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Chemical contamination of the Hudson-Raritan Estuary as a result of the attack on the World Trade Center: Analysis of polycyclic aromatic hydrocarbons and polychlorinated biphenyls in mussels and sediment

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Abstract

The September 11, 2001 attack on the World Trade Center (WTC) resulted in a massive plume of dust and smoke that blanketed lower Manhattan and part of the Hudson-Raritan Estuary (HRE). The NOAA National Status and Trends Mussel Watch Program has long-term monitoring sites in the area and thus had an opportunity to assess the effect of the WTC attack on PAH and PCB contamination of the surrounding estuary. Seven additional sites were added in the Upper HRE to attain higher sampling resolution for comparison with regularly sampled Mussel Watch Project HRE sites. Elevated background levels of PCBs and PAHs in mussel tissue and sediments were high enough before the WTC attack that concentrations were not measurably changed by WTC derived contaminant input.

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Keywords: World Trade Center; Mytilus edulis; Sediments; PAHs; PCBs and Hudson-Raritan Estuary

1. Introduction

The National Oceanic and Atmospheric Administration's Mussel Watch Project (MWP) was established in 1986 to assess ambient contamination in coastal waters nationally and to determine temporal trends for contaminants of concern. Five of the 280 established core MWP monitoring sites are located in the Hudson-Raritan Estuary (HRE) where on September 11, 2001 the World Trade Center (WTC) attack resulted in the dispersion of building material and pollutants over lower Manhattan (Table 1; Fig. 1). The initial plume was dispersed in all directions from ground zero, and later to many outdoor and indoor locations downwind towards Brooklyn (Offenberg et al., 2003). The fine dust released by the WTC collapse contained asbestos, crushed cement, and organic contaminants such as dioxin and PAHs (Offenberg et al., 2003; Butt et al., 2004). Studies found that concentrations of collapse and combustion related contaminants were distributed in a plume, and that concentrations of organic contaminants decreased with distance from ground zero (Butt et al., 2004). Litten et al. (2003) assessed several contaminants including PCBs in runoff, sediment, and sewage sludge, and concluded the WTC disaster did not significantly impact ambient concentrations of target chemicals in New York Harbor.

The Mussel Watch Project, in contrast to other post WTC attack assessments, gives a unique temporal perspective of the attacks by comparing measurements taken in the same time frame as the WTC attacks to measurements taken before and after the attacks. Mussels, used as sentinel organisms by the Mussel Watch Project, are filter feeders

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Table 1

Location of Mussel Watch monitoring sites collected in the Hudson-Raritan Estuary (HRE) to assess contaminant input as a result of the attack on the WTC

Site location	Site	Matrix		Latitude	Longitude
		2002	2004		
Core monitoring sites					
HRE Jamaica Bay, NY	HRJB	M,S	М	40.576°	-73.895°
HRE Upper Bay, NY (Liberty Is.)	HRUB	M,S	М	40.689°	-74.043°
HRE Lower Bay, NY (Swinburne Is.)	HRLB	M,S	М	40.566°	-74.051°
HRE Raritan Bay, NY (Staten Is.)	HRRB	M,S	М	40.519°	-74.185°
Sandy Hook, NJ	NYSH	M,S	М	40.488°	-74.033°
Special WTC monitoring	sites				
HR Holland Tunnel, NY	HRHT	S		40.726°	-74.015°
HR World Trade Center, NY	HRWT	S		40.719°	-74.015°
HR Battery Park, NY	HRBP	Μ	Μ	40.705°	-74.018°
HRE Governors Island, NY	HRGI		М	40.693°	-74.019°
HRE Ellis Island, NY	HREI	M,S		40.699°	-74.043°
HRE Shore Road, NY	HRSR	М	Μ	40.608°	-74.035°
HRE Fort Wadsworth, NY	HRFW	М	М	40.615°	-74.061°

M = Mytilus edulis; S = sediment; this matrix information is applicable to samples collected during the 2002 and 2004 sampling efforts. More detailed information about core sites and data can be found at http:// ccma.nos.noaa.gov/cit/data/welcome.html.



