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Concentrations of organic contaminants in *Mytilus edulis* from the Hudson-Raritan Estuary and Long Island Sound

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Abstract

In this paper, we present the findings of 2-, 3-, 4-, and 5-ring polynuclear aromatic hydrocarbons (PAH), selected polychlorinated biphenyl (PCB) congeners, the pesticide DDT and its degradation products, and the marine paint antifouling agent tributyltin (TBT), in the tissues of *Mytilus edulis* collected from 10 sites in the Hudson-Raritan Estuary and 10 sites in Long Island Sound. In the estuary, contaminant concentrations were highest in the Upper Bay, and systematically decreased moving south and east into the New York Bight. Near equal distributions of tetra-, penta-, and hexa-chlorinated PCBs in the Upper Bay systematically shifted to a composition dominated by more heavily chlorinated PCBs in the New York Bight. In Long Island Sound, contaminant concentrations were highest in the heavily populated southwest region of the Sound, while the lowest bivalve contaminant levels were observed near the eastern-most tip of Long Island. PAH distributions generally were reminiscent of complex mixtures of combustion products and refined fuel products. PCB congener distributions exhibited similar changes in composition as those observed in the Hudson-Raritan Estuary.

Keywords: Polynuclear aromatic hydrocarbons (PAH); Polychlorinated biphenyl congeners (PCB); DDT; Tributyltin (TBT); *Mytilus edulis*; Hudson-Raritan Estuary, New York; Long Island Sound, New York

1. Introduction

The objective of the National Oceanic and Atmospheric Administration (NOAA) National Status and Trends (NS&T) Mussel Watch Project is to determine the presence and amounts of selected trace metal and organic contaminants in the tissues of sessile bivalve mollusks collected from near-shore aquatic environments of the

United States, contrast the levels of contaminants on a site-to-site and regional basis, and determine temporal changes in bivalve contaminant burdens at each site. The types and concentrations of contaminants found in the bivalves reflect man's activities near the shoreline or major waterways that feed into the affected coastal areas, and to a lesser extent, the degree of atmospheric input of certain contaminants to the aquatic environment (Rapaport et al., 1985; Wittlinger and Ballschmiter, 1987; Rapaport and Eisenreich, 1988). The rationale for using nonmobile marine

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organisms for monitoring contaminants in the aquatic environment has been discussed previously (Philips, 1980; Farrington, 1983). Bivalve mollusks are pervasive, sedentary biointegrators of particulate-bound and dissolved trace metal and organic contaminants. Since they are immobile, they reflect the bioavailability of contaminants in their immediate habitat. Because they have very primitive enzymatic systems, they do not metabolize accumulated contaminants to any significant degree. Year-to-year variations in contaminant concentrations due to gonadal development stage (e.g. lipid-soluble contaminants) are minimized by collecting bivalves from a given region at the same time each year.

The NS&T Mussel Watch Project has collected data on bivalve contaminant levels in most major coastal areas of the United States annually since 1986. A broad, synoptic summary of the majority of these findings has been presented elsewhere (NOAA, 1989; O'Connor, 1990). In this paper, we present the results of analyses of selected trace organic contaminants in the blue mussel *Mytilus edulis* from the Hudson-Raritan Estuary and Long Island Sound.

1.1. The study areas

The Hudson-Raritan Estuary and Long Island Sound are two major bodies of water that border on metropolitan New York, northeastern New Jersey, and southeastern Connecticut — the most populated urban area on the East Coast of the United States. The Hudson-Raritan Estuary is fed primarily by the Hudson River, but also receives input from the Raritan, Hackensack, and Passaic rivers (Steimle and Caracciolo-Ward, 1989). These rivers drain the lower New York State area and much of northeastern New Jersey. The Long Island Sound is fed by numerous rivers, and drains a large basin of more than 16 000 square miles, including all of Connecticut, parts of Massachusetts, New Hampshire, Vermont, and in New York parts of Westchester County, Suffolk County, and New York City (EPA, 1990). Recent hydrographic evidence suggests that the Long Island Sound also receives input from the New York Bight, with water parcels entering the mouth of the sound after traveling from the

southeast along Long Island (Wong, 1990). In addition to river contributions, both the Hudson-Raritan Estuary and Long Island Sound receive significant input from nonpoint runoff and combined sewer overflow (CSO) (Mueller et al., 1982; EPA, 1989). The environmental status of these two bodies of water, bordered by densely populated urban areas and industrial facilities, is of key concern to local and Federal environmental regulators. Fig. 1 presents the study area with the NS&T Mussel Watch collection sites noted.

1.2. Organic compounds monitored

Organic contaminants measured in the Mussel Watch Project include selected polynuclear aromatic hydrocarbons (PAH), selected polychlorinated biphenyl (PCB) congeners, the pesticide DDT and its degradation products, and the marine paint antifouling agent tributyltin (TBT) and its degradation products (Table 1).

