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Sediment Toxicity in the Hudson-Raritan Estuary: Distribution and Correlations With Chemical Contamination

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ABSTRACT: The Hudson-Raritan Estuary is one of several United States coastal areas where chemical data have suggested a potential for contaminant-related biological effects, and multiyear intensive bioeffects surveys have been conducted by the National Oceanic and Atmospheric Administration. The severity and spatial patterns in sediment toxicity were determined in an estuary-wide survey during spring 1991 using amphipods, bivalve larvae, and luminescent bacteria as test organisms. Spatial patterns in toxicity corresponded to the distributions of a number of toxic chemicals in the sediments. Areas that exhibited the greatest sediment toxicity included the upper East River, Arthur Kill, Newark Bay, and Sandy Hook Bay. The lower Hudson River adjacent to Manhattan Island, upper New York Harbor, lower New York Harbor off Staten Island, and parts of western Raritan Bay generally showed lower toxicity. Supporting chemical analyses of the sediments, including acid-volatile sulfide and simultaneously-extracted metals, suggested that metals were generally not the cause of the observed toxicity, with the possible exception of mercury. Among all contaminants analyzed, toxicity was most strongly associated with polynuclear aromatic hydrocarbons, which were substantially more concentrated in toxic samples than in nontoxic samples, and which frequently exceeded sediment quality criteria.

Introduction

The highly urbanized Hudson-Raritan Estuary is heavily impacted by a variety of anthropogenic activities, including dredging and channelization for navigation, filling of natural shorelines and wetlands, commercial and residential development of coastal margins, and chemical contamination from runoff, wastewater treatment facilities, industrial plants, illegal dumping, and accidental spills (National Oceanic and Atmospheric Administration 1988a). Numerous sources of data have shown that concentrations of various categories of potentially toxic chemicals are elevated in this system, including polynuclear aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), DDT and other pesticides, dioxins, and many trace metals (Bretler 1984; Olsen et al. 1984; Belton et al. 1985; City of New York 1987; Bopp and Simpson 1989; Pruell et al. 1990; Tong et al. 1990; Bopp et al. 1991; Squibb et al. 1991). Wastewater treatment facilities were estimated to contribute 40–60% of the total input of several trace metals into the Hudson-Raritan Estuary, compared with 20–40% from tributary rivers, and 10–30% from urban runoff. Wastewater and tributaries were each estimated to contribute about 40% of the total load of PCB to the estuary (Hydroqual 1989).

Since 1984, the National Status and Trends (NS&T) Program of the United States National Oceanic and Atmospheric Administration has collected and analyzed samples of sediments and tissues of mussels (*Mytilus edulis*) and various finfish from up to seven locations within the Hudson-Raritan Estuary and seven more along coastal New Jersey for a broad suite of chemical contaminants (National Oceanic and Atmospheric Administra-

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tion 1987, 1988b, 1989, 1991; Gottholm et al. 1993). The concentrations of many contaminants in samples from sites within the Hudson-Raritan Estuary have consistently been among the highest measured at about 250 sites nationwide. Many of the Hudson-Raritan Estuary sites exhibited toxicant concentrations in sediments that exceeded known toxicity thresholds, and among all National Status and Trends sites nationwide, sites in the Hudson-Raritan Estuary were ranked as number 1, 3, 5, and 7 in terms of toxicity potential (Long and Morgan 1990; O'Connor and Ehler 1991). For many contaminants, including Pb, Hg, PCBs, and PAHs, the concentrations at most of the Hudson-Raritan Estaury sites fell into the upper 10th–15th percentile of sites nationwide, and many exceeded their corresponding Effects Range-Median (ERM) values. Sediments containing contaminant concentrations above the ERMs frequently elicit adverse biological effects in tests of sediment quality (Long and Morgan 1990; Long et al. 1995b). In a review of data available from numerous studies of the Hudson-Raritan Estuary, Squibb et al. (1991) performed similar analyses with a more extensive database, and concluded that numerous sediment contaminants in the Hudson-Raritan Estuary exceeded the Long and Morgan (1990) ERM values, especially in regions of Newark Bay, Arthur Kill, Gowanus Canal, Hackensack River, and the East River.