Fig. 1. Shown are the locations of the five (core) Mussel Watch Project monitoring sites (squares) and the seven special World Trade Center (WTC) sampling sites (circles) within the Hudson-Raritan Estuary (HRE). Core sites have been sampled for more than a decade as part of the Mussel Watch Project. Special WTC sampling sites were sampled in 2002 and 2004 to enhance spatial resolution.

that provide an integrated biotic perspective to WTC attack contamination assessment. This information adds to the body of knowledge that can be used to mitigate and assess catastrophic events.

Due to their hydrophobic nature, persistent organic contaminants such as PAHs and PCB accumulate in sediments. Surface sediment samples are used by many monitoring and assessment programs to characterize contamination in estuaries and bays (USEPA, 2002, 2004) resulting in spatial characterizations that can be used to identify areas of high contamination in a system and compare sediment concentrations nationally. Surface sediments integrate several years of contaminant input. For this reason MWP does not collect sediments as often as tissue samples. The national perspective provided by sediments is a valuable component of MWP because it gives a complete national perspective of contamination that compliments the mussel and oyster data that are limited by the extent of organism distribution.

Due to urban sources (LaFlamme and Hites, 1978; Cundy et al., 1997) and intense industrialization, the HRE has had elevated contaminant levels for decades. Before the collapse of the WTC, levels of contamination in the HRE were among the highest in the country (Ayers and Rod, 1986). Input of PAHs and PCBs from the WTC attacks were characterized using MWP measurements from before and after the attacks (core sites), and samples collected in the Upper Bay after the attack (special collection sites). Additionally, PAH fingerprinting was done to assess any source changes that may have occurred for PAHs as a result of the WTC attacks.

2. Methods

The mussel species used for historic comparison and in the special 2002 WTC sampling was *Mytilus edulis*. Mussels were also collected a second time at the same sites in 2004. Special WTC samples were taken in the field using MWP methods to allow for comparison with established regional and national temporal contaminant trends. During the early part of the MWP (1986–1992) collections took place annually; however, since 1992, bivalve sampling has occurred on a biennial basis. Bivalve mollusks were collected nationally in the winter months, except in the Great Lakes where the samples were collected during September. Winter sampling reduced the potential effect that spawning has on the organism's body burdens of contaminants. Repeat site sampling occurred within 3 weeks of the prescribed sampling date (Lauenstein et al., 1997).

The time frames for the majority of MWP sediment site collections nationwide were 1986–1987 and 1996–1997. When the special 2002 WTC bivalve samples in the HRE were collected, sediments were also sampled. Only sediments containing at least 20% fine grained material (silt and clay) were analyzed for contaminants. A discussion of the effect of grain size on sediment contaminant levels can be found in O'Connor (1996a). At sites where no bivalves could be found, only fine-grained sediments were collected (Table 1).

Sediment samples were generally limited to recent deposition. This was achieved by avoiding areas of wave and tidal scouring, and by limiting the sample to the top 2 cm. Nationwide collection of sediment samples makes it possible to create a national ranking of sites that was used to identify areas with elevated sediment concentrations. In addition, sediments were used in the HRE to compare the Upper and Lower Bays. Because sediment samples were not collected biennially like bivalves, they were not used to assess the before and after effects of the WTC attacks on contaminant input to the HRE.

Mussels were shucked with only the soft tissues analyzed for contaminants. Approximately 30 mussels were composited for each analysis. Additional information on the standardized field work methodologies can be found in Lauenstein and Cantillo (1993a). Detailed methods information for the majority of the chemical analyses performed in the MWP are reported in NS&T methods documents (Lauenstein and Cantillo, 1993a,b,c,d, 1998).