The PAH are relatively refractory organic compounds that are components of raw and refined petroleum products, and of combustion-related waste. These compounds are ubiquitous contaminants in marine sediments and organisms (Brown and Weiss, 1978; Hites et al., 1980; Takada et al., 1990). PAH are excellent indicators of, for example, contamination from petroleum spills, fugitive petroleum, creosote leachate, atmospheric input, and urban runoff of soot from combustion sources. PCB have been recognized as pervasive environmental contaminants for over 25 years (Cairns et al., 1986; Alford-Stevens, 1986). In the United States, PCB have been introduced into the environment as commercial mixtures of Aroclors. The mixtures have a wide variety of uses as heat-transfer fluids, solvent extenders, flame retardants, and dielectric fluids (Mullin et al., 1984). The compound DDT (*p,p'*-dichlorodiphenyltrichloroethane) was patented in 1942, and was a widely used agricultural, commercial and residential pesticide. From the time of its patenting through the 1960s, DDT was the dominant insecticide used in the United States. Technical grade DDT normally consists of about 70% 4,4'-DDT, 20% 2,4'-DDT, and the remainder small amounts of 4,4'-DDD, 2,4'-DDD, 4,4'-DDE and 2,4'-DDE (Stetter, 1983). Although banned in the United

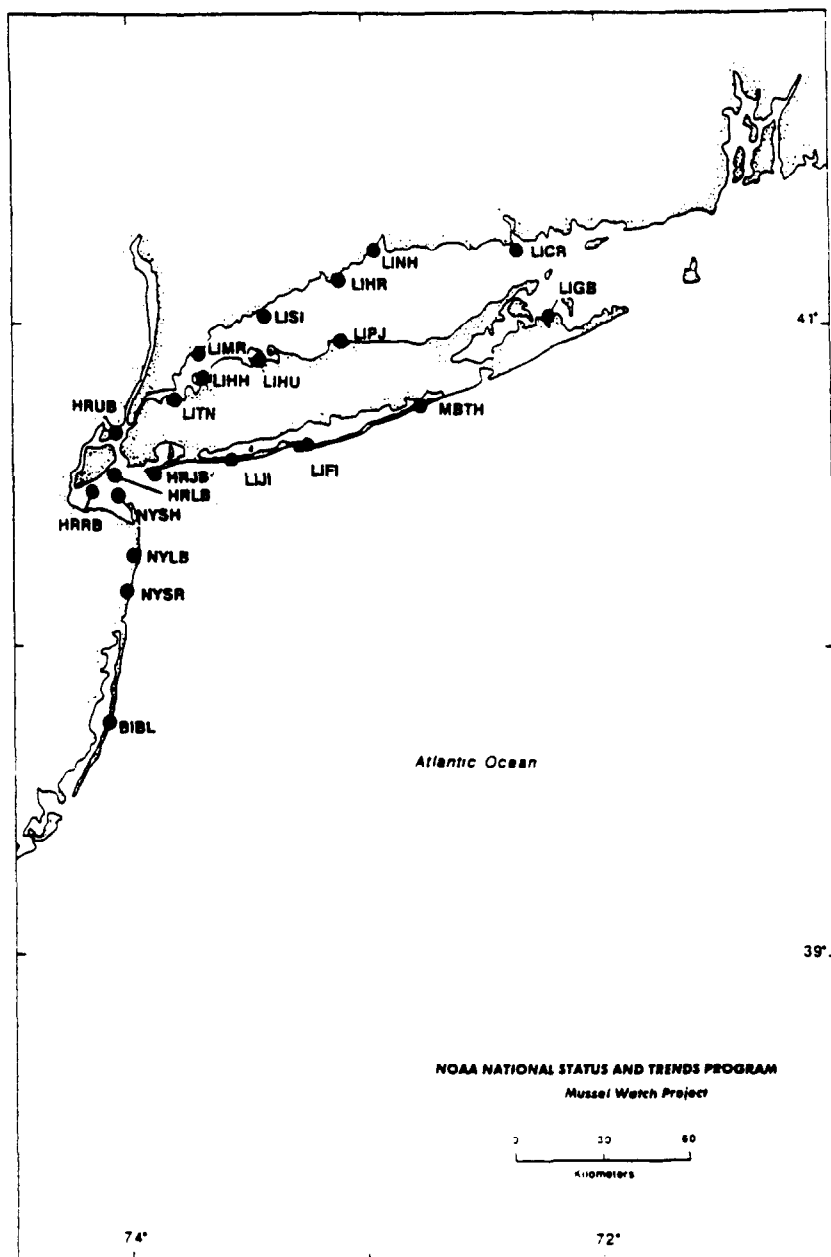


Fig. 1. NS&T Mussel Watch Project sites in Long Island Sound and the Hudson-Raritan Estuary.

