. It is important to note, however, that there has been no reported incidence of stomach tumors in bioassays of Aroclor 1260, Clophen A60, or Clophen A30. Further, the incidence of stomach tumors in Aroclor 1254-exposed animals was not significantly greater than in controls, was not dose related, and was so low that even if stomach tumors were considered, they would have no effect on the tumorigenic potency estimates derived in this document.

e) Clophen A30

Clophen A30, a German commercial PCB mixture similar to Aroclor 1242, was also tested in the previously-cited study by Schaeffer et al. (1984). Male Wistar rats received dietary concentrations of 100 ppm Clophen A30 over a period of 832 days. Liver cancer (hepatocellular carcinoma) incidence in the treated group was 3% (4/130), while in the control group the incidence of hepatocellular carcinoma was 0.76% (1/131). Non-cancerous tumors (neoplastic nodules) of the liver were 29% (38/130) and 3.8% (5/131) in the Clophen A30 and control groups, respectively. The incidence of neoplastic nodules but not the incidence of hepatocellular carcinoma (malignant tumors), in the Clophen A30 group was statistically significantly increased compared to controls.

f) Kanechlors 300, 400, and 500

Three rodent studies of Kanechlors 300, 400, and 500 add qualitative support to the variation of tumorigenic potency among the PCB mixtures. These include rat studies by Kimura and Baba (1973) and Ito et al. (1974) and mouse studies by Ito et al. (1973).

Kimura and Baba (1973) exposed male and female Donryu rats to Kanechlor 400; initial exposure was 38.5 ppm in diet but was increased to the very high dose of 616 ppm to keep pace with body weight gain. When severe body weight loss was observed, the dose was reduced to 462 ppm. The total Kanechlor consumption in females ranged from 700 to 1,500 mg and in males from 450 to 1,800 mg. Non-cancerous (adenomatous) nodules were observed in 6/10 of the females consuming more than 1,200 mg of Kanechlor 400; no such lesions were observed in the males. EPA (1988) concluded that this study was too short and the exposure level too high (treated animals received doses exceeding the maximum tolerated dose) to provide a good experimental basis for the determination of the carcinogenic potential of Kanechlor 400.

In a second rat study, Ito et al. (1974) exposed male Wistar rats (via feed) to 100; 500; or 1,000 ppm of either Kanechlor 300, 400, or 500 for 28 weeks to one year. Nodular hyperplasia was observed in all of the Kanechlor 500 dose groups and in the 100 and 1,000 ppm dose groups exposed to Kanechlor 400 and Kanechlor 300. The incidence of this nodular hyperplasia increased with dose as well as with percent chlorine content. EPA (1988) stated that this study does not demonstrate tumorigenicity, but it cannot be considered evidence of non-tumorigenicity because of the short duration and small number of subjects per group limit the ability of the study to

detect tumorigenicity. EPA also concluded that the nodular hyperplasia, which appeared as early as 40 weeks, further precludes considering this study a negative finding. In addition, this study did not include female rats, which in light of the Kimura and Baba (1973) study results, may be more sensitive than males.

In a series of mouse studies by Ito et al. (1973), male mice were exposed to either Kanechlor 500, 400, or 300 in feed at concentrations of either 500, 250, or 100 ppm for 32 weeks. Although liver weight increase in all treatment groups was greater than controls, liver cancer (hepatocellular carcinomas) and increase in the number of liver cells (nodular hyperplasia) were induced in only the high dose (500 ppm) group exposed to Kanechlor 500. Forty-two percent (5/12) of the high-dose Kanechlor 500 group showed hepatocellular carcinomas, while 58% (7/12) showed hyperplastic nodules. Amyloid degeneration[1/] of the liver was observed in mice fed Kanechlor 500 or 400 at 250 ppm or 100 ppm, but not in the 500 ppm groups; however, according to the authors, the effects seen in the Kanechlor 500 group (nodular hyperplasia and hepatocellular carcinomas) could have masked any amyloid degeneration. Some mice fed Kanechlor 300 in the 500 ppm, 250 ppm, and 100 ppm dose groups also showed amyloid degeneration. None of the controls showed hepatocellular carcinomas, nodular hyperplasia, or amyloid degeneration. For evaluating carcinogenicity, interpretation of this study is

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[1/]This is a type of tissue or organ degeneration that is characterized by the deposition of a starchlike substance (amyloid) in the tissues.

limited by several factors including short study duration (52 weeks), lack of data on female mice, the small number of mice per dose group, and a lack of dose-response.

In conclusion, the Kanechlor data seem to indicate tumorigenic potential of these mixtures in rodents. The limitations of study design suggest that these data should not be used to derive a cancer potency factor; however, they do qualitatively support the liver as a site of action for PCBs in rodents.

g) Conclusions Regarding Animal Data on Tumorigenicity

There is clear evidence indicating that some of the highly chlorinated commercial PCB mixtures are tumorigenic in some animals. The responses are mostly limited to the livers in rats and mice, although there is a suggestion that some PCB mixtures may also affect the stomach of rats and monkeys.

There is uncertainty as to whether or not Aroclors 1248, 1242, and 1232 are tumorigenic in animals. Because there are no valid cancer bioassays for these mixtures, a comparison with other commercial PCB mixtures based on comparative composition is the only basis for evaluation. The best evidence for comparison comes from the study by Schaeffer et al. (1984) in which male rats were exposed to either Clophen A60 or Clophen A30. As previously explained, the Clophen A60 rats showed a 48% incidence of hepatocellular carcinoma, while the Clophen A30 rats showed only a 3% incidence of hepatocellular carcinoma that was not statistically significant. Although these results are not evidence for tumorigenicity for the lower-chlorinated Aroclors or Clophens, the data can be used to derive a

preliminary and conservative estimate of relative cancer potency. If we assume Clophen A60 parallels the cancer potency of Aroclor 1260 and Clophen A30 parallels that of Aroclor 1242, then we can conclude that the cancer potency of Aroclor 1242 is much lower (at least 16 times lower) than that of Aroclor 1260. The data for Aroclor 1254 qualitatively indicate an even lower potency than Aroclor 1260 than indicated by the Clophen data. These data, however, are not as well suited for use in quantitative estimation of cancer potency as the Clophen data because the data for Aroclor 1254 are from a different strain of rats than the data for Aroclor 1260.

It must be emphasized that reliance on the rodent liver-tumor data to estimate effects in humans may be conservative. As previously noted, the relevance of liver tumors in rodents to humans has been questioned because of the high and variable incidence of liver tumors in various strains of mice (e.g., Butler and Newberne 1975, Nutrition Foundation 1983, Clayson 1981) and the high spontaneous incidence in the livers of rats of preneoplastic cells that can be induced by promoting agents to produce tumors (e.g., Ogawa et al. 1981, Ward 1983, Schulte-Hermann et al. 1983).

Indeed, in a review of proliferative hepatocellular (liver) lesions of the rat, EPA (1986) has stated that, although neoplastic nodules are increased in animals receiving carcinogens and some neoplastic nodules may have "malignant potential," others may only be "hyperplastic" lesions and still others may regress following cessation of exposure. Thus, EPA (1986) stated that "the exact contribution of neoplastic nodules to the overall incidence of hepatocellular tumors in the rats is unclear at this

time." Nonetheless, despite the skepticism that must surround reliance on observations of hepatocellular tumors in rats as indicators of tumorigenic effects in humans, the standard regulatory practice is to assume these data are accurate predictors of carcinogenic potency in humans. This approach must be seen as possibly resulting in exaggeration of the hazards of PCBs.

E. Cancer Potency Differences Among PCB Congeners

1. Importance of Differences Among Mixtures of PCB Congeners

In light of the limitations in the available animal test data, a cautious approach would be to classify the various Aroclors as potential animal tumorigens. The considerations that lead to this position may be briefly summarized:

(i) There are no valid test data on "Aroclors" other than Aroclor 1260 and Aroclor 1254.

(ii) It is not clear which specific congeners are responsible for Aroclor 1260-induced tumorigenicity.[1/] Figure 2 reveals that all commercial PCB products have some congeners in common. Thus, it is possible that all Aroclors contain some tumorigenic congeners.

(iii) The tests of Aroclors 1260 and 1254 involved different strains of rats, and the different

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[1/] There are strong reasons to believe that substantial differences exist in the toxicity and tumorigenicity of various PCB congeners.

outcomes could possibly reflect differences in experimental design.

(iv) Clophen A30 produces excess benign tumors in the rat liver, thereby suggesting a tumorigenic response for a mixture of congeners similar to that associated with Aroclor 1242.

For the above reasons the possibility of animal tumorigenicity cannot be ruled out for Aroclors other than Aroclor 1260. Nevertheless, the available data reveal clear differences in tumorigenic potencies among these sets of congeners. (By "potency" we refer to the incidence of tumors, i.e., risk, associated with a specific PCB dose.) These potency estimates are critical to an evaluation of PCB risks and are discussed in the next section.

There is some evidence that, for certain noncancer endpoints, the biological activity of PCBs increases with increasing chlorine content (see section III.E.3 for discussion). Studies by Ito et al. (1973) and Koller (1977) have shown that the degree of liver cell proliferation and pathologic alterations is much higher in mice chronically exposed to commercial PCB mixtures of higher chlorine content than to those with lower chlorine content. Hepatic microsomal enzyme induction potency also increases with increasing chlorine content (Litterst et al. 1972). PCBs containing 54% or greater chlorine content appear to be the most potent at inducing these effects. The differences in the ability of various PCB mixtures to ellicit biological changes other than cancer may be important indicators of differences in tumorigenic potency as well. Some of these endpoints, such as liver cell proliferation, may be associated with cellular events that might affect the rate of tumor formation.

Thus, the observed differences in carcinogenic potency and in other biological effects among the PCB mixtures may be correlated, but a definitive causal relationship has not been established.

2. Relative Potencies of Aroclors

The most compelling evidence for potency differences among the commercial PCBs is derived from the studies of Clophen A60 and Clophen A30. These products were tested in experiments of identical design and yielded quite different outcomes. As previously shown, the incidence of hepatocellular carcinoma in the Clophen A60 rats was 16 times greater than the incidence in the Clophen A30 rats (Schaeffer et al. 1984). Combining the incidence of both hepatocellular carcinomas and neoplastic nodules yields a smaller difference in cancer potency between Clophen A60 and Clophen A30. Specifically, the tumorigenic potency of Clophen A30 proved to be at least 10 times less than that of Clophen A60. This comparison is based on the highly conservative assumption that the excess benign tumors observed in the Clophen A30 experiment should be given equal weight to the malignant tumors produced by Clophen A60. If the benign and malignant tumors are weighted differently, the potency difference is even greater.

Because of the strong chemical similarities between Clophen A60 and Aroclor 1260 and between Clophen A30 and Aroclor 1242, the data on the Clophens can be used to estimate the potency of (untested) Aroclor 1242 relative to that of Aroclor 1260. Specifically, we propose to assign a potency of 0.1 to Aroclor 1242 relative to 1.3 for Aroclor 1260 (see section III.F.2.b for discussion).

The potency difference observed for Clophens is supported by the results of the experiments involving Aroclors 1260 and 1254. Although the difference in potency between Aroclors 1260 and 1254 appears to be even greater than that between Clophens A60 and A30, it must be

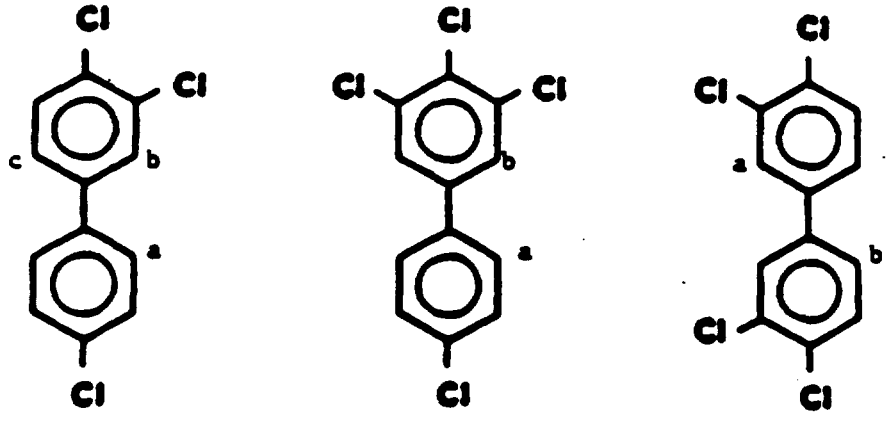
recognized that the two Aroclors were assayed in different rat strains, whereas the two Clophens were tested in identical experiments. The difference in potencies between Aroclors 1260 and 1254 nonetheless suggests that our reliance on the potency differential between the Clophens is highly conservative, i.e., highly overestimates the tumorigenic potency of the less chlorinated Aroclors.

3. Information on Mechanism of PCB-Induced Toxicity Supports Potency Differences

The role of structure on the potencies of PCB isomers and congeners has been extensively investigated (Safe 1984). The most potent compounds, namely 3,3',4,4'-tetra-; 3,3',4,4',5-penta-; and 3,3',4,4',5,5'-hexachlorobiphenyl are all coplanar (i.e., flat) in structure and bind with high affinity to the aryl hydrocarbon (Ah) receptor.[1/] These compounds, however, are either not detectable or are present in only trace levels in the lower chlorinated PCB mixtures.

Several studies have demonstrated that the responses caused by 11 monoortho analogs (i.e., specific congeners) of the coplanar PCBs resemble those described for the higher chlorinated commercial PCBs. This group of 11 congeners (see figure 3), although they are not coplanar, bind with low to moderate affinity to the Ah receptor but are much less potent than the coplanar PCBs. These 11

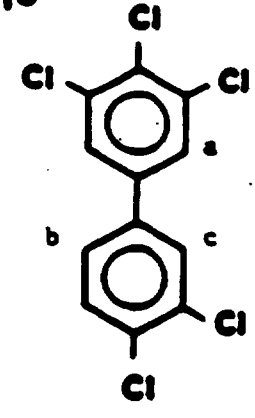
1/The Ah receptor is a protein molecule that binds a variety of chlorinated hydrocarbons such as PCBs, polychlorinated dibenzofurans, and polychlorinated dibenzo-p-dioxins. Experiments have shown: (1) the strength of the binding varies among the isomers of each of these classes of compounds and (2) inbred strains of animals that have high levels of this receptor are more sensitive to some of the toxic effects of these chemicals than strains that have low levels of the receptor.



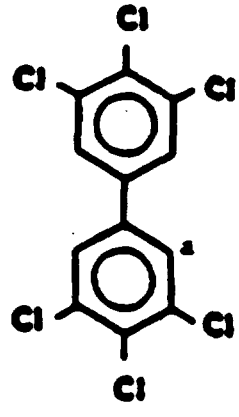
a: 2,3',4,4'
b: 2,3,4,4'
c: 2,4,4',5

a: 2',3,4,4',5
b: 2,3,4,4',5

a: 2,3,3',4,4'
b: 2,3',4,4',5



a: 2,3,3',4,4',5
b: 2,3',4,4',5,5'
c: 2,3,3',4,4',5'



a: 2,3,3',4,4',5,5'

Figure 3. Monoortho substituted tetra-, penta-, hexa-, and heptachlorobiphenyls. Letters a, b, and c are alternate positions for chlorine substitution. For example in the first structure, the molecule is 2,3',4,4'-tetrachlorobiphenyl if a chlorine atom is in the "a" position.

PCBs, however, have been identified in the commercial Aroclors and are a major contributor to the activity of these mixtures.

The tumorigenic potencies of individual PCBs have not been determined; however, mechanistic studies indicate that PCBs and related halogenated aryl hydrocarbons act as tumor promoters. Moreover, at least in the skin model for carcinogenesis using hairless mice,[2/] the observed structure-activity relationships confirm the role of the Ah receptor in this process. If one accepts the hypothesis that the mechanism of PCB tumorigenicity involves interaction with the Ah receptor, the structure-toxicity relationships that are also dependent on relative binding affinities for the Ah receptor protein can also be used to estimate tumorigenic potencies of individual PCBs and PCB mixtures.

F. Estimation of Potencies of PCB Mixtures

1. EPA Approach to Estimating Cancer Potency Factors

EPA (1986) has developed specific guidelines for risk assessment involving carcinogens. These guidelines require the derivation of a cancer potency factor (CPF) through the application of a mathematical model to extrapolate the observed dose-response data to very low doses at which humans are exposed (typically, hundreds of thousands of times lower than those used experimentally). It is not known whether the model is accurate; in fact,

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2/The skin of (genetically) hairless mice has been used as a model system for evaluating the potential of some chemicals to promote cancer. Usually, the cancer initiator is either injected or painted on the skin, followed by repeated applications of the suspected promoter. Appearance of skin lesions, including tumors, is recorded.

according to EPA, its use is designed to produce an upper limit on risks. The true risk, according to EPA (1986), is likely to be lower and could be zero.

EPA's CPFs are derived using the 95% upper bound of the slope of the linearized, multistage model for extrapolation to low doses. This model is based on certain assumptions about the action of carcinogens that may or may not be appropriate for PCBs. Furthermore, this model is one of the more conservative extrapolation models, i.e., it usually estimates a higher CPF than other models. Even though the analysis that follows is based on EPA's CPF, a further review of the scientific data may justify a different procedure for extrapolating to low doses.

EPA (1988) has calculated a CPF for PCBs of 7.7 $(mg/kg/day)^{-1}$ based on the Norback and Weltman (1985) study.[1/] Prior to this, EPA (1984) had determined the CPF for PCBs to be 4.34 $(mg/kg/day)^{-1}$ based on the Kimbrough et al. study. Both of EPA's CPFs are based on studies in which rats were exposed to Aroclor 1260, and the agency has suggested that this CPF should be used for all PCBs. EPA (1988) has published, however, a "preliminary calculation" indicating a CPF of 2.6 $(mg/kg/day)^{-1}$ for Aroclor 1254 based on the 1978 NCI bioassay data. EPA states that, although the Aroclor 1260 data are the best for estimating the cancer potency of PCBs as a whole class of compounds, it is appropriate to ask whether existing data on other PCB mixtures are adequate for making separate cancer potency estimates. Citing limitations in the data for calculating separate

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1/The units on CPF are "risk per unit dose", where dose is expressed in mg/kg body weight/day. Multiplying "risk per unit dose" by the "estimated lifetime average daily dose" (in units of mg/kg body weight/day) yields an upper-bound estimate of lifetime risk.

cancer potency estimates for each PCB mixture, EPA has made a policy choice to use the CPF from Aroclor 1260 to characterize the upper limits on risks for all other PCB mixtures. As discussed below, EPA's approach is not supported by the available scientific information.

2. Modification of EPA Approach Based on Relative Potency Adjustment

EPA's cancer potency estimate for PCBs should be modified in two significant ways: a) the EPA potency estimate (based on Aroclor 1260) should be changed to reflect the lower potencies of Aroclor 1254, Aroclor 1248, Aroclor 1242, and Aroclor 1232; and b) the interspecies extrapolation factor used by EPA should be changed from a dosage per surface area scaling factor (i.e., $mg/m^2/day$) to a dose per unit body weight scaling factor (i.e., mg/kg/day).

a) Interspecies Scaling

The interspecies scaling (i.e., extrapolation) of dose is necessary to compensate for differences between humans and laboratory animals for such factors as size, lifespan, and basal metabolic rate. The most commonly used measures of dose are milligrams of chemical per kilogram of body weight of the animal per day (mg/kg/day) and milligrams of chemical per square meter body surface area per day ($mg/m^2/day$). Debate over the choice of dosage unit has centered on the appropriate measure for body size (kg body weight or m^2 body surface area) and on the temporal descriptor (per day or per lifetime) (cf. Hoel et al. 1975, Crump et al. 1980, Food Safety Council 1980, Allen et al. 1987). For carcinogenic compounds, both scaling factors have been used in risk assessment by different federal agencies, and both scaling factors were considered valid when

reviewed by the Office of Science Technology and Policy (OSTP 1985). For example, the EPA uses $mg/m^2/day$ while the Food and Drug Administration uses mg/kg/day. (EPA recently published a notice in which, among other matters, it requested comments on whether to modify its approach.)

The use of $mg/m^2/day$ as a scaling factor tends to give higher risk estimates per unit of dose than does mg/kg/day. (Risk is presumed in the linear no-threshold model to be directly proportional to dose.) For example, in extrapolating from mouse to man, the use of $mg/m^2/day$ will result in a risk estimate (per unit of dose) that is approximately 12 times greater than the estimate obtained using mg/kg/day. In extrapolating from rat to man the risk estimate is approximately 7 times greater when surface area scaling ($mg/m^2/day$) is used as opposed to mg/kg/day.

There are a number of reasons why extrapolation should be undertaken on a body weight basis. First, consider the basis of the surface area scaling factor. Hoel et al. (1975) proposed the use of dosage units in $mg/m^2/day$ on the basis of studies of the acute toxicity of anticancer drugs in humans and animals. In these studies, the acutely toxic level was similar in mouse, rat, hamster, dog, monkey, and man when dosage was expressed as $mg/m^2/day$. This finding is not unexpected. In many cases toxic substance are detoxified by the metabolic processes of the organism. The body surface area of an animal is an indirect measure of the animal's basal metabolic rate. It is this relationship between body surface area and metabolic rate that explains the interspecies similarity in dosages when expressed on a surface area basis. But this relationship for acute toxic effects does not

necessarily apply for other effects. The relationship between dose and body surface area, a priori means very little when considering chronic effects such as cancer.

In contrast, the Scientific Committee of the Food Safety Council (1980) favored the use of body weight as the basis for extrapolation. The council explained that "with long experience of the value of extrapolation on body weight basis, we recognize this as the most satisfactory procedure." Crump et al. (1980) and Allen et al. (1987) determined, based on an analysis comparing the carcinogenic potency of 13 chemicals in humans and rodents, that the unit of dosage measurement giving the closest correlation between species was mg/kg/day.

A similar conclusion was reached by Crouch (1983), after examining a large data set on chemicals that had been tested for carcinogenicity in more than one species. Some of the chemicals in this data set had also been studied epidemiologically in humans. Crouch (1983) found that he could derive a range of scaling factors to extrapolate among species, strains, or sexes, but argued that a body-weight scaling factor value of 1 (i.e., mg/kg/day) should be chosen for general extrapolation from rodents to humans.

In the absence of good evidence for the use of a more complex procedure, we believe that the use of mg/kg/day is the most appropriate basis for interspecies dosage comparison. In addition to its relative simplicity, this procedure appears to have the best empirical support (Crump et al. 1980, Allen et al. 1987).

b) Potency Differences

Based on the above discussion of interspecies scaling factors and the earlier discussion of observed potency differences, it becomes possible to derive potency factors for each commercial PCB mixture. EPA derived a potency factor of 7.7 per mg/kg/day for Aroclor 1260 based on the study by Norback and Weltman (1985). In deriving a potency value for this report, we preserve EPA's conservative linearized multistage low-dose extrapolation model, but modify the interspecies scaling procedure by about six-fold,[1/] as discussed above. This leads to a potency factor of 1.3 per mg/kg/day for Aroclor 1260.

As noted earlier, Clophen A60 is at least ten times more potent than Clophen A30 in studies of identical design. Because Aroclor 1260 is similar in composition to Clophen A60 and Aroclor 1242 is similar to Clophen A30, it is reasonable to assume that Aroclor 1242 should exhibit a potency no more than one-tenth that of Aroclor 1260. We thus assign a potency to Aroclor 1242 of 0.13 per mg/kg/day. It should be noted, however, that there are no studies that show a statistically significant increase in tumors for any mixture of PCBs other than Aroclor 1260 and Clophen A60. The potency of the less chlorinated mixtures of PCBs may thus be appreciably less than our estimate.

A simple interpolation procedure can be used to assign potencies to the other Aroclors. The

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[1/] The potency value is based on a study in rats. EPA's estimate is 7.7 per mg/kg/day. We used actual body weight data from the Norback and Weltman study rather than the generic factor of 7.

procedure assumes that potency declines proportionally with chlorine content. This can be approached either through an analysis by percent chlorine content of a particular Aroclor type or through an analysis by average chlorine number by Aroclor type. For example, if potencies of 1.3 and 0.13 are assigned to Aroclor 1260 (60% chlorine) and Aroclor 1242 (42% chlorine), respectively, then potencies can be assigned to Aroclors 1254, 1248, and 1232 based on their respective percent chlorine: 54%, 48%, and 32%. Similarly, the potencies of Aroclor 1260 and Aroclor 1242 can be used as the basis for interpolation of potency factors for Aroclors 1254, 1248, and 1232 from the average number of chlorine atoms in each Aroclor.

Because commercial PCBs are complex hydrocarbon mixtures, which are not completely identical with respect to specific isomer content, chlorine number can vary within Aroclor type. In order to derive a potency value based on actual chlorine number, rather than percent chlorine, an average value for chlorine number per Aroclor type must be determined. Several approaches have been used to derive the average chlorine number: a probabilistic (pseudo-stochastic) approach in which chlorine number is calculated based on the relationship between percent chlorine and chlorine number in individual PCB congeners; an approach calculating the average chlorine number based on weight percentages of congeners in commercial PCBs; and an approach calculating the average chlorine number based on data present in an ENVIRON (1987) report listing amount (percent mass fraction) of specific congeners in Aroclors 1260, 1254, and 1242. Slight variations in the average chlorine number per Aroclor type are seen among these three different approaches. When these alternative

values of chlorine number are each used to derive a cancer potency estimate, however, virtually no difference in potency exists. Therefore, the values for cancer potency are virtually the same no matter which approach is used to derive average chlorine number.

The tumorigenic potency values derived for various Aroclors using a percent chlorine approach and an average chlorine number approach are presented in table 1. All potencies except the potency for Aroclor 1260 (that provides the basis for the other estimates) are rounded to one significant figure. It is evident that the cancer potency values derived using a percent chlorine approach are virtually identical to those using an average chlorine number approach. Because it is our hypothesis that the cancer potency of PCBs varies with chlorine number, it is probably more accurate to rely on the mean chlorine number.

It is important to note that the TPFs in table 1 have been developed by combining the incidence data for benign and malignant tumors. Some individuals have suggested that only malignant tumors should be used to estimate cancer potency. If the data on the incidence of animals with malignant liver tumors (carcinoma or adenocarcinoma) only, i.e., excluding neoplastic nodules, were used, the potency factor for Aroclor 1260 would be 0.98 per mg/kg/day. Based on an analysis of malignant tumors only, Clophen A60 is about 13 times more potent than Clophen A30. Hence, the potency of Aroclor 1242, which is similar in composition to Clophen A30, should exhibit a potency no more than one-thirteenth that of Aroclor 1260, if malignant tumors only are considered. This would reduce the potency value of Aroclor 1242 to 0.075 per mg/kg/day. These values are slightly lower than those estimated in table 1.

TABLE 1

Alternative Estimation of Tumorigenic Potency (TPF) Factors For Various PCB Mixtures -- Benign and Malignant Tumors Combined[1/]

PCB Mixture	% Chlorine	TPF $(mg/kg/d)^{-1}$ Based on % Chlorine	TPF $(mg/kg/d)^{-1}$ Based on Mean Chlorine Number
Aroclor 1260	60	1.3	1.3
Aroclor 1254	54	0.9	0.8
Aroclor 1248	48	0.5	0.4
Aroclor 1242	42	0.1	0.1
Aroclor 1232	32	0.08	0.07

1/Based on a comparision of data from Clophens A60 and A30, combining benign and malignant tumors as discussed in the text.

3. Uncertainties and Limitations

There are several important limitations that tend to contribute to the conservative nature of this analysis:

1. All environmental PCB mixtures are assumed to be potential animal tumorigens, even though data are available to support such a conclusion only for those closely resembling Aroclor 1260.

2. Those PCB mixtures that are known or potential animal tumorigens are assumed to have the potential to cause tumors in humans, notwithstanding the absence of evidence of a causal relationship from all available epidemiology data.

3. A linear, no-threshold, low-dose extrapolation model is used to estimate potencies for all PCBs, notwithstanding the fact that PCBs do not exhibit many of the characteristics of carcinogens for which such models were developed. Indeed, it is possible that there is a threshold dose that must be exceeded before PCBs could pose any cancer risk, whatsoever.

G. Conclusions

Although some workers have been exposed to high levels of PCBs for long periods of time, several studies of such populations have not provided information establishing that PCBs cause cancer in human beings. If PCBs were potent human carcinogens, it is likely that such an increase in cancers among these several worker populations would have been observed. Therefore, based solely on data from exposure of people, it is not possible to conclude that PCBs are carcinogenic to humans.

Of the PCB mixtures that have been tested in animals, those that are 60% chlorinated (i.e., Aroclor 1260 and Clophen A60) are carcinogenic in rats. Tests in other species have not been adequate to demonstrate or rule out carcinogenicity. PCB mixtures that are less chlorinated (i.e., Aroclor 1254, 54% chlorinated, and Clophen A30, 42% chlorinated) were not carcinogenic in rats, but the latter increased the incidence of non-malignant tumors. Two conclusions can be reached from these data:

1. Highly (60%) chlorinated PCBs are carcinogenic in one animal species.

2. Less chlorinated PCBs are either not carcinogenic or are substantially less potent than the more highly chlorinated mixtures.

Guided by these conclusions, it is possible to make several conservative hypotheses:

1. If 60% chlorinated PCBs are carcinogenic in one mammalian species, it is assumed they may be carcinogenic in others, including humans, i.e., Aroclor 1260 and Clophen A60 may be carcinogenic in humans. It must be noted that this is a conservative assumption, based on current regulatory practice.

2. Because there is some overlap in the congener composition of the commercially available PCB mixtures and because no data exist to determine unequivocally which of the congeners are responsible for the animal carcinogenicity, other mixtures of PCBs may be viewed as carcinogenic. Again, this is an assumption that is consistent with current regulatory practice.

3. If all PCBs are assumed to be tumorigenic in humans, the maximum potencies of less chlorinated PCBs can be estimated from the available bioassay data.

These hypotheses form the basis of the estimates of human carcinogenic potency that are derived in this report. As is discussed herein, however, these estimates constitute a conservative upper bound on cancer potency. In the case of PCBs, the carcinogenic risks to humans from environmental exposure are almost certainly less than our estimates and, in fact, could well be zero. EPA (1986) has also acknowledged the conservative nature of upper-bound cancer potency estimates.

E. REFERENCE MATERIALS FOR SECTION 2.0

1. Figures for Section 2.2

   a. Figure 2.2-1 — Conceptual Diagram of PCB Interactions in Sediment, Water, Air and Biota

   b. Figure 2.2.2.2.-1 — Henry's Constants for PCBs in relation to chlorine number.

2. Xu, Y.J. 1991. Transport Properties of Fine-Grained Sediments, Ph.D. dissertation Abstract, UCSB.

3. Gailini, J., C.K. Ziegler, W. Lick. 1991a. The Transport of Suspended Solids in the Lower Fox River, J. of Great Lakes Research, in press.

4. Ziegler, C.K., J. Lick and W. Lick. 1990. SEDZL: A User-Friendly Numerical Model for Determining the Transport and Fate of Fine-Grained, Cohesive Sediments, UCSB report.

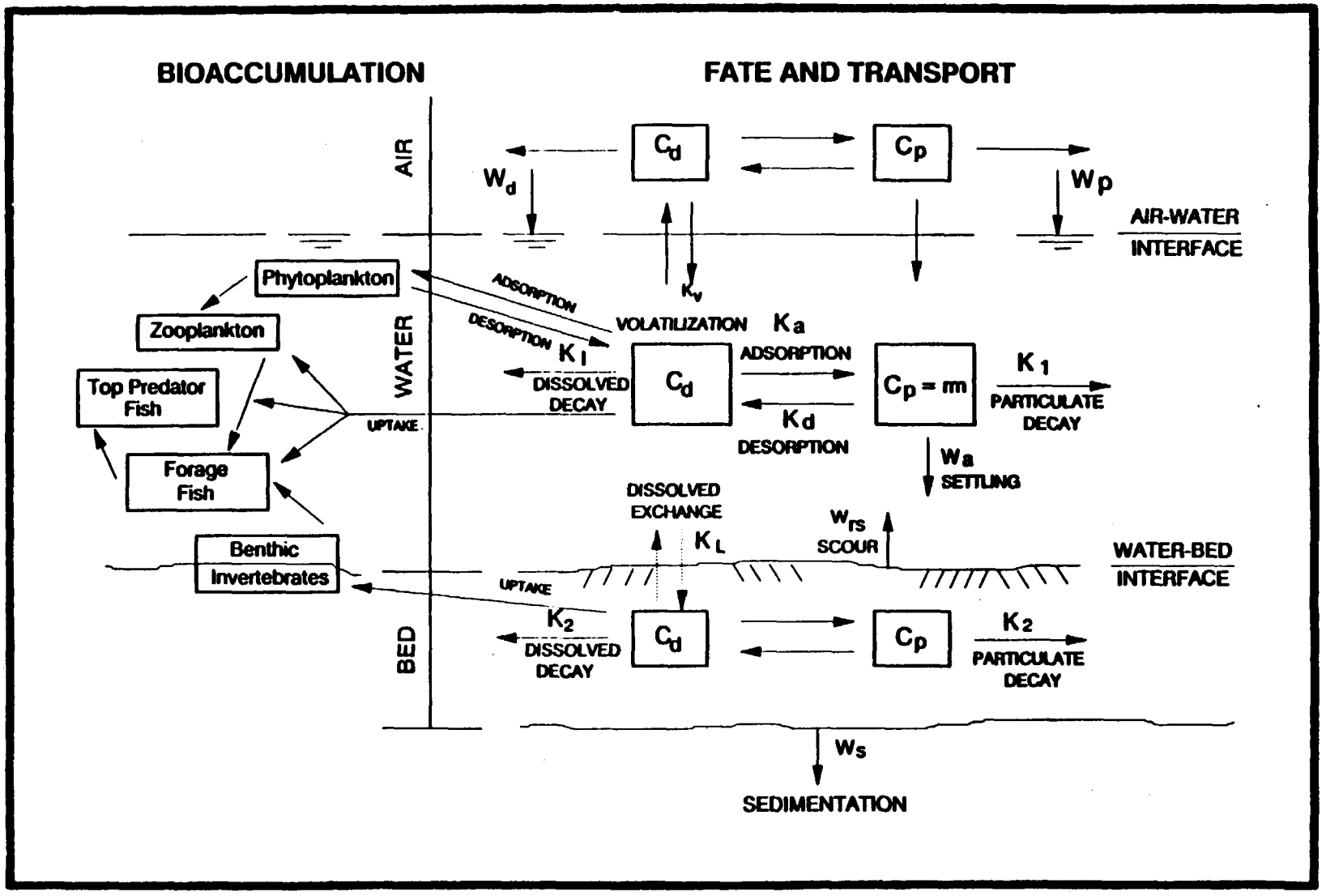
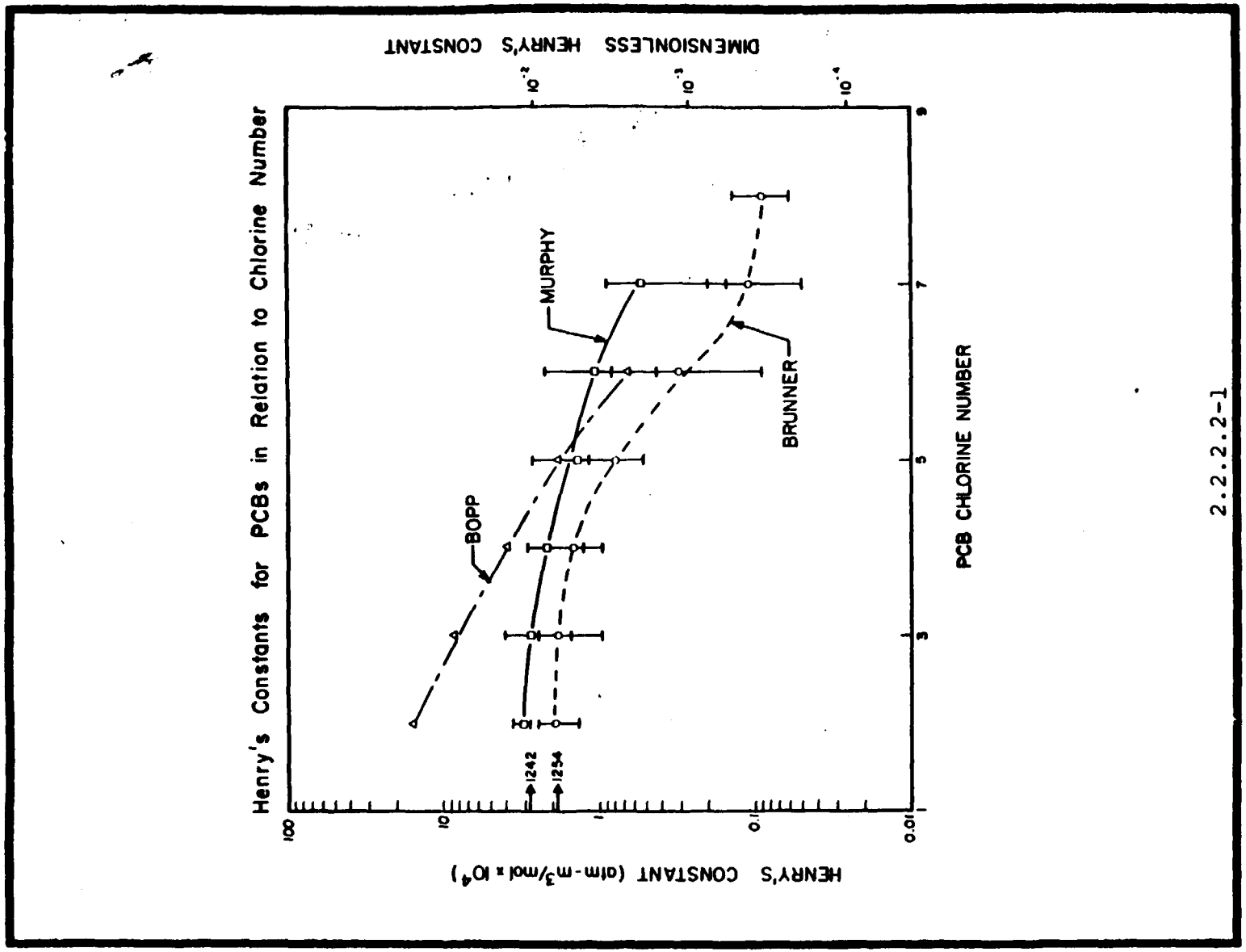


Figure 2.2-1

Conceptual Diagram of PCB Interactions
in Sediment, Water, Air, and Biota


Henry's Constants for PCBs in Relation to Chlorine Number
DIMENSIONLESS HENRY'S CONSTANT
HENRY'S CONSTANT (atm-m³/mol x 10⁴)
PCB CHLORINE NUMBER
MURPHY
BRUNNER
BOPP
1242
1254
100
10
1
0.1
0.01
3
5
7
9

2.2.2.2-1

UNIVERSITY OF CALIFORNIA

Santa Barbara

TRANSPORT PROPERTIES OF FINE-GRAINED SEDIMENTS

A dissertation submitted in partial satisfaction

of the requirements for the degree of

Doctor of Philosophy

in

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by

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January 1991

ABSTRACT

Transport Properties of Fine-Grained Sediments

by

Yao-Jun Xu

Resuspension, flocculation, and settling experiments were conducted on fine-grained sediments from the Fox River, Green Bay, and the Buffalo River, and on bentonite clay. The purpose was to quantitatively determine the transport properties of these sediments, the parameters on which these properties depend, and their variation within the aquatic system.

In annular flume experiments, the amount of sediment resuspended was quantitatively determined as a function of the fluid shear and bed compaction. It was shown that the higher the fluid shear and the shorter the consolidation time, the larger the amount of sediment resuspended. Field measurements with a shaker on undisturbed sediments were done to supplement the laboratory studies. Preliminary work on the influence of a current on the resuspension of sediment was also done. In this case, the amount of sediment resuspended was significantly greater than for sediments deposited in a quiescent body of water.

Flocculation experiments were done with a Couette viscometer. The dependence of median floc sizes on the fluid shear and the concentration were quantitatively obtained. The results have shown that the lower the fluid shear and the lower the concentration, the larger the floc diameters in the steady state and the longer the time required to reach steady state. The average density and the density function have been derived from the diameters

of flocs and the size distribution.

The settling speeds of flocs were measured in a rigid-mounted, thermally-isolated tube by microscopy. It was demonstrated that the settling speeds of flocs depend on the conditions of formation. The higher the fluid shear and the higher the concentration, the higher the settling speeds that the flocs formed in such conditions have. Larger flocs were observed to have greater settling speeds. The drag coefficients have been computed from the experimental data in different ways. It was found that Stokes' law underpredicts the drag coefficient significantly for large flocs.

This data is of importance in accurately predicting the transport of these sediments and the contaminants associated with these sediments. The data is presently being used in numerical models of sediment and contaminant transport in the Fox River, Green Bay, the Buffalo River, and as preliminary data for the transport of drilling muds from off-shore oil platforms.

THE TRANSPORT OF SUSPENDED SOLIDS IN THE LOWER FOX RIVER

by

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Running Head: Transport of Suspended Solids in the Fox River

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ABSTRACT

The general purpose of the present study was to study the transport of the sediments and associated contaminants in the lower Fox River and from the Fox into Green Bay. For this reason, a numerical model of the transport and fate of suspended solids in the lower Fox River has been developed and verified. The model consists of a two-dimensional, vertically-integrated, time-dependent hydrodynamic and transport model coupled with a three-dimensional, time-dependent model of the sediment bed and its properties. Settling speeds and sediment resuspension parameters needed in the model were determined from laboratory and field tests.

In the description of the transport of suspended solids, three components of solids are considered, i.e., fine (zero settling speed), medium (moderate settling speed affected by flocculation), and coarse (large settling speed). It is assumed that the sediment bed is layered in the vertical direction. The properties of each layer depend on time after deposition (and therefore with depth) and composition (relative fractions of medium and coarse sediments), and the number of layers and their thicknesses can be arbitrarily specified at the beginning of the calculation. The thickness of the surface layer changes with time depending on the rates of resuspension and deposition.

Calculations were made for steady flows at high, medium, and low flow rates as well as for real, time-varying flow events. In particular, two flow events were modeled in detail, the first from May 22, 1989 to June 20, 1989 (this included a once in a ten year high flow as well as moderate to low flows) and the second from March 24, 1989 to April 10, 1989. For these events, calculated sediment concentrations at the river mouth were compared with observations. Good agreement between the calculations and observations was obtained, thereby validating the model and the description of the physical processes implied in the modeling. In particular, the presence and effect of an easily resuspendable surficial layer was demonstrated.

Key words: Suspended Solids, Transport, Fox River

INTRODUCTION

The Fox River in Wisconsin is 56 km long and runs from Lake Winnebago in the south to Green Bay in the north. The valley through which the Fox runs is heavily industrialized and contains the largest concentration of pulp and paper industries in the world. Because of this, the waters and sediments in the Fox are heavily polluted. In the present study, the particular concern is with the lower Fox River which extends to Green Bay from a dam at DePere, 11 km upstream from the mouth of the river.

The purpose of this study was to develop a quantitative, predictive model of the resuspension, deposition, and transport of fine-grained sediments in the Fox River and especially the transport of these sediments from the river into Green Bay. Many contaminants, e.g., PCBs, are associated with fine-grained sediments, and it is the transport of these contaminants from the river into Green Bay which is of major concern. This transport is an essential component of the Green Bay/Fox River Mass Balance Project, a study of the sources, pathways, and sinks of toxics in a well-defined, medium-size, aquatic system. Three sessions at the 34th Great Lakes Conference were devoted to this topic, and the papers presented there should be consulted for general details on the project.

A bathymetric map of the lower Fox River (Figure 1) shows that the upstream portion (defined as upstream of the Ft. Howard Paper Company) is wide with many shallow pool-like areas less than two meters in depth. These areas are characterized by low water velocities and large areas of plant growth. This portion is no longer dredged, but previous dredging has established a channel up to five meters deep which is still present today. The depths used to develop Figure 1 were obtained from National Oceanographic and Atmospheric Administration (NOAA) maps and from U.S. Army Corp of Engineers dredging maps supplied by Robert Mundelius of the Kewaunee branch. The bottom sediments in this upstream portion are essentially all fine-grained, cohesive sediments, predominantly silts and clays. Measurements of the resuspension and transport properties of these sediments as well as those in the downstream portion of the river have been made

(Xu, 1991). From these studies, it can be shown that all sediments in the upstream portion have reasonably similar properties typical of fine-grained, cohesive sediments.

The river narrows near its midpoint at Fort Howard and remains narrow down to its mouth at Green Bay. The U.S. Army Corp of Engineers dredges this narrow portion as needed to allow large ship passage. Sediments in the deeper channels of this downstream area are generally coarse-grained and sandy. In the shallow, near-shore areas, the sediments are again fine-grained and cohesive. The only significant tributary to the lower Fox River is the East River which joins the Fox approximately 2 km upstream from the mouth. The junction of the two rivers has been widened and dredged to form a turning basin for large ships. The flow in the East River has been estimated to be approximately ten percent of the flow in the Fox River (a median flow of 105 m^3/s). Point sources along the lower Fox River, such as paper mills and sewage treatment plants, have been determined to add less than 3% to the total suspended solids load in the river (Mark Velleux of the Environmental Protection Agency, personal communication) and have therefore not been included in the calculations.

The flow in the Fox is controlled primarily by two dams upstream of the lower Fox. This flow can be increased so as to allow continued navigation in the river and to maintain water quality, or decreased so as to allow additional water storage in Lake Winnebago. Because of this control, extreme flow variations are now less than before control and less than in most other comparable rivers without control. A moderately high flow event can be caused either by a storm when the river is allowed to flow freely or by the opening of a sluice gate on one of the dams. Also, a large storm may not necessarily result in greatly increased flow rates if most of the water is retained behind the dams.

Fluctuations in the lake level in Green Bay have a significant impact on the flow velocity in the Fox River near the mouth. Increasing lake levels during low flow periods often reverse the direction of flow in the river, while falling lake levels, especially during high flow periods, may significantly increase the speed of the currents. As will be shown later, this has a significant effect on the resuspension and deposition of sediments in the lower Fox. Typical lake level changes are about twenty centimeters during a twelve hour

period, and changes of sixty-five centimeters during a twelve hour period will occur several times during a year. Hourly lake level data was supplied by Peter Hughes of the U.S. Geological Survey (USGS).

During 1989, daily concentration measurements were made at the DePere dam and at the river mouth by the Wisconsin Department of Natural Resources (DNR) and the USGS. An automatic sampler took samples three times a day at the mouth and four times a day at the dam to obtain averaged daily concentrations. The concentration data was supplied by Leo House of the USGS. The USGS also takes daily averaged flow rate measurements at the dam; this data was supplied by Barry Holmstrom of the USGS and Mark Velleux of the EPA.

A widely used numerical model for evaluating sediment transport processes in rivers has been HEC-6 (U.S. Army Corps of Engineers, 1977). This model has proven to be quite useful in a number of studies; however, it does have its limitations. First, HEC-6 is a one-dimensional hydrodynamic model so that lateral variations in the river flow cannot be resolved. This limitation is especially important when studying rivers such as the Fox. Secondly, HEC-6 does not model erosion of fine-grained, cohesive sediments; only deposition of clays and silts is allowed in the version of the model presently available. A new, unreleased version of HEC-6 accounts for the erosion of clays and silts (U.S. Army Corps of Engineers, 1990). The work of Partheniades (1965) and Ariathurai and Krone (1976) has been incorporated into this new, untested version of HEC-6.

The application of two-dimensional and three-dimensional models of cohesive sediment transport has been reviewed by the Task Committee of ASCE on Fine Sediment Transport Processes (1989). Various models were discussed with an emphasis on the STUDH model, which is a two-dimensional, vertically-integrated model employing the sediment dynamics of Ariathurai and Krone (1976). Reviewed applications were primarily limited to cohesive sediment transport in estuaries and coastal waters, with no presentation of studies in rivers. Application of a vertically-integrated, finite element model for cohesive sediment transport, CSTM-H, to simulation of sedimentation in a small yacht basin was

presented by Hayter and Mehta (1986).

Both of the sediment transport models mentioned above, STUDH and CSTM-H, do not adequately describe the resuspension of fine-grained, cohesive sediments and the variation of resuspension properties with depth. These models also do not account for the effects of flocculation on the deposition rate. A sediment transport model which does possess these attributes has been developed by us and has been applied to the Fox River using the data previously discussed for input and verification. For the hydrodynamics and sediment transport, a two-dimensional vertically-integrated, time-dependent model was used (Ziegler and Lick, 1986; Ziegler et al, 1989); this is briefly described in the following section. In order to describe the resuspension and deposition adequately, this model was then coupled with a three-dimensional, time-dependent model of the sediment bed and its properties. Settling speeds and sediment resuspension parameters needed in the model were determined from laboratory and field tests. The properties and dynamics of the sediment bed are described in the third section.

For a basic understanding of the hydrodynamics and sediment transport in the Fox, calculations were made for steady flows at high, medium, and low flow rates. In addition, for calibration and verification of the model and for further understanding, calculations of real, time-varying flows were made and also compared with observations. In particular, two flow events were modeled; the first was from May 22, 1989 to June 20, 1989 while the second was from March 24, 1989 to April 10, 1989. These calculations were made with the best estimates of sediment properties available (Xu, 1991), and generally good agreement between the calculations and observations was obtained, especially at moderate flows. However, at very high and low flows and during rapidly increasing flows, there were significant differences between the calculations and observations that indicated that the description of the resuspension/deposition processes and especially the parameters necessary for this description needed further refinements. After these refinements were made, excellent agreement between calculations and observations was obtained. The modifications to the model, the results of the calculations, and comparisons of the calculations with observations are discussed in the fourth section. A summary and

concluding remarks are presented in the final section.

Hydrodynamics and Sediment Transport

In order to simply describe the hydrodynamics and sediment transport in the river, vertically integrated equations of motion were used. These equations are a valid approximation in shallow waters when the horizontal velocities and suspended sediment concentrations are approximately independent of depth, i.e., when there is almost complete mixing of the water column in the vertical direction. For the Fox River, which is only 1 to 7 meters deep, this is almost always true except perhaps during a few low flow, highly stratified events. Continuous velocity readings for current meters located at the mouth at five and sixteen feet depths for 1989 and 1990 (Peter Hughes of the USGS in Madison, Wisconsin, personal communication) indicated that at almost all flow rates the velocities are independent of depth at the mouth.

The vertically integrated hydrodynamic equations are:

$$\frac{\partial \eta}{\partial t} + \frac{\partial U}{\partial x} + \frac{\partial V}{\partial y} = 0 \tag{1}$$

$$\frac{\partial U}{\partial t} + gh\frac{\partial \eta}{\partial x} = \tau_x^w - \tau_x^b + A_H\left[\frac{\partial}{\partial x}\left(\frac{\partial (U/h)}{\partial x}\right) + \frac{\partial}{\partial y}\left(\frac{\partial (U/h)}{\partial y}\right)\right] - \frac{\partial (U^2/h)}{\partial x} - \frac{\partial (UV/h)}{\partial y} \tag{2}$$

$$\frac{\partial V}{\partial t} + gh\frac{\partial \eta}{\partial y} = \tau_y^w - \tau_y^b + A_H\left[\frac{\partial}{\partial x}\left(\frac{\partial (V/h)}{\partial x}\right) + \frac{\partial}{\partial y}\left(\frac{\partial (V/h)}{\partial y}\right)\right] - \frac{\partial (UV/h)}{\partial x} - \frac{\partial (V^2/h)}{\partial y} \tag{3}$$

where the total water depth is $h = h_o + \eta$, $h_o(x,y)$ is the equilibrium water depth, $\eta(x,y)$ is the surface displacement from that equilibrium, U and V are vertically integrated velocities defined by $U = \int_{-h_o}^{\eta} u\,dz$, $V = \int_{-h_o}^{\eta} v\,dz$, u and v are velocities in the x and y directions respectively, z is the vertical coordinate, τ^w is the wind stress, τ^b is the bottom stress whose components are given by $\tau_x^b = c_f q u$ and $\tau_y^b = c_f q v$, where $q = (u^2 + v^2)^{1/2}$, c_f is a friction factor, A_H is the constant horizontal eddy viscosity, and g is the acceleration due to gravity. The coefficient c_f has been assumed to be 0.003, a value typically used in flow problems of this type in rivers, lakes, and oceans.

The vertically integrated transport equation for the concentration of suspended solids of each size-class k, C_k, is

$$\frac{\partial(hC_k)}{\partial t} + \frac{\partial(UC_k)}{\partial x} + \frac{\partial(VC_k)}{\partial y} = D_H\left[\frac{\partial}{\partial x}\left(h\frac{\partial C_k}{\partial x}\right) + \frac{\partial}{\partial y}\left(h\frac{\partial C_k}{\partial y}\right)\right] + Q_{s,k} \qquad (4)$$

where D_H is the eddy diffusivity and $Q_{s,k}$ is the sediment flux at the sediment-water interface of the k'th component of the sediment. Only suspended sediment transport is considered here; bed load transport is not explicitly modelled.

Sizes of sediment particles in the Fox vary from less than 1 μm to over 1 mm but are predominantly in the clay and silt size range, i.e., from 1 to 100 μm. In order to accurately model sediment transport, it was found that a minimum of three size-classes of sediments were required: fine, medium, and coarse. Fine-grained sediments (generally less than 10 μm) have negligible settling speeds and are transported through the river without appreciable deposition on the bottom. These fine sediments make up a significant fraction of the suspended solids only during low flow periods. The medium size sediments (on the order of 10 to 100 μm) are present during all flows and generally are the major fraction of the suspended solids. It is assumed that these sediments are cohesive and therefore flocculate, with their settling speed dependent upon their state of flocculation. Coarse-grained sediments (generally greater than 100 μm) are a small fraction of the total load at

low and medium flow rates. However, during large storms and high run-off events, large amounts of coarse-grained material can be washed out of the surrounding drainage basin into the river. When this occurs, coarse-grained material may become a significant fraction of the total suspended solids load in the river. It is assumed that this coarse grain material is non-cohesive and does not flocculate.

Resuspension and Deposition

The resuspension properties of bottom sediments are significantly affected by particle size variations and also by cohesion between particles. At low stresses, only the finer particles on the surface of the bed can be resuspended while the larger particles are left behind and armor the bed. In addition, cohesion of particles and the resulting compaction of the bed cause the resuspension rate to vary with depth and with time after deposition. Because of this, sediments near the surface are less compacted and are relatively easy to resuspend, while sediments further down are more compacted and more difficult to resuspend. The result of all this is that, for fine-grained sediments at any particular stress, only a finite and relatively small amount of sediment can be resuspended as opposed to noncohesive, uniform-size sediments, which have a uniform rate of resuspension (Massion, 1982; Tsai and Lick, 1987).

For cohesive sediments from both lakes and oceans, experimental work (Krone, 1962; Partheniades, 1972; Mehta, 1973; Lee et al, 1981; Lick and Kang, 1987; Tsai and Lick, 1987; MacIntyre et al, 1990) has determined that the resuspension rate and the total amount of sediment e that can be resuspended at a particular stress are functions of (a) the turbulent stress at the sediment-water interface and (b) the water content of the deposited sediments (or the time after deposition). In particular, it can be demonstrated that the rate of bed compaction (and the associated reduction in resuspension) is greatest immediately after the bottom sediments are deposited and then decreases with time. In most cases, the effects of benthic organisms must also be considered. These organisms continually rework the sediments. When these organisms are present and active, the properties of the sediment

bed change rapidly in the first few days after deposition, as in the absence of organisms, but then are essentially constant after five to seven days (Tsai and Lick, 1987).

A formula for ε which approximates the existing experimental data can be written as

$$\varepsilon = \frac{a_0}{t_d^n}\left[\frac{\tau - \tau_0}{\tau_0}\right]^m \qquad \text{for } \tau > \tau_0$$

$$= 0 \qquad \text{for } \tau \le \tau_0 \tag{5}$$

where ε is the net amount of resuspended sediment per unit surface area in gm/cm^2, a_0 is approximately equal to 8×10^{-3}, t_d is the time after deposition in days, n is approximately equal to 2, m is approximately equal to 3, τ is the shear stress (dynes/cm^2) produced by wave action and currents, and τ_0 is an effective critical stress which varies from approximately 0.1 dynes/cm^2 for freshly deposited sediments to approximately 1 dyne/cm^2 for t_d greater than one day. Each of the parameters τ_0, a_0, m, and n is dependent on the particular sediment (and the effects of benthic organisms) and needs to be determined experimentally. However, the above values are reasonable first approximations for fine-grained sediments. A more detailed discussion of the physical properties of the sediments in the Fox River and Green Bay and a determination of the values of the parameters in Eq. (5) is given by Xu (1991).

Experimental results, summarized by Eq. (5), have only been obtained for a very limited range of parameters and conditions. Limitations on the experimental data and Eq. (5) are as follows. (1) Data is only available for low to moderate shear stresses, generally below about 10 dynes/cm^2. (2) Data is only available for times of compaction of 1 to 30 days. Most importantly, insufficient data is available for very recently deposited sediments. (3) Data is only available for sediments depositing in a quiescent fluid. This is a valid approximation for sediments depositing in a lake, for example, where currents are generally small. However, in a river, currents are generally much larger and continuously present. The effects of currents on deposition can be shown to be of major significance

(Xu, 1991), but have not been sufficiently investigated. Although Eq. (5) is a valid description of existing experimental data and is useful as a first approximation to describe the variation of ε in many realistic situations, it will be demonstrated below that significant additional information is needed for an adequate description of sediment resuspension in the Fox and other similar rivers.

The above formula is for the net resuspension. The total amount of sediment is not resuspended instantaneously but over a period of time on the order of an hour. In our present computations, it was assumed that the resuspension rate was constant and equal to its initial value until all available sediment was resuspended and was then zero until further sediment was deposited and available for resuspension, or until the shear stress increased further.

In our present model, in order to accurately describe the resuspension of the bottom sediments, the sediment bed is assumed to consist of layers in the vertical direction; the properties of each layer depend on time after deposition and composition (relative fractions of medium and coarse sediments) and are allowed to vary in the horizontal direction. An arbitrary number of layers and their thicknesses can be defined initially. In the present calculations, 11 layers were assumed. This is more than sufficient. The top layer is assumed to be newly deposited sediment less than three hours old. It has a very high water content and is more like a thick soup rather than a mud. Because of this characteristic, it has a very low critical shear stress ($\tau_0 = 0.1$ dynes/cm^2) and is easily resuspended. Below this fresh layer are three, six, and twelve hour old layers with critical shear stresses increasing with age ($\tau_0 = 0.2$, 0.4, and 0.8 dynes/cm^2 respectively). Below these layers are 7 layers one to seven days old with age increasing with depth. For these layers, it has been assumed that $\tau_0 = 1.0$ dynes/cm^2. As a first approximation, all layers compact with time with ε for each layer given by Eq. (5) above.

The program monitors changes in the sediment bed for each cell in the river model. Deposited material is added to and sediment is resuspended from the surface layer. The program also monitors the aging of the bed. The fresh layer material is moved into the three hour old layer every three hours, the three hour old material is moved into the six

hour old layer every six hours, and so forth. For a given cell in the model, any or all layers may be empty. Frequently the bottom layer (seven days old) is the only layer containing sediment, and therefore it is the surface layer.

The net sediment flux is of course given by the difference between the resuspension rate (ε/3600 s) and the deposition rate. Deposition is assumed to be due to settling of the suspended particles, so that the deposition rate D is given by

$$D = \sum_{k=1}^{3} w_{sk} C_k \qquad (6)$$

where w_{sk} is the effective settling speed for the k'th class of particles. As noted above, the settling speed is different for each of the three classes of sediment. For the fine grain class of sediments, $w_s = 0$.

For the medium sized class of sediments, which are assumed to be cohesive and therefore flocculate, w_s depends on the floc diameter and a variable effective density which in turn depends on the conditions (fluid shear and sediment concentration) under which the floc was formed (Burban et al, 1990). This is different from non-cohesive particles whose settling speed depends only on the particle diameter and a constant density. For flocs in quasi-equilibrium with the local flow conditions, experimental data has shown that the average floc diameter can be approximated by

$$C\, d_m^2\, G = \alpha_0 \qquad (7)$$

where d_m is the median floc diameter (cm), G is the fluid shear stress (dynes/cm^2), and α_0 is an experimentally determined constant (Lick and Lick, 1988). For fine-grained, cohesive sediments in fresh water, $\alpha_0 = 10^{-8}$ gm^2/cm^3 s^2. From laboratory experiments on flocculated, cohesive sediments (Burban et al, 1990), a valid first approximation for w_s is then

$$w_s = B_1(CG)^{-0.85} d_m^{-[0.8+0.5 \log (CG-B_2)]} \quad (8)$$

where $B_1 = 9.6 \times 10^{-4}$, $B_2 = 7.5 \times 10^{-6}$, and both are experimentally determined constants. Settling speeds for these flocs range from 60 μm/s to 160 μm/s and are always less than that for a solid particle of equal diameter, generally by several orders of magnitude. It can be seen that w_s increases as either C or G increases.

The coarse class size of particles are non-cohesive and have a faster settling speed than the medium class size. For this class of particles, an average settling speed of 1 mm/s has been assumed.

Numerical Calculations

The above sediment bed model was combined with the hydrodynamic and sediment transport model described previously. The resulting model is valid for rivers, lakes, and near-shore areas of the ocean whenever the vertically-averaged approximation is valid.

The specific application here is to the lower Fox River (Figure 1). In these calculations, rectangular grid elements, 90 m by 30 m, were used. As input data, the model requires specified flows and suspended solids concentrations at the DePere Dam and the East River. It also allows specified changes in water surface elevations at the mouth of the Fox due to seiches in Green Bay while, at the same time, allowing waves and other surface motions to propagate out of the Fox into the Bay without reflection at this boundary. With this treatment of the boundary, reverse flows from Green Bay into the Fox are allowed and are correctly modeled when they occur.

For purposes of calibration and verification, calculations were made for a wide range of real, time-varying flows and were then compared with field measurements. These calculations are presented and discussed below. However, for purposes of quantitatively understanding some of the main features of the flows and the effects of flow rate, several calculations of steady-state flows were also made. The results of these latter calculations will be discussed first.

Steady-State Flows

In the following calculations, it has been assumed that the flows are steady, that all sediments are medium size, and that the properties of the sediment bed are initially independent of depth and correspond to a seven day old bed with ε given by Eq. (5). The flow in the East River was assumed to be 10% of the flow into the Fox over the DePere Dam with the same solids concentration in the East River as at the Dam.

In the first calculation, a steady-state flow of 105 m^3/s, corresponding to the median flow in the Fox, and a suspended solids concentration at the Dam of 30 mg/ℓ, typical of median flows, were assumed. Calculated results for the depth-averaged velocities are shown in Figure 2a. The flow generally follows the deeper parts of the river channel except near DePere Dam where the inertia of the flow causes higher currents in shallow regions of the river. Compared to the upstream part of the river, currents are significantly higher in the downstream part of the river where it is relatively deep but narrow.

Bottom shear stresses corresponding to this flow are shown in Figure 2b. The shear stresses are generally comparatively low. In the upstream part of the river, shears are almost always below 0.5 dynes/cm^2 while, in the downstream part, shears are somewhat greater, especially in the deep and narrow part of the river where shears range from 0.25 dynes/cm^2 to above 1.0 dynes/cm^2 in a few isolated areas.

Depositional and erosional areas are shown in Figure 2c. Because of the low shear stresses, erosion occurs at only a few, small, isolated areas. Deposition occurs in the shallow, near-shore areas with much greater deposition in the upstream portion than in the downstream portion. More specifically, large amounts of deposition occur in the shallow regions near DePere Dam, near Fort Howard, and near the mouth of the East River. It should be noted that there are large areas where neither net resuspension nor net deposition occur.

The concentration of suspended solids is shown in Figure 2d. The concentration decreases more rapidly in the upstream portion because of the large deposition of sediments in the shallow areas near the Dam and at Fort Howard. In the downstream portion, the

concentration is fairly constant since there is little deposition in this portion of the river. Of the total amount of solids input to the river, approximately 80% is deposited in the river.

A calculation was also made for a 99.7 percentile flow of 280 m^3/s. The suspended solids concentration at the Dam was assumed to be 75 mg/ℓ, a typical concentration for a flow of this magnitude. Currents are shown in Figure 3a and are qualitatively similar to those shown in Figure 2a, except higher of course. Bottom shear stresses are shown in Figure 3b. In the upper portion of the river, large bottom areas now have stresses above 1 $dyne/cm^2$ with a maximum a little over 4 $dynes/cm^2$. In the lower portion of the river, most of the channel now has stresses greater than 1 $dyne/cm^2$ with considerable areas having stresses of 2 to 5 $dynes/cm^2$.

Calculated results for erosion and depositon are shown in Figure 3c. It can be seen that, because of increased stresses, large parts of the river now show erosion while deposition is more confined to the near-shore, shallow regions.

The suspended solids concentrations are shown in Figure 3d. Again, the most rapid decrease in concentration occurs in the upstream part of the river where most depositon occurs. The solids concentration in the lower part of the river is almost constant. Of the total amount of solids input to the River, approximately 24% is deposited in the River.

From the data on flow rates and sediment concentrations, rates of sediment transport and deposition can be calculated. From this data, it follows that much more sediment is transported to Green Bay during a high flow (17.56×10^3 gm/s) than during a medium flow (0.71×10^3 gm/s), but also (and somewhat surprisingly) more sediments are deposited in the river during a high flow (5.54×10^3 gm/s) than during a medium flow (2.75×10^3 gm/s).

Time-Varying Flow Events

For purposes of calibration and verification of the model and for further understanding, calculations of several real, time-varying flows were made and compared with observations. In particular, two flow events were modeled; the first was from May 22, 1989 to June 20, 1989 while the second was from March 24, 1989 to April 10, 1989.

The first period included low and medium flows as well as a once-in-ten-year high flow (maximum flow of 432 m^3/s). The second period included medium and moderately high flows.

For the first event, the flow rate and measured sediment concentrations at DePere Dam and at the mouth of the river at Green Bay are shown in Figure 4a. At the beginning, there are three days of low flow (less than 75 m^3/s). The flow then increases rapidly to about 150 to 200 m^3/s. On May 30, the flow increases rapidly again and reaches a peak of 432 m^3/s, stays reasonably constant for about five days, and then gradually declines over a period of 10 to 12 days to about 150 to 175 m^3/s.

In the beginning during low flows, suspended solids concentrations at both DePere and at the river mouth are relatively low. As the flow increases on May 25, solids concentrations at both places increase rapidly to a peak and then decline rapidly. As the flow increases again on May 30, the solids concentrations at both DePere and the mouth again increase rapidly and then decline with a more rapid decline initially at the mouth than at the Dam. During high flows, the solids concentrations at the mouth are a significantly smaller fraction of the solids concentrations at DePere than during low flows. The high solids concentration at DePere on June 14 is unexplained and probably is a measurement error.

With the flow rate and solids concentration at the Dam as input, calculations were made of currents, bottom shear stresses, areas and amounts of resuspension/deposition, and suspended solids concentrations. In our first calculations, two simple, but critical, assumptions were made. One was that all sediments were medium size, while the second was that the properties of the sediment bed were initially constant with depth and corresponded to those of a seven-day old bed although they could change later as resuspension/deposition occurred. For accurate results, both of these assumptions need to be modified as will be demonstrated below.

When calculations were made with these approximations, reasonably good agreement between the calculated and observed suspended solids concentration at the mouth of the

river was obtained (Figure 4b). However, it is also evident from this figure that, at high flows, the predicted concentrations were higher than observed and, at very low flows, the predicted concentrations were lower than observed.

The reasons for this are assumed to be as follows. During large storms and hence high flows, more of the larger, coarse-grained material in the surrounding drainage basin can be washed into the river. These large particles have much greater settling speeds than do medium-size particles, are more difficult to resuspend, and hence cause lower suspended solids concentrations than if they were medium size. However, as noted above, only medium size particles were included in the initial calculations. If it is assumed that large particles are present during high flows with their fraction of the total load increasing with flow rate, much better agreement between the computed and measured concentrations is obtained during high flows.

Similarly, for low flows, a significant fraction of the total load must be fine-grained particles which have very small settling speeds and essentially travel through the system without settling. Because of this, their concentrations would be greater than those of medium size sediments under the same flow conditions. Therefore, if it is assumed that a fraction of the total suspended solids load at low flows is fine-grained, the discrepancy between the observed and calculated solids concentrations is reduced.

To accommodate the entire flow range, three size classes of sediments were therefore assumed with the relative proportion of each dependent on the flow rate Q. (See Figure 5.) At very low flows, almost all of the load can be fine-grained material with no coarse material present while, at high flows, the dominant material is medium-grained sediments with some coarse but no fines present. For large flows greater than 250 m^3/s, it was assumed that, after the maximum flow was reached, the medium size fraction was given by 0.9 - .0026 (Q - 150), and the rest was coarse-grain material with no fine-grained material present.

With this modification, the agreement between calculated and observed solids concentrations was significantly improved; especially good agreement was obtained when the flow was steady or declining in magnitude. However, when the flow rates increased

rapidly (for example, on May 25 and May 31), it can be seen from Figure 4a that large peaks in solids concentrations occurred. Although these peaks were present in the calculations, they were not reproduced well, especially the first peak.

The reason for these peaks is resuspension of an easily resuspendable surficial layer, the presence of this layer was not taken into account in the above calculations. In order to reproduce the first peak on May 25 reasonably well, a surficial layer with a mass of 0.5 gm/cm^2 and an a_0 (see Eq. (5)) of 0.1 was added to the sediment bed. With this modification, the results as shown in Figure 4c were produced. As can be seen, the agreement between the calculated and measured concentrations is excellent.

It should be noted that water level changes, or seiche effects, in Green Bay are very significant in modifying the currents, shear stresses, and sediment erosion in the Fox River during this period. Due to seiche effects, the flow velocity in the Fox can be retarded or increased by as much as 20 cm/s, and changes in shear stresses during a twelve hour period can be as great as 15 $dynes/cm^2$. During the period of maximum flow, from May 30 to June 3, typical shear stresses in the lower part of the river may be as high as 27 $dynes/cm^2$. Without the inclusion of seiche effects, these shear stresses would be decreased to 19 $dynes/cm^2$. This reduction in shear stress would decrease the erosion in parts of the river by as much as a factor of three.

For further verification, a second calculation, which included the period from March 24, 1989 to April 10, 1989, was also made. At the beginning of this period, the flow rates (Figure 5a) were about 150 m^3/s, then increased to about 200 to 250 m^3/s before declining near the end of the period. The solids concentrations were low at the beginning of the period. As in the previous calculation, when the flow rate increased rapidly, the solids concentrations at both DePere and at the mouth of the river increased rapidly to a peak and then declined rapidly. After this, the concentrations declined slowly for the rest of the period.

The parameters used in this second calculation were the same as in the previous calculation. A comparison of the calculated and measured solids concentrations at the mouth of the River is shown in Figure 5b. It can be seen that the agreement is very good.

In this calculation the seiche effect also resulted in greater erosion in the lower portion of the river. Shear stresses which would have been approximately 9.5 dynes/cm^2 had the lake level been kept constant increased to a peak value of 12.9 dynes/cm^2. This resulted in up to 2.5 times as much erosion.

Although not shown here, calculations were also made for low flow events. Generally good agreement between calculated and observed solids concentrations was obtained.

Concluding Remarks

A numerical model of the transport and fate of suspended solids in the lower Fox River has been developed and verified by comparison of the calculated and measured suspended solids concentrations at the mouth of the river. These comparisons were made and the model was verified for two long-term events of 30 days and 18 days (which included low, medium, and high flows) and several short term low flow events. In preliminary calculations of time-varying events, two critical assumptions were made. These were that the sediments were all medium size and the sediment properties were initially uniform with depth. With these assumptions, reasonable agreement between calculated and measured solids concentrations was obtained. However, at very high and low flows and during rapidly increasing flows, there were significant differences between the calculations and observations. These differences were attributed to and reduced by modifying the two assumptions mentioned above, i.e., (1) three size classes of sediments were assumed with the relative proportion of each dependent on the flow rate, and (2) an easily resuspendable surficial layer was assumed to be present at the beginning of the event. With these modifications, the agreement between the calculated and measured solids concentrations increased markedly, and excellent approximations to the suspended solids concentrations and the flux from the Fox into Green Bay were obtained.

These modifications are realistic but nevertheless are somewhat annoying in that they could not be quantitatively determined a priori. The fact that the median size of suspended sediments in a river flow increases as the flow rate increases is well-known. However, the specific variation of size with flow rate is not known and especially so for the Fox where size distribution as a function of flow rate was never measured. Similarly, the presence of an easily resuspendable surficial layer in both rivers and lakes is well-known from field and laboratory observations. However, the thickness and amount of solids in this layer is not known nor are the parameters on which this layer depends. Because of this, an accurate a priori evaluation of all parameters needed in the model was not possible. Nevertheless, after all is said and done, it should be emphasized that the present model, even with the assumptions of constant properties, describes the suspended solids concentration and flux to Green Bay quite accurately.

Although the effects of the easily resuspendable surficial layer are quite evident in the concentration vs time record, the resuspension of this layer (because the total amount of material in this layer is relatively small) does not contribute more than 10 to 20% of the total flux of solids to Green Bay during the periods being modeled. In other periods, where the flow is changing more slowly, the effects of this surficial layer should be even less. Because of this and as a first approximation, it may be valid to ignore this layer in long-term calculations.

Nevertheless, the presence of this layer is obvious, and its effects are intriguing and need to be understood, especially in terms of contaminant flux from the sediments. Before this layer is adequately understood, considerable work needs to be done on the deposition of fine-grained sediments in a flowing stream. Also, as mentioned above, the variation of the suspended solids size distribution as a function of flow rate is not quantitatively understood and needs investigation.

The shear stress coefficient c_f was assumed to be constant and equal to 0.003. It is well known that this coefficient varies with flow rate, depth, and bed roughness scale length (Christoffersen and Jonsson, 1985). However, the characteristics of the bottom sediments in the Fox are not well known, and it was felt that the assumption of a constant

coefficient of 0.003 was adequate at the present state of the investigation.

In summary, a numerical model of the transport and fate of suspended solids in the lower Fox has been developed and verified. As a first approximation, the suspended solids concentration and flux to Green Bay can be calculated on the basis of sediment parameters determined a priori from field and laboratory measurements. Because of this, it should be possible to apply this model to other rivers and lakes, again with sediment parameters determined only and a priori from laboratory and field measurements. Little or no field calibration should be necessary. Of course, this needs to be verified for other rivers and lakes. Further refinements of the model were based on calibrating the model by comparison of the calculated and measured suspended solids concentration.

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FIGURE 1

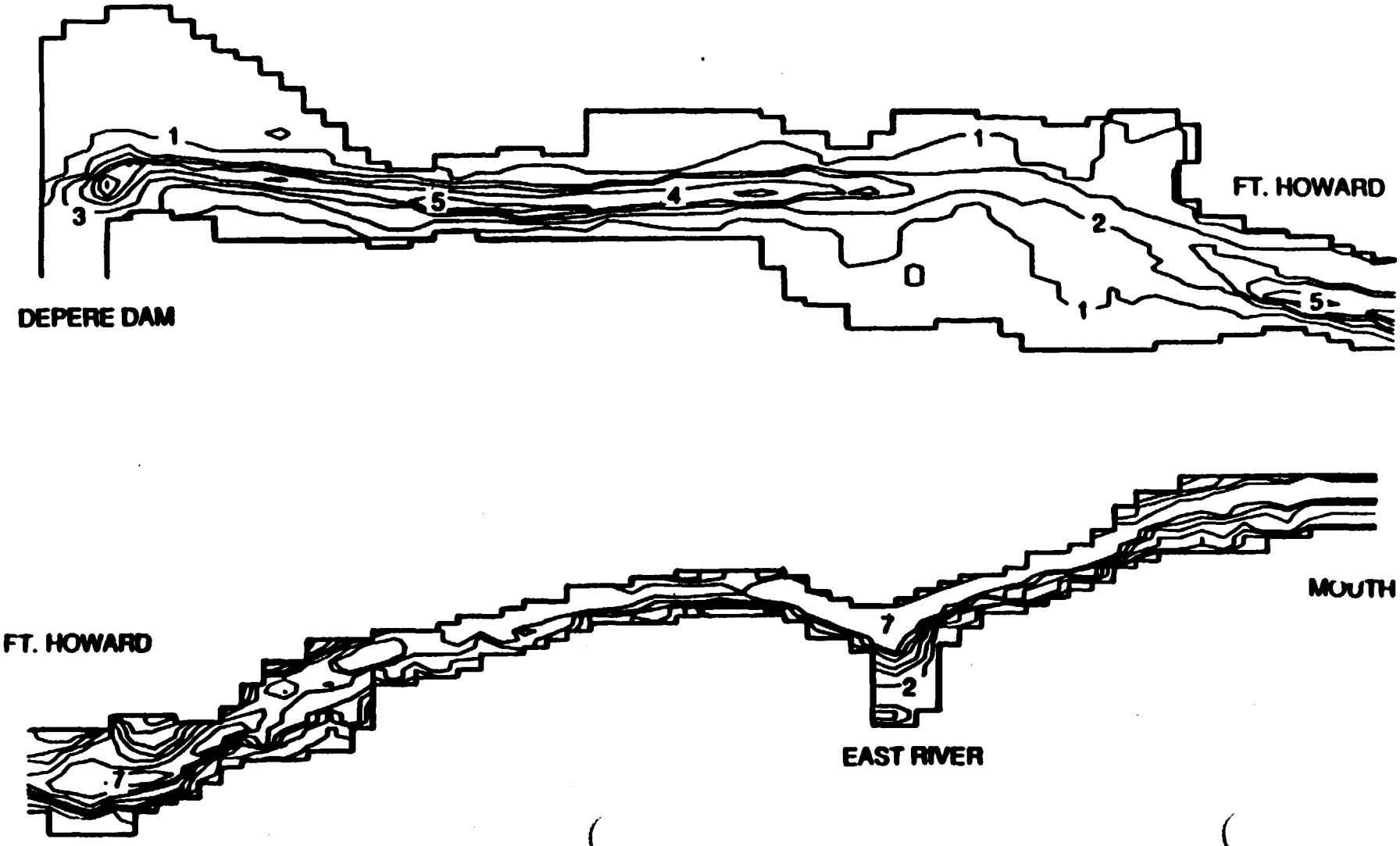
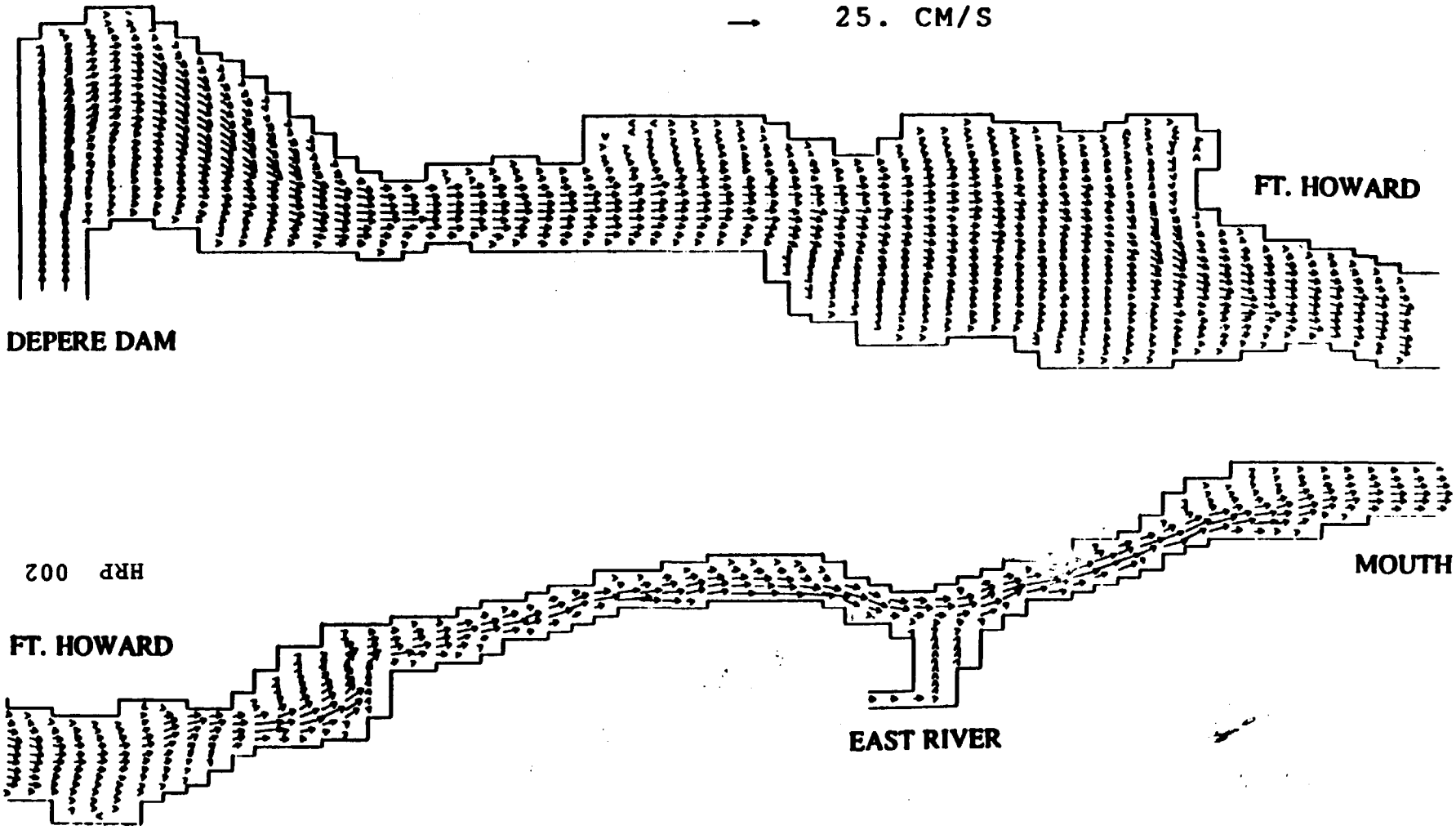


FIGURE 2a



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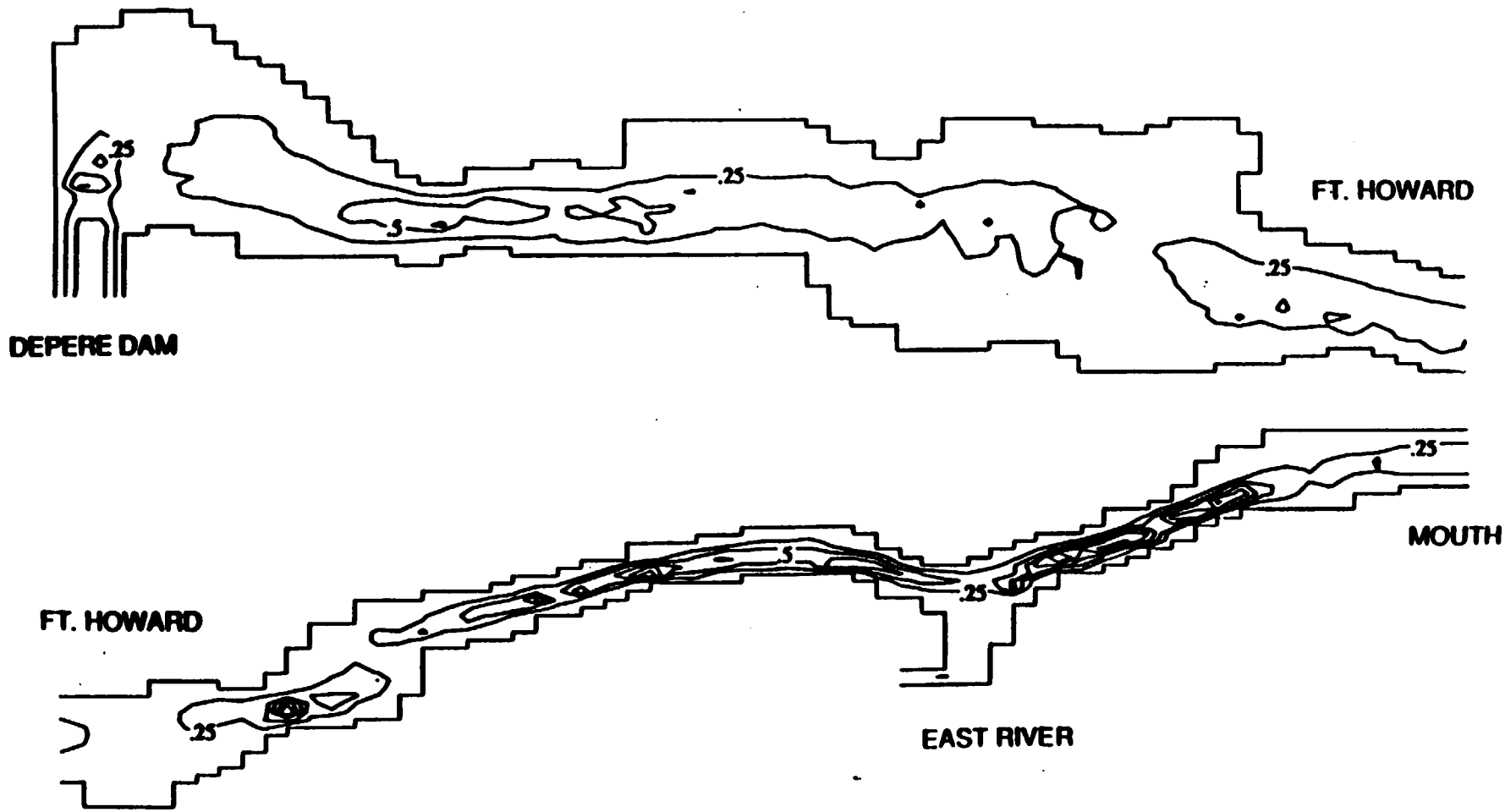


FIGURE 2c

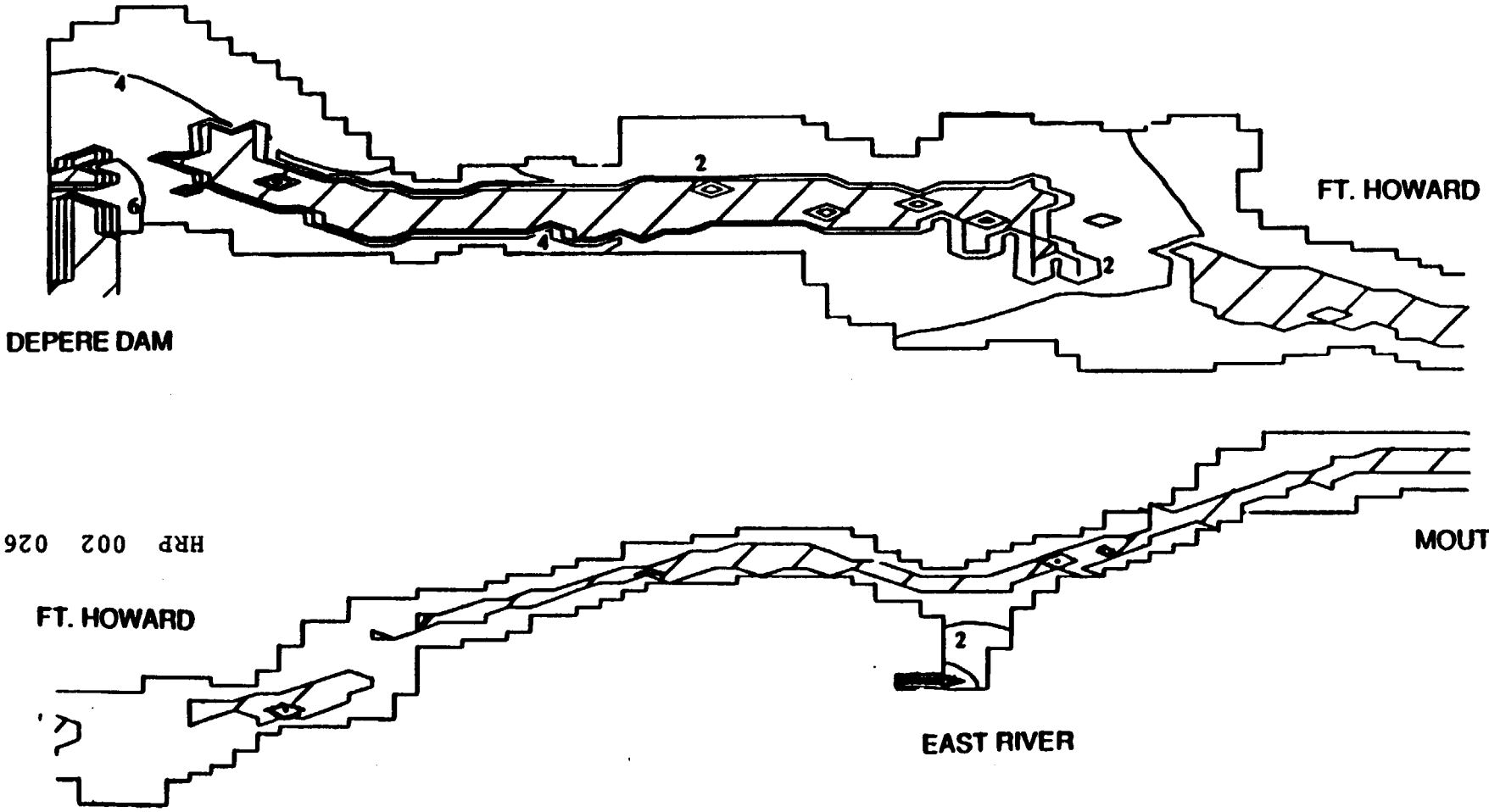
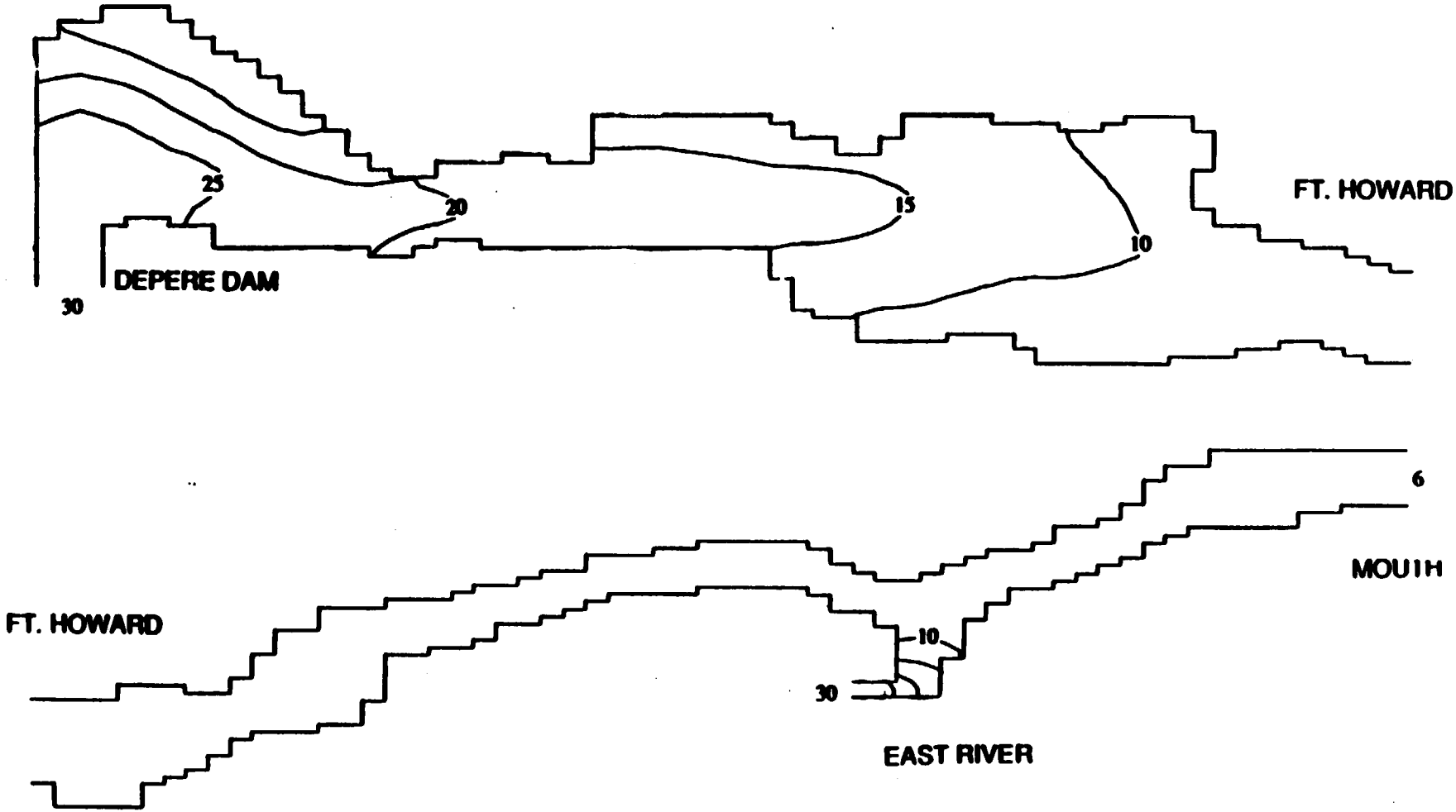


FIGURE 2d



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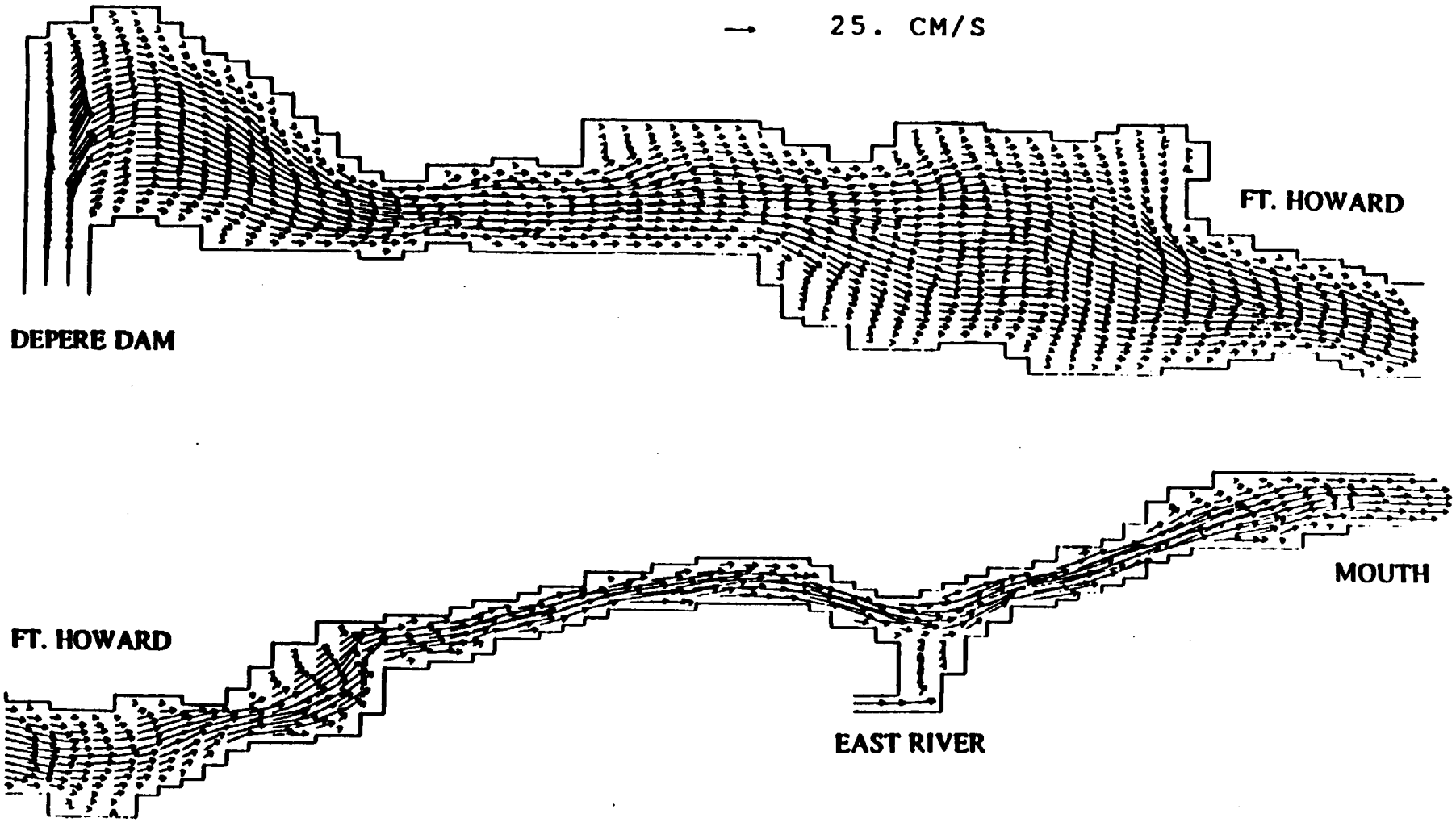
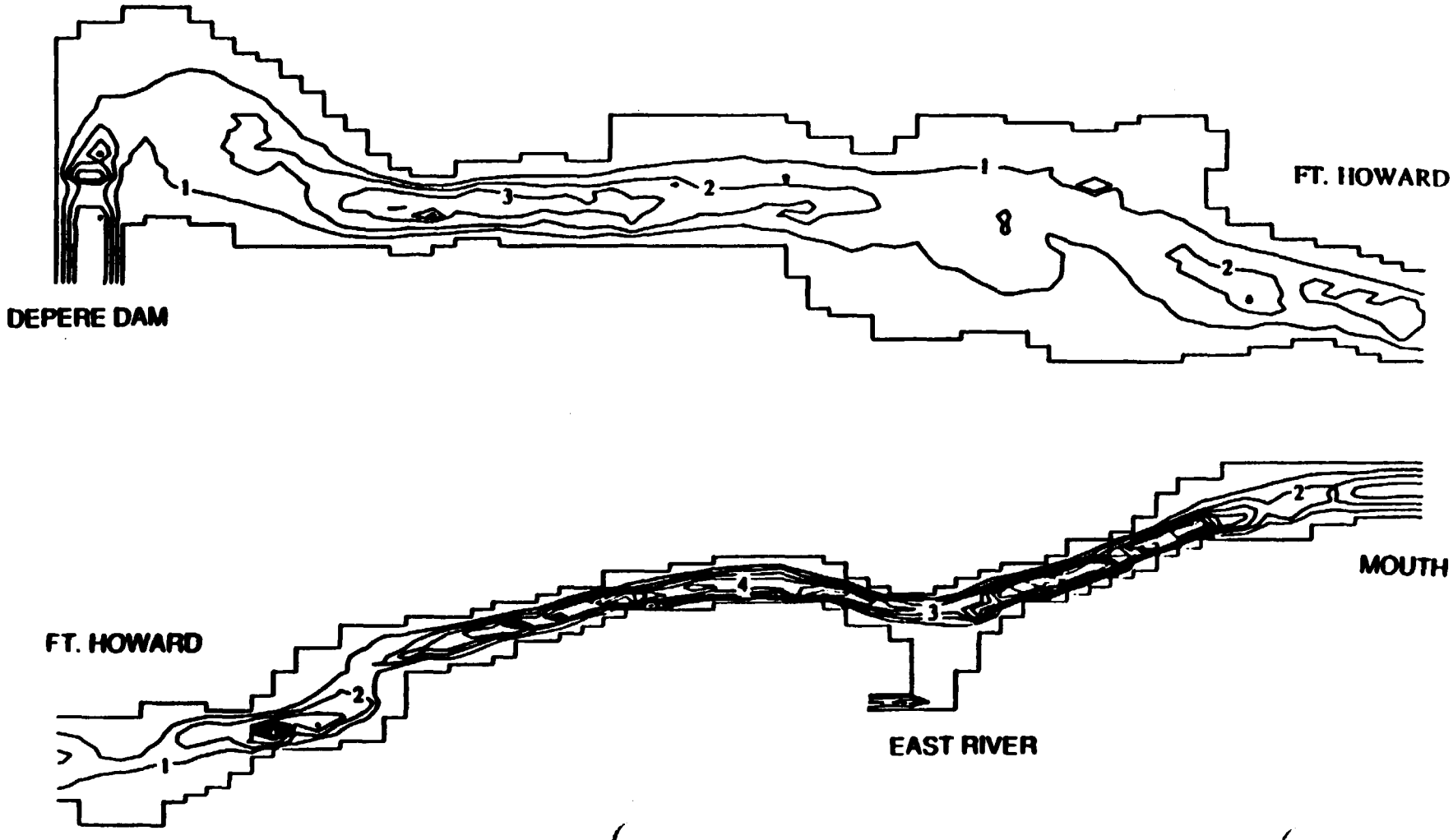


FIGURE 3b



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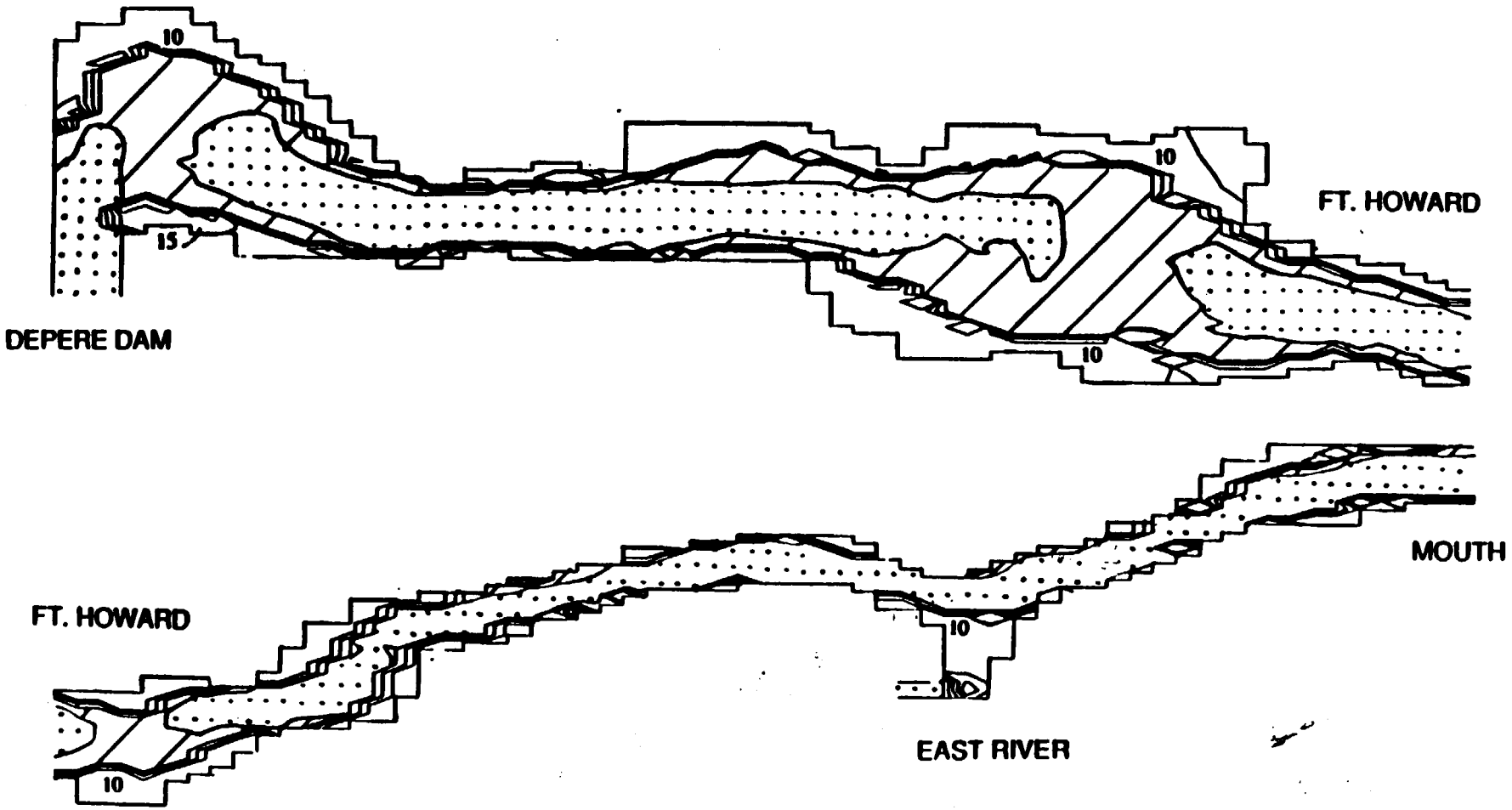
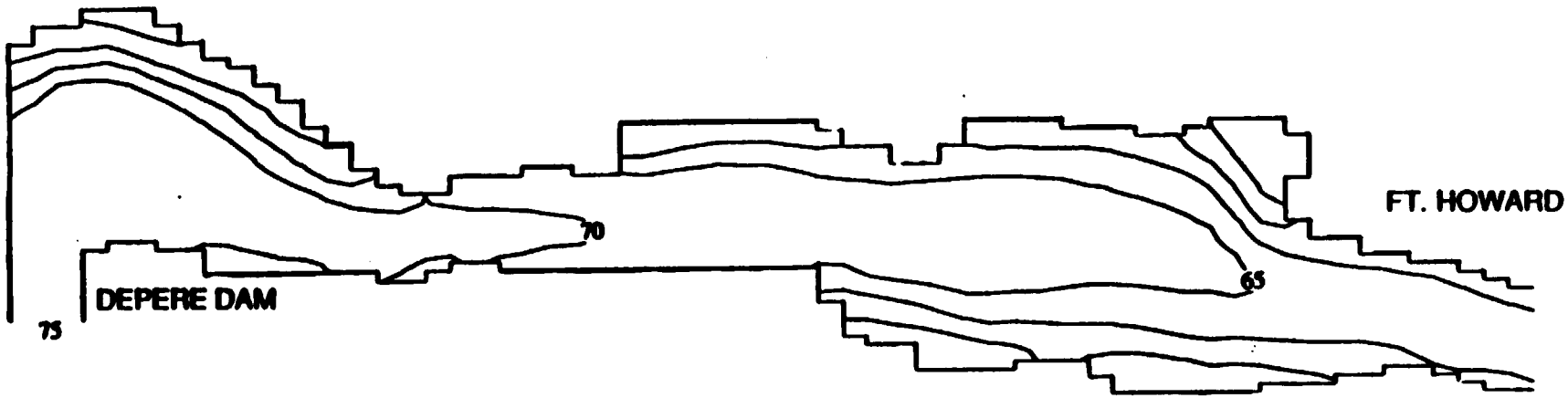


FIGURE 3d



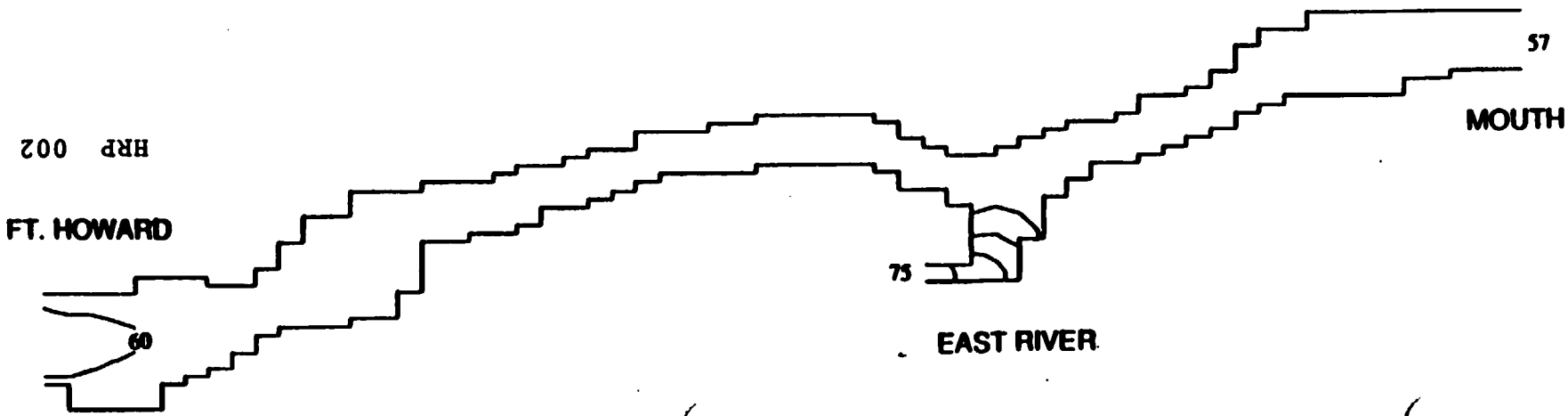
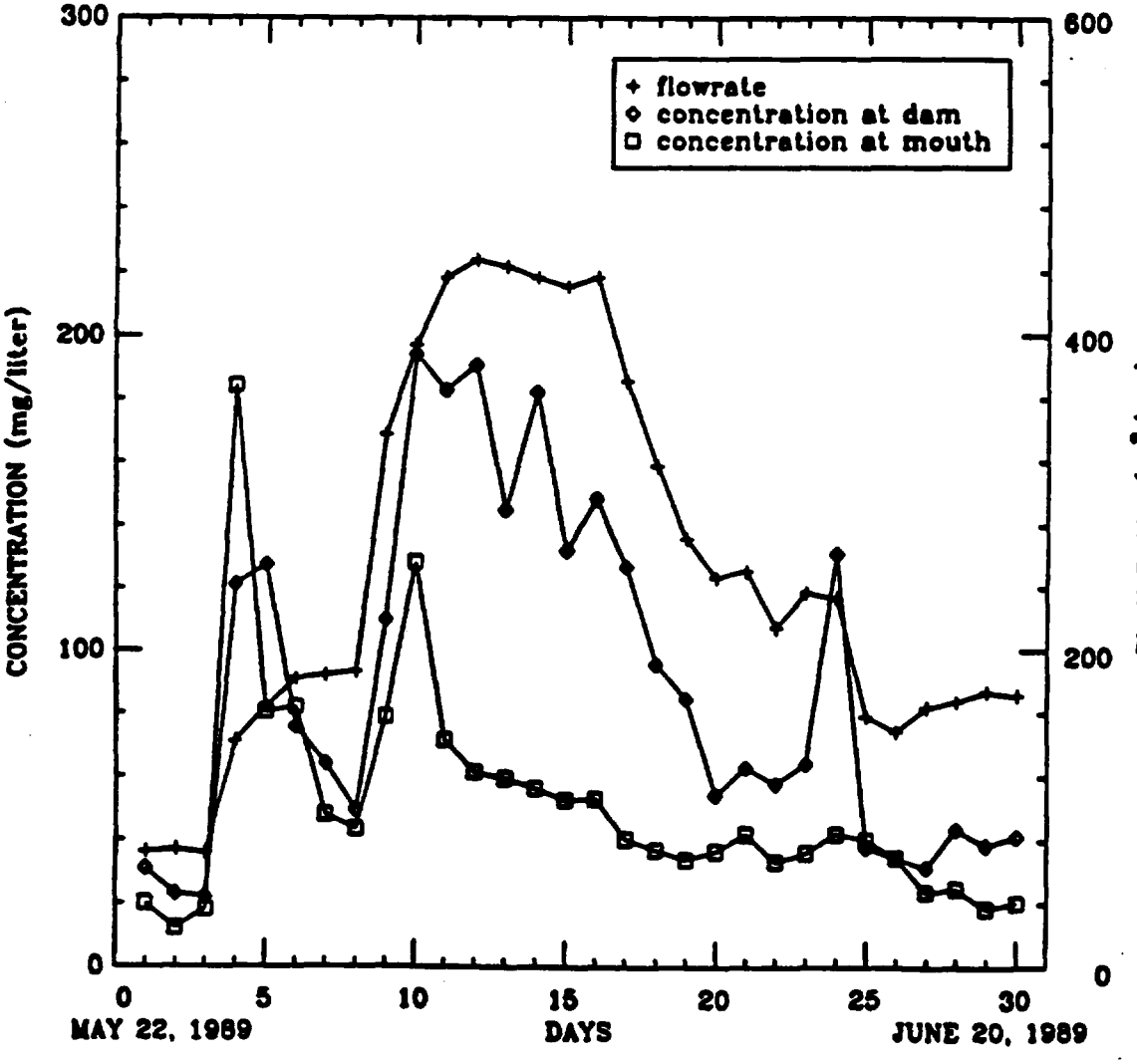


FIGURE 4a



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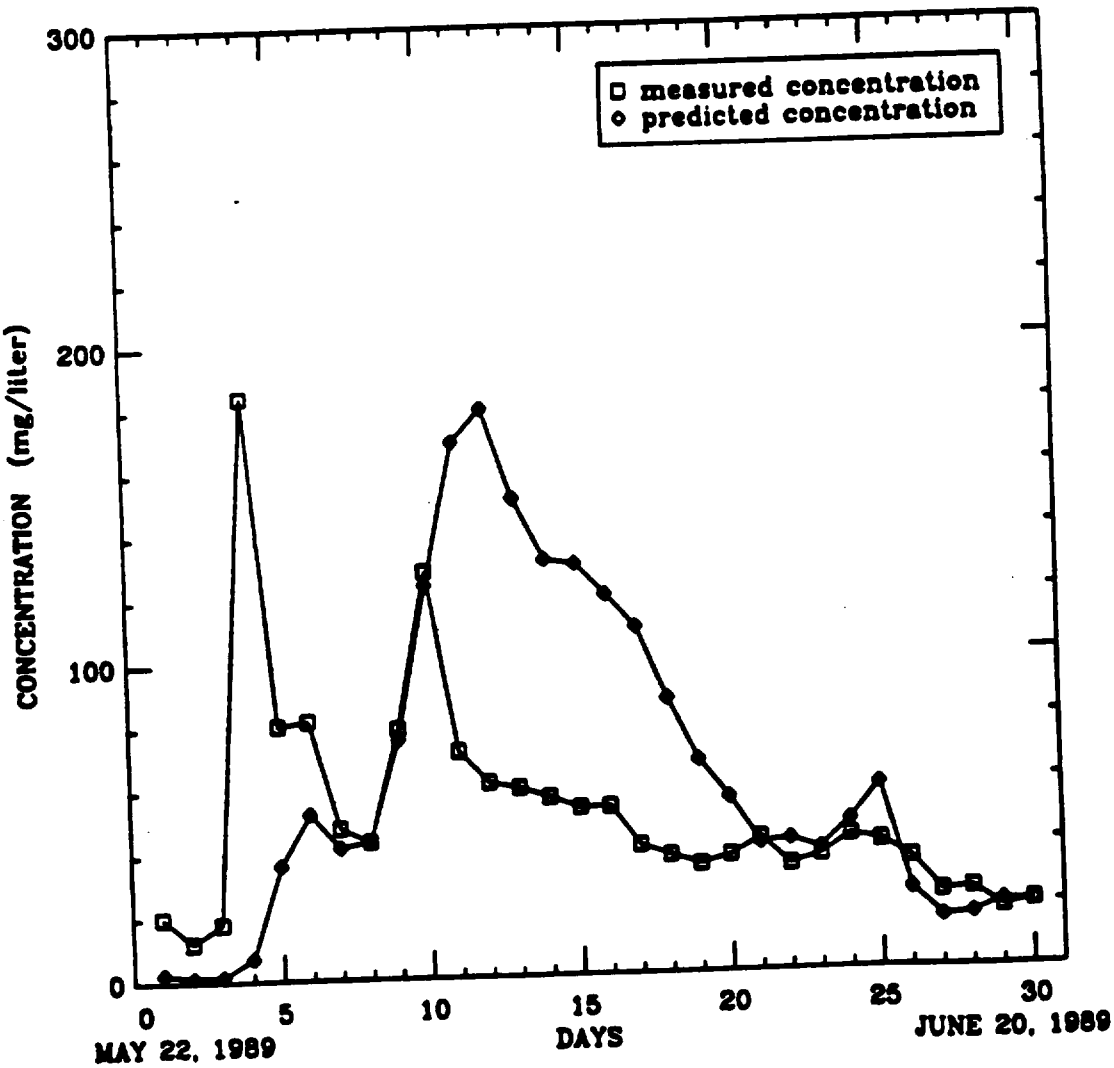
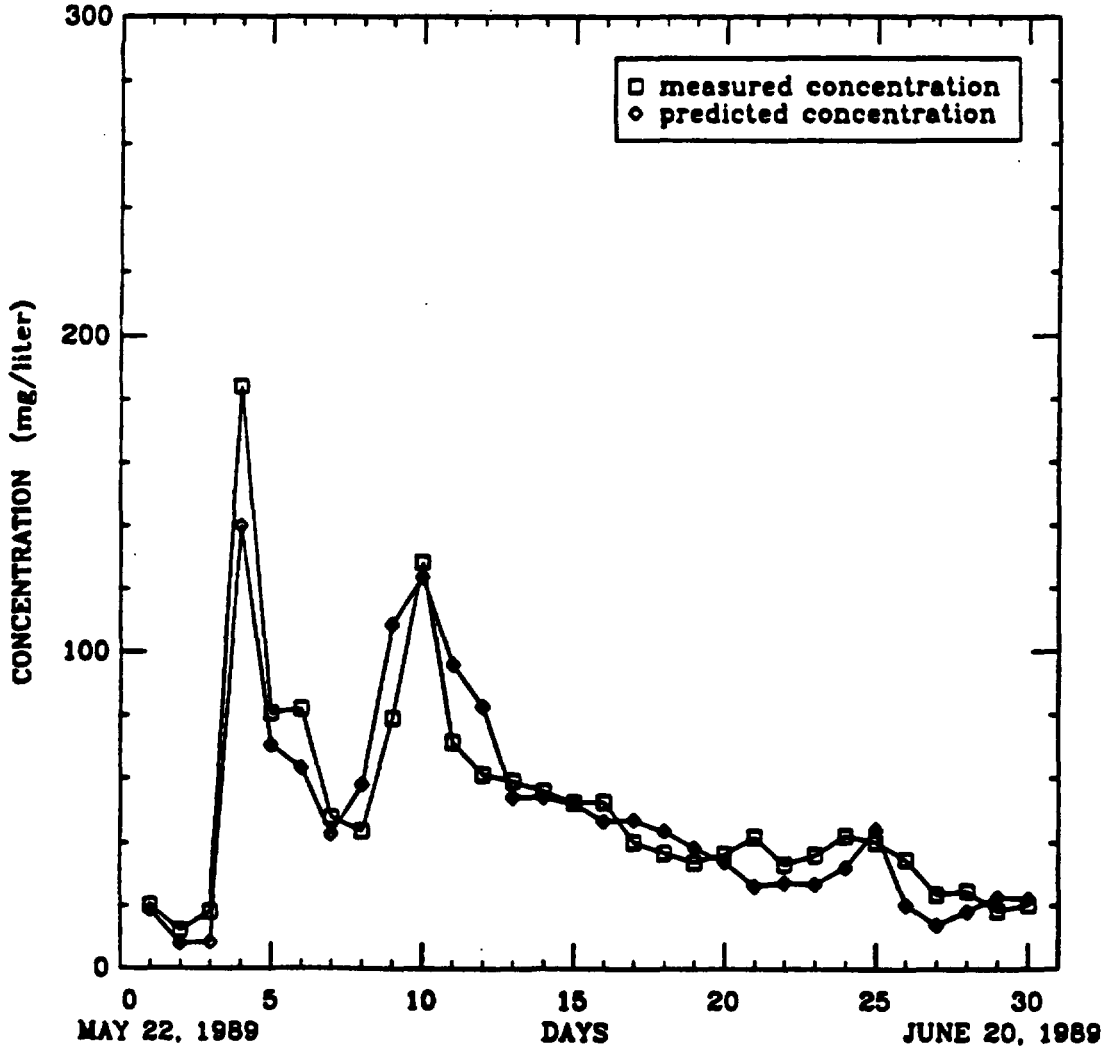


FIGURE 4c



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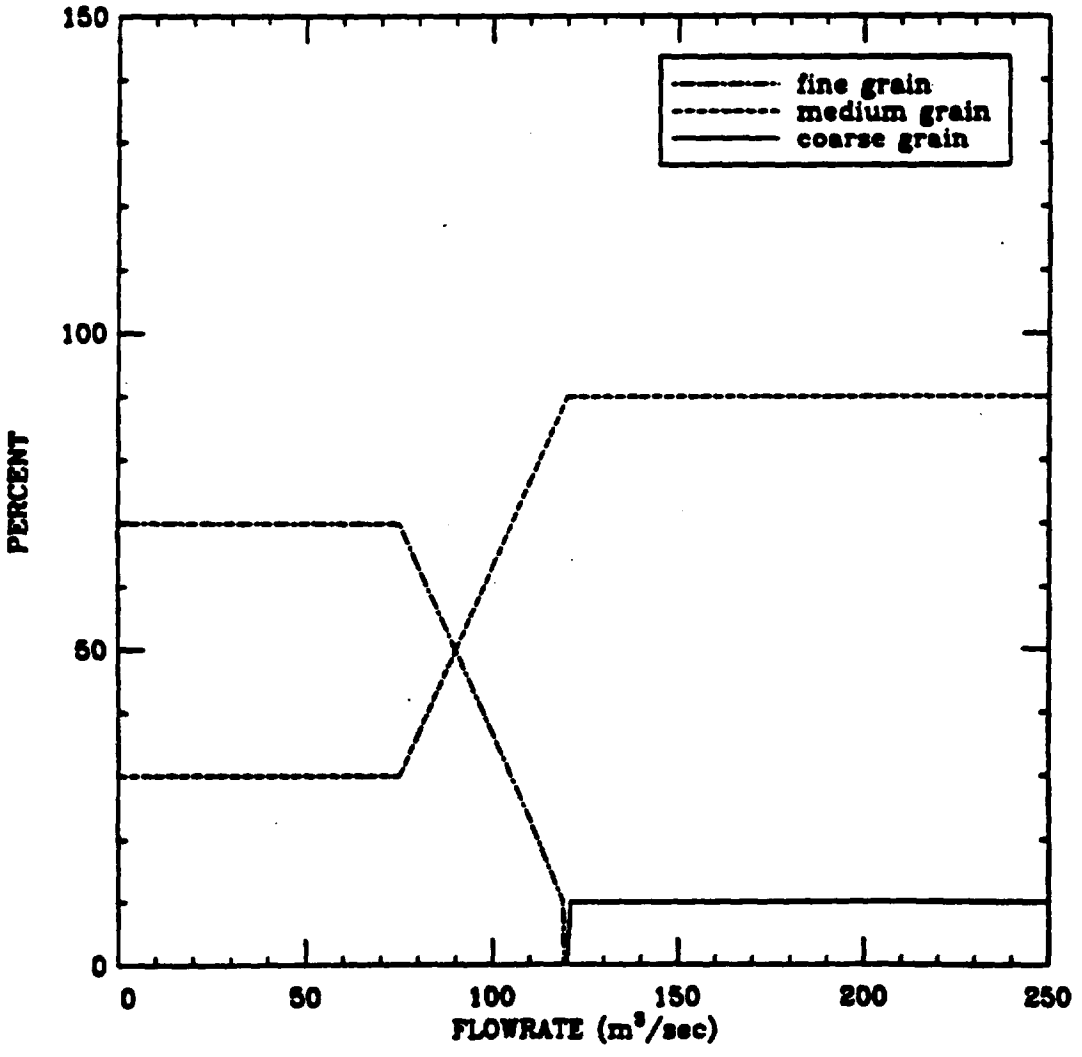


FIGURE 6a

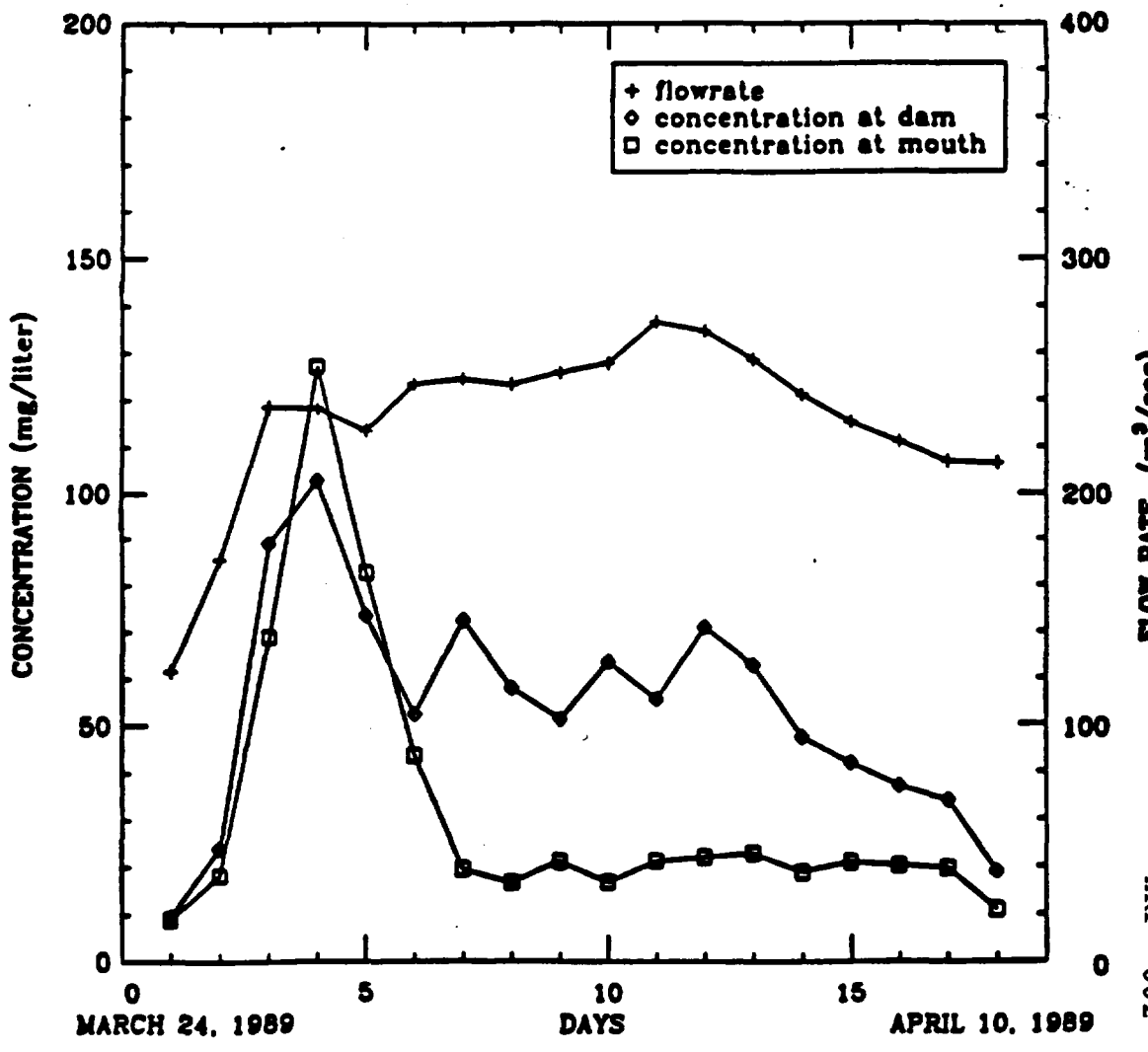
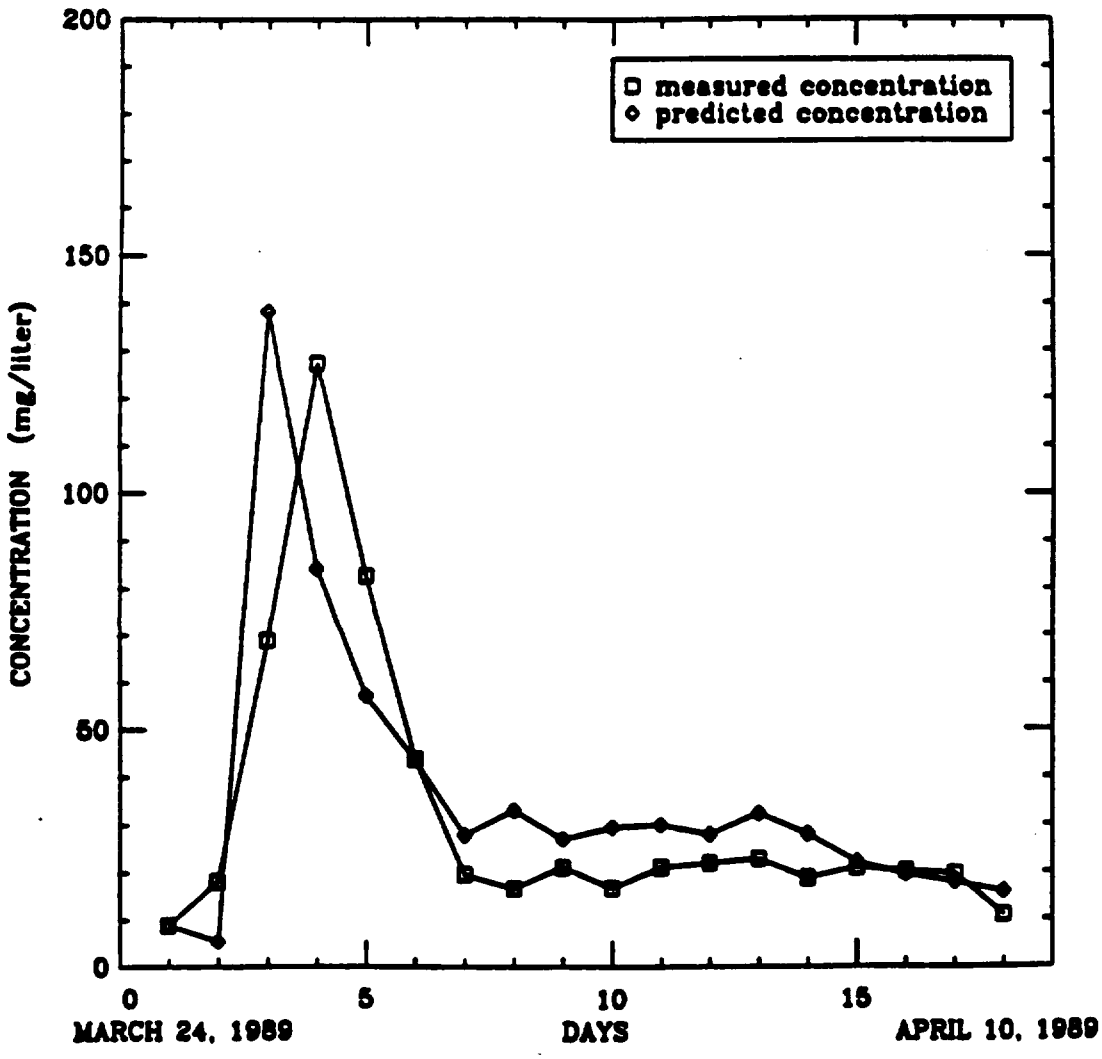


FIGURE 6b



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SEDZL : A USER-FRIENDLY NUMERICAL MODEL FOR DETERMINING THE TRANSPORT AND FATE OF FINE-GRAINED, COHESIVE SEDIMENTS

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DISCLAIMER

The present report is a rough draft for the purposes of early dissemination of a working program on sediment transport and to receive criticisms and helpful suggestions for improvement to the program. This is not a final report; it will be modified and hopefully improved with time. SEDZL is not to be used or referenced without the authors' consent.

ACKNOWLEDGEMENTS

The writing of this report, as well as the numerical calculations, experimental work, and theoretical development on which the present report is based were supported by the U.S. Environmental Protection Agency, first through the Large Lakes Research Station at Grosse Ile, Michigan and later by the Great Lakes National Program Office, Chicago, Illinois.

Table of Contents

1. INTRODUCTION

The sediment transport model, SEDZL, can be a very useful tool in determining the transport and fate of fine-grained, cohesive sediments in shallow waters. Use of SEDZL can be complicated, however, due to the generality and flexibility built into the model. The purpose of this report is to provide the user with instructions which make the use of SEDZL a straight-forward process. Included with the SEDZL code is a user-friendly interface called *setup* which makes input file creation and modification relatively easy.

The following topics are discussed in Section 2: the governing equations which are solved numerically by the model along with a description of the sediment dynamics included in the model; numerical parameters and stability limits; capabilities of the model; computer requirements; and software installation. Section 3 deals with auxilliary input files which must be created in order to analyze a particular problem. Section 4 describes in detail the input file and use of *setup* for easy creation and modification of the input file. Finally, two example problems, the Lower Fox River and Green Bay, are presented in Section 5. Included with both examples are all of the input files used in the calculations and the resulting output files.

SEDZL and *setup* have been extensively debugged and tested; however, the possibility exists of minor errors still existing in the programs. The user is encouraged to contact the authors of this report if errors or bugs in the programs are discovered. In addition, user comments on program improvements and modifications are also welcome.

2. MODEL DESCRIPTION

2.1 VERTICALLY INTEGRATED SEDIMENT TRANSPORT MODEL

Vertically integrated hydrodynamic and sediment transport equations have been used in SEDZL in order to simplify the numerical analysis (Ziegler and Lick, 1986). The primary assumptions used to derive these equations are: hydrostatic pressure variation, complete mixing of the water column in the vertical direction, and the horizontal velocities and suspended sediment concentrations are approximately independent of depth. These equations are valid approximations for situations where the water is relatively shallow and where the vertical stratification of the water column is weak. The resulting hydrodynamic equations are:

$$\frac{\partial \eta}{\partial t} + \frac{\partial U}{\partial x} + \frac{\partial V}{\partial y} = 0 \tag{2.1}$$

$$\frac{\partial U}{\partial t} + gh\frac{\partial \eta}{\partial x} = \tau_x^w - \tau_x^b + A_H\left[\frac{\partial}{\partial x}\left[\frac{\partial (U/h)}{\partial x}\right] + \frac{\partial}{\partial y}\left[\frac{\partial (U/h)}{\partial y}\right]\right] - \frac{\partial (U^2/h)}{\partial x} - \frac{\partial (UV/h)}{\partial y} + fV \tag{2.2}$$

$$\frac{\partial V}{\partial t} + gh\frac{\partial \eta}{\partial y} = \tau_y^w - \tau_y^b + A_H\left[\frac{\partial}{\partial x}\left[\frac{\partial (V/h)}{\partial x}\right] + \frac{\partial}{\partial y}\left[\frac{\partial (V/h)}{\partial y}\right]\right] - \frac{\partial (UV/h)}{\partial x} - \frac{\partial (V^2/h)}{\partial y} - fU \tag{2.3}$$

where the total water depth is $h = h_o + \eta$, $h_o(x,y)$ is the equilibrium water depth, $\eta(x,y)$ is the surface displacement from that equilibrium, U and V are vertically integrated velocities defined by

$$U = \int_{-h_o}^{\eta} u \, dz,$$

$$V = \int_{-h_o}^{\eta} v \, dz,$$

u and v are velocities in the x and y directions, respectively, z is the vertical coordinate, τ^w is the wind stress, τ^b is the bottom stress whose components are given by $\tau_x^b = c_f \, u|u|$ and $\tau_y^b = c_f \, v|v|$ where c_f is a friction factor, A_H is the constant horizontal eddy viscosity, f is the Coriolis parameter,

and g is the acceleration due to gravity.

The vertically integrated transport equation for suspended sediment concentration of size-class k, C_k, is

$$\frac{\partial(hC_k)}{\partial t} + \frac{\partial(UC_k)}{\partial x} + \frac{\partial(VC_k)}{\partial y} = D_H \left[\frac{\partial}{\partial x}\left[h\frac{\partial C_k}{\partial x}\right] + \frac{\partial}{\partial y}\left[h\frac{\partial C_k}{\partial y}\right]\right] + Q_{s,k} \qquad (2.4)$$

where D_H is the constant eddy diffusivity and $Q_{s,k}$ is the sediment flux at the sediment-water interface of sediment particle size-class k. Equation 2.4 must be solved separately for each sediment size-class being modelled. Only suspended sediment transport is considered here; bed load transport is not modelled.

The transport and fate of three different sediment size-classes can be modelled. A fine size-class which does not deposit on the sediment bed, i.e., zero effective settling speed, represents very small sediment particles. Fine-grained, cohesive sediments which flocculate are modelled by a medium size-class. Finally, coarser, non-cohesive sediments comprise the coarse size-class. The medium and coarse size-classes can be deposited on to and resuspended from the sediment bed.

Sediment dynamics are the crucial element in a transport model for accurately and realistically determining the fate of fine-grained, cohesive sediments. The sediment dynamics incorporated into SEDZL are based on valid laboratory and field studies concerning the deposition and resuspension of fine-grained, cohesive sediments (Fukuda and Lick, 1980; Lee et al., 1981; Lick, 1982; Tsai et al., 1987; Tsai and Lick, 1988; Burban et al., 1989, 1990; Xu, 1990). A brief review of the sediment dynamics used in the model will now be presented; a more detailed discussion can be found in Gailani et al., 1990.

The sediment flux at the sediment-water interface of sediment particle size-class k, $Q_{s,k}$, is simply the difference between the resuspension rate, R_k, and the deposition rate, D_k. For the fine particle size-class, $D_1 = 0$ and $R_1 = 0$. The deposition rates for the medium and coarse size-classes are given by

$$D_k = \beta_k C_k \quad , \quad k = 2, 3 \qquad (2.5)$$

where β_k is the effecive settling speed of size-class k. The coarse size-class is assumed to be non-

flocculating so this class has a constant effective settling speed, β_3. In SEDZL, β_3 is specified by the user through the variable BETA, see Line 14 of the input file in Section 4.1.

The medium size-class flocculates and laboratory results have been used to construct an approximate flocculation model. These experiments indicate that the following relationship is a valid first approximation

$$d_m = \left[\frac{\alpha_o}{C\,G} \right]^{1/4} \tag{2.6}$$

where d_m is the median floc diameter, C is the sediment concentration of the medium size-class, G is the local fluid shear stress, and α_o is an experimentally determined constant. For sediments in fresh water, $\alpha_o = 10^{-8}$ $gram^2/cm^3\text{-}s^2$ and C is in $grams/cm^3$, d_m is in cm, and the units of G are $dynes/cm^2$. Thus, the median floc diameter, d_m, at any point in the solution domain of a particular problem can be deterimined once the medium size-class concentration and fluid shear stress at that point are calculated.

Once d_m is deterimined, the effective settling speed of the medium size-class, β_2, can be determined. Experiments have shown that β_2 primarily depends on the floc diameter but the concentration and shear stress at which the floc was created are important secondary effects (Burban et al., 1990). Based upon floc settling speed experiments, a valid first approximation for β_2, in cm/s, is:

$$\beta_2 = B_1\,(CG)^{-0.25}\,d_m^{-\left[0.8\,+\,0.5\,\log(CG\,-\,B_2)\right]} \tag{2.7}$$

where $B_1 = 9.6 \times 10^{-4}$ and $B_2 = 7.5 \times 10^{-6}$. At the present time, experimental limitations restrict the maximum median floc diameter to 0.0200 cm (200 μm). This restriction creates minor errors in the determination of settling speeds. As more experimental data becomes available, this restriction can be removed. The maximum floc diameter is controlled in the model by the variable DMAX, see Line 14 of the input file in Section 4.1. Using Eqs. 2.6 and 2.7, calculated settling speeds range from 60 $\mu m/s$ to 160 $\mu m/s$. Comparison of numerical results with field data has also indicated that no deposition of medium class-size sediments occurs when the bottom shear stress is greater than 0.1 $dynes/cm^2$. Variable TCRDEP, see Line 14 of the input file, controls the value of the maximum shear stress for medium size-class deposition.

The resuspension properties of fine-grained, cohesive sediments differ significantly from non-cohesive sediments, i.e., sand. Non-cohesive sediments will be resuspended at a constant rate if the sediment bed is subjected to a uniform shear stress, τ, which is greater than some critical shear stress, τ_c. As long as there is a supply of non-cohesive sediment, resuspension will occur. However, laboratory and field experiments clearly indicate that only a finite amount of fine-grained, cohesive sediment can be resuspended under the same conditions (Tsai and Lick, 1988). Based upon these experimental results, the total amount of sediment, ε ($grams/cm^2$), which can be resuspended at a particular bottom shear stress is

$$\varepsilon = \frac{a_o}{t_d^3}\left[\frac{\tau-\tau_c}{\tau_c}\right]^n \quad , \tau > \tau_c \tag{2.8}$$

$$= 0 \quad , \tau \le \tau_c$$

where τ is the bottom shear stress due to currents and waves ($dynes/cm^2$), τ_c is the critical shear stress which is site-specific and varies with depth in the sediment bed ($dynes/cm^2$), t_d is the time after deposition of the sediments in the sediment bed ($days$), a_o is a site-specific constant ($gm-day^3/cm^2$), and $n = 3$. The parameters a_o and n are user specified and are input through the variables A0IN and RESEXP, respectively, see Line 14 of the input file in Section 4.1. Experiments have also shown that the resuspension process takes approximately one hour. The assumption is made in the model that the resuspension rate is constant during this period.

A realistic model of the sediment bed structure is also necessary if the transport processes are to be modelled properly. A layered sediment bed model has been created and it is depicted in Figure 2.1. The effects of compaction on resuspension are accounted for by the parameter t_d in Eq. 2.8, which increases with depth in the sediment bed. The critical shear stress also increases with depth until $t_d \ge 1$ *day* after which time it is assumed to be constant. Experimental results have shown that compaction effects begin to become negligible for $t_d > 6$ to 7 days, approximately.

Model input variables controlling the structure of the sediment bed model are discussed further in Sections 2.3 and 4.1. Recommended values for deposition and resuspension parameters are summarized in Table 2.1. Values of the parameters used in the lower Fox River and Green Bay examples are given

in Figures 5.4 and 5.6.

A volume integral method was used to derive finite difference equations which are used to numerically solve Eqs. 2.1, 2.2, 2.3, and 2.4 (Ziegler and Lick, 1986). Interior and boundary point equations were developed which have the following attributes: explicit, second-order accurate, locally conservative, and boundary conditions are treated correctly. A unique feature of this model is its successful treatment of open boundary conditions (Lick et al., 1986; Lick et al. 1987).

2.2 NUMERICAL PARAMETERS AND STABILITY LIMITS

Accurate and stable numerical solutions of Eqs. 2.1 to 2.4 depend on the grid element size ($\Delta x, \Delta y$), timestep size (Δt), eddy viscosity (A_H), and eddy diffusivity (D_H). The values of these parameters which yield accurate and stable solutions are controlled by several dimensionless relations. Generally, the computational grid for a particular problem is first constructed, see Section 3.1, and this determines the values of Δx and Δy. The bathymetry of the aquatic system, $h_o(x,y)$, is also discretized and input to the model, see Section 3.2. Let $H_{max} = h_{o,max}$ + DELTAH, where DELTAH is a correction for long term changes in the mean water depth, see Line 22 of the the input file in Section 4.1. The maximum wave speed in the solution domain is then $c_{o,max} = (gH_{max})^{1/2}$. Now, the Courant number, $\sigma = \frac{c_o \Delta t}{\delta}$, where $\delta = 0.5(\Delta x + \Delta y)$, must be less than approximately 0.6 for stable hydrodynamic solutions. This stability limit restricts the maximum timestep size to

$$\Delta t_{max} \approx 0.6 \frac{\delta}{c_{o,max}}. \tag{2.9}$$

A similar stability limit exists for the sediment transport equation except that stability is based on the maximum current velocity. Thus, if the sediment transport timestep is denoted by Δt_{sed}, then

$$\Delta t_{sed,max} \approx 0.6 \frac{\delta}{u_{max}}. \tag{2.10}$$

where u_{max} is the maximum current velocity occurring in the calculation. The sediment transport timestep is input as an integer multiple of the hydrodynamic timestep, see Line 6 of the input file.

In addition to numerical stability, realistic and accurate solutions depend on the value of A_H used.

Generally, one should use the smallest A_H possible as this will usually produce the most realistic solution. However, there are numerical limitations on the minimum value of A_H, and these restrictions depend not only on the grid size but also on the velocity field of a particular problem. For accurate solutions, the theoretical value of the cell Reynolds number, $Re_{cell} = \frac{u_{max}\Delta_{min}}{A_H}$, where $\Delta_{min} = MIN\ (\Delta x, \Delta y)$, should be less than approximately 2. Thus, the theoretical minimum A_H is given by

$$A_{H,min} \approx 0.5\ \frac{\Delta_{min}}{u_{max}}\ . \tag{2.11}$$

A similar relation holds for the eddy diffusivity in Eq. (2.4):

$$D_{H,min} \approx 0.5\ \frac{\Delta_{min}}{u_{max}}\ . \tag{2.12}$$

Practically, much lower values of A_H and D_H can be used than the minimum values given in Eqs. 2.11 and 2.12. Generally, one can obtain realistic solutions with Re_{cell} values of up to 10. Numerical experimentation is the best method for determining the minimum values of A_H and D_H for a particular problem and Eqs. 2.11 and 2.12 should be used as guidelines. Also, a good assumption for most problems is that $A_H = D_H$.

Two other hydrodynamic parameters must also be specified, c_f and f. The value of the bottom friction factor can range from 0.002 for clay/silt sediment beds to 0.004 for sandy sediment beds. In nearly all problems to be solved using SEDZL, using $c_f = 0.002$ is a very good approximation. The effect of bed forms on c_f is not accounted for here.

In evaluating the Coriolis terms in Eqs. (2.2) and (2.3), the f-plane approximation is used , i.e., f is constant over the solution domain, and this approximation is valid for problems in which the North-South extent of the solution domain is less than about 100 kilometers (Pond and Pickard, 1978). The value of f is then determined from

$$f = 2\ \Omega \sin\phi \tag{2.13}$$

where Ω is angular velocity of the Earth ($\Omega = 7.272 \times 10^{-5}\ s^{-1}$) and ϕ is the latitude at the center of the solution domain.

2.3 CAPABILITIES OF THE NUMERICAL MODEL

SEDZL is a flexible and general sediment transport model but it does have limitations as to the types of aquatic systems it can be applied to and the kinds of problems it can solve. This section will broadly describe the capabilities and limitations of the model. Detailed descriptions of various program options and inputs are provided in Sections 3 and 4.

The numerical model can be applied to rivers, partially-enclosed bays, shallow lakes, and partially-enclosed well-mixed estuaries. Examples of systems which have been modelled using SEDZL are: Venice Lagoon (Italy), Trenton Channel of the Detroit River, Lower Fox River, Lake Erie, and Green Bay. The main restriction on the geometry is that two open boundaries cannot be joined at a 1/4 element intersection, see Section 3.1 and Figure 3.5. This limitation means that flow from a river into an unbounded lake or ocean cannot be modelled with this version of SEDZL. The modeller must also be sure that the assumptions concerning vertical mixing of the water column made in deriving the governing equations are valid. Shallow lakes which are strongly thermally stratified and estuaries with a well-defined salt wedge should not be modelled using SEDZL.

A total of 24 program options are available for controlling program execution, input, and output. These options are described in Section 4.1 and summarized in Table 4.2. The model is capable of performing three types of calculations: (1) time-dependent hydrodynamics, (2) time-dependent hydrodynamics and sediment transport, or (3) time-dependent sediment transport using steady-state hydrodynamics. As mentioned in Section 2.1, a maximum of three sediment particle size-classes can be modelled (fine, medium, and coarse). The modeller has the option of simulating the transport of a one, two, or three sediment component system, see Line 9 of the input file in Section 4.1. Resuspension of fine-grained sediments can be caused by currents and wind waves. A wind wave model is built into SEDZL (Ziegler et al., 1989) for determining bottom stresses due to wind waves for either a constant or time-varying wind field.

The numerical model uses a constant, rectangular grid element for constructing the computational grid, see Section 3.1 and Line 3 of the input file. The physical boundaries of the aquatic system being examined are approximated using this rectangular grid and this geometry is controlled by an auxiliary

input file called *node* which is described in Section 3.1. The computer program does not have to be modified and recompiled in order to model different geometries. Proper modifications of the *node* file are all that is necessary in order to study different problems. The variable bathymetry of the water body is also input from an auxiliary input file, see Section 3.2. Long-term variations in mean water depth can also be accounted for by SEDZL, see Line 22 of the input file in Section 4.1.

The present version of the code will work for a maximum problem size of 2100 grid points, $MX_{max} = 127$, and $MY_{max} = 50$, which is slightly larger than the Fox River example in Section 5. Many problems can be solved using these limits and it is not advised that larger sized problems be attempted unless the modeller has access to either a mini-supercomputer or supercomputer. See Section 2.5 for further discussion of code size modification and computer requirements.

A variety of program options are available for specifying the hydrodynamic forcing for a particular problem. A time-varying, spatially-constant wind stress can be input, see Section 3.3.1. Time-varying inlet flow rates, from multiple inlets, can be specified, see Section 3.3.2. For example, this option could be used to model a river with two tributaries, independently varying the flow rate at each of the three inlets. The flow rates can also be expressed as velocities (cm/s) or volumetrically (m^3/s or cfs). Tidal/seiche forcing can also be used to drive the system, see Section 3.3.3 and Line 49 of the input file. Specified incoming flows at an open boundary can also be superposed on outgoing flows generated in the interior of the solution domain, see Section 3.1.

Similar to the inlet flow rates, time-variable sediment loading at inlets can be specified, see Section 3.4. The composition of the total sediment load can be controlled by setting the inlet concentration of each sediment size-class.

Options exist for designating the initial composition and structure of the layered sediment bed. The total number of layers in the sediment bed is set by variable LAYMAX (Line 15 of the input file) and it can have a maximum value of 14. The initial composition of the sediment bed for a multi-component sediment system consists of a mixture of medium and coarse particles; the mixture distribution can be uniform throughout the solution domain, see Line 19 of the input file, or spatially-varying, see Section 3.5. The initial 'thickness' of each sediment bed layer can also be specified by the user.

see Line 17 of the input file. Generally, a sediment calculation is started with the oldest sediment layer, L=LAYMAX, having sediment in it and all other layer 'thicknesses' initially set to zero, see the examples in Section 5. Resuspension properties of each sediment bed layer are also set by the modeller. The time after deposition, t_d, of bed layer L corresponds to input variable FTIME(L), see Line 16 of the input file in Section 4.1. The critical stress also increases with depth, and τ_c of layer L is specified by variable TAUCR(L), see Line 18 of the input file in Section 4.1. Recommended values for FTIME(L) and TAUCR(L) are given in Table 2.2 and illustrated in Figure 2.1.

2.4 PROGRAM OUTPUT

Output from numerical calculations can come in several forms. Hard copy or graphics output of solution domain distributions is available for velocities (u, v), surface displacement (η), suspended sediment concentrations (C_k), change in sediment bed 'thickness', and sediment bed composition, see Lines 52 to 59 of the input file. Output of these distributions at periodic times during program execution is user controlled, see Line 60 of the input file. Time-histories at specific grid points in the solution domain of η, C_k, sediment bed thickness, and total bottom shear stress can also be generated, see Lines 62 to 107 of the input file.

Structure of the various graphics and time-history output files will now be discussed. The user may need to write a computer program to convert these output files to any special format required by the user's graphics software. The velocity graphics output file, *uvgraph*, contains the $u_{i,j}$ and $v_{i,j}$ velocities necessary for generation of a vector plot of the velocity field. The velocities are output in units of *cm/s*. All velocities at solid boundaries and outside of the solution domain are zero. The format of the file is 20F6.1 and its structure is shown in Figure 2.2a. If MX is greater than 20, then each line will wrap around; for a given j value, $1 \le i \le 20$ will be on the first line, then $21 \le i \le 40$ will be on the next line, and so on until i=MX is reached. See Section 3.1 for definitions of MX and MY. The first line is free format and it contains the TIME1 value which is the time, in hours, at which the u and v values immediately below it were output. Lines 2 to MY+1 contain u values at TIME1 and Lines MY+2 to 2*MY+1 contain v values at TIME1. The program has the option of outputting intermediate

data, INPOPT(19)=1, so TIME2 = TIME1 + OUTINT, see Line 60 of the input file. The u and v values at TIME2 are output on the lines immediately below it, Lines 2*MY+3 to 4*MY+2. At each intermediate output interval, the TIME, u, and v data are written to the end of file *uvgraph* in a similar manner.

The data in the graphics output files *etagraph, cgraph1, cgraph2, cgraph3, cgraph4, bedgraph,* and *fracgraph* are intended to be used to generate contour plots of the various variables, see Lines 53 to 59 of the input file in Section 4.1. File *etagraph* contains surface displacements, $\eta_{i,j}$, throughout the solution domain. The units of output η values are in centimeters. The *cgraph* files contain suspended sediment concentration distributions which have units of *mg/liter*.

The file *bedgraph* contains the sediment bed 'thickness' distribution, which needs some further discussion. The units of this 'thickness', Φ, are *grams/cm*^{2} and one would need to specify the porosity of the sediment in order to obtain the actual thickness of the sediment bed. The following formula can be used to determine the sediment thickness (Ziegler et al., 1989)

$$T = \frac{\Phi}{(1-\gamma)\,\rho_s} \tag{2.14}$$

where T is the sediment thickness in *cm*, Φ is expressed in *grams/cm*^{2}, γ is the volume fraction of water in the sediment bed (porosity), and ρ_s is the density of the solid sediment. For the fine-grained sediments examined here, ρ_s = 2.65 *grams/cm*^{3}. The output 'thickness', Φ, is the deviation of the sediment bed with respect to the initial total sediment 'thickness' specified by the modeller, i.e.,

$$\Phi = \sum_{L=1}^{LAYMAX} \phi_L - \sum_{L=1}^{LAYMAX} \phi_{o,L} \tag{2.15}$$

where ϕ_L is the present calculated 'thickness' of layer L and $\phi_{o,L}$ is the initial specified 'thickness' of layer L, $\phi_{o,L}$ = TSED0(L). Areas of net erosion occur where Φ is negative and positive values of Φ correspond to depositional areas. The *fracgraph* file contains the spatial distribution of medium sediment in the sediment bed. The output in this file ranges in value from 0 to 1 with 0 representing a pure coarse sediment and 1 indicates a pure medium sediment.

All of the above contour plot output files have the same structure, which is illustrated in Figure 2.2b, and the format of these files is 15E10.4. If MX is greater than 15, than each line will wrap

around; for a given j value, $1 \le i \le 15$ will be on the first line, then $16 \le i \le 30$ will be on the next line, and so on until i=MX is reached. The first line is free format, and it contains the TIME1 value which is the time, in hours. Similar to file *svgraph*, the contour plotting data can be output at intermediate time intervals.

The files containing time-history output data at specific grid points, *etahist*, *chist*, *sedhist*, and *tauhist*, all have the same format of 21E10.4. The *etahist* file structure is as follows:

Line	Time Period	TIME	ETA(1)	...	ETA(META)
1	1	.xxxxE-xx	.xxxxE-xx	...	.xxxxE-xx
2	2	.xxxxE-xx	.xxxxE-xx	...	.xxxxE-xx
3	3	.xxxxE-xx	.xxxxE-xx	...	.xxxxE-xx
	.	.	.	.	.
	.	.	.	...	.
	.	.	.	...	.

where TIME is in hours, ETA(1) represents $\eta_{i_{meta}(1),j_{meta}(1)}$, ETA(META) represents $\eta_{i_{meta}(meta),j_{meta}(meta)}$, and η values are in centimeters, see Lines 63 to 83 of the input file. The *sedhist* and *tauhist* files are structured in the same manner as file *etahist*, except that the data values are output at ICT(M),JCT(M), M=1,...,MCT. The *chist* file is set up slightly differently in order to account for the concentrations of different sediment size-classes. This file's structure is

Line	Time Period	Size-class	TIME	C(1)	...	C(MCT)
1	1	Fine	.xxxxE-xx	.xxxxE-xx	...	.xxxxE-xx
2	1	Medium	.xxxxE-xx	.xxxxE-xx	...	.xxxxE-xx
3	1	Coarse	.xxxxE-xx	.xxxxE-xx	...	.xxxxE-xx
4	2	Fine	.xxxxE-xx	.xxxxE-xx	...	.xxxxE-xx
5	2	Medium	.xxxxE-xx	.xxxxE-xx	...	.xxxxE-xx
6	2	Coarse	.xxxxE-xx	.xxxxE-xx	...	.xxxxE-xx
7	3	Fine	.xxxxE-xx	.xxxxE-xx	...	.xxxxE-xx
8	3	Medium	.xxxxE-xx	.xxxxE-xx	...	.xxxxE-xx
9	3	Coarse	.xxxxE-xx	.xxxxE-xx	...	.xxxxE-xx
	.	.	.	.	.	...
	.	.	.	...	.	
	.	.	.	...	.	

where it has been assumed that KCT=3, C(1) represents $C_{ist(1),jst(1)}$, and C(MCT) represents $C_{ist(mst),jst(mst)}$. For KCT=2, medium size-class concentrations are output before coarse size-class concentrations at each time interval.

Output of the values of all dependent variables at the end of a calculation to a file called *icout* is also possible, see INPOPT(20) in Section 4.1. This file can then be used as the initial conditions to

start a future run. This option is very useful when a series of calculations need to be made. Se the Lower Fox River example in Section 5.1.

The generality and flexibility that has been built into SEDZL make it a very powerful computer program but these attributes also create the need for an input file which can be quite complex. A user-friendly interface called *setup* has been developed which greatly simplifies input file creation and modification. Installation and use of *setup* are discussed in Sections 2.6 and 4.2, resepectively.

2.5 COMPUTER REQUIREMENTS

The source code for SEDZL consists of a main driver program (ctl.main.f) which calls 83 subroutines and 2 functions. The source code files are divided into three categories:

ctl.*.f - controls input/output, calculates constants;

h.*.f - calculates hydrodynamics;

s.*.f - calculates sediment transport.

The modular design of the source code makes it easy to modify the code. However, code modifications should not be made unless the user is extremely familiar with the structure of the code and its underlying theory.

The code is written in FORTRAN 77, and it has been compiled and executed on the following machines: DEC MicroVAX II, Sun Sparcstation 1, Sun Sparcstation 330, and Convex C-1. The only device-dependent statement in the program is on line 129 of ctl.main.f:

CALL ABRUPT_UNDERFLOW

This statement is specifically for execution on Sun Sparcstations. It should be deleted before the program is compiled on a different computer system. This statement disables the IEEE conformance floating point recalculation. On IEEE conforming computers, this code will take up to twice as long to run, unless the recalculation is turned off. If the user's system has this problem, there may be a similiar command to turn off the recalculation.

The matrices and arrays in the source code are sized to run a problem with a maximum of 2100 grid points and MX= 127 and MY= 50, see Section 3.1 for definitions of MX and MY. This size of

program generates an executable file that requires 8 Mbytes of internal memory. If larger arrays are needed in order to solve a particular problem, then each source code file must be edited. Each matrix that is presently sized 127,50 must be changed to the desired MX,MY values in the REAL, DOUBLE PRECISION, and INTEGER declaration statments of each source code file. Similarly, all arrays currently set at 2100 must be resized to a new maximum total grid point size. Note that the maximum number of grid points is the number actually in the solution domain; grid points outside of the domain *(node-number = 0)* are not counted in this total. The user must keep in mind that increasing array/matrix size will also increase the internal memory requirements of the executable code.

2.6 INSTALLATION OF SEDZL AND setup

Only a few simple commands are needed to compile and install SEDZL and *setup*. The software provided with the SEDZL package is ready to compile on any SunOS machine which has SysV tools and Sun FORTRAN installed on it. If the user has a different computer system, then small changes in two different *Makefiles* may be necessary prior to compiling and installing the software. Please read and understand this entire section before attempting to install these programs. If the user is uncomfortable about installing the software after reading this section, see if there is a System Administrator or someone else available at your site to help you install the codes.

The SEDZL software package is provided in a directory called *sedzl.dir* which contains the four sub-directories *src.dir*, *setup.dir*, *fox.dir*, and *gbay.dir*. The sub-directory *src.dir* contains all of the source code files for SEDZL that were discussed in Section 2.5. Files for the *setup* program are contained in the *setup.dir* sub-directory. Input and output files for the Lower Fox River and Green Bay examples presented in Section 5 are in the *fox.dir* and *gbay.dir* sub-directories, respectively.

2.6.1 USE OF Makefiles

Both SEDZL and *setup* come with *Makefiles* which allow automatic compilation and installation of the software with the *make* program. The *make* program is standard on UNIX systems, and it is also available on many other operating systems as well. The first section of each *Makefile* contains definitions of which compiler to use, what to call the compiled program, where to put it, etc. Thus, it

is fairly easy to modify the *Makefile* included with the SEDZL package to reflect the software available on the user's system.

2.6.2 INSTALLING SEDZL

To compile SEDZL, the user's computer system needs to have a FORTRAN compiler. If *make* is available on the user's system, it can be used to automatically install the software. If *make* is unavailable, simply use the FORTRAN compiler on your system to compile all of the source code files in *sedzl.dir/src.dir*, which are the files ending with *.f*, and place the resulting compiled program in the directory in which it is to be used.

2.6.3 INSTALLING SEDZL USING make

The user should be in the directory *sedzl.dir/src.dir* when executing the instructions in this section. The software package comes with a *Makefile* set up for SEDZL installation on a SunOS system which has the Sun FORTRAN compiler. If you are on such a system, the *Makefile* should not need to be changed. Be sure to read this entire section to make sure that *Makefile* modification is unnecessary. If you are convinced that the provided *Makefile* is compatible with your computer system, simply type *make* and then press the *RETURN/ENTER* key. This command will start the compilation process, which can take up to 30 minutes on a Sun Sparcstation 1. If *make* generates any error messages, the *Makefile* will have to be edited. Modifying the *Makefile* is discussed below. The error messages should give some indication of the nature of the problem. Contacting a System Administrator to help the user diagnose the problem may be necessary.

If no errors are encountered during the *make* process, the compiled program will be ready for installation. Super-user access is usually required for this step. Once super-user access has been obtained, type *make install* and then press the *RETURN/ENTER* key. The SEDZL executable file, *run_sedzl.x*, will then be installed into the proper directory and be available for use.

If the user does not have a SunOS computer system, or if errors were encountered during the *make* process discussed above, then the *Makefile* provided with the SEDZL software package will need to be modified. Only the first section of the *Makefile* will need to be changed in most cases. Do not

edit below the WARNING line unless you know what you are doing. Always make a backup copy of the *Makefile* before making any changes.

The first section of the *Makefile* is where variables are defined to be used in the second section, which contains the rules for compiling the program. To change the variables, simply edit the line in the *Makefile* so that proper execution occurs on your system.

The first variable is *FP*, which defines the FORTRAN compiler used on the system. Most systems use *f77* although some use *fc* instead. The variable *FP* is set by default to *f77*. *FFLAGS* defines what flags to use with the compiler. If there are any special settings for your FORTRAN compiler, put them on this line. Most compilers have some sort of optimization. The highest level of optimization should be used, as the savings in run-time will far outweigh the extra compile-time needed. The optimization flag should be set here. This flag is currently set to *-O3* which is the highest level of optimization available with the Sun FORTRAN compiler. Check your manual for details if you have a different compiler. The flag *-O* works with most compilers, but usually does not produce the best optimization.

The following variables define the directories in which the software and the manual pages are to be put. The variable *BINDIR* is the directory where *make* will put the compiled program. Most systems have a */usr/local/bin* directory where locally installed software is kept. This directory must be in the user's account path in order to run the software. If this directory is not present on your machine, or you wish to place the software in a different directory, change this line. Installing software into this directory usually involves getting super-user access. If you cannot get this access, change *BINDIR* to one of your own directories.

MANDIR is the directory where *make* will put the on-line manual page. On almost every UNIX system, this directory is */usr/man*. *Make* will put the manual page in the *man1* directory just below this directory. Installing the online manual page usually involves getting super-user access. If you cannot get this access, *make* will generate an error message when it tries to install the manual page. This error message can be ignored. The variable *MAN1* defines the manual pages to install in section 1 (user programs) of *Makefile*. Only one manual page is currently available, *sedsl.1*.

The variable *BINNAME* is the name of the compiled SEDZL program. *BINNAME* is currently set to *run.sedzl.x*, but the user can change the name of the executable file if so desired. Using the default setting of *BINNAME*, the example in Section 5.1 could be run by using the command

run.sedzl.x < in.fox.1 > out.fox.1

and all of the output files for run *fox.1* would be created.

You should not need to make any changes in the *Makefile* past this point. Once you have made any changes, save the file and type *make* from the UNIX prompt. As discussed above, this command will start the compilation process.

If *make* gives you any errors at this point, you probably need to edit the *Makefile* again and correct the errors. The error message should give some indication of what the problem is. If you are not sure, try to find a System Administrator to help you diagnose the problem.

Once you successfully complete this step, you can install the SEDZL executable code into the proper directory. Super-user access is usually required for this step. Once super-user access has been obtained, type *make install* and the software will be installed into the proper directories, as discussed above.

At this point, the SEDZL software should be ready to use. Make sure that the *BINDIR* directory is in the user's account path. If you use *csh* as your shell, you may need to type the command *rehash* before the software is executable. If you use a different shell, you may need to logout and login again before using the program.