Based on the foregoing observations, an intensive survey of sediment toxicity was initiated in the Hudson-Raritan Estuary in 1991 to determine the severity and spatial extent of contaminant-related toxicity in this region. This study is one of a series of intensive bioeffects surveys implemented by the National Oceanic and Atmospheric Administration in areas indicated by the National Status and Trends Program and by other data as having significant potential for contaminant-related biological effects (Long et al. 1990, 1992, 1994, 1995a, c; Wolfe et al. 1992, 1993, 1994; Bricker et al. 1993).

Materials and Methods

Sediment Sampling

Sediment samples were collected for sediment toxicity testing and chemical analysis within 13 contiguous zones (A–M, Fig. 1). Based on previous studies, these zones were expected to be relatively homogeneous in depth and sediment texture. Three sampling sites were selected (Fig. 1) at broadly spaced locations in each zone, and three stations, usually equidistant from each other and approximately 250 m apart, were sampled at each site. In a few cases, dictated by site characteristics, stations were located in a straight line instead of a triangular configuration. Samples were collected with a modified 0.1-m^2 Van Veen grab from the research vessel *Mysidopsis* during five separate periods: March 18–22, April 1–5, April 15–18, April 28–May 2, and May 13–16, 1991.

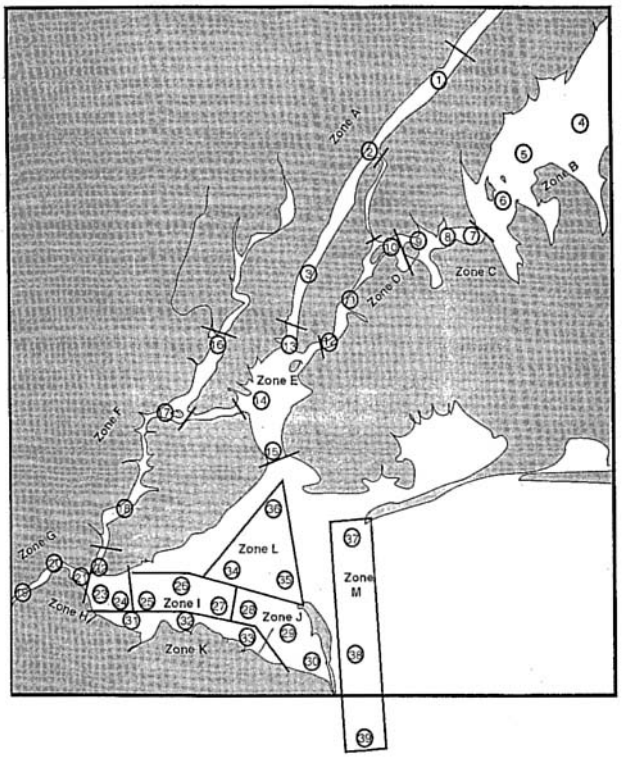

Fig. 1. Locations of Hudson-Raritan sampling sites for sediment toxicity testing, and zones used for estimation of areal extents of toxicity.

At each station, approximately 5 l of surficial (2 cm) sediments were accumulated from successive deployments of the grab for sediment toxicity tests and for analyses of inorganic and organic contaminants, total organic carbon (TOC), and grain size. The sediments for toxicity tests were homogenized thoroughly upon collection and transported (4°C) in polyethylene jugs to the laboratory. Portions of the samples were removed, chilled (not frozen), and tested for toxicity usually within 10 d. Other portions were frozen for later testing with Microtox^R. Also at each station, subsamples for later chemical analysis were placed into 500-ml precleaned glass jars with teflon lids, refrigerated at 4°C, and frozen within 2 d. The grab and sampling utensils were washed with seawater and acetone between sites, and with seawater alone between stations.