States and Canada in 1972, DDT and its breakdown products are still present in the environment (Sericano et al., 1990). Tributyltin (TBT) has been used extensively for almost 30 years in marine paint formulations to prevent the accumulation of barnacles and slime on boat hulls. TBT is in the water, sediment and biota near harbors,

marinas, and drydocks (Seligman et al., 1986; Unger et al., 1986; Wade et al., 1988; Uhler et al., 1989, 1993). Acute and chronic toxicity of TBT at very low aqueous levels (less than 20 ng/l) prompted a partial Federal ban on TBT in 1988 in an attempt to lower average ambient aqueous TBT levels to less than 10 ng/l. Although sus-

Table 1
Organic contaminants and auxiliary parameters measured in the NS&T Mussel Watch Project

Polynuclear aromatic hydrocarbons	Polychlorinated biphenyls
Naphthalene (N)	2,4-Cl ₂ (8)
2-Methylnaphthalene (2MN)	2,2',5-Cl ₃ (18)
1-Methylnaphthalene (1MN)	2,4,4'-Cl ₃ (28)
Biphenyl (B)	2,2',3,5'-Cl ₄ (44)
2,6-Dimethylnaphthalene (DMN)	2,2',5,5'-Cl ₄ (52)
Acenaphthylene (AC)	2,3',4,4'-Cl ₄ (66)
Acenaphthene (ACN)	3,3',4,4'-Cl ₄ (77)
1,6,7-Trimethylnaphthalene (TNM)	2,2',4,5,5'-Cl ₅ (101)
Fluorene (F)	2,3,3',4,4'-Cl ₅ (105)
Phenanthrene (PH)	2,3',4,4',5-Cl ₅ (118)
Anthracene (AN)	3,3',4,4',5-Cl ₅ (126)
1-Methylphenanthrene (1MP)	2,2',3,3',4,4'-Cl ₆ (128)
Fluoranthene (FL)	2,2',3,4,4',5'-Cl ₆ (138)
Pyrene (PY)	2,2',4,4',5,5'-Cl ₆ (153)
Benz[<i>a</i>]anthracene (BAN)	2,2',3,3',4,4',5-Cl ₇ (170)
Chrysene (CY)	2,2',3,4,4',5,5'-Cl ₇ (180)
Benzo[<i>b</i>]fluoranthene (BBF)	2,2',3,4,5,5',6-Cl ₇ (187)
Benzo[<i>k</i>]fluoranthene (BKF)	2,2',3,3',4,4',5,6-Cl ₈ (195)
Benzo[<i>e</i>]pyrene (BEP)	2,2',3,3',4,4',5,5',6-Cl ₉ (206)
Benzo[<i>a</i>]pyrene (BAP)	Decachlorobiphenyl-Cl ₁₀ (209)
Perylene (PER)	Organotins
Indeno[1,2,3- <i>c,d</i>]pyrene (IN)	Tributyltin
Dibenz[<i>a,h</i>]anthracene (DBA)	Dibutyltin
Benzo[<i>g,h,i</i>]perylene (BP)	Monobutyltin
Pesticides	Tetrabutyltin
Hexachlorobenzene	Auxiliary parameters
Lindane	
Heptachlor	Gonadal index (selected (bivalves))
Aldrin	Lipid content
Heptachlorepoide	Dry weight
<i>cis</i> -Chlordane	Shell size
<i>trans</i> -Nonachlor	
Dieldrin	
Endrin	
Mirex	
2,4'-DDD	
4,4'-DDD	
2,4'-DDE	
4,4'-DDE	
2,4'-DDT	
4,4'-DDT	

pected to be a relatively short-lived contaminant, determination of TBT in bivalve tissue will provide one measure of the effectiveness of recent

legislation to limit input of the compound to the marine environment.

2. Experimental

2.1. Field methods

The bivalve species *Mytilus edulis* (blue mussel) was collected at all sites shown in Fig. 1 in Long Island Sound and the Hudson-Raritan Estuary during November and December, 1989. Site-center positions were determined and are verified using multiple methods, including electronic navigation with Loran-C, visual positioning using a compass or sextant to record relative bearings to charted landmarks or aids to navigation, and/or photo-documentation.

Samples from three stations within each site were obtained, with each station sample representing a replicate sample from that site. A minimum of 65 mussels were collected at each station: 30 for organic analysis, 30 for trace element analysis, and 5 for gonadal index determination. Collection-station locations were always within a 400-m radius or 100-m linear transect of the documented site center. Depending on the station depth and environmental conditions at a site, one of several collecting techniques was employed to obtain bivalves. The majority of bivalves were collected by hand or using a stainless steel skip dredge. Polyethylene gloves were worn while handling bivalves, and noncontaminating techniques were used for all sample handling and processing. Bivalves intended for organic and trace element analyses were rinsed with site water and placed on dry ice as soon as possible after sampling and processing. Mussels intended for gonadal index determination and histopathological examination were opened, placed in a jar containing Dietrich's preservative, and returned to the laboratory where they were transferred to 80% ethanol and stored at ambient temperature until processed for histological examination.