2.6.4 INSTALLING *setup*

To install *setup*, you need to have a C compiler and a *curses* library. Both are standard on most UNIX systems. C compilers are available on nearly every operating system and *curses* libraries are available on other systems, although they are not standard. If the user's operating system does not have a C compiler and *curses* library, then you will be limited to manual editing of SEDZL input files.

2.6.5 INSTALLING setup USING make

The user should be in the directory *sedzl.dir/setup.dir* when executing the instructions in this section. The software package comes with a *Makefile* configured for *setup* installation on a SunOS system which has the SysV tools. If you are on such a system, the *Makefile* should not need to be changed. Be sure to read this entire section to make sure that *Makefile* modification is unnecessary. If you are convinced that the provided *Makefile* is compatible with your computer system, then simply type *make* and then press the *RETURN/ENTER* key. This command will start the compilation process, which takes about 5 minutes on a Sun Sparcstation 1. If *make* generates any error messages, the *Makefile* will have to be edited. Modifying the *Makefile* is discussed below. The error messages should give some indication of the nature of the problem. Contacting a System Administrator to help the user diagnose the problem may be necessary.

If no errors are encountered during the *make* process, the compiled program will be ready for installation. Super-user access is usually required for this step. Once super-user access has been obtained, type *make install* and then press the *RETURN/ENTER* key. The executable file *setup* will then be installed into the proper directory and be available for use.

If the user does not have a SunOS computer system or if errors were encountered during the *make* process discussed above, then the *Makefile* provided with the SEDZL software package will need to be modified. Only the first section of the *Makefile* will need to be changed in most cases. Do not edit below the WARNING line unless you know what you are doing. Always make a backup copy of the *Makefile* before making any changes.

The first section of the *Makefile* is where variables are defined to be used in the second section, which contains the rules for compiling the program. To change the variables, simply edit the line in the *Makefile* so that proper execution occurs on your system.

The first variable is *CCP*, which defines the C compiler used on the system. This variable is set to *cc*, which is what most systems use. The variable *CFLAGS* defines what flags to use with the compiler. If there are any special settings you need to give your C compiler, put them on this line. This variable is currently set to *-O* which will run it through the standard optimization. Optimization is not

critical to this program. Be sure your compiler supports this flag.

The following variables define the directories in which some of the support files and compiled programs are to be put after compilation. The variable *BINDIR* is the directory where *make* will put the compiled program. Most systems have a */usr/local/bin* directory where locally installed software is kept. This directory must be in your path in order to run the software. If this directory is not present on your machine, or you wish to place the software in a different directory, change this line. Installing software into this directory usually involves getting super-user access. If you cannot get this access, change this to one of your own directories.

The variable *MENUDIR* is the directory where the configurable menus for the *setup* program will be kept. This variable is currently set to */usr/local/lib/setup*. If the */usr/local/lib* directory is not present on your machine, or you wish to place the menus in a different directory, change this line. Installing files into this directory usually involves getting super-user access. If you cannot get this access, change this to one of your own directories.

The variable *MANDIR* is the directory where *make* will put the on-line manual pages. On almost every UNIX system, this directory is */usr/man*. *Make* will put the manual pages in the *man1* and *man5* directories just below this directory. Installing the online manual pages usually involves getting super-user access. If you cannot get this access, *make* will generate an error message during installation of the manual pages. This error message can be ignored. The variable *MAN1* defines the manual pages to install in section 1 (user programs). Only one manual page is provided in this section, *setup.1*. The variable *MAN5* defines the manual pages to install in section 5 (file formats). There is only one manual page in this section, *menu.5*.

The variable *BINNAME* is the name of the compiled *setup* program. This variable is currently set to *setup*, but you can change it to anything you want.

You should not need to make any changes in the *Makefile* past this point. Once you have made any changes, save the file and type *make* from the UNIX prompt. This command will start the compilation process, which was discussed above.

If *make* generates any error messages at this point, you probably need to edit the *Makefile* again and correct the errors. The error message should give some indication of what the problem is. If you are not sure, try to find a System Administrator to help you diagnose the problem.

Once you successfully complete this step, you can install the *setup* executable code into the proper directory. Super-user access is usually required for this step. Once super-user access has been obtained, type *make install* and the software will be installed into the proper directories, as discussed above.

At this point, the *setup* software should be ready to use. Make sure that the *BINDIR* directory is in the user's account path. If you use *csh* as your shell, you may need to type the command *rehash* before the software is executable. If you use a different shell, you may need to logout and login again before using the program.

3. PROBLEM SETUP

Modelling the sediment transport processes in a particular aquatic system requires preparation of various auxiliary input files which are used by the computer program during a simulation. Two files which are required for any type of run are the *node* file and the *depth* file. *Node* is used to specify the geometry of the water body while *depth* contains its bathymetry. Optional files can also be constructed so that time-varying hydrodynamic forcing and sediment loading as well as spatially-varying sediment bed properties can be specified. Details concerning the construction of these files will be presented in this section.

3.1 GEOMETRY: NODE FILE CONSTRUCTION

The *node* file is used to input the geometry of a particular aquatic system, i.e., river, lake, or estuary. Different water bodies can be modelled by simply changing the *node* file; no modification of the computer code is necessary. Each grid point in a problem has a particular number *(node-number)* associated with it. The *node-number* determines what type of numerical element surrounds the grid point. The computer program then uses the *node-numbers* to apply the correct equations to each grid point during calculations.

The use of four basic numerical elements makes it possible to accurately approximate complex geometries even though a uniform, rectangular grid is used. A full element is used for interior points, see Figure 3.1a. Boundaries are defined by using 1/4 elements for 90° corners (Figure 3.1b), 1/2 elements for straight boundaries (Figure 3.1c), and 3/4 elements for 270° corners (Figure 3.1d). Each of these boundary elements can be oriented in 4 different directions with respect to the *x-y* coordinate system. All possible element orientations are illustrated in Figures 3.2 to 3.4. The *node-numbers* corresponding to each element type and orientation are listed in Table 2.1.

Boundaries involving inflow/outflow need some further explanation. A 1/2 element with a specified inflow is a boundary where only incoming flow is allowed. Any outgoing flow generated in the interior of the solution domain will be perfectly reflected by this type of element. Examples of situations where this element type is appropriate are a dam or the upstream limit of a river. A 1/2

element at an open boundary allows both incoming and outgoing flow to pass across it. The incoming flow is specified but the outflow is calculated with negligible reflection and distortion. The total flow is then determined by superposition. A river mouth which is affected by tidal/seiche motion is an example where an open boundary should be used. Partially-open 1/4 elements are used to connect the specified-inflow or open boundary 1/2 elements with other 1/2 element solid boundaries.

Certain element combinations are not allowed due to the structure of the computer code. Elements which *cannot* be next to each other are:

1) full element and element outside solution domain, e.g., 1 0;

2) two 3/4 elements, e.g., 31 32;

3) a 3/4 element and a partially-open 1/4 element, e.g., 34 62.

Other combinations are not possible due to geometric constraints. As discussed in Section 2.3, two open boundaries cannot be joined together by a partially open corner, see Figure 3.5. The computer code checks the *node* file for errors at the beginning of each run. If *node-number* errors are detected, messages are displayed describing the error and its location. Program execution is then halted. An error-free *node* file results in no action or response.

The format of the *node* file is 2013. The file must be structured as follows:

Line	j	i= 1	i= 2	...	i= MX
1	MY	xx	xx	...	xx
2	MY-1	xx	xx	...	xx
		.	.	...	.
		.	.	...	.
		.	.	...	.
MY-1	2	xx	xx	...	xx
MY	1	xx	xx	...	xx

The *i* index increases in the positive *x* -direction while the *j* index increases in the positive *y* -direction. The maximum *i* and *j* indices are MX and MY, respectively. If MX is greater than 20, then each line

will wrap around; for a given j value, $1 \le i \le 20$ will be on the first line, then $21 \le i \le 40$ will be on the next line, and so on until i=MX is reached. Orienting the numerical grid such that either the x or y axis is parallel to the North-South map direction is recommended. Examples of *node* files for the lower Fox River and Green Bay are shown in Section 5.

The size of the rectangular grid element determines the values of MX and MY. Let the overall horizontal lengths of the solution domain in the x- and y- directions be denoted by L_x and L_y, respectively. The total number of grid points in the x- direction is then

$$MX = \frac{L_x}{\Delta x} + 1$$

and in the y- direction

$$MY = \frac{L_y}{\Delta y} + 1$$

where Δx is the grid element size in the x- direction and Δy is the grid element size in the y- direction. Generally, use of a square grid element, i.e., $\Delta x = \Delta y$, produces the best numerical results. Use of a rectangular grid is permissible but the ratio between the two element dimensions should not exceed 5. A rectangular element with a high aspect ratio, greater than 5, can result in inaccurate numerical solutions. The total number of grid elements in the x- and y- directions are $MX-1$ and $MY-1$, respectively.

3.2 BATHYMETRY: DEPTH FILE CONSTRUCTION

Accurately determining the bathymetry of an aquatic system is one of the most important steps in producing a realistic model of the system. Bottom depths must be determined at each grid point in the solution domain and then entered into the *depth* file, which is similar in structure to the *node* file discussed previously. Generally, U.S. Army Corps of Engineers sounding maps are the most accurate bathymetric maps available, and bottom depth data should be taken from them whenever possible. Areas which lack navigation channels, however, may not be mapped by the Corps of Engineers, so one must then rely on NOAA maps, usually. Note that the water depths on these maps are referenced to some datum, e.g., Great Lakes datum or mean low water. Seasonal or tidal variations may change the actual mean water depth considerably. The computer program has an option by which adjustments can

be made for these long-term mean depth changes, see Line 22 of the input file in Section 4.1. Thus, it is recommended that the *depth* file contain the datum-referenced water depths from the maps used.

The format of the *depth* file is 20F6.0. Similar to the *node* file, the *depth* file must be structured as follows:

Line	j	i= 1	i= 2	...	i= MX
1	MY	xxx.	xxx.	...	xxx.
2	MY-1	xxx.	xxx.	...	xxx.
		.	.	...	.
		.	.	...	.
		.	.	...	.
MY-1	2	xxx.	xxx.	...	xxx.
MY	1	xxx.	xxx.	...	xxx.

Examples of *depth* files for the lower Fox River and Green Bay are included with the SEDZL software package.

All depths must be input in centimeters and be positive. The maximum depth possible is 99999 cm (999.99 meters or 3280 feet). Zero depths are not permitted. The computer program will check the *depth* file to make sure that each grid point in the solution domain has a depth assigned to it. If a grid point has zero depth, program execution will stop and an error message stating the location of the zero depth will be printed out. No action or response results from an error free *depth* file.

3.3 TIME-VARYING HYDRODYNAMIC FORCING

Real rivers, lakes, and estuaries are subjected to forces which are continually changing. In order to properly model the transient nature of these systems, it is possible to construct files used by the computer program which specify time-varying wind stresses, flow rates, and water level variations (tide/seiche). Construction of these files, *wind*, *inflow*, and *tide*, will now be discussed. The program options which enable the use of these files are discussed in Section 4.1.

3.3.1 WIND STRESSES

Shear stresses due to time-varying winds can be imposed by using the *wind* file. The wind shear stress is assumed to be spatially constant over the entire solution domain. Three wind parameters are specified in the *wind* file: direction, velocity, and air temperature. The velocity must be input in centimeters/second and be positive. The air temperature is used to adjust for air density variations when calculating the wind shear stress and it must be expressed in degrees Celsius.

Sixteen different wind directions can be specified with a difference of 22.5° between each direction. Each direction is denoted by a number, ranging from 1 to 16. The orientation of each direction with respect to the *x*-*y* coordinate system of the numerical grid is shown in Figure 3.6. Note that the orientation of the numerical grid with respect to North must be determined in order to correctly convert wind directions obtained from meteorological data to the directions used by the computer program. The wind direction specified means that the wind is blowing from that direction.

The *wind* file is constructed such that the direction, velocity, and temperature at a specific time are entered on a line. Each line of the file represents the wind conditions at a different time and it is assumed that the data are uniformly spaced in time. Specification of the time interval, assumed to be in hours, between each wind data point is discussed in Section 4.1. The *wind* file must be structured as follows:

Line	Time period	DIR.	VEL.	TEMP.
1		NWIND		
2	1	xxx.	xxx.	xxx.
3	2	xxx.	xxx.	xxx.
	.	.	.	
	.	.	.	
	.	.	.	
NWIND+1	NWIND	xxx.	xxx.	xxx.

NWIND is the total number of time periods listed in the file. For the program to function properly, NWIND must be one time period longer than the actual length of the calculation. For example, if a 96 hour long run was to be made and the time interval between each wind data input was 1 hour, then NWIND should be 97 and one additional line of wind data is needed after hour 96. The format of each data line is 3F6.0 and columns 1 through 3 contain direction, velocity, and temperature, respectively. A *wind* file is included with the Green Bay example, see Section 5.2.

3.3.2 FLOW RATES

Time-varying flow rates, either in m^3/s or *cfs*, can be specified by using the *inflow* file. Input flow rates must be uniformly spaced in time and the time interval between each value must be specified in hours. The program can vary the flow rate between two consecutive time periods either by a discontinuous step-function or a linear ramping. Details on this subject are presented in Section 4.1.

The format of the *inflow* file is 20F6.0, and it is constructed as follows:

Line	Time period				
1		FLOW			
2		QOLD			
3	1 - 20	xxx.	xxx.	...	xxx.
4	21 - 40	xxx.	xxx.	...	xxx.
5	41 - 60	xxx.	xxx.	...	xxx.
		.	.	.	
		.	.	.	
		.	.	.	

Flow rate values are entered in *inflow* as shown above until the last time period is reached. One additional flow rate value is necessary after the last time period value for proper ramping of flow rates at the end of the calculation. All flow rates must be positive and can be expressed either as m^3/s or *cfs*. The variable FLOW on the first line of *inflow* determines the units of the flow rates in the file. Enter *cfs* on the first line for flow rates in cfs and set FLOW equal to *meter* for flow rates in m^3/s. The

variable QOLD on the second line of *inflow* is the flow rate value at the start of the calculation, see Figure 4.2. A *inflow* file is included with the lower Fox River example, see Section 5.1.

3.3.3 WATER LEVEL VARIATIONS (TIDE/SEICHE)

The water level, with respect to h_o + *DELTAH*, at an open boundary can be specified by using the *tide* file. The time-varying water level, $\eta(t)$, must be specified uniformly in time with the interval between each $\eta(t^n)$ value being measured in hours. The units of $\eta(t^n)$ are in centimeters and it is measured with respect to the equilibrium depth at the open boundary where it is specified. Thus, $\eta(t^n)$ can have both positive and negative values. At the open boundary, η is varied linearly between the values of $\eta(t^n)$ and $\eta(t^{n+1})$.

The *tide* file is constructed as follows:

Line	Time period				
1		NLINE	TTIME		
2	1 - 30	xxx.	xxx.	...	xxx.
3	31 - 60	xxx.	xxx.	...	xxx.
4	61 - 90	xxx.	xxx.	...	xxx.
		.	.	.	
		.	.	.	
		.	.	.	
NLINE+1		xxx.	xxx.	...	xxx.

where NLINE is the total number of lines of data to read and TTIME is the time period between each $\eta(t^n)$ in hours. The first line is free format while the water level data lines have a format of 20F5.0. If the last data line has less than 20 data entries in it, then zeros must be added after the last time period value in line NLINE+1 so that the line has 20 entries in it. One additional water elevation must added after the last data point for the program to function correctly. A *tide* file is included with the lower Fox River example, see Section 5.1.

3.4 TIME-VARYING SEDIMENT LOADING

Time-varying sediment concentrations, in mg/liter units, can be specified in much the same manner as the flow rates discussed in Section 3.3.2 by using a file called *sedload*. Specified concentrations in *sedload* are uniformly spaced in time with the interval between each concentration being in hours. The concentration profile between two consecutive time periods can be varied either as a step-function or a linear ramping, see Section 4.1 for further details.

Sediment suspended in the water column consists of a wide range of particle sizes. As discussed in Section 2.3, the computer program can model the transport of three size classes of particles: fine, medium, and coarse. The fraction of the total sediment load (concentration) of each of these size classes can be specified in the *sedload* file.

The format of the *sedload* file is 20F6.0 and it is constructed as follows:

Line	Time period	Size-class				
1			COLD(k), k=1,...,KCT			
2	1 - 20	Fine	xxx.	xxx.	...	xxx.
3	1 - 20	Medium	xxx.	xxx.	...	xxx.
4	1 - 20	Coarse	xxx.	xxx.	...	xxx.
5	21 - 40	Fine	xxx.	xxx.	...	xxx.
6	21 - 40	Medium	xxx.	xxx.	...	xxx.
7	21 - 40	Coarse	xxx.	xxx.	...	xxx.
8	41 - 60	Fine	xxx.	xxx.	...	xxx.
9	41 - 60	Medium	xxx.	xxx.	...	xxx.
10	41 - 60	Coarse	xxx.	xxx.	...	xxx.
	.	.	.			
	.	.	.			
	.	.	.			

Concentration values are entered in *sedload* as shown above until the last time period is reached. Similar to an *inflow* file, one additional concentration for each sediment size-class is necessary after the last time period value for proper ramping of concentrations at the end of the calculation. All concentrations must be expressed in *mg/liter* and the variable COLD(k) on the first line of the file is the value of the concentration of sediment size-class *k* at the start of the calculation, see Figure 4.2. If KCT=1, then only medium size-class concentrations are sprecified. If KCT=2, then k=1 denotes medium size-class and k=2 denotes coarse size-class. If KCT=3, then k=1 denotes fine size-class, k=2 denotes medium size-class, and k=3 denotes coarse size-class. The example shown is for KCT=3. Lines 2 to 4 are the sediment concentrations for the three size-classes during time periods 1 to 20. Lines 5 to 7 are for time periods 21 to 40 and so on. If KCT=2, then only lines 2 and 3 would represent time periods 1 to 20 with line 2 being concentrations for the medium size-class and line 3 being for the coarse size-size class. For KCT=1, then only line 2 would be present for the medium size-class during time periods 1 to 20. An example of a *sedload* file can be found in Section 5.1.

3.5 SPATIALLY-VARYING SEDIMENT BED PROPERTIES

The sediment bed consists of a mixture of medium and coarse particles. The initial spatial distribution of the fraction of medium sediment particles in the sediment bed can be specified by using the *sedbed* file. The fraction of medium particles in the sediment bed can range from 0 to 1, with 0 being a pure coarse bed and 1 being a pure medium bed. The format of *sedbed* is 15E10.4 and its structure is similar to the *node* and *depth* files, i.e.,

Line	j	i= 1	i= 2	...	i= MX
1	MY	.xxxxE-xx	.xxxxE-xx	...	.xxxxE-xx
2	MY-1	.xxxxE-xx	.xxxxE-xx	...	.xxxxE-xx
		.	.	...	.
		.	.	...	.
		.	.	...	.
MY-1	2	.xxxxE-xx	.xxxxE-xx	...	.xxxxE-xx
MY	1	.xxxxE-xx	.xxxxE-xx	...	.xxxxE-xx

4. INPUT FILE SETUP

The generality and flexibility of SEDZL make it a very powerful tool for analyzing sediment transport processes in rivers, lakes, and estuaries. However, these same attributes necessitate the use of an input file which can be rather complex. A user-friendly interface called *setup* has been created in order to make input file construction and modification an easy task.

This section first describes all of the input variables that can be included in the input file on a line-by-line basis. Various program options are also discussed. Next, *setup* use is presented in detail. Finally, manual input file setup is described for SEDZL users who do not have access to a computer with a UNIX operating system.

4.1 INPUT FILE PARAMETERS AND PROGRAM OPTIONS

The input file can have a possible length of 107 lines and input/output to 27 different files. A total of 24 options are available for controlling program execution. Summaries of the input file structure and program options can be found in Tables 4.1 and 4.2, respectively.

Each input line will now be described. Lines 1 to 8 of the input file must always be included. All lines past Line 8 are optional, meaning that each line is included only if the option variable, INPOPT(n), for that particular line is set to the value indicated in Table 4.1. If INPOPT(n) = 0 for a particular line, then that line is not to be included. The FORTRAN data type of each variable is shown in parantheses.

Line 1: RUNNO

Label for this run. (CHARACTER*64)

Line 2: INPOPT(n), n =1,...,24

Program option variables, 24 total. (INTEGER)

Line 3: MX MY XSTP YSTP

MX = maximum i index in x-direction (INTEGER)
MY = maximum j index in y-direction (INTEGER)
XSTP = grid element size in x-direction, in centimeters (REAL)
YSTP = grid element size in y-direction, in centimeters (REAL)

- Generally, the ratio between XSTP and YSTP should be less than 5, see Section 3.1.

Line 4: *node* FILE NAME

See Section 3.1 for a description of this file. (CHARACTER*64)

Line 5: *depth* FILE NAME

See Section 3.2 for a description of this file. (CHARACTER*64)

Line 6: TSTP IRTSTP TMAX

TSTP = hydrodynamic timestep, in seconds (REAL)
IRTSTP = integer to multiply TSTP by to determine sediment transport timestep (INTEGER)
TMAX = maximum time for this run, in hours (REAL)

- If the hydrodynamic timestep is 10 s and a sediment transport timestep of 50 s is desired, then IRTSTP = 5.
- IRTSTP ≥ 1

Line 7: AH CF

AH = eddy viscosity, in cm^2/s (REAL)
CF = friction factor for bottom stress (REAL)

Line 8: U0 UL V0 VL

U0 = constant specified inflow at Plus x 1/2 element, node-number=41 (REAL)
UL = constant specified inflow at Minus x 1/2 element, node-number=42 (REAL)
V0 = constant specified inflow at Plus y 1/2 element, node-number=43 (REAL)
VL = constant specified inflow at Minus y 1/2 element, node-number=44 (REAL)

- See Figure 4.1 for orientations of U0, UL, V0, VL.
- Positive values of U0, UL, V0, VL produce incoming flows as shown in Figure 4.1
- Units of U0 - VL are in cm^2/s (integrated velocities) unless INPOPT(11) > 0, see INPOPT(11) for details.
- These inflows are not time-variable unless INPOPT(12) is used, see Lines 26 to 29.

Line 9: DH KCT (include only if INPOPT(1)=1)

DH = eddy diffusivity, in cm^2/s (REAL)
KCT = total number of sediment particle size-classes, maximum of 3 (INTEGER)

- If KCT = 1, then medium size-class particles only.
- If KCT = 2, then medium and coarse size-class particles.
- If KCT = 3, then fine, medium, and coarse size-class particles.

Line 10: CX0(k), k=1,...,KCT (include only if INPOPT(1)=1)

Specified concentration of size-class k, in mg/liter, at Plus x 1/2 element (REAL)

- If KCT = 1, then medium size-class particles only.
- If KCT = 2, then k=1 is medium size-class and k=2 is coarse size-class.
- If KCT = 3, then k=1 is fine size-class, k=2 is medium size-class and k=3 is coarse size-class.
- These concentrations are not time-variable.
- These concentrations are generally applied at a specified inflow, *node-number* =41. However, using these concentrations at an open boundary, *node-number* =51, will result in the value of CX0(k) being applied if the velocity at the open boundary is incoming, i.e., reversed flow.

Line 11: CXL(k), k=1,...,KCT (include only if INPOPT(1)=1)

Specified concentration of size-class k, in mg/liter, at Minus x 1/2 element (REAL)

- If KCT = 1, then medium size-class particles only.
- If KCT = 2, then k=1 is medium size-class and k=2 is coarse size-class.
- If KCT = 3, then k=1 is fine size-class, k=2 is medium size-class and k=3 is coarse size-class.
- These concentrations are not time-variable.
- These concentrations are generally applied at a specified inflow, *node-number* =42. However, using these concentrations at an open boundary, *node-number* =52, will result in the value of CXL(k) being applied if the velocity at the open boundary is incoming, i.e., reversed flow.

Line 12: CY0(k), k=1,...,KCT (include only if INPOPT(1)=1)

Specified concentration of size-class k, in mg/liter, at Plus y 1/2 element (REAL)

- If KCT = 1, then medium size-class particles only.
- If KCT = 2, then k=1 is medium size-class and k=2 is coarse size-class.
- If KCT = 3, then k=1 is fine size-class, k=2 is medium size-class and k=3 is coarse size-class.
- These concentrations are not time-variable.
- These concentrations are generally applied at a specified inflow, *node-number* =43. However, using these concentrations at an open boundary, *node-number* =53, will result in the value of CY0(k) being applied if the velocity at the open boundary is incoming, i.e., reversed flow.

Line 13: CYL(k), k=1,...,KCT (include only if INPOPT(1)=1)

Specified concentration of size-class k, in mg/liter, at Minus y 1/2 element (REAL)

- If KCT = 1, then medium size-class particles only.
- If KCT = 2, then k=1 is medium size-class and k=2 is coarse size-class.
- If KCT = 3, then k=1 is fine size-class, k=2 is medium size-class and k=3 is coarse size-class.
- These concentrations are not time-variable.

- These concentrations are generally applied at a specified inflow, *node-number* =44. However, using these concentrations at an open boundary, *node-number* =54, will result in the value of CY0(k) being applied if the velocity at the open boundary is incoming, i.e., reversed flow.

Line 14: A0IN ALPHA DMAX RESEXP BETA CB TCRDEP (include only if INPOPT(1)=1)

A0IN = a_o in resuspension equation, Eq. (2.8) (REAL)
ALPHA = α in flocculation equation, Eq. (2.6) (REAL)
DMAX = maximum floc diameter, in microns (REAL)
RESEXP = exponent 'n' in resuspension equation, Eq. (2.8) (REAL)
BETA = settling speed of coarse size-class particles, in microns/second (REAL)
CB = bottom shear stress factor for boundary elements (REAL)
TCRDEP = critical bottom shear stress for no deposition of medium size-class (REAL)

- For a boundary element, $\tau^b = CB\ \tau_i^b$ where τ_i^b is the bottom shear at the nearest interior grid point and $0 \le CB \le 1$. RECOMMENDATION: use CB = 1.0.
- See Sections 2.1 and 5 for discussions on appropriate values of a_o, α, DMAX, RESEXP, BETA, and TCRDEP.

Line 15: LAYMAX (include only if INPOPT(1)=1)

- Total number of layers in sediment bed, see Sections 2.1, 2.3 and 5 (INTEGER)
- Maximum value of LAYMAX is 14.

Line 16: FTIME(L), L=1,...LAYMAX (include only if INPOPT(1)=1)

FTIME(L) = time after deposition of sediment bed layer 1 (REAL)

- FTIME has units of days and is t_d in Eq. (2.8), see Sections 2.1, 2.3 and 5.

Line 17: TSED0(L), L=1,...LAYMAX (include only if INPOPT(1)=1)

TSED0(L) = initial 'thickness' of sediment bed layer L (REAL)

- TSED0 has units of *grams/cm*^{2} so the actual thickness of the layer can be determined once the porosity of the layer is specified, see Sections 2.1, 2.3 and 5.

Line 18: TAUCR(L), L=1,...LAYMAX (include only if INPOPT(1)=1)

TAUCR(L) = critical bottom shear stress of sediment bed layer L (REAL)

- TAUCR has units of *dynes/cm*^{2} and is τ_o in Eq. (2.8), see Sections 2.1, 2.3 and 5.

Line 19: P0 (include only if INPOPT(1)=1 and KCT>1)

P0 = fraction of medium size-class particles in initial sediment bed (REAL)

- Values of P0 must be such that $0 \le P0 \le 1$. For P0=1, the initial sediment bed consists of pure medium-size class particles. For P0=0, the initial sediment bed is 100% coarse-size class particles.
- P0 is assumed to be constant throughout the initial sediment bed, i.e., for each sediment bed layer at all grid points.

Line 20: CTIC(k), k=1,...KCT (include only if INPOPT(1)=1)

CTIC(k) = *initial concentration of size-class k, in mg/liter* (REAL)

- CTIC(k) is constant throughout the solution domain.
- If KCT = 1, then medium size-class particles only.
- If KCT = 2, then k=1 is medium size-class and k=2 is coarse size-class.
- If KCT = 3, then k=1 is fine size-class, k=2 is medium size-class and k=3 is coarse size-class.

Line 21: *icin* FILE NAME

icin is a file generated at the end of a previous run. This file contains all of the hydrodynamic and sediment transport data needed to start the current calculation from the point where the previous run ended. File *icin* for the current run is the same as file *icout* of the previous run, see Line 61 and INPOPT(20). (CHARACTER*64)

Line 22: DELTAH (include only *if* INPOPT(3)=1)

DELTAH = constant depth added to all bottom depths, in centimeters (REAL)

- Generally, bottom depths used in the *depth* file have been measured with respect to some datum. Long-term lake level or tidal variations may cause the actual mean water depth to be significantly different from the depths specified in the *depth* file. DELTAH can be used to adjust the bathymetry so that the correct mean water depth is being used.
- Positive DELTAH adds to the bottom depths while negative DELTAH subtracts from the bottom depths. DELTAH is constant throughout the solution domain.

Line 23: *tide* FILE NAME (include only if INPOPT(9)=1)

See Section 3.3.3 for a description of this file. (CHARACTER*64)

Line 24: *wind* FILE NAME (include only if INPOPT(6)=1)

See Section 3.3.1 for a description of this file. (CHARACTER*64)

Line 25: TWIND (include only if INPOPT(6)=1 and INPOPT(7)=1)

TWIND is the time, in hours, between each wind measurement used in the *wind* file, see Section 3.3.1. (REAL)

Line 26: NRAMP(1) (include only if INPOPT(12)=1)

NRAMP(1) = number of hydrodynamic timesteps to linearly increase U0 (INTEGER)

- This option is to be used only when a constant inflow at a Plus x 1/2 element, *node-number* =41, is desired, see Line 8.
- The flow rate will be linearly ramped from 0 at the start of the calculation to U0 over a time period of NRAMP*TSTP seconds. Use of this option will solve numerical problems associated with sharp wave fronts generated by a discontinuous increase in flow rate, especially when large U0 values are used.

Line 27: NRAMP(2) (include only if INPOPT(12)=1)

NRAMP(2) = number of hydrodynamic timesteps to linearly increase UL (INTEGER)

- This option is to be used only when a constant inflow at a Minus x 1/2 element, *node-number* =42, is desired, see Line 8.

- The flow rate will be linearly ramped from 0 at the start of the calculation to UL over a time period of NRAMP*TSTP seconds. Use of this option will solve numerical problems associated with sharp wave fronts generated by a discontinuous increase in flow rate, especially when large UL values are used.

Line 28: NRAMP(3) (include only if INPOPT(12)=1)

NRAMP(3) = number of hydrodynamic timesteps to linearly increase V0 (INTEGER)

- This option is to be used only when a constant inflow at a Plus y 1/2 element, *node-number* =43, is desired, see Line 8.
- The flow rate will be linearly ramped from 0 at the start of the calculation to V0 over a time period of NRAMP*TSTP seconds. Use of this option will solve numerical problems associated with sharp wave fronts generated by a discontinuous increase in flow rate, especially when large V0 values are used.

Line 29: NRAMP(4) (include only if INPOPT(12)=1)

NRAMP(4) = number of hydrodynamic timesteps to linearly increase VL (INTEGER)

- This option is to be used only when a constant inflow at a Minus y 1/2 element, *node-number* =44, is desired, see Line 8.
- The flow rate will be linearly ramped from 0 at the start of the calculation to VL over a time period of NRAMP*TSTP seconds. Use of this option will solve numerical problems associated with sharp wave fronts generated by a discontinuous increase in flow rate, especially when large VL values are used.

Line 30: NQIN (include only if INPOPT(13)=1)

Total number of *inflow* files to input, see Section 3.3.2. (INTEGER)

Line 31: *inflow1* FILE NAME (include only if INPOPT(13)=1)

See Section 3.3.2 for a description of this file. (CHARACTER*64)

Line 32: *inflow2* FILE NAME (include only if INPOPT(13)=1 and NQIN ≥ 2)

See Section 3.3.2 for a description of this file. (CHARACTER*64)

Line 33: *inflow3* FILE NAME (include only if INPOPT(13)=1 and NQIN ≥ 3)

See Section 3.3.2 for a description of this file. (CHARACTER*64)

Line 34: *inflow4* FILE NAME (include only if INPOPT(13)=1 and NQIN=4)

See Section 3.3.2 for a description of this file. (CHARACTER*64)

Line 35: NODEQ(1) NTS(1) TS(1) TR(1) (include only if INPOPT(13)=1)

NODEQ(1) = node-number at which to specify time-variable inflow (INTEGER)
NTS(1) = total number of inflow values to read from 'inflow1' file (INTEGER)
TS(1) = time between each inflow value, in hours (REAL)
TR(1) = ramping time, in hours (REAL)

- NODEQ(1) can equal 41, 42, 43, or 44 depending on which specified inflow 1/2 element that *inflow1* is to be applied at.

- TR(1) is used to control how the specified inflow varies between two consecutive values. Now, $0 \le TR(1) \le TS(1)$. For TR(1)=0, the specified inflow varies discontinuously. For TR(1)=TS(1), the specified inflow varies linearly between the two consecutive values, see Figure 4.2.

Line 36: NODEQ(2) NTS(2) TS(2) TR(2) (include only if INPOPT(13)=1 and NQIN ≥ 2)

NODEQ(2) = node-number at which to specify time-variable inflow (INTEGER)
NTS(2) = total number of inflow values to read from 'inflow2' file (INTEGER)
TS(2) = time between each inflow value, in hours (REAL)
TR(2) = ramping time, in hours (REAL)

- NODEQ(2) can equal 41, 42, 43, or 44 depending on which specified inflow 1/2 element that *inflow2* is to be applied at.
- NODEQ(1) ≠ NODEQ(2)
- TR(2) can be used to control how the specified inflow varies between two consecutive values. Now, $0 \le TR(2) \le TS(2)$. For TR(2)=0, the specified inflow varies discontinuously. For TR(2)=TS(2) the specified inflow varies linearly between the two consecutive values, see Figure 4.2.

Line 37: NODEQ(3) NTS(3) TS(3) TR(3) (include only if INPOPT(13)=1 and NQIN ≥ 3)

NODEQ(3) = node-number at which to specify time-variable inflow (INTEGER)
NTS(3) = total number of inflow values to read from 'inflow3' file (INTEGER)
TS(3) = time between each inflow value, in hours (REAL)
TR(3) = ramping time, in hours (REAL)

- NODEQ(3) can equal 41, 42, 43, or 44 depending on which specified inflow 1/2 element that *inflow3* is to be applied at.
- NODEQ(1) ≠ NODEQ(2) ≠ NODEQ(3)
- TR(3) can be used to control how the specified inflow varies between two consecutive values. Now, $0 \le TR(3) \le TS(3)$. For TR(3)=0, the specified inflow varies discontinuously. For TR(3)=TS(3), the specified inflow varies linearly between the two consecutive values, see Figure 4.2.

Line 38: NODEQ(4) NTS(4) TS(4) TR(4) (include only if INPOPT(13)=1 and NQIN=4)

NODEQ(4) = node-number at which to specify time-variable inflow (INTEGER)
NTS(4) = total number of inflow values to read from 'inflow4' file (INTEGER)
TS(4) = time between each inflow value, in hours (REAL)
TR(4) = ramping time, in hours (REAL)

- NODEQ(4) can equal 41, 42, 43, or 44 depending on which specified inflow 1/2 element that *inflow4* is to be applied at.
- NODEQ(1) ≠ NODEQ(2) ≠ NODEQ(3) ≠ NODEQ(4)
- TR(4) can be used to control how the specified inflow varies between two consecutive values. Now, $0 \le TR(4) \le TS(4)$. For TR(4)=0, the specified inflow varies discontinuously. For TR(4)=TS(4),the specified inflow varies linearly between the two consecutive values, see Figure 4.2.

Line 39: NCIN (include only if INPOPT(1)=1 and INPOPT(13)=1)

Total number of *sedload* files to input, see Section 3.4. (INTEGER)

Line 40: *sedload1* FILE NAME (include only if INPOPT(1)=1 and INPOPT(13)=1)
See Section 3.4 for a description of this file. (CHARACTER*64)

Line 41: *sedload2* FILE NAME (include only if INPOPT(1)=1 and INPOPT(13)=1 and NCIN ≥ 2)
See Section 3.4 for a description of this file. (CHARACTER*64)

Line 42: *sedload3* FILE NAME (include only if INPOPT(1)=1 and INPOPT(13)=1 and NCIN ≥ 3)
See Section 3.4 for a description of this file. (CHARACTER*64)

Line 43: *sedload4* FILE NAME (include only if INPOPT(1)=1 and INPOPT(13)=1 and NCIN=4)
See Section 3.4 for a description of this file. (CHARACTER*64)

Line 44: NODEC(1) NTC(1) TSC(1) TRC(1) (include only if INPOPT(1)=1 and INPOPT(13)=1)

NODEC(1) = node-number at which to specify time-variable concentration (INTEGER)
NTC(1) = total number of concentration values to read from 'sedload1' file (INTEGER)
TSC(1) = time between each concentration value, in hours (REAL)
TRC(1) = ramping time, in hours (REAL)

- NODEC(1) can equal 41, 42, 43, or 44 depending on which specified inflow 1/2 element that *sedload1* is to be applied at.
- TRC(1) can be used to control how the specified concentration varies between two consecutive values. Now, $0 \le TRC(1) \le TSC(1)$. For TRC(1)=0, the specified concentration varies discontinuously. For TRC(1)=TSC(1), the specified concentration varies linearly between the two consecutive values, see Figure 4.2.

Line 45: NODEC(2) NTC(2) TSC(2) TRC(2) (include only if INPOPT(2)=1 and INPOPT(13)=1 and NCIN ≥ 2)

NODEC(2) = node-number at which to specify time-variable concentration (INTEGER)
NTC(2) = total number of concentration values to read from 'sedload2' file (INTEGER)
TSC(2) = time between each concentration value, in hours (REAL)
TRC(2) = ramping time, in hours (REAL)

- NODEC(2) can equal 41, 42, 43, or 44 depending on which specified inflow 1/2 element that *sedload2* is to be applied at.
- NODEC(1) ≠ NODEC(2)
- TRC(2) can be used to control how the specified concentration varies between two consecutive values. Now, $0 \le TRC(2) \le TSC(2)$. For TRC(2)=0, the specified concentration varies discontinuously. For TRC(2)=TSC(2), the specified concentration varies linearly between the two consecutive values, see Figure 4.2.

Line 46: NODEC(3) NTC(3) TSC(3) TRC(3) (include only if INPOPT(3)=1 and INPOPT(13)=1 and NCIN ≥ 3)

NODEC(3) = node-number at which to specify time-variable concentration (INTEGER)
NTC(3) = total number of concentration values to read from 'sedload3' file (INTEGER)
TSC(3) = time between each concentration value, in hours (REAL)
TRC(3) = ramping time, in hours (REAL)

- NODEC(3) can equal 41, 42, 43, or 44 depending on which specified inflow 1/2 element that *sedload3* is to be applied at.

- NODEC(1) ≠ NODEC(2) ≠ NODEC(3)
- TRC(3) can be used to control how the specified concentration varies between two consecutive values. Now, 0 ≤ TRC(3) ≤ TSC(3). For TRC(3)=0, the specified concentration varies *discontinuously*. For TRC(3)=TSC(3), the specified concentration varies linearly between the two consecutive values, see Figure 4.2.

Line 47: NODEC(4) NTC(4) TSC(4) TRC(4) (include only if INPOPT(4)=1 and INPOPT(13)=1 and NCIN = 4)

NODEC(4) = node-number at which to specify time-variable concentration (INTEGER)
NTC(4) = total number of concentration values to read from 'sedload4' file (INTEGER)
TSC(4) = time between each concentration value, in hours (REAL)
TRC(4) = ramping time, in hours (REAL)

- NODEC(4) can equal 41, 42, 43, or 44 depending on which specified inflow 1/2 element that *sedload4* is to be applied at.
- NODEC(1) ≠ NODEC(2) ≠ NODEC(3) ≠ NODEC(4)
- TRC(4) can be used to control how the specified concentration varies between two consecutive values. Now, 0 ≤ TRC(4) ≤ TSC(4). For TRC(4)=0, the specified concentration varies discontinuously. For TRC(4)=TSC(4), the specified concentration varies linearly between the two consecutive values, see Figure 4.2.

Line 48: FCOR (include only if INPOPT(8)=1)

FCOR is the Coriolis parameter, f, and has units of s^{-1}. (REAL)

Line 49: ASEI OMSEI (include only if INPOPT(10)=1)

ASEI = amplitude of sinusoidally-varying tide/seiche, in centimeters (REAL)
OMSEI = period of sinusoidally-varying tide/seiche, in hours (REAL)

- For use at an open boundary where $\eta(t) = A \sin \omega t$ with A = ASEI and $\omega = 2\pi/(3600 OMSEI)$.

Line 50: NODTID (include only if INPOPT(9)=1 or INPOPT(10)=1)

NODTID is the *node-number* of the open boundary at which to apply either a specified time-varying tide/seiche (INPOPT(9)=1) or a sinusoidally-varying tide/seiche (INPOPT(10)=1). NODTID can equal 51, 52, 53, or 54. (INTEGER)

Line 51: *sedbed* FILE NAME (include only if INPOPT(14)=1)

See Section 3.5 for a description of this file. (CHARACTER*64)

Line 52: *uvgraph* FILE NAME (include only if INPOPT(15)=1 or 3)

Velocity graphics output file, see Section 2.4 for a description of this file. Units of output velocities are *cm/s*. (CHARACTER*64)

Line 53: *etagraph* FILE NAME (include only if INPOPT(16)=1 or 3)

Surface displacement graphics output file, see Section 2.4 for a description of this file. Units of output η values are *cm*. (CHARACTER*64)

Line 54: *cgraphl* FILE NAME (include only if INPOPT(17)=1 or 3)

Total concentration graphics output file, see Section 2.4 for a description of this file. Units of output concentrations are *mg/liter*. (CHARACTER*64)

Line 55: cgraph2 FILE NAME (include only if INPOPT(17)=1 or 3 and KCT=3)
Line 56: cgraph3 FILE NAME (include only if INPOPT(17)=1 or 3 and KCT>1)
Line 57: cgraph4 FILE NAME (include only if INPOPT(17)=1 or 3 and KCT>1)

- *cgraph2* is the graphics output file for fine size-class concentration. (CHARACTER*64)
- *cgraph3* is the graphics output file for medium size-class concentration. (CHARACTER*64)
- *cgraph4* is the graphics output file for coarse size-class concentration. (CHARACTER*64)
- See Section 2.4 for a description of these files. Units of output concentrations are *mg/liter*.

Line 58: *bedgraph* FILE NAME (include only if INPOPT(18)=1 or 3)

Sediment bed thickness graphics output file, see Section 2.4 for a description of this file. Units of output sediment bed 'thickness' are $grams/cm^2$.(CHARACTER*64)

Line 59: *fracgraph* FILE NAME (include only if INPOPT(18)=1 or 3 and KCT>1)

Graphics output file for fraction of medium size-class particle in the sediment bed, see Section 2.4 for a description of this file. (CHARACTER*64)

Line 60: OUTINT (include only if INPOPT(19)=1)

Time interval, in hours, between each intermediate output, either graphics or hard copy. (REAL)

Line 61: *icout* FILE NAME (include only if INPOPT(20)=1)

icout is a file generated at the end of the current run. This file contains all of the necessary hydrodynamic and sediment transport data to start a future calculation from the point where the current run ended. File *icout* for the current run is the same as file *icin* of a future run, see Line 21 and INPOPT(3). (CHARACTER*64)

Line 62: *etahist* FILE NAME (include only if INPOPT(21)=1)

- Output file for time-history of surface displacement, $\eta(t)$, at specific grid points, see Section 2.4 for a description of this file. (CHARACTER*64)
- $\eta(t)$ at each timestep during the calculation will be output.

Line 63: META (include only if INPOPT(21)=1)

Total number of grid points at which to output time history of η. Maximum of 20 grid points can be output. (INTEGER)

Lines 64 to 83: IETA(m) JETA(m) (include only if INPOPT(21)=1)

IETA(m) = i index of point m at which to output $\eta(t)$ (INTEGER)
JETA(m) = j index of point m at which to output $\eta(t)$ (INTEGER)

- Each grid point (i,j) at which $\eta(t)$ is to be output is denoted by (IETA(m),JETA(m)). A total of META lines, each containing a single (IETA(m),JETA(m)) pair, must be included.

Line 84: *chist* FILE NAME (include only if INPOPT(22)=1)

Output file for time-history of sediment concentrations, $C_k(t)$, at specific grid points, see Section 2.4 for a description of this file. (CHARACTER*64)

Line 85: MCT CTOUT (include only if INPOPT(22)=1)

MCT = total number of grid points at which to output $C_k(t)$ (INTEGER)
CTOUT = time interval, in hours, between each $C_k(t)$ output (REAL)

- Maximum of 20 grid points can be output.

Lines 86 to 105: ICT(m) JCT(m) (include only if INPOPT(22)=1)

ICT(m) = i index of point m at which to output $C_k(t)$ (INTEGER)
JCT(m) = j index of point m at which to output $C_k(t)$ (INTEGER)

- Each grid point (i,j) at which $C_k(t)$ is to be output is denoted by (ICT(m),JCT(m)). A total of MCT lines, each containing a single (ICT(m),JCT(m)) pair, must be included.

Line 106: *sedhist* FILE NAME (include only if INPOPT(22)=1 and INPOPT(23)=1)

Output file for time-history of sediment bed thickness at specific grid points. This option must be used in conjunction with *chist* output, INPOPT(22)=1. Thicknesses are output at the same grid points and times specified for $C_k(t)$. See Section 2.4 for a description of this file. (CHARACTER*64)

Line 107: *tauhist* FILE NAME (include only if INPOPT(22)=1 and INPOPT(24)=1)

Output file for time-history of total bottom shear stress at specific grid points. This option must be used in conjunction with *chist* output, INPOPT(22)=1. Bottom shear stresses are output at the same grid points and times specified for $C_k(t)$. See Section 2.4 for a description of this file. (CHARACTER*64)

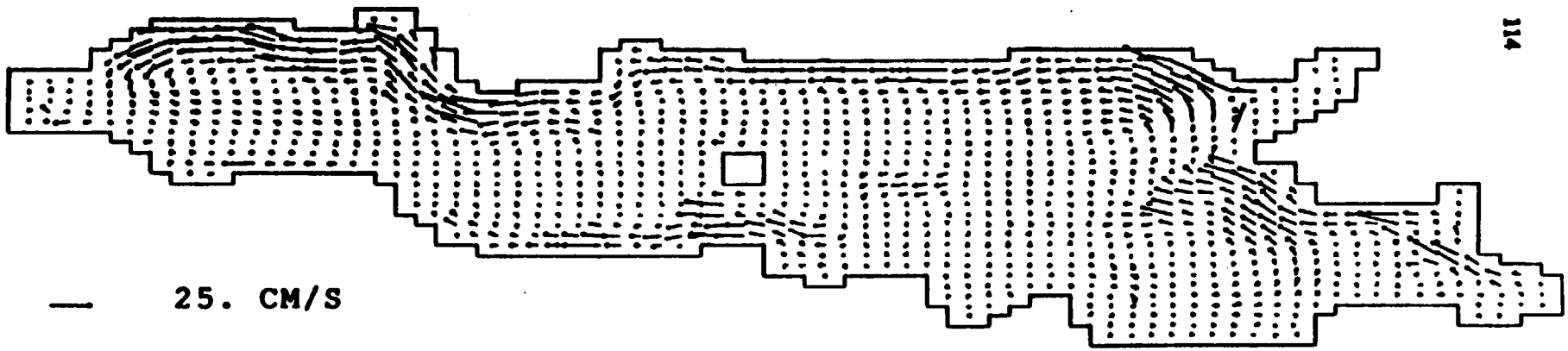

Figure 5.8 Velocity field at 48 hours for run *gbay.1*.

Program options are controlled by use of the INPOPT(n) variable as shown above. A total of 24 options are available to control various program functions. See Table 4.2 for a summary of INPOPT(n) values and effects. The general purpose, as well as any particular details, of each option will now be discussed.

INPOPT(1)

Controls whether a hydrodynamic only or a hydrodynamic and sediment transport calculation is to be performed.

INPOPT(2)

Performs a sediment transport calculation which assumes that the hydrodynamics are at a steady-state. This calculation will require much less computer time than a calculation using time-dependent hydrodynamics. In order to use this option, a previous hydrodynamic run must have been performed in order to generate an *icin* file to use as input for the current run. Care must be taken that the previous calculation was run long enough for the hydrodynamics to approach a steady-state. Using an *icin* file for which the hydrodynamics are not close to steady-state will result in a sediment transport calculation which does not conserve mass.

INPOPT(3)

Input *icin* generated from a previous run.

INPOPT(4)

Controls whether constant or variable bathymetry is to be specified. Generally, a constant bottom depth is only used for program debugging or numerical experiments. If a constant bottom depth is desired (INPOPT(4)=0), then the *depth* file will contain only one line which will be the constant bottom depth, in centimeters.

INPOPT(5)

Adds DELTAH to all bottom depths, see Line 22 of the input file.

INPOPT(6)

Determines if wind stresses, for both hydrodynamics and sediment transport, are to be included in this calculation.

INPOPT(7)

Specifies whether the wind stresses are to be constant or time-variable. For constant wind stresses (INPOPT(7)=0), the *wind* file consists of only one line. In the *wind* file, The values of τ_x^w and τ_y^w are entered on the first line and free format is used. The units of the wind stresses are *dynes/cm*^{2} and their orientation with respect to the *x-y* coordinate system is shown in Figure 4.3.

INPOPT(8)

Determines whether or not to include effects of the Coriolis acceleration. Generally, the Coriolis acceleration is negligible in rivers and bodies of water with dimensions less than 50 kilometers. Accounting for the Coriolis acceleration will increase the execution time of the hydrodynamic portion of the model.

INPOPT(9)

Input specified time-varying tide/seiche from *tide* file, see Section 3.3.3.

INPOPT(10)

Apply sinusoidally time-varying tide/seiche at open boundary, see Line 49 of the input file.

INPOPT(11)

Controls units of constant specified inflows. If INPOPT(11) = 0, then units of U0,UL,V0,VL (Line 8) are in cm^2/s (integrated velocity). If INPOPT(11)=1, units of U0,UL,V0,VL are in cm/s. If INPOPT(11) = 2, then units of these inflows are in m^3/s. If INPOPT(11) = 3, then units are in cfs.

INPOPT(12)

Linearly ramp constant inflow, see Lines 26 to 29 of the input file.

INPOPT(13)

Input time-variable flow rates and sediment loads from *inflow* and *sedload* files, see Sections 3.3.2 and 3.4 for descriptions of these files.

INPOPT(14)

Input spatially-variable sediment bed properties from the *sedbed* file, see Section 3.5 for a description of this file.

INPOPT(15)

Controls output of velocity field. If INPOPT(15)=0, no output of velocity field. If INPOPT(15)=1, then graphics output is written to file *uvgraph*, see Section 2.3.6 for a description of this file. If INPOPT(15)=2, then only hard copy is written to standard output. If INPOPT(15)=3, then both graphics and hard copy output is generated. For most real problems, hard copy output is not advised since rather large output files can result.

INPOPT(16)

Controls output of surface displacement (η) distribution. If INPOPT(16)=0, no output of η distribution. If INPOPT(16)=1, then graphics output is written to file *etagraph*, see Section 2.3.6 for a description of this file. If INPOPT(16)=2, then only hard copy is written to standard output. If INPOPT(16)=3, then both graphics and hard copy output is generated. For most real problems, hard copy output is not advised since rather large output files can result.

INPOPT(17)

Controls output of sediment concentration distribution. If INPOPT(17)=0, no output of concentration distribution. If INPOPT(17)=1, then graphics output is written to the *cgraph* files, see Section 2.3.7 and Lines 54-57 of the input file for a description of these files. If INPOPT(17)=2, then only hard copy is written to standard output. If INPOPT(17)=3, then both graphics and hard copy output is generated. For most real problems, hard copy output is not advised since rather large output files can result.

INPOPT(18)

Controls output of sediment bed thickness and composition distributions. If INPOPT(18)=0, no output of sediment bed distributions. If INPOPT(18)=1, then graphics output is written

to the *bedgraph* and *fracgraph* files, see Section 2.3.7 and Lines 58-59 of the input file for a description of these files. If INPOPT(18)=2, then only hard copy is written to standard output. If INPOPT(18)=3, then both graphics and hard copy output is generated. For most real problems, hard copy output is not advised since rather large output files can result.

INPOPT(19)

Controls output of intermediate results, see Line 60 of the input file.

INPOPT(20)

Controls output of *icout* file for use in a future run.

INPOPT(21)

Controls output of time-history of η at specific grid points, see Lines 62-83 of the input file.

INPOPT(22)

Controls output of time-history of concentration at specific grid points, see Lines 84-105 of the input file.

INPOPT(23)

Controls output of time-history of sediment bed thickness at specific grid points, see Line 106 of the input file. This option must be used in conjunction with INPOPT(22).

INPOPT(24)

Controls output of time-history of bottom shear stress at specific grid points, see Line 107 of the input file. This option must be used in conjunction with INPOPT(22).

4.2 USE OF setup TO CREATE AND MODIFY INPUT FILES

Setup was created in order to give an easy, error-proof way of creating and modifying input files for SEDZL. A series of menus are used in *setup* to input the variables needed to create the input file. If the input file already exists, *setup* will read the existing file and use the values in it as defaults.

4.2.1 setup MENUS

Three types of menus are used in *setup*. The first type is a selection menu, such as the Main Menu (Figure 4.5). In this type of menu, sub-menus are selected by pressing the letter code that appears in front of the choice. Pressing *RETURN* or *SPACE BAR* will select the next letter in the menu.

The second type of menu is the option menu, e.g., Menu A in Figure 4.6. In this kind of menu, you press *SPACE BAR* to toggle between values and *RETURN* to proceed to the next input line. Most options are either a 0 or 1 value, but some options will go up to 3. *Setup* will only let you select valid option values.

The third type of menu is the input menu, e.g., Menu C in Figure 4.8. In this type of menu, you can press *RETURN* to accept the current variable value and proceed to the next input variable. If you wish to change the value of the variable, press *SPACE BAR* causing the current variable value to disappear. A new value can then be typed in after which *RETURN* is pressed in order to go on to the next input line. There is currently no provision for restoring the old value once you have pressed *SPACE BAR* to enter a new value. If an incorrect variable value or file name is input in any menu, simply repeat that menu and make the correction.

For input menus, the program will only let you enter values if the proper option has been set, as well as any other necessary variables having the appropriate values. If these conditions are not met, the program will write the label *(Option not used)* where the variable would be and go on to the next variable. With certain option settings, an entire menu will not be needed. In this case, on fast terminals, the screen will just blink quickly and return to the previous menu. Only on slower terminals will you see the *(Option not used)* labels for the entire menu.

When running *setup* to generate a new input file, alphanumeric strings, e.g., a *node* file name for use in Menu H (Figure 4.17), will be empty by default. Be sure to input a file name for each file that *setup* asks for. If you exit from *setup* before inputting each requested file name, *setup* will write a blank line to the input file. Because of this blank line, neither *setup* nor SEDZL will be able to use the input file without error. To recover from this condition, you can manually edit the input file and replace any blank lines with an alphanumeric string and then run either *setup* or SEDZL using the modified input file.

4.2.2 RUNNING setup

Before running *setup*, you need to make sure that the terminal type is set. If you use *csh*, you can set it by typing *set term=type*, where *type* is the termcap code for the terminal you are using, and press *RETURN*. Popular terminal types include sun, sun-cmd, xterm, vt100, etc. The terminal type must be known to your system. They are usually defined in the file */etc/termcap*. Read the *termcap* manual page for more information. *Setup* will not run properly if the incorrect terminal type is defined. Other tools, such as *vi* also rely on this setting. If *vi* works, then *setup* should also work.

To start *setup*, type *setup* and then press *RETURN*. You can specify the run number to be used by typing the run number on the command line. For example, if you wanted to use the run number *test.1*, you would type *setup test.1*. If run number is not specified on the command line, the program will first ask for this to be input. It will then open a file of the form *in.{run_number}*, e.g., *in.test.1*. This is a file formatted to the SEDZL input file specification.

You will then be presented with the Main Menu (Figure 4.5). From the Main Menu, you can select Menus A through I or quit by pressing J. Press *RETURN* or *SPACE BAR* to sequentially go through each menu, from A through I. Menus should be completed in their given order in most cases, as option settings in the first menus affect whether or not certain variables are input later in the program.

Menus A and B (Figures 4.6 and 4.7) are where the options are set. Menu A contains the options controlling input while Menu B is for setting the options controlling program output. Most of these

options determine whether or not other variables are to be input in Menus C through I. Hence, Menus A and B should be completed before using the other menus.

Menus C through F (Figures 4.8 through 4.11) are input menus for various input variables.

Menu G (Figure 4.12) contains four sub-menus. Like the Main Menu, you can access the sub-menus by pressing the keys A through D or E to quit. Menu G.a (Figure 4.13) asks for input of the LAYMAX variable. The value entered here determines the number of values input in Menus G.b through G.d (Figures 4.14 through 4.16). Pressing E from Menu G will return you to the Main Menu.

Names of input and output files are specified in Menu H (Figure 4.17).

Menu I (Figure 4.18) contains nine sub-menus. You can access further menus by pressing the keys A through I, or J to quit. Menus I.a through I.i (Figures 4.19 through 4.27) are all input menus. When you press J, you will be returned to the Main Menu.

Pressing J from the Main Menu will store all of the input variables in file *in.{run_number}* and then exit the *setup* program. This process will overwrite the old input file, if it exists. Prior to modifying an existing input file, you should make a backup copy of the original input file in case you make a mistake.

This input file can then be fed directly into the SEDZL program.

4.3 MANUAL CREATION/MODIFICATION OF INPUT FILES

The user can use a text editor to manually create or modify a SEDZL input file. The order and content of each line of the input file must be as described in Section 4.1 with optional lines only included if the proper value of INPOPT(n) is set on Line 2. SEDZL reads input using free format so only a single space needs to separate each variable on a particular line. An input file which is incorrectly constructed will cause SEDZL to abort during execution. The user must be sure that all necessary lines have been included in the input file for the specified option/variable settings of a particular run.

5. EXAMPLE CALCULATIONS

Two example problems will be presented in this section in order to demonstrate how to use SEDZL. The Lower Fox River and Green Bay are the areas for which example calculations have been made. The primary purpose of these examples is to provide the user with samples of input and output files to complement the instructions provided in Sections 2, 3, and 4. While typical hydrodynamic forcing and sediment loading values, as well as recommended sediment property parameters, have been used, it must be emphasized that these calculations are only examples and their results are not necessarily realistic or accurate.

The input files for the example calculaions, *in.fox.1*, *in.fox.2*, and *in.gbay.1* were all generated using *setup*. The user is encouraged to practice using *setup* by utilizing these input files as models for comparison. Be sure to use dummy file names when testing *setup* so that the example input files are not accidentally modified. The user should also repeat the example runs using the provided input files, but generate output files with different names than the output files provided in the SEDZL software package, e.g., *out.fox.1.new* instead of *out.fox.1* and *out.srv.fox.1.new* in place of *out.srv.fox.1*. These newly created output files should be compared with the provided output files to make sure that there is no difference between them. If there are significant differences between the two sets of output files, then SEDZL probably has not been installed correctly on the user's computer system. Different computer systems may produce slightly different numerical results for identical input files due to round-off error.

If SEDZL has been installed as discussed in Section 2.6 and the executable file *run.sedzl.x* has been put in the directory */usr/local/bin*, then the user can execute *run.sedzl.x* from any directory desired. To perform the *fox.1* calculation discussed below, simply enter

run.sedzl.x < in.fox.1 > out.fox.1 &

and the hard copy output file *out.fox.1* will be created. Use of the & puts the job in background which should always be done when running SEDZL due to the length of the runs.

5.1 LOWER FOX RIVER EXAMPLE

The Lower Fox River comprises the 11 km reach from Depere Dam to the river mouth at Green Bay in Wisconsin. Two example calculations were performed for the Lower Fox River; a 6 hour long hydrodynamic run and a hydrodynamic/sediment transport calculation with a duration of 12 hours. For a detailed discussion of the sediment transport processes in the Lower Fox River see Gailani et al., 1990.

The bathymetry and computational grid boundaries for the Lower Fox River are shown in Figure 5.1. The *node* file used for the Lower Fox River is called *node.fox* and the file is illustrated in Figure 5.2a and 5.2b in matrix form. Note that the structure of *node.fox* is different from what is shown in Figure 5.2, which is for illustrative purposes only. For this problem, MX=125 and MY=50 with the i subscript increasing as one travels from the dam to the mouth. The x-y coordinate system is oriented such that North is 30° from the positive x- axis. Specified inflows have been placed at Depere Dam (*node-number* = 43) and at the East River inlet (*node-number* = 41). An open boundary (*node-number* = 52) exists at the river mouth. The file *depth.fox* contains bathymetry data for the Lower Fox River, and the water depths in this file are referenced to the Great Lakes Datum of 576.8 feet. A grid element size of Δx = 9000 cm and Δy = 3000 cm was used here, which allowed a timestep size of 3 seconds when a DELTAH of 50 cm was added to the bottom depths.

The first calculation performed was a 6 hour hydrodynamic run. Performing a short hydrodynamic run prior to doing sediment transport calculations is highly recommended. By doing this, most unwanted hydrodynamic transients due to model startup will be eliminated. The input file for this run is *in.fox.1* and it was generated using *setup*. The auxiliary input files *seiche.fox.1* and *inflow.fox.1* were used in this calculation. The file *seiche.fox.1* is a *tide* file (see Section 3.3.3) and it is applied at the open boundary at the river mouth. The time period between seiche values in this file is 1 hour, and there are a total of 7 values. Note that the initial water elevation is zero. This initial value for starting a tide/seiche driven problem is recommended so that large waves are not generated due to a sharp discontinuity at the first timestep of the run. The file *inflow.fox.1* is an *inflow* file (see Section 3.3.2) and it contains the time-varying inflow data at Depere Dam in m^3/s. The QOLD value was set at zero

in order to smoothly start the flow in the river. The time interval between the inflow values is 3 hours with a ramping time of 1 hour. The East River inlet is assumed to have a constant specified inflow of 15 *cm/s* and this flow rate was initially ramped over 600 timesteps (0.5 hour). Wind stresses and Coriolis effects were assumed to be negligible in this problem.

The file *out.fox.1* contains the hard copy output of this run and this file is shown in Figure 5.3. A listing of all input parameters is included in this file. Note that minimum and maximum values of velocity and surface displacement at each intermediate output are included in this file. The graphics output files *out.uv.fox.1 (uvgraph)* and *out.eta.fox.1 (etagraph)* were generated along with file *out.etahist.fox.1 (etahist)* which contains $\eta(t)$ at three grid points. A plot of the velocity field at the end of run *fox.1* in shown in Figure 5.4. Note that only every other velocity vector in this illustration has been plotted for clarity. In addition, the file *out.ic.fox.1* was created, and this is an *icout* file containing the values of the hydrodynamic variables at the end of the run. This file will be used as the initial conditions for the next calculation.

A second calculation modelled the sediment transport processes in the Lower Fox River. The input file for this run is *in.fox.2*. The file *out.ic.fox.1* will be the *icin* file for this calculation. A sediment transport timestep of 15 seconds was used so IRTSTP=5. The *tide* file for this run is *seiche.fox.2*, and its values have a time period between them of 1 hour. Note that the first value in *seiche.fox.2* is the same as the last value used in *seiche.fox.1*. These two values must be the same when doing this type of serial calculation so that no discontinuity in surface elevation occurs at the start of the second calculation. The *inflow* file for the sediment transport run is *inflow.fox.2*. This file contains 5 flow values with a time interval of 3 hours and a 1 hour ramping time. Note that the first value in *inflow.fox.2* is the same as the last value in *inflow.fox.1* and that the value of QOLD in *inflow.fox.2* was set at 46 m^3/s which is the average of the last two inflow values in *inflow.fox.1*. Use of these values ensures that a continuous flow rate at the start of the *fox.2* run will result. For this run, KCT=3, so time-varying sediment loads for the fine, medium, and course size-classes are specified at the dam. The file *sedload.fox.2* contains sediment load input for this calculation. There are four values for each sediment size-class in this file, and the time interval between each concentration value is 4 hours with a

ramping time of 1 hour. The initial concentration values at the dam (COLD(k)) for the fine, medium, and coarse size-classes are 5, 10, and 5 mg/liter, respectively. Constant specified concentrations of 5, 15, and 7 mg/liter were applied at the East River inlet.

All hydrodynamic and sediment transport parameters for this run are included in the hardcopy output file, *out.fox.2*, shown in Figure 5.5. Note that output for this run was generated at times of 12 and 18 hours. The time at the end of run *fox.1* was 6 hours which was the value output to file *out.ic.fox.1;* this value was then the time at the start of run *fox.2*.

Velocity and surface displacement graphics files, *out.uv.fox.2* and *out.eta.fox.2*, were generated by this run. The graphics output files for the sediment concentrations are *out.ctot.fox.2 (cgraph1), out.ct.cfin.fox.2 (cgraph2), out.ct.cmed.fox.2 (cgraph3),* and *out.ccor.fox.2 (cgraph4).* The file *out.bed.fox.2 (bedgraph)* contains graphics output for the change in sediment bed 'thickness', Φ, while *out.frac.fox.2 (fracgraph)* has graphics data for the bed composition. Time-histories at three grid points for concentrations, sediment bed 'thickness', and bottom shear stress were also generated.

The above two calculations were done on a Sun Sparcstation 330 computer which is a 16 MIPS, 2.5 Mflops machine. The *fox.1* run took 25 cpu minutes while *fox.2* took 71 cpu minutes.

5.2 GREEN BAY EXAMPLE

The second example calculation concerns wind-driven circulation in Green Bay and sediment resuspension due to wind waves. The assumptions have been made that sediment loading from the lower Fox River and interchange between Green Bay and Lake Michigan are negligible. The resulting enclosed computational grid for Green Bay is shown in Figure 5.6. This illustration of the *node* file for Green Bay, *node.gbay*, has the mouth of the Lower Fox River located at approximately i=1 and j=26. Inclusion of Chambers Island in central Green Bay is also shown. For this problem, MX=77 and MY=34 with North being inclined by 30° with respect to the x- axis. A grid element size of Δx = 243,840 cm (8000 feet) and Δy = 121,920 cm (4000 feet) was used. Green Bay bathymetry, with respect to Great Lakes Datum of 576.8 feet, is provided in *depth.gbay*, and a DELTAH of 100 cm has been added to these bottom depths. The hydrodynamic timestep size is 45 seconds, and the sediment

transport timestep is 225 seconds. A total of 96 hours (4 days) was modelled.

The wind forcing for this run is given in file *wind.gbay*. Note that the time interval between each wind data point is 1 hour and that there is one additional value, hour 97, in the file, as discussed in Section 3.3.1.

The purpose of this run was to examine sediment resuspension due to wind waves and subsequent transport and deposition. Therefore, a two component sediment size-class system was employed in run *gbay.1*, i.e., KCT=2. The sediment bed was assumed to be initially composed of a 50-50 mixture of medium and coarse size-class particles (P0 = 0.5). The initial sediment thicknesses, TSED0(L), are given in Figure 5.7, file *out.gbay.1*, along with all of the other parameters used in this calculation. Initial suspended sediment concentrations were set to zero so that the sediment bed was the source of all sediment in this run. The various output files generated by the run are also listed in this file. A plot of the velocity field midway through the calculation, at 48 hours, is shown in Figure 5.8.

One interesting result of this calculation was the generation of negative sediment concentrations, which are physically impossible. Negative concentrations indicate Re_{cell} problems, see Section 2.2, and this means that the value of D_H needs to be increased. The negative concentrations calculated in this run are very small, however, and cause negligible errors.

This calculation was carried out on a Sun Sparcstation 330 and it took 28 cpu minutes.

6. REFERENCES

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TABLES

TABLE 2.1: RECOMMENDED SEDIMENT PROPERTIES

PARAMETER	EQUATION	VARIABLE	RECOMMENDED VALUE
a_o	2.8	A0IN	0.008 $gram-day^2/cm^2$
α_o	2.6	ALPHA	10^{-6} $gram^2/cm^3-s^2$ (Fresh water)
Maximum floc diameter		DMAX	200 μm (0.0200 cm)
n	2.8	RESEXP	3
Coarse size-class settling speed		BETA	1000 $\mu m/s$
Boundary element shear stress factor		CB	0.5
Critical τ_o for no deposition of medium size-class		TCRDEP	0.1 $dyne/cm^2$
Maximum number of sediment bed layers		LAYMAX	10
Initial fraction of medium size-class in sediment bed		P0	0.5

TABLE 2.2: RECOMMENDED SEDIMENT BED STRUCTURE

LAYER L	TIME AFTER DEPOSITION FTIME(L) (days)	CRITICAL SHEAR STRESS TAUCR(L) (*dynes/cm*^{2})
1	0.005	0.1
2	0.125	0.2
3	0.25	0.4
4	0.5	0.8
5	1	1.0
6	2	1.0
7	3	1.0
8	4	1.0
9	5	1.0
10	6	1.0

TABLE 3.1 : NODE-NUMBER DEFINITIONS

ELEMENT TYPE/ORIENTATION	NODE-NUMBER
Outside solution domain	0
Full element	1
1/2 element: solid boundary	
Plus x	11
Minus x	12
Plus y	13
Minus y	14
1/4 element: solid boundary	
Quadrant 1	21
Quadrant 2	22
Quadrant 3	23
Quadrant 4	24
3/4 element: solid boundary	
Quadrant 1	31
Quadrant 2	32
Quadrant 3	33
Quadrant 4	34
1/2 element: specified-inflow	
Plus x	41
Minus x	42
Plus y	43
Minus y	44
1/2 element: open-boundary	
Plus x	51
Minus x	52
Plus y	53
Minus y	54
1/4 element: partially-open	
Quadrant 1	61
Quadrant 2	62
Quadrant 3	63
Quadrant 4	64

TABLE 4.1 : INPUT FILE STRUCTURE

NOTE: INPOPT = *** indicates that this line is NOT OPTIONAL

LINE	INPOPT	VALUE	VARIABLE(S)
1	***		RUNNO
2	***		INPOPT(n), n=1,...,24
3	***		MX MY XSTP YSTP
4	***		node FILE NAME
5	***		depth FILE NAME
6	***		TSTP IRTSTP TMAX
7	***		AH CF
8	***		U0 UL V0 VL
9	1	1	DH KCT
10	1	1	CX0(k), k=1,...,KCT
11	1	1	CXL(k), k=1,...,KCT
12	1	1	CY0(k), k=1,...,KCT
13	1	1	CYL(k), k=1,...,KCT
14	1	1	A0IN ALPHA DMAX RESEXP BETA CB TCRDEP
15	1	1	LAYMAX
16	1	1	FTIME(l), l=1,...,LAYMAX
17	1	1	TSED0(l), l=1,...,LAYMAX
18	1	1	TAUCR(l), l=1,...,LAYMAX
19	1	1	P0 (include only if KCT > 1)
20	1	1	CTIC(k), k=1,...,KCT

TABLE 4.1 (cont.)

LINE	INPOPT	VALUE	VARIABLE(S)
21	3	1	icin FILE NAME
22	5	1	DELTAH
23	9	1	tide FILE NAME
24	6	1	wind FILE NAME
25	6 and 7	1	TWIND
26	12	1	NRAMP(1)
27	12	1	NRAMP(2)
28	12	1	NRAMP(3)
29	12	1	NRAMP(4)
30	13	1	NQIN
31	13	1	inflow1 FILE NAME (NQIN files)
32	13	1	inflow2 FILE NAME
33	13	1	inflow3 FILE NAME
34	13	1	inflow4 FILE NAME
35	13	1	NODEQ(1) NTS(1) TS(1) TR (1) (NQIN lines)
36	13	1	NODEQ(2) NTS(2) TS(2) TR (2)
37	13	1	NODEQ(3) NTS(3) TS(3) TR (3)
38	13	1	NODEQ(4) NTS(4) TS(4) TR (4)
39	1 and 13	1	NCIN
40	1 and 13	1	sedload1 FILE NAME (NCIN files)
41	1 and 13	1	sedload2 FILE NAME
42	1 and 13	1	sedload3 FILE NAME
43	1 and 13	1	sedload4 FILE NAME
44	1 and 13	1	NODEC(1) NTC(1) TSC(1) TRC(1) (NCIN lines)
45	1 and 13	1	NODEC(2) NTC(2) TSC(2) TRC(2)
46	1 and 13	1	NODEC(3) NTC(3) TSC(3) TRC(3)
47	1 and 13	1	NODEC(4) NTC(4) TSC(4) TRC(4)
48	8	1	FCOR
49	10	1	ASEI OMSEI
50	9 or 10	1	NODTID

TABLE 4.1 (cont.)

LINE	INPOPT	VALUE	VARIABLE(S)
51	14	1	sedbed FILE NAME
52	15	1 or 3	uvgraph FILE NAME
53	16	1 or 3	etagraph FILE NAME
54	17	1 or 3	cgraph1 FILE NAME (total concentration)
55	17	1 or 3	cgraph2 FILE NAME (fine conc., only if KCT = 3)
56	17	1 or 3	cgraph3 FILE NAME (medium conc., only if KCT > 1)
57	17	1 or 3	cgraph4 FILE NAME (coarse conc., only if KCT > 1)
58	18	1 or 3	bedgraph FILE NAME
59	18	1 or 3	fracgraph FILE NAME (include only if KCT > 1)
60	19	1	OUTINT
61	20	1	icout FILE NAME
62	21	1	etahist FILE NAME
63	21	1	METAS
64 to 83	21	1	IETA(m) JETA (m) (m=1,...,METAS lines)
84	22	1	chist FILE NAME
85	22	1	MCT CTOUT
86 to 105	22	1	ICT(m) JCT(m) (m=1,...,MCT lines)
106	22 and 23	1	sedhist FILE NAME
107	22 and 24	1	tauhist FILE NAME

TABLE 4.2 : PROGRAM OPTIONS

INPOPT	VALUE	DESCRIPTION
1	0	Hydrodynamic calculation only
	1	Hydrodynamic and sediment transport calculation
2	1	Sediment transport calculation with steady-state hydrodynamics
3	1	Input 'icin' file from previous run
4	0	Constant bottom depth
	1	Variable bathymetry
5	1	Add DELTAH to all bottom depths
6	0	No wind stresses used in calculation
	1	Wind stresses used in calculation
7	0	Constant wind stress
	1	Time-variable wind stress
8	1	Include Coriolis acceleration
9	1	Input specified time-varying tide/seiche from 'tide' file
10	1	Use sinusoidally time-varying tide/seiche
11	1	Constant specified inflows are velocities (cm/s)
	2	Constant specified inflows are in cubic meters/second
	3	Constant specified inflows are in cfs

TABLE 4.2 (cont.)

INPOPT	VALUE	DESCRIPTION
12	1	Linearly ramp constant specified inflows
13	1	Input time-variable flow rates and sediment loads from 'inflow' and 'sedload'
14	1	Input spatially-variable sediment bed from 'sedbed' file
15	 0 1 2 3	Velocity output control No output Graphics only ('uvgraph') Hard copy only (standard output) Graphics and hardcopy
16	 0 1 2 3	Surface displacement output control No output Graphics only ('etagraph') Hard copy only (standard output) Graphics and hard copy
17	 0 1 2 3	Concentration output control No output Graphics only ('cgraph') Hard copy only (standard output) Graphics and hard copy
18	 0 1 2 3	Sediment bed output control No output Graphics only ('bedgraph' and 'fracgraph') Hard copy only (standard output) Graphics and hard copy
19	1	Output intermediate results
20	1	Ouput 'icout' file to use for future run

TABLE 4.2 (cont.)

INPOPT	VALUE	DESCRIPTION
21	1	Output time-history of surface displacement at specific grid points
22	1	Output time-history of concentration at specific grid points
23	1	Output time-history of sediment bed thickness at specific grid points
24	1	Output time-history of bottom shear stress at specific grid points

FIGURES

FIGURE 2.1: SEDIMENT BED MODEL

WATER

---

t_d = 0.005 *day*, τ_o = 0.1 *dyne/cm*^{2}

---

t_d = 0.125 *day*, τ_o = 0.2 *dyne/cm*^{2}

---

t_d = 0.25 *day*, τ_o = 0.4 *dyne/cm*^{2}

---

t_d = 0.5 *day*, τ_o = 0.8 *dyne/cm*^{2}

---

t_d = 1 *day*, τ_o = 1 *dyne/cm*^{2}

---

t_d = 2 *days*, τ_o = 1 *dyne/cm*^{2}

LAYERS 3, 4, 5, 6

---

t_d = 7 *days*, τ_o = 1 *dyne/cm*^{2}

FIGURE 2.2: STRUCTURE OF THE GRAPHICS OUTPUT FILES

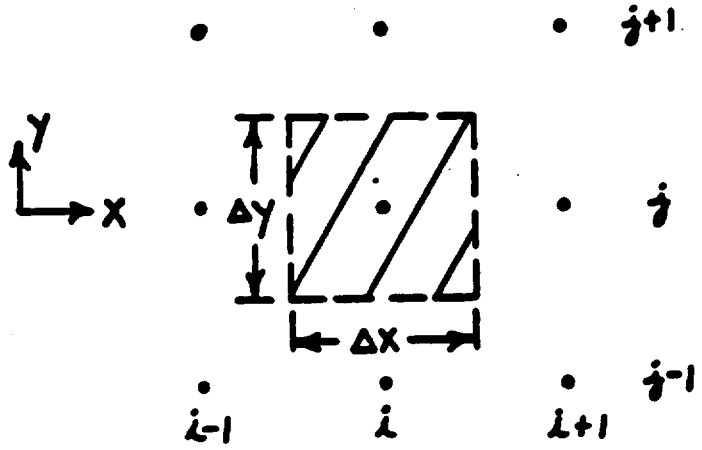
a. VELOCITY FIELD FILE

Line	j	i= 1	i= 2	...	i= MX
1		TIME1			
2	MY	uu.u	uu.u	...	uu.u
3	MY-1	uu.u	uu.u	...	uu.u
		.	.	...	.
		.	.	...	.
		.	.	...	.
MY+1	1	uu.u	uu.u	...	uu.u
MY+2	MY	vv.v	vv.v	...	vv.v
MY+3	MY-1	vv.v	vv.v	...	vv.v
		.	.	...	.
		.	.	...	.
		.	.	...	.
2*MY+1	1	vv.v	vv.v	...	vv.v
2*MY+2		TIME2			
2*MY+3	MY	uu.u	uu.u	...	uu.u
2*MY+4	MY-1	uu.u	uu.u	...	uu.u
		.	.	...	.
		.	.	...	.
		.	.	...	.
3*MY+2	1	uu.u	uu.u	...	uu.u
3*MY+3	MY	vv.v	vv.v	...	vv.v
3*MY+4	MY-1	vv.v	vv.v	...	vv.v
		.	.	...	.
		.	.	...	.
		.	.	...	.
4*MY+2	1	vv.v	vv.v	...	vv.v

b. CONTOUR PLOTTING FILE

Line	j	i= 1	i= 2	...	i= MX
1		TIME1			
2	MY	xxxxE-xx	xxxxE-xx	...	xxxxE-xx
3	MY-1	xxxxE-xx	xxxxE-xx	...	xxxxE-xx
		.	.	...	.
		.	.	...	.
		.	.	...	.
MY+1	1	xxxxE-xx	xxxxE-xx	...	xxxxE-xx
MY+2		TIME2			
MY+3	MY	xxxxE-xx	xxxxE-xx	...	xxxxE-xx
MY+4	MY-1	xxxxE-xx	xxxxE-xx	...	xxxxE-xx
		.	.	...	.
		.	.	...	.
		.	.	...	.
2*MY+2	1	xxxxE-xx	xxxxE-xx	...	xxxxE-xx

a. Full Element



b. ½ Element

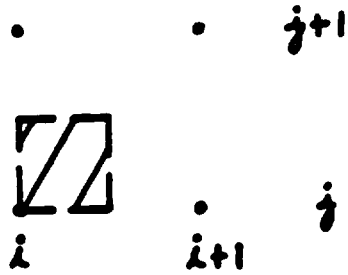
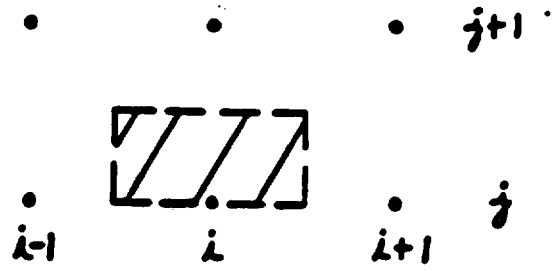


Figure 3.1 Computational grid elements.

c. ½ Element



d. 3/4 Element

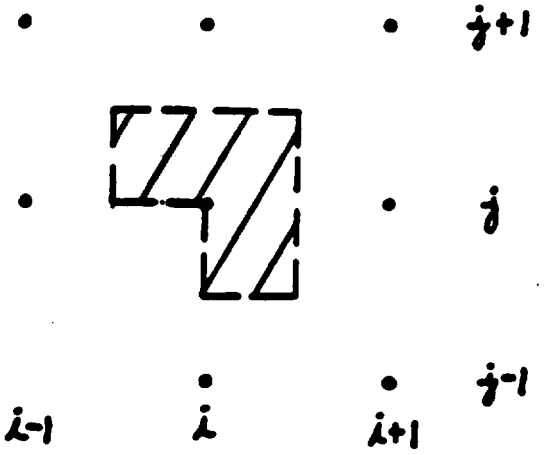
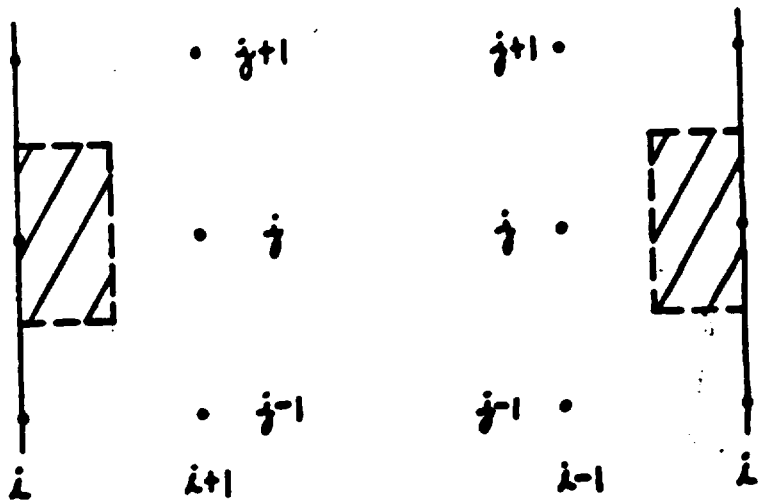


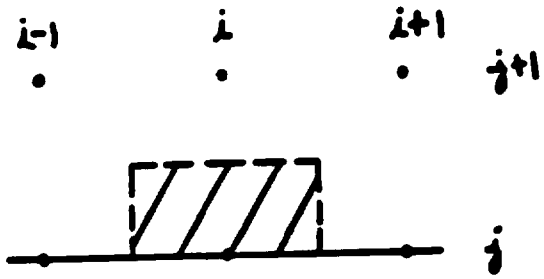
Figure 3.1 (cont.) Computational grid elements.

a. Plus x orientation.

b. Minus x orientation.



c. Plus y orientation.



d. Minus y orientation.

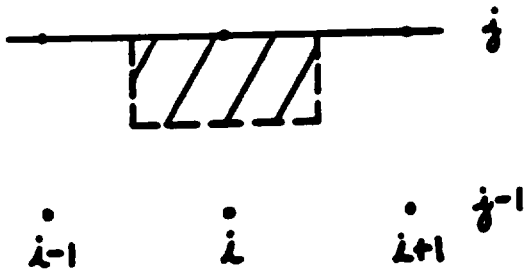
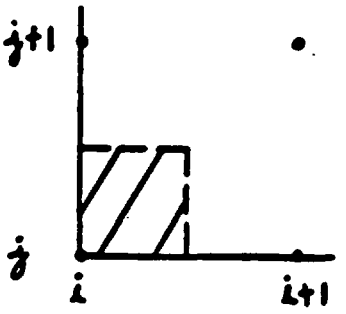
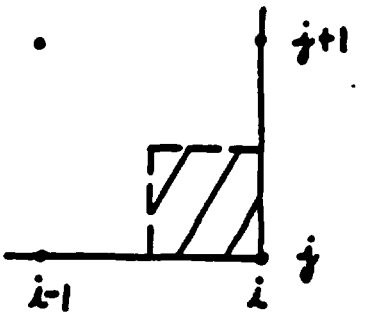


Figure 3.2 1/2 element orientations.

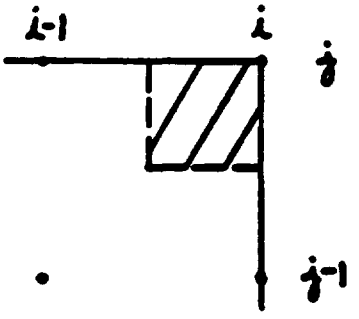
a. Quadrant 1



b. Quadrant 2



c. Quadrant 3



d. Quadrant 4

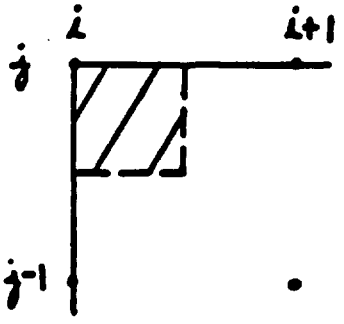
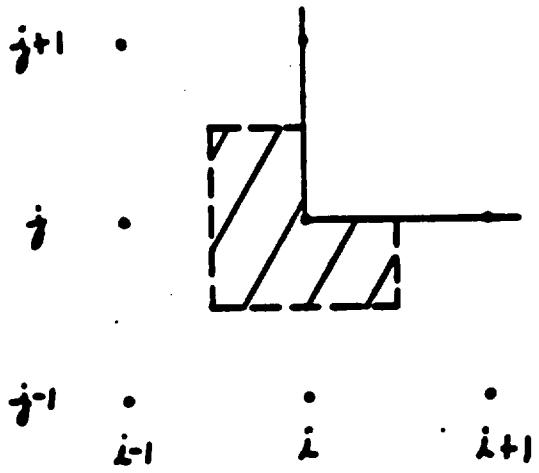


Figure 3.3 1/4 element orientations.

a. Quadrant 1



b. Quadrant 2

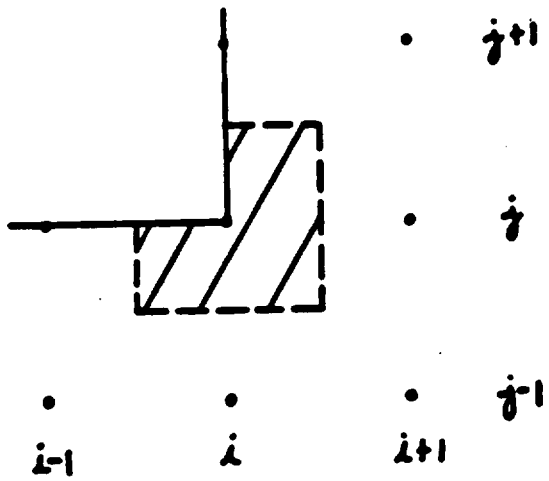
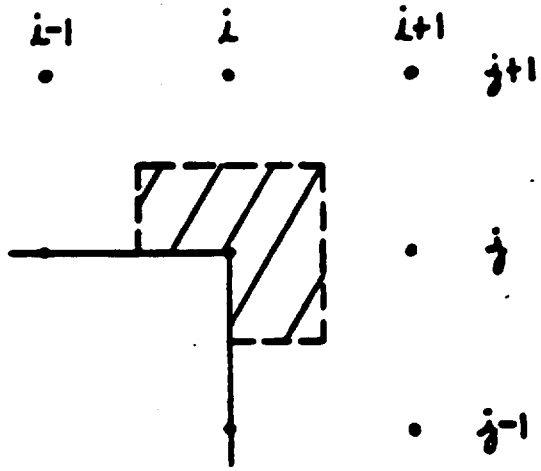


Figure 3.4 3/4 element orientations.

c. Quadrant 3



d. Quadrant 4

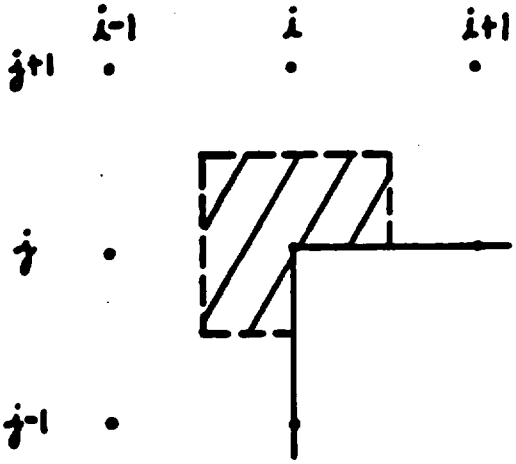


Figure 3.4 (cont.) 3/4 element orientations.

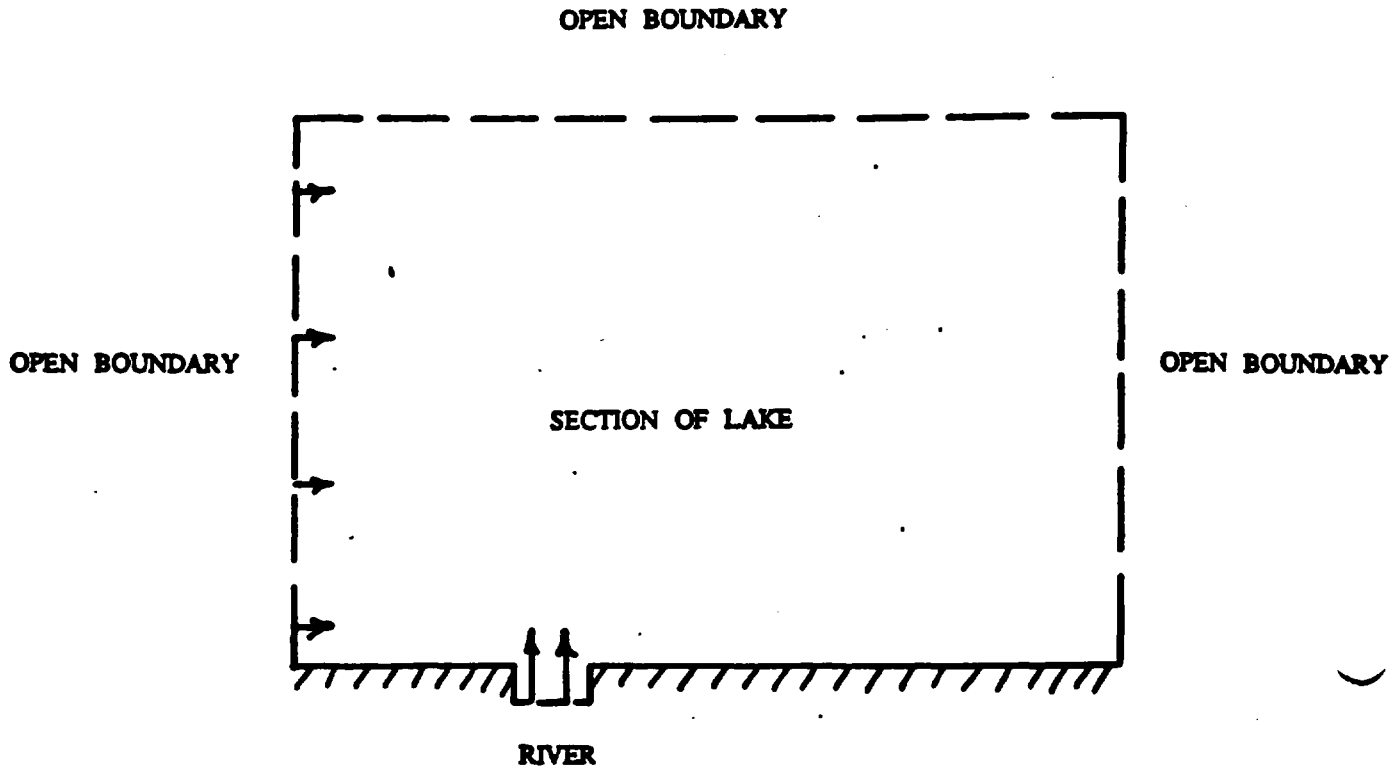


Figure 3.5 Example of a disallowed open boundary element configuration.

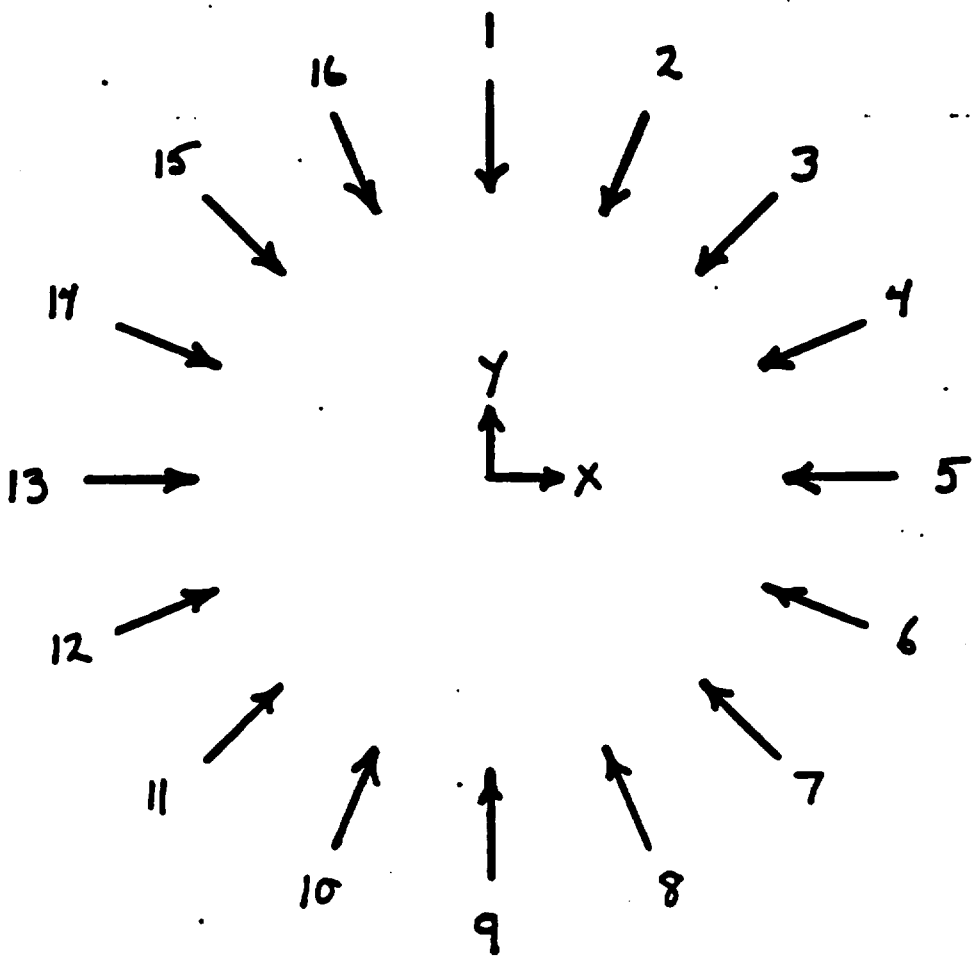


Figure 3.6 Wind direction numbers with respect to *x-y* coordinate system.

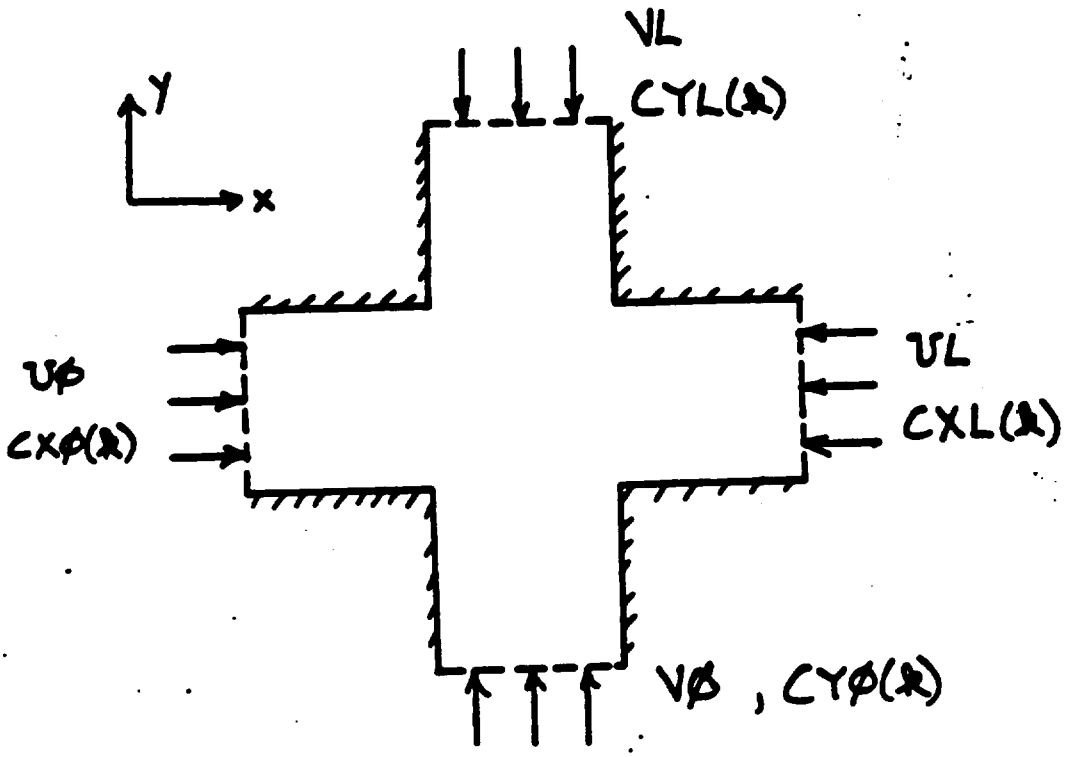


Figure 4.1 Orientation of specified constant inflows and inlet concentrations.

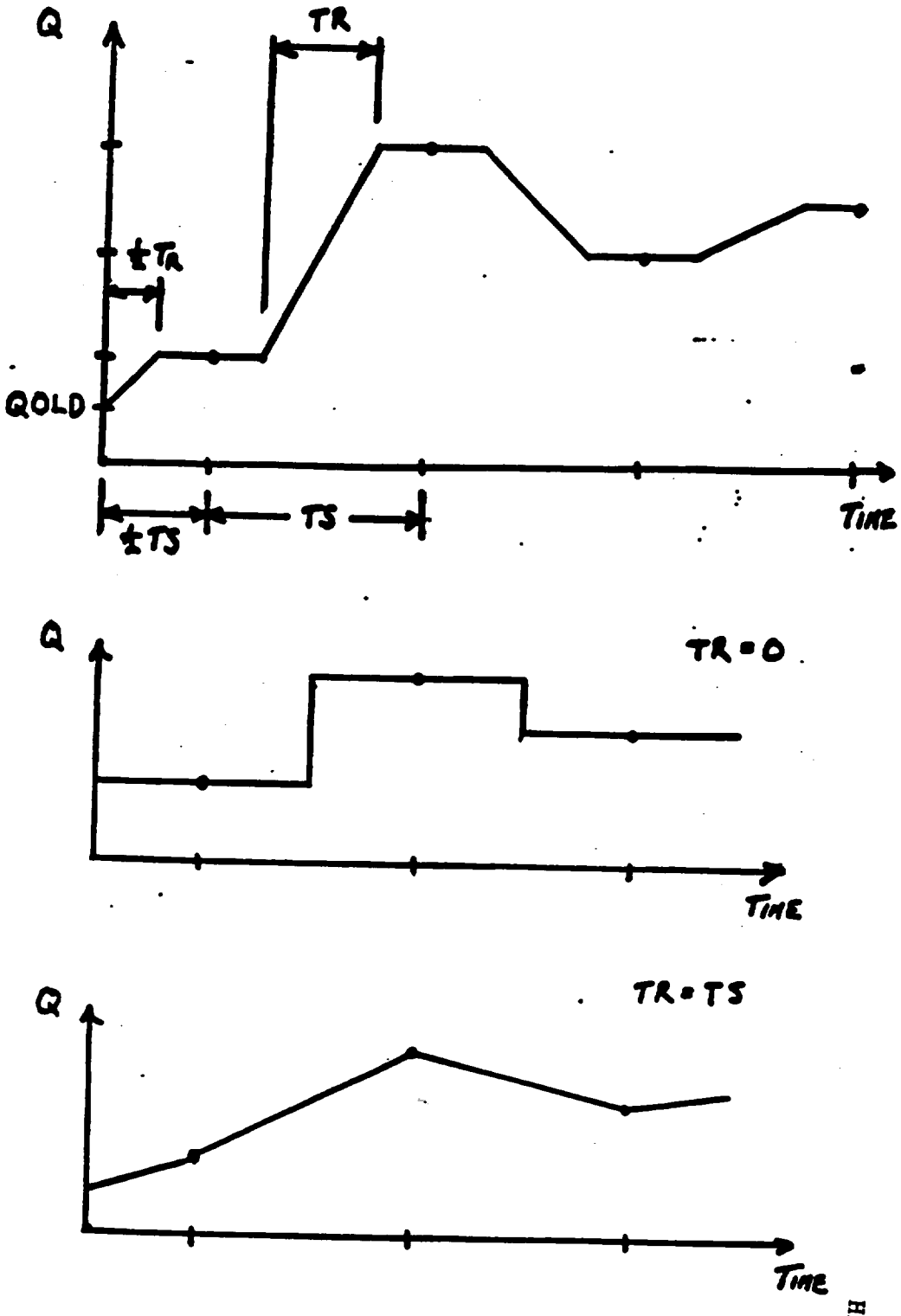
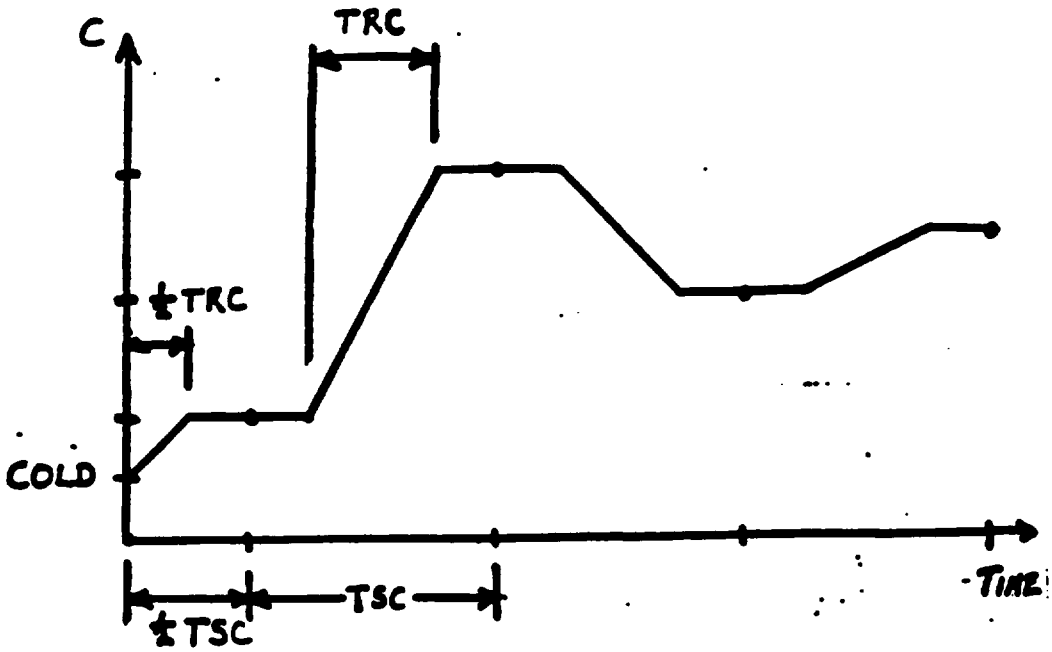
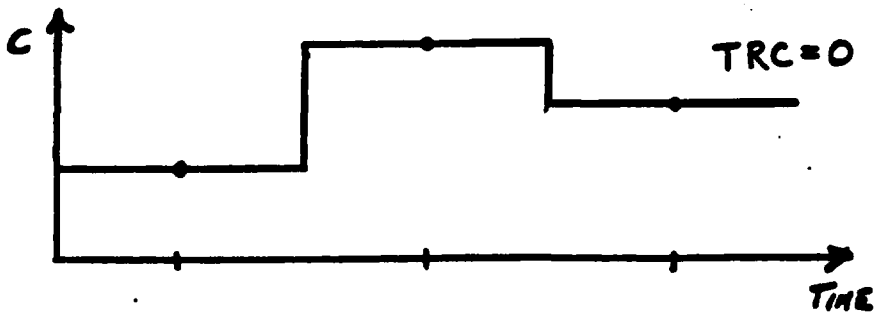


Figure 4.2 Time-varying inflow specification.





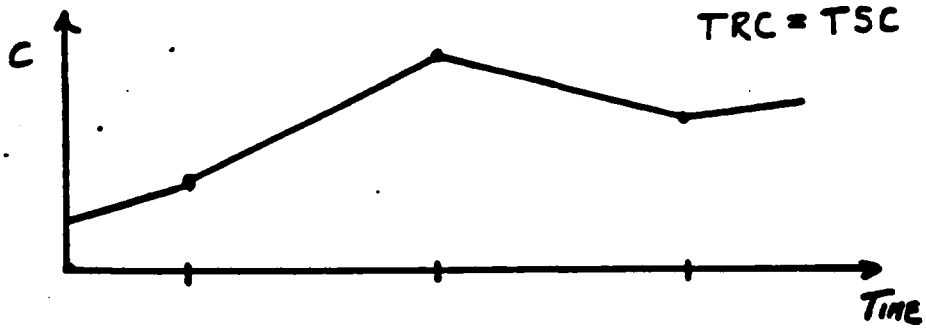


Figure 4.3 Time-varying inlet concentration specification.

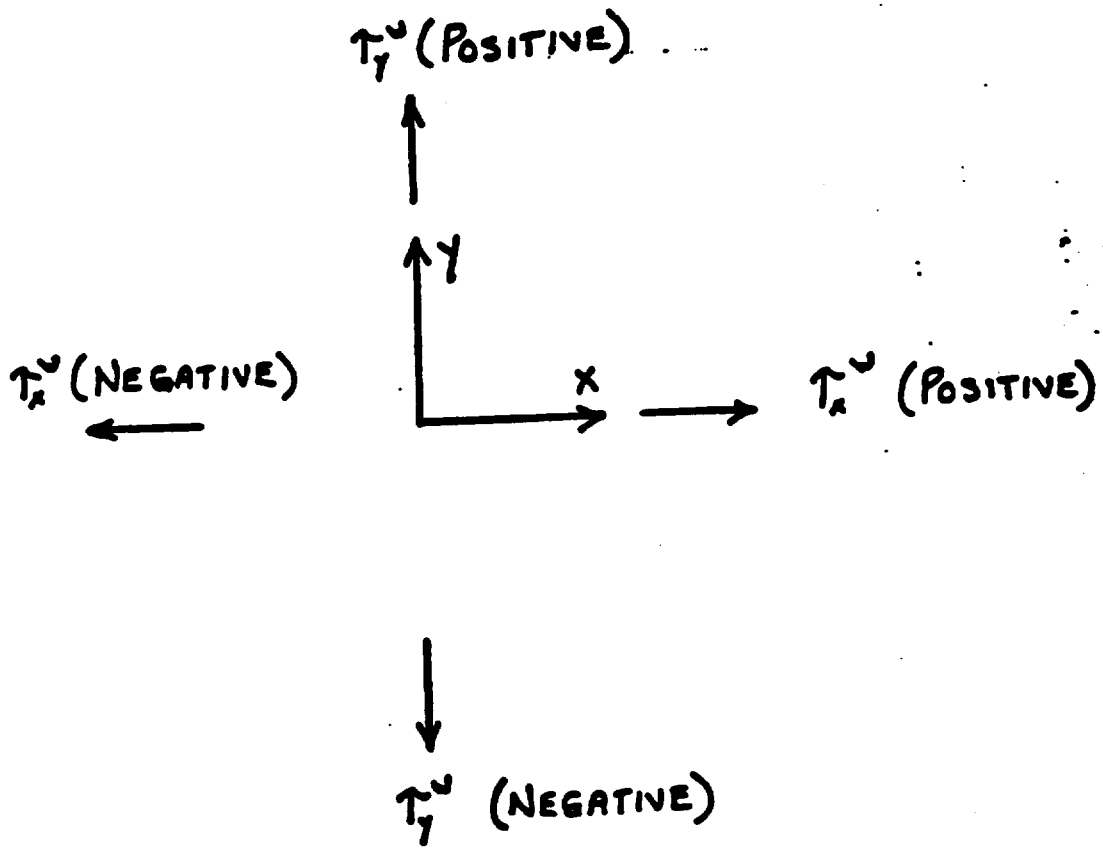


Figure 4.4 Constant wind stress orientation.

```
MAIN MENU: SEDZL INPUT FILE SETUP

        A. INPUT OPTIONS
        B. OUTPUT OPTIONS
        C. GEOMETRIC PARAMETERS
        D. TIME PARAMETERS
        E. HYDRODYNAMICS
        F. SEDIMENT TRANSPORT
        G. SEDIMENT BED STRUCTURE
        H. INPUT/OUTPUT FILE NAMES
        I. ADDITIONAL INPUT/OUTPUT

        J. QUIT

      MAKE YOUR SELECTION:
```

Figure 4.5 Main menu for *setup*.

```
   MENU A: OPTIONS CONTROLLING PROGRAM INPUT (INPOPT(n))

SEDIMENT TRANSPORT IN ADDITION TO HYDRO (1):
      SEDIMENT TRANSPORT WITH S-S HYDRO (2):
            READ icin FROM PREVIOUS RUN (3):
   READ VARIABLE BATHYMETRY FROM depth (4):
         ADD deltah TO BOTTOM DEPTHS (5):
                  USE WIND STRESSES (6):
    READ TIME VARIABLE WINDS FROM wind (7):
  INCLUDE CORIOLIS ACCELERATION EFFECTS (8):
   READ VARIABLE TIDE/SEICHE FROM tide (9):
          USE SINUSOIDAL TIDE/SEICHE (10):
  UNITS OF CONSTANT SPECIFIED INFLOWS (11):
LINEARLY RAMP CONSTANT SPECIFIED INFLOW (12):
 TIME VARIABLE INFLOW AND CONCENTRATION (13):
 READ VARIABLE SEDIMENT BED FROM sedbed (14):
```

Figure 4.6 Menu A for *setup*.

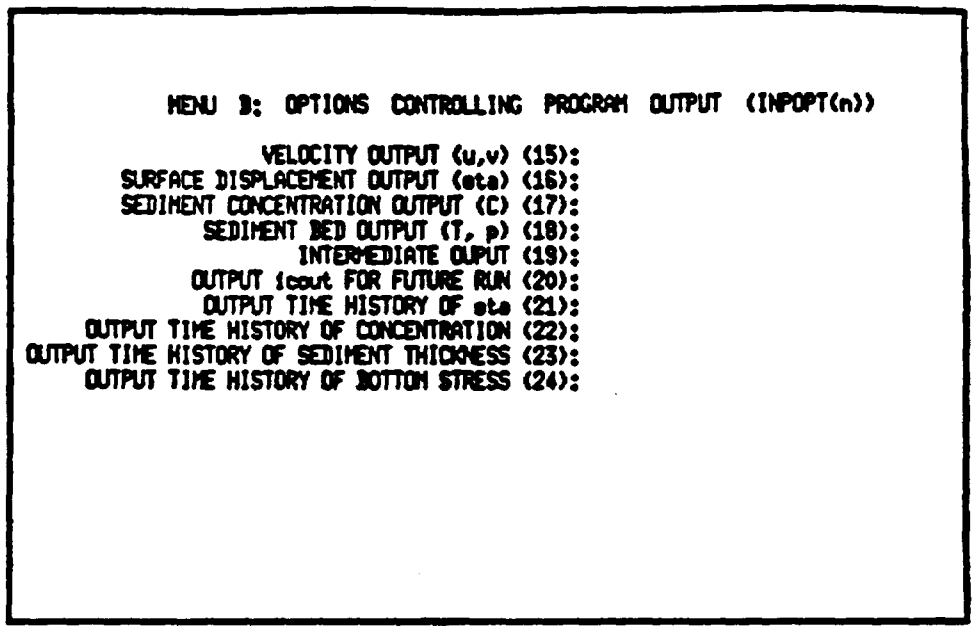


Figure 4.7 Menu B for *setup*.

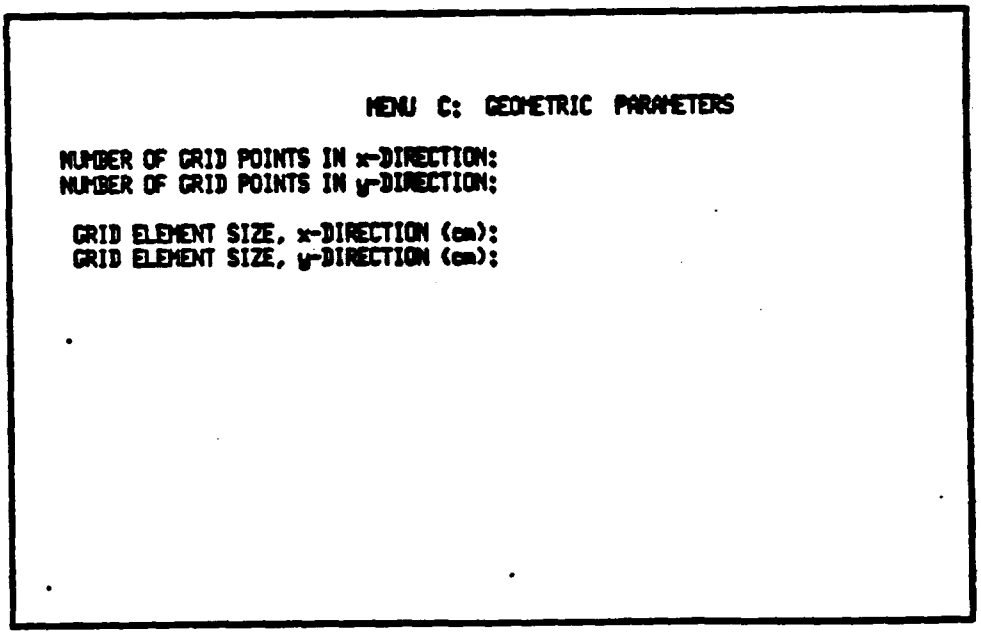


Figure 4.8 Menu C for *setup*.

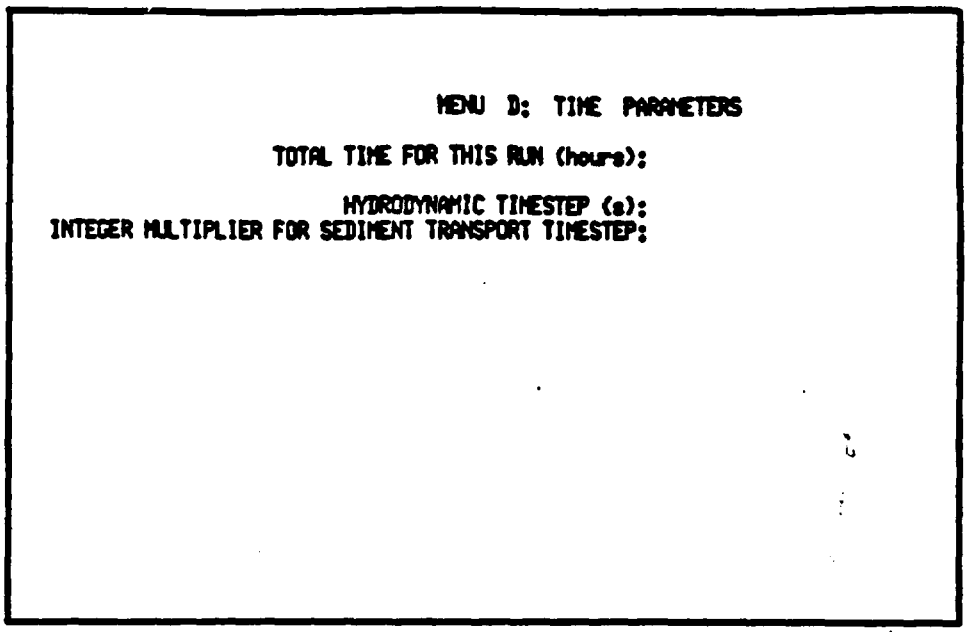


Figure 4.9 Menu D for *setup*.

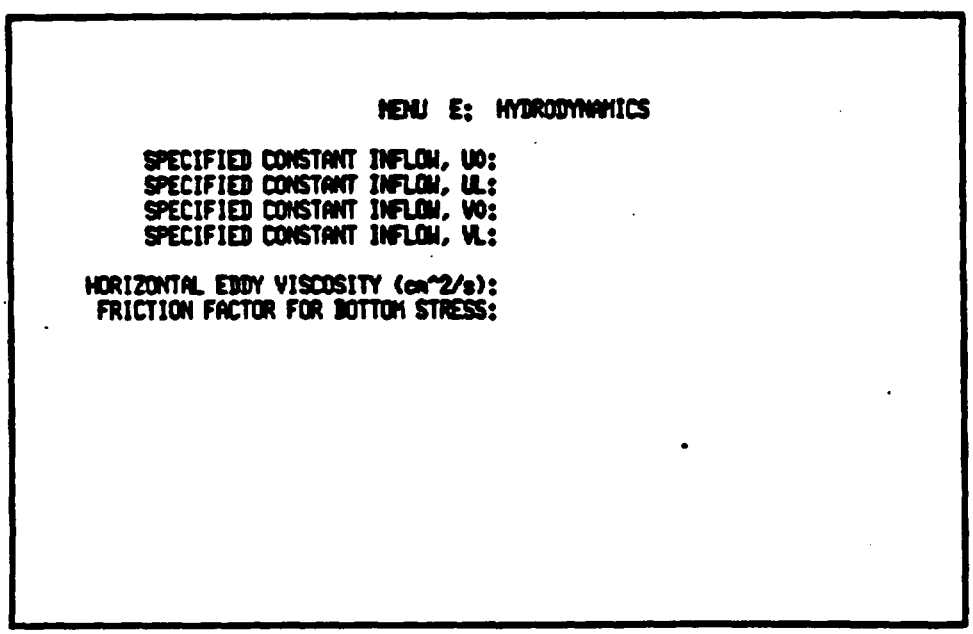


Figure 4.10 Menu E for *setup*.

```
                    MENU F: SEDIMENT TRANSPORT

      HORIZONTAL EDDY DIFFUSIVITY (cm^2/s):

    NUMBER OF SEDIMENT PARTICLE SIZE-CLASSES:

                RESUSPENSION CONSTANT a0in:
              FLOCCULATION CONSTANT alpha:
          MAXIMUM FLOC DIAMETER (microns):
             RESUSPENSION EXPONENT reaexp:
COARSE SIZE-CLASS SETTLING SPEED (microns/s):
            BOTTOM SHEAR STRESS FACTOR cb:
 CRITICAL SHEAR STRESS FOR MEDIUM SIZE-CLASS:
```

Figure 4.11 Menu F for *setup*.

```
          MENU G: SEDIMENT BED STRUCTURE

             A. MAXIMUM NUMBER OF LAYERS, laymax
             B. TIME AFTER DEPOSITION OF BED LAYERS, ftime(1)
             C. INITIAL 'THICKNESS' OF BED LAYERS, tbed0(1)
             D. CRITICAL SHEAR STRESS OF BED LAYERS, taucr(1)

             E. QUIT

        ENTER YOUR CHOICE:
```

Figure 4.12 Menu G for *setup*.

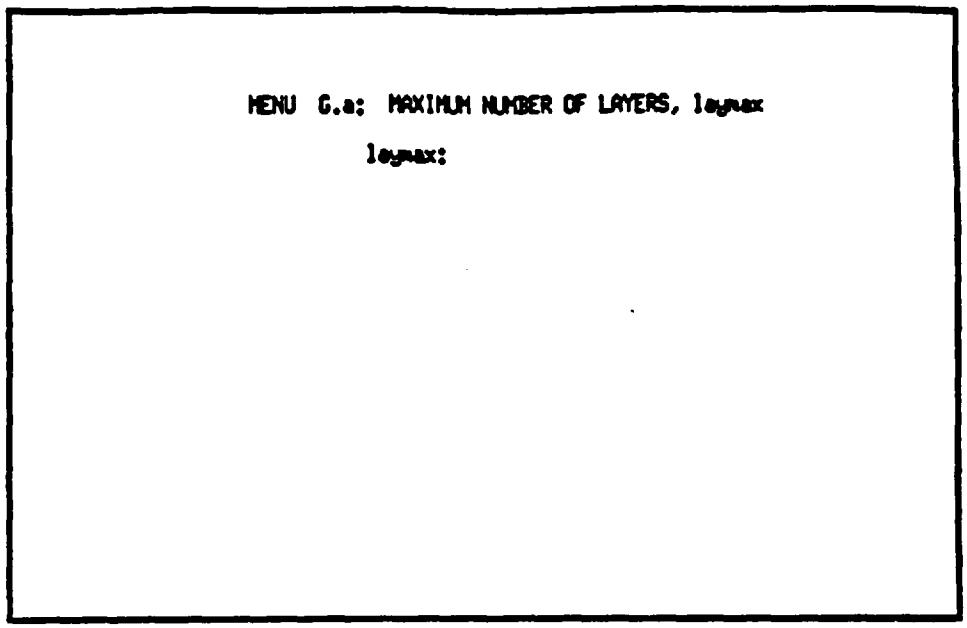


Figure 4.13 Menu G.a for *setup*.

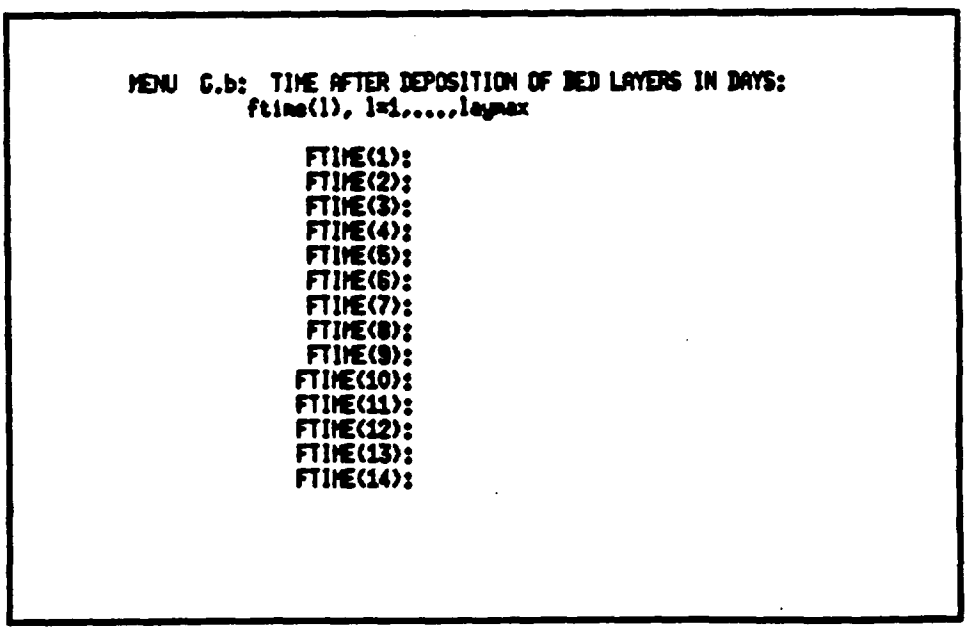


Figure 4.14 Menu G.b for *setup*.

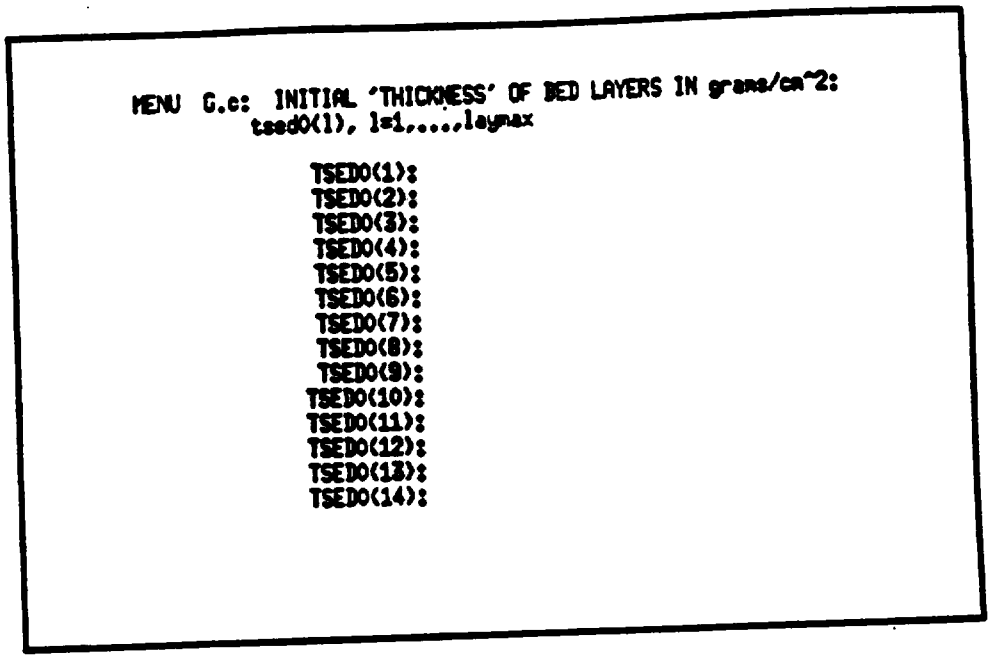


Figure 4.15 Menu G.c for *setup*.

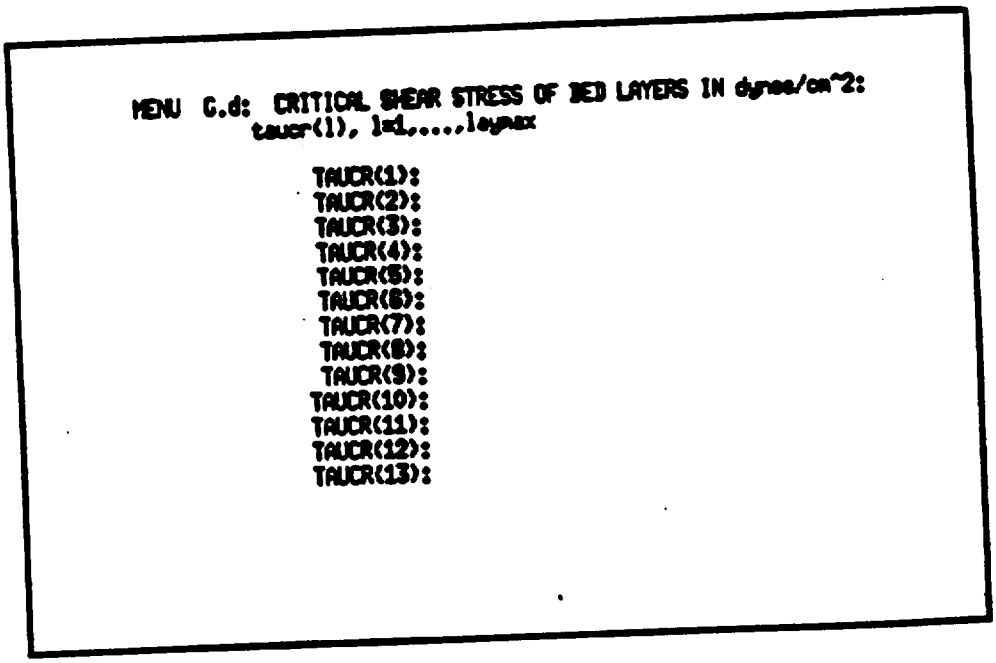


Figure 4.16 Menu G.d for *setup*.

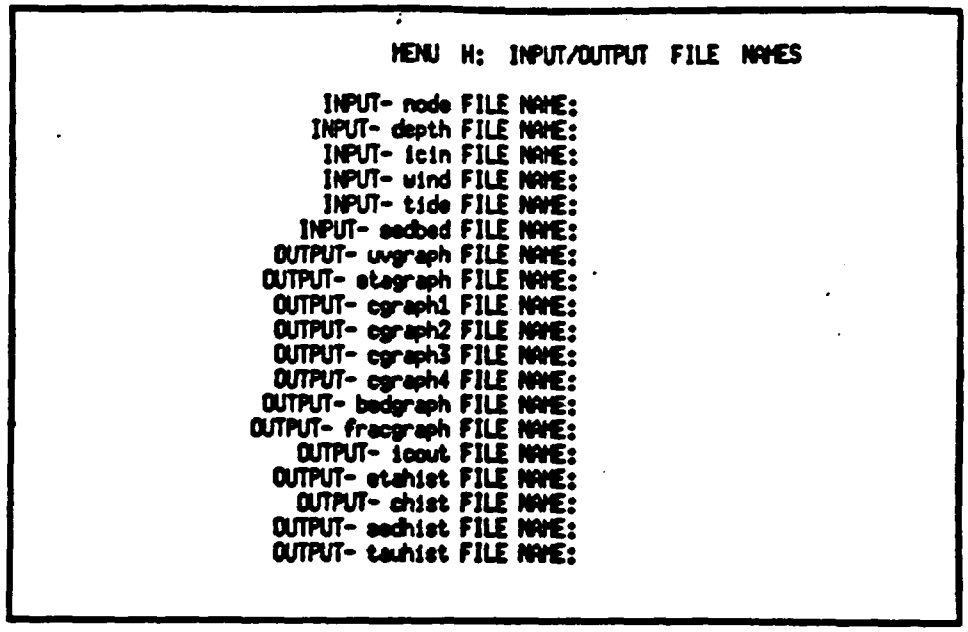


Figure 4.17 Menu H for *setup*.

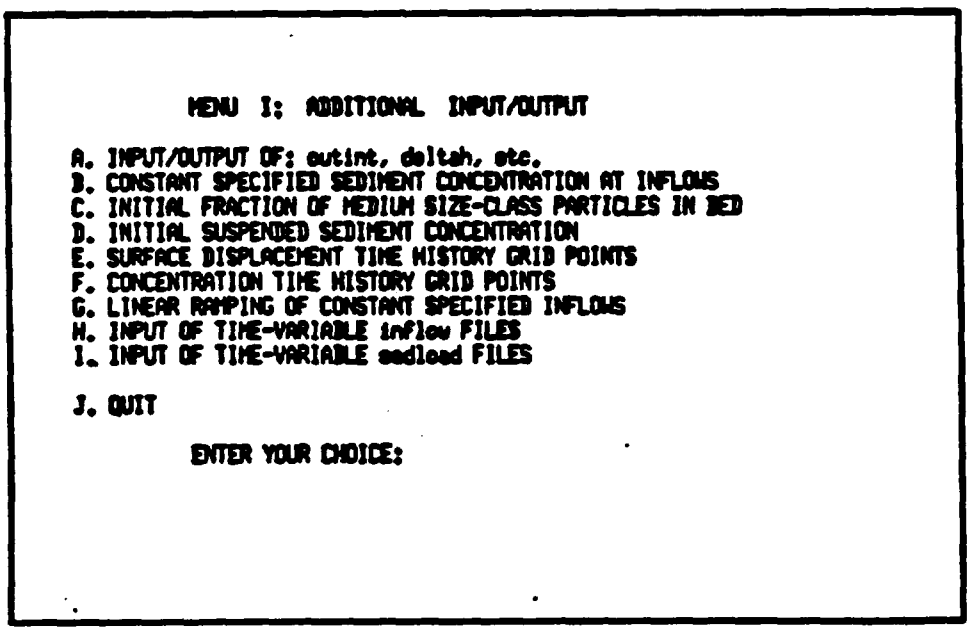


Figure 4.18 Menu I for *setup*.

```
                    MENU I.a: ADDITIONAL VARIABLES

          TIME BETWEEN INTERMEDIATE OUTPUT outint (hours):

     CONSTANT DEPTH ADDED TO ALL BOTTOM DEPTHS deltah (cm):

TIME INTERVAL BETWEEN VALUES IN wind FILE - twind (hours):

          NUMBER OF GRID POINTS FOR ETA TIME HISTORY neta:

 NUMBER OF GRID POINTS FOR CONCENTRATION TIME HISTORY nct:
 TIME INTERVAL OF CONC. TIME HISTORY OUTPUT ctout (hours):

                        CORIOLIS PARAMETER fcor (1/s):

              SINUSOIDAL TIDE/SEICHE AMPLITUDE ampl (cm):
            SINUSOIDAL TIDE/SEICHE PERIOD cmpl (hours):

      NODE-NUMBER OF OPEN BOUNDARY FOR TIDE/SEICHE nodtid:
```

Figure 4.19 Menu I.a for *setup*.

```
MENU I.b: CONSTANT SPECIFIED SEDIMENT CONCENTRATION AT INFLOW IN mg/liter

                          CXO(1):
                          CXO(2):
                          CXO(3):

                          CXL(1):
                          CXL(2):
                          CXL(3):

                          CYO(1):
                          CYO(2):
                          CYO(3):

                          CYL(1):
                          CYL(2):
                          CYL(3):
```

Figure 4.20 Menu I.b for *setup*.

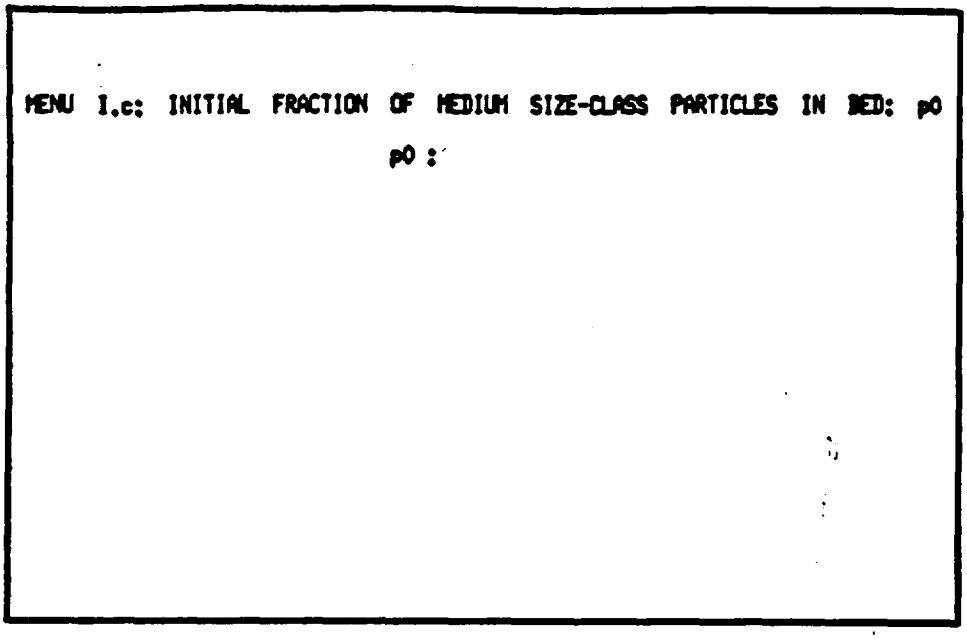


Figure 4.21 Menu I.c for *setup*.

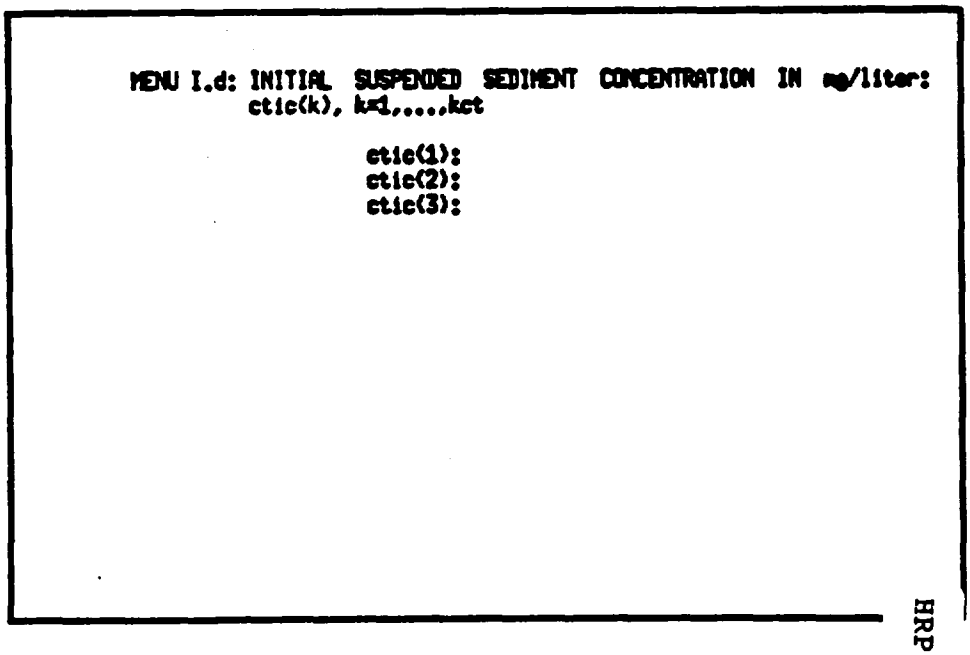


Figure 4.22 Menu I.d for *setup*.

```
MENU I.e: SURFACE DISPLACEMENT TIME HISTORY GRID POINTS

 IETA(1):                          JETA(1):
 IETA(2):                          JETA(2):
 IETA(3):                          JETA(3):
 IETA(4):                          JETA(4):
 IETA(5):                          JETA(5):
 IETA(6):                          JETA(6):
 IETA(7):                          JETA(7):
 IETA(8):                          JETA(8):
 IETA(9):                          JETA(9):
IETA(10):                         JETA(10):
IETA(11):                         JETA(11):
IETA(12):                         JETA(12):
IETA(13):                         JETA(13):
IETA(14):                         JETA(14):
IETA(15):                         JETA(15):
IETA(16):                         JETA(16):
IETA(17):                         JETA(17):
IETA(18):                         JETA(18):
IETA(19):                         JETA(19):
IETA(20):                         JETA(20):
```

Figure 4.23 Menu I.e for *setup*.

```
MENU I.f: CONCENTRATION TIME HISTORY GRID POINTS

 ICT(1):                           JCT(1):
 ICT(2):                           JCT(2):
 ICT(3):                           JCT(3):
 ICT(4):                           JCT(4):
 ICT(5):                           JCT(5):
 ICT(6):                           JCT(6):
 ICT(7):                           JCT(7):
 ICT(8):                           JCT(8):
 ICT(9):                           JCT(9):
ICT(10):                          JCT(10):
ICT(11):                          JCT(11):
ICT(12):                          JCT(12):
ICT(13):                          JCT(13):
ICT(14):                          JCT(14):
ICT(15):                          JCT(15):
ICT(16):                          JCT(16):
ICT(17):                          JCT(17):
ICT(18):                          JCT(18):
ICT(19):                          JCT(19):
ICT(20):                          JCT(20):
```

Figure 4.24 Menu I.f for *setup*.

```
        MENU I.g: LINEAR RAMPING OF CONSTANT SPECIFIED INFLOWS

NUMBER OF TIMESTEPS TO LINEARLY INCREASE UO - nramp(1):
NUMBER OF TIMESTEPS TO LINEARLY INCREASE UL - nramp(2):
NUMBER OF TIMESTEPS TO LINEARLY INCREASE VO - nramp(3):
NUMBER OF TIMESTEPS TO LINEARLY INCREASE VL - nramp(4):
```

Figure 4.25 Menu I.g for *setup*.

```
        MENU I.h: INPUT OF TIME-VARIABLE inflow FILES

        TOTAL NUMBER OF inflow FILES TO INPUT - nqin:
                                    inflow1 FILE NAME:
                                    inflow2 FILE NAME:
                                    inflow3 FILE NAME:
                                    inflow4 FILE NAME:
            node-number FOR inflow1 FILE - nodeq(1):
            node-number FOR inflow2 FILE - nodeq(2):
            node-number FOR inflow3 FILE - nodeq(3):
            node-number FOR inflow4 FILE - nodeq(4):
  NUMBER OF INFLOW VALUES TO READ FROM inflow1 - nts(1):
  NUMBER OF INFLOW VALUES TO READ FROM inflow2 - nts(2):
  NUMBER OF INFLOW VALUES TO READ FROM inflow3 - nts(3):
  NUMBER OF INFLOW VALUES TO READ FROM inflow4 - nts(4):
TIME BETWEEN EACH INFLOW VALUE IN inflow1 - ts(1) (hours):
TIME BETWEEN EACH INFLOW VALUE IN inflow2 - ts(2) (hours):
TIME BETWEEN EACH INFLOW VALUE IN inflow3 - ts(3) (hours):
TIME BETWEEN EACH INFLOW VALUE IN inflow4 - ts(4) (hours):
  RAMPING TIME BETWEEN VALUES IN inflow1 - tr(1) (hours):
  RAMPING TIME BETWEEN VALUES IN inflow2 - tr(2) (hours):
  RAMPING TIME BETWEEN VALUES IN inflow3 - tr(3) (hours):
  RAMPING TIME BETWEEN VALUES IN inflow4 - tr(4) (hours):
```

Figure 4.26 Menu I.h for *setup*.

```
            MENU I.1: INPUT OF TIME-VARIABLE sedload FILES

            TOTAL NUMBER OF sedload FILES TO INPUT - ncin:
                                        sedload1 FILE NAME:
                                        sedload2 FILE NAME:
                                        sedload3 FILE NAME:
                                        sedload4 FILE NAME:
                node-number FOR sedload1 FILE - nodsc(1):
                node-number FOR sedload2 FILE - nodsc(2):
                node-number FOR sedload3 FILE - nodsc(3):
                node-number FOR sedload4 FILE - nodsc(4):
TOTAL NUMBER OF CONC. VALUES TO READ FROM sedload1 - ntc(1):
TOTAL NUMBER OF CONC. VALUES TO READ FROM sedload2 - ntc(2):
TOTAL NUMBER OF CONC. VALUES TO READ FROM sedload3 - ntc(3):
TOTAL NUMBER OF CONC. VALUES TO READ FROM sedload4 - ntc(4):
TIME BETWEEN EACH CONC. VALUE IN sedload 1 - tsc(1) (hours):
TIME BETWEEN EACH CONC. VALUE IN sedload 2 - tsc(2) (hours):
TIME BETWEEN EACH CONC. VALUE IN sedload 3 - tsc(3) (hours):
TIME BETWEEN EACH CONC. VALUE IN sedload 4 - tsc(4) (hours):
    RAMPING TIME BETWEEN VALUES IN inflow1 - trc(1) (hours):
    RAMPING TIME BETWEEN VALUES IN inflow2 - trc(2) (hours):
    RAMPING TIME BETWEEN VALUES IN inflow3 - trc(3) (hours):
    RAMPING TIME BETWEEN VALUES IN inflow4 - trc(4) (hours):
```

Figure 4.27 Menu I.1 for *setup*.

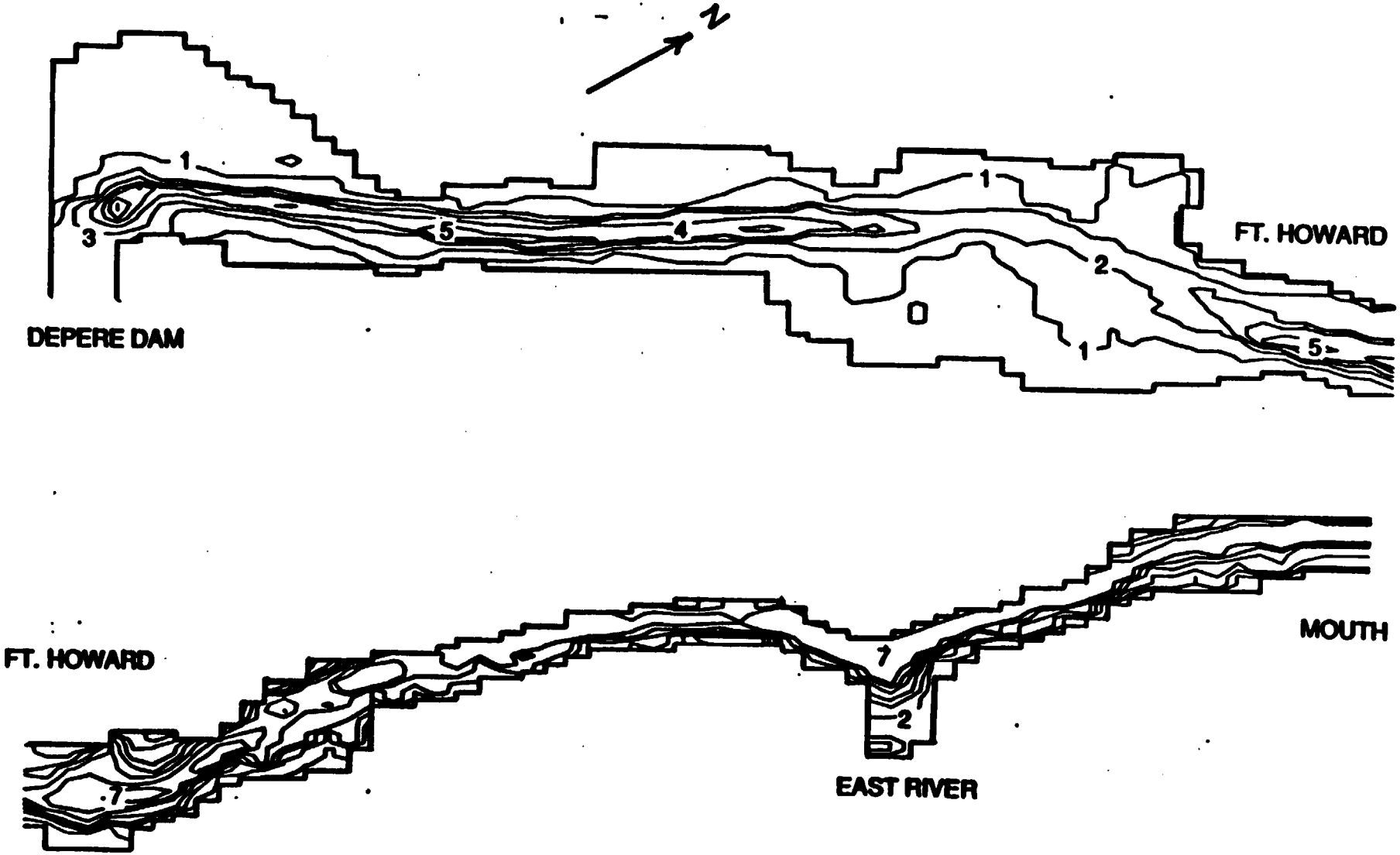


Figure 5.1 Computational grid and bathymetry for Lower Fox River.

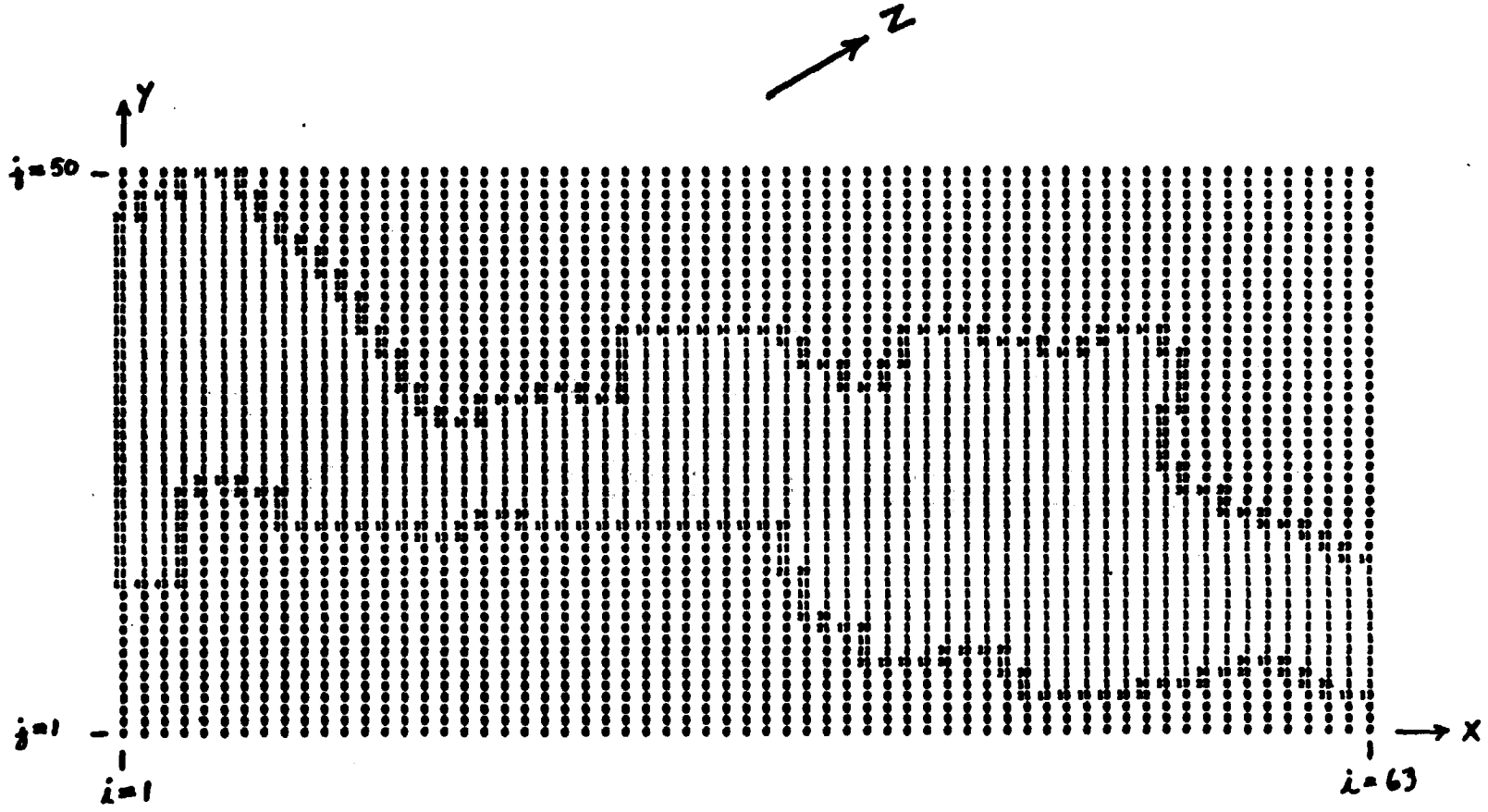


Figure 5.2a *node-number* matrix for upper portion of Lower Fox River computational grid.

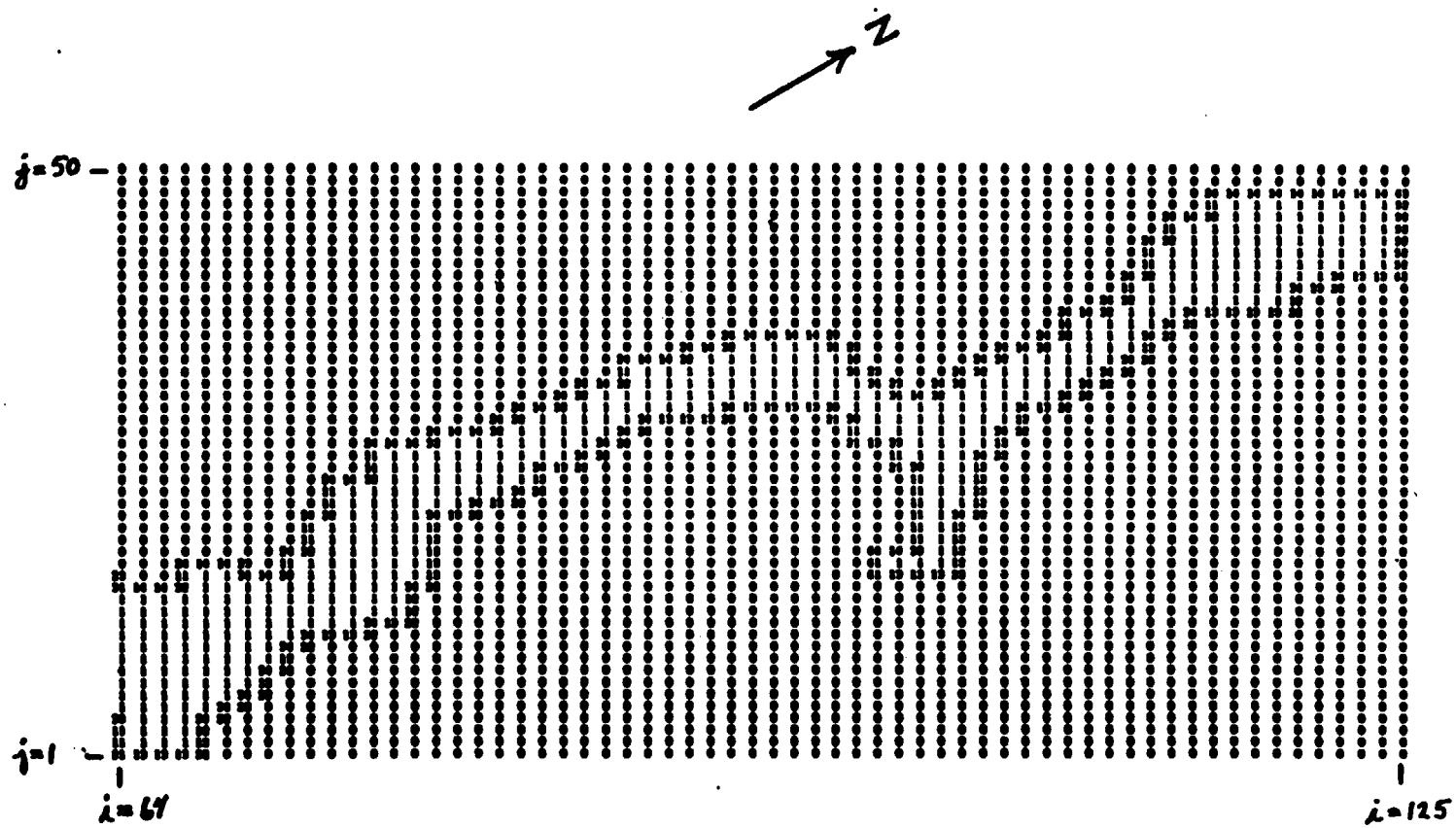


Figure 5.2b *node-number* matrix for lower portion of Lower Fox River computational grid.

```
      ######  SEDZL :  VERSION 1.0  ######

                    JUNE, 1990  RELEASE

*****************************************

              RUN NUMBER:    fox.1

*****************************************

  PROGRAM  OPTION  SETTINGS

      INPOPT( 1) =  0
      INPOPT( 2) =  0
      INPOPT( 3) =  0
      INPOPT( 4) =  1
      INPOPT( 5) =  1
      INPOPT( 6) =  0
      INPOPT( 7) =  0
      INPOPT( 8) =  0
      INPOPT( 9) =  1
      INPOPT(10) =  0
      INPOPT(11) =  1
      INPOPT(12) =  1
      INPOPT(13) =  1
      INPOPT(14) =  0
      INPOPT(15) =  1
      INPOPT(16) =  1
      INPOPT(17) =  0
      INPOPT(18) =  0
      INPOPT(19) =  1
      INPOPT(20) =  1
      INPOPT(21) =  1
      INPOPT(22) =  0
      INPOPT(23) =  0
      INPOPT(24) =  0

*****************************************

    INPUT - node FILE NAME :      node.fox

    INPUT - depth FILE NAME :      depth.fox

    INPUT - tide FILE NAME :      seiche.fox.1

    INPUT - inflow FILE NAMES :
        inflow.fox.1

*****************************************
```

Figure 5.3 Output file *out.fox.1* for Lower Fox River example.

```
   OUTPUT - uvgraph FILE NAME :      out.uv.fox.1

   OUTPUT - etagraph FILE NAME :      out.eta.fox.1

   OUTPUT - icout FILE NAME :      out.ic.fox.1

   OUTPUT - etahist FILE :      out.etahist.fox.1
****************************************

      ***** INPUT PARAMETERS (DIMENSIONS ARE IN cgs UNITS) *****

   MAXIMUM TIME FOR THIS RUN (hours) :        6.0

   MX, MY, XSTP, YSTP :    125  50        9000.        3000.

   TSTP, IRTSTP :       3.00    1

   AH, CF :       10000.  0.0020

   U0, UL, V0, VL :         15.0          0.0          0.0          0.0

   DELTAH :      50.

   NRAMP(1) :     600
   NRAMP(2) :       0
   NRAMP(3) :       0
   NRAMP(4) :       0

   NQIN :    1

      N, NODEQ, NTS, TS, TR :      1    43     3    3.00    1.00

   NODTID :    52

   OUTINT (hours) :       3.0

   METAS :     3

      M, IETA, JETA :      1     4    14
      M, IETA, JETA :      2    63    10
      M, IETA, JETA :      3   125    45

****************************************

      MAXIMUM COURANT NUMBER OF  0.521  OCCURS AT i,j =   78   24

****************************************

      TOTAL NUMBER OF GRID POINTS FOR THIS PROBLEM:    2078

****************************************
```

Figure 5.3 (cont.) Output file *out.fox.1* for Lower Fox River example.

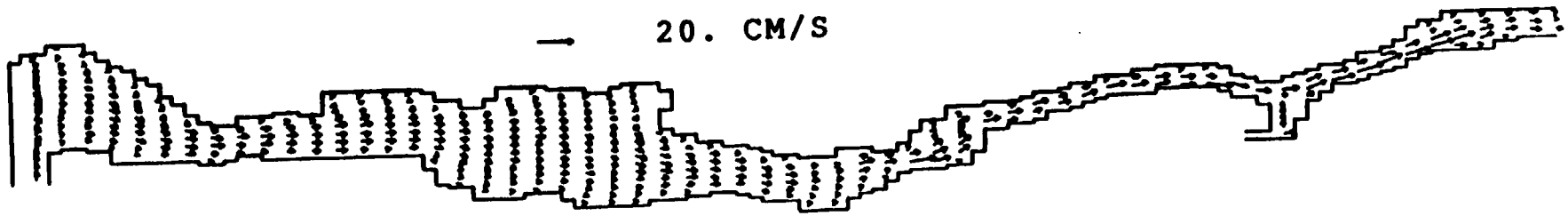
```
##### TIME =    0.3000E+01  HOURS

MINIMUM VELOCITY AT  119  46 IS      0.0
MAXIMUM VELOCITY AT  102  17 IS     17.5
  MINIMUM ETA AT  125  41 IS     12.0
  MAXIMUM ETA AT  104  17 IS     20.5

##### TIME =    0.6000E+01  HOURS

MINIMUM VELOCITY AT   65   2 IS      0.0
MAXIMUM VELOCITY AT  107  32 IS     18.9
  MINIMUM ETA AT  125  41 IS     11.0
  MAXIMUM ETA AT  104  17 IS     24.8
```

Figure 5.3 (cont.) Output file *outfox.1* for Lower Fox River example.



HRP 002 0380

Figure 5.4 Velocity field at end of run *fox.1*.

```
      ######  SEDZL :  VERSION 1.0  ######

                JUNE, 1990  RELEASE

* ****************************************

            RUN NUMBER:   fox.2

* ****************************************

   PROGRAM  OPTION  SETTINGS

      INPOPT( 1) =  1
      INPOPT( 2) =  0
      INPOPT( 3) =  1
      INPOPT( 4) =  1
      INPOPT( 5) =  1
      INPOPT( 6) =  0
      INPOPT( 7) =  0
      INPOPT( 8) =  0
      INPOPT( 9) =  1
      INPOPT(10) =  0
      INPOPT(11) =  1
      INPOPT(12) =  0
      INPOPT(13) =  1
      INPOPT(14) =  0
      INPOPT(15) =  1
      INPOPT(16) =  1
      INPOPT(17) =  1
      INPOPT(18) =  1
      INPOPT(19) =  1
      INPOPT(20) =  0
      INPOPT(21) =  0
      INPOPT(22) =  1
      INPOPT(23) =  1
      INPOPT(24) =  1

*****************************************

   INPUT - node FILE NAME :     node.fox

   INPUT - depth FILE NAME :     depth.fox

   INPUT - icin FILE NAME :     out.ic.fox.1

   INPUT - tide FILE NAME :     seiche.fox.2

   INPUT - inflow FILE NAMES :
       inflow.fox.2
```

Figure 5.5 Output file *out.fox.2* for Lower Fox River example.

```
     INPUT - sedload FILE NAMES :
          sedload.fox.2

* ****************************************

     OUTPUT - uvgraph FILE NAME :      out.uv.fox.2
     OUTPUT - etagraph FILE NAME :      out.eta.fox.2
     OUTPUT - cgraph1 FILE NAME (TOTAL CONC.) :      out.ctot.fox.2
     OUTPUT - cgraph2 FILE NAME (FINE CONC.) :      out.cfin.fox.2
     OUTPUT - cgraph3 FILE NAME (MEDIUM CONC.) :      out.cmed.fox.2
     OUTPUT - cgraph4 FILE NAME (COARSE CONC.) :      out.ccor.fox.2
     OUTPUT - bedgraph FILE NAME :      out.bed.fox.2
     OUTPUT - fracgraph FILE NAME :      out.frac.fox.2
     OUTPUT - chist FILE NAME :      out.chist.fox.2
     OUTPUT - sedhist FILE NAME :      out.sedhist.fox.2
     OUTPUT - tauhist FILE NAME :      out.tauhist.fox.2
** ****************************************

          *****  INPUT PARAMETERS (DIMENSIONS ARE IN cgs UNITS)  *****
     MAXIMUM TIME FOR THIS RUN (hours) :       12.0
     MX, MY, XSTP, YSTP :   125  50       9000.       3000.
     TSTP, IRTSTP :       3.00    5
     AH, CF :       10000.  0.0020
     U0, UL, V0, VL :         15.0         0.0         0.0         0.0
     DH, KCT =     20000.    3
     CX0(k), k=1,..,KCT (mg/liter):       5.0    15.0     7.0
     CXL(k), k=1,..,KCT (mg/liter):       0.0     0.0     0.0
     CY0(k), k=1,..,KCT (mg/liter):       0.0     0.0     0.0
     CY0(k), k=1,..,KCT (mg/liter):       0.0     0.0     0.0
     A0IN, ALPHA, DMAX (microns), RESEXP, BETA (microns/s), CB, TCRDEP :
          0.0080  0.100E-07  200.0    3.0 1000.0   1.00  0.1000
```

Figure 5.5 (cont.) Output file *out.fox.2* for Lower Fox River example.

```
 LAYMAX :    10

 FTIME (l), l=1,...,LAYMAX (days) :
      0.005  0.125  0.250  0.500  1.000  2.000  3.000  4.000  5.000  6.000

 TSED0(l), l=1,...,LAYMAX :
       0.0    0.0    0.0    0.0    0.0    0.0    0.0    0.0    0.0   10.0

 TAUCR(l), l=1,...,LAYMAX :
       0.10   0.20   0.40   0.80   1.00   1.00   1.00   1.00   1.00   1.00

 P0 :    0.50

 CTIC(k), k=1,...,KCT (mg/liter) :        5.0    10.0     5.0

 DELTAH :     50.

 NQIN :    1

    N, NODEQ, NTS, TS, TR :      1    43     9    3.00    1.00

 NCIN :    1

    N, NODEC, NTC, TSC, TRC :      1    43     7    4.00    1.00

 NODTID :    52

 OUTINT (hours) :       6.0

 MCT, CTOUT (hours) :      3    1.0

    M, ICT, JCT :      1     4    14
    M, ICT, JCT :      2    63    10
    M, ICT, JCT :      3   125    45

*****************************************

      MAXIMUM COURANT NUMBER OF  0.521  OCCURS AT i,j =   78   24

*****************************************

      TOTAL NUMBER OF GRID POINTS FOR THIS PROBLEM:    2078

*****************************************


 ##### TIME =    0.1200E+02  HOURS

 MINIMUM VELOCITY AT   65   2 IS      0.0

 MAXIMUM VELOCITY AT  107  32 IS     29.1
```


Figure 5.5 (cont.) Output file *out.fox2* for Lower Fox River example.

```
 MINIMUM ETA AT  113  42 IS     -9.6

 MAXIMUM ETA AT    4  22 IS      3.3

  TOTAL SEDIMENT CONCENTRATION
    MINIMUM CONCENTRATION AT I, J =   3  37 IS   0.6043E+01
    MAXIMUM CONCENTRATION AT I, J =   1  14 IS   0.2300E+02

  FINE SEDIMENT CONCENTRATION
    MINIMUM CONCENTRATION AT I, J = 100  17 IS   0.0000E+00
    MAXIMUM CONCENTRATION AT I, J =   1  14 IS   0.6000E+01

  MEDIUM SEDIMENT CONCENTRATION
    MINIMUM CONCENTRATION AT I, J =   5  50 IS   0.2530E+01
    MAXIMUM CONCENTRATION AT I, J = 100  17 IS   0.1500E+02

  COARSE SEDIMENT CONCENTRATION
    MINIMUM CONCENTRATION AT I, J = 108  33 IS  -0.1793E+00
    MAXIMUM CONCENTRATION AT I, J =   1  14 IS   0.1200E+02

    MINIMUM SEDIMENT THICKNESS AT I, J = 107  32 IS  -0.3444E-04

    MAXIMUM SEDIMENT THICKNESS AT I, J = 100  18 IS   0.1732E-01

##### TIME =    0.1800E+02  HOURS

MINIMUM VELOCITY AT   49   5 IS      0.0

MAXIMUM VELOCITY AT  102  17 IS     17.4

  MINIMUM ETA AT  124  42 IS     -3.6

  MAXIMUM ETA AT  104  17 IS      3.2

  TOTAL SEDIMENT CONCENTRATION
    MINIMUM CONCENTRATION AT I, J = 125  42 IS   0.4660E+01
    MAXIMUM CONCENTRATION AT I, J =   1  14 IS   0.3000E+02

  FINE SEDIMENT CONCENTRATION
    MINIMUM CONCENTRATION AT I, J = 100  17 IS   0.0000E+00
    MAXIMUM CONCENTRATION AT I, J =   1  14 IS   0.8000E+01

  MEDIUM SEDIMENT CONCENTRATION
    MINIMUM CONCENTRATION AT I, J =  11  43 IS   0.9758E+00
    MAXIMUM CONCENTRATION AT I, J = 100  17 IS   0.1500E+02

  COARSE SEDIMENT CONCENTRATION
    MINIMUM CONCENTRATION AT I, J =  46   4 IS   0.1665E-08
    MAXIMUM CONCENTRATION AT I, J =   1  14 IS   0.1600E+02

    MINIMUM SEDIMENT THICKNESS AT I, J =  73  13 IS   0.1338E-03

    MAXIMUM SEDIMENT THICKNESS AT I, J =   1  14 IS   0.4148E-01
```

Figure 5.5 (cont.) Output file *out.fox2* for Lower Fox River example.

