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Polychlorobiphenyl (PCB) Congeners in Striped Bass (*Morone saxatilis*) from Marine and Estuarine Waters of New York State Determined by Capillary Gas Chromatography

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Abstract. Edible filets of striped-bass samples (60) from the Hudson River estuary, the Atlantic shore of Long Island, and Long Island Sound were analyzed by high-resolution glass-capillary gas chromatography for 74 polychlorobiphenyl (PCB) compounds. The fish, a non-random subset of a larger sample, contained the following concentrations of total PCB residue (mean ± the standard error of the mean). The corresponding mean values for the entire survey (N = 717) are given in parentheses: eastern Long Island Sound 1.8 ± 0.4 (1.91 ± 0.1) mg/kg; western Long Island Sound, 1.9 ± 0.2 (2.51 ± 0.1) mg/kg; eastern Atlantic shore, 3.0 ± 0.5 (2.45 ± 0.2) mg/kg; western Atlantic shore, 7.5 ± 1.9 (3.04 ± 0.2) mg/kg; Hudson River, 15 ± 3 (6.19 ± 0.4) mg/kg. All the samples contained PCB residues derived from Aroclors 1242, 1016, 1254, and 1260 but none of the residue present came from Aroclor 1221 as evidenced by the complete absence of 4-chlorobiphenyl from the fish. Fish from the Hudson River and the majority from the Atlantic shore of Long Island contained 2,2′- and 2,6-dichlorobiphenyls indicating recent exposure to Hudson River PCBs. On this basis, there was evidence that a subpopulation of fish was not exposed to the Hudson River PCB source. The detailed PCB congener make-up of the typical residue found in these fish differs from that found in the majority of human foodstuff in that it contains a proportionately large amount of mono- through tetra-chlorinated PCB. The mean concentration of the more toxicologically suspect congeners which are not readily degraded by mammals and birds (those with two 4-substituents) comprise less than 1 ppm in Long Island Sound fish.

Polychlorobiphenyl (PCB) contamination of the Hudson River and the bioaccumulation of PCBs in species like the striped bass (*Morone saxatilis*) began nearly thirty years ago when PCBs were first discharged into the river, and is still evident today. The entire Hudson River estuary has been contaminated by PCBs (Horn *et al.* 1979) in excess of 2.5×10^5 kg. The majority of the PCB contamination of the Hudson River estuary, from Aroclors® 1016, 1242, 1254, and 1260 resulted from a solvent washing process by an electrical capacitor manufacturer. High levels of PCB were found in the edible flesh of the striped bass in 1978 (mean 18.1 ppm total PCB) (Horn and Sloan 1985). Earlier findings of similar concentrations (Spagnoli and Skinner 1977) led to the 1976 closure of the commercial striped bass gill net fishery, and the issuance of annual fish consumption health advisories to recreational fishermen (Sloan and Horn 1986). Sediment core samples taken from areas in the upper Hudson River are highly contaminated by PCBs, many at levels exceeding 50 ppm total PCB (Brown *et al.* 1985; Bush *et al.* 1987).

Apiezon L and the related synthetic C-87 hydrocarbon Apolane are the only stationary phases which resolve 3- and 4-dichlorobiphenyl and 2,2′- and 2,6-dichlorobiphenyls (Erickson 1986; Bush *et al.* 1985a). These congeners are components of Aroclors 1221, 1016, 1242, and 1248 and analysis on these stationary phases has indicated characteristic PCB patterns in water samples from the Hudson River. At low-flow periods in the summer, more than 50% of the PCB transported downstream via the water column was made up of 2-chlorobiphenyl and 2,2′- and 2,6-dichlorobiphenyls (Bush *et al.* 1985b). Similar PCB patterns were reproduced in laboratory experiments by pumping water very slowly over PCB-contaminated sediment in a continuous sediment release reactor (Wood *et al.* 1987). The PCB patterns that are found in water tend to have greater concentrations of the less chlorinated PCB homologs (congeners with one to three chlorine substitutions) than those with a greater degree of chlorination. Most of the recent samples of water, sediment and biota contain very little 4-chlorobiphenyl which comprises 25% of Aroclor 1221 (Bush *et al.* 1985a; Sissons and Welti 1971). This is indicative of the probable elimination of Aroclor 1221 as a component of current PCB pollution in the river (Novak *et al.* 1988; Bush *et al.* 1985b, 1987; Sloan *et al.* 1983, 1984, Armstrong and Sloan 1988; Sloan and Armstrong 1988).

The large number of data points anticipated from this

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Table 1. Recovery of PCBs from a spiked fish sample (in quadruplicate)

PCB Congener	ng recovered	ng in unspiked fish	Mean ng in spiking solution	Recovery %	Lotus[a] Ref #
2	3323	0	4144	81 ± 3	1
2,2′	785	2	822	95 ± 2	2
2,6	99	1	111	88 ± 2	3
4	2025	0	1842	110 ± 1	4
2,5	115	0	115	100 ± 2	5
2,4	154	0	151	102 ± 2	6
2,3′	411	0	433	95 ± 2	7
2,6,2′[b]	207	2	221	93 ± 2	8
2,4′	1446	3	1504	96 ± 3	9
2,5,2′	950	9	1015	93 ± 3	10
Hexachlorobenzene	207	2	231	89 ± 4	11
2,4,2′	326	3	345	94 ± 4	12
2,3,2′	391	4	463	84 ± 4	13
2,6,4′	304	6	407	73 ± 4	14
4,4′	639	2	762	84 ± 4	15
2,4,6,2′	78	2	84	90 ± 3	16
2,5,3′[c]	344	6	357	95 ± 4	17
2,4,3′	85	1	103	82 ± 6	18
2,4,2′6′	105	1	114	91 ± 4	19
2,5,4′	731	7	768	94 ± 4	20
3,4,2′	428	1	466	92 ± 3	21
2,4,4′	665	8	778	84 ± 4	22
2,5,2′5′	1019	42	1137	86 ± 7	24
2,4,2′5′	676	28	705	92 ± 4	25
2,3,2′5′	722	17	785	90 ± 4	26
2,4,2′4′	265	19	272	90 ± 5	27
2,3,2′4′	237	11	228	99 ± 2	28
2,3,2′3′[d]	390	0	486	80 ± 6	29
2,3,6,3′	512	20	547	90 ± 6	30
2,3,6,4′	728	20	788	90 ± 4	31
2,3,5,2′6′	679	0	777	87 ± 3	32
Octachlorostyrene	218	6	234	91 ± 3	33
2,3,4,2′6′	369	6	371	98 ± 3	34
2,3,6,2′3′	401	17	401	96 ± 4	35
2,5,3′4′	595	7	609	97 ± 4	36
2,4,5,4′	182	4	185	96 ± 5	37
2,4,3′4′	190	6	190	97 ± 4	38
3,4,2′3′	224	4	223	99 ± 4	39
2,3,6,2′3′6′	393	18	389	96 ± 4	40
2,4,5,2′5′	799	28	842	92 ± 3	41
2,4,5,2′4′	310	12	323	92 ± 3	42
2,4,5,2′3′	346	8	368	92 ± 2	43
2,3,4,2′5′	501	20	529	91 ± 3	44
2,3,4,2′4′	360	8	362	97 ± 3	46
2,3,4,2′3′	1153	19	1229	92 ± 3	47
2,3,6,3′4′	192	3	227	83 ± 14	48
2,3,5,6,2′5′	486	14	518	91 ± 3	49
2,3,5,6,2′3′	127	4	134	92 ± 8	50
2,3,6,2′4′5′	692	15	732	92 ± 5	51
2,3,4,2′3′6′[e]	297	6	439	66 ± 1	54
2,3,5,6,2′3′6′[f]	245	6	266	92 ± 7	55
2,4,5,3′4′	616	16	612	98 ± 6	56
2,3,5,2′4′5′	230	7	227	98 ± 4	58
2,3,4,3′4′	415	6	346	119 ± 13	59
2,4,5,2′4′5′	1880	86	1698	106 ± 1	60
2,3,4,2′3′5′	122	4	115	103 ± 4	61
2,3,5,6,3′4′	216	3	201	107 ± 7	62
2,3,4,2′4′5′	1217	41	1212	97 ± 4	64
2,3,5,6,2′3′5′	231	4	231	97 ± 6	65
2,3,4,2′3′4′	245	6	234	103 ± 5	66
2,3,5,6,2′4′5′	562	23	542	99 ± 3	67
2,3,4,6,2′4′5′	909	8	870	104 ± 4	68

Table 1. (cont'd)

