POLYCHLORINATED BIPHENYL (PCB's) IN LONG ISLAND SOUND

A Review

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ABSTRACT

This report contains a review of currently available information on the distribution patterns and behavior of PCB's in sediments, water and biota in Long Island Sound, paying particular attention to the striped bass (*Morone saxatilis*). PCB's in sediments are widespread in harbors and rivers along the north shore with concentrations up to 810 ppb, which is towards the lower range of polluted aquatic areas. Concentrations offshore range up to 480 ppb which is comparable to similar offshore locations elsewhere. The evidence suggests that these concentrations have originated from urban areas in such waters as sewage and urban run-off and are transported offshore in dredging spoil. Application of bioconcentration theory together with calculated water concentrations for the Sound indicates that PCB concentrations in the striped bass increase with age due to the slow rate of bioconcentration of the higher molecular weight PCB's. The PCB's in Long Island Sound fish arise from two sources. Firstly bioconcentration of PCB's in the Sound itself and secondly the movement of PCB contaminated fish from the Hudson River. The decline of PCB's in the striped bass is not amenable to treatment by first order kinetics but current trends suggest the decline will occur at a comparatively slow rate. PCB's also occur in significant concentrations in a range of other Long Island Sound biota with the highest concentrations in Atlantic menhaden (up to 3.45 ppm), and the bluefish (up to 1.89 ppm).

1. INTRODUCTION

Long Island Sound is one of the most heavily urbanized water bodies in the United States. It extends in an approximately northeasterly direction from New York City for a distance of about 150 km and is about 20 km wide at its widest point. The coastline of Connecticut forms its northern shore while the north Long Island coastline forms it's southern shore (see Fig. 1).

The immediate environs of the Sound contains numerous urban centers including the New York City boroughs of the Bronx and Queens, the New York counties of Nassau, Suffolk and Westchester as well as the Connecticut counties of Fairfield, Middlesex, New Haven and New London. The populations in these centers use the Sound for swimming, recreational boating, sport fishing, commercial fishing, commercial transport, sand and gravel mining, shellfish aquaculture, disposal of sewage, dredging wastes and other wastes (Koppelman et al, 1976).

Several rivers discharge into the Sound possibly containing detrimental concentrations of nutrients, pesticides and other potentially deleterious chemicals. Also substantial discharges of PCB's have been made to the geographically adjacent Hudson River by factories located upstream at Fort Edward and Hudson Falls, resulting in contamination of sediment and water downstream for about 306 km to New York Harbor (Brown et al, 1985). Biota in the river and estuary have accumulated relatively high concentrations of these substances such that the river was closed to all fishing for about 70 km downstream from the PCB sources to Troy in 1976. In addition, the commercial fisheries based on the striped bass (*Morone saxatilis*) were closed throughout the remaining length downstream. The New York State Department of

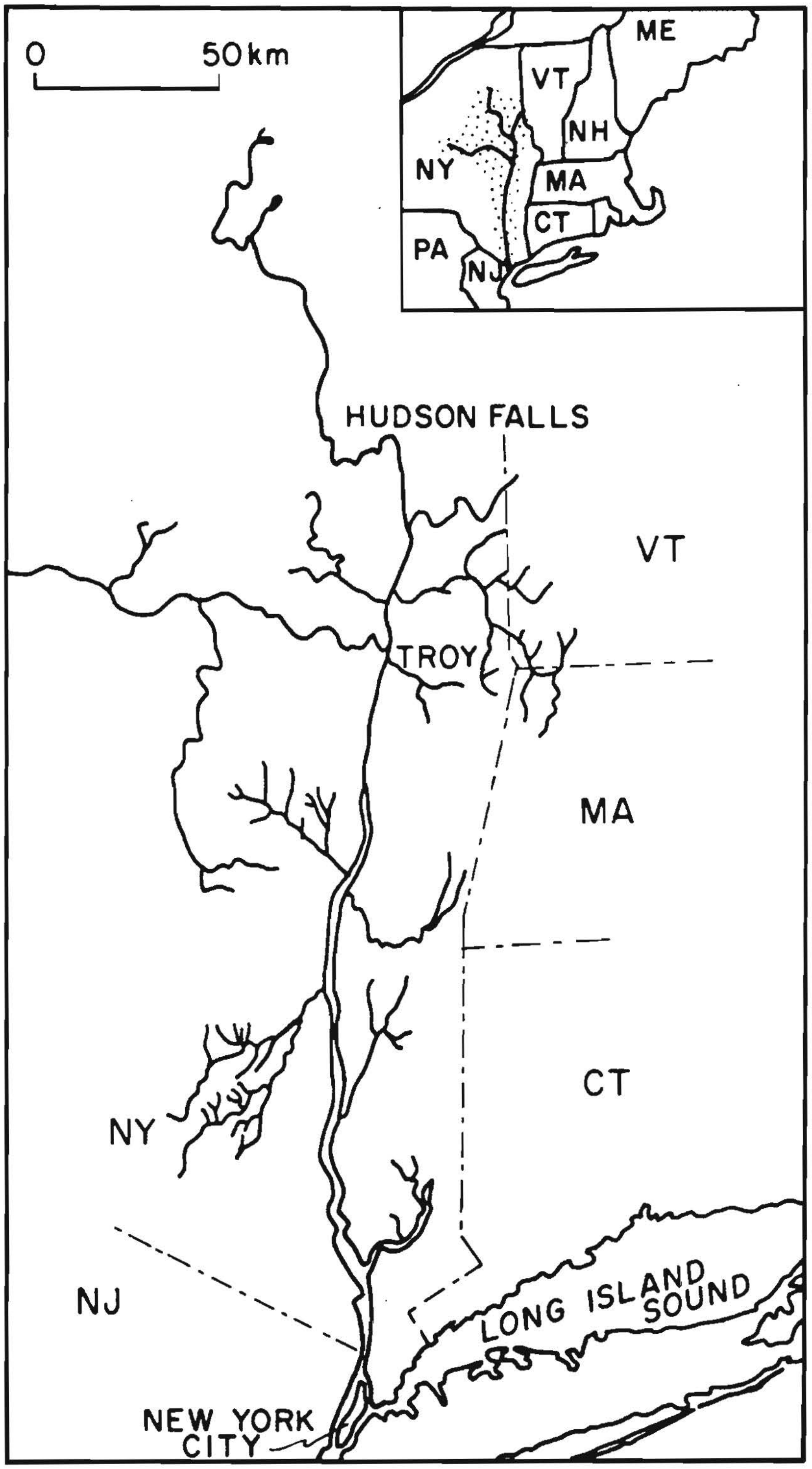

Fig. 1 - The Hudson River and Long Island Sound

Environmental Conservation has extensively investigated this problem as described by Brown et al (1985) and in publications referred to by them.

Long Island Sound is connected to the Hudson River at the Upper New York Bay through the East River (see Fig. 1). Some exchange of contaminated waters from the East River, presumably at least partially derived from Upper New York Bay, with the western part of the Sound has been documented by Jay and Bowman (1975). Also the occurence of contaminated fish in Long Island Sound may be due to the movement of fish between the Hudson River and the Sound (Anon, 1978).

In 1985 the commercial striped bass fishery was closed in western Long Island Sound and New York Harbor due to the occurrence of PCB concentrations greater than the FDA limit of 2.0 ppm in bass of legal size, ie. greater than 24 in. total length (Brown et al, 1985). The origin of the PCB's in the Long Island Sound fish is of concern for management purposes. Also the patterns of distribution of PCB's in the biotic and abiotic components of the Sound are important in developing an overall prospective regarding the significance of these substances as environmental contaminants.

In accord with this general background information the objectives of this current review were to:

1) investigate the distribution patterns of PCB's in the Sound.

2) interpret these distribution patterns in terms of bioaccumulation theory paying particular attention to the striped bass.

3) evaluate possible sources of PCB's in the Sound system.

TABLE 1. Estimates of Sediment Sources to Long Island Sound

	Riverborne sediment (thousands of tons/yr)	Sewage effluents (thousands of tons/yr)	Barged waste disposal (thousands of tons/yr) 1960-63	1964-68
East River	–	15	–	–
Western	30	5	400	900
Central		4	60	50
Housatonic River	16			
Eastern		1	100	300
Connecticut River	90			
Thames River	10			
TOTAL	146	25	560	1250

From: Koppelman et al, 1976.

2. SOME IMPORTANT PHYSICAL CHARACTERISTICS OF LONG ISLAND SOUND

2.1 Catchment of the Sound

The catchment of the Sound occupies an area of about 39,000 km^2 which contains three major river drainage basins. The largest of these, the Connecticut, has an area of 29,100 km^2 and extends from the Canadian border to the northern shore of the Sound. The other two rivers also discharge to the northern shore and have a total catchment area of 7,860 km^2. The southern shore catchment accounts for comparatively minor stream discharges.

2.2 Sediment Characteristics and Origin

These are no large sources of sediment discharging to the Sound since the river discharges are comparatively low in suspended sediment (Koppelman et al, 1976). However, barged solid waste, containing dredging spoil from various harbors, has been deposited at many sites over a considerable period (see Fig. 2). These quantities of barged sediments are considerably in excess of those derived from the river catchments (see Table 1).

Fine grained silts, which are rich in organic matter as indicated by the development of hypoxic conditions on occasions, cover about one third of the bottom in the deepest parts of the Sound. This area is reported to have a low density of food organisms (Koppelman et al, 1976) but organic enrichment suggests a high population of a limited number of invertebrate species (Connell and Miller, 1984). In shallower areas the bottom consists of fine and course sands. The western area of the Sound contains most of the silt deposits whereas the sands tend to become coarser towards the open sea, where more turbulent conditions exist.