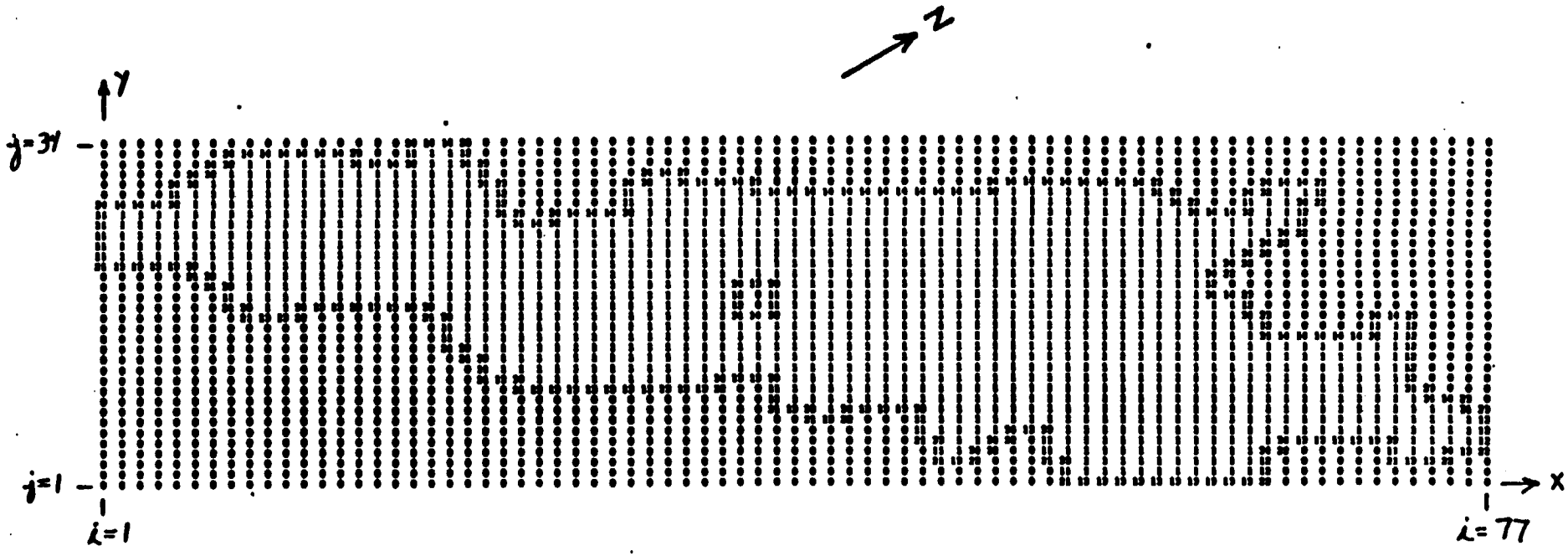


Figure 5.6 *node-number* matrix for Green Bay computational grid.

```
      ######  SEDZL :  VERSION 1.0  ######

                    JUNE, 1990  RELEASE
*****************************************

                RUN NUMBER:    gbay.1

*****************************************

     PROGRAM  OPTION  SETTINGS

        INPOPT( 1) =  1
        INPOPT( 2) =  0
        INPOPT( 3) =  0
        INPOPT( 4) =  1
        INPOPT( 5) =  1
        INPOPT( 6) =  1
        INPOPT( 7) =  1
        INPOPT( 8) =  1
        INPOPT( 9) =  0
        INPOPT(10) =  0
        INPOPT(11) =  0
        INPOPT(12) =  0
        INPOPT(13) =  0
        INPOPT(14) =  0
        INPOPT(15) =  1
        INPOPT(16) =  1
        INPOPT(17) =  1
        INPOPT(18) =  1
        INPOPT(19) =  1
        INPOPT(20) =  0
        INPOPT(21) =  0
        INPOPT(22) =  0
        INPOPT(23) =  0
        INPOPT(24) =  0
*****************************************

     INPUT - node FILE NAME :      node.gbay

     INPUT - depth FILE NAME :      depth.gbay

     INPUT - wind FILE NAME :      wind.gbay.1
*****************************************

     OUTPUT - uvgraph FILE NAME :      out.uv.gbay.1

     OUTPUT - etagraph FILE NAME :      out.eta.gbay.1
```

Figure 5.7 Output file *ouu.gbay.1* for Green Bay example.

```
   OUTPUT - cgraph1 FILE NAME (TOTAL CONC.) :      out.ctot.gbay.1
   OUTPUT - cgraph3 FILE NAME (MEDIUM CONC.) :      out.cmed.gbay.1
   OUTPUT - cgraph4 FILE NAME (COARSE CONC.) :      out.ccor.gbay.1
   OUTPUT - bedgraph FILE NAME :      out.bed.gbay.1
   OUTPUT - fracgraph FILE NAME :      out.frac.gbay.1
** ****************************************

      *****  INPUT PARAMETERS (DIMENSIONS ARE IN cgs UNITS)  *****
   MAXIMUM TIME FOR THIS RUN (hours) :       96.0
   MX, MY, XSTP, YSTP :     77  34     243840.     121920.
   TSTP, IRTSTP :      45.00    5
   AH, CF :      500000.  0.0020
   U0, UL, V0, VL :          0.0          0.0          0.0          0.0
   DH, KCT =  2000000.    2
   CX0(k), k=1,..,KCT (mg/liter):        0.0      0.0
   CXL(k), k=1,..,KCT (mg/liter):        0.0      0.0
   CY0(k), k=1,..,KCT (mg/liter):        0.0      0.0
   CY0(k), k=1,..,KCT (mg/liter):        0.0      0.0
   AOIN, ALPHA, DMAX (microns), RESEXP, BETA (microns/s), CB, TCRDEP :
        0.0080  0.100E-07  200.0    3.0 1000.0   0.50  0.1000
   LAYMAX :     10
   FTIME (l), l=1,...,LAYMAX (days) :
        0.005  0.125  0.250  0.500  1.000  2.000  3.000  4.000  5.000  6.000
   TSED0(l), l=1,...,LAYMAX :
         0.0   0.0   0.0   0.0   1.0   1.0   1.0   1.0   1.0 100.0
   TAUCR(l), l=1,...,LAYMAX :
         0.10   0.20   0.40   0.80   1.00   1.00   1.00   1.00   1.00   1.00
   P0 :     0.50
   CTIC(k), k=1,...,KCT (mg/liter) :        0.0      0.0
   DELTAH :     100.
```

Figure 5.7 (cont.) Output file *out.gbay.1* for Green Bay example..

```
     TWIND (hours) :       1.0

     FCOR :   0.00010

     OUTINT (hours) :     12.0

** ****************************************

       MAXIMUM SIGMA OF  0.385  OCCURS AT i,j =   58   19

** ****************************************

       TOTAL NUMBER OF GRID POINTS FOR THIS PROBLEM:    1491

** ****************************************


     ##### TIME =    0.1200E+02  HOURS

     MINIMUM VELOCITY AT   47   6 IS      0.0

     MAXIMUM VELOCITY AT   61  19 IS     10.6

       MINIMUM ETA AT    1  22 IS     -2.1

       MAXIMUM ETA AT   67  26 IS      2.1

        TOTAL SEDIMENT CONCENTRATION
          MINIMUM CONCENTRATION AT I, J =  54   1 IS   0.0000E+00
          MAXIMUM CONCENTRATION AT I, J =  54   1 IS   0.0000E+00

        MEDIUM SEDIMENT CONCENTRATION
          MINIMUM CONCENTRATION AT I, J =  54   1 IS   0.0000E+00
          MAXIMUM CONCENTRATION AT I, J =  54   1 IS   0.0000E+00

        COARSE SEDIMENT CONCENTRATION
          MINIMUM CONCENTRATION AT I, J =  54   1 IS   0.0000E+00
          MAXIMUM CONCENTRATION AT I, J =  54   1 IS   0.0000E+00

          MINIMUM SEDIMENT THICKNESS AT I, J =  54   1 IS   0.0000E+00

          MAXIMUM SEDIMENT THICKNESS AT I, J =  55   1 IS   0.0000E+00


     ##### TIME =    0.2400E+02  HOURS

     MINIMUM VELOCITY AT   43  28 IS      0.0

     MAXIMUM VELOCITY AT   13  19 IS      4.3

       MINIMUM ETA AT    1  28 IS     -1.2
```

Figure 5.7 (cont.) Output file *out.gbay.1* for Green Bay example.

```
 MAXIMUM ETA AT   72   3 IS      1.1

  TOTAL SEDIMENT CONCENTRATION
    MINIMUM CONCENTRATION AT I, J =  54   1 IS   0.0000E+00
    MAXIMUM CONCENTRATION AT I, J =  54   1 IS   0.0000E+00

  MEDIUM SEDIMENT CONCENTRATION
    MINIMUM CONCENTRATION AT I, J =  54   1 IS   0.0000E+00
    MAXIMUM CONCENTRATION AT I, J =  54   1 IS   0.0000E+00

  COARSE SEDIMENT CONCENTRATION
    MINIMUM CONCENTRATION AT I, J =  54   1 IS   0.0000E+00
    MAXIMUM CONCENTRATION AT I, J =  54   1 IS   0.0000E+00

    MINIMUM SEDIMENT THICKNESS AT I, J =  54   1 IS   0.0000E+00

    MAXIMUM SEDIMENT THICKNESS AT I, J =  55   1 IS   0.0000E+00

##### TIME =    0.3600E+02  HOURS

MINIMUM VELOCITY AT   24  12 IS      0.0

MAXIMUM VELOCITY AT   36  15 IS      9.4

 MINIMUM ETA AT   77   4 IS     -2.8

 MAXIMUM ETA AT    8  33 IS      3.3

  TOTAL SEDIMENT CONCENTRATION
    MINIMUM CONCENTRATION AT I, J =  54   1 IS   0.0000E+00
    MAXIMUM CONCENTRATION AT I, J =  54   1 IS   0.0000E+00

  MEDIUM SEDIMENT CONCENTRATION
    MINIMUM CONCENTRATION AT I, J =  54   1 IS   0.0000E+00
    MAXIMUM CONCENTRATION AT I, J =  54   1 IS   0.0000E+00

  COARSE SEDIMENT CONCENTRATION
    MINIMUM CONCENTRATION AT I, J =  54   1 IS   0.0000E+00
    MAXIMUM CONCENTRATION AT I, J =  54   1 IS   0.0000E+00

    MINIMUM SEDIMENT THICKNESS AT I, J =  54   1 IS   0.0000E+00

    MAXIMUM SEDIMENT THICKNESS AT I, J =  55   1 IS   0.0000E+00

##### TIME =    0.4800E+02  HOURS

MINIMUM VELOCITY AT   67  29 IS      0.0

MAXIMUM VELOCITY AT   24  24 IS     36.5

 MINIMUM ETA AT   76   9 IS    -11.3
```

Figure 5.7 (cont.) Output file *ou.gbay.1* for Green Bay example.

```
 MAXIMUM ETA AT    1  25 IS     13.2

  TOTAL SEDIMENT CONCENTRATION
    MINIMUM CONCENTRATION AT I, J =  25  24 IS  -0.1765E+00
    MAXIMUM CONCENTRATION AT I, J =  24  24 IS   0.1249E+02

  MEDIUM SEDIMENT CONCENTRATION
    MINIMUM CONCENTRATION AT I, J =  25  24 IS  -0.8832E-01
    MAXIMUM CONCENTRATION AT I, J =  24  24 IS   0.6248E+01

  COARSE SEDIMENT CONCENTRATION
    MINIMUM CONCENTRATION AT I, J =  25  24 IS  -0.8818E-01
    MAXIMUM CONCENTRATION AT I, J =  24  24 IS   0.6237E+01

    MINIMUM SEDIMENT THICKNESS AT I, J =  24  24 IS  -0.5192E-02

    MAXIMUM SEDIMENT THICKNESS AT I, J =  24  25 IS   0.7749E-04

##### TIME =    0.6000E+02  HOURS

MINIMUM VELOCITY AT   49   6 IS      0.1

MAXIMUM VELOCITY AT   24  24 IS     22.1

 MINIMUM ETA AT   73  17 IS     -4.6

 MAXIMUM ETA AT    1  22 IS      6.0

  TOTAL SEDIMENT CONCENTRATION
    MINIMUM CONCENTRATION AT I, J =  68  28 IS  -0.4290E-09
    MAXIMUM CONCENTRATION AT I, J =  36  12 IS   0.1829E+00

  MEDIUM SEDIMENT CONCENTRATION
    MINIMUM CONCENTRATION AT I, J =  54   1 IS   0.7744E-13
    MAXIMUM CONCENTRATION AT I, J =  36  12 IS   0.1486E+00

  COARSE SEDIMENT CONCENTRATION
    MINIMUM CONCENTRATION AT I, J =  16  32 IS  -0.3065E-03
    MAXIMUM CONCENTRATION AT I, J =  35  14 IS   0.3564E-01

    MINIMUM SEDIMENT THICKNESS AT I, J =  24  24 IS  -0.5189E-02

    MAXIMUM SEDIMENT THICKNESS AT I, J =  24  25 IS   0.7027E-03

##### TIME =    0.7200E+02  HOURS

MINIMUM VELOCITY AT   48   4 IS      0.0

MAXIMUM VELOCITY AT   47  28 IS     10.3

 MINIMUM ETA AT   66  30 IS     -1.6
```

Figure 5.7 (cont.) Output file *out.gbay.1* for Green Bay example.

```
  MAXIMUM ETA AT    2  22 IS      3.6

   TOTAL SEDIMENT CONCENTRATION
     MINIMUM CONCENTRATION AT I, J =  57   1 IS   0.2788E-08
     MAXIMUM CONCENTRATION AT I, J =  36  11 IS   0.5687E-01

   MEDIUM SEDIMENT CONCENTRATION
     MINIMUM CONCENTRATION AT I, J =  57   1 IS   0.2766E-08
     MAXIMUM CONCENTRATION AT I, J =  36  11 IS   0.5595E-01

   COARSE SEDIMENT CONCENTRATION
     MINIMUM CONCENTRATION AT I, J =  14  33 IS  -0.7096E-04
     MAXIMUM CONCENTRATION AT I, J =  35  15 IS   0.1882E-02

     MINIMUM SEDIMENT THICKNESS AT I, J =  24  24 IS  -0.5192E-02

     MAXIMUM SEDIMENT THICKNESS AT I, J =  24  25 IS   0.7110E-03


##### TIME =    0.8400E+02  HOURS


MINIMUM VELOCITY AT   47   6 IS      0.0

MAXIMUM VELOCITY AT   47  28 IS     12.8

  MINIMUM ETA AT   76   9 IS     -2.2

  MAXIMUM ETA AT    1  22 IS      3.5

   TOTAL SEDIMENT CONCENTRATION
     MINIMUM CONCENTRATION AT I, J =  57   1 IS   0.1128E-06
     MAXIMUM CONCENTRATION AT I, J =  35  11 IS   0.2797E-01

   MEDIUM SEDIMENT CONCENTRATION
     MINIMUM CONCENTRATION AT I, J =  57   1 IS   0.1127E-06
     MAXIMUM CONCENTRATION AT I, J =  35  11 IS   0.2792E-01

   COARSE SEDIMENT CONCENTRATION
     MINIMUM CONCENTRATION AT I, J =  13  33 IS  -0.2876E-04
     MAXIMUM CONCENTRATION AT I, J =  34  16 IS   0.1451E-03

     MINIMUM SEDIMENT THICKNESS AT I, J =  24  24 IS  -0.5192E-02

     MAXIMUM SEDIMENT THICKNESS AT I, J =  24  25 IS   0.7155E-03


##### TIME =    0.9600E+02  HOURS


MINIMUM VELOCITY AT   48   4 IS      0.0

MAXIMUM VELOCITY AT   58  28 IS      8.4

  MINIMUM ETA AT   77   4 IS     -1.1
```

Figure 5.7 (cont.) Output file *out.gbay.1* for Green Bay example.

```
MAXIMUM ETA AT     8  33 IS       1.7

 TOTAL SEDIMENT CONCENTRATION
   MINIMUM CONCENTRATION AT I, J =  59   1 IS   0.7899E-06
   MAXIMUM CONCENTRATION AT I, J =  35  10 IS   0.1717E-01

 MEDIUM SEDIMENT CONCENTRATION
   MINIMUM CONCENTRATION AT I, J =  59   1 IS   0.7898E-06
   MAXIMUM CONCENTRATION AT I, J =  35  10 IS   0.1716E-01

 COARSE SEDIMENT CONCENTRATION
   MINIMUM CONCENTRATION AT I, J =  12  33 IS  -0.1641E-04
   MAXIMUM CONCENTRATION AT I, J =  33  17 IS   0.1629E-04

   MINIMUM SEDIMENT THICKNESS AT I, J =  24  24 IS  -0.5192E-02

   MAXIMUM SEDIMENT THICKNESS AT I, J =  24  25 IS   0.7176E-03
```

Figure 5.7 (cont.) Output file *out.gbay.1* for Green Bay example.

F. REFERENCE MATERIALS FOR SECTION 3.0

1. Figures for Section 3.1

a.	Figure 3.1-1	Time Trend Graph of Decreasing PCB Concentrations in the Upper Hudson River Water Column
b.	Figure 3.1.3-1	Time Trend Graph of Total Lipid Based PCB Concentration in Large Mouth Bass

2. Figures for Section 3.2

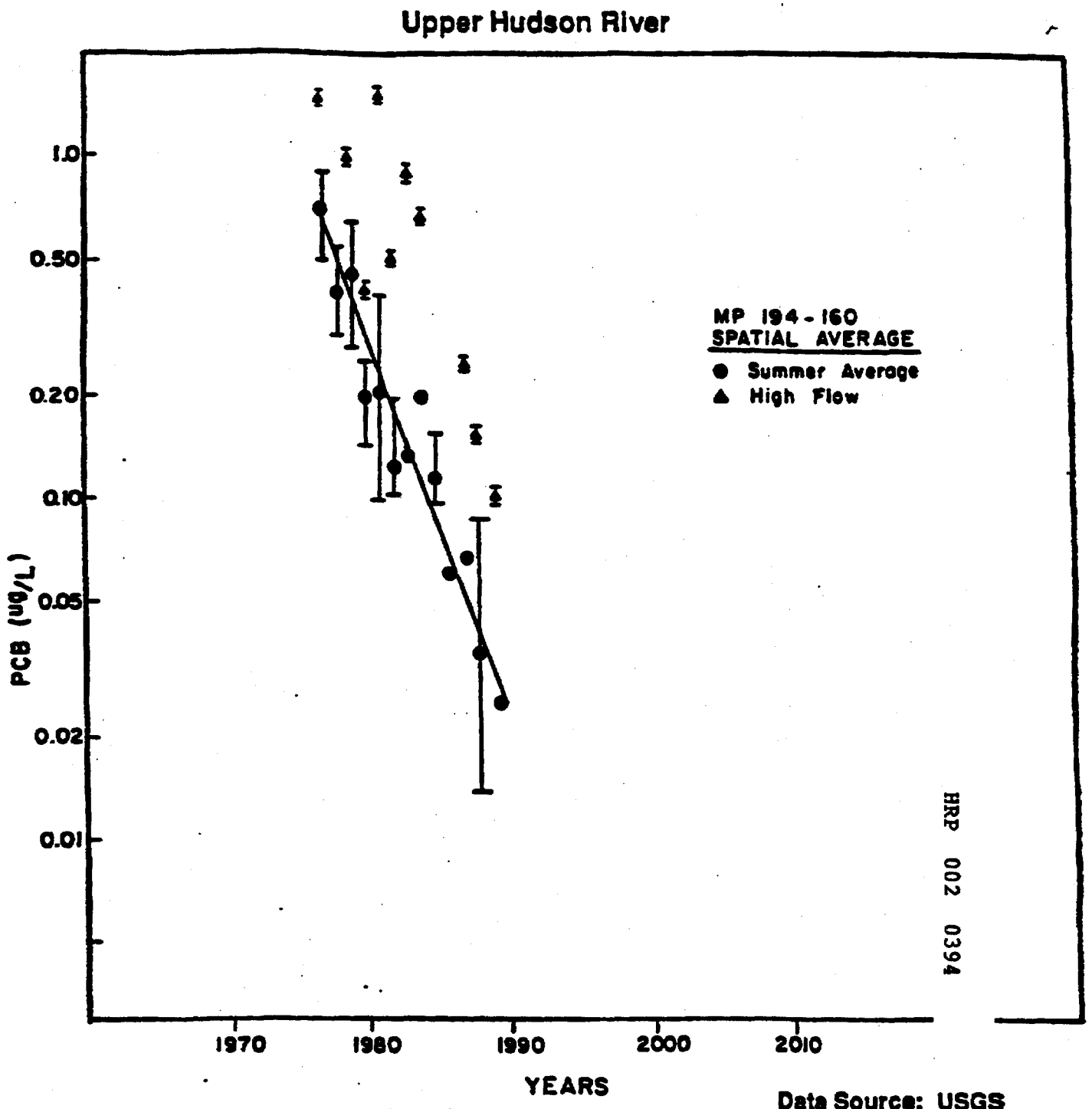
a.	Figure 3.2.1-1	Mean and Standard Deviation of Birth Weight: High Dose Group and Controls from Barsotti and Van Miller (1984), and Historical Measurements from Van Wagen and Catchpole (1956)
b.	Figure 3.2.2-1	Monte Carlo Simulation (10,000 iterations)

3. Tables for Section 3.2

a.	Table 3.2.2-1	Summaries of Studies Regarding Recreational Angler Fish Consumption
b.	Table 3.2.2-2	Species-specific Angler Effort
c.	Table 3.2.2-3	LADD Distributions (mg/kg/day) for Thirty Year Exposure
d.	Table 3.2.2-4	Average Percent Reductions in the Concentrations of PCBs in Fish Filets by Various Cooking Methods

PCB Concentration - Time Trend

Figure 3.1-1



Data Source: USGS

From Update on PCB Biodegredation and the Hudson River, Presented to Com. Axelrod, NYSDOH, by Dr. Daniel A. Abramowicz

- PCB Concentration Decreasing For All Flows
- Year-To-Year Variations But Trend Is 50% Decrease Every 3 Years

Figure 3.1-1

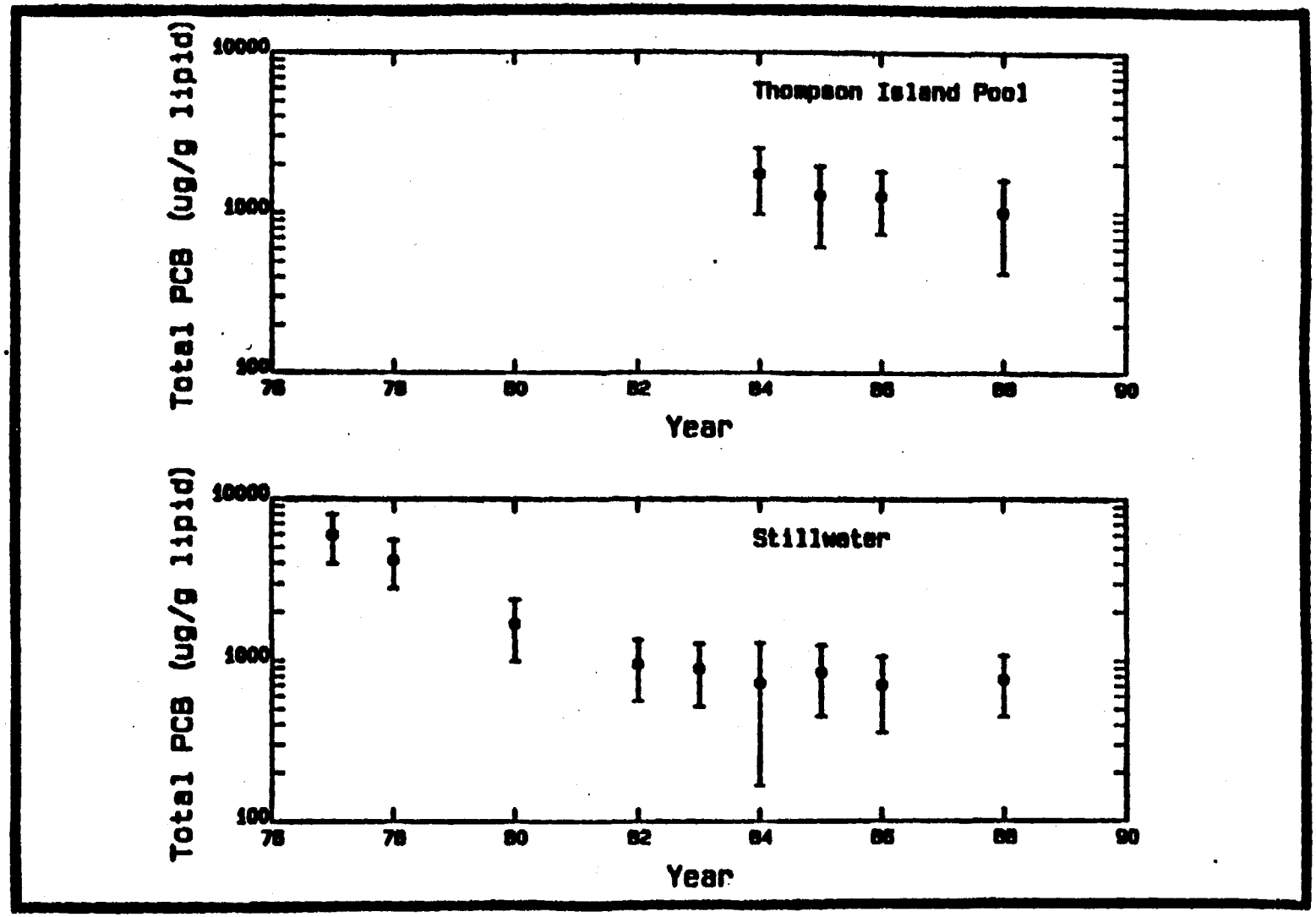
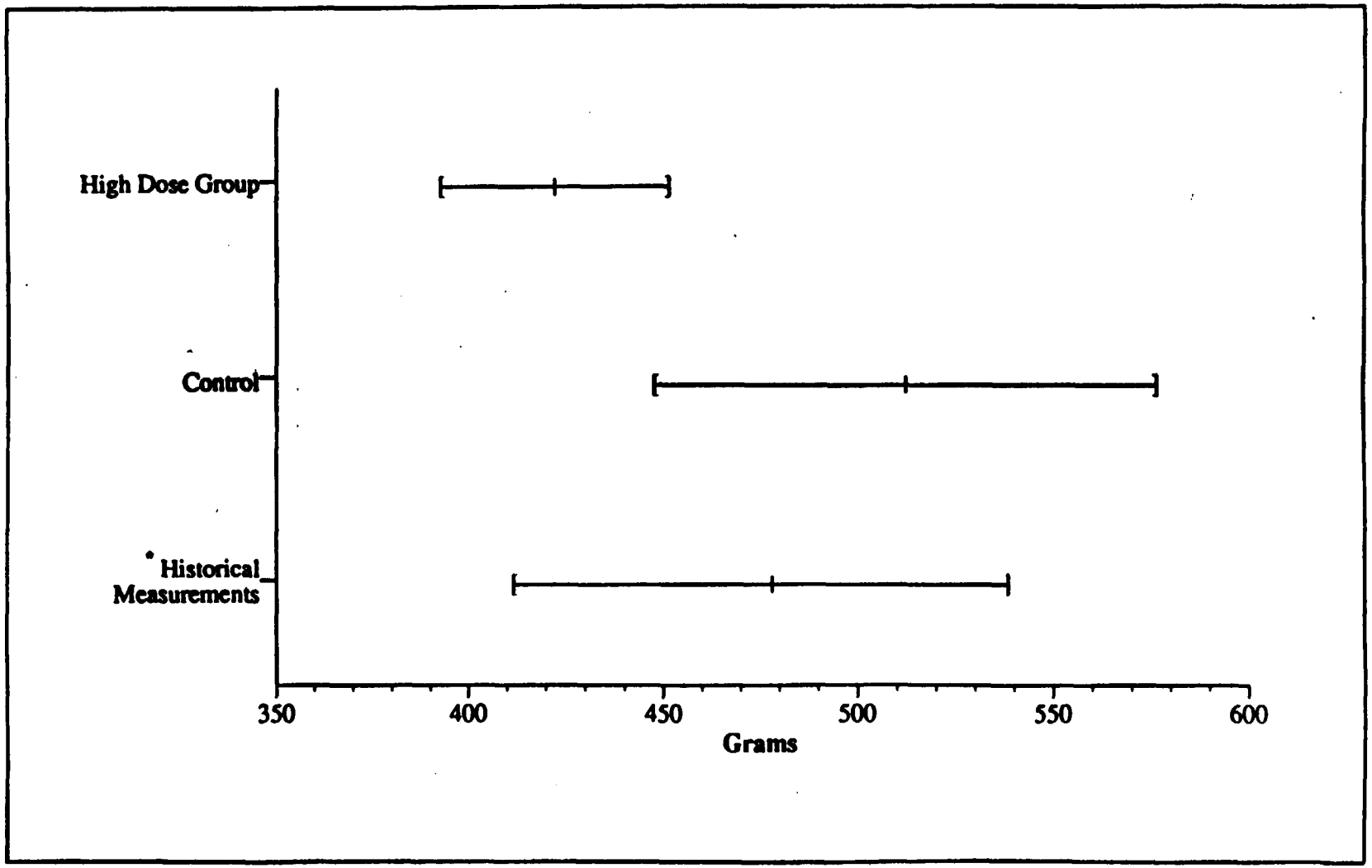


Figure 3.1.3-1

HRP 002 0395

Figure 3.1.3-1
Total Lipid Based PCB Concentration in Large Mouth Bass
in the Thompson Island Pool and at Stillwater (NYSDEC)

Figure 3.2.1-1. Mean and Standard Deviation of Birth Weight: High Dose Group and Controls from Barsotti and Van Miller (1984), and Histroical Measurements from Van Wagen and Catchpole (1956)



* Average of male and female mean and standard deviation

Figure 3.2.1-1

Monte Carlo Simulation (10,000 iterations)

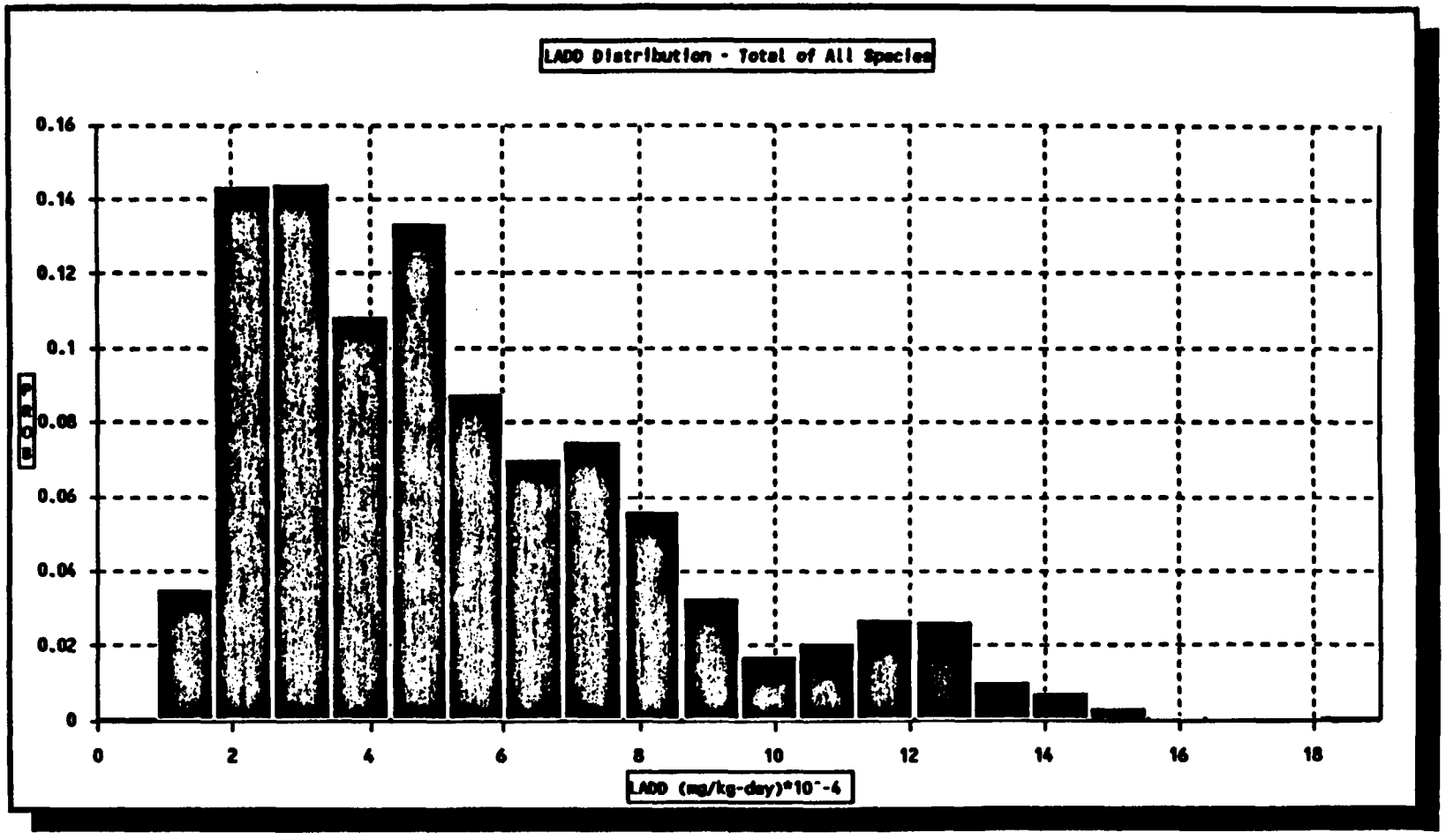


Figure 3.2.2-1

Table 3.2.2-1. Consumption of Fish by Recreational Anglers (g/day)

Study	Sport-Caught Fish		Commercial and Sport-Caught Fish	
	Median	Mean	Median	Mean
Puffer et al., 1981	37	--	--	--
Pierce et al. 1981	23	--	--	--
NYSDEC, 1990 (statewide)	--	--	--	28
NYSDEC, 1990 (Lake Ontario)	--	4.4	--	--
ChemRisk, 1991a	2.0[a] 0.99[b]	6.4[a] 3.7[b]	-- --	-- --
Wisconsin Division of Health, 1987	6.2	12.3	--	--
West et al., 1989	--	7	--	--
Honstead et al., 1971	--	7.7	--	--
Soldat, 1970	--	1.8	--	--
Turcotte, 1983	--	6.2 - 18.6	--	--

a. All waterbodies including lakes, ponds, rivers and streams.
b. Rivers and streams only.

Table 3.2.2-2. Species-specific Angler Effort for "Other Species" from the Upper Hudson River

Species	Angler Effort[a] (angler days)	Percent of Total Effort	Estimated Consumption (meals/year)
Yellow Perch	1,738,440	18.28	1.99
Walleye	2,388,360	25.11	2.73
Northern Pike	1,107,710	11.65	1.27
Brook Trout	1,389,500	14.61	1.59
Bullhead	1,150,110	12.09	1.32
Chain Pickerel	319,780	3.36	0.367
Sunfish	823,800	8.66	0.943
Other[b]	593,120	6.24	
American Eel			0.225
White Perch			0.225
Goldfish			0.225
Totals	9,510,820	100	10.88

a. From Table 6; NYSDEC, 1990.
b. "Other Species" category from Table 6; NYSDEC, 1990.

LADD Distributions (mg/kg-day) for Thirty Year Exposure

Simulation Statistics									
Date: 10/18/91 at 7:39									
Iterations: 10000									
Simulations: 1									
Worksheet: GE.XLS									
Output Range: LADDs									
Cell:	$PS1	$PS2	$PS3	$PS4	$PS5	$PS6	$PS7	$PS8	$PS9
Minimum=	7.7E-06	3.77E-07	1.32E-06	2.13E-06	1.72E-06	1.17E-06	3.63E-07	2.27E-06	7.91E-05
Maximum=	3.45E-05	0.000515	8.7E-05	0.000633	2.89E-05	8.38E-06	0.000108	0.000676	0.001836
Mean=	2.07E-05	7.91E-05	3.74E-05	0.000176	1.13E-05	4.09E-06	2.99E-05	0.000188	0.000546
Std Deviation=	6.8E-06	6.18E-05	2.05E-05	0.000147	7.03E-06	1.83E-06	2.05E-05	0.000157	0.000313
Variance=	4.63E-11	3.82E-09	4.19E-10	2.17E-08	4.95E-11	3.36E-12	4.22E-10	2.47E-08	9.78E-08
Skewness=	0.194227	2.345189	0.059762	1.038171	0.809954	0.583264	0.967329	1.038171	0.974943
Kurtosis=	3.759067	10.69646	2.822648	3.39878	2.83163	2.991466	3.360702	3.39878	3.35956
Percentile Values									
5Perc=	7.7E-06	1.7E-05	2.62E-06	1.05E-05	3.02E-06	1.17E-06	5.27E-06	1.12E-05	0.000179
10Perc=	7.7E-06	2.42E-05	3.08E-06	2.82E-05	3.3E-06	1.87E-06	7.7E-06	3.01E-05	0.000213
15Perc=	1.92E-05	3.04E-05	8.66E-06	3.38E-05	4.63E-06	2.53E-06	9.69E-06	3.6E-05	0.000241
20Perc=	1.92E-05	3.61E-05	2.38E-05	4.99E-05	4.92E-06	2.53E-06	1.16E-05	5.33E-05	0.000267
25Perc=	1.92E-05	4.13E-05	2.38E-05	5.79E-05	5.89E-06	3E-06	1.37E-05	6.17E-05	0.000295
30Perc=	1.92E-05	4.6E-05	3.09E-05	6.3E-05	6.25E-06	3.09E-06	1.61E-05	6.72E-05	0.000327
35Perc=	1.92E-05	4.99E-05	3.21E-05	7.6E-05	7E-06	3.09E-06	1.86E-05	8.11E-05	0.000365
40Perc=	1.99E-05	5.44E-05	3.22E-05	0.000109	7.75E-06	3.28E-06	2.08E-05	0.000116	0.000406
45Perc=	1.99E-05	5.84E-05	3.53E-05	0.000132	8.08E-06	3.36E-06	2.31E-05	0.000141	0.000443
50Perc=	2.02E-05	6.32E-05	3.69E-05	0.00014	9.3E-06	3.92E-06	2.49E-05	0.00015	0.000474
55Perc=	2.02E-05	6.87E-05	3.76E-05	0.000157	1.1E-05	3.92E-06	2.71E-05	0.000168	0.000506
60Perc=	2.02E-05	7.35E-05	4.34E-05	0.00017	1.14E-05	4.7E-06	3.02E-05	0.000182	0.000546
65Perc=	2.12E-05	7.97E-05	4.59E-05	0.000191	1.17E-05	4.94E-06	3.4E-05	0.000203	0.000603
70Perc=	2.12E-05	8.65E-05	4.79E-05	0.000245	1.57E-05	4.94E-06	3.76E-05	0.000262	0.000668
75Perc=	2.34E-05	9.53E-05	5.24E-05	0.000262	1.57E-05	5.27E-06	4.1E-05	0.00028	0.000726
80Perc=	2.34E-05	0.000108	5.26E-05	0.000285	1.83E-05	5.68E-06	4.55E-05	0.000304	0.000785
85Perc=	2.34E-05	0.000124	5.49E-05	0.000325	1.86E-05	5.68E-06	5.11E-05	0.000347	0.000862
90Perc=	3.45E-05	0.000147	6.02E-05	0.000396	2.29E-05	6.15E-06	6.25E-05	0.000422	0.001027
95Perc=	3.45E-05	0.000202	7.1E-05	0.000499	2.55E-05	8.38E-06	7.35E-05	0.000532	0.001217

Table 3.2.2-3

HRP 002 0400

Col.1=Am.Eel,2=B.Bullhead,3=Goldfish,4=Bass,5=Sunfish,6=W.Perch,7=Brown Trout,8=Other,9=TOTAL

Table 3.2.2-4. Average Percent Reductions in the Concentrations of PCBs in Fish Fillets by Various Cooking Methods

Cooking Method	Aroclor 1254 [a]	Aroclor 1254, 1242 [b]	Aroclor 1254, 1242 [c]	Total PCBs [d]	Aroclor 1254, 1242 [e]
Broiled	53 %	--	--	25 %	--
Roasted	34 %	--	--	20 %	11 - 16 %
Microwave	26 %	--	--	33 %	--
Pan Fried	--	65 %	28 %	25 %	--
Poached	--	--	--	27 %	2 - 8 %

a. Lake trout fillets from Lake Superior (Zabik et al., 1979).
b. White croaker fillets from Santa Monica Bay, Ca. (Puffer and Gossett, 1983).
c. White croaker fillets from Orange County, Ca. (Puffer and Gossett, 1983).
d. Carp fillets from Lake Superior (Zabik et al., 1982).
e. Chinook and Coho Salmon from Lake Michigan (Smith et al., 1973).

G. REFERENCE MATERIALS FOR SECTION 4.0

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Technical Notes

To provide a place within ASCE for publication of technical ideas that have not advanced to the point where they warrant publication as a technical paper in a journal, the publication of technical notes was authorized by the Board of Direction on October 16–18, 1967, under the following guidelines:

1. Four copies of an original manuscript are to be submitted to the Journals Department, ASCE, 345 East 47th Street, New York, NY, 10017-2398, along with a request by the author that it be considered as a technical note.
2. One of the copies will be sent to an appropriate technical division or council for review.
3. If the division or council approves the contribution for publication, it will be returned to Society Headquarters with appropriate comments.
4. The journal staff will prepare the material for use in the earliest possible issue of the journal, after proper coordination with the author.
5. Each technical note is not to exceed 2,500 word-equivalents. As an approximation, each full manuscript page of text, tables, or figures is the equivalent of 900 words.
6. The technical notes are grouped in a special section of the journal.
7. A 175-word information retrieval abstract and key words are necessary for technical notes.
8. The final date on which a discussion should reach the Society is given as a footnote with each technical note.
9. Technical notes will be included in *Transactions*.
10. Technical notes will be included in ASCE's annual and cumulative subject and author indexes.

The manuscripts for technical notes must meet the following requirements:

1. Titles must have a length not exceeding 70 characters and spaces.
2. The manuscript should be typed double spaced on one side of 220 mm by 280 mm paper. Four copies of all figures and tables must be included.
3. The author's full name, Society membership grade (if applicable), and a footnote stating present employment must appear on the first page of the note. Authors need not be Society members.
4. All mathematics must be typewritten and special symbols must be properly identified. The letter symbols used must be defined where they first appear, in figures, tables, or text.
5. Standard definitions and symbols must be used. Reference must be made to the lists published by the American National Standards Institute and to the *ASCE Authors' Guide to Journals, Books, and Reference Publications*.
6. Figures must be drawn in black ink on one side of 220 mm by 280 mm paper. Because figures will be reproduced with a width of between 76 mm to 110 mm, the lettering must be large enough to be legible at this width. Photographs must be submitted as glossy prints. Explanations and descriptions must be made within the text for each figure.
7. Tables must be typed on one side of 220 mm by 280 mm paper. An explanation of each table must appear in the text.
8. References cited in text must be typed double spaced at the end of the technical note in alphabetical order in an Appendix. References.
9. Each author may use the International System of Units (SI), and units acceptable in SI or other units. When SI units are used, other units are required. When other units are used, the SI units shall be given in parentheses, in a supplementary or dual unit table, or in an appendix.

Scour-Depth Prediction under Armoring Conditions*

By Deva K. Borah,[1] Member, ASCE

Introduction

Accurate prediction of scour depth may assist in engineering planning and design involving water flow over an alluvial bed. Procedures for computing scour depths in long, contracted river reaches and near spur dikes, cofferdams, and drop structures are available in the literature (Vanoni 1975, Petersen 1986). These procedures are based on empirical relations predicting equilibrium sediment discharge under steady-state flow conditions. The procedures are not simple and proper applications require clear understanding of the underlying assumptions and the fluvial processes.

Bed scour in alluvial streams can also be predicted by using a simulation model. The models are based on mass conservation and equilibrium sediment discharge relations and often require calibration with measured data. Learning and using a simulation model is a time-consuming task. Search for simpler procedure of bed scour computation must continue so that practicing engineers can use such a procedure with minimum effort.

In this note, a simple procedure is proposed to compute scour depth on an alluvial bed having graded materials under certain flow conditions that can produce an armor layer at the bed surface. The procedure is validated with laboratory and field conditions, namely Little and Mayer's (1972) flume and downstream of the Clavius Point Dam in the Missouri River (Karim and Kennedy 1982), respectively. The proposed procedure will be useful in computing scour depths downstream from hydraulic structures and in new unlined canals, and for selecting materials for canal beds.

Bed Scour and Formation of Armor Layer

Bed scour on an alluvial bed is the result of imbalance between the sediment transport capacity of the flow and the amount of sediment coming along with the flow. If the sediment transport capacity is higher than the incoming sediment rate, bed scour will take place until these two become equal.

In the presence of graded materials, the bed scour may be restricted by formation of an armor layer. Such a layer is formed by the continuous action of water flow having a transport capacity continually higher than the sediment supply. In this case, bed scour continues due to the entrainment of transportable particles already exposed or getting exposed at the bed surface.

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Note. Discussion open until March 1, 1989. To extend the closing date one month, a written request must be filed with the ASCE Manager of Journals. The manuscript for this paper was submitted for review and possible publication on October 25, 1988. This paper is part of the *Journal of Hydraulic Engineering*, Vol. 115, No. 10, October, 1989. ©ASCE, ISSN 0733-9429/89/0010-1421/$1.00 + $.15 per page. Paper No. 23936.

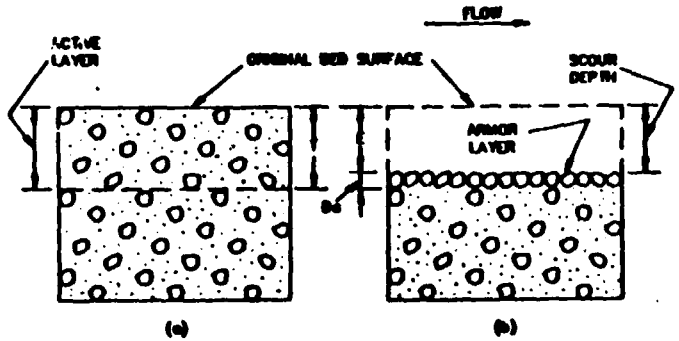

FIG. 1. Formation of Armor Layer: (a) Well-Mixed Original Bed Material; and (b) Armor Layer with Underlying Bed Material

The larger and heavier particles, which are nontransportable under the flow conditions, remain on the bed and gradually occupy the entire bed surface. As soon as this is the case, bed scour stops. This layer of coarse particles is called the armor layer. Such a layer will protect the underlying finer particles until it is destroyed by higher flow conditions. Formation of an armor layer is schematically illustrated in Fig. 1, where the well-mixed original bed material is shown in Fig. 1(*a*) and the armor layer with the underlying bed material is shown in Fig. 1(*b*).

Scour-Depth Calculation Procedure

Under the armoring condition discussed in the preceding section, the scour depth can be calculated from an upper bed layer known as the active layer, which has enough nontransportable particles to cover the entire bed surface by one layer, after the finer particles present within get exposed at the bed surface and are washed away (Fig. 1). [The concept of the active layer was earlier discussed by Thomas and Prasuhn (1977).] Borah et al. (1982) introduced an expression for the active-layer thickness, which may be expressed as

$$T = \frac{D_s}{(1 - e)P_s} \quad \ldots\ldots (1)$$

in which T = the thickness of the active layer; D_s = the smallest armor size or size of the smallest nontransportable particle present in the bed material; e = the porosity of the bed material; and P_s = the fraction of all the armor sizes present in the bed material. In deriving this equation, it is assumed that the bed layer is well mixed and all the particles have the same specific gravity.

The scour depth is computed as (Fig. 1)

$$E = T - D_s \quad \ldots\ldots (2)$$

in which E = the scour depth. In using Eq. 2 to estimate the scour depth, the hiding finer particles within the active layer is ignored, which is conservative and keeps the procedure simple. In reality, some of the finer particles will be trapped within the armor particles and, as a result, the armor layer will not be as horizontal as shown in Fig. 1(*b*), and the scour depth will be less than that predicted by Eq. 2. In order to account for this, the term D_s in Eq. 2 may be multiplied by a hiding factor, similar to Einstein (1950), having values greater than 1.

The size of the smallest armor particle D_s is approximately equal to a critical particle size given by the Shields curve for initiation of motion. All the sizes bigger than this are nontransportable and will contribute to the armor layer. Alonso et al. (1981) used certain relations to represent the Shields curve (Vanoni 1975) and compute critical shear stress. These are: $T_* = 0.08/R_*^{0.4}$ for $R_* \leq 10$; $T_* = 0.022R_*^{0.16}$ for $10 < R_* \leq 500$; and $T_* = 0.06$ for $R_* > 500$; T_* = the dimensionless shear stress and R_* = the boundary Reynolds Number. From these relations, the following expressions for smallest armor size are derived:

$$D_s = 68\left(\frac{dS_f}{S - 1}\right)^{1.67}\left(\frac{U_*}{\nu}\right)^{0.67}, \quad \text{for } \frac{U_*D_{50}}{\nu} \leq 10 \quad \ldots\ldots (3a)$$

$$D_s = 27\left(\frac{dS_f}{S - 1}\right)^{0.86}\left(\frac{\nu}{U_*}\right)^{0.14}, \quad \text{for } 10 < \frac{U_*D_{50}}{\nu} \leq 500 \quad \ldots\ldots (3b)$$

$$D_s = 17\left(\frac{dS_f}{S - 1}\right), \quad \text{for } \frac{U_*D_{50}}{\nu} > 500 \quad \ldots\ldots (3c)$$

in which U_* = the shear velocity equal to $(gdS_f)^{0.5}$; g = the acceleration due to gravity; d = the flow depth; S_f = the energy slope; D_{50} = the median particle size; ν = the kinematic viscosity of water; and S = the specific gravity of sediment.

The bed porosity e, the fraction of the armor particles P_s, and the median particle size D_{50} are estimated from analyses of bed material samples. The porosity may be estimated by using the following empirical relation given by Komura and Simmons (1967):

$$e = 0.245 + \frac{0.0864}{(0.1D_{50})^{0.21}} \quad \ldots\ldots (4)$$

In this equation D_{50} is in millimeters.

Validation with Laboratory Data

The procedure is validated with runs 3-4 and 6-1 of Little and Mayer's (1972) experimental data. The experiments were conducted in a recirculating laboratory flume of 0.6 m wide with different mixtures of crushed quartz spread uniformly over a length of 12.19 m with a depth of 101.67 mm. The particle sizes ranged from 0.125 to 8 mm in both the runs. Median diameter D_{50} for both the mixtures was 1 mm. Clear water was continuously discharged into the flume from upstream and the eroded sediment was trapped at the downstream end for continuous monitoring and analysis. Run 3-4 was continued for more than three days and run 6-1 for approximately six days. Both the runs resulted in development of complete armor layers.

Table 1 shows the hydraulic parameters in the experimental runs and the

TABLE 1. Comparisons of Predicted and Observed Scour Depths

Data source (1)	Flow rate (m^3/s) (2)	Flow depth (m) (3)	Energy slope (4)	Smallest armor size (mm) (5)	Fraction of armor material (6)	Bed porosity (7)	Predicted scour depth (mm) (8)	Observed scour depth (mm) (9)
Little and Mayer (1972); run 3-1	0.016	0.066	0.0049	1.70	0.70	0.305	0.56	5.62
Little and Mayer (1972); run 6-1	0.013	0.056	0.0030	1.61	0.34	0.305	0.17	4.62
Gavins Point Dam (Missouri River	1,303	2.545	0.000220	6.48	0.0891	0.426	1,193	1,585

predicted and observed scour depths. In computing the smallest armor sizes for both the runs, kinematic viscosity of water is taken as 9.84×10^{-7} m^2/s. The observed scour depths are calculated from the cumulative dry mass of eroded materials, 66.958 kg for run 3-1 and 55.004 kg for run 6-1, and taking a value of 1,000 kg/m^3 for the density of water and a value of 2.65 for the specific gravity of sediment. Bed porosities are calculated using Eq. 4. The fractions of armor sizes for the two runs are estimated from particle size distributions reported by the investigators. As can be seen in Table 1, the predicted values are conservative because the hiding effect is not considered.

Validation with Field Data

Karim and Kennedy (1982) and Holly and Karim (1986) applied their simulation model IALLUVIAL to the 195-mi (314-km) reach of the Missouri River between Gavins Point Dam and Omaha to predict bed elevation changes during a 20-year period. The river reach suffered severe erosion during this period due to the elimination of upstream sediment supply by the trapping effect of Gavins Point Dam and other upstream multipurpose developments. The investigators used certain flow values and hydraulic and sediment parameters based on U.S. Army Corps of Engineers' records. These flow values and parameters are used to validate the procedure, presented in this technical note, by predicting scour depth at the downstream of Gavins Point Dam.

Karim and Kennedy (1982) used two sets of average discharges in their simulation. The higher average discharge of 1,303 m^3/s and a width of 549 m are used in the present calculations. Table 1 shows the hydraulic parameters and the comparison of predicted and observed scour depths. The flow depth is calculated by solving the Manning's equation with a Manning's n value of 0.030. The median diameter of bed material D_m and the kinematic viscosity of water ν are taken as 0.297 mm and 1.567×10^{-6} m^2/s, respectively. Bed porosity is estimated using Eq. 4.

As seen in Table 1, the predicted scour depth of 1,193 mm (3.9 ft) is lower than the observed value of 1,585 mm (5.2 ft). This is expected because of the use of an average flow throughout the 20-year period. Use of the highest or a representative high flow occurred during the 20-year period would improve the result. Holly and Karim (1986) also reported lower prediction, approximately 3.6 ft (1,097 mm), from their simulation results using IALLUVIAL.

Conclusions

A simple procedure is presented for calculating the scour depth on an alluvial bed under armoring conditions. It was used to predict scour depths in Little and Mayer's flume and downstream from Gavins Point Dam. The procedure is applicable to only graded bed materials with a size distribution having adequate quantity of critical and bigger size particles to form an armor layer. Calculation of scour depth requires flow depth, bed slope, bed porosity, particle size distribution of the bed material, and few commonly available constants. The procedure can serve as a useful tool for field and design engineers in making conservative estimates of scour depths downstream of hydraulic structures and in new unlined canals as well as in selecting materials for canal beds. The active-layer thickness (Eq. 1) provides a simple theoretical base for future work in further verifying the proposed procedure with reliable laboratory and field data and improving it by incorporating the hiding effect.

Acknowledgments

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Equipment Choices for Dredging Contaminated Sediments

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Michael R. Palermo is a research civil engineer in the experimental laboratory at the U.S. Army Engineer Waterways Experiment Station in Vicksburg, MS.

This article describes the selection and field evaluation of dredging equipment and techniques for removal of highly contaminated sediments from the upper estuary of the Acushnet River, a portion of the New Bedford Harbor Superfund Project. Site conditions as related to dredge selection and operation, factors considered in selection of equipment, and various dredge types considered for use are described. Each of the dredge types is ranked according to the following criteria: availability, safety, potential for sediment resuspension, maneuverability, cleanup precision, cost and production, flexibility, required water depth for operation, ability to access the site, and compatibility with disposal options. A field pilot study comparing three dredge types indicated that dredging could be conducted at the site without a significant increase in the contaminant release from the upper estuary to the lower harbor.

Bottom sediments in many of the nation's harbors and waterways have become polluted because of municipal and industrial discharges and non-point sources. Sediments act as a sink for many contaminants, and sediment removal is being considered in some of the most seriously contaminated areas. For example, cleanup of contaminated sediments is proposed at several Superfund sites; in most cases, the cleanup alternatives under consideration involve dredging for sediment removal prior to treatment or disposal. The selection of appropriate dredging equipment is crucial for successful cleanup. One such site is the Acushnet River Estuary and harbor adjacent to New Bedford, Massachusetts, where the sediments are highly contaminated with polychlorinated biphenyls (PCBs) and heavy metals.

In August 1984, the United States Environmental Protection Agency (EPA) reported on the Feasibility Study of Remedial Action Alternatives for the upper Acushnet River Estuary above the Coggeshall Street Bridge in New Bedford, Massachusetts (NUS 1984). EPA received extensive comments on the proposed remedial action alternatives from other federal, state, and local officials, potentially responsible parties, and individuals. Responding to these comments, EPA chose to conduct additional studies to better define available cleanup methods. Because dredging was associated with all of the removal alternatives, EPA requested the United

States Army Corps of Engineers (USACE) to conduct an Engineering Feasibility Study (EFS) of dredging and disposal alternatives. A major emphasis of the EFS was placed on evaluating the potential for contaminant releases from both dredging and disposal operations.

This article describes the selection and field pilot evaluation of appropriate dredging equipment and techniques for removal of contaminated sediments from the upper estuary of New Bedford Harbor.

This article describes the selection and field pilot evaluation of appropriate dredging equipment and techniques for removal of contaminated sediments from the upper estuary of New Bedford Harbor. The USACE has conducted extensive research on dredging equipment and methods to reduce sediment resuspension and contaminant release during dredging operations (Barnard, 1978; Raymond, 1984; Hayes, 1986; McLellan et al., 1989). The equipment selection process described in this article is based on a review of these techniques and equipment, along with their applicability to the dredging conditions in the Acushnet River Estuary near New Bedford. Additional detailed information is available on the equipment selection process (Palermo and Pankow, 1988) and the field pilot evaluation (New England Division 1990) for the New Bedford project.

SITE DESCRIPTION

The area of concern, called the upper estuary, is north of New Bedford Harbor, Massachusetts. It extends from Coggeshall Street Bridge to Saw Mill Dam, a distance of less than two miles (See **Figure 1**). This section of the river averages about .2 miles wide and is approximately .4 miles wide at its widest part. The total surface area of the upper estuary at mean tide level is approximately 187 acres. This portion of the Acushnet River is relatively shallow, with a channel depth progressing from 15 to 7 feet and overbank areas of less than one foot to three feet mean low water. The mean tide range is 3.7 feet, and expanses of mud flats are exposed at low tide. Freshwater inflow at the Saw Mill Dam was measured between 1972 and 1974 by the USGS and ranged from a monthly maximum of 26 cfs to a monthly minimum of .55 cfs. Some ungaged storm sewers also drain into the upper harbor. The average water depths at mean low tide are indicated in Figure 1.

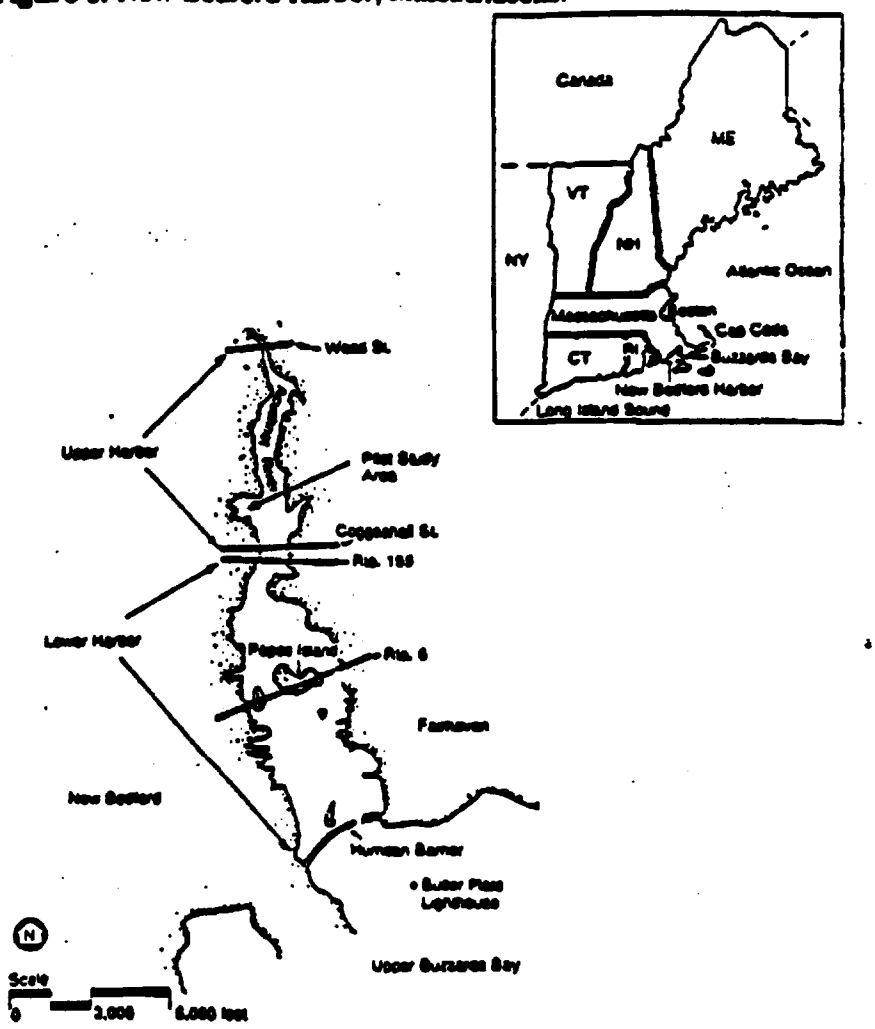
The salinities in the upper harbor are typically in the range of 26 to 30 ppt, with less than 1 ppt difference from top to bottom except after heavy rains, when the surface salinity can be much less. Current velocities above the Coggeshall Street Bridge average roughly .3 fps, with a maximum of .85 fps; ebb currents are generally stronger than flood currents.

Another feature of the upper Acushnet River is the restriction at the Coggeshall Street Bridge. The bridge is fixed with an eight-foot-high vertical clearance and a sixty-two-foot-wide horizontal clearance at mean low water. This limits the size of equipment that can be floated into the upper river.

Sediment Properties

The sediments to be removed and treated as contaminated are generally classified as silts and clays with a significant fraction of fine sand. There is little physical difference between the surficial contaminated layers and the underlying layers to a depth of approximately four feet. Field

Figure 1. New Bedford Harbor, Massachusetts.



inspections have indicated that there is little debris within the sediment mass, with the exception of visible cobbles and rock fragments and other debris near the shoreline. Contaminant concentrations in sediment decrease with depth and are generally restricted to the upper one foot of sediment. PCB sediment concentrations vary within the upper estuary from less than 50 ppm to as high as 100,000 ppm, but sampling conducted for the EFS indicated that the median PCB concentration of samples taken from the upper one foot was approximately 100 ppm (Averett et al., 1989).

Dredging Volumes

The contaminated material (the upper one foot of sediment for most of the area) plus approximately one foot of underlying clean material is being considered for removal. This two-foot removal thickness over the entire cleanup area corresponds to approximately 600,000 cubic yards of

in situ material. All this material would be considered contaminated, requiring appropriate treatment and disposal.

Dredging Requirements

The most important dredging requirement associated with this project is that the sediment must be removed and transported to the disposal site with a minimum of sediment resuspension and associated contaminant release. The dredging process must also be conducted at a reasonable speed so that the time period during which resuspension could occur would be held to the minimum. When contaminated sediments are disturbed, as in dredging operations, contaminants may be transferred to the water column through resuspension of the sediment solids, dispersal of interstitial water, or desorption from the resuspended solids. An investigation of PCB-laden sediments (Fulk et al., 1975) showed that almost all the contaminants transferred to the water column are due to the resuspension of solids. The release of contaminants can therefore be decreased by reducing the resuspension of sediment during dredging and disposal operations.

Because all contaminated dredged material must be placed in disposal sites with costly treatment and controls, precision of the dredging process is critical.

Because all contaminated dredged material must be placed in disposal sites with costly treatment and controls, precision of the dredging process is critical. Contaminated sediments must be removed to the extent that little contaminated material is left behind. At the same time, removal of cleaner underlying material (overdredging) should be avoided to the extent possible because it would require the same costly treatment.

DREDGING EQUIPMENT AND TECHNIQUES CONSIDERED

All major dredging equipment types were initially considered for this evaluation. This included equipment using the three basic dredging processes (i.e., mechanical, hydraulic, and pneumatic), the equipment types normally employed for conventional dredging operations, and equipment considered to be special-purpose. The dredges included in the evaluation are operational and proven dredges. A brief description of the basic equipment types considered is given in the following paragraphs. Operational characteristics of hydraulic/pneumatic dredges considered are summarized in **Table 1**. Resuspension characteristics of conventional and some specialty dredges considered are summarized in **Tables 2 and 3**. More detailed descriptions of operating characteristics of the dredge types, especially as related to sediment resuspension, are given by Raymond (1984).

Mechanical Dredging

Excavation of sediment using such devices as clamshell dredges, dipper dredges, draglines, grab buckets, and in some instances, front-end loaders and backhoes is commonly referred to as mechanical dredging. Mechanical dredging normally yields dredged material high in solids content, and removal from the dredging site involves the use of barges and tugs to transport the material to the disposal site. Mechanical dredges can be operated from the shore if the area to be dredged is near the water's

Table 1. Operational Characteristics of Hydraulic/Pneumatic Dredges.*

Dredge	Percent Solids by Weight	Range of Production Rates cu yd/hr	Vertical Dredging Accuracy +ft	Horizontal Dredging Accuracy +ft
Bucket	Up to 100	30-600	2	1
Suction	10-15	25-5,000	1	2-3
Dustpan	10-20	25-5,000	0.5	2-3
Cutterhead	10-20	25-5,000	1	2-3
Hopper	10-20	500-2,000	2	10
Horizontal Auger	10-40	60-150	0.5	0.5
PNEUMA	Up to 80	60-300	1	1
Oozer	Up to 80	450-650	1	2-3
Clean-up	30-40	500-2,000	1	2-3
Refresher	30-40	200-1,300	1	2-3

*Adapted from Phillips and Malek (1984).

Table 2. Resuspension Characteristics of Conventional Dredges.*

Dredge Type	Downcurrent Distance Suspended Solids Concentration, mg/l** Within 100 ft	Within 200 ft	Within 400 ft
Cutterhead	25-250	20-200	10-150
Hopper			
With overflow	250-700	250-700	250-700
Without overflow	25-200	25-200	25-200
Clamshell			
Open bucket	150-900	100-600	75-350
Enclosed bucket	50-300	40-210	25-100

*From Hayes (1986).
**Suspended solids concentrations were adjusted for background concentrations.

edge, or operated from a barge that is anchored in position. Some mechanical dredges, such as a clamshell bucket dredge, leave an irregular, cratered bottom and can generate a large amount of turbidity throughout the water column (Barnard, 1978; Raymond, 1984) as compared with other

Table 3. Resuspension Characteristics of Specialty Dredges.*

Name of Dredge	*Reported Suspended Sediment Concentrations***
PNEUMA pump	48 mg/l, 3 ft above bottom 4 mg/l, 23 ft above bottom (16 ft in front of pump)
Clean-up system	1.1 to 7.0 mg/l 10 ft above suction 1.7 to 3.5 mg/l at surface
Oozer pump	Background level (6mg/l), 10 ft from head
Refresher system	4 to 23 mg/l, 10 ft from head

* From Hayes (1986) and Herbich and Brahme (in preparation).
** Suspended solids concentrations were adjusted for background concentrations.

dredge types. The turbidity is a result of sediment resuspension as the bucket makes impact on and is pulled off the bottom, water and sediment spillage from the bucket as it is pulled up through the water column and breaks the water surface, and spillage of material as it is loaded into the scow.

Turbidity can be reduced with the use of a watertight bucket and with carefully controlled operation. The watertight bucket has interlocking jaws that seal when the bucket is closed; the top is also covered so that the dredged material cannot escape once the bucket is closed. A comparison of a 1-m^3 bucket with a watertight clamshell bucket indicates that the watertight bucket generates 30 percent to 70 percent less turbidity in the water column than the typical open clamshell bucket (Barnard, 1978). Turbidity levels generally decrease rapidly with distance from the dredge, and the major cause of resuspension appears to be the impact of the bucket hitting the bottom.

Each step of the mechanical dredging operation from initial dredging to final placement is subject to spillage and splashing, which allow sediment to return to the water. There are additional causes of sediment resuspension when scows are used to transport sediment. Propeller wash from tug boats can stir up sediment, and, in extremely shallow areas, a loaded scow may touch bottom, causing considerable sediment resuspension.

Hydraulic Dredging

Hydraulic dredges use a centrifugal water pump to create a vacuum at the dredgehead; atmospheric pressure acts to force water and sediments through the suction pipe. The dredged materials are usually hydraulica[illegible]

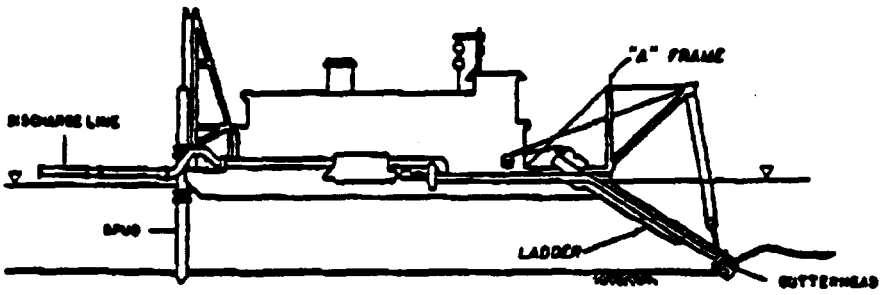
pumped via a pipeline to the disposal site, which can be a confined disposal facility (CDF) or an open-water area. The material can also be pumped into barges for transportation to the disposal site. Examples of hydraulic dredges are plain suction, cutter-suction, dustpan, sidecast, and trailing hopper dredges. The simplest form of hydraulic dredge, the plain suction dredge, is used for excavating free-flowing sandy material. The cutterhead, also called the cutter-suction dredge (Figure 2) is the most commonly used dredge in the United States. In this dredge, the suction head is fitted with a rotating basket that can have blades or teeth, depending on the type of material to be removed. As the cutter rotates, it mechanically loosens the bottom sediment and moves it toward the flow field around the dredge suction.

Most turbidity associated with a cutterhead dredging operation is in the immediate vicinity of the rotating cutterhead. The amount of resuspended sediment decreases rapidly from the cutter to the water surface. Depending on the sediment type, the operational conditions, and the current velocity, turbidity levels also decrease rapidly with distance from the cutter. Turbidity can be reduced by controlling the cutterhead rotation speed, the swing speed of the ladder, and the procedures for operation of the cutterhead. Undercutting, cutting into the swing of the cutterhead, produces less resuspended sediment than overcutting, cutting away from the swing direction of the cutterhead (Koba and Shiba, 1982). The avoidance of large sets and very thick cuts and the use of close concentric swings to reduce the occurrence of windrows between cuts are other operational procedures that can be employed to reduce the resuspension of sediment (Raymond, 1984). Attempts to reduce turbidity in the area of the cutterhead have resulted in the use of hoods, shields, or covers of various types. These shields seem to increase velocities and turbulence near the bottom, causing increased entrainment, and help prevent turbid water from reaching the surface (Herbich and Brahme, 1991).

Pneumatic Dredging

Pneumatic dredge systems use compressed air to pump slurry through a pipeline (Richardson et al., 1982). The principle under which the pump operates is the pressure differential between the pressure in the chamber

Figure 2. Hydraulic pipeline cutterhead dredge.



and the hydrostatic pressure of water outside the pump. The chamber is lowered into position and the inside air is released to the atmosphere, producing atmospheric pressure in the chamber. The pressure difference between the inside and the outside of the chamber forces water and sediment into the chamber. The entrance valve is then closed, and air is pumped into the chamber, increasing pressure and forcing the slurry out the discharge valve.

Pneumatic dredging systems have been developed in Italy and Japan. The Italian device called the PNEUMA pump consists of three chambers, each connected to a common discharge line above the pressure vessels. The chambers are operated so that the filling and emptying cycles are out of phase but overlap enough to minimize discharge surging (Richardson et al., 1982).

Although there are three major types of dredges, many dredges combine more than one operational principle to produce a dredge suited to specific conditions.

Turbidity levels around the PNEUMA dredge are extremely low, and high concentrations of low-viscosity materials can be dredged. The PNEUMA dredge is mounted on a barge with a crane to raise and lower the pump body. The pump is placed in position and pulled through the sediment as necessary. Different attachments can be used to suit the dredging method and sediment being removed.

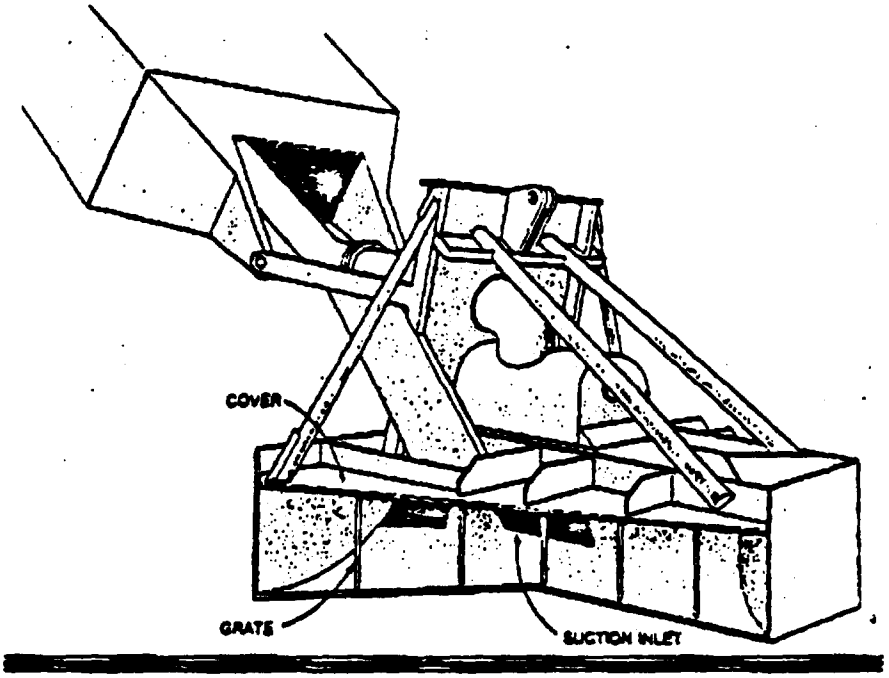
The Japanese have developed a pneumatic dredge called the Oozer (Koba et al., 1975), which is ladder-mounted, similar to a hydraulic cutter-suction dredge. Recent versions of the PNEUMA dredge also are ladder-mounted.

Specialty Dredges

Although there are three major types of dredges, many dredges combine more than one operational principle to produce a dredge suited to specific conditions. Many of the features incorporated in specialty dredges are attempts to reduce sediment resuspension at the dredgehead. Examples of specialty dredges are the cleanup (Sato, 1976) and refresher dredges, which were developed in Japan, and the matchbox (d'Angremond et al., 1984), which was developed in the Netherlands. The matchbox dredge has been used in Holland for dredging contaminated sediment. It is designed to dredge fine-grained sediments at near in situ density and keep resuspension to a minimum. The matchbox is a plain suction dredgehead enclosed in a housing that resembles a matchbox. The housing collects escaping gas bubbles, and valved openings on each side of the suction head allow the leeward opening on each swing to be closed to avoid an influx of water (Figure 3).

A comparison test of sediment resuspension of a matchbox suction head and a cutterhead was conducted by the USACE in Calumet Harbor, Illinois, on Lake Michigan. As the matchbox head is new to this country, and the comparison was short-term, some of the recommended instrumentation was omitted. This made it difficult for an inexperienced operator to determine the location of the head, a factor that affected the operation of the dredge and the resuspension of sediment. In general, the report concluded that the matchbox is capable of removing sediment with very

Figure 3. Matchbox head.



little resuspension; it also concluded, however, that the cutterhead was equally capable when operated properly (Hayes, 1988).

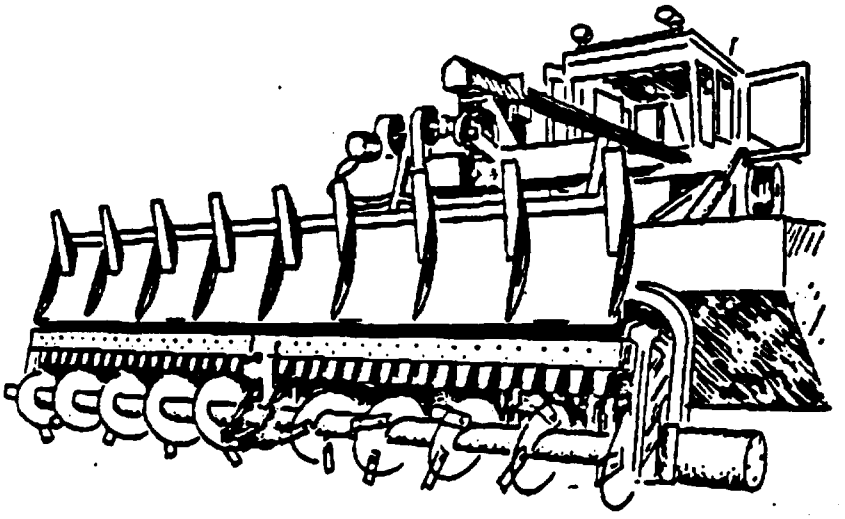
Horizontal Auger Dredge

Small portable horizontal auger dredges are equipped with cutter knives or spiral augers that cut and move the material toward the suction. The mudcat (Figure 4) and VMI dredges are examples of this type of equipment. Designed to remove fine-grained sediments, small portable dredges can float in water as shallow as twenty-one inches (.5 m). Movement of the dredge through the water is controlled by winching along a cable anchored on the shore. In some designs the cutterhead is surrounded by a mud shield that is effective in minimizing turbidity by entrapping suspended sediment. Manufacturers claim that discharges with as much as 60 percent solids have been obtained. This cutter design can remove a layer of material eight feet (2.4 m) wide, leaving the bottom flat and free of windrows and ridges that are typical of cutterhead and clamshell dredge operations (Barnard, 1978).

SELECTION OF DREDGING EQUIPMENT

Selection of equipment for the pilot study was based on a consensus rating by USACE personnel knowledgeable in the operation and capabilities of the various dredge types. Nine dredge types or groupings were identified for rating, as listed in Table 4. The clamshell dredge was the only

Figure 4. Horizontal cutterhead of the Mudcat dredge showing cutter knives and spiral auger (courtesy of Mudcat Division, National Car Rental Systems, Inc.).



mechanical dredge rated. All others identified for rating were hydraulic or pneumatic. Small cutterhead and shrouded cutterhead dredges were rated as separate categories. Dustpan and matchbox dredgeheads, considered adaptations of conventional hydraulic dredges, were rated separately. The horizontal auger, although considered a portable hydraulic dredge, was rated separately because of its unique auger dredgehead and cable anchoring system. The Dubuque, a twelve-inch (30 cm) hydraulic dredge

Table 4. Equipment Ratings.

		Rating by			
Dredge Type	*Total Rating*	*Availability*	*Safety*	*Resuspension*	*Maneuverability*
Horizontal Auger	32	3	3	3	3
Small Cutter	29	2	3	2	2
Matchbox	29	1	3	3	2
Shrouded cutter	27	1	3	2	2
Dustpan	26	1	3	2	2
Japanese	23	1	3	3	2
Dubuque	21	1	3	2	1
PNEUMA	21	1	3	3	2
Clamshell	20	3	2	1	2

used in demonstrations at Indiana Harbor, was rated separately because it was owned by the USACE and had fabricated cutter and matchbox heads. The Oozer, Refresher, Clean-up, and other specialty dredges were grouped under the classification "Japanese" because of similarities in availability.

Factors and Ratings

The specific factors considered important in the selection of equipment are also listed in Table 4. Each dredge type was given a comparative rating between one and three for each factor, three being the best rating. For this evaluation, each factor was considered to be of equal importance. A description of the factors, the ratings given (shown in parentheses), and a discussion of how the rating for each piece of equipment was determined are presented in the following paragraphs.

It should be noted that the dredge groupings were identified and rated in 1986, before the availability of final results from the Calumet field demonstrations (Hayes, McLellan, and Truitt, 1988). If the results of the Calumet study and of the New Bedford pilot study were considered, the ratings given below would differ somewhat.

Availability

All cleanup dredging and related construction were likely to be accomplished by contracted efforts. Specifying readily available equipment simplifies the contracting process. If specialized equipment is required, it must be obtainable or constructable by contractors. Use of equipment designed or constructed in the United States or for which there is a United States licensee is also a factor in availability, because of certain legal restrictions on use of foreign equipment.

(Table 4 cont'd)

Individual Factor						
	Cost and	*Flexi-*	*Compatibility*			
Cleanup	*Production*	*bility*	*CAD*	*CDF*	*Draft*	*Access*
3	3	2	3	3	3	3
2	3	3	3	3	3	3
3	2	3	3	3	3	3
2	2	3	3	3	3	3
3	2	2	3	3	2	3
2	1	2	3	3	2	1
2	2	3	3	2	1	1
2	1	1	3	2	1	2
1	1	3	1	2	2	2

Ratings for availability were:

Horizontal auger and clamshell: Rated (3)
Numerous contractors in the general area have this equipment.
Small cutterhead: Rated (2)
Available but located farther away.
All other dredges: Rated (1)
Equipment is either a specialty item or located a considerable distance from New Bedford.

Safety

Safety of the dredging/construction personnel and the surrounding population was a major consideration. Volatilization of contaminants is a possibility when sediments resuspended during dredging and transport operations increase contaminant concentrations in the water column. If exposure of dredged material to the atmosphere is minimized, volatilization will also be minimized.

Ratings for safety were:

Hydraulic and specialty dredges and PNEUMA: Rated (3)
Removes the material in a slurry form and delivers it directly to the disposal site.
Clamshell: Rated (2)
Exposes the material to the atmosphere during the dredging process and requires the rehandling of the dredged material at the disposal site.

Resuspension

Release of contaminants into the water column occurs to some degree when the sediments are resuspended by the dredging operation. Selection of dredging equipment and operational techniques that have low potential for resuspension at the dredgehead is a major requirement for the New Bedford project. Other sources of resuspension, such as propeller wash from work boats, and grounding of scows should also be considered.

Ratings for resuspension were:

Horizontal auger and matchbox: Rated (3)
Matchbox and hood over the auger may reduce resuspension.
PNEUMA: Rated (3)
No mechanical action to stir the material although some sediment may be disturbed during movement of the dredge through the sediment.
Japanese dredges: Rated (3)
Dredging action is similar to a PNEUMA or some type of hooded cutterhead.
Cutterhead and dustpan: Rated (2)
Action of the cutterhead or water jets may cause some

resuspension (note that the pilot study later indicated the cutterhead would receive the highest rating).

Clamshell dredge: Rated (1)

Dredging process results in higher sediment resuspension than hydraulic or pneumatic equipment, material must also be rehandled prior to final disposal, and necessary scows and workboats would stir up additional material when working in shallow areas.

Maneuverability

Cleanup may be required over the entire upper estuary area, encompassing approximately 187 acres (76 hectares) of varying depth. Complete coverage of the bottom area by the dredging operation is required. Maneuverability of the equipment within the area is essential and should be accomplished with a minimum of sediment resuspension by work boats, cables, etc. Anything that hits the bottom with force will resuspend sediment. The spuds and anchors used in the positioning, moving, and anchoring of the dredge will stir up sediment in the area of impact. The use of a land-anchored winching system could minimize this problem.

Ratings for maneuverability were:

Horizontal auger: Rated (3)

Ability to work in shallow water and to work off a cable system rather than spuds, digging in a straight line instead of an arc.

Clamshell, cutterhead, dustpan, PNEUMA, matchbox, and Japanese dredges: Rated (2):

Operation uses either spuds or anchors and requires deeper water.

Dubuque: Rated (1)

Relatively large size and draft requirements.

Cleanup Precision

No existing dredge type is capable of dredging a thin surficial layer of contaminated material without leaving behind a portion of that layer and/or mixing a portion of the surficial layer with underlying clean sediment. Equipment selected for the New Bedford project should be capable of dredging layers of one foot (.3 m) (generally equivalent to the minimum depth of contamination for most of the estuary) with acceptable precision, assuming that a second one-foot (.3 m) layer would subsequently be dredged to remove any residual contaminated sediment and deeper pockets of contamination.

Ratings for cleanup precision were:

Horizontal auger: Rated (3)

Operates off a cable system with greater control over the depth of cut.

Matchbox and dustpan: Rated (3)
Good control over the dredging operation.
Cutterhead dredges: Rated (2)
Less control due to action of the rotating cutter.
PNEUMA: Rated (2)
Dredging operation may leave some spots undredged.
Japanese dredges: Rated (2)
Lack of experience in their operation in shallow water.
Clamshell dredge: Rated (1)
Dredging operation may leave spots undredged; control is less with respect to overdredging, and sloughing of the banks may leave contaminated sediment on the bottom.

Cost and Production

Although dredging for this project is for the purpose of cleanup and will be done in a controlled fashion, dredging production is an important consideration. Acceptable production rates will lessen the time during which sediments will be resuspended by the operation, minimizing the duration of associated exposures. At the same time, the production must be accomplished with minimal overdredging of clean sediment, since all material removed during the cleanup must be disposed of as contaminated material with associated higher cost. The rate at which the dredge can complete the dredging project is dependent on the percent solids, the volume of slurry, and the amount of time the dredge is pumping. It is also a function of the accuracy and control of the vertical and horizontal movement of the dredgehead and the ability to dredge an area with the minimum number of passes.

If debris is present in the sediment, it may have to be mechanically removed before a small-diameter hydraulic dredge can work in the area. Dredges with small suction lines (six to ten inches inside [15 to 25 cm] diameter) are easily clogged. The resulting downtime is nonproductive and costly. The entire dredge cycle of advancement, positioning, dredging, and cleanup should be evaluated and designed with a minimum amount of nondredging time.

The ratings for production and cost were:

Horizontal auger and small cutterhead dredges: Rated (3)
Contractors with this equipment are located in the general area, the equipment can work effectively in the shallow-water conditions, and operation can be controlled to reduce overdredging.
Dubuque, dustpan, shrouded cutterhead, and matchbox: Rated (2)
Increased cost to transport the equipment to New Bedford, plus the cost of fabricating and installing these to a dredge.
Clamshell dredge: Rated (1)
Rehandling of the material is required and more overdredging would be likely.

Japanese dredges: Rated (1)
Cost of transporting the equipment to New Bedford.
PNEUMA: Rated (1)
None are located in the area and production rate is low compared with the other equipment.

Flexibility

Equipment and operating techniques must be flexible to adjust to changes in water depths, sediment types, and disposal conditions. Equipment with which the dredging process can be adjusted offers an advantage over equipment that is limited to one method of operation.

The ratings for flexibility were:

Clamshell dredge: Rated (3)
Can work off a barge or off land, different size buckets could be used, and buckets could be modified.
Cutterhead and matchbox: Rated (3)
Ability to adjust the width and depth of the cut.
Horizontal auger and dustpan: Rated (2)
Only makes a straight cut.
Japanese dredges: Rated (2)
Lack of information regarding flexibility of operation.
PNEUMA: Rated (1)
Operates in only one way.

Compatibility with Disposal Options

Two available disposal options, use of a diked area or confined disposal facility (CDF) and contained aquatic disposal (CAD), require that the material be transported to a disposal site. CAD involves placement of contaminated material in a pre-excavated subaqueous pit, followed by capping with clean material. Equipment selected for dredging must be compatible with the transport and placement of material at the disposal site. Mechanical excavation and transport in barges would require rehandling (most likely by slurrying) to place material at either a CDF or CAD site. Hydraulic transport would not require rehandling.

The ratings for compatibility with the CAD disposal option were:

All hydraulic, PNEUMA, and Japanese dredges: Rated (3)
Material is pumped at a controllable rate directly to the site, where it can be placed with some degree of control.
Clamshell dredge: Rated (1)
Material must be rehandled for disposal and controlled placement of cap material would be difficult.

The ratings for compatibility with the CDF disposal option were:

Small hydraulic and Japanese dredges: Rated (3)
Material can be pumped directly to the site at a desirable rate.

Clamshell dredge: Rated (2)
Material must be rehandled.
Dubuque: Rated (2)
Size with regard to the pilot study site may overload capacity without intermittent operation.
PNEUMA: Rated (2)
Low production rate.

Draft

The required draft of the dredge and its attendant work boats is a major constraint in the upper estuary. Although a portion of the estuary has water depths exceeding three feet at low tide, a majority of the area to be dredged has a water depth less than three feet (.9 m) at low tide. Equipment with draft requirements exceeding two feet (.6 m) would be constrained to operate during only a portion of the tidal cycle. Draft is also an important factor for the auxiliary vessels that service or reposition the dredge. If the water is shallow, the propeller wash will stir the bottom sediment.

The ratings for draft were:

Horizontal auger, matchbox, and small cutterhead dredges: Rated (3)
Can operate in very shallow water.
Dustpan: Rated (2)
Larger vessel size requires greater depth.
Clamshell: Rated (2)
Equipment operates off a barge whose movement would be restricted as would the scows required for transport of material.
Japanese dredges: Rated (2)
Size and draft requirements.
PNEUMA: Rated (1)
Does not operate well in shallow water.
Dubuque: Rated (1)
Size of equipment would restrict operations.

Access

Equipment must be able to pass through restricted bridge openings to enter the upper estuary or be capable of being transported by truck and launched from the shore.

The ratings for access were:

Horizontal auger and small cutterhead: Rated (3)
Would not be restricted by the bridge clearance and are transportable by truck.
Clamshell: Rated (2)
Would be restricted by the bridge clearance and the lack of waterfront facilities for loading the crane onto a barge

PNEUMA: Rated (2)
Not readily available.
Japanese dredges: Rated (1)
Relative size of the equipment and the distance to New Bedford.
Dubuque: Rated (1)
Increased distance to New Bedford and the size of the equipment.

Selected Equipment

... the horizontal auger dredge received the highest rating ... followed by the small cutterhead and the matchbox...

As shown in Table 4, the horizontal auger dredge (Figure 4) received the highest rating (thirty-two points of a possible thirty-three), followed by the small cutterhead and the matchbox (Figures 2 and 3) (each with a rating of twenty-nine). Based on the ratings, these three hydraulic dredge types were selected for demonstration during a pilot field study. A summary of the major advantages of the hydraulic dredging process and these specific dredgeheads is given in the following paragraphs.

Hydraulic Dredging Operations

Hydraulic dredging (horizontal auger, cutterhead, and matchbox) offers several major advantages over mechanical dredging. Small hydraulic dredges can be used more effectively in the shallow water of the upper estuary, with greater maneuverability, flexibility, and production. The material can be pumped directly to the disposal site, without need for rehandling or exposure to the atmosphere. Hydraulic dredges can remove contaminated material with higher precision and lower sediment resuspension than mechanical dredges.

Horizontal Auger Dredge

The horizontal auger dredge uses a cable and winch system, anchored on shore, which would allow the dredge to work in parallel and overlapping cuts, leaving a flat-bottomed cut with no windows. This operation has a greater potential for control and precision removal of contaminated material. The horizontal auger has an auger-type head with attached hood, which can potentially reduce sediment resuspension and increase the solids content of the slurry.

Small Cutterhead

The cutterhead, considered to be a "standard" hydraulic dredge type, is widely available. Cutterhead operation can be optimized with respect to resuspension by varying cutter rotation, swing speed, and cutter burial. Since the cutterhead is most frequently used, many experienced, capable operators are available. The skill of the dredge operator is a major factor in minimizing sediment resuspension.

Matchbox

The dredgehead design has been reported to generate lower sediment

resuspension than a conventional cutterhead. The matchbox could be easily compared with the cutterhead using the same hydraulic dredge.

COSTS

The costs of dredging contaminated sediments vary according to a number of factors. Costs are generally comparable to those for conventional navigational dredging projects, however, unless extraordinary control measures are required. For the New Bedford project, the estimated costs for full-scale cleanup dredging ranged from $9 to $12 per cubic yard (Averett et al., 1988).

PILOT STUDY

For the New Bedford project, the estimated costs for full-scale cleanup dredging ranged from $9 to $12 per cubic yard.

Early in the evaluations it was determined that a pilot study would be conducted to provide field data on the performance of dredging equipment and the feasibility of disposal alternatives. The dredging requirements for the pilot study were equivalent to those for a full-scale cleanup project. Therefore, the selection of dredging equipment for the pilot study was aimed at identifying several promising dredge types considered most appropriate for a full-scale cleanup. A detailed description of the overall pilot study is available (New England Division 1990).

The pilot study was conducted in the fall of 1988, in a small cove of the upper estuary (See Figure 1). PCB concentrations in the cove sediments ranged from 150 to 600 ppm in the upper six inches of sediment and were not detected below the two-foot depth. Approximately 10,000 cubic yards of sediment were dredged during the pilot study, of which approximately 2,900 cubic yards was contaminated. The water depths in the cove were .5 feet at low water with a tidal range of four feet. These conditions were typical of a large portion of the upper estuary.

The cutterhead, matchbox, and horizontal auger dredgeheads were evaluated in the pilot study, each using separate hydraulic dredge plants. The pilot study was designed to evaluate the ability of the dredges to remove the layer of contaminated sediment while minimizing overdredging, minimize resuspension of sediment during operation, and operate in the shallow water conditions of the upper estuary.

An extensive program of monitoring and sampling was conducted as a part of the pilot study. This program included an array of stations located around the dredges, in the vicinity of the cove, and at the constriction of the Coggeshall Street Bridge. All of the dredges successfully removed contaminated sediments without excessive overdredging of clean material. The dredges removed the sediment in two passes of the dredgehead, leaving PCB concentrations of less than 10 ppm in the sediments. Also, a defined plume of resuspended material never developed, and no unacceptable concentrations of resuspended sediment or released PCBs were observed during the dredging process for any of the dredges. The rates of sediment resuspension and contaminant release at the operating dredgeheads are summarized in Table 5. Based on the results of the pilot, the cutterhead dredge was recommended for future cleanup of the upper estuary. ■

Table 5. Sediment Resuspension and Contaminant Release Rates During New Bedford Pilot Study Dredging.

Dredge	Resuspension Rate (g/sec)	Total Suspended Solids (mg/l)	Total PCB (ppb)
Cutterhead	12	82	7.0
Matchbox	46	319	2.6
Horizontal Auger	329	1610	54.9

Notes:

1. Background levels for suspended solids and PCBs are less than 10 mg/l and .5 to 1.0 ppb, respectively.
2. The resuspension rate is calculated using the following factors: water depth, length of dredgehead, swing speed, and average suspended solids in the water column adjacent to the operating dredgehead.
3. Samples used to obtain this data were taken from a sampling device installed at the dredgehead. Samples were drawn from 6 sampling ports positioned around the dredgehead.

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Environmental Toxicology

PCB AVAILABILITY ASSESSMENT OF RIVER DREDGING USING CAGED CLAMS AND FISH

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Abstract—The effects of dredging to remove sediments contaminated with polychlorinated biphenyls (PCBs) were studied in the South Branch of the Shiawassee River in south-central Michigan. Caged fingernail clams, *Sphaerium striatinum* (Lamarck), and fathead minnows, *Pimephales promelas* Rafinesque, were used to monitor the bioavailability of PCBs, and these data were compared with changes in water column concentrations before dredging, during dredging and up to six months after dredging was completed. Monitoring of water, clams and fish during dredging confirmed that significant amounts of PCBs were released from the sediments. At all locations downstream and in the area of the dredging, there were increases in the availability of PCBs for at least six months. PCB concentrations in clams and fish in the dredged zone increased from 64.5 to 87.95 μg/g dry weight and from 13.82 to 18.30 μg/g dry weight, respectively, after dredging. Downstream (11 km), post-dredge uptake was also higher; however, clams showed less uptake than fish. This implied different uptake routes for the two organisms. There was no noticeable change in total PCB concentration in the water after dredging.

In situ experiments were run to determine uptake curves and time to uptake equilibrium for both clams and fish. Results of tests for equilibrium uptake by fatheads agreed with literature values. Uptake maxima occurred within 20 to 30 d. For clams, maximum uptake for Aroclor 1242 occurred after 9 d. Log bioconcentration factors for clams ranged from 2.6 to 4.5 for Aroclor 1242 and from 3.0 to 4.4 for Aroclor 1254; for fish, they ranged from 3.0 to 4.4 for Aroclor 1242 and from 4.5 to 5.5 for Aroclor 1254.

Keywords—Polychlorinated biphenyls (PCBs) Dredging Fingernail clams Uptake Fathead minnows

INTRODUCTION

Many of the streams and rivers in south-central Michigan have long histories of contamination from foundry operations, oil refineries and chemical manufacture [1,2]. With the exception of visible pollutants, however, more subtle contamination was not of concern until the early to mid-1970s. Initial data on polychlorinated biphenyl (PCB) contamination of sediments in the South Branch of Shiawassee River was obtained in Michigan Department of Natural Resource (MDNR) surveys in 1974 [3]. A follow-up survey by the MDNR in 1977 resulted in the closing of the river for fishing in 1978 [3], and two surveys in 1981 provided further evidence of widespread sediment PCB contamination, primarily by Aroclor 1242 [4]. The source of PCBs was identified as the Cast Forge Company of Howell, Michigan, and, as a result of a court settlement in August 1983, the company was assessed a fine awarded to the state of Michigan for cleaning up the river.

The state of Michigan (i.e., MDNR) decided that dredging was the best way to remove PCB-laden sediments, and A-1 Disposal Corporation of Plainwell, Michigan, was contracted to carry out the dredging during the summer and fall of 1982. To coincide with cleanup operations, the University of Michigan monitored the impact and results of dredging through studies of PCB uptake by caged fingernail clams and fathead minnows; concentrations of dissolved PCBs and PCBs associated with suspended sediments were also monitored.

Dredging, as a means of removal of sediment-bound contaminants, has been implemented or proposed for several river systems [2,5,6], but to date there is little information on either the efficiency of dredging as a cleanup measure or its potential for increasing toxicant concentrations and bioavailability downstream. Hafferty et al. [6] monitored the possible increased release of PCBs during vacuum dredging of a transformer spill

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(pure Aroclor 1242) in the Duwamish River in Seattle, Washington. As here, their study involved pre-dredge, during-dredge, and post-dredge sampling of the water and suspended solids. During dredging, they noted a direct increase in concentration at the dredge site as well as a shift in the PCB homologue mixture, but there was no evidence of a significant change ($\alpha = 0.05$) in PCB concentrations 15 to 70 m downstream either during or after dredging. However, their values were always higher at the downstream sites than at upstream reference stations. Tofflemire et al. [7] studied the movement of PCBs during maintenance dredging of a contaminated section of the Hudson River. In their study, as in the Duwamish River study, there was a definite increase in waterborne concentrations of PCBs at the site during dredging (from 0.36 to 3.91 μg/L). PCB levels decreased, however, to near-background concentrations approximately 1.6 km downstream from the dredge site (0.714 μg/L), even during dredging. Tofflemire et al. [7], in examining the rate of loss of PCBs during high- and low-flow periods, concluded that losses from dredging continued to act as a constant source, with mean PCB concentrations lower during high flow because of dilution.

The use of caged organisms to monitor environmental quality in aquatic environments is not new [8]. The emphasis in the majority of such tests, however, has been on testing for toxicity and not for residue accumulation. These tests should provide a rapid means of integrating residue availability and are directly applicable to the biota of the stream. Skea et al. [9] caged different species of Hudson River fish to monitor PCB availability downstream of a suspected PCB spill. Werner [10] used caged clams to monitor PCB availability during dredging of a PCB hotspot in the Hudson River. Caged mussels were used to monitor PCB availability at a New London, Connecticut, dump site [11]. Caged sunfish and yellow perch were used to measure PCB availability at the Waukegan PCB spill area in Lake Michigan [12]. Studies using caged organisms to test for toxicity and availability of other chemicals have been reviewed by Seelye and Mac [13].

Background

The sediment of the South Branch of the Shiawassee River, from just below the Cast Forge facility to Oak Grove Road (approximately 17 km downstream), was described in the MDNR surveys of 1974 and 1977 as having "extremely high" concentrations of PCBs, i.e., more than 5,000 μg/kg [3,14]. Elevated concentrations of PCBs (>5 ppm) were also reported in fish in the stream reac extending from the Cast Forge facility as far a 50 km downstream. The highest concentrations o PCBs in the sediment appeared to be confined t the region extending from just below Cast Forg to about 1.6 km downstream, i.e., about 600 r downstream from Bowen Road (Fig. 1). Thi stretch of the river was then targeted for dredgin by the A-1 Disposal Corporation [15]. The Jul 1981 MDNR [4] survey of this stretch of river wa used as the basis for estimates of the followin PCB sediment concentrations: an average of 533. μg/g for km 0 to 0.4, starting just below the Cas Forge outfall; 24.4 μg/g for km 0.4 to 1.6, endin at Bowen Road; and 21.3 μg/g for the 600-r stretch downstream of Bowen Road. In addition a large buildup of PCBs (>500 μg/g) was detecte several meters away from the channel in a floo plain just upstream from Bowen Road.

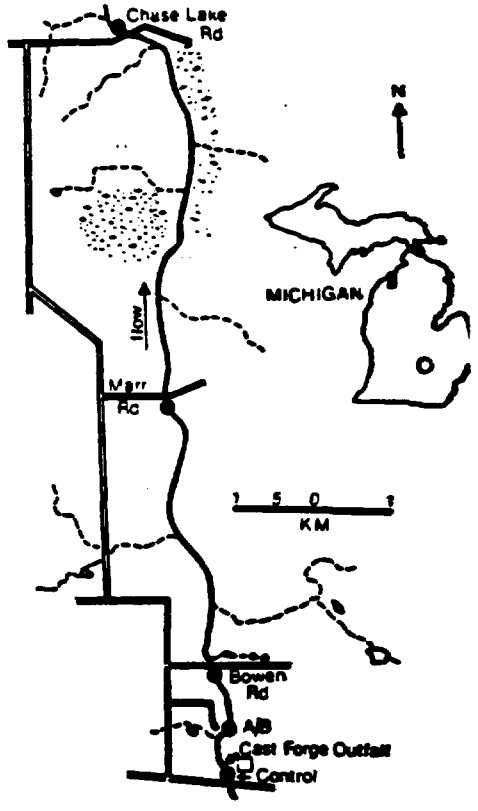

Fig. 1. Map of South Branch of the Shiawassee River i south-central Michigan, showing sample sites, road small tributaries and marshy areas. Inset: Study area i Michigan.

Vacuum dredging was used for most of the in-river sediment removal, beginning in August 1982. Final dredging was finished on or about 11 November 1982. The dredging operations reportedly removed 1,148 kg PCBs contained in 1,509 m^3 of river sediment; 3,445,000 liters of waste liquid was generated from the vacuuming [15]. We have no data to determine what fraction of the total PCB contamination or of the original inputs was removed.

MATERIALS AND METHODS

Study site

The reach of the South Branch of the Shiawassee River slated for dredging was a third- to fourth-order sand-bottomed stream with few distinct riffles and pools. Late summer water depths ranged from 0.25 to 2 m, and widths ranged from 8 to 15 m. While the predominant substratum was sand, deeper pools had silt accumulations of up to 0.6 m deep. Base discharge was approximately 1 m^3/s. High flows have been recorded in June and July of up to 3 m^3/s, and the stream is subject to periodic flash flooding. Much of the surrounding land is used for agriculture, although there are areas of second-growth beech-maple forest and marsh.

One control and four study sites were monitored (Fig. 1). The control site was just above the outfall at Cast Forge (120 m downriver from road M-59). The study sites were selected based on river access and the potential extent of dredging: site A/B was 0.4 km downstream; Bowen Road, 1.6 km downstream; Marr Road, 5.3 km downstream; Chase Lake Road, 11 km downstream. Water was regularly sampled at the control site and at Bowen Road and Chase Lake Road. Caged fish were placed at the control site and at Bowen Road and Chase Lake Road. Caged clams were placed at all sites, and water parameters and discharge were regularly measured at all sites.

River water characterization

River discharge was monitored on each sample date. Both stream depth to a fixed marker (gauge or bridge abutment) and direct velocity were measured with a hand-held Teledyne Pygmy Gurly Meter. Also measured were pH, temperature, dissolved oxygen and specific conductance. For most of the sampling trips, suspended solids were collected at each site. Suspended solids were collected by suction filtering 1 liter of water through preweighed 47-mm, 0.1-m Millipore filters. For the determination of the solids concentrations, the tared filters plus retained particulates were dried to a constant weight at 60°C.

Sampling for PCBs

Stream water was collected every 2 to 3 weeks before and during dredging in 1982 and then again in the following spring and summer of 1983. The usual volumes collected ranged from 11 to 15 liters. Samples were collected by dipping a 7.5-liter galvanized steel bucket into the stream and immediately vacuum-filtering the water through preignited Gelman Type A/E 143-mm-diameter glass fiber filters rated to retain particles 1 μm or more in diameter. Filters containing particulate matter were then carefully folded face-inward, wrapped in aluminum foil packets and frozen. The filtrate was collected and stored in 20-liter glass carboys.

Water extraction

The suspended matter retained on each of the filters was processed by 24-h solvent extraction using a Soxhlet apparatus. An acetone:hexane (1:1, v/v) solvent mixture was used in the Soxhlet, with the acetone added first to ensure complete solvent contact with the wet filters. After extraction, acetone was removed by washing the filters with PCB-free water. The extract then was dried by passage through a 15-g column of ignited Na_2SO_4. The extracts were adjusted in volume for subsequent cleanup and fractionation by evaporative concentration using Kuderna Danish concentrators.

Dissolved PCBs were extracted from the filtrate portion of samples using solvent extraction with pesticide-free methylene chloride. To assure adequate mixing with methylene chloride, samples were vigorously stirred for 15 min using a power drill to rotate a metal rod with a chain attached to its end. The water and methylene chloride layers were allowed to separate, then the upper water layer was siphoned off. To remove the final few milliliters of water, some of which was in an emulsion, the extract was transferred to a separatory funnel and allowed to stand until most of the emulsion disappeared. The methylene chloride was drained from the bottom of the separatory funnel through a 15-g column containing ignited Na_2SO_4 and then concentrated by evaporation. Hexane was added to replace the lower-boiling methylene chloride by evaporative concentration.

Clams

The fingernail clams, *Sphaerium striatinum* (Lamarck), used for this study were collected from Fleming Creek where the stream flows through the

University of Michigan Botanical Gardens. Fleming Creek and the South Branch of the Shiawassee River are approximately the same size, with similar substrata; both contain natural populations of *S. striatinum*. Fleming Creek is not known to have any PCB contamination, and preliminary tests showed extremely low background concentrations in the clams' tissues. Clams were maintained in aerated aquaria in clean Lake Michigan sand and Lake Michigan water at approximately 15°C until they were transferred to the South Branch of the Shiawassee River (usually after 2 to 6 d). The clams ranged from 6 to 10 mm in length, with an average of 8 mm.

The cages were made of standard hardware cloth (heavy wire screen) fashioned into 10-cm-deep rectangular boxes enclosing a 43 × 21 × 2.5-cm deep baking sheet. At the river, each baking sheet was filled with wet Lake Michigan sand and about 200 clams. The cages were held in place by tying them to metal pipes driven into the river bottom. Two cages were set at each site. Sampling was accomplished by taking cages from the stream, removing the baking sheets and hand-picking clams from the sand, usually 10 clams per cage for each collection. The intervals of exposure varied from 19 to 68 d in 1982, and from 14 to 45 d in 1983. A short-term uptake experiment was conducted in 1983 to verify when equilibrium concentrations were attained. In this experiment, samples were taken after 1, 3, 7, 9 and 14 d of exposure.

All sampled clams were wrapped whole in aluminum foil packets, labeled and stored frozen in the laboratory prior to extraction. To prepare the clams for extraction, the meat of all the clams in the sample was removed, weighed as a composite and freeze-dried for 24 h in preweighed glass scintillation vials loosely covered with aluminum foil-lined lids. Freeze-dried samples were ground in a mortar, dry weights were determined and then the samples were stored in scintillation vials.

Fish

Hatchery-reared fathead minnows, *Pimephales promelas* Rafinesque, were obtained from the Kutz Fish Hatchery in Elverson, Pennsylvania, 1 to 2 d before being caged. The cages were constructed of standard hardware cloth fashioned into triangular boxes 45 cm in height and 60 cm along each face. For the first year, the cages were partitioned into two compartments of equal size (one above the other). In the second year, the horizontal partition was moved down to 8 cm from the bottom. The cages were placed in the stream with the apex of the triangle anchored to a metal spike embedded in the river bottom. Sampling was accomplished through a door toward the rear of the cages that was wired closed between samplings. In the pre-dredge tests, both the upper and lower cages were stocked with fish, for the purpose of comparing the availabilities of PCBs to the fish in contact with the substratum with that to fish in contact only with waterborne PCBs. In the post-dredge fish uptake studies, only the upper cages were used. Approximately 100 fish, 5 to 10 cm long, were placed in each compartment. Two sets of cages were placed in the deepest areas at each sampling location, the second cage about 10 m downstream from the first.

Three fish were removed from the upper and lower compartments of each cage at each sampling time. Each group of three fish was wrapped in aluminum foil, labeled and returned to the laboratory to be stored frozen until preparation for extraction and analysis. To determine the rate of uptake and final equilibrium concentrations of PCBs accumulated by the fish, sampling was conducted at various intervals during a 29-d exposure period (8 May to 5 June 1982) and a 62-d exposure period (17 May to 17 July 1983). Also, samples of unexposed fish, replicates and spikes were prepared for quality assurance tests. The general condition of the fish was determined visually during site visits and by calculation of wet versus dry weights and lipid content.

In preparation for extraction, the combined wet weight of each three-fish sample was determined, and then the sample was placed frozen into a freeze-drying jar. The jars were plugged by soft polyurethane foam installed in the glass tube attached to the main chamber of the freeze-drying compartment. The foam plugs were analyzed to confirm that no PCBs passed either into the sample from the freeze dryer or out of the sample via volatilization during drying. The samples were freeze-dried for 24 h and reweighed. Samples were then ground to a fine powder using a Bunn coffee grinder and transferred to glass scintillation vials.

Clam and fish extractions

The ground freeze-dried tissue was weighed into precleaned Soxhlet thimbles and extracted for 12 to 24 h with pesticide-grade hexane. Kontes Micro Soxhlets were used for clam tissue extractions since the total sample weights were small (<1 g). Fish tissue was extracted in larger 250-ml Soxhlet assemblies. After extraction, 10% of the

fish extract and 40% of the clam extract were removed for gravimetric determination of lipid residue content, which involved solvent evaporation and drying of the extracts at 60°C.

Extract cleanup, fractionation and analysis

All of the extracts were cleaned up by column chromatography, using an alumina cleanup procedure [16]. For selective fractionation of the PCBs from interfering peaks (chlordanes, DDTs, toxaphenes, etc.), a silicic acid [17] procedure was used for about half the analyses. For the remainder of the samples (most post-dredge samples and all of the fish), a silica gel fractionation procedure was used [18].

Fractions for PCB analysis were analyzed with a Varian Model 3700 electron-capture gas chromatograph. Two 2.5 m long × 2 mm i.d. borosilicate glass columns were used: one a mixed-phase column packed with 1.5% SP-2251/1.95% SP-2401 on 100/120 mesh Supelcoport (Supelco Inc., Bellefonte, Pennsylvania) and the second a single-phase column packed with 5% SP-2100 on 100/120 mesh Supelcoport. The heated zones were as follows: 220°C injector, 190°C column and 320°C detector. The instrument operating conditions are described by Rice et al. [2], with peak areas corrected using the response factor correction procedure proposed by Sawyer [19]. Sawyer [20] and Webb and McCall [21] demonstrated the advantages of using response-factor-weighted values to achieve both better accuracy and precision in PCB analysis. We have found this to be especially true of PCB chromatographic patterns that are altered from the patterns of the standards used for matching. The response factor technique also provided accurate data on sample homologue composition.

The blanks averaged 0.02 μg/L PCB for the water and 0.13 and 0.12 μg/g PCB for the clams and fish, respectively. With the exception of the results for the filtered water, these blank values were used to correct all analytical data. Clam and fish tissue background checks were conducted to verify the zero-time background concentrations of the PCBs. These values averaged 0.10 μg/g for both Aroclor 1242 and Aroclor 1254 for both clams and fish. The duplicate performance for total PCB in water showed a relative error of 6.2†. For clams and fish, the errors averaged 21.4 and 6.6†, respectively. The limits of detection for both of the Aroclor mixtures (1242 and 1254) were 0.004 μg/L for the water analyses and 0.1 μg/g for the biological tissue analyses.

RESULTS

Field data

A tabulation of all the physicochemical field data for this study can be found elsewhere [22]. Most values were within the ranges for third- and fourth-order streams in south-central Michigan and were not significantly altered by the dredging. The pH most often was about 8.0 and varied from 7.4 to 8.6, dissolved oxygen was at saturation and midsummer temperatures were near 30°C. Specific conductance was always high (>500 μmhos/cm), particularly in late July through October 1982 (>1,000 μmhos/cm), which was attributed to both low flow and dredging. Suspended solids (3 to 25 mg/L) did not show any correlation with discharge for either 1982 or 1983. The lack of correlation between suspended solids and discharge could be attributed to dilution factors and to agricultural cycles within the river's drainage basin.

Timed uptake experiments

Timed uptake studies with the clams were conducted at the control site, site A/B and at Bowen and Chase Lake Roads. While the magnitude of uptake varied, the patterns were generally similar among the sites (Fig. 2), i.e., rapid and almost linear uptake until the fourteenth day for both Aroclor 1242 and 1254, then uptake of the Aroclor 1242 mixture reached equilibrium while uptake of the Aroclor 1254 mixture continued to climb at a steady but slower rate until the experiment was terminated (Fig. 2). The lipid weights per organism (average 4.57% lipid on a dry weight basis) did not vary in any consistent pattern during the test (Table 1).

For clams, Aroclor 1242 equilibrium occurred after about 14 d. Fortunately, all of the clam exposures lasted longer than 19 d. Establishing the equilibrium point for the fish was more difficult. Generally, tissue concentrations increased after exposure, reached a brief plateau and then declined (Fig. 3). For 1982 data, uptake studies were conducted for only 29 d, and it appeared that the concentrations in tissues might be declining after 19 d. For our purposes, we assumed that the average of the three highest values in the 1982 uptake study represented equilibrium (13, 16 and 19 d). In 1983, uptake tests were conducted for 62 d, with declines in PCB concentrations not occurring until after 44 d; therefore, the mean of the concentration values for days 16, 23 and 44 was used to represent equilibrium.

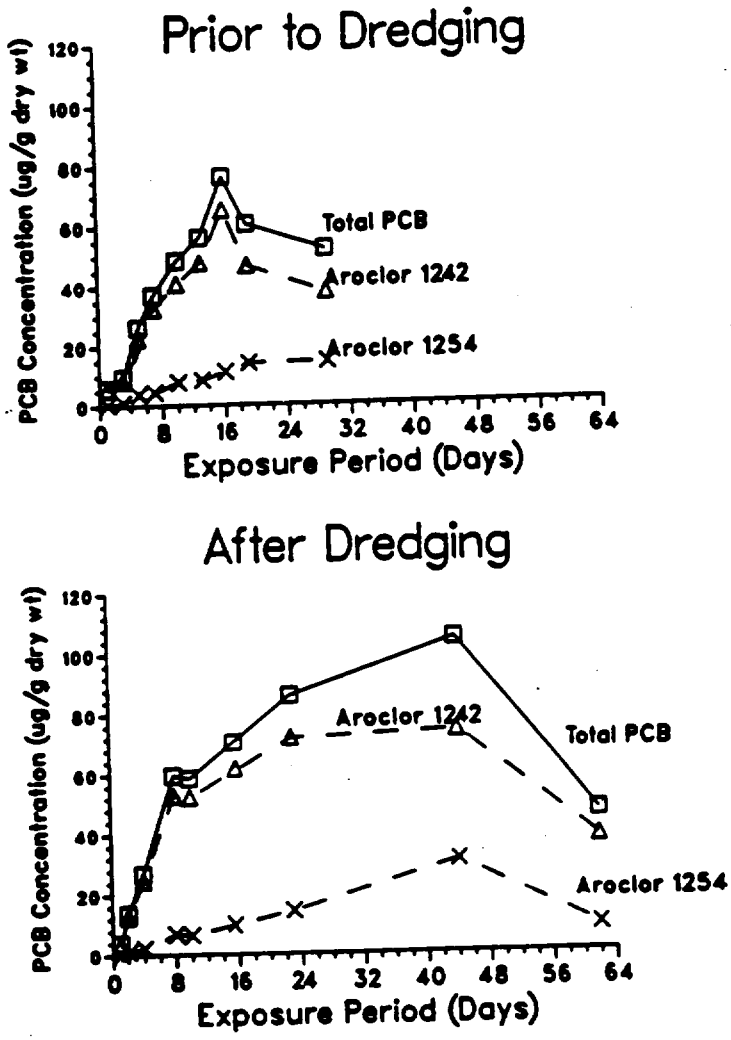

Fig. 2. Timed uptake of PCBs, expressed on a dry weight basis, by fathead minnows exposed at Bowen Road before (1982) and after (1983) dredging.

In pre-dredging tests begun on 8 May of fish caged at Bowen Road, there appeared to be little or no difference between PCB tissue concentrations in the upper and lower cage compartments on any date, particularly days 0 to 7 (Fig. 4); therefore, the results were averaged for both compartments (Fig. 3). Uptake of Aroclor 1242 by fish at Bowen Road was steady and rapid for the first 10 d, but then, excluding the high value at day 16, appeared to level off until after day 19, when it declined (Fig. 3). The uptake of Aroclor 1254 reached a plateau after day 19 but did not decrease thereafter.

The lipid content of the fish at all sites in 198 started from initially low levels (0.54% lipid on a wet weight basis) and rose rapidly 2 d later to a average of 2.6% lipid, whereupon it decline slowly at all locations (average of 0.40% lipid o the last day, day 29) (Table 1). The percent freeze dried weight per fish also declined slowly over th exposure period. Given the worsening condition c the fish over time, we cannot explain the gain i lipid content.

The 1983 post-dredging fish equilibrium stud lasted 62 d. The total PCB concentrations ir creased rapidly through the eighth day, then mo

Table 1. Biological condition (lipid content) of clams and fish during timed uptake experiments

Days exposed	Percent fresh wt.			Percent dry wt.		
	Pre-dredge fish	Post-dredge fish	Clams	Pre-dredge fish	Post-dredge fish	Clams
1	0.54(3)	2.34(3)	0.41(3)	20.9(3)	21.2 (3)	10.0 (3)
2		2.51(3)			22.7 (3)	
3	2.76(3)		0.39(3)	22.8(3)		10.3 (3)
4		1.96(2)			20.6 (2)	
5	1.64(3)			20.9(3)		
7	1.75(5)		0.33(3)	20.9(3)		8.06(3)
8		1.65(3)			20.73(3)	
9			0.38(2)			7.89(3)
10	1.10(3)	1.65(3)		20.3(3)	20.4 (3)	
13	1.29(2)			20.2(3)		
14			0.37(3)			10.4 (3)
16	1.07(3)	1.33(3)		19.5(3)	20.3 (3)	
19	0.82(3)			19.4(3)		
23		1.09(3)			19.7 (3)	
29	0.40(3)			1.85(2)		
44		0.82(2)			18.3 (2)	
45			0.25(3)			5.79(3)
62		0.83(3)			19.6 (3)	

Number of samples averaged in parentheses.

slowly through the forty-fourth day, after which the concentrations declined at all locations (Fig. 3). Concentrations declined more sharply for Aroclor 1242 than for Aroclor 1254 after 44 d. Uptake of Aroclor 1242 was rapid over the first 8 to 10 d, then more gradual up to day 23 when equilibrium seemed to have been achieved. The concentration of Aroclor 1254 rose more gradually and peaked later, at 44 d. Similar to the 1982 tests, there was a gradual and steady decline in lipid content in the fish after the second to eighth days of the exposure period; initially the content rose slightly at Bowen and Chase Lake Roads (Table 1).

Effects of dredging

The equilibrium values, as calculated above, were used for comparisons with the results of the three major study intervals of pre-dredging (21 March to 29 June 1982), during dredging (1 July to 2 October 1982) and post-dredging (16 May to 17 July 1983) (Table 2, Figs. 5 to 7). From the water column data for Bowen and Chase Lake Roads (Fig. 5), there was a significant increase in PCB concentrations after 1 July at all locations downstream of the Cast Forge. At Bowen Road, the concentration rose gradually from 1.7 μg/L total PCBs on 1 July (average pre-dredge background concentration 1.0 μg/L) to a maximum of 13.7 μg/L on 27 July. This was the highest concentration of PCBs measured in water at any location during dredging. The PCB concentrations then decreased through August; thereafter, they fell to 1.9 μg/L on 2 October, the last sampling date. At Chase Lake Road, the maximum concentration of PCBs measured in the water column was 1.4 μg/L on 1 July, which was before dredging was started (Fig. 5). It should be noted that the high concentrations recorded on 27 July at Bowen Road were not observed farther downstream at Chase Lake Road.

Generally, the patterns of PCB concentrations in the clams sampled at sites A/B and Bowen Road (Fig. 6) were similar to each other but different from patterns for Marr Road and Chase Lake Road samples (Fig. 7). For A/B and Bowen, the first samples after dredging showed a dramatic increase in PCB uptake, whereas at Marr and Chase, concentrations in the first samples after dredging actually appeared to be lower than pre-dredging. These data may reflect the relative distances from the dredging area of sites A/B and Bowen as compared with Marr and Chase Lake Roads (Table 2).

Comparison of 1982 and 1983 data for water column, clams and fish

For all collections, caged clams and fish closest to the Cast Forge outfall area showed the highest amounts of PCBs in their tissues. During dredging, PCB concentrations showed a significant (α =

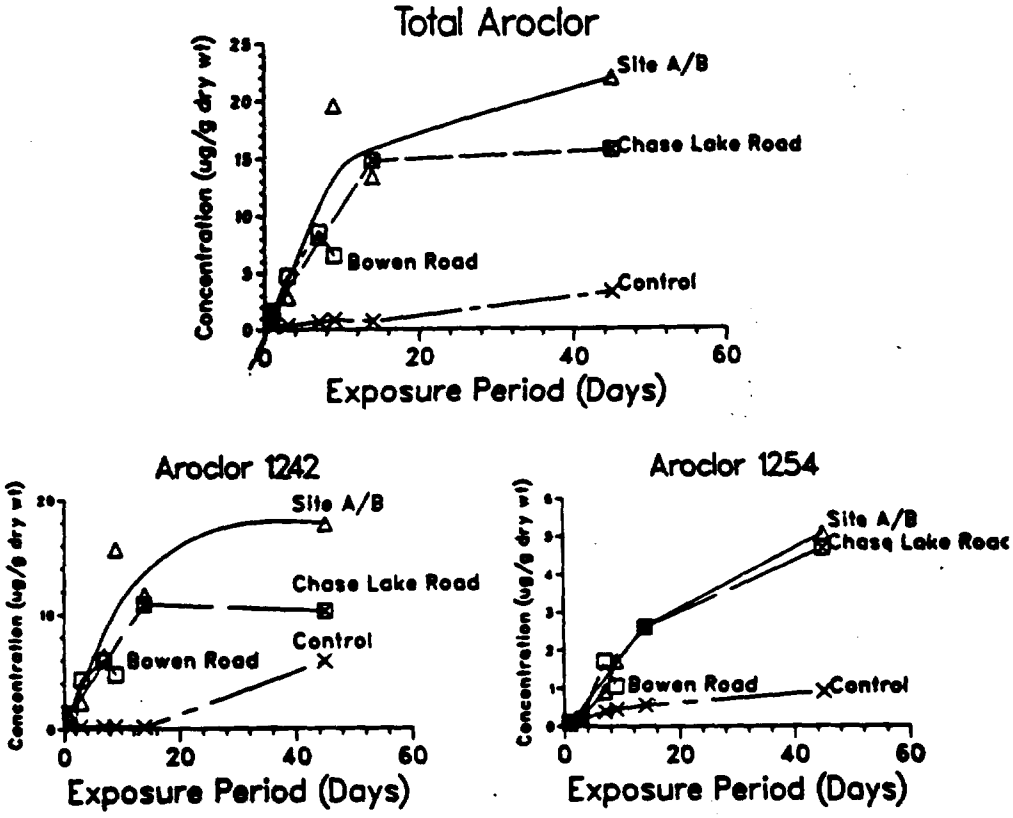

Fig. 3. Timed uptake of total Aroclor, Aroclor 1242 and Aroclor 1254 by clams exposed during post-dredge tes ing at each of the sampling locations.

0.05) increase at site A/B and at Bowen Road in clams and at Bowen Road and at Marr Road in water (dissolved and particulate). The concentration of PCBs in the water column at Chase Lake Road (the site farthest downstream) was only slightly higher (not significant at the 5% level) than during pre-dredge studies, and there were no changes in clam tissue concentrations at this site. The clams also showed no significant change in PCB concentrations at Marr Road, which was even closer than Chase Lake Road to the Cast Forge outfall.

All of the post-dredge concentrations of PCBs for all of the samples collected downstream from Cast Forge were higher than those collected before dredging. One exception to this was the decrease in clam PCB concentrations at Bowen Road; however, this decrease appeared to have been partially the result of problems with siltation from dredging at this location that partly buried the cages. Only the fish sampled at Chase Lake Road co tained statistically greater amounts of PCBs. A of the post-dredge PCB concentrations were ge erally lower than during-dredge concentrations.

DISCUSSION

As expected, there was an increase in the co centration of PCBs in the water column duri dredging (Table 2), which declined rather quic farther downstream. The sampled clams a showed this trend. For example, at Marr Ro which was only 4.0 km from Bowen Road, th was no increase in PCBs in the clams. Nor w there any observable increase at Chase Lake Ro which was 9.25 km downstream of Bowen Ro It appeared from the water column and clam d that PCBs released during dredging were mobilized very far downstream (at least not o the short term), and they produced only locali

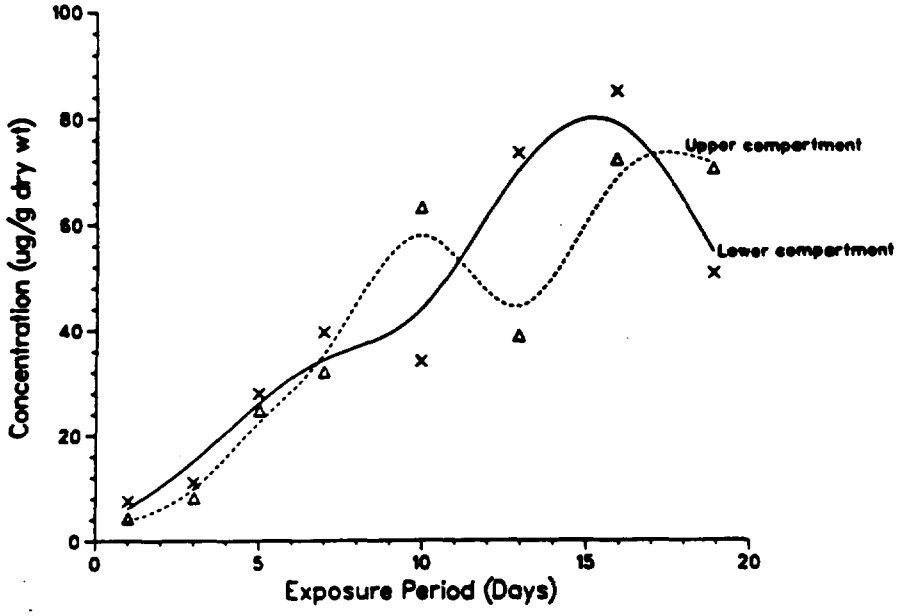

Fig. 4. Uptake of total PCBs by fathead minnows in cages positioned on the river bottom and in cages positioned above the bottom in a pre-dredge test carried out at the Bowen Road site.

Table 2. Total PCB (±SD) measured in the water column, clams and fish before, during and 1 year after dredging in the South Branch of the Shiawassee River

River mile (site)	Pre-dredge	During dredging	Post-dredge
Water (μg/L)			
0.0 (control site)	0.047 ± 0.34(3)	0.029 ± 0.015(5)	0.037 ± 0.015(5)
1.0 (Bowen Rd.)	1.10 ± 0.37(6)	4.67 ± 3.76(9)	1.11 ± 0.43(5)
3.5 (Marr Rd.)	0.68(1)	2.83(1)	—
6.8 (Chase Lake Rd.)	0.65 ± 0.20(5)	1.03 ± 0.26(5)	0.522 ± 0.095(5)
Clams (μg/g dry wt.)			
0.0 (control site)	0.78(1)	1.18 ± 2.57(5)	1.36 ± 1.25(4)
0.25 (site A/B)	13.82 ± 2.31(2)	59.08 ± 33.3(4)	18.30 ± 4.38(3)
1.0 (Bowen Rd.)	40.02(1)	69.34 ± 16.1(4)	6.49*(1)
3.3 (Marr Rd.)	44.27(1)	43.54 ± 9.6(4)	—
6.8 (Chase Lake Rd.)	13.21(1)	12.55 ± 3.68(3)	15.26 ± 0.56(2)
Fish (μg/g dry wt.)			
0.0 (control site)	1.78 ± 0.08(3)	—	1.62 ± 0.40(2)
1.0 (Bowen Rd.)	64.54 ± 10.70(3)	—	87.95 ± 15.77(3)
6.8 (Chase Lake Rd.)	32.09 ± 7.08(3)	—	61.14 ± 0.23(2)

Number of analyses in parentheses.
*These cages were silted over; therefore, this result is unusually low.

increases in PCBs. These data conform to those of Hafferty et al. [6] and Tofflemire et al. [7].

As the goal of dredging was the removal of contaminants, it was expected that, after some period of time, there would be a noticeable post-dredge decrease in available PCBs downstream. Decreases in PCB concentrations, however, did not occur within the study period. In fact, there was a possible increase in PCB availability. At Chase Lake Road, for example, there was a sig-

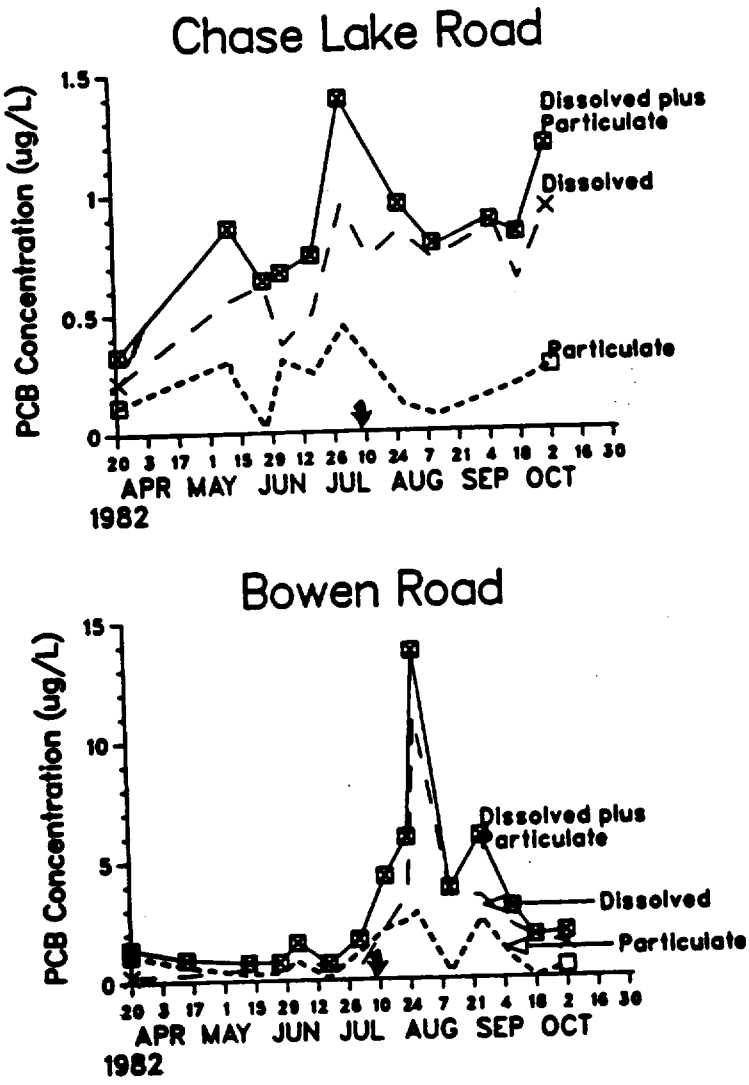

Fig. 5. Changes in concentration of PCBs in water sampled at Bowen Road and Chase Lake Road sites before and during dredging. Arrow indicates start of dredging.

nificant ($\alpha = 0.05$) increase in PCBs accumulated by the caged fatheads (Table 2). Fish at Bowen Road also showed significantly ($\alpha = 0.10$) increased uptake. Furthermore, uptake by clams after dredging was also slightly higher at sites A/B and Chase Lake Road. The obvious conclusion is that dredging appears to have worsened the problem of contamination, at least over the short term.

The different results for PCB availability to clams versus fish during the post-dredge uptake studies may indicate an important distinction between the routes of uptake in these two organisms. The fingernail clams seem to be able to integrate locally occurring PCB concentrations at the sediment-water interface; however, they appear unable to integrate the bulk of the PCBs passing by in the water column. Increased concentrations of PCBs from dredging were measurable in the water column at Chase Lake Road and Marr Road, but data from the clam assays did not reflect any elevated concentrations at these locations (Table 2).

Some unique features of the South Branch of

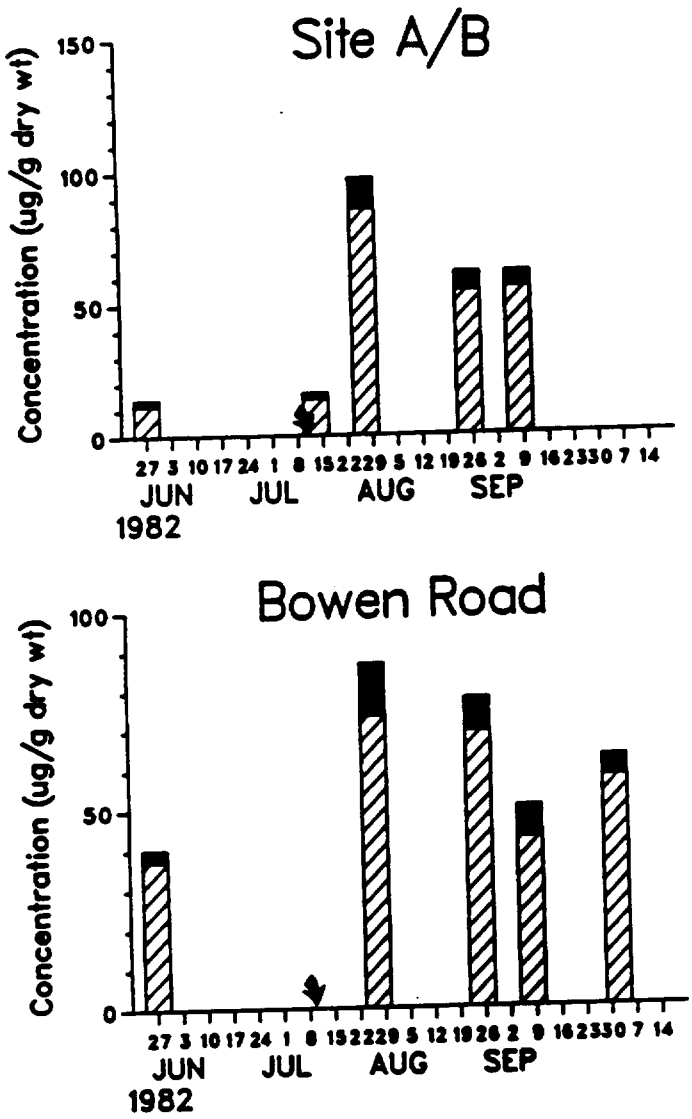

Fig. 6. Concentration of PCBs in clams exposed at site A/B and Bowen Road site before and during dredging. Arrow indicates start of dredging; black area, Aroclor 1254; hatched area, Aroclor 1242.

the Shiawassee River study area should be mentioned, since they bear on the results of this study. Visual inspection showed numerous oily areas along the river bank. Inspection of some trial cores also indicated that much of the PCBs existed in oily deposits layered into various-sized lenses in the sandy substrata of the river. Fine, erodable, distinctly organic silt was often found to occur along with the large PCB deposits. The sediment of the river was essentially lacking in clay. These factors tend to make the PCBs more available than would be the case in well-mixed, sand-silt-clay types of sediment typically found in larger rivers.

Comparisons with other uptake and equilibration studies

Data from fathead minnow studies [23-26] have been widely used to derive constants for uptake of PCBs by fish from water and from food. Halter and Johnson [23] used fathead min-

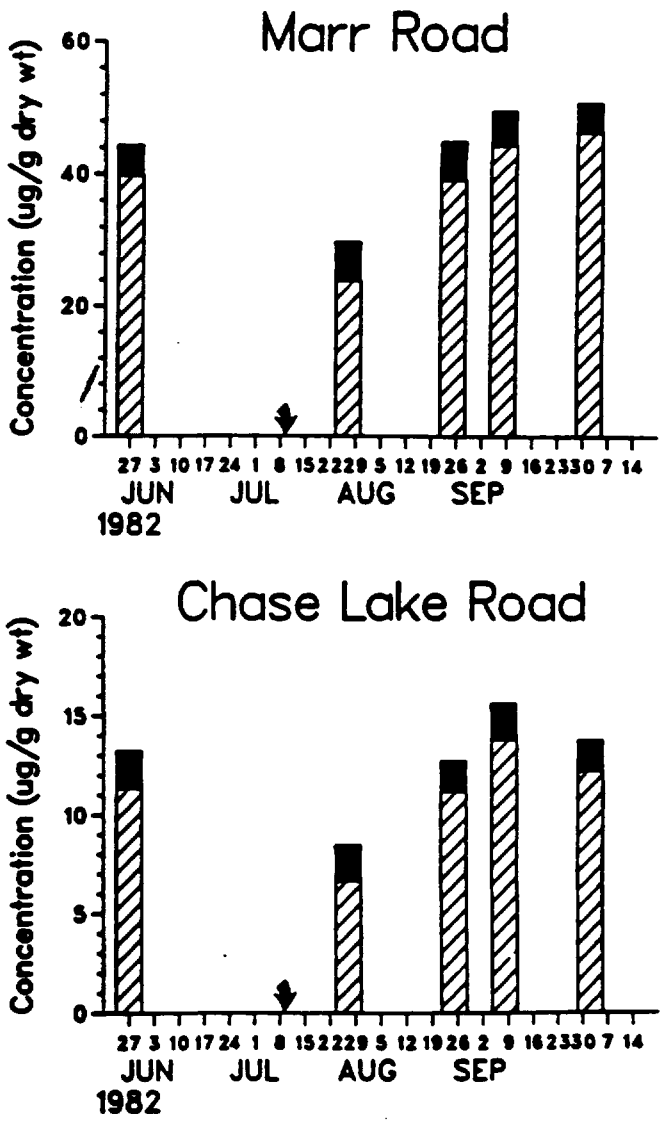

Fig. 7. Concentration of PCBs in clams exposed at Marr Road and Chase Lake Road sites before and during dredging. Arrow indicates start of dredging; black area, Aroclor 1254; hatched area, Aroclor 1242.

nows to measure the availability of PCBs derived from sediment. They reported transfer coefficients (hourly rate of PCB removal from water by fish) based on the homologues of PCBs. The coefficients were derived by dividing the slope for the regression for uptake of each PCB homologue by its concentration in water. Halter and Johnson's [23] laboratory values for uptake were 59.1 h^{-1} and 62.5 h^{-1} for two concentration doses of tetrachlorobiphenyl; our uptake value was 31.07 h^{-1} on the basis of field data (Table 3). For the penta homologue, our transfer coefficient was 72.04 h^{-1}, that of Halter and Johnson [23] ranged from 64.2 h^{-1} to 88.6 h^{-1}.

Another, more often cited technique for measuring biological uptake potential is the bioconcentration factor (BCF). Halter and Johnson [23] did not measure an uptake equilibrium for the fish in

Table 3. Transfer coefficients for uptake of dichloro- to hexachlorobiphenyl homologues by caged fathead minnows at Bowen Road in 1983

Homologue number	Whole water concentration (ng/L)	Regression equation for homologue uptake by fish (uptake units: ng g [wet wt.]$^{-1}$ h^{-1})	r^2	Transfer coefficient[a] (h^{-1})
2	151.8	y = 1.88x + 8.23	0.89	12.42
3	491.1	y = 16.06x + 38.2	0.91	32.70
4	278.3	y = 8.64x + 67.29	0.93	31.07
5	44.7	y = 3.22x + 2.62	0.99	72.04

[a]Equals slope of regression line (ng g^{-1} h^{-1}) divided by water PCB homologue concentration (ng L^{-1}).

their system; therefore, a BCF could not be calculated. Our study was designed to examine equilibrium concentrations of PCBs in both clams and fatheads. By using average values for the fathead equilibrium concentrations, the BCF was calculated as follows:

$$\text{BCF} = \frac{C_f}{C_w}$$

where C_w is the dissolved PCB concentration in the water and C_f is the PCB concentration in the fish. The use of C_w, the dissolved concentration, rather than the total water concentration was suggested by Eaton et al. [27]. For comparison with our BCFs, the BCFs reported by Veith et al. [25] are also listed in Table 4. Because results for Aroclor 1242 were not reported by Veith et al., the log BCFs for Aroclor 1016 and Aroclor 1248, which span the range of chemical and physical properties of Aroclor 1242, are shown. There appears to be relatively good agreement between our field-derived values and those of Veith et al. [25].

Defoe et al. [24] observed that female fathead minnows accumulated approximately twice as much PCB as did their male counterparts. We did not determine the sexes of the fish used in this study, so some of our analytical variation may have been caused by this random selection of fish.

Table 4. Comparison of bioconcentration factors (BCFs) for fathead minnows in this study versus BCFs determined by Veith et al. [21]

Location	$C_{w(\text{dissolved})}$ (μg/cm^3 × 10^{-3})	C_f (μg/g wet wt.)	BCF	Log BCF (this study)	Log BCF [21]
Aroclor 1242					
Control site					
1982	0.0187	0.029	1,551	3.19	4.63–4.85
1983	0.0071	0.008	1,101	3.04	
Bowen Rd.					
1982	0.468	10.53	22,496	4.35	
1983	0.709	13.21	18,632	4.27	
Chase Lake Rd.					
1982	0.431	5.46	12,664	4.10	
1983	0.441	10.91	24,740	4.39	
Aroclor 1254					
Control site					
1982	0.0019	0.32	168,420	5.23	5.00
1983	0.0081	0.29	35,802	4.55	
Bowen Rd.					
1982	0.014	2.20	157,140	5.20	
1983	0.010	3.55	355,210	5.55	
Chase Lake Rd.					
1982	0.026	0.87	33,462	4.52	
1983	0.007	1.99	284,290	5.45	

$C_{w(\text{dissolved})}$, concentration in water; C_f, concentration in fish. BCF = $C_f/C_{w(\text{dissolved})}$.

Table 5. PCB concentration factors (BCFs) for clams placed in cages in the South Branch of the Shiawassee River, before (1982) and after (1983) dredging

Location	$C_{w(dissolved)}$ ($\mu g/cm^3 \times 10^{-3}$)	C_f ($\mu g/g$ wet wt.)	BCF	Log BCF
Aroclor 1242				
Control site				
1982	0.0187	0.210	11,230	4.05
1983	0.0071	0.014	1,972	3.29
Bowen Rd.				
1982	0.4680	2.700	5,769	3.76
1983	0.7090	0.317	447	2.65
Chase Lake Rd.				
1982	0.4310	0.190	441	2.64
1983	0.4410	0.945	2,143	3.33
Aroclor 1254				
Control site				
1982	0.0019	0.060	31,579	4.50
1983	0.0081	0.046	5,679	3.75
Bowen Rd.				
1982	0.0140	0.205	14,643	4.17
1983	0.0100	0.069	6,900	3.84
Chase Lake Rd.				
1982	0.0260	0.032	1,231	3.09
1983	0.0070	0.293	41,857	4.62

$C_{w(dissolved)}$, concentration in water; C_f, concentration in clams. BCF = $C_f/C_{w(dissolved)}$.

BCFs were calculated for the clams (Table 5). As before, only the dissolved fraction was used for the water concentration. There was much more variability in the values for clam tissue than for fish tissue. Excluding the control and Bowen Road sites, the average BCF for Aroclor 1242 was 2,784 ± 2,721. This is lower than the estimated 7,500× factor derived by Werner [10] for Aroclor 1016 accumulation in a large freshwater mussel, *Ellipto complanatus*. Risebrough et al. [28] reported BCFs for Aroclor 1254 of from 69,000 to 690,000 for *Mytelus* spp. from various marine locations. Their values are also higher than the Aroclor 1254 concentration factors we found (Table 4). Hartley and Johnston [29] reported some BCFs for pesticides in the sphaerium-sized Asiatic clam, *Corbicula manilensis* (*C. fluminea*), in an Illinois stream. Their BCFs, normalized for lipid content, ranged from 2,300 to 13,390. BCFs normalized for lipids in our study ranged from 100,000 to 3,270,000, values considerably higher than those of Hartley and Johnson [25].

McFarland et al. [30] examined the uptake of dichlorobiphenyl and trichlorobiphenyl by *C. fluminea* from Hudson River sediment in laboratory flow-through chambers. The log BCFs for dichlorobiphenyl varied from 3.39 to 3.75 for six separate sediment tests and from 3.81 to 4.10 for trichlorobiphenyl with the same sediments. Similarly calculated values from our study were 3.20 for log BCF for dichlorobiphenyl and 3.04 for log BCF for trichlorobiphenyl at site A/B.

Equilibrium was attained after about 14 d in clams exposed to Aroclor 1242 in our experiments. However, equilibrium was not achieved with Aroclor 1254 even after 44 d (Fig. 6). The Aroclor 1242 data are similar to those reported by Bedford et al. [31] and Curry [32], who noted that equilibrium was reached in 2 to 4 weeks for a variety of chlorinated pesticides, including PCBs, in several species of unionid mussels. In the study by McFarland et al. [30], equilibrium of dichlorobiphenyl uptake was reached after 15 d in *C. fluminea* and was approached on the last day of uptake (day 28) for trichlorobiphenyl. Hartley and Johnston [29] studied the rates of uptake of some organochlorine pesticides by *C. fluminea* and found that equilibrium appeared to occur after 10 to 20 d for heptachlor and after 10 to 40 d for lindane (the greater range for lindane might have been due simply to the lack of a sample at 20 d). Vreeland [33] found that the concentration of six individual PCBs, applied in water solutions to oysters (*Crassostrea virginica*), reached equilibrium in

about 28 d and then remained constant for the following 65 d. Lowe et al. [34], however, found that equilibrium was never reached in oysters exposed to 5 ppb Aroclor 1254 for 24 weeks (168 d). Parrish et al. [35] found that it took 8 weeks (56 d) for equilibrium to occur when oysters were exposed to 10 ppt Aroclor 1254. Thus, our data for uptake and equilibrium by *Sphaerium striatinum* seem quite reasonable when compared with the results of most studies using bivalves of similar size, particularly *Corbicula*, and indicate the usefulness of the fingernail clam as a biomonitor.

SUMMARY AND CONCLUSIONS

Among water, clams and fish, there was no one ideal monitor for the true bioavailability of PCBs in the South Branch of the Shiawassee River. Grab-sample monitoring of water appeared to be too episodic to detect small differences in exposure concentrations for organisms in the stream. Exposure uptake by fathead minnows appeared to reflect water column concentrations moving downstream. The fish were very sensitive indicators of changes in availability of the PCBs even more than 6 miles downstream from the dredging site. Exposure uptake by fingernail clams (*Sphaerium striatinum*) appeared to reflect local conditions at the sediment-water interface, probably influenced by near-surface sediment and detritus, but was not a sensitive indicator of conditions more than 1 mi downstream.

Uptake rates and BCFs derived from field experiments with fathead minnows and fingernail clams were similar to constants derived during laboratory studies.

Dredging of PCBs from deposits that are poorly integrated into sand/silt-bottomed streams may, in fact, increase bioavailability of PCBs to stream organisms, at least over the short term (0 to 6 months following dredging).

Acknowledgement—We are indebted to Ms. Diane Nelson and Mr. Paul Shulec for all their efforts and interest in collection of field and laboratory data, often under unfavorable environmental conditions. We acknowledge the assistance of Ms. Patty Arscot for sample preparation and analysis and Dr. Mila Simmons for input on experimental and sampling design. This study was made possible through a contract from the Michigan Department of Natural Resources.

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Information Circular 9008

Analysis of Dredge Safety Hazards

By Stephen A. Swan

UNITED STATES DEPARTMENT OF THE INTERIOR
Donald Paul Hodel, Secretary

BUREAU OF MINES
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TABLES

UNIT OF MEASURE ABBREVIATIONS USED IN THIS REPORT

ft	foot	yr	year
pct	percent		

ANALYSIS OF DREDGE SAFETY HAZARDS

By Stephen A. Swan [1]

ABSTRACT

Bureau of Mines research has not included analyses of mining dredge safety since 1948. Because of the increasing use of dredges, 63 fatal accidents and several hundred nonfatal injury accidents involving dredges were examined. Also, 31 working dredges and 3 dredge manufacturers were visited to acquire qualitative information.

Drowning accidents represented 59 pct of the fatalities and are delineated in detail. Also, countermeasures against other hazards are discussed.

[1]Mining engineer, Twin Cities Research Center, Bureau of Mines, Minneapolis, MN.

INTRODUCTION

The Bureau of Mines concern about mining dredge safety is longstanding. S. B. Ash studied safety practices related to California gold in the early 1930's.[2] In the 1940's, R. W. Fatzinger visited hydraulic and dredge mining operations in Alaska and California. He also reviewed 10 yr of accident data pertinent to such mining in the United States. The data, covering the period 1933 to 1942, included those related to 36 fatalities and 3,700 injuries in dredge mining.[3]

It is generally assumed that dredge operations are "safe"--certainly much safer than other mining and construction operations to which they are usually compared. It is true that personal injury rates for dredge workers, normalized in terms of worker-hours, dredge operating hours, or tons of material moved, are lower than those for many other mining operations.[4] Still, over the 10-1/2-yr period that ended in June 1983 63 people were killed and almost a thousand injured in mining dredge operations in the United States.

The research discussed here included field visits to a selected sample of mining dredge operations, an examination of dredge-related injury data in the files of the Mine Safety and Health Administration's Health and Safety Analysis Center in Denver, and field visits to a selected number of nonmining dredge operations. In all, field visits were made to 31 dredge operations. Three companies whose principal business is the manufacture of dredges and dredge components were also visited. Design considerations for safety were discussed, as were training and product liability questions and the accident experience of company field representatives during erection, initial operations, and maintenance of delivered dredges.

The information was analyzed in five separate categories which appear to be the most important in terms of developing countermeasures for these accidents. These categories include drowning, slips and falls, electrical, mechanical, and fire.

DREDGE FATALITIES

To try to quantify the hazards, data were taken from the Mine Safety and Health Administration's accident records since January 1973. Table 1 shows the fatal accidents that occurred on U.S. mining dredges during those years. Six people (average) died each year. There was also an annual average of more than 95 nonfatal injury accidents, as well as several hundred noninjury accidents. Note that about 59 pct of the fatalities in the table are drowning accidents. There were more drownings from boats and walkways (mostly pipeline walkways) than from the dredges themselves. Machinery accounted for a smaller number. An example of such an accident is one in which a worker was repairing a dredge-to-shore conveyor belt. The belt was started while he was working on it, and he was knocked into the water. Five of the fatal accidents were from electrocutions. Twenty-one fatalities involved shore- or dredge-based support equipment, including a rollover of a front-end loader, a falling crane boom, a fall into a hopper, and conveyor accidents on land.

[2]Ash, S. H. Safety Practices in California Gold Dredging. BuMines B 352, 1932, 31 pp.

[3]Fatzinger, R. W. Safety Practices in Dredging and Hydraulic Mining. BuMines B 450, 1948, 76 pp.

[4]Mine Safety and Health Administration. Annual and Quarterly Injury Reports.

DREDGE OPERATIONS

The majority of the dredges visited were extracting sand or gravel. Others in the sample included units engaged in recovering phosphate, gold, ilmenite, and coal. Dredges involved in land reclamation after mining and settling pond maintenance--for example, in recreational area development, specifically units engaged in making lakes for fishing and water sports--were also included in the analysis.

Of the 31 dredges visited, 18 were built by manufacturing firms whose principal business is the design and manufacture of dredges. Thirteen units were built by the owners or by local machine shops not primarily engaged in dredge design and construction. The 31 dredge operations included in this analysis were located in 10 States (table 2). Three of the four Alaskan dredges were mining gold.

TABLE 1. - Mining dredge fatalities, January 1973-June 1983

	Total fatalities
Drowning:	
Boat	11
Dredge	10
Walkway	6
Machinery	5
Other	5
Total	37
Electrocution (shore or dredge)	5
Mobile machinery (shore)	8
Fixed machinery (shore or dredge)	13
Total	63

TABLE 2. - Location of dredges included in hazard analysis

	Sites visited
Alaska	4
Colorado	1
Florida	6
Illinois	1
Nebraska	4
New Jersey	4
North Carolina	2
Ohio	5
Pennsylvania	3
Wyoming	1
Total	31

GENERAL HAZARDS ANALYSIS

Many dredges depend upon a workboat to transport personnel and materials between the dredge and shore (fig. 1). It is important to note that most of the dredge operations that depend upon a boat had only one such boat. Several fatal accidents might have been avoided had a second boat been immediately available to enable persons who might have rendered aid to get to the dredge quickly. In these accidents the only available boat was tied up at the dredge, and the victim was the only person on the dredge.

Some dredges have workboats but do not depend upon them to transport people between dredge and shore as a normal practice. The crew moves between the dredge and shore by walking the pipeline. Figure 2 is an example of such a pipeline. Several dredgemasters reported that they had "knocked a guy off the pipeline" when they started the pumps. If access between the shore and the dredge depends primarily upon walking the pipeline, it follows that a safe walkway must be provided. Figure 3 shows a safe walkway separated from the pipeline. Infrequently, however, the design of the walkway is such that there are large gaps between one section and another (fig. 4). In adverse weather negotiating a gap of this size can be particularly difficult and hazardous undertaking for the dredge worker.

FIGURE 1. - Single workboat.

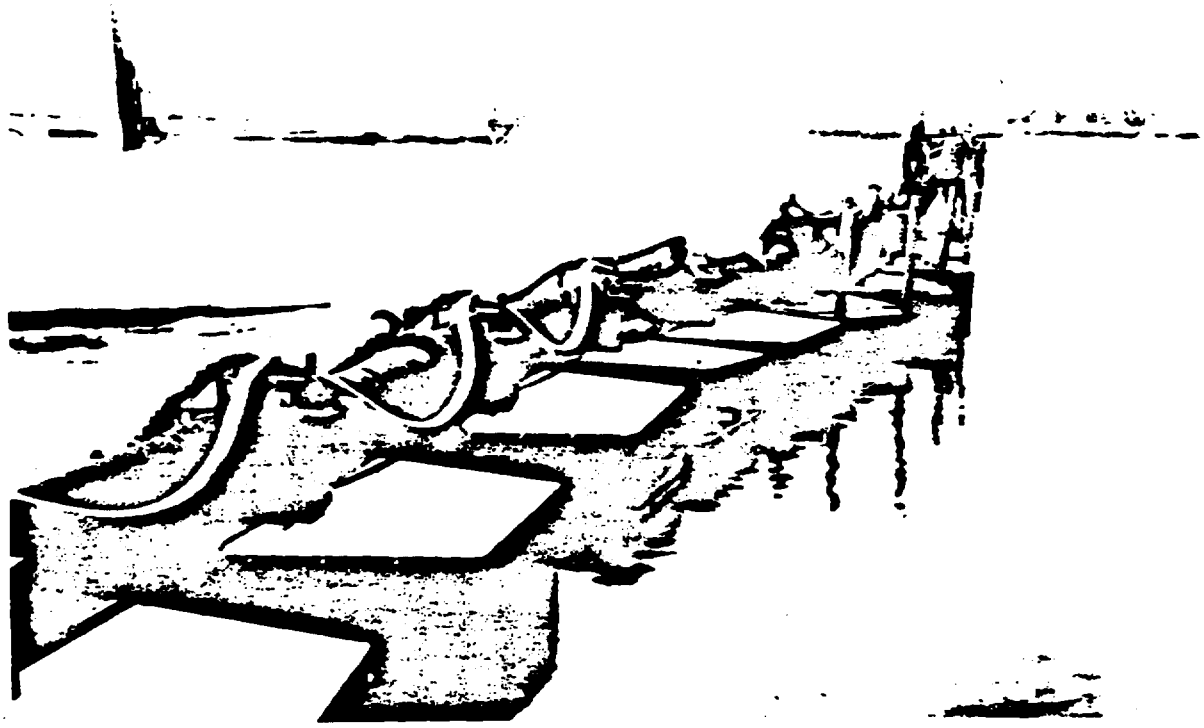

FIGURE 2. - Pipeline without walkway.

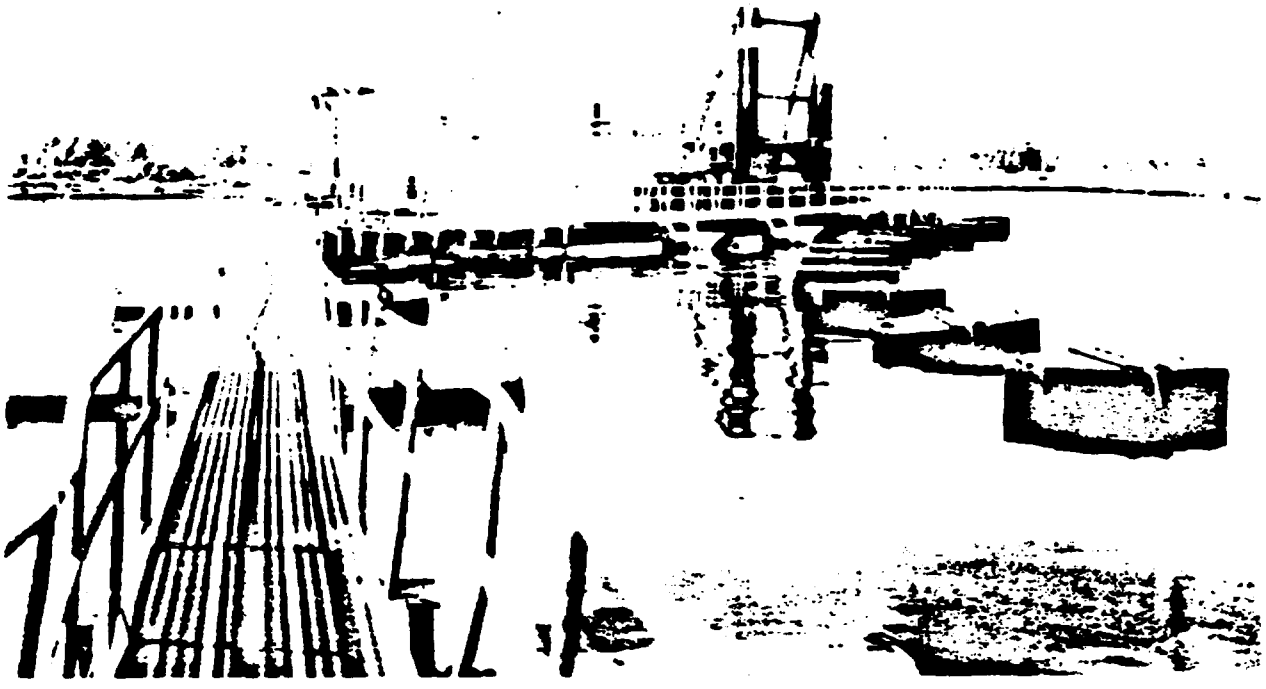

FIGURE 3. - Safe walkway.

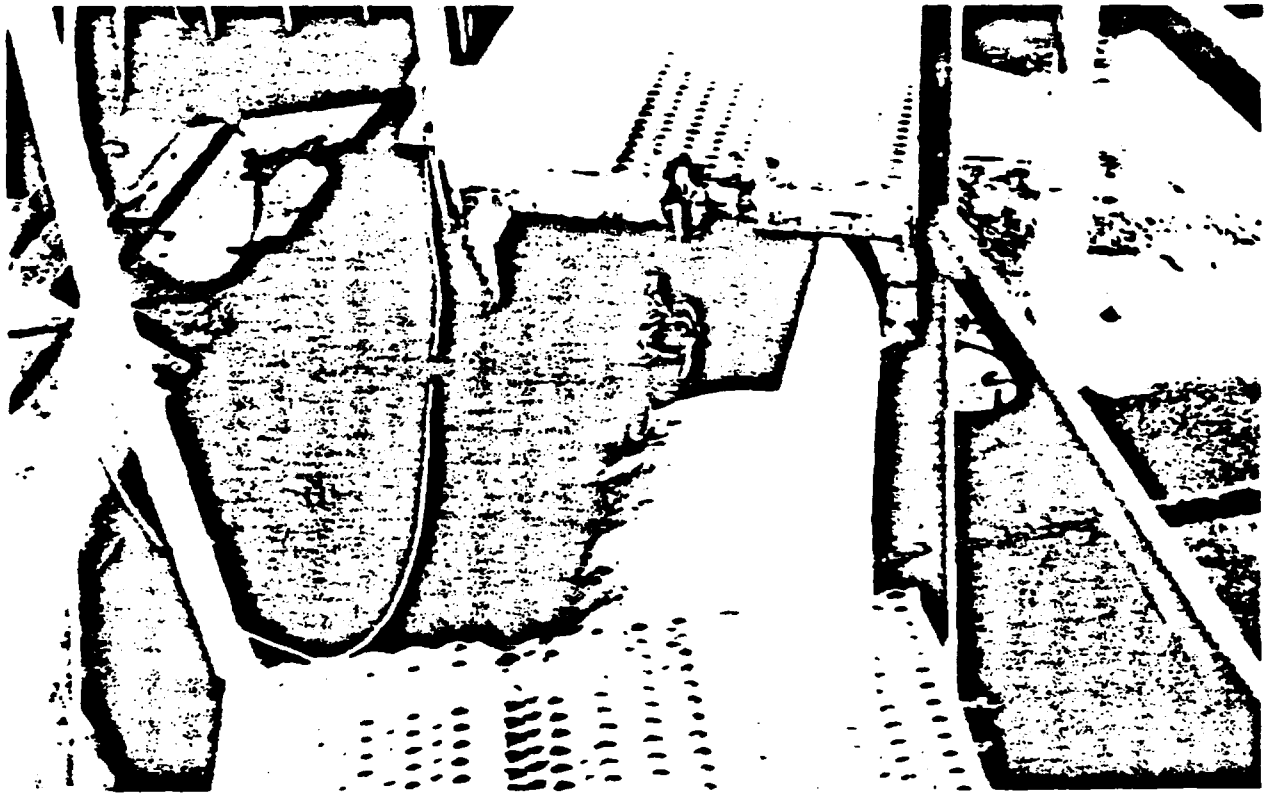

FIGURE 4. - Walkway safety hazard.

