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CRITICAL ISSUES REGARDING THE BIOACCUMULATION OF PCBS FROM THE WATER COLUMN AND SEDIMENT OF THE UPPER HUDSON RIVER

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CONTENTS

FIGURES

TABLES

SECTION 1

INTRODUCTION

Predictions of the changes in PCB concentrations in fish in the upper Hudson River for both natural recovery and various remedial scenarios are needed as a basis for sound regulatory judgments. This paper reviews a number of important issues related to constructing a bioaccumulation model for the upper Hudson River and describes an approach that if adopted by EPA should provide the capability to simulate the PCB levels in fish under various future conditions.

PCB concentrations in Hudson River fish are expected to decline due to natural attenuation of exposure concentrations. A goal of the EPA Hudson River Superfund Reassessment Remedial Investigation and Feasibility Study (RRI/FS) is to estimate what the rate of decline will be and whether the rate can be significantly accelerated by feasible remedial actions. Such estimation is complicated because the fish can obtain their PCBs from multiple media, including the water column, surficial sediment, and subsurface sediment. The contributions of these media to fish PCBs vary in space and time depending on the structure of the food web and the relative PCB concentrations in the media. Thus, relationships between fish PCB and media PCB concentrations can vary widely by location and over time. The relationship in Thompson Island Pool, for example, is different from the relationship in Stillwater, and both have changed over the historical record (Figure 1-1). Further, the relationships can be expected to change in the future as PCB concentrations in the various media decline at differing rates that depend on future trends, on the action considered, and location within the river system.

Fish:media PCB concentration ratios also vary because of the slow response of the fish to changes in exposure concentrations. For example, the fish did not achieve steady-state with the high water column concentrations that occurred in late 1991 and 1992, and will require a few years to recover from these events.