2.2. Bivalve tissue processing and analysis for PAH, pesticides, and PCB

Bivalve tissue was processed and analyzed according to methods presented in Peven and Uhler

(1993). Briefly, bivalves from each station were sized, shucked, and homogenized in preparation for sample processing. An aliquot of approximately 30 g (wet wt.) was taken for organic compound analysis. Simultaneously, a 5-g aliquot of the homogenate was removed for percent dry-weight determination. With each batch of 12–15 field samples, one procedural blank sample and one control material (National Institute of Standards and Technology Standard Reference Material 1974, a matrix spike sample, or an in-house control sample) were extracted and analyzed to act as quality control samples. Each sample was spiked with surrogate internal standards (SIS) and serially extracted three times with a Tekmar Tisumizer, using dichloromethane as the extraction solvent. The extract was passed through an alumina cleanup column and concentrated. The concentrated extract was fractionated by size exclusion high-performance liquid chromatography (HPLC). The final extract was concentrated, spiked with recovery internal standards (RIS), and analyzed by gas chromatography with mass

spectrometry (GC/MS) for PAH, and gas chromatography with ^{63}Ni electron capture detection (GC/ECD) for chlorinated hydrocarbons.

2.3. Bivalve tissue processing and analysis for butyltins

The measurements of tributyltin (TBT) and its degradation products dibutyltin (DBT) and monobutyltin (MBT), as well as the TBT-manufacturing impurity tetrabutyltin (TTBT) in bivalve tissues were carried out following methods described by Uhler et al. (1989) and Peven and Uhler (1993). Tissue composites were extracted with hexane and the chelating agent tropolone, the extracted butyltins converted to inert n-hexyl derivatives, and analyzed by gas chromatography with flame photometric detection.

3. Results

The chemical contaminant burdens determined in the tissues of bivalves from this study are presented in Table 2.

Table 2

Summary of organic contaminants in mussels from Long Island Sound and the Hudson-Raritan Estuary

Site name (NOAA code)	Total DDT (ng/g)	Total PCB (ng/g)	Total PAH (ng/g)	TBT (ng/g)
Long Island Sound, Connecticut River (LICR)	60.3	238	935	217
Long Island Sound, New Haven (LINH)	69.5	315	1020	159
Long Island Sound, Housatonic River (LIHR)	81.3	374	859	0
Long Island Sound, Sheffield Island (LISI)	64.9	213	466	456
Long Island Sound, Mamaroneck (LIMR)	145	329	1040	725
Long Island Sound, Throgs Neck (LITN)	222	730	2160	380
Long Island Sound, Hempstead Harbor (LIHH)	73.9	213	613	417
Long Island Sound, Huntington Harbor (LIHU)	68.8	202	302	635
Long Island Sound, Port Jefferson (LIPJ)	58.5	167	399	1130
Long Island Sound, Gardiners Bay (LIGB)	28.6	42.2	295	72
Long Island Sound, Jones Inlet (LIJI)	33.8	118	449	81
Long Island Sound, Fire Island (LIFI)	50.7	188	806	199
Hudson-Raritan Estuary, Jamaica Bay (HRJB)	172.5	525	2610	504
Hudson-Raritan Estuary, Upper Bay (HRUB)	456	1980	6380	1910
Hudson-Raritan Estuary, Lower Bay (HRLB)	316	1030	2640	963
Hudson-Raritan Estuary, Raritan Bay (HRRB)	202	582	1100	0
New York Bight, Sandy Hook (NYSH)	231	922	1700	897
New York Bight, Long Branch (NYLB)	103	407	695	143
New York Bight, Shark River (NYSR)	102	426	1380	492
Barnegat Inlet, Barnegat Light (BIBL)	46.1	184	290	94

3.1. PAH

The highest total PAH (sum of the 24 PAH in Table 1) concentrations in the Hudson-Raritan Estuary were found in the Upper Bay (HRUB) and Lower Bay (HRLB) of the Hudson River, and near the mouth of the estuary in Jamaica Bay (HRJB). Generally, a decreasing trend in total PAH concentration was found moving south from the Upper Bay to sites in the New York Bight in southern New Jersey. Total PAH concentrations also declined moving east out of the estuary along the southwestern shore of Long Island. The highest total PAH concentrations in Long Island Sound mussels were found at LITN (2155 ng/g) and LIMR (1041 ng/g). Generally, these levels are four- to fivefold lower than the highest concentrations found in the Hudson-Raritan Estuary.