PCB Congener	ng recovered	ng in unspiked fish	Mean ng in spiking solution	Recovery %	Lotus[a] Ref #
2,3,4,5,2'4'6'	261	7	261	97 ± 6	69
2,3,5,6,2'3'4'	302	8	308	95 ± 2	70
2,3,4,6,2'3'4'	203	10	217	89 ± 6	71
2,3,4,5,2'3'5'6'[g]	94	1	111	85 ± 11	72
2,3,4,5,3'4'	237	4	211	110 ± 2	74
2,3,4,5,2'3'5'	132	3	133	97 ± 4	75
2,3,4,5,2'4'5'	1286	31	1243	104 ± 6	76
2,3,4,5,2'3'4'	557	10	526	105 ± 6	77
2,3,4,6,2'3'5'6'	296	8	290	99 ± 3	78
2,3,4,5,6,3'4'	148	3	148	98 ± 2	79
2,3,4,5,2'3'4'6'	157	4	159	96 ± 1	80
2,3,4,5,6,2'4'5'	181	7	195	89 ± 5	81
2,3,4,5,6,2'3'4'	181	7	158	109 ± 8	82
2,3,4,5,2'3'4'5'	309	8	286	105 ± 5	83
TOT PCB	37917	3439	38680	89 ± 3	

[a] Serial number of congener as used in Lotus 1-2-3 Bar Charts
[b] Also includes a small percentage of 2,3
[c] Also includes a small percentage of 2,6,2'
[d] Also includes a small percentage of 2,3,4,2'
[e] Also contains 3,4,3'4'
[f] Also includes a small percentage of 2,3,5,6,2'4'6'
[g] Also includes a small percentage of 2,3,4,5,3'4'5'

study mandated a careful investigation of chemometric tools for converting the data into understandable information (Meglen 1988). Two main tools are available, those based on the General Linear Model and data organization using computer graphics. All data are transferred electronically from the gas chromatograph (Bush *et al.* 1983) either to a VAX 780 main-frame computer or using KERMIT to a personal computer. Two widely available programs were employed; the Biomedical Statistics Data Package (BMDP) (Dixon 1981) and Lotus 1,2,3 (Lotus Development Corp., Cambridge, MA). The BMDP software has been used extensively for discerning data trends and for quality assurance (Bush *et al.* 1983), also available in the program package are principal component and cluster analysis which have been used for investigating PCB data (Stalling *et al.* 1985) and step-wise regression which we have used for relating human fertility to PCB concentration (Bush *et al.* 1986). The alternative of visualization of the data (Cloake 1988; Ouchi 1988) has several advantages over the more mathematically oriented general linear model approach as is illustrated in Stalling *et al.* (1985) which uses principal component analysis and graphics presentation on a map. It is needed here for several purposes: ordering data with distance from a putative source; synthesizing PCB patterns from previously established Aroclor patterns; comparing PCB patterns between fish. One approach has not been relied on exclusively, in order to maintain the freedom to examine minor components of the data set which are known to the analyst *a priori* to be of probable importance.

Polychlorinated biphenyl levels in the river's biota are monitored by annual collections and analyses of multiplate biota samples and caddisfly larvae found in the riffle areas of the upper Hudson River (McDowell 1986). These macroinvertebrate samples have been analyzed by both high resolution capillary and packed column gas chromatography yielding yearly results on the transport of Hudson River PCB's through suspended sediments and bioaccumulation (Novak *et al.* 1988). The PCB residue that is characteristic of the biota samples analyzed in this monitoring effort contains a characteristic pair of gas chromatographic (g.c.) peaks 2,2'- and 2,6-dichlorobiphenyl, but not 4-chlorobiphenyl. This pair of congeners has also been found in other fish sampling studies (unpublished data). Previous studies of fish in the Hudson River have not utilized glass capillary procedures and hence congeneric information was not available (Sloan *et al.* 1983; Brown *et al.* 1985; Sloan and Armstrong 1988; Armstrong and Sloan 1988). Both 2,2' and 2,6-dichlorobiphenyl made up a large percentage of PCB pattern accumulated in caged midge larvae (*Chironomus tentans*) in the initial part of an uptake study, though the more chlorinated PCB congeners bioaccumulated to a greater extent over a more lengthy exposure period (Novak and Reilly 1986; Novak *et al.* 1988). The presence of this pair of PCBs in striped bass sampled in Long Island Sound and the Atlantic Coast should imply PCB exposure in the Hudson River. This may correspond with the tagging study done by Clark (1968), which indicated the Hudson River as a major spawning and nursery area for striped bass in the northeastern United States.

There were several objectives of this study. The most important was to delineate clearly the PCB congener composition of the residues in this species of fish, since it is a popular human food and is also under environmental pressure. Subsidiary objectives were to determine whether high resolution analysis could yield information on sources of pollution, and possibly migration patterns of the fish themselves.

Table 2. Striped bass sample parameters with comparison of analytical methods

Site	Season	Area[a]	Distance (Km) from Troy Dam	Length (mm)	Weight (g)	Sex	22′ 26[b]	Percent lipids	DEC tot PCB[c]	OH tot PCB[d]	Lotus ref number[e]
Lilco A	spring	NW	248	688	3292	F		7.00	4.65	2.22	19
Lilco B	spring	NW	248	682	3691	M	√	5.84	2.43	2.49	21
Little Neck Bay	spring	NW	248	729	3746	M	√	4.35	3.81	3.53	20
Eatons Neck A	summer	NW	241	660	2951	F		8.05	5.66	3.08	15
Eatons Neck B	summer	NW	241	635	2610	M		5.20	2.03	1.30	18
Eatons Neck C	summer	NW	241	713	3291	F		7.10	3.45	1.92	12
Eatons Neck D	fall	NW	241	641	2724	M		6.00	2.93	2.64	16
Eatons Neck E	fall	NW	241	616	2270	M	√	8.70	3.64	3.50	13
Eatons Neck F	fall	NW	241	661	2951	F		2.10	1.47	1.01	17
Eatons Neck G	fall	NW	241	647	2724	M		3.80	3.91	2.81	14
Plum Gut A	spring	NE	382	694	3289	M	√	7.40	2.68	3.52	54
Little Gull Is	spring	NE	382	1173	20412	F		6.09	1.25	0.88	55
Orient Pt. A	spring	NE	375	651	2948	M	√	6.21	1.61	2.06	52
Orient Pt. B	summer	NE	375	651	3007	F		4.17	1.16	1.10	50
Orient Pt. C	summer	NE	375	615	2383	F		0.91	1.07	0.24	49
Orient Pt. D	summer	NE	375	643	2043	F		1.22	0.69	0.25	51
Orient Pt. E	fall	NE	375	654	3064	F		2.80	1.05	1.47	47
Orient Pt. F	fall	NE	375	655	3518	F		8.40	2.30	1.91	53
Orient Pt. G	fall	NE	375	693	3745	M		2.10	2.52	2.30	48
Far Rockaway	spring	SW	275	637	2724	F	√	8.07	2.14	2.04	26
Breezy Pt. A	spring	SW	268	703	3742	M	√	2.75	6.68	9.21	25
Breezy Pt. C	summer	SW	268	619	2834	M		8.12	1.82	5.84	24
Breezy Pt. D	summer	SW	268	620	2497	M	√	7.86	4.89	4.53	22
Breezy Pt. E	summer	SW	268	814	4994	M	√	5.48	3.94	4.11	23
Rckwy Inlet A	fall	SW	275	987	10699	F	√	7.40	31.10	16.10	31
Rckwy Inlet B	fall	SW	275	1080	15890	F	√	7.50	3.46	3.71	29
Rckwy Inlet C	fall	SW	275	1098	15890	F	√	6.50	2.84	2.46	27
Rckwy Inlet D	fall	SW	275	662	3291	M		9.00	4.28	2.74	28
Rckwy Inlet E	fall	SW	275	622	2696	F	√	5.00	13.00	14.70	30
Fire Island B	spring	SE	342	824	6690	F	√	7.10	1.50	1.16	37
Fire Island C	spring	SE	342	1015	11000	M		6.69	9.17	4.36	40
Fire Island D	spring	SE	342	843	6577	M	√	4.60	5.65	4.60	36
Fire Island E	summer	SE	342	703	3742	M		3.89	8.12	4.54	38
Fire Island F	summer	SE	342	774	4794	F	√	2.15	1.63	5.76	41
Fire Island G	summer	SE	342	932	8456	F		1.58	0.83	1.79	39
Fire Is Inlet	summer	SE	328	675	3374	M		4.40	4.63	2.85	34
Jones Beach	fall	SE	302	1060	16117	F	√	17.80	4.29	3.15	32
Fire Island H	fall	SE	342	629	3064	M		7.70	3.88	2.29	35
Fire Is Inlet B	fall	SE	328	1154	17237	F		2.40	1.04	0.30	33
Whmptn Beach A	spring	SE	348	668	3062	M	√	1.91	2.93	1.77	42
Whmptn Beach B	spring	SE	348	678	3130	M	√	8.39	3.37	2.72	43
Amagannsett A	spring	SE	395	655	2948	M	√	3.17	2.71	2.13	58
Shinnecock	summer	SE	389	1240	25388	F		7.15	1.40	1.11	56
Amagannsett B	summer	SE	395	769	4653	F		10.06	10.56	6.69	57
Moriches Bay A	summer	SE	362	836	6583	F	√	10.40	13.45	10.90	44
Montauk A	fall	SE	422	889	6810	M		6.40	2.48	2.18	60
Montauk B	fall	SE	422	745	3972	F		20.00	5.44	0.34	59
Moriches Inlet A	fall	SE	362	615	2610	F		6.50	0.98	0.79	45
Moriches Inlet B	fall	SE	362	1198	21792	F		6.90	2.01	1.06	46
Gw Bridge	spring	HUD	214	697	3820	F		11.10	3.57	1.52	11
Tapzee	spring	HUD	190	651	2970	F	√	5.49	3.66	3.20	10
Alb/Troy A	fall	HUD	10	645	2460	M	√	4.41	23.41	24.10	4
Alb/Troy B	fall	HUD	10	573	2240	M	√	6.96	27.98	21.70	5
Croton Pt. A	FALL	HUD	168	745	4500	F	√	12.84	4.96	1.31	9
Croton Pt. B	fall	HUD	168	665	3360	F	√	16.83	17.49	18.80	8
Croton Bay A	fall	HUD	168	734	4640	F	√	8.30	2.87	2.24	6
Croton Bay B	fall	HUD	168	665	3360	F	√	8.40	19.50	32.10	7
Troy A	fall	HUD	0	593	2400	M	√	15.40	9.39	15.0	1
Troy B	fall	HUD	0	667	3000	M	√	10.40	37.17	30.3	2
Troy C	fall	HUD	0	710	—	F	√	7.20	19.90	20.8	3