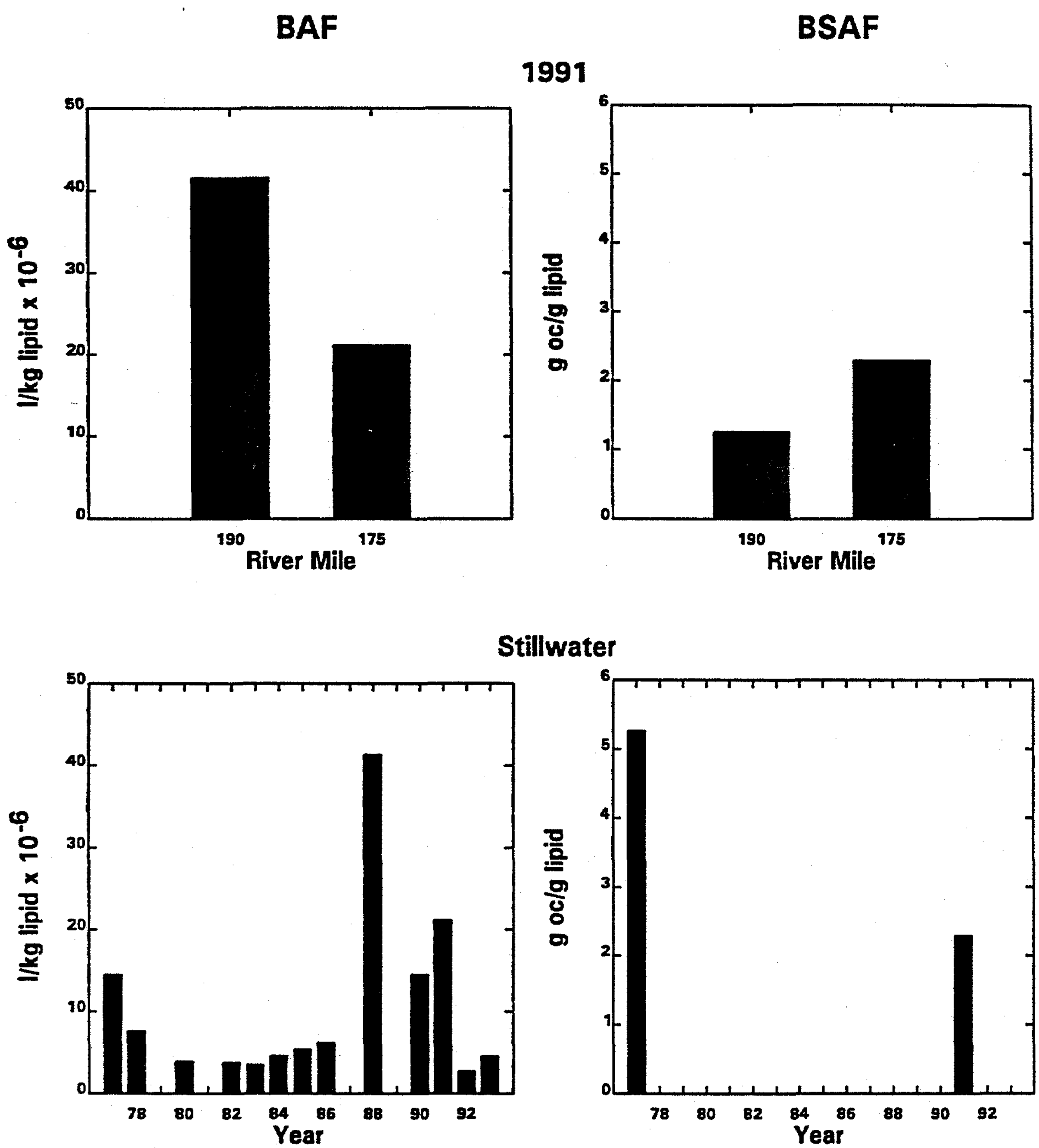

Figure 1-1 Bioaccumulation Factors (BAF) and Biota-Sediment Acculumation Factors (BSAF) for Largemouth Bass from Upper Hudson River. Upper panels: 1991 factors for TIP and Stillwater, bottom panels: 1977-93 factors for Stillwater.

Finally, physiological variations in the fish, specifically changes in fat content, cause changes in fish PCBs that can occur independently of changes in media PCB concentrations. For example, mean lipid content of Stillwater largemouth bass varies by as much as a factor of two between successive years and by over an order of magnitude over the period of record. These changes in lipid content alter the PCB excretion rate and cause increases or decreases in PCB body burden that occur even if the media concentrations remain constant.

Simple relationships, such as BAF and BSAF, between Upper Hudson River fish and media PCB concentrations, if they exist, are misleading, because they describe an average of the historical relationships that may not be applicable to future conditions. For this reason, a bioaccumulation model developed by statistical analysis of the historical data will have little predictive power. The main deficiency of a statistical approach is that it lacks causality. Causality is achieved by explicit descriptions of the mechanisms of PCB bioaccumulation in the river. Such descriptions are embodied in dynamic food web bioaccumulation models that have been successfully developed and used at other sites. Examples are listed in Table 1-1. These models quantitatively characterize the contaminant transfer pathways and the rates of uptake and loss that account for the dynamics of the fish:media PCB ratios. They can interpret the changes in fish:media PCB relationships that result from spatial and temporal changes in media PCB concentrations and changes in fish physiology.

This paper begins with the assertion that credible evaluations of future fish PCB concentrations can be made only using a food web bioaccumulation model. The development of such a model is predicated on the ability to define the food web (i.e., the contaminant transfer pathways), characterize the bioenergetics and toxicokinetics of the food web components, and calibrate and test the model. The purpose of this paper is to 1) identify the components of the food web and the interaction of those components, and 2) to identify elements within the existing data base which can be used to develop and test the model. Bioenergetics and toxicokinetics are not discussed. Previous experience indicates that these components of the model can be effectively characterized by readily available published data.