The PAHs and PCBs monitored at MWP sites around the US are shown in Table 2. In order to discuss the groups of contaminants, concentrations were summed such that total polycyclic aromatic hydrocarbons (total PAH) is the

> Years Tissue

1988-

2004 1989–

2004

Sediment

2002

2002

Table 2

Chemicals quantified in Mytilus edulis and sediment in the Hudson-Raritan Estuary				
Compound class				
Total PCBs	PCB8, PCB18, PCB28, PCB44, PCB52, PCB66, PCB101, PCB105, PCB118, PCB128, PCB138, PCB153, PCB170, PCB180, PCB187, PCB195, PCB206, PCB209			
Total PAHs	naphthalene; 2-methylnaphthalene; 1-methylnaphthalene; biphenyl; 2,6-dimethylnaphthalene; acenaphthene; acenaphthylene; 1,6,7-trimethylnaphthalene; fluorene; phenanthrene; 1-methylphenanthrene; anthracene;			

	benzo[e]pyrene, benzo[a]pyrene, perylene, dibenz[a,h]anthracene, indeno[1,2,3-cd]pyrene, benzo[ghi]perylene	
Fingerprint	biphenyl, naphthalene, C1-naphthalenes, C2-naphthalenes, C3-naphthalenes, C4-naphthalenes,	1995–
PAHs	acenaphthylene, acenaphthene, fluorene, C1-fluorenes, anthracene, phenanthrene, C1-phenanthrenes/	2004
	anthracenes, C2-phenanthrenes/anthracenes, C3-phenanthrenes/anthracenes, C4-phenanthrenes/	
	anthracenes, pyrene, fluoranthene, C1-fluoranthenes/pyrenes, benz[a]anthracene, chrysene, C1-chrysenes,	
	C2-chrysenes, C3-chrysenes, C4-chrysenes, $benzo[b]$ fluoranthene, $benzo[k]$ fluoranthene, $benzo[e]$ pyrene,	
	benzo[a]pyrene, dibenzo[a,h]anthracene, indeno[1,2,3-c,d]pyrene, benzo[g,h,i]perylene	
	benzo[a]pyrene, dibenzo[a,h]anthracene, indeno[1,2,3-c,d]pyrene, benzo[g,h,i]perylene	

fluoranthene, pyrene, benz[a]anthracene, chrysene, benz[b]fluoranthene, benz[k]fluoranthene,



Fig. 2. Hudson-Raritan Estuary (HRE) sediment characterization from the 2002 World Trade Center sampling. There is no significant difference between fines (clay and silt) percentage in the Upper and Lower HRE. Sediment total organic carbon varies by less than 4% in 2002 HRE samples. Legend divisions represent quantiles.

sum of 24 PAHs; total PCBs represents the sum of 18 PCBs analyzed (Table 2). Polycyclic aromatic hydrocarbons, unlike the other organics quantified in the MWP, can occur naturally, and can also enter the environment as a result of human activities such as the combustion of fossil fuels or the release of petroleum products (LaFlamme and Hites, 1978). Fingerprinting, a comparison of compounds with varying ring sizes and substitutions, was used to distinguish between petrogenic and pyrogenic sources (Kimbrough and Dickhut, 2006).

Sample preparation and extraction techniques were the same as those documented in Sericano et al. (1990) and Lauenstein and Cantillo (1998). Bivalve concentrations were reported on a dry weight basis. The core suite of MWP organic contaminants (Table 2) were reported at the parts per billion level (ng/g dry wt.).

Five long-term core sites (1989–2004) were used to characterize historical PAH and PCB contamination in the HRE. The Liberty Island, Upper Bay site is located in the Upper Estuary, where the WTC attack occurred and was used as a proxy for historic characterization of the immediate area. All of the special WTC sites were located in the Upper Bay and were compared to historic contaminant concentrations in order to determine if contaminant input magnitude or sources changed as a result of the collapse of the WTC (Fig. 1). The remaining core sites which were not in the Upper Bay (Swinburne Island, Staten Island, Sandy Hook and Jamaica Bay) represent dispersal pathways and are used for comparison purposes (Fig. 1).

Pyrogenic and petrogenic sources of PAHs can be distinguished from one another by comparing the proportion of high and low molecular weight compounds and by



Fig. 3. Total PAH (A) and total PCB (B) concentrations in sediments (2002). Legend divisions represent quantiles.

comparing unsubstituted (parent) to substituted (alkylated) compounds (Wang and Fingas, 2003; Page et al., 1996). Specifically, petrogenically derived PAHs have a higher proportion of substituted and low molecular weight (LMW) PAHs (Neff, 1979; Page et al., 1999).