[a]Areas: NW Western Long Island Sound; NE Eastern Long Island Sound; SW Western Atlantic Shore; SE Eastern Atlantic Shore; HUD Hudson River Estuary
[b] Hudson River indicating congeners present
[c] Environmental Conservation Department determination
[d] This determination
[e] Reference number on Lotus bar charts

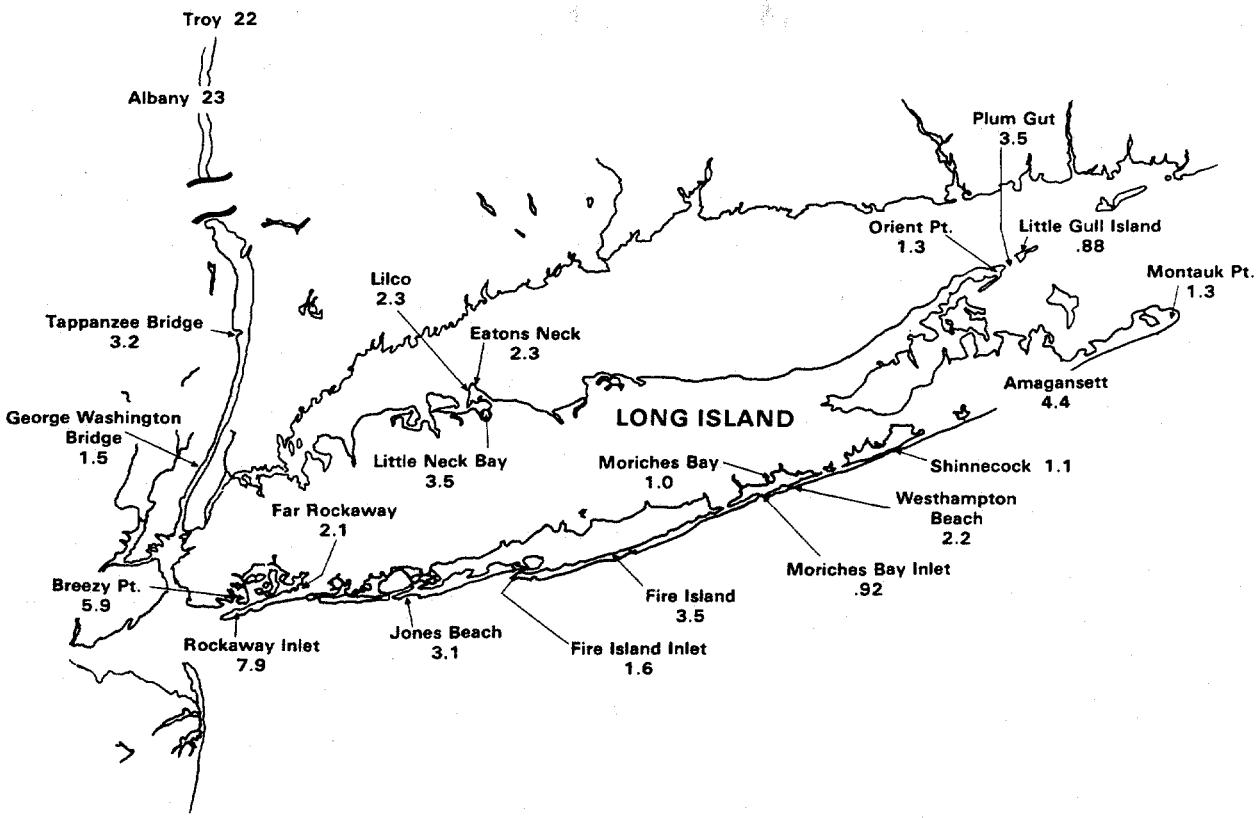

Fig. 1. Sampling points in the New York Bight with mean PCB concentrations of each point (mg/kg wet weight)

Materials and Methods

In 1984 and 1985, attention on PCBs in the Hudson River shifted to include the Marine District in New York State as a consequence of the U.S. Food and Drug Administration's change in the tolerance level from 5 ppm to 2 ppm for PCBs in fish destined for interstate commerce (Sloan and Horn 1985, 1986). This change led to a more extensive sampling effort in the Atlantic Ocean and Long Island Sound for striped bass, which also coincided with the annual Hudson River effort. The 60 fish samples used in the analyses reported herein were selected from the larger 1985 set (total of 717) collected by commercial fishermen by gill netting and by recreational anglers during the spring, summer and fall months at the time of spawning and coastal migrations and analyzed by packed-column gas chromatography (Sloan and Horn 1986; Sloan *et al.* 1986). Analyses of samples used in these earlier reports were conducted by Hazleton Laboratories America, Inc., Madison, Wisconsin using standard fillets (*i.e.*, one side of the fish with skin intact but scales removed) as established by sampling methods developed by the New York State Department of Environmental Conservation (Sloan *et al.* 1984). The individuals used to determine congeneric PCB distribution were chosen to reflect an array of total PCB concentrations, but they tended to be skewed toward higher total PCB levels.

Each of the 60 striped bass samples was reground to the consistency of a fine paste using a food processor to achieve sample homogeneity. A subsample of approximately one gram was weighed into a 20 mL screw top liquid scintillation vial. Two grams of sodium sulfate were added to remove water from the sample. The samples and sodium sulfate were then ground together with 10 mL of hexane in a Tekmar (Cincinnati, Ohio) tissuemizer using a ¼ inch blade for a period of one minute. The extract was pipetted into a 250 mL Kuderna-Danish evaporator. Two more extractions with 10 mL of hexane each were performed, using the tissuemizer and added to the Kuderna-Danish evaporation flask.