TABLE 1-1. EXAMPLES OF PREVIOUS APPLICATIONS OF FOOD WEB BIOACCUMULATION MODELS			
System	Contaminant	Food Web Leading to	Reference
Lake Michigan	PCBs	lake trout	Thomann & Connolly, 1984
Lake Ontario	PCBs	lake trout	Connolly & Thomann, 1992
James River Estuary	Kepone	striped bass	Connolly & Tonelli, 1985
Hudson River Estuary	PCBs	striped bass	Thomann et al., 1989
New Bedford Harbor	PCBs, Cd, Cu, Pb	winter flounder, lobster	Connolly, 1991
Green Bay	PCBs	walleye, brown trout	Connolly et al., 1992
Southern California Bight	DDE, PCBs	white croaker, Dover sole, kelp bass	HydroQual, 1994
Lake Ontario	dioxins, PCBs	lake trout	Endicott & Cook, 1994
Canadian rivers	dioxin	whitefish, suckers	Muir et al., 1992

SECTION 2

REPRESENTATION OF THE FOOD WEB

The food web describes the pathways of contaminant transfer from the various media to the fish. To predict the transfer of PCBs from buried and surficial sediments and the water column to species of interest, species can generally be grouped according to the prey consumed, foraging area, and physiological characteristics, in particular lipid content and net growth efficiency. The fish community of the upper Hudson River includes species that can feed on fish, on benthic invertebrates, and on invertebrates living on plant surfaces. Young of many species may feed on plankton. Thus, a basic food web consisting of a top predator, forage fish, and invertebrates encompasses the range of trophic levels in the upper Hudson River.

Critical elements in a quantitative food web analysis for the upper Hudson are the links to the various media: subsurface sediments, surface sediments, and water column particulates. The invertebrate level provides these links. Some invertebrates feed on suspended particles or on periphyton, others feed on the surface sediment, and still others feed on deeper sediment. For a bioaccumulation model, representative invertebrate groups are based upon these three feeding strategies. The relative contributions of each of these types of invertebrates to the diet of the forage fish, in combination with the relative concentrations of PCBs in the particulate matter they ingest, determine the relative importance of each pathway.

When deposit-feeding invertebrates are a component of the food web, a series of issues arise because of the large areal and vertical variations in sediment PCB concentrations. Among these are the following two: 1) the depth of sediment over which the invertebrates feed, and 2) the rate at which sediments below the maximum depth of feeding may be incorporated into the feeding zone by mixing processes. These issues have implications for predicting temporal changes in fish PCB levels. The rate of decline in invertebrate PCBs due to sediment burial will be inversely related to the depth of bioavailable sediments. The shallower this depth, the faster the rate at which invertebrate PCB levels will decline.

The key issues associated with more precisely defining the food web for contaminant transfer in the upper Hudson River are discussed below. The most important questions relate to:

- choosing a species to represent each trophic level
- determining foraging location

2.1 CHOOSING SPECIES TO REPRESENT EACH TROPHIC LEVEL

2.1.1 Top Predators

By default, any analysis of bioaccumulation in the Upper Hudson River must focus on largemouth bass. It is the only species of interest from a human health risk assessment perspective for which a substantive PCB database exists. Fortunately, largemouth bass is reasonably representative of most of the top predators of the upper Hudson River. This predator feeds largely on fish, but also on other large prey such as crayfish (Carlander 1977). This mixed diet is also characteristic of most of the important predators listed in Table 2-1, including smallmouth bass, chain pickerel, yellow perch, black crappie and walleye (Smith 1985). One exception is the northern pike, which is more specialized as a piscivore (Smith 1985).

TABLE 2-1. FISH SPECIES OF THE UPPER HUDSON RIVER	
COMMON NAME	SCIENTIFIC NAME
American eel	Anguilla rostrata
Northern pike	Esox lucius
Chain pickerel	Esox niger
Common carp	Cyprinus carpio
Silvery minnow	Hybognathus nuchalis
Comely shiner	Notropis amoenus
Satinfin shiner	Notropis analostanus
Emerald shiner	Notropis atherinoides
Bridle shiner	Notropis bifrenatus
Golden shiner	Notemigonus crysoleucas
Spottail shiner	Notropis hudsonius
Bluntnose shiner	Pimephales notatus
Fallfish	Semotilus corporatis
White sucker	Catostomus commersoni
Brown bullhead	Ictalurus nebulosus
Rock bass	Ambloplites rupestris
Redbreast sunfish	Lepomis auritus
Green sunfish	Lepomis cyanellus
Pumpkinseed	Lepomis gibbosus
Smallmouth bass	Micropterus dolomieui
Largemouth bass	Micropterus salmoides
Black crappie	Pomoxis nigromaculatus
Yellow perch	Perca flavescens
Walleye	Stizostedion vitreum

2.1.2 Forage Fish

Predatory fish are generally size-specific predators, implying that forage fish availability will determine what species are eaten. The potential forage fish listed in Table 2-1 exhibit a range of behaviors and may spend time both in deeper waters and in shallow weedy areas. They consume a mixture of plankton, benthos and vegetation-dwelling invertebrates.