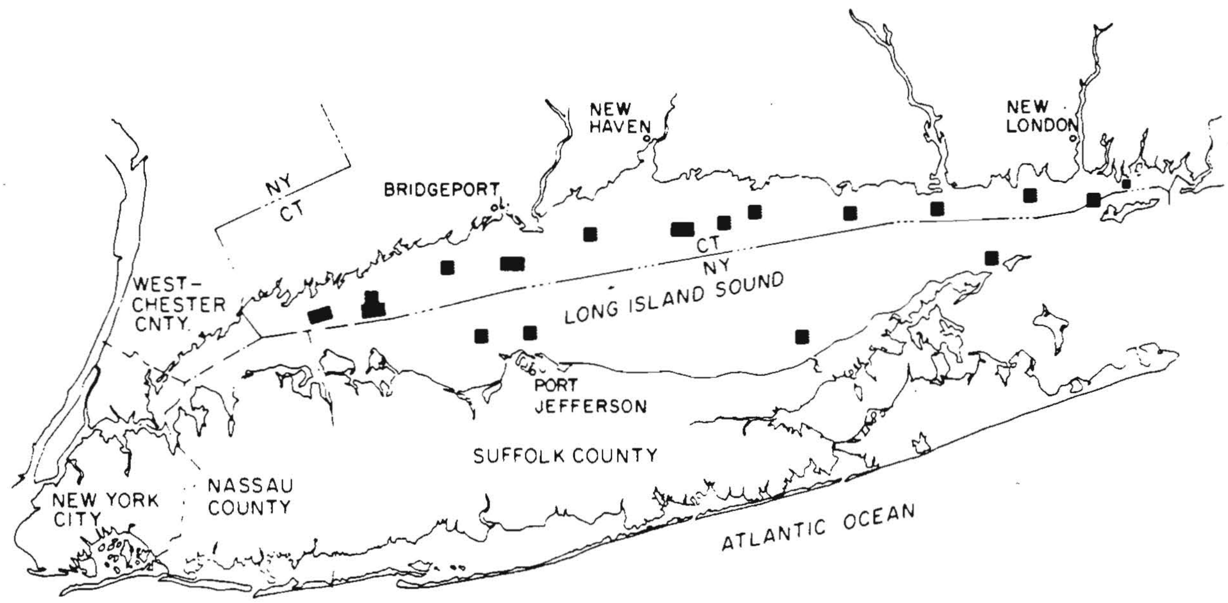

Fig. 2 - Waste Disposal Sites in Long Island Sound (From: Koppelman, 1976)

2.3 Water Quality and Movement

Salinity varies with seasonal conditions but on average decreases from about 34.5 ppt in the east to 20 ppt near New York City. Local reductions in surface salinity also occur around the major river mouths on the north shore. During the summer stratification of substantial parts of the Sound occurs resulting in dissolved oxygen depletion in the bottom waters (Hardy and Weyl, 1971). In addition blooms of phytoplankton occur periodically due to high nutrient concentrations (Koppelman et al, 1976). Jay and Bowman (1975) have calculated that estuarine and dispersal mechanisms in the East River, which receives a substantial proportion of New York City's sewage discharges, transport a considerable fraction of this sewage to the western Sound. Presumably toxic components in sewage, such as PCB's, will move in general accord with this pattern.

3. PROCESSES AFFECTING THE DISTRIBUTION OF LIPOPHILIC CHEMICALS

3.1 Overall Environmental Distribution of Lipophilic Chemicals

Lipophilic chemicals are a group of compounds which exhibit a high level of fat, or lipid, solubility. Converse this lipophilicity of a chemical confers on it, at the same time, a low level of water solubility, termed hydrophobicity. So lipophilicity and hydrophobicity are alternative descriptions which describe the same basic property of a molecule, ie. low polarity. A wide range of important environmental pollutants, such as the PCB's, DDT and so on, fall into this group.

The distribution of lipophilic chemicals in the environment is a highly complex process controlled by the physicochemical properties of the chemical and the environment itself. As a first step in understanding these processes the environment can be simplified by considering it to be divided into separate phases or compartments within which a chemical behaves in a uniform manner. The phases most commonly used are air, soil, water, sediments, and biota (see Fig. 3). It is possible to subdivide some of these phases further, for example sediments into bottom and suspended sediments, if further refinement is required. However, this requires that data are available on the phases and the behavior of the chemical in them. When a chemical is discharged to the aquatic environment it distributes in the pattern diagrammatic shown in Fig. 3. At equilibrium the concentrations in the phases (water, air etc,) are constant and the equilibrium between each pair of phases is characterized by an equilibrium constant, ie.e. K_{sew}, K_b etc. Mackay and Patterson (1981) have shown how the fugacity, or "escaping tendency" can be calculated for chemicals in the environment and used to calculate the potential distribution.

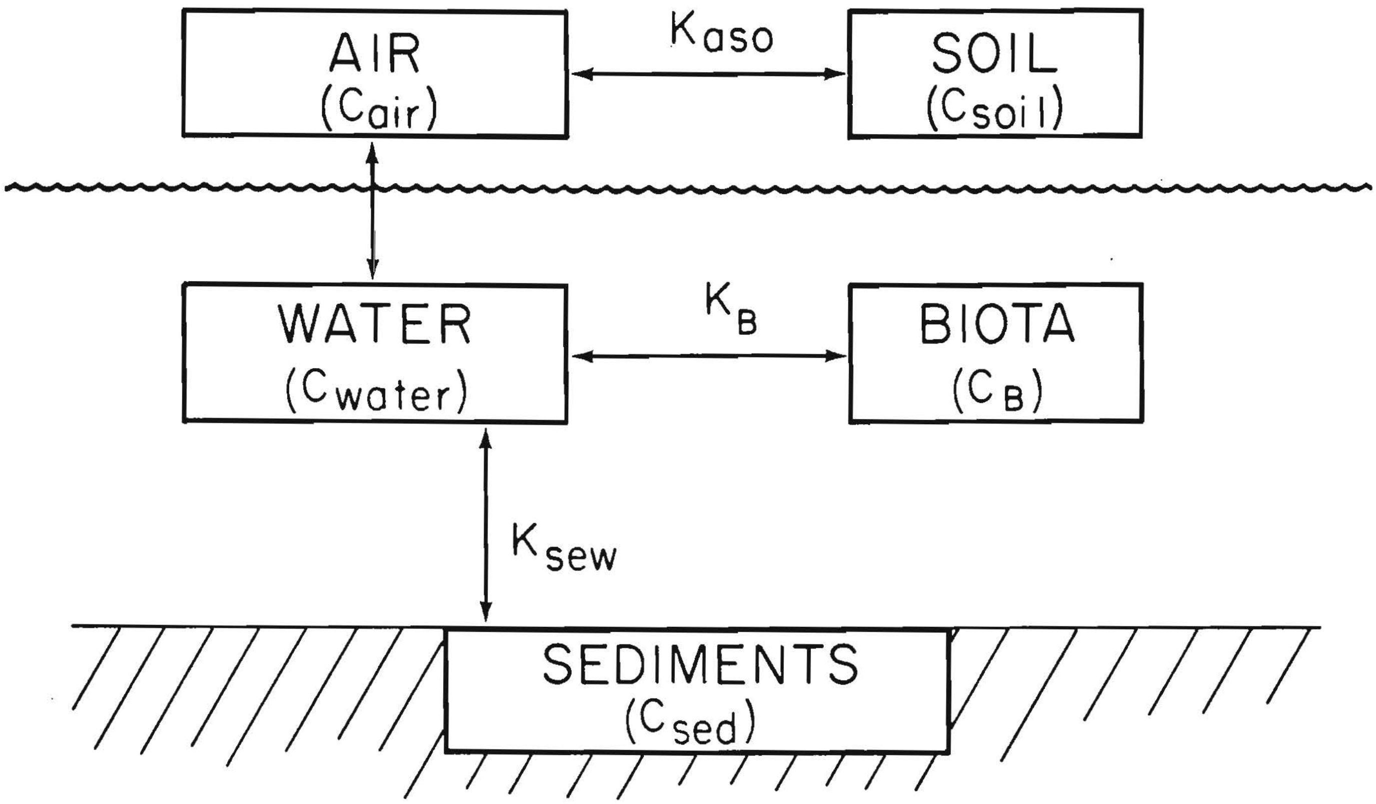

Fig. 3 - Distribution of a Chemical Between Phases in the Aquatic Environment

3.2 The Sediment to Water Exchange

The sediment is of critical importance because sediment contains relatively high concentrations and acts as a reservoir which maintains low concentrations of lipophilic contaminants in the water mass. From the water mass these substances can be taken up by biota. Karickhoff (1981) has shown that in marine systems the water to sediment equilibrium generally follows the Freundlich equation which is characterized by the linear relationship

$$C_{sed} = K_{sew} \, C_w$$

where C_{sed} is equal to the concentration in the sediment, K_{sew} the distribution coefficient between water and sediment and C_W the concentration in the water (see Fig. 3). The key characteristic in this equation is the sediment to water partition coefficient, K_{sew}, and this can be calculated utilizing the following equation

$$K_{sew} = K_{oc} \quad f_{oc}$$

where K_{oc} is the water to sediment distribution coefficient in terms of organic carbon and f_{oc} the fraction of organic carbon present in the sediment. The K_{oc} value can be calculated from the octanol to water partition coefficient, K_{ow}, using the following equation (Jury et al, 1983)

$$\log (K_{oc}/1000) = 1.029 \log (K_{ow}/1000) - 0.18$$

Thus, from a knowledge of the K_{ow} value of the compound and the f_{oc} for the sediment its K_{sew} can be calculated. This can then be used to calculate possible concentrations in overlying water, C_w, which result from differing lipophilic chemical concentrations in sediments with different organic carbon contents. But it should be kept in mind that this only applies when equilibrium is established and for dynamic systems in the natural environment this is rarly attained.