PIPELINE HAZARDS

A more specific hazard associated with pipelines is related to moving the line and making repairs where the job of making connections between sections involves the danger of crushing or pinching injuries. Although several dredges provided work platforms on the pipeline buoyancy structures, many do all the work from workboats, sometimes using people who have little skill in boat operation or safe mooring.

Another hazard area related to the pipeline involves power cables carried along the pipeline. One obviously unsatisfactory arrangement is shown in figure 5. If the insulators that carry the line are inadequate, or if the line droops into the water, chances of breaking or electrical shorts are greatly increased

RAILING HAZARDS

As on the pipeline walkways, railings make an important safety contribution on dredge decks. Many dredges had strong, well-maintained deck railings (fig. 6). Some, however, had no railings at all (fig. 7). A need for guardrails inboard was evident on the type of dredge that mounts a dragline in a deck well. Two of the dredges visited had arrangements similar to that show in figure 8. Note that as the dragline swings, a pinch point is created between the cab and the deck. Several accidents have occurred when maintenance people were not aware of this

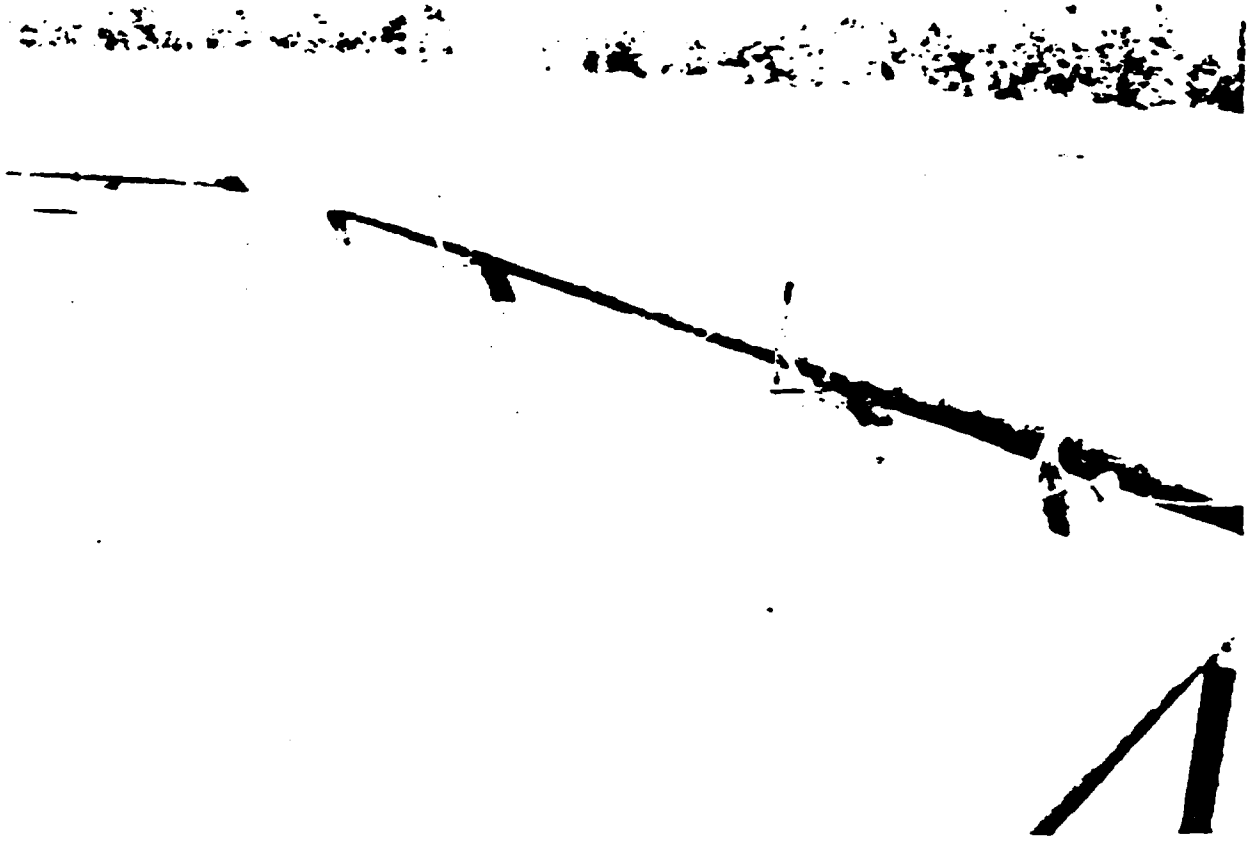

FIGURE 5. - Power cable along pipeline.

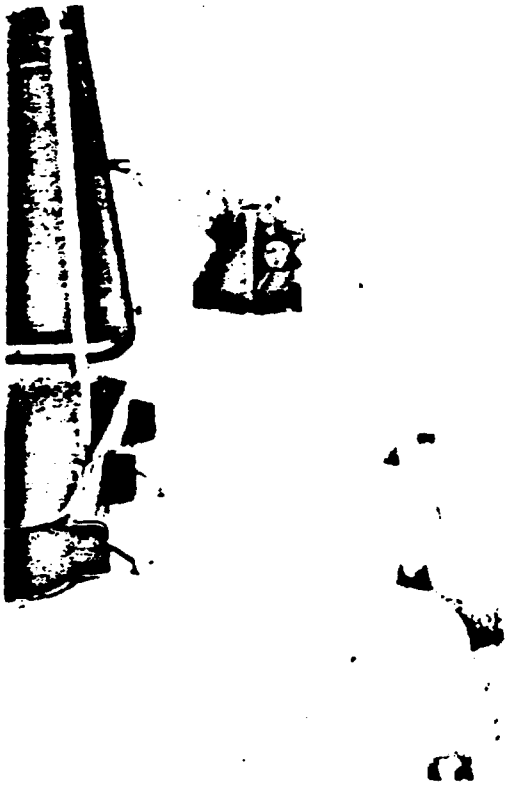

FIGURE 6. - Well-maintained railing.

FIGURE 7. - No railings.

potential hazard. The problem is not one of having maintenance done during dragline operations, because most companies require that the dragline be stopped during maintenance. The accidents that have occurred involved people doing deckwork and inadvertently stepping into the well.

LADDER HAZARDS

Figure 9 is an example of a ladder (or stairway) into a lower hull area of an otherwise well-constructed dredge. Note the absence of handrails. Tripping and bumping hazards are also visible in the stairwell area.

Ladders for abovedeck maintenance are frequently designed with little thought about the safety of their use. Figure 10 shows such a ladder, made of round stock welded to an I-beam. The spacing is inappropriate, the toe room is inadequate, and the footing is poor. Often it was not a cost problem that led to poor ladders, but simply lack of attention to the details of safety design. Figure 11 shows another ladder welded to an A-frame. The rungs are not evenly placed, and the ladder is quite difficult to negotiate. Some of the lower rungs have been removed because they constituted another hazard to people working near the base of the A-frame.

FIGURE 8. - Dragline pinch point.

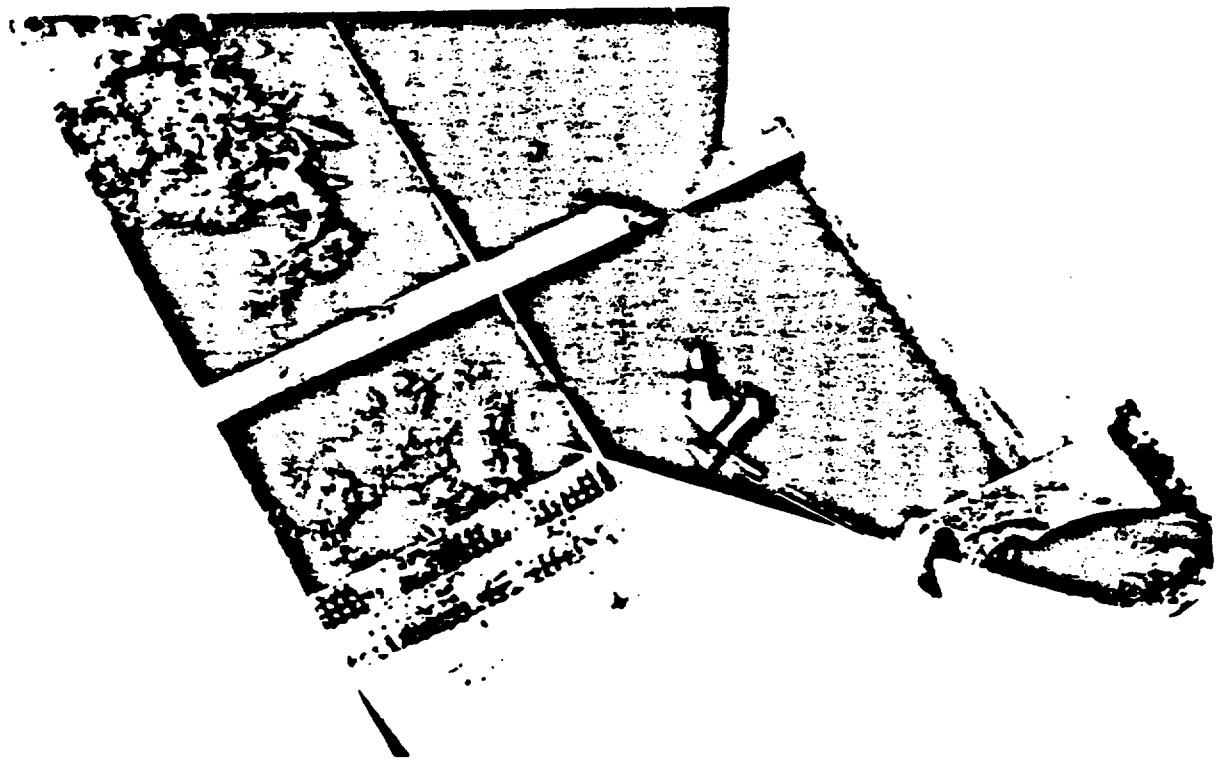

FIGURE 9. - Ladder or stairway.

FIGURE 10. - Poorly constructed ladder.

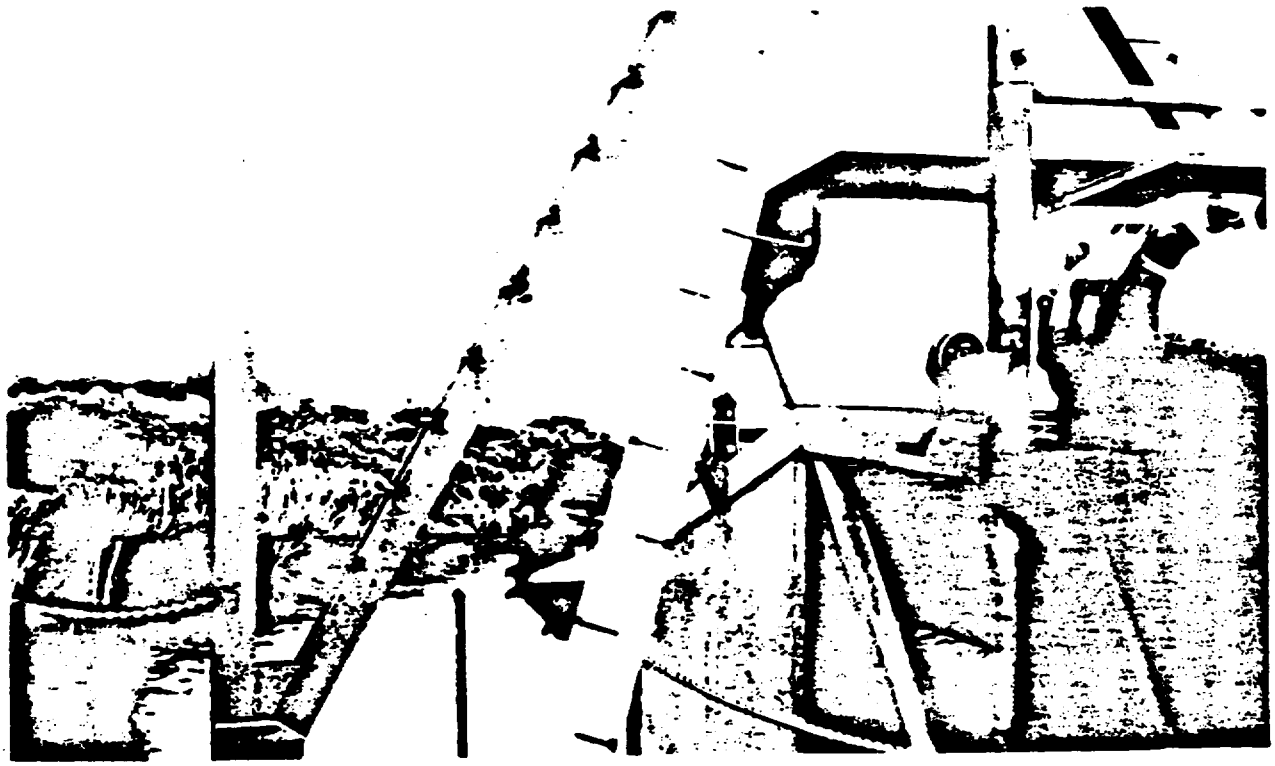

FIGURE 11. - Unsafe ladder.

ELECTRICAL HAZARDS

Electrical hazards are common on many smaller dredges. Figure 12 shows poor handling of a power cable. Even on larger dredges, electrical safety is sometimes given inadequate attention. Distribution boxes are poorly placed and improperly mounted. Cables are routed with little regard for insulation protection, not to mention the trippling hazards created. Common safety practice must be followed.

WIRE ROPE HAZARDS

Wire rope breakage on dredges is not uncommon. Both broken positioning lines and broken digging ladder control lines were observed during field visits. On one occasion, the end of a snapped rope violently struck and entered the operator's compartment. It would undoubtedly have injured the dredgemaster had he been in his usual position at the time. There are several countermeasures. One is the proper handling of the equipment so that the rope is not overloaded. Another is regular inspection and replacement of work or damaged rope. A third is more careful design to ensure that the rope size is appropriate to the loads experienced during the majority of the operations. Finally, proper guarding of the cables and drums can be invaluable to ensure that a broken cable does not injure

FIGURE 12. - Power cable.

a worker. Although some of the ropes and sheaves observed during the study were well guarded, most were not. Many drums, including drums immediately adjacent to the operator's compartment, were not guarded.

FIRE HAZARDS

Fires on dredges do not appear to be a common occurrence, but when one did occur, the results were frequently extremely expensive and dangerous. In most of the operations visited, the fire protection practices appeared to be substantially less well developed than those in shore-based plants. Scenes like that shown in figure 13 were very common. Flammable debris (oily rags, cartons, and fluids) were frequently observed in areas where ignition was possible. The training given to most dredge workers relative to fire prevention and fire suppression was minimal. The available fire suppression equipment was not always easily accessible and properly maintained.

PERSONAL FLOTATION DEVICE

Perhaps the single greatest hazard on dredges is related to the simple fact that work is done over water. Only one of the 31 dredges vistied required water survival training and demonstrated water survival proficiency as conditions of employment. All except two of the dredge operations required the wearing of a personal flotation device (PFD), according to the written safety rules of each company. However, in only a few cases was the wearing of PFD's observed to be enforced.

In the majority of operations visited, no one was wearing his PFD. Scenes like that depicted in figure 14 were common. On some of the very smallest and oldest dredges it was apparent that the available PFD's were never worn. On some of the largest it was not uncommon to find PFD's stored as in figure 15, not well-maintained, not inspected, not fitted, and not assigned to individuals. Sometimes the PFD's, even new ones, were lying in dirt and grease as shown in figure 16.

A few of the dredges had the PFD's assigned to individuals; these PFD's were well maintained and regularly inspected and were worn while working over water at all times except when the person was inside the dredge control house. However, at most operations the dredge workers complained that the PFD was uncomfortable, too old to be of any use, or a work hazard because it tended to catch on things. In short, a multitude of reasons were offered in defense of not wearing a PFD.

FIGURE 13. - Fire hazard.

FIGURE 14. - Failure to wear personal flotation devices.

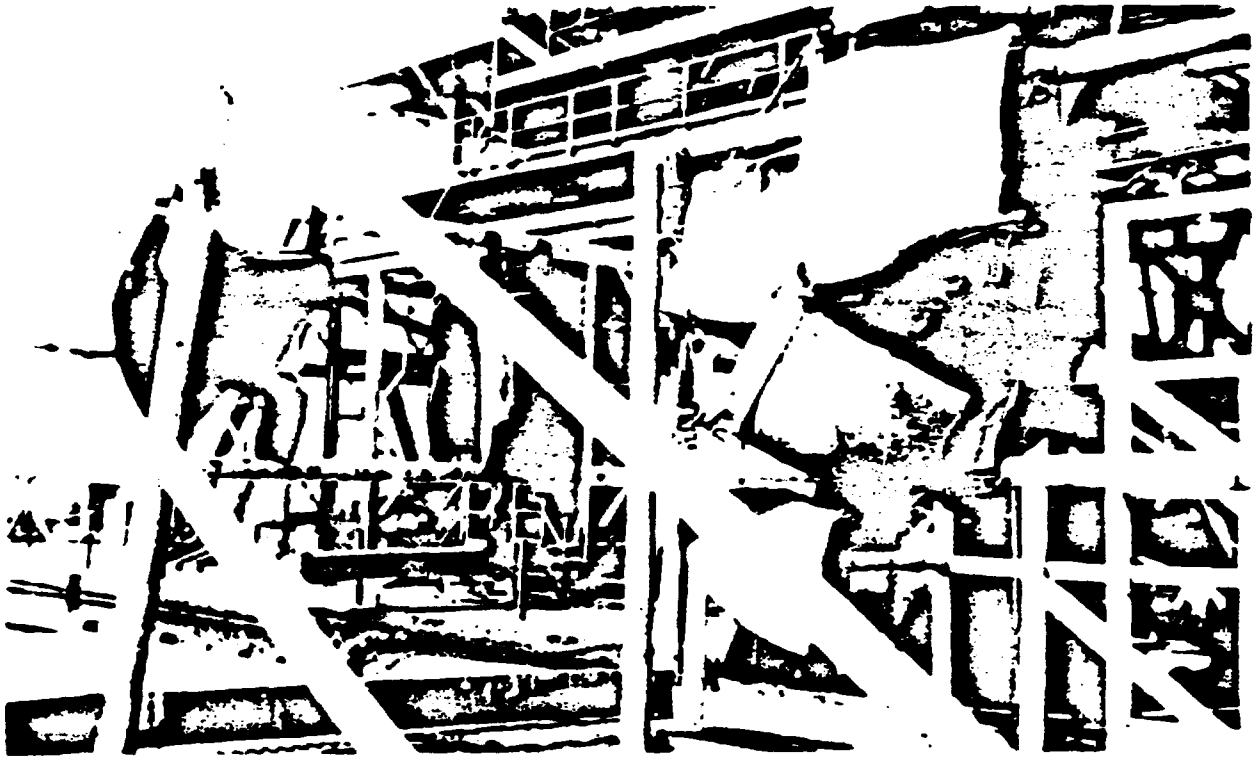

FIGURE 15. - Improper storage of personal flotation devices.

FIGURE 16. - Abuse of personal flotation devices.

CONCLUSIONS

More attention needs to be given to published safety requirements, including those in American National Standards Institute Standard A10.15 and in the Code of Federal Regulations. The three dredge manufacturers visited appeared to have studied and considered the standards in terms of design improvements. Beyond that, all three expressed the general view that safety is the responsibility of the operator, not the manufacturer. Considerable interest was expressed in obtaining information about effective safety practices, including the development of safe job procedures, at many of the operations visited.

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H. REFERENCE MATERIALS FOR SECTION 5.0

1. Figures for Section 5.2

a.	Figure 5.2.2-1	Dechlorination of PCBs in the Upper Hudson River. Open circles represent samples with the ratio of peak 70 to peak 47 less than or equal to 0.5. Crosses are sample location.
b.	Figure 5.2.3-1 (A-C)	Capillary column GC chromatograms from sediment samples from site H7 in the Upper Hudson River. Figure A to C represent samples obtained in progressively later years.
c.	Figure 5.2.3-2	Average PCB concentrations in sediment samples from the H7 site in the Upper Hudson River. Based on data obtained in 1990.
d.	Figure 5.2.3-3	Average number of chlorine atoms per biphenyl molecule in sediment samples from the H7 site in the Upper Hudson River. Based on samples obtained in 1990.

2. Tables for Section 5.2

a.	Table 5.2.2-1	Dechlorination status of PCBs for different concentration ranges on the Upper Hudson River. Based on an analysis of chromatographic peaks (70 and 47) from the New York 1984 data base.

b.	Table 5.2.2-2	Percent of samples from the Upper Hudson River showing significant declorination as a function of concentration. Based on an analysis of chromatographic peaks (70 and 47) from the New York 1984 data base.
c.	Table 5.2.3-1	Average number of chlorine atoms per biphenyl molecule in selected "Hot Spots" sediment samples in the Upper Hudson River. Based on data obtained during 1990.

3. Abramowicz, D.A. (1990) Aerobic and Anaerobic Biodegradation of PCBS: A Review. V. 10 I.3, pp. 241-251 Critical Reviews in Biotechnology.

4. Bedard, D.L. (1990) Bacterial Transformation of Polychlorinated Biphenyls. V. 4 Biotechnology and Biodegradation.

5. General Electric Company Corporate Research and Development (1990) Research and Development Program for the Destruction of PCBS: Ninth Progress Report

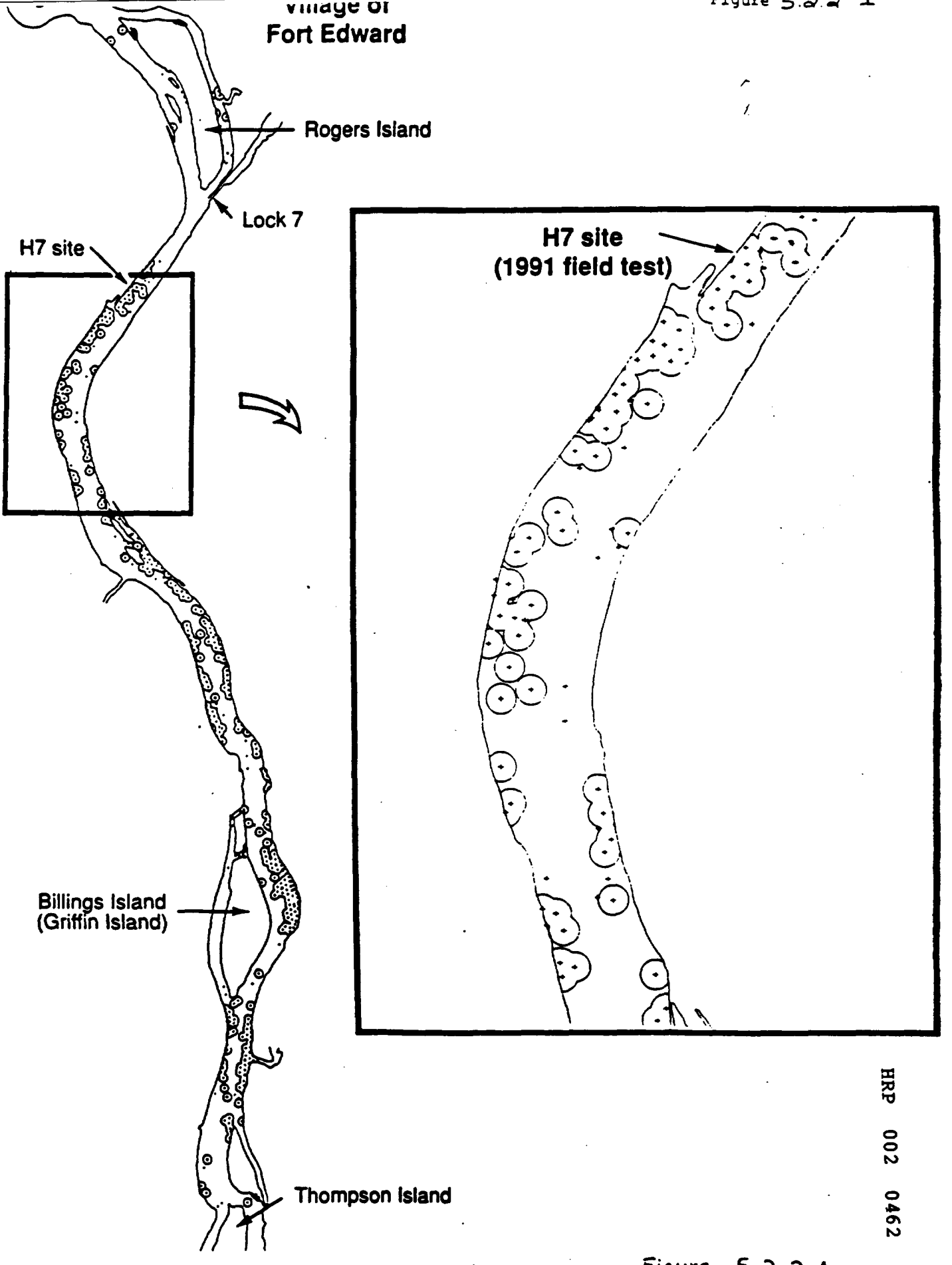
Village of
Fort Edward
Rogers Island
Lock 7
H7 site
Billings Island
(Griffin Island)
Thompson Island
H7 site
(1991 field test)

Figure 5.2.2-1

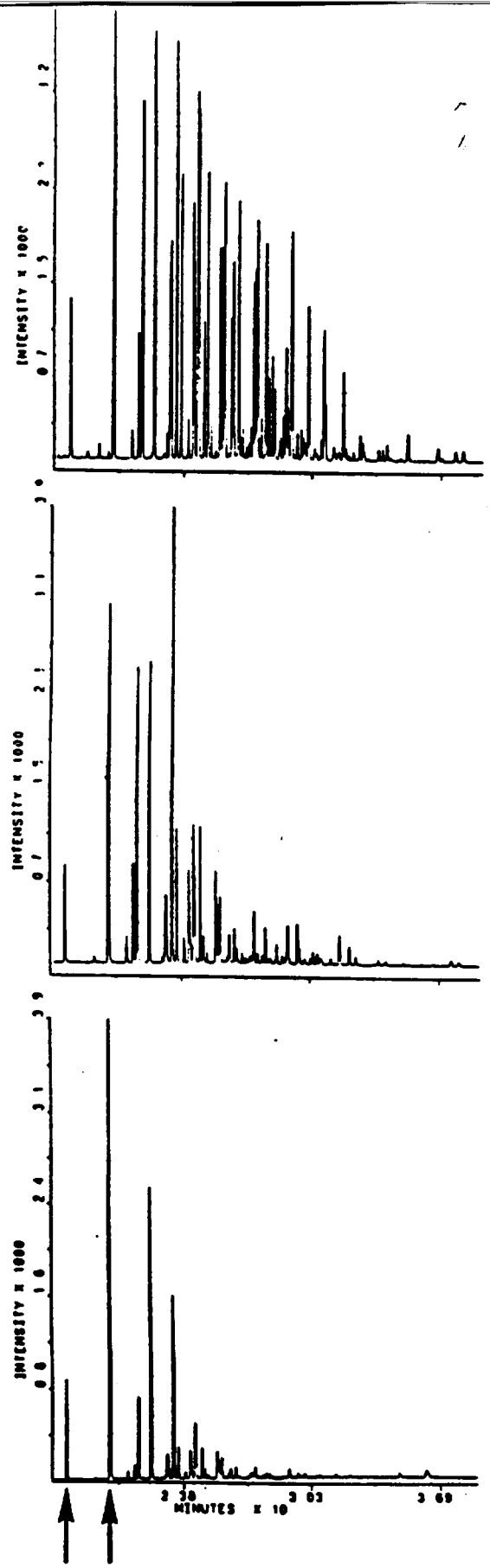

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Figure 5.2.3-1

12' x 12' Sampling

CORE DEPTH (IN.)
- 0
- 2
- 4
- 6
- 8
- 10
- 12
- 14

H-7 SITE CORING GRID

ppm Total PCBs

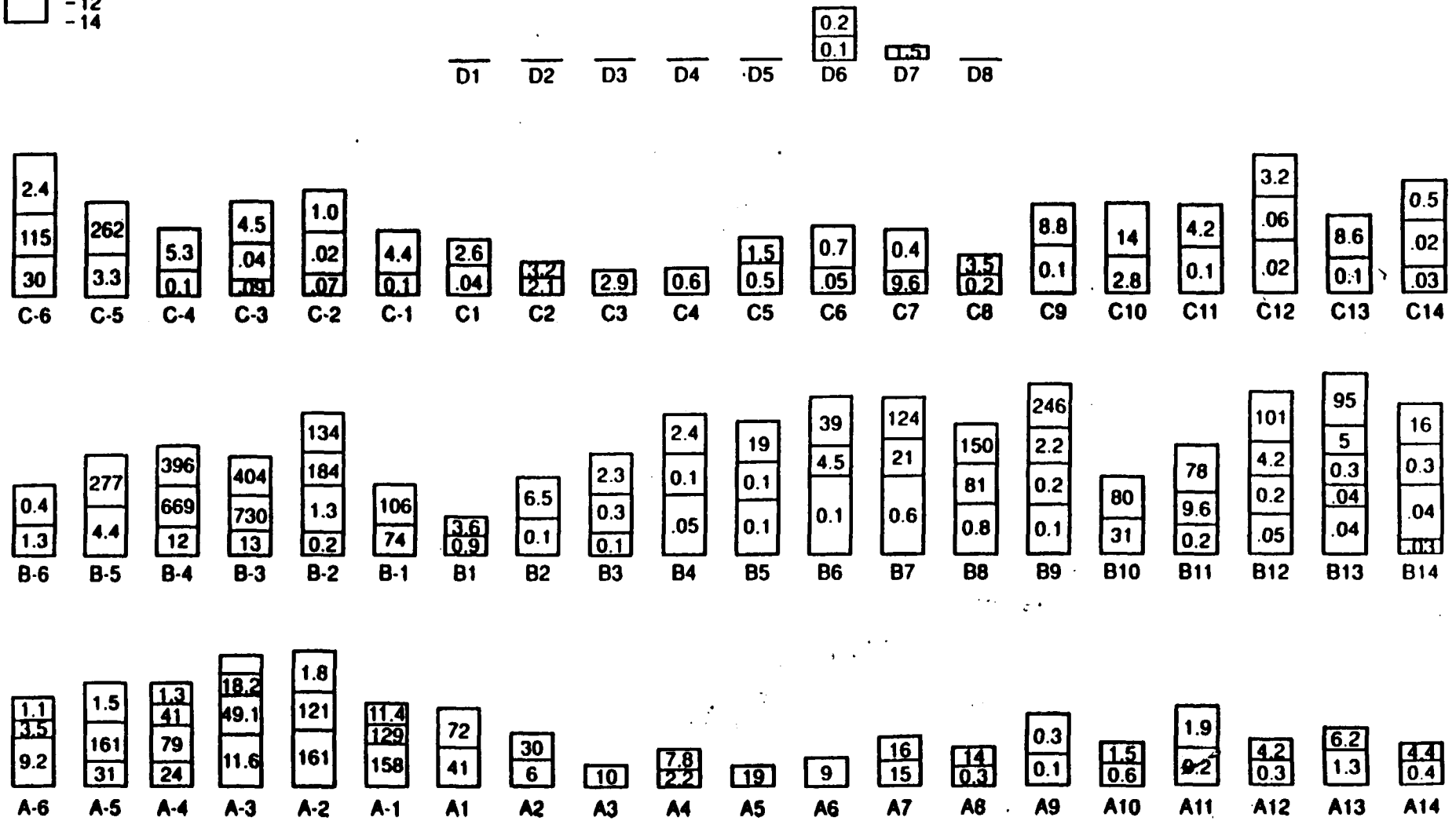

Figure 5.e.3-2

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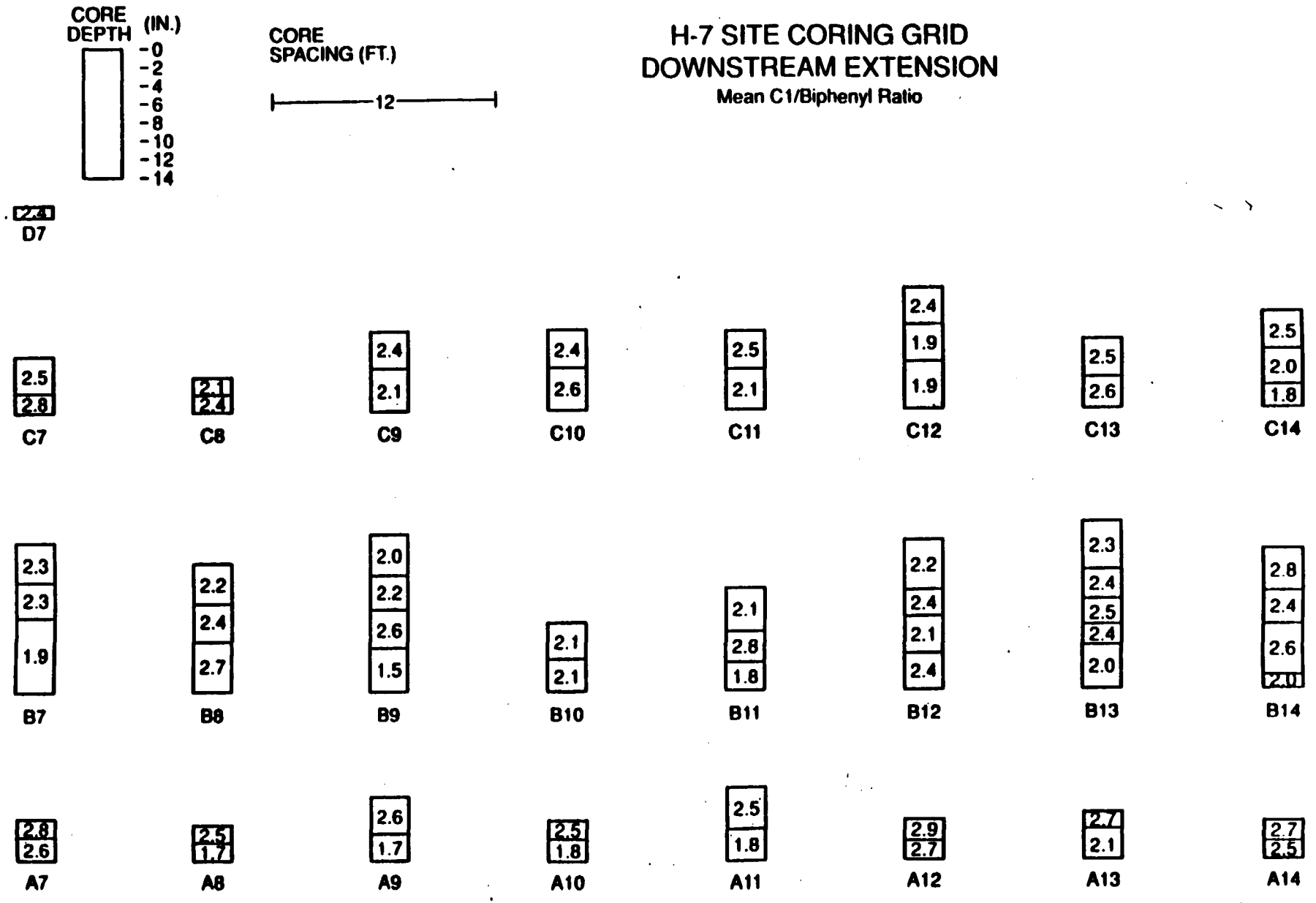


Figure 5.0.3-3

Dechlorination in Hudson River (1984 DEC data)

[PCB]	#Samples $0 < \frac{\text{Peak 70}}{\text{Peak 47}} \leq 1$	Avg [PCB]	$\frac{\text{Peak 70}}{\text{Peak 47}}$ (Avg)
≥ 5	447	83	0.49
≥ 10	384	95	0.48
≥ 20	314	114	0.44
≥ 50	190	167	0.40
≥ 100	111	232	0.36

Table 5.2.2-1

Dechlorination in Hudson River (1984 DEC data)

PCB Range	# Samples	# Samples $0 < \frac{\text{Peak 70}}{\text{Peak 47}} \leq 1$	%
5-10 ppm	100	63	63%
10-20 ppm	108	70	65%
20-50 ppm	179	124	69%
50-100 ppm	92	79	86%
>100 ppm	119	111	93%

Table 5.2.2-1

Average Cl/biphenyl (1990 Survey)

Site #	Location	Sample 1	Sample 2	Sample 3
1	Bakers Falls	--	--	--
2	Near Remnant 1	--	--	--
3	Near Remnant 4	--	--	--
4	Hotspot 6	2.3	2.4	2.4
5	Hotspot 14	2.2	2.2	2.3
6	Hotspot 16	2.1	2.3	2.3
7	Hotspot 18	2.4	2.3	2.4
8	Hotspot 19	2.4	2.3	--
9	Hotspot 28	2.8	3.1	2.9
10	Hotspot 31	3.0	3.1	3.2
11	Hotspot 36	3.3	3.5	3.3
12	Hotspot 39	2.3	2.7	3.2
13	Hotspot 40	2.7	2.8	2.8
14	Rogers Island	2.8	--	--
15	Hotspot 5	--	--	--
16	Hotspot 8	--	--	--
17	Hotspot 9	2.4	--	--
18	Griffin Island	2.8	--	--

Aerobic and Anaerobic Biodegradation of PCBs: A Review

Daniel A. Abramowicz

ABSTRACT

This review summarizes recent research results on the biodegradation of polychlorinated biphenyls (PCBs). These compounds, commonly believed to be indestructible, have repeatedly been shown to biodegrade under a variety of conditions. Two distinct classes of bacteria have now been identified that biodegrade PCBs by different mechanisms. The focus of this manuscript is current research involving the aerobic biodegradation of PCBs (natural strains, recombinant organisms, and soil applications) and the dramatic new results demonstrating microbial reductive dechlorination of even highly chlorinated PCBs under anaerobic conditions.

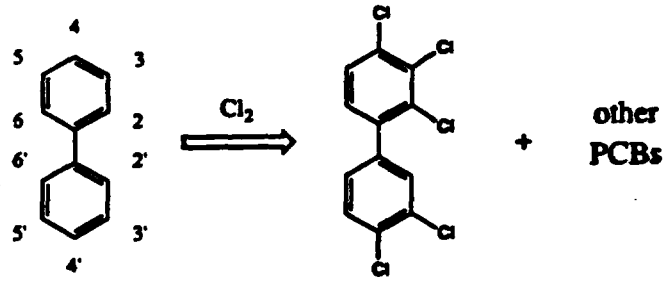
These two PCB-degradative systems include aerobic bacteria which live in oxygenated environments and anaerobic bacteria which live in oxygen free environments such as aquatic sediments. The aerobes attack PCBs oxidatively, breaking open the carbon ring and destroying the compounds. Anaerobes, on the other hand, leave the biphenyl rings intact while removing the chlorines. This anaerobic dechlorination degrades highly chlorinated compounds into less chlorinated derivatives. These two naturally occurring processes are complementary, and a two step treatment may permit the biological destruction of nearly all of the PCB mixtures commonly used.

I. INTRODUCTION

A. Definition

Polychlorinated biphenyls (PCBs) are a family of compounds produced commercially by the direct chlorination of biphenyl using ferric chloride and/or iodine as the catalyst.[81] The biphenyl molecule is made up of two connected rings of six carbon atoms each (see Figure 1), and a PCB is any molecule having multiple chlorines attached to the biphenyl nucleus. Chlorines can be placed at any or all of the ten available sites, with 209 different PCB compounds theoretically possible, varying in the number and position of the attached chlorines. The individual isomers and homologs are generically referred to as congeners. Of the 209 possible congeners, only about half are actually produced in the synthesis due to steric hindrance. The position of the chlorines is indicated by the numbering scheme shown in Figure 1. The reaction shown in Figure 1 would produce a large number of different PCB structures; only the 2,3,4,3',4'-pentachlorobiphenyl (2,3,4,3',4'-CB) is drawn as an illustration.

PCBs were manufactured and sold as complex mixtures differing in their average chlorination level. The crude mixtures



biphenyl 2,3,4,3',4'-pentachlorobiphenyl
2,3,4,3',4'-CB

FIGURE 1. The synthesis of PCBs via the direct chlorination of biphenyl. Note that this method produces nearly 100 different PCB products, with the congener 2,3,4,3',4'-CB included as an example.

resulting from the chlorination were fractionally distilled to produce commercial mixtures with the desired properties. The products range from light oily fluids (di-, tri-, and tetra-chlorobiphenyls) to heavy, honey-like oils (penta-chlorobiphenyls), to greases and waxes (more highly chlorinated). The manufacturers of PCBs sold the materials under various trade names: "Aroclor" (Monsanto, U.S.); "Phenoclor", and "Pyralene" (Prodelec S.A., France); "Clophen" (Farbenfabriken Bayer AG, Germany); and "Kanechlor" (Kanegafuchi Chemical Industrial Co. Ltd, Japan). The manufacturers also assigned product numbers that usually reflected the degree of chlorination by either the average number of chlorines/biphenyl or the weight percent chlorine in the mixture. For example, Aroclor 1242 (12 carbon atoms and 42% chlorine), Clophen A 30 (3 chlorines/biphenyl), and Kanechlor 300 (3 chlorines/biphenyl) all contain 42% chlorine by weight which corresponds to three chlorines/biphenyl on average. Likewise, Aroclor 1260 and Clophen A 60 contain 60% chlorine and 6 chlorines/biphenyl on average.

B. Properties

The desirable physical and chemical properties of PCBs led to their widespread use. The most important physical properties of the mixtures are that they are liquids, have low vapor pressures, low water solubility, and excellent dielectric properties. Chemical properties include stability to oxidation, flame resistance, and relative inertness. Because of excellent flam-

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mability, electrical, and stability properties, PCBs found application in a wide variety of industrial uses including heat transfer fluids, hydraulic fluids, solvent extenders, plasticizers, flame retardants, organic diluents, and dielectric fluids.[51]

In a 50-year period approximately 1.4 billion pounds of PCBs were produced. Such extensive application of these chemically and thermally stable compounds has resulted in widespread contamination.[20,52,88] It is estimated that several hundred million pounds have been released to the environment.[20] The lipophilic nature of PCBs contributes to their tendency to accumulate in fatty deposits and results in a magnification in the food chain.[80]

C. Health Risk

This accumulation of PCBs in organisms and the past exposure of some industrial workers was initially a cause for concern.[25,71] But the toxicity associated with PCBs has recently been re-evaluated.[20,57,58,64] It has now been concluded that "... the only observed acute effects have generally been minor. So far, no significant chronic health effects have been causally associated with exposure to PCBs or PBBs."[58]

Another health risk commonly associated with PCBs involves their role as suspected human carcinogens. This premise stems from early reports that high levels of Aroclor 1260 caused liver cancer in rats.[26] But a study by the National Cancer Institute (1978) concluded that Aroclor 1254, a mixture of PCBs having a slightly lower level of chlorination than Aroclor 1260, was not carcinogenic.[70] In addition, a recent thorough review of the epidemiological literature stated that "No conclusive evidence thus far reported shows that occupational exposure to PCBs causes an increased incidence of cancer."[58]

Most reviews concerning the biological and toxic effects of PCBs note that the relative potency generally correlates with the degree of chlorination.[32,55] These results suggest that the toxicities of the mixtures are variable, and it is therefore reasonable that the activities of individual congeners may also differ considerably. Valuable data involving structure/activity relationships for individual congeners is now available.[73,81,82] Safe has concluded from animal studies carried out in his laboratories that the most toxic PCB congeners contain two *para* and at least two *meta* chlorines, and the addition of *ortho* chlorines reduces this effect significantly.[81]

II. AEROBIC BIODEGRADATION OF PCBs

A. Enrichments

Most of the environmental contamination by PCBs is in the form of complex commercial mixtures (e.g., Aroclor 1242) containing >60 different congeners with varying degrees of chlorination. Biodegradation of this large number of distinct substrates therefore requires broad enzymatic specificity. In addition, chlorinated organic materials frequently resist microbial degradation.[6] Although these complex chlorinated mixtures can be difficult to biodegrade, the aerobic bacterial biodegradation of PCBs is known and has been well studied.[5,10,11,14,39,40,59,62,66,73,86] Previous reviews on the aerobic biodegradation of these materials have been published,[41,42] and this review concentrates on research results reported after their publication.

Using a rapid screening procedure, Bedard et al.[9] isolated natural aerobic bacteria capable of degrading PCBs in nearly every contaminated soil they tested. Soil and sediment samples were collected from PCB-contaminated sites and cultures were enriched on biphenyl as the sole carbon and energy source available to the bacteria. The bacterial enrichments obtained were assayed for their ability to degrade defined mixtures of PCBs. Using this approach, a diverse group of 25 strains of PCB degrading bacteria were isolated and characterized.[9,92] This method allowed the rapid determination of PCB competence for a large number of isolates. In addition, the use of defined PCB mixtures in place of complex Aroclors permitted investigations into the nature of the enzymatic specificity observed. The results of this screening technique are shown in Figure 2.[1,9] Note that all of the organisms isolated are capable of degrading the lightly chlorinated PCBs. Characterization (genus and species) for some of the PCB degrading organisms isolated by several different workers is shown in Table 1. These results indicate that naturally occurring organisms can degrade PCBs, are quite common in the environment, and that the organisms consist of many different microbiological types. It is interesting to note that nearly two-thirds of the organisms represented in this survey are members of the genus *Pseudomonas*.

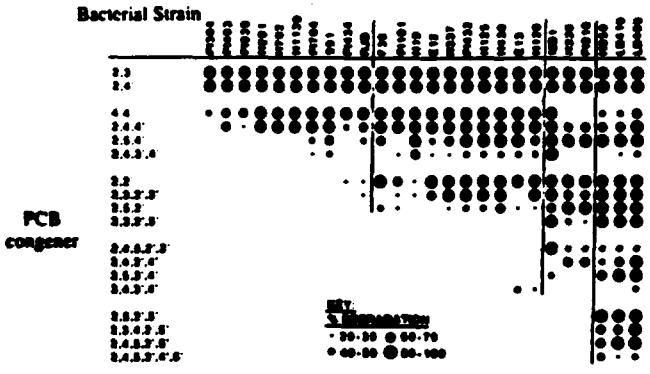

FIGURE 2. Comparison of the PCB-degrading competence of environmental bacterial isolates. [O] indicates that H850 degraded less than 20% of this congener (2,4,5,2',4',5'-CB), but a metabolite was isolated. (Adapted from Bedard, D. L., Unterman, R., Bopp, L. H., Brennan, M. J., Haberl, M. L., and Johnson, C., *Appl. Environ. Microbiol.*, 51, 761, 1986 and Abramowicz, D. A., *Hazardous Waste Treatment: Biosystems for Pollution Control*, Air and Waste Management Assoc., Pittsburgh, 1989, 301. With permission.)

B. Metabolic Pathway

The actual biochemical steps involved in the aerobic bio-

Table 1
Partial Listing of Aerobic Environmental Isolates Capable of PCB Biodegradation

Organism	Strain designation
Achromobacter sp.	BP, pCB
Acinetobacter sp.	P6, LS241
Alcaligenes sp.	KF708, Y42,BM—2
Alcaligenes eutrophus	H850
Alcaligenes faecalis	Pi434
Arthrobacter sp.	M5, B1B
Corynebacterium sp.	MB1
Pseudomonas sp.	LB400, LB410, KF714, JB1, IS140, 7509, WR912
Pseudomonas (*Acidovorans* group)	Pi939, H1130, Pi304, H702, Pi101
Pseudomonas cepacia	H201, Pi704, RJB
Pseudomonas paucimobilis	Q1
Pseudomonas pseudoalcaligenes	KF707
Pseudomonas putida	KF715, OU83
Pseudomonas testosteroni	H128, H336, H430

degradation of PCBs have been previously determined. In general, attack involves initial addition of O_2 at the 2,3- position by a dioxygenase enzyme, with subsequent dehydrogenation to the catechol followed by ring cleavage (see Figure 3).

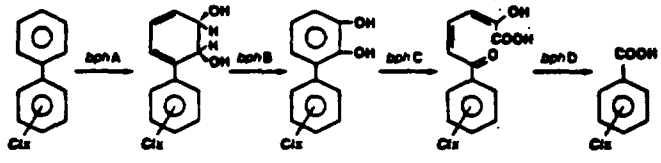

FIGURE 3. Degradation of biphenyl and chlorobiphenyls by the 2,3-dioxygenase pathway in *Pseudomonas* strain LB400. Gene designations: *bphA*, biphenyl 2,3-dioxygenase; *bphB*, dihydrodiol dehydrogenase; *bphC*, 2,3-dihydroxybiphenyl dioxygenase; *bphD*, 2-hydroxy-6-oxo-6-phenylhexa-2,4-dienoic acid (*meta*-cleavage product) hydrase. (From Mondello, F. J., *J. Bacteriol.*, 171, 1725, 1989. With permission.)

This pathway is similar to the degradation pathways for other aromatic substrates deduced for biphenyl[47] and for toluene.[34] The first two steps in the metabolism of biphenyl involve dioxygenase attack at the 2,3- position with subsequent dehydrogenation to the catechol.[47] The next step involves fission of the ring to the *meta*-cleavage product.[25] These authors also proposed that this ring fission product was further metabolized to benzoic acid, as this metabolite was identified from crude cell-free mixtures incubated with 2,3-dihydroxybiphenyl. This cleavage to benzoic acid was later confirmed.[72]

In the earliest reported isolation of PCB-degrading strains, Ahmed and Focht[5] identified both the *meta*-cleavage product and *p*-chlorobenzoic acid as metabolites of the degradation pathway. These authors postulated that the PCB degradation pathway is the same as that determined earlier for biphenyl and other aromatic hydrocarbons. This hypothesis was confirmed by Furukawa et al.[36] with the identification of the *meta*-cleavage product and chlorobenzoic acids as metabolites of PCBs.

In general, most PCB degrading aerobic bacteria are able to degrade only the lower chlorinated PCB congeners (e.g., mono- to tetra- substituted).[5,7,39,91] It is possible that higher chlorination levels result in steric hindrance of 2,3-dioxygenation by chlorine substitution at either of these two positions.[74] But several aerobic bacterial strains have demonstrated the exceptional ability to degrade an even larger range of congeners, up to and including penta-, hexa-, and even several heptachlorobiphenyls (*Pseudomonas* strain LB400,[14] *Alcaligenes eutrophus* H850,[10,11] *Corynebacterium* strain MB1,[10,12] and *Acinetobacter* strain P6.[39,40] One of these organisms has demonstrated the capacity to degrade more than 90% of the PCBs present in the mixture Aroclor 1242 (LB400).[66]

Although these organisms use the 2,3-dioxygenase degradative pathway described above, it is possible that PCBs are also metabolized through other routes. It is known that congeners containing a 2,5-chlorophenyl ring are preferentially degraded by strains H850[11] and LB400.[14] In addition, the production of different metabolites led to the proposal that a significant mechanism for PCB metabolism in these organisms involves a novel 3,4-dioxygenase attack.[9,11] This proposed 3,4-dioxygenase attack has been confirmed by Gibson[69] in both H850 and LB400 by identification of the expected *cis*-dihydrodiol intermediate from 2,5,2',5'-CB. This additional dioxygenase pathway may partially explain the exceptional range of PCB-degrading activity demonstrated by *A. eutrophus* H850 and *Pseudomonas* sp. LB400.

It is not currently known if the 2,3- and 3,4-dioxygenase activities originate from the same enzyme. It is clear, however, that the congener specificity indicates two distinct classes of dioxygenases. The dioxygenase type present in *Acinetobacter* P6 and *Corynebacterium* MB1 is particularly active against congeners containing double *para*- substitution, while the enzyme from *Alcaligenes* H850 and *Pseudomonas* LB400 prefers 2,5- substitution patterns. In general, these specificities are complementary and treatment with an organism from each class results in even greater PCB degradation.[92]

C. Optimization

It has been demonstrated that growth on biphenyl as the sole carbon source is required for optimal PCB degradative activity (LB400).[66] This is a disadvantage in soil applications where other carbon sources are available. The degradation of PCBs bound to soil has been investigated.[63,92] Although PCBs are degraded in these systems, the rates decrease significantly (more than 50-fold) compared to the biphenyl assays. One possible explanation is that biphenyl is required as the sole carbon source for maximal induction of the PCB degrading enzymes. The importance of biphenyl in the soil degradation of PCBs has been investigated by Focht[19,35] and enhanced degradation of

PCBs on soil were observed upon the addition of biphenyl as a carbon source. In addition, the PCB-degrading activity of growing cells was significantly greater for *Acinetobacter* sp. P6 and *Arthrobacter* sp. B1B than the activity observed with resting-cell suspensions.[38a] Biphenyl was utilized as the carbon source, and it is reasonable to conclude that biphenyl is required for maximal PCB degradative competence as an inducer of this dioxygenase pathway.

The PCB degradation pathways described earlier produce chlorobenzoates that are not further metabolized by these strains, although other organisms are known to mineralize these compounds.[39,40] Although many organisms can grow on monochlorobiphenyls (LB400, H850, KF715, KF707, KF708, Q1, M5, BM-2, MB1), microorganisms which could use complex PCB mixtures as a carbon source may perform better in soil applications. Strains which can degrade monochlorobiphenyls and further metabolize the chlorobenzoates have been reported. A *Pseudomonas* strain JB1 was isolated that can grow on monochlorobiphenyls, degrade mono-chlorobenzoates, and can cometabolize other congeners.[74] In addition, Focht and Huang[36] have developed a new strain which is also capable of degrading monochlorobiphenyls and metabolizing the chlorobenzoate intermediates, resulting in growth on 3-CB as the sole carbon source. This strain was generated via a method that facilitates the rapid exchange of genetic material between two parent strains. The development of new strains that could grow on the more highly chlorinated PCBs would represent a major advance in the aerobic biodegradation of PCBs.

Other methods to enhance the aerobic bacterial biodegradation of PCBs have also been reported. The addition of the aminopolysaccharide polymer chitin has been observed to increase the rate of PCB degradation.[76,77] The effects of polymer addition are shown in Table 2. Note that this method generally resulted in a twofold increase in the degradation rate by the indigenous soil microorganisms. The chitin appears to act as a solid substrate for growth as well as an efficient sorbing component for the PCBs, and therefore increases the bioavailability of these hydrophobic compounds. The addition of adapted PCB degrading bacteria resulted in even greater soil degradation rates (Table 2, microbe addition).

Table 2
Half-Life Estimates for Specific PCB Congeners with and without the Addition of Chitin or Chitin Plus Adapted Microbes

Toxicant	Polymer	Microbe	Half-life (days)
4,4'-CB	–	–	1.42 +/– 0.41
4,4'-CB	+	–	0.98 +/– 0.21
4,4'-CB	+	+	0.46 +/– 0.33
2,4,5,2',5'-CB	+	–	1.32 +/– 0.4
2,4,5,2',5'-CB	+	+	0.80 +/– 0.7
Aroclor 1232	–	–	61.4 +/– 3.6
Aroclor 1232	+	–	33.4 +/– 0.9
Aroclor 1232	+	+	26.8 +/– 0.7
Aroclor 1248	–	–	77.6 +/– 8.2
Aroclor 1248	+	–	38.6 +/– 2.4
Aroclor 1248	+	+	31.9 +/– 3.6
Aroclor 1254	–	–	81.9 +/– 7.2
Aroclor 1254	+	–	36.4 +/– 3.8
Aroclor 1254	+	+	35.5 +/– 2.2

From Portier, R. and Fujisaki, K., *Aquatic Toxicology and Hazard Assessment*, Vol. 10, ASTM STP971, Adams, W. J., Chapman, G. A., Landis, W. G., Eds., ASTM, Philadelphia, 1988, 517. With permission.

D. Genetic Engineering

The genes encoding bacterial degradation of PCBs have been isolated and utilized to construct recombinant organisms capable of degrading PCBs.[43,54,65] These studies utilized soil microorganisms from the genus *Pseudomonas* that degraded PCBs via the 2,3-dioxygenase pathway discussed earlier (see Figure 3). Furukawa and Miyazaki obtained the genes from *P. pseudoalcaligenes* strain KF707,[43] an organism known to degrade mono- to tri-CB, including 4-CB, 2,3-CB, 3,4-CB, 2,4'-CB, 2,4,5-CB, and 2,4,4'-CB. The genes were then cloned into a broad-host-range plasmid, and a transformant was isolated that was capable of degrading PCBs.[43] The researchers discovered that the genes encoding three of the four enzymes involved in PCB degradation (*bphA* through *bphC*) were localized on a small DNA fragment (7.9 kb). In addition, the 2,3-dihydroxybiphenyl dioxygenase (*bphC*) was isolated[44] and sequenced from two different organisms.[45,67] Mondello obtained the PCB-degradative genes from a *Pseudomonas* strain LB400,[65] an organism known to degrade mono- to hexa-CB, including 2,3-CB, 2,4'-CB, 2,2'-CB, 2,4,4'-CB, 2,5,2'-CB, 2,3,2',5'-CB, 2,4,2',4'-CB, 2,4,5,2',5'-CB, 2,3,4,2',5'-CB, and 2,4,5,2',4',5'-CB. The genes were then cloned into a broad-host-range vector, and a number of *E. coli* transformants capable of degrading PCBs were isolated.[65] All four of the PCB-degradative genes were isolated on a 12.4 kb DNA fragment, and one recombinant strain FM4560 demonstrated a PCB competence remarkably similar to the wild-type LB400 (see Figure 4). Note that the same congeners that are slowest to degrade in the wild-type organism display comparable kinetics in the recombinant organism. This similarity requires that the enzymes are expressed, functional, and catalyze reactions with the same congener specificity in the *E. coli* recombinant and *Pseudomonas* wild-type organisms. This somewhat unexpected result suggests that the PCB-degradative genes may be functional in a broad range of different microorganisms. Kahn and Walia obtained the PCB degradative genes from a *Pseudomonas putida* strain OU83 and localized the *bphC* and *bphD* genes onto a 2.4 kb DNA fragment.[54] The authors determined that the amount of *bphC* produced in the recombinant *E. coli* strain was 20-fold greater than that measured in the parent strain.

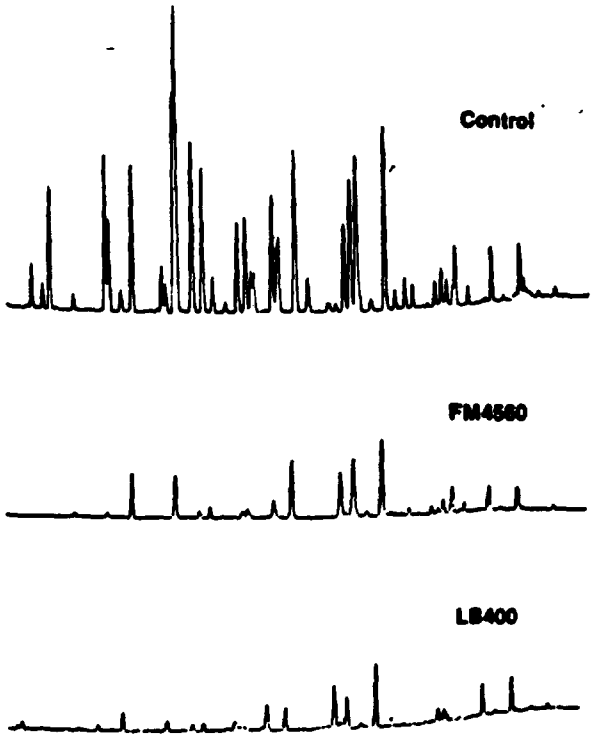

FIGURE 4. Biodegradation of Aroclor 1242 by *E. coli* FM4560 and *Pseudomonas* strain LB400. (Top) Aroclor 1242 incubated with mercury-killed cells; (middle and bottom) Aroclor 1242 (10 ppm) incubated at 30°C for 24 h with cells (optical density at 615 nm of 1.0) of FM4560 and LB400, respectively. FM4560 was grown on succinate; LB400 was grown on biphenyl. (From Mondello, F. J., *J. Bacteriol.*, 171, 1725, 1989. With permission.)

The correspondence between the PCB-degrading organisms KF707 and LB400 includes a similar metabolic pathway and similar gene organization. In addition, DNA hybridization studies revealed that the PCB-degrading genes in two very different organisms (*Pseudomonas* sp. LB400 and *Alcaligenes eutrophus* H850) were strongly conserved.[94] Additionally, the *bphABC* cluster has now been observed in five *Pseudomonas* strains and one additional *Alcaligenes* strain.[46] The high correspondence among all of these distinct organisms implies that the PCB-degradative genes did not evolve independently and the genes must have been acquired through some form of DNA transfer. This result may have important implications as it demonstrates that natural organisms can transfer and have transferred the PCB-degradative genes in the environment. These mobile genes enable the organisms to attack a broad range of PCB congeners.

Although the recombinant FM4560 can degrade PCBs no better than the wild-type LB400 under the conditions described in Figure 4, it may afford unique advantages in soil remediation applications. In addition to faster growth rates to higher cell densities, the recombinant demonstrates superior viability and temperature resistance.[65] Indeed, recombinant *E. coli* strains have been reported to survive 27 d on soils,[27] significantly better than the 2 d survival observed in the field with LB400.[63] More important is the fact that FM4560 does not require growth on biphenyl as the sole carbon source for optimal PCB-degradative competence.[66] Therefore, on soils where other organic material is readily available, the recombinant should display superior PCB-degradative competence.

E. Fungi

Microorganisms other than the bacteria, notably fungi, have also been reported to aerobically degrade PCBs. The filamentous fungus *Aspergillus niger*, used as a model of mammalian aromatic hydroxylation, has been shown to degrade the lower chlorinated PCBs in the commercial mixture Clophen A 30.[20] The wood-decay white-rot fungus *Phanerochaete chrysosporium* has also been utilized in the degradation of PCBs at very low concentrations.[21,22,31] It is believed that the same enzymes involved in lignin degradation are responsible for attack on the PCBs through the production of hydroxy radicals. This reactive intermediate should react with a wide number of organic compounds and *P. chrysosporium*, as well as other wood-decaying fungi, have been extensively studied in the degradation of a range of chlorinated organic compounds, including lindane, DDT, and chlorinated dioxins, with mineralization to $^{14}CO_2$ as the assay. The application to PCBs has demonstrated mineralization of even highly chlorinated congeners, including 3,4,3′,4′-CB,[22] 2,4,5,2′,4′,5′-CB,[21] and the mixture Aroclor 1254.[31] The results indicate that *P. chrysosporium* is capable of the complete degradation of highly chlorinated PCBs, but activity has only been observed at very low concentrations (250 ppb Aroclor 1254,[31] 5.5 ppb or 19 n*M* 3,4,3′,4′-CB).[22] Similar activities on highly chlorinated congeners have been observed with the aerobic bacteria previously described, but at much higher concentrations (10 ppm Aroclor 1254 with H850,[10] 1.8 ppm or 5 μ*M* 2,4,5,2′,4′,5′-CB with LB400,[14] and 15 ppm or 50 μ*M* 3,4,3′,4′-CB with P6.[40] The successful application of white-rot fungus to the biodegradation of PCBs will require demonstrated activity at the 50 to 1000 fold higher concentrations currently handled by bacterial systems.

F. Summary

A large number of naturally occurring, aerobic microorganisms have been isolated from many different locations and studied for their ability to degrade PCBs. The organisms range from common soil bacteria to more complex fungi. Some of the major findings follow. (1) Most soils contaminated with PCBs contain organisms with some level of PCB-degrading ability. (2) These microorganisms display congener specificity and therefore degrade individual congeners at different rates. (3) Most aerobic bacteria that have been isolated degrade only the lightly chlorinated congeners, although some bacteria have been isolated that are capable of attacking congeners containing

as many as seven chlorines. (4) For the known cases, the 2,3-dioxygenase pathway is common and quite similar in otherwise unrelated organisms. (5) Similarities in the genes encoding PCB degradation imply that these genes are being transferred between bacteria in the environment. (6) In general, the effect of aerobic bacterial PCB biodegradation is to remove the less chlorinated congeners. (7) No aerobic microorganisms have been reported that degrade the more highly chlorinated commercial mixtures Aroclor 1260 or Clophen A 60.

III. ANAEROBIC BIODEGRADATION OF PCBs

A. Environmental Evidence

Despite the extensive research on the aerobic biodegradation of PCBs, little was known about their fates in anaerobic environments such as river or lake sediments until very recently. Early studies indicated that anaerobic fermentations did not alter PCB concentrations with organisms from silage[37] or marine sediments.[38] But more recently, alterations of the PCBs present in anaerobic river and lake sediments have been observed.[15-17] These alterations involve the extensive removal of highly chlorinated PCB congeners with corresponding increases in congeners containing only a few chlorines (mono- and dichlorobiphenyls).

Several different patterns or alterations were observed for Hudson River sediments originally contaminated with Aroclor 1242 (see Figure 5). All these patterns showed markedly lower levels of most tri-, tetra-, and pentachlorobiphenyls and increased levels of mono- and dichlorobiphenyls.[16] Note that the detector response displayed in the chromatogram is non-linear and particularly poor for the congeners containing very few chlorines with short elution times.[60] Quantitation of the individual capillary chromatogram peaks indicated that in all sediments the levels of 2,6,2'-, 2,6,3'- and all dichlorobiphenyls increased 2- to 6-fold, and the level of the monochlorobiphenyl 2-CB increased 7- to 70-fold.

The observed transformations are congener specific, demonstrating selective removal of *meta* and *para* chlorines and increases in the expected partially dechlorinated PCB congeners. No known transformation processes such as evaporation or aerobic degradation could account for the striking changes observed, and it was therefore proposed that anaerobic microorganisms present in the sediments were reductively dechlorinating the PCBs.[16] In addition, transformation of even the highly chlorinated Aroclor 1260 had been observed in the environment.[15]

Anaerobic dechlorination of chlorinated aromatic compounds is not unprecedented. Tiedje and coworkers identified an anaerobic sulfidogenic bacterium strain DCB-1 capable of reductively dechlorinating dichlorobenzoates.[50] This organism represents the first and only anaerobe in pure culture capable of aromatic reductive dechlorination. It was isolated from an anaerobic consortium capable of mineralizing chlorobenzoates.[51] A review of the anaerobic dehalogenation of pesticides has recently been completed.[61] This review discusses the dechlorination of a number of aromatic substrates, including chlorobenzoates, chlorophenols, chloroanisoles, and herbicides. Reductive dechlorination of aromatics has also been reported with aerobic bacteria (chlorinated phenols and chlorinated quinones).[48,49]

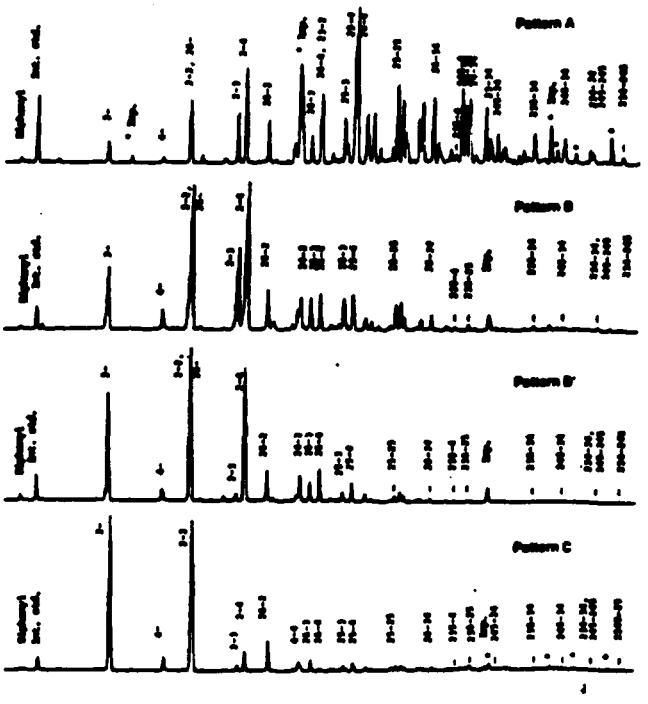

FIGURE 5. DB-1 Capillary chromatograms (plots of detector response vs. elution time) of upper Hudson River sediments that show surface pattern A (largely unchanged Aroclor 1242) and subsurface patterns B, B', and C. A flame ionization detector was used so that PCB peak response was nearly proportional to molar concentration, however non-PCB impurities also produced observable peaks (designated * and "Imp."). (From Brown, J. F., Jr., Bedard, D. L., Brennan, M. J., Carnahan, J. C., Feng, H., and Wagner, R. E., *Science*, 236, 709, 1987. With permission.)

B. Laboratory Confirmation

The proposed microbial dechlorination in anaerobic river sediments was confirmed in the laboratory.[76] The result of anaerobic dechlorination of Aroclor 1242 by microorganisms in Hudson River sediments is shown in Figure 6. Note the dramatic loss of the highly chlorinated congeners with corresponding increases in the less chlorinated products. These microorganisms dechlorinate the PCB mixture so extensively that it is converted from 85% tri- and tetra-chlorinated PCBs to 88% mono- and dichlorinated products. The end result of this natural process is the conversion of the more highly chlorinated PCBs into congeners of low toxicity that are degraded by a large number of aerobic bacteria.

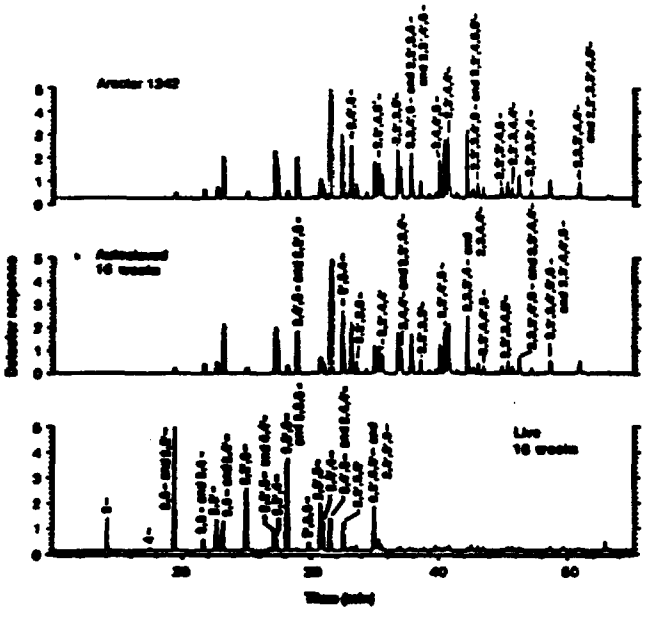

FIGURE 6. Capillary gas chromatograms showing the anaerobic dechlorination of Aroclor 1242 by Hudson River microorganisms. All chromatograms were normalized so that the highest peak had a height of 5. An electron capture detector was used. From Quensen, J. F., III, Tiedje, J. M., and Boyd, S. A., *Science*, 242, 752, 1988. With permission.)

The dechlorination was found to represent selective removal of *meta* and *para* chlorines as well, confirming that this natural process observed in the lab is the same as the dechlorination found in the environment. Therefore an additional benefit of this anaerobic dechlorination is the removal of the *meta* and *para* chlorines known to contribute to PCB toxicity.[81] The similarity between environmental and laboratory changes can be seen by comparing Figures 5 and 6.

Methods to accelerate this desirable natural process have also been identified.[2,4] The addition of a simple minimal medium (first described by Shelton and Tiedje)[84] containing nutrients and trace minerals results in a significantly more rapid activity compared to that of unsupplemented sediments. Other factors which increase the rate of dechlorination include the addition of a complex carbon source (fluid thioglycollate medium with beef extract) or a detergent (Triton X-705).[2] These effects are additive and a combination of variables results in even greater enhancement. Importantly, the dechlorination of the highly chlorinated Aroclor 1260 has also been observed in the laboratory.[4,78a] The concentration of the most highly chlorinated congeners (hexa-, hepta-, and octachlorobiphenyls) has been decreased by more than one-third the original level in Aroclor 1260.[4] Anaerobic dechlorination of Aroclor 1260 has recently been observed by others as well.[8,67,90]

C. Single Congener

Methods for the synthesis of individual PCB congeners[23] and commercially available material have provided single compounds for a more detailed study of this anaerobic dechlorination process.[3,4] The timecourse of the dechlorination observed with Hudson River sediments on the congener 2,3,4,3',4'-CB is shown in Figure 7. The dechlorination activity observed demonstrates a sequential pathway from the penta- (2,3,4,3',4'-CB) to tetra-(2,4,3,',4'-CB), tri- (2,4,3'-CB), di-(2,3'-CB), and mono- (2-CB) chlorinated biphenyls (major products shown in parentheses). The result of this process is the conversion of one of the more toxic congeners (2,3,4,3',4'-CB) into a monochlorobiphenyl (2-CB) which has low toxicity and is easily metabolized by aerobic bacteria and higher organisms.

This result again confirms that chlorines are removed from only the *meta* and *para* positions as was observed for the river sediment itself.[16] The selectivity of each individual step is surprising; for example 2,4,3',4'-CB is the only detectable tetrachlorinated product observed from 2,3,4,3',4'-CB, and it is produced in stoichiometric amounts.

This microbial selectivity for only *meta* and *para* chlorines is different than that observed from the direct electrochemical reduction of PCBs. Farwell et al.[33] determined that although the PCB reduction pathways were complex, dechlorination via voltammetric reduction was observed from the *ortho*, *meta*, and *para* positions. In some cases the *ortho* dechlorinated species was the major product, for example 2,3-CB yields 89% 3-CB and only 11% 2-CB.[33] This can be contrasted to the microbial dechlorination of 2,3'-CB, where 2-CB is the only observable product.[4]

The dechlorination of single congeners with higher toxicity has also been demonstrated by Tiedje et al.[89] These investigators found that 2,3,4,3',4'-CB and 3,4,3',4'-CB were dechlorinated at rates comparable to other penta- and tetrachlorobiphenyls, even in the presence of the complex PCB mixture Aroclor 1242.

From such studies utilizing single PCB congeners, one can prove that microbial reductive dechlorination is occurring in the sediments. The stoichiometric production of PCBs containing fewer chlorines demonstrates the substitution of hydrogen in place of the chlorine. It is believed that the anaerobic microorganisms are utilizing the chlorine as the terminal electron acceptor, involving the addition of the electron to the carbon-chlorine bond, followed by chloride loss and subsequent hydrogen abstraction (see Figure 8). The compound from which the hydrogen is ultimately abstracted is unknown, and potential primary electron donors include water, hydrogen, or an organic compound. The availability of hydrogen in anaerobic microbial systems may make it the most likely primary source of reducing equivalents.

Recently it has been shown by Hogenkamp and coworkers[68] that vitamin B_{12} can catalyze the reductive dechlorination of carbon tetrachloride and other chlorinated methanes. Vitamin B_{12} is a known hydride transfer agent and this result suggests an alternative dechlorination mechanism involving a single step

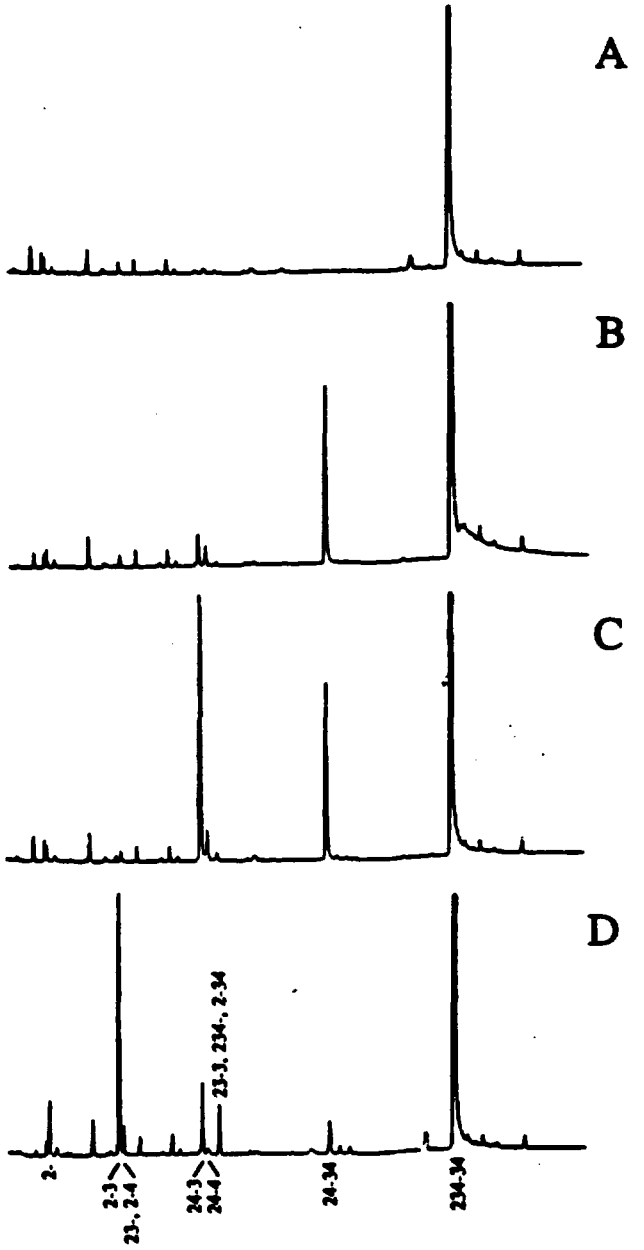

FIGURE 7. Stepwise dechlorination of 2,3,4,3',4'-CB (500 ppm) by anaerobic Hudson River sediments. Panel A, autoclaved control; Panel B, 4 weeks; Panel C, 8 weeks; Panel D, 12 weeks. From Abramowicz, D. A., Brennan, M. J., Van Dort, H. M., and Gallagher, E. L., *Chemical and Biochemical Detoxification of Hazardous Waste II*, Glasser, J., Ed., Lewis Publishers, Chelsea, MI, 1990. With permission.)

$$\text{Ar-Cl} + e^- + \text{R-H} \Longrightarrow \text{Ar-H} + Cl^- + \text{R}^\cdot$$
$$(H_2O) \qquad\qquad (HO^-)$$
$$(H_2) \qquad\qquad (H^\cdot)$$

$$\text{Ar-Cl} \xrightarrow{e^-} \text{Ar-Cl}^{-\cdot} \xrightarrow{Cl^-} \text{Ar}^\cdot \xrightarrow{\text{R-H}} \text{Ar-H} + \text{R}^\cdot$$

FIGURE 8. Possible mechanism for reductive dechlorination catalyzed by anaerobic microorganisms. In the proposed scheme, the organisms utilize PCBs as an electron acceptor, with addition of the electron to the carbon-chlorine bond, chloride loss, and hydrogen abstraction from an unknown species.

concerted process catalyzed by this cobalamin cofactor or other corrinoids present in these anaerobic microorganisms.

D. Anaerobic Degradation

The dechlorination process described earlier does degrade highly chlorinated PCBs, but the organisms leave the biphenyl nucleus untouched and less chlorinated PCBs are formed. Although this dechlorination represents actual biodegradation of highly chlorinated PCBs, it is being distinguished here from processes that do attack the biphenyl ring, resulting in potential mineralization of the PCB. Such a process has recently been reported by Rhee and coworkers.[26,79] In this work, bacterial populations from Hudson River sediments were reported to anaerobically degrade the lightly chlorinated congeners in PCB mixtures. No metabolites were identified, and no evidence for the dechlorination process described earlier was observed by these authors. Although the dechlorination and biodegradation results reported here both utilized sediments from the Hudson River, CO_2 was provided to the dechlorinating systems as bicarbonate, but it was absent in the biodegradation studies. It is interesting to speculate that CO_2 may be important in determining the type of anaerobic activity observed on PCBs. It is possible that in the absence of CO_2, a selection is imposed favoring organisms capable of degrading PCBs to obtain CO_2 and/or low molecular weight metabolites as electron acceptors.

E. Summary

This environmental dechlorination of PCBs has now been observed in a large number of contaminated anaerobic sediments. Sites include many locations in the Hudson River (New York), Silver Lake (Pittsfield, Massachusetts), New Bedford Harbor (Massachusetts), Escambia Bay (Pensacola, Florida), Woods Pond (Massachusetts), the Housatonic River (Con-

necticut), the Sheboygan River (Wisconsin), Waukegan Harbor (Illinois), and the Hoosic River (North Adams, Massachusetts).[13,18] The widespread occurrence of this natural process indicates that it is a general phenomenon.

These changes observed in PCB-contaminated anaerobic river sediments led to the proposed microbial reductive dechlorination of PCBs.[15-17] This process has now been confirmed in a number of laboratories with sediments from many distinct aquatic systems.[2-4,7b,7da,89,93] Some of the most significant findings from current anaerobic dechlorination experiments follow. (1) Dechlorination has been observed in a large number of sediments and the process is widespread in the environment. (2) Although congener preferences are demonstrated, in general the organisms present in Hudson River sediments exhibit broad dechlorination activity on the more highly chlorinated PCBs. (3) These anaerobic microorganisms are capable of dechlorinating even the previously recalcitrant, highly chlorinated PCB congeners contained in Aroclor 1260. (4) All results to date involve primary cultures and pure PCB dechlorinating strains have not yet been isolated. (5) Dechlorination selectively removes *meta* and *para* chlorines, significantly reducing any toxicity associated with PCBs. (6) The less chlorinated congeners that are produced are known substrates for aerobic bacterial systems.

IV. CONCLUSIONS

This paper has focused on recent progress (since 1985) in the aerobic bacterial biodegradation of PCBs and the new anaerobic dechlorination process recently discovered. The large number of researchers cited have repeatedly demonstrated that PCBs, commonly believed to be indestructible, are degraded by a number of diverse microorganisms.

Two separate and complementary natural biological systems have been the focus of this report on the biodegradation of PCBs. Anaerobic bacteria, present in river and lake sediments, remove chlorine from even the most highly chlorinated PCBs. This process is relatively broad, attacking a large array of highly chlorinated PCBs. The resultant lightly chlorinated compounds are less toxic, and are known substrates for aerobic bacterial biodegradation. Such aerobes have been identified in nearly all PCB-contaminated areas and are widespread in the environment. The obvious complementarity of these biological processes leads to the combined treatment scheme shown in Figure 9 (only one PCB congener is shown as an illustration). Successful application of this sequential treatment may enable the bioremediation of nearly all types of PCB contamination.

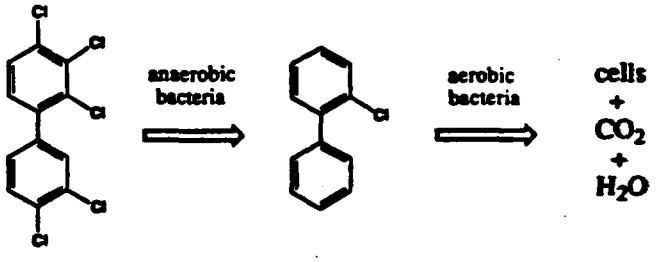

FIGURE 9. Two-step combined anaerobic/aerobic process to biodegrade PCBs. In this scheme, initial anaerobic treatment converts highly chlorinated PCBs to lightly chlorinated derivatives. Subsequent aerobic treatment destroys the remaining material.

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19

Bacterial Transformation of Polychlorinated Biphenyls

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Aerobic bacteria are proving versatile and effective agents for biodegrading polychlorinated biphenyls (PCBs). Four natural isolates have been shown to degrade many tetra- and pentachlorobiphenyls and some hexachlorobiphenyls. The enzymes responsible for PCB metabolism in these organisms fall into two genetically distinct classes that differ markedly in congener reactivity preferences. Because the reactivities complement each other, treating Aroclor 1242 with a mixture of two bacteria representing each class of enzymes has been particularly effective. *In situ* bioremediation of Aroclor® 1242 (Monsanto, St. Louis, MO) has been demonstrated.

Extensive reductive dechlorination of PCBs in anerobic sediments of lakes, rivers, and harbors has been documented. Laboratory experiments have demonstrated that this dechlorination proceeds by step-wise removal of *meta* and *para* chlorines and results in the accumulation of mono- to trichlorobiphenyls that are easily degraded by aerobic bacteria. It appears that natural microbial populations already have the capacity to biodegrade most if not all of the PCBs that contaminate the environment. Research aimed at stimulating the activity of these organisms should make it possible to accelerate biodegradation of PCB contaminants

Introduction

Polychlorinated biphenyls (PCBs) have attracted concern because of their persistence, their bioaccumulation, and their possible health effects. PCBs were used worldwide for a wide range of applications for more than 50 years. Major uses included transformer oil, capacitor dielectric fluid, heat transfer fluid, fire retardants, and plasticizers. The properties that made PCBs useful industrial chemicals; thermal and chemical stability, resistance to chemical corrosion, and general inertness, have contributed to their widespread persistence in nature near the sites of their production, use, storage, or disposal. Because of their hydrophobic nature, PCBs have accumulated primarily in soils and aquatic sediments where they adsorb strongly to organic matter.

PCBs are a family of compounds (congeners) consisting of a biphenyl nucleus carrying one to 10 chlorines; hence, there are 209 possible PCB congeners that differ in the number and position of the chlorines. Because the commercial PCBs (Aroclors) commonly contain more than 60 to 80 congeners, they pose a particulary difficult challenge as candidates for bioremediation.

Bacterial Oxidation of PCBs

There have been numerous reports of bacteria that are capable of degrading PCBs. The literature prior to 1982 has been thoroughly reviewed,[1] but there have been a number of reports of bacteria with exceptional ability to degrade PCBs since that date, and several reports of novel dioxygenase attack on PCBs. Four strains of bacteria are particularly noteworthy for their ability to degrade a broad spectrum of PCBs: *Acinetobacter* sp. P6,[2] *Corynebacterium* sp. MB1[3,4] *Alcaligenes eutrophus* H850,[3-5] and *Pseudomonas* sp. LB400.[6]

Acinetobacter sp. P6 was extensively characterized by Furukawa and co-workers.[1,2] When grown on biphenyl or 4-chlorobiphenyl (4-CB), *Acinetobacter* sp. P6 can oxidize a broad range of PCB congeners, including many tetrachlorobiphenyls and some penta- and

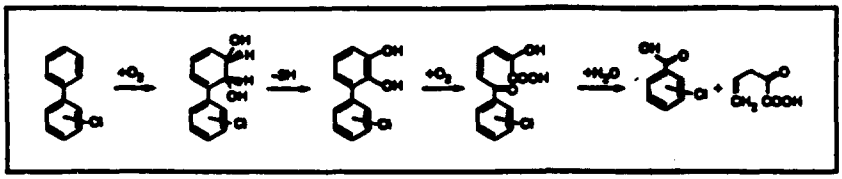

Figure 1. Pathway for the degradation of PCBs.

hexachlorobiphenyls.[7] *Corynebacterium* sp. MB1 was isolated as a contaminant from a culture of *Acinetobacter* sp. P6. Although never compared side by side, the two strains appear to have very similar PCB-degradative competence. Both oxidize PCBs via dioxygenase attack at carbon positions 2,3 followed by dehydrogenation, *meta* ring-fission between carbons 1and 2, and cleavage to generate chlorobenzoic acid and a five carbon fragment (Figure 1). Both can oxidize some dichlorophenyl rings, most notably those with chlorine substituents at carbon positions 2,3 and 3,4, but neither strain has been shown capable of oxidizing a trichlorophenyl ring.

A. eutrophus H850 and *Pseudomonas* sp. LB400 were isolated at different times and from different locations, yet they have remarkably similar congener specificity. Both strains have a superior ability to degrade PCBs via attack on 2-, 2,4-, 2,5-, 2,3,6- and 2,4,5-chlorophenyl rings, all of which are common substituents in the commercial PCB mixtures (Aroclors). Because of this, they degrade many tetra- and pentachlorobiphenyls and some hexachlorobiphenyls. These strains also metabolize biphenyl and PCBs via a 2,3-dioxygenase pathway (see Figure 1), but in addition they metabolize PCBs containing a 2,5-chlorophenyl ring via 3,4-dioxygenase attack (Figure 2, A).[8,4] It appears that the 3,4-dihydrodiol cannot be dehydrogenated or further metabolized in these microorganisms except by a second 3,4-dioxygenase attack to generate a *bis*-diol.[8]

Both *A. eutrophus* H850 and *Pseudomonas* sp. LB400 can oxidize 2,4,5,2',4',5'-CB to a single-ring product, 2',4',5'-trichloroacetophenone,[4,6,8] but the route of degradation of this congener has not been understood as it has no unchlorinated 2,3- or 3,4- position available for dioxygenase attack. However, we have recently obtained evidence that the 2,3-dioxygenase in these organisms can

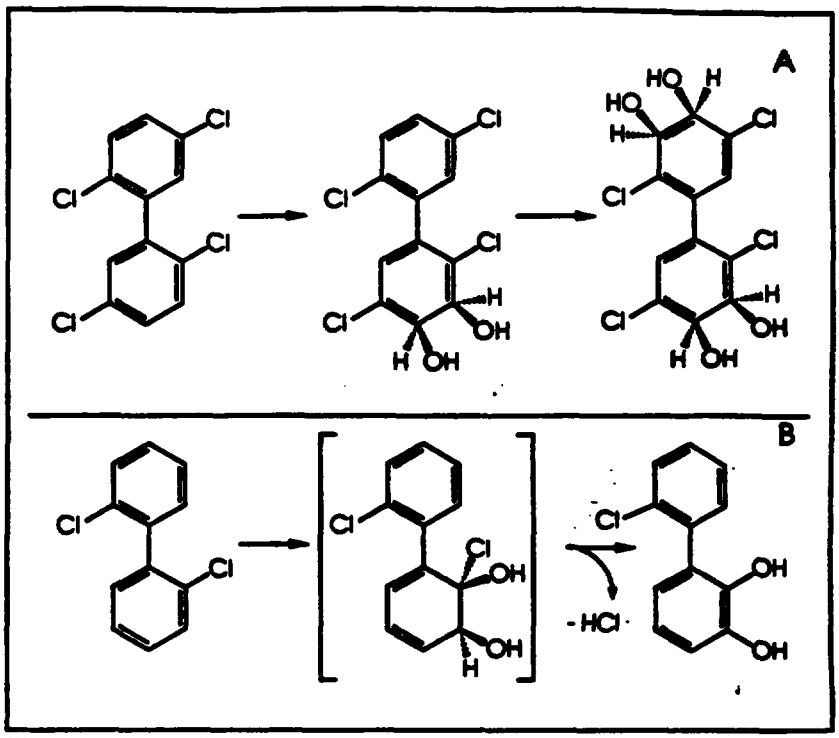

Figure 2. Alternative dioxygenase attacks on PCBs that occur in *Alcaligenes eutrophus* H850 and *Pseudomonas* sp. LB400: (A) 3,4 dioxygenase attack; (B) 2,3-dioxygenase attack at an *ortho*-chlorinated carbon.

attack at an *ortho*-chlorinated carbon and may even exhibit a preference for attack at a chlorinated position in PCB congeners containing chlorines at carbon positions 2,2'.[9] Figure 2, panel B illustrates the proposed reaction. Instead of a dihydrodiol, an unstable intermediate would form that would spontaneously lose a chloride ion to form a dihydroxy-chlorobiphenyl. The dihydroxy-chlorobiphenyl could then be further metabolized via the biphenyl 2,3-dioxygenase pathway. Dioxygenase attack at the *ortho*-chlorine of the 2,5-chlorophenyl ring of 2,5,2',5'-CB would result in loss of the *ortho*-chlorine and would generate 2,5,3'-trichloro-5',6'-dihydroxybiphenyl, the same intermediate that would result from conventional 2,3-dioxygenase attack on the 3-chlorophenyl ring of 2,5,3'-CB. In other words, once the 2-

chlorine of a 2,5-chlorophenyl ring is removed, the ring would be metabolized exactly like a 3-chlorophenyl ring. Because the oxidation of a 3-chlorophenyl ring of a PCB generates chloroacetophenones,[4,10] this would explain the production of chloroacetophenones from the metabolism of PCBs containing a 2,5-chlorophenyl ring. Presumably 2,4,5,2',4',5'-CB could be metabolized in much the same way.

It is not yet clear whether a single PCB–biphenyl dioxygenase with relaxed specificity is responsible for all three types of dioxygenase attacks that have been observed in *A. eutrophus* H850 and *Pseudomonas* sp. LB400. It is clear, however, that there are at least two classes of PCB/biphenyl dioxygenases that differ markedly in congener reactivity preferences. The type of dioxygenase found in *Acinetobacter* sp. P6 and *Corynebacterium* sp. MB1 is particularly well suited to the degradation of the more planar PCB congeners such as 4,4'-CB and 3,3'-CB and to congeners with a single *ortho*-chlorine, such as 2,4,5,4'-CB. The class of dioxygenase typified by *A. eutrophus* H850 and *Pseudomonas* sp. LB400 preferentially degrades congeners containing an *ortho*-chlorine on each ring (2,2') and congeners containing a 2-, 2,4-, 2,5-, 2,3,6-, or 2,4,5-chlorophenyl ring, but has a limited ability to degrade congeners containing 4-chlorophenyl rings. This is clearly illustrated by the fact that the latter two strains cannot degrade 2,4,5,4'-CB but can degrade 2,4,5,2',4',5'-CB. The PCB congener reactivity preferences of these two dioxygenase classes complement each other so well that treatment of Aroclor 1242 with both *Corynebacterium* sp. MB1 and *Pseudomonas* sp. LB400 resulted in total degradation of almost all congeners in the Aroclor.[11]

Aroclor 1254 is a more difficult substrate because it is more highly chlorinated, but substantial degradation of this Aroclor by several bacteria has now been demonstrated in the laboratory. Most PCB degradation assays have been conducted with resting-cell suspensions because this permits the correlation of degradation activity to cell number and therefore provides a means of more direct comparisons between strains. However, because the bacteria probably gain little or no energy from the metabolism of the more highly chlorinated congeners, the metabolism of PCBs would not be optimal under

resting-cell conditions unless nutrients and cofactors were replenished. Recently, a direct comparison was made of the ability of two bacterial strains, *Acinetobacter* sp. P6 and *Arthrobacter* sp. B1B, to oxidize Aroclor 1254 as resting cell suspensions and as cells actively growing on biphenyl.[7] The authors found that for both bacterial strains, growing cells were superior to resting cells in terms of the total amount of PCB degraded, the extent of depletion of specific congeners, and the diversity of congeners that were degraded. Resting cells of *Acinetobacter* sp. P6 degraded 17 percent of Aroclor 1254 (10ppm) and the components of 19 of the 40 capillary gas-chromatographic peaks of Aroclor 1254. Cells of the same strain growing on biphenyl degraded 32 percent of the Aroclor and the components of six additional peaks.[7] The congeners that were degraded by growing cells included 2,5,2',5'-CB and several other congeners containing a 2,5-chlorophenyl ring. These are congeners against which this strain has been reported to have only weak activity.[2] At this point, only resting cell data are available for *A. eutrophus* H850. However, even under these conditions H850 degraded the components of 21 of 44 capillary peaks to effect a degradation of 35 percent of Aroclor 1254.[5] Because the congeners that were selectively degraded by *Acinetobacter* sp. P6 and *A. eutrophus* H850 differed, treatment of Aroclor 1254 with both strains should result in even better degradation.

Genetics of PCB Degrading Bacteria

Until recently little was known about the genes that encode the enzymes responsible for PCB degradation, but the genes encoding the entire PCB degradation pathway have now been cloned from *Pseudomonas* sp. LB400, *A. eutrophus* H850, and *P. putida* OU83,[12-14] and the genes for part of the pathway have been cloned for several other *Pseudomonas* strains.[15,16] The recombinant *Escherichia coli* containing the insert from *Pseudomonas* sp. LB400 has been shown to degrade Aroclor 1242 nearly as well as the donor strain, but unlike *Pseudomonas* sp. LB400, the recombinant did not require growth on biphenyl to achieve high levels of degradative activity.[12] In addition,

DNA-DNA hybridization experiments comparing the genes for PCB degradation from *Pseudomonas* sp. LB400 with those of seven other PCB-degrading strains showed that these genes are closely related in *Pseudomonas* sp. LB400 and *A. eutrophus* H850 but that they are genetically distinct from the other six strains.[17] The six strains included *Corynebacterium* sp. MB1 and four other species with PCB congener selectivity preferences similar to albeit narrower than that of *Corynebacterium* sp. MB1. These results confirm the existence of at least two distinct classes of genes encoding PCB degradation.[17] In the region of DNA encoding PCB metabolism, *A. eutrophus* H850 and *Pseudomonas* sp. LB400 showed a strong conservation of restriction sites, yet no other sequence similarities were detected in the two genomes. The authors inferred that these genes must have been acquired through some form of DNA transfer and that the genes for PCB degradation can be spread within bacterial populations in the environment.[17] However, in these two strains the PCB degradation pathway does not appear to be plasmid-encoded.[17]

Degradation of PCBs in Soil

There is little evidence for PCB degradation in soil in nature, yet bacteria capable of degrading PCBs are easily isolated from PCB-contaminated soils by enrichment with biphenyl. The natural substrate for the biphenyl–PCB-degradative enzymes has not been identified, but at present it appears that biphenyl or a monochlorobiphenyl is required as a growth substrate in order to achieve maximum activity of the enzymes involved in PCB degradation. Our laboratory studies have shown that *A. eutrophus* H850, *Pseudomonas* sp. LB400, and *Corynebacterium* sp. MB1 all have reduced PCB-degrading activity when grown on carbon sources such as glucose, succinate, glutamate, histidine, or Luria broth rather than biphenyl. The PCB congeners that persist in soil generally have three or more chlorines and will not support the growth of any of the PCB-degrading bacterial strains described in the literature. This is a major obstacle to PCB degradation *in situ*. Because the PCB congeners that are present at spill sites will

not support growth, we know of no way to enrich or sustain a PCB-degrading population unless biphenyl is added.

Some work has been done on the degradation of PCBs in soil. Two papers describe laboratory experiments investigating the conditions under which Aroclor 1242 (100ppm) in soil could be degraded.[18,19] In these experiments it was found that no significant degradation occurred unless biphenyl was added (3.3mg/kg). The addition of biphenyl alone (without bacterial inoculum) resulted in approximately 60 percent degradation of the PCB in 49 days. When a single inoculum of *Acinetobacter* sp. P6 was also added, 70 percent of the PCB was degraded, including a higher proportion of tetrachlorobiphenyl. Some congeners such as 2,5,2',5'-CB were refractory.

Plating assays were conducted to determine the growth kinetics of the bacterial population in the soil capable of using biphenyl as a carbon source. After the addition of biphenyl, the population increased rapidly for 10 to 15 days, then declined exponentially when the biphenyl was exhausted.[19] This suggests that sustained degradation of PCBs would require repeated applications of biphenyl or some other substrate that would sustain the activity of the PCB-degrading population. It was also established that 72.5 percent of the degraded PCB was mineralized to carbon dioxide, carbonate, and bicarbonate. This result indicates that microorganisms indigenous to the soil were capable of mineralizing the by-products of PCB degradation: chlorobenzoic acids and chlorinated organic acids.

The experiments just described were conducted with clean soil that was treated with PCBs in the laboratory. However, actual spills are often contaminated with other organic pollutants such as oil. If the PCBs are sequestered in dispersed oils or waxes, their bioavailability may be quite limited. In addition, the nature of the soil (sand, clay, organic content) affects the sorption of the PCBs and hence their bioavailability.

Recently, a series of laboratory experiments was conducted using PCB-contaminated soil obtained from a drag strip in South Glenns Falls, New York.[11,20] The PCB was partially evaporated (depleted in di- and trichlorobiphenyls) Aroclor 1242 at a concentration of 525ppm. Initial studies were conducted under optimal condi-

tions to determine if the PCB could be degraded: 0.4g of soil was incubated with 2ml of *Pseudomonas* sp. LB400 (1 $O.D._{615nm}$, 30°C, 250rpm for 3 days). Under these conditions, 50 percent of the PCBs were degraded. However, percolation experiments of the same soil dosed three times per week with *Pseudomonas* sp. LB400 showed a much lower rate of degradation. In undisturbed soil, 50 percent of the PCB in the top centimeter was degraded in 15 weeks, but only 10 percent of the PCBs below a depth of one centimeter was degraded. When thoroughly mixed after each application of bacteria, 35 percent degradation at all depths was achieved in 23 weeks.

Following these laboratory studies, a field test was conducted in a site at the drag strip in South Glenns Falls from June to late October, 1987.[20] *Pseudomonas* sp. LB400 was again applied three times a week. Half of the 3m x 3m test plot was left undisturbed and half was rototilled weekly. At the end of the 19-week test, 25 percent degradation had occurred in the top centimeter, but significantly less degradation occurred below the surface. In the half that was mixed, 19 percent of the PCB was degraded throughout the 15cm depth. Undoubtedly, cell viability, temperature conditions, and moisture control all affected the outcome of the field test, but the oil and other organic pollutants in the soil were probably also a major factor.

Organic pollutants appeared to be the major obstacle to biodegradation of Aroclor 1242 in an industrial sludge from a settling tank that we investigated.[21] The sludge was composed of oily coarse sand containing about 500ppm of Aroclor 1242 with small amounts of Aroclors 1221, 1016, and 1254. Additional organic compounds present in the sludge included trichlorobenzenes, di(2-ethylhexyl) phthalate (DEHP), mineral oil, kerosene, and No. 2 fuel oil. The PCBs in the sludge were completely refractory to degradation by high cell densities of either *Pseudomonas* sp. LB400 or *A. eutrophus* H850. When subsequent experiments ruled out water-soluble inhibitory agents as the source of the problem, we suspected the other organic pollutants in the sludge. The most prominent of these was DEHP, which we estimated was present at approximately a 150:1 ratio relative to the PCB (w:w). DEHP is an oily substance that is extensively used as a plasticizer. Aroclor 1242 has a very low solubility in

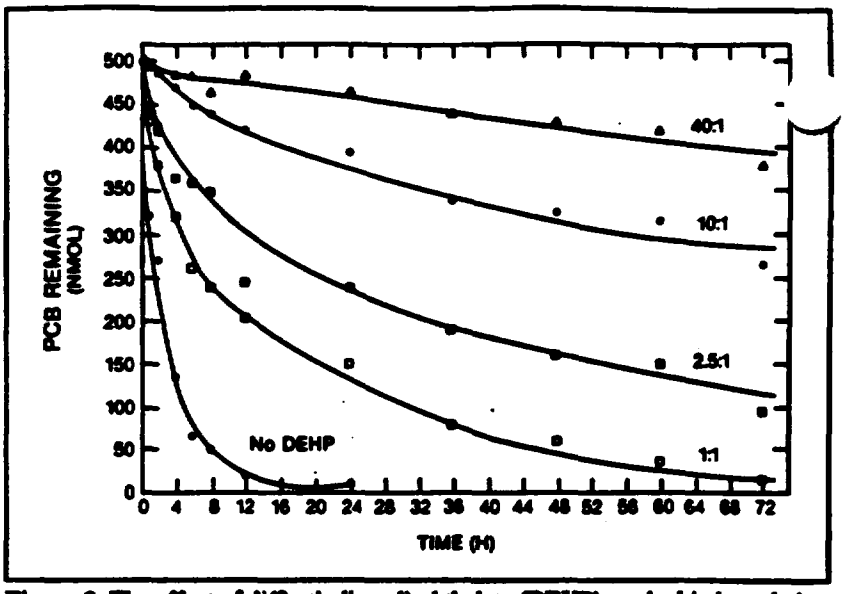

Figure 3. The effect of di(2-ethylhexyl) phthalate (DEHP) on the biodegradation of 2,4'-CB by *Alcaligenes eutrophus* H850. The 2,4'-CB was incubated with resting cells of *A. eutrophus* H850 in the presence of various amounts of DEHP. The ratios on the graph refer to the molar ratio of DEHP to PCB.

water (288ppb) but is readily soluble in oil. In fact, the octanol:water partition coefficient for Aroclor 1242 is 196,500.[22] The partitioning of PCB into an oil phase such as DEHP would be expected to depress its availability for biodegradation.

In a resting-cell assay[23] we compared the biodegradation of Aroclor 1242 (10ppm) by *A. eutrophus* H850 in the presence or absence of a 150-fold excess of DEHP. The sample without DEHP was 78 percent degraded in 72 hours, but no degradation of the PCB was detected in the presence of DEHP. A second experiment examined the effect of DEHP on the biodegradation of 2,4'-CB. The results are shown in Figure 3. In the absence of DEHP, the PCB was nearly 50 percent degraded in two hours, but when DEHP was added at the same molar concentration as the PCB, it took six hours to degrade half the PCB. With a 10-fold molar excess of DEHP, the PCB was still not 50 percent degraded at 72 hours, and with a 40-fold molar excess, less than 25 percent of the PCB was degraded in 72 hours.

It is apparent from these experiments that DEHP, even at low concentrations, had a strong negative effect on PCB degradation. We have established that *A. eutrophus* H850 can grow in the presence of 10 percent DEHP, so the observed effect on PCB biodegradation cannot be attributed to toxicity. Other possible explanations are (1) the DEHP might compete with PCB for, or otherwise affect, the PCB-degrading enzymes, or (2) the DEHP might sequester the PCB and make it unavailable for degradation. We favor the latter possibility.

Reductive Dechlorination of PCBs in Anaerobic Sediments

In 1984 Brown *et al.*[24] reported that extensive dechlorination of PCBs had occurred in the anaerobic sediments of the Hudson River south of Hudson Falls, New York. Existing usage records indicate that this PCB was originally almost entirely Aroclor 1242 that was released from a capacitor manufacturing plant between 1952 and 1971. Aroclor 1242 is primarily composed of tri- and tetrachlorobiphenyls (see Figure 4) and contains only 0.7 percent 2-chlorobiphenyl and 11.5 percent dichlorobiphenyl, but the PCB extracted from the anaerobic river sediments was composed of 10 to 43 percent 2-chlorobiphenyl and 21 to 50 percent dichlorobiphenyls. Capillary gas chromatography showed particularly prominent peaks of lower congeners with *ortho*-chlorines: 2-, 2,2'-, 2,6-, 2,3'-, 2,3-, 2,4-, 2,6,2'-, 2,6,3'-, and 2,6,4'-CB.[24-26] The authors concluded that anaerobic microorganisms were selectively removing chlorines from the *meta* and *para* positions of the more highly chlorinated PCB congeners, and this resulted in the accumulation of *ortho*-chlorinated mono-, di-, and trichlorobiphenyls. Furthermore, because several distinct dechlorination patterns were seen, the authors concluded that several different populations of microorganisms were involved.

Extensive dechlorination of Aroclor 1260 (primarily hexa- and heptachlorobiphenyls) was seen in the anaerobic sediments of Silver Lake (Pittsfield, Massachusetts), which is located next to a transformer manufacturing plant. The PCB extracted from the sediments showed a 90 to 98 percent loss of the hexa- and heptachlorobiphenyl

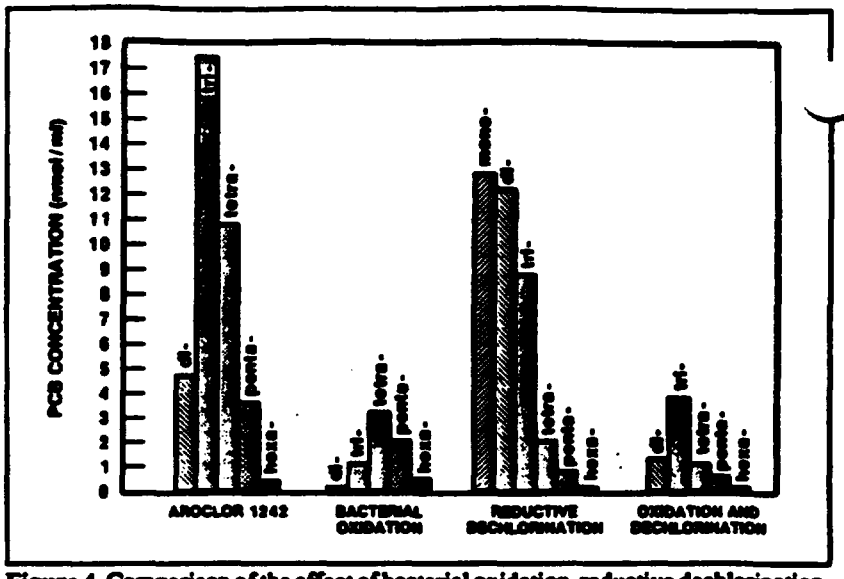

Figure 4. Comparison of the effect of bacterial oxidation, reductive dechlorination, or both on Aroclor 1260. The data were taken from Figure 1 and Table 1 in Bedard and co-workers[5] and were compiled by Vogel, Nies, and Anid.[32] Bacterial oxidation: resting-cells of *Alcaligenes eutrophus* H850 were incubated with 10ppm of Aroclor 1242 at 30°C for 48 hours. Reductive dechlorination: environmentally dechlorinated Aroclor 1242 was extracted from Hudson River sediment. Oxidation and dechlorination: environmentally dechlorinated Aroclor 1260 from Hudson River sediment was incubated with resting-cells of *A. eutrophus* H850 as previously described.

peaks of Aroclor 1260 and the appearance of large amounts of tri- tetrachlorobiphenyls. (These homologs account for less than one percent of the total PCB in Aroclor 1260, but they account for 57 to 82 percent of the total PCB in the altered Aroclor extracted from the sediments.[25,26] Evidence for dechlorination of PCBs in anaerobic sediments at several other spill sites was also found. These included freshwater sediments from Waukegan Harbor, Illinois; the Hoosic River (North Adams, Massachusetts); the Sheboygan River (Sheboygan, Wisconsin); and sediments from two marine sites: the Acushnet Estuary (New Bedford, Massachusetts), and Escambia Bay (Pensacola, Florida).[25-27]

Experiments in several laboratories have now confirmed that re-

ductive dechlorination of PCBs occurs in PCB-contaminated sediments cultured under anaerobic conditions.[28-32] Quensen, Tiedje, and Boyd[28] transferred anaerobic microorganisms from a PCB-contaminated Hudson River sediment sample that showed evidence of extensive dechlorination to an autoclaved PCB-free sediment that had been amended with 700ppm of Aroclor 1242 in the laboratory. After 16 weeks' incubation in a minimal medium under methanogenic conditions, 53 percent of the total chlorine was removed. Furthermore, 2-CB, a congener not present in Aroclor 1242, represented 63 percent of the total PCB at the end of the incubation, and 2,2'- and 2,6-CB increased from 1 percent to 14 percent. The observed dechlorination pattern was very similar to environmental dechlorination pattern C, one of the three dechlorination patterns most commonly seen in the region of the Hudson River from which the inoculum was obtained.[25,26]

In more recent experiments[29] conducted under similar conditions (again with Hudson River sediment), extensive dechlorination of Aroclors 1242 and 1248 (500ppm on the basis of dry sediment weight) was seen in only eight weeks. By 12 weeks, most tetra- and pentachlorobiphenyls were 80 to 90 percent depleted and *ortho*-substituted mono- and dichlorobiphenyls had accumulated. Dechlorination of Aroclors 1254 and 1260 was also observed but at much slower rates. When the same sediments were incubated with biphenyl, 2-CB, 2,2'-CB, or 2,6-CB, no dechlorination or degradation was observed, indicating that these compounds are terminal dechlorination products for the methanogenic consortia. Two of the more toxic PCB congeners, 3,4,3',4'- and 2,3,4,3',4'-CB were individually added to sediment incubations with Aroclor 1242. Both were completely removed by dechlorination.

Two other laboratories have also observed dechlorination of Aroclors 1242[31,32] and 1260[31] in laboratory experiments with Hudson River sediment incubated under methanogenic conditions. In these experiments, several distinct dechlorination patterns were observed under slightly different culture conditions. Taken in various combinations, these dechlorination patterns account for the environmental dechlorination patterns previously reported for Hudson River sedi-

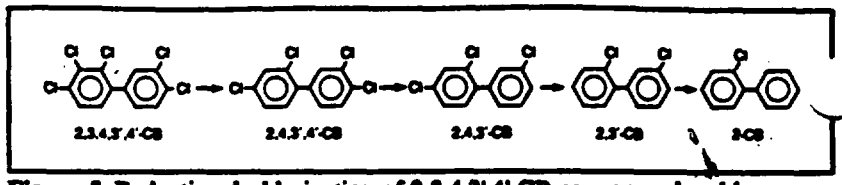

Figure 5. Reductive dechlorination of 2,3,4,3',4'-CB as proposed and later confirmed in the laboratory.

ments[25,26,33] and suggest that the Hudson River sediments contain several different populations of anaerobes that are able to dechlorinate PCBs.

Dechlorination of Aroclor 1242 in Silver Lake sediment has now been observed in the laboratory, but at slower rates than in Hudson River sediment.[29,31]

Dechlorination studies with single congeners have also been initiated. These studies have confirmed that the dechlorination of 2,3,4,3',4'-CB (one of the more toxic congeners) occurs by stepwise removal of all the *meta*- and *para*-chlorines, resulting in the formation of 2-CB, a congener that is easily metabolized by aerobic bacteria and by many higher organisms, including man. Several different sequences of dechlorination of this congener have been seen depending on the incubation conditions, particularly the carbon source and the reduction potential of the medium.[30] The sequence most often seen in Hudson River sediment (Figure 5) is in fact the pattern of dechlorination predicted for this congener based on analysis of the dechlorinated PCBs in Hudson River sediments.[34] The major intermediates are 2,4,3',4'-, 2,4,3'-,and 2,3'-CB

Biodegradation of PCBs in Anaerobic Sediments

Rhee and co-workers[35] have recently reported evidence for the biodegradation of endogenous PCBs (in untreated anaerobic sediments from the Hudson River) incubated in the laboratory under an N_2 atmosphere for seven months.[35] Statistically significant decreases on the order of 33 to 63 percent were observed in congeners ranging from

mono- to tetrachlorobiphenyls. Notably, the PCBs in the sediment used in this experiment were already extensively dechlorinated. Sixty-one percent of the PCB was composed of only three congeners: 2-, 2,2'- and 2,6-CB. These congeners have been repeatedly described as terminal dechlorination products in the Hudson River sediments.[24-26,28,29] In the untreated sediment, 2-CB was decreased by 63 percent and 2,2'-CB by 49 percent over the course of the experiment. Amending the sediment with biphenyl had little effect on the degradation of the mono- and dichlorobiphenyls but significantly enhanced the biodegradation of the more highly chlorinated congeners.

No biodegradation products have been identified from the experiments just described.[35] Clearly, this needs to be done. There are various reports of the anaerobic degradation of other halogenated compounds, but in all cases the biodegradation occurred only after complete dehalogenation.[36-39] Although the authors found no evidence of dechlorination, it is possible that 2-CB, 2,2'-CB, and other congeners in these sediments were completely dechlorinated to biphenyl prior to biodegradation. Regardless of the mechanism, these results suggest that the bacterial population indigenous to the Hudson River sediments has the ability to mineralize PCBs.

It should be noted that all reported observations of reductive dechlorination of PCBs occurred with methanogenic consortia. In contrast, the anaerobic incubations that resulted in biodegradation were not methanogenic, although the reduction potential was very low (-210 to -310mv as indicated by a platinum electrode versus a silver/silver chloride electrode).

Discussion

Research over the past decade has demonstrated that aerobic bacteria are versatile and effective agents for biodegradation of PCBs. Four strains of bacteria representing four different species have been shown to oxidize many tetra- and pentachlorobiphenyls and some hexachlorobiphenyls. It has been shown that at least two genetically distinct classes of PCB-degradative enzymes exist and that the PCB

congener reactivity preferences for these two classes complement each other. Thus, treatment of Aroclor 1242 with a mixture compo of one organism of each class degrades nearly all of the componen of the Aroclor. Laboratory experiments have demonstrated that both *Acinetobacter* sp. P6 and *A. eutrophus* H850 can each degrade approximately one third of the total PCB in Aroclor 1254. Because the congener selectivity patterns of these two strains complement each other, it is to be expected that a mixture of the two organisms would be even more effective.

There are at least four primary factors limiting the degradation of PCBs in soil:

1. Degree of chlorination of the PCB. Generally, PCBs with five or more chlorines are more refractory than the lower congeners, yet recent studies on the biodegradation of Aroclor 1254 show considerable promise.[5,7] Genetic engineering may permit the development of recombinant organisms with increased degradative capabilities.

2. Solubility and bioavailability of the PCB. In order to be degraded, the PCBs must come in contact with the cells. This problem can be minimized under incubation conditions that involve rapidly mixing or shaking the contaminated soil with cell suspensions under controlled conditions (essentially a small bioreactor). Mixing may also be effective for *in situ* treatment. The presence of oil or other organic pollutants may increase the problem of availability due to partitioning of the PCB into the organic phase. This could be alleviated through identification of effective bacterial strains for degradation of these contaminants.

3. Inability of the PCB-degrading bacteria to use PCBs (except monochlorobiphenyls) as growth substrates. This means that the PCB-degraders have no selective advantage over other soil bacteria. Furthermore, high levels of PCB-degradative activity have not been achieved unless the cells are grown on biphenyl. In fact, the best activity has been found when cells are actively growing on biphenyl. There are two ways that this

problem might be overcome. First, biphenyl or another suitable carbon source might be added to the contaminated soil. Research would be required to identify alternative growth substrates. Second, genetic engineering might be used to combine the pathways for PCB biodegradation and chlorobenzoic acid degradation in a single organism in order to develop recombinant strains capable of mineralizing PCBs.

4. *Temperature and moisture conditions. In situ* treatment of PCBs will require developing ways of preventing wide variations in temperature and moisture. Some organisms may be more tolerant than others of such fluctuations and might be good candidates for the basis of new strains developed through genetic engineering.

The prospects for PCB removal in aquatic sediments are excellent. It appears that anaerobic dechlorination of PCBs in both freshwater and marine sediments is widespread. Furthermore, it appears that several populations of microorganisms may be involved. Like the aerobic PCB-degrading bacteria, these populations exhibit different congener selectivity patterns.

Reductive dechlorination of PCBs in anaerobic sediments has now been confirmed in three different laboratories. Efforts are needed to determine the optimal culture conditions for dechlorination (carbon source, mineral requirements, temperature) and the conditions under which the organisms responsible can be enriched and possibly isolated. Ideally, the dechlorinating organisms should be isolated and characterized. At a minimum, conditions must be established for maintaining stable dechlorinating consortia, preferably on a defined medium in the absence of sediment.

There is a need to monitor the progress of dechlorination in sediments where it is known to be occurring (such as the Hudson River) in order to identify the limitations of the dechlorinating system that is operative in the sediment. Because different populations exhibit distinct congener selectivity patterns, it may be possible to inoculate sediment with a second population of PCB-dechlorinating bacteria that will aid the activity of the endogenous population.

In addition, efforts are needed to analyze other aquatic sediments for evidence of reductive dechlorination of PCBs. If evidence of dechlorination is not found or if it is not occurring rapidly enough, laboratory experiments should be done to determine if there are conditions under which reductive dechlorination of PCBs can be stimulated in the sediment. This may require the addition of an essential nutrient (carbon source, phosphate, nitrogen source, trace metals) or may require inoculation with dechlorinating microorganisms from another sediment.

At the time of this writing no evidence has been found for dechlorination of the *ortho*-chlorinated congeners that accumulate as a result of the removal of *meta*- and *para*-chlorines from more highly chlorinated congeners. These congeners (2-, 2,2'-, 2,6-, 2,6,2'-CB) can all be degraded by naturally occurring aerobic bacteria that were originally isolated from the same sediments in which the dechlorination occurred.[3,5] Therefore, it should be possible to develop conditions to promote aerobic degradation of these congeners by introducing oxygen. The benefit of such a sequential anaerobic-aerobic treatment is apparent from Figure 4. Further efforts in this area are already in progress.[32] Alternatively, anaerobic populations that can completely dechlorinate or biodegrade the *ortho*-chlorinated lower congeners may already exist in the sediments. Rhee and co-workers[35] observed significant depletion of these congeners in unamended Hudson River sediment incubated in the laboratory. Further experiments are needed to clarify the mechanism of the depletion of these congeners.

In summary, it appears that natural microbial populations ready have the capacity to biodegrade most if not all of the PCBs that contaminate the environment. Through research directed at understanding the growth requirements of these populations, it should be possible to accelerate biodegradation of the PCBs that persist as environmental contaminants.

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Discussion

Kamely: Have you studied the fate of the biphenyl-degrading bacteria? What happens to them when they finish degrading the biphenyl? Do they die off or can you recover them anytime you add biphenyl again?

Bedard: That experiment has not been done. That was Brunner and Focht's work. They did not report on trying to revive the bacteria.

Kamely: Has anybody tried to add nutrient or fertilizers or enhancers with these PCB-degrading bacteria to see if they can enhance the rate of degradatir

Bedard: No, they have not. One point that I did not make is that the P degraders will not degrade PCBs optimally unless they are grown on biphenyl. That is, the cells are not fully "turned-on" unless they are grown on biphenyl. Biphenyl is the only substrate that we know that can induce maximal PCB-degradation activity. Focht and Brunner found the same thing. When they inoculated the soil with bacteria and did not add biphenyl, they got minimal degradation of PCBs.

Ribbons: I was very interested in the dihydroxylation you showed of the 2,5,2',5'-tetrachlorobiphenyl molecule. You described it as a 3,4-dihydroxylation. That is very important for chemists. I would suggest to you that, in fact, it is still just a 2,3-dihydroxylation with respect to the chlorines as opposed to the other aromatic ring, and so it really looks like the toluene system with a great big substituent at the bottom.

Bedard: Yes, it could be thought of that way, but it is a novel position of attack on the PCB.

RESEARCH AND DEVELOPMENT PROGRAM FOR THE DESTRUCTION OF PCBs

Ninth Progress Report
June 1, 1989 – July 31, 1990

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Submitted by

Herman L. Finkbeiner and Stephen B. Hamilton

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EXECUTIVE SUMMARY

The organization of this year's report represents a change from past reports. For the first time, research on anaerobic reductive dechlorination is presented first. This change was made both to reflect the increased amount of research activity in this area over the past several years and because anaerobic dechlorination is the first step in a two-step anaerobic-aerobic process sequence with the potential to degrade completely even the most highly chlorinated Aroclor mixtures. The anaerobic research presented here continues to focus on the characterization and isolation of the individual microorganisms which make up the PCB-dechlorinating consortia that have now been found in many anaerobic environments. Efforts in this area include the characterization of dechlorination patterns of consortia found in different sediments and the development of techniques to transfer activity from one sediment matrix to another. In addition, work on measuring the reduction in toxicity of PCB mixtures effected by reductive dechlorination is also presented. A second goal of this year's research is to prepare for the eventual field application of this technology. In that light, the dechlorination rate of endogenous PCBs is studied and bioavailability issues explored. Preliminary results from a laboratory reactor designed to model the conditions in an actual river are also presented. Laboratory results of sequential anaerobic-aerobic processes are presented and provide a transition back into aerobic biodegradation. Genetic studies of aerobes indicate that engineered organisms may have significant advantages over natural isolates in stability of the genes that encode for PCB-degrading activity. Research on PCB-degrading aerobes that can survive in anaerobic conditions is also presented, along with work aimed at gaining a clearer understanding of the aerobic metabolic pathway. Finally, a progress report is presented on a state-of-the-art analytical method for quantitating PCB congeners present at low levels in Aroclor mixtures which cannot be measured by traditional GC analysis. Of particular significance here is the ability to quantitate the more toxic congeners.

ANAEROBIC

A Systematic Study of Reductive Dechlorination of Trichlorobiphenyls in River Sediments. This report describes a study of the dechlorination patterns of six added trichlorobiphenyls by anaerobic consortia in sediments from the Hudson River, Woods Pond (Lenox, MA) and Silver Lake (Pittsfield, MA). The amount of depletion of added congener, the number of chlorines removed, and the dechlorination pattern observed were shown to vary with both the sediment type and added congener. Anaerobes in Woods Pond sediment were able to dechlorinate even the *ortho* chlorines on 246-CB. The addition of a single congener also stimulated the dechlorination of the endogenous PCBs in Hudson River and Woods Pond sediments. In addition, successful transfer of PCB dechlorination activity to an uncontaminated sediment is demonstrated.

Reductive Dechlorination of Aroclors by Anaerobic Microorganisms. This work focuses on the dechlorination of the more highly chlorinated Aroclors, especially Aroclor 1260. Silver Lake sediments dechlorinated all four Aroclors (1242, 1248, 1254, 1260), but the rate and extent was lowest for 1260. Still, these microorganisms proved more capable of dechlorinating 1260 than did Hudson River consortia. This limited dechlorination ability of Hudson River microorganisms on 1260 is probably not due to inhibition by any of the more heavily chlorinated congeners in that Aroclor. In other experiments, the PCB congeners in Aroclor

1242 with greater dioxin-like toxicity were effectively dechlorinated in laboratory assays, reducing the toxicity of the mixture as measured by EROD induction assays by 75%.

Establishment and Characterization of an Anaerobic Aroclor 1242-Dechlorinating Culture. Anaerobic enrichment cultures were established as a first step in obtaining pure cultures or stable consortia capable of dechlorinating PCBs. Pyruvate was demonstrated to stimulate dechlorination of Aroclor 1242 and greatly improve transferability of dechlorination activity by reducing the lag period before the onset of dechlorination. High sediment loadings and the presence of natural organic carbon in the sediment matrix were shown to be important in supporting the dechlorination consortium.

Reductive Dechlorination of PCBs in Sediments from the Hudson River and New Bedford Harbor. The dechlorination of PCBs in these sediments was studied under both methanogenic and sulfate reducing conditions. Dechlorination of added Aroclor 1242 was observed in both sediments under methanogenic conditions, although it occurred much faster in Hudson River sediment. The addition of a mixture of fatty acids as an auxiliary carbon source greatly enhanced the rate of 1242 dechlorination in Hudson River sediment, but had no impact on the dechlorination rate in New Bedford sediment. No dechlorination activity has been observed to date under sulfate-reducing conditions in either sediment.

Anaerobic Dechlorination of Endogenous PCBs in Woods Pond Sediment. Modest natural dechlorination of the endogenous PCBs in Woods Pond sediment has occurred. The goal of this work was to establish conditions which would stimulate greater dechlorination activity in this sediment. This was accomplished through the addition of single congeners (25-34- or 23456-CB) to the sediment in laboratory assays. After addition of the congeners two distinct dechlorination patterns, designated as Pattern H (*para* dechlorination) and Pattern N (*meta, para* dechlorination) of the endogenous PCBs emerged. Dechlorination of endogenous PCBs could not be stimulated by the addition of nutrients unless a PCB congener was added. These results suggest that the growth and activity of a bacterial population capable of dechlorinating PCBs may be severely limited by poor bioavailability of the endogenous PCBs.

Anaerobic and Aerobic Biodegradation of Endogenous PCBs. A two-fold increase in the rate of anaerobic dechlorination in Hudson River sediments was observed upon the addition of a simple trace metals mixture to the sediment. This result suggests that low levels of a trace metal in the sediment may limit the rate of PCB dechlorination in the environment today. Experiments on several PCB-contaminated soils and sediments indicate that this anaerobic process will effectively attack endogenous PCB contamination at rates comparable to those for spiked samples. Sequential anaerobic-aerobic degradation was demonstrated on sediments that had been extensively dechlorinated both naturally and in the laboratory, resulting in significant reductions in total PCB concentrations.

Sequential Anaerobic-Aerobic Biodegradation of PCBs. A sequential anaerobic-aerobic biodegradation process has the potential to completely degrade even highly chlorinated Aroclor mixtures. This work demonstrates that lack of organic substrate may be a limiting factor for reductive dechlorination in some sediments. Dechlorination activity was observed in sediments with no past history of PCB contamination, indicating that bacterial populations capable of dechlorinating PCBs may be widely dispersed in the environment. Biphenyl-degrading aerobes were also isolated from several sites, indicating they too are ubiquitous. Although

their PCB-degrading competence was not great, PCB levels in sediment samples were significantly reduced in an anaerobic-aerobic laboratory trial using one of these isolated aerobes.

Hudson River Model. ***In situ*** **application is the ultimate aim of most laboratory bioremediation studies. A large-scale (0.8 tons of sediment) model reactor has been set up in an attempt to stimulate dechlorination in PCB-contaminated sediment while at the same time simulating *in situ* conditions. Methanol injection has been able to initiate anaerobic conditions in the reactor without the use of a reducing agent. Active dechlorination is taking place spatially and temporally within the reactor. The similarity of these results to batch incubations suggest it will be possible to translate small-scale work to *in situ* conditions.**

Differentiation of Anaerobic Microbial Dechlorination Processes. The PCB congener distribution alteration patterns by PCB-contaminated aquatic sediments or anaerobic cultures inoculated within such sediments have already suggested the operation of some 13-15 different anaerobic microbial dechlorination systems. In order to provide objective criteria for distinguishing among such systems, the specific differences in the range of PCB congeners attacked by each have now been tabulated. Thus far, it would appear that each PCB-contaminated drainage basin exhibits its own distinctive assemblage of PCB-dechlorinating microorganisms, presumably in response to the availability of PCBs as terminal electron acceptors for anaerobic metabolism.

AEROBIC

Genetic Studies of Bacterial PCB Degradation: 1. Bacterial Survival on PCB-Containing Soil 2. Analysis of *BPH* Genes. The ability to develop a practical process for aerobic PCB biodegradation depends largely upon obtaining organisms with the ability to survive and maintain activity in PCB-containing soils. Experiments with both naturally occurring LB400 and recombinant FM4560 organisms showed good survivability (detectable populations after 28 days) on soil in laboratory tests. While instability of the *bph* genes were noted in the LB400, FM4560 was found to be stable if antibiotic selection for the plasmid was maintained. Evidence for at least two separate promoters in the Bbph region suggest that these genes are not organized as a operon.

PCB Biodegradation and Nitrate Reduction. Several organisms which can grow on biphenyl or a biphenyl/Aroclor mixture and use oxygen or nitrate as an electron acceptor have been isolated. One of these organisms, a *Comamonas testosteroni*, grows to high densities on biphenyl/Aroclor 1242 under aerobic conditions, with the formation of chlorobenzoic acids as metabolic products. Efforts are underway to understand the relationship between this metabolic pathway and the one which mediates nitrate reduction.

Availability of PCBs in Soils and Sediments to Surfactant Extraction and Aerobic Biodegradation. An analogy between surfactant extraction and aerobic biodegradation is used to study the effect of natural organic matter (NOM) on PCB availability in soils and sediments. PCB equilibrium partitioning into the NOM phase is demonstrated to have a detrimental effect on the extent and efficiency of PCB removal for both of these processes. Oil and grease also form a secondary phase in soils and sediments, and are shown to have a similar negative impact on biodegradation. A treatment is presented which can enhance PCB

removal by modifying the NOM present in the soil or sediment matrix.

Synthesis of Proposed Intermediates in the Aerobic Degradation of PCBs; Ring-Fission Products. A major pathway for the destruction of PCBs by aerobic microorganisms closely resembles the pathway for oxidation of biphenyl itself, proceeding through a ring-fission intermediate which is further degraded in at least two different ways. Alternatively, the oxidation of 23-3-CB is proposed to proceed through a tri-keto acid intermediate. These intermediates have now been chemically synthesized allowing their reactions under non-enzymic conditions, as well as their transformation by microorganisms, to be explored. To date, non-enzymic hydrolysis of these intermediates has yielded both benzoic acid and acetophenone, which are the major aromatic products in the aerobic degradation of many PCBs, and oxalic acid, which has not previously been isolated as a metabolite of biphenyl or PCBs.

ANALYTICAL

The Quantitation of Polychlorinated Biphenyls by Gas Chromatography and Tandem Mass Spectrometry. Using a selected reaction monitoring mode of operation in a GC/MS/MS experiment and monitoring the exchange reaction of oxygen for chlorine, PCB congeners are able to be quantitated even when not chromatographically resolved. The method couples the high specificity of MS/MS with the excellent detection limits of selected ion monitoring coupled with negative chemical ionization. This ability is critical to the quantitation of toxic congeners present at low levels in PCB mixtures, particularly when other congeners co-elute with these in standard GC/MS methods.

Chapter 1

A SYSTEMATIC STUDY OF REDUCTIVE DECHLORINATION OF TRICHLOROBIPHENYLS IN RIVER SEDIMENTS

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INTRODUCTION

In 1989, Abramowicz et al., reported dechlorination of added polychlorinated biphenyl (PCB) congeners in river sediments containing a background level of a complex PCB mixture [GE Report, 1989]. They determined the pattern of dechlorination for each added congener and quantified the dechlorination rate. They also noted a specific stepwise dechlorination of 2,3,4-3,4-CB to 2-CB within Hudson River sediment.

This report describes a systematic study of the dechlorination of added trichlorobiphenyls with all of the chlorines on one ring by sediments from the Hudson River, Woods Pond and Silver Lake. An earlier study of 2,3,4-CB added to Woods Pond sediment indicated that reductive dechlorination had occurred, and at a rate considerably more rapid than dechlorination of an added Aroclor mixture in Hudson River sediment [H. Van Dort, personal communications]. The goal of this work was to understand the patterns and rates of dechlorination of these single congeners by the microbial populations in the river sediments listed above. The results of the study may help to characterize the microorganisms capable of mediating reductive dechlorination of PCBs and have led to successful passage of PCB-dechlorinating activity.

RESULTS AND DISCUSSION

Experimental Procedure

PCB-contaminated river sediments from the Hudson River downstream of Fort Edward, NY (H7 site), Silver Lake (Pittsfield, MA) and Woods Pond (Lenox, MA) were collected and stored until use in sealed vessels at 4°C. Cultures were prepared and sampled within a glove box containing an oxygen-free atmosphere (95% N_2, 5% H_2). Batches (300-500 mL) of sediment and RAMM mineral salts reduced with 0.1% cysteine hydrochloride were prepared at a 2:3 ratio (volume:volume). The batches were stirred vigorously and 30 mL volumes were removed to 50 mL serum vials. A trichlorobiphenyl stock solution (70 mM in acetone) was added to each serum vial, making the concentration of a single congener addition at 350 μM. The added congeners are shown in Figure 1-1. After congener addition, each vial was

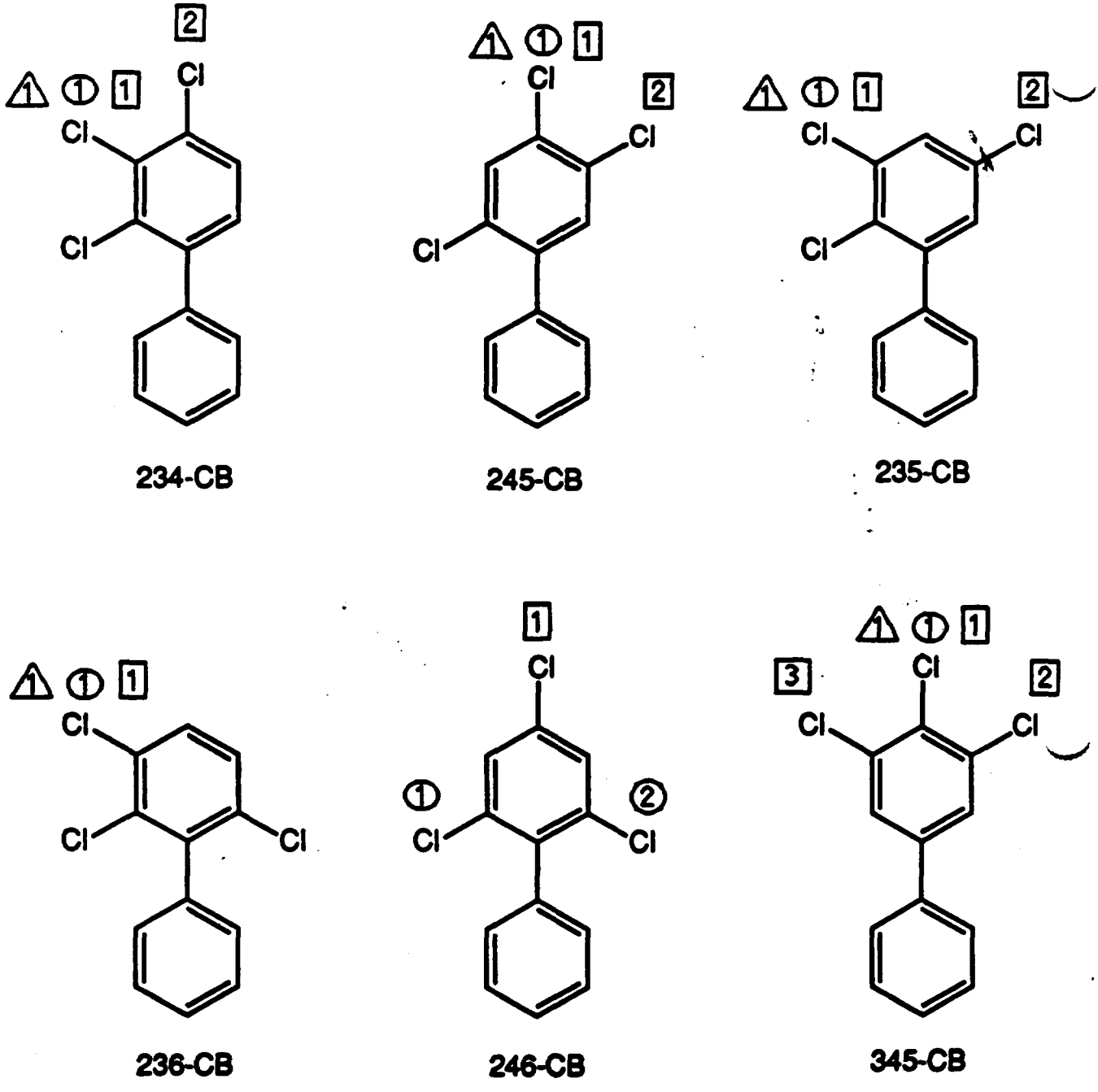

Figure 1-1. Reductive dechlorination of trichlorobiphenyls by river sediments. The number above each chlorine indicates the order of chlorine removal within the particular river sediment. The boxed numbers represent Hudson River sediment; the circled numbers represent Woods Pond sediment; and the triangulated numbers represent Silver Lake sediment.

vortexed for 2 minutes and two 1 mL samples were removed as time zero points and stored at -20°C. All vials were crimp sealed and removed from the glove box.

Four vials were prepared for each added congener and river sediment. Two of each group of four were immediately autoclaved for 3 hours (the autoclaved control samples). All vials were stored stationary at 24°C in the dark. At particular timepoints, vials were placed in the glove box, vortexed for 30 seconds, immediately uncapped and 1 mL samples were placed in 8 mL glass vials. The culture vials were then crimp sealed and returned for further incubation. Each 8 mL glass vial was extracted with 5 mL of diethyl ether and ~0.5 mL mercury, added to precipitate the molecular sulfur in the sediment sample. The ether extract was analyzed by capillary gas chromatography (GC) using a fused silica capillary column (30 m x 0.25 mm ID) coated with a 0.25 μm bonded liquid phase of DB-1 (polydimethylsiloxane, J&W Scientific, Folsom, CA) and an electron capture detector at 300°C.

For five of the six added trichlorobiphenyls, the results are reported as the percent peak area of each congener product relative to the total peak areas for the added congener and its products (no account was made for the response factor of each congener). The results in experiments with 3,4,5-CB were determined using standard solutions of 3,4,5-CB and each of its congener products. This was done because 3-CB has an extraordinarily low response factor relative to the other congeners and would result in a significant underprediction of 3-CB in the mixture if the percent peak area method was used.

Dechlorination of Added Congener Within Hudson River Sediment

Figure 1-1 is a descriptive diagram of the dechlorination patterns for the added congeners by each river sediment. Dechlorination of every added trichlorobiphenyl was observed in Hudson River sediment collected downstream of Fort Edward, NY. However, only *meta* and *para* chlorines were removed. Since 3,4,5-CB does not contain *ortho* chlorines, all of the chlorines were removed in Hudson River sediment resulting in the accumulation of biphenyl (confirmed by mass spectra (MS) analysis after gas chromatography). Figure 1-2 shows the pattern of 3,4,5-CB dechlorination. The figure shows a sequential dechlorination of the trichlorobiphenyl to dichlorobiphenyl, then of the dichlorobiphenyl to monochlorobiphenyl, and finally of the monochlorobiphenyl to biphenyl.

The apparent rate of trichlorobiphenyl dechlorination was very rapid as compared to what has been seen with the addition of the Aroclor mixture [GE Report, 1989]. Dechlorination of every added congener was observed by the first timepoint at 2 weeks (a lag time <2 weeks) and >80% of the added trichlorobiphenyl was converted to product by 5 weeks. The pattern of chlorine removal seemed to be dictated by the spatial arrangement of the chlorines in the PCB molecule, such that the inner chlorine was preferentially removed first (e.g., 2,4,5-CB goes to 2,5-CB, 3,4,5-CB goes to 3,5-CB).

Dechlorination of the endogenous PCBs in Hudson River sediment occurred concomitant with dechlorination of the added congener. 2,3,4-CB, 2,4,5-CB, and 2,4,5-CB all induced a

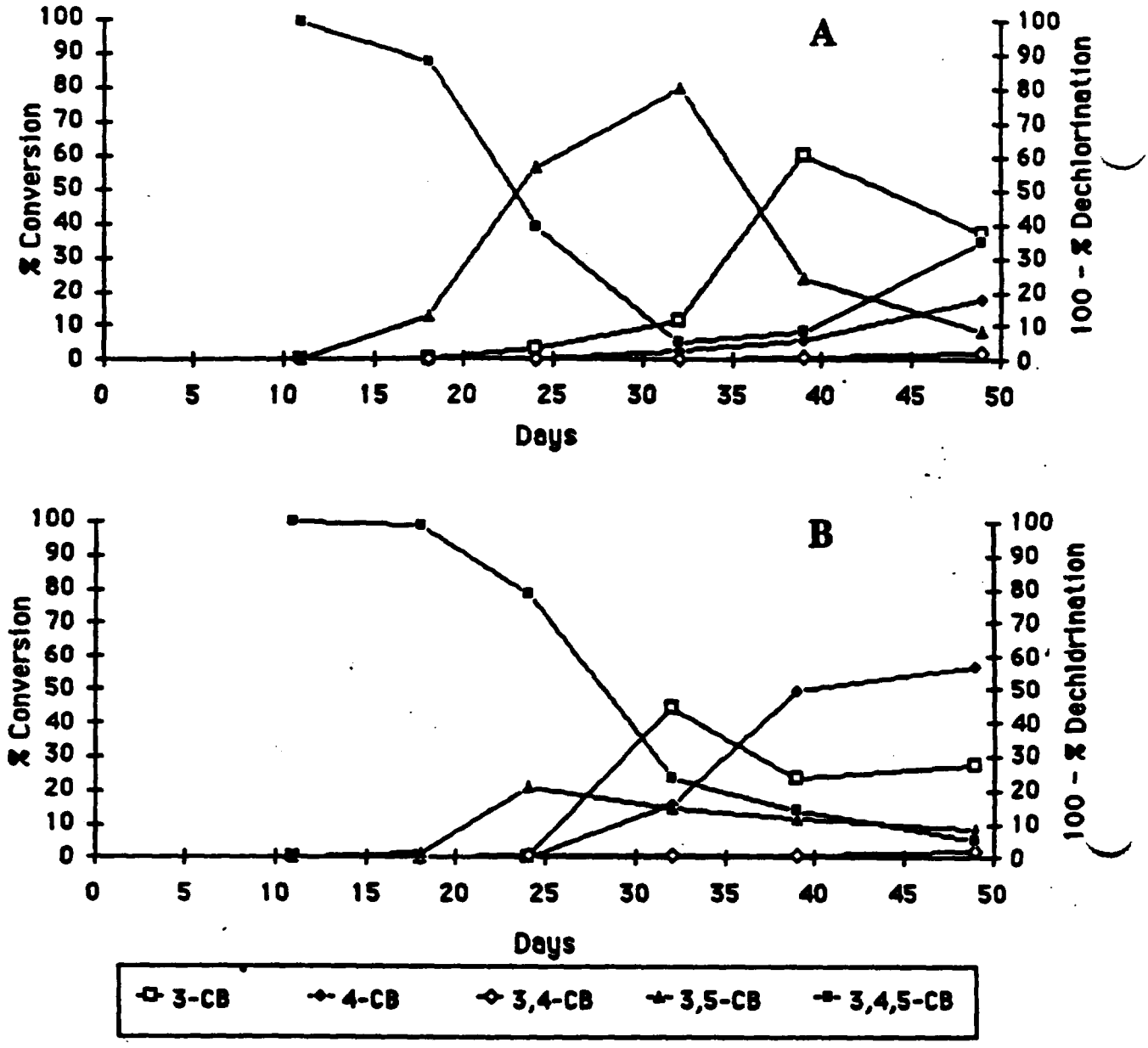

Figure 1-2. Stepwise dechlorination of added 3,4,5-CB in PCB-contaminated Hudson River sediment. Panels A and B are duplicate experiments showing stepwise dechlorination of added 3,4,5-CB in PCB-contaminated Hudson River sediment. The graphs showing the conversion of 3,4,5-CB to the products are defined in the box below Panel B and relate to the left ordinate in each panel. Biphenyl, which is the ultimate dechlorination product, it is not described in this figure because it could not be measured under the GC analysis conditions. The data shown above at 48 days after 3,4,5-CB addition does not take into account the significant amount of biphenyl that has accumulated from dechlorination.

Pattern Q activity [GE Report, 1989], whereas 2,3,6-CB induced a Pattern M activity. Unfortunately, the Hudson River sediment used in the 2,3,5-CB and 3,4,5-CB experiments came from a batch of sediment collected more recently from the H7 site. This sediment contained such a highly dechlorinated endogenous PCB content so as to preclude the observation of any added congener-induced dechlorination pattern.

Two groups of experiments were run using Hudson River sediment collected from a spot upstream of the PCB deposits (Spier Falls, Hudson River). The groups differed by the added congener; one group had 2,3,6-CB and the other group had 2,4,6-CB. After a 3-week lag time, 2,3,6-CB dechlorination to 2,6-CB was observed and dechlorination was >80% complete by 8 weeks. No dechlorination of 2,4,6-CB has been observed after 8 weeks of incubation with Spier Falls sediment. The dechlorination of 2,3,6-CB in Spier Falls sediment is surprising, since no dechlorination activity was observed in this sediment in the past when an Aroclor mixture was added [GE Report, 1989].

Dechlorination of Added Congener Within Silver Lake Sediment

A descriptive diagram of the dechlorination patterns for the added congeners in Silver Lake sediment is shown in Figure 1-1. This sediment can be described as a black mayonnaise with a high content of organic contaminants, including Aroclor 1254 and 1260. Dechlorination of the endogenous PCBs has been noted in the environment [Brown et al., 1987b] as well as in the laboratory [GE Report, 1989]. Dechlorination of five of the six added trichlorobiphenyls was observed in Silver Lake sediment. No dechlorination of 2,4,6-CB has been observed after over 8 months of incubation at room temperature. For each of the added congeners that were dechlorinated, only one chlorine was removed, with accumulation of the dichlorobiphenyl product. As seen in Hudson River sediment, the inner chlorine was preferentially removed (e.g., 2,4,5-CB goes to 2,5-CB). The lag time and rate of the added congener dechlorination varied. Dechlorination of 2,3,4-CB to 2,4-CB and 3,4,5-CB to 3,5-CB was observed by the first timepoint at 2 weeks and was >80% complete by 5 weeks (similar to the results in the Hudson River sediment). Lag times of 4 weeks for 2,4,5-CB dechlorination, 8 weeks for 2,3,6-CB dechlorination, and 14 weeks for 2,3,5-CB dechlorination were observed. For each of these congeners dechlorination was >80% complete by 5-7 weeks after the lag period ended. The dechlorination of the added congeners seemed to have little observable effect on the dechlorination of the endogenous PCBs.

Dechlorination of Added Congener Within Woods Pond Sediment

A descriptive diagram of the dechlorination patterns for the added congeners in Woods Pond sediment is shown in Figure 1-1. In several respects the dechlorination patterns were very similar to those observed within Silver Lake sediment. However, there were some notable exceptions. Most often, as was the case with Silver Lake sediment cultures, added trichlorobiphenyl was only dechlorinated to dichlorobiphenyl. The lag time for dechlorination

of 2,3,4-CB, 2,4,5-CB and 3,4,5-CB was 2-3 weeks, with >80% dechlorination of each congener by 4-5 weeks. Lag times of 5 weeks for 2,3,6-CB dechlorination and 12 weeks for 2,3,5-CB dechlorination were observed, with >80% dechlorination of each congener by 3-4 weeks after the onset of dechlorination. It was noted that 2,3,6-CB addition stimulated significant dechlorination of the endogenous PCBs in Woods Pond sediment. Until very recently, little natural dechlorination of the endogenous PCBs had been observed [GE Report, 1990]. Dechlorination of the endogenous PCBs in Woods Pond sediment has also been observed in the laboratory by Bedard and co-workers after the addition of a single congener.

Recently, it was discovered that 2,4,6-CB is being dechlorinated by Woods Pond sediment after a lag time of ~24 weeks. Most surprising was that both the *ortho* chlorines were being removed. Figure 1-3 shows mass spectra analysis after gas chromatography of the 2,4,6-CB culture at time zero days and after 220 days. *Ortho* dechlorination has previously been noted in Silver Lake sediment [Brown et al., 1987b] and in Woods Pond sediment after the addition of 2,3,5,6-CB [D. Bedard, personal communications].

An experiment was done with Woods Pond sediment to see if heating for a brief period of time (pasteurization) would selectively bring up a PCB-dechlorinating microbial population. Four serum vials containing Woods Pond sediment, RAMM mineral salts reduced with cysteine hydrochloride, and 350 μM 2,3,4-CB were prepared in the same fashion as described above. The four vials were heated in an 80°C water bath for 30 minutes and then kept stationary in the dark at 24°C. Two other serum vials were prepared in the same fashion, but without heating. Two of the four heated vials and the two unheated vials were sampled every 10 days starting at week 2. In the unheated experiments, 2,3,4-CB was dechlorinated to predominantly 2,4-CB with <10% 2,3-CB and >80% 2,4-CB by 5 weeks (lag time of ~2 weeks). In the heated vials, 2,3,4-CB was dechlorinated exclusively to 2,3-CB with >80% 2,3-CB by 10 weeks followed by further dechlorination to 2-CB (lag time of ~5 weeks). Figure 1-4 is a diagram of 2,3,4-CB dechlorination by heat-treated Woods Pond sediment.

Passage of PCB-Dechlorination Activity

Stable PCB dechlorinating cultures have been successfully passed in the laboratory. Dechlorination activity of PCB-contaminated sediments from the Hudson River, Woods Pond, and Silver Lake have been passed to a culture medium of Spier Falls sediment. The cultures were passed at 10% by volume to a medium containing dried river sediment from Spier Falls premixed with RAMM mineral salts reduced with 0.1% cysteine hydrochloride (2:3 ratio, volume:volume) and autoclaved for 3 hours. Passage of PCB-dechlorination activity to a river sediment medium previously uncontaminated with PCBs allows a more accurate quantitation of dechlorination activity. In Figure 1-5, a typical passage of activity from a Woods Pond culture to a medium containing Spier Falls sediment is depicted. Upon passage there was a decrease in the dechlorination lag time of about 1 week. A lag time of ~1-2 weeks for a 10% by volume passage was a typical result for passages from all three

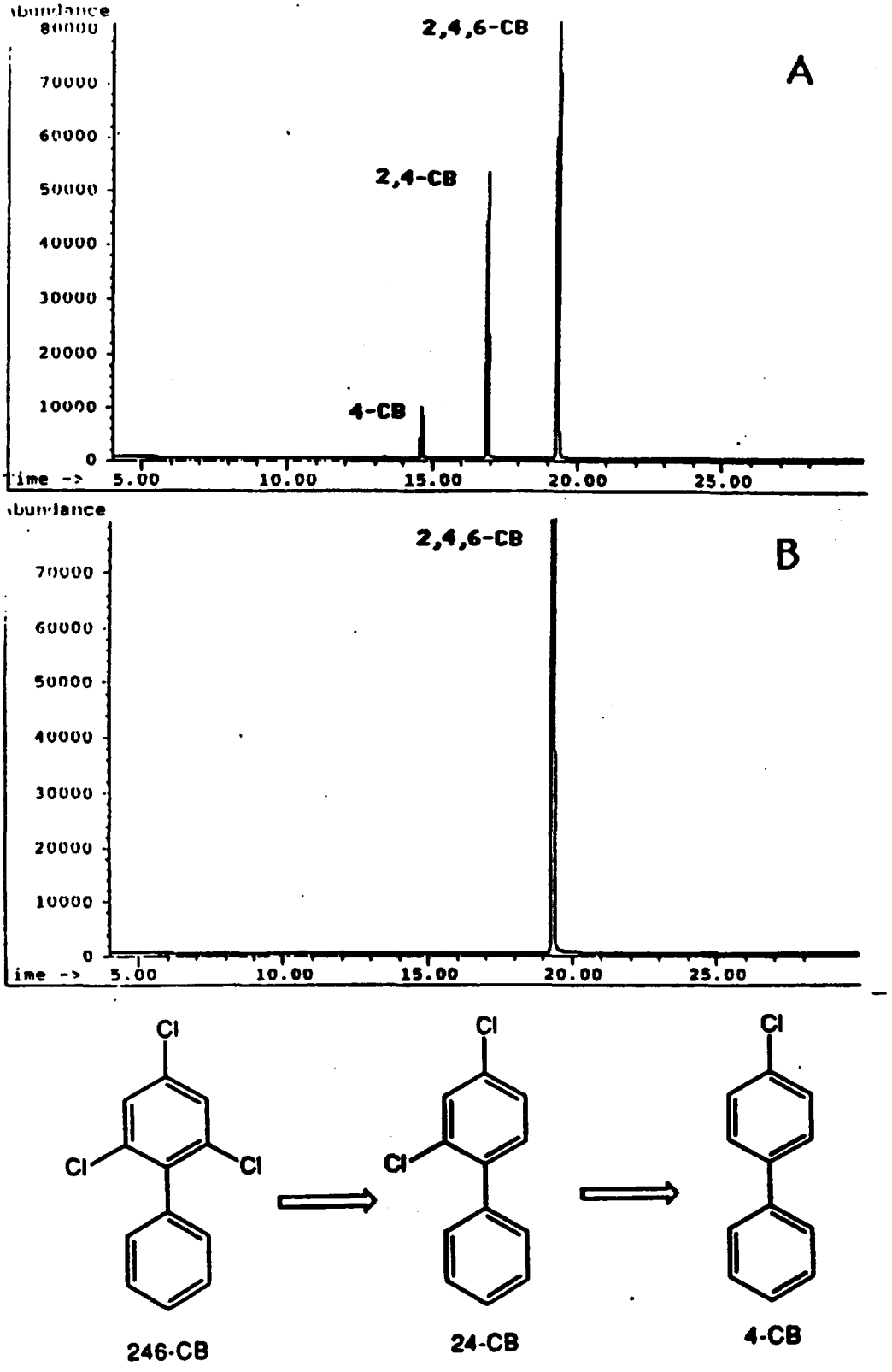

Figure 1-3. *Ortho* dechlorination of 2,4,6-CB added to Woods Pond sediment. Panel A shows ion selective mass spectra analysis after gas chromatography of the dechlorination products of 2,4,6-CB 220 days after addition of Woods Pond sediment. Panel B shows the same analysis of a sample of the experiment taken at time zero days. Below the spectra is a diagram of the dechlorination of 2,4,6-CB.

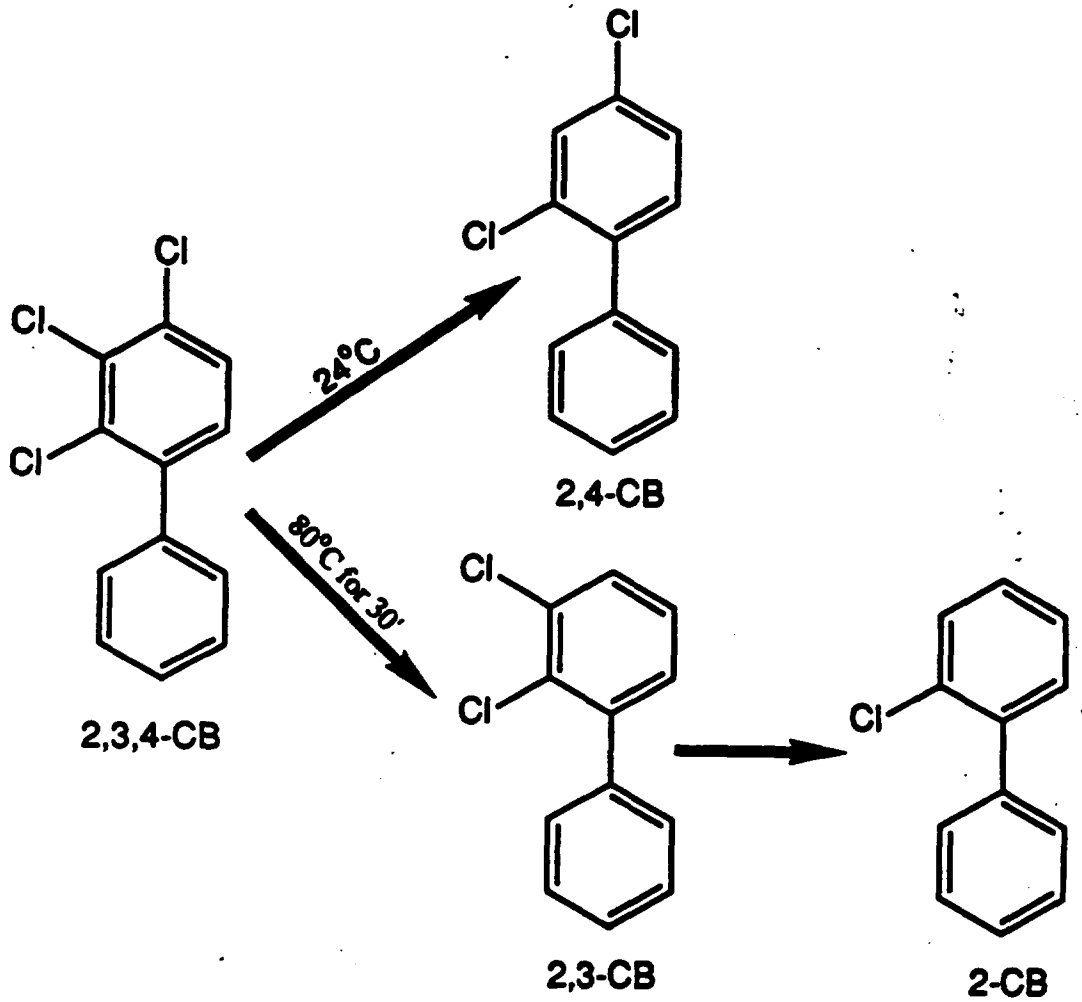

Figure 1-4. Differential dechlorination of added 2,3,4-CB by differential treatment of woods Pond sediment. Dechlorination of 2,3,4-CB added to Woods Pond sediment and incubated at 24°C resulted in conversion to 2,4-CB. A different dechlorination pattern resulted when 2,3,4-CB added to Woods Pond sediment was incubated at 80°C for 30 minutes followed by incubation at 24°C.

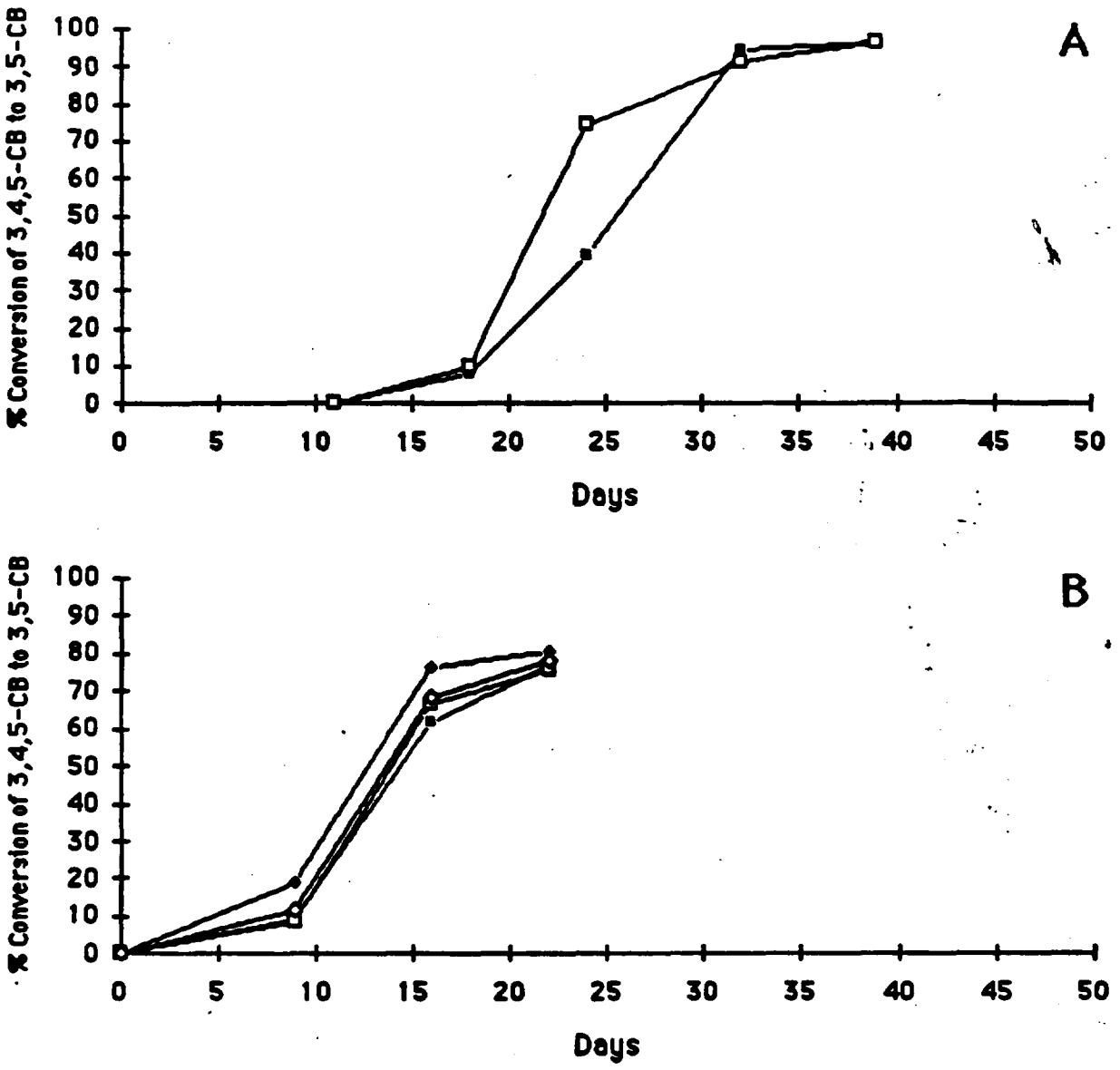

Figure 1-5. Passage of PCB reductive dechlorination activity. Panel A shows dechlorination activity on added 3,4,5- CB in Woods Pond sediment. Panel B describes the same dechlorination activity as shown in Panel A, after a 10% by volume pass from Woods Pond sediment to a Hudson River sediment medium which previously was uncontaminated with PCBs. The multiple curves represent duplicate samples.

sediments. The minimum lag time for passages originating from any of the three PCB-contaminated sediments was ~1 week. Successful multiple passes have been done starting with PCB-contaminated Hudson River sediment and carried with each of the added trichlorobiphenyls. This has resulted in a >1000-fold dilution of the original sediment [M. Brennan, personal communications].

SUMMARY AND CONCLUSIONS

The dechlorination rate of an added trichlorobiphenyl within a river sediment is rapid as compared to the Aroclors. Generally, in cultures incubated at room temperature the pattern of trichlorobiphenyl dechlorination resulted in removal of the inner chlorine first. Microorganisms in the Hudson River which are located in PCB-contaminated sediment can mediate reductive dechlorination of all six trichlorobiphenyls to either biphenyl or *ortho* chlorine-containing biphenyls. The addition of a single congener to a PCB-contaminated river sediment will stimulate dechlorination of the endogenous PCBs, with different congeners stimulating different dechlorination patterns. Microorganisms in Hudson River sediment which is not contaminated with PCBs can mediate dechlorination of 2,3,6-CB to 2,6-CB. This implies that extended exposure of a microbial population to PCBs may not be necessary to bring about microbial PCB-reductive dechlorination.