The extract was concentrated to approximately 2 mL on a steam bath using a 3-ball Snyder column and was quantitatively pipetted onto a 1-cm diameter glass chromatography column containing 10 g of 2% deactivated Florisil® with a two-g layer of sodium sulfate on top of the Florisil. The column was eluted with 55 mL of hexane and the first 40 mL of the eluate was collected in a 50 mL Erlenmeyer flask. The 40 mL fraction was concentrated to 1 mL, using a Kuderna-Danish evaporator and transferred via pipette to a 1 mL Wheaton GC vial for analysis. The samples were analyzed on a Hewlett-Packard 5840A gas chromatograph equipped with a Ni-63 electron capture detector using a 60 m glass capillary column coated with Apiezon L. The oven temperature was held at 60°C for 2 min then raised to 10°C/min for 6 min, then 1°C/min to 250°C. The carrier gas used was helium 25 psi, the makeup gas was 5% argon-methane (40 ml/min). The temperature of the injection port was 250°C, the electron capture detector was maintained at 250°C. The calibration mixture that was used to quantitate the PCB level in the samples was a 1:1:1:1 mixture of 1 μg/mL each of Aroclors 1221, 1016, 1254, 1260 with Mirex and *p,p'*-DDE at concentrations of 50 ng/mL, and hexachlorobenzene and octachlorostyrene at 26 ng/mL (Bush *et al.* 1985a). Total PCB was determined by summing the individual PCB congener concentrations. The analytical method yielded recoveries of >95% with a precision < ±5% relative standard deviation (RSD) for the 74 major congeners present in the samples (Table 1).

Data analysis was carried out with a VAX 780 computer using the Biomedical Statistics Data Package (Dixon 1981) and a Leading Edge Model D computer system (Canton, MA), using "Lotus 1-2-3" software (Lotus Development Corp., Cambridge, MA). Quality control for the project was ensured by running replicates of a sample in every batch of ten samples that were analyzed (twenty-

Table 3. Mean proportions of congeners comprising >0.6% in the PCB residues of 60 striped bass

Site[a]	% of Total PCB				
Congener	NW	NE	SW	SE	HUD
2,5,4′	.8	.6	1.5	1.1	1.9
2,4,4′	1.1	.6	1.4	1.0	1.4
2,5,2′5′	4.0	3.7	4.6	4.1	4.4
2,4,2′5′	3.5	3.2	4.6	3.9	4.7
2,3,2′5′	2.0	1.8	2.5	2.1	2.3
2,4,2′4′	2.4	2.3	3.7	2.8	4.0
2,3,2′4′	2.3	1.8	2.8	2.5	2.8
2,3,6,3′	2.0	1.8	2.8	2.6	3.1
2,3,6,4′	2.6	2.4	3.6	3.0	4.1
2,3,6,2′3′	2.3	2.3	2.2	2.6	2.4
2,5,3′4′	2.2	2.1	2.2	2.2	2.4
2,4,5,4′	1.6	1.4	1.6	1.5	1.6
2,4,3′4′	2.6	2.3	2.6	2.4	2.4
2,3,6,2′3′6′	2.6	2.8	2.2	2.3	2.4
2,4,5,2′5′	4.1	3.9	2.9	3.3	3.0
2,4,5,2′4′	3.7	3.5	2.8	2.9	2.8
2,4,5,2′3′	1.8	1.8	1.5	1.7	1.5
2,3,4,2′4′	1.8	2.3	2.3	2.8	1.9
2,3,4,2′3′	4.1	3.9	3.6	3.8	3.8
2,3,5,6,2′5′	1.6	1.6	1.2	1.4	1.3
2,3,6,2′4′5′	2.2	2.3	1.7	2.1	1.6
2,4,5,3′4′	4.1	3.6	3.1	3.1	2.5
2,3,4,2′4′5′	1.8	1.8	1.5	1.5	1.2
2,3,4,3′4′	0.8	0.8	1.0	1.1	0.8
2,4,5,2′4′5′	9.3	11.1	7.0	8.0	7.0
2,3,4,2′4′5′	5.7	6.0	4.1	4.9	4.5
2,3,4,2′3′4′	1.0	0.9	0.9	0.8	1.1
2,3,5,6,2′4′5′	2.4	2.4	1.6	2.5	1.7
2,3,4,6,2′4′5′	1.0	1.0	0.9	1.0	1.0
2,3,4,5,2′4′5′	3.4	3.5	3.0	2.9	3.1
2,3,4,5,2′3′4′	1.4	0.4	1.1	1.0	1.2
2,3,4,5,2′3′4′5′	0.9	1.5	0.7	1.2	0.6
		Total PCB mg/kg			
This sample	1.9	1.8	7.5	3.0	15
Large sample (N = 717)	2.5	1.9	3.0	2.4	6.2

[a] Areas: NW Western Long Island Sound; NE Eastern Long Island Sound; SW Western Atlantic Shore; SE Eastern Atlantic Shore: HUD Hudson River Estuary

four sample replicates were analyzed as part of the project) to ensure the consistency of sample extraction. Statistical analysis, of the replicates yielded RSD's of <5% for the replicate samples for the project. PCB substitution patterns are given throughout this work numbering each ring sequentially since in considering structure activity, this ring-by-ring numbering is most descriptive.

Results and Discussion

The mean length of the 60 striped bass was 755 cm with a standard error of the mean of ±22 cm (Table 2). They were collected by gill netting which is a size selective sampling process resulting in the collection of fish of a fairly uniform length and weight dependent upon gill net mesh size. Dew (1980), in his study of the biological characteristics of commercially caught striped bass, found that of the female striped bass that were netted over 60% of the females were sexually mature at a total length of 651–750 cm and 92% of the males were mature at this total length. The mean length for the male striped bass sampled in this project is 691 cm and the mean length for the females is 790 cm which in keeping with Dew's findings, would indicate that the fish represent a subgroup of the available striped bass population that is at or near sexual maturity.

The 60 striped bass contained a mean total PCB level of 5.5 μg/g ± 0.9 μg/g wet weight of the filets. Table 2 allows the independent analysis carried out by the more conventional packed g.c. column analysis to be compared with the present congener specific analysis. The packed column method gave a mean concentration of 6.31 mg/kg. The correlation coefficient is 0.91, the standard error of the estimate 3.3, the y intercept (where y = OH) is 0.97 and the regression coefficient (slope) is 0.95. There is one outlier which is obvious from the residuals plot given by the BMDP (Rockaway Inlet fish A, Lotus #31) which is also an unusually polluted fish with a peculiar PCB congener pattern (*vide infra*). Removal of this fish from the population increased the correlation coefficient marginally to 0.93 with corresponding improvements of the other model fit parameters.

Figure 1 shows a map of the New York Bight indicating the sampling locations for this study and the mean total PCB concentrations for the striped bass sampled in each area. Nine samples from the western part of Long Island Sound contained (mean ± standard error of the mean) 1.9 ± 0.2 μg/g, ten from eastern Long Island Sound 1.8 ± 0.4 μg/g, ten from the west Atlantic shore area 7.5 ± 1.9 μg/g, twenty from the eastern Atlantic shore area 3 ± 0.5 μg/g, and eleven taken in the Hudson River estuary 15 ± 3 μg/g (Table 3). Stratifying samples to discriminate differences in the PCB concentrations between different sampling seasons or between sexes was not the intent of this project.

Figure 2 shows the total PCB (ng/g) found in each of the fish plotted in order of increasing distance from the Troy Dam, assuming free passage from Long Island Sound to the Hudson River. The Troy Dam is near the upstream limit of the spawning migration. The figure shows a general trend in reduction of the amount of PCB with increased distance from the upper Hudson.

Initial examination of the data in Table 3 shows that the fish from the Long Island Sound area exhibited differences as a group from the fish sampled in the Hudson and those collected in the Atlantic Shore area of Long Island in total PCB contamination. Congener by congener composition differences are not, however, very pronounced. The total PCB levels are consistent with the results from the larger data set from which these samples were drawn. The average total PCB concentration (± standard error) for the Hudson River at all locations and collection periods was 6.19 ppm ± 0.36 (N = 408) (Sloan and Horn 1986). Similarly, the average total PCB concentrations were for western Long Island Sound (NW) 2.51 ppm ± 0.13 (N = 90); eastern Long Island Sound (NE) 1.91 ppm ± 0.10 (N = 90); western south shore of Long Island (SW) 3.04 ppm ± 0.20 (N = 89); and eastern south shore of Long Island (SE) 2.45 ppm ± 0.21 ppm (N = 85). The New York Harbor area with an average total PCB concentration of 3.54 ppm ± 0.36 (N = 90) represents a transition zone between the Hudson River samples and those from the western end of Long Island, but samples from the Harbor area were not included in the congeneric