The nonparametric Spearman's rank correlation test was used to test for PCB and PAH temporal trends. Specifically, it was used to determine if bivalve tissue concentra-

Table 3

Most long-term core Mussel Watch sites in the Hudson-Raritan Estuary have no distinguishable total PAH temporal trend

Total PAH Spearman's correlation test				
Site	Spearman ρ	Probability	Year range	
HRUB	-0.27	0.39	1989-2004	
HRLB	-0.72	0.03	1989-2004	
HRRB	0.00	1.00	1990-2004	
NYSH	-0.37	0.29	1989-2004	
HRJB	-0.27	0.39	1989-2004	

Only one site, Swinburne Island (HRLB), exhibited a distinguishable temporal trend, that was decreasing.

tions have been increasing, decreasing or exhibiting no temporal trend at sites with a minimum of ten years of data. A detailed discussion of this method and previous MWP applications was published by O'Connor (1996b). The nonparametric Wilcoxon statistical test was used to assess potential differences among years of data and between the Upper and Lower Bays. Nonparametric statistical tests were used as MWP data are not normally distributed.

3. Results and discussion

3.1. Sediment characterization

Organic contaminant concentrations reported for sediments were not grain size normalized. Sediments that contained less than 20% fines were not analyzed because of the relatively small surface area associated with larger particles and the resulting lower contaminant concentrations (O'Connor, 1996a). The smaller relative surface area provides fewer binding sites for lipophilic contaminants. Fines



Fig. 4. Total PAH tissue concentration for core sites. Line represents the national biennial 75th percentile for *Mytilus edulis*. The three year period 2002–2004 utilizes 2002–2003 75th percentile.

is defined as the sediment fraction containing silt and clay with the remaining sediment fractions characterized as sand and gravel. All 2002 sediment data from the special sampling contained at least 35% fines (Fig. 2A–D). No significant difference was found between the Lower and Upper Estuary fines ($\chi^2 = 3.0$, $\rho = 0.08$) or total organic carbon ($\chi^2 = 2.1$, $\rho = 0.15$) as a percent of total sediment using the Wilcoxon statistical test.

3.2. Total polycyclic aromatic hydrocarbons in sediment

Sediment samples were used to compare the Upper and Lower Bays and to characterize concentrations relative to national MWP sediment concentrations (Fig. 3A). All HRE total PAH sediment concentrations exceeded the national 75th percentile of 2,100 ng/g dry wt. that was derived from the MWP nationwide assessment in 1996-1997. The highest concentration for total PAHs in sediments in the Estuary was reported for the Liberty Island site in 1989 at over 47,000 ng/g dry weight. The second highest reported concentration (over 25,000 ng/g dry wt.) was found at the World Trade Center site during the 2002 WTC special sampling. Sediment PAH concentrations were significantly higher in the Upper Bay relative to the Lower Bay ($\chi^2 = 5.3$, $\rho = 0.02$). It is difficult to conclude that these high concentrations found in the HRE are solely the result of the attack on the WTC because HRE total PAH concentrations have always been among the highest concentrations nationally.

3.3. Total polychlorinated biphenyl in sediment

Upper Bay sediment total PCB concentrations were significantly higher than Lower Bay sites ($\chi^2 = 10$; $\rho = 0.01$). The elevated total PCB concentrations in the HRE have been documented (Achman et al., 1996; Wolfe et al., 1996). The HRE total PCB measurements from 2002 all exceeded the 75th percentile for the most recent (1996– 1997) national sediment sampling. The elevated sediment concentrations in the Upper Bay is thought to be part of a historical pattern and not the result of the WTC attack (Fig. 3B).

3.4. Temporal polycyclic aromatic hydrocarbon characterization using mussels

Measurements from all HRE core sites were used to assess long-term contaminant tends. All sites except one, Swinburne Island, showed no discernable temporal trend for total PAHs (Table 3), which is similar to total PAH temporal trends found at the national level (O'Connor and Lauenstein, 2006). Urban sources of PAHs include petroleum derived materials, automobile exhaust, coal fired power plants and other sources of incomplete combustion. High interannual variability and inconsistent input result in no discernable temporal trend at most sites (Fig. 4).