Previous studies have suggested that the PAH in the New York region are derived from urban runoff, sewage treatment plant effluents, CSOs, oil spillage in the vicinity of major shipping areas, dumping of dredge spoils, and atmospheric input (Tanacredi, 1977; Anderson, 1982; Kneip et al.,

1982; O'Connor et al., 1982). The distribution of PAH compounds reflects the source of the material and the degree of physical or biological weathering that has taken place since introduction of the PAH to the marine environment.

The distribution of PAH in mussels at the sites in and near the Hudson-Raritan Estuary are shown in Fig. 3. The composition of PAH at HRUB and HRLB and at Sandy Hook (NYSH) is dominated by the pyrogenic compounds pyrene and fluoranthene, with significant amounts of other 4-, 5-, and 6-ring PAH in a distribution pattern indicative of creosote contamination (Douglas, 1991). This is reasonable, considering the abundance of docks and other structures preserved with creosote in the immediate area of the Upper and Lower Bays. South of the mouth of the estuary, near Shark River (NYSR), the PAH in bivalves are dominated by fluoranthene (4-ring), pyrene (4-ring), and phenanthrene (3-ring), respectively. There are proportionally less 4-, 5- and 6-ring PAH at this site relative to those locations within the estuary. The distribution of PAH at

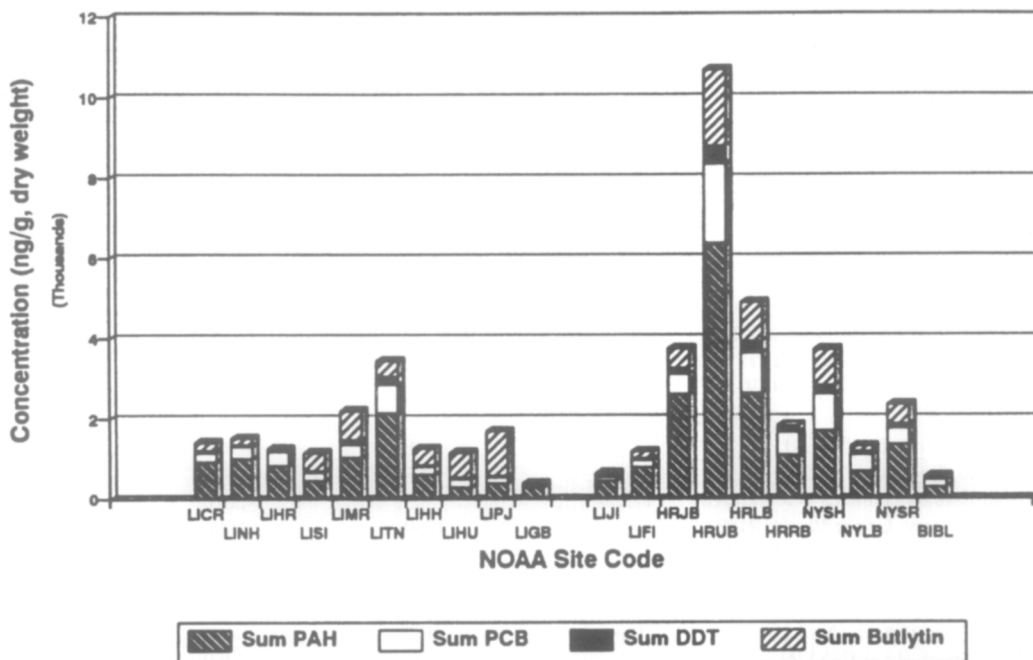


Fig. 2. Relative distribution of organic contaminants in the study area.

NYSR is that normally associated with soot, with little significant input from petroleum-type materials.

Representative PAH distribution in bivalves from Long Island Sound are shown in Fig. 4. In this Figure, one site at the mouth of the Connecticut River (LIAR), one from the poorly flushed Throgs Neck Bridge (LITN) area, one near the mid-point of Long Island (LIPJ), and one near the tip of Long Island (LIGB) are presented. The PAH distributions at LITN and LIAR are quite similar. The PAH are dominated by pyrene and fluoranthene, and show significant amounts of other 4-, 5-, and 6-ring PAH — again indicative of both pyrogenic and creosote- or coal tar-related contamination. The PAH at LISI and LIPJ, dominated by fluoranthene and pyrene, respectively, suggest that soot from combustion processes is the major source of PAH in these areas.