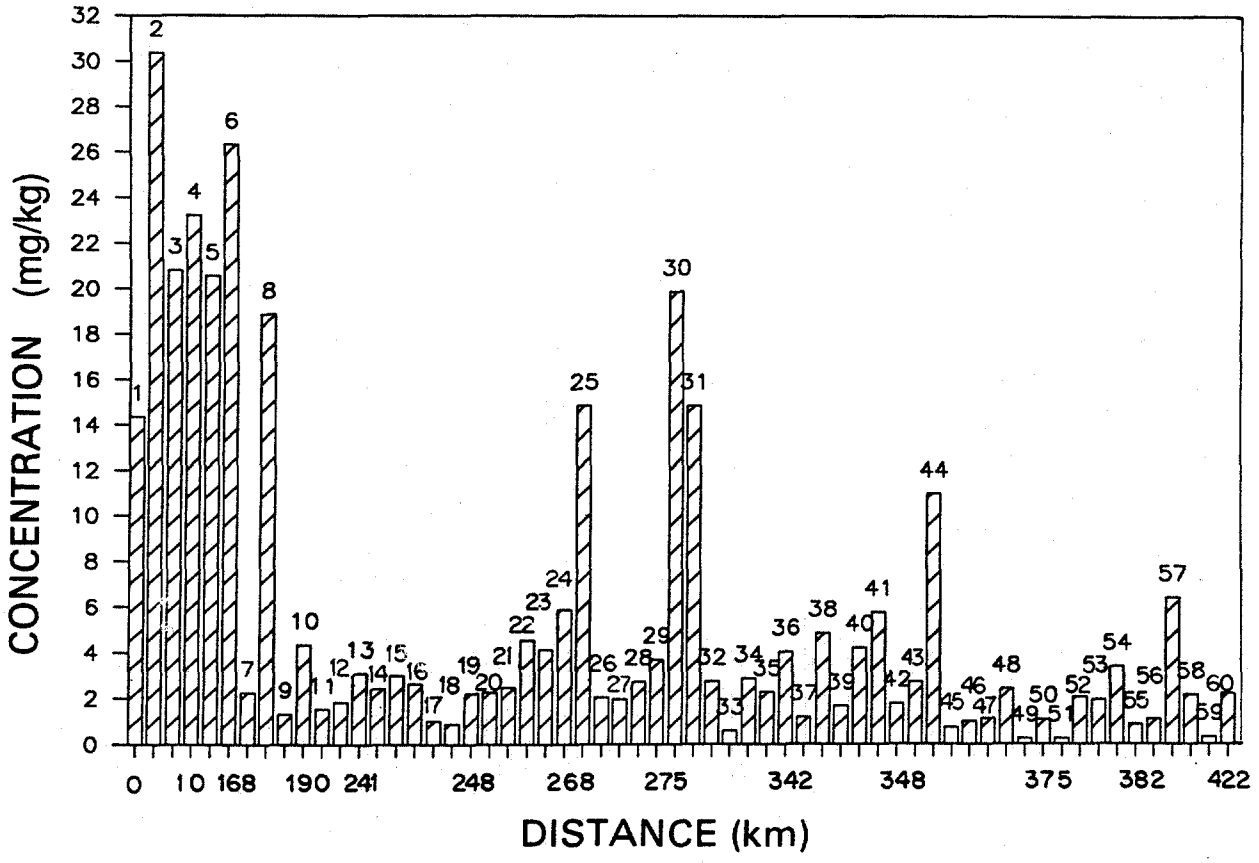

Fig. 2. Total PCB concentration in order of increasing distance from the Troy Dam. Fish numbers from Table 2 are shown on each bar

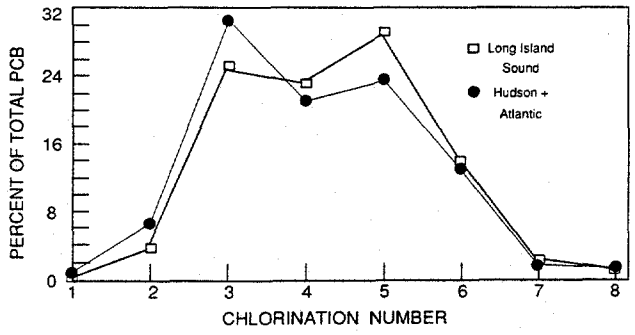

Fig. 3. Diagrammatic representation of the normalized concentration of mono through octachlorobiphenyls in fish from Long Island Sound (□) and the Hudson River and Atlantic shore of Long Island combined (+)

analyses reported herein. The averages of the data from the limited survey are fairly close to the averages for the whole survey with the exception of the Hudson River (Table 3). In this case, a whole set of samples for N.Y. Harbor were not included in the smaller samples.

Figure 3 is a diagram illustrating the pattern differences on a chlorination pattern basis. It shows the percent of the total PCB represented by each chlorination group plotted against the chlorination group for the two sets of striped bass samples (Hudson River and Atlantic Shore area of Long Island vs. Long Island Sound). This form of data presentation makes it clear that the striped bass collected in the Hudson River and the Atlantic Shore area of Long Island have more trichlorobiphenyls as a percentage of their total residue than do those from Long Island Sound. The figure also shows the Hudson River and Atlantic shore fish to be poorer in pentachlorobiphenyls than the Long Island Sound fish. The percent of total (normalized) concentrations were compared using t tests with separate variance (BMDP Program P7D). Discernable differences between the two groups (Figure 3) were found for total dichlorobiphenyl ($p < .0002$), total trichlorobiphenyl ($p < .0017$), and total pentachlorobiphenyl ($p < .0001$). The differences in the chlorination pattern of the two groups might indicate that fish from the Long Island Sound area were, for the most part, not exposed to the Hudson River Estuary, alternatively they could have been absent from the estuary for an extended time so that the less chlorinated congeners had a chance to disappear. This would agree with the tagging study done by Clark (1968) that showed that not all of the fish in this area enter the Hudson River but migrate to other areas.

For more detailed comparison of PCB residue patterns, the normalized data are plotted as bar charts. Figure 4 is a bar-chart representation of the chromatogram of a 1:1:1:1 mixture of Aroclors 1221, 1016, 1242, 1254 and 1260. There are some congeners which are unique to particular Aroclor products, these are listed in Table 4, which also shows minor contributions from other Aroclor products. Until now it has been impossible to distinguish pollution by similar Aroclor