Since 1992 national sampling changed from yearly to biennially, two sampling year's data were combined to form national quartiles (Figs. 4 and 5). The years 2004 and 2005 represent one biennial period; however, 2005 was not available during the drafting of this manuscript, hence, the quartile from the 2002 to 2003 period was used for comparison with 2004. Tissue measurements above the 75th percentile were among the highest in the country measured in *M. edulis*. Only three tissue measurements taken at HRE core sites were less than their respective 75th percentile over the past 15 years. All Upper Bay measurements were well above the 75th percentile (Figs. 4 and 5).

A comparison of total PAH in mussels for the years 2001, 2002 and 2004 (core and WTC sites) using the Wilcoxon statistical test found no difference between the years $(\chi^2 = 1.2, \rho = 0.76)$. Input of PAHs from WTC did not result in significantly higher concentrations for the 2002 (WTC special sampling) year relative to the other years. Also, for many of the HRE core sites, the highest total PAH measurements were recorded the year before the WTC attack. In the case of Liberty Island site, the 2001 total PAH measurement was an order of magnitude higher than the 2002 total PAH measurement made shortly after the attacks on the WTC. The Battery Park and Fort Wadsworth sites exhibited high interannual variability. However, most of the WTC measurements were below the median concentration when compared to the Liberty Island samples collected from 1989 to 2004 (Fig. 5). A temporal characterization of the HRE indicated a high level of variability in PAH tissue concentrations, resulting no statistically significant temporal trend.



3.5. Polycyclic aromatic hydrocarbon spatial characterization using mussels

The Upper Bay exhibited higher total PAH tissue concentrations than the Lower Bay. When comparing historical tissue measurements, the only core site located in the Upper Bay, the Liberty Island site, was significantly higher than the Lower Bay Core sites ($\chi^2 = 14$, $\rho < 0.01$). A comparison of 2002–2004 data from Lower and Upper Bays (WTC and core sites included) also indicated that Upper Estuary sites had significantly higher tissue concentrations relative to Lower Bay sites ($\chi^2 = 7.9$, $\rho = 0.01$). When compared to the Liberty Island historic trend, WTC Upper Bay sites are low. However, the higher concentrations found in the Upper Bay are among the highest nationally for *M. edulis*, masking the input of PAHs to the HRE that occurred as a result of the WTC collapse (Figs. 4 and 5).

3.6. Polycyclic aromatic hydrocarbon fingerprinting using mussels

Polycyclic aromatic hydrocarbon fingerprint data from the Liberty Island site (1995–2004) was compared to assess temporal PAH source variability (Fig. 6). In 2001 the highest total PAH measurement at the Liberty Island site coincided with PAH data that deviated from the dominant fingerprint (Fig. 6). Higher proportions of LMW PAHs are evidence that the increase was associated with a petrogenic source such as an oil spill.

Sites from the special 2002 WTC sampling were compared to 1995–2004 Liberty Island site PAH fingerprints



Fig. 6. Normalized concentration (compound/all PAHs by site) plot of parent and substituted compounds for the Liberty Island site. Biphenyl (BIPHENYL), naphthalene (NAPH), C1-naphthalenes (C1NAPH), C2-naphthalenes (C2NAPH), C3-naphthalenes (C3NAPH), C4-naphthalenes (C4NAPH), acenaphthylene (ACENTHY), acenaphthene (ACENTHE), fluorene (FLUORENE), C1-fluorenes (C1FLUOR), anthracene (ANTHRA), phenanthrene (PHENANTH), C1-phenanthrenes/anthracenes (C1PHEN), C2-phenanthrenes/anthracenes (C2PHEN), C3-phenanthrenes/anthracenes (C3PHEN), C4-phenanthrenes/anthracenes (C1PHEN), C2-phenanthrenes/anthracenes (C2PHEN), C3-phenanthrenes/anthracenes (C3PHEN), C4-phenanthrenes/anthracenes (C4PHEN), pyrene (PYRENE), fluoranthene (FLUORANT), C1-fluoranthenes/pyrenes (C1FLUPY), benz[a]anthracene (BENANTH), chrysene (CHRYSENE), C1-chrysenes (C1CHRY), C2-chrysenes (C2CHRY), C3-chrysenes (C3CHRY), C4-chrysenes (C4CHRY), benzo[b]fluoranthene (BENZOBFL), benzo[k]fluoranthene (BENZOKFL), benzo[c]pyrene (BENAPY), dibenzo[a,h]anthracene (DIBENZ), indeno[1,2,3-c,d]pyrene (INDENO) and benzo[g,h,i]perylene (BENZOP) are arrange by increasing molecular weight from left to right. Black bars represent parent compounds and grey bars represent substituted compounds.