Microorganisms in Woods Pond and Silver Lake mediate reductive dechlorination of five of the six added trichlorobiphenyls to dichlorobiphenyls. Dechlorination of 2,4,6-CB in Woods Pond sediment has also been observed after long incubation periods. Since Woods Pond and Silver Lake sediments contain more highly chlorinated PCBs (Aroclor 1254 and 1260) than does Hudson River sediment, the active PCB-dechlorinating microbial populations may not have the reducing power necessary to mediate dechlorination of a dichlorobiphenyl to a monochlorobiphenyl. Heat treatment of Woods Pond sediment will bring up different dechlorination patterns of the same trichlorobiphenyl. A PCB-dechlorinating microbial population has been brought up which is either resistant to or activated at a temperature of 80°C. This suggests a spore former is present.

FUTURE PLANS

Since successful passage of PCB-dechlorination activity has been achieved, an effort is currently underway to chemically fractionate the Spier Falls sediment to determine the necessary nutrients for dechlorination. Another effort is underway to distinguish cultures brought up at different temperatures. Considerable interest has been taken in the *ortho* declorinating population brought up on 2,4,6-CB in Woods Pond sediment. Can this activity be passed to PCB-contaminated Hudson River sediment where >90% of the PCBs are *ortho* chlorine containing mono-, di- or trichlorobiphenyls [M. Stephens, personal communications]? All of these efforts are aimed at the enrichment, isolation, and characterization of microorganisms capable of mediating reductive dechlorination of PCBs.

Chapter 2

REDUCTIVE DECHLORINATION OF AROCLORS BY ANAEROBIC MICROORGANISMS

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INTRODUCTION

During the past year our research on PCB dechlorination has advanced on several fronts. We have devoted a major effort to the dechlorination of the more heavily chlorinated Aroclors, especially Aroclor 1260. We have examined the dechlorinating capabilities of Silver Lake microorganisms which were environmentally exposed to Aroclor 1260, and attempted to enhance Aroclor 1260 dechlorination by mixing inocula from different sources. We have also determined that the limited ability of Hudson River microorganisms to dechlorinate Aroclor 1260 is not likely due to inhibition by any Aroclor 1260 components. We have also begun to quantify the toxicity reduction that occurs as a result of anaerobic PCB dechlorination, investigated the bioavailability of PCBs in a contaminated soil, and determined the suitability of alternatives to the natural sediments we normally use in our dechlorination assays.

RESULTS

Aroclor Survey

Microorganisms from site F3 in Silver Lake [Brown et al., 1984] were found to dechlorinate all four Aroclors (1242, 1248, 1254, and 1260) tested. Dechlorination of Aroclors 1242 and 1248 was first evident after 4 weeks of incubation, but leveled off shortly thereafter with approximately one *meta* plus *para* chlorine remaining (Figure 2-1). In this respect dechlorination was inferior to that typically obtained with Hudson River microorganisms, which leave as few as 0.2 *meta* plus *para* chlorines when dechlorinating Aroclor 1242 [GE Report, 1989; Quensen et al., 1990]. This difference in performance can be understood from a comparison of dechlorination patterns. The Hudson River microorganisms removed chlorines from both *meta* and *para* positions with the result that the primary dechlorination products (2-CB, 2-2-CB and/or 26-CB, 26-2-CB and 26-26-CB) were substituted in only the *ortho* position(s). The Silver Lake microorganisms, however, removed chlorines primarily from the *meta* positions with the result that *para* substituted products (especially 2-4-CB, 24-2-CB, 24-4-CB and 24-24-CB) accumulated in addition to *ortho*-only substituted products. The Silver Lake microorganisms, however, dechlorinated Aroclor 1260 sooner and more extensively than had previously been observed for Hudson River microorganisms [GE Report, 1989; Quensen et al., 1990]. Thus they seem capable of more extensively dechlorinating the

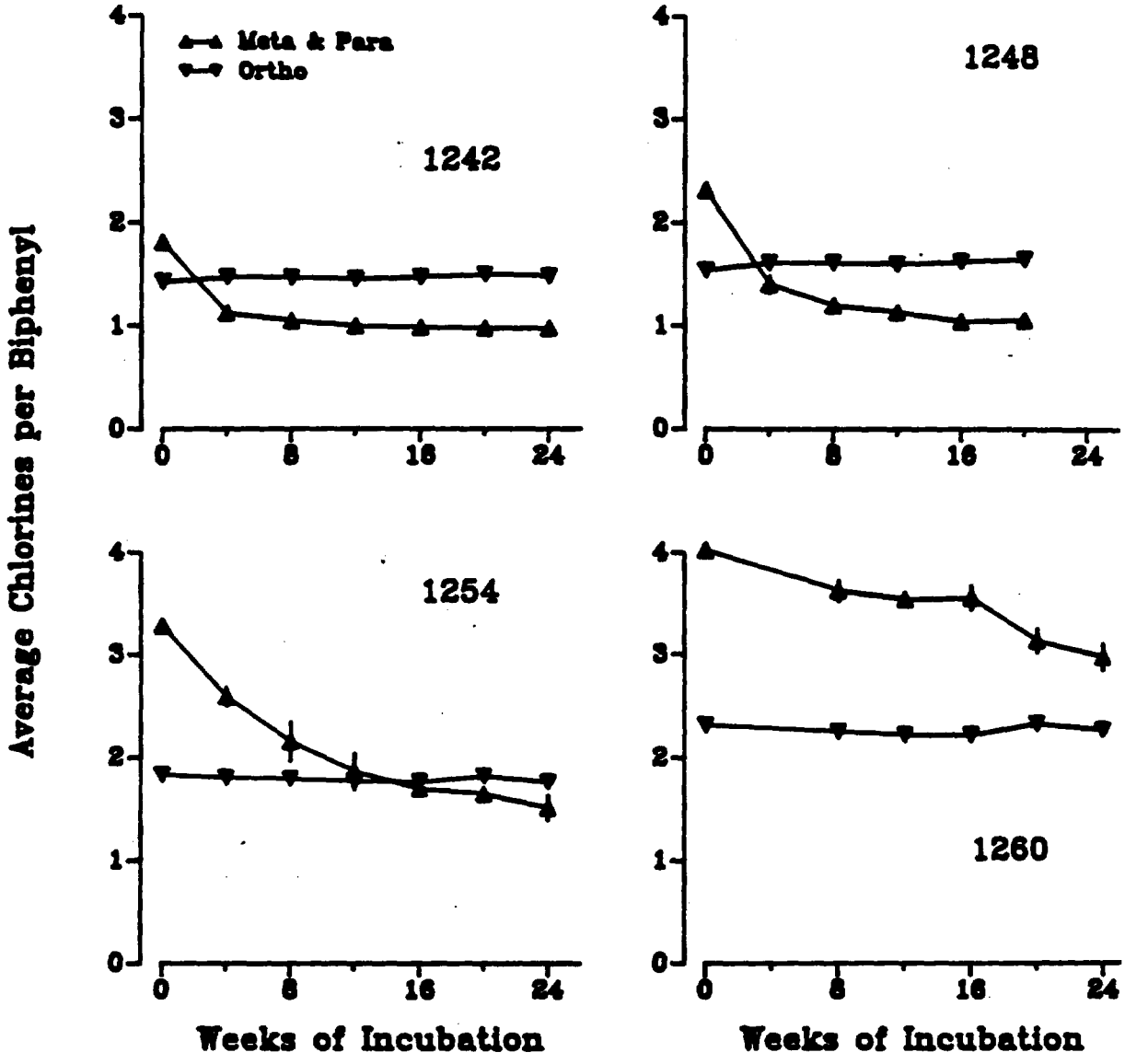

Figure 2-1. Decrease in the average number of chlorines by position for four Aroclors.

more heavily chlorinated PCB congeners. The extent of dechlorination by Silver Lake microorganisms for the congeners represented by each chromatographic peak is given in Table 2-1.

Mixed Inocula

An attempt was made to affect greater overall dechlorination of Aroclor 1260 by using a mixture of Hudson River and Silver Lake microorganisms. The results of the Aroclor surveys [above and GE Report, 1989] demonstrated the complementary nature of the dechlorination activities of microorganisms from these two sources. The Silver Lake organisms more quickly and extensively dechlorinated Aroclor 1260 but left *para* substituted products that should be readily dechlorinated by Hudson River microorganisms. Thus we expected that a mixture of the two organisms should give enhanced dechlorination of Aroclor 1260.

Surprisingly, this did not prove to be the case. The extent of dechlorination of Aroclor 1260 by a mixture of organisms from these two sources was not much greater than for Hudson River microorganisms alone (Table 2-2). An examination of the dechlorination patterns obtained for each treatment suggested that it was primarily the Silver Lake microorganisms that were responsible for dechlorination in the mixed inoculum. These results may be due to competition between the dechlorinating microorganisms from the two sites. Perhaps the Hudson River microorganisms in the mixed inoculum depleted some limiting factor, thereby inhibiting dechlorination by the Silver Lake microorganisms, and at the same time contributed little to overall dechlorination themselves. It was not readily apparent that the Hudson River microorganisms dechlorinated any products from the Silver Lake microorganisms.

TABLE 2-2. Average number of chlorines removed from Aroclor 1260 over time by Hudson River (HR) and Silver Lake (SL) microorganisms alone and in combination.

	Source of Inoculum		
Week	HR	SL	HR & SL
8	0.04	0.14	0.07
16	0.20	0.76	0.19
24	0.24	1.22	0.53

Mixed Aroclors

Because the Hudson River microorganisms dechlorinate Aroclor 1260 so much more slowly than the lesser chlorinated Aroclors, we wondered if any of the PCB congeners present in Aroclor 1260 inhibited dechlorination by these organisms. To investigate this possibility we compared the extent of dechlorination over time for Aroclors 1242 and 1260 added separately and in combination. To allow a determination of quantitative recovery, these experiments

TABLE 2-1. Net decrease (average % for 3 replicates) of each chromatographic peak representing more than 0.1 mole % of each Aroclor after 20 weeks of dechlorination by Silver Lake microorganisms.

Peak	Structure	Aroclor 1242	1248	1254	1260
1	2	+[a]	+	+	+
2	4				
3	2-2 26	+	+	+	+
4	24 25	+	+	+	
5	2-3	45	+	+	
6	2-4 23	+	+	+	
7	26-2	+	+	+	+
8	34 3-4	+	+		
9	25-2 4-4	94	86	0	+
10	24-2	+	+	+	+
11	26-3 236	+	+	+	+
12	23-2 26-4	10	+	+	+
13	35-2 235 26-26	+	+	+	
14	245	78	+	+	+
15	25-3	+	+	+	
16	24-3	+	+	+	+
17	25-4	+	+	+	
18	24-4 246-2	+	+	+	+
19	34-2 23-3 234 25-26	91	76	+	+
20	23-4 24-26	87	91		
21	236-2	88	97		
22	23-26	84	76	+	+
23	25-25 26-35	79	82	59	+
24	24-25	68	66	+	+
25	24-24	31	24	+	+
26	245-2 246-4	nr	nr	nr	nr
27	34-3				+
28	23-25	93	96	93	
29	23-24 236-3 34-4	89	90	11	
30	26-34 234-2 236-4 25-35	88	96	53	+
31	236-26		89		
32	23-23	85	85	+	+
33	245-3 246-24 235-3 246-25	92	63	+	+
34	23-35 235-4	+	+	+	
35	245-4 235-26	97	95	81	+
36	25-34 345-2	98	96	94	
37	236-25 245-26 24-34	93	93	89	70
38	236-24 234-3	57	88	85	+
39	23-34 234-4 236-23 235-25	93	91	67	+
40	245-25 235-24	82	82	81	21
41	245-24	95	91	89	+
42	236-246 2356-3 246-34				

[a] + indicates a net increase

nr = not resolved

TABLE 2-1 (Cont'd)

Peak	Structure	Aroclor 1242	1248	1254	1260
43	245-23 2345-2 2356-26	97	91	91	24
44	234-25 2346-4	89	89	86	54
45	234-24	96	94	92	
46	236-236			79	60
47	34-34 236-34	84	92	93	64
48	2356-25	86	86	85	50
49	235-236 345-25 2346-25	66	80	84	57
50	245-34 236-245 2345-3	33	+		75
51	2356-23 2345-26 2345-4			53	49
52	345-23 2346-23 235-235		91	89	65
53	235-245 2356-35	76	80	43	
54	245-245	71	76	78	50
55	234-236 234-34	87	90	90	59
56	2345-25			81	54
57	2356-236			61	40
58	2345-24		91	80	0
59	2346-236			82	47
60	234-245 2356-34	52	67	80	51
61	2346-34		71	81	46
62	2356-235			79	34
63	2346-235			88	52
64	2356-245 2345-246		31	58	29
65	2346-245			67	35
66	245-345			86	53
67	23456-25				42
68	2345-236 23456-24			68	38
69	2356-234			67	39
70	2346-234 2345-34 2356-2356			78	36
71	2346-2356 23456-246 23456-23			80	32
72	2345-235 23456-35			66	33
73	2345-245			62	47
74	2356-345				23
75	2346-345				
76	23456-236				29
77	2345-234			64	34
78	23456-34				19
79	2356-2345			23	15
80	2345-2346 23456-245			44	18
81	2345-345				
82	23456-234				19
83	23456-2356				
84	2345-2345				
85	23456-345				14
86	23456-2345				
87	Internal Standard (Octachloronaphthalene)				
88	23456-23456				

[a] + indicates a net increase

nr = not resolved

were performed using Balch tubes and a tube was sacrificed for each observation. Two μmole equivalents of Aroclor 1242 or 1260 were added per tube to the treatments receiving only one Aroclor. Two μmole equivalents of each Aroclor (1242 and 1260) were added to the treatment receiving both Aroclors.

There was no evidence that the presence of Aroclor 1260 inhibited dechlorination by the Hudson River microorganisms. The amount of chloride released was additive in the treatment receiving both Aroclors (Table 2-3). It appears that the dechlorination of the Aroclor 1260 congeners by these organisms is inherently slower than that of lesser chlorinated congeners. This is the opposite of what is to be expected from a purely thermodynamic standpoint and suggests that rate of congener uptake by the microorganisms and/or enzyme specificity strongly influence(s) the rate of dechlorination of specific PCB congeners.

TABLE 2-3. Comparison of the extent of dechlorination over time of Aroclors 1242 and 1260 separately and in combination by Hudson River microorganisms.

	μgm Atoms Cl^- Removed from Aroclor		
Weeks	1242	1260	1242 & 1260
8	1.50	0.04	1.40
16	1.74	0.30	2.24
24	2.40	0.72	3.19

Toxicity Reduction

The most toxic of the PCB congeners are generally considered to be the coplanar congeners 34-34-CB, 345-34-CB, and 345-345-CB. In a coplanar configuration, these congeners are structurally similar to 2378-tetrachlorodibenzodioxin (TCDD) and exhibit similar toxicity effects. PCB congeners like these but with a single *ortho* chlorine also have similar toxicity effects but are much less potent.

Because these toxicologically important PCB congeners coelute with other PCBs, it is not possible to directly determine the decrease in their concentrations effected by reductive dechlorination using conventional gas chromatography (GC) with an electron capture detector. Recently, Lopshire and Enke have developed a sensitive GC/MS/MS technique to directly quantify these toxic PCB congeners [Chapter 14, this report]. This new method allowed us to determine the percent reduction of each of the toxic congeners after a 16 week incubation of Aroclor 1242 with the Hudson River microorganisms (Table 2-4).

TABLE 2-4. Percent reduction in concentrations of toxic isomers in Aroclor 1242 as a result of 16 weeks of dechlorination by Hudson River microorganisms under laboratory conditions. See Lopshire and Enke, Chapter 14, for analytical methodology.

Congener	% Reduction
34-34-CB	>96.5
345-34-CB	83.3
345-345-CB	>90.0
245-34-CB	80.0
234-34-CB	85.8
2345-34-CB	46.6

The dioxin-like toxicity of compounds has been correlated with their potential to induce P_{450} enzymes such as aryl hydrocarbon hydroxylase (AHH) and ethoxy resorufin O-deethylase (EROD) and the toxicities of various PCB congeners have been estimated based on their potential to induce these enzymes [Safe, 1987; Sawyer and Safe, 1982]. Using such toxicity estimates we calculated an 85% reduction in toxicity in 16 weeks as a result of the dechlorination of Aroclor 1242 by Hudson River microorganisms.

EROD induction assays were performed on the PCB extracts from live and autoclaved treatments to directly determine the toxicity reduction resulting from 16 weeks of dechlorination of Aroclor 1242. A 75% reduction was determined by this method, in good agreement with our calculations.

Bioavailability

Bioavailability to microorganisms is of special concern to the implementation of a bioremediation process for PCB destruction. The PCB residues in contaminated soil or sediment may not be as available as the PCBs freshly added in our dechlorination assays. It is commonly believed that over long periods of time PCBs diffuse into soil or sediment particles, that only aqueous phase PCBs can be biotransformed, and that diffusion into the aqueous phase from inside particles is likely to be slow and rate limiting.

We conducted dechlorination assays to assess the availability of PCBs in Aroclor 1242 contaminated Glens Falls dragstrip soil to dechlorinating microorganisms. We compared the amount of dechlorination of selected late eluting congeners in the contaminated soil to the dechlorination of freshly added 23456-CB. A parallel assay in which Aroclor 1242 and 23456-CB were added to clean sediment was also conducted for comparison.

The results did suggest that the PCBs in the contaminated soil were less available than freshly added PCBs. When standardized on the amount of 23456-CB dechlorination that occurred in the two assays, less of the 25-34-CB and/or 345-2-CB (coeluting isomers) in the

dragstrip soil was dechlorinated (Table 2-5). However, overall dechlorination activity was much greater in the dragstrip soil assay with the result that 84% of the 25-34-CB/2-345-CB was dechlorinated compared to only 31% of the 25-34-CB/2-345-CB added to the clean sediments. In this case at least, substantial dechlorination of environmental PCB residues can occur under appropriate conditions and bioavailability does not appear to be a limiting factor.

TABLE 2-5. Dechlorination of selected congeners in a bioavailability experiment with Glens Falls dragstrip soil (GFDS) during 16 weeks of incubation.

Treatment	Congener	Initial Amount (nmoles/tube)	Amount Dechlorinated (nmoles/tube)	% Dechlorinated
GFDS	25-34/2-345-*	51	43	84
	23456-	90	62	69
Clean Sediment	25-34/2-345-*	72	22	31
	23456-	91	12	13

* Present from the historical PCB contamination; the other PCBs were freshly added

Solid Supports

We have investigated the effect of using several alternatives to the sediments normally included in our dechlorination assays. We have found that little or no dechlorination occurs in the absence of sediments. Sand, sawdust, clay, or vermiculite substituted for sediments did not support dechlorination. Dechlorination was observed, however, in the presence of peat and mixtures of peat (20% by weight) with sand or vermiculite or clay. Of these alternative solid supports, mixtures of peat with vermiculite or clay gave the best results, but were inferior to natural sediments. These experiments were repeated with several of the alternative solid supports. In addition, Aroclor 1242 was added in either acetone or as an emulsion with ligno sulfonate [Liu, 1980]. This time the peat/vermiculite mixture gave results comparable to natural sediments (Table 2-6), while the method of PCB addition had no apparent effect.

The difference in the extent of dechlorination obtained with each of these alternative supports was related to the dechlorination pattern that occurred. Three of the dechlorination patterns described by John Brown [GE Report, 1989] are M (predominantly *meta* dechlorination), Q (predominantly *para* dechlorination), and C (both *meta* and *para* dechlorination).

TABLE 2-6. Average number of chlorines removed from Aroclor 1242 by Hudson River microorganisms in 25 weeks.

Solid Support	Aroclor 1242 Added in Acetone	Aroclor 1242 Added in Ligno Sulfonate
Sediments	1.3	1.4
No Support	---	0.0
Peat	0.6	0.6
Peat/Vermiculite	1.2	1.1
Peat/Clay	0.5	0.8

The treatments with sediments and peat/vermiculite mixtures began as Pattern M and became Pattern C. The peat treatment yielded only Pattern Q, and the peat/clay mixture yielded only Pattern M. Patterns were consistent among all replicates within treatments. These alternative supports may select for different subsets of dechlorinating strains initially present in the PCB-contaminated sediments, but the mechanism of selection is unclear.

SUMMARY AND CONCLUSIONS

1. In laboratory assays, microorganisms eluted from Silver Lake sediments dechlorinated all four Aroclors tested (1242, 1248, 1254, and 1260), but the rate and extent of dechlorination was less for Aroclor 1260. Still, these microorganisms proved more capable of dechlorinating Aroclor 1260 than Hudson River microorganisms. In these assays the Silver Lake dechlorinators accumulated *ortho* and *para* substituted products in addition to the *ortho*-only substituted products so typical of the Hudson River dechlorinators.

2. Despite the complementary dechlorination activities of the Hudson River and Silver Lake microorganisms, mixing them together did not enhance overall Aroclor 1260 dechlorination. It is possible that competition between organisms in the two sediments limited dechlorination.

3. The limited ability of Hudson River microorganisms to dechlorinate Aroclor 1260 is probably not due to inhibition by any of the more heavily chlorinated congeners present in Aroclor 1260.

4. The PCB congeners in Aroclor 1242 with greater dioxin-like toxicity were effectively dechlorinated despite their low concentrations in this PCB mixture. EROD induction potency, which correlates with dioxin-like toxicity, was also shown to decrease as a result of PCB dechlorination.

5. Under appropriate environmental conditions, substantial dechlorination of PCB residues in contaminated soils can be achieved despite somewhat decreased bioavailability.

This may be because the generally longer incubation times required for anaerobic dechlorination also allows more time for the PCBs to become available.

6. Of the alternative solid supports investigated, peat mixed with vermiculite gave the best results. The results suggest that both mineral surfaces and organic matter are required for optimal dechlorination activity.

FUTURE PLANS

Our proposed objectives for the coming year are:

1. To compare the toxicity reduction of different Aroclors as a result of dechlorination by inocula giving different dechlorination patterns, and to estimate the extent of detoxification that has occurred as a result of *in situ* dechlorination at different sites. Toxicity reduction will be assessed by quantifying the congeners with dioxin-like toxicity and by EROD induction assays. Additional toxicity assays may also be adopted.
2. To continue to look for microorganisms more capable of Aroclor 1260 dechlorination using different sediments and different incubation conditions. The dechlorination pattern obtained in assays with Silver Lake microorganisms is more limited than has apparently occurred *in situ*, implying only a subset of the organisms present in the sediment is active under our present incubation conditions.
3. To continue our bioavailability experiments using other contaminated soils and sediments. PCB desorption rates will also be determined directly for each.
4. To continue our attempts to identify the physiological group of microorganism(s) responsible for PCB dechlorination and isolate pure cultures.

Chapter 3

ESTABLISHMENT AND CHARACTERIZATION OF AN ANAEROBIC, AROCLOR 1242-DECHLORINATING CULTURE

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INTRODUCTION

Anaerobic enrichment cultures were established as a first step in obtaining pure cultures or stable consortia capable of reductively dechlorinating PCBs. The objective was to determine conditions which allow dechlorination activity as well as growth of the responsible organisms. The bases for conditions tested were previous studies of anaerobic dechlorination of PCBs and of a pure culture which dehalogenates benzoates (strain DCB-1) [Quensen et al., 1988; Mohn and Tiedje, 1990a and b; DeWeerd et al., 1990, in press]. Cosubstrates were included with PCBs to serve several possible functions, including (1) providing reducing potential for dechlorination, (2) providing carbon and energy for dechlorinating organisms, and (3) supporting other organisms which might establish and maintain an environment favorable for dechlorination. Aroclor 1242 was added at a concentration adequate to support significant growth of any organisms capable of utilizing PCBs as catabolic electron acceptors (1 mg/g dry sediment).

RESULTS

Test of Cosubstrates

Initially four conditions were tested (1) no cosubstrate (except any provided by the uncontaminated sediment), (2) formate, (3) formate plus bromoethanesulfonate to prevent electron flow to methanogenesis, and (4) pyruvate. The former three were in bicarbonate-buffered medium to avoid CO_2 as a potential electron acceptor. The cultures with no cosubstrate and those with pyruvate had the highest dechlorination rates. In transfers of those two treatments, those with pyruvate had much higher dechlorination rates (or shorter lag periods) than those with no cosubstrate (Figure 3-1). Pyruvate therefore stimulates dechlorination activity and greatly improves transferability of dechlorination activity.

Serial Transfers

Dechlorination activity has now been maintained through eight monthly transfers on pyruvate medium. This medium contains RAMM mineral salts, Wolin and DeWeerd vitamins,

[DeWeerd et al., 1990, in press], $NaHCO_3$ (30 mM), cysteine (1 mM), titanium citrate (0.1 mM) and pyruvate (20 mM). Aroclor 1242 was added at a concentration of 1 mg/g dry sediment. A 2:1 (v:w) ratio of liquid medium to clean Spier Falls sediment was used in serum bottles. The transfer technique involved shaking the serum bottle, letting the sediment settle for ~ 10 minutes, and removing the supernatant for inoculum. The two month lag before dechlorination in the primary enrichments was not observed in subsequent transfers on pyruvate-containing medium. All inoculum transfers were 20%, but only slightly lower activity was observed with a 1% inoculum transfer. Thus, the dechlorinating microorganisms appear to grow in these enrichment cultures. Dechlorination occurred primarily from the *meta* position, and resembled Pattern M previously observed (Figure 3-2) [GE Report, 1989].

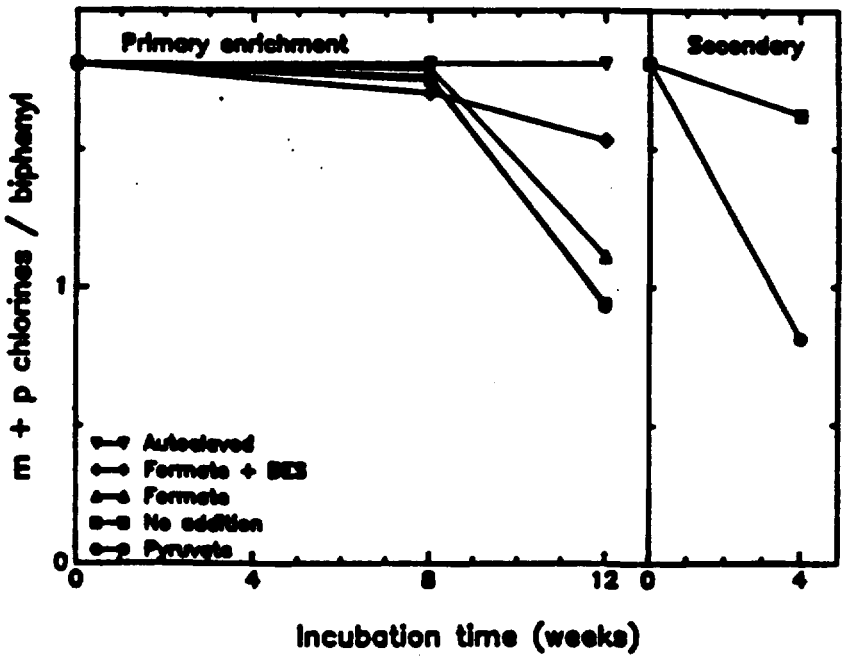

Figure 3-1. Primary and secondary enrichment cultures.

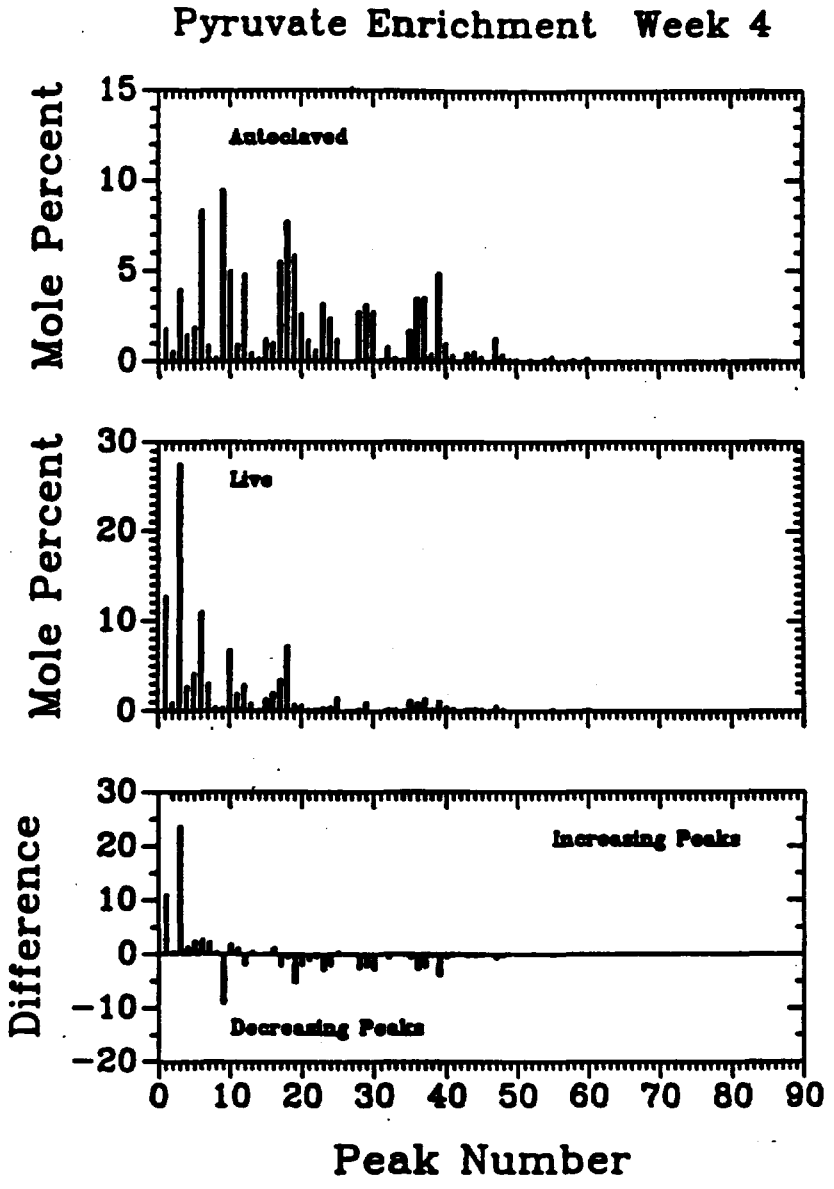

Figure 3-2. Aroclor 1242 dechlorination pattern by pyruvate enrichment at week 4. (Refer to Table 3-1, this report for peak number assignments).

Pyruvate Metabolism

Pyruvate was rapidly consumed by cultures (within 1 week), acetate was a transient product, and methane and CO_2 were final products (Figure 3-3). Dechlorination began simultaneously with the onset of methane production (during week 2 of incubation). Pyruvate was not required for activity but reduced the acclimation period preceding dechlorination. The requirement of the dechlorinating community for reducing potential is apparently not highly specific, as pyruvate could be replaced with H_2 or acetate (the latter supported a somewhat lower dechlorination rate) (Figure 3-4).

Sediment Studies

In all of our studies, uncontaminated Spier Falls sediment was included in the medium, and was required for dechlorination activity. Sediment could not be replaced by defined materials, such as sand and synthetic organoclay, Chromosorb, or a hexadecane phase above the culture. Particle size analysis of the sediment yielded a 90% sand and 10% silt plus clay content. In order to understand which fraction of the sediment was important in supporting the dechlorinating consortium, the sediment was separated into crude fractions. The sediment fractions were separated by sonication and wet-sieving, and the sediment, sediment sand fraction, and sediment silt plus clay fraction were treated with 30% H_2O_2 to remove organic components (Table 3-1). The silt plus clay fraction contained ~ 10 times more organic carbon by weight than the sand fraction. Since 10% of the sediment was silt plus clay, only 2.5 g of this fraction was added to bottles, compared to 22.5 g of the sand fraction. Peroxidized sediment and sediment fractions did not support dechlorination (Figure 3-5). Additionally, varying ratios of sediment to liquid medium were tested (Figure 3-6). A minimal ratio of sediment to liquid medium (> 10 g sediment/50 mL medium) was required to support dechlorination.

TABLE 3-1. Total carbon content of Spier Falls sediment and sediment fractions following peroxidation.

Sediment Fraction	% Total Carbon
sediment	1.47
sediment (H_2O_2 treatment)	0.28
sand	0.72
sand (H_2O_2 treatment)	0.21
clay + silt	8.09

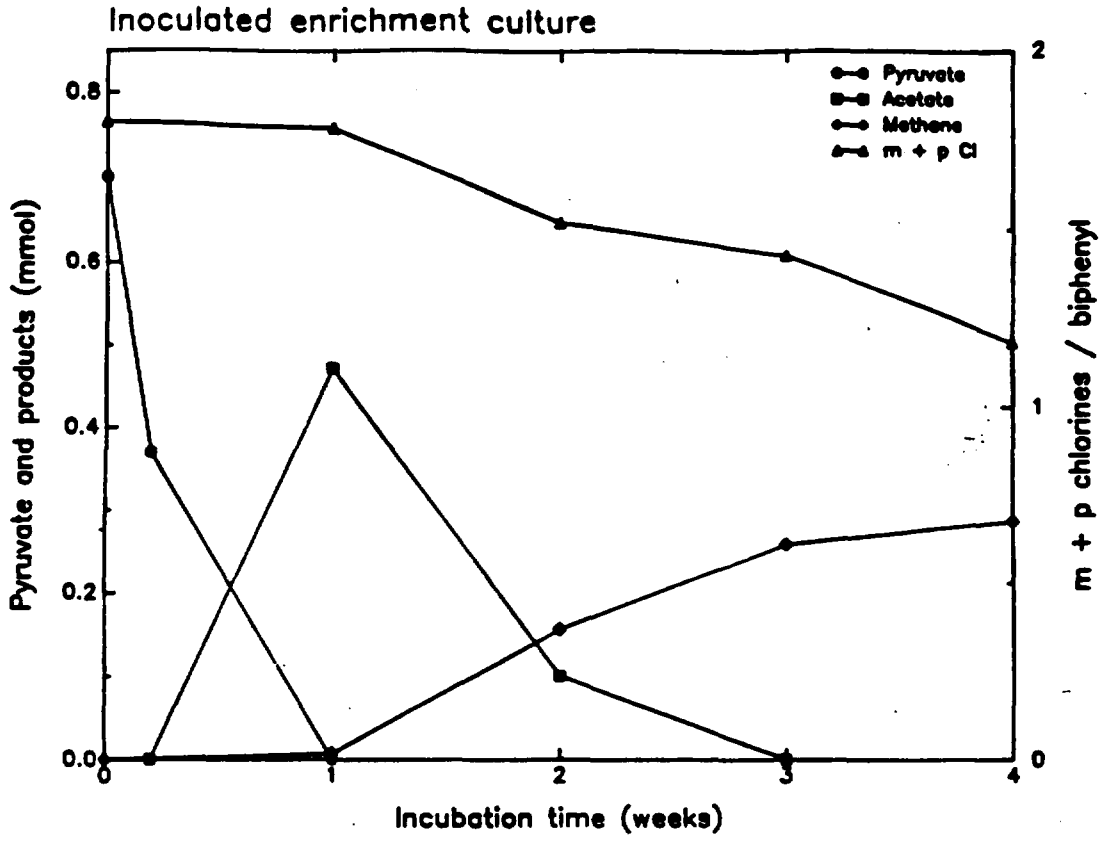

Figure 3-3. Metabolism of pyruvate, methane production, and dechlorination of Aroclor 1242 by anaerobic pyruvate enrichment culture in 4 weeks.

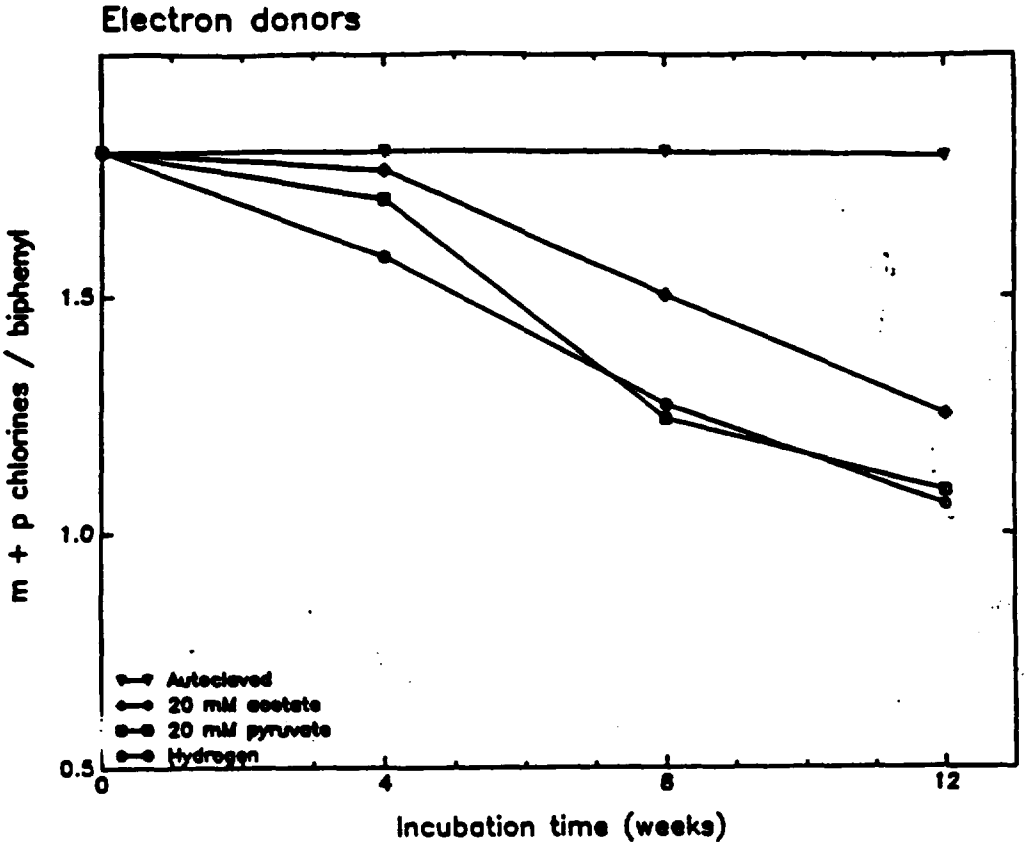

Figure 3-4. Effect of selected electron donors on reductive dechlorination.

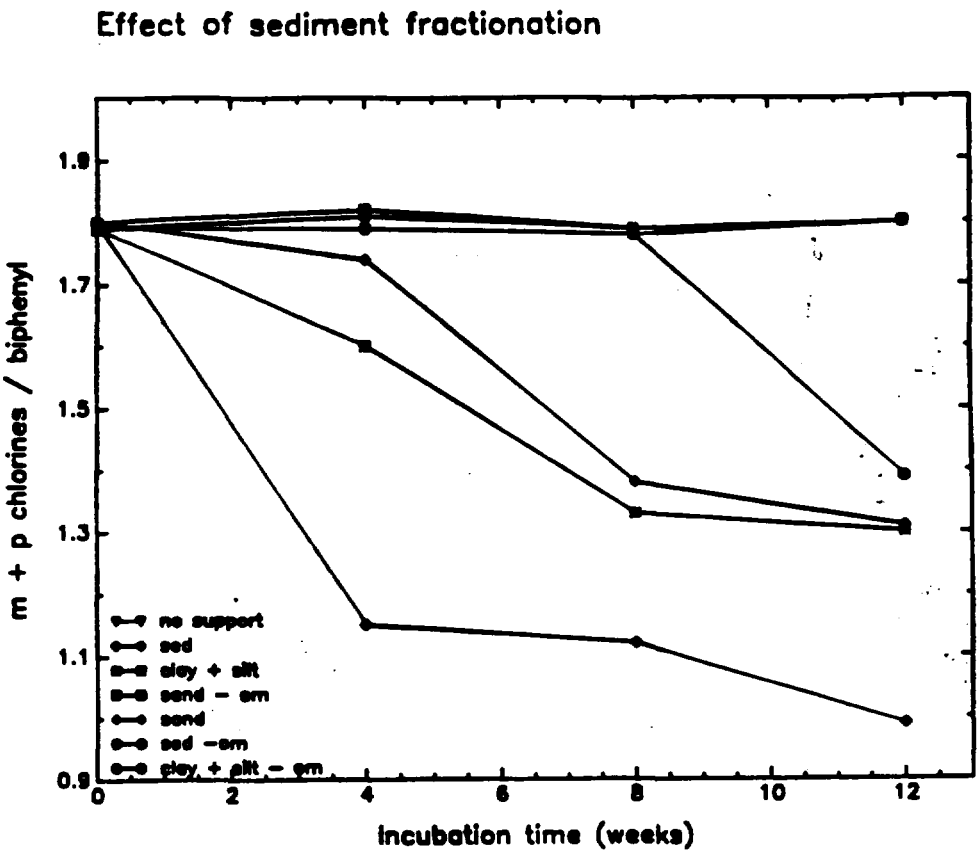

Figure 3-5. Effect of sediment fractionation and peroxidation on dechlorination activity. Treatments included no solid support (50 mL medium), Spier Falls sediment (25 g/50 mL medium), peroxided sediment (25 g/50 mL medium), clay plus silt fraction (2.5 g/50 mL medium), peroxided clay plus silt fraction (2.5 g/50 mL medium), sand fraction (22.5 g/50 mL medium), and peroxided sand fraction (22.5 g/50 mL medium).

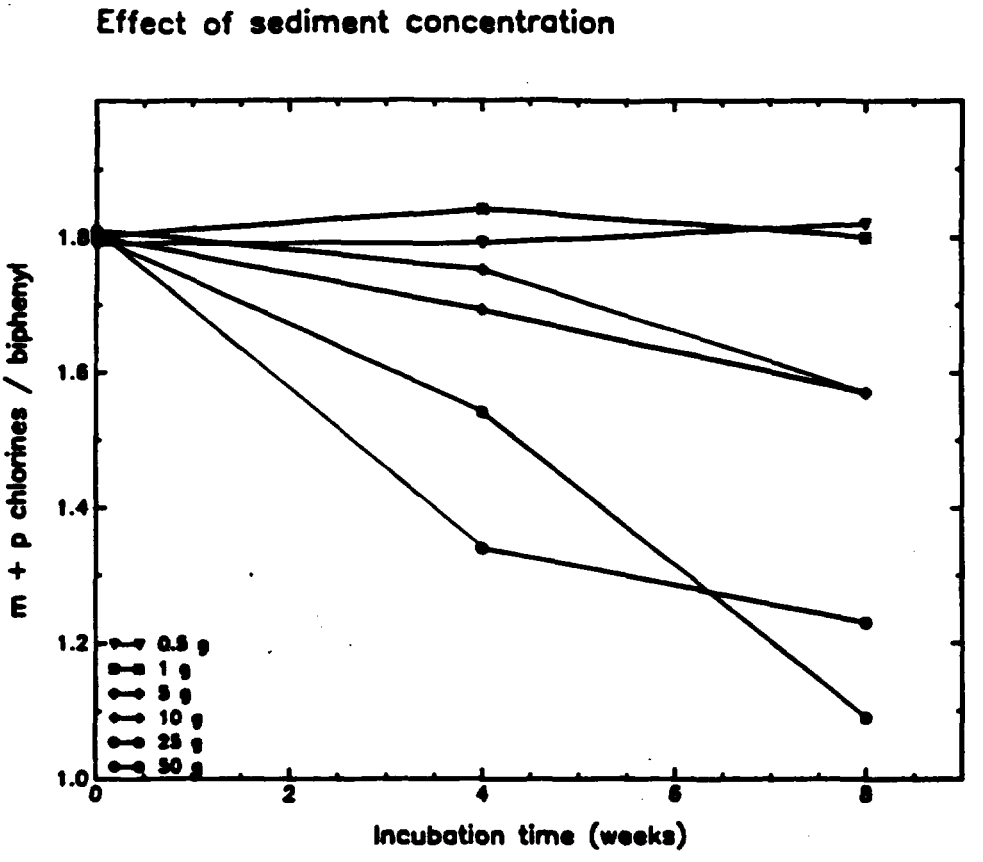

Figure 3-6. Effect of sediment concentration on dechlorination activity. Sediment concentration was varied while the amount of liquid medium remained constant (50 mL).

SUMMARY AND CONCLUSIONS

1. A transferable enrichment culture was established which dechlorinated Aroclor 1242 within 2 weeks of transfer.
2. Pyruvate stimulated the initial dechlorination rate.
3. Hydrogen could replace pyruvate as an electron donor, supporting a higher initial dechlorination rate.
4. High sediment concentrations were optimal.
5. Removal of organic matter by peroxidation inhibited the onset of dechlorination.

FUTURE PLANS

1. To examine the function of the clean sediment in these anaerobic communities, and to find defined materials which can replace the sediments.
2. To further characterize the anaerobic-dechlorinating community, and to attempt isolation of the organism(s) involved.

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Chapter 4

REDUCTIVE DECHLORINATION OF PCBs IN SEDIMENTS FROM THE HUDSON RIVER AND NEW BEDFORD HARBOR

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INTRODUCTION

The occurrence and persistence of PCBs in the environment has been a matter of concern over the last few decades. In aquatic systems they can be removed through adsorption to or partitioning into abiotic and biotic settling particles and subsequent sedimentation. Especially for hydrophobic organic chemicals with high affinity for solids, the sediments act as a sink. But through resuspension and mixing by aquatic organisms the surface sediment can also act as a source for these contaminants, increasing the residence time in the ecosystem [Eisenreich et al., 1989].

High concentrations of hydrophobic organic chemicals hint that these compounds may show mobility and bioavailability in the sediments [Capel and Eisenreich, 1990]. PCBs, especially the higher chlorinated congeners, have been shown to undergo reductive microbial dechlorination [Bopp et al., 1984; Brown et al., 1984; Brown et al., 1987a and 1987b; and Quensen et al., 1988]. It has been hypothesized that reductive dechlorination takes place because the high redox potential of these compounds allows anaerobic bacteria to use PCBs as a terminal electron acceptor [Brown et al., 1987b].

To our knowledge, reductive dechlorination of PCBs has only been observed under methanogenic conditions. It has been suggested [Suflita and Miller, 1985; Gibson and Suflita, 1986] that reductive dechlorination of aromatic compounds could not occur under sulfate-reducing conditions because sulfate would be a better electron sink than the chloroaromatics. Chlorophenols, however, can be degraded under such conditions, their mineralization being coupled to sulfate reduction [Häggblom and Young, submitted]. We therefore also chose to study the dechlorination of PCBs under sulfate-reducing conditions.

Objectives

In an attempt to better understand the conditions which control the reductive dechlorination in sediments, we had the following objectives: A) a systematic study of the microbial activity in sediments under two different reducing conditions; B) the study of different sediments; C) the effect of organic substrates on dechlorination; D) the comparison of possible differences in the fate of endogenous and freshly added PCB; and E) studies with a selected congener mixture.

Sulfate reduction and methanogenesis are the major metabolic processes in anaerobic environments. Sulfate reduction is usually the predominant metabolic process in marine sediments or other sulfate-rich habitats, while methanogenesis is predominant in sulfate-poor environments, such as fresh water habitats [Widdel, 1988]. In marine environments a wide variety of haloaliphatic and haloaromatic compounds are produced biologically by marine organisms [King, 1988; Neidleman and Geigert, 1986]. Therefore, anaerobic marine sediments may potentially allow for the selection and enrichment of anaerobic dehalogenating organisms. Different sites, with possible differences in microbial populations, salinity, content of organic matter, porosity of the natural particles, and historical contamination with different Aroclors mixtures, may lead to different dechlorination activities.

Addition of a mixture of fatty acids (acetate, propionate, butyrate and hexanoic acid) was performed in order to support growth and provide accessible energy. Relatively low concentrations were chosen in order to prevent selection for the fast growing organisms and to more closely approximate the situation in the environment. This mixture of fatty acids was chosen because acetate, propionate and butyrate are probably the most important products from the fermentative decomposition of biomass in sediments [Widdel, 1988]. On one hand, further degradation of propionate and butyrate to methane is possible only by syntrophism between methanogens and hydrogen-producing bacteria, since the former do not directly use organic acids higher than acetate [Widdel, 1988]. On the other hand, propionate and butyrate can directly be oxidized by sulfate reducers. Also, a syntrophic association between sulfate reducers and hydrogen-producing acetogens is most likely in sediments of high sulfate concentration, as is probably the case in New Bedford Harbor. In such a situation, methanogenesis can take place in addition to sulfate reduction, suggesting the presence of syntropic acetogenic bacteria [Widdel, 1988].

In addition to studying dechlorination of the endogenous PCBs or added Aroclor mixtures, studies with a specific selection of congeners were performed for a clearer identification of the dechlorination products.

RESULTS AND DISCUSSION

Sediments

In our studies two different sediments were used as inoculum, both with a history of PCB contamination. These were from the Hudson River (NY) and New Bedford Harbor (MA). Samples collected for this study were from the top 20 cm of the sediment. The Hudson River sediment (H7) is a sandy silt with a moderate content of organic carbon [7-8%, M. Harkness, GE, personal communication], and a PCB composition of dechlorinated Aroclor 1242. Much of the organic carbon is present as wood tailings or other poorly biodegradable natural material. The New Bedford Harbor sediment is a marine, silty mud, rich in organic carbon with a PCB contamination consisting of a 300-400 ppm mixture of Aroclor 1242 and 1254 [Alford-Stevens et al., 1988; Brownawell and Farrington, 1986].

Culture Set-up

A heavy sediment inoculum (35% v/v) was added to methanogenic [Healy and Young, 1979] or sulfidogenic [Häggblom and Young, submitted] mineral salts medium supplemented with vitamins to an end volume of 50 mL in 65 mL flasks with CO_2/N_2 (30%/70%) as the head space gas. The flasks were sealed with Teflon-coated butyl rubber stoppers and aluminum crimp sealers. Methanogenic and sulfate-reducing conditions were promoted by addition of excess carbonate or sulfate, respectively. The sediments of Hudson River and New Bedford Harbor were spiked with Aroclor 1242 in acetone (0.26 g/L) to a final concentration of 100 ppm. For the studies with a congener mixture we added 25-4- (66 ppm), 34-34- (30 ppm) and 234-34-CB (6 ppm) in acetone (1.9 g/L).

Fatty acids were added as a carbon source to one set of cultures, initially to 500 mg/L and then monthly to 250 mg/L. All the cultures were incubated statically at 30°C. Controls were prepared in the same way and autoclaved twice. Subsamples were taken after 0, 1, 2, 4 and 7 months by shaking the batches vigorously, removing the stopper and removing 2 mL of sample slurry with an upside-down Pasteur-pipette. The head space was flushed with N_2/CO_2 and the flask recapped. The subsamples were frozen until extraction and analysis. The head space gas was monitored for methane as described [Bossert and Young, 1986; Häggblom and Young, submitted].

Sample Extraction and Analysis

The subsample vial was used as the extraction vessel. The water was replaced first with 1.5 mL acetone, then with 3 mL of 50/50 acetone/hexane with octachloronapthalene as the internal standard. The vial was then shaken overnight on a mechanical shaker. The organic phase was replaced by 2 mL hexane and the sediment was shaken again for 4 hours. The organic and water phases were combined and, after the addition of sodium chloride, back-extracted twice. The organic extract was dried by addition of a few grams of sodium sulfate, concentrated to ~1 mL with a gentle flow of Argon, and cleaned-up in a micro Florisil column. The PCBs were eluted from the Florisil column in the first 4 mL of hexane. Elemental sulfur was removed by the addition of tetrabutylammonium hydrogen sulfate and sodium sulfite [Jensen et al., 1977]. The samples were analyzed by gas chromatography equipped with a ^{63}Ni electron capture detector (DB-5, 30 m x 0.32 mm I.D.).

Over an incubation period of 7 months we observed the following results:

Hudson River:

In Hudson River sediment cultures under methanogenic conditions dechlorination of added Aroclor 1242 was observed after 1 to 2 months. Loss of higher chlorinated congeners was observed, with *meta* and *para* dechlorination and enrichment of *ortho*-substituted mono- (2-CB), di- (2-2- + 26-CB and 2-4- + 23-CB) and tri- (26-2-CB and 236- + 26-3-CB) chlorobiphenyls (Figure 4-1A). The addition of a fatty acid mixture as an auxiliary carbon source greatly enhanced the rate of dechlorination (Figure 4-1B). After 2 months, the

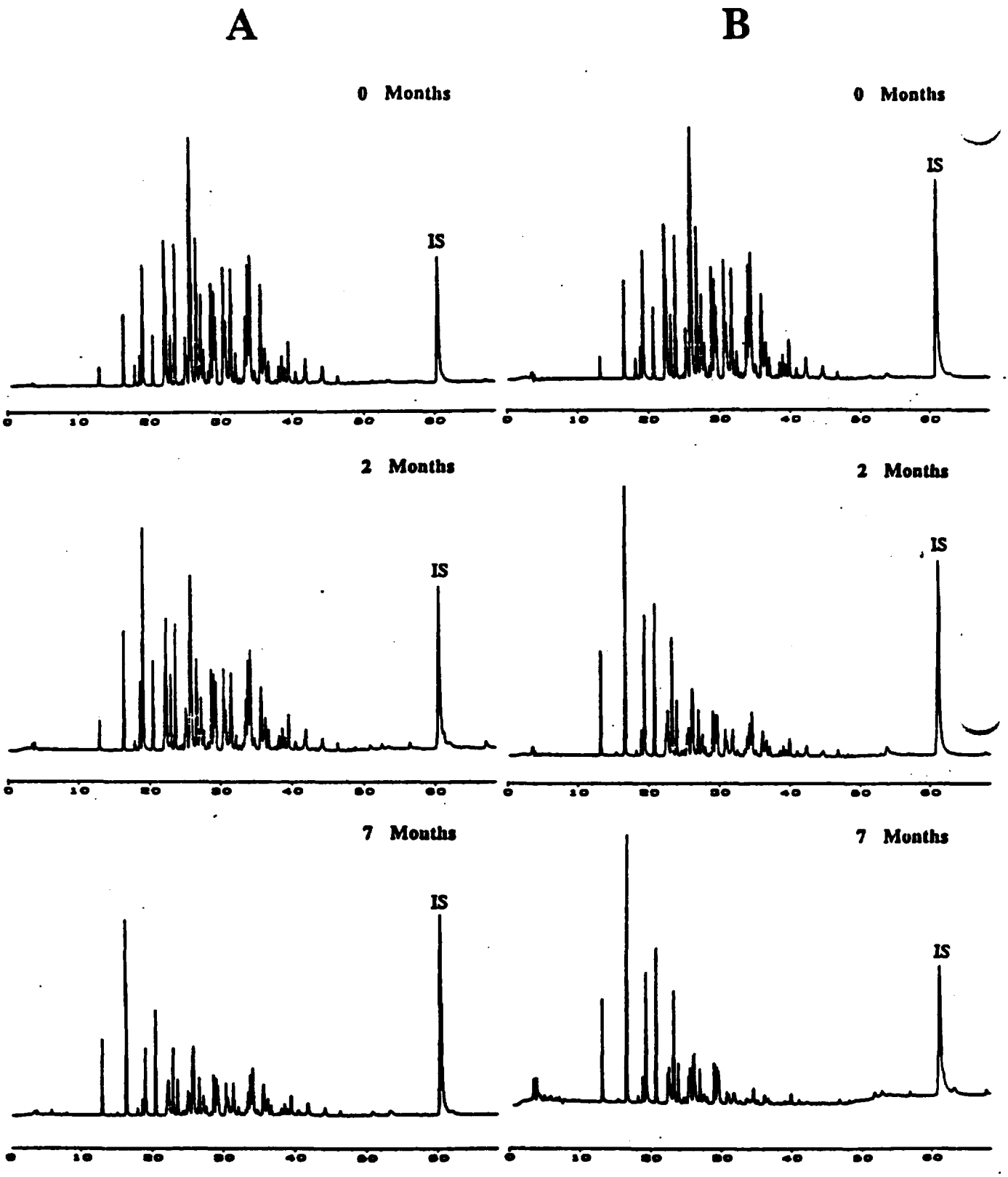

Figure 4-1. Transformation of added Aroclor 1242 in Hudson River sediment: A) no auxiliary substrate added; B) with a fatty acid mixture added monthly. IS: internal standard.

transformation of added Aroclor 1242 generated a pattern of dechlorinated PCBs identical to the endogenous pattern (Figure 4-2). Cultures without the addition of fatty acids showed a significantly longer lag period and slower rates of dechlorination. No transformation of the endogenous PCBs was observed. With the congener mixture the GC-chromatograms show evidence for sequential dechlorination of 25-4-CB to 2-4-CB and 2-CB, and dechlorination of 234-34-CB first, to 24-34-CB and via two possible pathways to 2-CB (Figure 4-3). No transformations have yet been seen of the added Aroclor 1242, endogenous PCBs, or congener mixture under sulfate-reducing conditions.

New Bedford Harbor:

In New Bedford Harbor sediment cultures under methanogenic conditions some trichlorobiphenyls of added Aroclor 1242 were dechlorinated after 2 months of incubation. But only after 4 months did the dechlorination become more pronounced. The overall rate of transformation was much slower than in Hudson River (Figure 4-4). Tetrachlorinated biphenyls were dechlorinated with formation of tri- and dichlorobiphenyls. Transformation of some congeners of tri- and tetrachlorobiphenyls of the endogenous PCBs was observed (Figure 4-4B). The addition of fatty acids as an auxiliary carbon source had no effect on the rate or extent of dechlorination. Under sulfate-reducing conditions no transformation of PCBs was observed.

SUMMARY AND CONCLUSIONS

1. Dechlorination of PCBs in different sediments occurred under methanogenic, but not under sulfate-reducing conditions.
2. Tri- and tetrachlorobiphenyls were transformed in Hudson River sediment with accumulation of *ortho* chlorinated mono-, di- and trichlorobiphenyls. This is consistent with earlier results [Brown et al., 1987a; Quensen et al., 1988].
3. Addition of an auxiliary carbon source enhanced dechlorination of PCBs in Hudson River sediment, a sandy sediment with moderate levels of poorly degradable organic carbon content, but had no effect on dechlorination observed in New Bedford Harbor sediment, a silty sediment which was already rich in organic matter. Lack of an organic substrate (energy and/or reducing power) may thus be limiting PCB-dechlorination in some sediments.
4. Dechlorination of PCBs was much faster in Hudson River than in New Bedford Harbor sediment. This may due to differences in the microbial populations. Another explanation might lie in bioavailability differences caused by differences in the sediment matrices. This could be a mechanism similar to the availability of the fumigant 1,2-dibromoethane (EDB) in soils as reported by Steinberg et al. [1987]. EDB is a volatile, moderately water soluble compound with a low affinity for soils. This compound

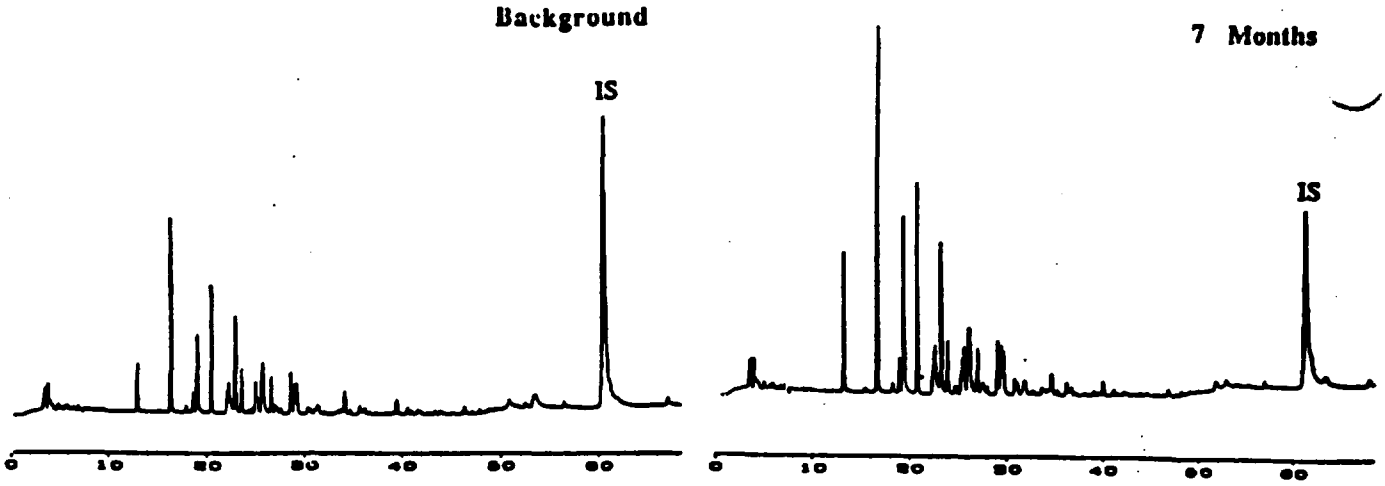

Figure 4-2. Background PCB contamination in Hudson River sediment and sediment with added Aroclor 1242 after 7 months incubation. IS: internal standard.

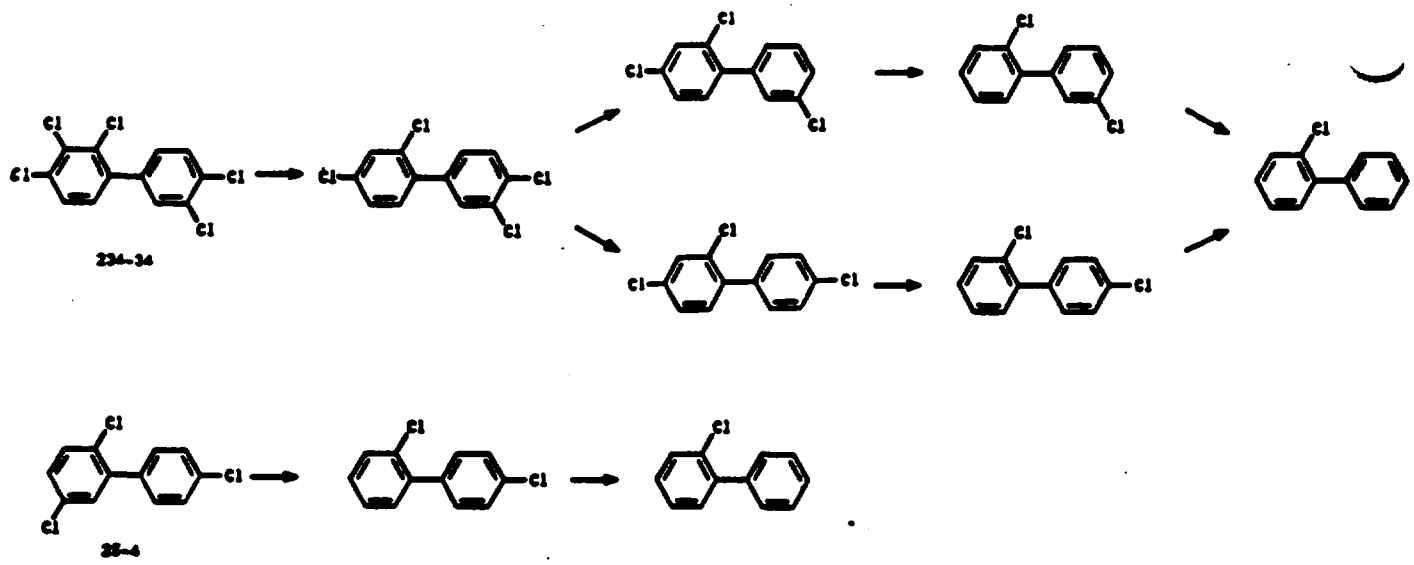

Figure 4-3. Possible pathways for dechlorination of 234-34-CB and 25-4-CB in Hudson River sediment.

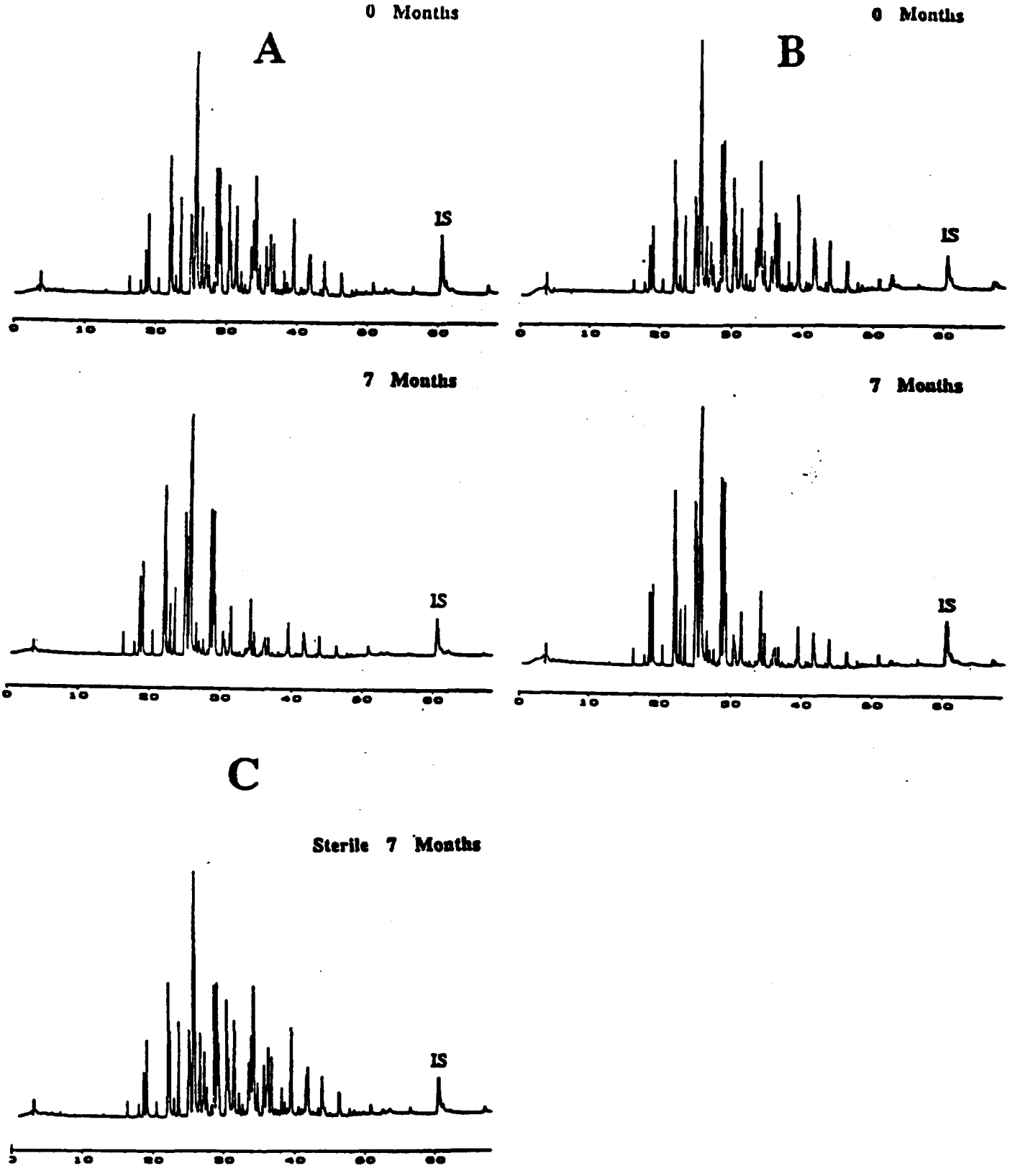

Figure 4-4. Transformations in New Bedford Harbor sediment: A) of added Aroclor 1242, with addition of a fatty acid mixture, B) of background PCB with no addition of an auxiliary substrate, C) sterile control with added Aroclor 1242 after 7 months incubation. IS: internal standard.

was found to persist in surface soil as long as 19 years after its last known application. This residual EDB was extremely resistant to mobilization (desorption into air and water) and biodegradation, in contrast to freshly added [^{14}C]-EDB at comparable concentrations that was rapidly removed and mineralized. Pulverization of the soil greatly enhanced the release of residual EDB. The inertness to biodegradation suggest that native EDB was entrapped in intraparticle micropores, making it inaccessible to bacteria and slow to equilibrate with air or water phases. Similar unavailability to biodegradation was also observed with "old" chlorophenols in some contaminated soils even though freshly added chlorophenols were degraded [Salkinoja-Salonen et al., 1989].

5. The pattern of the dechlorination of added Aroclor 1242 that we observed in the Hudson River sediment is consistent with that found in the environment. The dechlorination in New Bedford Harbor is apparently still in process after 7 months of incubation. It is premature to predict whether the dechlorination pattern is different from that of the Hudson River sediment due to congener specificity of the dechlorination consortia or to dechlorination rate differences.

FUTURE PLANS

Comparison of the anaerobic activity of Hudson River with that of New Bedford Harbor will be made in order to determine whether there are two different anaerobic populations at the two sites. This effort will be extended to examine whether sediments of New Bedford Harbor are inhibitory to PCB transformation. In addition, attempts to propagate active cultures in order to maximize the possibility of isolating pure cultures will be undertaken. Further, the cultures under sulfate-reducing conditions will periodically be monitored to see if a PCB-dechlorinating consortia will evolve under these conditions.

Chapter 5

ANAEROBIC DECHLORINATION OF ENDOGENOUS PCBs IN WOODS POND SEDIMENT

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INTRODUCTION

Woods Pond (Lenox, MA) is a shallow impoundment on the Housatonic River system located 10.5 miles downstream of Silver Lake (Pittsfield, MA). Water depths range from 1-3 feet with the exception of channels created by the flow of the Housatonic River. The sediments are composed of a mixture of black organic matter, sand, and silt and are contaminated with an unidentified hydrocarbon oil (1 to 5%) and with Aroclor 1260 (50 to 150 ppm) to depth of ~18". The Aroclor 1260 in Silver Lake sediments has undergone extensive dechlorination via loss of *ortho*, *meta*, and *para* chlorines [Brown et al., 1987a, 1987b], but the PCBs in Woods Pond sediments show evidence of only slight dechlorination via loss of *meta* and *para* chlorines. The striking difference in the type and extent of environmental dechlorination at these two sites led us to question the underlying cause(s): Do Woods Pond sediments harbor microorganisms capable of dechlorinating PCBs? Do the Woods Pond sediments lack some nutrient that is essential for the growth and/or activity of these microbes? Does some component of these sediments inhibit the growth of dechlorinating bacteria? Are the PCBs in the Woods Pond sediments accessible to dechlorinating microorganisms? Our objective was to determine whether conditions could be found to stimulate the biological dechlorination of the endogenous PCBs in Woods Pond sediment.

RESULTS AND DISCUSSION

Sediment samples were collected along the eastern and western shores of Woods Pond for analysis of PCBs and assessment of biological dechlorination activity. Typically, core samples were collected to the point of refusal using a Lexan® tube (2" diameter). Samples to be used for cultures were transferred to glass jars, topped with site water, sealed, and stored at 4°C. In all, samples were collected from 88 locations: 28 along the eastern shore, 58 along the western shore, and two at shallow points in the middle. The PCBs were extracted from the sediment samples, analyzed by capillary gas chromatography (GC), and compared with standards of Aroclors 1254 and 1260. A comparison of the congener distribution of the PCBs in the sediment with the Aroclors indicates that the contaminant was virtually all Aroclor 1260. We based this estimate primarily on the proportions of hepta- and octachlorobiphenyls, especially GC peaks 88, 100, 109, 110, and 115 (Figure 5-1). Sediment PCBs from all 88 locations showed evidence of some, albeit only slight, environmental dechlorination. Two

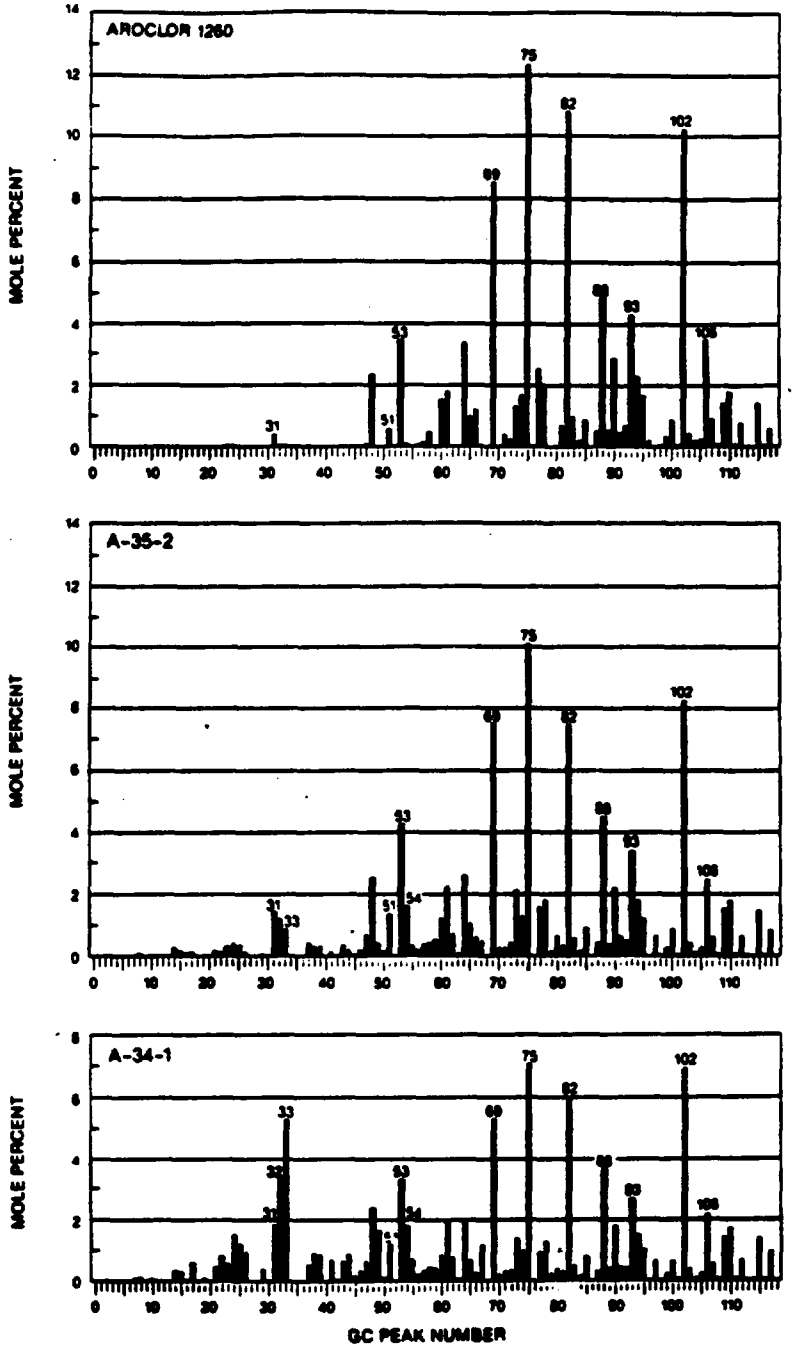

Figure 5-1. Comparison of the proportions of PCB congeners in Aroclor 1260 and in two Woods Pond sediments. Congener peaks were identified and quantified as previously described [Brown et al., 1987b].

sediment samples taken only 15 feet apart on the western shore of Woods Pond illustrate the range of environmental dechlorination seen in the sediments. At site A-35-2 the PCBs showed the least dechlorination, yet four major peaks, 75, 82, 102, and 106 (245-245-, 234-245-, 2345-245-, and 2345-234-CB, respectively) showed decreases of 16 to 33% relative to Aroclor 1260 (Figure 5-1). At the same time, peaks corresponding to expected *meta, para* dechlorination products of these congeners were quite evident: peaks 31, 32, 33, 51, 53, and 54 (25-25-, 24-25-, 24-24-, 235-25-, 245-25-, and 245-24-CB, respectively). At site A-34-1, which shows the most advanced dechlorination we have seen in Woods Pond sediment, peaks 75, 82, 102, and 106 were decreased by 33 to 45% relative to Aroclor 1260, and peaks corresponding to dechlorination products, especially 31, 32, and 33, were quite prominent (Figure 5-1). Table 5-1 compares the proportions of 25-25-, 24-25-, 24-24-, and 245-24-CB (GC peaks 31, 32, 33, and 54, respectively) in Aroclor 1260 and in these two sediment samples. The four congeners are present at only negligible amounts in Aroclor 1260 but are substantial components of the sediment PCBs. These data indicate that the 25-25-, 24-25-, 24-24-, and 245-24-CB in the Woods Pond sediments must have been formed by the dechlorination of components of Aroclor 1260.

TABLE 5-1. Proportions of four PCB congeners in Aroclor 1260 and in two Woods Pond sediments.

Sample	Mole Percent			
	25-25-CB	24-25-CB	24-24-CB	245-24-CB
Aroclor 1260	0.39	0.03	0.05	0.07
A-35-2	1.41	1.12	0.88	1.61
A-34-1	1.79	3.32	5.21	1.75

Although it is clear that some *meta* and *para* dechlorination of the Aroclor 1260 in Woods Pond sediments has occurred, it is not clear when or where this occurred. The dechlorination may have taken place upstream before the sediments were deposited in Woods Pond, or it may have taken place in Woods Pond at some time in the past. The only way to determine if the microoganisms responsible for the dechlorination were still present in Woods Pond sediments was to demonstrate dechlorination in these sediments in controlled laboratory experiments. Our first experiments involved the addition of more Aroclor, either Aroclor 1260 or a 1:1 mixture of Aroclors 1254 and 1260 at two different concentrations (100 and 500 ppm) to slurries of Woods Pond sediment prepared in reduced anaerobic minimal medium (RAMM) [Shelton and Tiedje, 1984]. We detected no dechlorination of the Aroclor over a 2-month period and decided to set up new slurries to which we would add a single PCB congener. The use of a single congener offered two advantages over the use of an Aroclor: (1) Even low levels of dechlorination of a single congener added at high concentration would be easier to detect because only a few products would be formed. (2) We could try a variety of PCB congeners in case some turned out to be better substrates for dechlorination than others.

When 25-34-CB (350 μM) was added to slurries of Woods Pond sediment, we observed loss of the *para* chlorine to yield 25-3-CB after 3 weeks of incubation at 22 to 24°C. This dechlorination continued until ~ 85% of the tetrachlorobiphenyl had been converted to 25-3-CB at 16 weeks. Beginning at 7 weeks, when ~ 30% of the 25-34-CB had been dechlorinated to 25-3-CB, we also began to see changes in the endogenous PCBs. Figure 5-2 compares the proportions of PCB congeners in a Woods Pond sediment at the start of the experiment and after a 12-week incubation. All major peaks except peak 88 were substantially decreased after incubation. At the same time, large increases occurred in some of the tetra- and pentachlorobiphenyl peaks, especially peaks 31, 32, 37, and 51 (25-25-, 24-25-, 23-25-, and 235-25-CB). Table 5-2 identifies the major congeners that have been dechlorinated, while Table 5-3 identifies the major congeners formed as a result of dechlorination. It appears from these data that PCB congeners having 245-, 234-, and 2345-chlorophenyl groups have each lost the chlorine in the *para* position, yielding products carrying 25-, 23-, and 235-chlorophenyl rings. *Para* chlorines were also removed from 34-chlorophenyl groups, but not from 24-chlorophenyl groups.

TABLE 5-2. Major PCB components of Aroclor 1260 decreased by Woods Pond dechlorination Pattern H.

DB-1 GC Peak No.	PCB Congener	Mole Percent			Percent Decrease	Proposed Dechlorination Products
		T_0	T_{12}	Net Loss		
53	245-25	3.97	2.12	1.85	47	25-25
54	245-24	1.97	0.33	1.64	83	24-25
69	245-34 236-245	8.31	6.38	1.93	23	25-34[a] → 25-3[a] 236-25
75	245-245	9.15	5.32	3.84	42	245-25 → 25-25
82	234-245	6.81	4.90	1.92	28	234-25 → 23-25
93	2345-236	3.11	2.19	0.91	29	235-236
102	2345-245	6.73	5.66	1.07	16	2345-25 → 235-25
106	2345-234	2.16	1.75	0.42	19	235-234 → 235-23

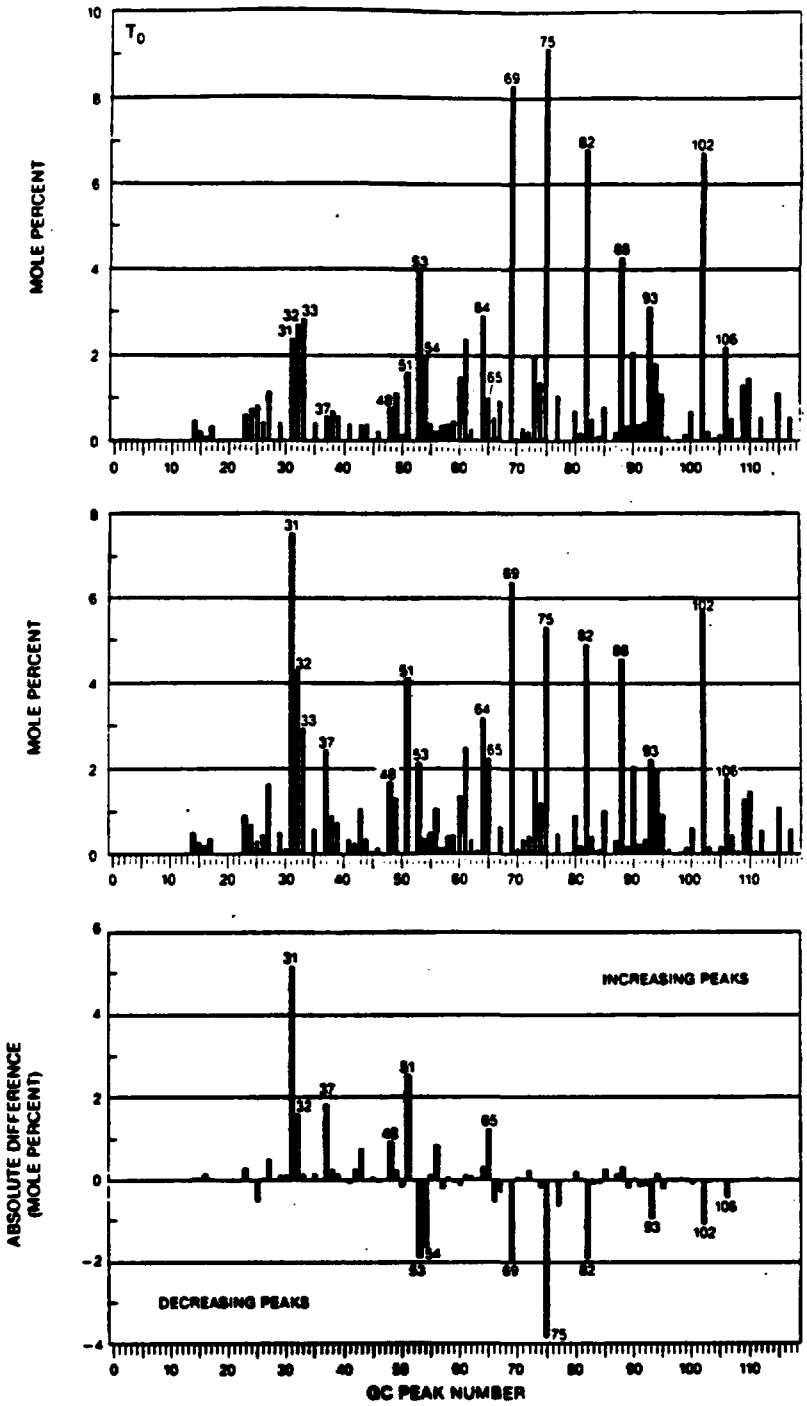

Figure 5-2. Pattern H dechlorination. Comparison of the PCB Congener distribution of PCBs from Woods Pond sediment before and after dechlorination. Peaks corresponding to 25-34-CB (peak 47) and its dechlorination product 25-3-CB (peak 21) were edited out of the chromatogram before calculating the mole percentages of the sediment PCBs. The data presented represents the average of duplicate samples. Top panel: Sediment PCBs at the beginning of the experiment. Center panel: Sediment PCBs after 12 weeks incubation. Bottom panel: Net change in individual PCB peaks after 12 weeks incubation.

TABLE 5-3: Major dechlorination products formed from Aroclor 1260 by Woods Pond Dechlorination Pattern H.

DB-1 GC Peak Number	PCB Congener	Mole Percent		
		T_0	T_{12}	Net Increase
31	25-25	2.36	7.52	5.17
32	24-25	2.70	4.30	1.60
37	23-25	0.58	2.39	1.81
48	236-25, 245-26	0.75	1.66	0.91
51	235-25, 236-23	1.57	4.10	2.53
56	235-23	0.24	1.07	0.83
65	235-236	0.99	2.21	1.22

This type of dechlorination is known as Pattern H. Pattern H dechlorination of Aroclor 1260 is most easily recognized on chromatograms by a large increase in peak 31 (25-25-CB), and moderate increases in peaks 32 and 51 (24-25-, and 235-25-CB) (Figure 5-2). Pattern H dechlorination of Aroclors 1242 and 1254 has been observed in environmental samples from New Bedford Harbor (Massachusetts), Escambia Bay (Pensacola, Florida), and some regions of the Hudson River [Brown and Wagner, 1990] and in laboratory incubations of Aroclor 1260 using inocula from the Hudson River [Quensen et al., 1990].

We subsequently incubated slurries of Woods Pond sediment with various other PCB congeners to determine whether microorganisms capable of removing chlorines from the *meta* positions were also present in Woods Pond sediments. When 23456-CB was added, it was dechlorinated primarily by loss of both *meta* chlorines to yield 246-CB (86%), and to a lesser extent by loss of the *para* chlorine to yield 2356-CB (13%). Small amounts of 236- (0.5%) and 26-CB (0.5%) were also formed. The endogenous Aroclor 1260 began to dechlorinate after most of the 23456-CB was dechlorinated. This time we observed extensive and almost indiscriminate decreases in hexa-, hepta-, and octachlorobiphenyls (peaks 69 through 115) and large increases in tri-, tetra-, and pentachlorobiphenyls (peaks 14 through 63). A single dechlorination product, 24-24-CB (peak 33), accounted for 21.3% of the total PCB (Figure 5-3). Table 5-4 lists the major PCB congeners in the sediment and shows the extent to which they were removed by dechlorination. Table 5-5 lists the major products formed by dechlorination.

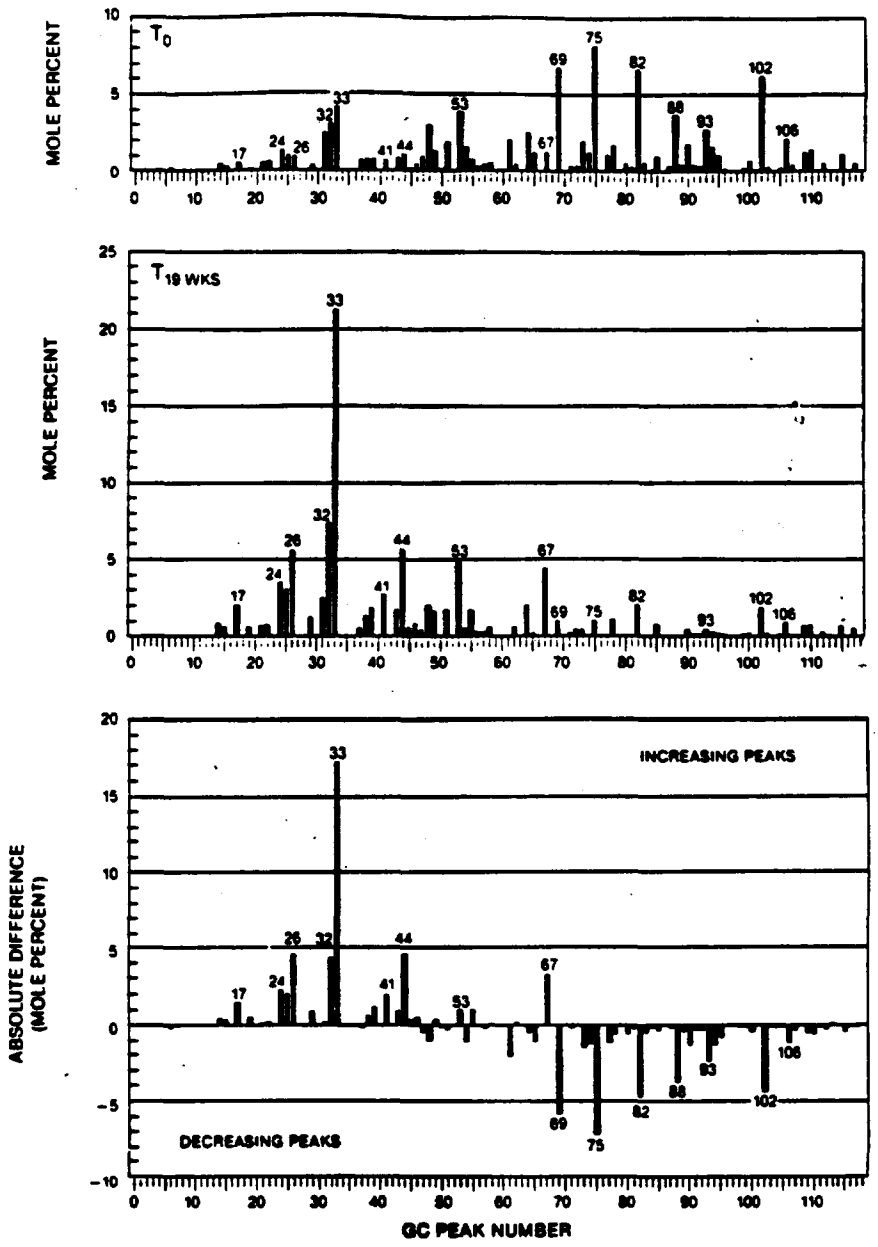

Figure 5-3. Pattern N dechlorination. Comparison of the PCB Congener distribution of PCBs from Woods Pond sediment before and after dechlorination. Peaks corresponding to 23456-CB (peak 60) and its dechlorination products were edited out of the chromatogram before calculating the mole percentages of the sediment PCBs. The peaks edited out were: 2356-CB (peak 52), 236-CB (peak 16), 246-CB (peak 11), and 26-CB (peak 5). Top panel: Sediment PCBs at the beginning of the experiment. Center panel: Sediment PCBs after 19 weeks incubation. Bottom panel: Net change in individual PCB peaks after 19 weeks incubation.

TABLE 5-4. Major PCB components of Aroclor 1260 decreased by Woods Pond dechlorination Pattern N.

DB-1 GC Peak Number	PCB Congener	Mole Percent		Net Loss	Percent Decrease	Proposed Dechlorination Products
		T_0	T_{19}			
54	245-24[a]	1.53	0.49	1.04	68	24-25
61	236-34	2.04	0	2.04	100	26-34 → 26-4
65	235-236	1.19	0.18	1.01	85	235-26 → 25-26
69	236-245 245-34	6.72	0.91	5.81	86	236-24 → 24-26 24-34 → 24-4
73	235-245	1.93	0.45	1.48	77	235-24 → 24-25
75	245-245	8.09	0.98	7.11	88	245-24 → 24-24
82	234-245	6.61	1.94	4.67	71	234-24 → 24-24
88	2356-245 2345-246	3.66	0	3.66	100	2356-24 245-246 → 246-24
90	2346-245	1.74	0.44	1.30	74	245-246 → 246-24
93	2345-236 23456-24	2.69	0.40	2.29	85	236-245 → 236-24 → 24-26 2346-24 → 246-24
94	2356-234	1.63	0.29	1.34	82	2356-24
102	2345-245	6.16	1.81	4.35	71	245-245 → 245-25 → 24-24 235-245 → 235-24 → 24-25
106	2345-234	2.05	0.91	1.14	55	245-234 → 234-24 → 24-24 235-234 → 235-24 → 24-25
TOTAL		46.04	8.80	37.24	81	

[a] 245-24-CB is not present in Aroclor 1260 but transiently accumulates in Woods Pond sediments, most likely as an intermediate in the dechlorination of 245-245-CB (peak 75) and 234-245-CB (peak 82).

TABLE 5-5. Major dechlorination products formed from Aroclor 1260 by Woods Pond dechlorination Pattern N.

DB-1 GC Peak Number	PCB Congener	Mole Percent		
		T_0	T_{19}	Net Increase
17	26-4	0.49	1.93	1.44
24	24-4[a]	1.32	3.52	2.20
25	25-26[a]	1.01	3.02	2.01
26	24-26[a]	0.93	5.52	4.59
32	24-25	2.96	7.30	4.34
33	24-24	4.03	21.28	17.25
41	236-26[ab], 24-35[a]	0.68	2.64	1.96
44	246-24[a]	1.05	5.66	4.61
67	2356-24[a]	1.18	4.34	3.16
TOTAL		13.65	55.21	41.56

[a] The identity of these products was confirmed by GC-MS.

[b] This product was probably derived primarily from 236-236-CB (peak 60) which coelutes with 23456-CB and was therefore edited out of the chromatogram.

The endogenous PCBs appear to have lost almost exclusively *meta* chlorines: 2345-, 234-, and 245-chlorophenyl groups were all dechlorinated to 24-chlorophenyl groups, and 2346-, 236-, and 34-chlorophenyl groups were dechlorinated to 246-, 26-, and 4-chlorophenyl, respectively. The 24-24-CB appears to have been almost totally derived from the dechlorination of four major peaks: 75, 82, 102, and 106 (245-245-, 234-245-, 2345-245-, and 2345-234-CB, respectively). The moderate increase in 24-25-CB (4.34 mole percent), however, indicates that some of the 2345-chlorophenyl group in 2345-245- and 2345-234-CB (peaks 102 and 106) must undergo *para* dechlorination to 235-chlorophenyl and subsequently *meta* dechlorination to 25-chlorophenyl (see Tables 5-4 and 5-5). Analysis of some of the smaller peaks (data not shown) also indicate that 235- and 2356-chlorophenyl groups were slowly dechlorinated, whereas 25-chlorophenyl groups were not. This type of dechlorination has been designated Pattern N and closely resembles the Pattern N previously seen in laboratory cultures of sediment incubated with Aroclor 1260 and an inoculum from Silver Lake [Quensen et al., 1990]. Pattern N dechlorination of Aroclor 1260 is most easily recognized on chromatograms by a large increase in peak 33 (24-24-CB), and moderate increases in peaks 32 and 67 (24-25- and 2356-24-CB, respectively) (Figure 5-3).

Figure 5-4 shows the shift in the PCB homolog distribution of the endogenous PCBs that occurred and underscores the effectiveness of the Pattern N dechlorination system in eliminating hexa-, hepta-, and octachlorobiphenyls. These three homolog classes were decreased by 70, 72, and 44%, respectively, while tri-, tetra-, and pentachlorobiphenyls increased to 82% of the total PCB content.

Our data demonstrate that microbial populations from Woods Pond sediments can effect two different types of dechlorination of Aroclor 1260: Pattern H (*para* dechlorination), and Pattern N (*meta, para* dechlorination). The major dechlorination products of both of these dechlorination systems, 25-25-CB for Pattern H, and 24-24-, and 24-25-CB for Pattern N, were found at all 88 locations sampled in Woods Pond, albeit in varying proportions. Pattern H dominates in sediments taken from the eastern shore. Along the western shore, sediment PCBs randomly display both Patterns H and N in various combinations ranging from predominantly Pattern H (25-25- >24-25- > >24-24-CB), to predominantly Pattern N (24-24- >24-25- > >25-25-CB). Sample A-35-2 (Figure 5-1) is an example of slight dechlorination by a mixture of Patterns H (dominant) and N, whereas sample A-34-1 illustrates moderate Pattern N dechlorination mixed with some Pattern H dechlorination (evidenced by peaks 31 and 51). Laboratory incubations with sediments taken from 30 different locations along the entire eastern and western edges of the pond all showed dechlorination activity, thus demonstrating that the microorganisms capable of dechlorination are widely distributed.

SUMMARY AND CONCLUSIONS

It is apparent from our data that two different dechlorination systems, Pattern H, *para* dechlorination, and Pattern N, *meta, para* dechlorination, are active throughout the sediments of Woods Pond. Given this information, it is not clear why only modest dechlorination of the

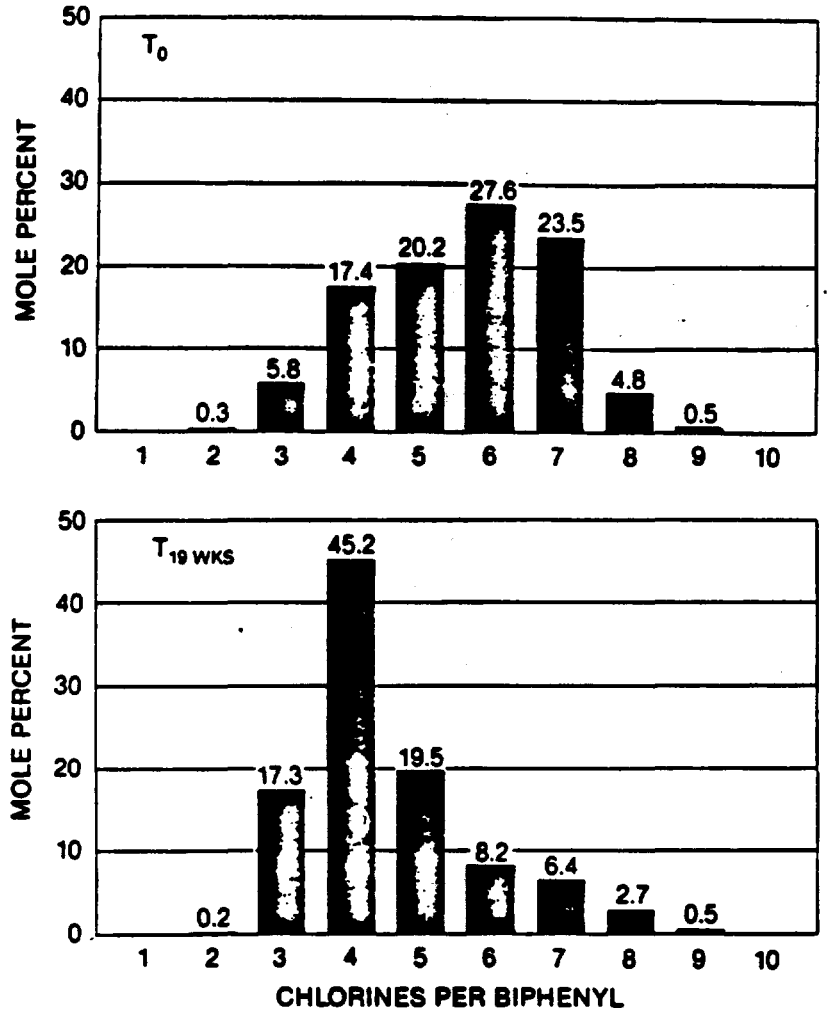

Figure 5-4. Homolog distribution of PCBs in Woods Pond sediment before and after dechlorination.

sediment PCBs has occurred. Even in the laboratory we have only been able to stimulate the dechlorination of endogenous PCBs by adding a high concentration (350 μM) of PCB. The addition of medium (RAMM) with or without various carbon sources, but without the addition of PCBs, stimulated methanogenesis but did not stimulate dechlorination of the endogenous PCBs. Collectively these data imply that the slow dechlorination in Woods Pond cannot be attributed to the lack of an essential nutrient or to the presence of a toxic inhibitor. A more likely explanation is that limited bioavailability of the Woods Pond sediment PCBs, caused by partitioning of the PCBs into both the highly organic sediment and the oil associated with the sediment, may severely limit the growth and activity of the appropriate microbial population(s). It has been proposed that the anaerobic bacteria responsible for dechlorination of PCBs may use the PCB as an electron acceptor and may therefore derive energy from the dechlorination of PCBs [Brown et al., 1987b; Quensen et al., 1988]. If this is the case, then the addition of a single congener in high concentration may stimulate the growth of these microorganisms by providing them with a readily accessible electron acceptor. The resulting high numbers of dechlorinating bacteria could then dechlorinate the endogenous PCBs. However, regardless of the reason, our laboratory experiments (Figures 5-3 and 5-4) clearly demonstrate that it *is possible* to extensively dechlorinate the hexa-, hepta-, and octachlorobiphenyls in the Woods Pond sediments.

FUTURE PLANS

Our objectives for the next year are:

1. To learn how to grow the microbial population responsible for Pattern N dechlorination.
2. To learn how to accelerate PCB dechlorination of endogenous PCBs without the addition of more PCBs.
3. To find means of further dechlorinating the major dechlorination products of Pattern N (24-24-, and 24-25-CB).

Chapter 6

ANAEROBIC AND AEROBIC BIODEGRADATION OF ENDOGENOUS PCBs

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INTRODUCTION

The altered distribution of residual PCB contamination observed in several aquatic sediments is evidence of widespread microbial reductive dechlorination of PCBs in the environment [Brown et al., 1984; 1987a; 1987b]. This process effects the specific removal of *meta* and *para* chlorines, resulting in the depletion of highly chlorinated PCB congeners with corresponding increases in lower chlorinated, *ortho* substituted PCBs. In addition, microbial anaerobic dechlorination of PCBs has been observed in the laboratory [Quensen et al., 1988; GE Report, 1989; Abramowicz et al., 1990], and has recently been reviewed [Bedard, 1990; Abramowicz, 1990]. This report will focus on recent findings involving the acceleration of dechlorination in Hudson River sediments, the dechlorination of endogenous PCB contamination, as well as the sequential anaerobic/aerobic treatment of contaminated sediments.

The acceleration of anaerobic dechlorination in Hudson River sediments was observed upon the addition of a complex nutrient mixture, surfactants, or a simple trace metal mixture. The latter result may indicate that low levels of a trace metal in the sediment may limit the rate of PCB dechlorination occurring in the environment today. Dechlorination of endogenous PCB contamination has been observed in three different soils and sediments. This result indicates that anaerobic microorganisms have access to PCBs in even "aged" soil environments. In addition, sequential microbial treatment via anaerobic dechlorination and aerobic biodegradation has been demonstrated on such endogenous PCB contamination.

RESULTS AND DISCUSSION

Rate Enhancement

The methods utilized in these experiments will be briefly discussed. The sediments were collected and stored until use in sealed vessels at 4°C. All subsequent operations to investigate anaerobic dechlorination were performed in an oxygen-free atmosphere (95% N_2, 5% H_2). The sediment was mixed with medium (35 g soil/ 50 mL medium) to produce a sediment slurry from which aliquots were removed while mixing to obtain reproducible sampling. Typically 10 μL of a concentrated PCB stock solution in acetone was added to spiked samples. Samples were crimp sealed, vigorously vortexed, and stored stationary in the dark at 23°C, unless otherwise indicated. Aerobic studies were performed by initially growing

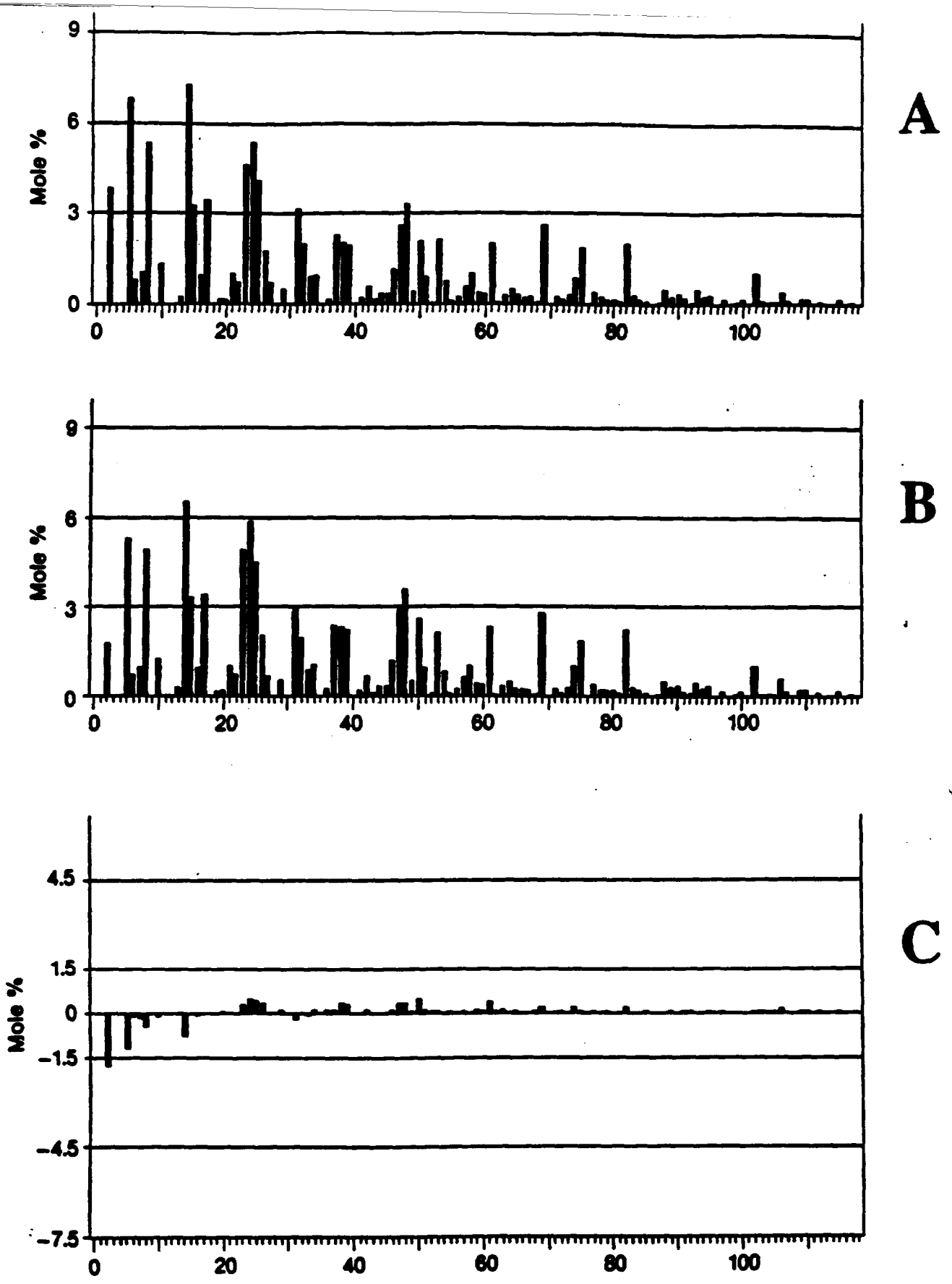
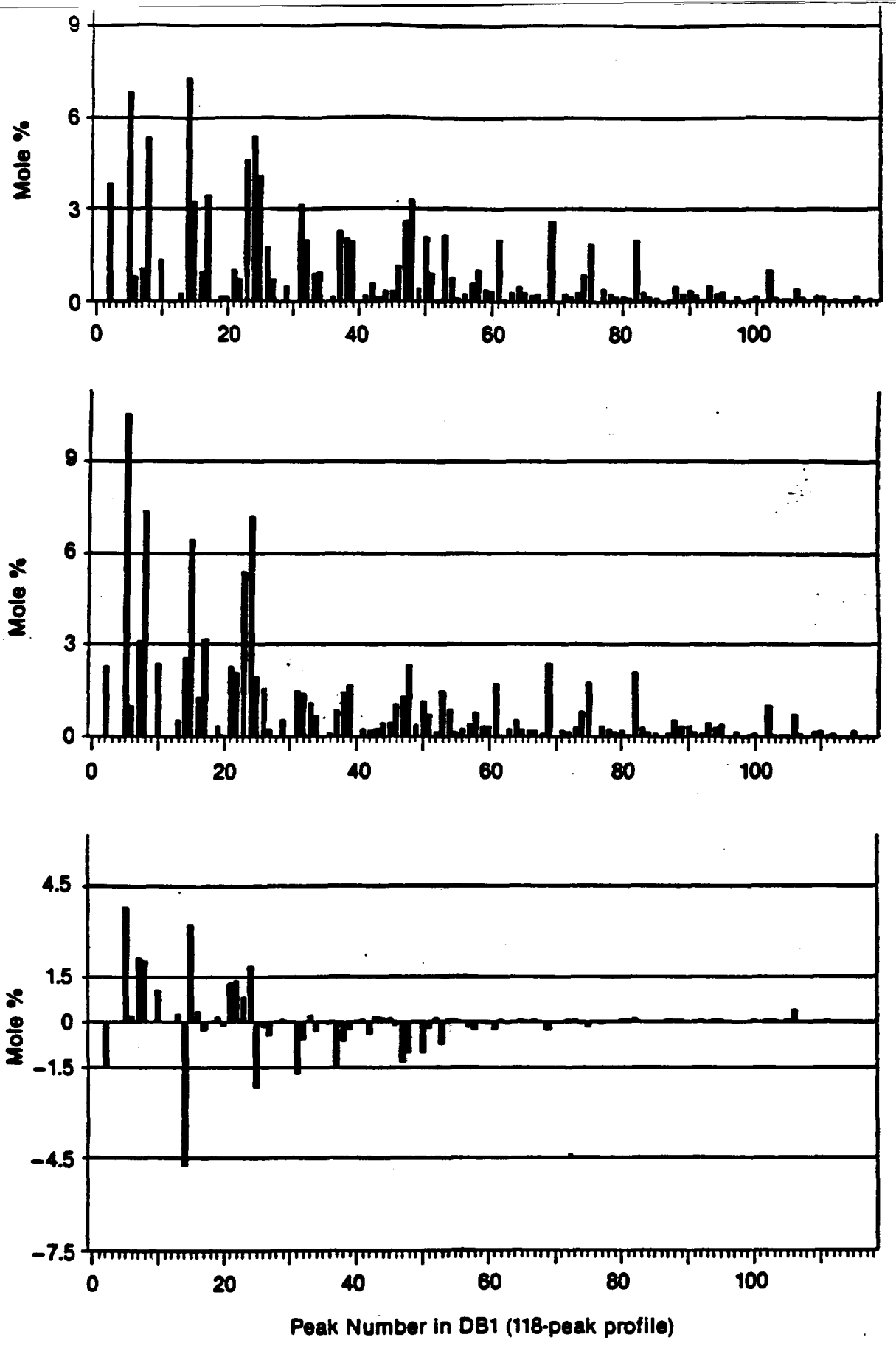


Figure 6-1: Acceleration of the reductive dechlorination of PCBs upon addition of nutrients (8 week timepoint). Panel A and D, autoclaved control; Panel B, includes distilled water; Panel C, difference (B-A); Panel E, includes RAMM; Panel F, difference (E-D). All samples contain 500 ppm PCB mixture.


D
Mole %
9
6
3
0
0
20
40
60
80
100
E
Mole %
9
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3
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40
60
80
100
F
Mole %
4.5
1.5
0
-1.5
-4.5
-7.5
0
20
40
60
80
100
Peak Number in DB1 (118-peak profile)

cultures of *Pseudomonas* strain LB400 and *Corynebacterium* strain MB1 on biphenyl as the sole carbon source. Cells were harvested, washed, and added to contaminated sediments at 1 OD with a 20% soil loading by weight. Individual vials were sacrificed for timepoints by extraction of the entire contents with three volumes of diethyl ether. Samples were then analyzed by high resolution gas chromatography with an electron capture detector on a fused silica capillary column (30 m by 0.25 mm ID) coated with a 0.25 μm bonded liquid phase of DB-1.

The addition of a minimal medium to the sediment slurry resulted in a dramatic increase in the observed rate of anaerobic dechlorination after 8 weeks (see Figure 6-1). The RAMM minimal medium contained nutrients, trace minerals, and bicarbonate [Shelton and Tiedje, 1984]. The control was autoclaved and incubated along with the samples; no change was observed in any of the heat-treated controls during the experiments. The control (Figure 6-1A and 6-1D), therefore, represents the PCB distribution in the original mixture added to the sediment (70% Aroclor 1242, 20% Aroclor 1254, 10% Aroclor 1260).

The sample mixed with distilled water in place of the minimal medium is shown in Figure 6-1B. Only slight activity was observed in this experimental sample after an 8-week incubation. This is contrasted by the significant change observed in the sample to which RAMM minimal medium had been added (Figure 6-1E). A direct comparison can be found by comparing the difference plots in Figure 6-1C and 6-1F. The control and water-only sample contains nearly the same average number of chlorines per biphenyl (Cl/BP; 3.75 and 3.78, respectively), indicating that no significant dechlorination had occurred at this timepoint. The sample with RAMM nutrients added contains 3.52 Cl/BP, indicating a loss of approximately 0.25 Cl/BP during the initial 2 months of the experiment. The selective *meta* and *para* dechlorination observed in this sample continued at later timepoints, and is consistent with the environmental changes observed in the Hudson River [Brown et al., 1987].

This result suggests that a limiting nutrient present in RAMM medium may be restricting the rate of dechlorination in upper Hudson River sediments. It should be noted that at later timepoints significant dechlorination was also observed in the sediment to which no nutrients were added. These changes were similar although less extensive than the sample to which nutrients were added. Therefore, nutrient addition can decrease the lag time before activity is initiated, as well as increase the extent of dechlorination observed.

The RAMM medium was subdivided into four different components to further investigate nutrient stimulation of this PCB dechlorination activity. Individual components were added in various combinations and concentrations; results are shown in Table 6-1. Note that the addition of the trace metals (Zn^{+2}, Cu^{+2}, Ni^{+2}, Se^{+2}, B^{+3}) correlates with nearly a two-fold increase in the rate of dechlorination of 234-34-CB. This effect suggests that one of these trace metals, added at less than the 0.02 ppm level, may represent the component that limits the PCB dechlorination rate in Hudson River sediments. Other agents that have been demonstrated to increase the rate and/or extent of PCB dechlorination in Hudson River sediments include non-ionic high molecular weight surfactants (e.g., Triton X-705) and the addition of a complex carbon source (e.g., yeast extract or fluid thioglycollate medium

TABLE 6-1. Effect of RAMM components on PCB dechlorination rate (500 ppm 234-34-CB).

Relative Dechlorination Rate	A	B	C	D
105%	+			
90	+	+		
105	+	+	+	
171	+	+	+	+
210	+	++	+	+
171	++	++	+	+
202	+	+		+
210	+	+	++	+
191	+	+		++

A = phosphate salts, cysteine, HCO_3^-
B = nitrogen + minerals (Ca^{+2}, Mg^{+2}, Fe^{+2})
C = Mn^{+2}, Mo^{+6}, Co^{+2}
D = trace metals (Zn^{+2}, Cu^{+2}, Ni^{+2}, Se^{+2}, B^{+3})

with beef extract). In addition, PCB dechlorination has been observed over a broad range of PCB concentrations (20-1500 ppm) and on Aroclor 1260 [Abramowicz et al., 1990].

It has been reported that corrinoids, including cobalamin vitamin B_{12}, can catalyze the reductive dechlorination of chlorinated methanes in the presence of an electron donor [Krone et al., 1989]. It is possible that these cobalt-heme coenzymes may play a role in anaerobic microbial dehalogenations of PCBs as well. Therefore, the effect of vitamin B_{12} addition on the rate of dechlorination was investigated at various concentrations, and the results are shown in Figure 6-2. The unexpected results display a classic inhibition curve for the dechlorination of PCBs upon vitamin B_{12} addition. The decreased dechlorination activity observed in these experiments can be explained by the known inactivation of enzymes with vitamin B_{12} in the presence of small molecular weight thiols [Harada et al., 1975a, 1975b, 1975c]. Harada and coworkers showed that μM concentrations of corrinoids like cyanocobinamide (Factor B) and cobalamin (Vitamin B_{12}) inactivate papain and yeast alcohol dehydrogenase via the formation of mixed disulfides between small molecular weight thiols and the enzymes. Our RAMM medium contains cysteine and the observed inhibition of PCB dechlorination may be explained by a similar mechanism. An alternate explanation could involve naturally produced thiols as the nucleophile responsible for the *in vivo* dechlorination. Thiols are known to be involved in the dechlorination of chlorobenzenes [Stiles, personal communication]. Therefore, the coenzyme could be inhibiting dechlorination by inactivating the natural thiol nucleophile via disulfide formation.

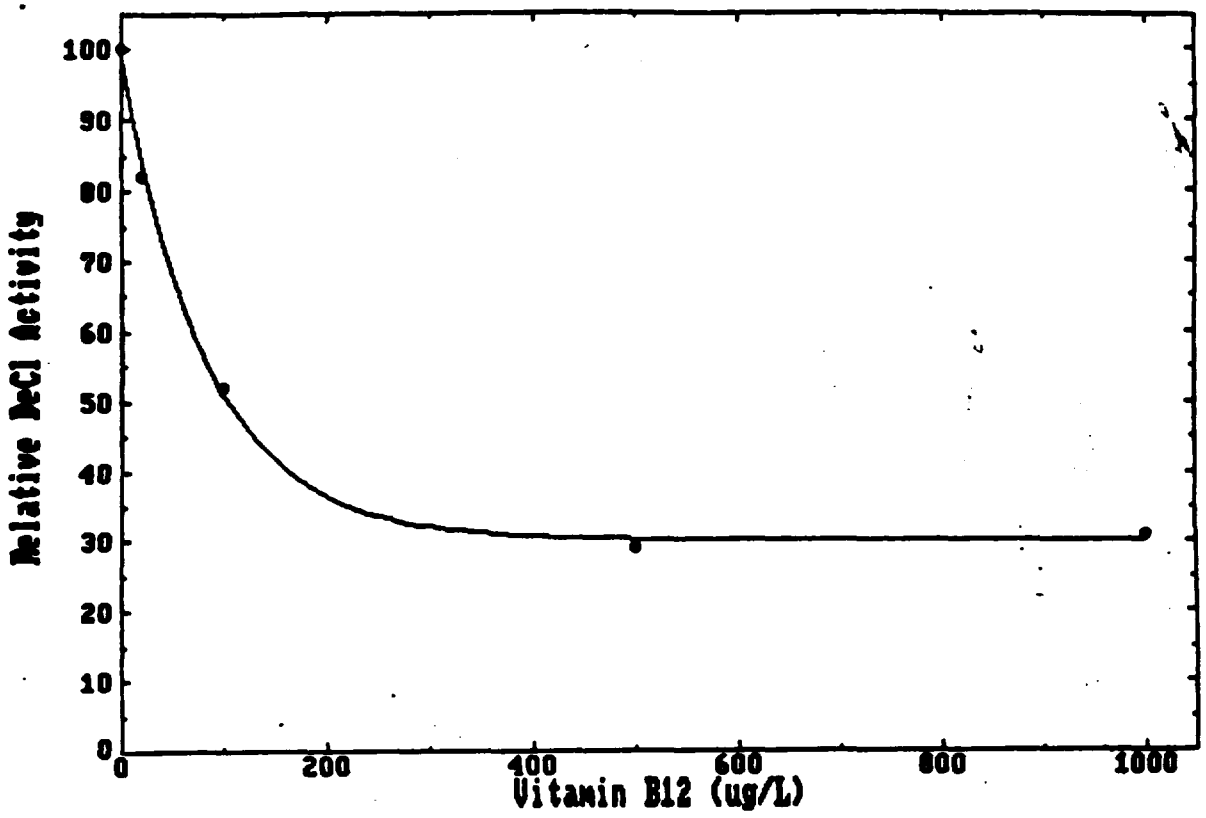

Figure 6-2: Effect of Vitamin B_{12} addition on the rate of dechlorination (100 ppm 234-34-CB).

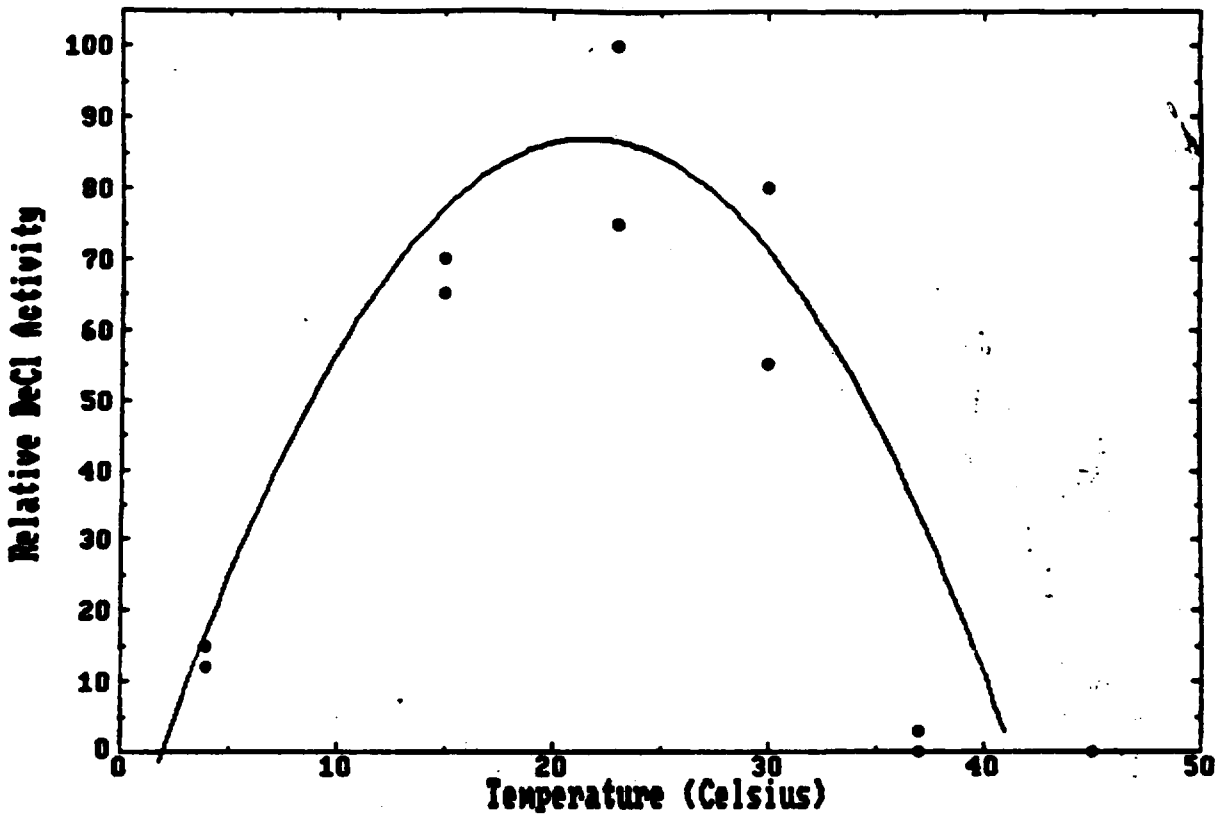

Figure 6-3: Temperature profile of dechlorination activity with Hudson River sediments. All samples contain 500 ppm PCB mixture.

Initial experiments to investigate the effect of temperature on dechlorination activity are shown in Figure 6-3. Note that the dechlorination occurred over a broad range of temperatures (4-30°C), indicating that this process may occur year-round in river sediments. The optimal dechlorination rate was found at 15-30°C.

Endogenous PCBs

It is possible that biodegradation studies on soils spiked with PCBs may not provide accurate kinetic data for similar experiments on endogenous, aged PCB contamination. It has been observed with South Glens Falls dragstrip soil that aerobic biodegradation rates can be limited due to bioavailability issues [GE Report, 1990]. Therefore, several different contaminated soils and sediments were investigated to directly monitor the PCB dechlorination rate of the endogenous contamination.

Upper Hudson River sediments contaminated >15 years ago have already been extensively dechlorinated in the environment [Brown et al., 1987]. Such sediments can be even further dechlorinated by the addition of RAMM nutrients (see Figure 6-4). Note that the environmental sample (Figure 6-4A) contains ~ 41% 2-CB (peak 2) and 2-2/26-CB (peak 5). The mole fraction represented by these congeners increases to >63% after RAMM addition (Figure 6-4B). This dechlorination activity attacks all of the more highly chlorinated congeners (difference plot in Figure 6-4C), and is similar to environmental Pattern C. The dechlorination rate is comparable to that observed in samples spiked with PCB. Endogenous PCB contamination can also be dechlorinated by anaerobic bacteria in Woods Pond sediments (Aroclor 1260, data not shown).

The endogenous PCBs bound to dragstrip soil were also available for dechlorination via anaerobic microorganisms (see Figure 6-5). In this experiment, 25% by weight Hudson River sediments were added to the dragstrip soil containing RAMM medium. Again, this microbial process successfully attacked the endogenous PCB contamination. The control sample contains on average 3.86 Cl/BP. This value has decreased to 3.64 in the live sample (Figure 6-5B). The difference plot (Figure 6-5C) displays strong losses of the more highly chlorinated congeners (5 or more Cl), with the production of tri- and di-CBs. This result is particularly encouraging since this same soil demonstrated bioavailability limitations upon aerobic treatment [Chapter 12, GE Report 1990].

Anaerobic microorganisms capable of PCB reductive dechlorination are widespread and exist in even uncontaminated river sediments, as shown in Table 6-2. The Spier Falls sediments are located >10 miles upstream of the source of contamination (river mile 205) and contain <1 ppm PCB. This is compared to the H7 site, located downstream of the source (river mile 193.5) and containing 45 ppm PCB. The dechlorinating organisms are found in such uncontaminated sediments, although they appear to be present at lower concentrations and dechlorination rates are about four-fold slower. This PCB dechlorination activity has been observed in a number of other aquatic sediments, including Woods Pond (Lenox, MA), Silver Lake (Pittsfield, MA), New Bedford Harbor (MA), Escambia Bay (Pensacola, FL), the

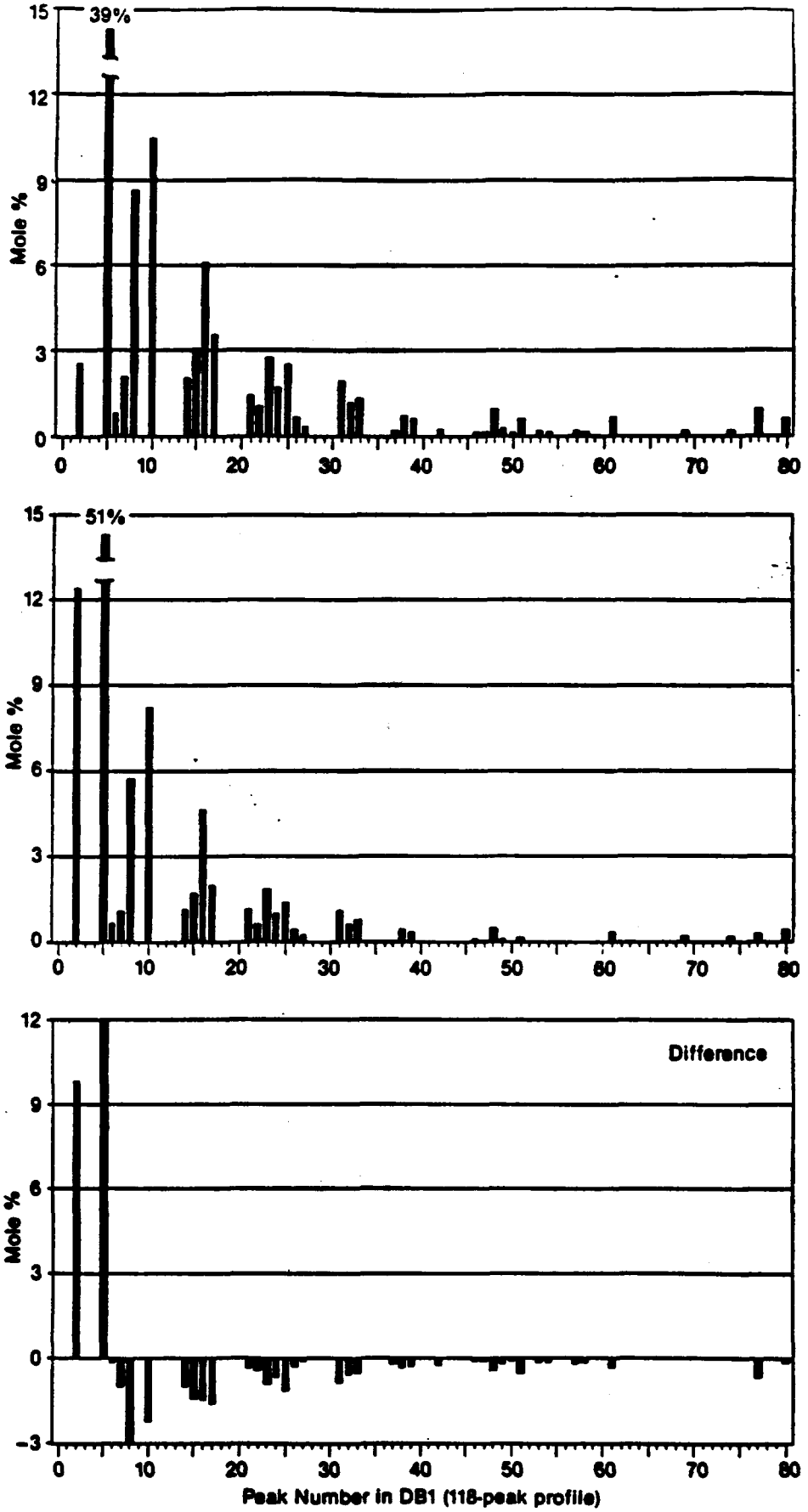

Figure 6-4: Dechlorination of endogenous PCB contamination in Hudson River sediments (24 week timepoint, H7 site). Panel A, autoclaved control; Panel B, experimental; Panel C, difference.

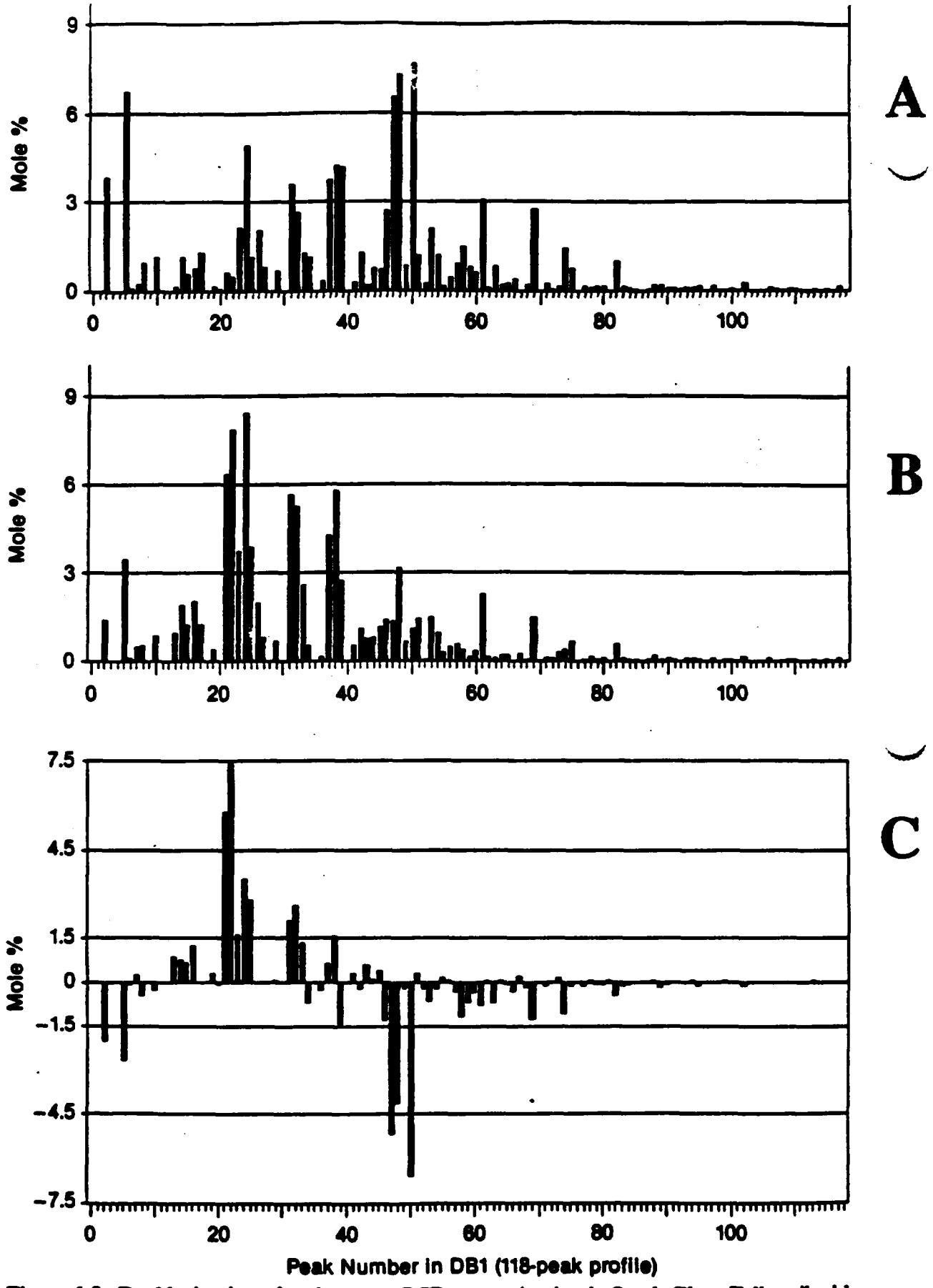

Figure 6-5: Dechlorination of endogenous PCB contamination in South Glens Falls soil with 25% Hudson River sediment (24 week timepoint). Panel A, autoclaved control; Panel B, experimental; Panel C, difference.

Housatonic River (CT), the Sheboygan River (WI), Waukegan Harbor (IL), the Hoosic River (North Adams, MA), and many different locations in the Hudson River (NY). This widespread occurrence of natural anaerobic dechlorination in different sediments indicates that it is a general phenomenon that occurs in environments that differ dramatically in their geography, seasonal temperature, PCB concentration, PCB composition, and organic content.

TABLE 6-2. Comparison of PCB dechlorination rates observed with PCB-contaminated (H7) and uncontaminated (Spier Falls) Hudson River sediments. (500 ppm 234-34-CB)

Time (wk)	% Dechlorination	
	Contaminated	Uncontaminated
0	0.0	0.0
4	5.0	0.0
8	19.7	0.0
12	23.6	0.0
18	ND	9.0
32	90.0	18.2

This natural dechlorination of endogenous PCB contamination is still occurring in the environment. The H7 Hudson River site has been repeatedly sampled over the last 10 years. A timecourse displaying the observed PCB distribution is shown in Figure 6-6. Note that even the early sample (Figure 6-6A) displays a significant amount of 2-CB and 2-2/26-CB (see arrows), indicating that dechlorination is already well advanced. In our most recent sampling (Figure 6-6C), the 2-CB and 2-2/26-CB now account for >85% of the total PCB in the sample. Six samples taken at this time at 20-foot intervals all displayed similar PCB distributions, indicating that nearly all of the *meta* and *para* chlorines have been removed to date (1.63 Cl/BP, including <0.1 non-*ortho* chlorines).

Sequential Anaerobic/Aerobic Treatment

The *ortho* substituted, lower chlorinated products of anaerobic dechlorination should be good substrates for subsequent aerobic biodegradation. Therefore, Hudson River sediments dechlorinated in the laboratory (as described above) were then subjected to aerobic treatment, and the results are shown in Figure 6-7. The control bars indicate the original PCB mixture added to the sample. Anaerobic treatment results in the classic redistribution previously described, and the total PCB concentration has not been significantly decreased. In contrast, subsequent aerobic treatment now reduces the PCB concentration 85% (mole percent values have been reduced by 85% for these bars to reflect this decrease).

Similar experiments have also been performed on endogenous PCB contamination. Hudson River sediments that had previously undergone environmental dechlorination were

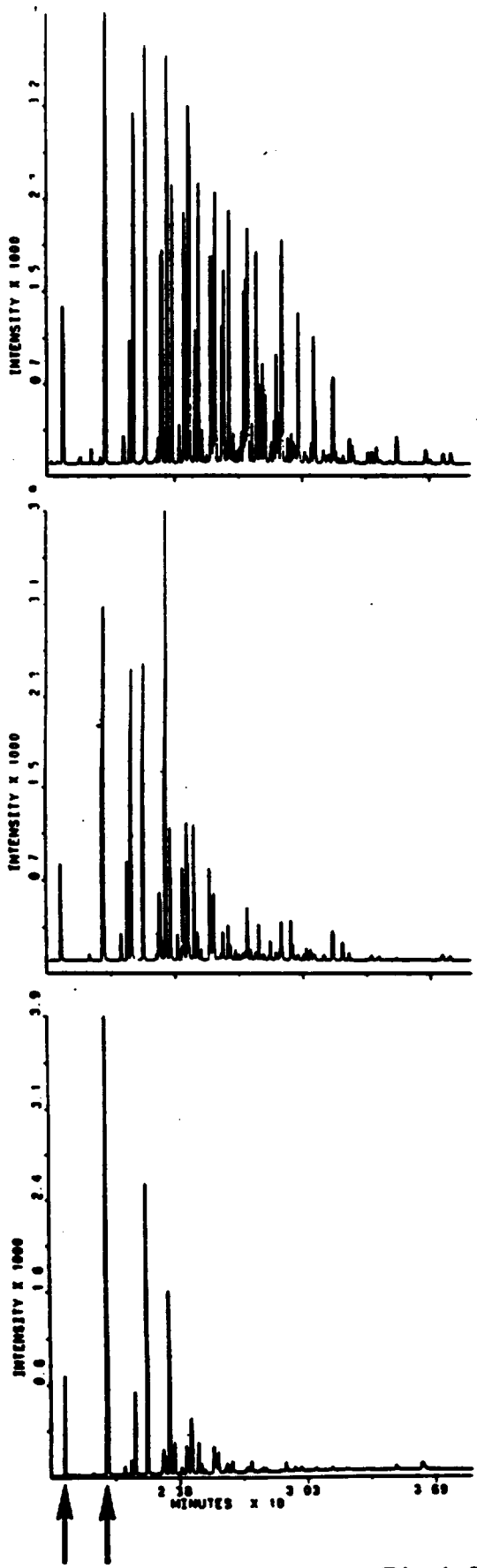

Figure 6-6: PCB distribution at H7 site (Hudson River). Panel A, year 1982; Panel B, year 1985; Panel C, year 1990.

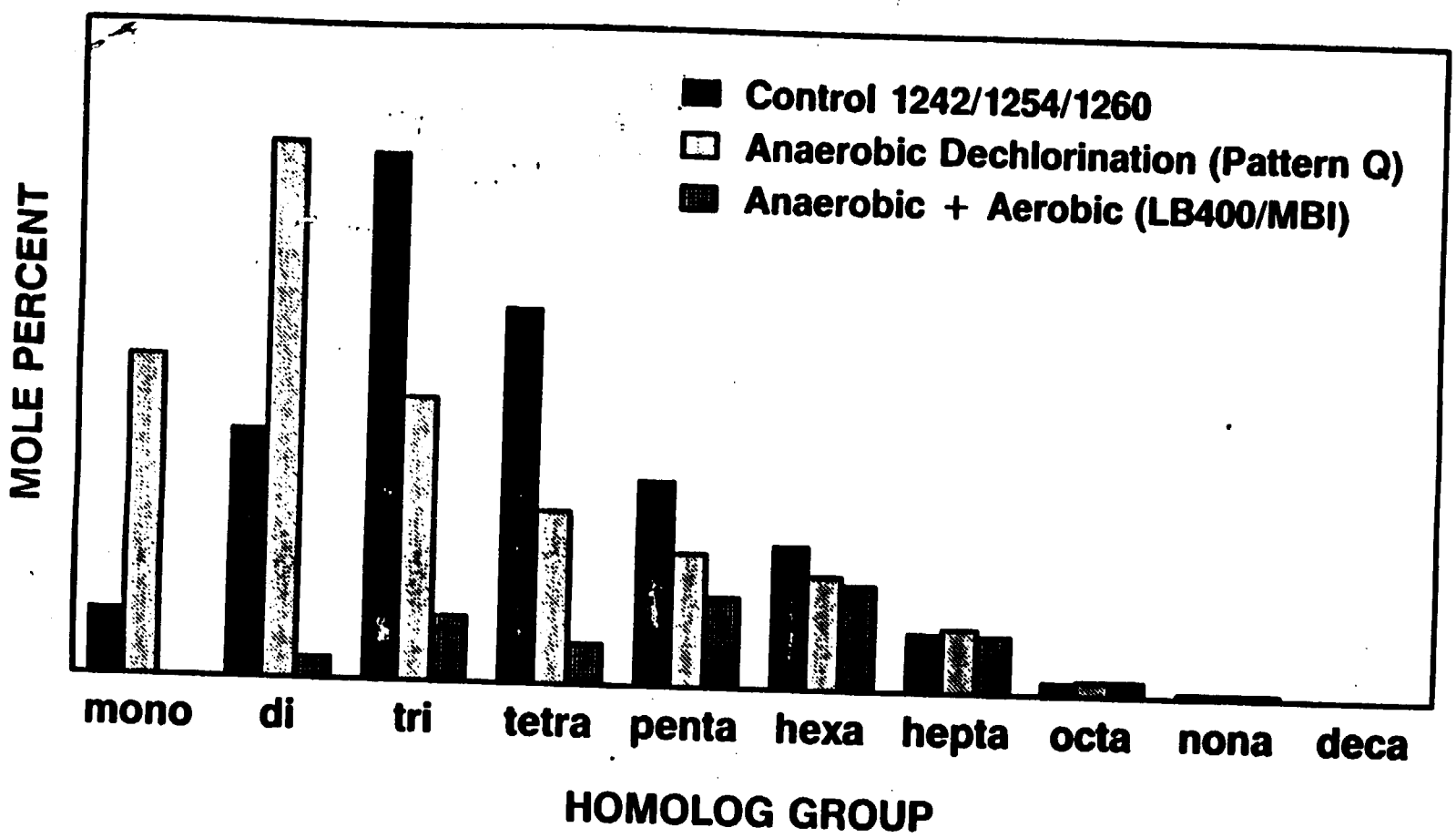

Figure 6-7: Sequential anaerobic/aerobic treatment of spiked PCB contamination in Hudson River sediments

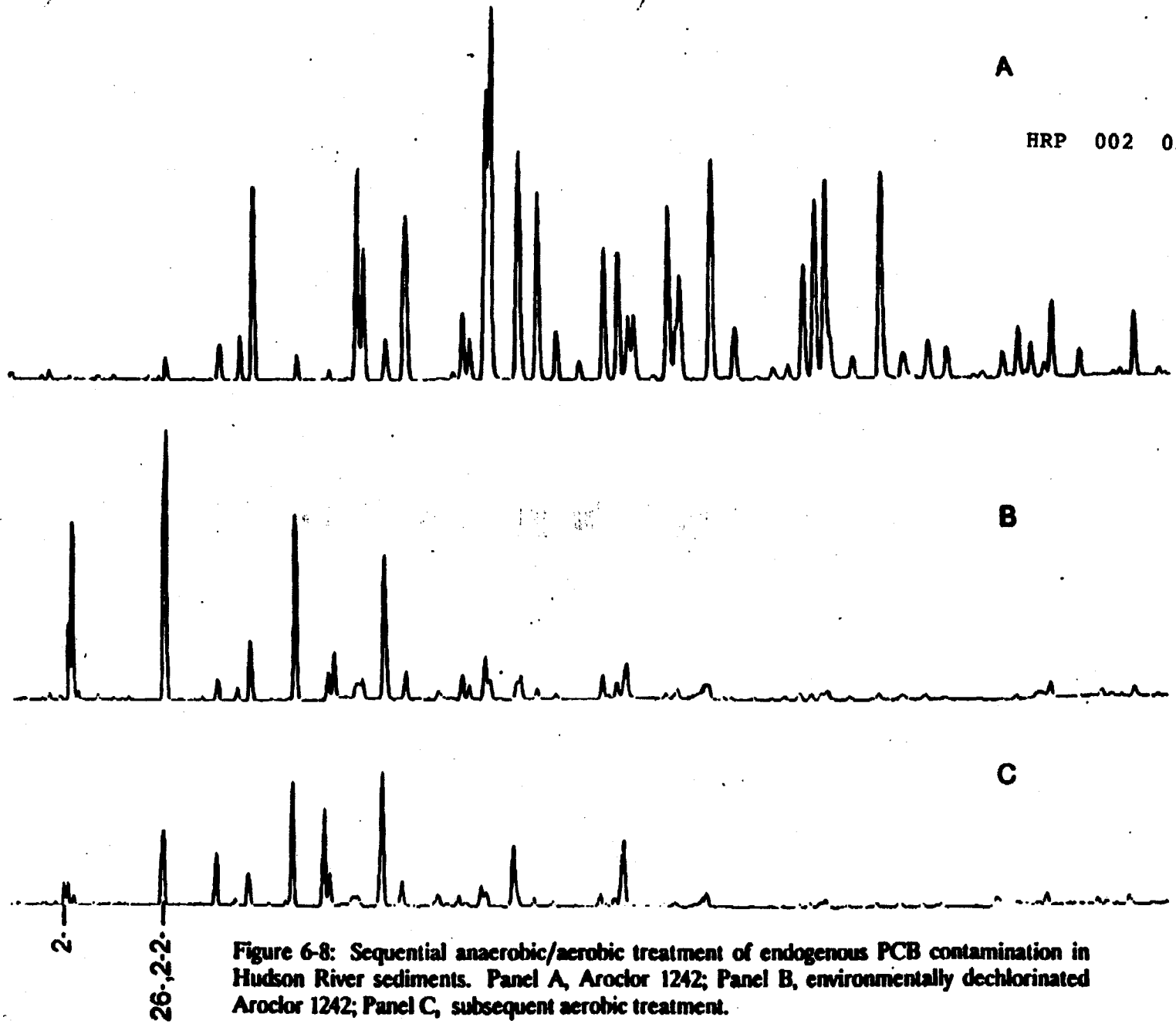

Figure 6-8: Sequential anaerobic/aerobic treatment of endogenous PCB contamination in Hudson River sediments. Panel A, Aroclor 1242; Panel B, environmentally dechlorinated Aroclor 1242; Panel C, subsequent aerobic treatment.

then treated by aerobic PCB-degrading organisms to demonstrate the effect of this combined process (see Figure 6-8). Figure 6-8A represents the original contamination (Aroclor 1242); Figure 6-8B displays a recently obtained sediment sample from the Hudson River that has been environmentally dechlorinated (>85% mono- and di-CB); Figure 6-8C displays the resulting chromatogram after aerobic treatment. This initial trial demonstrated ~80% reduction in the PCB concentration after 1 day of aerobic treatment. Therefore, this two-stage process has resulted in a dramatic change in the PCB distribution to lower chlorinated products, as well as a marked reduction in the PCB concentration.

SUMMARY AND CONCLUSIONS

Hudson River sediments contain anaerobic microorganisms capable of extensively dechlorinating PCB mixtures. The observed rate of laboratory dechlorination can be stimulated by the addition of a trace metal mixture or detergents at low concentrations. This dechlorination occurs over a broad range of temperatures (4-30°C), and the organisms are even capable of attacking the highly chlorinated Aroclor 1260.

Experiments on a variety of PCB-contaminated soils have demonstrated that this anaerobic process will effectively attack endogenous PCB contamination. No significant rate difference was observable for endogenous or spiked PCB samples. In addition, sequential anaerobic/aerobic treatment of the PCB contamination present in Hudson River sediments have resulted in a dramatic shift in PCB distribution to lower chlorinated, *ortho* substituted products and an 80% reduction in total PCB concentrations.

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Chapter 7

SEQUENTIAL ANAEROBIC-AEROBIC BIODEGRADATION OF PCBs

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INTRODUCTION

Evidence for the reductive dechlorination of PCBs under anaerobic conditions has accumulated from studies of PCB congener pattern changes of commercial Aroclors in natural sediments [Brown et al., 1984, 1987a,b; GE Reports, 1987, 1988]. Recent laboratory studies confirm that the anaerobic dechlorination of PCBs is due to biological activity [Quensen, 1988]. Several laboratory studies have shown that a broad range of PCB congeners can be aerobically degraded by microorganisms [Bedard et al., 1986, 1987a,b]. Aerobic degradation is usually faster for congeners with fewer chlorine substituents. Although sequential anaerobic dechlorination and aerobic degradation of PCBs have not been observed in natural sediments, extensive aerobic degradation of anaerobically transformed Aroclors has been shown [Bedard et al., 1987b]. Initial attempts at a sequential anaerobic-aerobic biodegradation of PCBs in our laboratory have also been relatively successful. These results provide evidence for the potential of complete *in situ* degradation of PCBs.

Last year we reported on the effects of substrate amendments for enhancing the anaerobic reductive dechlorination of Aroclor 1242 in Hudson River sediments. Acetone and methanol, compounds often used as solvents for the addition of PCBs in laboratory studies, and glucose were all shown to have a beneficial effect on the rate of dechlorination relative to incubations receiving no substrate amendments. These incubations were done in non-autoclaved sediments [GE Report, 1989; Nies and Vogel, 1990]. In addition, Fluid Thioglycollate Medium with Beef Extract (FTMBE) or sediment extract (organic solubilized by autoclaving) were also shown to enhance dechlorination activity [GE Report, 1989].

Since last year, we have completed a more extensive study of substrate amendments by varying the concentration of the different substrates added. These more recent anaerobic studies have all been done without the use of any oxygen scavenging reducing agents, in order to model more closely the environmental conditions that would be encountered during *in situ* bioremediation. We have demonstrated that a sequential anaerobic-aerobic biodegradation scheme is possible in the laboratory by inducing anaerobic conditions in sediments where PCBs were reductively dechlorinated, and then aerobically degrading many of the less chlorinated PCB congeners in those sediments. We have transferred anaerobic Hudson River organisms from sediments to incubations with sterile sand or glass beads, while maintaining dechlorinating activity. We have also shown that anaerobic river sediments that had not previously been contaminated with PCBs can dechlorinate Aroclor 1242 without long acclimation times.

RESULTS AND DISCUSSION

Anaerobic Substrate Effects

Anaerobic batch incubations of Hudson River sediments were started to begin the first phase of the anaerobic-aerobic biodegradation sequence. In order to test independently the effects of different organic substrates on the dechlorinating microorganisms, the sediments were respiked with Aroclor 1242 without the addition of a solvent [GE Report, 1989; Nies and Vogel, 1990]. The batches were inoculated with microorganisms that had been acclimated to the particular substrate being tested. The organic substrates tested were acetate, acetone, butanol, glucose, and methanol. Each substrate was tested at three different concentrations (high, medium, low) across three orders of magnitude (Table 7-1). The sediments were not autoclaved prior to inoculation, as this would solubilize large amounts of organic carbon and potentially mask differences between the substrates being tested. Control batches were autoclaved. Anaerobic conditions were induced by substrate addition and subsequent microbial consumption of oxygen. Batches that had been started previously, without the addition of any organic substrate, had sodium sulfide added to maintain anaerobic conditions.

TABLE 7-1. Substrate amounts added to anaerobic incubations[a]

	Acetate	Acetone	Butanol
H	0.78%	0.67%	0.54%
M	0.078%	0.067%	0.054%
L	0.0078%	0.0067%	0.0054%
	Glucose	**Methanol**	
H	0.93%	0.67%	
M	0.093%	0.067%	
L	0.0093%	0.0067%	

[a] - Substrate amount given as weight percent of substrate per wet weight of sediment.

H-High level M-Medium level L-Low level

High levels of acetone and methanol, and medium levels of glucose, all enhanced reductive dechlorination with the extent and pattern being nearly identical (Figure 7-1a). High

TABLE 7-2: Congener assignments for chromatographic peak numbers in this report.

Peak number	Congeners in peak		
1	2		
2	2-2	2,6-	
3	2,5-		2,4-
4	2-3		
5	2-4	2,3-	
6	2,6-2		
7	2,5-2	4-4	
8	2,4-2		
9	2,6-3	2,3,6-	
10	2,3-2	2,6-4	
11	2,5-3		
12	2,4-3		
13	2,5-4		
14	2,4-4	2,4,6-2	
15	2-3,4	2,3-3	2,3,4-
16	2,5-2,6		
17	2,4-2,6		
18	2,3-4		
19	2,3,6-2		
20	2,3-2,6		
21	2,5-2,5	3,5-2,6	
22	2,4,-2,5		
23	2,4-2,4		
24	2,4,5-2	2,4,6-4	
25	2,3-2,5		
26	3,4-4		
27	2,3-2,4	2,3,6-3	
28	2,6-3,4	2,3,6-4	2,3,4-2
29	2,3-2,3		
30	2,4,5-4	2,3,5-2,6	
31	2,5-3,4	2-3,4,5	
32	2,4-3,4	2,4,5-2,6	
33	2,3,6-2,5		
34	2,3,6-2,4		
35	2,3-3,4	2,3,4-4	
36	2,4,5-2,5	2,3,5-2,4	
37	2,3,6-3,5		
38	2,4,5-2,3	2,3,4,5-2	2,3,5,6-2,6
39	2,3,4-2,5	2,3,4,6-4	
40	3,4-3,4	2,3,6-3,4	
41	2,3,5,6-2,5		
42	2,4,5-3,4	2,3,4,5-3	2,3,6-2,4,5
43	2,4,5-2,4,5		
44	2,3,4-3,4	2,3,4-2,3,6	

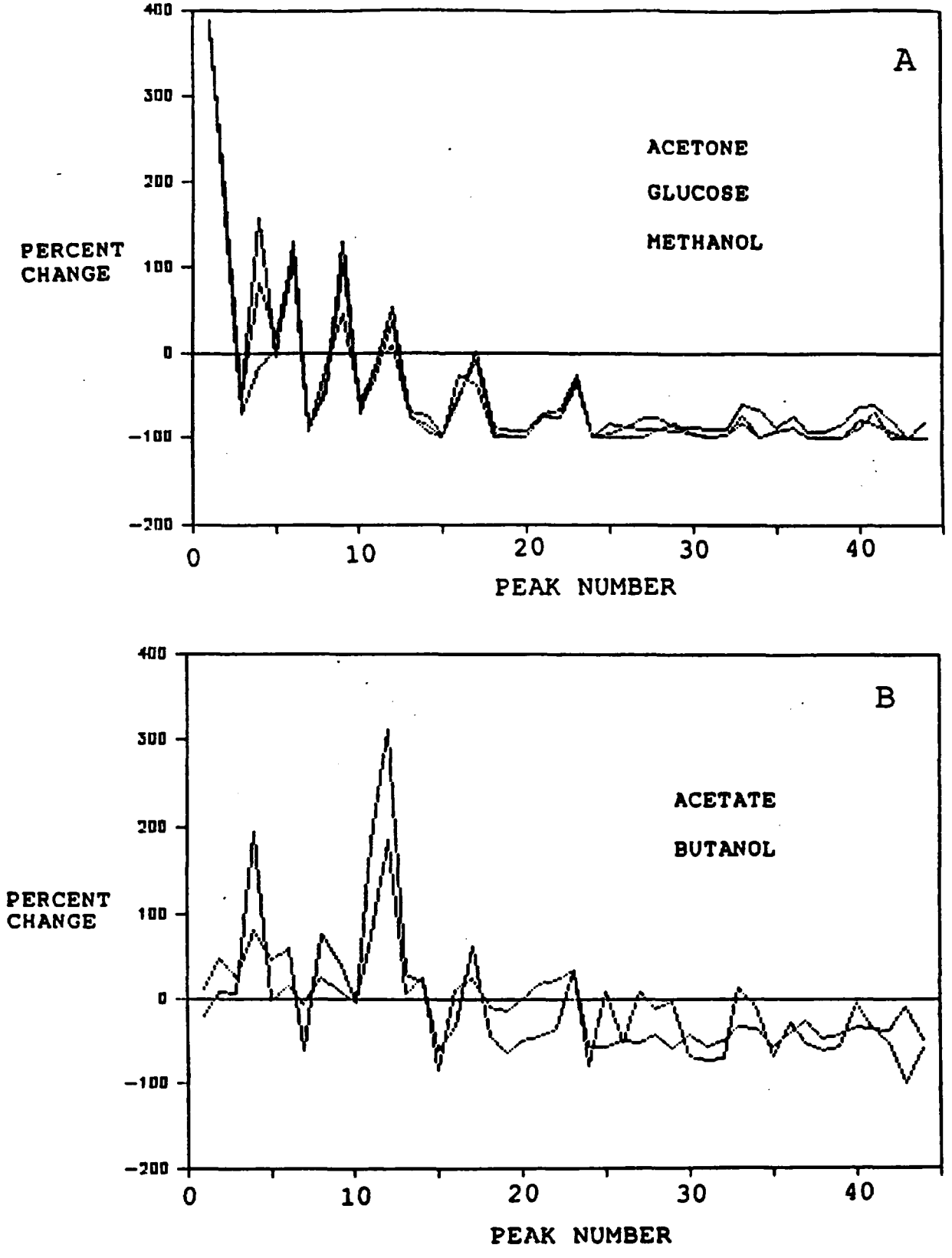

Figure 7-1. Difference plot of anaerobic substrate-fed batches at 20 weeks: a) acetone-, glucose-, and methanol-fed batches; b) acetate- and butanol-fed batches. Note: congener peaks are discrete points, lines are drawn between points for ease of comparison.

levels of glucose caused a drop in pH in those batches, which inhibited dechlorination activity. Acetate and butanol also enhanced dechlorination, relative to batches receiving no substrate, although the patterns were different and dechlorination was not as extensive as the other substrates tested, as indicated by the accumulation of peak number 12 (2,4-3-CB) relative to acetone-, glucose-, and methanol-fed batches (Figure 7-1b). The congener assignments for the peak numbers in Figures 7-1 and 7-3, are given in Table 7-2. After more than 54 weeks incubation, slight dechlorination has been observed in batches receiving no additional organic substrate.

All substrate-fed batches showed an increase in relative rates and extent of reductive dechlorination with an increase in substrate concentration from low to medium levels. The transformation of Aroclor 1242 by reductive dechlorination is illustrated in Figure 7-2 by the decrease in tri-, tetra-, penta-, and hexachlorobiphenyl homologs, and the increase in the mono- and dichlorobiphenyl homologs. After 20 weeks incubation, the mono- and dichlorobiphenyls account for ~ 80% of the total PCBs present. Dechlorination in acetone- and methanol-fed batches also increased when substrate was increased from medium to high levels. An example of the increase in rate and extent of dechlorination with increased substrate levels is shown in Figure 7-3. Results from acetate-, butanol-, and glucose-fed batches indicate that the high substrate levels may have some slight inhibitory effect on dechlorination relative to medium substrate levels.

Microbial Enrichments

Serum bottles with sterile sand or glass beads (4 mm) and mineral medium were spiked with Aroclor 1242 dissolved in acetone. Dechlorinating Hudson River microbial enrichments, acclimated to acetone, were used as an inoculum. Reductive dechlorination has been observed in all batches, roughly coinciding with the onset of methanogenesis. These consortia of microorganisms are only one transfer removed from Hudson River sediments. If successive attempts to transfer these dechlorinating enrichments to inert substratum are successful, this will provide a unique opportunity to study the microorganisms responsible for reductive dechlorination of PCBs in a defined environment.

Dechlorination in Non-PCB-Contaminated Sediments

Aroclor 1242 was added to anaerobic sediments taken from the Saline River, Michigan. Acetone, glucose, and methanol were added to different batches as substrate. After 8 weeks, dechlorination activity was observed in all batches. This sediment has no detectable background levels of PCBs, and no history of previous PCB-contamination. It appears that dechlorination of PCBs by microorganisms does not require long (years) acclimation times. There may be PCB dechlorinating capability across broad classes of microorganisms, rather than adaptation to dechlorination of PCBs by unique organisms.

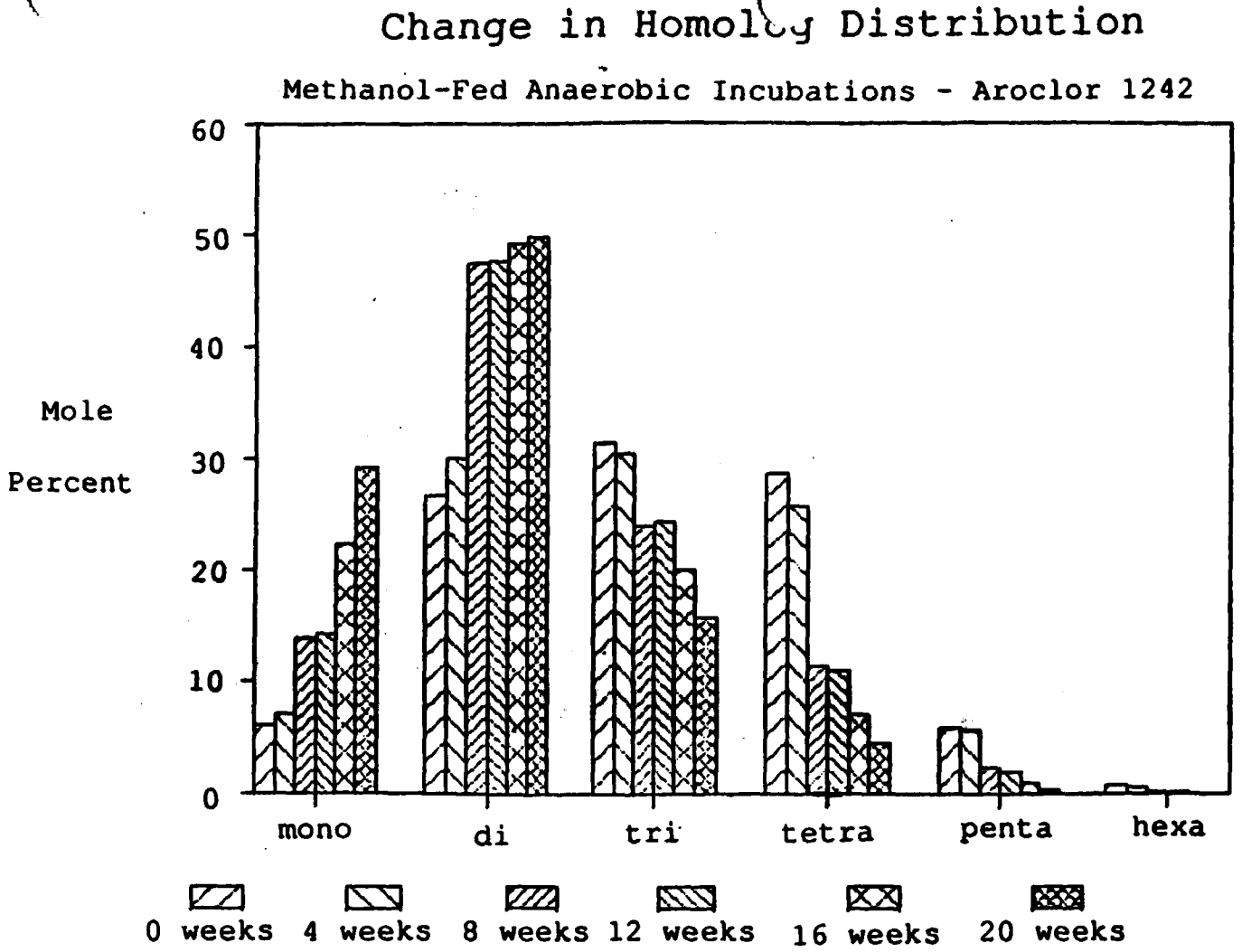

Figure 7-2. Change in homolog distribution of Aroclor 1242 in methanol-fed (high level) batches through 20 weeks.