The most complete PCB data set for any forage fish of the upper Hudson River exists for pumpkinseed. Therefore, a food web bioaccumulation model which uses the pumpkinseed to represent the forage fish trophic level provides the best opportunity for calibration. Pumpkinseed is appropriate as an indicator species for forage fish of the upper Hudson River. It is similar to most of the other forage species in that its diet includes a mix of invertebrates that may be receiving their PCB loads from both the water column and surface and buried sediments. It is known to consume both planktonic and benthic invertebrates (Keast and Welsh 1968, Mittelbach 1984, Collins and Hinch 1993). In a study of gut contents of pumpkinseed in Thompson Island Pool, Feldman (1992) found 85 percent snails, which are generally surface grazers. This surface material may contain algae, invertebrates or detritus and may include particulates that have been in the sediment bed for some time as well as newly settled material and growing algae. It should be noted that the fish were collected as part of a single sampling in August, 1988, and therefore provide only a limited picture of the long-term average diet of the pumpkinseed.

2.1.3 Invertebrates

The invertebrate community includes species that can potentially accumulate PCBs from the subsurface and surface sediments and the water column. Surface sediment is potentially important because of the dominance of chironomids in the five surveys of the benthic invertebrate fauna of the upper Hudson River conducted since 1972 (Tables 2-2 and 2-3). Chironomids feed on the surface of the bed. Further, caddisflies, another common invertebrate in the surveys, feed from water column particulates, some of which could be resuspended surface sediment particles. The surveys indicate that oligochaetes are also an important component of the benthic fauna. These invertebrates feed within the sediment bed and can make subsurface PCBs available to the food web. Finally, young fish may feed on plankton, which derive their PCBs from the water column.

TABLE 2-2. SURVEYS OF INVERTEBRATES IN THE UPPER HUDSON RIVER			
Dates of Collection	Locations	Method of Collection	Reference
Aug - Oct 1972	12 stations, upstream of Corinth to Troy	multiplate samplers	Simpson 1976
Summer 1976 and Summer 1977	8 stations, Hudson Falls to Troy	multiplate samplers	Hetling et al. 1978
Summers, 1978-1985	10 stations, Hudson Falls to Troy	multiplate samplers	Novak et al. 1988
November, 1990	1 station in Moreau Pool and 3 in Thompson Island Pool	Petite Ponar sampler (6" X 6")	survey performed by Law Environmental
Summer 1988	2 stations in Thompson Island Pool: one *Vallisneria* bed and one *Trapa* bed, in 1.5 m deep water.	Invertebrates from plant surfaces collected with polyethylene tube	Feldman, RS. 1992.

TABLE 2-3. INVERTEBRATES IN THE UPPER HUDSON RIVER			
Oligochaetes	Chironomids	Source of material	Reference
14 %	70 %	multiplate samplers	Simpson 1976
15 -20 %	60 -70 %	multiplate samplers	Hetling et al. 1978
chironomids, oligochaetes and caddisflies were the most abundant invertebrates [a]		multiplate samplers	Novak et al. 1988
38 %	23 %	sediment bed	survey performed by Law Environmental
0 %	38, 60 % (Vall.) 8, 26 % (Trapa)	plant surfaces	Feldman, RS. 1992.
Note: [a] data not reported. Text is based on discussion in the text of the reference			

Based on fish feeding behavior, PCBs in the food web are more likely to originate in surface sediments and water column particulates than in subsurface sediments, but the latter cannot be ruled out. Chironomid larvae, which are surface sediment feeders, form a significant portion of the food of fishes in general (Pennak 1978). In addition, fish often use visual or hydromechanical cues to locate prey, so invertebrates feeding at the surface may be expected to be more available than those feeding at depth. A possible exception to this for some fish in some environments may be conveyor-belt-feeding oligochaetes, the ends of which stick up from the sediment into the water column.