(Fig. 7). Shore Road was the only site that deviated from the predominant Upper Bay fingerprint, all other sites had a similar fingerprint to that exhibited by Liberty Island samples taken in 1995, 2002, 2003, and 2004; samples taken from before and after the WTC attacks. Hence, there is little evidence to support the conclusion that WTC attacks had a significant affect on PAH input to the HRE.

3.7. Temporal polychlorinated biphenyl characterization using mussels

All HRE core sites exhibit a decreasing temporal trend for total PCBs. The Staten Island site was the only core site whose decreasing temporal trend was not significant (Table 4). The HRE total PCB temporal trends from 1988 to 2004 are consistent with the overall decreasing temporal trend



Fig. 7. Normalized concentration (compound/all PAHs) plot of parent and substituted compounds for special WTC sampling sites. Biphenyl (BIPHENYL), naphthalene (NAPH), C1-naphthalenes (C1NAPH), C2-naphthalenes (C2NAPH), C3-naphthalenes (C3NAPH), C4-naphthalenes (C4NAPH), acenaphthylene (ACENTHY), acenaphthene (ACENTHE), fluorene (FLUORENE), C1-fluorenes (C1FLUOR), anthracene (ANTHRA), phenanthrene (PHENANTH), C1-phenanthrenes/anthracenes (C1PHEN), C2-phenanthrenes/anthracenes (C2PHEN), C3-phenanthrenes/anthracenes (C3PHEN), C4-phenanthrenes/anthracenes (C1PHEN), C2-phenanthrenes/anthracenes (C2PHEN), C3-phenanthrenes/anthracenes (C3PHEN), C4-phenanthrenes/anthracenes (C4PHEN), pyrene (PYRENE), fluoranthene (FLUORANT), C1-fluoranthenes/pyrenes (C1FLUPY), benz[a]anthracene (BENANTH), chrysene (CHRYSENE), C1-chrysenes (C1CHRY), C2-chrysenes (C2CHRY), C3-chrysenes (C3CHRY), C4-chrysenes (C4CHRY), benzo[b]fluoranthene (BENZOBFL), benzo[k]fluoranthene (BENZOKFL), benzo[c]pyrene (BENAPY), dibenzo[a,h]anthracene (DIBENZ), indeno[1,2,3-c,d]pyrene (INDENO) and benzo[g,h,i]perylene (BENZOP) are arrange by increasing molecular weight from left to right. Black bars represent parent compounds and grey bars represent substituted compounds. Since there was no Governors Island (HRGI) 2004 sample is presented.

Table 4 Most long term core sites in the HRE exhibit decreasing total PCB temporal trends

Total PCB Spearman's correlation test				
Site	Spearman ρ	Probability	Year range	
HRUB	-0.92	< 0.01	1988-2004	
HRLB	-0.85	< 0.01	1988-2004	
HRRB	-0.64	0.12	1990-2004	
NYSH	-0.93	< 0.01	1988-2004	
HRJB	-0.85	< 0.01	1988–2004	

Only one site Staten Island (HRRB) did not have a significant decreasing temporal trend.

that is seen nationwide and that is attributed to the 1979 ban on PCB production in the US (Table 4; Fig. 8).

Fig. 8 is arranged by distance from the WTC attack site, with the Liberty Island site being the closest core site. The sites in or close to the Upper Bay exhibit the highest concentration relative to those sites that are dispersed throughout the Lower Bay. Biennial national 75th percentiles for *M. edulis* were compared to HRE total PCB measurements. None of the sites located closest to the WTC attack site exhibited concentrations below the 75th percentile.