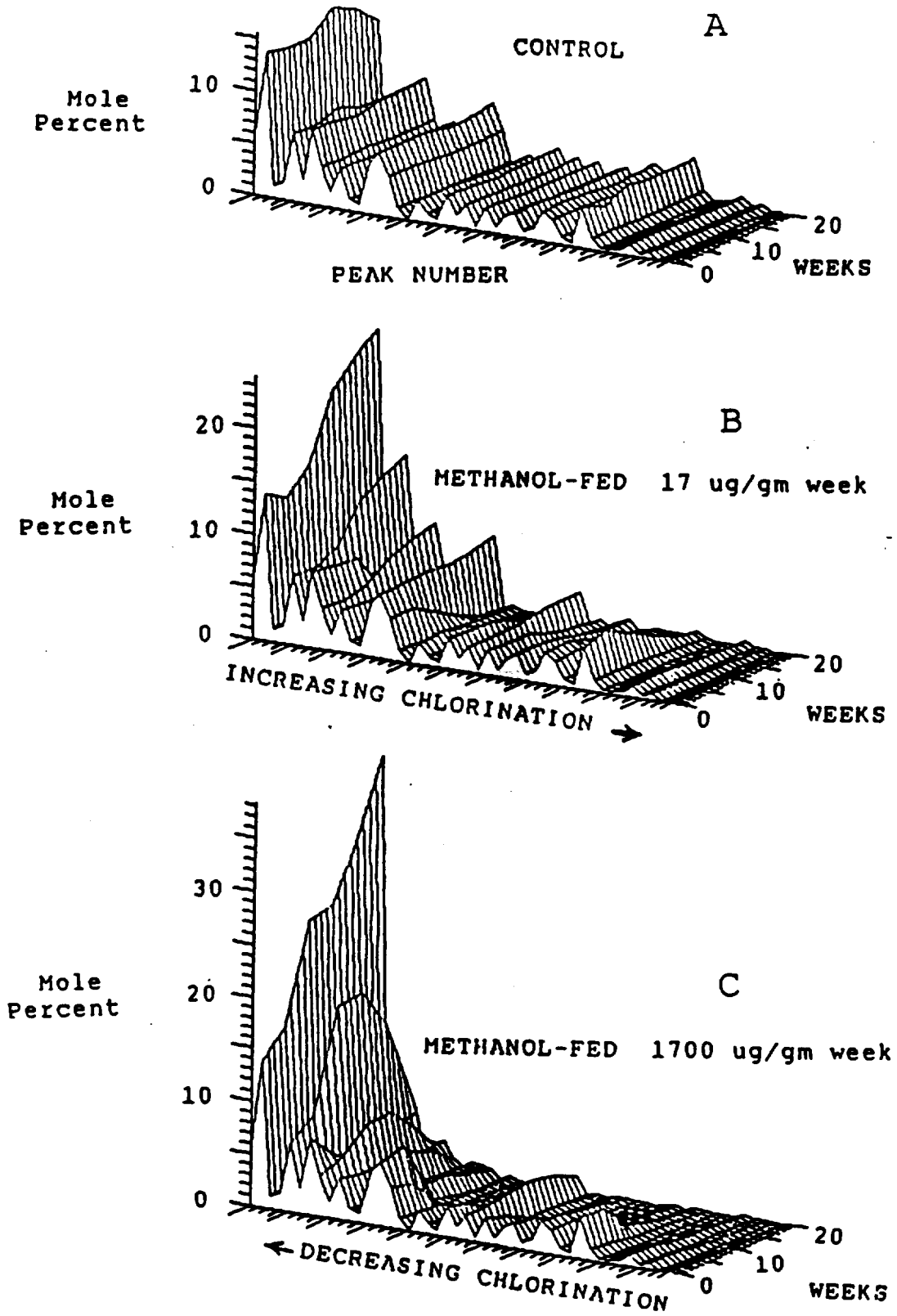

Figure 7-3. Change in congener distribution of Aroclor 1242 in methanol-fed batches through 20 weeks: a) autoclaved controls; b) low methanol level; c) high methanol level.

AEROBIC DEGRADATION

Rapid Assay

Biphenyl-degrading microorganisms were enriched from more than 20 sediment and soil samples. Although some of the samples had no detectable PCBs present, all samples yielded cultures of biphenyl degraders. These cultures were then screened for PCB degradative competence using the rapid assay procedure [Bedard et al., 1986] with the congener mixture shown in Table 7-3 at a concentration of 5 μM/congener. The enrichments varied in their ability to degrade the dichlorobiphenyl congeners, and none of the isolates showed any ability to degrade tetrachlorobiphenyl congeners. The rapid assay results of the two most competent isolates are shown in Table 7-3, along with the results using the pure culture LB400 for comparison. Note that significantly better degradation has been observed with LB400 under similar conditions by others [Mondello, 1989; Bedard et al., 1986]. The Hudson River isolate, S3, was selected to test for aerobic degradation of PCB congeners on anaerobically dechlorinated sediment.

TABLE 7-3. Rapid assay degradation of PCBs by biphenyl grown microorganisms (percent degraded 96 hours)[a]

	Culture		
Congener Mix	16	S3	LB400
2-2	100	100	100
2,3-	100	100	100
4-4	100	100	0
2,5-2,6	0	0	17
2,5-2,5	0	0	75
2,4-2,5	0	0	43
2,4-2,4	0	0	0
3,4-3,4	0	0	0
3,5-3,5[b]	0	0	0
2,4,5-2,4,5	0	0	0

[a] - Degradation of less than 10% is not considered significant and is shown as 0%.

[b] - Internal standard.

Aerobic PCB Degradation in Sediments

Anaerobic incubations (dechlorinated sediments and mineral medium) were purged with pure oxygen for 5 minutes. Approximately 4 mLs of freshly grown S3 was then added to the incubations. After 96 hours, live and autoclaved control samples were extracted. Significant degradation of all mono- and dichlorobiphenyl congeners occurred, except for the di*ortho* substituted 2-2:2,6-CB (Table 7-4). The total PCB concentration in the sediment was reduced from 300 ppm to less than 50 ppm by the sequential anaerobic-aerobic treatment. No trichlorobiphenyls were significantly degraded aerobically. These preliminary results can be improved through longer aerobic treatment, use of a more competent organism, or more extensive dechlorination during the anaerobic phase.

TABLE 7-4. Aerobic degradation of anaerobically dechlorinated PCBs on sediment

Congener	Percent Degraded
2-	85
4-	100
2-2 : 2,6-	18
2,5- : 2,4-	94
2-3	72
2-4 : 2,3-	75

SUMMARY AND CONCLUSIONS

Organic substrate may be a limiting factor for anaerobic reductive dechlorination in some sediments. Anaerobic batches with non-autoclaved sediments, receiving inorganic nutrients and vitamins, showed only slight dechlorination after more than 1 year incubation.

Aerobic biphenyl-degrading microorganisms are extremely common in soils and sediments, although their PCB degradative competence varies considerably. However, it is possible that more discriminatory enrichment techniques could select for more competent PCB degraders.

Using aerobic organisms with only "average" PCB-degrading ability, it is possible to significantly reduce PCB levels on sediments using a sequential anaerobic-aerobic biodegradation process.

FUTURE PLANS

The proposed objectives for the coming year will focus on the transition between the anaerobic and aerobic phases of PCB biodegradation. Anaerobic sediments are relatively

high in organic matter and very reduced, therefore, the sediments would be expected to have a large oxygen demand. The use of pure oxygen or hydrogen peroxide for inducing aerobic conditions will be tested. Methods that are potentially applicable to *in situ* bioremediations will be modeled in the laboratory.

Once a suitable aerobic environment has been established, methods to initiate aerobic PCB degradation will be examined. Ideally, there would be indigenous bacteria throughout the sediment that survived the anaerobic phase. Enrichment techniques that select for the most competent PCB degraders will be tested. In addition, the reintroduction of previously enriched PCB-degrading organisms will be tested in order to determine the optimum conditions for effective aerobic PCB degradation *in situ*.

Chapter 8

HUDSON RIVER MODEL

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INTRODUCTION

In situ application is the ultimate aim of most laboratory bioremediation studies. The goal of these studies is to better understand the naturally occurring processes that might aid in the remediation of a contaminated site. This can help in developing strategies that enhance the degradative abilities of the indigenous microorganisms. Laboratory studies have shown that PCBs can be dechlorinated under both anaerobic and aerobic conditions [GE Report,1988; Brown et al., 1987; Quensen et al., 1988] and aerobically oxidized by mixed cultures and pure cultures of microorganisms [Bedard et al., 1987; Bedard et al., 1986; Bopp, 1986; GE Report, 1987]. Generally, anaerobic microbes reduce the chlorine number and aerobic microbes degrade the lightly chlorinated PCBs that remain.

Anaerobic dechlorination and aerobic degradation have not been shown to occur in sequence within the same natural system. One experiment on a PCB-contaminated soil has been attempted that clearly showed the potential of *in situ* methods to remove PCBs using aerobic organisms, although the degradation efficiency was higher under laboratory conditions [GE Report, 1988]. Unfortunately, an *in situ* method using both anaerobic and aerobic processes has not been attempted yet. However, illustration of the sequential anaerobic-aerobic degradation of PCBs in batch reactors using river sediments is well underway and might provide evidence for the potential of *in situ* biodegradation. Determining whether this technique is likely to succeed depends on results from bench-scale experiments that simulate river conditions.

Because of its past history of PCB contamination, the Hudson River sediments represent an ideal site for testing the feasibility of this *in situ* bioremediation technique. Using both anaerobic and aerobic processes, the objectives of this research are: (1) To model the Hudson River bottom sediment environment in order to determine whether PCBs can be degraded under sequential anaerobic-aerobic conditions using substrate addition to enhance that degradation. (2) To examine the design parameters that influence the degradation process and to relate those parameters to real *in situ* conditions.

RESULTS AND DISCUSSION

The river model consists of a closed glass reactor (1.8 x 0.6 x 0.6 m) (Figure 8-1), filled with 0.8 tons of a lightly PCB-contaminated Hudson River sediment (<5 ppm of mono-, di-, and trichlorobiphenyl congeners). Additional Aroclor 1242 was added and mixed into the sediment to a final concentration of ~ 300 ppm on a wet basis. Anaerobic conditions were created in the sediment by adding methanol in two increments of 6.67 mg/L/week, along with a mineral media, through eight injection wells connected to a multichannel pump. In order to accurately model the flow in the river bed, a flowing stream of water on top of the sediment was permanently recirculated. The flow rate (0.15 L/second) was low enough so that no sediment refluidization occurred. The liquid was circulated for the duration of the experiment, which to date has been 8 weeks. The reactor was kept at room temperature.

In order to monitor dechlorination, eight samples of sediments were collected every 4 weeks using a special coring device through eight top access ports and analyzed. Samples were taken from the bottom, middle, and top of the sediment. Batch incubations were prepared in parallel by adding 15 g of the same sediment (after Aroclor 1242 was added) to 120-mL serum bottles. Methanol was added in one increment to all bottles except three, along with the anaerobic media. Triplicate bottles were kept at 12°C, 25°C and 30°C, respectively. Control bottles were prepared identically and autoclaved twice at 24-hour intervals. All bottles were sealed with Teflon-coated stoppers and aluminum crimp caps. Gas production by the batch incubations was measured by inserting the needle of a wetted, glass-barreled syringe through the stopper of the serum bottle and measuring gas volume. Sample extraction, PCB analysis, and Aroclor peak identification are described elsewhere [Nies and Vogel, 1990].

In order to study the transport and distribution of the substrate into the sediment, a study using bromide as a conservative tracer was also performed on a smaller, 5-gallon model reactor. Water with bromide tracer at 600 ppm was injected into the reactor through a central injection well and the bromide monitored as it moved with the flow past multilevel sampling wells located at varying distances from the injection well. The pumping rate was such that the reactor fluid volume was replaced every 6 hours. The bromide concentration was measured by ion chromatography (IC). The distribution of the bromide tracer into the smaller reactor varied with time and distance from the injection port. Early after pumping began, bromide was found in locations close to the injection well. Total breakthrough was quickly achieved in the bottom of the reactor, whereas it was only after 24 hours of pumping that bromide reached near maximal concentration in the top sand.

Methanol addition to the river model quickly initiated anaerobic conditions within the reactor, as evidenced by the intense methane bubbling at the top of the flowing water. However, considerably less gas was observed whenever the methanol was apparently depleted. After 4 weeks of operation, a shift of the congener pattern from the highly chlorinated congeners to the less chlorinated PCBs was already observed, indicating that a dechlorination process had started. The extent of dechlorination is the removal of chlorine relative to the control. Table 8-1 gives the homolog distribution for Aroclor 1242 at different levels in the

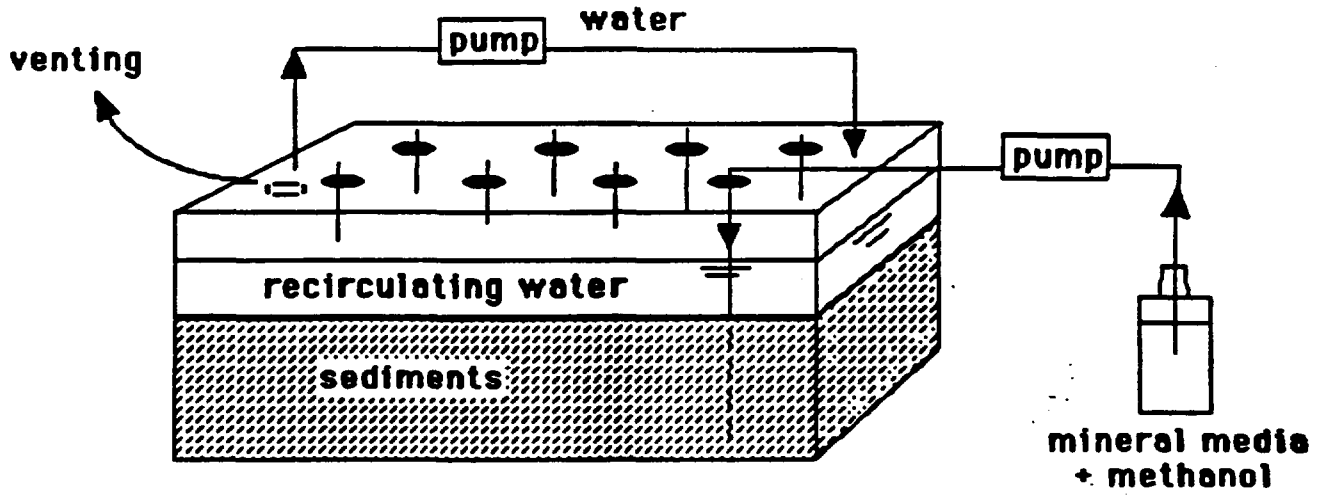

Figure 8-1. Schematic description of the Hudson River model.

TABLE 8-1. Homolog distribution (mol%) and percent change for Aroclor 1242 in the Hudson River model sediment.

Homolog	Week 0[a]	Top Week 4	Top Week 8	Middle Week 4	Middle Week 8	Bottom Week 4	Bottom Week 8
Mono-	2.55	2.94 (+15.1%)	4.13 (+61.6%)	6.94 (+171%)	2.76 (+8.0%)	4.02 (+57.4%)	3.52 (+38.1%)
Di	19.50	23.06 (+18.2%)	24.28 (+24.4%)	28.38 (+45.4%)	24.80 (+27.1%)	24.96 (+27.9%)	27.31 (+40.0%)
Tri-	34.31	33.14 (-3.4%)	31.27 (-8.8%)	31.20 (-9.0%)	33.34 (-2.8%)	32.30 (-5.8%)	32.25 (-6.0%)
Tetra-	35.08	32.54 (-7.2%)	31.78 (-9.4%)	26.97 (-23.0%)	30.84 (-12.0%)	30.74 (-12.3%)	29.37 (-16.2%)
Penta	7.57	7.32 (-3.2%)	7.57 (+0.07%)	5.79 (-24.4%)	7.34 (-3.0%)	6.97 (-7.8%)	6.69 (-11.6%)
Hexa-	0.96	0.97 (+1.35%)	0.94 (-1.96%)	0.68 (-28.6%)	0.90 (-6.5%)	0.98 (+2.4%)	0.80 (-13.5%)

[a] The homolog distribution in controls without methanol addition remained unchanged after 8 weeks.

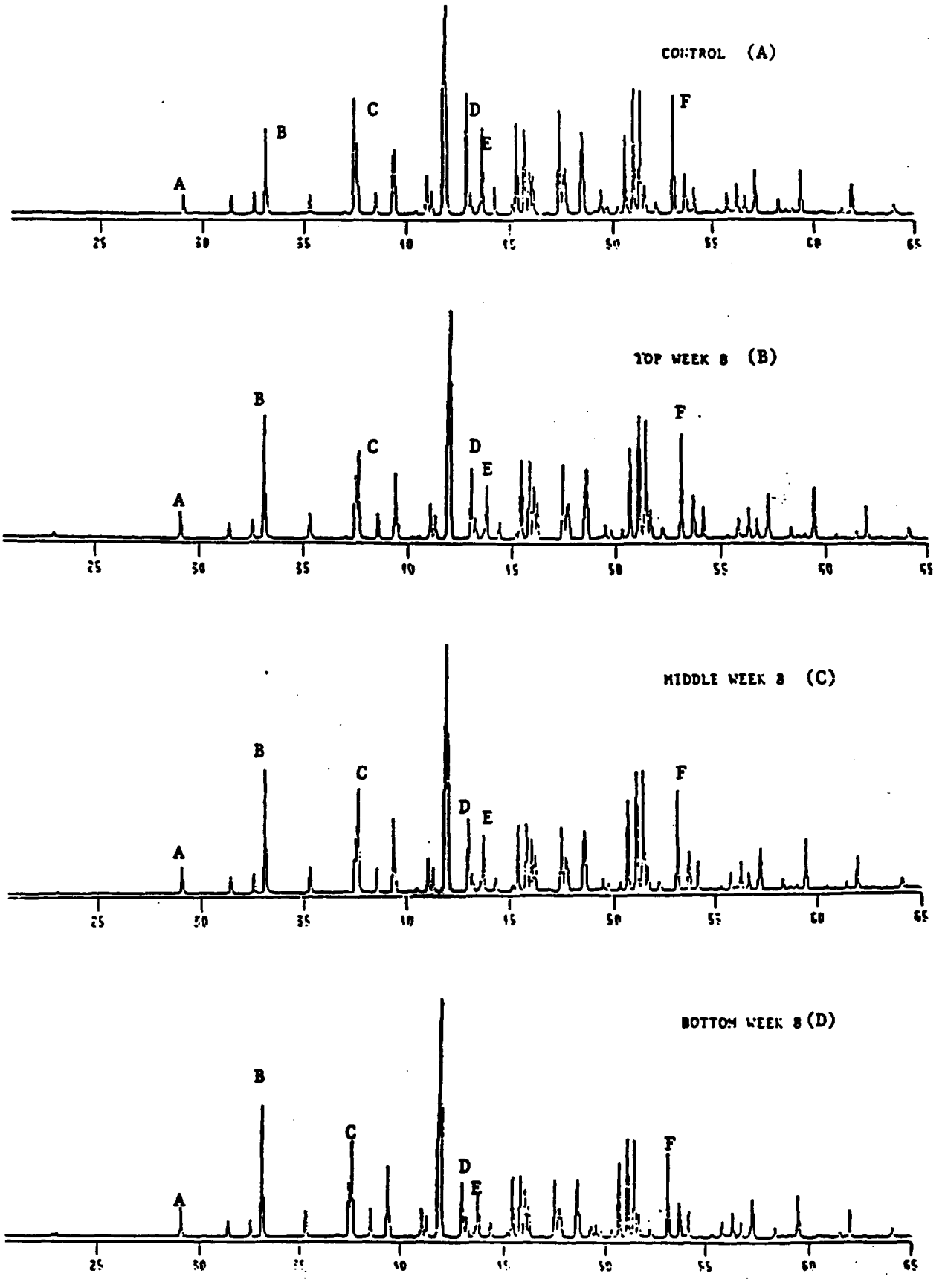

Figure 8-2. Capillary gas chromatograms showing the anaerobic dechlorination of Aroclor 1242 after 8 weeks of operation in the Hudson River model sediments. A) control; B) surface of sediment; C) middle of sediment; D) bottom of sediment.

reactor after 4 and 8 weeks of operation. An increase in the mono- and dichlorobiphenyls was observed relative to levels of week zero. Those levels were unchanged in autoclaved controls. In addition, a decrease in tri-, tetra-, penta- and hexachlorobiphenyl relative to levels in controls was also observed. In general, dechlorination was highest in the middle and bottom of the reactor. The individual congeners that underwent dechlorination are shown in Figure 8-2. In general, congeners 2,3-3,4 and 2,3,4-4-CB (peak F), 2,3-4-CB (peak E), 2,3-3- and 2,3,4-CB (peak D), 4-4- and 2,5-2-CB (peak C) were first dechlorinated as indicated by a reduction in their peak height relative to the control. Congeners 2-4- and 2,3-CB (peak B) as well as 2-2- and 2,6-CB (peak A), were the major dechlorination products after 8 weeks of operation.

No dechlorination was observed in batches incubated at 12°C, whereas incubations at 30°C and at room temperature were similar to the Hudson River model sediment in the extent and pattern of dechlorination. Gas production was monitored in the batch incubations. In general, the volume of gas measured was lowest in the 12°C incubations and highest in batches kept at room temperature and at 30°C.

SUMMARY AND CONCLUSIONS

So far, anaerobic reductive dechlorination has been demonstrated in laboratory batch experiments. In PCB-contaminated sediments, anaerobic microbial communities seem to mediate the dechlorination reaction. However, in nature dechlorination is a slow process. This study is the first attempt to initiate dechlorination through substrate addition while simulating at the same time *in situ* conditions. Methanol injection was able to initiate anaerobic conditions and dechlorination without using a reducing agent. The pattern of the shift in congener distribution is indicative of active dechlorinating activity taking place spatially and temporally within the reactor. In addition, the similarity of the results between the batch incubations and the river model sediment suggests that it might be possible to translate small-scale work to *in situ* conditions.

FUTURE WORK

The focus will be on the aerobic phase of the study. Once an appreciable amount of dechlorination products have been obtained, the addition of methanol will be stopped and the anaerobic layer will be oxygenated by adding hydrogen peroxide through the injection wells. Eventually, most of the sediment should be aerobic. At this point, the upper aerobic layer of the sediment that contains aerobic organisms adapted to PCB degradation will be pumped into the reactor, introducing PCB degraders into the bulk of the sediment. It is hoped this will mediate the oxidation of the less chlorinated congeners.

Chapter 9

DIFFERENTIATION OF ANAEROBIC MICROBIAL DECHLORINATION PROCESSES

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INTRODUCTION

It has long been recognized that the gas chromatographic patterns of the PCB mixtures recovered from environmental samples often differed from those of the commercially used compositions (e.g., Aroclors) originally released, indicating that changes in PCB congener distribution had occurred. These changes were originally attributed to "weathering" of the environmental PCBs. Several years ago we began to recognize that "weathering" encompassed a wide variety of physico-chemical and (usually) biochemical alteration processes, each of which was associated with a characteristic pattern of selection among the various PCB congeners present in the commercial PCB product originally released. As a result, each environmental alteration process leaves a distinctive distribution of residual PCBs, and hence a characteristic pattern of alteration in the gas chromatogram. Since then, we have been collecting "weathered" PCB samples and chromatograms from all accessible sources, and studying them to determine the nature of the transformation processes that had been underway. These surveys have shown that the environmental processes capable of altering PCB congener distributions included inter-phase partitioning, aerobic microbial biodegradation, and aerobic microsomal metabolism, all of which were known prior to our studies, and anaerobic microbial dechlorination, which was not.

The reductive dechlorination of PCBs that is effected by anaerobic microbial action was originally recognized in the gas chromatograms of aquatic sediments from a variety of sources [GE Reports, 1984-1989; Brown et al., 1984; 1987a; 1987b; 1990], but has more recently been duplicated in several laboratories [GE Reports, 1987-1990; Quensen et al., 1988]. From the very beginning it was recognized that anaerobic microbial reductive dechlorination is not a single process, as if carried out by just one dechlorinase enzyme system in just one strain of bacteria, but instead a group of related processes, each characterized by its own pattern of selection among PCB congeners as substrates, and hence yielding a mixture of residual and partially dechlorinated PCB congeners which exhibits a characteristic gas chromatographic profile. These profiles were given letter designations which reflected either the order of their discovery or their source. Thus far, 11 different GC profiles (B, B′, B*, C, E, F, G, H′, H*, N, and W) have been seen repeatedly in environmental samples, and at least two others (K and L) seem indicated by limited data from the St. Lawrence. Six of these environmental profiles (B, C, E, H, H′, and N) have now been duplicated in laboratory cultures. In addition, the laboratory cultures have often yielded alteration patterns (e.g., H, H′, J, M, N, or Q) which were different from those exhibited by the sediments used as

inocula. The reasons for this appear to be two-fold. First, the particular strain of PCB-dechlorinating microbe that becomes predominant under conditions of laboratory culture may not be the one that was predominant when the bulk of the environmental dechlorination occurred. Second, some of the patterns observed in the environmental samples result from the action of two or more individual dechlorination systems, which may be segregated under the appropriate conditions of laboratory culture. Thus, the environmental Pattern C of upper Hudson River sediments has been found to result from the combined action of the dechlorination systems responsible for Patterns M and Q [GE Report, 1989] and Pattern H′ probably results from the action of System H with a small contribution from B, M, or J, depending on the site.

In order to differentiate the observed dechlorination processes, it appeared important to develop objective criteria for characterizing the congener selection patterns seen. Accordingly, a detailed analysis of the congener selection patterns in the 15 best-documented dechlorination systems was undertaken.

RESULTS AND DISCUSSION

The resulting Table 9-1 shows that the 15 dechlorination systems examined differ in four general characteristics. First, in their relative preferences for attacking chlorines located in the *ortho*, *meta*, and *para* positions on the ring. Systems M and N have a strong preference for removing *meta* chlorines; Q and probably E for removing *para* chlorines; while the remainder attack chlorines in either position with comparable ease. However, only the systems from the Housatonic River drainage (F, G, and certain new cultures described elsewhere in this report) have thus far been found to remove *ortho* chlorines.

Second, the observed systems differ in their preferences for attacking lightly, moderately, or heavily chlorinated PCB congeners. Most of them appear to have mild preferences for attacking the more lightly chlorinated congeners, in line with the mildly inhibitory effects of opposite ring substitution which have been previously detailed [Brown et al., 1987b; 1990]. However, Systems E and F seem to prefer to attack the higher congeners, and G and N show an intermediate behavior. These sort of preferences are most marked for System F, which attacks most heavily chlorinated PCBs both preferentially and indiscriminately, and for System M, which attacks only a few congeners carrying more than four chlorines, but can be very active on the lower congeners.

Third, the systems differ in their overall selectivity pattern in attacking higher PCB congeners, both in terms of substitution pattern on the ring being attacked and the sensitivity of the process to *ortho* substitution on the opposite ring. These patterns have been worked out in detail for Systems B, H, and H′ [Brown et al., 1987b; 1990], but can be summarized in more general terms for the other systems as in Table 9-1.

Table 9-1. Congener selectivity patterns shown by environmental PCB dechlorination systems[‡]

System	Source	Position and Range of Attack	Higher PCB Removal Pattern	Range of Reactivities for PCBs with Indicated Mono- or Dichlorophenyl Group					
				3,4-	2,3-	2,5-	2,4-	4-	3-
B	Upper + mid-Hudson R (NY) + cultures	*m,p*-L	III-4	4	4	2b	1	0?	1?
B′	Upper Hudson R (NY)	*m,p*-L	III-4	4	4	2-3b	2	0?	2
B*	Sheboygan R (WI)	*m,p*-L	III-4	4	4	3b	1	0?	2
C	Upper Hudson R (NY) + cultures	*m,p*-L	III-4	4	4	4	3-4	4	2
E	Upper Hudson R (NY) + cultures	*m,p*-H	II-4	?	?	?	?	?	?
F	Silver Lake (Pittsfield, MA)	*o,m,p*-H	III-4	4	<3	0	0	0	0
G	Silver Lake (Pittsfield, MA)	*o,m,p*-M	III-4	4	4	2-3b	0?	?	2
H	(Several sites[†] + cultures)	*m,p*-L	II-3	3	2	0	0	0	0
H′	(Several sites[†] + cultures)	*m,p*-L	II-3	3	3	0	0	0	0
H*	Sheboygan R + Harbor (WI)	*m,p*-L	II$^+$-3	3	2	0	0	0	0
J	New Bedford (MA) cultures	*m,p*-L?	III-4	4a	3	0	0	0	0
M	Upper Hudson R (NY) cultures	*m*,*p*-LL	I-4	4a	4	4	0	0	2
N	Woods Pond (MA) + cultures (on 1260)	*m*,*p*-M	III-4	?	?	?	0	?	?
Q	Upper Hudson R (NY) cultures	*m,p*-L	III-4	4	4	0?	3-4	4	0?
W	Waukegan Harbor (IL)	*m,p*-L?	II$^+$-3	3-4	3-4	2b	0	0?	2

[‡]KEY. Chlorine positions attacked: *m,p* = *meta, para*; *o,m,p* = *ortho, meta, para*; underline indicates preference. Chlorination range attacked: L, M, H = mild preference for lower, medium, or higher congeners, respectively; LL = strong preference for lower congeners. Pattern of attack on higher congeners: I = 236-XY > 234-XY >> 245-XY, etc. II = 234-XY > 245-XY > 235-XY >> 236-XY; 2345-XY, 2346-XY >> 2356-XY. II$^+$ = ditto except 234-XY $\simeq$ 245-XY. III = 234-XY $\geq$ 245-XY > 236-XY > 235-XY but range of relative reactivities small; 2345-XY, 2346-XY, 2356-XY all reactive. Effect of opposite ring substitution pattern on reactivity of indicated chlorophenyl group (or 245-chlorophenyl group in a higher congener): 0 = indicated group never dechlorinated; 1 = only congeners lacking 2′ substitution attacked; 2 = congeners substituted 2′ but not 2′X′ attacked; 3 = congeners substituted 2′X′ but not 2′6′ attacked; 4 = all congeners carrying indicated group attacked. Modification of usual 2′6′ substitution effects on reactivity: a = 2′6′ substituted congeners attacked at least as easily as less *ortho* substituted types. b = indicated *ortho* substituted type may be more reactive than non-*ortho* substituted species (e.g., 25-3, 25-4 CB).

[†]New Bedford (MA); Pensacola (FL); Bridgeport (CT); Woods Pond (MA); mid-Hudson R. (NY); upper Hudson cultures.

Finally, and most characteristically, the various systems differ in their selectivity patterns for attacking the lower PCB congeners, again in terms of both the substitution pattern on the ring being attacked and the sensitivity to various types of substitution on the opposite ring (Table 9-1, last six columns). Basically, most systems can attack 3,4- or 2,3-dichlorophenyl groups in most of the lower congeners, but encounter increasing limitations with 2,5-, 2,4-, 3-, or 4-chlorophenyl groups. The limitations are manifest mainly in the range of PCB congeners containing such groups which can undergo attack; increasing substitution on the opposite ring, especially *ortho* substitution, limits the range of reactivity.

Incidentally, on Table 9-1 no data is given in the last six columns for System E because so far it has only been observed in environmental samples which also exhibited dominant System B or C dechlorination in the lower congener range, or in early cultures on Aroclor 1260, which contains almost no lower congeners to start with. Data has also been omitted for the action of System N or lower congeners, in this case because we are not absolutely sure whether the system responsible for transforming Aroclor 1260 in culture (and observed in Woods Pond) is identical to that responsible for transforming Aroclor 1242 in parallel cultures examined by Quensen. The latter system is very similar to System M in its lower congener reactivity preferences, but also (unlike M) attacks the higher congeners present (at low levels) in 1242 as well.

SUMMARY AND CONCLUSIONS

In summary, as a result of the differences in range of PCB congeners attacked it is possible to distinguish among, and thereby characterize, at least 13 different anaerobic microbial dechlorination systems (allowing Systems C and H′ as composites) and to recognize that several others may be already hinted at from fragmentary chromatographic data. Thus far, only Patterns H and H′ have appeared in either sediments or cultures from more than one drainage basin. Otherwise, it would appear that each of the more heavily studied drainage basins (e.g., the Hudson, Housatonic, Acushnet, Sheboygan, and Waukegan) has produced its own distinctive assemblage of dechlorination systems in response to the availability of PCBs as terminal electron acceptors for anaerobic microbial metabolism. Whether these assemblages arise from local mutations to produce the requisite dechlorinases, or from natural selection guided by local environmental features, is still unknown.

Chapter 10

GENETIC STUDIES OF BACTERIAL PCB DEGRADATION: 1. BACTERIAL SURVIVAL ON PCB-CONTAINING SOIL 2. ANALYSIS OF *bph* GENES

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INTRODUCTION

The ability to develop a practical process for aerobic PCB biodegradation depends largely upon obtaining organisms with suitable characteristics. Two of the most important attributes are (1) high levels of degradative activity against a wide variety of PCB congeners, and (2) the ability to survive and retain activity on PCB-containing soils long enough for significant degradation to occur. The survivability of naturally occurring and genetically engineered PCB-degrading bacteria on contaminated soil is being examined in the laboratory. In addition, other properties which may influence the utility of these strains are being investigated.

We are also continuing our efforts to develop recombinant bacteria with superior PCB-degrading abilities. Biphenyl dioxygenase (encoded by the *bphA* gene) is the enzyme primarily responsible for PCB degradation. Our approach is to construct a wide-host-range plasmid in which *bphA* activity is controlled from a high expression promoter. This should lead to the increased production of biphenyl dioxygenase and thereby an increase in PCB-degrading activity. In order to accomplish this goal the nucleotide sequence of the *bphA* gene is being determined and the influence of adjoining DNA regions on PCB degradation is being examined.

RESULTS AND DISCUSSION

Survival of Genetically Engineered Bacteria on PCB-Containing Soil

The need to repeatedly add organisms to contaminated soil severely reduces the utility of either *in situ* or reactor-based bioremediation processes. The use of soil isolates in such processes does not guarantee that these organisms will survive on PCB-laden soil. For example, in the field study conducted by Unterman et al., [1988a], a soil isolate (LB400) with good activity against many PCB congeners, exhibited poor soil survivability and therefore had to be repeatedly applied to the site [GE Report, 1988]. Previous studies have demonstrated that the genetically engineered *Escherichia coli* stain FM4560 (containing the LB400 *bph A, B* and *C* genes), degrades PCBs nearly as well as LB400 while displaying characteristics which may be advantageous in a bioremediation process [Mondello, 1989; GE Report, 1988]. Among the potential advantages was greater survivability in laboratory media containing PCBs.

Recently, the survivability of FM4560 and LB400 were compared on PCB-contaminated soil from the Glens Falls dragstrip site. This material contains ~ 550 ppm of highly evaporated Aroclor 1242 and thus is similar in composition to Aroclor 1248. A series of 2 dram vials containing 0.2 g of dragstrip soil were inoculated with 0.1 mL of either FM4560 or LB400 ($\sim 10^8$ cells/vial), sealed, and incubated at 23°C without shaking. Separate vials were sampled at each timepoint by the addition of 2.0 mL of phosphate buffer (50 mM, pH 7.5), and vortex mixing at maximum speed for 2.0 minutes. Samples were serially diluted and spread onto minimal medium plates containing biphenyl, 0.02% succinate and 5 μg/mL tetracycline (for LB400 samples). FM4560 cells were plated onto both MaConkey and Luria agar plates containing 200 μg/mL ampicillin. Colony formation was used to measure cell survival. The presence of active *bph* genes in LB400 was determined by growth on biphenyl, while *bph* gene activity in FM4560 colonies was demonstrated by their ability to produce yellow *meta* cleavage product when sprayed with an ether solution of biphenyl or 2,3-dihydroxybiphenyl.

Similar survival curves were obtained for FM4560 and LB400 (Figure 10-1). Early timepoints show a significant increase in cell numbers resulting from growth on stored intracellular nutrients. After 72 hours the cell numbers returned to their original levels and continued to decrease such that by 8 days ~ 20% of the cells remained viable. Of the initial inoculum, 2% of the cells were found to survive after incubation for 28 days.

The presence of active *bph* genes in the surviving cells was examined at each timepoint. For LB400 the percentage of viable cells unable to grow using biphenyl remained relatively stable for the first 8 days at ~ 2.5% and then increased to 4.2% for the remainder of the experiment. Virtually all (99.8%) of the FM4560 colonies contained active *bph* genes. Those that did not were restreaked onto selective media and tested again. In each instance the cells were found to contain active *bph* genes. An examination of the plasmids in these cells revealed no alterations in their *Eco*RI restriction patterns indicating that no substantial DNA rearrangements or deletions had occurred.

The survival of LB400 for long periods on PCB-contaminated soil was unexpected as previous investigators have reported that LB400 cells rapidly lost viability upon exposure to soil from the dragstrip site. The different results obtained in the previous experiment may be due to the use of a different experimental protocol. In those experiments bacterial cultures were added to the soil and allowed to air dry. The more recent studies were conducted in sealed vials where the moisture level was constant. Experiments are in progress to determine the effect of drying on cell viability. Somewhat less surprising was FM4560's ability to survive on PCB-containing soil. Several studies had demonstrated long-term survivability of recombinant *E. coli* strains on soils; however, none of this work involved PCB-contaminated material [Devanas and Stotsky, 1986; Trevors, 1987].

Instability of LB400 *bph* Genes

In 1989, we reported that cultures of LB400 grown without biphenyl contained an unusually large number of cells which had lost the ability to degrade PCBs [GE Report,

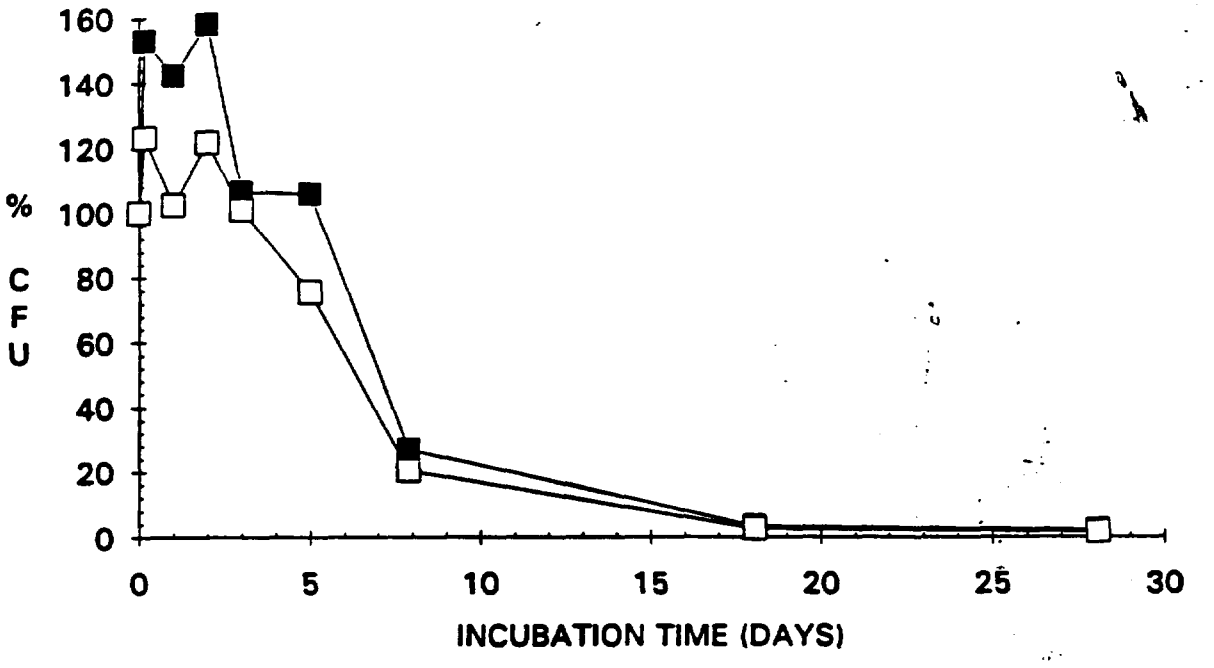

Figure 10-1. Survival of FM4560 and LB400 on PCB-contaminated soil. FM4560, Closed Boxes; LB400, Open Boxes.

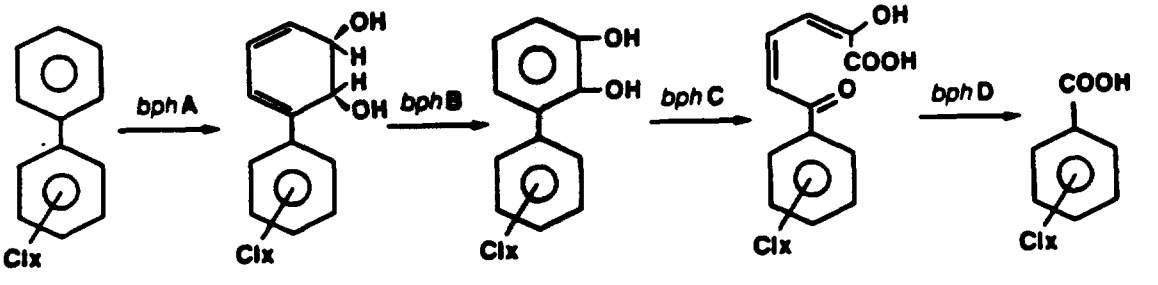

Figure 10-2. Degradation of biphenyl and chlorobiphenyls by the 2,3-dioxygenase pathway in *Pseudomonas* strain LB400. Gene designations: *bphA*, biphenyl 2,3-dioxygenase; *bphB*, dihydrodiol dehydrogenase; *bphC*, 2,3-dihydroxybiphenyl dioxygenase; *bphD*, 2-hydroxy-6-oxo-6-phenylhexa-2,4-dienoic acid (*meta*-cleavage compound product) hydrase.

1989]. Analysis of these spontaneous mutants revealed that many had lost all of the enzymatic activities encoded by *bph A, B, C,* and *D* (Figure 10-2). Southern hybridization experiments revealed that these strains had undergone a deletion of their *bph* structural genes. The potentially serious effects of this phenomenon on LB400's utility in a PCB bioremediation process has led us to investigate this phenomenon in more detail.

1. Bph Mutation Frequency

Biphenyl grown colonies of LB400 were inoculated into *Pseudomonas* minimal medium (PMM) containing 0.5% sodium succinate as the carbon source. The cells were maintained in logarithmic growth phase for 100 generations by serial passage into fresh media. The ratio of Bph^-/Bph^+ cells was determined by colony counts on minimal medium containing biphenyl and 0.05% succinate. The results of six independent experiments are shown in Table 10-1. The percentage of mutant strains in the cultures ranged from 0.6 to 3.2 and with an average of 1.5%. Of the 64 mutant strains isolated, 62 (96.9%), were stable (reversion frequency $<1/10^{10}$). Southern hybridization analysis of of these mutant strains confirmed that they had lost the *bph* structural genes.

TABLE 10-1. Spontaneous mutagenesis of the LB400 *bph* genes

Experiment	# of Colonies	# Bph^-	# Stable[a]	% Loss[b]
1	452	9	8	1.8
2	310	10	10	3.2
3	591	6	6	1.0
4	1067	6	6	0.6
5	1515	22	21	1.4
6	1287	11	11	0.9

Average = 1.5%

[a] these mutants reverted to the Bph^+ phenotype at a frequency of less than 1 in 10^{10}

[b] % loss determined by number of stable mutants/number of colonies examined

2. Deletion Specificity in the LB400 Genome

Wild-type cultures of LB400 can be grown on minimal salts medium and succinate without the addition of growth factors such as amino acids, nucleotides, or vitamins. A mutation in any of the >100 genes involved in the biosynthetic pathways for these compounds will result in an auxotrophic strain unable to grow in a minimal medium. To determine whether

the high spontaneous mutation frequency is specific for the *bph* genes, the number of auxotrophic mutants appearing after growth of LB400 on a complete medium was examined. A culture of LB400 was maintained in logarithmic growth phase on Luria-Bertani medium for 100 generations by serial passage into fresh medium. The percentage of auxotrophic mutants in this population was determined by replica plating colonies of these cells grown on Luria agar plates onto minimal agar plates supplemented with 0.2 % succinate. Of the 987 colonies tested, none were found to be auxotrophic, indicating that the deletion phenomenon does not occur throughout the LB400 genome and may be *bph* specific.

3. Growth Rate Comparison

If the Bph^- mutant strains grow faster than the wild-type they will eventually dominate the culture and PCB degrading activity will be lost. In order to determine whether the Bph^- strains had a selective advantage in growth rate over the Bph^+ cells we compared the growth curves of the two organisms in PMM + 0.5 % succinate. As shown in Figure 10-3, both LB400 (Bph^+) and LS2 (Bph^- mutant of LB400) have nearly identical growth curves and therefore the ratio of Bph^-/Bph^+ cells in a mixed culture will tend to remain constant.

4. *Bph* Gene Stability in Recombinant Bacteria

Escherichia coli strain FM4560 contains the recombinant plasmid pGEM456 (ampicillin resistance [Ap^r], *bphA, B,* and *C*) and has PCB-degrading ability similar to that of LB400 [Mondello, 1989]. In order to evaluate the ability of this strain to stably retain its PCB-degrading ability, FM4560 was maintained in logarithmic growth phase for 72 hours with and without antibiotic selection in Luria-Bertani broth. The presence of active *bph* genes was determined by the ability to accumulate *meta* cleavage product when treated with either solutions of biphenyl or 2,3-dihydroxybiphenyl. When grown in the presence of ampicillin (to select for plasmid-containing cells), all colonies tested throughout the experiment contained active *bph* genes. In the absence of antibiotic selection, an average of 8.3% of the cells had lost both antibiotic resistance and *bph* activity by 21 hours (Figure 10-4). The loss of Bph^+/Ap^r cells in the population decreased exponentially and by 72 hours was reduced to an average of 31.7%. The loss of *bph* gene activity was always accompanied by the loss of antibiotic resistance suggesting that the entire pGEM456 plasmid had been lost from these cells. An examination of 24 Bph^-/Ap^s strains for plasmid content confirmed the absence of plasmid DNA in these cells. These data indicate that the pGEM456 plasmid is lost or maintained as a single unit and is not subject to the deletion phenomenon associated with the *bph* genes in LB400. Similar results were also observed in the soil survival experiment described above. The loss of plasmid DNA from cells growing in the absence of antibiotic selection is common and results from the faster growth rate of plasmid-free cells. These data suggest that in the recombinant strain, PCB-degrading activity can be stably maintained through simple antibiotic selection.

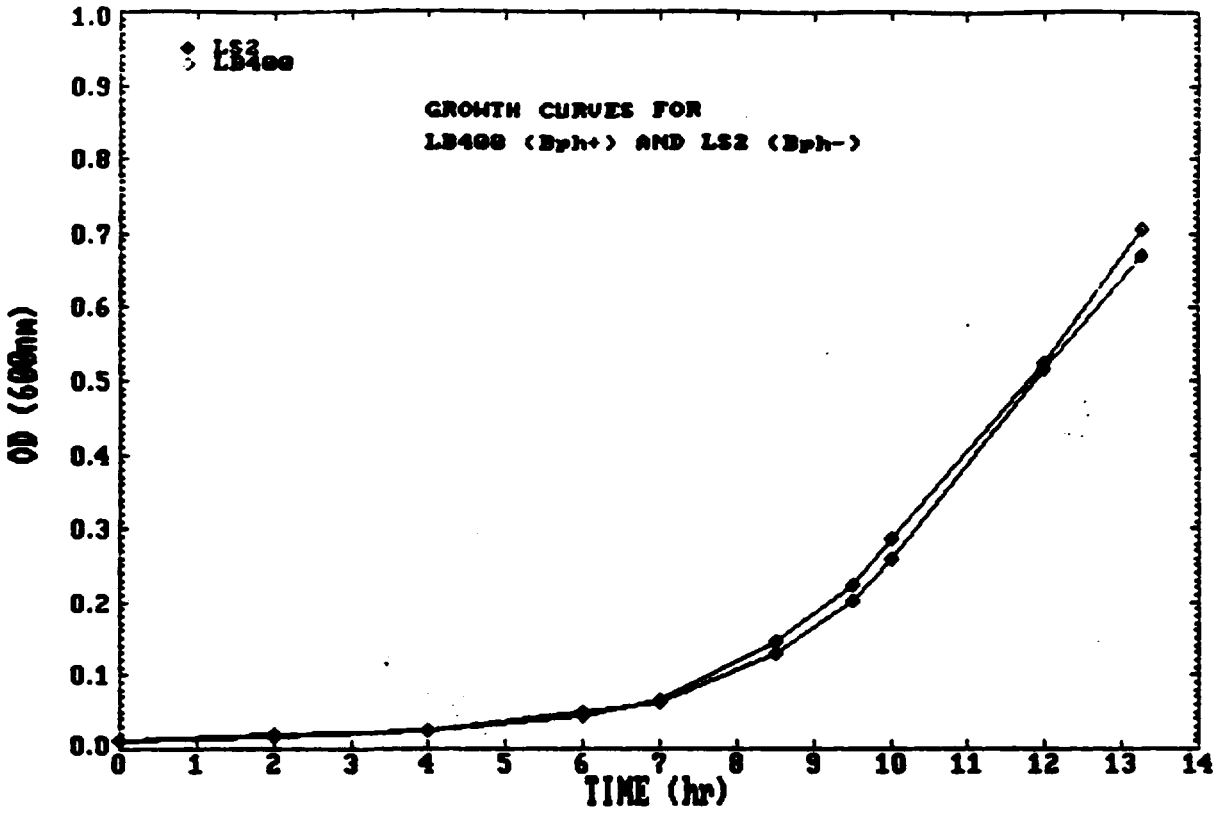

Figure 10-3. Comparison of the growth curves for strains LB400 and LS2.

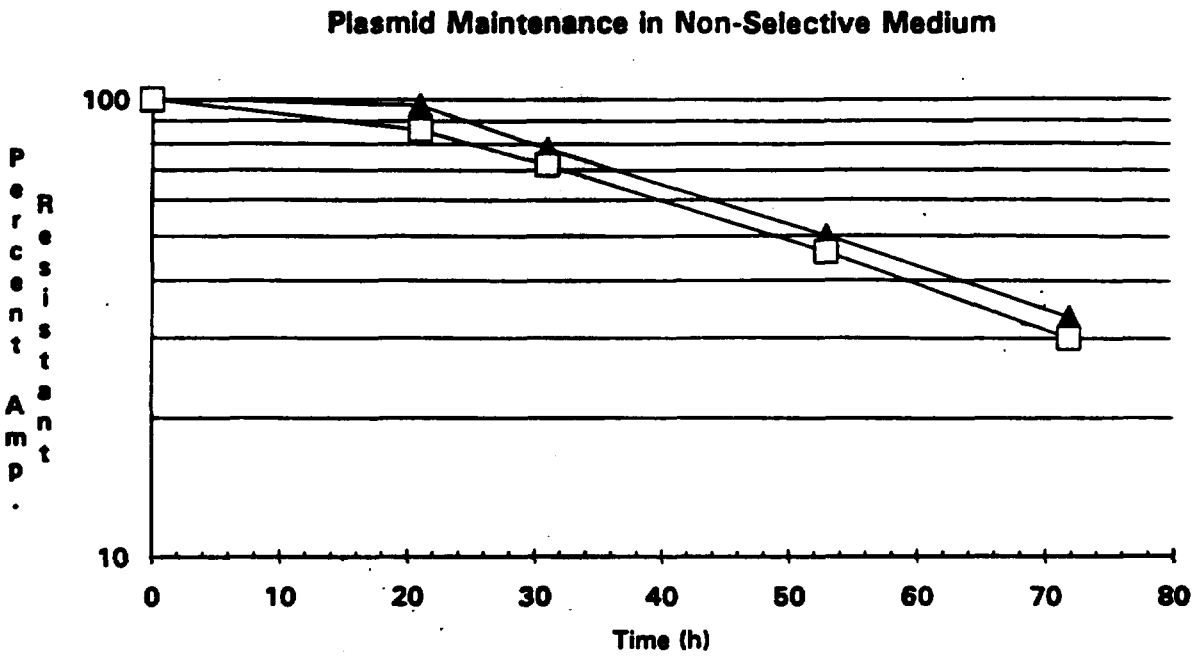

Figure 10-4. Plasmid stability in *Escherichia coli* FM4560 grown in nonselective medium. Data represent results of two independent experiments.

Analysis of the LB400 *bphA* Gene

1. Sequencing the *bphA* gene

To date, ~ 50% of the LB400 *bphA* gene has been sequenced. The procedure involved the use of Exonuclease III and S1 nuclease to construct a series of nested deletion clones from a plasmid containing the 2.9 kilobase *Eco*RI fragment from pGEM456. Sequencing was performed using T7 DNA polymerase and dideoxy nucleotide termination reactions with [^{35}S]dATP as the label. The reactions were analyzed on 6% polyacrilamide wedge gels containing 8 M urea. Two contiguous sections of DNA sequence have been obtained and work is currently underway to fill the gap between the two segments.

2. Evidence for Multiple Bph Promoters

Several lines of evidence suggest that the LB400 *bph* genes are not arranged as an operon. Previously reported transposon mutagenesis studies have found none of the polar effects commonly observed in operons (i.e., the inactivation of downstream genes). The absence of these effects usually indicates that the downstream genes contain their own promoter regions. The existence of multiple Bph promoters is also suggested by recent cloning experiments in which a 3.7 kilobase DNA fragment (encoding the *bphA* gene) and a 2.0 kb fragment (containing *bphB* and *C*) were inserted into pUC-13. In both cases, the *bph* genes were expressed regardless of fragment orientation, consistent with the presence of a promoter on each fragment.

3. Wide-Host-Range Plasmids Encoding the *bphA* Gene

The cloning of the *bphA* gene onto broad-host-range plasmids allows this gene to be transferred to a variety of bacterial strains from diverse environments. Once this is accomplished the strain-specific expression of *bphA* can be studied. A 3.7 kb DNA fragment encoding the LB400 *bphA* gene was inserted into cloning vectors pMMB66EH and pKT240. These plasmids are capable of being transferred to and maintained in many different genera of bacteria. The recombinant plasmids (designated pGEM438 and pGEM240 for the pMMB66EH and pKT240 derivatives, respectively) have thus far been mobilized into strains of *E. coli*, *Alcaligenes eutrophus*, and *Pseudomonas*. In all cases, PCB-degrading activity in these strains (as measured by resting-cell assays on PCB mixes 1B and 2B), were much lower than that of FM4560. Among the possible reasons for this are (1) the lower copy number of the vector plasmids, (2) the absence of a structural or regulatory region of DNA required for high activity, or (3) the accumulation of an inhibitory intermediate from PCB degradation.

SUMMARY AND CONCLUSIONS

Both the naturally occurring and recombinant PCB-degrading bacteria showed better than expected survivability on PCB-containing soil, and were easily detectable even after 28 days. This may mean that given the proper conditions, multiple additions of cells to the soil

would not be necessary. Instability of the *bph* genes in LB400 has been further characterized and found to occur in greater than 1.5% of the population. The phenomenon appears to be *bph* specific and does not produce cells with a growth rate advantage over the wild-type strain. The *bph* genes in the recombinant strain FM4560 were found to be stable if antibiotic selection for the plasmid was maintained.

Evidence for at least two separate promoters in the *bph* region suggest that these genes are not organized as an operon. This may influence our strategy for constructing bacteria with enhanced PCB-degrading ability.

FUTURE PLANS

Once the complete *bphA* gene sequence is available, a computer analysis will be performed to determine nucleotide and amino acid homology with sequences contained in the Genbank database. In addition, transcriptional analysis of the *bph* region will be performed to identify and confirm the existence of potential regulatory sites (i.e., promoters, ribosome binding sites and terminators, etc.). This information will be used to construct a recombinant plasmid in which the *tac* promoter is used to control the expression of *bphA*.

Laboratory studies to evaluate the ability of genetically engineered bacteria to degrade PCBs on soil have been initiated. These investigations will help determine whether currently available recombinant strains have significant advantages over LB400 for PCB degradation.

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Chapter 11

PCB BIODEGRADATION AND NITRATE REDUCTION

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INTRODUCTION

It has become increasingly evident that future bioremediation processes for PCBs will employ consortia of microorganisms, either naturally constituted, reconstituted, or genetically engineered. Such cooperative interactions may prove especially useful for contaminated sites which contain pools of PCBs under aqueous deposition. Towards this end, we have initiated a program designed to obtain assemblages of bacteria which may facultatively utilize nitrate as well as oxygen as electron acceptors for PCB dissimilation. Thus, enrichment cultures were made from municipal sewage and PCB-contaminated sites, including Hudson River sediments. Samples from these sources were inoculated into mineral salts media amended either with biphenyl, hexachlorobenzene, Aroclor 1242, or mixtures, and were incubated in air or with nitrate under reduced oxygen tensions. At this point, we have isolated a number of organisms which are capable of biphenyl or selected Aroclor congener metabolism, and which also demonstrate nitrate reduction. Current endeavors are directed toward the linkage of these two major metabolic functions.

RESULTS AND DISCUSSION

A compilation of results which relate to the salient gross physiological properties of these microorganisms thus far obtained is encompassed in Table 11-1. As can be seen from these data, a number of consortia and single isolates would appear to utilize biphenyl or PCBs and concomitantly reduce nitrate or produce gas. While it is generally recognized that oxygen at μMolar concentrations represses the synthesis and activity of nitrate and nitrite reductase [Coyne and Tiedje, 1990], there has been at least one report [Krul, 1976] of a constitutive nitrate reductase in denitrifying *Alcaligenes* sp. From among this array of isolates, we have chosen initially to investigate two consortia, "TS-1" and "95-SW", in more detail.

Properties of Consortium "TS-1"

The identity of the individual microbial components present within TS-1 (Table 11-1) are as follows: *Citrobacter freundii, Pseudomonas fluorenscens, Pseudomonas putida,* and *Comanonas testosteroni.* Other reports [Pettigrew et al., 1990] and earlier references therein have also indicated that the latter organism possesses aromatic degradative capabilities. We will concentrate our efforts on the biochemistry of this isolate.

TABLE 11-1. Qualitative screen for nitrate reduction in the presence of biphenyl or Aroclor 1242

Microbial Isolate	Nitrate[3] Reduction Plates (3 days)	Nitrate[3] Reduction Tubes[1] (stab) (3 days)	Biphenyl[1] NO_3 Tubes	Aroclor1242-NO_3[2] NO_3 tubes
Hexachlorobenzene isolate	+	gas/-	+	-
P. putida isolate	+	gas/±	+	-
Hudson R. isolate #1	+	++	++(strong)	+
P. fluorescens isolate	+	gas/-	+	-
TS-1 consortium	+	gas/-	++	+
Comonoas testosteroni	+	++(strong)++	-	
Sludge aroclor consortium	+	gas/-	(gas)/++	++
Isolate E-4y	-	-		
Hudson R. isolate #5			+	+
Hudson R. isolate #16		+	+	+
Sludge isolate #15			+	++
Gull Creek isolate #2			±	-
Hudson R. isolate #17[4]			+	-
Hudson R. isolate #18		+	-	
Sludge isolate #16			+	+++(very strong)
Sludge isolate #2			+	-

[1] PAS salts with 0.5% biphenyl, 4 mM KNO_3

[2] PAS salts with 200 ppm Aroclor 1242, 4 mM KNO_3

[3] Yeast extract-peptone medium

[4] Intense yellow-orange colony

Comamonas testosteroni reached cell densities between 10^7 to 10^8 per mL when grown on biphenyl, Aroclor, or both after 24 hours (Figure 11-1). Even in the absence of yeast extract, the culture demonstrated comparable cell densities when grown aerobically on a mixture of Aroclor and biphenyl (data not shown). Capillary GC analyses of *C. testosteroni* activity on Aroclor 1242 (Figures 11-2A and 11-2B) indicate changes in concentration of several congeners (indicated by arrows). Five dichlorobiphenyls (25-, 24-, 2-3-, 2-4-, and 23-CB), three trichlorobiphenyls (25-3, 35-2-, and 23-4-CB), and one tetrachlorobiphenyl congener (24-26-CB) underwent degradation in this experiment.

Growth of *C. testosteroni* in the presence of 2-4- and 4-4-CB indicated that the metabolic transformations of these compounds during 48 hours of growth resulted in the formation and accumulation of chlorobenzoic acids (Figure 11-3). A recent report by Pettigrew et al. [1990], who investigated an isolate identified as *Comamonas testosteroni* (LPS10A), also determined that these compounds are an endpoint of PCB metabolism.

The aerobic metabolism of biphenyl carried out by this microorganism, as established by GC-MS of intermediary metabolites, provided proof that the earlier proposed *meta* cleavage pathway [Smith and Ratledge, 1989] is operational (Figure 11-4). The following intermediates were unequivocally identified (data not shown): 2-hydroxy-6-oxo-6-phenylhexa-2,4-dienoate, 2-oxo-penta-4-enoate, γ-benzoic acid, γ-benzoyl-butyraldehyde, and γ-benzoyl-butyric acid.

Consortium 95-SW

This consortium is a mixture of two microorganisms isolated from aquatic sediments, tentatively identified as a *Pseudomonas* sp. and a *Bacillus* sp. (Table 11-2). The individually resolved isolates do not show nitrate reductase activity with biphenyl. Using resting-cell assays [Bedard et al., 1986] in screw cap vials, complete metabolism of biphenyl was observed after 6 hours of incubation. Assays with individual congeners demonstrated significant degradation (95% loss of 2-4-CB and 20% loss of 4-4-CB in 48 hours; 50% loss of 23-234-CB in 96 hours). Preliminary experiments with Aroclor 1242 indicate a decrease in several congeners, especially 24- and 25-CB and 245-CB (data not shown).

TABLE 11-2. Nitrate reductions by 95-SW consortium

Microorganism	Nitrate medium, Plates, [Beef Ext. & Peptone]	Nitrate medium, Slants [Biphenyl]
95-S *Pseudomonas*	(-)	(-)
95-W *Bacillus*	(+)	(-)
95-S & W Mix of both	(+)	(+)

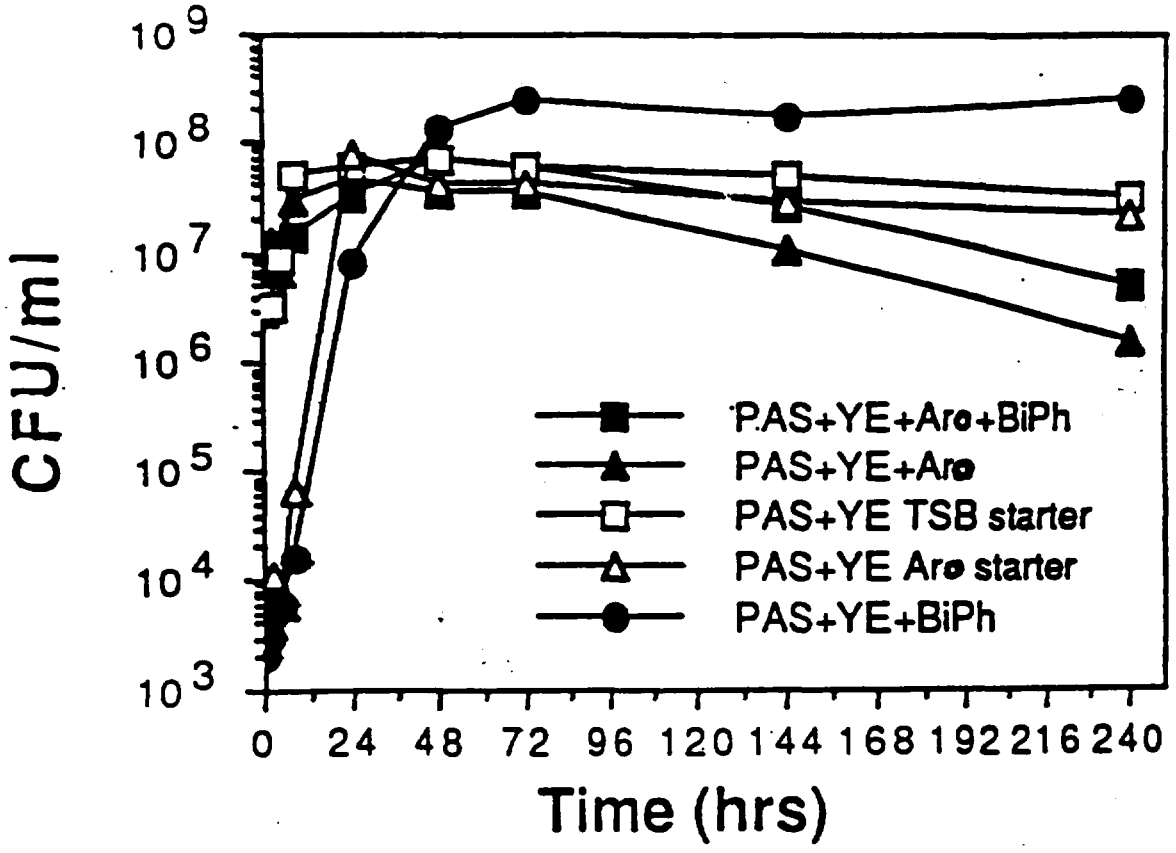

Figure 11-1. *C. testosteroni* **grown on PAS and yeast extract. (BiPh = biphenyl, Aro = Aroclor 1242)**

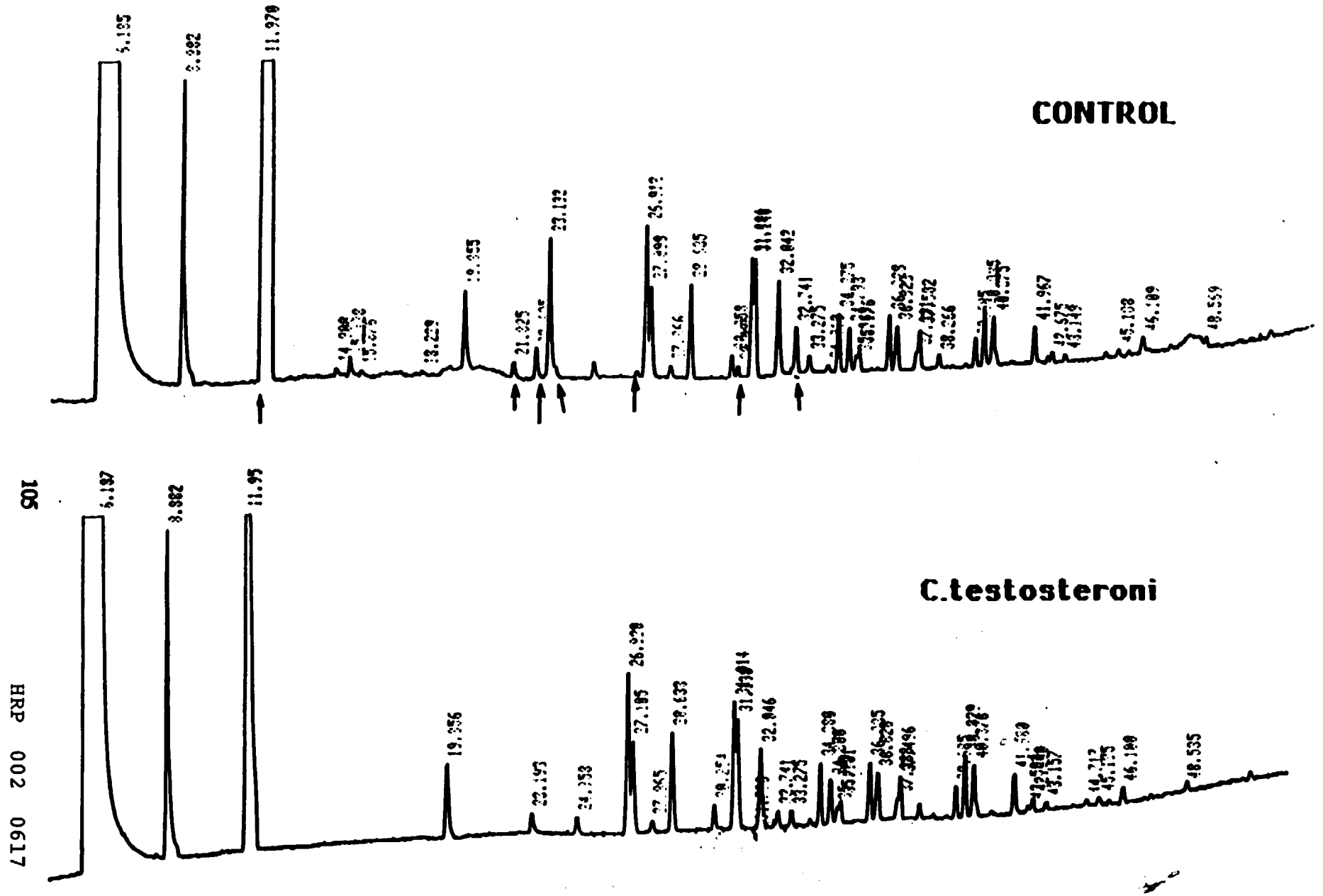

Figure 11-2. Capillary GC analyses of 1242 after incubation with *C. testosteroni*. Upper curve indicates control. Lower curve indicates post-metabolism. Arrows indicate individual congeners which appear to have been transformed.

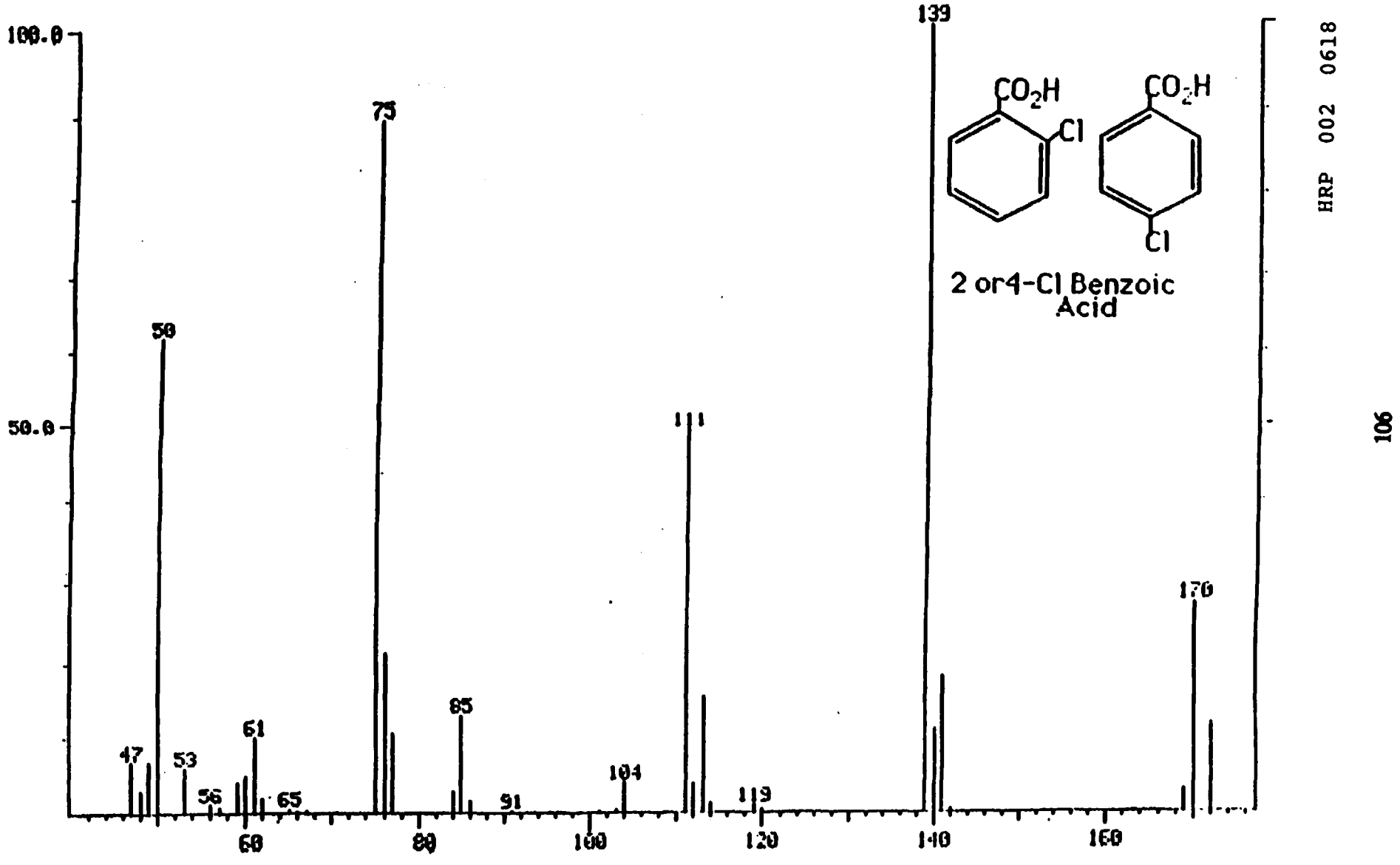

Figure 11-3. Mass spectra of the metabolic products of *C. testosteroni* grown in the presence of 2-4- and 4-4-CB.

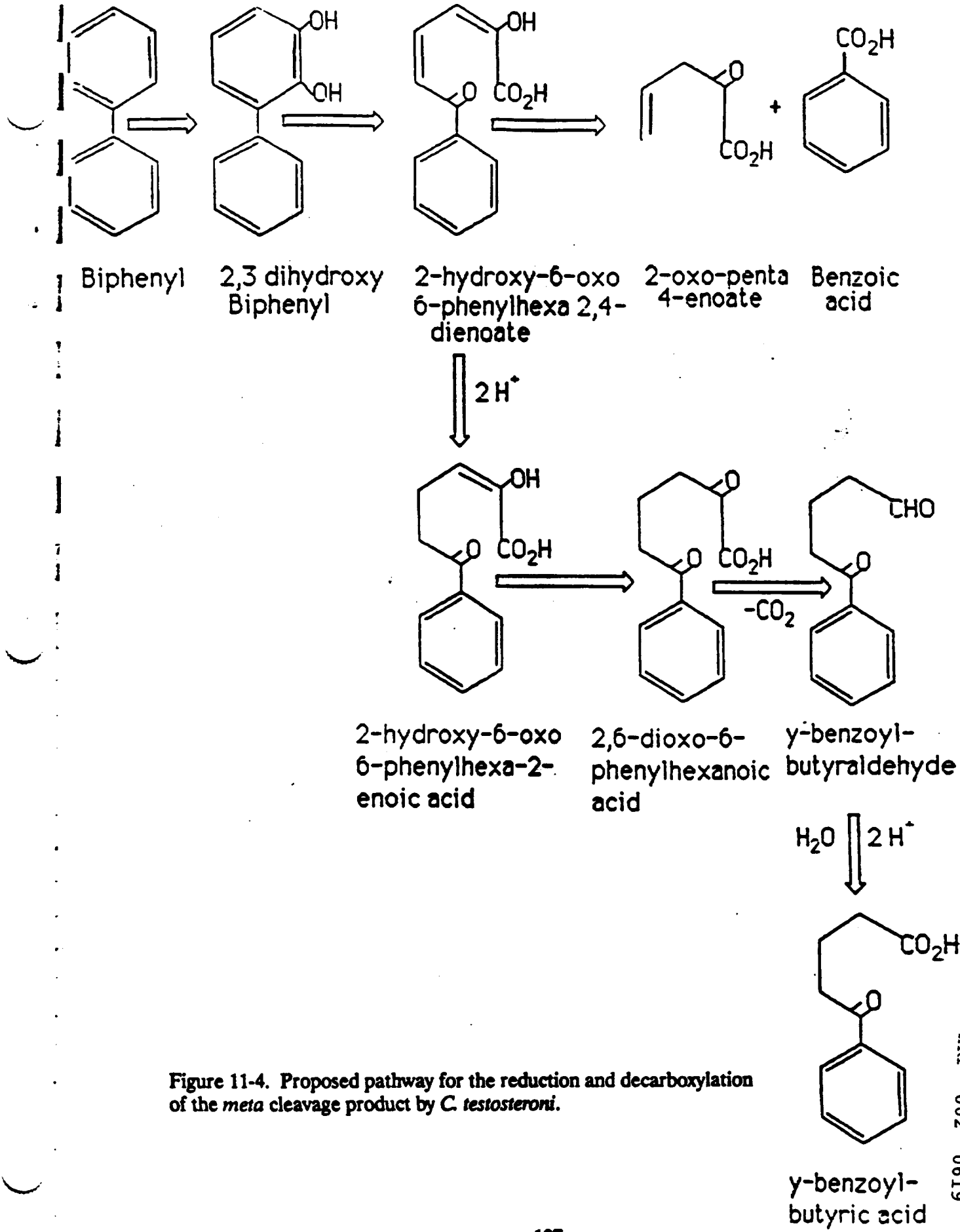

Figure 11-4. Proposed pathway for the reduction and decarboxylation of the *meta* cleavage product by *C. testosteroni*.

SUMMARY AND CONCLUSIONS

Enrichment cultures were made from municipal sewage as well as from a number of PCB-contaminated sites, including Hudson River sediments. Samples from these sources were inoculated into mineral salts amended either with biphenyl, Aroclor 1242, or both, and were grown in air or with nitrate as electron acceptor. Pure isolates include a *Comamonas testosteroni*, a Pseudomonad, and a *Bacillus* sp. Cell densities of the former approaching 3 x 10^8/mL can be achieved when this organism is grown on a biphenyl-Aroclor mixture. High performance liquid chromatography (HPLC) analysis following extraction and workup of growth supernatants from either substrate revealed at least five major metabolites. From biphenyl, further GC/MS analysis identified these compounds: 2-oxo-penta-4-enoic acid, 2-hydroxy-6-oxo-6-phenylhexa-2-enoic acid, γ-benzoylbutyric acid, γ-benzoylbutyraldehyde, and benzoic acid. The most likely biochemical explanation for the appearance of these intermediates involves the initial activity of oxygenase and *meta* cleavage enzymes, followed by a series of oxido-reduction states. *C. testosteroni* also accumulates a melange of yellow-colored compounds when grown on Aroclor 1242. The exact nature of these remains to be established.

Several of the above enrichments reduce nitrate in the presence of biphenyl and/or Aroclor 1242 when grown anaerobically. Such putative anaerobic dissimilatory routes must initially involve different activation and ring-cleavage steps. However, in facultative strains, these intermediates may be convergent with later steps found in the aerobic metabolic route.

FUTURE PLANS

Our proposed objectives for the coming year are:

1. To continue to probe the enzymology of catabolizing enzymes involved in ring fission of chlorinated biphenyls and to link these studies with possible biological dechlorination.
2. To continue to resolve and study the physiology of biphenyl, haloaromatic, and Aroclor-transforming bacterial isolates with special regard to those which possess N-oxide-reducing activities.
3. To select from among the former strains or combinations thereof, those which are either constitutive for nitrate reductase or which can express such enzyme activity in the presence of reasonably low oxygen tensions.
4. To attempt to link Aroclor biotransformation under anoxic conditions with nitrate reduction by introducing such reconstituted assemblages into a laboratory aquatic setup. For such studies, the experimental procedures developed by Kuhn et al. [1988] will be followed.

Chapter 12

AVAILABILITY OF PCBs IN SOILS AND SEDIMENTS TO SURFACTANT EXTRACTION AND AEROBIC BIODEGRADATION

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INTRODUCTION

Surfactant extraction and aerobic biodegradation of aged PCB contamination in soils and sediments can be viewed analogously. In both processes the hydrophobic PCB molecules must move from the soil/sediment matrix and transfer into the aqueous phase before uptake can occur. In surfactant systems this uptake occurs by adsorption into surfactant micelles. In biological systems it occurs by partitioning into microorganisms [GE Report, 1988]. For either process, phases which exist in the soil/sediment matrix and compete for the PCB molecules can substantially hinder the rate and extent of transport observed.

This chapter presents research examining the effect of two such phases, naturally occurring organic matter (NOM) and oil, on surfactant extraction and aerobic biodegradation of PCBs in soils and sediments. A soil pretreatment is presented which enhances both processes by modifying the NOM present in the soil or sediment matrix.

RESULTS AND DISCUSSION

Surfactant Extraction

Sodium dodecylbenzene sulfonate (SDBS) has been used successfully to extract PCBs from Oakland soil [GE Report, 1989]. This soil is primarily montmorillite clay with little organic carbon content. Montmorillite is considered difficult to extract due to its high internal surface area. The soil is contaminated with Aroclor 1260 to a level of 3500 mg/kg, a high PCB loading. Despite these facts, better than 95% extraction of PCBs was achieved in less than 45 minutes with a 1% aqueous solution of SDBS at 80°C. These extractions were run at a 4:1 solution to soil ratio with agitation. Solution PCB loadings approached 1000 mg/L.

Glens Falls dragstrip soil has been characterized as a Windsor Loamy Fine sand. The soil is predominantly a fine sand with some silt, clay, and organic matter present. The organic carbon content is 2-3% by weight as measured by combustion of the organic carbon. The soil is contaminated with partially evaporated Aroclor 1242 at a level of 100 mg/kg. The expectation was that this low level of PCBs would be removed easily from the sandy loam soil by a SDBS treatment comparable to that given the Oakland soil.

This was not the case. Only about 40% of the PCBs present in the dragstrip soil were removed after 2 hours of extraction with the surfactant. The kinetic results are compared with those from Oakland soil in Figure 12-1. The numbers plotted represent the percentage of total PCBs present in the supernatant at any time. These results indicate little difference in the percentage of PCBs removed from the two soils at early timepoints, but clearly demonstrate a difference in final extent of extraction. The absolute amount of PCB solubilized was also different. Final solution PCB loadings in the dragstrip soil extraction only reached 25 mg/L at a 2:1 solution to soil ratio, far below that achieved with the Oakland soil.

Several experiments were run to examine the mechanism causing the poor extraction performance of the dragstrip soil. The data presented in Figure 12-2 show the effects of three levels of surfactant concentration on the kinetics of extraction from this soil. The extractions were run at 80°C, again with a 2:1 solution to soil ratio and agitation. Increasing the SDBS concentration in aqueous solution from 1 to 3% increased the amount of total PCB solubilized from 40% to 85% after 2 hours. The initial rates of extraction were not significantly altered by the increased surfactant concentration.

The results of three consecutive extractions on a single soil sample are given in Table 12-1. One percent surfactant solutions were used with the conditions just described. After each extraction the slurry was centrifuged, the supernatant withdrawn, and fresh surfactant solution added to make up the volume removed. About 75% of the PCBs were removed by the three extractions combined. The ratio of solution PCB concentration to soil PCB concentration stayed relatively constant for all three extractions. The results of these two experiments make it clear that the extraction is not being limited by surfactant loading or by kinetic rate. Instead, the process appears to be equilibrium limited.

TABLE 12-1. Consecutive extractions from dragstrip soil using a 1% SDBS surfactant solution

	Sol'n [PCB]	Soil [PCB]	Soil/Sol'n [PCB]
Start	---	99 mg/kg	---
Extraction 1	20 mg/L	66 mg/kg	3.3
Extraction 2	15 mg/L	40 mg/kg	2.7
Extraction 3	9 mg/L	25 mg/kg	2.8

There is literature [Means et al., 1980; Chiou et al., 1983; Gschwend and Wu, 1985] indicating that nonpolar molecules such as PCBs undergo equilibrium partitioning into NOM when both are present in an aqueous medium. This is much like the distribution of these molecules between the two phases which form when octanol and water are mixed. NOM exists in the form of large macromolecules in many soils and sediments, the result of a partial breakdown of organic material, primarily lignin, deposited over decades. These macromolecules are largely nonpolar, although they are heterogenous and may contain polar

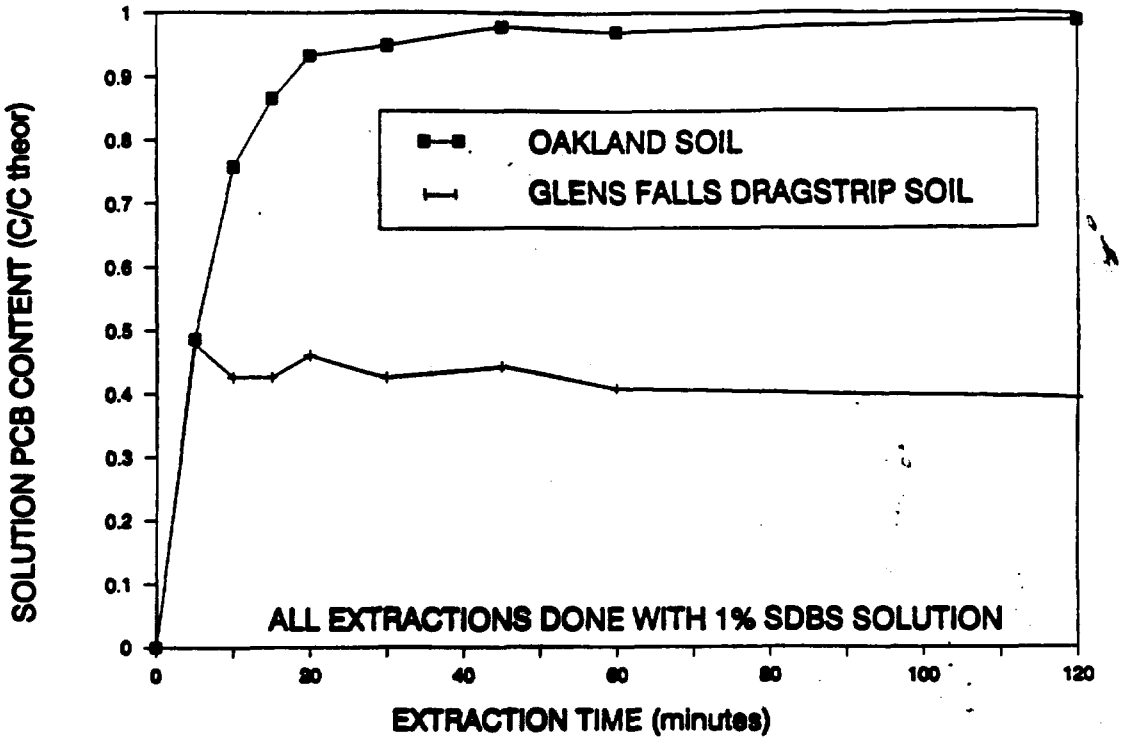

Figure 12-1. Kinetic data for surfactant extraction of PCBs from two soils

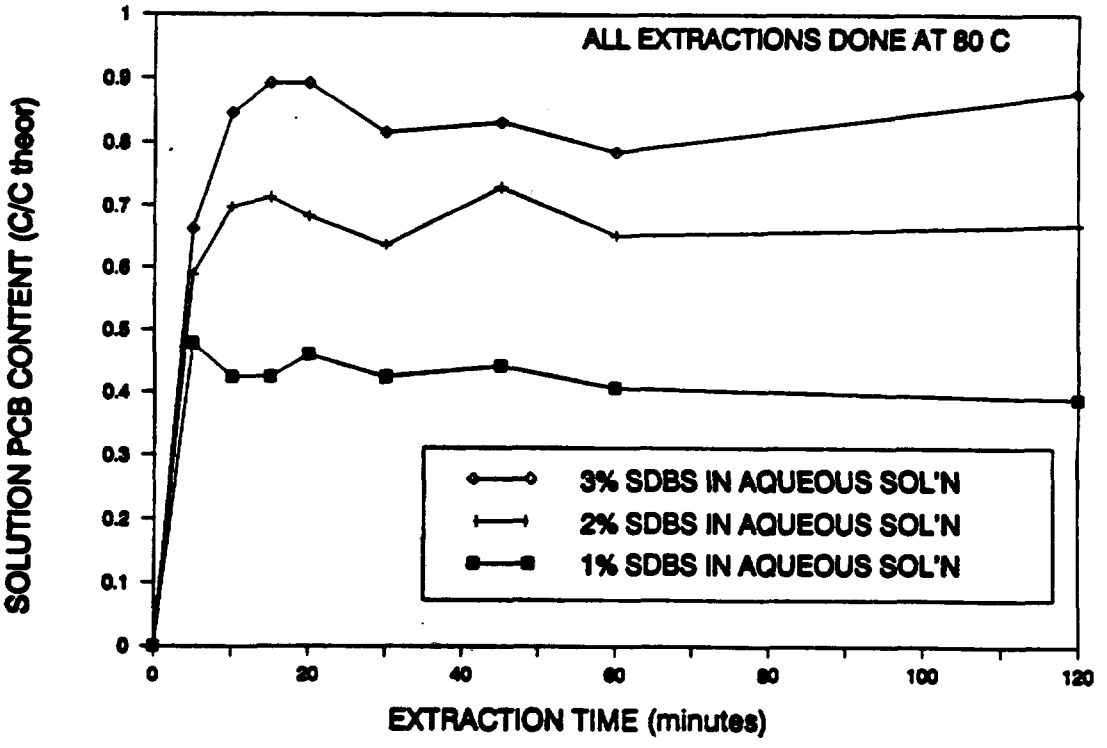

Figure 12-2. The effect of surfactant concentration on the extraction kinetics of PCBs from Glens Falls soil

constitutive groups. When present with PCBs in aqueous media, they act as a sorptive phase for the hydrophobic PCB molecules, shielding them from the polar aqueous environment.

Partitioning into NOM is generally described by an equilibrium partition coefficient, K_{oc}, defined as the concentration of solute present in the natural organic carbon phase in mg/kg divided by the concentration of solute present in solution in mg/L. Typical K_{oc} values for PCBs in water range from 10^5 to 10^8 L/kg [Karickhoff et al., 1979], depending on the average number of chlorine atoms attached to the biphenyl molecule. In natural systems the organic carbon will make up only a fraction of the total soil or sediment mass. In this case, a general partitioning constant, K_p, is defined as

$$K_p = K_{oc} f_{oc} \qquad [1]$$

where f_{oc} is the mass fraction organic carbon in the soil or sediment.

The organic carbon partition coefficient of a hydrophobic organic molecule is strongly correlated with its solubility [Chiou et al., 1979]. The addition of surfactant above the critical micelle concentration creates a second organic phase in a water/soil or water/sediment system. Surfactant micelles and soil/sediment NOM now compete for hydrophobic molecules. Increasing the surfactant concentration increases the number and volume of micelles in the system, increasing its capacity to take up these molecules. There is a shift from molecules partitioned into the soil NOM to molecules partitioned into the surfactant micelles. This phenomenon explains why varying the SDBS concentration had such a significant effect on the extent of extraction of PCBs from dragstrip soil.

A series of experiments using dragstrip soil and soil extract were run to further confirm that equilibrium partitioning is controlling the extraction process. Five gram samples of dragstrip soil were spiked with from 0 to 2 mL of concentrated (1000 mg/L) PCB extract obtained from a hexane/acetone soxhlet extraction of the soil. The hexane and acetone were allowed to evaporate and 15 mL volumes of 1% SDBS solution were added to the soil. The samples were placed in an ultrasonic bath at 50°C for 1 hour, then allowed to stand and approach equilibrium for 24 hours. The samples were centrifuged and the supernatant assayed for PCB content. The soil was dried, extracted, and also analyzed for PCB content. The final soil PCB concentration was adjusted for the contribution of residual surfactant solution dried onto the soil.

In addition to these trials, another series of experiments were run to extend the range of equilibrium measurements to low PCB concentrations. In this case, sample soil loadings were varied from 1-5 g and then combined with 15 mL of 1% SDBS solution. No PCB spiking was done. The samples were processed as described above.

The results of both series of experiments are combined in the curve in Figure 12-3. In an ideal equilibrium partitioning situation a plot of soil PCB concentration versus solution PCB concentration at equilibrium should appear as a straight line through the origin with K_p as the slope. This is generally the case here. The fact that the line does not appear to go through

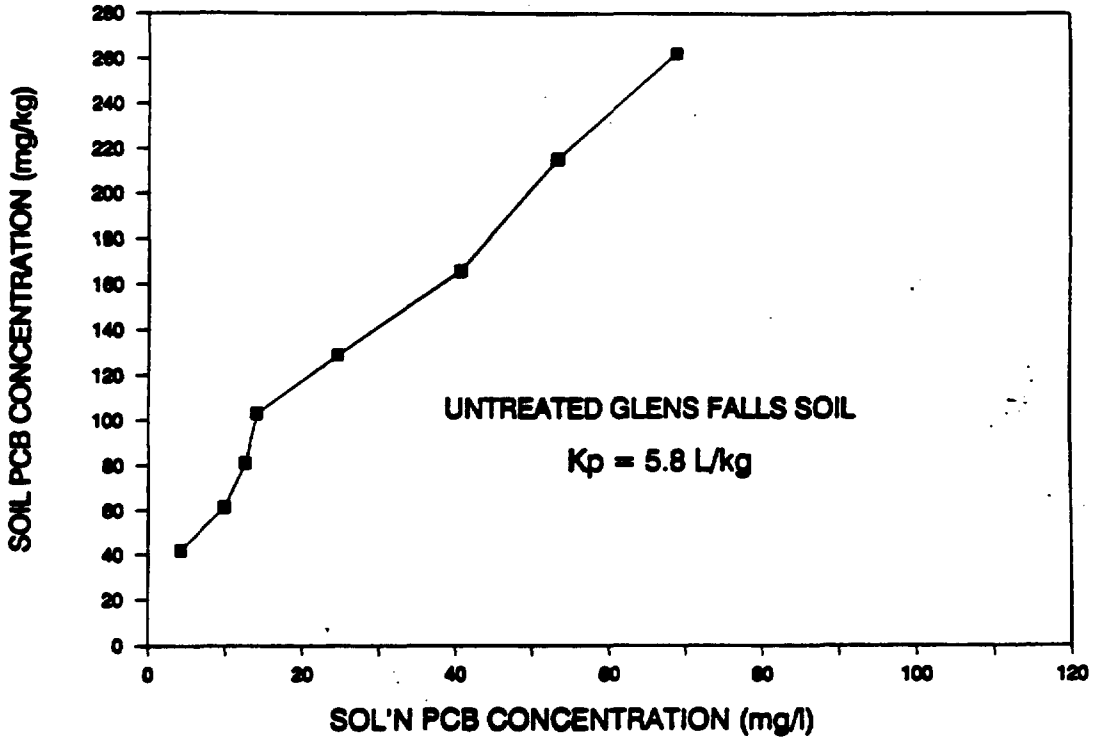

Figure 12-3. Equilibrium partitioning of PCBs between Glens Falls soil and surfactant solution

the origin may reflect diffusional resistance in the soil matrix. True equilibrium may not have been reached in the 24-hour equilibration periods. However, the general equilibrium hypothesis is supported by these data.

Aerobic Biodegradation

It is generally assumed that aerobic microorganisms require substrates in soluble form in order for biotransformation to occur. The transport of slightly soluble substate into the aqueous phase can be rate limiting in many biological processes. Substrates sequestered in NOM macromolecular matrices are also presumably not available to the organisms. As in surfactant systems, the transport of hydrophobic molecules from this phase, through an aqueous medium, and into a second lipophilic phase, this time in a microorganism, is of significant importance to biodegradative processes.

The bioavailability of aged PCBs in soils and sediments can be assessed by removing the PCBs from the soil/sediment matrix via soxhlet extraction, followed by biodegradation of the extract. The extent of biodegradation observed can then be compared to that in the original matrix under similar conditions. These experiments have been done for PCBs in Glens Falls dragstrip soil.

Soxhlet extractions were performed using hexane/acetone as the extraction solvent. The biodegradation studies shown were done with *Pseudomonas* sp. JB1P3C organisms [GE Report, 1989] in 72-hour resting-cell assays. The results appear in Table 12-2. The presence of a large difference in starting PCB concentration in the assays makes it difficult to compare the degradation numbers directly. Yet given that aerobic biodegradation generally proceeds more efficiently (i.e., degrades a higher percentage of the material present) at lower concentrations, these results indicate significant limitations in PCB bioavailability do exist in the soil, consistent with the surfactant extraction results.

TABLE 12-2. PCB bioavailability in Glens Falls dragstrip soil

Soil Sample	Starting [PCB] (ppm)	Cell OD	Mass Sediment (g)	% Degraded
GF dragstrip	20.0	5.0	0.2 (wet)	35%
GF (soxhlet)	130.0	5.0	---	54%

A simple trial was performed to show that this bioavailability limitation could be due to the NOM present in the dragstrip soil. One hundred microgram portions of Aroclor 1242 dissolved in hexane were deposited into a series of vials, with the hexane allowed to evaporate. To these vials were added neutralized Aldrich humic acid dissolved in 0.5 mL of water in ratios of 10:1, 50:1, 100:1, and 200:1 by weight to the Aroclor. The vials were shaken for 24 hours to allow the PCBs to come into equilibrium with the humic acid, then JB1P3C cells were added and standard resting-cell assays performed.

The results are shown in Figure 12-4. Inhibition of both the rate and extent of biological activity was observed at humic acid ratios of 100:1 and above. This equates to 1% humic acid in the assay. Duplicate vials were checked for pH shifts that might have occurred over the course of the experiment, but none were found. It has been noted in the literature that hydrophobic compounds tend to partition more strongly into commercial humic acid than they do into NOM [Chiou et al., 1987]. Therefore, this experiment qualitatively demonstrates the potential of NOM to account for the limited biodegradation results observed in dragstrip soil.

Natural organic matter is not alone in acting as a sorptive phase for PCBs in the environment. In a recent communication, Boyd and Sun [1990] showed experimentally that oil and grease can be up to 10 times more effective than NOM as a sorptive phase for hydrophobic compounds. They reformulated the expression for the generalized partition constant in equation [1] to include the contribution of this additional phase

$$K_p = K_{oc} f_{oc} + K_{oil} f_{oil} \qquad [2]$$

where f_{oil} is now the oil and grease fraction in the soil or sediment. Thus the extent of PCB partitioning can be expressed as a combined function of both the organic carbon and oil/grease content of the soil/sediment matrix.

Both NOM and oil/grease are present in many environmental samples [GE Report, 1988]. Table 12-3 shows a compositional analysis of two sediment samples, one from the upper Hudson River and one representing a composite of several samples from Woods Pond in Massachusetts. Both samples are high in organic carbon, as measured by combustion of the dry sediment. This is especially true of the Woods Pond sample. The fraction of oil and grease present in each was determined by performing a soxhlet extraction on the sediment and measuring the loss in mass. Both straight hexane and hexane/acetone were used as extraction solvents, yielding similar results. This indicates oil and grease make up a substantial part of the extract. Samples of the extract were then submitted for infrared (IR) analysis.

TABLE 12-3. Compositional analysis of selected sediments

	Upper Hudson	Woods Pond
Water Content (%)	41 - 46	73 - 77
Extractable Organic (%) *	0.55	2.7
Combustible Fraction (%) *	7.8	23.1
PCB Content (mg/kg)	57	98

* indicates analysis done on a dry sediment weight basis

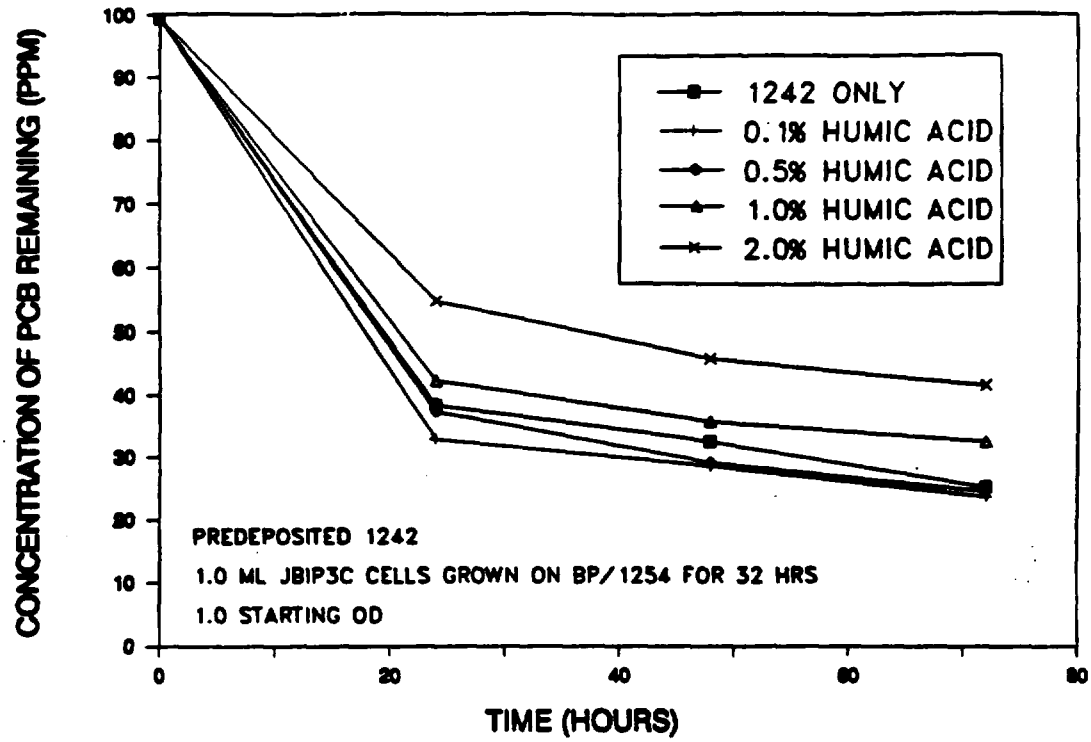

Figure 12-4. The influence of Aldrich humic acid on PCB biodegradation by JB1P3C

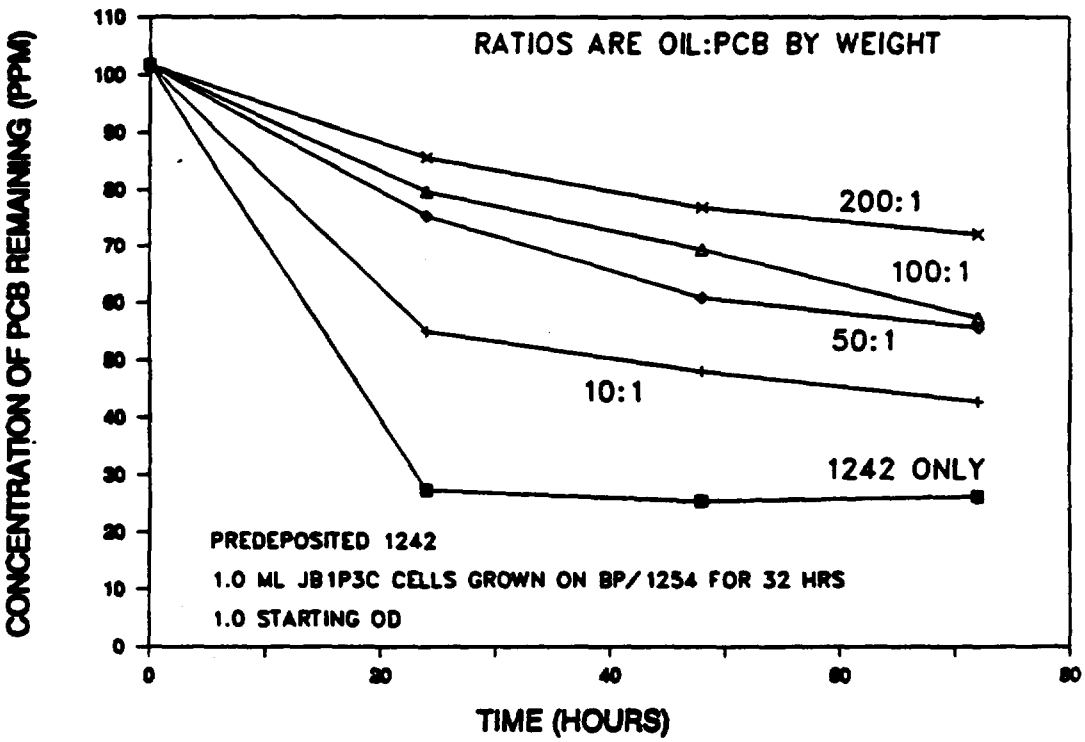

Figure 12-5. The influence of mineral oil on PCB biodegradation by JB1P3C

The IR analyses indicate the presence of an aliphatic hydrocarbon oil in both samples. The Woods Pond sample probably contains mostly mineral oil, based upon the large amount of this material historically used at nearby manufacturing sites. There is no single likely candidate for the type of oil found in the Hudson River sediment.

General Electric has demonstrated in the past that the presence of oils can make PCBs unavailable for aerobic biodegradation [GE Report, 1988; Bedard, 1990]. To further confirm those results, an experiment was carried out where light mineral oil was added with Aroclor 1242 to small vials in ratios of 10:1, 50:1, 100:1 and 200:1 by weight. Water was added and the vials were shaken for 24 hours to allow the oil and PCB to approach equilibrium. JB1P3C cells were then added to the vials and standard resting-cell assays performed.

The results appear in Figure 12-5. The inhibition of biodegradation is much more severe here than observed with the Aldrich humic acid. Inhibition begins at the lowest concentration of oil and increases progressively as additional oil is added to the system. Partitioning of the PCBs into the oil phase is the logical explanation for these observed results. The magnitude of these effects indicate strong partitioning and support Boyd's findings that the partitioning coefficients for oil phases may be much greater than those for NOM.

These results have been obtained with aerobes, but there is no biological or physical basis for expecting this phenomenon to depend upon type of organism. Anaerobic dechlorination may also be affected by partitioning-related bioavailability issues. However, several factors should be taken into consideration.

Partitioning is ideally a reversible, dynamic equilibrium process. If a system at equilibrium is perturbed, such as by removing PCBs from the aqueous phase, it will return to equilibrium given sufficient time. This time to equilibrium is determined by the rate of transport of the PCB out of the partitioning phase and may be limited by adsorption or diffusion phenomena in that phase. In aerobic biodegradation trials in the laboratory, organisms are usually present at high concentrations and degradation is fast. The organisms act as sinks for PCBs, with depletion of some PCB congeners occurring on a time-scale of hours. If the transport of PCBs out of the partitioning phase cannot keep up, the system operates under nonequilibrium conditions, with PCBs depleted in the aqueous phase. Bioavailability limitations result. If cells run out of energy or die over time, as commonly occurs in resting-cell assays, biodegradation may cease altogether.

In anaerobic dechlorination, both in the laboratory and in nature, organisms are generally present at much lower cell concentrations. Anaerobes transform PCB molecules into lower chlorinated congeners, rather than oxidizing them as in the aerobic case. They act over time-scales of months, rather than hours [GE Reports, 1988; 1989]. These observations suggest that anaerobic systems should operate at conditions much nearer to equilibrium concentrations of PCB in the aqueous phase than do the aerobic systems we have studied. However, in cases such as Woods Pond, whose sediment contains high levels of both NOM and oil, it is conceivable that the equilibrium concentration of PCBs is so low that it cannot even maintain the active organism population necessary to carry out anaerobic dechlorination. This may

explain the modest natural dechlorination seen to date on that site [GE Report, 1990].

Enhanced Partitioning

The partitioning of PCBs into soil humic materials adds significant difficulty and cost to a surfactant extraction process and will impair attempts at biological remediation as well. One method of addressing this problem is to seek either to remove the NOM from the soil or sediment matrix or to change the character of that NOM so that it no longer is a strong sorptive phase for hydrophobic compounds.

Certain organic components of soils can be solubilized with dilute caustic solutions [Mortensen, 1979]. Humic and fulvic acids, which make up most of the dragstrip natural organic fraction, are in this category. A third component of NOM, humin, is not solubilized by this treatment. Sodium hydroxide in concentrations of 0.1 N to 0.5 N is commonly used to remove these solubilized fractions in soil analysis. If heat is added with the caustic, the acids undergo partial hydrolysis. Both solubilization and hydrolysis of the humic material can be expected to have a positive effect on the partitioning of PCBs with regard to extraction.

This thesis was tested in a pair of experiments involving caustic-treated dragstrip soil. In the first experiment 0.5 N NaOH caustic solution was added to dragstrip soil in a 4:1 ratio and shaken for 1 hour. The contents were then centrifuged and the dark brown supernatant containing the dissolved humic material drawn off. A second volume of clean caustic was then added to the soil and this procedure repeated until the supernatant was no longer deeply colored. Approximately 10% of the PCBs originally present in the soil were removed in the supernatant. The caustic-treated soil was then neutralized, dried, and 5 g samples were spiked with concentrated dragstrip extract (0-2 mL). Fifteen mL volumes of 1% SDBS solution were added to each sample and equilibrium partitioning experiments were carried out as previously described.

In the second experiment, the 0.5 N caustic solution was added to the soil in a 1:1 ratio and the slurry was refluxed for 3 hours using a heating mantle and condenser. This time the supernatant was not removed. Instead, the slurry was neutralized and air dried. The dissolved organic fraction remained with the soil. Five gram samples were then spiked with concentrated soil extract as before and the partitioning experiments performed.

The results of these experiments are given in Figure 12-6. Soil from both caustic treatments show a significant decrease in PCB partitioning compared to the untreated standard (K_p = 1.3 and 0.9 L/kg versus 5.8 L/kg for the standard). Soil subjected to both caustic and heat treatment showed the least partitioning despite the fact that its solubilized organic component was not removed. The organic phase must be altered in order for this to occur, making the PCBs more available for extraction.

A kinetic study was carried out using the caustic and heat-treated dragstrip soil to verify this result. The soil was slurried with surfactant and the PCBs extracted (1% SDBS, 80°C, 2:1 solution to soil ratio) as the supernatant was monitored for PCB concentration. The results

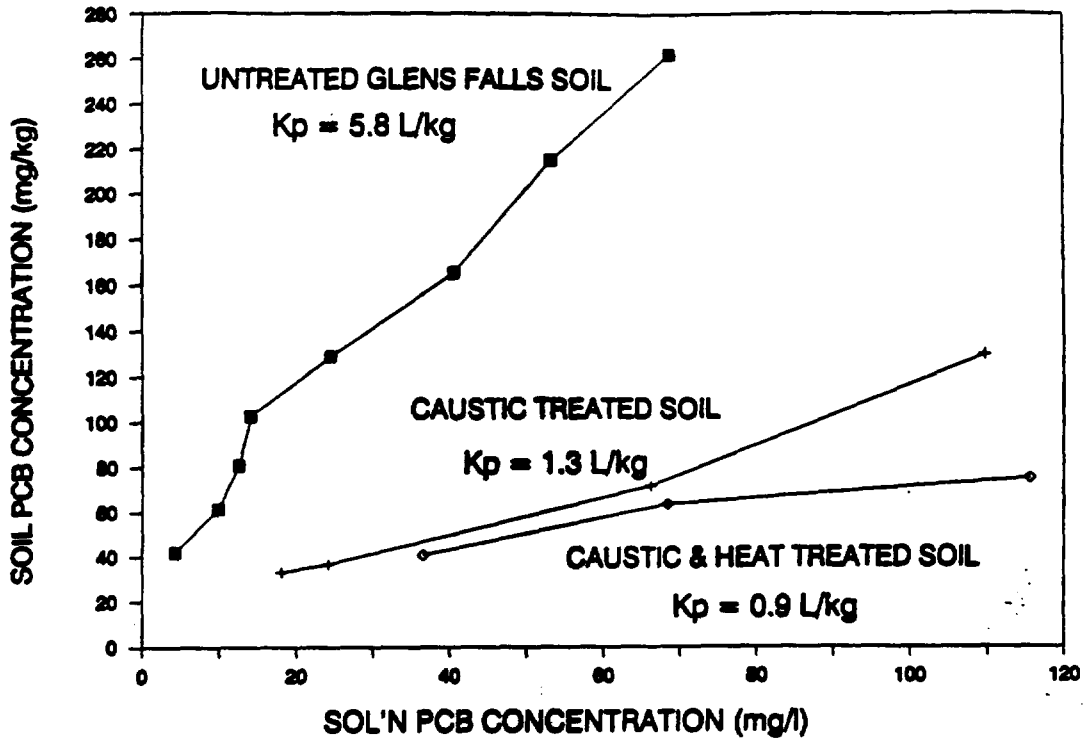

Figure 12-6. The effect of soil pretreatment on the partitioning of PCBs between Glens Falls soil and surfactant solution.

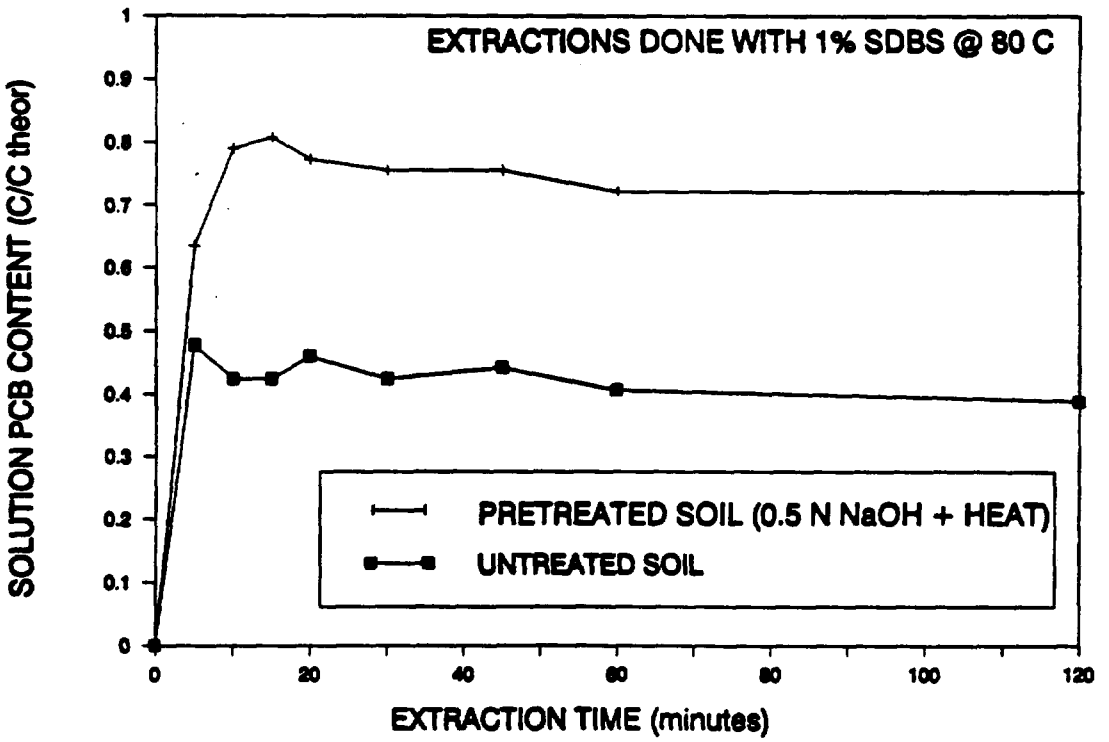

Figure 12-7. The effect of soil pretreatment on the kinetics of surfactant extraction from Glens Falls soil

are shown in Figure 12-7. About 75% of the PCBs in the soil are solubilized when the pretreatment is used, as opposed to <40% for untreated soil. It appears the ability of the soil humic matter to sorb PCBs is decreased by the pretreatment, but not eliminated.

Biodegradation experiments were also performed using the treated soils. Reliable data are available only on the soil with the soluble organic material removed. Fifty-five percent of the PCBs in the NOM-depleted soil were biodegraded by JB1P3C in resting-cells assays. This contrasts with 35% PCB biodegradation by JB1P3C observed using untreated dragstrip soil and is comparable to the soil-free degradation value given in Table 12-2.

SUMMARY AND CONCLUSIONS

The analogy between surfactant extraction and aerobic biodegradative processes is used to elucidate the effect of natural organic matter on PCB availability in soil and sediment matrices. PCB equilibrium partitioning into the NOM phase is demonstrated to have a detrimental effect on the extent and efficiency of PCB removal for both of these processes. Oil and grease also form a secondary phase in soils and sediments, and are shown to have a similar detrimental effect on biodegradation. The presence of one or both of these phases in most soils and sediments requires that their impact be better understood and factored into any extractive or biodegradation studies undertaken.

Partitioning into NOM can be reduced by pretreating the soil with a mild caustic (0.5 N NaOH) solution and heat. The treatment solubilizes and hydrolyzes the humic and fulvic fractions in the soil, limiting the extent to which PCBs can sorb into these fractions. The resulting enhanced PCB availability is beneficial to both extractive and biodegradative processes.

FUTURE WORK

We plan to continue to explore the PCB bioavailability issue as it impacts the aerobic bioremediation of river sediments. The caustic soil treatment may be beneficial as a pretreatment for a bioreactor system, but is of limited value if an *in situ* approach is desired. Greater understanding of the interaction between equilibrium partitioning and biological uptake will be required to achieve more effective bioremediation strategies.

Chapter 13

SYNTHESIS OF PROPOSED INTERMEDIATES IN THE AEROBIC DEGRADATION OF PCBs; RING-FISSION PRODUCTS

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INTRODUCTION

A major pathway for the destruction of PCBs by aerobic microorganisms [Furukawa et al., 1979; GE Reports, 1984-1989] closely resembles the pathway elucidated earlier for oxidation of biphenyl itself [Catelani et al., 1973; Gibson et al., 1973]. Oxidative cleavage of biphenyl-2,3-diol yields the ring-fission product (I, Ar = phenyl) which is further degraded in at least two different ways (Figure 13-1). Isomerization to the di-keto tautomer (Ik) and cleavage by hydrase leads to benzoic acid via path A. Path B has been proposed [Omori et al., 1988] to explain the isolation of keto-acid II as a metabolite of I.

Both paths A and B (Figure 13-1) have also been observed in PCB metabolism. Chlorobenzoic acids (Cbas) are major metabolites of PCBs in numerous organisms. Keto-acid II (Ar = 2-chlorophenyl) has been found as a metabolite of both 2-CB and 2,2′-CB in a recombinant strain of *E. coli* that harbors a plasmid (from *Pseudomonas* sp. LB400) encoding the first three enzymes of the biphenyl/PCB pathway but not the hydrase needed for cleavage of I to benzoic acid [Bedard, GE Report, 1989].

There is some indication of a third way (path C) by which the ring-fission product (I) can be degraded [Barton and Crawford, 1988]. Oxidation of 4-CB by a *Pseudomonas* sp. was reported to produce large amounts of 4-chloroacetophenone (4-CA). The corresponding ring-fission product (I, Ar = 4-chlorophenyl) was not shown to be an intermediate in this case, so we cannot consider that path C is firmly established.

Oxidation of 23-3-CB by several different strains led to attack on the 3-chlorophenyl ring and isolation of both 2,3-CA and 2,3-Cba [Bedard et al., 1987; Bedard and Haberl, 1990, in press]. Other congeners possessing a 3-chlorophenyl ring behaved similarly. It was proposed [GE Report, 1987] that the pathway in this case involved the tri-keto acid III (Figure 13-2). Attack at either C_4 or C_6 could lead to cleavage at the C_4C_5 bond or the C_5C_6 bond, respectively, to produce the observed products. Many instances are known in which unsymmetrical β-diketones undergo non-enzymic hydrolytic cleavage in both possible directions [Hauser et al., 1948].

The goals of the present research were to develop chemical syntheses for compounds I and III, and to explore their reactions under non-enzymic conditions, as well as their transformation by appropriate microorganisms. In this report we describe the preparation of I (Ar = phenyl) and of six compounds of type III, and preliminary results of a study of their non-enzymic cleavage reactions.

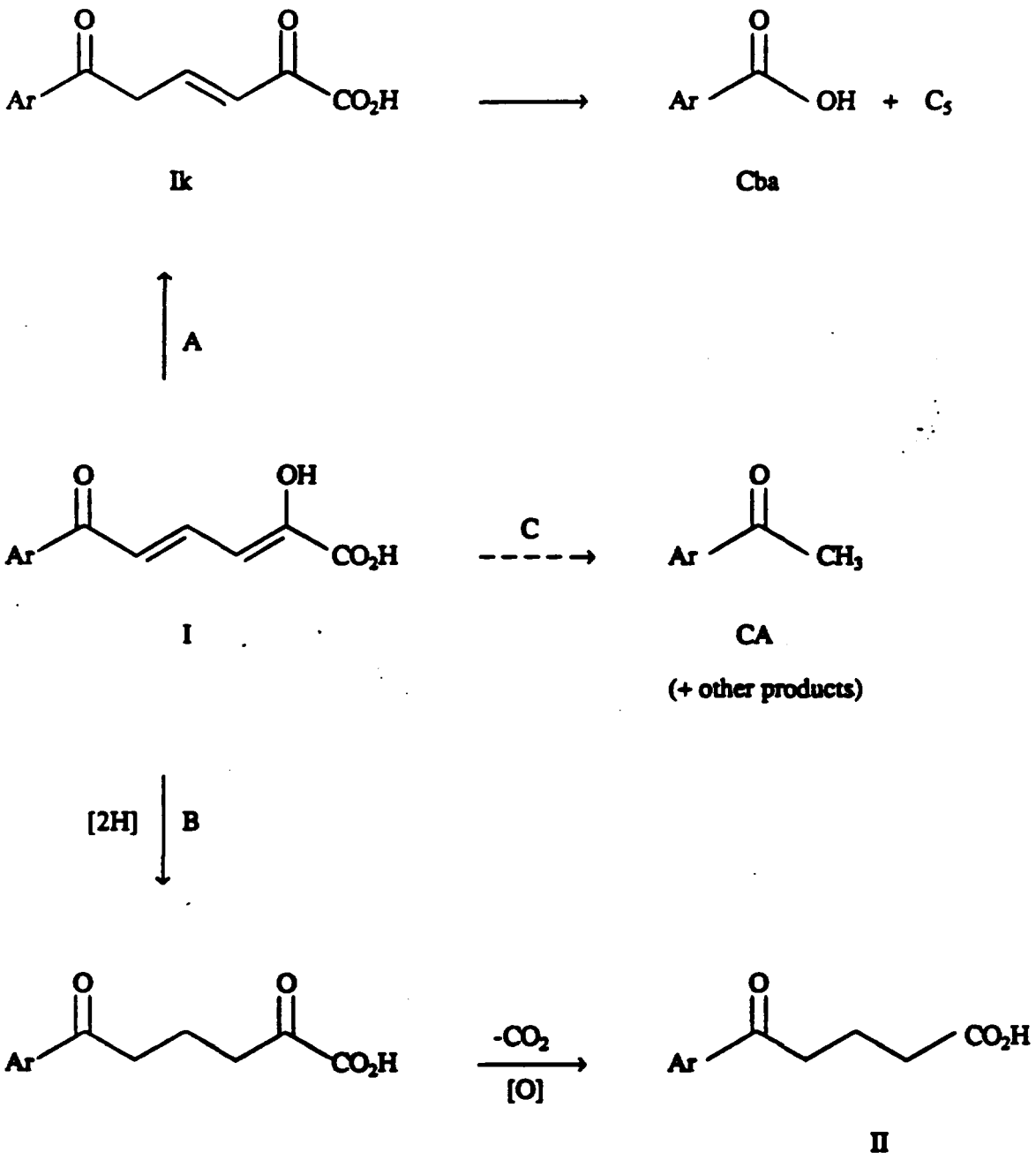


Figure 13-1. Paths for metabolism of the ring-fission product (I).

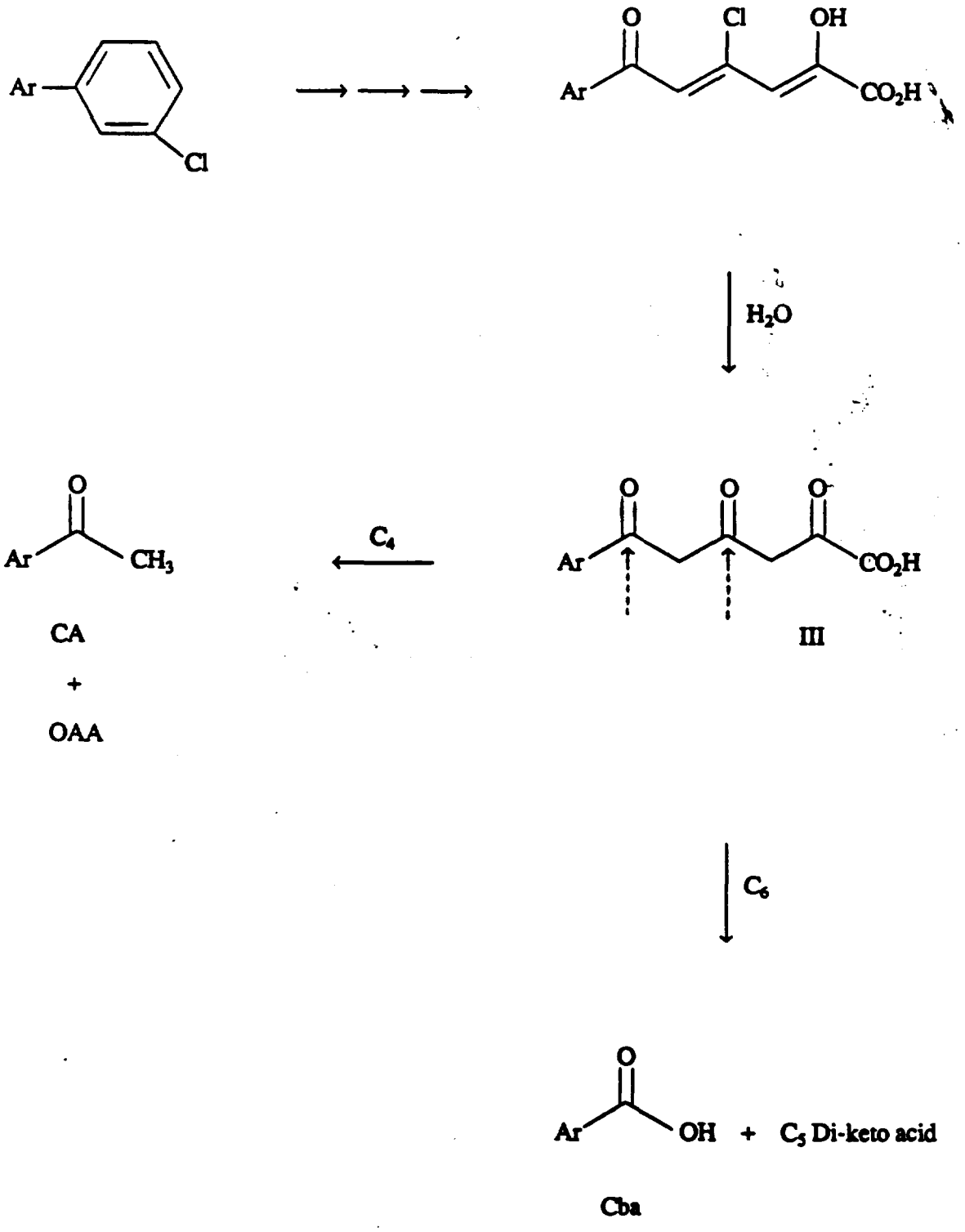


Figure 13-2. Proposed pathway for degradation of a 3-chlorophenyl ring. Compound III can exist as a mixture of tautomeric forms in which one or two of the three keto groups is in the enol form. The numeral III is used throughout the text to refer to any one or all of these species.

RESULTS AND DISCUSSION

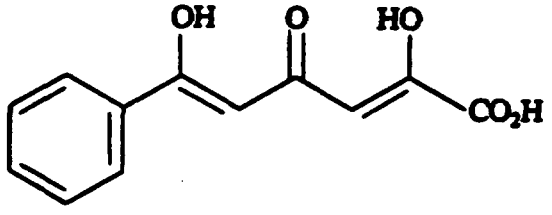
Synthesis of the Ring-Fission Product (I)

The ring-fission product from biphenyl (I, Ar = phenyl) has been isolated from microbial cultures and characterized [Catelani et al., 1973] but no chemical synthesis of the compound has been reported. We have found that the methyl ester is readily prepared from crotonophenone (1-phenyl-2-buten-1-one) and dimethyl oxalate (DMO). Hydrolysis in aqueous LiOH, followed by acidification, affords the free acid as yellow crystals which darken on storage.

Comparison of the physical properties of our product and that described by Catelani reveals satisfactory agreement (Table 13-1). Samples of our product decomposed upon heating at temperatures somewhat higher than he reported, but decomposition points are notoriously dependent on particle size and heating rates, and the observed difference may not be of much significance.

Synthesis of Tri-Keto Acids (III)

The prototype tri-keto acid III (Ar = phenyl) was recently synthesized for the first time in this laboratory [Stiles, unpublished]. Condensation of benzoylacetone with dimethyl oxalate (DMO) in the presence of magnesium methyl carbonate (MMC) provided yellow crystalline product in yields above 50%. X-ray crystallographic analysis [Selegue, unpublished] indicated



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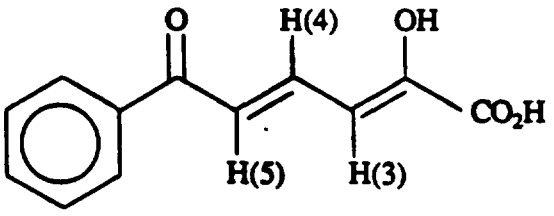
that the crystalline substance exists as the tautomer IIIA. NMR spectra of solutions in acetone-d_6 indicated the presence of small quantities of the two possible monoenols in equilibrium with the dienol illustrated. No evidence was seen of the completely ketonized tautomer.

We have now succeeded in preparing five chloro substituted derivatives of III, starting in each case with the appropriate chloro substituted benzoylacetone. Table 13-2 summarizes these synthetic results.

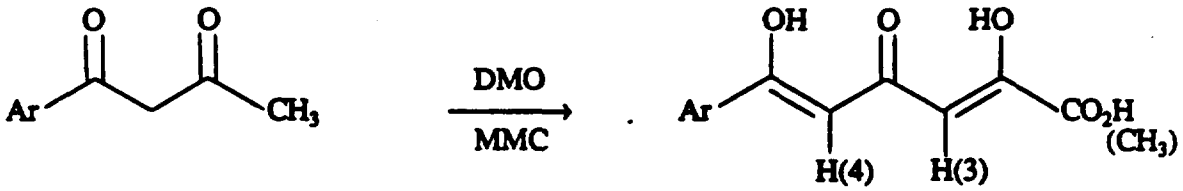
TABLE 13-1. Physical properties of the ring-fission product



		Present Work	Catelani et al., 1973
NMR: (δ)	H(3)	6.53 d (J = 12)	6.52 (J = 12, 0.5)
	H(4)	7.91 dd	7.91
	H(5)	7.39 d (J = 15)	7.34 (J = 16, 0.5)
M.S.:		218 202 201 173	218 202 173
		157 128 105 77	157 105
U.V.:[a]	pH 2	337 nm (4.2)	336 nm (4.3)
	pH 12	435 nm (4.5)	435 nm (4.5)
M.P.:		ca 134° dec.	112° dec.

[a]All of our samples have exhibited a weak absorption of variable intensity near 250 nm. We consider it probable that this absorption is due to an impurity.

TABLE 13-2. Yield and NMR data for the tri-keto acids III-VIII



Ar		NMR δ (acetone-d_6)		Yield (%)[a]
		H_3	H_4	
III	C_6H_5	6.44	6.62	82
IV	2-ClC_6H_4-	6.43	6.34	79
V	4-ClC_6H_4-	6.44	6.66	91
VI	2,3-$Cl_2C_6H_3$-	6.41	6.31	81
VII	3,4-$Cl_2C_6H_3$-	6.41	6.75	71
VIII	2,4-$Cl_2C_6H_3$-	[b]	[b]	high

[a]Free acid plus methyl ester.

[b]Compound VIII was badly contaminated with a product containing one methoxyl group in place of one of the chlorines. It was not possible to assign the NMR signals in the 6-7 δ region with certainty.

Non-Enzymic Cleavage of the Tri-Keto Acids

Treatment of tri-keto acid III (Ar = phenyl) with 1 M NaOH at room temperature resulted in slow (half-time ca. 8 hours) hydrolytic cleavage to form benzoic acid, acetophenone, oxalic acid, and acetic acid. The 4-chloro derivative (V) behaved similarly. Yields are summarized in Table 13-3. A reaction scheme is presented in Figure 13-3. The products can be accounted for on the basis of attack at either C_2 or C_6 of the tri-keto acid. Products that would result from attack at C_4 (benzoylacetic acid, oxaloacetic acid) were not found.

In separate experiments we confirmed previous reports [Hauser et al., 1948; Lehninger and Witzemann, 1942] that the diketones IX and X undergo the indicated reactions at rates that are rapid compared to that which we observe for the tri-keto acids. Measurement of the ratio CA/Cba formed from the diketones IX and IXa (Ar = 4-chlorophenyl) gave values of 2.5 and 1.2, respectively. From these data and the yields given in Table 13-3, the ratio C_6/C_2 for the two modes of attack on the tri-keto acids III and V can be calculated to be 1.0 and 1.1, respectively. The effect of *p*-chloro substituent thus has only a small effect on the point of attack. Studies of the more highly chlorinated derivatives VI-VIII will be instructive on this point.

Non-Enzymic Cleavage of the Ring-Fission Product (I)

The ring-fission product (I) proved more resistant to alkali than the tri-keto acid (III). At 50-55° in either K_2CO_3 or NaOH (1 M) a slow reaction could be readily observed by following the decay in absorbance at 435 nm. However, after complete disappearance of the 435 peak (several days were required) analysis of products did not give a satisfactory material balance. Oxalic acid (>50% yield) benzoic acid, and acetophenone were readily identified, but the ratio of the latter two varied widely in different experiments. Efforts to improve the analysis of this reaction mixture are continuing.

Although oxalic acid is a major product of the non-enzymic hydrolysis of both I and III, this substance has not been reported to be a metabolite of biphenyl or PCBs. Evidently, the enzyme directs attack on the ring-fission products at points more remote from the ionized carboxyl group than C_2. An analogous situation exists with acetylpyruvic acid (X). Non-enzymic hydrolysis leads quantitatively to oxalic acid and acetone, as shown in Figure 13-3, but the compound is metabolized to acetate and pyruvate [Lehninger and Witzemann, 1942; Meister and Greenstein, 1948].

SUMMARY AND CONCLUSIONS

The ring-fission product (I) has been prepared for the first time by chemical synthesis. Studies of PCB metabolism may be facilitated by the availability of this compound in gram quantities.

TABLE 13-3. % Yields of products from alkaline hydrolysis of tri-keto acids III and V

	$ArCO_2H$	$ArCOCH_3$	Oxalate[a]	Acetate[b]
III	69[c]	37[d]	80	n.d.
V	67[bc]	21[c]	82	+
	70[d]	25[d]		

[a]By permanganate titration following isolation of Ca salt.

[b]Identified by ^{13}C-NMR.

[c]Determined from weight of isolated product.

[d]Determined by ^{1}H-NMR.

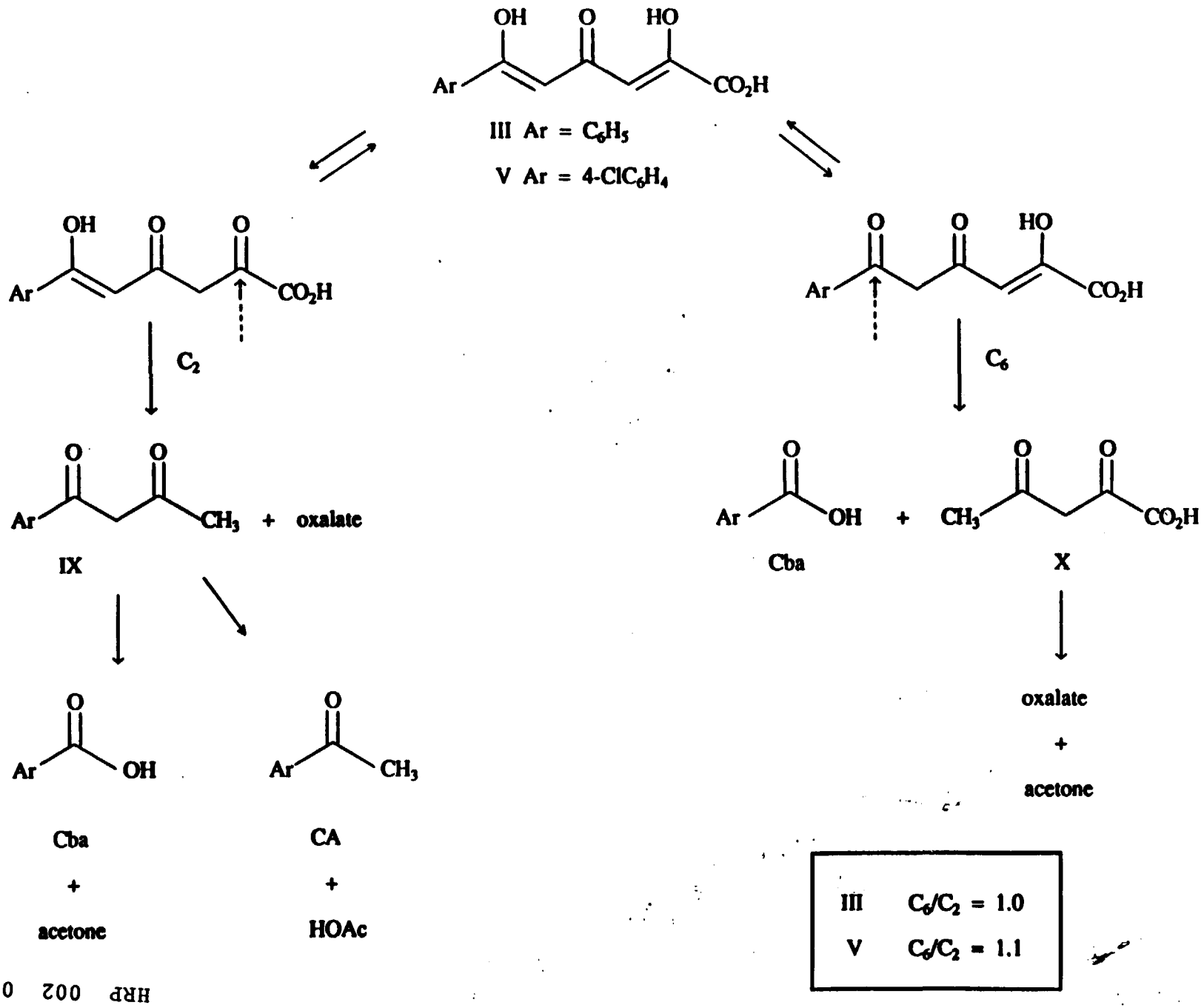


Figure 13-3. Scheme for the alkaline hydrolysis of III and V. Acetone was not determined; yields of other products are given in Table 13-3.

The tri-keto acid III (Ar = phenyl) and five chloro substituted derivatives (IV-VIII) have been synthesized.

Non-enzymic hydrolysis of I, III, and V yielded both benzoic acid and acetophenone (4-Cba and 4-CA in the case of V). These compounds are the major aromatic products formed in the aerobic degradation of many PCBs, and this result may thus lend support to the proposed hydrolytic pathways involving I and III. However, alkaline hydrolysis may not be a good model for the hydrase reaction, and experiments with I and III in microbial cultures should be completed before firm conclusions can be drawn.

Oxalic acid is a major product of the non-enzymic hydrolytic cleavage of compounds I, III, and V. This compound has not been found as a metabolite of biphenyl or PCBs. We suggest that oxalate may be a key substance in delineating the differences between enzymic and non-enzymic mechanisms of attack on this class of compound.

FUTURE PLANS

Chloro derivatives of I, and additional derivatives of III, will be prepared as need arises. Examples will be chosen which correspond to those PCB congeners that are of interest as substrates for aerobic degradation.

Studies of the alkaline cleavage of I and III, including the chloro derivatives, will be continued. We propose to study the effects of pH and of ring substitution on the rate of this reaction, which may be viewed as a crude model for the hydrase reaction.

Efforts will be made to observe the hydrolysis of synthetically produced I and III by cultures of LB400, H850, and/or such other strains as may be available for the purpose.

Chapter 14

THE QUANTITATION OF POLYCHLORINATED BIPHENYLS BY GAS CHROMATOGRAPHY AND TANDEM MASS SPECTROMETRY

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INTRODUCTION

The focus of this research has been to develop methods of analysis for PCB congeners involved in anaerobic dechlorination studies [Brown et al., 1987a; Chen et al., 1988; Quensen et al., 1988]. A major problem with detecting this dechlorination is that with routine gas chromatography (GC) and mass spectrometric (MS) methods, the PCB congeners of interest are often not resolved. Electron capture detection of chromatographically separated PCB isomers is a highly sensitive technique, but coeluting and overlapping components are difficult, if not impossible to identify and quantitate [Duinker et al., 1988; Schulz et al., 1989]. Although MS detection offers enhanced specificity of analysis, detection limits are often not adequate for the samples of interest. Thus, it is desirable to develop methods which will allow the accurate quantitation of individual congeners which may or may not be chromatographically resolvable from other congeners.

It has long been known that chlorinated aromatic molecular anions will undergo an exchange of a chlorine atom for an oxygen atom in the presence of an oxygen diradical in the source of a mass spectrometer [Hunt et al., 1975; Hass et al., 1979; Mitchum et al., 1980]. By using a triple-stage quadrupole mass spectrometer [Yost and Enke, 1983], this reaction can also be observed when oxygen is introduced into the second quadrupole (collision cell) in an MS/MS experiment [Kostiainen and Auriola, 1988 and 1990]. In addition, the oxygen-chlorine exchange reaction is specific for odd-electron molecular anions which allows for the elimination of many of the interferences (especially from more highly chlorinated congeners) present in other methods. As such, the monitoring of this reaction may be used to quantitate PCB congeners which coelute with congeners having a different degree of chlorination.

By using this GC/MS/MS method of analysis, river sediment samples containing Aroclor 1242 which have been subjected to anaerobic activity have been quantitated. Also, river sediment samples which have undergone apparent *in situ* dechlorination have been analyzed. This method offers excellent detection limits, comparable to electron capture detection, while simultaneously providing the high specificity of MS/MS techniques.

EXPERIMENTAL

Sample Preparation

River sediment samples were spiked with Aroclor 1242 and then autoclaved. Three samples were then inoculated and allowed to incubate for 16 weeks. After incubation, three live and three autoclaved aliquots were each extracted with 50/50 acetone/hexane. Each extract was subjected to Florisil cleanup and then shaken with mercury for sulfur removal and finally concentrated to 10 mL. Prior to analysis, the three live samples were combined into a single sample, as were the three autoclaved samples. Sediment samples from the H7 Hudson River site and from Silver Lake were each extracted and prepared in a similar fashion. The Aroclor 1242 standard was provided by John Quensen, and all other individual congeners and standards were purchased from Ultra Scientific Company, Rhode Island.

Instrumental

All GC/MS and GC/MS/MS methods were performed with a Finnigan TSQ-70B mass spectrometer using a Varian 3400 gas chromatograph equipped with a 30 meter x 0.250 μ column of 0.25 μ DB-5 phase (J & W Scientific). The GC was temperature programmed at 140°C for 2 minutes, 140°C to 320°C at 6°C per minute and 320°C for 2 minutes. Helium was used as the GC carrier gas, ammonia was used as the chemical ionization reagent gas, and pure oxygen was introduced into the collision cell. The instrument utilizes a 20 kV dynode which was set at 5 kV. All reactions were generated using electron energies of 70 eV, collision offset energies of 2-3 eV (laboratory), and collision pressures from 0.5 mTorr to 1.5 mTorr. The instrument was tuned for unit resolution in both Q1 and Q3 modes of operation.

RESULTS AND DISCUSSION

Selectivity of MS/MS Detection

The goal of this work is to develop methods of analysis for quantitating PCB congeners which chromatographically coelute with more highly chlorinated congeners. Of particular interest are the six congeners which have been shown to exhibit dioxin-like toxicity; 34-34-CB, 234-34-CB, 245-34-CB, 345-34-CB, 2345-34-CB, and 345-345-CB [Kannan et al., 1988]. It is desirable to be able to quantitate these congeners, in particular 34-34-CB and 234-34-CB, in river sediment samples containing Aroclors. While electron capture detection offers excellent sensitivity, this method is not of use in distinguishing coeluting congeners. Though MS methods offer better selectivity with reduced sensitivity, coeluting structural isomers are still indistinguishable. In addition, quantitation of congeners in the presence of more highly chlorinated isomers by MS is difficult, if not impossible due to the lack of uniqueness of fragment ions. This is illustrated in Figure 14-1 which shows the negative chemical ionization (NCI) mass spectra of both a tetrachloro- and pentachlorobiphenyl. With NCI only a limited amount of fragmentation is observed as opposed to other modes of ionization such as

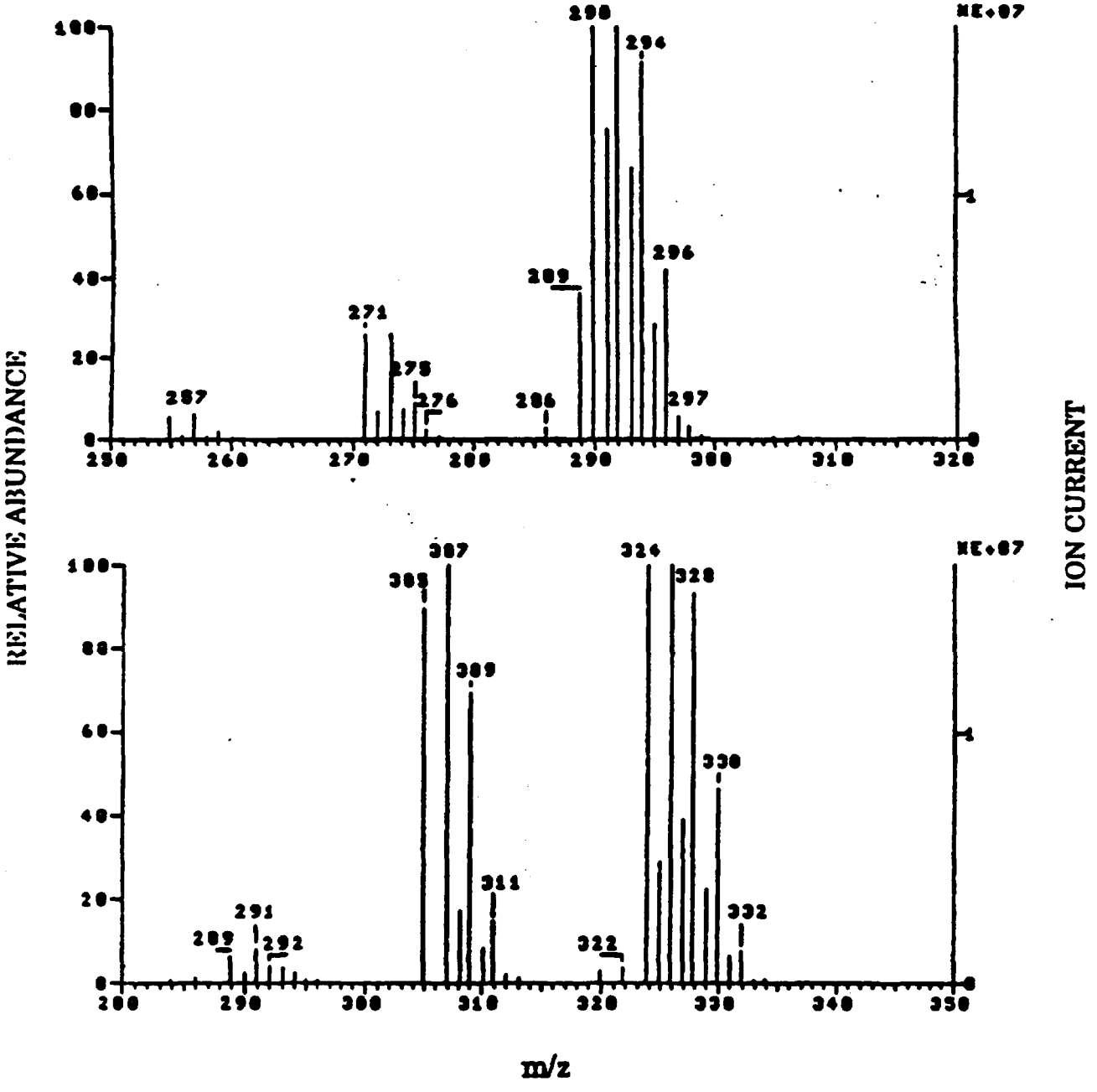

Figure 14-1. The primary (parent) mass spectra of a tetrachlorobiphenyl (top) and a pentachlorobiphenyl generated with negative chemical ionization.

electron impact (EI) ionization [Dougherty et al., 1972; Field, 1980; Daishima et al., 1989]. Also, the most abundant ions in the spectrum of the tetrachlorobiphenyl (the molecular ion region) also appear in the spectrum of the pentachloroisomer. It is for this reason then, that MS/MS methodology was explored for the purpose of enhanced specificity of analysis.

By forming the parent ion [M^-] under conditions of electron capture negative chemical ionization and selecting it in the first quadrupole of a triple-stage quadrupole mass spectrometer and introducing oxygen into the second quadrupole (collision cell), the exchange reaction of oxygen for chlorine may be observed. The m/z 290 ion which is the ^{35}Cl molecular anion of tetrachlorobiphenyls is also present in the primary mass spectrum of pentachlorocongeners as a result of the loss of chlorine from the ^{13}C molecular anion (Figure 14-1). The oxygen-chlorine exchange reaction, however, is specific for the odd-electron molecular anion. Therefore, the MS/MS detection of m/z 290 forming m/z 271 will be completely free of interference from congeners with other degrees of chlorination.

Since the degree of electron affinity of the [M^-] ion relative to that of the [$M\text{-}H^-$] ion is dependent upon chlorination of the aromatic ring and to some extent the positions of this chlorination, not all PCBs form [M^-] ions in the source and are thus not amenable to this reaction. With lower degrees of chlorination, the [$M\text{-}H^-$] ion is an even electron moiety and will not undergo chlorine exchange with oxygen. As a result, none of the mono or di substituted congeners and only a few of the tri substituted congeners are able to be analyzed in this fashion. Thus, shown in Figure 14-2 are the total ion current chromatograms for an Aroclor 1242 sample analyzed by monitoring the reaction products of the molecular anions of the tri- through nonachlorobiphenyls with oxygen. Since the congeners of particular interest are the aforementioned six toxic congeners, and these all consist of at least tetrachloro substitution, this method is of great utility in their analyses.

Choice of Chromatographic Internal Standard

Many PCB analyses are performed chromatographically using octachloronaphthlene as an internal standard. For ECD or MS routines this is a reasonable choice since it is chromatographically resolvable from PCB congeners and exhibits a response factor of the same order of magnitude as that of the PCB congeners. The response factor of an analyte is defined as the ratio of its detector response to its concentration. The response factors for several congeners with respect to octachloronaphthalene are listed in Table 14-1. This internal standard may not be the best choice, however, for the oxygen-chlorine exchange reaction. While octachloronaphthalene will undergo chlorine exchange with oxygen, it is not nearly as responsive as the PCB congeners. Response factors for PCB congeners vary up to three orders of magnitude with respect to this internal standard. The apparent reason for this low response of octachloronaphthalene is that fully-chlorinated aromatics exhibit a lesser tendency to undergo this reaction. This low response may also be one reason for not being able to observe a decachlorobiphenyl congener in the Aroclor 1242 sample. Since the response factors vary dramatically with the degree and position of chlorine substitution, it may be of interest to

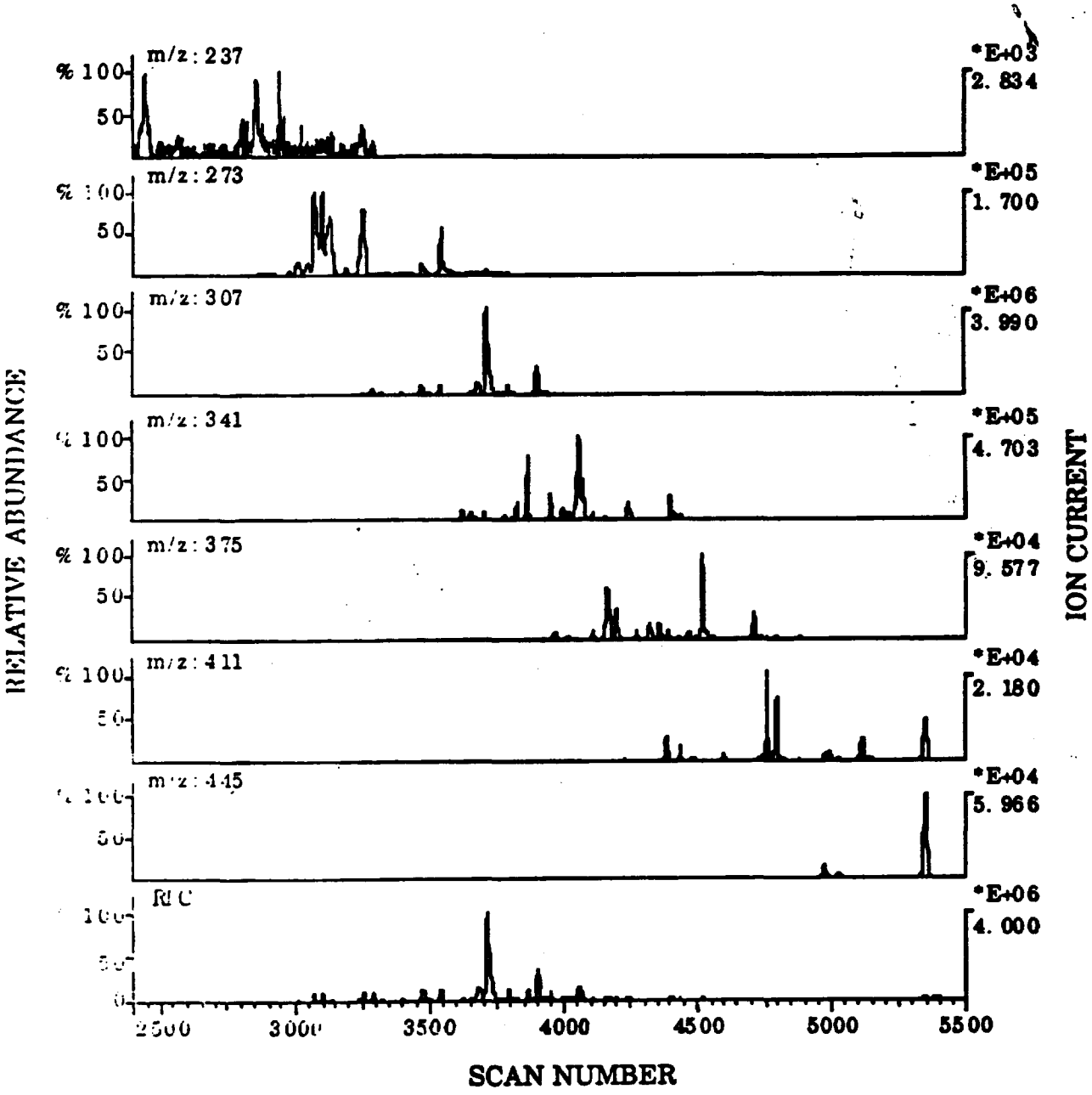

Figure 14-2. The total ion current chromatograms for the tri- through nonachlorobiphenyls of an Aroclor 1242 standard.

TABLE 14-1. Relative response factors (RRF) with respect to octachloronaphthalene and minimum detectable quantities (MDQ - in femtograms) for selected PCB congeners.

CB	RRF	MDQ
34-34	1.6	1200
234-24	290	4
234-34	18	110
236-34	88	12
245-34	11	180
345-34	14	140
234-236	94	11
236-245	194	5
245-245	134	8
2345-34	14	140
345-345	6.0	320
2346-236	48	22
2346-245	499	2
2346-2346	133	8
2345-2356	577	2
23456-345	164	6
23456-2345	234	5
23456-2346	222	5
23456-2356	122	9

explore using other or multiple internal standards for these analyses.

Detection Limits

A primary concern with the development of this method is the achievement of detection limits and sensitivities adequate for the samples of interest. Typical sediment samples contain anywhere from a few parts per million total PCBs to several hundred ppm. This means that many of the individual congeners of interest are on the order of parts per billion or less. While negative chemical ionization offers much better sensitivity and detection limits than EI/MS, it approaches the performance of an ECD only in selected-ion monitoring (SIM) modes of operation. For PCBs this involves the monitoring of chloride in particular. Using SIM, however, offers very little advantage in selectivity over the use of an ECD. By monitoring constant neutral losses of m/z 19 in an GC/MS/MS experiment, a sample may be specifically analyzed for chlorinated aromatics using the oxygen-chlorine exchange reaction. This technique is a scanning technique and as such has detection limits two to three orders of magnitude higher than that of an ECD or SIM. By monitoring only one reaction (for instance, m/z 324 to m/z 305), the high specificity of MS/MS is coupled with the low detection limits of SIM. A comparison of detection limits for these modes of operation for several congeners are also given in Table 14-1.

As is shown, detection limits for the more highly chlorinated are exceptionally low. Of particular interest are the detection limits for the nonachlorobiphenyls. These congeners are generally not observed in a quantifiable manner in an Aroclor 1242 sample; and yet, by monitoring the selected reaction m/z 466 ($M+4^{-}$) to m/z 447 in a chromatographic run, three (and only three) significant peaks are observed in quantities well above the detection limits. The quantities of the three nonachlorobiphenyls in an Aroclor 1242 standard were calculated to be 0.059% for 23456-2345-CB, 0.0066% for 23456-2346-CB, and 0.022% for 23456-2356-CB.

In Table 14-2 are listed the quantitative results of the six selected toxic congeners for the autoclaved and inoculated river sediment samples and for sediment samples taken from contaminated sites. As is shown, for all but one congener, ~ 80% dechlorination was observed. The advantage of this method is apparent from the fact that for two of these congeners (34-34-CB and 234-34-CB), quantitation is difficult by GC/MS due to the presence of co-eluting congeners (236-34-CB and 234-236-CB, respectively) in amounts one to two orders of magnitude greater. By using this selected reaction method, however, the co-elutates do not present an interference (see Figure 14-3).

SUMMARY AND CONCLUSIONS

Using a selected reaction monitoring mode of operation in a GC/MS/MS experiment and monitoring the exchange reaction of oxygen for chlorine, PCB congeners are able to be quantitated even when not chromatographically resolved. This method couples the high specificity of MS/MS with the excellent detection limits of selected-ion monitoring coupled with

TABLE 14-2. Results of analysis of PCB samples for the toxic congeners. See text for explanation of samples. Values listed are amounts in mg/L and percents of total PCBs.

CB	34-34	234-34	245-34	345-34	2345-34	345-345	
H7:AUTO	0.29	0.86	0.67	0.006	0.17	0.003	mg / L
H7:LIVE	0.00	0.12	0.14	0.001	0.009	0.00	mg / L
HUDSON RIVER	0.17	0.14	0.72	0.001	0.18	0.00	mg / L
	0.10	0.08	0.42	0.001	0.11	0.00	%
SILVER LAKE	0.19	0.25	2.20	0.048	7.00	0.069	mg / L
	0.04	0.02	0.16	0.003	0.50	0.005	%
H7:JUNE	1.5	0.65	1.7	0.037	0.35	0.010	mg / L
	0.6	0.26	0.68	0.015	0.14	0.004	%

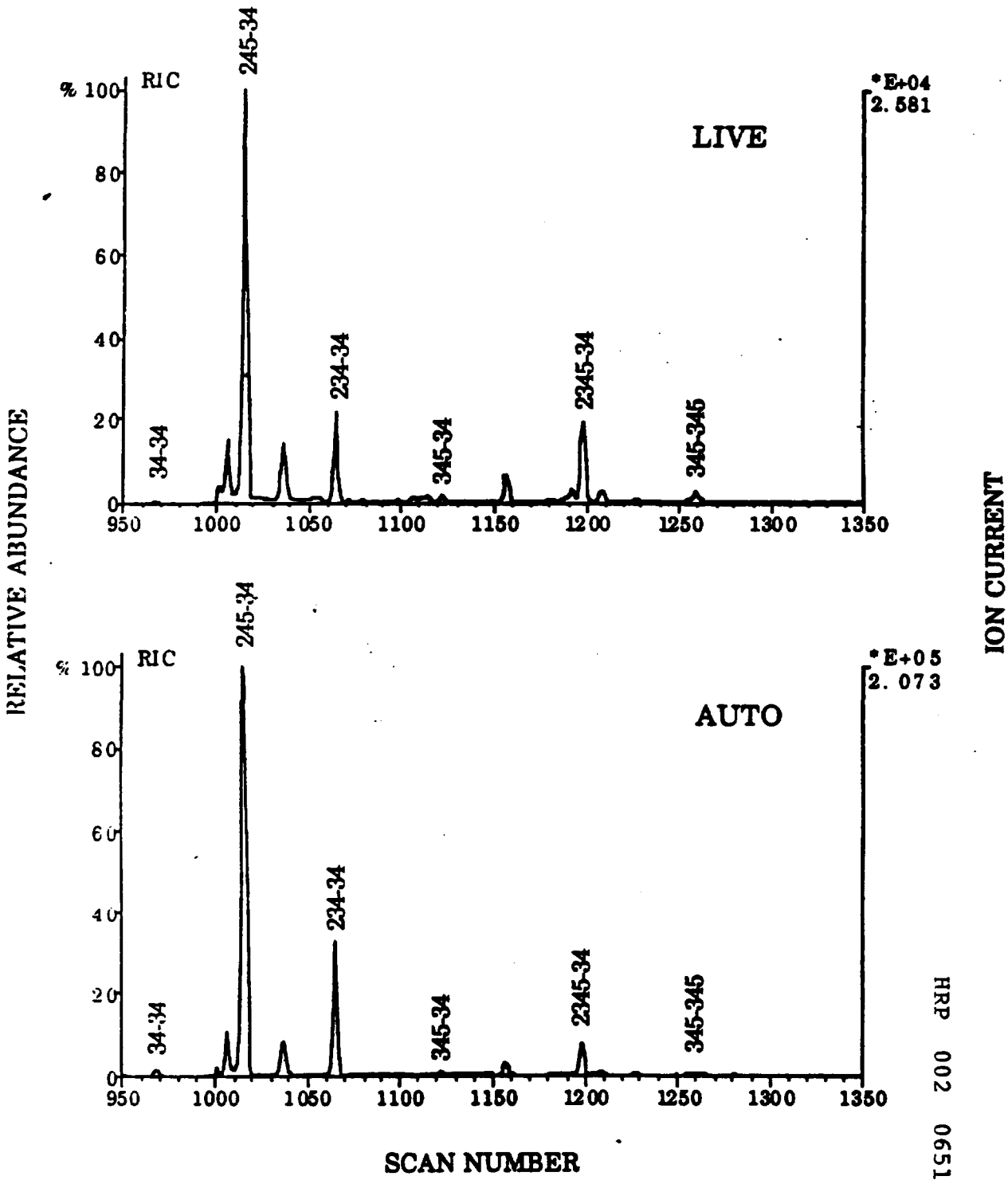

Figure 14-3. The total ion current chromatograms generated by monitoring the selected reactions for the toxic congeners (see text) in the analysis of two river sediment samples spiked with Aroclor 1242 with one inoculated (Live) with bacteria from the H7 site under anaerobic conditions and the other autoclaved (Auto). Note the difference in intensities for the two chromatograms.

negative chemical ionization. For the more highly chlorinated congeners, detection limits are even lower than that of an ECD. Due to the highly selective nature of this technique, individual PCB congeners may be quantitated in chromatographic analysis times of 30 minutes or less.

FUTURE WORK

1. Quantitation. The concentrations of individual congeners within a standard Aroclor mixture need to be determined. In addition, the precision and accuracy of the method need to be defined.
2. Internal Standards. The use of internal standards in conjunction with, or in addition to, octachloronaphthalene needs to be explored.
3. Instrumental Modifications. The instrument (TSQ-70B) needs to be modified in order to more accurately control the gas flow into the source and collision cell.
4. Other Oxidants. Reactants other than oxygen may prove to be useful for this method.
5. Other Aroclors. This method will be used for other Aroclors such as 1016, 1254, and 1260.
6. Other PCB Reactions. It would be desirable to explore other reactions of PCB ions in MS/MS experiments which could be used to distinguish isomers of the same mass.
7. Other Compounds in PCB Samples. This method will be used to detect and quantitate other chlorinated species such as PCDDs and PCDFs in PCB containing samples.

Chapter 15

RESEARCH PLANS FOR 1990-1991

A sequential biological process combining anaerobic reductive dechlorination and aerobic oxidation continues to show great promise for degrading even highly chlorinated Aroclor mixtures. Now that this process has been demonstrated in the laboratory, the challenge remains to apply it in the field under real environmental conditions. Future research plans are focused on gaining a greater understanding of these complementary modes of PCB biotransformation and the factors which influence their activity in the environment.

Anaerobic

The characterization and isolation of single strains or consortia of PCB-dechlorinating cultures remains as a significant goal in this research. Other areas of interest include further identification of consortia capable of *ortho* dechlorination and additional quantitation of the toxicity reduction in dechlorinated PCB mixtures. Research to examine the factors which influence the rate and extent of the dechlorination process on endogenous PCBs in different soils and sediments will continue, with an increased emphasis on bioavailability issues. Efforts to scale up the anaerobic process in anticipation of *in situ* application of this technology will increase.

Aerobic

The potential advantages of recombinant PCB-degrading strains over natural isolates will continue to be explored in the laboratory. These efforts include sequencing the *bph* genes and overexpression of these genes in the recombinants. Further characterization of the aerobic degradative pathway via both chemical and biological routes will also continue. Finally, laboratory aerobic degradation results using natural isolates will be extended and scaled up. The results of this work will be applied toward developing an aerobic process to degrade the endogenous PCBs found in extensively dechlorinated sediments.

Other

Efforts in the analytical area will include improving the precision and accuracy of the GC/MS/MS method and extending the method to detect other chlorinated species, such as PCDDs and PCDFs. Other nonbiological methods of removing PCBs from soils, such as radio frequency heating, will continue to be explored in the coming year.