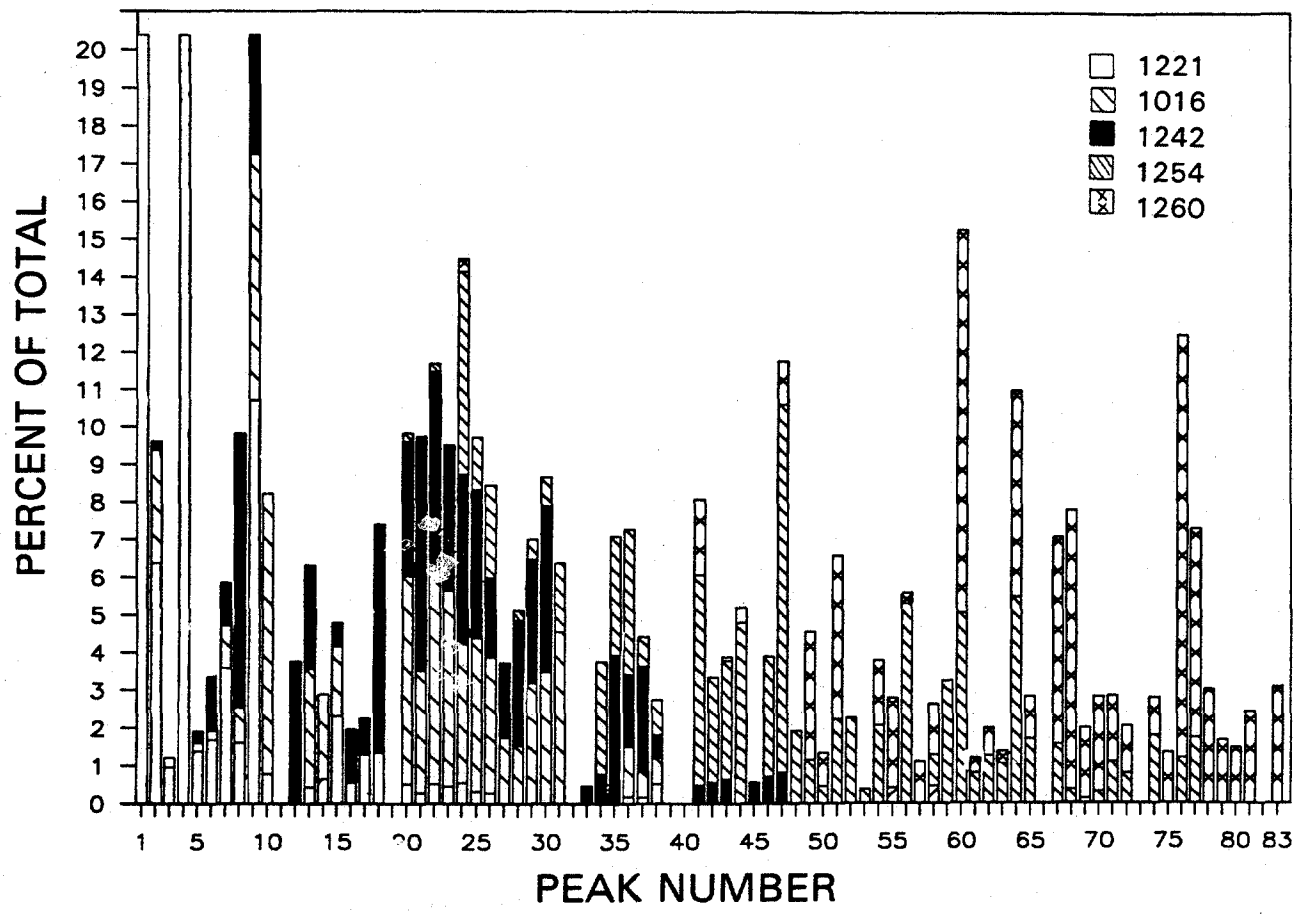

Fig. 4. A synthesis of the normalized chromatograms of Aroclors 1221, 1016, 1242, 1254 and 1260

Table 4. PCB congeners which are unique to Aroclor products and so may be used to indicate their presence in an environmental residue

Aroclor	Congener	% of Total	Interference from other Aroclors (%)	
1221	2	40	1242	3
	4	25	1242	1
1242	2,6,2′	7	1221	2
			1016	1
	2,4,2′	4	none	
1016	2,5,2′	7	1221	1
	2,3,6,4′	5	1254	2
1254	2,4,5,3′4′	5	1260	<1
	2,3,6,3′4′	2	none	
1260	2,3,4,5,2′4′5′	11	1254	1
	2,3,4,6,2′3′5′6′	3	none	
	2,3,4,5,2′3′4′5′	3	none	

products such as Aroclors 1016 and 1242 because the major congeners in each were indistinguishable; the high resolution of the present capillaries now differentiates these mixtures. Relative quantities of the various Aroclor mixtures in an environmental residue (assuming no physicochemical or metabolic modification) may be determined from the proportion of these indicator congeners, a mixture may then be synthesized using Lotus 1-2-3 to match the environmental residue. Figure 5A shows a computer-simulated bar chart of the chromatogram of a fish from the Troy Dam (Troy A) and the bar chart of the fish's actual chromatogram (Figure 5B). The simulation is not entirely accurate, especially in the trichlorobiphenyl region but it fits the fish residue sufficiently well to indicate that the approximate make-up of the fish residue: Aroclor 1016 (20%), Aroclor 1242 (35%), Aroclor 1254 (24%), Aroclor 1260 (13%). Some of the congener concentrations in the computer simulated bar chart are greater than in the fish, and some are smaller. Further investigation of these differences may delineate factors which have modified the residue while in the environment such as selective amplification of congeners by physicochemical interaction between water and sediments, liver metabolism or by partitioning via the gills.

Figure 6 shows the chromatograms of two fish, one of which came from the Hudson River and the other from the Atlantic Ocean near Orient Point on eastern Long Island. The Figure clearly shows the 2,2′ and 2,6-dichlorobiphenyl present in the fish from the Hudson River but not present in the Orient Point sample. 2,2′- and 2,6-dichlorobiphenyl are very minor components of Aroclors 1242 and 1016, but we have demonstrated recently (Wood *et al.* 1987) that these congeners are selectively dissolved from contaminated sediment into the water column. Along with 2-monochlorobiphenyl they represent 50% of the PCB transported from the

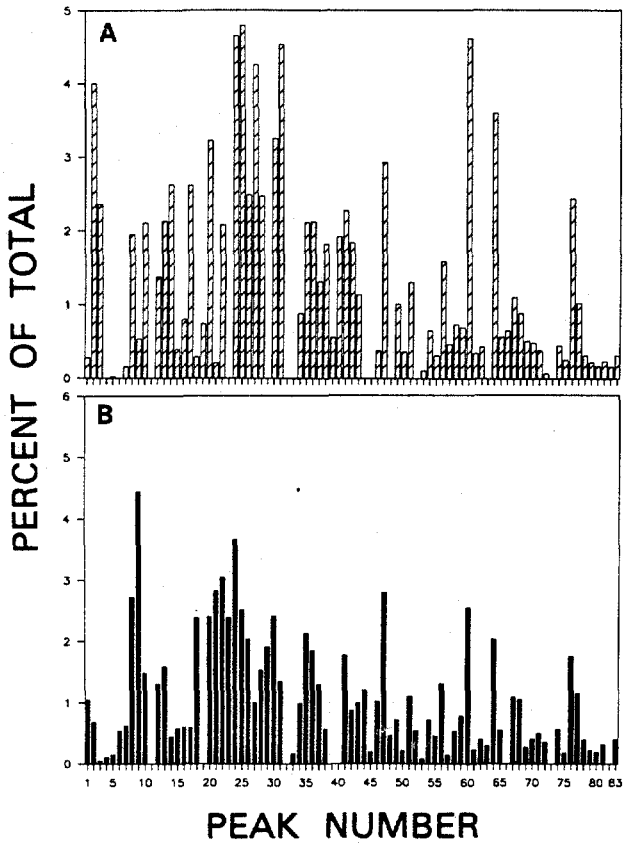

Fig. 5. Bar chart representation of the normalized concentrations of PCB in a fish from the Hudson River at Troy (A) and a simulation of the chromatogram made up of Aroclor chromatograms (B) (see text)

pollution site at Fort Edward, N.Y. during times of low flow, approximately 300 kg/annum (Bush *et al.* 1985b). A high degree of environmental mobility has been observed in uptake studies with caged midge larveae (*Chironomus tentans*) where the congeners 2,2′ and 2,6 accounted for a large proportion of the total accumulated residue early in the study (Novak and Reilly 1986) though the more chlorinated PCB congeners were shown to bioaccumulate to a greater extent later in the exposure. A similar phenomenon was observed at the same sampling sites in an *in situ* study using caged fathead minnows (*Pimephales promelas*) in 1985 (P. Jones, personal communication). Hence although these two congeners may indicate exposure to the Hudson River environment, the indication may disappear relatively rapidly after termination of exposure.

All of the fish sampled in the Hudson River had these congeners in their residue as did five fish from Long Island Sound, and 16 fish from the Atlantic shore area. Their presence in only some of the fish sampled in Long Island Sound and the Atlantic Shore area may indicate the intermingling of two or more populations of striped bass present in these areas at the time of sampling, which is consistent with the findings of Fabrizio *et al.* (1985). Tagging studies by Clark (1968) of striped bass from Long Island Sound and the New York Bight area showed that some of the fish that occupy these areas at some point during the year, winter and spawn in rivers other than the Hudson, presumably to the south near the Chesapeake Bay area or other spawning grounds in the Southern United States. Clark (1968) identified a group of fish that, based on recaptures of fish tagged in the Long Island Sound area, were part of a group of striped bass that wintered in Long Island Sound and moved to southern river areas to spawn. Some of the fish tagged in the Long Island Sound area were recaptured in coastal areas ranging from Chesapeake Bay to Rhode Island. The presence of these fish would explain the lack of the 2,2′ and 2,6-dichlorobiphenyl signature pattern delineating fish exposed to Hudson River PCB contamination in some of the fish sampled in this study, particularly those from Long Island Sound and the eastern part of the Atlantic Shore of Long Island.

To discover whether the Aroclor products all followed the same geographical trend, which would indicate one major source of pollution, the indicating congeners (Table 4) were plotted vs. distances. Also included in the plots are results for fish from above the Troy Dam (brown bullhead, *Ictalurus nebulosus*). The residue in this species would not be expected to be identical to that of striped bass because of their different diet and habitat, but it would indicate whether there was a continuum of pollution right up to the putative polluter's geographic location. Figure 7 shows plots for 2,2′-dichlorobiphenyl, 2,6,2′- and 2,5,2′-trichlorobiphenyl, 2,4,5,3′,4′-pentachlorobiphenyl and 2,3,4,5,2′,4′,5′-heptachlorobiphenyl. The Aroclor 1016 indicator, 2,5,2′-trichlorobiphenyl, appears generally higher than the Aroclor 1242 indicator 2,6,2′-trichlorobiphenyl, which is consistent with the more recent use of the former in the capacitor industry. It is clear that the pollution is continuous for each Aroclor mixture from the upper Hudson River to Montauk Point.