Fig. 9. A comparison of 2002 and 2004 WTC sites to historic Liberty Island PCB measurements. Box and whiskers plots represent Liberty Island (HRUB) total PCB measurements from 1989 to 2004. Points on box and whiskers plot represent all Upper Bay sites (core and special WTC sampling) taken in 2002 and 2004. The lines represent national biennial 75th percentile for *Mytilus edulis* (2002–2003).

Sites found farther away exhibited concentrations below the 75th percentile, especially in more recent years.



Fig. 8. Total PCB concentrations in tissue for HRE core sites. The lines represent National biennial 75th percentile for Mytilus edulis (2002-2003).

3.8. Polychlorinated biphenyl spatial characterization using mussels

Upper Bay measurements were used to determine if samples collected in 2002 and 2004 were different than the Liberty Island site, a regularly sampled MWP site. When compared to total PCB measurements taken at the Liberty Island site from 1988 to 2004, WTC sites from 2002 and 2004 were all below the median value with many being below the lowest Liberty Island site value previously recorded (Fig. 9). However, the 2002 special WTC samples are among the highest nationally when compared to the MWP national 75th percentile (Fig. 9). Therefore there is no evidence that the WTC collapse had a significant affect on PCB input to the HRE.

4. Conclusion

While the attack on the WTC released organic contaminants to the HRE, the general pattern of improving environmental conditions continues back to the time that MWP began monitoring in the HRE in 1986. Data from 19 years of monitoring indicate that with minor exceptions, current contaminant concentrations are at or below the values reported previously. However, concentrations for total PAHs and total PCBs are still near or above the 75th percentiles for the national MWP data. The high background levels of PAHs and PCBs in the HRE masked the contaminant input associated with the WTC collapse and obscured its input to the Estuary.

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References

- Achman, D.R., Brownawell, B.J., Zhang, L.C., 1996. Exchange of polychlorinated biphenyls between sediment and water in the Hudson River Estuary. Estuaries 19, 950–965.
- Ayers, R.U., Rod, S.R., 1986. Patterns of pollution in the Hudson-Raritan Basin. Environment 28 (14–20), 39–43.
- Butt, C.M., Diamond, M.L., Truong, J., Ikonomou, M.G., Helm, P.A., Stern, G.A., 2004. Semivolatile organic compounds in window films from Lower Manhattan after the September 11th World Trade Center Attacks. Environmental Science and Technology 38, 3514–3524.
- Cundy, A.B., Croudace, I.W., Thomson, J., Lewis, J.T., 1997. Reliability of salt marshes as "geochemical recorders" of pollutant input: a case study from contrasting estuaries in southern England. Environmental Science and Technology 31, 1093–1101.