3.3 The Bioaccumulation of Dissolved Contaminants

The other equilibrium of particular concern in the aquatic environment is the water to biota equilibrium (see Fig. 3.) The bioconcentration of chemicals from the water and biomagnification of chemicals in food by organisms are of particular importance in determining the biological effects of chemicals in the aquatic environment. In general, organisms take up chemicals from two principle sources, directly from the ambient environment or in food. But contaminants in food must ultimately have been derived from the ambient environment. Recent literature (e.g. Davies and Dobbs, 1984) indicates that irrespective of the source of chemicals the final concentrations in organisms approach that which would be expected from water alone, i.e. bioconcentration. Thus this process alone can be considered to control the concentrations of contaminant chemicals in organisms and the deleterious effects on biota.

The atmosphere contains very low concentrations of contaminants compared with water bodies, such as the oceans, lakes, rivers, etc. and as a result the water to organism relationships have received most attention. The bioaccumulation process can be seen as a balance between two first order kinetic processes, uptake and clearance characterized by rate constants k_1 and k_2 respectively (Hawker and Connell, 1986). These processes are depicted diagramatically in Fig. 4. Thus the concentration in an organism will be at equilibrium when the rates of

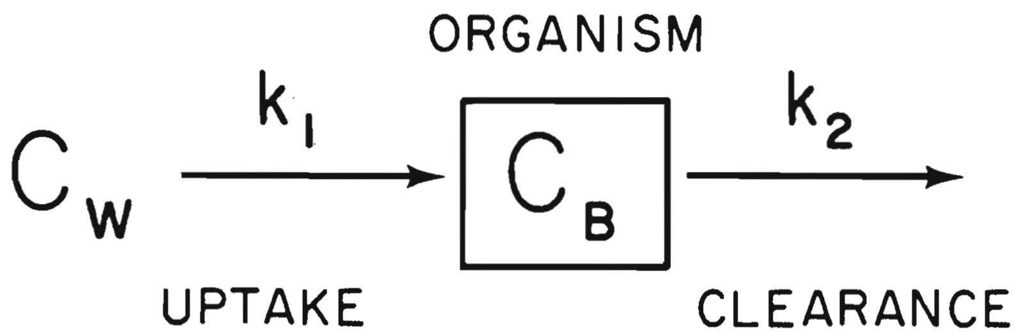

Fig. 4 - Single Compartment Model for the Upstake and Loss of a Lipophilic Chemical by an Organism

uptake and clearance are equal. The bioaccumulation which occurs can be characterized by the bioaccumulation factor (K_B) which is the ratio of concentration in the biota (C_b) to concentration in the water (C_w). For a particular compound and particular organism, or groups of organisms, e.g. fish, K_B has been shown to approach a constant value (e.g. Mackay, 1982). A knowledge of the K_B value and the concentration of a chemical in water will allow estimation of the concentrations likely to occur in organisms.

Mackay (1982) has demonstrated that for lipophilic compounds the major concentrating phase in biota is the lipid phase and the aqueous tissues have little effect. Thus bioconcentration can be viewed as essentially the establishment of an equilibrium between biota lipid and the surrounding water. For example, Mackay (1982) has found the following relationship with fish

$$\log K_B = \log K_{ow} - 1.320 \qquad \text{Eqn. 1}$$

Also Hawker and Connell (1986) have found the following relationships

$$\log K_B = 0.844 \log K_{ow} - 1.235 \text{ (molluscs)}$$

$$\log K_B = 0.898 \log K_{ow} - 1.315 \text{ (daphnids)}$$

Since k_1/k_2 is directly related to the K_B value and K_B is directly related to K_{ow}, it is not entirely unexpected that there is a relationship between K_{ow} and k_1 and k_2. Hawker and Connell (1985) have found that for fish the following relationships apply between the rate constants and K_{ow}

$$\log 1/k_2 = 0.663 \log K_{ow} - 0.947 \qquad \text{Eqn. 2}$$

$$\log k_1 = 0.337 \log K_{ow} = 0.373 \qquad \text{Eqn. 3}$$

Thus, the rate constants with fish can be calculated for a particular compound provided its K_{ow} value is known. These rate constants together with a knowledge of C_W and C_B will allow the actual rates of uptake and clearance of chemicals from fish to be calculated.

4. OCCURRENCE OF PCB'S IN SEDIMENTS AND WATER

4.1 Sediment Concentrations

Results from analyses of PCB's in sediments in harbors and rivers along the north shore of Long Island Sound are listed in Table 2. Each of these values represent a mean concentration of between 1 and 11 analyses which were carried out by EPA in the period from 1975 - 1985 and those in Table 2 for which no standard deviation is listed are the result of one analysis. In addition Table 2 contains corresponding figures on the population in the local area associated with the harbor or river location listed. Table 2 also lists PCB concentrations in sediments collected offshore in the Sound by Chytalo (1979) and results obtained by NOAA (1982). The same set of results reported in Table 2 are plotted in Fig. 5 to indicate the geographical spread of the data.

The results indicate that there are significant concentrations of PCB's in the harbors and rivers along the north shore of Long Island Sound (see Table 2 and Fig. 5). A general association of PCB's with the urban populations is evident from this data. For example, five out of the eight highest concentrations were in areas with the highest local population, generally greater than 70,000. On the other hand the seven lowest concentrations all had populations less than 70,000. A close correlation between urban populations and PCB's concentrations would not be expected due to the different nature of urban runoff patterns and the different specific locations and types of industries. It is important to note that sediments high in organic carbon would be expected to be efficient accumulators of PCB's compared with those which are low in carbon. Therefore the PCB to TOC (total organic carbon) ratio would be

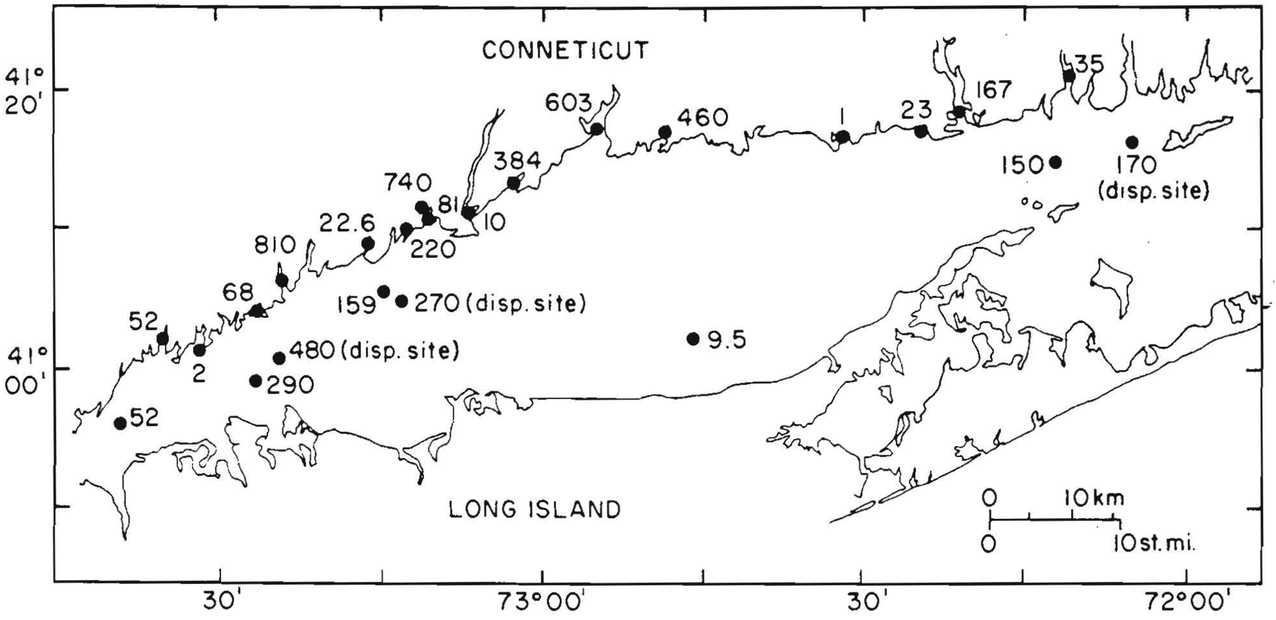

Fig. 5 - Concentrations of PCB's in Sediments in Long Island Sound (ppb)

TABLE 2. Concentrations of PCB's in Sediment and Calculated for Water in Long Island Sound*

Location	Mean Sediment Concentration ppb(SD)	PCB/TOC	Local Government Area	Population	Calculated Maximum Water Concentration (ppb)**
Clinton Harbor	1	-	Clinton Town	12,000	
Housatonic River	10	-	Stratford Town	50,000	
North Cove Old Saybrook	22(30)	6.4	Old Saybrook Town	9,800	0.016
Mill River	22	-	Fairfield Town	53,000	
New London Harbor	35(28)	11.9	New London City	29,000	0.028
Mianus River	52(37)	11.2	Greenwich Town	59,000	0.028
Five Mile River	68(64)	13.3	Old Saybrook Town	9,800	0.033
Bridgeport Harbor	87***	-	Bridgeport Town	>142,000***	
Connecticut River	167	-	Lyme Town	2,000	
Blackrock Harbor	220(382)	67.2	Bridgeport City	142,000	0.17
Milford Harbor	38(85)	103.4	Milford City	52,000	0.26
Branford Harbor	460	312.9	Branford Town	25,000	0.78
New Haven Harbor	603(398)	157.8	West Haven City	178,000	0.39
Bridgeport Harbor	736(837)	204.5	Bridgeport City	142,140	0.51

(Continued)

TABLE 2 (Continued)

Norwalk Harbor	810(566)	-	Norwalk City	78,000	--
Eatons Neck Disposal Site	480(200)	110			0.2
Eatons Neck Control	290(30)	30			0.07
Bridgeport Disposal Site	270(140)	50	Offshore		0.12
Bridgeport Control	150(20)	20			0.05
New London Disposal Site	170(90)	30			0.07
New London Control	150(10)	20			0.05
Western Sound	52	--	Offshore		0.017
Eastern Sound	9.5	--			0.003

* based on 1. EPA 1975-85 data for onshore harbor and river results in upper section of the table. 2. Chytalo, 1979 for disposal and control sites offshore. 3. NOAA, 1982 for offshore Sound results.