Pumpkinseeds in particular are likely to feed on invertebrates that consume surface sediment and water column particulates (Keast and Welsh 1968). Pumpkinseeds feed extensively on vegetation-dwelling prey and on gastropods (Mittelbach 1984). In addition, planktonic feeding by pumpkinseeds can be important (Collins and Hinch 1993). Invertebrates dwelling on vegetation are most likely receiving their PCBs from water column sources, perhaps *via* periphyton. Snails scrape the surfaces of macrophytes and the surface sediment. Chironomids may feed to a depth of 0.5 to 1.0 cm (Pennak 1978). Thus, a bioavailable depth of about 1 cm is suggested. Oligochaetes were found to compose less than one percent of pumpkinseed diets in Lac Vert, Quebec (Beaulieu et al. 1979), and were not mentioned as important prey for pumpkinseeds by Mittelbach (1984).

To the extent that oligochaetes are important in the diet of some forage fish, deeper sediments may contribute some PCBs to the food web. Pennak (1978) remarks that particles may be ingested by oligochaetes at a depth of 2 to 3 cm. McCall and Fisher (1979), analyzing data of Davis (1974) for profundal lake sediments, found that 70 percent of the tubificids were found within the top 3 cm of the bed, but because bigger worms dig deeper, the maximum tubificid feeding activity occurred at 3 to 6 cm depth.

Deeper sediments contribute PCBs to the food web only if there is sufficient vertical mixing to bring material within the biologically active zone. This can occur in two ways: (1) bioturbation due to the activities of subsurface deposit feeders, and (2) periodic turbulent mixing under flood flow conditions. The extent of such mixing in the Hudson is not known. Analysis of the vertical profiles of short-lived radiotracers such as ^{7}Be as well as of PCB congeners could provide some assessment, and the EPA high-resolution cores provide an opportunity to explore this possibility. However, it would be difficult to collect sufficient data to define reasonable averages for the river. The best estimate of vertical mixing can probably be obtained from the EPA PCB fate model currently being developed. The rate at which water column PCBs decline in such a model would be dependent on the rate of decline of surface sediment PCBs. This rate would, in turn, be dependent on the sedimentation rate and mixing with lower layers of sediment. The sedimentation rate is independently estimated by the change in the PCB mass within the full sediment column (HydroQual 1995).

In conclusion, the base of the food web is most likely to consist of a community of invertebrates that feed on a mixture of surface sediments and water column particulates. These two invertebrate groups provide a basis for developing a realistic quantitative model of the bioaccumulation of PCBs (Figure 2-1). The extent to which subsurface sediments contribute can be determined in conjunction with a fate model.

2.2 DETERMINING FORAGING LOCATIONS

Two major points are presented here. First, it is suggested that most feeding occurs in shallow weedy areas, and that in particular, pumpkinseeds are likely to feed from these areas. Second, it is shown that even if feeding location varies between fish species, the type of areas in which forage fish feed is not likely to make a difference to the accumulation in the food web, because concentrations on an organic carbon basis are similar in both coarse and fine sediments.

Most organic material inputs to the food web are likely to be from shallow areas and from the water column. Forage fish can range in general from protected areas to deeper waters (Smith 1985). However, it is likely that they do most of their feeding in areas of greater productivity. The primary sources of organic material in the upper Hudson River are macrophytes, periphyton, plankton, and allochthonous inputs. Macrophytes occur in shallow areas and are the main substrate on which periphyton grow. Allochthonous inputs of organic material (leaves, runoff) are likely to deposit primarily in shallow areas. Planktonic production can occur throughout the river. Planktonic material, including drift