As mentioned earlier, advantages of using bivalve mollusks as indicators of near-shore aquatic contamination include the fact that the mussels

integrate exposure to toxics over time, unlike a water column grab sample, which represents the condition of the sampling location at a given point in time. Using bivalve mollusks to evaluate environmental conditions decreases the likelihood of obtaining a nonrepresentative depiction of average water column contaminant concentrations. However, comparison of bivalve contaminant data to water column data is useful, primarily to evaluate representativeness of bivalve body burdens to water column contaminant concentrations. In this study, water column samples were obtained from the Hudson River Upper Bay area in April 1991 during a survey of ambient concentrations of trace metal and organics in the New York Bight. This sampling provided an opportunity to briefly examine the relationship between PAH concentrations in the water column and nearby bivalve mollusks. It should be noted that there is about a 3-month difference in collection time between the mussels (collected in January) and the water column samples, and average water column PAH composition and concentration may

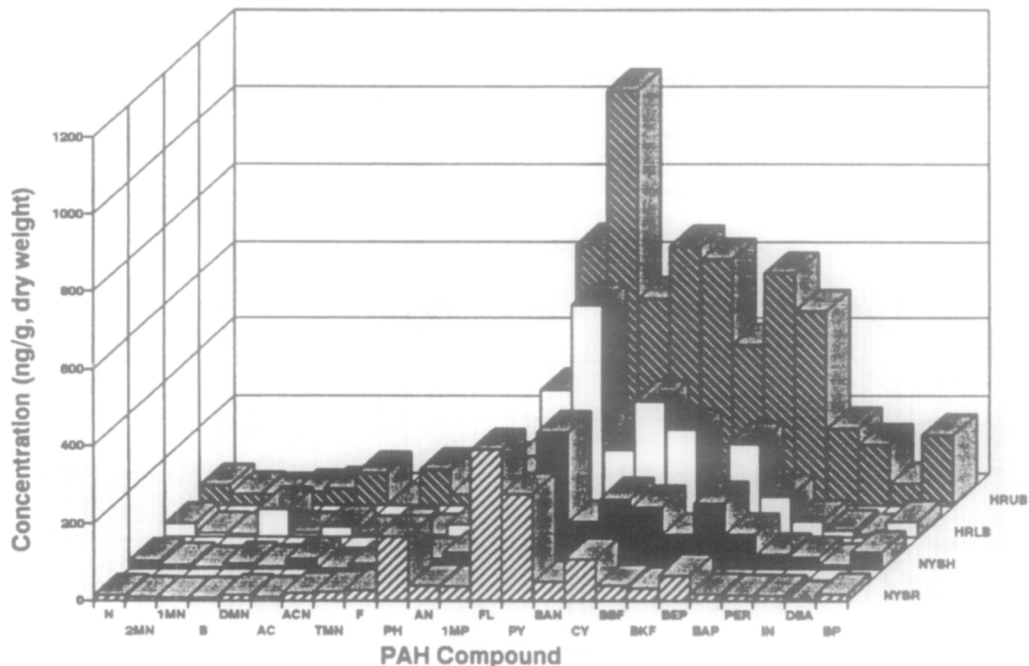


Fig. 3. PAH distributions in mussels from selected sites in Hudson-Raritan Estuary Region.

be different than found for this one sample reported here. None the less, a striking relationship between water column and bivalve tissue PAH concentrations can be seen. Fig. 4 presents a plot of the bivalve PAH concentration versus water PAH concentration as a function of the PAH compounds listed in Table 1. The apparent bioconcentration factor for the 2-, 3-, 4- and most 5-ring PAH increase from about 10^4 for naphthalene to about 10^6 for benzo[*k*]fluoranthene. There is a decrease in the apparent bioconcentration factor for the higher molecular weight 5- and 6-ring PAH. This phenomena has been reported previously for PAH uptake from sediment (Farrington, 1983), the suggestion being that pyrogenic high molecular weight PAH are sorbed tightly to soot or other particles, and were unavailable for biouptake. This bioconcentration data may support the position that high molecular weight PAH — sorbed or dissolved — may be assimilated across biological membranes with difficulty relative to the lower molecular weight PAH. More detailed investigations of in situ

biouptake of contaminants are needed to address this issue fully.

3.2. PCBs

During the period between 1952 and 1977, primarily Aroclor 1242 was released into the upper Hudson River from capacitor manufacturing facilities (Brown et al., 1987; Bush et al., 1987). During the 1970s Aroclor discharges into the Hudson ranged from 0.001 to 15 kg/day (Kneip et al., 1982). Continued release of such material from landfills, dredged materials, atmospheric fallout and contaminated river sediments which continually flow downstream into New York Harbor and into the Bight contribute to the high concentrations of PCB currently present in these areas (O'Connor et al., 1982; Rapaport and Eisenreich, 1988; Chevreuil et al., 1989).