The plot of total PCB concentration with distance from the Troy Dam (Figure 2) shows that some fish do not fit the general downward trend. These were two fish from Breezy Point jetty, two fish from Rockaway Inlet and one from Moriches Bay. When the bar-chart presentation is used to examine the residues from these locations (Figure 8), it is clear that the patterns in these fish are similar to each other but different from the Hudson River pattern, although all contain the 2,2′- and 2,6′-chlorobiphenyls. The major part of those residues may be derived from another source of PCB pollution which deserves further investigation. Pizza and O'Connor (1983) fed striped bass ^{14}C-labelled Aroclor 1254 and showed some reduction in labelled PCB body burden with time, although different congeners could not be distinguished. Figure 7 suggests that the lower-chlorinated indicating congeners depurate rapidly from the fish whereas the higher-chlorinated congeners do not. This agrees with preliminary data produced with fathead minnows in the laboratory (unpublished data).

One purpose of this study was to produce detailed analyses by congeneric isomer. This allows the mammalian toxicity of the PCB residue to be better assessed. It is clear from Table 3 that the residue pattern is fairly uniform over the whole geographical region, although there are statistically discernable differences such as those shown in Figure 3. The fish residue is composed of tri-, tetra-, and pentachlorobiphenyls (29%, 22% and 25% of each) with 13% of hexachlorobiphenyls (the standard error of these mean concentrations is <1%). This is an unusual pattern of PCB pol-

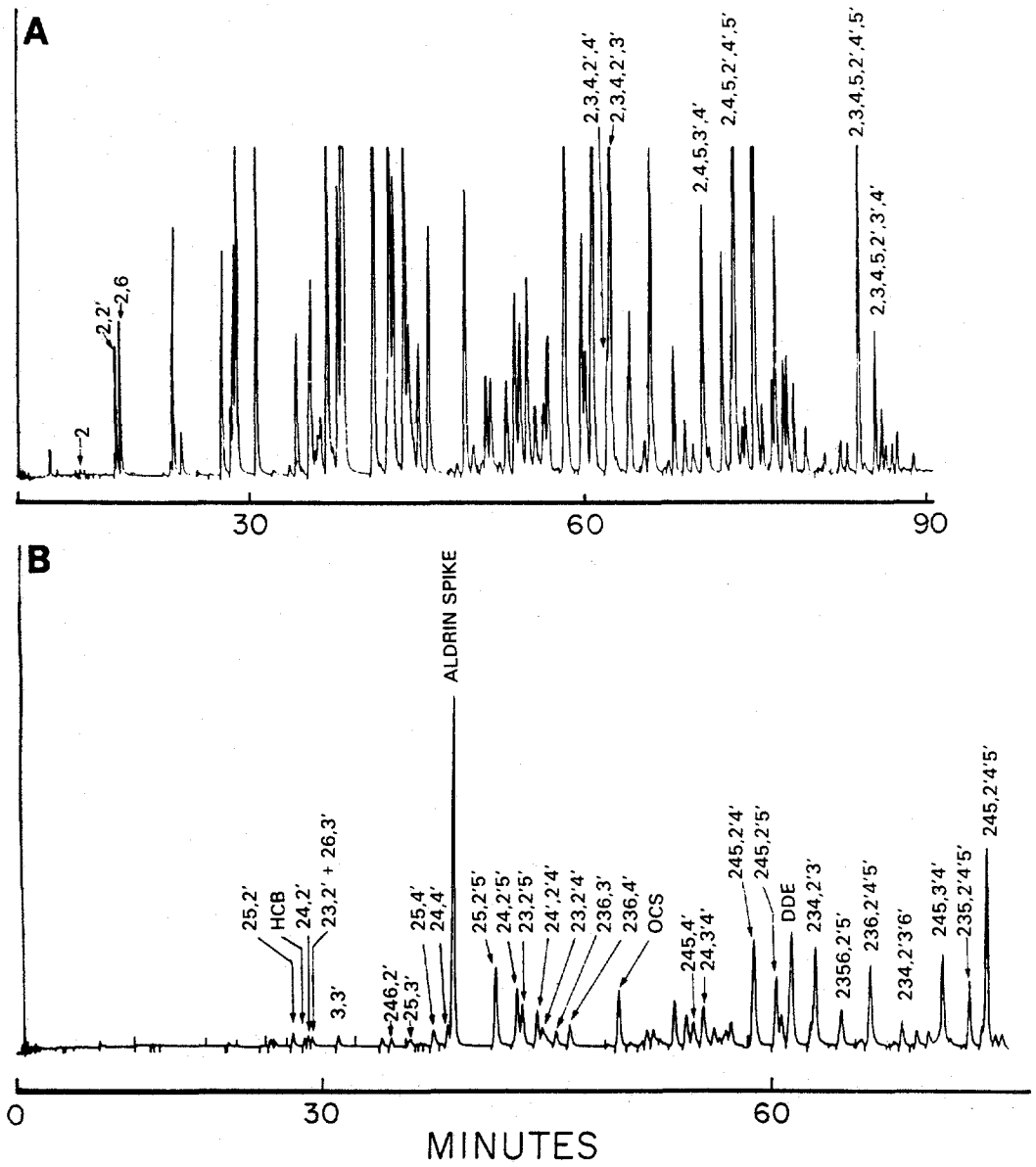

Fig. 6. Gas chromatograms of a fish from the Hudson River at Troy (A) and a fish from Montauk Point (B)

lution. In Lake Ontario for example there are few PCB congeners less retentive on gas chromatography than 2,5,2'5'-tetrachlorobiphenyl, because the major pollutants are derived from Aroclor 1254 and 1260 (Spagnoli and Skinner 1977). In foodstuffs derived from mammals (Shain *et al.* 1985) and birds (Bush *et al.* 1974) there is little quantifiable residue with g.c. retention less than 2,4,5,3'4'-pentachlorobiphenyl. Warm blooded animals metabolize PCB congeners which do not have chlorine substitution at both 4 positions in the molecule (Shain *et al.* 1985) and there is a large currently unexplained increase in accumulation for all such congeners with g.c. retentions longer than this pentachlorobiphenyl. This is also observed in humans (Bush *et al.* 1985c). Neurotoxicity as indicated by alteration of brain neurotransmitter concentrations, following acute and chronic exposure to PCB mixtures, is attributable to these persistent congeners which are not rapidly eliminated by warm blooded animals (Seegal *et al.* 1985).

Effects on liver function are probably the most important toxicological property of PCB, because induction of the mixed function oxidases may potentiate the mutagenic effects of PCB themselves and other possible carcinogens which may be present in the organism (Safe and Hutzinger 1987; Turner and Collins 1984). Congeners which lack chlorine substitution at the 2- and 6-positions of the PCB molecule, such as 3,4,3'4'-tetrachlorobiphenyl or 3,4,5,3'4'-pentachlorobiphenyl, are the most potent liver toxins (Kimbrough 1981; Safe *et al.* 1981; Kannan *et al.* 1988). This is