** based on the 1254 grade

*** in an area of lower population

expected to compensate for different PCB concentrations as effected by the TOC levels present. So on observing Table 2 it can be seen that areas with PCB to TOC ratios greater than 30.29 are associated with the higher populations and those with PCB to TOC ratios less than 13.29 are associated with lower populations.

The origin of the PCB's in these locations is difficult to demonstrate in a convincing manner since specific point sources are few in the general area. The Housatonic River has received discharges of PCB's from an plant at Pittsfield in Massachusetts. This was sufficiently serious in 1977 to cause fish in downstream Connecticut to accumulate concentrations in excess of 5 ppm (Anon, 1978). However residues detected by EPA (1975-85) amount to 10 ppb in the Housatonic River sediments towards the mouth and place this area in one of the areas having comparatively low contamination. This data suggests that most have originated from the urban area generally, probably as trace components in such discharges as urban runoff and sewage discharges. Subsequently these trace components have been adsorbed on organic matter in the sediments and accumulated over time due to their high level of environmental persistence.

Concentrations in the offshore sediments by Chytalo (1979) are comparatively closely related to the data on harbors and rivers along the northshore. This is not surprising since a proportion of the sediments present in the dumpsites (see Fig. 2 and Fig. 5) has originated from the disposal of dredging spoil derived from the inshore harbors. The offshore data shows a general trend of declining concentrations from west to east in the Sound (see Fig. 5). This would be expected for at least two reasons. Firstly, the New York metropolitan complex is located at the western end of the Sound and associated discharges of sewage and urban run-off would be expected to contain trace concentrations of PCB's which would be deposited in sediments. Secondly, tidal flushing is greater at the eastern of the Sound than at the western end allowing higher concentrations of contaminants to build up in the western part of the Sound.

The control sites in the work by Chytalo (1979) are not in the dumping areas and in all cases exhibit lower concentrations than the adjacent dumping sites. In addition, the concentrations which occur throughout the six sites studied by Chytalo range from 480 - 150 ppb. This is a fairly narrow range of concentrations within an area covering several hundred square miles. To obtain a more precise evaluation of distribution, taking into account organic carbon content of the sediments the PCB to TOC ratio was calculated for these sediments and is entered into Table 2. Excluding the value for the Eatons Neck Disposal Site, the other PCB to TOC ratios occur in the range 20 to 50 suggesting a reasonably consistent low level of distribution of PCBs throughout the sectors of the Sound covered by these determinations. This may indicate a degree of mixing of bottom sediments throughout large portions of the Sound. But sectors of the Sound where sediments are sand and gravel would be expected to be low in organic carbon and thus low in PCB concentration. The results in Table 2 reported by NOAA (1982) support the east-west decline in PCB concentrations apparent in the results of Chytalo (1979) but are generally of a lower order of magnitude.

4.2 Calculated Water Concentrations

The water concentrations which would result from equilibrium between water and sediments, and having the PCB and TOC concentrations noted in Table 2, were calculated from the relationships established by

Jury et al (1983) (see Section 3). The water concentrations were calculated based on the Aroclor Grade 1254 with an average K_{ow} value of 6.5 and are indicated in Table 2.

It should be noted that these figures result from calculations assuming equilibrium between water and sediment has been established. This state most probably has not been reached in the Sound due principally to evaporation of PCB's from the water surface. However, the values are probably reasonable representations of concentrations which exist in bottom waters in close contact with the PCB containing sediment. The overlying waters would be expected to be lower than these calculated values. On the other hand, although these figures are calculated they have some potential advantages over actual measurements made on the water mass. Firstly, the actual determinations often not only include dissolved PCB's but also include PCB's adsorbed onto fine particulate matter. In considerations of bioaccumulation of PCB's by biota it is only the dissolved fraction which is normally taken up. Secondly, the water concentration values are usually extremely low and can be below or near the detection limit of measurement techniques. Thus actual measurement may not be able to provide any values at all or may provide values which are not accurate.

4.3 Comparison of Long Island Concentrations with Those Elsewhere

To place these sedimentary concentrations in perspective Table 3 includes broad summaries of PCB's concentrations from the Sound and from other areas as a basis for comparison. The harbors and river data for the north shore of the Sound are most appropriately compared with the Hudson River data and the data for U.S. estuaries and marine areas. This suggests that the Long Island Sound harbors and rivers lie in the

TABLE 3. Comparison of Long Island Sound Sediment PCB Concentrations with Values from Elsewhere

Area	Year	Concentration (ppb)	Reference
Upper Hudson River	1977	>50,000 (average in forty areas) max >1,000,000	Brown et al, 1985.
Upper Hudson River	1981	>200,000 (in some areas)	Brown et al, 1985
New York Bight	1980	150->1	NOAA, 1980
Great Lakes	1979	100-4	Connell and Miller, 1984
U.S. estuaries and marine areas	1979	up to 486,000 close to discharges	Connell and Miller, 1984
L.I.S. harbors and rivers	1975-85	810-1	Table 2
L.I.S. offshore	1979-80	480-9.5	Table 2

lower range of the PCB contaminated areas in the coastal zone. The Long Island Sound offshore data is probably best compared to the concentrations in the New York Bight and the Great Lakes. In this instance the concentrations lie in a similar range indicating that the Long Island Sound sediments have a similar level of PCB contamination to other somewhat related areas.

5. PCB's IN THE STRIPED BASS (*Morone saxatilis*)

5.1 Composition and Occurrence of PCB's

The New York Department of Environmental Conservation (Sloan et al, 1986) has published a considerable volume of data on the concentrations of PCB in the Hudson River striped bass and from elsewhere. Table 4 is selected from that data and shows the mean concentration variation with length in New York Harbor and Long Island Sound during 1985. It was decided to examine data from 1985 as a typical example rather than include data from other years as well. The other years have similar patterns of concentration increase with length but the inclusion of this with the 1985 data would be expected to lead to variations due to year to year environmental factors.

The overall concentration of PCB's in striped bass from 1978 to 1981 exhibited a general decline as shown in Fig. 6 and in addition the composition of the PCB's in the fish changed. It would be expected that the lower molecular weight PCB's, which are more volatile and water soluble, would be removed from the PCB mixture in the early stages after release leaving the higher molecular weight substances. Table 5 shows the composition of Aroclor 1016 and Aroclor 1254. It can be seen that Aroclor 1016 contains di-, tri-, and tetra-chlorobiphenyls as its major constituents whereas Aroclor 1254 contains tetra-, penta-and hexa-chlorobiphenyls as its major components. Thus as weathering occurs the lowest molecular fractions of the mixture, which are the lower compounds in the 1016 grade i.e., the di-and tri-chloro compounds, would be removed. This is illustrated by the information in Fig. 6 which shows a decline in the proportion of the 1016 grade as compared to the 1254 grade over time. Thus the 1016 grade would produce residues containing

TABLE 4. PCB in Striped Bass in New York Harbor and Long Island Sound

Area	Size (in)	All Seasons Mean Concentration (ppm)
New York Harbor	≥18	2.78
	≥24	3.53
	≥30	3.97
West Long Island Sound	≥18	2.32
	≥24	2.48
	≥30	2.84
East Long Island Sound	≥18	1.83
	≥24	2.01
	≥30	2.45