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I. Reference Materials for Section 6.0

1. Tables for Section 6.2

a. Tables 6.2-1 and 2	Data Collected by Harza Engineering Co. (1988-90); Sample location sites.
b. Table 6.2.1-1	Bopp et al. (1982) Cs-137/PCB Core Data.
c. Table 6.2.1-2	Annual PCB Environmental Load by Aroclor Type. Versar, at 288 (1976).
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2. Tables for Section 6.3

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3. Tables for Section 6.4

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Sampling Station	Location	Date Sampled	Norza ID	NEA ID	Aroclor 1221 ug/g	Aroclor 1221 (%)	Aroclor 1242 ug/g	Aroclor 1242 (%)	Aroclor 1254 ug/g	Aroclor 1254 (%)	Aroclor 1260 ug/g	Aroclor 1260 (%)	Aroclor 1268 ug/g	Aroclor 1268 (%)	Total PCB
HR-1	Troy Dam	6/1/90	2365	90000899	0.1239	32.5	0.2244	58.9	0.0282	7.4	0.0041	1.1	0.0007	0.2	1.86
HR-1	Troy Dam	6/1/90	2366	90000900	0.0645	15.5	0.3216	77.2	0.0274	6.6	0.0029	0.7	0.0003	0.1	2.16
HR-1	Troy Dam	10/19/90	5665	90002320	0.0245	17.9	0.0582	42.6	0.0309	22.6	0.0195	14.3	0.0036	2.6	1.21
HR-1	Troy Dam	10/19/90	5666	90002321	0.0387	14.2	0.2121	78.1	0.0181	6.7	0.0025	0.9	0.0002	0.1	1.36
HR-1	Troy Dam	9/25/89	269T	89000608	0.0138	12.2	0.0811	71.8	0.0131	11.6	0.0041	3.6	0.0009	0.8	0.75
HR-1	Troy Dam	9/25/89	270T	89000609	0.0019	7.6	0.0192	78.7	0.0022	9.2	0.0009	3.8	0.0002	0.7	0.15
HR-1	Troy Dam	7/25/88	HR1S	90000411	0.0419	10.9	0.2819	73.2	0.0477	12.4	0.0121	3.1	0.0017	0.4	2.10
HR-1	Troy Dam	9/25/89	271T	89000610	0.0000	0.0	0.0067	84.9	0.0008	10.6	0.0004	4.6	0.0000	0.0	0.06
HR-2	Watervliet	9/25/89	266T	89000611	0.0140	13.1	0.0762	71.3	0.0108	10.1	0.0048	4.5	0.0011	1.0	0.72
HR-2	Watervliet	9/25/89	267T	89000612	0.0092	11.1	0.0563	67.4	0.0130	15.6	0.0043	5.2	0.0007	0.8	0.51
HR-2	Watervliet	9/25/89	268T	89000613	0.0928	20.1	0.3308	71.7	0.0324	7.0	0.0049	1.1	0.0008	0.2	2.64
HR-3	Albany	7/25/88	HR3S	90000541	0.0036	14.5	0.0173	68.8	0.0029	11.7	0.0010	4.2	0.0002	0.8	0.14
HR-4	North Albany Turning Basin	7/26/88	HR4S	90000544	0.0373	16.2	0.1529	66.4	0.0271	11.8	0.0108	4.7	0.0022	0.9	1.32
HR-5	South Albany Turning Basin	9/25/89	211	89000614	0.0401	15.7	0.1717	67.3	0.0304	11.9	0.0107	4.2	0.0023	0.9	1.45
HR-5	South Albany Turning Basin	9/25/89	212	89000616	0.0705	26.2	0.1670	62.0	0.0251	9.3	0.0057	2.1	0.0011	0.4	1.47
HR-5	South Albany Turning Basin	9/25/89	214	89000615	0.0440	17.1	0.1916	74.6	0.0162	6.3	0.0043	1.7	0.0009	0.3	1.58
HR-5	South Albany Turning Basin	9/25/89	215	89000617	0.0126	12.7	0.0657	66.3	0.0116	11.7	0.0075	7.6	0.0016	1.6	0.66
HR-5	South Albany Turning Basin	9/25/89	216	89000618	0.0000	0.0	0.0087	74.1	0.0020	16.8	0.0009	7.8	0.0001	1.2	0.07
HR-5	South Albany Turning Basin	9/25/89	259T	89000619	0.0138	12.0	0.0755	65.8	0.0137	12.0	0.0096	8.3	0.0022	1.9	0.80
HR-5	South Albany Turning Basin	9/25/89	260T	89000620	0.0102	3.2	0.2632	83.3	0.0325	10.3	0.0083	2.6	0.0018	0.6	1.89
HR-5	South Albany Turning Basin	9/25/89	261T	89000621	0.0434	14.7	0.2006	68.1	0.0298	10.1	0.0171	5.8	0.0036	1.2	1.66
HR-5	South Albany Turning Basin	9/25/89	262T	89000622	0.0457	7.2	0.4982	78.3	0.0710	11.2	0.0186	2.9	0.0025	0.4	3.82
HR-5	South Albany Turning Basin	9/25/89	263T	89000623	0.0496	19.0	0.1730	66.4	0.0296	11.4	0.0071	2.7	0.0014	0.5	1.47
HR-5	South Albany Turning Basin	9/25/89	264T	89000624	0.0205	9.8	0.1557	74.2	0.0186	8.8	0.0125	6.0	0.0024	1.1	1.21
HR-6	Campbell Island	7/26/88	HR6S	90000416	0.0041	9.7	0.0269	63.3	0.0099	23.2	0.0014	3.3	0.0002	0.5	0.24
HR-7	Ravena	7/27/88	HR7S	90000402	0.0000	0.0	0.0136	84.8	0.0013	7.9	0.0011	6.6	0.0001	0.7	0.07
HR-8	Coxsackie	9/19/89	169	89000598	0.0098	13.2	0.0261	35.0	0.0095	12.7	0.0241	32.4	0.0050	6.7	0.75
HR-8	Coxsackie	9/19/89	170	89000599	0.0102	16.7	0.0448	73.0	0.0050	8.2	0.0011	1.8	0.0003	0.3	0.36
HR-8	Coxsackie	9/19/89	171	89000600	0.0058	15.8	0.0224	61.3	0.0062	17.0	0.0018	5.0	0.0003	0.8	0.29
HR-8	Coxsackie	9/19/89	172	89000601	0.0655	15.4	0.3130	73.4	0.0392	9.2	0.0072	1.7	0.0015	0.3	2.14
HR-8	Coxsackie	9/19/89	173	89000602	0.0230	12.9	0.1285	72.2	0.0179	10.1	0.0070	3.9	0.0015	0.8	1.16
HR-8	Coxsackie	9/19/89	174	89000603	0.0190	16.5	0.0851	73.6	0.0073	6.3	0.0035	3.0	0.0007	0.6	0.71
HR-8	Coxsackie	9/19/89	175	89000604	0.0021	12.8	0.0121	74.4	0.0015	9.2	0.0005	3.1	0.0001	0.4	0.11
HR-8	Coxsackie	9/19/89	176	89000605	0.0070	12.3	0.0376	66.0	0.0058	10.2	0.0054	9.4	0.0012	2.0	0.37
HR-8	Coxsackie	9/19/89	177	89000606	0.0029	1.2	0.2046	85.4	0.0259	10.8	0.0052	2.2	0.0009	0.4	1.54
HR-8	Coxsackie	9/19/89	178	89000607	0.0020	10.2	0.0148	76.7	0.0018	9.3	0.0007	3.4	0.0001	0.5	0.12
HR-9	Stockport Creek	7/27/88													
HR-10	Hudson-Athens	7/27/88	HR10S	90000422	0.0037	7.2	0.0398	77.9	0.0048	9.4	0.0019	3.8	0.0009	1.7	0.29
HR-11	Catskill Creek	7/28/88	HR11S	90000539	0.0068	5.5	0.0999	80.9	0.012	9.7	0.0039	3.2	0.0009	0.7	0.63

Table 6.2-1

Sampling Station	Location	Date Sampled	Nerza ID	NEA ID	- Aroclor 1221 - ug/g	(%)	- Aroclor 1242 - ug/g	(%)	- Aroclor 1254 - ug/g	(%)	- Aroclor 1260 - ug/g	(%)	- Aroclor 1268 - ug/g	(%)	Total PCB
HR-12	Inbocht Bay	7/28/88	HR12S	90000277	0.0023	7.0	0.0265	81.4	0.0028	8.5	0.0008	2.5	0.0002	0.5	0.17
HR-13	Esopus Creek	7/28/88	HR13S	90000274	0.0071	4.9	0.1185	81.9	0.0137	9.5	0.0044	3.0	0.0010	0.7	0.75
HR-14	South Bay-Annandale	7/28/88	HR14S	90000540	0.0063	3	0.1806	85.4	0.019	9	0.0047	2.2	0.0009	0.4	1.12
HR-15	Kingston	9/14/89	153	89000589	0.0046	4.2	0.0889	81.8	0.0111	10.2	0.0034	3.1	0.0007	0.6	0.66
HR-15	Kingston	9/14/89	155	89000590	0.0031	1.3	0.1801	78.2	0.0367	15.9	0.0088	3.8	0.0017	0.7	1.41
HR-15	Kingston	9/14/89	156	89000591	0.0109	1.8	0.5204	83.7	0.0762	12.3	0.0119	1.9	0.0021	0.3	3.27
HR-15	Kingston	9/14/89	158	89000592	0.0048	6.2	0.0624	81.0	0.0074	9.5	0.0021	2.7	0.0004	0.5	0.47
HR-15	Kingston	9/14/89	159	89000593	0.0098	8.0	0.0950	77.0	0.0114	9.2	0.0059	4.8	0.0012	1.0	0.74
HR-15	Kingston	9/14/89	160	89000594	0.0000	0.0	0.0199	78.6	0.0043	16.9	0.0012	4.5	0.0000	0.0	0.14
HR-15	Kingston	9/14/89	161	89000595	0.0000	0.0	0.0065	42.8	0.0068	44.5	0.0018	11.8	0.0001	0.9	0.13
HR-15	Kingston	9/14/89	162	89000596	0.0000	0.0	0.0705	81.3	0.0133	15.4	0.0026	3.0	0.0003	0.3	0.49
HR-15	Kingston	9/14/89	168	89000597	0.0036	2.9	0.1067	86.0	0.0106	8.6	0.0026	2.1	0.0005	0.4	0.70
HR-16	Esopus Meadows Point	7/29/88	HR16S	90000423	0.0047	1.9	0.1995	81.8	0.0325	13.3	0.0061	2.5	0.0010	0.4	1.30
HR-17	Hyde Park	10/4/89	315T	89000679	0.0107	10.0	0.0834	77.5	0.0094	8.7	0.0034	3.2	0.0007	0.7	0.64
HR-17	Hyde Park	10/4/89	316T	89000680	0.0094	7.6	0.0985	79.7	0.0108	8.8	0.0039	3.2	0.0010	0.8	0.72
HR-17	Hyde Park	10/4/89	317T	89000681	0.0084	8.1	0.0815	78.3	0.0098	9.4	0.0036	3.5	0.0008	0.8	0.62
HR-17	Hyde Park	10/4/89	318T	89000682	0.0000	0.0	0.1536	86.4	0.0205	11.5	0.0033	1.8	0.0004	0.2	0.87
HR-17	Hyde Park	10/4/89	319T	89000683	0.0000	0.0	0.0007	82.1	0.0001	9.5	0.0001	8.3	0.0000	0.0	0.01
HR-17	Hyde Park	10/4/89	320T	89000684	0.0037	3.8	0.0818	85.2	0.0077	8.0	0.0023	2.4	0.0005	0.6	0.59
HR-17	Hyde Park	10/4/89	321	89000685	0.0099	7.2	0.1087	79.8	0.0123	9.1	0.0044	3.2	0.0010	0.7	0.81
HR-17	Hyde Park	10/4/89	322	89000686	0.0000	0.0	1.0350	90.8	0.0828	7.3	0.0188	1.6	0.0037	0.3	6.51
HR-17	Hyde Park	10/4/89	323	89000687	0.0063	6.0	0.0843	81.4	0.0089	8.6	0.0033	3.2	0.0007	0.7	0.61
HR-17	Hyde Park	10/4/89	324	89000688	0.0124	5.5	0.1855	82.1	0.0190	8.4	0.0073	3.2	0.0017	0.7	1.36
HR-18	North Poughkeepsie	7/29/88	HR18S	90000275	0.0149	5.5	0.1898	70.2	0.0413	15.3	0.0204	7.5	0.0039	1.4	1.48
HR-19	Poughkeepsie	10/4/89	305T	89000669	0.0064	6.7	0.0757	79.5	0.0091	9.6	0.0033	3.5	0.0007	0.7	0.58
HR-19	Poughkeepsie	10/4/89	306T	89000670	0.0083	7.6	0.0867	78.9	0.0103	9.4	0.0038	3.5	0.0008	0.7	0.65
HR-19	Poughkeepsie	10/4/89	307T	89000671	0.0093	7.4	0.0985	78.1	0.0127	10.1	0.0046	3.6	0.0010	0.8	0.80
HR-19	Poughkeepsie	10/4/89	308T	89000672	0.0090	7.3	0.0982	79.7	0.0113	9.2	0.0039	3.2	0.0007	0.6	0.73
HR-19	Poughkeepsie	10/4/89	309T	89000673	0.0093	5.4	0.1395	80.9	0.0162	9.4	0.0061	3.5	0.0014	0.8	1.00
HR-19	Poughkeepsie	10/4/89	310T	89000674	0.0085	5.6	0.1180	77.3	0.0203	13.3	0.0047	3.1	0.0010	0.7	0.83
HR-19	Poughkeepsie	10/4/89	311T	89000675	0.0146	1.8	0.6733	81.2	0.1159	14.0	0.0224	2.7	0.0034	0.4	4.72
HR-19	Poughkeepsie	10/4/89	312T	89000676	0.0000	0.0	0.4694	87.3	0.0510	9.5	0.0144	2.7	0.0028	0.5	3.29
HR-19	Poughkeepsie	10/4/89	313T	89000677	0.0080	6.9	0.0895	77.1	0.0113	9.8	0.0058	5.0	0.0014	1.2	0.70
HR-19	Poughkeepsie	10/4/89	314T	89000678	0.0095	7.0	0.1086	80.0	0.0115	8.5	0.0051	3.7	0.0010	0.7	0.76
HR-20	New Hamburg	7/30/88	HR20S	90000425	0.0018	3.3	0.0433	78.8	0.0082	15.0	0.0014	2.6	0.0002	0.3	0.28
HR-21	Newburgh	7/30/88	HR21S	90000391	0.0050	3.6	0.1030	74.7	0.0235	17.0	0.0055	4.0	0.0010	0.7	0.79
HR-22	Cornwall on Hudson	7/30/88	HR22S	90000427	0.0000	0.0	0.0114	88.3	0.0009	7.3	0.0006	4.4	0.0000	0.0	0.05
HR-23	Foundry Cove	7/30/88	HR23S	90000420	0.0106	3.1	0.2459	73.0	0.0637	18.9	0.0142	4.2	0.0022	0.6	1.84
HR-24	Con Hook	8/25/89	258	89000535	0.0071	1.3	0.4477	81.8	0.0705	12.9	0.0183	3.3	0.0038	0.7	2.93

6.2-1 (cont.)

Sampling Station	Location	Date Sampled	Narga ID	NEA ID	- Aroclor 1221 - ug/g	(%)	- Aroclor 1242 - ug/g	(%)	- Aroclor 1254 - ug/g	(%)	- Aroclor 1260 - ug/g	(%)	- Aroclor 1268 - ug/g	(%)	Total PCB
HR-24	Con Hook	8/25/89	397	89000530	0.0057	2.3	0.2063	83.0	0.0308	12.4	0.0046	1.9	0.0012	0.5	1.30
HR-24	Con Hook	8/25/89	398	89000531	0.0000	0.0	0.2864	84.5	0.0386	11.4	0.0115	3.4	0.0025	0.7	1.86
HR-24	Con Hook	8/25/89	399	89000532	0.0000	0.0	0.0239	21.4	0.0093	8.3	0.0609	54.5	0.0176	15.8	0.65
HR-24	Con Hook	8/25/89	400	89000533	0.0000	0.0	0.0062	71.8	0.0020	22.9	0.0004	4.3	0.0001	0.9	0.05
HR-24	Con Hook	8/25/89	257	89000534	0.0000	0.0	0.0633	85.1	0.0089	11.9	0.0019	2.5	0.0003	0.4	0.37
HR-25	Iona Island	7/29/88	HR25S	90000414	0.0000	0.0	0.2788	82.4	0.0457	13.5	0.0119	3.5	0.0021	0.6	1.75
HR-26	Peekskill Bay	7/29/88	HR26S	90000555	0.0000	0.0	0.0590	73.8	0.0158	19.8	0.0041	5.2	0.0009	1.2	0.46
HR-27	Indian Point	7/29/88	HR27S	90000554	0.0022	2.7	0.0630	76.4	0.0133	16.1	0.0032	3.9	0.0008	0.9	0.42
HR-28	Stony Point Bay	8/25/89	393	89000504	0.0078	2.1	0.2755	75.8	0.0621	17.1	0.0146	4.0	0.0036	1.0	1.94
HR-28	Stony Point Bay	8/25/89	394	89000505	0.0000	0.0	0.0660	49.6	0.0470	35.3	0.0172	12.9	0.0030	2.2	1.17
HR-28	Stony Point Bay	8/25/89	395	89000506	0.0120	3.6	0.2977	76.8	0.0513	15.3	0.0125	3.7	0.0022	0.7	1.81
HR-28	Stony Point Bay	8/25/89	408	89000503	0.0000	0.0	0.0065	78.2	0.0015	17.6	0.0003	4.2	0.0000	0.0	0.05
HR-28	Stony Point Bay	8/25/89	259	89000536	0.0087	4.0	0.1762	80.3	0.0271	12.3	0.0058	2.6	0.0017	0.8	1.20
HR-28	Stony Point Bay	8/25/89	260	89000515	0.0000	0.0	0.2590	83.5	0.0395	12.7	0.0100	3.2	0.0019	0.6	1.70
HR-28	Stony Point Bay	8/25/89	261	89000516	0.0087	3.7	0.1834	78.3	0.0328	14.0	0.0077	3.3	0.0016	0.7	1.19
HR-28	Stony Point Bay	8/25/89	262	89000517	0.0000	0.0	0.0519	81.3	0.0093	14.6	0.0023	3.6	0.0004	0.6	0.29
HR-28	Stony Point Bay	8/25/89	263	89000518	0.0000	0.0	0.1416	80.9	0.0255	14.6	0.0065	3.7	0.0014	0.8	0.94
HR-28	Stony Point Bay	8/25/89	264	89000519	0.0133	3.4	0.3186	81.6	0.0468	12.0	0.0097	2.5	0.0020	0.5	1.95
HR-29	Haverstraw	7/29/88	HR29S	90000546	0.0054	1.3	0.3192	74.2	0.0858	19.9	0.0170	4.0	0.0003	0.7	2.44
HR-30	Croton Bay	7/27/88	HR30S	90000285	0.0065	5.0	0.0931	72.4	0.0231	18.0	0.0049	3.8	0.0010	0.8	0.71
HR-31	Tarrytown	7/27/88	HR31S	90000276	0.0040	2.9	0.0988	73.9	0.0229	17.0	0.0070	5.2	0.0014	1.0	0.74
HR-32	Tappan Zee Bridge - Nyack	7/27/88	HR32S	90000545	0.0090	2.5	0.2652	73.1	0.0704	19.4	0.0155	4.3	0.0028	0.8	2.10
HR-33	Piermont	7/27/88	HR33S	90000542	0.0027	3.4	0.0590	74.5	0.0107	13.5	0.0051	6.4	0.0018	2.3	0.49
HR-34	Yonkers	7/28/88	HR34S	90000282	0.0083	3.6	0.1649	71.2	0.0408	17.6	0.0144	6.2	0.0032	1.4	1.27
HR-35	Spuyten Devvil Creek	7/28/88	HR35S	90000279	0.0088	2.6	0.2364	68.5	0.0545	15.8	0.0358	10.4	0.0093	2.7	1.96
HR-36	George Washington Bridge	7/28/88	HR36S	90000401	0.0105	4.7	0.1520	68.5	0.0411	18.5	0.0155	7.0	0.0030	1.4	1.27
HR-37	North Bergen	7/28/88	HR37S	90000417	0.0077	2.4	0.2327	72.0	0.0580	17.9	0.0204	6.3	0.0045	1.4	1.86
HR-38	The Battery	8/19/89	285	89000309	0.0049	5.4	0.0599	65.7	0.0149	16.4	0.0089	9.8	0.0025	2.8	0.66
HR-38	The Battery	8/19/89	286	89000310	0.0000	0.0	0.1383	63.9	0.0479	22.2	0.0236	10.9	0.0064	3.0	1.42
HR-38	The Battery	8/19/89	287	89000311	0.0000	0.0	0.2050	74.3	0.0501	18.2	0.0168	6.1	0.0038	1.4	1.61
HR-38	The Battery	8/19/89	288	89000312	0.0000	0.0	0.0103	32.1	0.0079	24.5	0.0095	29.5	0.0045	13.9	0.50
HR-38	The Battery	8/19/89	289	89000313	0.0000	0.0	0.2908	73.4	0.0790	19.9	0.0221	5.6	0.0043	1.1	2.55
HR-38	The Battery	8/19/89	290	89000314	0.0000	0.0	0.0137	63.9	0.0055	25.7	0.0019	9.1	0.0003	1.3	0.15
HR-38	The Battery	8/19/89	291	89000315	0.0000	0.0	0.0370	47.8	0.0187	24.1	0.0172	22.2	0.0046	5.9	0.66
HR-38	The Battery	8/19/89	292	89000520	0.0000	0.0	0.0320	50.2	0.0218	34.1	0.0088	13.7	0.0013	2.0	0.42
HR-38	The Battery	8/21/89	293	89000525	0.0000	0.0	0.1015	69.2	0.0284	19.3	0.0139	9.5	0.0030	2.1	0.93
HR-38	The Battery	8/21/89	294	89000526	0.0000	0.0	0.1613	75.0	0.0344	16.0	0.0158	7.4	0.0034	1.6	1.26
NYH-1	Bayonne	7/28/88	NYH-1S	90000281	0.0176	12.9	0.0527	38.6	0.0288	21.1	0.0293	21.5	0.0081	5.9	0.88

6.2-1 (cont.)

Sampling Station	Location	Date Sampled	Norza ID	NEA ID	- Aroclor 1221 - ug/g	(%)	- Aroclor 1242 - ug/g	(%)	- Aroclor 1254 - ug/g	(%)	- Aroclor 1260 - ug/g	(%)	- Aroclor 1268 - ug/g	(%)	Total PCB
NYH-2	Gowanus Canal	8/21/89	441	89000507	0.0000	0.0	0.0535	54.8	0.0260	25.6	0.0169	16.6	0.0029	2.9	0.72
NYH-2	Gowanus Canal	8/21/89	443	89000509	0.0000	0.0	0.1112	43.9	0.0893	35.2	0.0421	16.6	0.0110	4.3	1.93
NYH-2	Gowanus Canal	8/21/89	444	89000510	0.0000	0.0	0.4117	65.6	0.1496	23.8	0.0564	9.0	0.0101	1.6	3.97
NYH-2	Gowanus Canal	8/21/89	445	89000511	0.1217	2.6	2.8109	59.8	1.1934	25.5	0.4878	10.4	0.0810	1.7	39.90
NYH-2	Gowanus Canal	8/21/89	446	89000512	0.0000	0.0	0.2176	56.9	0.0882	23.1	0.0660	17.2	0.0109	2.8	2.60
NYH-2	Gowanus Canal	8/21/89	447	89000513	0.0000	0.0	0.1396	53.7	0.0681	26.2	0.0440	16.9	0.0082	3.2	1.99
NYH-2	Gowanus Canal	8/21/89	448	89000514	0.0000	0.0	0.0429	63.2	0.0142	20.9	0.0089	13.2	0.0018	2.7	0.46
NYH-2	Gowanus Canal	8/21/89	449	89000521	0.0000	0.0	0.0832	64.8	0.0272	21.2	0.0150	11.7	0.0030	2.3	0.84
NYH-3	Verrazano Narrows Bridge	8/23/89	426	89000556	0.0000	0.0	0.0578	61.1	0.0227	24.1	0.0117	12.4	0.0022	2.4	0.62
NYH-3	Verrazano Narrows Bridge	8/23/89	427	89000557	0.0000	0.0	0.0338	55.8	0.0175	29.0	0.0079	13.1	0.0013	2.1	0.43
NYH-3	Verrazano Narrows Bridge	8/23/89	429	89000559	0.0000	0.0	0.0328	27.2	0.0748	62.0	0.0111	9.2	0.0019	1.6	1.14
NYH-3	Verrazano Narrows Bridge	8/23/89	430	89000560	0.0000	0.0	0.0317	64.5	0.0116	23.7	0.0050	10.1	0.0008	1.7	0.32
NYH-3	Verrazano Narrows Bridge	8/23/89	433	89000474	0.0000	0.0	0.0384	57.3	0.0203	30.3	0.0071	10.6	0.0012	1.9	0.53
NYH-3	Verrazano Narrows Bridge	8/23/89	435	89000476	0.0067	1.6	0.2419	59.4	0.0822	20.2	0.0653	16.0	0.0111	2.7	2.79
NYH-3	Verrazano Narrows Bridge	8/23/89	438	89000478	0.0000	0.0	0.0469	63.1	0.0176	23.6	0.0084	11.3	0.0015	2.0	0.53
NYH-3	Verrazano Narrows Bridge	8/23/89	439	89000553	0.0000	0.0	0.0152	49.2	0.0058	18.7	0.0078	25.3	0.0021	6.8	0.21
NYH-3	Verrazano Narrows Bridge	8/23/89	440	89000554	0.0000	0.0	0.1050	65.1	0.0349	21.6	0.0177	11.0	0.0037	2.3	1.09
NYH-4	Midland Beach	8/23/90	401	89000466	0.0000	0.0	0.0542	59.3	0.0234	25.6	0.0115	12.6	0.0023	2.5	0.65
NYH-4	Midland Beach	8/23/89	403	89000467	0.0000	0.0	0.0644	59.2	0.0286	26.2	0.0134	12.3	0.0025	2.3	0.77
NYH-4	Midland Beach	8/23/89	406	89000470	0.0000	0.0	0.0398	59.2	0.0169	25.1	0.0089	13.3	0.0017	2.5	0.49
NYH-4	Midland Beach	8/23/89	431	89000463	0.0000	0.0	0.0648	62.9	0.0239	23.2	0.0119	11.5	0.0024	2.3	0.68
NYH-4	Midland Beach	8/23/89	432	89000465	0.0000	0.0	0.0749	61.2	0.0030	24.5	0.0147	12.0	0.0029	2.3	0.89
NYH-4	Midland Beach	8/21/89	451	89000523	0.0000	0.0	0.0032	58.2	0.0013	24.5	0.0009	17.2	0.0000	0.0	0.04
NYH-5	Coney Island	8/23/88	NYH5S	90000437	0.0000	0.0	0.0438	55.7	0.0230	29.3	0.0086	10.9	0.0032	4.1	0.52
NYH-6	Great Kills Harbor	8/16/89	305	89000320	0.0000	0.0	0.0071	56.9	0.0039	31.5	0.0013	10.6	0.0001	1.0	0.10
NYH-6	Great Kills Harbor	8/16/89	306	89000321	0.0000	0.0	0.0252	64.4	0.0088	22.6	0.0043	11.1	0.0008	2.0	0.26
NYH-6	Great Kills Harbor	8/16/89	313	89000328	0.0000	0.0	0.0423	62.0	0.0167	24.5	0.0078	11.4	0.0014	2.1	0.45
NYH-6	Great Kills Harbor	8/16/89	314	89000329	0.0000	0.0	0.0526	63.1	0.0199	23.8	0.0093	11.1	0.0016	2.0	0.56
NYH-6	Great Kills Harbor	8/16/89	315	89000330	0.0000	0.0	0.0486	61.3	0.0208	26.3	0.0081	10.2	0.0018	2.3	0.54
NYH-6	Great Kills Harbor	8/16/89	316	89000331	0.0000	0.0	0.0043	58.2	0.0020	26.7	0.0010	13.3	0.0001	1.9	0.05
NYH-6	Great Kills Harbor	8/16/89	318	89000333	0.0000	0.0	0.0418	65.3	0.0145	22.6	0.0065	10.2	0.0012	1.9	0.42
NYH-6	Great Kills Harbor	8/16/89	319	89000334	0.0000	0.0	0.0045	69.5	0.0011	17.0	0.0007	11.2	0.0002	2.3	0.04
NYH-7	Seguine Point	8/30/88	NYH7S	90000385	0.0000	0.0	0.0062	73.7	0.0016	19.7	0.0005	6.6	0.0000	0.0	0.06
NYH-8	Raritan River mouth	8/16/89	297	89000300	0.0000	0.0	0.0061	43.3	0.0055	39.2	0.0020	14.1	0.0005	3.4	0.14
NYH-8	Raritan River mouth	8/16/89	298	89000301	0.0000	0.0	0.0637	56.2	0.0324	28.6	0.0148	13.0	0.0025	2.2	0.92
NYH-8	Raritan River mouth	8/16/89	299	89000302	0.0000	0.0	0.0202	48.5	0.0133	32.0	0.0068	16.2	0.0014	3.3	0.43
NYH-8	Raritan River mouth	8/16/89	308	89000323	0.0000	0.0	0.0332	58.5	0.0150	26.4	0.0073	12.9	0.0012	2.2	0.44
NYH-8	Raritan River mouth	8/16/89	309	89000324	0.0000	0.0	0.0428	13.0	0.2100	63.7	0.0655	19.9	0.0116	3.5	3.15
NYH-8	Raritan River mouth	8/16/89	311	89000326	0.0000	0.0	0.0308	51.7	0.0186	31.2	0.0090	15.1	0.0012	2.0	0.45

6.2-1 (con't)

Sampling Station	Location	Date Sampled	Marza ID	NEA ID	- Aroclor 1221 - ug/g	(%)	- Aroclor 1242 - ug/g	(%)	- Aroclor 1254 - ug/g	(%)	- Aroclor 1260 - ug/g	(%)	- Aroclor 1268 - ug/g	(%)	Total PCB
NYH-8	Raritan River mouth	8/16/89	312	89000327	0.0000	0.0	0.0116	51.6	0.0065	28.7	0.0038	17.0	0.0006	2.7	0.23
NYH-9	Woodbridge Creek	8/18/89	276	89000293	0.0038	3.4	0.0617	55.6	0.0318	28.6	0.0116	10.4	0.0022	2.0	1.14
NYH-9	Woodbridge Creek	8/18/89	278	89000295	0.0000	0.0	0.0999	51.1	0.0658	33.7	0.0254	13.0	0.0044	2.2	1.51
NYH-9	Woodbridge Creek	8/18/89	279	89000296	0.0000	0.0	0.4550	64.1	0.1836	25.8	0.0612	8.6	0.0104	1.5	5.59
NYH-9	Woodbridge Creek	8/18/89	281	89000298	0.0000	0.0	0.0691	48.8	0.0457	32.3	0.0219	15.5	0.0048	3.4	1.12
NYH-9	Woodbridge Creek	8/18/89	282	89000299	0.0000	0.0	0.0586	45.7	0.0549	42.8	0.0127	9.9	0.0020	1.6	1.08
NYH-9	Woodbridge Creek	8/18/89	283	89000308	0.0000	0.0	0.0335	50.0	0.0232	34.7	0.0088	13.1	0.0015	2.2	0.55
NYH-10	Fresh Kills	8/23/88	NYH10S	90000412	0.0000	0.0	0.0301	46.7	0.0264	41.0	0.0068	10.6	0.0010	1.6	0.36
NYH-11	Pralls Island	9/19/88	NYH11S	90000407	0.0033	3.6	0.0634	68.2	0.0000	0.0	0.0216	23.2	0.0047	5.0	0.82
NYH-12	Newark Bay	8/18/89	265	89000305	0.0000	0.0	0.0144	53.3	0.0083	30.9	0.0037	13.9	0.0005	2.0	0.20
NYH-12	Newark Bay	8/18/89	266	89000306	0.0000	0.0	0.0287	54.9	0.0150	28.7	0.0072	13.8	0.0014	2.6	0.38
NYH-12	Newark Bay	8/18/89	267	89000307	0.0000	0.0	0.0417	57.6	0.0180	24.9	0.0098	13.6	0.0029	4.0	0.51
NYH-12	Newark Bay	8/18/89	268	89000284	0.0000	0.0	0.1102	57.0	0.0516	26.7	0.0267	13.8	0.0050	2.6	1.50
NYH-12	Newark Bay	8/18/89	269	89000285	0.0000	0.0	0.1041	59.4	0.0448	25.6	0.0220	12.6	0.0043	2.5	1.26
NYH-12	Newark Bay	8/18/89	270	89000286	0.0000	0.0	0.0586	56.6	0.0251	24.2	0.0165	15.9	0.0033	3.2	0.80
NYH-12	Newark Bay	8/18/89	271	89000287	0.0000	0.0	0.1022	58.1	0.0441	25.1	0.0244	13.9	0.0052	2.9	1.29
NYH-12	Newark Bay	8/18/89	273	89000288	0.0000	0.0	0.1042	55.9	0.0526	28.2	0.0249	13.4	0.0047	2.5	1.45
NYH-12	Newark Bay	8/18/89	274	89000289	0.0000	0.0	0.0713	61.0	0.0292	25.0	0.0139	11.9	0.0025	2.1	0.87
NYH-12	Newark Bay	8/18/89	275	89000291	0.0000	0.0	0.0584	55.3	0.0320	30.3	0.0128	12.1	0.0024	2.3	0.77
NYH-13	Wallabout Bay	8/25/88	NYH13S	90000399	0.0000	0.0	0.0593	56.1	0.0255	24.1	0.0170	16.1	0.0039	3.7	0.73
NYH-14	Newtown Creek	8/22/89	413	89000540	0.0000	0.0	0.1216	63.0	0.0444	23.0	0.0221	11.6	0.0048	2.5	1.26
NYH-14	Newtown Creek	8/22/89	414	89000541	0.0000	0.0	0.1063	58.7	0.0450	24.9	0.0248	13.7	0.0049	2.7	1.25
NYH-14	Newtown Creek	8/22/89	415	89000542	0.0000	0.0	0.0844	50.0	0.0356	21.1	0.0409	24.2	0.0080	4.7	1.11
NYH-14	Newtown Creek	8/22/89	416	89000543	0.0000	0.0	0.1906	58.5	0.0789	24.2	0.0490	15.0	0.0077	2.4	2.01
NYH-14	Newtown Creek	8/22/89	417	89000544	0.0000	0.0	0.1228	64.7	0.0428	22.5	0.0200	10.6	0.0041	2.2	1.27
NYH-14	Newtown Creek	8/22/89	423	89000471	0.0000	0.0	0.1163	58.7	0.0440	22.2	0.0323	16.3	0.0056	2.8	1.24
NYH-14	Newtown Creek	8/22/89	424	89000472	0.0083	8.9	0.0489	52.1	0.0246	26.2	0.0106	11.3	0.0015	1.6	0.69
NYH-14	Newtown Creek	8/22/89	452	89000527	0.0000	0.0	0.0868	63.3	0.0283	20.6	0.0180	13.1	0.0041	3.0	0.89
NYH-15	Wards Island	8/22/89	409	89000552	0.0000	0.0	0.0960	60.9	0.0387	24.6	0.0190	12.0	0.0039	2.5	1.08
NYH-15	Wards Island	8/22/89	410	89000537	0.0000	0.0	0.0129	72.2	0.0025	14.0	0.0020	11.3	0.0004	2.5	0.08
NYH-15	Wards Island	8/22/89	411	89000538	0.0045	2.0	0.1160	51.9	0.0632	28.3	0.0340	15.2	0.0058	2.6	1.82
NYH-15	Wards Island	8/22/89	412	89000539	0.0000	0.0	0.1381	67.1	0.0399	19.4	0.0228	11.1	0.0050	2.4	1.28
NYH-15	Wards Island	8/22/89	453	89000528	0.0000	0.0	0.1648	82.2	0.0194	9.7	0.0137	6.8	0.0027	1.3	1.32
NYH-15	Wards Island	8/22/89	454	89000549	0.0000	0.0	0.0030	38.3	0.0017	21.8	0.0028	35.5	0.0003	4.4	0.06
NYH-15	Wards Island	8/22/89	455	89000550	0.0000	0.0	0.0895	56.6	0.0373	23.6	0.0260	16.5	0.0052	3.3	0.97
NYH-15	Wards Island	8/22/89	456	89000551	0.0000	0.0	0.0485	53.0	0.0241	26.3	0.0155	17.0	0.0034	3.7	0.65
NYH-16	Harlem River	8/25/88	NYH16S	90000271	0.0062	3.2	0.1215	62.9	0.0415	21.5	0.0192	9.9	0.0048	2.5	1.14
NYH-17	College Point	8/25/88	NYH17S	90000424	0.0035	2.1	0.0772	46.0	0.0519	31.0	0.0294	17.6	0.0056	3.3	1.03
NYH-18	Throgs Neck Bridge	8/25/88	NYH18S	90000280	0.0000	0.0	0.0477	52.0	0.0259	28.3	0.0150	16.3	0.0031	3.3	0.61

6.2-1 (con't)

Sampling Station	Location	Date Sampled	Nerze ID	NEA ID	- Aroclor 1221 - ug/g	(%)	- Aroclor 1242 - ug/g	(%)	- Aroclor 1254 - ug/g	(%)	- Aroclor 1260 - ug/g	(%)	- Aroclor 1268 - ug/g	(%)	Total PCB
NYH-19	Sandy Hook	10/12/90	5014	90002062	0.002	4.3	0.0265	56.1	0.0114	24.2	0.006	12.6	0.0013	2.7	0.38
NYH-19	Sandy Hook	10/12/90	5013	90002049	0	0	0.0017	56.9	0.0008	26.7	0.0004	14.5	0.0001	3.9	0.03
NYH-19	Sandy Hook	10/12/90	5020	90002050	0	0	0.0369	58.8	0.015	23.9	0.0088	14	0.0021	3.3	0.53
NYH-19	Sandy Hook	10/12/90	5019	90002051	0	0	0.0031	61.4	0.001	19.1	0.0008	15.9	0.0002	3.5	0.04
NYH-19	Sandy Hook	10/12/90	5015	90002064	0	0	0.008	59.1	0.0034	25	0.0019	13.6	0.0003	2.3	0.1
NYH-20	Lower Bay														
NYH-21	Rockaway Inlet	10/30/90	5154	90002157	0	0	0.0044	62.5	0.0015	21.5	0.0009	13.3	0.0002	2.7	0.06
NYH-21	Rockaway Inlet	10/30/90	5156	90002159	0	0	0.0153	59	0.0049	19	0.0045	17.4	0.0012	4.5	0.21
NYH-21	Rockaway Inlet	10/30/90	5155	90002158	0	0	0.0302	60.2	0.0104	20.7	0.008	15.9	0.0016	3.1	0.41
NYH-21	Rockaway Inlet	10/30/90	5164	90002160	0	0	0.0375	57.4	0.0152	23.3	0.0105	16.1	0.0021	3.2	0.56
NYH-21	Rockaway Inlet	10/30/90	5165	90002161	0	0	0.0123	56.1	0.0058	26.7	0.0032	14.8	0.0005	2.4	0.18
NYH-21	Rockaway Inlet	10/30/90	5166	90002162	0	0	0.0132	54.5	0.0072	29.7	0.0033	13.6	0.0005	2.3	0.2
NYH-22	Jamaica Bay-Beach Channel	09/11/90	3699	90001581	0	0	0.0255	55.3	0.0103	22.4	0.0076	16.4	0.0028	6	0.35
NYH-22	Jamaica Bay-Beach Channel	09/10/90	3209	90001576	0	0	0.0198	52.8	0.0099	26.5	0.0064	17	0.0014	3.7	0.33
NYH-22	Jamaica Bay-Beach Channel	09/10/90	3210	90001577	0.0017	8.8	0.0099	50.2	0.0046	23.3	0.0028	14.4	0.0007	3.3	0.17
NYH-22	Jamaica Bay-Beach Channel	09/10/90	3211	90001578	0	0	0.0089	55.5	0.0038	24	0.0027	16.6	0.0006	3.9	0.14
NYH-22	Jamaica Bay-Beach Channel	09/10/90	3690	90001579	0	0	0.0124	53.7	0.0064	27.7	0.0035	15.2	0.0008	3.5	0.22
NYH-22	Jamaica Bay-Beach Channel	09/10/90	3691	90001580	0	0	0.0143	55.3	0.0059	22.8	0.0046	17.8	0.001	4.1	0.22
NYH-23	Jamaica Bay - Canarsie	10/16/90	5056	90002063	0	0	0.0044	38.1	0.0033	29	0.0032	27.9	0.0006	5	0.09
NYH-23	Jamaica Bay - Canarsie	10/16/90	5061	90002069	0	0	0.0183	55.8	0.0078	23.9	0.0055	16.8	0.0012	3.5	0.27
NYH-23	Jamaica Bay - Canarsie	10/16/90	5063	90002070	0	0	0.0054	57.7	0.002	20.9	0.0017	17.7	0.0003	3.7	0.08
NYH-23	Jamaica Bay - Canarsie	10/16/90	5059	90002072	0	0	0.0104	59.7	0.0033	18.8	0.0031	17.6	0.0007	3.9	0.15
NYH-23	Jamaica Bay - Canarsie	10/16/90	5060	90002065	0	0	0.0151	50.5	0.0081	27.2	0.0056	18.7	0.0011	3.6	0.26
NYH-23	Jamaica Bay - Canarsie	10/16/90	5062	90002068	0	0	0.0053	49.8	0.0024	22.7	0.0024	22.8	0.0005	4.8	0.09
NYH-23	Jamaica Bay - Canarsie	10/16/90	5064	90002071	0	0	0.0024	55.1	0.001	21.6	0.0009	19.4	0.0002	3.9	0.04
NYH-24	Harlem River - 207th St.	10/26/90	5151	90002060	0.0086	3.6	0.1799	74.6	0.034	14.1	0.0155	6.4	0.0032	1.3	1.42
NYH-24	Harlem River - 207th St.	10/26/90	5149	90002061	0.0091	4.5	0.1351	66.2	0.0383	18.8	0.0179	8.8	0.0037	1.8	1.41
NYH-24	Harlem River - 207th St.	10/26/90	5148	90002058	0	0	0.0721	58.7	0.0169	13.8	0.0276	22.5	0.0062	5.1	0.8
NYH-24	Harlem River - 207th St.	10/26/90	5150	90002059	0	0	0.0898	63.8	0.0281	19.9	0.0183	13	0.0047	3.3	0.98
NYH-24	Harlem River - 207th St.	10/26/90	5145	90002057	0	0	0.075	68.7	0.0155	14.3	0.0152	14	0.0034	3.1	0.69
NYH-24	Harlem River - 207th St.	10/26/90	5146	90002066	0.0000	0.0	0.1044	67.1	0.0264	17.0	0.0198	12.8	0.0049	3.1	0.98
NYH-24	Harlem River - 207th St.	10/26/90	5147	90002067	0	0	0.1118	69.7	0.0254	15.8	0.0183	11.4	0.005	3.1	0.96
LIS-1	Little Neck Bay	8/24/89	381	89000495	0.0000	0.0	0.0065	46.3	0.0042	30.2	0.0028	19.9	0.0005	3.6	0.10
LIS-1	Little Neck Bay	8/24/89	382	89000496	0.0000	0.0	0.0313	52.8	0.0159	26.9	0.0099	16.7	0.0021	3.6	0.46
LIS-1	Little Neck Bay	8/24/89	383	89000497	0.0000	0.0	0.0052	55.2	0.0025	26.9	0.0014	15.3	0.0003	2.7	0.06
LIS-1	Little Neck Bay	8/24/89	384	89000498	0.0000	0.0	0.0336	51.5	0.0194	29.7	0.0103	15.7	0.0021	3.1	0.41
LIS-1	Little Neck Bay	8/24/89	386	89000500	0.0000	0.0	0.0269	48.7	0.0146	26.5	0.0117	21.2	0.0020	3.6	0.36
LIS-1	Little Neck Bay	8/24/89	387	89000501	0.0000	0.0	0.0317	69.5	0.0099	21.6	0.0034	7.4	0.0007	1.5	0.33
LIS-1	Little Neck Bay	8/24/89	388	89000502	0.0000	0.0	0.0580	56.9	0.0267	26.2	0.0144	14.1	0.0029	2.8	0.69

6.2-1 (cont.)

Sampling Station	Location	Date Sampled	Narza ID	NEA ID	Aroclor 1221 ug/g	Aroclor 1221 (%)	Aroclor 1242 ug/g	Aroclor 1242 (%)	Aroclor 1254 ug/g	Aroclor 1254 (%)	Aroclor 1260 ug/g	Aroclor 1260 (%)	Aroclor 1268 ug/g	Aroclor 1268 (%)	Total PCB
LIS-2	Eastchester Bay	9/17/88	LIS2S	90000553	0.0000	0.0	0.0236	40.4	0.0214	36.7	0.0109	18.6	0.0025	4.3	0.42
LIS-3	Manhasset Bay	8/26/88	LIS3S	90000373	0.0000	0.0	0.0092	33.3	0.0098	35.2	0.0063		0.0024	8.6	0.22
LIS-4	Sands Point	8/24/89	143	89000485	0.0000	0.0	0.0063	46.0	0.0039	28.3	0.0028	20.5	0.0007	5.2	0.12
LIS-4	Sands Point	8/24/89	144	89000486	0.0000	0.0	0.0250	42.5	0.0208	35.5	0.0108	18.4	0.0021	3.5	0.51
LIS-4	Sands Point	8/24/89	377	89000491	0.0000	0.0	0.0149	44.2	0.0120	35.7	0.0052	15.4	0.0016	4.6	0.29
LIS-4	Sands Point	8/24/89	378	89000492	0.0000	0.0	0.0233	42.2	0.0191	34.5	0.0108	19.4	0.0021	3.9	0.42
LIS-4	Sands Point	8/24/89	379	89000493	0.0000	0.0	0.0146	41.9	0.0114	32.9	0.0072	20.6	0.0016	4.6	0.29
LIS-5	Hempstead Harbor	8/23/88	LIS5S	90000389	0.0000	0.0	0.0062	33.2	0.0082	44.0	0.0035	18.9	0.0007	3.9	0.13
LIS-6	Matinecock Point	8/27/88	LIS6S	90000419	0.0000	0.0	0.0004	40.0	0.0003	32.6	0.0002	23.2	0.0000	4.2	0.01
LIS-7	Bayville	8/27/88	LIS7S	90000410	0.0000	0.0	0.0024	29.9	0.0035	42.6	0.0019	22.7	0.0004	4.8	0.07
LIS-8	Oyster Bay	8/27/88	LIS8S	90000415	0.0000	0.0	0.0010	33.4	0.0011	39.4	0.0006	20.2	0.0002	7.0	0.02
LIS-9	Lloyd Point	8/26/88	LIS9S	90000289	0.0000	0.0	0.0014	33.6	0.0018	41.8	0.0009	20.4	0.0002	4.2	0.04
LIS-10	Target Rock	8/22/89	LIS10S91	89000393	0.0000	0.0	0.0014	29.1	0.0025	52.0	0.0009	18.9	0.0000	0.0	0.05
LIS-10	Target Rock	8/22/89	LIS10S92	89000394	0.0000	0.0	0.0069	42.2	0.0061	37.0	0.0029	17.6	0.0005	3.2	0.13
LIS-10	Target Rock	8/22/89	LIS10S93	89000395	0.0000	0.0	0.0117	38.3	0.0132	43.5	0.0046	15.1	0.0010	3.1	0.22
LIS-10	Target Rock	8/22/89	LIS10S94	89000396	0.0000	0.0	0.0136	51.2	0.0093	35.0	0.0031	11.7	0.0006	2.2	0.16
LIS-10	Target Rock	8/22/89	LIS10S95	89000397	0.0000	0.0	0.0052	38.8	0.0054	40.7	0.0023	17.2	0.0004	3.2	0.10
LIS-10	Target Rock	8/22/89	LIS10S96	89000398	0.0000	0.0	0.0033	38.7	0.0033	37.9	0.0016	19.1	0.0004	4.4	0.07
LIS-10	Target Rock	8/22/89	LIS10S97	89000399	0.0000	0.0	0.0239	58.9	0.0117	28.7	0.0043	10.6	0.0008	1.8	0.25
LIS-10	Target Rock	8/22/89	LIS10S98	89000400	0.0000	0.0	0.0327	59.7	0.0148	27.0	0.0062	11.4	0.0010	1.8	0.31
LIS-10	Target Rock	8/22/89	LIS10S99	89000401	0.0000	0.0	0.0039	42.0	0.0036	39.3	0.0014	15.7	0.0003	3.0	0.07
LIS-10	Target Rock	8/22/89	LIS10S910	89000402	0.0000	0.0	0.0073	51.5	0.0043	30.5	0.0021	15.1	0.0004	2.9	0.11
LIS-11	Eatons Neck	8/26/88	LIS11S	90000531	0.0000	0.0	0.0025	32.9	0.0030	40.0	0.0017	22.2	0.0004	4.9	0.07
LIS-12	Nissequogue River	8/26/88	LIS12S	90000524	0.0000	0.0	0.0010	21.3	0.0030	65.3	0.0005	10.5	0.0001	2.9	0.02
LIS-13	Stony Brook Harbor	8/26/88	LIS13S	90000286	0.0000	0.0	0.0081	47.6	0.0049	28.8	0.0035	20.7	0.0005	2.9	0.13
LIS-14	Port Jefferson Harbor	8/26/88	LIS14S	90000392	0.0000	0.0	0.0004	32.4	0.0006	42.6	0.0002	16.9	0.0001	8.1	0.01
LIS-15	Shoreham	8/25/88	LIS15S	90000400	0.0000	0.0	0.0003	48.3	0.0002	29.3	0.0001	15.5	0.0000	6.9	0.01
LIS-16	Glen Island	8/17/88	LIS16S	90000396	0.0000	0.0	0.0047	32.7	0.0058	40.6	0.0032	22.3	0.0006	4.4	0.11
LIS-17	Echo Bay - New Rochelle	8/17/89	LIS1789S1	89000266	0.0000	0.0	0.0247	31.3	0.0355	44.9	0.0160	20.3	0.0027	3.5	0.61
LIS-17	Echo Bay - New Rochelle	8/17/89	LIS1789S3	89000267	0.0000	0.0	0.0622	40.8	0.0576	37.8	0.0271	17.8	0.0055	3.6	1.04
LIS-17	Echo Bay - New Rochelle	8/17/89	LIS1789S4	89000268	0.0000	0.0	0.0281	13.3	0.0767	36.3	0.0916	43.4	0.0147	7.0	1.74
LIS-17	Echo Bay - New Rochelle	8/17/89	LIS1789S2	89000269	0.0000	0.0	0.0145	32.7	0.0186	41.9	0.0093	21.0	0.0019	4.3	0.36
LIS-18	Mamaroneck Harbor	8/17/89	LIS1889S1	89000258	0.0000	0.0	0.0136	48.5	0.0097	34.5	0.0040	14.3	0.0007	2.6	0.22
LIS-18	Mamaroneck Harbor	8/17/89	LIS1889S2	89000259	0.0000	0.0	0.1110	69.8	0.0327	20.6	0.0128	8.0	0.0026	1.6	0.67
LIS-18	Mamaroneck Harbor	8/17/89	LIS1889S3	89000260	0.0000	0.0	0.1157	62.1	0.0466	25.1	0.0198	10.6	0.0041	2.2	1.07
LIS-18	Mamaroneck Harbor	8/17/89	LIS1889S4	89000261	0.0000	0.0	0.0088	26.4	0.0082	24.8	0.0148	44.3	0.0015	4.5	0.21
LIS-18	Mamaroneck Harbor	8/17/89	LIS1889S5	89000262	0.0000	0.0	0.0175	42.6	0.0142	34.6	0.0077	18.9	0.0016	4.0	0.29
LIS-18	Mamaroneck Harbor	8/17/89	LIS1889S6	89000263	0.0000	0.0	0.0019	37.2	0.0020	39.6	0.0009	17.6	0.0003	5.5	0.04
LIS-18	Mamaroneck Harbor	8/17/89	LIS1889S7	89000264	0.0000	0.0	0.0251	43.5	0.0199	34.5	0.0102	17.7	0.0024	4.2	0.41

6.2-1 (con't)

Sampling Station	Location	Date Sampled	Marza ID	NEA ID	Aroclor 1221 ug/g	Aroclor 1221 (%)	Aroclor 1242 ug/g	Aroclor 1242 (%)	Aroclor 1254 ug/g	Aroclor 1254 (%)	Aroclor 1260 ug/g	Aroclor 1260 (%)	Aroclor 1268 ug/g	Aroclor 1268 (%)	Total PCB
LIS-18	Mamaroneck Harbor	8/17/89	LIS188988	89000265	0.0000	0.0	0.0064	21.9	0.0178	59.8	0.0048	16.0	0.0008	2.6	0.27
LIS-19	Port Chester Harbor	8/19/88	LIS198	90000934	0.0000	0.0	0.0063	11.6	0.0280	51.5	0.0193	28.4	0.0046	8.5	0.42
LIS-20	Flat Neck Point	8/19/88	LIS208	90000290	0.0000	0.0	0.0011	32.6	0.0015	42.9	0.0007	20.6	0.0001	3.8	0.03
LIS-21	Stamford Harbor	8/22/89	LIS218981	89000403	0.0000	0.0	0.3615	39.5	0.2426	26.5	0.2687	29.3	0.0431	4.7	6.96
LIS-21	Stamford Harbor	8/22/89	LIS218982	89000404	0.0314	3.2	0.4014	41.4	0.2811	29.0	0.2236	23.0	0.0328	3.4	7.97
LIS-21	Stamford Harbor	8/22/89	LIS218983	89000405	0.0000	0.0	0.2886	37.6	0.3707	48.3	0.0912	11.9	0.0179	2.3	5.58
LIS-21	Stamford Harbor	8/22/89	LIS218984	89000406	0.0000	0.0	0.0261	27.1	0.0434	45.1	0.0223	23.7	0.0039	4.0	0.89
LIS-21	Stamford Harbor	8/22/89	LIS218985	89000407	0.0000	0.0	0.0145	28.0	0.0227	43.7	0.0126	24.2	0.0022	4.2	0.44
LIS-21	Stamford Harbor	8/22/89	LIS218986	89000408	0.0000	0.0	0.0126	25.3	0.0225	45.3	0.0124	24.9	0.0022	4.5	0.41
LIS-21	Stamford Harbor	8/22/89	LIS218987	89000409	0.0000	0.0	0.0142	18.6	0.0485	63.8	0.0117	15.4	0.0017	2.2	0.64
LIS-22	Long Neck Point	8/19/88	LIS228	90000394	0.0000	0.0	0.0011	29.3	0.0016	42.4	0.0008	22.8	0.0002	5.4	0.03
LIS-23	Norwalk River Mouth	8/19/88	LIS238	90000528	0.0000	0.0	0.0011	30.0	0.0017	44.5	0.0008	20.8	0.0002	4.7	0.03
LIS-24	Southport	8/22/88	LIS248	90000291	0.0000	0.0	0.0046	25.7	0.0076	42.3	0.0048	26.7	0.0010	5.3	0.14
LIS-25	Black Rock Harbor	8/25/89	LIS2589S12	89000413	0.0000	0.0	0.3390	37.7	0.4146	46.1	0.1224	13.6	0.0237	2.6	7.70
LIS-25	Black Rock Harbor	8/25/89	LIS2589S11	89000412	0.0000	0.0	0.3394	39.5	0.3683	42.9	0.1252	14.6	0.0258	3.0	6.90
LIS-25	Black Rock Harbor	8/25/89	LIS2589S2	89000411	0.0000	0.0	0.0080	39.8	0.0090	45.0	0.0028	14.0	0.0002	1.2	0.18
LIS-25	Black Rock Harbor	8/25/89	LIS2589S21	89000414	0.0000	0.0	0.2760	49.4	0.2213	39.6	0.0509	9.1	0.0110	2.0	3.31
LIS-25	Black Rock Harbor	8/25/89	LIS2589S22	89000415	0.0000	0.0	0.3491	54.7	0.2154	33.8	0.0612	9.6	0.0123	1.9	4.41
LIS-25	Black Rock Harbor	8/25/89	LIS2589S31	89000416	0.0000	0.0	0.0830	43.0	0.0784	40.6	0.0264	13.7	0.0052	2.7	1.63
LIS-25	Black Rock Harbor	8/25/89	LIS2589S32	89000417	0.0000	0.0	0.0882	38.6	0.1014	44.4	0.0330	14.5	0.0056	2.5	1.69
LIS-25	Black Rock Harbor	8/25/89	LIS2589S41	89000418	0.0000	0.0	0.0721	38.5	0.0827	44.1	0.0269	14.4	0.0057	3.0	1.44
LIS-25	Black Rock Harbor	8/25/89	LIS2589S42	89000419	0.0000	0.0	0.1102	36.7	0.1425	47.4	0.0397	13.2	0.0082	2.7	2.36
LIS-25	Black Rock Harbor	8/25/89	LIS2589S51	89000410	0.0000	0.0	0.0049	39.7	0.0052	42.3	0.0019	15.2	0.0003	2.8	0.10
LIS-26	Bridgeport Harbor	8/23/89	LIS2689S1	89000385	0.0000	0.0	0.0264	38.5	0.0312	45.5	0.0096	14.0	0.0014	2.0	0.52
LIS-26	Bridgeport Harbor	8/23/89	LIS2689S2	89000386	0.0000	0.0	0.3105	57.3	0.1767	32.6	0.0447	8.2	0.0101	1.9	3.95
LIS-26	Bridgeport Harbor	8/23/89	LIS2689S3	89000387	0.0000	0.0	0.3818	67.7	0.1467	26.0	0.0289	5.1	0.0063	1.1	3.83
LIS-26	Bridgeport Harbor	8/23/89	LIS2689S4	89000388	0.0000	0.0	0.4226	72.3	0.1218	20.8	0.0325	5.6	0.0073	1.3	3.82
LIS-26	Bridgeport Harbor	8/23/89	LIS2689S5	89000389	0.0000	0.0	0.0160	61.5	0.0067	25.8	0.0027	10.6	0.0005	2.1	0.18
LIS-26	Bridgeport Harbor	8/23/89	LIS2689S6	89000390	0.0000	0.0	0.1151	27.1	0.0723	17.0	0.1720	40.5	0.0654	15.4	3.95
LIS-26	Bridgeport Harbor	8/23/89	LIS2689S7	89000391	0.0000	0.0	0.0269	51.8	0.0161	31.2	0.0070	13.5	0.0018	3.5	0.38
LIS-26	Bridgeport Harbor	8/23/89	LIS2689S8	89000392	0.0000	0.0	0.0002	46.0	0.0002	30.0	0.0001	24.0	0.0000	0.0	0.01
LIS-27	Housatonic River mouth	8/22/88	LIS278	90000525	0.0000	0.0	0.0279	78.6	0.0000	0.0	0.0058	16.5	0.0018	5.0	0.24
LIS-28	Milford Harbor	8/23/88	LIS288	90000284	0.0000	0.0	0.0033	29.8	0.0042	37.7	0.0030	26.5	0.0007	6.1	0.09
LIS-29	Stratford Shoal	8/25/88	LIS298	90000283	0.0000	0.0	0.0009	35.6	0.0009	35.6	0.0005	22.2	0.0002	6.7	0.02
LIS-30	New Haven Harbor	8/24/89	LIS3089S1	89000378	0.0000	0.0	0.0646	28.5	0.0717	31.6	0.0760	33.6	0.0143	6.3	1.87
LIS-30	New Haven Harbor	8/24/89	LIS3089S2	89000379	0.0000	0.0	0.9912	69.1	0.3389	23.6	0.0871	6.1	0.0171	1.2	11.81
LIS-30	New Haven Harbor	8/24/89	LIS3089S3	89000380	0.0000	0.0	0.0331	48.7	0.0237	34.9	0.0093	13.6	0.0019	2.8	0.46
LIS-30	New Haven Harbor	8/24/89	LIS3089S4	89000381	0.0000	0.0	0.0067	35.0	0.0076	39.4	0.0042	21.9	0.0007	3.7	0.18
LIS-30	New Haven Harbor	8/24/89	LIS3089S5	89000382	0.0000	0.0	0.0269	29.5	0.0130	14.2	0.0425	46.5	0.0089	9.8	0.67

6.2-1 (con't)

Sampling Station	Location	Date Sampled	Norta ID	NEA ID	• Aroclor 1221 • ug/g	(%)	• Aroclor 1242 • ug/g	(%)	• Aroclor 1254 • ug/g	(%)	• Aroclor 1260 • ug/g	(%)	• Aroclor 1268 • ug/g	(%)	total PCB
LIS-30	New Haven Harbor	8/24/89	LIS308986	89000383	0.0000	0.0	0.2603	54.5	0.1231	25.8	0.0739	15.5	0.0204	4.3	3.29
LIS-30	New Haven Harbor	8/24/89	LIS308987	89000384	0.0000	0.0	0.1883	58.9	0.1006	31.5	0.0262	8.2	0.0048	1.5	2.28
LIS-31	Central Dumping Ground	8/23/88	LIS31S	90000288	0.0000	0.0	0.0025	32.6	0.0032	41.4	0.0017	22.3	0.0003	3.7	0.06
LIS-32	Western Dumping Ground	9/27/88	LIS32S	90000550	0.0000	0.0	0.0103	24.1	0.0186	43.6	0.0113	26.5	0.0025	5.9	0.31

6.2-1 (con't)

Sampling Station	Location	Date Sampled	Harza ID	NEA ID	Homologs, Weight %												
					Mono	Di	Tri	Tetra	Penta	Hexa	Hepta	Octa	Nona	Deca	oCl/BP	m+pCl/BP	Tot Cl/B
HR-1	Troy Dam	6/1/90		2365 90000899	6.66	24.48	32.56	22.70	8.89	3.15	1.03	0.16	0.24	0.12	1.46	1.53	2.99
HR-1	Troy Dam	6/1/90		2366 90000900	2.98	23.12	37.88	24.27	7.89	2.56	0.62	0.12	0.32	0.23	1.43	1.66	3.09
HR-1	Troy Dam	10/19/90		3665 90002320	2.55	8.14	14.94	15.52	15.26	12.32	6.09	2.62	11.27	11.28	1.97	2.91	4.88
HR-1	Troy Dam	10/19/90		3666 90002321	3.78	23.21	37.59	23.03	7.79	3.00	0.77	0.19	0.40	0.24	1.38	1.69	3.07
HR-1	Troy Dam	9/25/89		2699 89000608	1.84	17.72	34.65	26.69	10.39	4.71	2.49	0.84	0.36	0.15	1.48	1.90	3.38
HR-1	Troy Dam	9/25/89		2701 89000609	1.22	17.94	35.36	29.76	10.05	3.49	1.42	0.35	0.22	0.00	1.41	1.99	3.40
HR-1	Troy Dam	7/25/88	HR1S	90000411	2.00	17.40	34.02	26.35	12.62	4.35	2.01	0.47	0.31	0.29	1.48	1.90	3.39
HR-1	Troy Dam	9/25/89		2711 89000610	0.00	22.61	34.78	27.60	8.75	3.53	2.47	0.25	0.00	0.00	1.49	1.79	3.28
HR-2	Watervliet	9/25/89		2661 89000611	1.96	18.09	32.21	27.21	10.16	5.60	2.74	0.93	0.79	0.32	1.57	2.03	3.60
HR-2	Watervliet	9/25/89		2671 89000612	1.80	15.47	27.75	27.21	15.67	7.06	3.29	0.62	0.59	0.55	1.38	1.66	3.04
HR-2	Watervliet	9/25/89		2681 89000613	3.51	25.36	34.69	25.75	7.25	2.62	0.70	0.14	0.00	0.00	1.51	1.94	3.45
HR-3	Albany	7/25/88	HR3S	90000541	2.64	17.95	29.94	26.81	11.49	5.93	3.90	0.67	0.59	0.09	1.47	1.97	3.44
HR-4	North Albany Turning Basin	7/26/88	HR4S	90000544	2.82	17.10	29.59	27.00	12.17	6.34	3.43	0.93	0.53	0.10	1.53	1.93	3.46
HR-5	South Albany Turning Basin	9/25/89		211 89000614	2.77	16.63	30.49	27.58	12.66	5.47	2.94	0.77	0.60	0.09	1.51	1.94	3.45
HR-5	South Albany Turning Basin	9/25/89		212 89000616	4.79	22.52	30.50	25.35	10.49	3.94	1.56	0.44	0.35	0.07	1.48	1.68	3.16
HR-5	South Albany Turning Basin	9/25/89		214 89000615	2.78	22.39	39.18	23.56	6.87	3.11	1.14	0.43	0.47	0.07	1.42	1.69	3.11
HR-5	South Albany Turning Basin	9/25/89		215 89000617	1.90	11.40	29.88	29.37	12.45	7.84	4.70	1.50	0.77	0.19	1.57	2.14	3.70
HR-5	South Albany Turning Basin	9/25/89		216 89000618	0.00	5.96	24.33	34.68	18.06	8.55	5.71	0.93	1.46	0.31	1.67	2.41	4.08
HR-5	South Albany Turning Basin	9/25/89		2591 89000619	1.72	16.40	29.07	25.18	12.19	7.95	4.94	1.55	0.81	0.18	1.51	2.09	3.61
HR-5	South Albany Turning Basin	9/25/89		2601 89000620	0.54	10.82	36.49	32.08	11.60	5.38	2.00	0.57	0.41	0.11	1.46	2.11	3.57
HR-5	South Albany Turning Basin	9/25/89		2611 89000621	2.61	18.47	31.14	25.10	11.05	6.25	3.76	1.04	0.49	0.08	1.51	1.91	3.42
HR-5	South Albany Turning Basin	9/25/89		2621 89000622	1.20	17.77	34.89	26.78	11.80	5.54	1.73	0.14	0.15	0.00	1.47	1.91	3.38
HR-5	South Albany Turning Basin	9/25/89		2631 89000623	3.38	16.56	31.18	28.70	12.54	4.70	1.95	0.56	0.36	0.07	1.53	1.84	3.37
HR-5	South Albany Turning Basin	9/25/89		2641 89000624	5.35	17.46	32.30	23.96	10.12	5.61	3.66	0.71	0.65	0.17	1.39	1.91	3.31
HR-6	Campbell Island	7/26/88	HR6S	90000416	1.69	12.00	26.15	25.04	22.33	9.37	2.46	0.45	0.37	0.13	1.57	2.19	3.76
HR-7	Revena	7/27/88	HR7S	90000402	0.00	16.35	41.61	23.81	8.79	4.36	3.99	0.84	0.24	0.00	1.39	2.03	3.42
HR-8	Coxsackie	9/19/89		169 89000598	1.30	6.41	11.57	11.64	9.13	13.58	14.31	4.61	11.73	15.72	2.22	3.32	5.55
HR-8	Coxsackie	9/19/89		170 89000599	2.82	24.33	34.22	22.10	8.82	4.26	1.65	0.33	0.75	0.72	1.31	1.87	3.18
HR-8	Coxsackie	9/19/89		171 89000600	2.00	18.50	25.67	24.42	13.09	7.79	3.17	0.68	1.98	2.69	1.55	2.10	3.64
HR-8	Coxsackie	9/19/89		172 89000601	3.07	20.20	36.28	25.10	9.69	3.50	1.43	0.42	0.28	0.04	1.44	1.76	3.20
HR-8	Coxsackie	9/19/89		173 89000602	1.99	17.63	34.50	26.12	10.19	5.57	2.48	0.82	0.52	0.17	1.50	1.89	3.39
HR-8	Coxsackie	9/19/89		174 89000603	2.68	22.56	37.75	22.84	7.34	3.81	1.96	0.59	0.35	0.13	1.42	1.74	3.16
HR-8	Coxsackie	9/19/89		175 89000604	1.82	21.45	35.01	25.87	8.63	4.38	2.43	0.26	0.15	0.00	1.48	1.77	3.25
HR-8	Coxsackie	9/19/89		176 89000605	1.88	16.54	29.46	23.97	11.24	8.34	5.99	1.72	0.70	0.16	1.52	2.09	3.61
HR-8	Coxsackie	9/19/89		177 89000606	0.19	9.80	38.56	34.05	10.61	4.68	1.44	0.43	0.20	0.05	1.48	2.07	3.55
HR-8	Coxsackie	9/19/89		178 89000607	1.59	17.99	34.76	27.14	9.62	5.39	2.63	0.43	0.31	0.15	1.47	1.91	3.37
HR-9	Stockport Creek	7/27/88															
HR-10	Hudson-Athens	7/27/88	HR10S	90000422	1.29	15.98	32.27	28.04	11.84	5.77	2.88	1.09	0.71	0.13	1.49	2.02	3.51
HR-11	Catskill Creek	7/28/88	HR11S	90000539	1.08	12.3	32.59	31.15	13.03	5.83	2.6	0.81	0.51	0.1	1.46	2.13	3.59

6.2-1 (con't)

Sampling Station	Location	Date Sampled	Nerze ID	NEA ID	Homologs, Weight % Mono	Di	Tri	Tetra	Penta	Hexa	Hepta	Octa	None	Deca	oCl/BP	m+pCl/BP	Tot Cl/B
HR-12	Inbocht Bay	7/28/88	HR12S	90000277	1.37	11.94	34.05	31.45	12.91	5.03	2.28	0.55	0.35	0.07	1.5	2.05	3.55
HR-13	Esopus Creek	7/28/88	HR13S	90000274	0.95	12.65	33.76	30.93	12.22	5.74	2.4	0.76	0.5	0.1	1.47	2.1	3.57
HR-14	South Bay-Annandale	7/28/88	HR14S	90000540	0.56	11.16	35.59	33.41	11.83	4.94	1.77	0.47	0.22	0.05	1.49	2.07	3.56
HR-15	Kingston	9/14/89	153	89000589	0.70	12.50	35.41	29.23	12.61	6.07	2.39	0.52	0.44	0.12	1.47	2.10	3.56
HR-15	Kingston	9/14/89	155	89000590	0.22	7.96	33.57	31.31	14.54	8.18	2.53	0.84	0.55	0.30	1.58	2.24	3.78
HR-15	Kingston	9/14/89	156	89000591	0.33	10.82	37.15	31.96	13.19	4.41	1.38	0.41	0.30	0.04	1.48	2.07	3.55
HR-15	Kingston	9/14/89	158	89000592	1.02	13.96	36.84	29.40	10.45	5.16	2.02	0.52	0.57	0.06	1.47	1.99	3.47
HR-15	Kingston	9/14/89	159	89000593	1.33	14.24	34.91	27.90	10.62	6.10	3.17	1.04	0.53	0.12	1.49	2.03	3.52
HR-15	Kingston	9/14/89	160	89000594	0.00	9.47	31.26	30.81	16.84	6.84	2.90	0.43	0.61	0.85	1.49	2.31	3.80
HR-15	Kingston	9/14/89	161	89000595	0.00	3.68	11.17	22.80	32.28	22.80	6.07	0.58	0.46	0.17	1.80	2.86	4.66
HR-15	Kingston	9/14/89	162	89000596	0.00	10.98	31.23	29.39	17.50	8.36	1.99	0.30	0.19	0.06	1.49	2.24	3.73
HR-15	Kingston	9/14/89	168	89000597	0.52	10.28	38.83	33.19	9.94	4.60	1.71	0.49	0.34	0.09	1.46	2.07	3.53
HR-16	Esopus Meadows Point	7/29/88	HR16S	90000423	0.36	8.84	34.34	33.01	13.99	5.87	1.77	0.56	1.03	0.24	1.53	2.17	3.70
HR-17	Hyde Park	10/4/89	315T	89000679	1.67	15.92	36.45	27.50	9.47	5.38	2.32	0.71	0.45	0.14	1.46	1.95	3.40
HR-17	Hyde Park	10/4/89	316T	89000680	1.30	14.17	35.84	28.03	10.74	5.97	2.42	0.84	0.56	0.14	1.46	2.03	3.49
HR-17	Hyde Park	10/4/89	317T	89000681	1.34	14.51	34.72	27.57	11.48	6.30	2.60	0.86	0.48	0.14	1.46	2.04	3.51
HR-17	Hyde Park	10/4/89	318T	89000682	0.00	10.13	37.10	34.38	12.53	4.10	1.33	0.25	0.19	0.00	1.44	2.12	3.57
HR-17	Hyde Park	10/4/89	319T	89000683	0.00	44.42	29.36	14.21	7.12	4.07	0.82	0.00	0.00	0.00	1.25	1.59	2.84
HR-17	Hyde Park	10/4/89	320T	89000684	0.63	12.77	39.29	29.94	9.67	4.67	1.74	0.65	0.52	0.12	1.50	1.97	3.47
HR-17	Hyde Park	10/4/89	321	89000685	1.22	14.02	35.13	28.46	11.21	5.99	2.31	0.81	0.68	0.17	1.46	2.05	3.51
HR-17	Hyde Park	10/4/89	322	89000686	0.00	9.74	42.26	31.89	9.65	4.43	1.27	0.41	0.27	0.06	1.50	2.01	3.51
HR-17	Hyde Park	10/4/89	323	89000687	1.02	13.50	36.80	29.20	10.37	5.35	2.36	0.77	0.51	0.12	1.47	2.02	3.49
HR-17	Hyde Park	10/4/89	324	89000688	0.91	13.27	37.00	29.29	10.38	5.35	2.25	0.83	0.63	0.14	1.49	2.01	3.50
HR-18	North Poughkeepsie	7/29/88	HR18S	90000275	1.01	10.83	27.94	27.09	14.82	9.66	5.44	1.44	1.60	0.18	1.56	2.30	3.87
HR-19	Poughkeepsie	10/4/89	305T	89000669	1.11	13.67	34.82	28.72	11.74	6.13	2.42	0.78	0.48	0.14	1.48	2.05	3.53
HR-19	Poughkeepsie	10/4/89	306T	89000670	1.28	14.45	35.56	28.37	10.09	6.32	2.58	0.77	0.46	0.12	1.47	2.02	3.49
HR-19	Poughkeepsie	10/4/89	307T	89000671	1.17	13.39	36.32	27.98	11.07	5.98	2.52	0.79	0.61	0.18	1.48	2.03	3.52
HR-19	Poughkeepsie	10/4/89	308T	89000672	1.23	13.65	35.98	28.72	10.96	5.93	2.23	0.69	0.47	0.13	1.48	2.02	3.50
HR-19	Poughkeepsie	10/4/89	309T	89000673	0.93	11.96	34.98	29.62	11.69	6.50	2.67	0.90	0.59	0.16	1.48	2.11	3.59
HR-19	Poughkeepsie	10/4/89	310T	89000674	1.02	12.62	35.21	29.10	12.22	5.56	2.58	0.88	0.65	0.16	1.46	2.10	3.56
HR-19	Poughkeepsie	10/4/89	311T	89000675	0.31	8.42	31.53	35.76	14.91	6.52	1.78	0.41	0.31	0.04	1.57	2.15	3.72
HR-19	Poughkeepsie	10/4/89	312T	89000676	0.00	9.10	38.48	32.24	11.30	5.89	1.88	0.61	0.42	0.09	1.54	2.08	3.62
HR-19	Poughkeepsie	10/4/89	313T	89000677	1.14	12.60	33.47	28.69	11.58	6.75	3.45	1.25	0.89	0.17	1.50	2.12	3.62
HR-19	Poughkeepsie	10/4/89	314T	89000678	1.26	13.73	36.98	28.32	10.47	5.43	2.52	0.73	0.56	0.00	1.46	2.02	3.48
HR-20	New Hamburg	7/30/88	HR20S	90000425	0.64	7.85	31.80	35.36	15.24	5.74	2.31	0.48	0.49	0.09	1.48	2.25	3.73
HR-21	Newburgh	7/30/88	HR21S	90000391	0.63	8.66	28.19	31.93	16.14	7.60	3.00	1.12	1.69	1.04	1.56	2.33	3.89
HR-22	Cornwall on Hudson	7/30/88	HR22S	90000427	0.00	3.08	36.25	34.78	14.24	6.58	4.24	0.82	0.00	0.00	1.49	2.37	3.87
HR-23	Foundry Cove	7/30/88	HR23S	90000420	0.58	7.99	29.49	31.12	17.75	8.66	2.90	0.77	0.56	0.19	1.56	2.29	3.85
HR-24	Con Hook	8/25/89	258	89000535	0.24	8.94	33.49	33.67	13.94	5.98	2.33	0.82	0.52	0.06	1.52	2.18	3.71

6.2-1 (cont)

Sampling Station	Location	Date Sampled	Marza ID	NEA ID	Homologs, Weight % Mono	Di	Tri	Tetra	Penta	Hexa	Hepta	Octa	Nona	Deca	oCl/BP	m+pCl/BP	Tot Cl/B
HR-24	Con Hook	8/29/89	397	89000530	0.44	8.94	33.11	35.44	13.95	5.68	1.96	0.43	0.38	0.07	1.48	2.19	3.67
HR-24	Con Hook	8/29/89	398	89000531	0.00	11.86	33.76	30.25	13.47	6.04	2.22	0.99	1.23	0.22	1.51	2.16	3.67
HR-24	Con Hook	8/25/89	399	89000532	0.00	3.66	9.66	11.01	10.11	17.08	32.32	12.92	3.17	0.07	2.11	3.48	5.58
HR-24	Con Hook	8/25/89	400	89000533	0.00	2.98	25.49	38.78	19.61	8.74	3.16	0.72	0.52	0.00	1.67	2.39	4.06
HR-24	Con Hook	8/29/89	237	89000534	0.00	9.10	34.55	33.04	14.05	5.94	2.53	0.50	0.27	0.00	1.55	2.13	3.69
HR-25	Iona Island	7/29/88	HR25S	90000414	0.00	4.27	36.52	36.57	14.26	4.86	2.40	0.67	0.40	0.07	1.53	2.25	3.78
HR-26	Peekskill Bay	7/29/88	HR26S	90000555	0.00	2.62	28.86	32.19	17.95	8.97	3.53	1.91	2.75	1.22	1.69	2.49	4.18
HR-27	Indian Point	7/29/88	HR27S	90000554	0.53	2.69	31.63	35.86	17.06	7.03	3.40	1.47	0.34	0.00	1.62	2.33	3.94
HR-28	Stony Point Bay	8/25/89	393	89000504	0.40	8.68	29.87	32.26	17.73	6.87	2.68	0.83	0.56	0.12	1.51	2.29	3.80
HR-28	Stony Point Bay	8/29/89	394	89000505	0.00	4.95	13.08	30.81	26.20	15.29	6.97	1.61	0.79	0.29	1.72	2.75	4.47
HR-28	Stony Point Bay	8/25/89	395	89000506	0.67	10.08	31.48	30.18	17.15	6.55	2.68	0.66	0.48	0.10	1.52	2.20	3.72
HR-28	Stony Point Bay	8/25/89	408	89000503	0.00	3.99	32.87	36.99	15.52	7.81	2.51	0.00	0.33	0.00	1.52	2.34	3.86
HR-28	Stony Point Bay	8/25/89	259	89000536	0.73	10.95	32.09	31.77	13.78	6.66	2.22	0.74	0.89	0.18	1.50	2.17	3.67
HR-28	Stony Point Bay	8/25/89	260	89000515	0.00	10.88	31.69	32.77	14.60	6.18	2.57	0.71	0.47	0.12	1.53	2.17	3.70
HR-28	Stony Point Bay	8/25/89	261	89000516	0.73	11.47	32.60	29.82	15.61	6.00	2.37	0.81	0.59	0.00	1.47	2.17	3.65
HR-28	Stony Point Bay	8/25/89	262	89000517	0.00	6.02	38.87	25.58	17.90	7.63	2.79	0.67	0.40	0.13	1.46	2.33	3.80
HR-28	Stony Point Bay	8/25/89	263	89000518	0.00	11.00	32.12	30.46	15.29	0.67	2.59	0.94	0.78	0.17	1.49	2.23	3.72
HR-28	Stony Point Bay	8/25/89	264	89000519	0.68	10.28	33.91	31.59	13.95	6.25	2.06	0.68	0.47	0.12	1.49	2.16	3.65
HR-29	Haverstraw	7/29/88	HR29S	90000546	0.22	6.13	28.00	34.38	18.57	8.36	2.80	0.80	0.55	0.19	1.57	2.36	3.93
HR-30	Croton Bay	7/27/88	HR30S	90000285	0.91	7.09	27.37	34.44	17.90	7.87	2.71	0.84	0.72	0.15	1.56	2.31	3.87
HR-31	Terrytown	7/27/88	HR31S	90000276	0.54	7.55	28.01	31.67	18.04	8.48	3.72	1.12	0.68	0.19	1.56	2.35	3.91
HR-32	Tappan Zee Bridge - Nyack	7/27/88	HR32S	90000545	0.43	6.69	27.38	34.59	18.10	8.40	2.89	0.76	0.60	0.17	1.57	2.34	3.91
HR-33	Piermont	7/27/88	HR33S	90000542	0.55	7.72	26.59	29.98	14.75	8.11	4.40	2.59	4.07	1.23	1.64	2.44	4.08
HR-34	Yonkers	7/28/88	HR34S	90000282	0.65	8.33	27.19	29.47	17.96	9.11	4.59	1.41	1.04	0.25	1.57	2.37	3.94
HR-35	Spuyten Devvil Creek	7/28/88	HR35S	90000279	0.45	7.66	27.36	27.06	16.10	9.55	7.42	2.91	1.31	0.17	1.62	2.44	4.06
HR-36	George Washington Bridge	7/28/88	HR36S	90000401	0.82	8.07	26.36	28.74	18.26	10.04	4.96	1.41	1.04	0.28	1.58	2.39	3.97
HR-37	North Bergen	7/28/88	HR37S	90000417	0.42	8.05	26.88	30.03	17.85	9.77	4.45	1.41	0.92	0.23	1.58	2.39	3.96
HR-38	The Battery	8/19/89	285	89000309	0.74	8.79	24.82	26.64	17.42	10.71	6.00	2.35	1.79	0.74	1.63	2.44	4.07
HR-38	The Battery	8/19/89	286	89000310	0.00	4.59	20.49	28.43	22.94	11.34	7.68	2.30	1.89	0.35	1.70	2.66	4.36
HR-38	The Battery	8/19/89	287	89000311	0.00	5.98	26.89	32.44	17.86	9.76	4.48	1.23	1.04	0.32	1.60	2.44	4.04
HR-38	The Battery	8/19/89	288	89000312	0.00	0.64	3.55	7.31	11.34	9.67	7.57	11.53	30.97	17.42	2.63	4.47	7.10
HR-38	The Battery	8/19/89	289	89000313	0.00	4.89	24.78	39.04	18.05	8.10	3.58	0.94	0.47	0.16	1.60	2.41	4.02
HR-38	The Battery	8/19/89	290	89000314	0.00	6.65	19.81	32.87	21.49	11.87	6.58	0.72	0.00	0.00	1.55	2.61	4.16
HR-38	The Battery	8/19/89	291	89000315	0.00	2.14	10.67	20.85	19.70	16.41	11.19	4.87	10.14	4.04	1.98	3.30	5.28
HR-38	The Battery	8/19/89	292	89000520	0.00	4.12	17.38	25.81	26.26	17.01	7.27	1.43	0.58	0.15	1.72	2.73	4.46
HR-38	The Battery	8/21/89	293	89000525	0.00	6.64	23.84	28.57	19.73	11.76	5.54	1.81	1.62	0.47	1.63	2.54	4.17
HR-38	The Battery	8/21/89	294	89000526	0.00	6.15	26.65	30.23	16.99	10.71	6.09	2.06	1.12	0.00	1.61	2.49	4.10
NYH-1	Bayonne	7/28/88	NYH-1S	90000281	2.00	2.31	14.90	18.04	16.37	19.00	12.45	5.77	6.08	3.08	1.87	3.10	4.97

6.2-1 (con't)

Sampling Station	Location	Date Sampled	Narza ID	NEA ID	Homologs, Weight %											
					Mono	Di	Tri	Tetra	Penta	Hexa	Hepta	Octa	Nona	Deca	oCl/BP	m+pCl/BP	Tot Cl/B
NYH-2	Gowanus Canal	8/21/89	441	89000507	0.00	3.95	18.40	25.49	22.67	17.66	8.78	1.85	0.93	0.26	1.72	2.77	4.49
NYH-2	Gowanus Canal	8/21/89	443	89000509	0.00	2.03	12.39	23.10	25.70	19.71	9.34	3.24	3.22	1.26	1.84	3.05	4.88
NYH-2	Gowanus Canal	8/21/89	444	89000510	0.00	4.02	21.00	26.87	18.92	21.63	5.09	1.41	0.76	0.31	1.87	2.51	4.38
NYH-2	Gowanus Canal	8/21/89	445	89000511	0.30	5.15	18.57	40.09	20.97	9.53	4.16	0.98	0.21	0.03	1.66	2.45	4.12
NYH-2	Gowanus Canal	8/21/89	446	89000512	0.00	4.29	18.53	25.30	22.39	15.14	11.85	1.75	0.75	0.00	1.82	2.67	4.49
NYH-2	Gowanus Canal	8/21/89	447	89000513	0.00	3.41	15.15	27.68	22.19	18.81	9.00	2.18	1.21	0.38	1.75	2.84	4.60
NYH-2	Gowanus Canal	8/21/89	448	89000514	0.00	6.09	20.75	27.26	20.21	14.41	7.07	2.17	1.50	0.54	1.66	2.65	4.32
NYH-2	Gowanus Canal	8/21/89	449	89000521	0.00	7.24	20.48	27.55	21.01	13.69	6.29	1.93	1.34	0.47	1.68	2.57	4.25
NYH-3	Verrazano Narrows Bridge	8/23/89	426	89000556	0.00	4.82	18.94	30.08	21.48	13.74	7.29	1.88	1.37	0.40	1.67	2.69	4.36
NYH-3	Verrazano Narrows Bridge	8/23/89	427	89000557	0.00	3.88	16.80	29.35	23.70	15.82	7.49	1.73	0.93	0.30	1.71	2.75	4.46
NYH-3	Verrazano Narrows Bridge	8/23/89	429	89000559	0.00	2.37	7.01	22.22	39.49	23.06	4.46	0.79	0.49	0.12	1.79	3.01	4.80
NYH-3	Verrazano Narrows Bridge	8/23/89	430	89000560	0.00	4.71	20.89	31.95	20.47	11.71	7.85	1.37	0.77	0.28	1.68	2.60	4.27
NYH-3	Verrazano Narrows Bridge	8/23/89	433	89000474	0.00	1.18	12.81	27.31	29.66	19.03	7.35	1.29	0.98	0.39	1.84	2.86	4.70
NYH-3	Verrazano Narrows Bridge	8/23/89	435	89000476	0.24	5.69	19.30	27.88	22.83	13.36	7.59	2.08	0.79	0.23	1.71	2.61	4.32
NYH-3	Verrazano Narrows Bridge	8/23/89	438	89000478	0.00	8.58	18.71	28.77	20.66	13.30	6.92	1.67	1.02	0.36	1.62	2.60	4.22
NYH-3	Verrazano Narrows Bridge	8/23/89	439	89000553	0.00	3.62	14.92	26.25	18.74	15.56	13.43	4.72	2.22	0.52	1.80	2.94	4.73
NYH-3	Verrazano Narrows Bridge	8/23/89	440	89000554	0.00	8.19	18.33	26.11	21.79	15.01	7.45	1.74	1.03	0.35	1.61	2.68	4.29
NYH-4	Midland Beach	8/23/90	401	89000466	0.00	4.23	17.62	26.99	22.54	16.07	8.23	2.15	1.64	0.53	1.73	2.76	4.50
NYH-4	Midland Beach	8/23/89	403	89000467	0.00	5.94	17.30	27.07	25.85	13.88	6.51	1.77	1.22	0.45	1.69	2.68	4.37
NYH-4	Midland Beach	8/23/89	406	89000470	0.00	3.68	16.46	27.31	22.85	17.52	8.31	1.87	1.53	0.48	1.73	2.82	4.54
NYH-4	Midland Beach	8/23/89	431	89000463	0.00	5.53	19.64	24.55	23.33	14.02	9.24	1.98	1.22	0.49	1.74	2.66	4.41
NYH-4	Midland Beach	8/23/89	432	89000465	0.00	4.81	19.22	26.50	23.78	14.30	7.93	1.83	1.19	0.44	1.73	2.68	4.40
NYH-4	Midland Beach	8/21/89	451	89000523	0.00	1.39	14.19	28.55	24.66	18.28	8.21	2.15	2.57	0.00	1.76	2.93	4.70
NYH-5	Coney Island	8/23/88	NYH5S	90000437	0.00	3.43	22.20	24.11	22.75	15.85	7.54	2.17	1.41	0.54	1.71	2.72	4.43
NYH-6	Great Kills Harbor	8/16/89	305	89000320	0.00	7.16	19.89	21.65	27.75	14.49	7.88	0.67	0.51	0.00	1.69	2.60	4.30
NYH-6	Great Kills Harbor	8/16/89	306	89000321	0.00	5.92	19.49	27.59	20.93	14.66	7.95	1.79	1.19	0.48	1.67	2.67	4.35
NYH-6	Great Kills Harbor	8/16/89	313	89000328	0.00	4.80	19.63	23.73	21.32	17.44	9.59	1.77	1.27	0.44	1.79	2.68	4.47
NYH-6	Great Kills Harbor	8/16/89	314	89000329	0.00	8.30	16.71	20.71	24.01	17.68	9.42	1.60	1.11	0.47	1.64	2.78	4.42
NYH-6	Great Kills Harbor	8/16/89	315	89000330	0.00	5.10	18.79	25.80	21.63	16.85	7.51	2.02	1.80	0.50	1.76	2.68	4.44
NYH-6	Great Kills Harbor	8/16/89	316	89000331	0.00	2.76	16.50	27.44	25.89	15.48	9.03	1.39	1.01	0.50	1.77	2.78	4.55
NYH-6	Great Kills Harbor	8/16/89	318	89000333	0.00	5.68	19.92	24.60	23.39	16.28	6.99	1.77	1.36	0.00	1.71	2.65	4.37
NYH-6	Great Kills Harbor	8/16/89	319	89000334	0.00	5.55	20.73	27.98	19.52	13.25	9.59	1.50	1.37	0.52	1.75	2.59	4.34
NYH-7	Seguine Point	8/30/88	NYH7S	90000385	0.00	11.58	22.36	26.67	20.06	13.33	4.81	0.66	0.53	0.00	1.72	2.26	3.98
NYH-8	Raritan River mouth	8/16/89	297	89000300	0.00	5.65	9.48	24.15	26.67	15.28	9.18	2.97	4.78	1.84	1.79	3.02	4.81
NYH-8	Raritan River mouth	8/16/89	298	89000301	0.00	2.29	15.99	29.25	23.41	18.47	7.53	1.87	0.91	0.28	1.79	2.78	4.56
NYH-8	Raritan River mouth	8/16/89	299	89000302	0.00	6.82	12.27	28.11	20.92	18.00	8.98	2.78	1.36	0.75	1.84	2.70	4.54
NYH-8	Raritan River mouth	8/16/89	308	89000323	0.00	8.22	14.68	27.93	22.51	16.63	6.64	1.96	0.98	0.44	1.68	2.68	4.35
NYH-8	Raritan River mouth	8/16/89	309	89000324	0.00	1.64	3.46	19.53	39.96	25.60	7.34	1.81	0.61	0.04	1.87	3.17	5.04
NYH-8	Raritan River mouth	8/16/89	311	89000326	0.00	3.06	14.90	27.14	23.57	17.04	11.51	1.78	0.71	0.29	1.85	2.77	4.62

6.2-1 (con't)

Sampling Station	Location	Date Sampled	Marza ID	NEA ID	Homologs, Weight %												
					Mono	Di	Tri	Tetra	Penta	Hexa	Hepta	Octa	Nona	Deca	oCl/BP	m+pCl/BP	Tot Cl/B
NYH-8	Raritan River mouth	8/16/89	312	89000327	0.00	15.99	10.64	23.36	24.32	16.91	9.77	1.81	0.96	0.24	1.68	2.50	4.18
NYH-9	Woodbridge Creek	8/18/89	276	89000293	0.33	5.19	20.80	35.82	19.77	10.92	4.21	1.31	0.77	0.68	1.71	2.45	4.16
NYH-9	Woodbridge Creek	8/18/89	278	89000295	0.00	2.26	14.38	31.34	28.01	15.80	6.34	1.56	0.09	0.22	1.76	2.75	4.51
NYH-9	Woodbridge Creek	8/18/89	279	89000296	0.00	2.58	22.73	37.61	21.91	9.28	4.25	1.01	0.45	0.18	1.71	2.46	4.17
NYH-9	Woodbridge Creek	8/18/89	281	89000298	0.00	4.42	13.42	26.32	27.28	15.24	9.43	1.90	1.31	0.27	1.80	2.77	4.57
NYH-9	Woodbridge Creek	8/18/89	282	89000299	0.00	3.21	12.97	34.24	28.68	14.44	4.78	1.14	0.43	0.10	1.72	2.72	4.44
NYH-9	Woodbridge Creek	8/18/89	283	89000308	0.00	4.30	14.09	31.30	25.37	14.66	7.38	1.68	0.88	0.33	1.73	2.74	4.47
NYH-10	Fresh Kills	8/23/88	NYH10S	90000412	0.00	3.91	19.32	29.18	24.10	14.27	6.75	1.28	0.84	0.36	1.72	2.65	4.37
NYH-11	Prails Island	9/19/88	NYH11S	90000407	0.41	3.71	16.20	28.76	19.90	16.36	10.32	2.75	1.19	0.42	1.77	2.76	4.54
NYH-12	Newark Bay	8/18/89	265	89000305	0.00	1.08	12.80	36.46	26.32	13.84	6.96	1.50	1.04	0.00	1.83	2.73	4.55
NYH-12	Newark Bay	8/18/89	266	89000306	0.00	4.41	17.92	28.02	26.97	12.58	6.68	1.90	1.10	0.42	1.73	2.67	4.40
NYH-12	Newark Bay	8/18/89	267	89000307	0.00	3.92	17.33	27.73	24.90	14.21	6.44	2.86	2.23	0.38	1.77	2.72	4.49
NYH-12	Newark Bay	8/18/89	268	89000284	0.00	5.07	18.18	30.38	24.79	12.36	6.35	1.72	0.84	0.31	1.73	2.60	4.33
NYH-12	Newark Bay	8/18/89	269	89000285	0.00	3.79	18.91	29.58	24.95	13.17	6.33	1.93	0.96	0.39	1.75	2.63	4.38
NYH-12	Newark Bay	8/18/89	270	89000286	0.00	3.58	17.39	30.42	21.61	14.91	8.38	2.31	1.23	0.16	1.74	2.72	4.47
NYH-12	Newark Bay	8/18/89	271	89000287	0.00	5.20	18.01	28.29	23.68	13.96	6.76	2.28	1.28	0.53	1.75	2.64	4.39
NYH-12	Newark Bay	8/18/89	273	89000288	0.00	5.01	15.54	31.33	25.95	12.99	6.23	1.74	0.92	0.30	1.71	2.68	4.38
NYH-12	Newark Bay	8/18/89	274	89000289	0.00	4.85	17.51	33.72	22.81	12.45	6.05	1.40	0.86	0.35	1.70	2.61	4.31
NYH-12	Newark Bay	8/18/89	275	89000291	0.00	3.57	16.43	29.99	25.72	14.58	6.31	1.64	1.30	0.46	1.75	2.71	4.46
NYH-13	Wallabout Bay	8/25/88	NYH13S	90000399	0.00	7.49	18.25	26.42	19.08	15.37	9.07	2.42	1.37	0.53	1.64	2.72	4.37
NYH-14	Newtown Creek	8/22/89	413	89000540	0.00	5.53	19.71	26.70	22.20	15.09	7.15	1.89	1.34	0.39	1.67	2.69	4.36
NYH-14	Newtown Creek	8/22/89	414	89000541	0.00	6.94	18.90	28.57	19.11	15.06	7.77	2.24	1.10	0.30	1.68	2.63	4.31
NYH-14	Newtown Creek	8/22/89	415	89000542	0.00	3.97	16.66	25.78	21.06	15.68	12.13	3.12	1.32	0.28	1.74	2.86	4.60
NYH-14	Newtown Creek	8/22/89	416	89000543	0.00	5.40	20.40	26.06	21.01	15.96	7.95	1.92	1.05	0.26	1.69	2.68	4.37
NYH-14	Newtown Creek	8/22/89	417	89000544	0.00	4.27	20.28	29.90	22.19	13.61	6.22	1.85	1.27	0.41	1.70	2.64	4.34
NYH-14	Newtown Creek	8/22/89	423	89000471	0.00	4.22	19.54	25.63	19.99	17.26	9.26	2.49	1.15	0.47	1.73	2.75	4.48
NYH-14	Newtown Creek	8/22/89	424	89000472	1.21	5.10	16.72	29.24	26.66	11.45	7.44	1.14	0.76	0.28	1.72	2.57	4.29
NYH-14	Newtown Creek	8/22/89	452	89000527	0.00	5.39	20.36	28.07	20.03	14.40	7.46	2.14	1.66	0.50	1.67	2.69	4.35
NYH-15	Wards Island	8/22/89	409	89000552	0.00	5.20	19.62	28.56	21.91	14.09	6.92	2.00	1.28	0.42	1.68	2.67	4.35
NYH-15	Wards Island	8/22/89	410	89000537	0.00	8.12	21.49	14.20	17.72	23.16	8.80	3.50	3.02	0.00	1.55	2.94	4.49
NYH-15	Wards Island	8/22/89	411	89000538	0.25	3.73	13.10	25.12	29.01	16.50	9.22	1.74	0.99	0.34	1.79	2.82	4.61
NYH-15	Wards Island	8/22/89	412	89000539	0.00	4.16	22.77	28.89	20.56	13.23	6.78	2.06	1.06	0.50	1.70	2.61	4.30
NYH-15	Wards Island	8/22/89	453	89000528	0.00	14.23	32.79	29.17	10.15	7.56	4.11	1.13	0.65	0.20	1.56	2.08	3.64
NYH-15	Wards Island	8/22/89	454	89000549	0.00	2.67	12.94	19.84	21.74	24.36	15.24	2.54	0.67	0.00	1.84	3.05	4.89
NYH-15	Wards Island	8/22/89	455	89000550	0.00	4.65	18.72	21.06	24.27	17.42	9.29	2.62	1.52	0.44	1.73	2.82	4.54
NYH-15	Wards Island	8/22/89	456	89000551	0.00	3.06	17.02	25.15	20.84	18.87	9.66	2.95	1.90	0.55	1.78	2.87	4.64
NYH-16	Harlem River	8/25/88	NYH16S	90000271	0.54	8.47	21.94	28.02	17.84	12.58	6.97	1.95	1.25	0.43	1.61	2.52	4.14
NYH-17	College Point	8/25/88	NYH17S	90000424	0.35	3.00	14.43	21.63	24.47	20.07	10.90	2.82	1.75	0.60	1.81	2.94	4.74
NYH-18	Throgs Neck Bridge	8/25/88	NYH18S	90000280	0.00	6.30	16.64	21.58	22.41	18.33	9.76	2.68	1.68	0.62	1.74	2.81	4.55

6.2-1 (cont.)

Sampling Station	Location	Date Sampled	Nerza ID	NEA ID	Homologs, Weight %												
					Mono	Di	Tri	Tetra	Penta	Hexa	Hepta	Octa	Nona	Deca	oCl/BP	m+pCl/BP	Tot Cl/B
NYH-19	Sandy Hook	10/12/90	5014	90002062	0.54	3.53	15.98	26.19	21.7	18.08	8.72	2.4	2.23	0.63	1.8	2.78	4.58
NYH-19	Sandy Hook	10/12/90	5013	90002049	0	1.97	12.95	29.02	25.61	18.67	7.87	2.43	1.48	0	1.85	2.82	4.67
NYH-19	Sandy Hook	10/12/90	5020	90002050	0	2.75	15.49	27.53	22.11	18.98	8.34	2.58	1.53	0.7	1.77	2.86	4.63
NYH-19	Sandy Hook	10/12/90	5019	90002051	0	2.23	13.79	25.56	23.25	19.76	10.36	2.41	1.92	0.72	1.87	2.89	4.76
NYH-19	Sandy Hook	10/12/90	5015	90002064	0	2.6	14.79	27.91	21.97	19.55	9.38	1.9	1.4	0.49	1.83	2.82	4.65
NYH-20	Lower Bay																
NYH-21	Rockaway Inlet	10/30/90	5154	90002157	0	3.04	14.58	25.78	25.68	18.97	8.68	1.63	1.13	0.5	1.81	2.82	4.63
NYH-21	Rockaway Inlet	10/30/90	5156	90002159	0	2.91	14.53	27.14	22.07	18.77	9.47	2.81	1.63	0.67	1.79	2.89	4.68
NYH-21	Rockaway Inlet	10/30/90	5155	90002158	0	3.48	15.7	23.93	19.06	23.94	10.19	2.15	1.13	0.43	1.83	2.85	4.68
NYH-21	Rockaway Inlet	10/30/90	5164	90002160	0	2.45	13.66	23.07	21.64	23.98	11.51	2.07	1.18	0.43	1.92	2.88	4.8
NYH-21	Rockaway Inlet	10/30/90	5165	90002161	0	2.98	13.89	23.49	21.52	27.49	7.71	1.56	1.04	0.32	1.84	2.88	4.72
NYH-21	Rockaway Inlet	10/30/90	5166	90002162	0	2.64	12.43	26.2	23.61	24.79	7.27	1.63	1.06	0.37	1.83	2.88	4.71
NYH-22	Jamaica Bay-Beach Channel	09/11/90	3699	90001581	0	2.65	14.65	22.09	22.69	18.38	10.44	4.56	3.89	0.64	1.92	2.93	4.85
NYH-22	Jamaica Bay-Beach Channel	09/10/90	3209	90001576	0	2.27	13.78	25.5	22.77	20.82	10.31	2.51	1.42	0.62	1.89	2.87	4.76
NYH-22	Jamaica Bay-Beach Channel	09/10/90	3210	90001577	1.03	6.73	18.82	21.36	23.06	18.01	7.29	2	1.29	0.42	1.76	2.59	4.35
NYH-22	Jamaica Bay-Beach Channel	09/10/90	3211	90001578	0	2.83	15.29	22.97	23.22	20.89	10.42	2.39	1.47	0.52	1.88	2.84	4.72
NYH-22	Jamaica Bay-Beach Channel	09/10/90	3690	90001579	0	2.01	21.58	27.1	22.72	16.09	6.6	1.94	1.3	0.64	1.77	2.69	4.46
NYH-22	Jamaica Bay-Beach Channel	09/10/90	3691	90001580	0	4.71	13.99	22.11	22.49	21.02	11.05	2.64	1.43	0.57	1.89	2.81	4.7
NYH-23	Jamaica Bay - Canarsie	10/16/90	5056	90002063	0	2.1	10.09	20.09	24.43	23.01	14.39	3.54	1.57	0.78	1.97	3.06	5.03
NYH-23	Jamaica Bay - Canarsie	10/16/90	5061	90002069	0	3.31	14.73	22.6	24.59	20.72	8.82	2.67	1.74	0.82	1.87	2.84	4.71
NYH-23	Jamaica Bay - Canarsie	10/16/90	5063	90002070	0	2.58	14.79	24.11	23.59	19.7	10.58	2.39	1.57	0.69	1.88	2.85	4.73
NYH-23	Jamaica Bay - Canarsie	10/16/90	5059	90002072	0	3.04	13.33	24.67	24.59	19.34	10.25	2.61	1.49	0.67	1.83	2.9	4.73
NYH-23	Jamaica Bay - Canarsie	10/16/90	5060	90002065	0	3	10.51	23.15	24.42	24.21	10.46	2.31	1.4	0.53	1.94	2.91	4.85
NYH-23	Jamaica Bay - Canarsie	10/16/90	5062	90002068	0	2.26	10.44	22.18	22.54	27.28	10.02	2.76	1.79	0.71	1.93	3.02	4.94
NYH-23	Jamaica Bay - Canarsie	10/16/90	5064	90002071	0	6.27	14.12	23.3	20.53	22.48	9.04	2.27	1.52	0.47	1.85	2.74	4.59
NYH-24	Harlem River - 207th St.	10/26/90	5151	90002060	0.61	7.57	27.57	30.81	16.44	10.05	4.76	1.28	0.71	0.2	1.6	2.35	3.95
NYH-24	Harlem River - 207th St.	10/26/90	5149	90002061	0.65	4.45	21.48	32.28	19.62	13.21	5.85	1.49	0.74	0.21	1.7	2.51	4.21
NYH-24	Harlem River - 207th St.	10/26/90	5148	90002058	0	3.54	18.76	25.99	16.81	16.09	13.07	3.83	1.55	0.37	1.79	2.82	4.61
NYH-24	Harlem River - 207th St.	10/26/90	5150	90002059	0	5.03	20.68	29.4	19.14	13.7	7.84	2.55	1.34	0.33	1.71	2.63	4.34
NYH-24	Harlem River - 207th St.	10/26/90	5145	90002057	0	4.18	22	30.12	17.5	13.28	8.57	2.5	1.46	0.39	1.71	2.64	4.35
NYH-24	Harlem River - 207th St.	10/26/90	5146	90002066	0.00	4.45	22.69	27.86	16.57	14.11	9.58	2.79	1.61	0.33	1.74	2.64	4.37
NYH-24	Harlem River - 207th St.	10/26/90	5147	90002067	0	4.84	23.51	28.95	16.6	12.3	8.99	2.75	1.73	0.33	1.73	2.58	4.31
LIS-1	Little Neck Bay	8/24/89	381	89000495	0.00	1.13	13.89	22.62	27.07	20.57	9.94	2.42	1.75	0.61	1.85	2.98	4.83
LIS-1	Little Neck Bay	8/24/89	382	89000496	0.00	3.90	15.52	23.73	24.67	18.24	9.32	2.35	1.65	0.61	1.79	2.84	4.63
LIS-1	Little Neck Bay	8/24/89	383	89000497	0.00	4.78	16.59	24.94	23.31	18.35	8.27	2.05	1.23	0.48	1.76	2.76	4.52
LIS-1	Little Neck Bay	8/24/89	384	89000498	0.00	4.28	17.25	18.54	26.77	18.45	9.32	2.90	1.85	0.64	1.80	2.85	4.65
LIS-1	Little Neck Bay	8/24/89	386	89000500	0.00	6.45	16.30	18.79	22.26	19.49	10.90	2.99	2.04	0.79	1.74	2.89	4.62
LIS-1	Little Neck Bay	8/24/89	387	89000501	0.00	10.42	25.24	28.96	17.73	11.14	4.84	1.04	0.63	0.00	1.61	2.33	3.94
LIS-1	Little Neck Bay	8/24/89	388	89000502	0.00	6.25	18.32	25.61	21.01	16.56	7.84	2.35	1.52	0.55	1.74	2.67	4.41

6.2-1 (con't)

Sampling Station	Location	Date Sampled	Nersa ID	NEA ID	Homologs, Weight %												
					Mono	DI	Tri	Tetra	Penta	Hexa	Hepta	Octa	Nona	Deca	eCl/BP	m+pCl/BP	Tot Cl/B
LIS-2	Eastchester Bay	9/17/88	LIS2S	90000553	0.00	2.09	11.64	20.89	24.81	20.01	14.59	3.30	1.95	0.80	1.89	3.08	4.97
LIS-3	Manhasset Bay	8/26/88	LIS3S	90000373	0.00	2.64	11.93	18.91	24.71	17.99	10.37	5.83	6.37	1.22	1.92	3.14	5.06
LIS-4	Sands Point	8/24/89	143	89000485	0.00	5.79	13.14	20.95	21.76	19.83	11.58	3.73	2.00	1.23	1.93	2.81	4.74
LIS-4	Sands Point	8/24/89	144	89000486	0.00	2.07	10.42	25.47	32.55	16.79	9.06	2.22	0.94	0.48	1.81	2.96	4.77
LIS-4	Sands Point	8/24/89	377	89000491	0.00	3.19	16.75	20.52	26.33	18.61	9.16	2.76	2.02	0.66	1.81	2.88	4.68
LIS-4	Sands Point	8/24/89	378	89000492	0.00	1.94	11.34	21.06	29.37	20.93	9.86	2.68	2.06	0.76	1.86	3.04	4.89
LIS-4	Sands Point	8/24/89	379	89000493	0.00	3.20	11.24	17.72	28.11	23.38	10.85	2.95	1.95	0.60	1.82	3.10	4.92
LIS-5	Hempstead Harbor	8/23/88	LIS5S	90000389	0.00	0.68	8.54	20.05	30.79	23.50	10.96	3.27	1.54	0.67	1.93	3.14	5.07
LIS-6	Matinecock Point	8/27/88	LIS6S	90000419	0.00	4.85	10.90	17.35	25.19	22.20	12.10	3.71	2.57	1.14	1.88	3.04	4.92
LIS-7	Bayville	8/27/88	LIS7S	90000410	0.00	1.14	6.59	16.07	35.17	23.90	12.66	2.46	1.27	0.74	1.93	3.22	5.15
LIS-8	Oyster Bay	8/27/88	LIS8S	90000415	0.00	3.03	7.23	17.12	25.55	21.06	14.36	8.28	2.45	0.93	2.03	3.18	5.22
LIS-9	Lloyd Point	8/26/88	LIS9S	90000289	0.00	3.16	9.16	22.50	27.05	20.46	10.78	3.55	2.29	1.04	1.86	3.07	4.93
LIS-10	Target Rock	8/22/89	LIS1089S1	89000393	0.00	4.62	11.36	15.78	34.30	24.61	7.49	1.17	0.67	0.00	1.73	3.01	4.74
LIS-10	Target Rock	8/22/89	LIS1089S2	89000394	0.00	3.76	11.78	22.41	25.74	23.87	8.94	2.00	0.99	0.52	1.77	2.98	4.75
LIS-10	Target Rock	8/22/89	LIS1089S3	89000395	0.00	1.84	10.54	24.16	30.17	23.17	7.17	1.86	0.89	0.19	1.78	3.02	4.80
LIS-10	Target Rock	8/22/89	LIS1089S4	89000396	0.00	0.72	14.91	23.65	27.83	21.34	8.02	1.95	1.25	0.34	1.81	2.95	4.75
LIS-10	Target Rock	8/22/89	LIS1089S5	89000397	0.00	2.20	12.10	22.04	28.26	23.58	8.58	2.36	0.71	0.17	1.74	3.05	4.80
LIS-10	Target Rock	8/22/89	LIS1089S6	89000398	0.00	2.83	11.22	17.66	28.95	25.14	9.96	2.77	1.07	0.41	1.83	3.07	4.90
LIS-10	Target Rock	8/22/89	LIS1089S7	89000399	0.00	3.60	19.42	24.06	25.40	17.02	7.07	1.69	0.74	1.00	1.79	2.69	4.48
LIS-10	Target Rock	8/22/89	LIS1089S8	89000400	0.00	2.72	20.60	21.44	27.42	18.36	7.12	1.68	0.60	0.15	1.77	2.73	4.50
LIS-10	Target Rock	8/22/89	LIS1089S9	89000401	0.00	1.09	12.93	22.53	25.97	23.89	9.82	2.20	1.57	0.00	1.88	2.97	4.85
LIS-10	Target Rock	8/22/89	LIS1089S10	89000402	0.00	2.90	13.98	27.54	25.74	17.19	9.37	2.06	0.99	0.25	1.81	2.82	4.62
LIS-11	Eatons Neck	8/26/88	LIS11S	90000531	0.00	2.99	9.66	17.17	27.42	22.54	12.65	3.80	2.72	1.05	1.89	3.16	5.05
LIS-12	Nissequogue River	8/26/88	LIS12S	90000524	0.00	2.02	7.23	20.85	34.08	20.68	8.24	3.21	2.59	1.10	1.89	3.12	5.01
LIS-13	Stony Brook Harbor	8/26/88	LIS13S	90000286	0.00	8.21	18.53	19.20	19.97	18.64	11.60	2.06	1.26	0.53	1.76	2.69	4.46
LIS-14	Port Jefferson Harbor	8/26/88	LIS14S	90000392	0.00	3.64	9.54	20.20	29.52	21.18	8.83	4.13	1.77	1.18	1.89	3.01	4.90
LIS-15	Shoreham	8/25/88	LIS15S	90000400	0.00	5.36	14.40	17.35	27.89	20.31	7.81	3.57	2.30	1.02	1.83	2.89	4.71
LIS-16	Glen Island	8/17/88	LIS16S	90000396	0.00	1.95	8.20	18.83	30.80	22.57	12.30	3.42	1.37	0.55	1.92	3.13	5.04
LIS-17	Echo Bay - New Rochelle	8/17/89	LIS1789S1	89000266	0.00	2.14	9.11	18.62	30.44	26.86	9.43	2.16	0.88	0.37	1.84	3.12	4.96
LIS-17	Echo Bay - New Rochelle	8/17/89	LIS1789S3	89000267	0.00	1.06	10.24	17.55	30.52	26.73	9.94	2.60	1.12	0.23	1.89	3.12	5.01
LIS-17	Echo Bay - New Rochelle	8/17/89	LIS1789S4	89000268	0.00	0.86	7.54	11.56	32.87	25.99	16.45	3.76	0.82	0.15	1.93	3.35	5.27
LIS-17	Echo Bay - New Rochelle	8/17/89	LIS1789S2	89000269	0.00	2.52	6.17	18.00	34.37	25.40	9.27	2.79	1.12	0.36	1.86	3.17	5.03
LIS-18	Mamaroneck Harbor	8/17/89	LIS1889S1	89000258	0.00	3.71	17.10	21.26	27.34	17.42	10.00	1.91	0.92	0.34	1.80	2.80	4.60
LIS-18	Mamaroneck Harbor	8/17/89	LIS1889S2	89000259	0.00	2.06	23.14	18.90	28.17	16.12	7.80	2.16	1.41	0.23	1.79	2.72	4.51
LIS-18	Mamaroneck Harbor	8/17/89	LIS1889S3	89000260	0.00	7.23	21.00	22.85	23.43	14.56	6.83	2.03	0.72	1.35	1.70	2.62	4.31
LIS-18	Mamaroneck Harbor	8/17/89	LIS1889S4	89000261	0.00	1.87	11.08	17.68	26.12	22.22	14.52	3.75	1.92	0.84	1.89	3.16	5.05
LIS-18	Mamaroneck Harbor	8/17/89	LIS1889S5	89000262	0.00	2.91	17.67	17.64	26.36	21.02	9.78	3.03	1.20	0.40	1.67	3.04	4.71
LIS-18	Mamaroneck Harbor	8/17/89	LIS1889S6	89000263	0.00	4.25	11.06	18.58	28.99	21.32	10.06	2.92	2.09	0.73	1.87	2.97	4.84
LIS-18	Mamaroneck Harbor	8/17/89	LIS1889S7	89000264	0.00	3.36	14.61	19.78	26.55	20.12	10.45	3.44	1.34	0.34	1.83	2.93	4.76

6.2-1 (con't)

Sampling Station	Location	Date Sampled	Norza ID	NEA ID	Homologs, Weight %												
					Mono	Di	Tri	Tetra	Penta	Hexa	Hepta	Octa	Nona	Deca	oCl/BP	m+pCl/BP	Tot Cl/B
LIS-18	Mamaroneck Harbor	8/17/89	LIS188988	89000265	0.00	1.57	5.96	21.69	37.82	24.35	6.47	1.65	0.48	0.00	1.81	3.11	4.92
LIS-19	Port Chester Harbor	8/19/88	LIS198	90000534	0.00	0.55	2.99	13.39	34.43	26.05	13.27	5.93	2.89	0.50	1.99	3.47	5.47
LIS-20	Flat Neck Point	8/19/88	LIS208	90000290	0.00	3.60	9.56	18.47	27.45	21.48	12.76	3.48	2.23	0.97	1.88	3.10	4.98
LIS-21	Stamford Harbor	8/22/89	LIS218981	89000403	0.00	3.23	12.11	18.71	24.66	23.64	14.13	2.85	0.59	0.07	1.84	3.04	4.88
LIS-21	Stamford Harbor	8/22/89	LIS218982	89000404	0.39	3.45	11.31	30.72	27.91	14.35	9.06	2.00	0.42	0.18	1.76	2.80	4.56
LIS-21	Stamford Harbor	8/22/89	LIS218983	89000405	0.00	2.27	11.44	25.44	33.29	18.59	6.78	1.49	0.55	0.12	1.82	2.86	4.68
LIS-21	Stamford Harbor	8/22/89	LIS218984	89000406	0.00	1.22	7.54	24.86	31.14	21.59	10.48	2.32	0.68	0.17	1.81	3.12	4.93
LIS-21	Stamford Harbor	8/22/89	LIS218985	89000407	0.00	1.37	7.09	19.50	33.15	24.67	10.62	2.56	0.85	0.20	1.87	3.17	5.04
LIS-21	Stamford Harbor	8/22/89	LIS218986	89000408	0.00	1.64	10.28	16.86	30.37	26.50	10.72	2.39	0.99	0.27	1.80	3.19	4.99
LIS-21	Stamford Harbor	8/22/89	LIS218987	89000409	0.00	0.77	4.18	16.93	40.03	29.21	6.95	1.28	0.58	0.07	1.87	3.23	5.10
LIS-22	Long Neck Point	8/19/88	LIS228	90000396	0.00	1.13	7.79	17.12	28.35	23.60	12.83	4.57	3.65	0.96	1.98	3.26	5.24
LIS-23	Norwalk River Mouth	8/19/88	LIS238	90000528	0.00	1.01	6.93	18.00	29.84	23.82	14.29	2.99	2.25	0.87	1.95	3.25	5.20
LIS-24	Southport	8/22/88	LIS248	90000291	0.00	0.56	6.47	16.23	29.07	27.42	13.44	4.15	2.15	0.51	1.96	3.32	5.29
LIS-25	Black Rock Harbor	8/25/89	LIS2589S12	89000413	0.00	2.09	11.00	25.22	31.38	20.63	6.25	2.36	0.97	0.10	1.80	2.94	4.74
LIS-25	Black Rock Harbor	8/25/89	LIS2589S11	89000412	0.00	2.58	12.67	26.55	28.68	19.73	6.57	1.99	1.13	0.12	1.77	2.89	4.66
LIS-25	Black Rock Harbor	8/25/89	LIS2589S2	89000411	0.00	2.62	10.85	24.56	33.84	18.84	7.29	1.26	0.74	0.00	1.78	2.91	4.69
LIS-25	Black Rock Harbor	8/25/89	LIS2589S21	89000414	0.00	3.44	16.01	24.44	27.66	19.37	5.94	1.72	1.22	0.20	1.74	2.81	4.55
LIS-25	Black Rock Harbor	8/25/89	LIS2589S22	89000415	0.00	3.59	16.06	25.54	29.70	17.52	5.14	1.40	0.91	0.15	1.72	2.78	4.49
LIS-25	Black Rock Harbor	8/25/89	LIS2589S31	89000416	0.00	2.23	13.20	22.32	29.04	24.21	6.15	1.76	0.95	0.13	1.78	2.95	4.72
LIS-25	Black Rock Harbor	8/25/89	LIS2589S32	89000417	0.00	1.79	10.13	21.60	31.16	24.49	7.81	1.84	1.04	0.14	1.79	3.07	4.86
LIS-25	Black Rock Harbor	8/25/89	LIS2589S41	89000418	0.00	1.68	10.11	20.79	33.81	23.07	6.83	2.02	1.55	0.33	1.80	3.08	4.88
LIS-25	Black Rock Harbor	8/25/89	LIS2589S42	89000419	0.00	1.49	9.26	21.45	34.40	23.49	6.28	1.75	1.53	0.35	1.80	3.08	4.88
LIS-25	Black Rock Harbor	8/25/89	LIS2589S51	89000410	0.00	4.47	10.83	25.09	29.57	21.10	6.73	1.47	0.75	0.00	1.72	2.91	4.63
LIS-26	Bridgeport Harbor	8/23/89	LIS2689S1	89000385	0.00	1.24	9.54	26.49	33.89	20.48	6.54	1.26	0.46	0.11	1.79	2.96	4.76
LIS-26	Bridgeport Harbor	8/23/89	LIS2689S2	89000386	0.00	3.52	16.07	26.88	28.68	17.80	4.89	1.47	0.65	0.03	1.71	2.76	4.47
LIS-26	Bridgeport Harbor	8/23/89	LIS2689S3	89000387	0.00	5.37	21.77	34.18	23.16	10.99	3.05	0.84	0.55	0.09	1.61	2.50	4.12
LIS-26	Bridgeport Harbor	8/23/89	LIS2689S4	89000388	0.00	6.45	24.62	35.34	19.10	9.91	2.96	0.95	0.59	0.09	1.57	2.43	4.00
LIS-26	Bridgeport Harbor	8/23/89	LIS2689S5	89000389	0.00	5.40	20.19	31.35	22.72	12.87	4.88	1.58	0.77	0.24	1.65	2.59	4.24
LIS-26	Bridgeport Harbor	8/23/89	LIS2689S6	89000390	0.00	3.23	5.84	12.16	24.79	25.15	15.58	8.37	4.57	0.32	2.04	3.37	5.41
LIS-26	Bridgeport Harbor	8/23/89	LIS2689S7	89000391	0.00	3.80	15.41	30.76	23.87	15.23	6.78	2.40	1.56	0.20	1.71	2.78	4.49
LIS-26	Bridgeport Harbor	8/23/89	LIS2689S8	89000392	0.00	28.64	11.91	30.87	18.33	4.53	4.37	1.33	0.00	0.00	1.63	1.89	3.52
LIS-27	Housatonic River mouth	8/22/88	LIS278	90000525	0.00	1.51	17.12	38.29	19.50	8.48	6.53	4.56	3.56	0.45	1.78	2.72	4.51
LIS-28	Milford Harbor	8/23/88	LIS288	90000284	0.00	0.69	7.59	13.95	29.37	27.51	12.99	4.57	2.71	0.62	2.00	3.29	5.30
LIS-29	Stratford Shoal	8/25/88	LIS298	90000283	0.00	1.77	8.19	20.17	25.78	20.75	14.91	4.22	2.90	1.32	1.99	3.16	5.15
LIS-30	New Haven Harbor	8/24/89	LIS3089S1	89000378	0.00	0.15	7.88	23.86	27.86	21.80	14.15	3.46	0.77	0.07	1.92	3.14	5.07
LIS-30	New Haven Harbor	8/24/89	LIS3089S2	89000379	0.00	1.18	19.93	40.18	23.78	10.71	2.85	0.85	0.49	0.04	1.71	2.53	4.24
LIS-30	New Haven Harbor	8/24/89	LIS3089S3	89000380	0.00	3.44	14.32	24.97	24.79	17.07	11.28	2.24	1.19	0.71	1.84	2.84	4.67
LIS-30	New Haven Harbor	8/24/89	LIS3089S4	89000381	0.00	0.00	8.19	22.80	30.86	25.56	10.53	1.14	0.90	0.00	1.91	3.09	5.00
LIS-30	New Haven Harbor	8/24/89	LIS3089S5	89000382	0.00	1.75	8.70	18.14	18.59	22.75	17.87	7.58	3.06	1.55	1.98	3.35	5.33

6.2-1 (cont.)

Sampling Station	Location	Date Sampled	Navsa ID	NEA ID	Homologs, Weight % Mono	Di	Tri	Tetra	Penta	Hexa	Hepta	Octa	Nona	Deca	aCl/BP	m+pCl/BP	Tot Cl/B
LIS-30	New Haven Harbor	8/24/89	LIS306956	89000383	0.00	2.31	19.57	34.31	20.54	9.91	7.92	3.74	1.51	0.19	1.84	2.57	4.42
LIS-30	New Haven Harbor	8/24/89	LIS306957	89000384	0.00	3.21	21.22	37.70	21.79	9.08	5.10	1.16	0.65	0.08	1.78	2.42	4.20
LIS-31	Central Dumping Ground	8/23/88	LIS31S	90000288	0.00	1.16	10.07	23.06	29.19	20.12	10.99	3.15	1.70	0.57	1.90	3.05	4.94
LIS-32	Western Dumping Ground	9/27/88	LIS32S	90000550	0.00	1.43	7.54	16.07	26.62	22.97	18.81	4.64	1.93	0.00	1.97	3.29	5.26

6.2-1 (con't)

Sampling Station	Location	Date Sampled	Harza ID	NEA ID	Heptachlo	Heptachlo Epoxide	Gamma chlordane	Alpha chlordane	Trans-nonachlor	Technical chlordane	Á-BHC	ß-BHC	4,4'-DDD	4,4'-DDE
HR-1	Troy Dam	6/1/90	2365	90000899										
HR-1	Troy Dam	6/1/90	2366	90000900										
HR-1	Troy Dam	10/19/90	5665	90002320										
HR-1	Troy Dam	10/19/90	5666	90002321										
HR-1	Troy Dam	9/25/89	269T	89000608	3.48	0.22	6.53	4.32	1.70	40.48	0.08	0.05	2.73	6.70
HR-1	Troy Dam	9/25/89	270T	89000609	0.66	0.05	1.94	0.92	0.30	10.19	0.02	0.02	0.48	1.30
HR-1	Troy Dam	7/25/88	HR1S	90000611										
HR-1	Troy Dam	9/25/89	271T	89000610	0.31	N.D.	0.72	0.31	0.10	3.65	0.03	0.03	0.13	0.40
HR-2	Watervliet	9/25/89	266T	89000611	3.57	2.55	8.58	4.02	1.40	45.16	0.08	0.05	2.17	6.50
HR-2	Watervliet	9/25/89	267T	89000612	1.24	0.15	1.96	2.43	2.10	20.94	0.06	0.04	3.06	4.70
HR-2	Watervliet	9/25/89	268T	89000613	4.92	0.78	0.07	0.14	0.50	2.29	0.04	0.03	2.40	2.80
HR-3	Albany	7/25/88	HR3S	90000541										
HR-4	North Albany Turning Basin	7/26/88	HR4S	90000544										
HR-5	South Albany Turning Basin	9/25/89	211	89000614	3.29	0.15	0.17	1.49	1.00	8.58	0.08	0.05	2.75	5.90
HR-5	South Albany Turning Basin	9/25/89	212	89000616	2.98	0.12	0.64	1.32	1.10	9.87	0.05	0.03	1.89	3.80
HR-5	South Albany Turning Basin	9/25/89	214	89000615	11.02	0.26	9.47	5.53	2.10	55.16	0.04	0.03	1.47	5.90
HR-5	South Albany Turning Basin	9/25/89	215	89000617	3.21	0.18	7.25	4.54	2.10	44.81	0.05	0.04	1.88	6.70
HR-5	South Albany Turning Basin	9/25/89	216	89000618	0.26	0.07	1.10	1.14	0.90	10.13	0.02	0.03	3.27	3.20
HR-5	South Albany Turning Basin	9/25/89	259T	89000619	3.81	1.01	9.22	5.70	2.50	56.19	0.08	0.04	2.70	8.50
HR-5	South Albany Turning Basin	9/25/89	260T	89000620	9.56	0.18	33.21	12.77	3.70	160.26	0.06	0.04	3.23	23.60
HR-5	South Albany Turning Basin	9/25/89	261T	89000621	3.35	0.31	0.11	1.26	0.80	7.00	0.07	0.05	3.04	5.30
HR-5	South Albany Turning Basin	9/25/89	262T	89000622	9.08	0.24	0.22	1.95	2.20	14.10	0.06	0.04	1.91	6.30
HR-5	South Albany Turning Basin	9/25/89	263T	89000623	3.57	0.30	1.39	1.88	1.20	14.42	0.07	0.04	2.59	4.80
HR-5	South Albany Turning Basin	9/25/89	264T	89000624	2.80	1.81	0.87	1.93	1.40	13.55	0.07	0.06	3.04	3.00
HR-6	Campbell Island	7/26/88	HR6S	90000416										
HR-7	Ravena	7/27/88	HR7S	90000402										
HR-8	Coxsackie	9/19/89	169	89000598	1.25	0.53	2.83	2.17	0.90	19.03	0.08	0.06	2.52	5.20
HR-8	Coxsackie	9/19/89	170	89000599	1.18	0.09	4.59	1.64	0.50	21.71	0.04	0.02	1.16	2.80
HR-8	Coxsackie	9/19/89	171	89000600	1.65	0.39	3.30	1.90	0.70	19.03	0.08	0.05	2.41	4.80
HR-8	Coxsackie	9/19/89	172	89000601	3.16	0.21	0.28	1.01	0.80	6.74	0.08	0.05	2.78	4.90
HR-8	Coxsackie	9/19/89	173	89000602	8.07	0.33	7.91	5.96	2.10	51.52	0.09	0.06	2.71	11.60
HR-8	Coxsackie	9/19/89	174	89000603	2.98	0.09	4.28	2.56	1.00	25.29	0.02	0.03	0.88	3.50
HR-8	Coxsackie	9/19/89	175	89000604	0.57	0.02	0.94	0.54	0.20	5.42	0.03	0.03	0.21	0.80
HR-8	Coxsackie	9/19/89	176	89000605	1.56	0.14	3.58	2.15	0.80	21.06	0.04	0.02	0.96	3.70
HR-8	Coxsackie	9/19/89	177	89000606	10.51	0.07	16.84	9.05	2.30	90.94	0.05	0.03	1.39	11.50
HR-8	Coxsackie	9/19/89	178	89000607	0.60	0.18	1.23	0.73	0.20	6.97	0.02	0.02	0.30	1.10
HR-9	Stockport Creek	7/27/88												
HR-10	Hudson-Athens	7/27/88	HR10S	90000422	N.D.	0.01	0.01	0.3	0.27	1.87	0.01	0.02	1.06	1.14
HR-11	Catskill Creek	7/28/88	HR11S	90000539	N.D.	N.D.	0.04	0.68	0.25	3.13	0.03	0.03	2.36	2.7

6.2-1 (con't)

Sampling Station	Location	Date Sampled	Horze ID	NEA ID	Heptachlo	Heptachlo Epoxide	Gamma chlordane	Alpha chlordane	Trans-nonachlor	Technical chlordane	Á-BHC	B-BHC	4,4'-DDD	4,4'-DDE
HR-12	Inbocht Bay	7/28/88	HR12S	90000277	N.D.	0.01	0.02	0.19	0.05	0.84	0.01	0.02	0.17	0.49
HR-13	Esopus Creek	7/28/88	HR13S	90000274	N.D.	N.D.	0.07	0.4	0.23	2.26	0.03	0.03	2.38	3.56
HR-14	South Bay-Annandale	7/28/88	HR14S	90000540	N.D.	N.D.	0.03	0.41	0.38	2.65	0.03	0.04	2.29	2.75
HR-15	Kingston	9/14/89	153	89000589	2.25	0.14	5.52	3.39	1.30	32.94	0.04	0.04	8.55	11.00
HR-15	Kingston	9/14/89	155	89000590	7.08	0.22	16.21	11.07	2.80	97.03	0.09	0.10	11.81	17.60
HR-15	Kingston	9/14/89	156	89000591	8.47	0.13	0.09	3.64	1.30	16.23	0.06	0.03	7.65	15.50
HR-15	Kingston	9/14/89	158	89000592	2.02	0.17	4.76	2.61	0.90	26.68	0.04	0.03	2.75	5.10
HR-15	Kingston	9/14/89	159	89000593	3.49	0.24	6.47	4.28	1.60	39.84	0.06	0.05	3.69	9.80
HR-15	Kingston	9/14/89	160	89000594	0.36	N.D.	0.78	0.54	0.40	5.55	0.02	0.03	4.86	6.60
HR-15	Kingston	9/14/89	161	89000595	0.59	0.06	1.32	0.95	1.00	10.55	0.02	0.04	10.44	12.30
HR-15	Kingston	9/14/89	162	89000596	0.81	0.06	0.04	0.42	0.20	2.13	0.02	0.02	1.75	3.00
HR-15	Kingston	9/14/89	168	89000597	3.38	0.06	9.10	3.84	1.10	45.29	0.07	0.03	3.13	7.10
HR-16	Esopus Meadows Point	7/29/88	HR16S	90000423										
HR-17	Hyde Park	10/4/89	315T	89000679	2.63	0.08	4.74	3.72	1.40	31.81	0.03	0.03	6.73	12.60
HR-17	Hyde Park	10/4/89	316T	89000680	2.54	0.16	5.58	4.22	1.40	36.13	0.06	0.05	5.32	11.40
HR-17	Hyde Park	10/4/89	317T	89000681	2.22	0.19	5.40	4.94	1.60	38.52	0.06	0.05	6.60	11.00
HR-17	Hyde Park	10/4/89	318T	89000682	1.36	0.18	4.58	2.34	0.60	24.26	0.04	0.05	3.58	5.90
HR-17	Hyde Park	10/4/89	319T	89000683	0.02	N.D.	0.42	0.10	N.D.	1.68	N.D.	0.01	0.07	0.20
HR-17	Hyde Park	10/4/89	320T	89000684	2.53	0.03	4.04	3.32	1.10	27.29	0.02	0.02	3.24	6.60
HR-17	Hyde Park	10/4/89	321	89000685	1.84	0.18	1.08	2.65	1.00	15.26	0.08	0.06	6.87	7.30
HR-17	Hyde Park	10/4/89	322	89000686	30.93	0.45	47.12	37.17	10.40	305.45	0.15	0.07	14.26	44.50
HR-17	Hyde Park	10/4/89	323	89000687	2.45	0.19	4.76	3.40	1.20	30.19	0.05	0.05	4.76	8.40
HR-17	Hyde Park	10/4/89	324	89000688	5.54	0.31	8.99	7.60	2.60	61.90	0.10	0.06	7.53	19.60
HR-18	North Poughkeepsie	7/29/88	HR18S	90000275										
HR-19	Poughkeepsie	10/4/89	305T	89000669	2.37	0.15	5.13	3.66	1.30	32.55	0.05	0.02	6.63	15.10
HR-19	Poughkeepsie	10/4/89	306T	89000670	2.51	0.08	5.15	4.02	1.50	34.42	0.04	0.04	5.80	12.20
HR-19	Poughkeepsie	10/4/89	307T	89000671	3.37	0.22	6.66	5.00	1.60	42.77	0.06	0.04	7.12	15.70
HR-19	Poughkeepsie	10/4/89	308T	89000672	3.45	0.71	5.87	4.53	1.80	39.35	0.08	0.06	7.39	16.50
HR-19	Poughkeepsie	10/4/89	309T	89000673	3.61	0.17	8.75	6.90	2.20	57.58	0.08	0.06	7.39	16.50
HR-19	Poughkeepsie	10/4/89	310T	89000674	2.69	0.19	7.54	5.58	1.90	48.45	0.08	0.06	7.45	15.50
HR-19	Poughkeepsie	10/4/89	311T	89000675	40.48	1.15	0.09	0.94	0.30	4.29	0.09	0.04	12.51	18.10
HR-19	Poughkeepsie	10/4/89	312T	89000676	15.13	0.36	21.94	19.54	6.00	153.16	0.07	0.05	13.15	27.70
HR-19	Poughkeepsie	10/4/89	313T	89000677	2.54	0.26	6.21	4.81	1.70	41.93	0.07	0.05	6.76	12.60
HR-19	Poughkeepsie	10/4/89	314T	89000678	2.61	0.13	6.26	4.50	1.30	38.90	0.05	0.04	4.60	11.70
HR-20	New Hamburg	7/30/88	HR20S	90000425										
HR-21	Newburgh	7/30/88	HR21S	90000391										
HR-22	Cornwall on Hudson	7/30/88	HR22S	90000427										
HR-23	Foundry Cove	7/30/88	HR23S	90000420										
HR-24	Con Hook	8/25/89	258	89000535	7.03	0.24	0.11	2.37	0.90	10.90	0.06	0.06	15.42	12.70

6.2-1 (con't)

Sampling Station	Location	Date Sampled	Marza ID	NEA ID	Heptachlo	Heptachlo Epoxide	Gamma chlordane	Alpha chlordane	Trans-nonachlor	Technical chlordane	A-BHC	B-BHC	4,4'-DDD	4,4'-DDE
HR-24	Con Hook	8/25/89	397	89000530	3.53	0.35	0.19	3.92	0.90	16.16	0.09	0.06	9.83	10.80
HR-24	Con Hook	8/25/89	398	89000531	2.58	0.28	0.35	2.55	1.20	13.23	0.08	0.06	8.96	11.60
HR-24	Con Hook	8/25/89	399	89000532	0.45	0.25	0.85	0.03	0.10	3.16	0.03	0.04	0.71	1.70
HR-24	Con Hook	8/25/89	400	89000533	0.09	0.04	0.15	0.14	0.10	1.26	0.02	0.06	1.94	0.70
HR-24	Con Hook	8/25/89	257	89000534	0.63	0.05	0.06	0.34	0.10	1.61	0.02	0.04	1.74	2.40
HR-25	Iona Island	7/29/88	HR25S	90000414										
HR-26	Peekskill Bay	7/29/88	HR26S	90000555										
HR-27	Indian Point	7/29/88	HR27S	90000554										
HR-28	Stony Point Bay	8/25/89	393	89000504	2.94	0.22	0.09	0.95	0.20	4.00	0.09	0.07	12.00	12.10
HR-28	Stony Point Bay	8/25/89	394	89000505	3.22	0.19	13.10	11.85	8.60	108.23	0.16	0.11	9.72	75.20
HR-28	Stony Point Bay	8/25/89	395	89000506	2.70	0.15	0.05	0.63	0.50	3.81	0.09	0.08	8.97	11.30
HR-28	Stony Point Bay	8/25/89	408	89000503	0.14	0.02	0.09	0.04	N.D.	0.42	0.02	0.04	1.33	0.30
HR-28	Stony Point Bay	8/25/89	259	89000536	2.82	0.17	0.65	4.08	1.30	19.45	0.07	0.06	8.87	14.50
HR-28	Stony Point Bay	8/25/89	260	89000515	2.45	0.31	0.41	2.10	1.10	11.65	0.13	0.14	11.24	11.90
HR-28	Stony Point Bay	8/25/89	261	89000516	1.91	0.33	0.26	2.18	0.90	10.77	0.09	0.05	7.42	9.50
HR-28	Stony Point Bay	8/25/89	262	89000517	0.47	0.01	0.05	0.32	0.20	1.84	0.04	0.04	1.83	2.10
HR-28	Stony Point Bay	8/25/89	263	89000518	1.63	0.16	0.15	2.48	0.90	11.39	0.08	0.05	7.08	8.00
HR-28	Stony Point Bay	8/25/89	264	89000519	2.63	0.20	0.08	2.74	0.90	12.00	0.08	0.05	7.88	12.70
HR-29	Haverstraw	7/29/88	HR29S	90000546										
HR-30	Croton Bay	7/27/88	HR30S	90000285										
HR-31	Tarrytown	7/27/88	HR31S	90000276										
HR-32	Tappan Zee Bridge - Nyack	7/27/88	HR32S	90000545										
HR-33	Piermont	7/27/88	HR33S	90000542	N.D.	N.D.	0.05	0.62	0.20	2.81	0.09	0.07	7.85	3.75
HR-34	Yonkers	7/28/88	HR34S	90000282										
HR-35	Spuyten Devvil Creek	7/28/88	HR35S	90000279										
HR-36	George Washington Bridge	7/28/88	HR36S	90000401										
HR-37	North Bergen	7/28/88	HR37S	90000417										
HR-38	The Battery	8/19/89	285	89000309	2.70	1.10	27.40	20.30	5.30	170.97	0.20	0.08	21.52	21.20
HR-38	The Battery	8/19/89	286	89000310	2.71	0.72	2.74	6.03	4.60	43.13	0.28	0.15	63.02	18.30
HR-38	The Battery	8/19/89	287	89000311	3.99	0.97	1.73	6.56	4.20	40.29	0.26	0.16	21.84	19.00
HR-38	The Battery	8/19/89	288	89000312	10.60	2.03	1.89	2.86	0.20	15.97	0.11	0.05	0.82	1.90
HR-38	The Battery	8/19/89	289	89000313	6.51	1.05	2.80	6.65	4.60	45.32	0.13	0.09	17.41	21.90
HR-38	The Battery	8/19/89	290	89000314	0.65	N.D.	0.63	0.72	0.60	6.29	0.06	0.06	2.69	2.90
HR-38	The Battery	8/19/89	291	89000315	1.33	5.81	4.66	4.68	1.70	35.61	0.14	0.09	12.74	9.50
HR-38	The Battery	8/19/89	292	89000520	0.75	0.02	0.73	0.91	1.20	9.16	N.D.	0.02	1.07	6.00
HR-38	The Battery	8/21/89	293	89000525	2.47	0.97	4.80	5.88	4.30	48.32	0.20	0.09	17.86	17.00
HR-38	The Battery	8/21/89	294	89000526	2.94	0.80	4.18	5.80	4.30	46.06	0.20	0.09	20.85	18.40
NYH-1	Bayonne	7/28/88	NYH-1S	90000281	0.82	0.37	6.09	2.64	14.48	74.87	0.44	0.17	471.62	293.45

6.2-1 (con't)

Sampling Station	Location	Date Sampled	Horze ID	NEA ID	Heptachlo	Heptachlo Epoxide	Gamma chlordane	Alpha chlordane	Trans-nonachlor	Technical chlordane	A-BHC	B-BHC	4,4'-DDD	4,4'-DDE
NYH-2	Gowanus Canal	8/21/89	441	89000507	2.14	5.08	5.91	4.52	3.90	46.23	0.25	0.09	29.99	18.20
NYH-2	Gowanus Canal	8/21/89	443	89000509	7.79	4.43	21.77	9.68	5.90	120.48	1.05	0.27	263.47	86.20
NYH-2	Gowanus Canal	8/21/89	444	89000510	32.89	3.83	92.38	91.98	37.10	714.39	0.40	0.33	556.63	117.00
NYH-2	Gowanus Canal	8/21/89	445	89000511	187.84	4.42	97.41	193.96	111.70	1300.23	0.40	0.41	93.60	283.00
NYH-2	Gowanus Canal	8/21/89	446	89000512	10.01	2.98	18.83	25.49	17.30	198.77	0.41	0.25	95.41	41.20
NYH-2	Gowanus Canal	8/21/89	447	89000513	6.25	2.47	9.65	12.49	9.10	100.77	0.34	0.12	46.10	33.20
NYH-2	Gowanus Canal	8/21/89	448	89000514	1.27	1.63	4.13	4.07	2.80	35.48	0.21	0.09	20.50	10.30
NYH-2	Gowanus Canal	8/21/89	449	89000521	1.70	0.99	7.18	7.07	5.40	63.39	0.20	0.09	18.15	18.30
NYH-3	Verrazano Narrows Bridge	8/23/89	426	89000556	1.71	3.93	2.03	2.99	2.70	24.90	0.16	0.09	14.15	14.80
NYH-3	Verrazano Narrows Bridge	8/23/89	427	89000557	0.91	0.84	1.37	2.68	2.10	19.84	0.13	0.06	12.45	9.10
NYH-3	Verrazano Narrows Bridge	8/23/89	429	89000559	8.71	3.82	1.03	4.32	1.70	22.74	0.21	0.07	9.76	10.10
NYH-3	Verrazano Narrows Bridge	8/23/89	430	89000560	0.54	0.42	0.59	1.12	1.10	9.06	0.06	0.04	5.41	5.30
NYH-3	Verrazano Narrows Bridge	8/23/89	433	89000474	1.59	1.44	3.46	4.49	4.20	39.19	0.09	0.06	9.18	9.80
NYH-3	Verrazano Narrows Bridge	8/23/89	435	89000476	8.43	1.81	13.73	16.05	14.50	142.84	0.22	0.07	24.49	41.90
NYH-3	Verrazano Narrows Bridge	8/23/89	438	89000478	1.17	0.57	2.54	3.78	3.00	30.06	0.14	0.08	12.48	10.80
NYH-3	Verrazano Narrows Bridge	8/23/89	439	89000553	0.38	0.31	0.80	1.14	0.80	8.84	0.08	0.05	4.76	3.40
NYH-3	Verrazano Narrows Bridge	8/23/89	440	89000554	3.20	1.69	7.06	9.66	6.20	73.94	0.51	0.17	29.02	24.70
NYH-4	Midland Beach	8/23/90	401	89000466	1.56	0.25	3.60	4.23	3.20	35.58	0.24	0.09	18.63	15.50
NYH-4	Midland Beach	8/23/89	403	89000467	1.71	0.24	3.86	4.90	3.90	40.84	0.29	0.09	18.94	15.90
NYH-4	Midland Beach	8/23/89	406	89000470	0.94	0.64	3.41	3.81	2.80	32.32	0.21	0.08	13.07	11.70
NYH-4	Midland Beach	8/23/89	431	89000463	2.15	0.54	7.26	7.17	5.00	62.68	0.24	0.10	17.00	16.20
NYH-4	Midland Beach	8/23/89	432	89000465	1.22	0.41	3.08	4.14	3.00	32.97	0.30	0.18	19.55	16.10
NYH-4	Midland Beach	8/21/89	451	89000523	0.11	N.D.	0.87	0.71	2.08	11.81	0.01	0.02	1.46	1.00
NYH-5	Coney Island	8/23/88	NYH5S	90000437										
NYH-6	Great Kills Harbor	8/16/89	305	89000320	0.11	0.05	0.14	0.21	0.20	1.77	0.02	0.02	1.21	1.30
NYH-6	Great Kills Harbor	8/16/89	306	89000321	0.45	0.33	1.42	1.96	1.30	15.10	0.12	0.08	6.50	5.40
NYH-6	Great Kills Harbor	8/16/89	313	89000328	1.22	0.69	5.96	6.16	5.00	55.23	0.31	0.08	17.90	9.20
NYH-6	Great Kills Harbor	8/16/89	314	89000329	4.73	0.91	6.16	6.63	4.50	55.77	0.41	0.13	18.69	11.70
NYH-6	Great Kills Harbor	8/16/89	315	89000330	1.97	0.74	9.43	9.38	6.80	82.61	0.39	0.13	22.27	14.80
NYH-6	Great Kills Harbor	8/16/89	316	89000331	0.13	0.27	0.79	0.68	0.50	6.35	0.04	0.05	2.17	1.30
NYH-6	Great Kills Harbor	8/16/89	318	89000333	1.34	0.38	5.77	6.39	4.60	54.06	0.22	0.08	16.40	10.70
NYH-6	Great Kills Harbor	8/16/89	319	89000334	0.03	0.04	0.14	0.22	0.10	1.48	0.01	0.02	0.67	0.60
NYH-7	Seguine Point	8/30/88	NYH7S	90000385										
NYH-8	Raritan River mouth	8/16/89	297	89000300	1.11	0.15	2.09	1.59	1.10	15.42	0.09	0.07	7.18	9.90
NYH-8	Raritan River mouth	8/16/89	298	89000301	3.69	1.08	1.86	7.13	5.20	45.77	0.42	0.13	21.36	21.50
NYH-8	Raritan River mouth	8/16/89	299	89000302	3.67	0.70	19.43	12.46	11.00	138.35	0.26	0.09	132.53	61.00
NYH-8	Raritan River mouth	8/16/89	308	89000323	0.70	0.67	3.41	4.24	2.20	31.77	0.19	0.06	17.25	13.40
NYH-8	Raritan River mouth	8/16/89	309	89000324	1.53	0.63	0.38	0.17	N.D.	1.77	0.12	0.05	19.27	6.70
NYH-8	Raritan River mouth	8/16/89	311	89000326	0.66	0.47	3.16	4.32	2.60	32.52	0.16	0.08	12.11	11.00

6.2-1 (con't)

Sampling Station	Location	Date Sampled	Marza ID	NEA ID	Heptachlo	Heptachlo Epoxide	Gamma chlordane	Alpha chlordane	Trans-nonachlor	Technical chlordane	Á-BHC	B-BHC	4,4'-DDD	4,4'-DDE
NYH-8	Raritan River mouth	8/16/89	312	89000327	0.57	1.75	6.24	5.58	2.00	44.58	0.89	0.25	40.96	5.60
NYH-9	Woodbridge Creek	8/18/89	276	89000293	9.26	1.47	29.73	13.47	9.70	170.65	0.26	0.08	80.18	91.60
NYH-9	Woodbridge Creek	8/18/89	278	89000295	6.43	1.34	10.54	12.86	10.80	110.32	0.40	0.15	109.11	74.80
NYH-9	Woodbridge Creek	8/18/89	279	89000296	34.30	4.52	1.49	7.90	10.10	62.87	0.34	0.15	50.39	114.00
NYH-9	Woodbridge Creek	8/18/89	281	89000298	4.12	2.06	10.22	12.01	9.90	103.65	0.28	0.11	55.67	49.30
NYH-9	Woodbridge Creek	8/18/89	282	89000299	3.69	0.85	4.72	9.57	6.60	67.39	0.09	0.05	72.32	33.20
NYH-9	Woodbridge Creek	8/18/89	283	89000308	1.51	0.56	5.67	4.83	5.10	50.32	0.33	0.12	40.59	37.20
NYH-10	Fresh Kills	8/23/88	NYH10S	90000412										
NYH-11	Prells Island	9/19/88	NYH11S	90000407										
NYH-12	Newark Bay	8/18/89	265	89000305	1.48	0.32	0.59	1.09	1.70	10.90	0.08	0.04	15.86	12.20
NYH-12	Newark Bay	8/18/89	266	89000306	1.56	0.35	3.82	4.56	2.90	36.39	0.07	0.04	28.96	19.60
NYH-12	Newark Bay	8/18/89	267	89000307	1.63	0.45	2.62	3.21	2.40	26.55	0.08	0.05	20.87	16.70
NYH-12	Newark Bay	8/18/89	268	89000284	5.10	1.69	6.27	10.34	7.50	77.77	0.23	0.12	35.25	41.30
NYH-12	Newark Bay	8/18/89	269	89000285	6.00	1.65	13.39	16.30	10.50	129.65	0.24	0.11	37.59	47.60
NYH-12	Newark Bay	8/18/89	270	89000286	3.06	1.81	6.76	7.27	4.40	59.45	0.13	0.06	28.94	34.70
NYH-12	Newark Bay	8/18/89	271	89000287	6.20	3.42	16.03	19.03	12.00	151.81	0.27	0.15	49.79	52.60
NYH-12	Newark Bay	8/18/89	273	89000288	4.45	1.70	5.73	9.22	6.70	69.84	0.19	0.08	27.73	45.60
NYH-12	Newark Bay	8/18/89	274	89000289	3.17	0.95	4.60	6.88	5.10	53.48	0.11	0.06	26.26	28.80
NYH-12	Newark Bay	8/18/89	275	89000291	3.14	1.49	5.65	6.22	4.30	52.16	0.21	0.10	39.44	33.60
NYH-13	Wallabout Bay	8/25/88	NYH13S	90000399										
NYH-14	Newtown Creek	8/22/89	413	89000540	3.63	1.22	5.72	9.12	7.20	71.10	0.24	0.12	27.04	21.20
NYH-14	Newtown Creek	8/22/89	414	89000541	3.49	2.00	11.31	11.02	7.70	96.87	0.37	0.16	36.85	26.50
NYH-14	Newtown Creek	8/22/89	415	89000542	2.60	1.33	4.67	5.31	4.90	48.00	0.27	0.13	24.60	17.70
NYH-14	Newtown Creek	8/22/89	416	89000543	8.07	3.48	15.85	18.04	14.10	154.81	0.24	0.12	41.85	34.70
NYH-14	Newtown Creek	8/22/89	417	89000544	2.24	1.18	8.50	9.50	7.00	80.65	0.24	0.13	24.44	23.80
NYH-14	Newtown Creek	8/22/89	423	89000471	3.08	1.85	6.19	8.02	7.00	68.42	0.34	0.18	38.97	24.60
NYH-14	Newtown Creek	8/22/89	424	89000472	1.95	1.79	8.13	7.31	5.10	66.26	0.23	0.11	41.06	15.90
NYH-14	Newtown Creek	8/22/89	452	89000527	2.40	0.81	9.52	9.09	6.90	82.29	0.26	0.11	24.90	21.00
NYH-15	Wards Island	8/22/89	409	89000552	3.00	1.18	7.63	9.26	6.20	74.48	0.95	0.12	22.02	22.70
NYH-15	Wards Island	8/22/89	410	89000537	2.77	2.88	0.12	0.54	1.10	5.68	0.07	0.05	1.76	0.70
NYH-15	Wards Island	8/22/89	411	89000538	4.25	2.12	11.78	15.63	8.90	117.13	0.29	0.13	37.59	33.10
NYH-15	Wards Island	8/22/89	412	89000539	3.89	1.68	8.75	10.50	7.30	85.65	0.36	0.11	22.77	23.40
NYH-15	Wards Island	8/22/89	453	89000528	5.16	0.82	1.66	3.63	3.30	27.71	0.21	0.07	12.07	11.70
NYH-15	Wards Island	8/22/89	454	89000549	0.07	4.32	0.32	0.32	0.10	2.39	0.10	0.07	0.92	0.50
NYH-15	Wards Island	8/22/89	455	89000550	2.57	1.46	10.01	10.00	8.20	91.00	0.23	0.13	24.06	22.00
NYH-15	Wards Island	8/22/89	456	89000551	2.19	1.21	7.48	7.11	5.40	64.48	0.19	0.11	19.59	14.70
NYH-16	Harlem River	8/25/88	NYH16S	90000271										
NYH-17	College Point	8/25/88	NYH17S	90000424										
NYH-18	Throgs Neck Bridge	8/25/88	NYH18S	90000280										

6.2-1 (cont.)

Sampling Station	Location	Date Sampled	Narza ID	NEA ID	Heptachlo	Heptachlo Epoxide	Gamma chlordane	Alpha chlordane	Trans-nonachlor	Technical chlordane	Á-BHC	ß-BHC	4,4'-DDD	4,4'-DDE
NYH-19	Sandy Hook	10/12/90	5014	90002062										
NYH-19	Sandy Hook	10/12/90	5013	90002049										
NYH-19	Sandy Hook	10/12/90	5020	90002050										
NYH-19	Sandy Hook	10/12/90	5019	90002051										
NYH-19	Sandy Hook	10/12/90	5015	90002064										
NYH-20	Lower Bay													
NYH-21	Rockaway Inlet	10/30/90	5154	90002157										
NYH-21	Rockaway Inlet	10/30/90	5156	90002159										
NYH-21	Rockaway Inlet	10/30/90	5155	90002158										
NYH-21	Rockaway Inlet	10/30/90	5164	90002160										
NYH-21	Rockaway Inlet	10/30/90	5165	90002161										
NYH-21	Rockaway Inlet	10/30/90	5166	90002162										
NYH-22	Jamaica Bay-Beach Channel	09/11/90	3699	90001581										
NYH-22	Jamaica Bay-Beach Channel	09/10/90	3209	90001576										
NYH-22	Jamaica Bay-Beach Channel	09/10/90	3210	90001577										
NYH-22	Jamaica Bay-Beach Channel	09/10/90	3211	90001578										
NYH-22	Jamaica Bay-Beach Channel	09/10/90	3690	90001579										
NYH-22	Jamaica Bay-Beach Channel	09/10/90	3691	90001580										
NYH-23	Jamaica Bay - Canarsie	10/16/90	5056	90002063										
NYH-23	Jamaica Bay - Canarsie	10/16/90	5061	90002069										
NYH-23	Jamaica Bay - Canarsie	10/16/90	5063	90002070										
NYH-23	Jamaica Bay - Canarsie	10/16/90	5059	90002072										
NYH-23	Jamaica Bay - Canarsie	10/16/90	5060	90002065										
NYH-23	Jamaica Bay - Canarsie	10/16/90	5062	90002068										
NYH-23	Jamaica Bay - Canarsie	10/16/90	5064	90002071										
NYH-24	Harlem River - 207th St.	10/26/90	5151	90002060										
NYH-24	Harlem River - 207th St.	10/26/90	5149	90002061										
NYH-24	Harlem River - 207th St.	10/26/90	5148	90002058										
NYH-24	Harlem River - 207th St.	10/26/90	5150	90002059										
NYH-24	Harlem River - 207th St.	10/26/90	5145	90002057										
NYH-24	Harlem River - 207th St.	10/26/90	5146	90002066										
NYH-24	Harlem River - 207th St.	10/26/90	5147	90002067										
LIS-1	Little Neck Bay	8/24/89	381	89000495	0.28	0.24	0.81	0.80	0.60	7.13	0.03	0.04	2.44	1.80
LIS-1	Little Neck Bay	8/24/89	382	89000496	1.46	0.63	8.33	7.94	5.70	70.87	0.14	0.09	13.69	12.00
LIS-1	Little Neck Bay	8/24/89	383	89000497	0.55	0.26	4.30	4.88	3.70	41.55	0.02	0.03	4.29	4.80
LIS-1	Little Neck Bay	8/24/89	384	89000498	2.64	1.13	19.81	18.84	13.40	167.90	0.13	0.08	18.56	16.50
LIS-1	Little Neck Bay	8/24/89	386	89000500	1.47	1.39	10.69	10.25	8.50	94.97	0.14	0.08	14.73	13.20
LIS-1	Little Neck Bay	8/24/89	387	89000501	0.60	0.21	0.98	1.32	1.10	10.97	0.03	0.04	5.91	3.30
LIS-1	Little Neck Bay	8/24/89	388	89000502	3.13	3.24	10.89	11.81	8.70	101.29	0.13	0.06	20.32	18.60

6.2-1 (con't)

Sampling Station	Location	Date Sampled	Horse ID	NEA ID	Heptachlo	Heptachlo Epoxide	Gamma chlordane	Alpha chlordane	Trans-nonachlor	Technical chlordane	A-BHC	B-BHC	4,4'-DDD	4,4'-DDE
LIS-2	Eastchester Bay	9/17/88	LI828	90000353										
LIS-3	Manhasset Bay	8/26/88	LI838	90000373										
LIS-4	Sands Point	8/24/89	143	89000485	0.58	0.35	5.28	4.78	3.30	43.10	0.11	0.08	5.97	8.20
LIS-4	Sands Point	8/24/89	144	89000486	1.47	0.75	10.12	6.82	5.30	71.74	0.09	0.07	10.51	14.90
LIS-4	Sands Point	8/24/89	377	89000491	2.43	0.37	7.76	7.05	4.90	63.58	0.12	0.08	7.87	13.20
LIS-4	Sands Point	8/24/89	378	89000492	0.66	0.86	4.67	3.59	2.70	35.35	0.13	0.09	7.90	11.10
LIS-4	Sands Point	8/24/89	379	89000493	0.81	0.69	3.50	3.53	3.80	34.94	0.11	0.06	10.64	8.70
LIS-5	Hempstead Harbor	8/23/88	LIS5S	90000389										
LIS-6	Matinecock Point	8/27/88	LIS6S	90000419										
LIS-7	Bayville	8/27/88	LIS7S	90000410										
LIS-8	Oyster Bay	8/27/88	LIS8S	90000415										
LIS-9	Lloyd Point	8/26/88	LIS9S	90000289										
LIS-10	Target Rock	8/22/89	LIS1089S1	89000393	0.20	0.03	1.01	0.82	0.70	8.16	0.01	0.02	3.32	2.00
LIS-10	Target Rock	8/22/89	LIS1089S2	89000394	0.63	0.28	4.37	3.87	3.30	37.23	0.08	0.05	8.97	7.30
LIS-10	Target Rock	8/22/89	LIS1089S3	89000395	1.48	0.76	9.99	7.39	6.50	77.03	0.05	0.04	20.52	16.40
LIS-10	Target Rock	8/22/89	LIS1089S4	89000396	1.28	1.14	10.81	10.05	7.60	91.81	0.04	0.04	28.14	14.10
LIS-10	Target Rock	8/22/89	LIS1089S5	89000397	0.41	0.52	2.15	1.49	1.10	15.29	0.05	0.04	5.17	5.20
LIS-10	Target Rock	8/22/89	LIS1089S6	89000398	0.17	0.08	1.35	0.99	0.80	10.13	0.03	0.03	2.79	4.80
LIS-10	Target Rock	8/22/89	LIS1089S7	89000399	5.48	1.87	23.46	24.44	21.10	222.58	0.05	0.05	65.14	14.80
LIS-10	Target Rock	8/22/89	LIS1089S8	89000400	10.00	1.94	47.22	39.64	29.80	376.32	0.07	0.06	68.49	22.20
LIS-10	Target Rock	8/22/89	LIS1089S9	89000401	0.13	0.18	1.49	1.18	1.10	12.16	0.05	0.03	2.43	4.40
LIS-10	Target Rock	8/22/89	LIS1089S10	89000402	0.34	0.36	2.21	1.73	1.30	16.90	0.06	0.05	2.98	5.30
LIS-11	Eatons Neck	8/26/88	LIS11S	90000531										
LIS-12	Nissequogue River	8/26/88	LIS12S	90000524										
LIS-13	Stony Brook Harbor	8/26/88	LIS13S	90000286										
LIS-14	Port Jefferson Harbor	8/26/88	LIS14S	90000392										
LIS-15	Shoreham	8/25/88	LIS15S	90000400										
LIS-16	Glen Island	8/17/88	LIS16S	90000396										
LIS-17	Echo Bay - New Rochelle	8/17/89	LIS1789S1	89000266	2.50	1.84	17.80	15.60	12.10	146.77	0.10	0.18	37.40	17.80
LIS-17	Echo Bay - New Rochelle	8/17/89	LIS1789S3	89000267	3.11	4.01	75.46	76.26	56.00	670.06	0.20	0.19	91.29	50.90
LIS-17	Echo Bay - New Rochelle	8/17/89	LIS1789S4	89000268	8.12	2.53	45.01	33.14	25.90	335.65	0.23	0.26	847.97	38.70
LIS-17	Echo Bay - New Rochelle	8/17/89	LIS1789S2	89000269	1.84	1.44	12.18	9.89	7.40	95.06	0.12	0.13	26.25	19.70
LIS-18	Mamaroneck Harbor	8/17/89	LIS1889S1	89000258	0.51	0.33	1.85	1.65	1.40	15.81	0.08	0.10	4.51	4.70
LIS-18	Mamaroneck Harbor	8/17/89	LIS1889S2	89000259	21.80	6.54	142.08	145.95	90.90	1222.35	1.67	3.91	129.47	58.20
LIS-18	Mamaroneck Harbor	8/17/89	LIS1889S3	89000260	12.80	4.53	64.61	57.73	36.80	513.35	0.49	0.84	93.41	36.10
LIS-18	Mamaroneck Harbor	8/17/89	LIS1889S4	89000261	1.27	0.63	10.51	8.41	5.90	80.06	0.44	0.95	27.70	16.90
LIS-18	Mamaroneck Harbor	8/17/89	LIS1889S5	89000262	3.19	1.01	15.41	14.16	10.20	128.29	0.28	0.34	28.57	16.90
LIS-18	Mamaroneck Harbor	8/17/89	LIS1889S6	89000263	0.25	0.13	1.33	1.11	0.70	10.13	0.03	0.05	2.64	2.40
LIS-18	Mamaroneck Harbor	8/17/89	LIS1889S7	89000264	3.85	1.14	24.54	22.35	15.20	200.29	0.32	0.52	46.55	25.50

6.2-1 (con't)

Sampling Station	Location	Date Sampled	Marza ID	NEA ID	Heptachlo	Heptachlo Epoxide	Gamma chlordane	Alpha chlordane	Trans-nonachlor	Technical chlordane	A-BHC	B-BHC	4,4'-DDD	4,4'-DDE
LIS-18	Hamaroneck Harbor	8/17/89	LIS188988	89000265	0.38	0.49	2.76	3.21	2.60	27.69	0.06	0.10	13.35	5.20
LIS-19	Port Chester Harbor	8/19/88	LIS198	90000534										
LIS-20	Flat Neck Point	8/19/88	LIS208	90000290										
LIS-21	Stamford Harbor	8/22/89	LIS218981	89000403	22.23	3.87	39.87	58.27	42.20	452.71	0.32	0.50	274.88	49.80
LIS-21	Stamford Harbor	8/22/89	LIS218982	89000404	24.38	3.46	24.95	49.60	41.80	379.32	0.18	0.21	140.83	52.80
LIS-21	Stamford Harbor	8/22/89	LIS218983	89000405	14.83	6.45	53.05	60.01	27.10	452.13	0.18	0.16	1744.95	151.50
LIS-21	Stamford Harbor	8/22/89	LIS218984	89000406	2.17	0.73	10.01	9.52	8.40	90.10	0.05	0.06	34.92	15.10
LIS-21	Stamford Harbor	8/22/89	LIS218985	89000407	1.51	2.59	7.04	6.06	5.10	58.71	0.09	0.09	32.13	9.80
LIS-21	Stamford Harbor	8/22/89	LIS218986	89000408	2.11	1.08	10.83	9.12	8.20	90.81	0.05	0.10	17.30	11.60
LIS-21	Stamford Harbor	8/22/89	LIS218987	89000409	2.69	0.99	6.73	10.31	7.00	77.55	0.06	0.05	20.49	7.20
LIS-22	Long Neck Point	8/19/88	LIS22S	90000394										
LIS-23	Norwalk River Mouth	8/19/88	LIS23S	90000528										
LIS-24	Southport	8/22/88	LIS24S	90000291										
LIS-25	Black Rock Harbor	8/25/89	LIS2589S12	89000413	22.57	5.22	25.02	45.08	25.40	308.06	0.25	0.17	451.55	81.80
LIS-25	Black Rock Harbor	8/25/89	LIS2589S11	89000412	18.87	1.36	39.28	62.12	33.7	435.81	0.72	0.14	504.93	87.9
LIS-25	Black Rock Harbor	8/25/89	LIS2589S2	89000411	0.32	0.08	0.24	0.41	0.40	3.39	0.02	0.02	1.35	1.90
LIS-25	Black Rock Harbor	8/25/89	LIS2589S21	89000414	20.51	6.07	73.85	66.60	45.20	598.87	0.15	0.30	1090.37	146.10
LIS-25	Black Rock Harbor	8/25/89	LIS2589S22	89000415	19.94	5.46	62.04	59.46	41.00	524.19	0.25	0.17	1304.87	154.60
LIS-25	Black Rock Harbor	8/25/89	LIS2589S31	89000416	5.37	2.25	21.60	24.15	19.00	208.87	0.07	0.10	40.85	34.70
LIS-25	Black Rock Harbor	8/25/89	LIS2589S32	89000417	2.15	1.51	18.28	22.47	17.60	188.23	0.05	0.13	32.45	32.70
LIS-25	Black Rock Harbor	8/25/89	LIS2589S41	89000418	4.27	1.12	2.97	3.28	3.20	30.48	0.05	0.05	39.15	30.00
LIS-25	Black Rock Harbor	8/25/89	LIS2589S42	89000419	9.04	1.47	4.83	2.30	3.40	33.97	0.07	0.06	71.93	43.10
LIS-25	Black Rock Harbor	8/25/89	LIS2589S51	89000410	0.16	0.11	0.25	0.42	0.40	3.45	0.02	0.03	7.98	1.30
LIS-26	Bridgeport Harbor	8/23/89	LIS2689S1	89000385	1.36	2.09	1.72	2.19	2.20	19.71	0.06	0.04	5.19	2.30
LIS-26	Bridgeport Harbor	8/23/89	LIS2689S2	89000386	10.34	2.47	11.64	34.06	21.90	218.06	0.23	0.11	44.25	28.30
LIS-26	Bridgeport Harbor	8/23/89	LIS2689S3	89000387	18.26	15.13	3.06	2.72	1.80	24.45	0.16	0.10	23.86	15.80
LIS-26	Bridgeport Harbor	8/23/89	LIS2689S4	89000388	8.07	0.59	0.34	2.32	1.00	11.81	0.09	0.09	15.98	15.30
LIS-26	Bridgeport Harbor	8/23/89	LIS2689S5	89000389	0.39	0.07	0.41	0.66	0.40	4.74	0.03	0.05	1.97	1.50
LIS-26	Bridgeport Harbor	8/23/89	LIS2689S6	89000390	10.31	2.15	35.44	43.58	25.20	336.19	0.08	0.08	29.73	8.90
LIS-26	Bridgeport Harbor	8/23/89	LIS2689S7	89000391	1.21	0.58	0.76	1.55	1.20	11.32	0.06	0.06	6.09	4.00
LIS-26	Bridgeport Harbor	8/23/89	LIS2689S8	89000392	0.22	N.D.	0.01	0.02	N.D.	0.10	N.D.	0.03	0.35	N.D.
LIS-27	Housatonic River mouth	8/22/88	LIS27S	90000525										
LIS-28	Milford Harbor	8/23/88	LIS28S	90000284										
LIS-29	Stratford Shoal	8/25/88	LIS29S	90000283										
LIS-30	New Haven Harbor	8/24/89	LIS3089S1	89000378	4.64	1.64	16.62	5.26	6.50	91.55	0.08	0.07	14.99	14.10
LIS-30	New Haven Harbor	8/24/89	LIS3089S2	89000379	33.04	1.03	58.54	31.27	27.90	379.71	0.11	0.10	50.67	45.20
LIS-30	New Haven Harbor	8/24/89	LIS3089S3	89000380	3.03	0.69	23.83	14.31	11.30	159.48	0.10	0.27	26.56	20.30
LIS-30	New Haven Harbor	8/24/89	LIS3089S4	89000381	0.74	0.62	8.42	2.52	2.70	44.00	0.06	1.30	3.25	4.00
LIS-30	New Haven Harbor	8/24/89	LIS3089S5	89000382	1.54	0.15	6.68	2.12	1.60	33.55	0.03	0.03	2.04	2.90

6.2-1 (con't)

Sampling Station	Location	Date Sampled	Norze ID	NEA ID	Heptachlo	Heptachlo Epoxide	Gamma chlordane	Alpha chlordane	Trans-nonachlor	Technical chlordane	Á-BHC	B-BHC	4,4'-DDD	4,4'-DDE
LIS-30	New Haven Harbor	8/24/89	LIS3089S6	89000383	8.23	2.03	38.80	12.65	15.10	214.68	0.08	0.06	23.20	18.80
LIS-30	New Haven Harbor	8/24/89	LIS3089S7	89000384	4.34	1.94	14.97	5.01	8.20	90.90	0.15	0.08	25.73	9.40
LIS-31	Central Dumping Ground	8/23/88	LIS31S	90000288										
LIS-32	Western Dumping Ground	9/27/88	LIS32S	90000550										

6.2-1 (con't)

Sampling Station	Location	Date Sampled	Normo ID	NEA ID	4,4'-DDT	DDD:DDE
HR-1	Troy Dam	6/1/90	2365	90000899		
HR-1	Troy Dam	6/1/90	2366	90000900		
HR-1	Troy Dam	10/19/90	5665	90002320		
HR-1	Troy Dam	10/19/90	5666	90002321		
HR-1	Troy Dam	9/25/89	269T	89000608	1.40	0.41
HR-1	Troy Dam	9/25/89	270T	89000609	0.30	0.37
HR-1	Troy Dam	7/25/88	HR1S	90000411		
HR-1	Troy Dam	9/25/89	271T	89000610	0.20	0.33
HR-2	Watervliet	9/25/89	266T	89000611	1.70	0.33
HR-2	Watervliet	9/25/89	267T	89000612	1.90	0.65
HR-2	Watervliet	9/25/89	268T	89000613	1.80	0.86
HR-3	Albany	7/25/88	HR3S	90000541		
HR-4	North Albany Turning Basin	7/26/88	HR4S	90000544		
HR-5	South Albany Turning Basin	9/25/89	211	89000614	1.20	0.47
HR-5	South Albany Turning Basin	9/25/89	212	89000616	1.20	0.50
HR-5	South Albany Turning Basin	9/25/89	214	89000615	1.60	0.25
HR-5	South Albany Turning Basin	9/25/89	215	89000617	2.10	0.28
HR-5	South Albany Turning Basin	9/25/89	216	89000618	0.60	1.02
HR-5	South Albany Turning Basin	9/25/89	259T	89000619	2.50	0.32
HR-5	South Albany Turning Basin	9/25/89	260T	89000620	3.70	0.14
HR-5	South Albany Turning Basin	9/25/89	261T	89000621	2.20	0.57
HR-5	South Albany Turning Basin	9/25/89	262T	89000622	1.20	0.30
HR-5	South Albany Turning Basin	9/25/89	263T	89000623	1.20	0.54
HR-5	South Albany Turning Basin	9/25/89	264T	89000624	1.00	1.01
HR-6	Campbell Island	7/26/88	HR6S	90000416		
HR-7	Ravena	7/27/88	HR7S	90000402		
HR-8	Coxsackie	9/19/89	169	89000598	4.40	0.48
HR-8	Coxsackie	9/19/89	170	89000599	1.20	0.41
HR-8	Coxsackie	9/19/89	171	89000600	1.10	0.50
HR-8	Coxsackie	9/19/89	172	89000601	3.20	0.57
HR-8	Coxsackie	9/19/89	173	89000602	2.40	0.23
HR-8	Coxsackie	9/19/89	174	89000603	0.90	0.25
HR-8	Coxsackie	9/19/89	175	89000604	0.20	0.26
HR-8	Coxsackie	9/19/89	176	89000605	1.20	0.26
HR-8	Coxsackie	9/19/89	177	89000606	2.40	0.12
HR-8	Coxsackie	9/19/89	178	89000607	0.20	0.27
HR-9	Stockport Creek	7/27/88				
HR-10	Hudson-Athens	7/27/88	HR10S	90000422	0.59	0.93
HR-11	Catskill Creek	7/28/88	HR11S	90000539	1.19	0.87

6.2-1 (cont.)