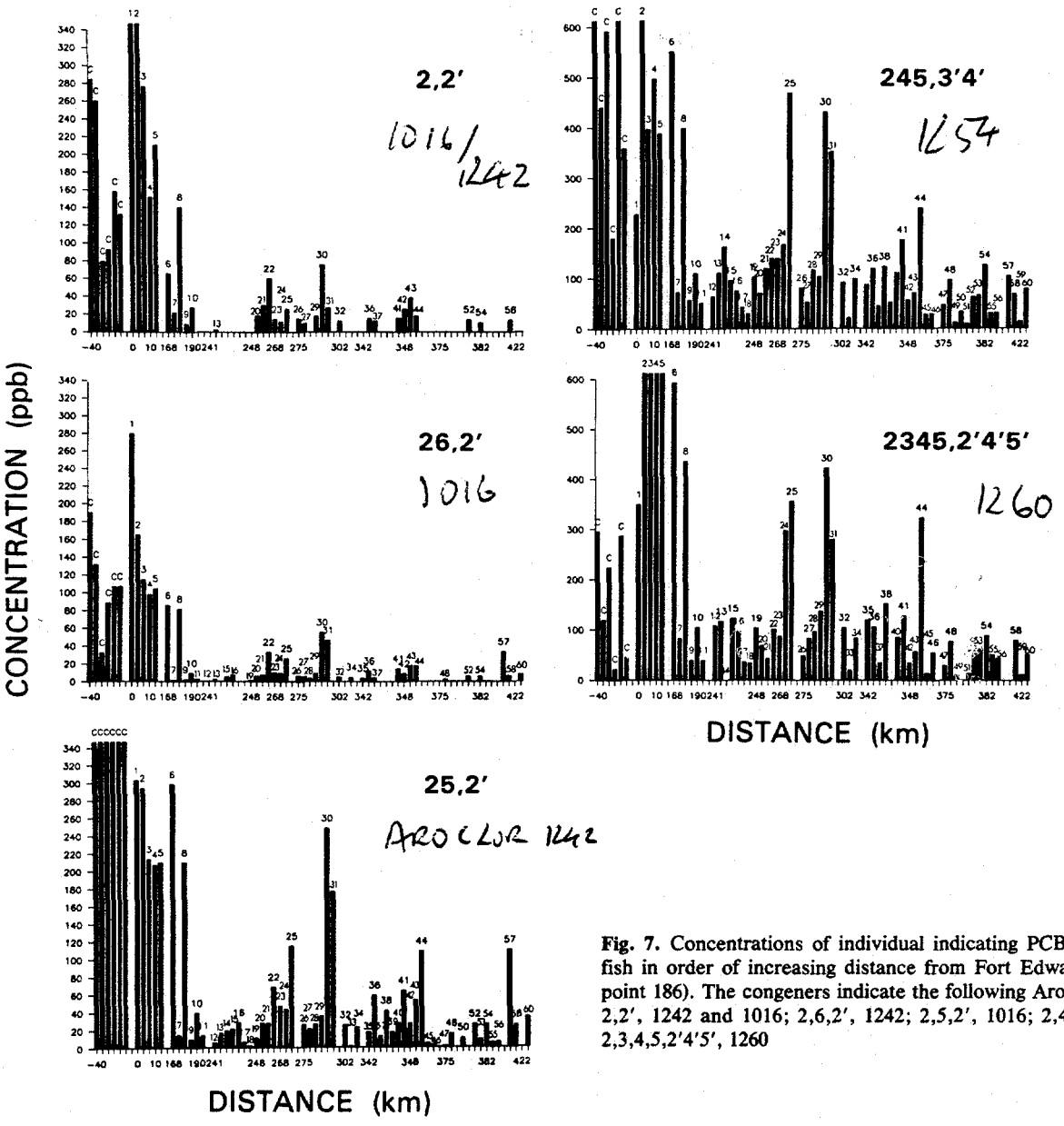

Fig. 7. Concentrations of individual indicating PCB congeners in fish in order of increasing distance from Fort Edward (river mile point 186). The congeners indicate the following Aroclor mixtures: 2,2', 1242 and 1016; 2,6,2', 1242; 2,5,2', 1016; 2,4,5,3'4', 1254; 2,3,4,5,2'4'5', 1260

also true for immunotoxicity (Silkworth and Grabstein 1982). These two congeners comprise less than 0.05% of Aroclor 1254 by a recent analysis of the product in this laboratory (unpublished result) and were not detected in the striped bass, moreover all the congeners in Table 3 have at least one 2-chloro substitution. However, congeners with no 2-substitution on one ring comprise approximately 15% of the residues in Table 3 and these have recently been shown to have considerable liver toxicity (Kannan *et al.* 1988). They also cause loss of body weight and thymus weight in young rats (Parkinson and Safe 1987). Persistent compounds with 4-substitution on both rings comprise the following proportion of the residues shown in Table 3: NW, 41% (0.77 mg/kg); NE, 41% (0.74 mg/kg); SW, 36% (2.7 mg/kg); SE, 37% (1.1 mg/kg) HUD, 36% (5.3 mg/kg). Thus, less than half of the typical residue is made up of the more toxicologically suspect PCB congeners and the risk of consuming Long Island Sound striped bass may be less than was apparent from previous low-resolution analysis.

Several major points can be deduced from this congener-specific analysis of Hudson River striped bass. The analytical method yields total PCB values which correlate well with conventional packed column analysis. The New York Bight is contaminated by Aroclors 1016, 1242, 1254 and 1260 but probably not Aroclor 1221 as indicated by the absence of 4-chlorobiphenyl. 2,2'- and 2,6-Dichloro-biphenyls may be used to indicate recent exposure to the Hudson River pollution. The typical PCB residue contains a smaller proportion

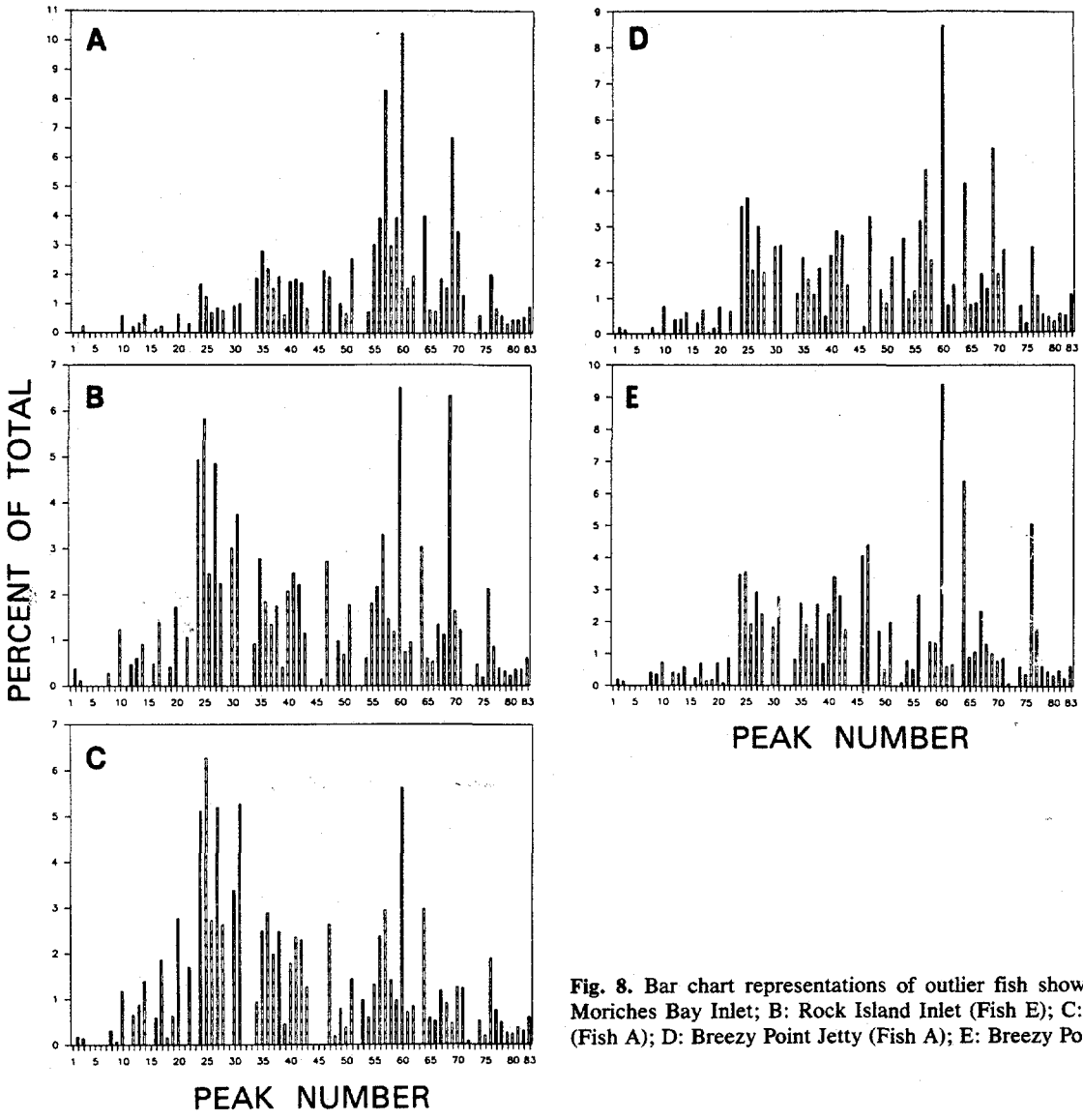

Fig. 8. Bar chart representations of outlier fish shown in Figure 2. A: Moriches Bay Inlet; B: Rock Island Inlet (Fish E); C: Rock Island Inlet (Fish A); D: Breezy Point Jetty (Fish A); E: Breezy Point Jetty (Fish C)

of metabolically persistent PCB congeners than is found in common human foodstuffs which are derived from warm blooded animals.

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