* from Table 4, Sloan et al., 1986

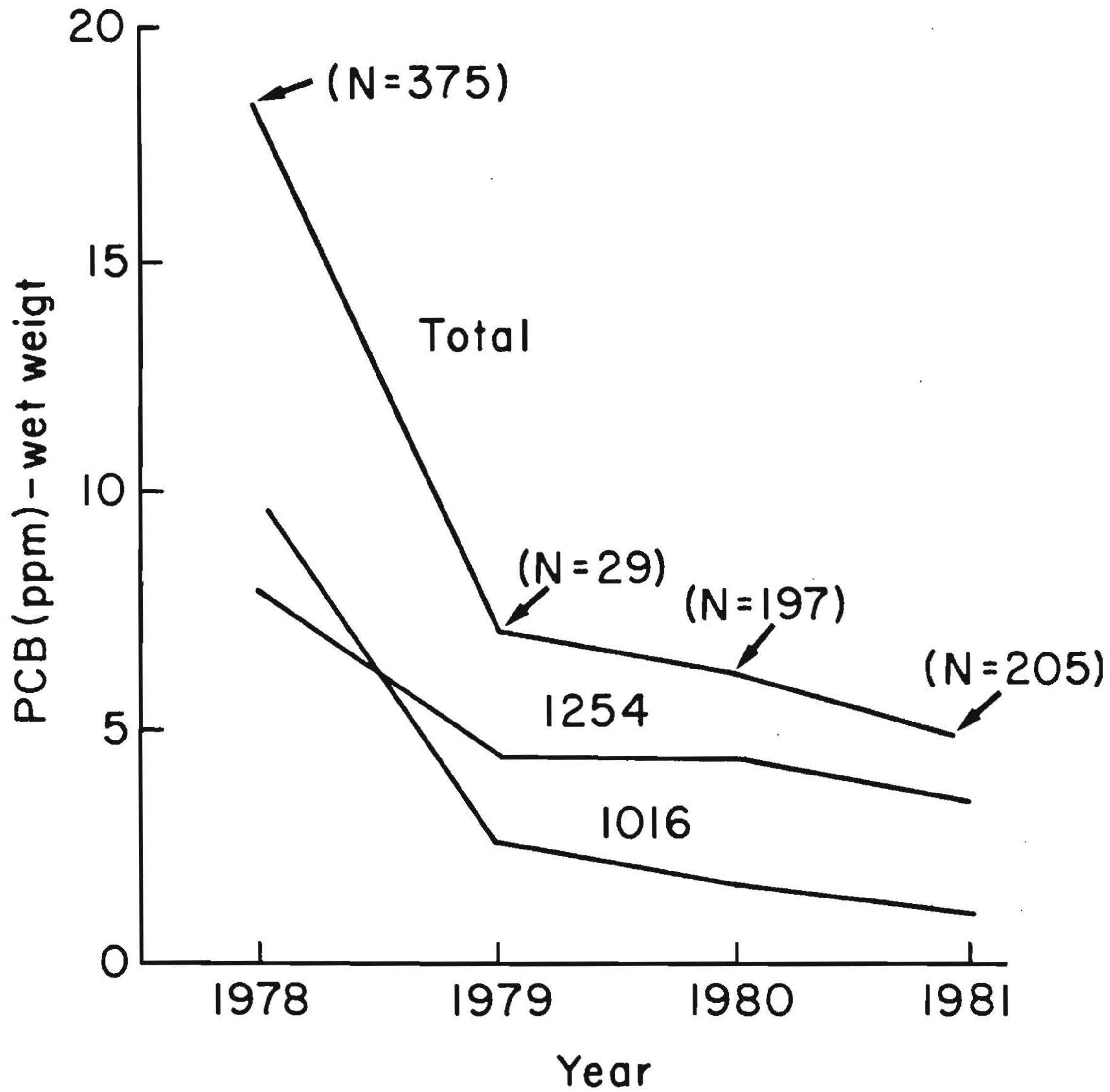

Fig. 6 - PCB Trends in Striped Bass Collected from Riverwide Locations in the Hudson River (From: Sloan and Armstrong, 1981)

TABLE 5. Composition of Aroclors 1016 and 1254*

Major chlorobiphenyl components	Grade 1016 (% Comp)	Grade 1254 (% Comp)
dichloro	20	0.5
trichloro	57	1
tetrachloro	21	21
pentachloro	2	48
hexachloro	-	23
heptachloro	-	6

* based on Hutzinger et al. 1974

mainly tetrachlorobiphenyls and the 1254 grade would degrade at slower rate and produce mainly penta-, hexa- and hepta-biphenyls. The weathered PCB residue remaining from both grades would be expected to have an overall composition of tetra-, penta-, hexa- and hepta chloro compounds with average properties centered around the hexa- to penta compounds (see Table 6). Table 6 contains data on bioconcentration factors (K_B) and kinetics derived from Mackay (1982) and Hawker and Connell (1986) which can be used in estimating likely bioaccumulation from water as described below.

5.2 Increase in PCB Concentration with Fish Length

The age of striped bass is required to enable calculations to be made of bioconcentration over time. The length of a fish is often related to its age, particularly within a limited geographical range. It was found that a plot of the age of striped bass against the length of the fish for the Hudson River and Chesapeake Bay populations gave a significant correlation (see Fig.7). Thus

length (in) = 2.6(age in years) + 10

$r = 0.934$

This equation allowed fish samples of different mean lengths to be aged and thus plots to be made of PCB concentration versus age (see Fig. 8). Fig. 8 is in fact the data in Table 4 set out in graphical form in which the age corresponding to the length has been calculated and noted on the graph.

Hawker and Connell (1987) have investigated the kinetics of bioconcentration of lipophilic compounds, including PCB's, by fish. In this work they found that the exposure time is of critical importance in influencing observed concentrations with compounds of relatively high

TABLE 6. Physiochemical and Biological Characteristics of the Biphenyls with Fish

Number of Chlorines	Approximate log K_{ow}	Calculated log K_B*	Calculated K_B*	Calculated k_2 (hr^{-1})	Half Life** (Days)
3	6.0	4.68	47,900	0.0009	14
4-5	6.2	4.9	79,400	0.0006	21
5	6.5	5.2	58,000	0.0004	31
5-6	7.0	5.68	480,000	0.00014	90
6-7	7.8	6.48	2,500,000	0.00006	209

* see Mackay, 1982

** see Hawker and Connell 1986

*** calculated from k_2

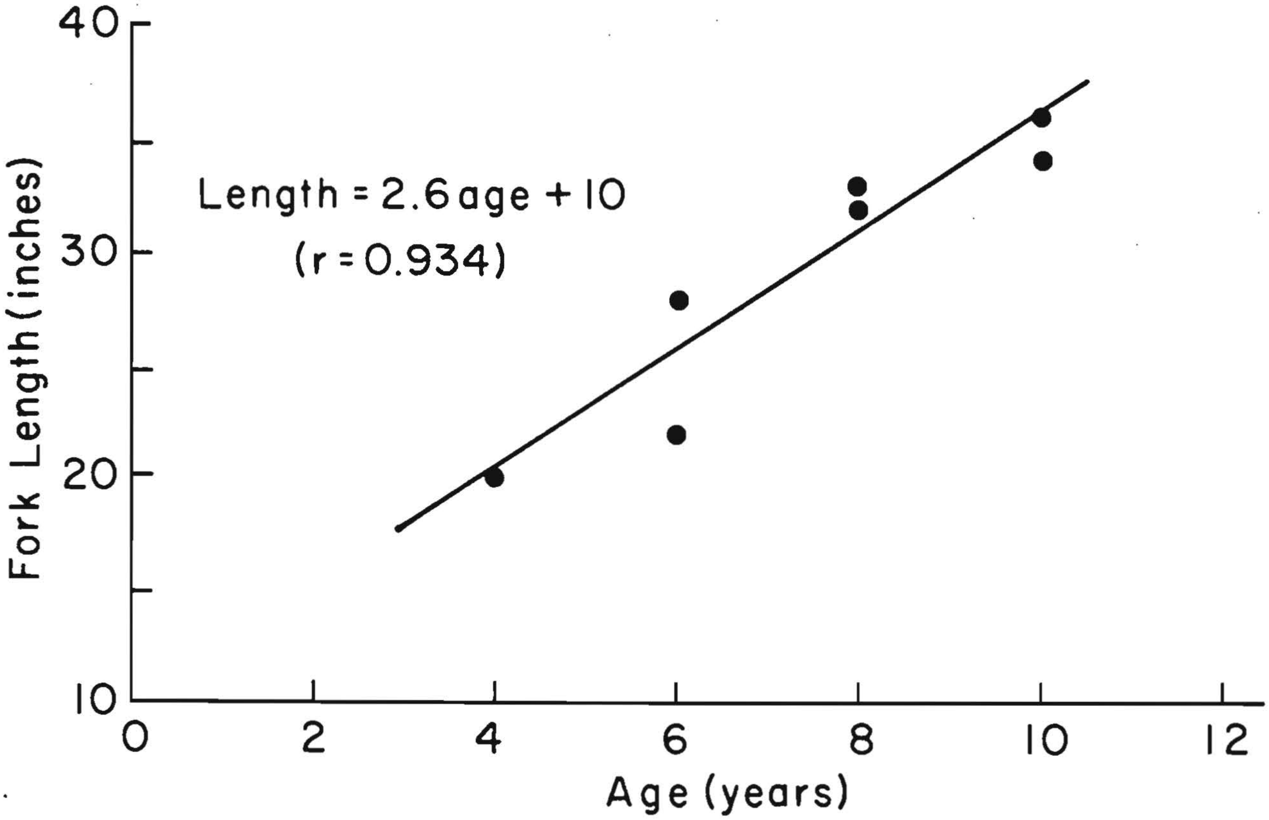

Fig. 7 - Plot of Age Against Length with the Striped Bass (Data from: Texas Instruments, 1973; Pearson, 1938)

K_{ow} values such as the PCB's in Grade 1016 and 1254. Equations were produced relating the K_{ow} values to k_1(the uptake rate constant) and k_2 (the clearance rate constant) for a range of fish species as outlined in Section 3, Eqns. 2 and 3. Using these relationships, plus that derived the K_{ow} to relationship K_B, Eqn. 1, the data in Table 6 was calculated and is generally applicable to the striped bass. In the previous section it was concluded that the PCB residue in 1985 would have properties centered about the penta- to hexachloro compounds. Thus the data in Table 6 for the compounds containing 5 to 6 chlorine atoms is believed to be applicable to the PCB residue in striped bass in 1985.

The uptake of PCB's would be expected to obey approximately first order kinetics. Thus

$$C_B = C_W . \ k_1/k_2 \ (1 - e^{-k_2 t})$$

where C_B is the concentration in the fish, C_W the concentration in the water, k_1 and k_2 the uptake and clearance rate constants and t the exposure time. But $k_1/k_2 = K_B$, thus

$$C_B = C_W . K_B (1 - e^{-k_2 t}) \qquad \text{Eqn. 4}$$

If values are available for C_W, K_B, k_2 and t then the equation above can be used to calculate values for the concentration in the organism after different periods of exposure. Values for K_B and k_2 have been calculated for the PCB residue most likely to occur, ie the penta- to hexa-chloro compounds, as shown in Table 6. However, data is also needed on the concentration of PCB's in water (C_W) and the likely exposure period. This data was obtained as outlined below.

Sloan et al. (1983) have produced a set of data on the concentration of PCB's in the lower Hudson River in the period from 1978 - 1981. The decline in PCB concentration in the water would be expected to approximate first order kinetics and thus produce a log/normal relationship between concentration and time. In fact, over the period from 1978 to 1981 this relationship applies but over more extended periods it would be expected to deviate increasingly from this for reasons outlined later in Section 5.4. Nevertheless an approximate water concentration can be obtained by extrapolation of the log/normal relationship. The regression equation between elapsed time in years and water concentration was found to be

$$\log C_W = -0.14 \text{ (elapsed time)} - 0.53$$

$$(r = -0.92)$$

By substituting into this equation the elapsed time of eight years a predicted value for 1985 of 0.02ppb is obtained.