Sampling Station	Location	Date Sampled	Horza ID	NEA ID	4,4'-DDT	DDD:DDE
HR-12	Inbocht Bay	7/28/88	HR12S	90000277	0.19	0.35
HR-13	Esopus Creek	7/28/88	HR13S	90000274	2.17	0.67
HR-14	South Bay-Annandale	7/28/88	HR14S	90000540	1.77	0.83
HR-15	Kingston	9/14/89	153	89000589	1.90	0.78
HR-15	Kingston	9/14/89	155	89000590	5.20	0.67
HR-15	Kingston	9/14/89	156	89000591	2.20	0.49
HR-15	Kingston	9/14/89	158	89000592	0.80	0.54
HR-15	Kingston	9/14/89	159	89000593	1.90	0.38
HR-15	Kingston	9/14/89	160	89000594	1.80	0.74
HR-15	Kingston	9/14/89	161	89000595	1.70	0.85
HR-15	Kingston	9/14/89	162	89000596	0.60	0.58
HR-15	Kingston	9/14/89	168	89000597	1.20	0.44
HR-16	Esopus Meadows Point	7/29/88	HR16S	90000423		
HR-17	Hyde Park	10/4/89	315T	89000679	2.30	0.53
HR-17	Hyde Park	10/4/89	316T	89000680	2.10	0.47
HR-17	Hyde Park	10/4/89	317T	89000681	2.10	0.60
HR-17	Hyde Park	10/4/89	318T	89000682	0.70	0.61
HR-17	Hyde Park	10/4/89	319T	89000683	N.D.	0.35
HR-17	Hyde Park	10/4/89	320T	89000684	1.20	0.49
HR-17	Hyde Park	10/4/89	321	89000685	1.00	0.94
HR-17	Hyde Park	10/4/89	322	89000686	8.70	0.32
HR-17	Hyde Park	10/4/89	323	89000687	1.30	0.57
HR-17	Hyde Park	10/4/89	324	89000688	3.10	0.38
HR-18	North Poughkeepsie	7/29/88	HR18S	90000275		
HR-19	Poughkeepsie	10/4/89	305T	89000669	2.10	0.44
HR-19	Poughkeepsie	10/4/89	306T	89000670	2.10	0.48
HR-19	Poughkeepsie	10/4/89	307T	89000671	2.30	0.45
HR-19	Poughkeepsie	10/4/89	308T	89000672	2.60	0.45
HR-19	Poughkeepsie	10/4/89	309T	89000673	2.60	0.45
HR-19	Poughkeepsie	10/4/89	310T	89000674	2.30	0.48
HR-19	Poughkeepsie	10/4/89	311T	89000675	2.50	0.69
HR-19	Poughkeepsie	10/4/89	312T	89000676	7.00	0.47
HR-19	Poughkeepsie	10/4/89	313T	89000677	9.60	0.54
HR-19	Poughkeepsie	10/4/89	314T	89000678	25.10	0.39
HR-20	New Hamburg	7/30/88	HR20S	90000425		
HR-21	Newburgh	7/30/88	HR21S	90000391		
HR-22	Cornwall on Hudson	7/30/88	HR22S	90000427		
HR-23	Foundry Cove	7/30/88	HR23S	90000420		
HR-24	Con Hook	8/25/89	258	89000535	7.70	1.21

6.2-1 (con't)

Sampling Station	Location	Date Sampled	Marza ID	MEA ID	4,4'-DDT	DDD:DDE
HR-24	Con Hook	8/25/89	397	89000530	2.50	0.91
HR-24	Con Hook	8/25/89	398	89000531	2.30	0.77
HR-24	Con Hook	8/25/89	399	89000532	2.80	0.42
HR-24	Con Hook	8/25/89	400	89000533	N.D.	2.77
HR-24	Con Hook	8/25/89	257	89000534	1.20	0.73
HR-25	Iona Island	7/29/88	HR25S	90000414		
HR-26	Peekskill Bay	7/29/88	HR26S	90000555		
HR-27	Indian Point	7/29/88	HR27S	90000554		
HR-28	Stony Point Bay	8/25/89	393	89000504	1.40	0.99
HR-28	Stony Point Bay	8/25/89	394	89000505	10.30	0.13
HR-28	Stony Point Bay	8/25/89	395	89000506	1.60	0.79
HR-28	Stony Point Bay	8/25/89	408	89000503	N.D.	4.43
HR-28	Stony Point Bay	8/25/89	259	89000536	3.80	0.61
HR-28	Stony Point Bay	8/25/89	260	89000515	2.80	0.94
HR-28	Stony Point Bay	8/25/89	261	89000516	1.30	0.78
HR-28	Stony Point Bay	8/25/89	262	89000517	0.10	0.87
HR-28	Stony Point Bay	8/25/89	263	89000518	1.60	0.89
HR-28	Stony Point Bay	8/25/89	264	89000519	2.20	0.62
HR-29	Haverstraw	7/29/88	HR29S	90000546		
HR-30	Croton Bay	7/27/88	HR30S	90000285		
HR-31	Tarrytown	7/27/88	HR31S	90000276		
HR-32	Tappan Zee Bridge - Nyack	7/27/88	HR32S	90000545		
HR-33	Piermont	7/27/88	HR33S	90000542	0.96	2.09
HR-34	Yonkers	7/28/88	HR34S	90000282		
HR-35	Spuyten Devvil Creek	7/28/88	HR35S	90000279		
HR-36	George Washington Bridge	7/28/88	HR36S	90000401		
HR-37	North Bergen	7/28/88	HR37S	90000417		
HR-38	The Battery	8/19/89	285	89000309	6.30	1.02
HR-38	The Battery	8/19/89	286	89000310	64.10	3.44
HR-38	The Battery	8/19/89	287	89000311	2.90	1.15
HR-38	The Battery	8/19/89	288	89000312	0.30	0.43
HR-38	The Battery	8/19/89	289	89000313	2.30	0.79
HR-38	The Battery	8/19/89	290	89000314	1.50	0.93
HR-38	The Battery	8/19/89	291	89000315	1.00	1.34
HR-38	The Battery	8/19/89	292	89000520	3.30	0.18
HR-38	The Battery	8/21/89	293	89000525	2.20	1.05
HR-38	The Battery	8/21/89	294	89000526	2.90	1.13
NYH-1	Bayonne	7/28/88	NYH-1S	90000281	43.89	1.61

6.2-1 (con't)

Sampling Station	Location	Date Sampled	Marza ID	NEA ID	4,4'-DDT	DDD:DDE
NYH-2	Gowanus Canal	8/21/89	441	89000507	4.60	1.65
NYH-2	Gowanus Canal	8/21/89	443	89000509	7.80	3.06
NYH-2	Gowanus Canal	8/21/89	444	89000510	61.90	4.76
NYH-2	Gowanus Canal	8/21/89	445	89000511	107.60	0.33
NYH-2	Gowanus Canal	8/21/89	446	89000512	11.40	2.32
NYH-2	Gowanus Canal	8/21/89	447	89000513	3.80	1.39
NYH-2	Gowanus Canal	8/21/89	448	89000514	1.50	1.99
NYH-2	Gowanus Canal	8/21/89	449	89000521	5.20	0.99
NYH-3	Verrazano Narrows Bridge	8/23/89	426	89000556	1.20	0.96
NYH-3	Verrazano Narrows Bridge	8/23/89	427	89000557	1.70	1.37
NYH-3	Verrazano Narrows Bridge	8/23/89	429	89000559	6.10	0.97
NYH-3	Verrazano Narrows Bridge	8/23/89	430	89000560	0.80	1.02
NYH-3	Verrazano Narrows Bridge	8/23/89	433	89000474	2.20	0.94
NYH-3	Verrazano Narrows Bridge	8/23/89	435	89000476	4.80	0.58
NYH-3	Verrazano Narrows Bridge	8/23/89	438	89000478	1.10	1.16
NYH-3	Verrazano Narrows Bridge	8/23/89	439	89000553	0.40	1.40
NYH-3	Verrazano Narrows Bridge	8/23/89	440	89000554	9.70	1.17
NYH-4	Midland Beach	8/23/90	401	89000466	1.80	1.20
NYH-4	Midland Beach	8/23/89	403	89000467	7.20	1.19
NYH-4	Midland Beach	8/23/89	406	89000470	1.70	1.12
NYH-4	Midland Beach	8/23/89	431	89000463	15.00	1.05
NYH-4	Midland Beach	8/23/89	432	89000465	3.00	1.21
NYH-4	Midland Beach	8/21/89	451	89000523	0.20	1.46
NYH-5	Coney Island	8/23/88	NYH5S	90000437		
NYH-6	Great Kills Harbor	8/16/89	305	89000320	N.D.	0.93
NYH-6	Great Kills Harbor	8/16/89	306	89000321	0.50	1.20
NYH-6	Great Kills Harbor	8/16/89	313	89000328	1.70	1.95
NYH-6	Great Kills Harbor	8/16/89	314	89000329	1.10	1.60
NYH-6	Great Kills Harbor	8/16/89	315	89000330	1.30	1.50
NYH-6	Great Kills Harbor	8/16/89	316	89000331	0.20	1.67
NYH-6	Great Kills Harbor	8/16/89	318	89000333	2.40	1.53
NYH-6	Great Kills Harbor	8/16/89	319	89000334	N.D.	1.12
NYH-7	Seguine Point	8/30/88	NYH7S	90000385		
NYH-8	Raritan River mouth	8/16/89	297	89000300	N.D.	0.73
NYH-8	Raritan River mouth	8/16/89	298	89000301	4.30	0.99
NYH-8	Raritan River mouth	8/16/89	299	89000302	5.90	2.17
NYH-8	Raritan River mouth	8/16/89	308	89000323	0.70	1.29
NYH-8	Raritan River mouth	8/16/89	309	89000324	9.60	2.88
NYH-8	Raritan River mouth	8/16/89	311	89000326	0.90	1.10

6.2-1 (cont.)

Sampling Station	Location	Date Sampled	Marza ID	NEA ID	4,4'-DDT	DDD:DDE
NYH-8	Raritan River mouth	8/16/89	312	89000327	0.90	7.31
NYH-9	Woodbridge Creek	8/18/89	276	89000293	14.20	0.88
NYH-9	Woodbridge Creek	8/18/89	278	89000295	142.30	1.46
NYH-9	Woodbridge Creek	8/18/89	279	89000296	102.80	0.44
NYH-9	Woodbridge Creek	8/18/89	281	89000298	14.10	1.13
NYH-9	Woodbridge Creek	8/18/89	282	89000299	33.60	2.18
NYH-9	Woodbridge Creek	8/18/89	283	89000308	4.50	1.09
NYH-10	Fresh Kills	8/23/88	NYH10S	90000412		
NYH-11	Pralls Island	9/19/88	NYH11S	90000407		
NYH-12	Newark Bay	8/18/89	265	89000305	6.00	1.30
NYH-12	Newark Bay	8/18/89	266	89000306	13.80	1.48
NYH-12	Newark Bay	8/18/89	267	89000307	4.00	1.25
NYH-12	Newark Bay	8/18/89	268	89000284	9.80	0.85
NYH-12	Newark Bay	8/18/89	269	89000285	4.10	0.79
NYH-12	Newark Bay	8/18/89	270	89000286	5.40	0.83
NYH-12	Newark Bay	8/18/89	271	89000287	7.60	0.95
NYH-12	Newark Bay	8/18/89	273	89000288	1.80	0.61
NYH-12	Newark Bay	8/18/89	274	89000289	6.70	0.91
NYH-12	Newark Bay	8/18/89	275	89000291	2.80	1.17
NYH-13	Wallabout Bay	8/25/88	NYH13S	90000399		
NYH-14	Newtown Creek	8/22/89	413	89000540	7.00	1.28
NYH-14	Newtown Creek	8/22/89	414	89000541	10.70	1.39
NYH-14	Newtown Creek	8/22/89	415	89000542	6.10	1.39
NYH-14	Newtown Creek	8/22/89	416	89000543	1.70	1.21
NYH-14	Newtown Creek	8/22/89	417	89000544	13.80	1.03
NYH-14	Newtown Creek	8/22/89	423	89000471	5.40	1.58
NYH-14	Newtown Creek	8/22/89	424	89000472	9.20	2.58
NYH-14	Newtown Creek	8/22/89	452	89000527	5.70	1.19
NYH-15	Wards Island	8/22/89	409	89000552	64.40	0.97
NYH-15	Wards Island	8/22/89	410	89000537	2.00	2.51
NYH-15	Wards Island	8/22/89	411	89000538	2.80	1.14
NYH-15	Wards Island	8/22/89	412	89000539	7.30	0.97
NYH-15	Wards Island	8/22/89	453	89000528	1.80	1.03
NYH-15	Wards Island	8/22/89	454	89000549	0.10	1.84
NYH-15	Wards Island	8/22/89	455	89000550	3.80	1.09
NYH-15	Wards Island	8/22/89	456	89000551	2.60	1.33
NYH-16	Harlem River	8/25/88	NYH16S	90000271		
NYH-17	College Point	8/25/88	NYH17S	90000424		
NYH-18	Throgs Neck Bridge	8/25/88	NYH18S	90000280		

9.2-1 (con't)

Sampling Station	Location	Date Sampled	Narza ID	NEA ID	4,4'-DDT	DDD:DDE
NYH-19	Sandy Hook	10/12/90	5014	90002062		
NYH-19	Sandy Hook	10/12/90	5013	90002049		
NYH-19	Sandy Hook	10/12/90	5020	90002050		
NYH-19	Sandy Hook	10/12/90	5019	90002051		
NYH-19	Sandy Hook	10/12/90	5015	90002064		
NYH-20	Lower Bay					
NYH-21	Rockaway Inlet	10/30/90	5154	90002157		
NYH-21	Rockaway Inlet	10/30/90	5156	90002159		
NYH-21	Rockaway Inlet	10/30/90	5155	90002158		
NYH-21	Rockaway Inlet	10/30/90	5164	90002160		
NYH-21	Rockaway Inlet	10/30/90	5165	90002161		
NYH-21	Rockaway Inlet	10/30/90	5166	90002162		
NYH-22	Jamaica Bay-Beach Channel	09/11/90	3699	90001581		
NYH-22	Jamaica Bay-Beach Channel	09/10/90	3209	90001576		
NYH-22	Jamaica Bay-Beach Channel	09/10/90	3210	90001577		
NYH-22	Jamaica Bay-Beach Channel	09/10/90	3211	90001578		
NYH-22	Jamaica Bay-Beach Channel	09/10/90	3690	90001579		
NYH-22	Jamaica Bay-Beach Channel	09/10/90	3691	90001580		
NYH-23	Jamaica Bay - Canarsie	10/16/90	5056	90002063		
NYH-23	Jamaica Bay - Canarsie	10/16/90	5061	90002069		
NYH-23	Jamaica Bay - Canarsie	10/16/90	5063	90002070		
NYH-23	Jamaica Bay - Canarsie	10/16/90	5059	90002072		
NYH-23	Jamaica Bay - Canarsie	10/16/90	5060	90002065		
NYH-23	Jamaica Bay - Canarsie	10/16/90	5062	90002068		
NYH-23	Jamaica Bay - Canarsie	10/16/90	5064	90002071		
NYH-24	Harlem River - 207th St.	10/26/90	5151	90002060		
NYH-24	Harlem River - 207th St.	10/26/90	5149	90002061		
NYH-24	Harlem River - 207th St.	10/26/90	5148	90002058		
NYH-24	Harlem River - 207th St.	10/26/90	5150	90002059		
NYH-24	Harlem River - 207th St.	10/26/90	5145	90002057		
NYH-24	Harlem River - 207th St.	10/26/90	5146	90002066		
NYH-24	Harlem River - 207th St.	10/26/90	5147	90002067		
LIS-1	Little Neck Bay	8/24/89	381	89000495	0.50	1.36
LIS-1	Little Neck Bay	8/24/89	382	89000496	6.50	1.14
LIS-1	Little Neck Bay	8/24/89	383	89000497	1.90	0.89
LIS-1	Little Neck Bay	8/24/89	384	89000498	10.10	1.12
LIS-1	Little Neck Bay	8/24/89	386	89000500	2.00	1.12
LIS-1	Little Neck Bay	8/24/89	387	89000501	0.60	1.79
LIS-1	Little Neck Bay	8/24/89	388	89000502	4.40	1.09

6.2-1 (con't)

Sampling Station	Location	Date Sampled	Norze ID	NEA ID	4,4'-DDT	DDD:DDE
LIB-2	Eastchester Bay	9/17/88	LIB2B	90000393		
LIS-3	Manhasset Bay	8/26/88	LIB3B	90000373		
LIS-4	Sands Point	8/24/89	143	89000485	1.20	0.73
LIS-4	Sands Point	8/24/89	144	89000486	0.60	0.71
LIS-4	Sands Point	8/24/89	377	89000491	1.70	0.60
LIS-4	Sands Point	8/24/89	378	89000492	1.90	0.71
LIS-4	Sands Point	8/24/89	379	89000493	1.80	1.22
LIS-5	Hempstead Harbor	8/23/88	LIS5S	90000389		
LIS-6	Matinecock Point	8/27/88	LIS6S	90000419		
LIS-7	Bayville	8/27/88	LIS7S	90000410		
LIS-8	Oyster Bay	8/27/88	LIS8S	90000415		
LIS-9	Lloyd Point	8/26/88	LIS9S	90000289		
LIS-10	Target Rock	8/22/89	LIS1089S1	89000393	0.40	1.66
LIS-10	Target Rock	8/22/89	LIS1089S2	89000394	0.80	1.23
LIS-10	Target Rock	8/22/89	LIS1089S3	89000395	1.90	1.25
LIS-10	Target Rock	8/22/89	LIS1089S4	89000396	3.30	2.00
LIS-10	Target Rock	8/22/89	LIS1089S5	89000397	0.90	0.99
LIS-10	Target Rock	8/22/89	LIS1089S6	89000398	1.20	0.58
LIS-10	Target Rock	8/22/89	LIS1089S7	89000399	1.50	4.40
LIS-10	Target Rock	8/22/89	LIS1089S8	89000400	5.40	3.09
LIS-10	Target Rock	8/22/89	LIS1089S9	89000401	0.60	0.55
LIS-10	Target Rock	8/22/89	LIS1089S10	89000402	0.80	0.56
LIS-11	Eatons Neck	8/26/88	LIS11S	90000531		
LIS-12	Nissequogue River	8/26/88	LIS12S	90000524		
LIS-13	Stony Brook Harbor	8/26/88	LIS13S	90000286		
LIS-14	Port Jefferson Harbor	8/26/88	LIS14S	90000392		
LIS-15	Shoreham	8/25/88	LIS15S	90000400		
LIS-16	Glen Island	8/17/88	LIS16S	90000396		
LIS-17	Echo Bay - New Rochelle	8/17/89	LIS1789S1	89000266	2.10	2.10
LIS-17	Echo Bay - New Rochelle	8/17/89	LIS1789S3	89000267	20.60	1.79
LIS-17	Echo Bay - New Rochelle	8/17/89	LIS1789S4	89000268	12.70	21.91
LIS-17	Echo Bay - New Rochelle	8/17/89	LIS1789S2	89000269	8.00	1.33
LIS-18	Mamaroneck Harbor	8/17/89	LIS1889S1	89000258	1.20	0.96
LIS-18	Mamaroneck Harbor	8/17/89	LIS1889S2	89000259	37.70	2.22
LIS-18	Mamaroneck Harbor	8/17/89	LIS1889S3	89000260	9.30	2.59
LIS-18	Mamaroneck Harbor	8/17/89	LIS1889S4	89000261	11.40	1.64
LIS-18	Mamaroneck Harbor	8/17/89	LIS1889S5	89000262	3.10	1.69
LIS-18	Mamaroneck Harbor	8/17/89	LIS1889S6	89000263	0.40	1.10
LIS-18	Mamaroneck Harbor	8/17/89	LIS1889S7	89000264	9.60	1.83

6.2-1 (cont.)

Sampling Station	Location	Date Sampled	Norza ID	NEA ID	4,4'-DDT	DDD:DDE
LIS-18	Mamaroneck Harbor	8/17/89	LIS18S8S8	89000265	1.70	2.57
LIS-19	Port Chester Harbor	8/19/88	LIS19S	90000534		
LIS-20	Flat Neck Point	8/19/88	LIS20S	90000290		
LIS-21	Stamford Harbor	8/22/89	LIS21S9S1	89000403	24.20	6.00
LIS-21	Stamford Harbor	8/22/89	LIS21S9S2	89000404	7.50	2.67
LIS-21	Stamford Harbor	8/22/89	LIS21S9S3	89000405	10.40	11.52
LIS-21	Stamford Harbor	8/22/89	LIS21S9S4	89000406	2.40	2.31
LIS-21	Stamford Harbor	8/22/89	LIS21S9S5	89000407	1.00	3.28
LIS-21	Stamford Harbor	8/22/89	LIS21S9S6	89000408	7.00	1.49
LIS-21	Stamford Harbor	8/22/89	LIS21S9S7	89000409	4.60	2.85
LIS-22	Long Neck Point	8/19/88	LIS22S	90000394		
LIS-23	Norwalk River Mouth	8/19/88	LIS23S	90000528		
LIS-24	Southport	8/22/88	LIS24S	90000291		
LIS-25	Black Rock Harbor	8/25/89	LIS2589S12	89000413	10.10	5.52
LIS-25	Black Rock Harbor	8/25/89	LIS2589S11	89000412	12.8	5.74
LIS-25	Black Rock Harbor	8/25/89	LIS2589S2	89000411	0.10	0.71
LIS-25	Black Rock Harbor	8/25/89	LIS2589S21	89000414	46.20	7.46
LIS-25	Black Rock Harbor	8/25/89	LIS2589S22	89000415	20.60	8.44
LIS-25	Black Rock Harbor	8/25/89	LIS2589S31	89000416	8.50	1.18
LIS-25	Black Rock Harbor	8/25/89	LIS2589S32	89000417	3.40	0.99
LIS-25	Black Rock Harbor	8/25/89	LIS2589S41	89000418	1.70	1.31
LIS-25	Black Rock Harbor	8/25/89	LIS2589S42	89000419	2.60	1.67
LIS-25	Black Rock Harbor	8/25/89	LIS2589S51	89000410	0.10	6.14
LIS-26	Bridgeport Harbor	8/23/89	LIS2689S1	89000385	1.60	2.26
LIS-26	Bridgeport Harbor	8/23/89	LIS2689S2	89000386	9.90	1.56
LIS-26	Bridgeport Harbor	8/23/89	LIS2689S3	89000387	4.10	1.51
LIS-26	Bridgeport Harbor	8/23/89	LIS2689S4	89000388	1.50	1.04
LIS-26	Bridgeport Harbor	8/23/89	LIS2689S5	89000389	0.20	1.31
LIS-26	Bridgeport Harbor	8/23/89	LIS2689S6	89000390	10.50	3.34
LIS-26	Bridgeport Harbor	8/23/89	LIS2689S7	89000391	0.70	1.52
LIS-26	Bridgeport Harbor	8/23/89	LIS2689S8	89000392	N.D.	
LIS-27	Housatonic River mouth	8/22/88	LIS27S	90000525		
LIS-28	Milford Harbor	8/23/88	LIS28S	90000284		
LIS-29	Stratford Shoal	8/25/88	LIS29S	90000283		
LIS-30	New Haven Harbor	8/24/89	LIS3089S1	89000378	3.40	1.06
LIS-30	New Haven Harbor	8/24/89	LIS3089S2	89000379	11.60	1.12
LIS-30	New Haven Harbor	8/24/89	LIS3089S3	89000380	10.70	1.31
LIS-30	New Haven Harbor	8/24/89	LIS3089S4	89000381	0.80	0.81
LIS-30	New Haven Harbor	8/24/89	LIS3089S5	89000382	1.40	0.70

6.2-1 (con't)

Sampling Station	Location	Date Sampled	Marza ID	NEA ID	4,4'-DDT	DDD:DDE
LIS-30	New Haven Harbor	8/24/89	LIS3089S6	89000383	10.30	1.23
LIS-30	New Haven Harbor	8/24/89	LIS3089S7	89000384	1.40	2.74
LIS-31	Central Dumping Ground	8/23/88	LIS31S	90000288		
LIS-32	Western Dumping Ground	9/27/88	LIS32S	90000550		

6.2-1 (con't)

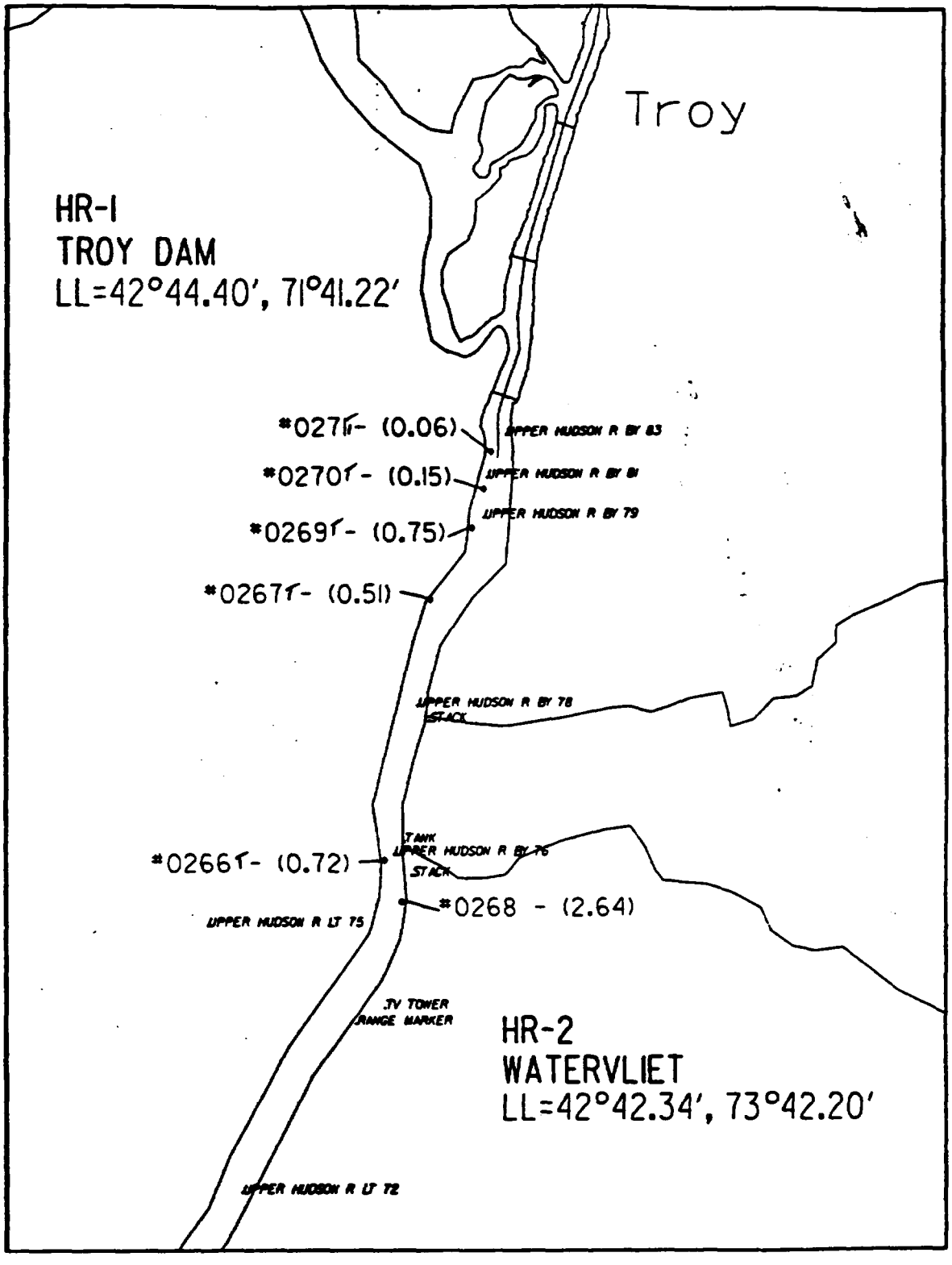
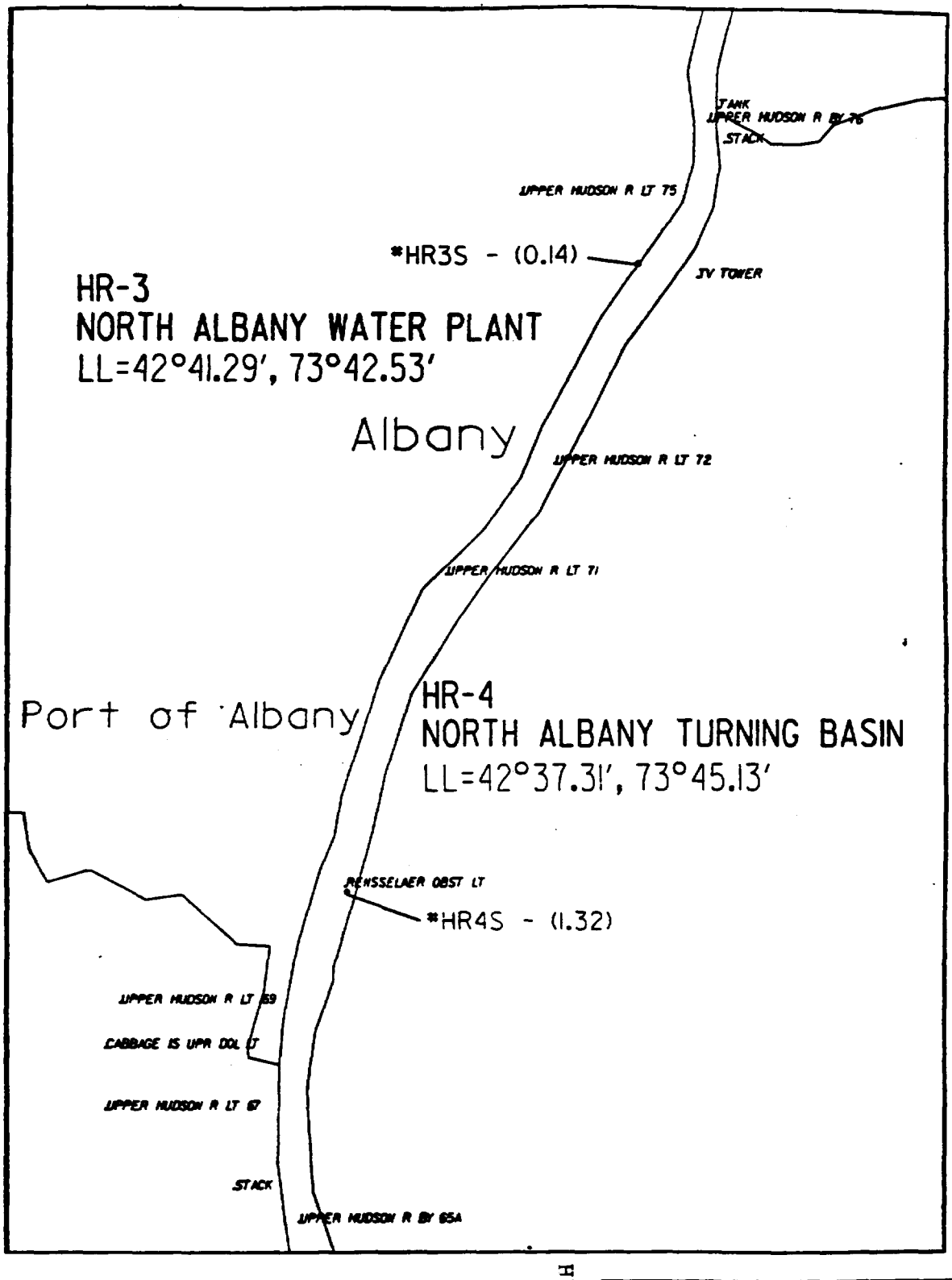


Table 6.2-2

HRP 002 0701

GENERAL ELECTRIC	
SEDIMENT SAMPLING SITES	
HARZA ENGINEERING CO., CHICAGO APPROVED_____	
DATE OCTOBER 1991	DWG. NO. HR-1 AND 2



6.2-2 (con't)

HRP 002 0702

GENERAL ELECTRIC	
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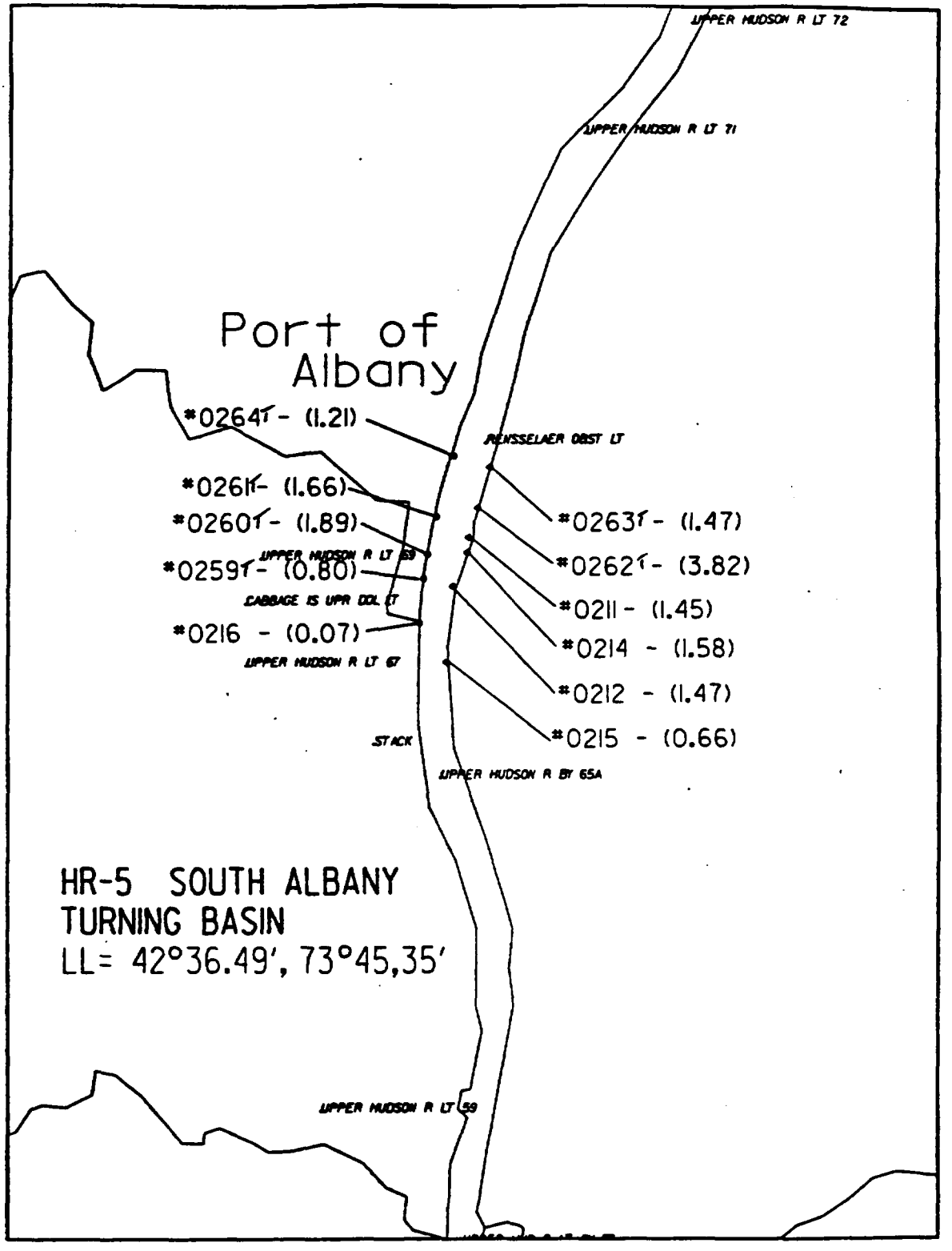
HARZA ENGINEERING CO., CHICAGO APPROVED_____	
DATE OCTOBER 1991	DWG. NO. HR-3 AND 4



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HRP 002 0703

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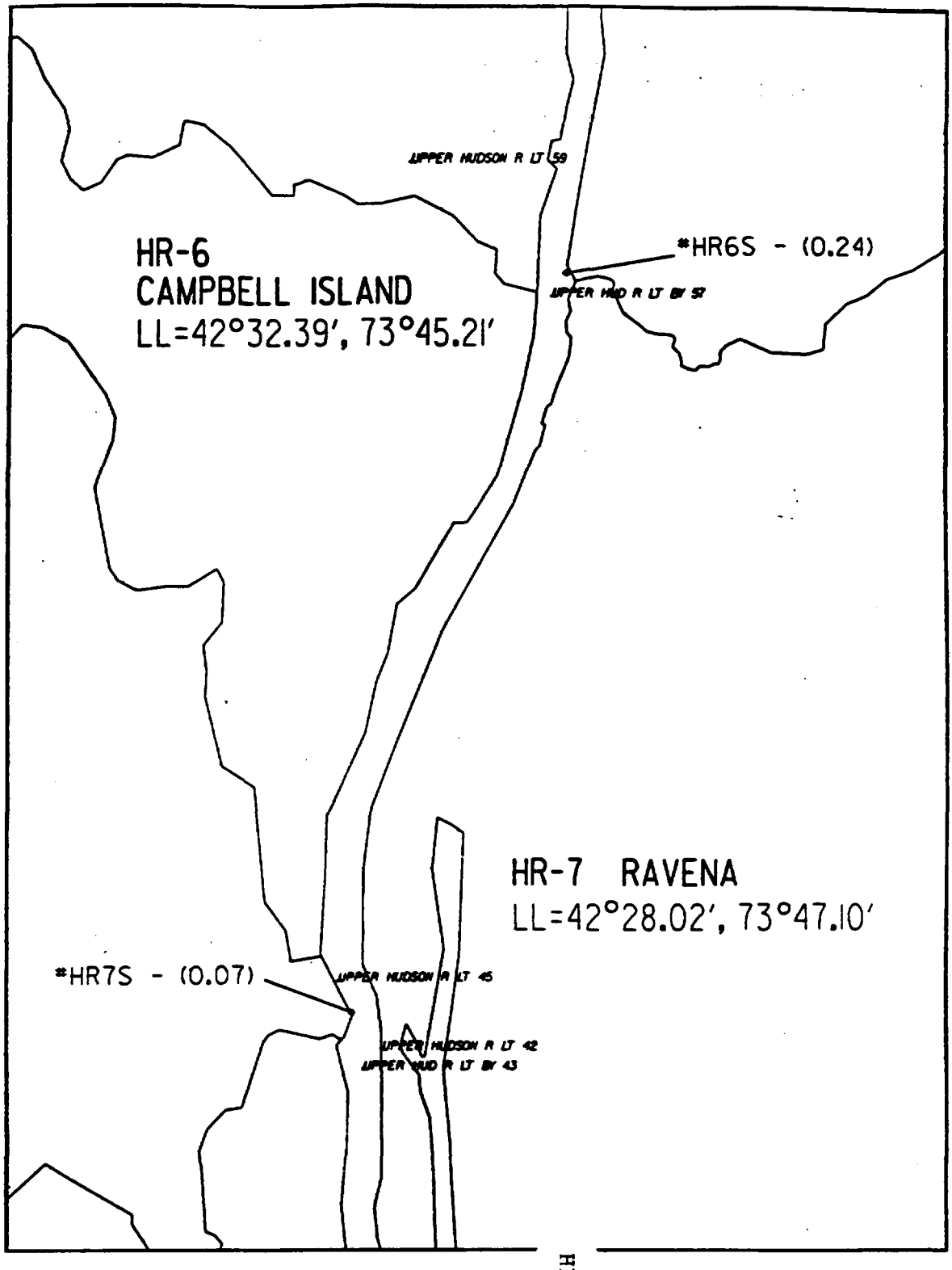
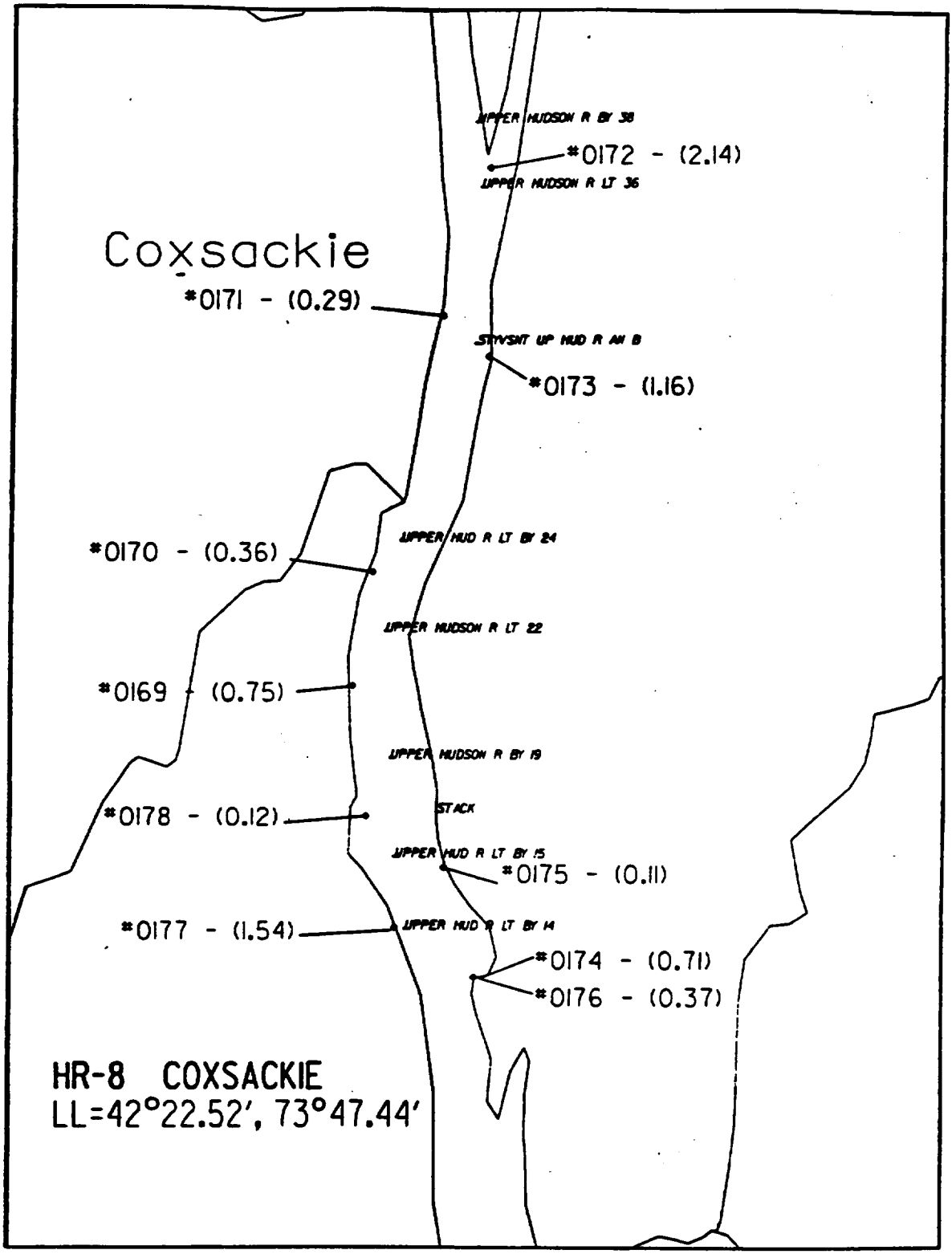
DATE OCTOBER 1991	DWG. NO. HR-5



6.2-2 (con't)

HRP 002 0704

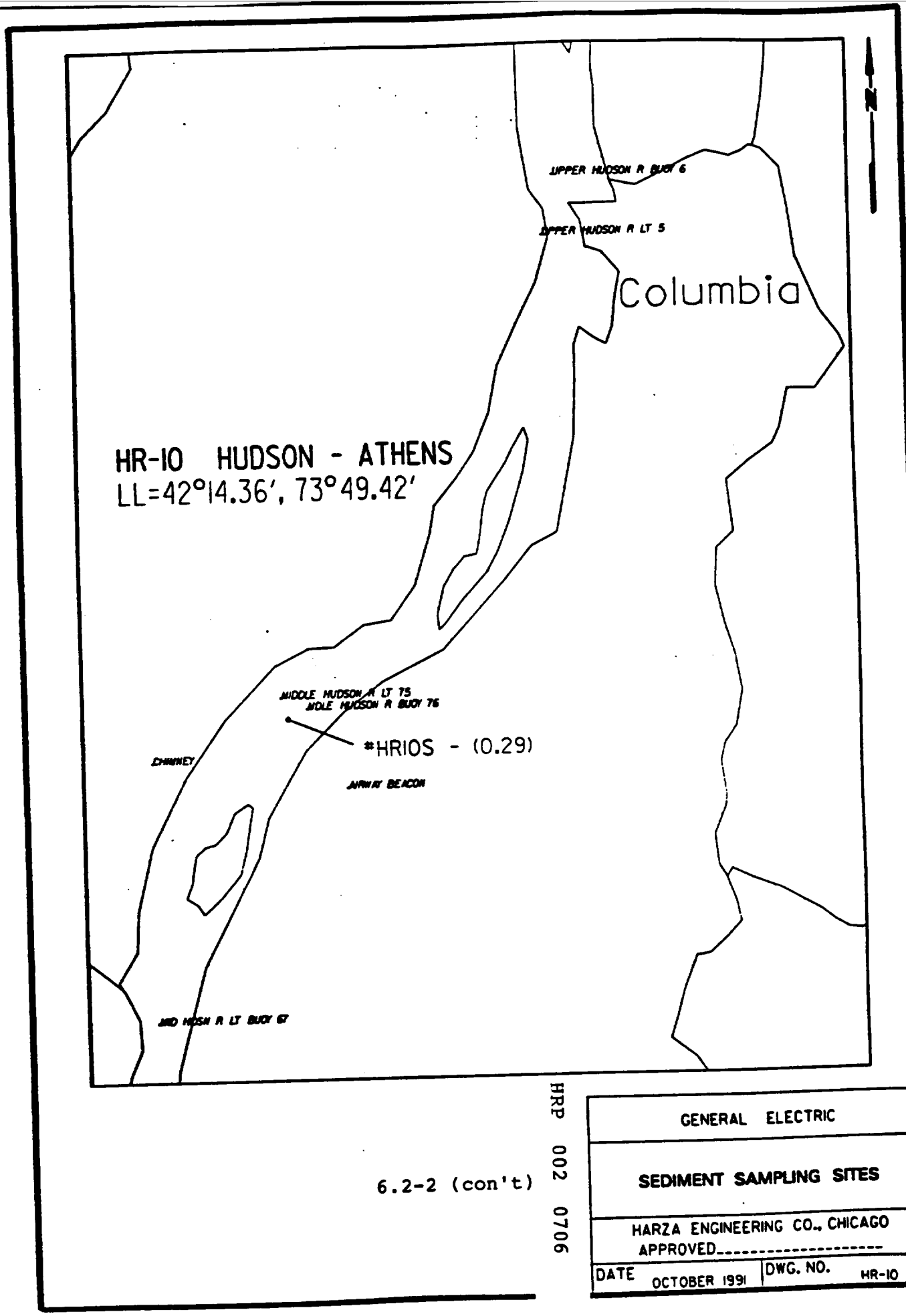
GENERAL ELECTRIC	
SEDIMENT SAMPLING SITES	
HARZA ENGINEERING CO., CHICAGO APPROVED_____	
DATE OCTOBER 1991	DWG. NO. HR-6 AND 7



6.2-2(con't)

HRP 002 0705

GENERAL ELECTRIC	
SEDIMENT SAMPLING SITES	
HARZA ENGINEERING CO., CHICAGO APPROVED_____	
DATE OCTOBER 1991	DWG. NO. HR-8



6.2-2 (con't)

HRP 002 0706

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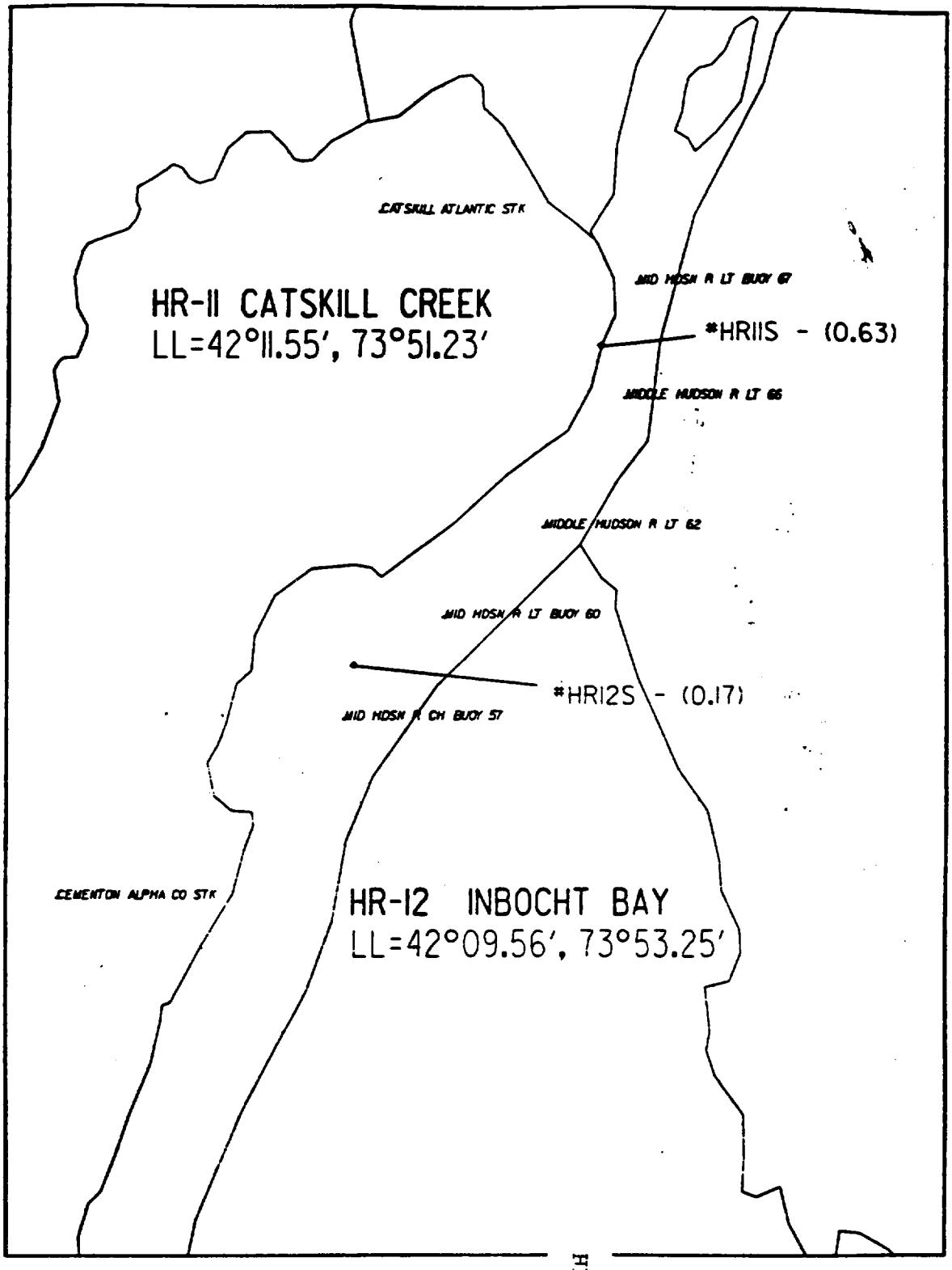
HARZA ENGINEERING CO., CHICAGO APPROVED_____	
DATE OCTOBER 1991	DWG. NO. HR-10



6.2-2 (con't

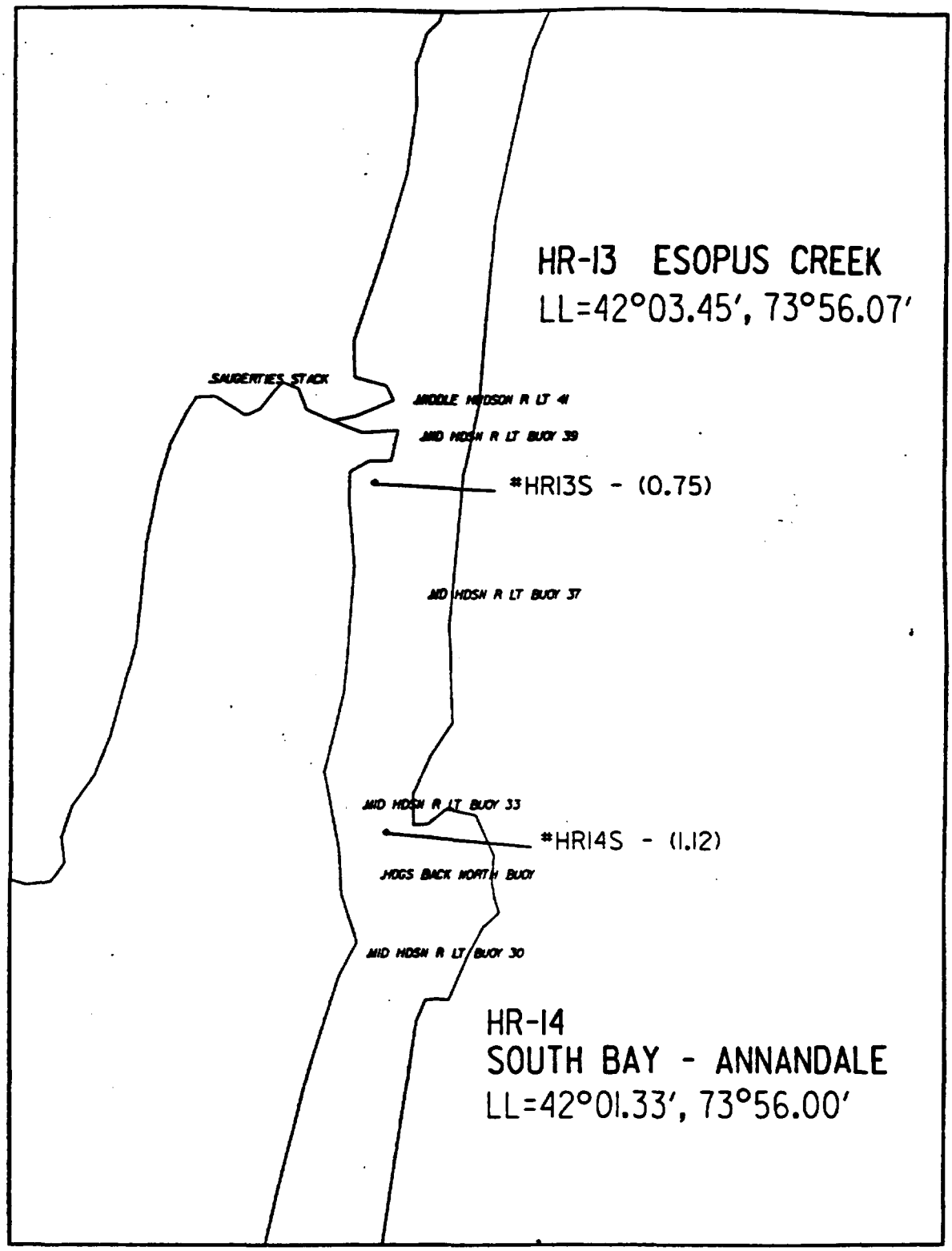
HRP 002 0707

GENERAL ELECTRIC

SEDIMENT SAMPLING SITES

HARZA ENGINEERING CO., CHICAGO
APPROVED_____

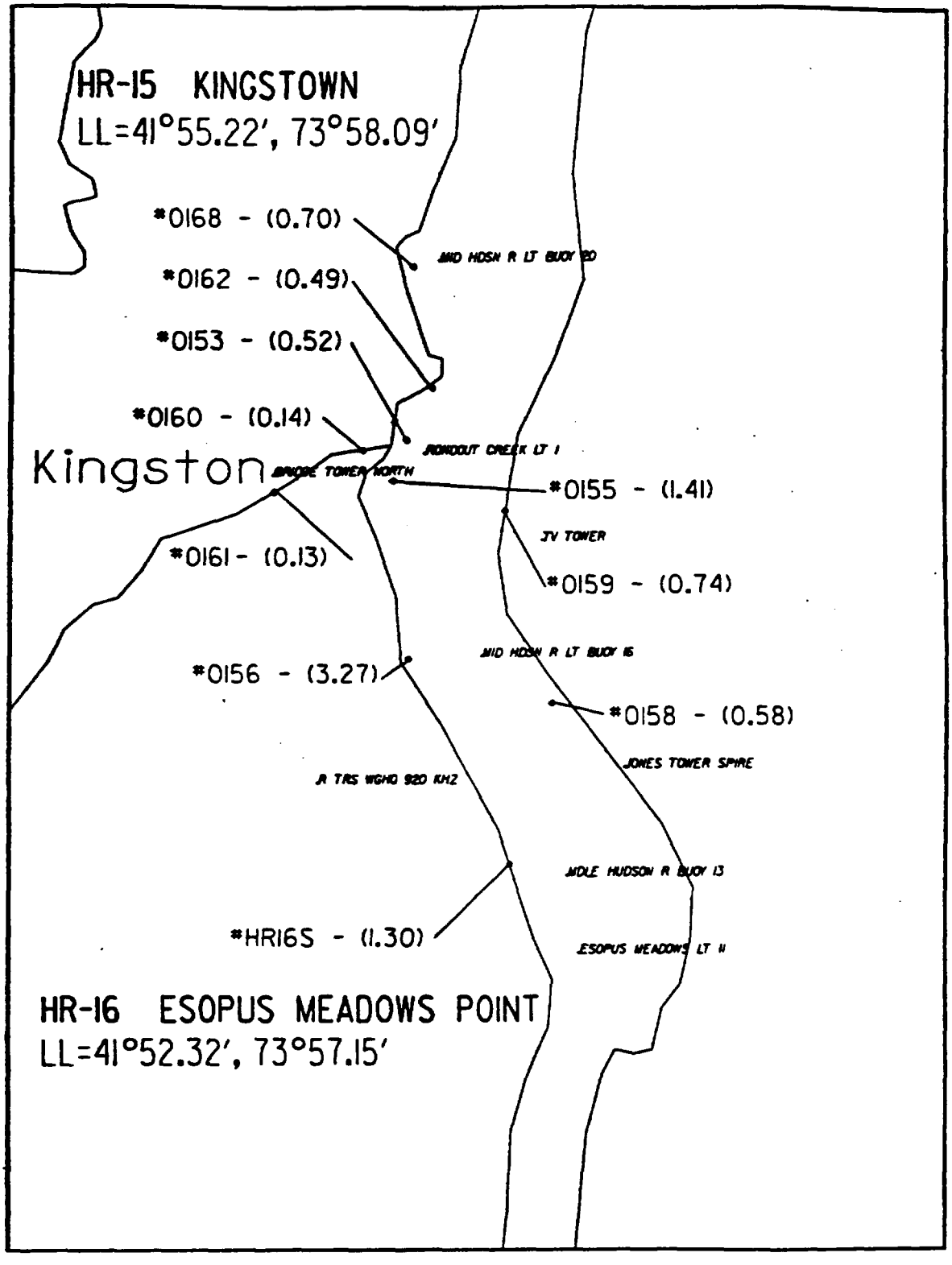
DATE OCTOBER 1991 | DWG. NO. HR-11 AND 12



6.2-2 (con't)

HRP 002 0708

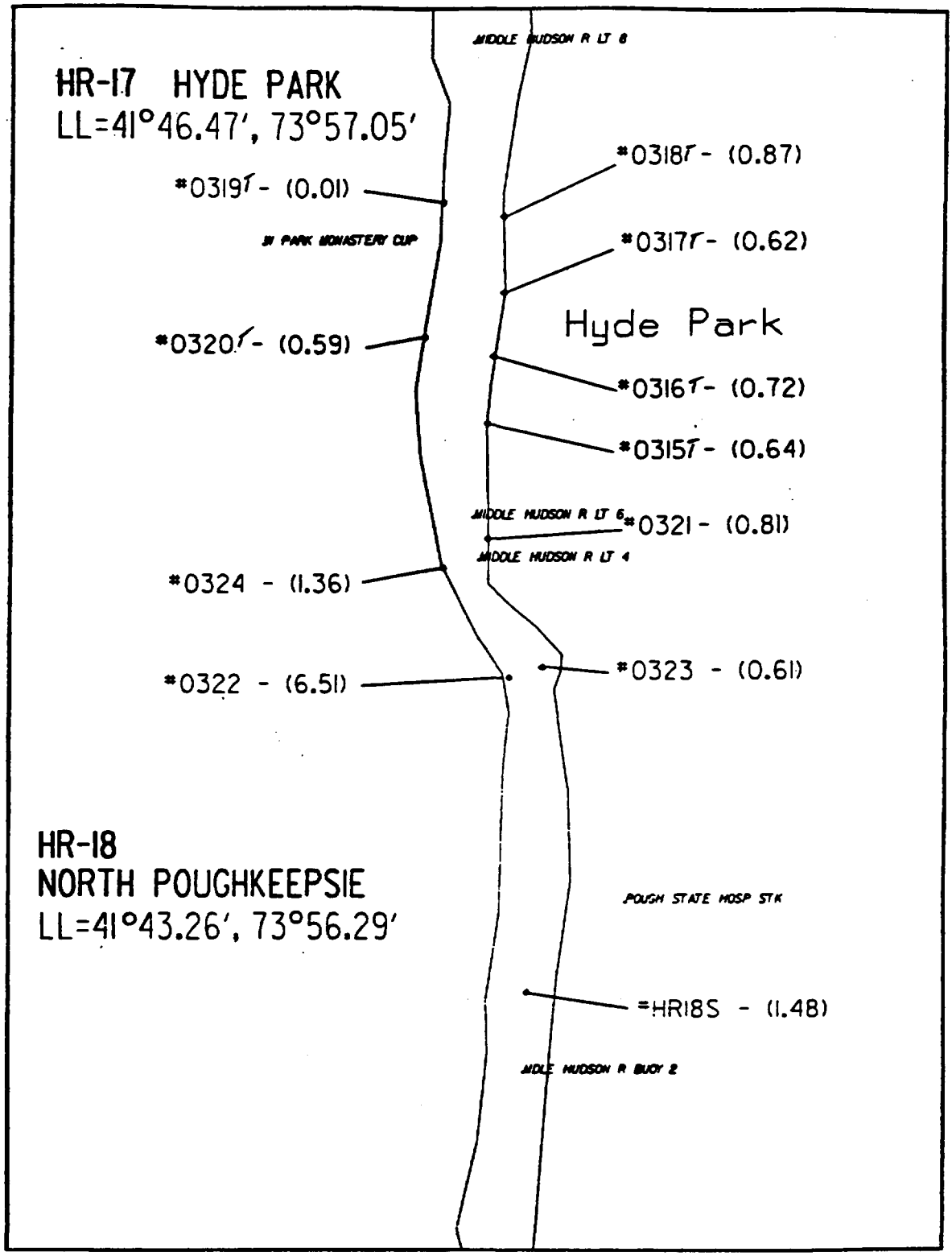
GENERAL ELECTRIC	
SEDIMENT SAMPLING SITES	
HARZA ENGINEERING CO., CHICAGO APPROVED_____	
DATE OCTOBER 1991	DWG. NO. HR-13 AND 14



6.2-2 (con't)

HRP 002 0709

GENERAL ELECTRIC	
SEDIMENT SAMPLING SITES	
HARZA ENGINEERING CO., CHICAGO APPROVED_____	
DATE OCTOBER 1991	DWG. NO. HR-15 AND 16



6.2-2 (con't)

HRP 002 0710

GENERAL ELECTRIC	
SEDIMENT SAMPLING SITES	
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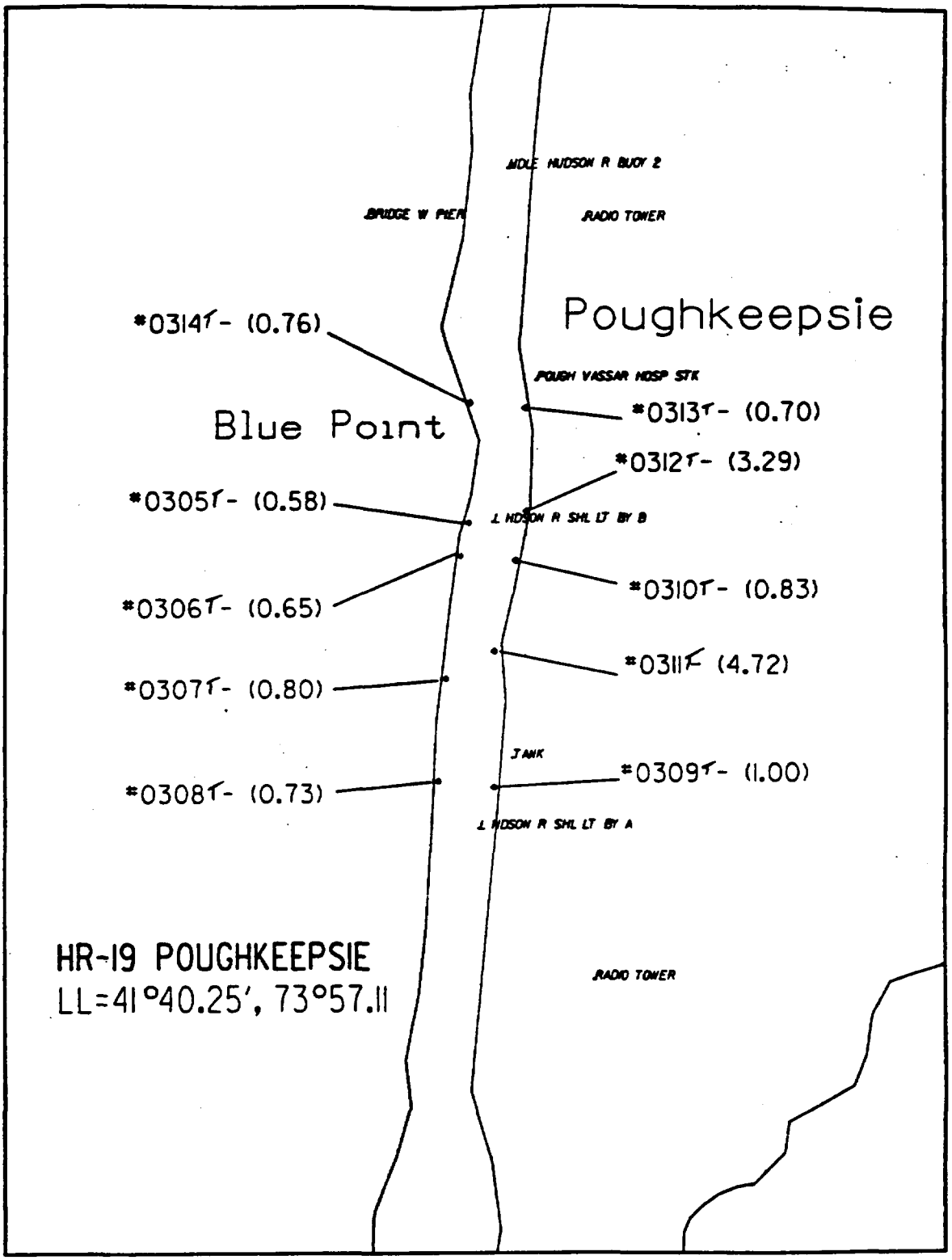
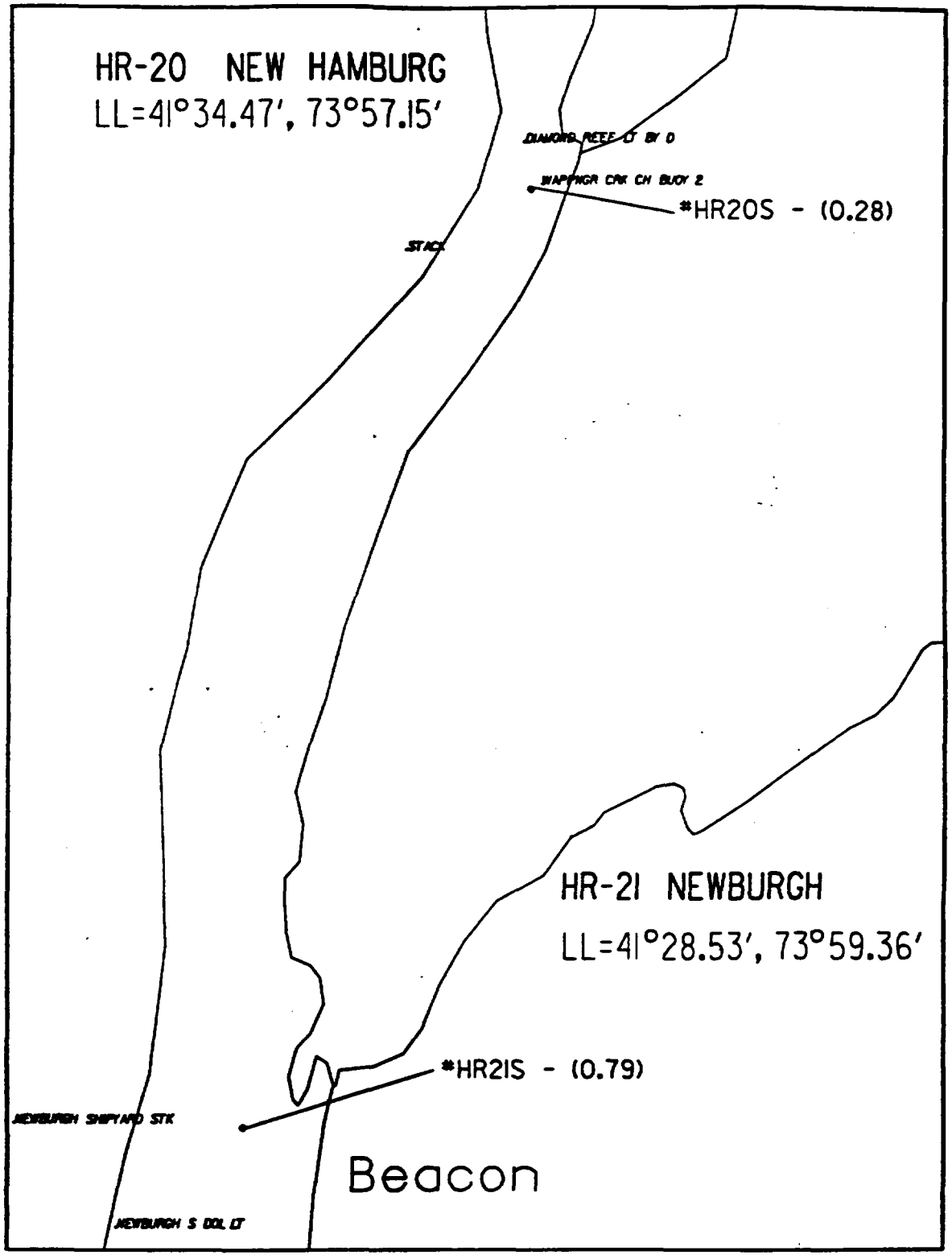
DATE OCTOBER 1991	DWG. NO. HR-17 AND 18



6.2-2 (con't)

HRP 002 0711

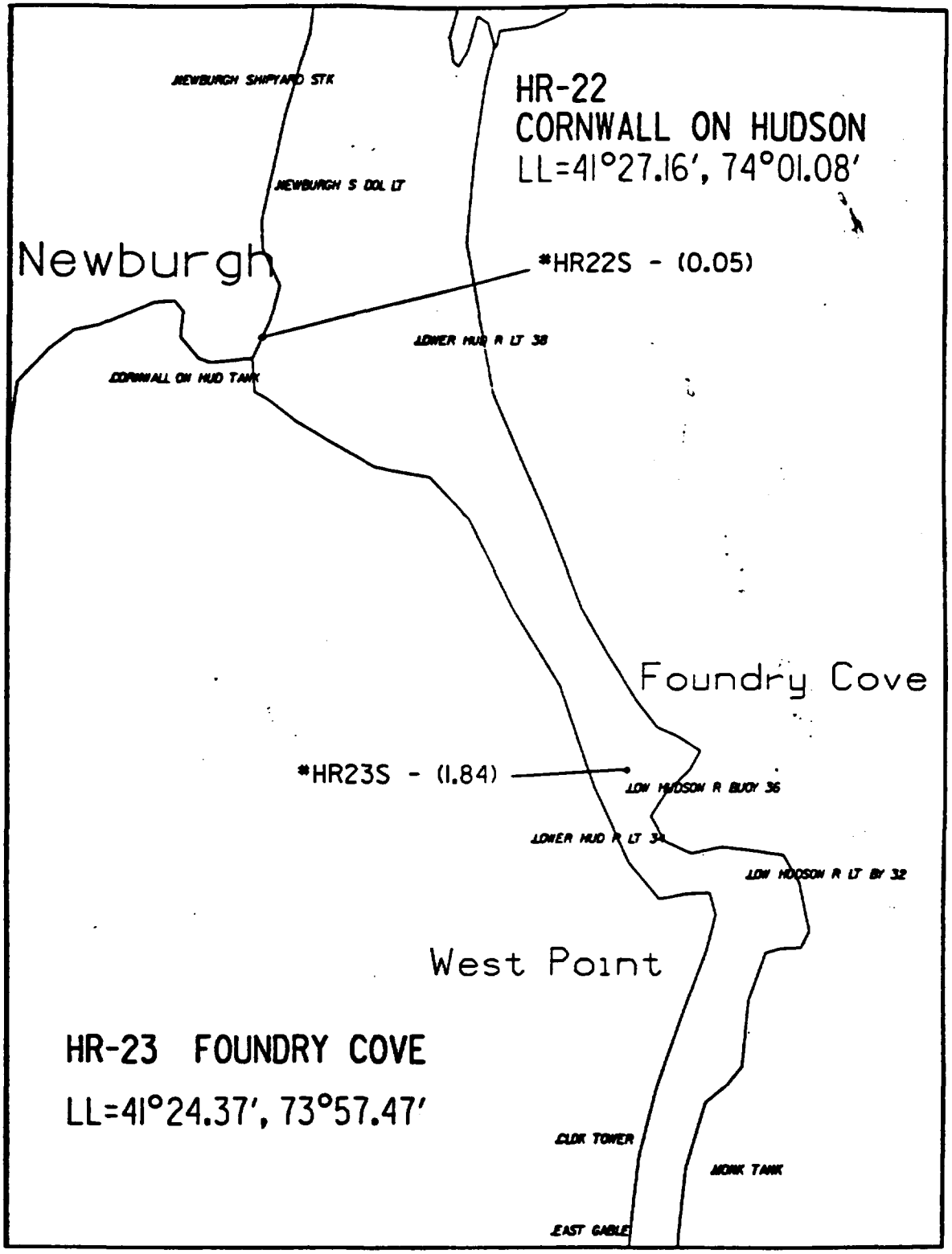
GENERAL ELECTRIC	
SEDIMENT SAMPLING SITES	
HARZA ENGINEERING CO., CHICAGO APPROVED_____	
DATE OCTOBER 1991	DWG. NO. HR-19



6.2-2 (con't)

HRP 002 0712

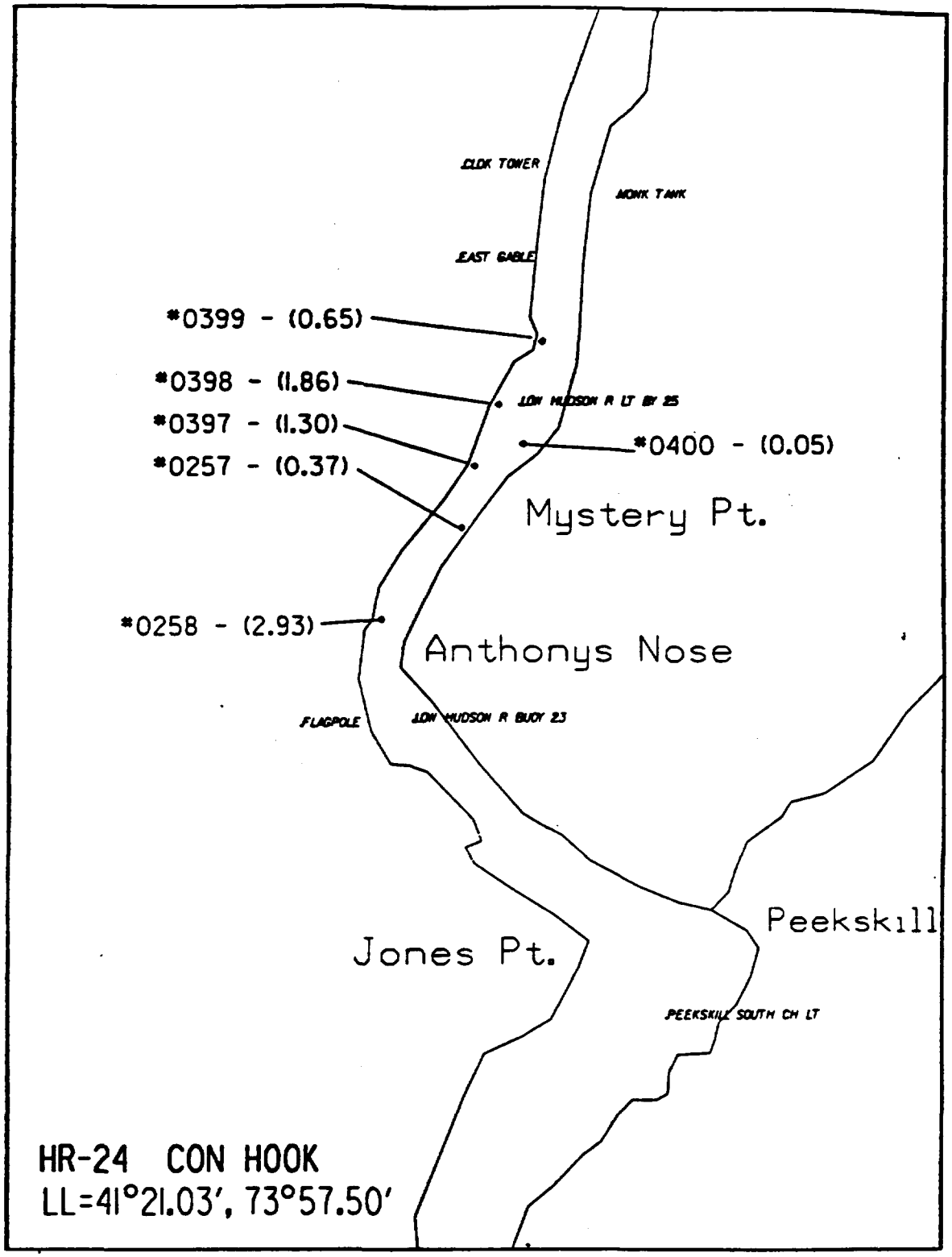
GENERAL ELECTRIC	
SEDIMENT SAMPLING SITES	
HARZA ENGINEERING CO., CHICAGO APPROVED_____	
DATE OCTOBER 1991	DWG. NO HR-20 AND 2



6.2-2 (con't)

HRP 002 0713

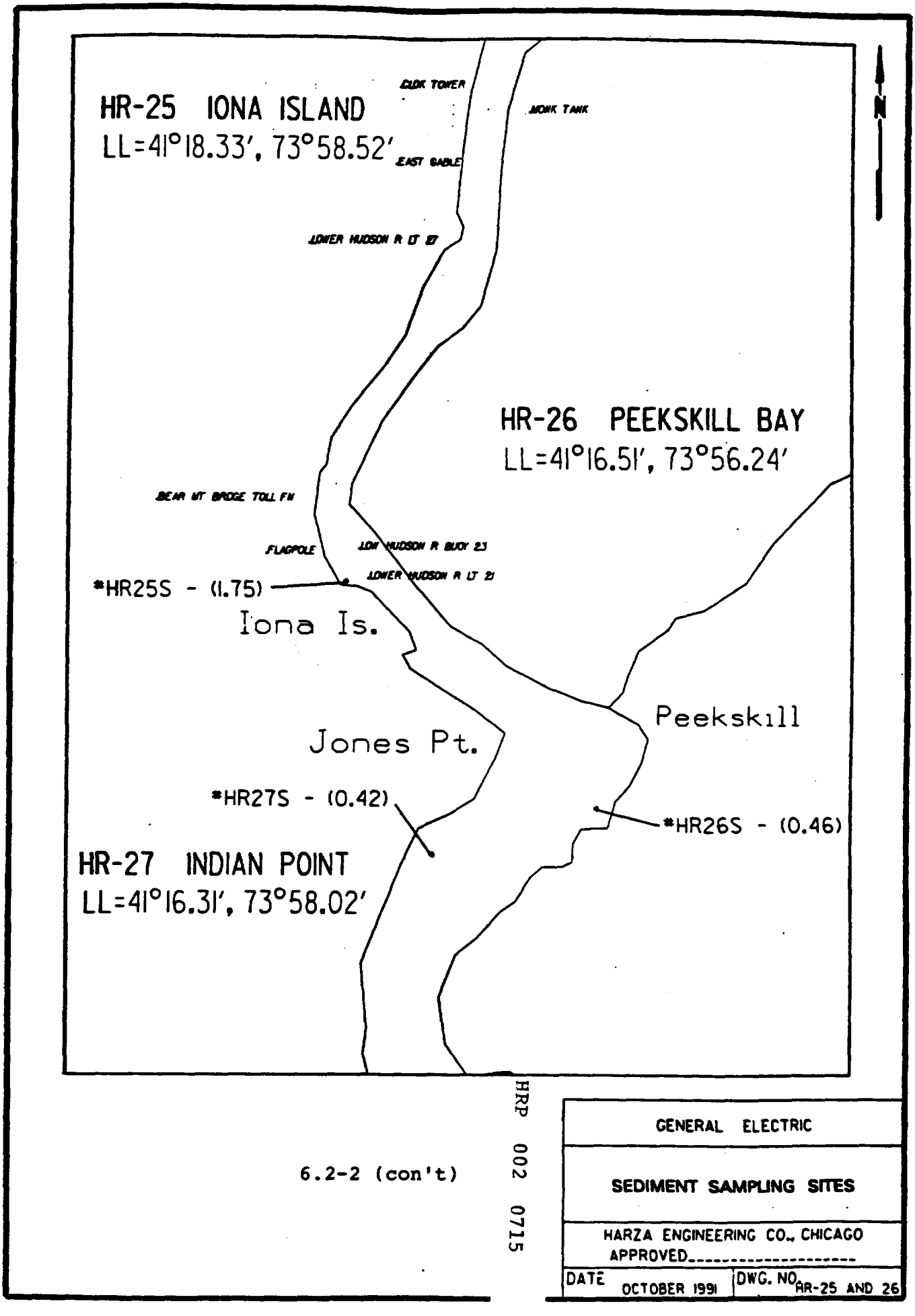
GENERAL ELECTRIC	
SEDIMENT SAMPLING SITES	
HARZA ENGINEERING CO., CHICAGO APPROVED_____	
DATE OCTOBER 1991	DWG. NO. HR-22 AND 23



6.2-2 (con't)

HRP 002 0714

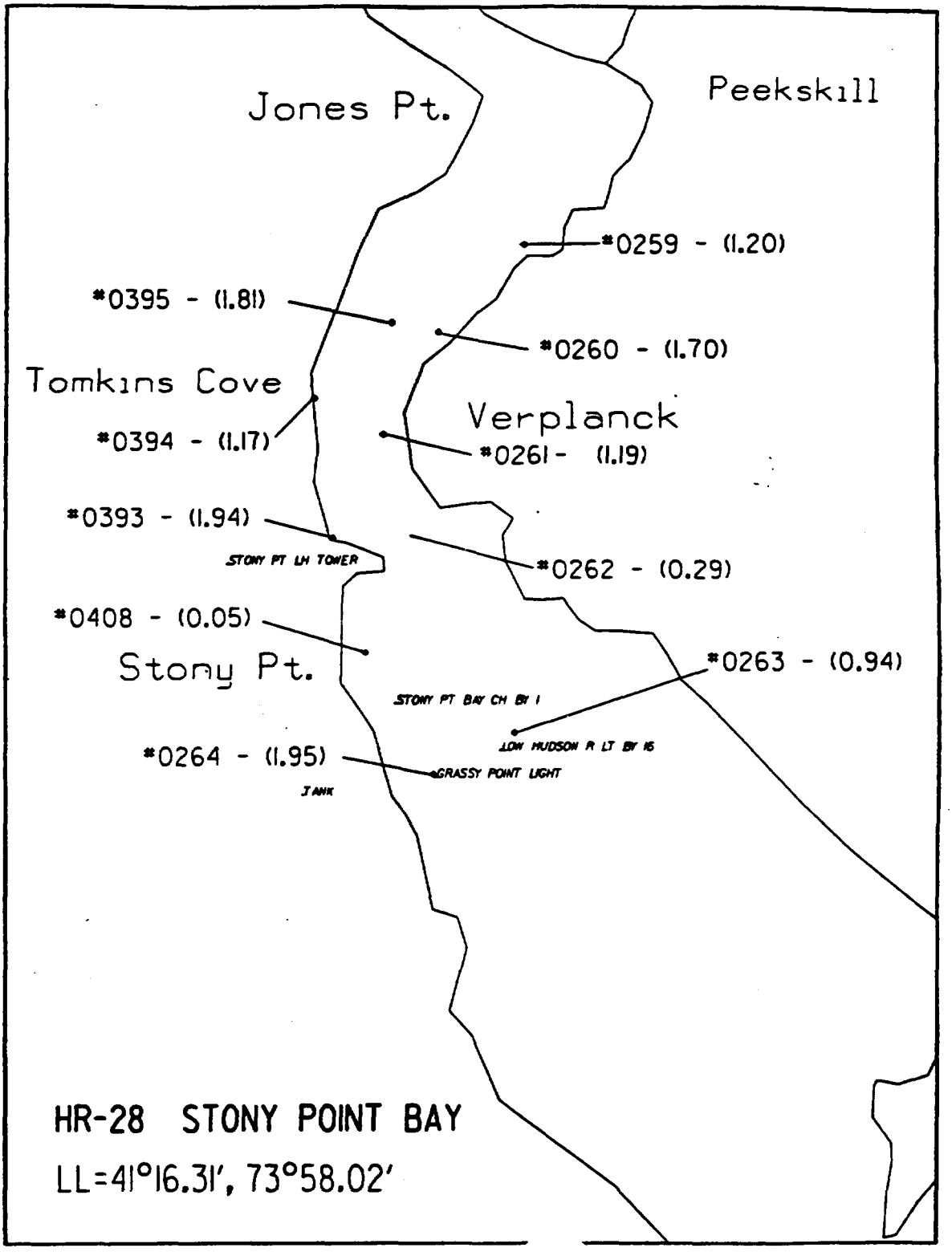
GENERAL ELECTRIC	
SEDIMENT SAMPLING SITES	
HARZA ENGINEERING CO., CHICAGO APPROVED_____	
DATE OCTOBER 1991	DWG. NO. HR-24



6.2-2 (con't)

HRP 002 0715

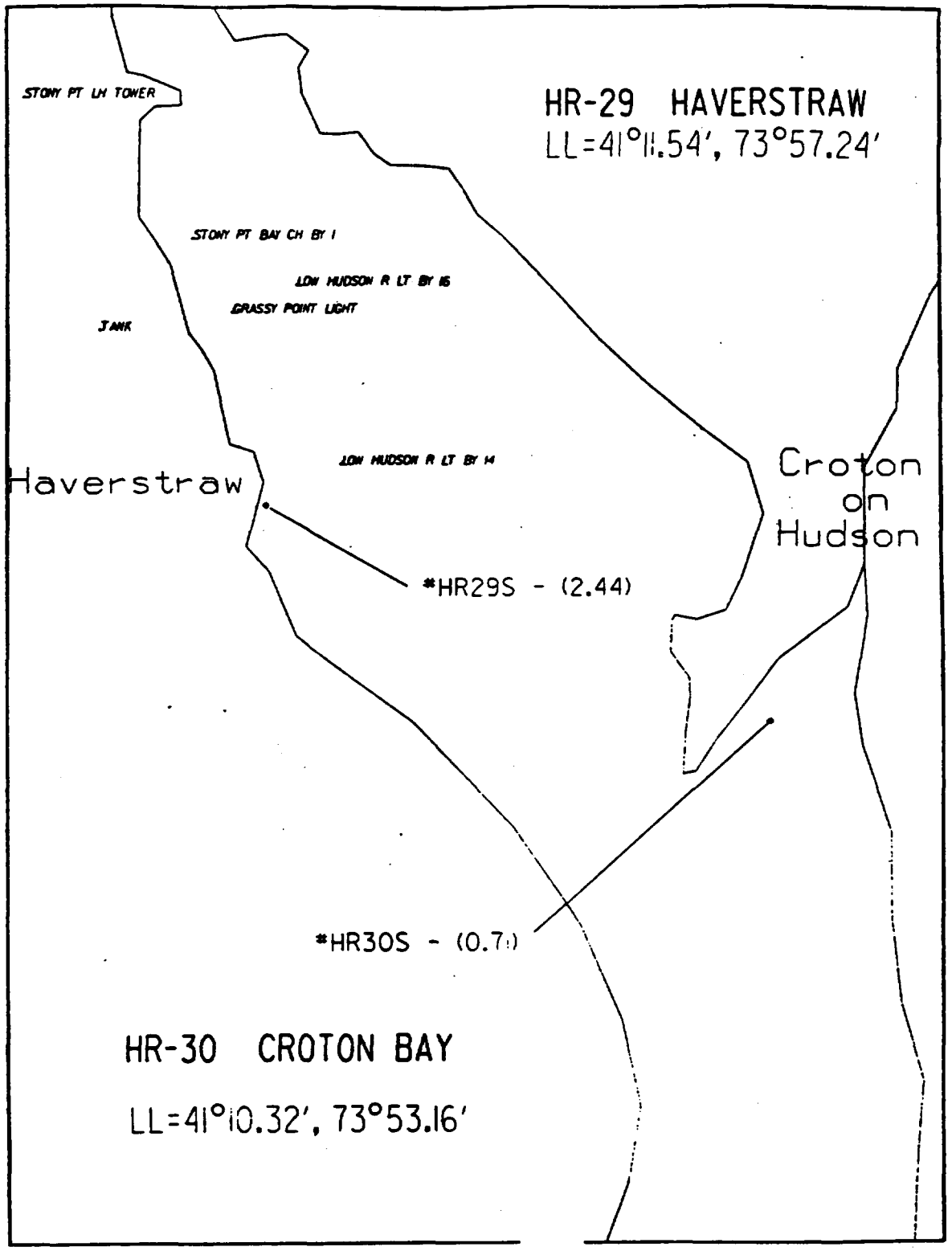
GENERAL ELECTRIC	
SEDIMENT SAMPLING SITES	
HARZA ENGINEERING CO., CHICAGO APPROVED_____	
DATE OCTOBER 1991	DWG. NO HR-25 AND 26



6.2-2 (con't)

HRP 002 0716

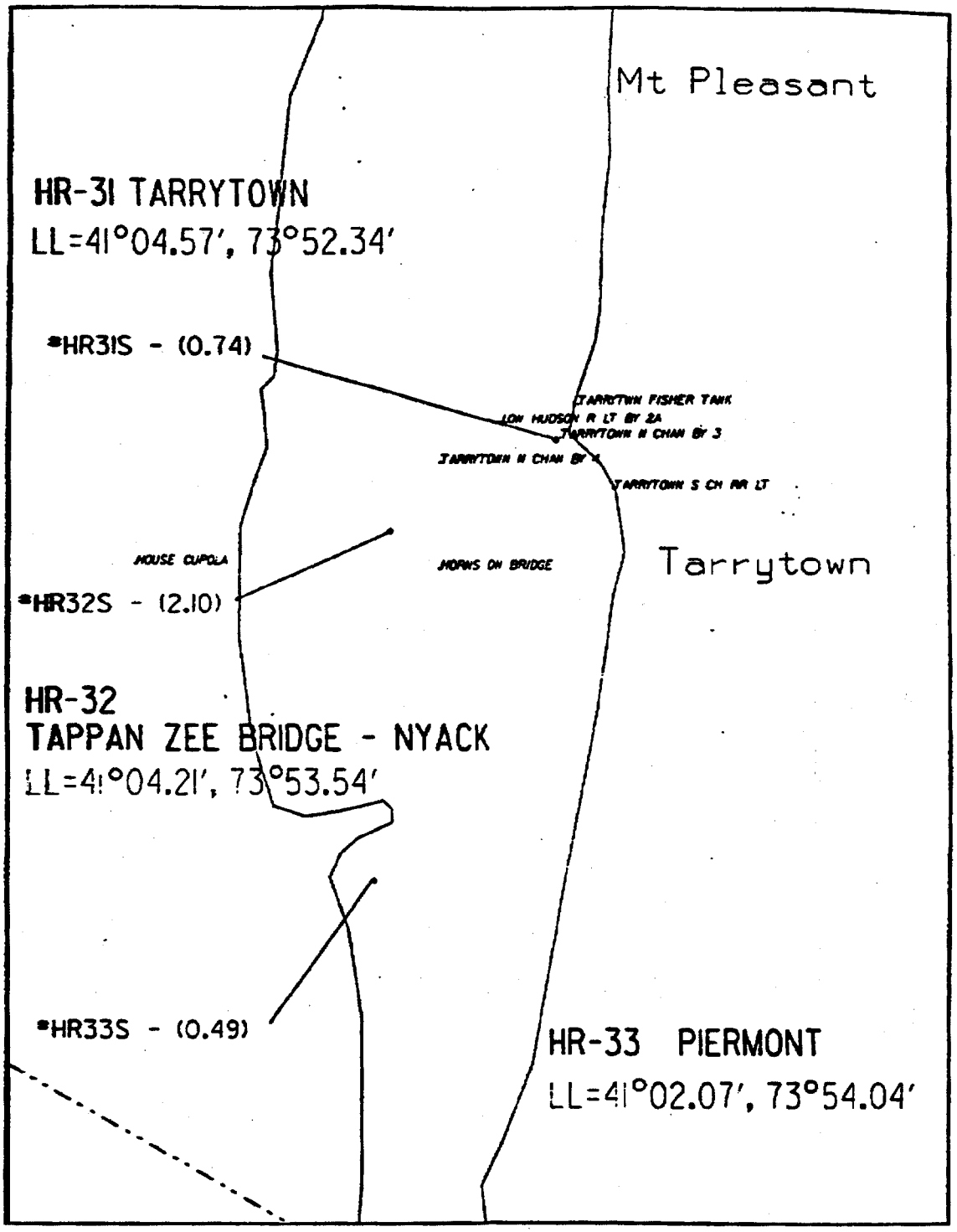
GENERAL ELECTRIC	
SEDIMENT SAMPLING SITES	
HARZA ENGINEERING CO., CHICAGO APPROVED_____	
DATE OCTOBER 1991	DWG. NO. HR-28



6.2-2 (con't)

HRP 002 0717

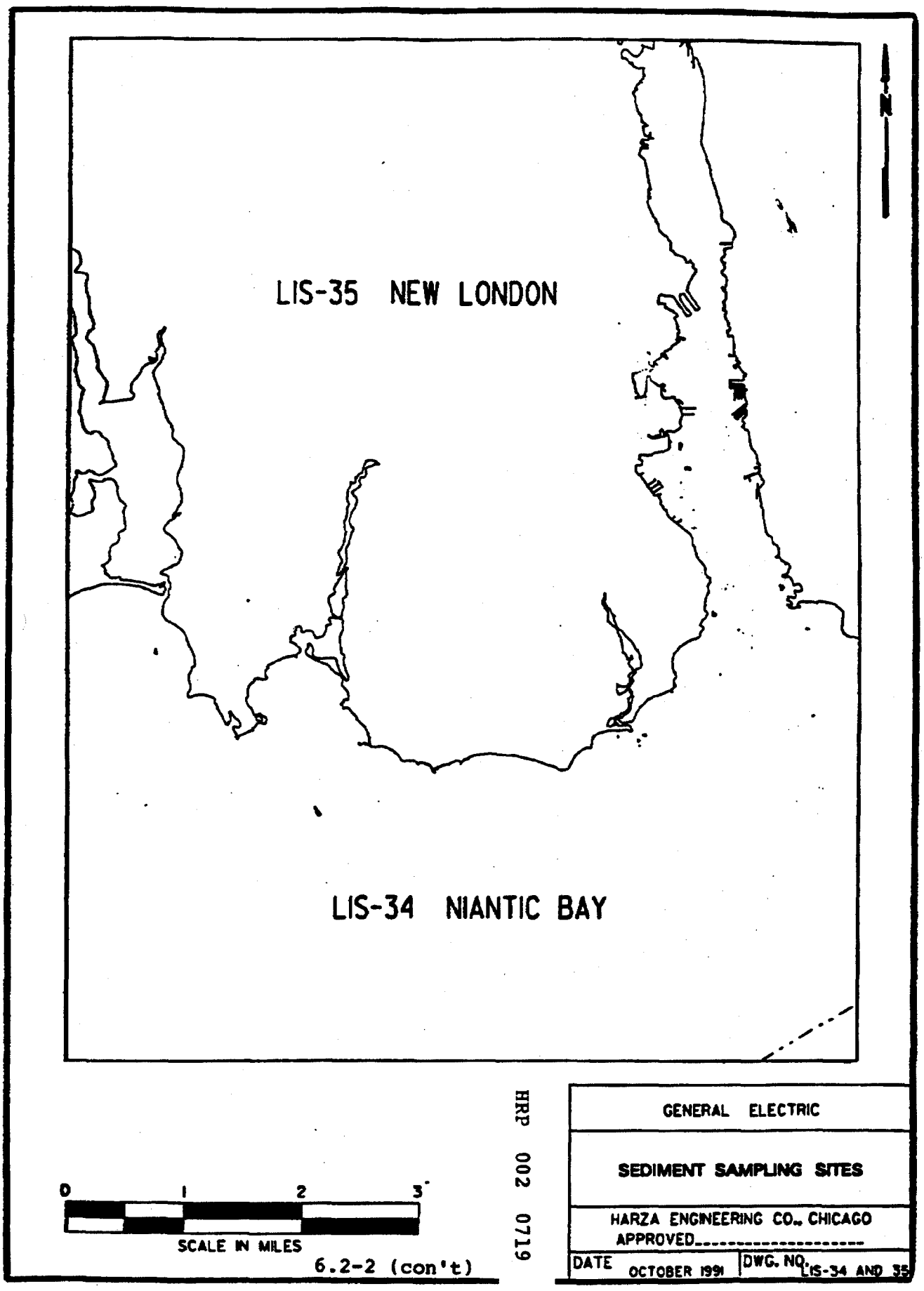
GENERAL ELECTRIC	
SEDIMENT SAMPLING SITES	
HARZA ENGINEERING CO., CHICAGO APPROVED_____	
DATE OCTOBER 1991	DWG. NO HR-29 AND 30



6.2-2 (con't)

HRP 002 0718

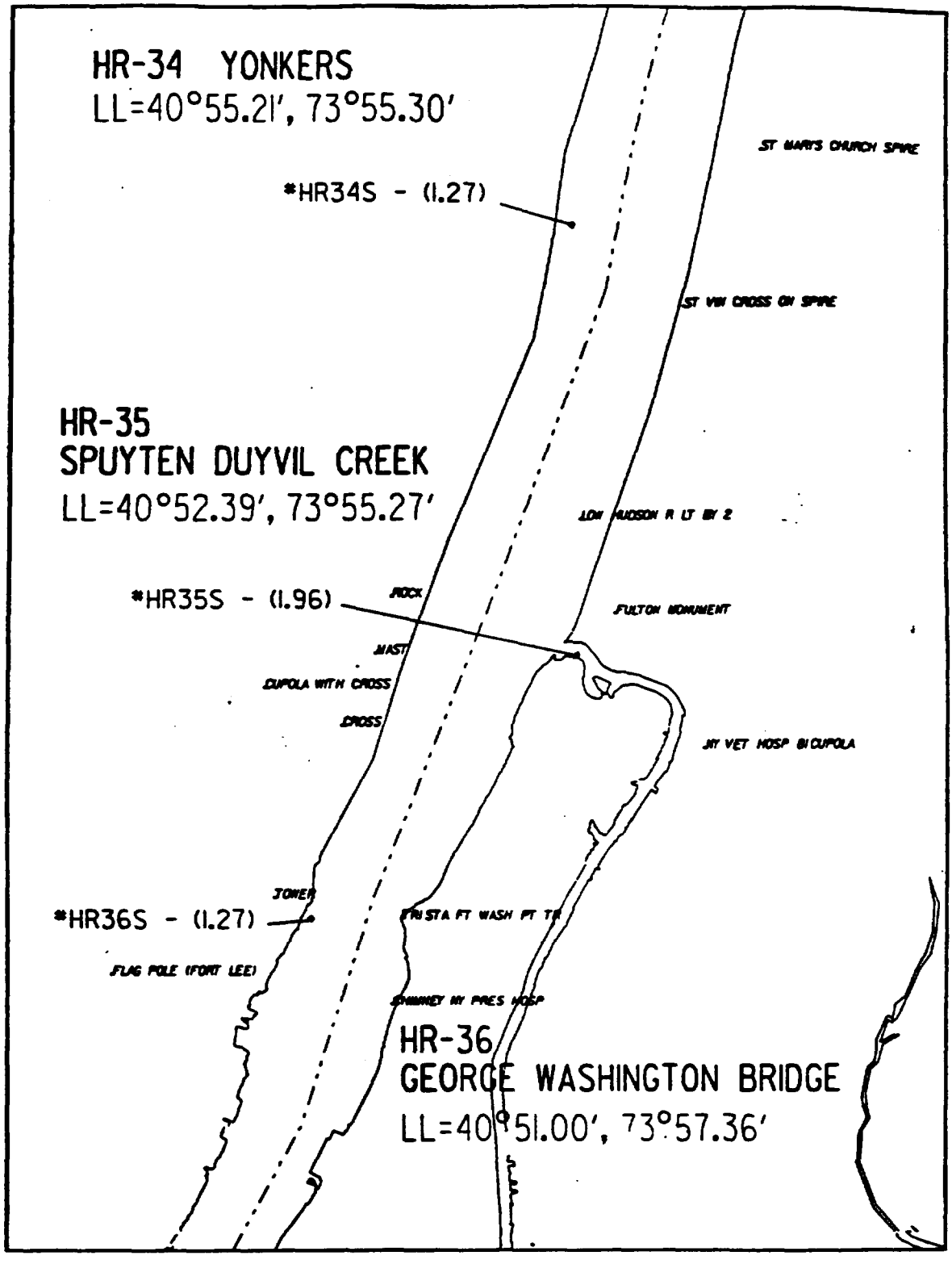
GENERAL ELECTRIC	
SEDIMENT SAMPLING SITES	
HARZA ENGINEERING CO., CHICAGO APPROVED_____	
DATE OCTOBER 1991	DWG. NO. HR-31, 32 & 33



6.2-2 (con't)

HRP 002 0719

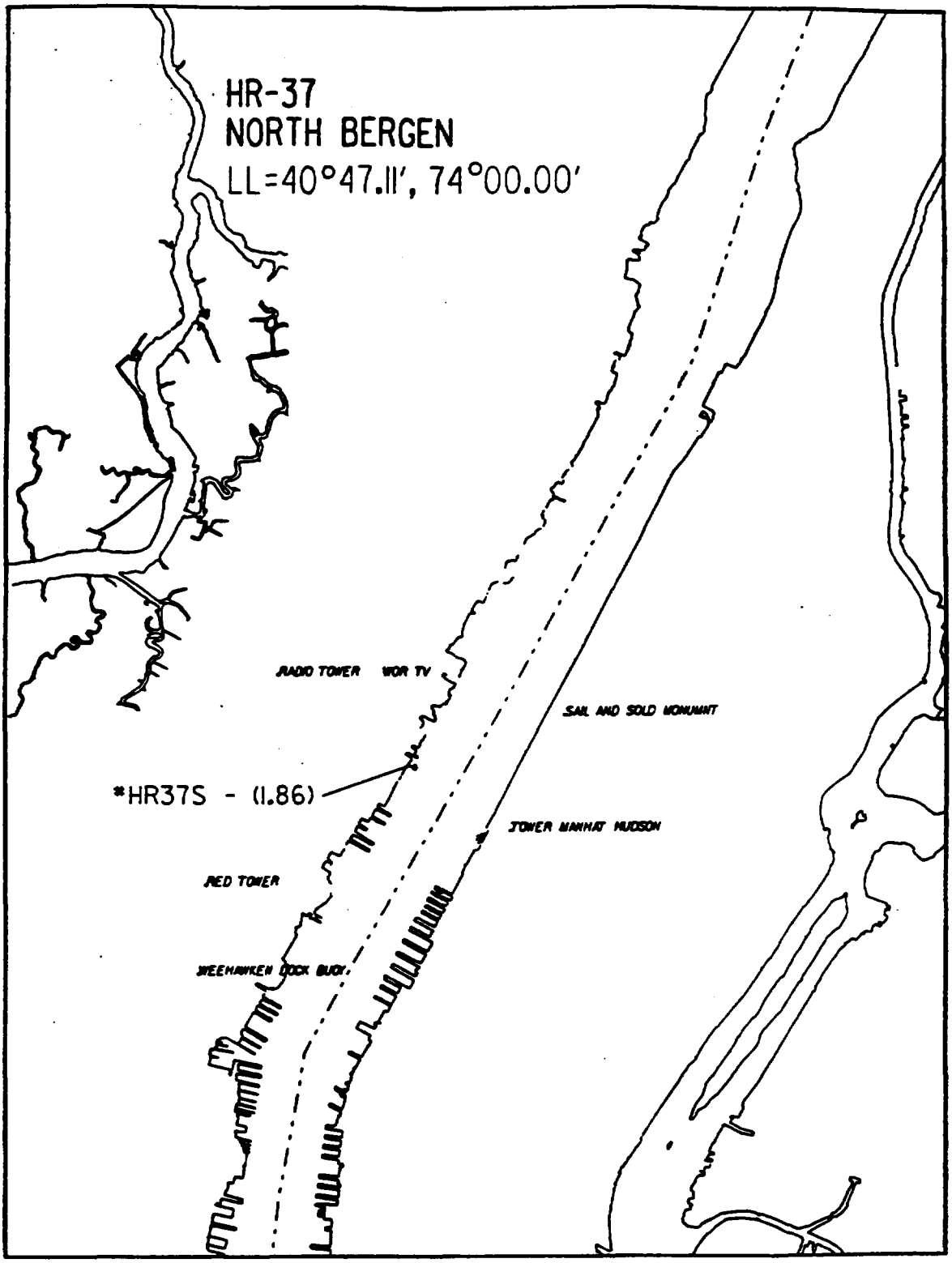
GENERAL ELECTRIC	
SEDIMENT SAMPLING SITES	
HARZA ENGINEERING CO., CHICAGO APPROVED_____	
DATE OCTOBER 1991	DWG. NO. LIS-34 AND 35



HRP 002 0720

GENERAL ELECTRIC	
SEDIMENT SAMPLING SITES	
HARZA ENGINEERING CO., CHICAGO APPROVED_____	
DATE OCTOBER 1991	DWG. NO. HR-34, 35 & 36

6.2-2 (con't)



6.2-2 (con't)

HRP 002 0721

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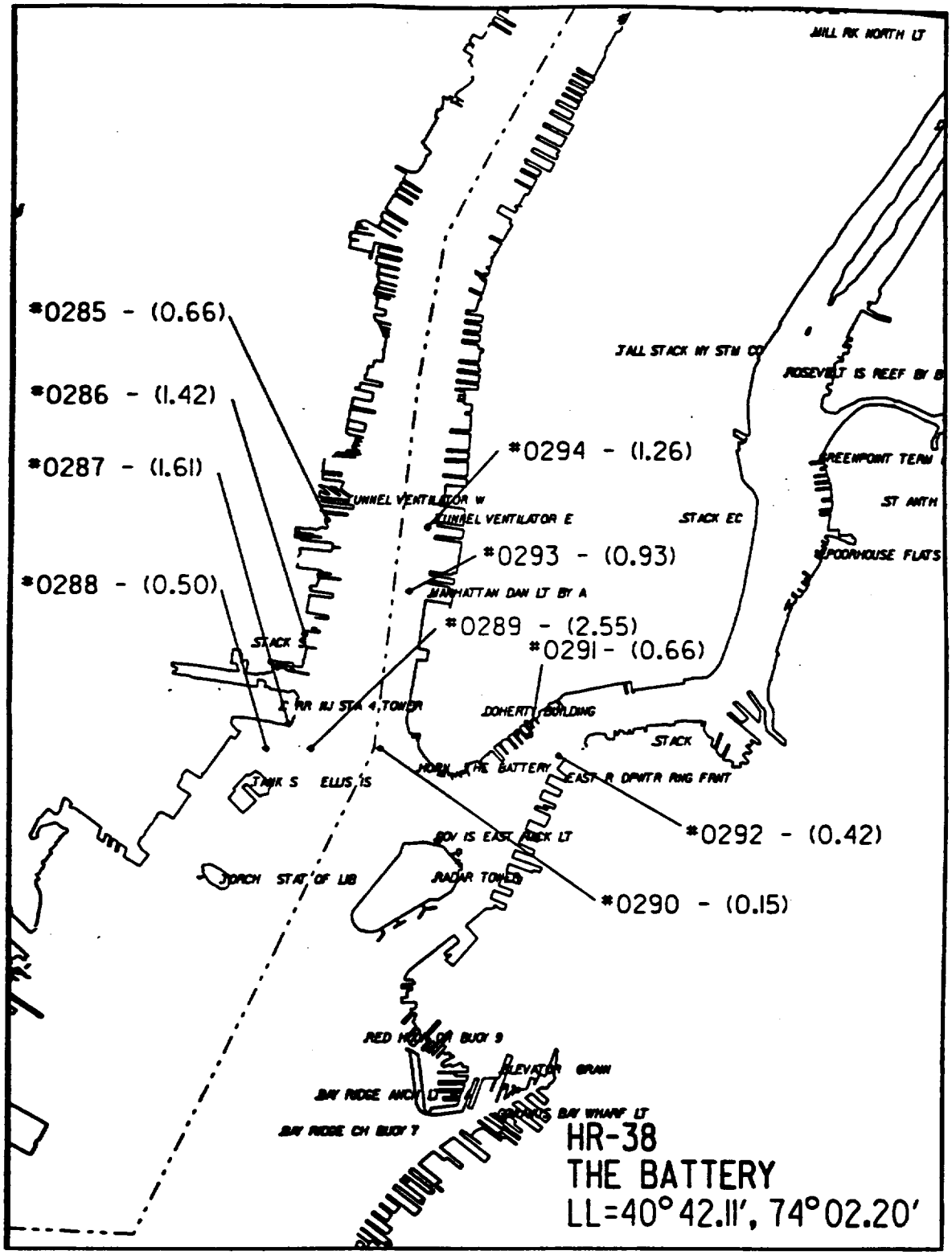
SEDIMENT SAMPLING SITES	
HARZA ENGINEERING CO., CHICAGO APPROVED_____	
DATE OCTOBER 1991	DWG. NO. HR-37



6.2-2 (con't)

0 1 2 3

SCALE IN MILES

HRP 002 0722

GENERAL ELECTRIC	
SEDIMENT SAMPLING SITES	
HARZA ENGINEERING CO., CHICAGO APPROVED_____	
DATE OCTOBER 1991	DWG. NO. HR-38

Footnote to Tables 6.2-1 and 6.2-2

All PCB concentrations are expressed as µg/g dry weight. PCB analyses performed by Northeast Analytical, Inc. (NEA), Schenectady, NY. Samples were analyzed using capillary column gas chromatography, which was capable of resolving 118 individual PCB peaks. Total PCB, Aroclor and homolog composition of the samples was also reported by NEA. Sediment samples were collected by either a Petite Ponar or Shipek dredge, which did not collect samples deeper than the top six inches of sediment. Almost all sediment samples collected during the course of this study were collected from nearshore sediment depositional areas, where fine particulate and/or organic rich sediments are found. Areas where sediments consisted of coarse particulates such as sand were avoided to the extent possible. Areas known to have been dredged, such as navigation channels, were also avoided.

Sample results for samples collected during field work in 1988 describe a five dredge haul composite sample analysis. The individual samples which made up the composite were collected within several hundred yards of each other, with samples being collected from both shorelines of the Hudson River, or scattered along the coastline for stations in New York Harbor and Long Island Sound. Samples were collected at 88 stations during 1988. Of these, 38 were located in the tidal Hudson River between the Troy Dam and The Battery, 18 were collected in New York Harbor (defined in this study as the East River between The Battery and Throgs Neck Bridge, Upper and Lower New York Bay, Raritan Bay, Newark Bay, the Arthur Kill and the Kill van Kull), and 32 stations were located in western Long Island Sound, west of a line between New Haven, CT and Shoreham, NY. One sample, from Hudson River station HR-9 (Stockport Creek mouth) was lost during laboratory analysis, and results are not reported.

During 1989, more detailed sediment sampling was performed at 28 stations which contained the highest PCB concentrations in either sediment or biota from samples collected in 1988. Ten of the stations were in the tidal Hudson River, while nine each were located in New York Harbor and Long Island Sound. Between 6-12 sediment samples were collected from each station, and individually analyzed for their Aroclor, homolog and congener content. Samples were collected along both shorelines of the Hudson River, and paralleled the coastal shoreline or the shoreline of embayments in New York Harbor and Long Island Sound. Concentrations of 11 pesticides were also determined on samples collected during 1989.

During 1990, individual sediment samples were collected and analyzed form five new stations (NYH-19, 21, 22, 23, 24) in New York Harbor, whose definition was expended to include Jamaica Bay, the Sandy Hook, NJ area and the Harlem River.

All of the above studies were funded by the General Electric Company, Fairfield, CT.

Table II. Sediment Activities of Fallout and Reactor Radionuclides and Concentrations of PCBs from Coves in the Low-Salinity Reach of the Hudson Estuary (1977)

depth, cm	^{137}Cs, pCi/kg	^{134}Cs, pCi/kg	^{60}Co, pCi/kg	^{239,240}Pu, pCi/kg	Aroclor, ppm 1242	Aroclor, ppm 1254
FOUNDRY COVE		Core Site C (kmp 86.6, 1/77, CN 1240)				
0-1	1530 ± 38	28 ± 13	73 ± 10	27.9 ± 1.4	7.48 ± 0.52	0.97 ± 0.22
1-2	1610 ± 44	47 ± 16	55 ± 13	28.6 ± 1.7	9.28 ± 0.65	1.45 ± 0.35
2-3	2000 ± 48	(75 ± 16)[b]	98 ± 13	30.9 ± 1.4	11.6 ± 0.5	1.42 ± 0.42
1971 → 3-4	(2400 ± 56)[b]	(75 ± 16)[b]	(145 ± 13)[b]	34.7 ± 1.9	19.3 ± 0.9	2.35 ± 0.54
4-5	2240 ± 55	30 ± 16	110 ± 14	35.4 ± 1.9	(26.0 ± 1.4)[a]	3.06 ± 0.82 ← PCB PEAK
5-6	2060 ± 56	−7 ± 13	89 ± 15	41.4 ± 2.7	20.8 ± 1.7	2.32 ± 0.60
6-7	2010 ± 48	5 ± 10	67 ± 12		21.6 ± 1.6	2.38 ± 0.61
7-8	2180 ± 54	12 ± 11	83 ± 12	47.5 ± 2.2	14.7 ± 1.3	1.77 ± 0.33
8-9	2130 ± 54	−7 ± 11	95 ± 13	53.4 ± 1.9	12.6 ± 0.9	1.64 ± 0.29
9-10	1970 ± 49	−7 ± 10	100 ± 12	49.6 ± 2.3	9.36 ± 0.87	1.49 ± 0.20
10-11	2120 ± 50	1 ± 10	70 ± 11	54.9 ± 2.3	6.31 ± 0.41	1.17 ± 0.11
11-12	2130 ± 59	−14 ± 13	41 ± 15	68.3 ± 2.6	5.17 ± 0.31	1.44 ± 0.13
12-13	2170 ± 57	−13 ± 12	67 ± 14	73.8 ± 2.0	5.57 ± 0.43	1.26 ± 0.09
13-14	2140 ± 68	−10 ± 16	47 ± 19	(76.6 ± 5.3)[c]	5.30 ± 0.33	1.39 ± 0.13
14-15	1990 ± 48	−18 ± 10	36 ± 11	55.6 ± 1.9	5.70 ± 0.60	1.60 ± 0.03
15-16	1680 ± 55	4 ± 16	43 ± 18	53.1 ± 2.1	2.60 ± 0.20	0.78 ± 0.06
16-17	1440 ± 38	2 ± 9	17 ± 10	48.6 ± 1.9	3.90 ± 0.30	1.21 ± 0.10
17-18	1160 ± 42	−10 ± 14	16 ± 15	39.8 ± 2.1	3.30 ± 0.23	1.10 ± 0.16
18-19	770 ± 29	−8 ± 11	−4 ± 14	25.9 ± 0.8	2.17 ± 0.18	0.92 ± 0.09
19-20	580 ± 29	−11 ± 9	−8 ± 10	20.4 ± 0.9	1.58 ± 0.15	0.90 ± 0.08
20-21	380 ± 21	−5 ± 11	24 ± 13	12.5 ± 0.6	1.20 ± 0.09	0.73 ± 0.08
21-22	200 ± 18	1 ± 11	13 ± 13	6.3 ± 0.4	0.72 ± 0.04	0.47 ± 0.06
22-23	105 ± 15	8 ± 10	26 ± 11	3.2 ± 0.3	0.51 ± 0.02	0.34 ± 0.06
23-24	22 ± 12	8 ± 9	−14 ± 11	0.8 ± 0.1	0.22 ± 0.03	0.23 ± 0.07
24-25	(19 ± 11)[d]	−6 ± 9	8 ± 9	(0.5 ± 0.1)[d]	0.33 ± 0.03	0.19 ± 0.04
25-26	−9 ± 13	−5 ± 10	−6 ± 10	0.2 ± 0.1	0.25 ± 0.07	0.09 ± 0.02
INDIAN POINT		Core Site D (kmp 69.5, 7/17/77, CN 1264)				
0-2	1090 ± 51	82 ± 25	96 ± 21	5.7 ± 0.5	3.95 ± 0.62	0.57 ± 0.04
2-4	1130 ± 54	54 ± 25	95 ± 21	8.2 ± 1.0	2.29 ± 0.37	0.34 ± 0.03
4-8	1730 ± 66	120 ± 28	195 ± 27	10.2 ± 0.6	7.41 ± 0.51	0.87 ± 0.09
1971 → 8-12	(4140 ± 93)[b]	(395 ± 29)[b]	(420 ± 20)[b]	10.0 ± 1.2	(10.06 ± 0.60)[a]	1.29 ± 0.19 ← PCB PEAK
12-16	3220 ± 70	255 ± 22	280 ± 17	9.8 ± 0.3	8.30 ± 0.53	1.07 ± 0.19
16-20	2200 ± 46	160 ± 12	175 ± 9	10.9 ± 0.9	7.23 ± 0.25	0.90 ± 0.13
20-24	1590 ± 43	78 ± 14	115 ± 14	11.0 ± 0.2	6.72 ± 0.27	0.88 ± 0.15
24-28	1280 ± 38	52 ± 15	190 ± 16	9.2 ± 0.4	7.34 ± 0.37	0.90 ± 0.12
28-32	970 ± 32	32 ± 13	99 ± 14	7.6 ± 0.4	4.90 ± 0.35	0.71 ± 0.06
32-36	860 ± 41	15 ± 16	100 ± 19	8.4 ± 0.4	3.70 ± 0.39	0.52 ± 0.03
36-40	1020 ± 35	7 ± 13	145 ± 16	9.8 ± 0.7	4.28 ± 0.39	0.53 ± 0.04
40-44	845 ± 35	19 ± 9	130 ± 12	6.8 ± 0.4	3.57 ± 0.28	0.40 ± 0.04
44-48	520 ± 35	4 ± 18	47 ± 19	5.3 ± 0.3	2.10 ± 0.34	0.31 ± 0.04
48-52	410 ± 19	8 ± 9	47 ± 11	4.2 ± 0.2	1.61 ± 0.14	0.25 ± 0.01
52-56	255 ± 21	11 ± 13	17 ± 16	1.7 ± 0.2	0.65 ± 0.09	0.11 ± 0.01
56-60	(11 ± 10)[e]	−9 ± 9	3 ± 9		<0.03	<0.02
60-64	−5 ± 10	14 ± 8	−5 ± 9		<0.03	<0.02

[a] We interpret the maximum concentration in PCBs to have resulted from removal of a dam just downstream of the release area in 1973. [b] Maximum activities in reactor nuclides (^{134}Cs, ^{60}Co, and part of the ^{137}Cs) are attributed to releases made in the year of maximum releases at the reactor site (1971). [c] We interpret the maximum in fallout nuclides to be associated with the year of highest fallout from nuclear weapons testing (1963). [d] We interpret the first appearance of fallout nuclides in this core to be associated with the beginning of measurable fallout from nuclear weapons testing (1954). [e] The first appearance of anthropogenic radionuclides in this core can most probably be associated with the last dredging episode at this site, which apparently occurred in the late 1960s.

PCB ENVIRONMENTAL LOAD BY AROCLOR TYPE

(In Thousands of Pounds)

	Aroclor Type					
	1242	1248	1254	1260	1016	Total PCBs
1930-56	18709	5395	5003	3559	-	32466
1957	1991	704	676	932	-	4303
1958	1431	1065	917	783	-	4196
1959	2298	1597	1142	1118	-	6155
1960	2955	1226	989	1191	-	6361
1961	4082	1745	1295	1347	-	8469
1962	3992	1452	1222	1258	-	7924
1963	3648	2062	1166	1503	-	8379
1964	4597	2160	1224	1664	-	9645
1965	5928	2260	1456	1096	-	10740
1966	7010	1932	1247	1041	-	11230
1967	7442	1794	1158	1111	-	11505
1968	7789	1881	1615	954	-	12239
1969	9182	2190	2172	997	-	14541
1970	10072	1536	2575	1044	-	15227
1971	3232	112	717	266	167	4494
1972	48	-	229	20	1045	1342
1973	310	-	399	-	1177	1886
1974	310	-	309	-	1098	1717
					Grand Total -	172.8 × 10^6 lbs

TABLE
(Taken from Versar, at 288 (1976))

Regression equations describing PCB trends in the tidal Hudson River with distance downstream from the Troy Dam.

Study	Regression equation	N[a]	r^{2}[b]	F[c]	P[d]
EPA 1977	[PCB] = -0.010*RIVERMILE + 7.121	24	0.001	0.024	0.877
Bopp 1979	[PCB] = 0.087*RIVERMILE + 0.621	22	0.247	6.552	0.019*
EPA 1981	[PCB] = 0.002*RIVERMILE + 2.721	10	0.000	0.003	0.955
Shephard et al. 1990	[PCB] = -0.001*RIVERMILE + 1.183	114	0.002	0.169	0.681

a - Number of detectable PCB samples included in regression analysis
b - Coefficient of determination
c - F statistic for the regression analysis
d - Statistical probability that a significant change in PCB concentrations with river mile is observed.
* - Statistically significant at P < 0.05 level

Table 6.2.4-1

Compositions of US PCB Production, of Standard and Evaporated Aroclors, and of Upper Hudson River Reach 9 Deposits and Reach 8 Redeposits

PCB and (% of 1957-77 US Production)		Wt.% Congeners with Indicated Chlorine Number									
		1	2	3	4	5	6	7	8	9	10
Aro. 1221 std.	(0.86)	65.9	26.0	5.4	2.2	0.2	0.2	0.0	0.0	0.0	0.0
Aro. 1232 std.	(0.24)	36.0	18.7	25.0	17.0	2.4	0.5	0.3	0.1?	0.0	0.0
Aro. 1016 std.	(12.88)	0.8	15.5	57.2	25.9	0.3	0.1?	0.0	0.0	0.0	0.0
1950-70 capacitor 1242[a]	--	1.5	11.4	48.9	32.9	4.2	0.88	0.22	0.05	0.01	0.00
Aro. 1242 std.	(51.76)	0.5	14.4	46.6	32.3	5.3	0.9	0.1?	0.0	0.0	0.0
" ", 16.7% evap.	--	0.0	10.4	45.3	36.9	6.5	0.95	0.05	0.01	0.0	0.0
" ", 31.4% evap.	--	0.0	5.1	43.8	42.3	7.5	1.14	0.06	0.03	0.0	0.0
Aro. 1248 std.	(6.76)	0.0	1.3	21.7	60.2	13.4	2.7	0.63	0.12?	0.0	0.0
Aro. 1254 std.	(15.73)	0.0	0.23	1.6	21.7	45.9	27.7	2.7	0.16	0.0	0.0
Aro. 1260 std.	(10.61)	0.0	0.0	0.0	2.0	10.2	42.2	37.3	7.4	0.79	0.0
Aro. 1262 std.	(0.83)	0.0	0.1?	0.5	1.4	5.2	25.8	45.8	19.6	3.5	0.02
Aro. 1268 std.	(0.33)	0.0	0.1?	0.1?	0.3?	0.0	1.9	2.5	24.8	67.9	2.5
Aro. 1270 (from environ.)[b]	--	--	--	--	--	--	--	--	~8	~57	~31
1957-77 US mean	(100.00)	1.6	8.2	34.4	28.6	11.4	9.3	4.9	1.1	0.3	0.01
UHR8 1977+ depos'n.[c]	--	2.9	12.9	33.8	35.5	10.7	3.7	0.58	0.14	0.06	0.00
UHR8 1976 depos'n.[d]	--	6.3	25.2	37.8	22.5	6.1	1.8	0.30	0.05	0.03	0.00
UHR9 pre-1974 depos'n.[e]	--	1.4	11.0	45.9	32.1	6.0	2.1	0.39	0.10	0.08	0.00

a. Estimated from 7 datable GE capacitors spanning 1950-70 time interval.
b. From sediment collected at Watervliet; no standard available.
c. From four 1984 and two 1989 sediment surface samples showing minimal dechlorination and concordant composition.
d. From upper four January, 1977 core sections, which also showed concordant compositions.
e. Estimated original composition based upon PCB congener distributions in 85 sections of six 1977-84 Reach 8 cores, after correction for dechlorination

Table 6.2.5-1

Table 20. Total PCB and other organochlorine concentrations in striped bass taken from the Hudson River in 1990.

Location	Month Collected	No. of fish	Average Length (mm)	Length Range (mm)	Average Weight (g)	Weight Range (g)	Average PCB (ppm)	PCB Range (ppm)	Average DDE (ppm)	DDE Range (ppm)	Average Nonachlor (ppm)	Nonachlor Range (ppm)	Average Dieldrin (ppm)	Dieldrin Range (ppm)
Albany/Troy (RM 153)	July/August	13	619	502-805	2636	1560-5200	6.90	3.13-11.75	0.25	0.08-0.51	0.07	<0.01-0.11	<0.01	<0.01-<0.01
Catskill (RM 112)	June	20	670	485-898	3712	1090-7820	3.55	0.60- 8.79	0.15	0.03-0.33	0.06	<0.01-0.11	<0.01	<0.01- 0.02
Poughkeepsie (RM 76	April	18	659	580-866	3665	2300-9120	3.18	1.00-10.64	0.17	0.05-0.62	0.08	0.03-0.23	<0.01	<0.01- 0.02
	May	31	628	545-820	2718	1620-5720	4.07	0.89-20.01	0.15	0.03-0.99	0.05	<0.01-0.12	<0.01	<0.01- 0.02
	All Dates	49	639	545-866	3066	1620-9120	3.74	0.89-20.01	0.16	0.03-0.99	0.06	<0.01-0.23	<0.01	<0.01- 0.02
Croton Pt. (RM 40)	April	21	704	607-810	4283	2640-7370	3.10	0.76-10.82	0.16	0.05-0.57	0.06	0.02-0.17	<0.01	<0.01- 0.08
	May	14	715	564-887	4539	1880-8390	3.18	0.40-11.33	0.10	0.03-0.55	0.02	<0.01-0.10	<0.01	<0.01- 0.05
	All Dates	35	709	564-887	4386	1880-8390	3.13	0.40-11.33	0.14	0.03-0.57	0.05	<0.01-0.17	<0.01	<0.01- 0.08
Tappan Zee Bridge (RM 27)	April	20	625	475-812	3129	1190-7410	2.72	0.60-14.74	0.12	0.04-0.37	0.04	<0.01-0.10	<0.01	<0.01- 0.02
	May	23	653	551-837	3360	1980-6790	1.50	0.62- 3.50	0.08	0.03-0.25	0.04	0.02-0.09	<0.01	<0.01- 0.01
	All Dates	43	640	475-837	3253	1190-7410	2.07	0.60-14.74	0.10	0.03-0.37	0.04	<0.01-0.10	<0.01	<0.01- 0.02
George Washington Bridge (RM 12)	April	20	672	551-807	3674	1880-6520	2.39	0.64- 9.16	0.11	0.04-0.31	0.05	0.02-0.12	<0.01	<0.01- 0.01
	May	17	722	645-797	4261	3160-5700	1.33	0.30- 5.89	0.08	0.03-0.35	0.04	0.01-0.13	<0.01	<0.01- 0.03
	All Dates	37	695	551-807	3944	1880-6520	1.90	0.30- 9.16	0.10	0.03-0.35	0.04	0.01-0.13	<0.01	<0.01- 0.03
Lower Estuary (RM 12-76)	Spring	164	667	475-887	3595	1190-9120	2.76	0.30-20.01	0.12	0.03-0.99	0.05	<0.01-0.23	<0.01	<0.01- 0.08
Croton Pt. (RM 34-40)	November	2	748	672-823	4690	3600-5780	4.45	4.06- 4.84	0.14	0.14-0.15	0.06	0.05-0.07	0.01	0.01- 0.01
	December	13	617	465-802	2797	1220-5740	4.85	0.51-15.16	0.18	0.03-0.68	0.04	<0.01-0.09	<0.01	<0.01- 0.02
	All Dates	15	634	465-823	3049	1220-5780	4.80	0.51-15.16	0.17	0.03-0.68	0.05	<0.01-0.09	<0.01	<0.01- 0.02

END-USES OF PCTs AND PCBs BY TYPE

End-Use	1016	1221	1232	1242	1248	1254	1260	1262	1268	PCTs
Existing Sales										
Capacitors	XX	X		XX through 1971		X				
Transformers				X		XX	X through 1971			
Sales Phased-Out										
Heat transfer				X						
Hydraulics/ lubricants										
. hydraulic fluids			X	X	X	X	X			
. vacuum pumps					X	X				
. gas-transmission turbines		X		X						
Plasticizers										
. rubbers		X	X	XX	X	X			X	
. synthetic resins					X	X	X	X	X	XX
. carbonless paper				XX						
Miscellaneous Industrial										
. adhesives		X	X	XX	X	X				XX
. wax extenders				XX		X			X	XX
. dedusting agents						X	X			
. inks						X				XX
. cutting oils						X				
. pesticide extenders						X				
. sealants & caulking compounds										XX

Notes: (1) X denotes use of a given Aroclor in a specific end-use, while XX denotes principal use
(2) PCTs denote series 25,44 & 54 Aroclors

Source: Monsanto Industrial Chemical Co.

TABLE 6.4.1-1
(Taken from Versar, at 206 (1976))

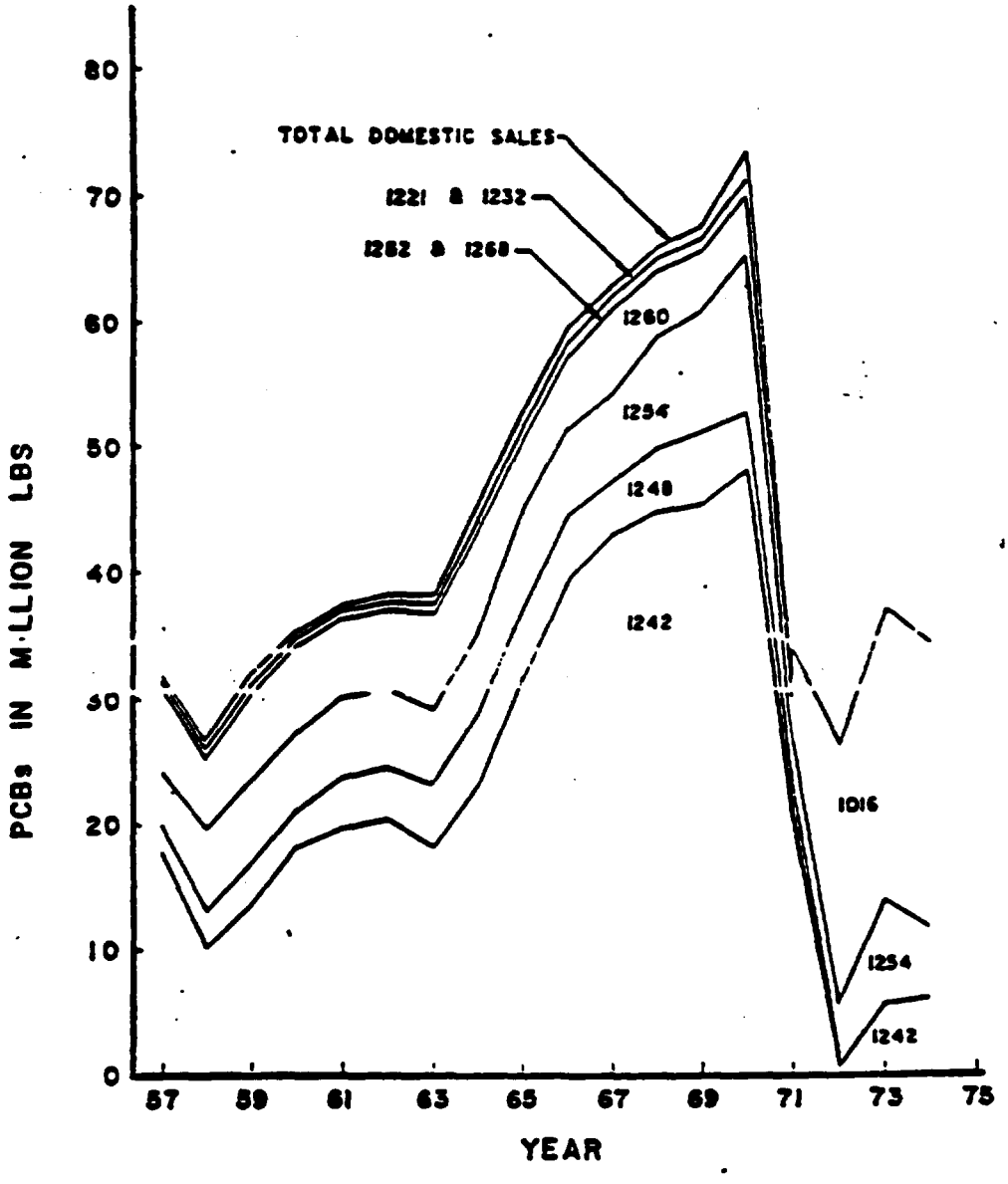
PCBs Concentration in the Effluents of the Machinery & Mechanical Products Manufacturing

Subcategory No.	Manufacturing Operations	PCBs Concentration mg/l (1)		
		Min.	Max.	Avg.
1	Casting & Molding of Nonferrous Metals	0.2	5.1	2.1
2	Mechanical Material Removal	0.2	63.3	6.997
3	Material Forming - All Materials Except Plastics	0.2	63.3	9.867
4	Physical Property Modification	0.2	100.0	12.842
5	Assembly Operations	0.2	104.4	15.553
6	Chemical-Electrochemical Operations	0.5	2.8	1.65
7	Material Coating	0.2	224.8	18.241
8	Smelting and Refining of Non-ferrous Metals	0.2	18.0	9.1
9	Molding and Forming - Plastics	-	None	-
10	Film Sensitizing	0.3	123.9	28.136
11	Dockside Ship Building Activities (2)	-	None	-
12	Lead Acid Battery Manufacture	7.5	30.0	18.75

Note: (1) Information obtained from the Development Document for Effluent Limitations Guidelines for the Machinery & Mechanical Products Manufacturing EPA Contract No. 68-01-2914, Vol. 3, June 1975.

(2) No water effluent; all solid wastes.

TABLE 6.4.1-2
(Taken from Versar, at 311 (1976))



U.S. Domestic Sales of PCBs by Type

FIGURE 6.2.1-1
(Taken from Versar, at 203 (1976))

Table III. Year of Peak Concentration and Appearance[a] of Chemicals in Lake Ontario Sediment Cores

station	peak concn or appearance[a]	DDT	mirex	PCBs	HCB	^{137}Cs
14	P	1967	1967	1971	1967	1968
	A	1945	1961	1950	1942	1953
403	P	1953	1962	1962	1965	1962
	A	1920	1942	1929	1920	1929
64	P	1959	1967	1967	1964	1962
	A	1935	1959	1935	1935	1952
peak productn/usage		1959	1965	1970	1969[b]	1963
start of prodn		1944	1959	1929	1915	1953

[a] Appearance at concentrations greater than 5% of maximum. [b] Total chlorobenzene production (HCB byproduct).

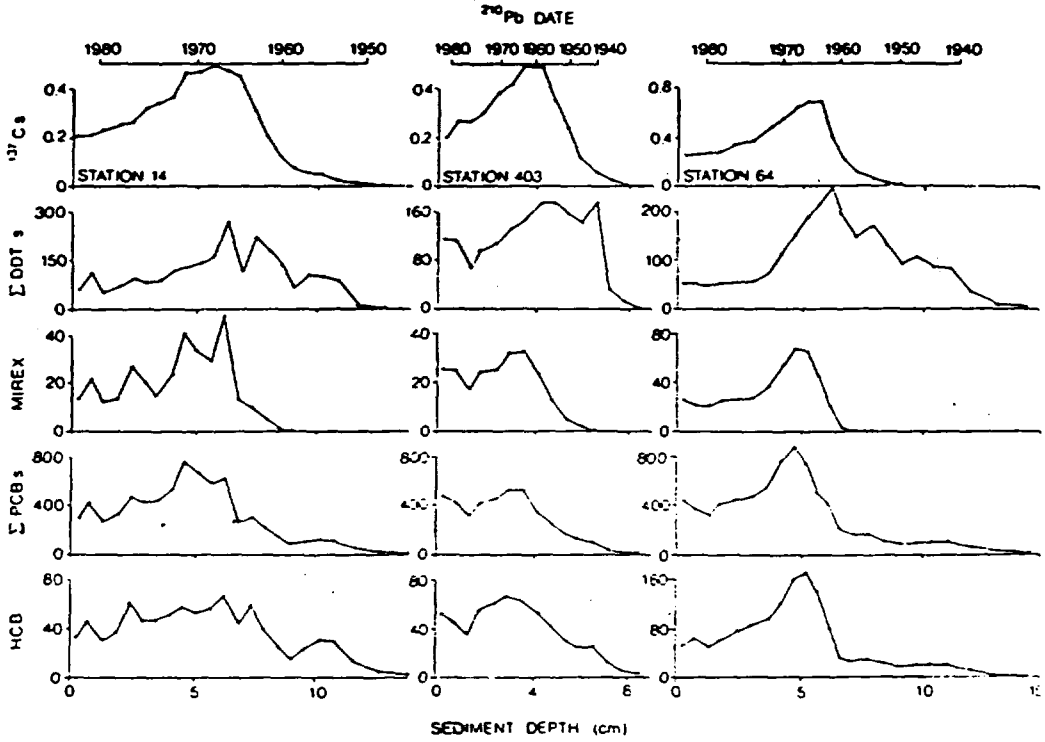


Figure 6. ^{137}Cs (Bq/g), ∑DDT, mirex, ∑PCBs, and HCB concentrations (ng/g) versus sediment depth and age for the three cores.

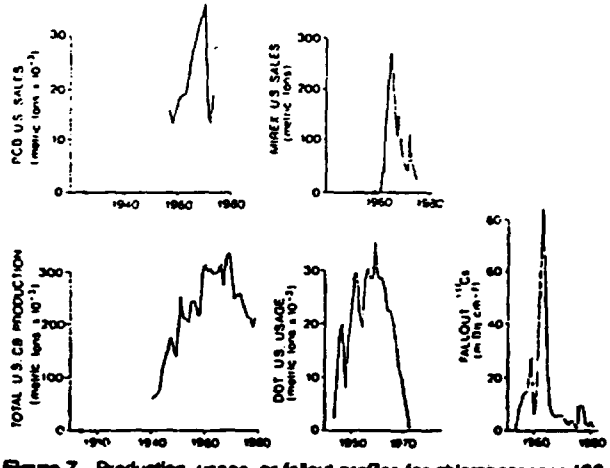


Figure 7. Production, usage, or fallout profiles for chlorobenzenes (*32*, *33*), PCBs (*34*), ∑DDT (*35*), mirex (*36*), and ^{137}Cs (*37*–*39*).

mirex, PCBs, and HCB occurred in the mid 1960s, and there is good agreement between the core record and the production or usage history. This is somewhat later than observed earlier for a single core near the mouth of the Niagara River, where these chemicals appeared to peak between 1959 and 1962 (*10*). The ∑DDT peak occurs in the late 1950s to early 1960s in good agreement with usage patterns. Interestingly, the three DDT pulses in the usage profile appear in cores 14 and 64 but not in the more smeared core 403. The appearance of the chemicals in the cores is reasonably consistent with the production history except in case 403 where the appearance predates the start of production. A downward smearing appears to occur for the organics as well as ^{137}Cs.

The typical behavior of *p,p'*-DDT, its aerobic metabolite *p,p'*-DDE, and its anaerobic breakdown product *p,p'*-DDD is shown for core 64 in Figure 8. DDT comprises only ~25% of the total DDT near the core surface. This observation is not surprising since DDT use was banned in 1972 and its reported half-life in soils is of the order of 3–5 years (*40*, *41*). The DDT abundance drops gradually to less than 2% at the bottom of the core, and a large relative concentration increase for DDD is observed. Similar trends were found in the other cores. The half-life of the first-order conversion of *p,p'*-DDT to *p,p'*-DDD in the sediments can be derived by plotting ln [DDT/(DDT + DDD)] versus time (Figure 9). The ratio must be used instead of the absolute DDT concentration because of the changing amount of total DDT in the various core segments. The linear correlation coefficients (r^2) for the plots

Figure

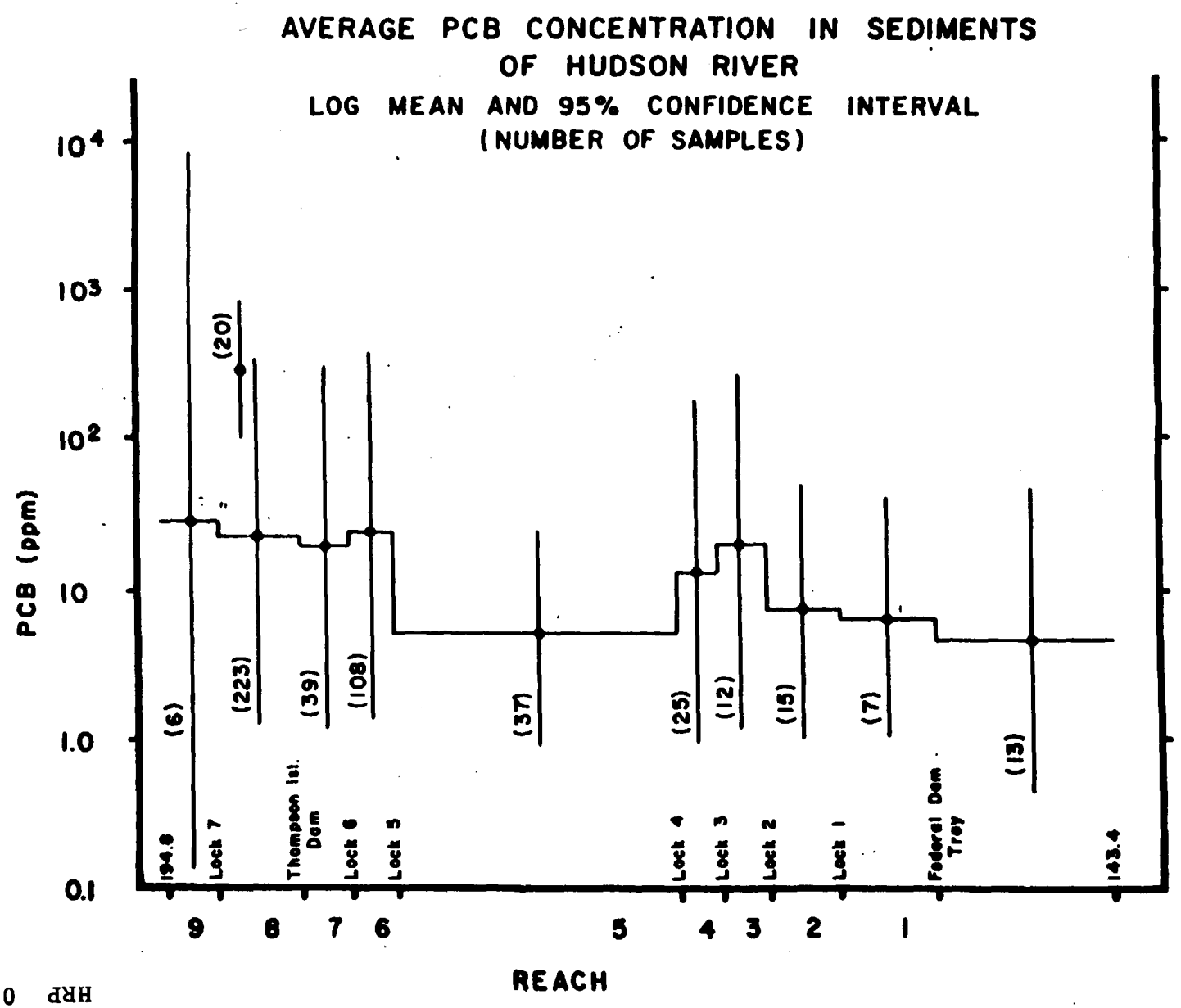


Figure 6.2.4-1

MEAN PCB LEVELS IN HUDSON RIVER GAMMARUS (1978-1981)
(from J.M. O'Connor)

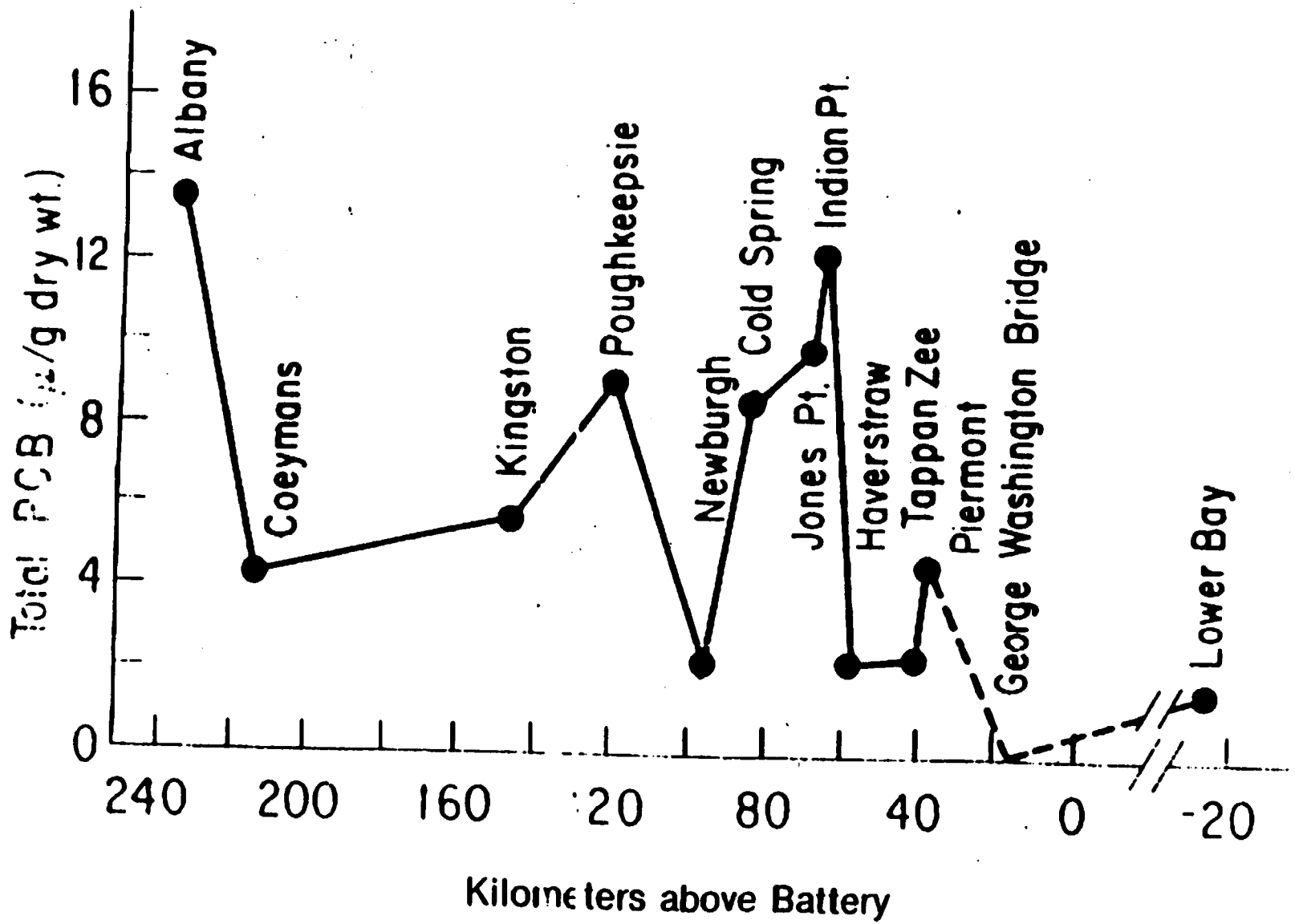


FIGURE 6.2.4-2

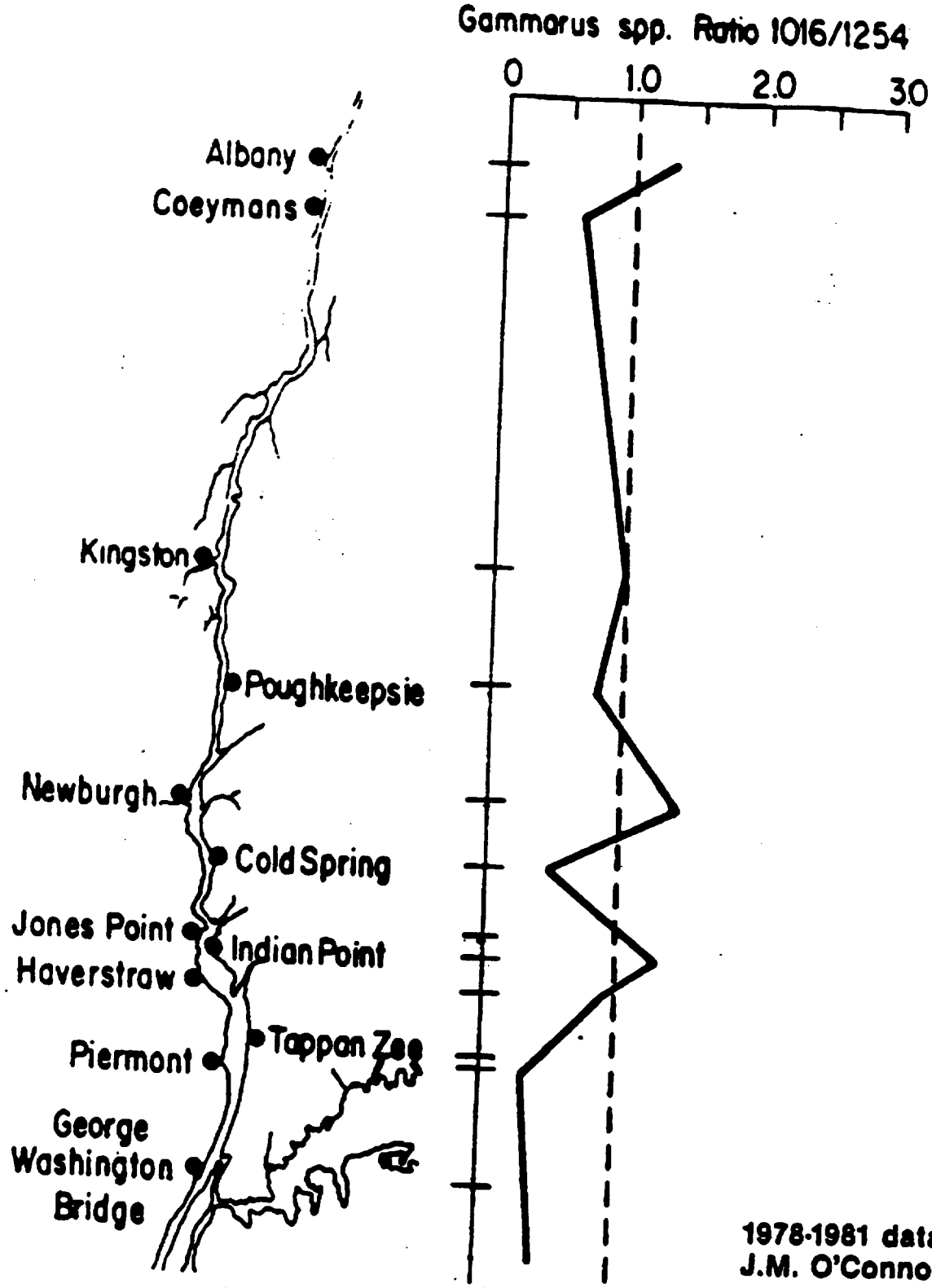


FIGURE 6.2.4-3

PCB Trends in Surficial Sediments
Hudson River

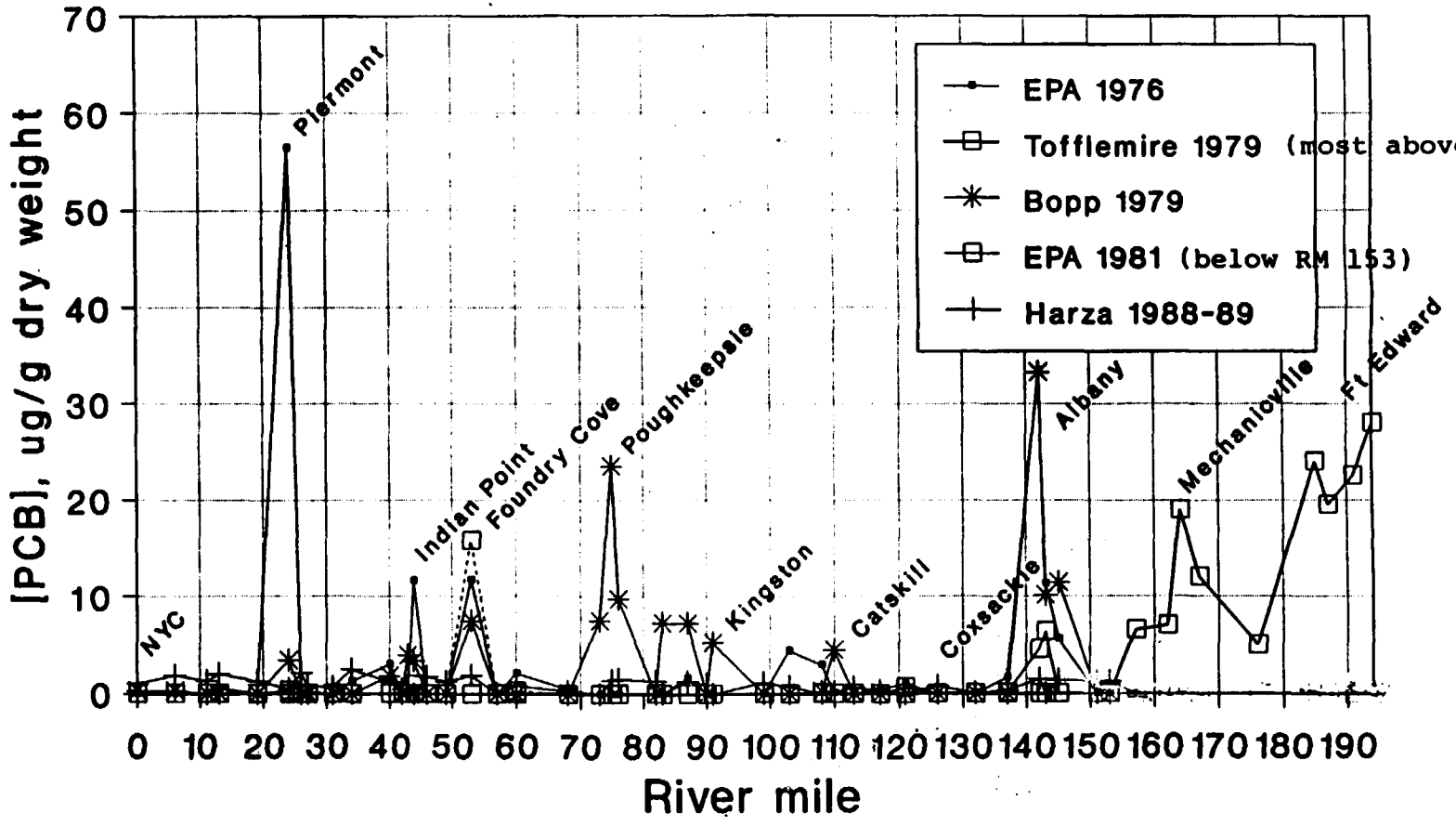


Figure 6.2.4-4