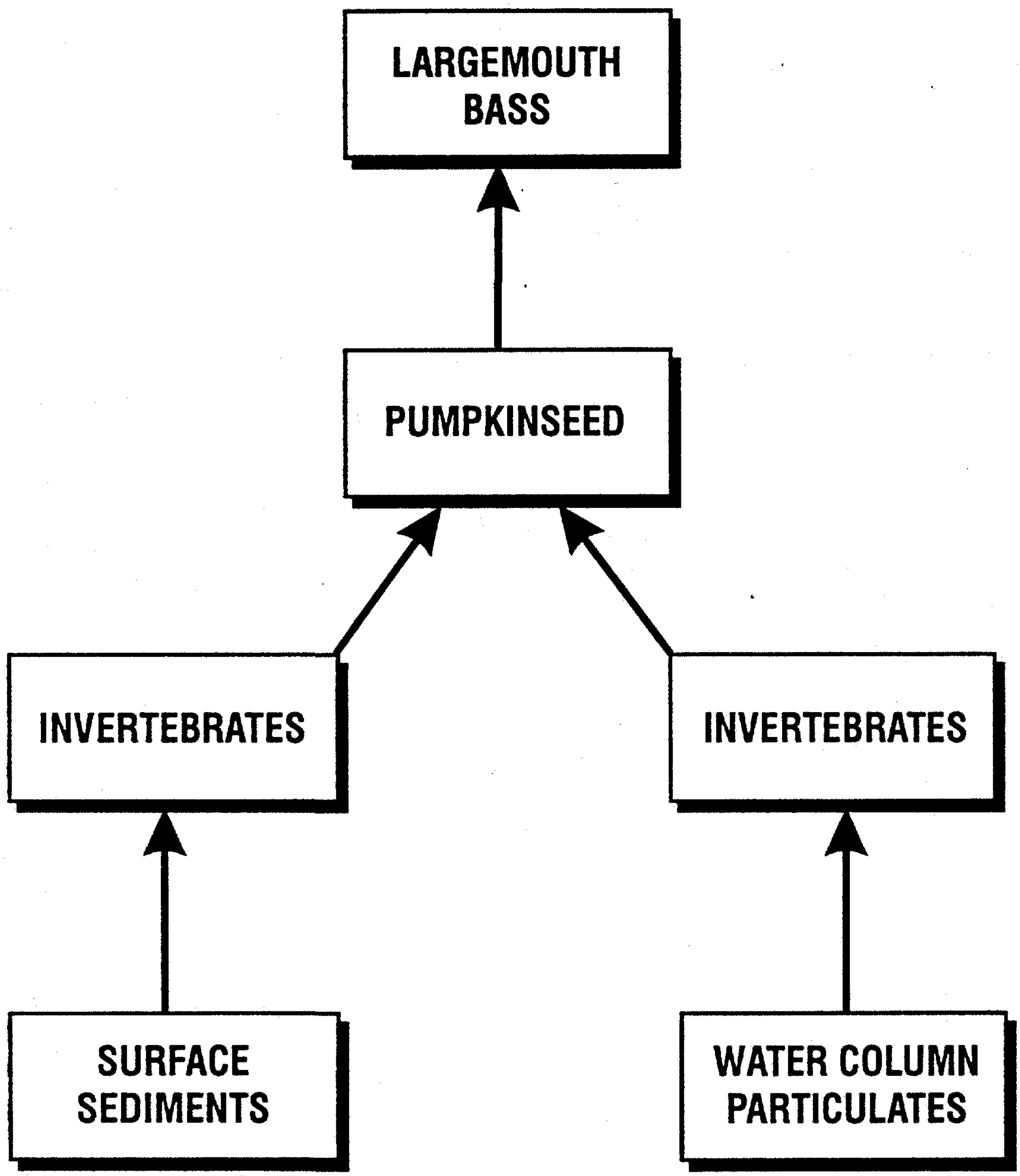

FIGURE 2-1. FOOD WEB FOR A BIOACCUMULATION MODEL

from upstream, can settle onto the sediment bed, a process that occurs primarily in depositional areas, that is, shallow vegetated areas. Thus, the base of the food web is likely to include sediments and water column particulates present in shallow, weedy areas.