Another calculation of C_w in 1985 can be made from other data. Brown et al. (1985) report concentrations for PCB's in water in the upper Hudson River from 1979 to 1983. By extrapolating these according a log/normal distribution a water concentration of 0.05ppb is obtained for 1985. In addition this paper reports that in the late 1970s the ratio of concentrations in the Upper Hudson River to the Lower Hudson River was 0.58/0.15. By applying this to the 1985 upper Hudson River figure of 0.05ppb a value of 0.012ppb is obtained for the water concentration of PCB's of the lower Hudson River in 1985. This is in reasonable agreement with the concentration obtained from the alternative set of data of 0.02ppb.

An approximate exposure period can be obtained from the age of the fish derived from its length. However some fish will be continuously resident in the Hudson River, others partial residents and a propostion will be migratory, spending a significant period at sea. The average fish would be expected to be exposed to Hudson River concentrations for considerably less than the full period of its life span. Thus exposure periods equivalent to half the age of the fish were substituted into Eqn. 4 for C_B above at ages of 3.08, 5.4 and 7.7 years. These concentrations are entered into Fig. 8 as concentrations in striped bass calculated for New York Harbor.

The concentrations obtained by calculation for New York Harbor fish can now be compared with the actual analyses of the PCB content of striped bass in the Harbor as shown in Fig. 8. The two curves show reasonable agreement considering the variety of factors which have been estimated or not taken into account at all. For example the concentration of PCB in the Lower Hudson River water is only an approximate estimate and has a major influence on the values obtained for PCB in fish. Age is accounted for by extended exposure times only utilizing the same kinetic rate constants. It would be expected that at different ages fish of different sizes would exhibit different rate constants. Different lipid contents are also possible which may also influence the final PCB concentration.

The slopes of the curves are in quite good agreement but the calculated curve indicates that the concentration is trending towards a maximum whereas the actual curve suggests further increases in concentrations with time. However, overall, this data suggests that the rate of bioaccumulation is a major factor in causing an increase in PCB concentration with increased length and age.

5.3 PCB Contaminants in Fish from Long Island Sound

The actual data obtained by Sloan et al. (1986) by analysis of fish from Long Island Sound on the change in PCB concentration with length age as shown in Fig. 8. There is a similar pattern of concentration

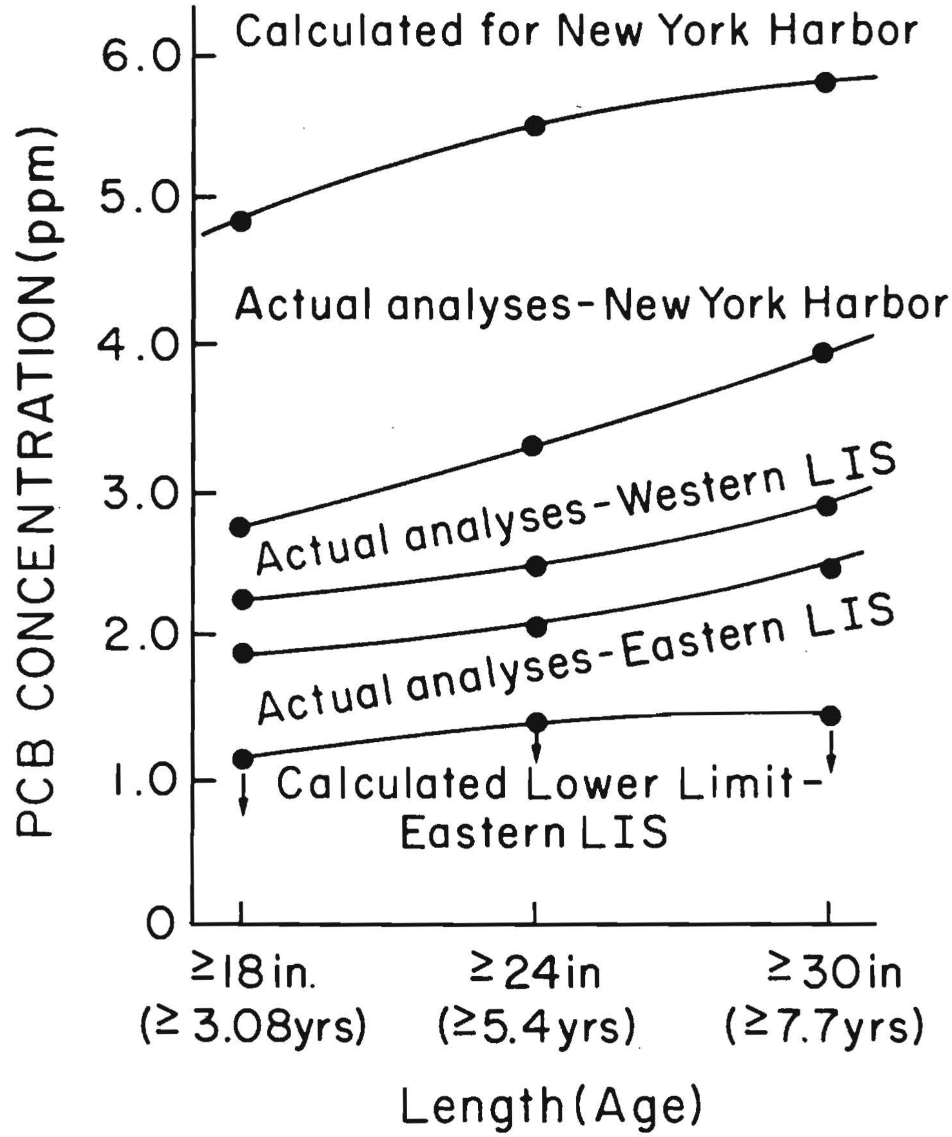

Fig. 8 - Calculated and Actual Measured PCB Concentrations Versus Age/Length for the Striped Bass from Different Areas

increase with length/age in east and west Long Island Sound as was observed with the lower Hudson River. However, the concentrations of Long Island Sound fish are lower than those observed for the New York Harbor area and the eastern Long Island Sound values are somewhat lower than the western Long Island Sound values.

Sloan et al. (1986) results indicate that 12 to 56% of the fish in western Long Island Sound have originated from the Hudson River and that 3 to 83% of fish in eastern Long Island Sound have the same origin. This illustrates that on occasions a high proportion of the mean PCB concentration observed in Long Island Sound fish is due to contaminated fish from the Hudson River. On the other hand Table 2 lists the sediment and calculated water concentrations at equilibrium in various parts of Long Island Sound. In fact these water concentrations would probably never be reached because equilibrium is not attained due to the evaporation of PCB from the water surface. But these calculations suggest that water concentrations in some parts of the Long Island Sound could approach, or exceed, those in the lower Hudson River, 0.012 to 0.02 ppb in 1985. Thus it could be suggested that an upper limit for PCB bioconcentration by fish which are continually exposed to the higher concentrations in Long Island Sound is about equivalent to the concentrations attained in the Hudson River (see Fig. 8). However, with the continuous movement of the fish it is unlikely that any single fish would be continuously exposed to these high concentrations so as to attain these relatively high concentrations. Thus these values could therefore be seen as a possible upper limit for PCB concentrations which could be attained by individual fish in Long Island Sound. At the other end of the scale a lower limit can be derived for PCB concentrations in fish exposed to the lowest concentration in Sound water. Table 1 records the possible lowest concentration at somewhere around a value of 0.03ppb at equilibrium. However, this high value is unlikely to be attained due to evaporation at the water surface and so this should be seen as a maximum value for the lower limit of concentration in the water.

Calculations for the likely PCB concentrations in fish at different ages have been made in using the same equation as was used in the previous section (Eqn. 4) and these concentrations shown in Fig. 7. Thus fish permanently resident in Long Island Sound could accumulate anywhere between these two values as a result of PCB concentrations in water at equilibrium, with the sedimentary mass. This indicates that striped bass in Long Island Sound could accumulate a significant proportion of their PCB load from PCB's in the area or, if permanently resident, could attain significant concentrations.

5.4 Rate of Decline in PCB's in Striped Bass with Time

The half lives of the various PCB's in fish are listed in Table 6. However, these are only apparent when the water concentration is zero and thus exposure has ceased. This is not the case with the striped bass in the Hudson River and Long Island Sound. In these situations there is continuous exposure in the water and persistence in the fish is controlled by the water concentration and not the half life of the compounds in the fish. The persistence of the PCB's in water is somewhat similarly controlled by the persistence of these substances in sediments rather than their persistence in the water mass. The Aroclor grades are all complex mixtures and the low molecular weight fractions are removed first leaving the more persistent fractions (see Fig. 6). In Table 6 the half lives of various PCB's in fish are listed and this illustrates the increasing persistence of the PCB's at higher molecular weights. So over time as the lower molecular weight fractions are removed the persistence in the sediments and also the water and fish, will increase.

This affect is illustrated by data from Brown et al. (1985) plotted in Fig. 9. In Hudson River water and the striped bass from the lower Hudson River and Long Island Sound there is an initial rapid rate of decline of PCB's from 1976 to about 1980 and then a slower rate of decline after that time. This reflects the change in composition in more recent years to higher molecular weight compounds with longer half lives and also higher K_B values (see Table 6). Thus although the concentrations in sediment and the water have declined over time the bioconcentration factor from water would be expected to increase. Accordingly plots of concentration in fish over time would not be expected to closely obey first kinetics which would provide a reasonably reliable means of estimating future concentrations.

The plots in Fig. 9 show that, in general terms, there is a relationship between PCB concentration in Hudson River water, Hudson River fish and Long Island Sound fish. The trend of the Hudson River fish is towards a slow decline with time and since many of the Long Island Sound fish originate from the Hudson river a similar slow decline with time would be expected with Long Island Sound fish. But fish accomulating PCB's in the Sound itself would be expected to respond to temporal changes within that environment. Thus it is suggested that PCB's in Long Island Sound fish will exhibit and comparatively slow rate of decline from the present concentration for some years into the future.