The highest total PCB concentration (sum of the 20 target congeners) in the Hudson-Raritan Estuary was found in mussels from the Upper Bay (HRUB: 1981 ng/g) and Lower Bay (HRLB: 1033 ng/g) of the Hudson River. Moving east out

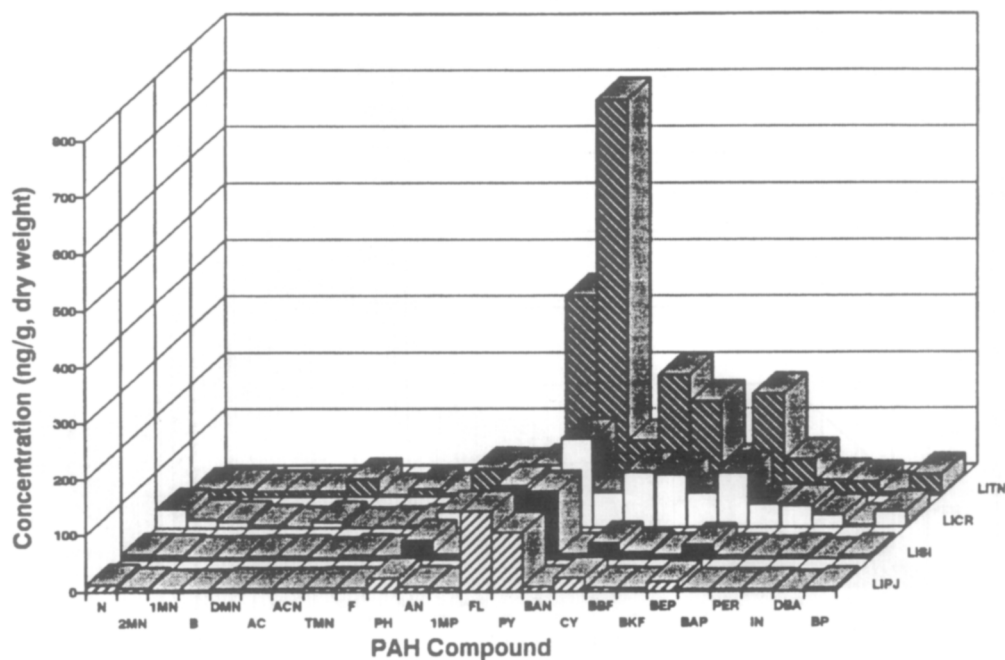


Fig. 4. PAH distributions in mussels from selected sites in Long Island Sound.

of the Bay around Long Island Sound and south to the New Jersey coast, total PCB concentrations generally decreased (Table 2).

The penta- and hexa-chlorobiphenyl isomers generally represented the greatest concentration of the target PCBs in the New York area. This trend is also true for most contaminated urban/industrial areas. In the mussels, hexachlorobiphenyls constituted more than 30% of the total PCB concentration (among the 20 congeners of interest). Fig. 5 graphically depicts the distribution of congeners at selected sites. The distribution of the levels of chlorination were typically hexachlorobiphenyls > pentachlorobiphenyls ≥ tetrachlorobiphenyls. However, the relative ratio of the levels of chlorination changes between sites. At those sites with the highest concentration of PCBs (and closest to the source of the contamination upriver), the relative concentration of hexachlorobiphenyls as compared with tetra- and penta-chlorobiphenyls differed by less than 10%. For example, at the Hudson-Raritan, Upper Bay site (HRUB), 25% of the total concentration is made up of tetrachlorobiphenyl

isomers, 25% pentachlorobiphenyl isomers, and 33% hexachlorobiphenyls.

Moving away from that central area, the relative concentration of hexachlorobiphenyls increases as compared with tetra- and penta-isomers. At the Long Island Fire Island (LIFI) site tetrachlorobiphenyls comprise 12% of the total concentration, pentachlorobiphenyls 29%, and hexachlorobiphenyls 45%. Additionally, the concentration of trichlorobiphenyl isomers constitutes a greater percentage of the total PCB concentration in the mussel samples from sites closest to the Hudson-Raritan sites. There are several possible explanations for this relative increase in the concentration of the lower chlorinated congeners. The composition of PCBs in the environment changes over time through various pathways, including selective evaporation and dissolution, possible bioaccumulation and degradation of selected congeners by marine organisms, and the mixing of different Aroclors from different sources. Brown et al. (1987) suggest that environmental dechlorination by enzymes associated with anaerobic organisms and/or solar photolysis