The pumpkinseed favors vegetated areas of rivers (Werner 1980), and the importance of snails in the diet of pumpkinseed is consistent with the presumed importance of shallow, weedy habitats. Thus, the pumpkinseed is a reasonable choice for a representative forage fish in a bioaccumulation model, because its tendency to forage in weedy areas is consistent with the likely importance of these areas as sources for PCBs in the food web.

Even if there is variation among fish in their foraging locations, and some feed in areas of coarse sediments and some in areas of fine sediment, the accumulation to the food web is likely to be similar. Sediment PCB concentrations vary greatly within the river. Variation on small spatial scales may be averaged out on the basis that the fish integrate exposure over a certain area. Dealing with larger scale variation that correlates with sediment type is generally considered to be more difficult. Typically, PCB concentrations are higher in fine sediments than in coarse sediments. These differences may not be important to the food web, because, within each pool of the river, expressing PCB levels as PCB mass per unit sediment organic carbon effectively eliminates differences among sediment types. This is illustrated in Figure 2-2 which presents the distributions of PCB concentrations (ug/g dry sediment and ug/g organic carbon) in fine and coarse surface sediments (0 to 5 cm) collected from the Thompson Island Pool in 1991. The elimination of differences among sediment types that occurs with carbon normalization reflects the common water source for all of the sediments. Particles depositing on the sediment have the same carbon-based PCB concentration that is defined by sorption processes in the water column. Areas of fine sediment have greater dry weight-based concentrations because of the greater particle flux to the sediment in these areas.

Given that carbon-based surface sediment PCB concentrations within a single pool of the river may be described by a single distribution, the PCB concentrations in deposit-

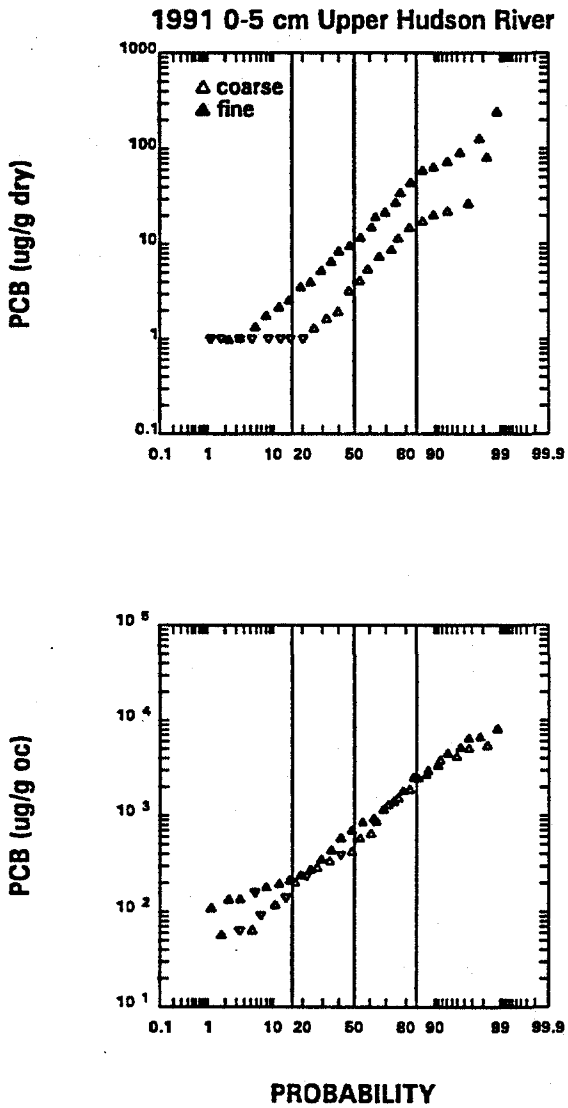

Figure 2-2 Distribution of PCB concentrations measured in surface sediments (0 to 5 cm) of Thompson Island Pool in 1991. Top panel: concentrations expressed on a dry weight basis. Bottom panel: concentrations expressed on an organic carbon weight basis.

feeding invertebrates may be treated as independent of location. All of the invertebrates receive the same dose because, for practical purposes, their rate of ingestion of organic carbon is independent of location. In other words, the invertebrates ingestion rate of sediment varies with the organic carbon concentration of the sediment in a manner that results in similar organic carbon ingestion rates and thus similar PCB ingestion rates.