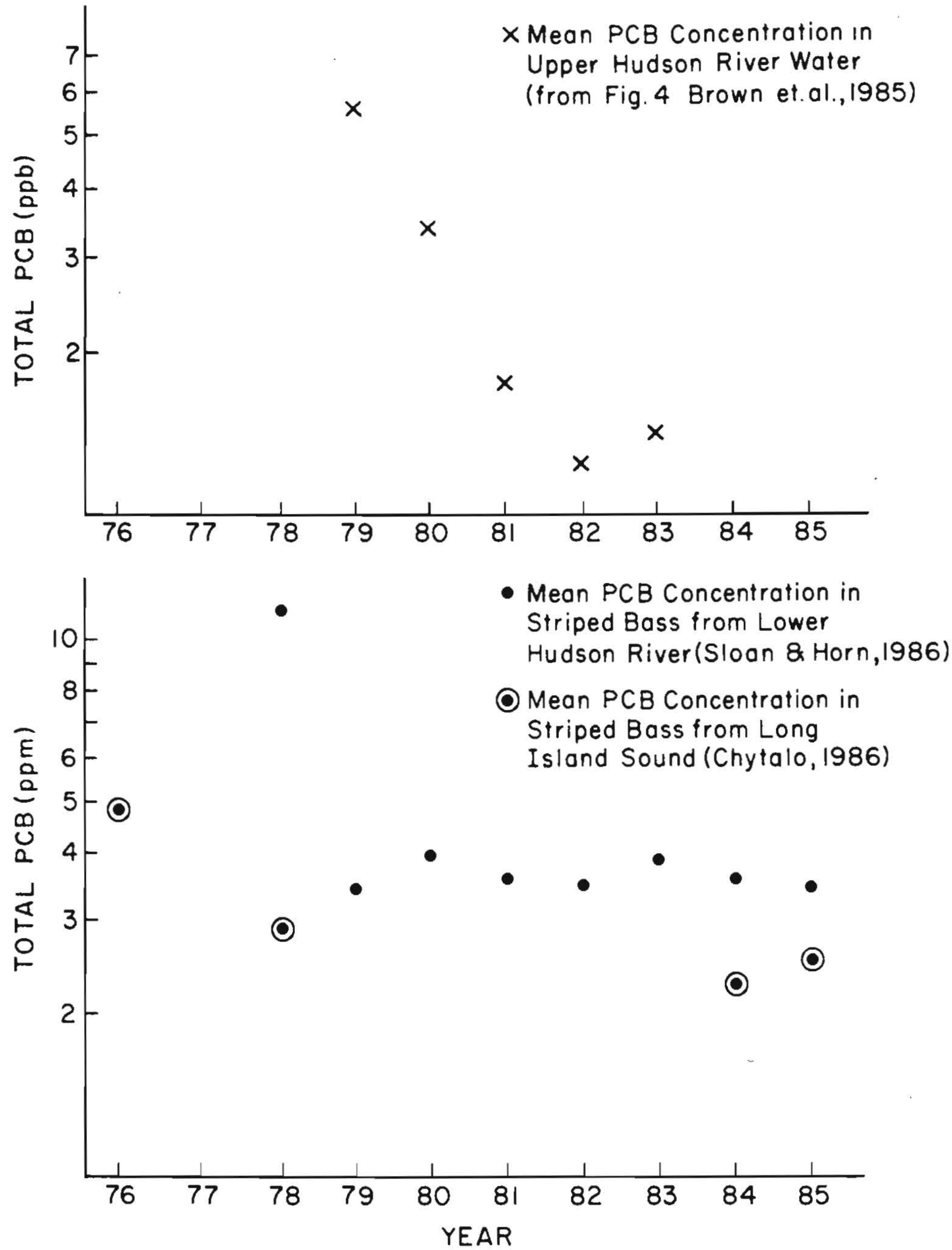

Fig. 9 - Trends of Mean PCB Concentration over Time in: (x) Upper Hudson River water (From: Fig. 4, Brown et al, 1985); (•) Striped Bass from the Lower Hudson River (From: Sloan and Horn, 1986); (◉) Striped Bass from Long Island Sound (From: Chytalo, 1986)

6. PCB'S IN OTHER BIOTA

6.1 Mussels

The mean concentrations in mussels (Mytilus edulis) obtained by the National Fisheries Service for locations along the northern shore in Long Island Sound are listed in Table 7 (Greig and Sennefelder, 1985). In some cases, where the organic carbon content of the sediments is known, the maximum concentrations in the overlying water have been calculated using methods previously described. These results are also included in Table 2 and allowed the calculation of the bioconcentration factor (K_B) as the ratio between the concentration in mussels and the concentration in water. An equation for calculating the K_B value with molluscs from K_{ow} has been derived by Hawker and Connell (1986) as

$$\log K_B = 0.844 \log K_{ow} - 1.235$$

The most common grade in Long Island Sound, particularly after extended exposure periods, is the Aroclor 1254 which has properties listed in Table 6 for the PCB's containing four to five chlorines. This grade has a log K_{OW} of 6.2 and using the equation above it should have log K_B of 3.99 which is equivalent to K_B of 9950. This is considerably higher than the K_B values listed in Table 2. This discrepancy could be due to errors of the estimation of C_W for reasons outlined previously.

6.2 Fish

The mean concentrations found in fish in different locations in Long Island Sound are shown in Table 8. In only one case, the windowpane flounder, was any information available on the PCB content in food. This indicates that a significant increase in concentration over

TABLE 7. Characteristics of PCB's in mussels in Long Island Sound

Location	Mean Mussel Concentration* (ppm wet wt)	Calculated Water Concentration** (ppb)	Approximate Bioconcentration Factor K_B (log K_B)
Thames River	0.049		
Connecticut River	0.068		
Hammondsset	0.084		
New Haven Harbor	0.098	0.39	251 (2.400)
Milford Harbor	0.067	0.26	257 (2.409)
Housatonic River	0.087		
Stratford Point	0.067		
Bridgeport	0.115	0.51	225 (2.353)
Norwalk Harbor	0.053		
Cos Cob Harbor	0.057		

* from Greig & Sennefelder, 1985.
** from Table 2

the concentration in food has not occurred with the flounder as a result of consuming PCB contaminated food. In fact the concentrations are of approximately the same order. This is in accord with the conclusions of Davies and Dobbs (1984) in a recent review who found that, irrespective of the source of PCB's ie food or water, the final concentration in fish is in agreement with the bioconcentration factor from water.

Approximate bioconcentration factors based on a limited amount of data on water reported in Table 2 have been calculated. These factors are different from the eastern to the western sectors of the Sound suggesting that the limited data base is probably insufficient for an accurate evaluation to be made.

A theoretical bioconcentration factor can be obtained, as a basis for comparison, from the equation produced by Mackay (1982) describing the relationship between K_{ow} and K_B for a variety of fish species. Thus

$$\log K_B = \log K_{ow} - 1.320$$

The Aroclor grade 1254 which is common in Long Island Sound has 4 to 5 chlorine atoms as its most common constituents and the K_{ow} values for this mixture is about 6.2 (see Table 6). Thus from the equation above $\log K_B$ equals 4.9 and K_B equals 79,400. The K_B values in Table 8 give a wide range of figures both above and below this value. This would be expected since the exposure to PCB's in water would vary with different species due to their different movement patterns and thus their contact with different water concentrations. In addition, lipid content, age, size and so on would need to be taken into account to provide more accurate values.

It's interesting to note that in some cases the western biota contain lower concentrations of PCB's than the eastern biota. This

TABLE 8. Characteristics of PCB's in Fish from in Long Island Sound (excluding striped bass)

Species	Location	Concentration* (ppm, wet wt)	Approximate Bioconcentration Factor, K_B (log K_B)	Mean Concentration* in Food (ppm, wet wt)
Windowpane	West Sound	0.04	2,350 (3.37)	0.14
Flounder	East Sound	0.23	76,000 (4.88)	0.13
Winter	West Sound	0.13	7,650 (3.88)	
Flounder	East Sound	0.08	26,700 (4.43)	
Red Hake	East Sound	0.11	36,700 (4.56)	
Atlantic	West Sound	3.07	180,600 (5.26)	
Menhaden	East Sound	3.54	1,200,000 (6.07)	
Bluefish	West Sound	1.08	63,500 (4.80)	
	East Sound	1.89	630,000 (5.80)	
Mummichog	West Sound	0.045	2,650 (3.43)	
	East Sound	0.10	33,300 (4.52)	

* based on NOAA, 1982; Greig et al, 1983 & Chytalo, 1986.
** estimated using calculated water concentrations of Aroclor 1254 shown in Table 2 ie 0.017 and 0.003 ppb for East and West Sound respectively

trend is contrary to the general trend of PCB concentrations in Long Island Sound sediments and water which is for the western sector of the Sound to have greater concentrations than the eastern Sound. However, the differences are not great and may not be significant in this situation.

6.3 Crustacea

The concentrations of PCB's in the rock crab and American lobster found by Chytalo (1987) as shown in Table 9. In this case the concentration trend is for biota from the western Sound to have greater concentrations than those in the eastern Sound, in accord with the sediment concentrations. But in fact the concentrations of all of the lobsters are similar and in a similar range to the rock crab.

6.4 American Eel

The mean concentrations of PCB's obtained by Chytalo (1987) in the American eel collected from inshore locations along the southern shore of Long Island Sound are shown in Fig. 10. These results show a general trend of decline from west to east in accord with the sediment concentrations offshore in the Sound and generally in accord with the decline in the size of urban centers in the same direction.