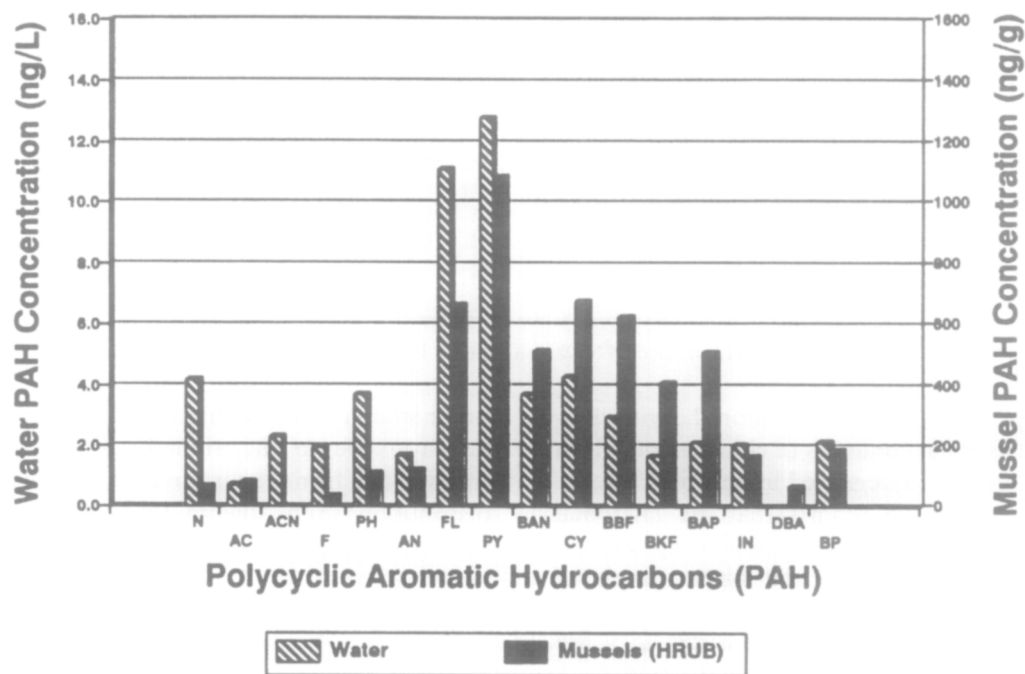


Fig. 5. Concentrations of individual PAH in water and mussel samples from the Hudson-Raritan Estuary.

Hudson River, sewage sludge, atmospheric input, and discharged dredge spoils (O'Connor et al., 1982; Rapaport et al., 1985). DDT concentrations in bivalves from the study area are presented in Table 2, and are reported as the sum of parent DDT and its metabolites (4,4'-DDT + 2,4'-DDT + 4,4'-DDD + 2,4'-DDD + 4,4'-DDE + 2,4'-DDE). The highest concentrations of total DDT were found in mussels from the Hudson-Raritan, Upper Bay and Lower Bay sites, where the concentrations were 456 ng/g and 316 ng/g, respectively, the highest concentrations of DDT detected on the East Coast. As with the PAH and PCB, the DDT concentrations decrease moving away from New York Harbor. Because of its breakdown kinetics, at least 50% of the total DDT concentration should consist of DDD or DDE if the pesticide was introduced into the environment before 1972 (Rapaport et al., 1985). On the average, 87% of the total DDT concentration is made up of DDD and DDE at these sites, suggesting little recent input of the pesticide.

3.4. Butyltins

The major source of TBT to the marine environment are boats docked in marinas or under repair in drydock facilities, and ships plying busy shipping channels and harbors (Maguire, 1982; Waldock and Miller, 1983). High levels of TBT and its degradation products in the marine environment are generally localized to these types of environments. The concentration of total butyltins (TBT + DBT + MBT) in the Hudson-Raritan region is highest in the Upper Bay (HRUB: 1910 ng/g), with a systematic decrease with distance from that site. This is consistent with the general observation that the highest concentration of vessels, both moored and under steam, are found in the Lower and Upper Bay regions. Additionally, the body of water comprising the Hudson-Raritan harbor estuary is better flushed farther out into the Atlantic, resulting in diluted butyltins and lower exposures to bivalves.

Total butyltins in Long Island Sound vary unsystematically, reflecting the strong point-source nature of the contamination. The highest concentration of butyltins in Long Island Sound was found at LIPJ (1130 ng/g), while the lowest was found at LIGB, the outermost site in Long Island

Sound. Unlike the Hudson-Raritan which is utilized regularly by commercial traffic, the majority of the boat traffic in Long Island Sound is recreational. The levels of butyltins found in Long Island Sound bivalves correlate strongly with proximity to recreational boating marinas, rather than showing a systematic geographic trend in concentrations as is seen in the Hudson-Raritan.

The ratio of TBT to its principal degradation product DBT in bivalve tissues is concentration-independent — approximately 2.5 for the bivalves collected both from the Hudson-Raritan and Long Island Sound. This is consistent with a reported average of 2.6 for *Mytilus edulis* from a national survey of butyltin contamination in bivalves (Uhler et al., 1989).

4. Discussion and conclusions

PAH, PCB, chlorinated pesticide, and butyltin contamination was evident in all bivalve mollusks sampled from both the Hudson-Raritan Estuary and Long Island Sound. The concentrations of all target analytes were highest in mussels from the Hudson Raritan Estuary Upper Bay site, the area in closest proximity to industrial and municipal discharges. It is evident that the anthropogenic activities around the Bay and along the shores of the Hudson River affect the contaminant loading in the municipal New York/New Jersey region. There is an apparent gradient effect taking place as one moves south from the mouth of the Hudson River area and eastward around Long Island. This gradient is evidenced by the changes in concentration and distribution of analytes moving away from the Upper Bay. Long Island Sound is more diverse in terms of many of the concentrations measured due to the distinct multiple point-sources of the contamination.

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