- Kimbrough, K.L., Dickhut, R.M., 2006. Assessment of polycyclic aromatic hydrocarbon input to urban wetlands in relation to adjacent land use. Marine Pollution Bulletin 52, 1355–1363.
- LaFlamme, R.E., Hites, R.A., 1978. The global distribution of polycyclic aromatic hydrocarbons in recent sediments. Geochmica Cosmochimica Acta 42, 289–303.
- Lauenstein, G.G., Cantillo, A.Y., 1993a. Sampling and analytical methods of the National Status and Trends Program National Benthic Surveillance and Mussel Watch Projects 1984–1992: overview and summary of methods. Volume I NOAA Technical Memorandum NOS ORCA 71, Silver Spring, MD.
- Lauenstein, G.G., Cantillo, A.Y. (Eds.), 1993b. Sampling and analytical methods of the National Status and Trends Program National Benthic Surveillance and Mussel Watch Projects 1984–1992: comprehensive descriptions of complementary measurements. Volume II NOAA Technical Memorandum NOS ORCA 71, Silver Spring, MD.
- Lauenstein, G.G., Cantillo, A.Y., (Eds.), 1993c. Sampling and analytical methods of the National Status and Trends Program National Benthic Surveillance and Mussel Watch Projects 1984–1992: comprehensive descriptions of elemental analytical methods. Volume III NOAA Technical Memorandum NOS ORCA 71, Silver Spring, MD.
- Lauenstein, G.G., Cantillo, A.Y., (Eds.), 1993d. Sampling and analytical methods of the National Status and Trends Program National Benthic Surveillance and Mussel Watch Projects 1984–1992: comprehensive descriptions of trace organic analytical methods. Volume IV NOAA Technical Memorandum NOS ORCA 71, Silver Spring, MD.
- Lauenstein, G.G., Cantillo, A.Y., 1998. Analytical methods of the National Status and Trends Program Mussel Watch Project – 1993– 1997 update. NOAA Technical Memorandum NOS ORCA 130.
- Lauenstein, G.G., Cantillo, A.Y., Kokkinakis, S., Jobling, J., Fay, R., 1997. Mussel Watch Project Site Descriptions, through 1997. NOAA Technical Memorandum NOS ORCA 112, Silver Spring, MD. 354pp.
- Litten, S., McChensney, D.J., Hamilton, M.C., Fowler, B., 2003. Destruction of the World Trade Center and PCBs, PBDEs, PCDD/ Fs, PBDD/Fs, and chlorinated biphenylenes in water, sediment, and sewage sludge. Environmental Science and Technology 37, 5502– 5510.
- Neff, J.M., 1979. Polycyclic Aromatic Hydrocarbons in the Aquatic Environment: Sources Fates and Biological Effects. Applied Science Publishers, London, pp. 22–43.
- O'Connor, T.P., 1996a. Coastal sediment contamination in the Northeast shelf large marine ecosystem. In: Sherman, K., Jaworsky, N.A., Smayda, T.J. (Eds.), The Northeast Shelf Ecosystem: Assessment Sustainability, and Management. Blackwell Science, Cambridge, MA, pp. 239–257.
- O'Connor, T.P., 1996b. Trends in chemical concentrations in mussels and oysters collected along the US Coast from 1986 to 1993. Marine Environmental Research 41, 183–200.
- O'Connor, T.P., Lauenstein, G.G., 2006. Trends in chemical concentrations in mussels and oysters collected along the US coast: update to 2003. Marine Environmental Research 62, 261–285.
- Offenberg, J.H., Eisenreich, S.J., Chen, L.C., Cohen, M.D., Chee, G., Prophete, C., Weisel, C., Loiyo, P.J., 2003. Persistent organic pollutants in the dusts that settled across Lower Manhattan after September 11, 2001. Environmental Science and Technology 37, 502– 508.
- Page, D.S., Boehm, P.D., Douglas, G.S., Bence, A.E., Burns, W.A., Mankiewicz, P.J., 1996. The natural petroleum hydrocarbon background in subtidal sediments of Prince William Sound, Alaska, USA. Environmental Toxicology and Chemistry 15, 1266–1281.
- Page, D.S., Boehm, P.D., Douglas, G.S., Bence, A.E., Mankiewicz, P.J., 1999. Pyrogenic polycyclic aromatic hydrocarbons in sediments record past human activity: a case study in Prince William Sound, Alaska. Marine Pollution Bulletin 38, 247–260.
- Sericano, J.L., Atlas, E.L., Wade, T.L., Brooks, J.M., 1990. NOAA's status and Trends Mussel Watch Program: chlorinated pesticides and PCBs in oyster (*Crassostrea virginica*) and sediments from the Gulf of Mexico, 1986-1987. Marine Environmental Research 29, 161–203.

- USEPA, 2002. Mid-Atlantic integrated assessment 1997–98 summary report, EPA/620/R-02/003. US Environmental Protection Agency, Atlantic Ecology Division, Narragansett, RI.
- USEPA, 2004. National coastal condition report II, EPA/620/R-03/002. US Environmental Protection Agency, Office of Research and Development/Office of Water, Washington, DC.
- Wang, Z., Fingas, M.F., 2003. Development of oil hydrocarbon fingerprinting and identification techniques. Marine Pollution Bulletin 47, 423–452.
- Wolfe, D.A., Long, E.R., Thursby, G.B., 1996. Sediment toxicity in the Hudson-Raritan Estuary: distribution and correlations with chemical contamination. Estuaries 19, 901–912.