7. CONCLUSIONS

7.1 Occurrence in PCB's in Sediments and Water and Their Origin

The Long Island Sound contains significant PCB concentrations in its sediments. The sediments in inshore areas such as rivers, harbors and so on lie in the lower range of polluted estuaries but concentrations up to 810ppb are known to occur. Significant quantities

TABLE 9. Characteristics of PCB's in Crustacea in Long Island Sound

Species	Location	Concentration* (ppm)	Approximate Bioconcentration Factor, K_B** (log K_B)
Rock Crab	West Sound	0.20	11,800 (4.07)
	East Sound	0.07	23,300 (4.37)
American Lobster	West Sound	0.17	10,000 (4.00)
	East Sound	0.14	46,700 (4.67)
	Central Sound (1980)	0.14	
	Central Sound (1986)	<0.10	

* based on Chytalo, 1986

** estimated as indicated on Table 8

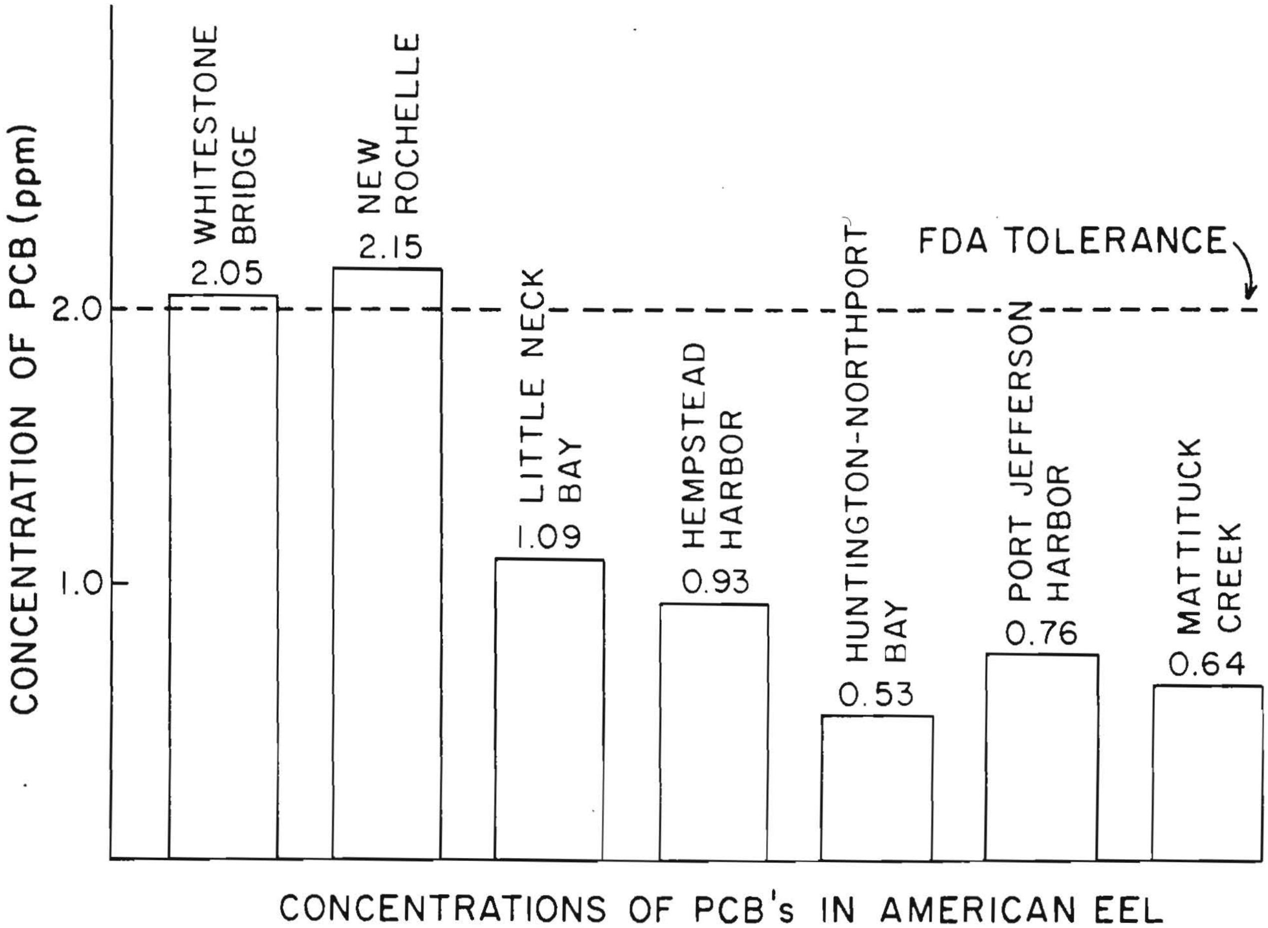

Fig. 10 - Concentrations of PCB's in the American Eel in Inshore Areas on the South Shore of Long Island Sound (From: Chytalo, 1987)

of these sediments have been transferred to offshore locations in the Sound where dispersal has occurred but removal to oceanic areas apparently occurs at a very slow rate. The PCB residues in these offshore areas are comparable in concentration to those in the New York Bight and similar locations elsewhere and range up to 480ppb.

The sediments in all locations provide a reservoir of PCB's from which the overlying water can become contaminated. Possible maximum concentrations in overlying water at equilibrium have been calculated. The PCB's in the water mass, although at extremely low concentrations, are highly dynamic and the major source for the contamination of biota.

In the absence of known significant point sources of PCB's except for industrial discharges to the Housatonic River, the available evidence suggests that PCB contamination offshore in Long Island Sound arises principally from the disposal of PCB contaminated dredging spoil from harbors. The source of the PCB's in the inshore harbors appears to be associated with general urban activities and would be expected to arise from the occurrence of trace amounts of these substances in urban run-off and sewage. In accord with this the inshore harbors and rivers along the northern shore of Long Island Sound show a general relationship between PCB concentration in sediment and the size of the adjacent urban population. The overall trend is for population to decline from the western end of the Sound, adjacent to New York City, towards the east. The offshore concentrations of PCB's in sediments tend to reflect this.

7.2 PCB's in the Striped Bass (*Morone saxitilis*)

It is well known that the PCB concentration in the striped bass increases with length. The age of striped bass corresponding with different lengths has been calculated from existing data. In addition by utilizing rate constants and other data from the literature on the kinetics of bioconcentration the concentration of a weathered Aroclor 1254 in striped bass of different ages has been calculated. The actual PCB concentration at different ages (lengths) was found to be in reasonable agreement with the calculated concentrations utilizing the bioconcentration data. This indicates that the principal factor involved in the increase in concentration with age is due to the slow rate of accumulation of PCB's thus requiring relatively long periods to reach the maximum concentration.

The origin of the PCB's in striped bass in Long Island Sound is of interest. It has been calculated by the methods indicated above that fish wholely resident in Long Island Sound could accumulate significant concentrations of PCB's. This calculation is supported, in a general sense, by the observation that other organisms which spend most of their time in the Sound, such as the rock crab, contain significant concentrations of PCB's which must have been acquired through the Long Island Sound system. Also other fish species with different patterns of movement and behavior exhibit somewhat similar concentrations to the striped bass. The calculations suggest that it is possible for fish in Long Island Sound to accumulate concentrations in the western sector of the Sound comparable to the concentrations which occur in Hudson river fish if they are resident for long periods of time in the areas of high water concentration.

It is suggested that uptake from water is the principal mechanism controlling PCB's in accord with conclusions in the current scientific literature. The current literature indicates that whether food or water are the source of PCB's the final concentration in the organism is equivalent to that obtained by bioconcentration from water. But the available information indicates that a high proportion, greater than 85%, of striped bass can, in some cases originate from the Hudson River population. So the PCB's in these fish could arise from bioconcentration in the Hudson River followed by movement into Long Island Sound. Thus, it can be concluded that there are two major origins for PCB's in Long Island Sound fish. Firstly, direct bioconcentration from PCB's dissolved in water in Long Island Sound and secondly, through PCB contaminated fish from the Hudson River entering the Sound. The actual proportion of fish in each category on any occasion will vary according to the movement patterns of the fish within that group.

The decline in PCB concentration in the striped bass is not amenable to treatment by simple first order kinetics which would provide a reasonably reliable method to predict loss over time. This is due to the fact that with passing time the less persistent fractions in the PCB mixtures are lost and so the PCB's become increasingly more persistent. The trend of existing data is in accord with the generalizations outlined above since in recent years the PCB concentrations have proved to be more persistent and a slowing in the rate of loss from various sectors of the system has become apparent. This suggests that the currently observed slow rate of decline in PCB's will continue for several years. Thus the PCB's in fish in the system are likely to persist at, or near, the current concentrations for the next few years.

7.3 PCB's in Other Biota

Significant concentrations of PCB's are widespread in aquatic biota in Long Island Sound. Mussels in harbors and rivers along the north shore contain concentrations up to 0.115ppm. There is little doubt that these residues have originated from sediments contaminating the overlying water and thus being available for uptake by the mussels. With fish there are relatively high concentrations in some species with the highest occurring in the Atlantic menhaden which has up to 3.54ppm and the bluefish which has up to 1.89ppm. These are comparable with the concentrations which occur in the striped bass which are up to about 2.5ppm. The windowpane flounder, winter flounder, red hake and mummichog generally have lower concentrations usually within the range 0.04 to 0.23ppm. The geographical origin of these PCB compounds is uncertain due to the lack of knowledge of the movement patterns of the fish. With Crustacea the concentrations range from 0.07 to 0.20ppm and since these are generally sedentary it could be expected that a substantial proportion of these PCB's have probably originated within the local habitat in Long Island Sound. The concentrations in these biota cannot be satisfactorily interpreted according to approximate bioconcentration factors which have been calculated. This is presumably due to errors in the estimation of the water concentration of PCB's and to the large amount of variability of exposure to PCB's which occurs with individual species in the Sound.

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