SECTION 3

CALIBRATION AND VALIDATION OF A FOOD-WEB BIOACCUMULATION MODEL

The wealth of PCB data that have been collected in the upper Hudson River provide an unusual opportunity to calibrate and validate a food web bioaccumulation model. During the 18 years of recorded data, there were long-term changes in sediment and water column concentrations and short-term pulses in water column concentrations. Spatial gradients in water and sediment concentrations have been observed. PCB concentrations have been measured in both pumpkinseeds and largemouth bass over 18 years in two locations.

Most importantly, the **relative** concentrations of PCBs in water and sediment varied temporally and spatially:

- Concentrations in sediments and water column exhibited declines during the period from the late 1970's to the 1990's. The rates of decline were not the same.

- Concentrations in surface sediment differ between Thompson Island Pool and Stillwater, whereas the water column concentrations do not.

- Concentrations in the water column exhibited a temporary spike in September 1991, followed by continued elevated levels for approximately two years.

This variation in the relationship between water and sediment concentrations can be used, with a time-variable model, to quantify the relative importance of each source to the fish. In fact, calibration to the full data set will overcome the initial uncertainty regarding the dietary composition of the forage fish (that is, the relative contributions from sediment and water column organic carbon). Thus, this data set provides the unique opportunity to develop and rigorously test a food web bioaccumulation model.

Confidence in the resulting food web bioaccumulation model can be established by using the data prior to September, 1991, when high PCB releases began, in the calibration and then using 1992 and 1993 data for model validation. Comparison of observed and computed '92 and '93 levels will provide checks on both the pathway specification and the kinetics of PCB uptake and loss. The kinetics are checked in this fashion because the duration of the high concentrations is much less than the time to steady-state in the food web.

In conclusion, these data provide an opportunity to quantitatively assess the relative importance of surface sediments and water column-associated particulates as ultimate PCB sources for the food web. Because the variations in each of these compartments occurs on scales ranging from daily to multi-year, a time-variable physiologically-based food web bioaccumulation model is necessary to take full advantage of this opportunity. By contrast, a statistical model will ascribe much of the temporal and spatial variability in fish:media PCB relationships to residual uncertainty. As a result, a statistical model is likely to have wider uncertainty bounds than the food web model and be less able to distinguish among various remedial alternatives.

SECTION 4

CONCLUSIONS

- Statistical models are inappropriate for predicting future PCB concentrations in upper Hudson River biota because they do not properly account for the dynamic nature of the relationship between PCBs in biota and their environment.

- A representative food web for calculating transfer of PCBs in the upper Hudson River should include a top predator, a forage fish, and invertebrates linking the food web to exposure sources.

- The largemouth bass is representative of many of the top predators in the upper Hudson.

- The pumpkinseed is an appropriate indicator species for the forage fish trophic level. The conclusion that its diet is likely to be a mix of surface sediment and water column particulates is consistent with the mix of available invertebrates and forage fish observed in the upper Hudson River.

- Based on natural history information, surface sediments (0 to 1 cm) and the water column are likely to be the primary sources for PCBs for Hudson River forage fish in general and for pumpkinseeds in particular.

- The relative importance of surface sediment and water column particulates cannot be determined based solely on natural history information.

- Pumpkinseeds forage in weedy areas, which are probably the primary sources of material for the fish food web. However, the availability of contaminants to the food web would not be affected if some forage fish feed in areas of coarse sediments. This is because carbon-based concentrations of PCBs are the same in fine and coarse sediments.

- The PCB data base for the upper Hudson River provides an unusual opportunity to develop and test a time-variable physiologically-based bioaccumulation model.

- Calibration of the model to the historical data base can be used to quantitatively define the link between the forage fish and the invertebrates and, thus, the relative importance of surface sediment and water column particulates

- The increases in concentration that occurred beginning in September 1991 provide an opportunity to validate the food web model.

SECTION 5

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