Toxicity Testing

The sediments from the Hudson-Raritan Estuary were tested for toxicity using three different ex-

posure modes and test organisms, and four different endpoints. A 10-d survival test using fresh, whole sediments was conducted with the amphipod *Ampelisca abdita,* following the protocols of the American Society for Testing and Materials (1990a). Sediment holding times were 2–9 d for 9 of the 10 test batches, and 27–28 d for the other. Test animals, collected from tidal flats in the Pettaquamscutt River, in Narragansett Bay, Rhode Island, were laboratory-acclimated for 2–10 d before testing. Test sediments were press-sieved (2.0 mm mesh) before addition to exposure containers. Five replicates were tested for each sediment sample. Each test chamber (1-l glass jars) had 200 ml of sediments covered with about 600 ml filtered seawater (20°C, 28 ‰) from Narragansett Bay. Twenty subadult amphipods were distributed randomly into each of the test chambers. Dead or moribund animals were recorded daily and removed. At the end of the 10-d test, surviving amphipods were enumerated. Temperature was measured daily, and salinity, dissolved oxygen, and pH were measured twice during each test. Control sediments were obtained from a Central Long Island Sound Reference Station (Wolfe et al. 1992). Tests were considered acceptable when control survival was at least 90%.

The 48-h test of survival and normal development of bivalve (*Mulinia lateralis*) embryos exposed to elutriates of test sediments followed standard protocols (American Society for Testing and Materials 1990b). Adult clams from a Narragansett Bay population were induced to spawn through temperature manipulation, and fertilization was allowed to proceed for at least 35 min before separation of the embryo stock.

Elutriates were prepared by addition of 500 ml seawater (28–30‰) to 100 g (wet weight) of homogenized sediment. The slurry was mixed for 30 min, allowed to settle for 1 h, then filtered (0.4 μm). Enough elutriate was filtered to produce five 15-ml test replicates for each sample. About 900 *Mulinia lateralis* embryos (0.75 ml well-mixed suspension) were added to each test vial, and the vials were incubated at 22°C for 48 h. Tests were terminated by addition of buffered formalin, and embryos were examined in 1-ml subsamples. Numbers of abnormally shelled embryos among surviving organisms were enumerated. Final percent survival was based on the mean initial embryo count. Tests were considered acceptable if at least 80% of the embryos survived in seawater controls and at least 80% of the survivors showed normal development.

Microtox^R tests were performed on methylene chloride extracts of the sediment samples, following an adaptation of the protocols recommended for the Puget Sound Estuary Program (1990). More consistent relationships to contaminant levels in sediments are obtained with Microtox^R on organic extracts than on saline extracts (Demuth et al. 1993). Sediments (3 g wet wt) were centrifuged to remove excess water; anhydrous Na_2SO_4 (15 g) was then added; and the samples were extracted with three successive 50 ml volumes of methylene chloride, which were combined, evaporated under N_2 to less than 1 ml and solvent-exchanged to 1 ml of ethanol for testing. Serial dilutions (representing 10 μl, 1.0 μl, 0.1 μl, and 0.01 μl of extract) with seawater were prepared in Microtox^R cuvettes, and reconstituted bacteria (*Photobacterium phosphoreum*) was added. Light emission was measured 5 min and 15 min later, and the results were compared to blanks containing the same dilutions of an ethanol reagent blank. These data were used to calculate the 50% inhibition concentrations (EC50).

Chemical Analyses

A subset of 38 of the sediment samples, representing 29 of the 39 sites and encompassing the full range of observed sediment toxicity, was selected for chemical analysis. Samples from site numbers 3, 15, 19, 20, 21, 27, 28, 31, 32, and 39 were not included among the samples analyzed chemically. Samples from two stations each were included from site numbers 7, 10, 12, 16, 17, 18, and 26; and all three samples from site number 30 were analyzed. Samples for chemical analysis were held at −20°C for about 2 yr before they were analyzed chemically. The concentrations of most analytes, including metals and organic constituents, are generally stable over this storage time (personal communications: H. Tatem, Army Corps of Engineers Waterways Experiment Station, Vicksburg Mississippi; Eric Crecelius, Battelle Pacific Northwest Laboratory, Sequim, Washington; Don Brown, National Marine Fisheries Service, National Oceanic and Atmospheric Administration, Seattle, Washington; Rick Swartz, United States Environmental Protection Agency, Environmental Research Laboratory, Newport, Oregon).

The suite of organic and inorganic chemicals measured in the sediment samples were those routinely measured by the National Status and Trends Program, including polycyclic aromatic hydrocarbons (PAHs), chlorinated pesticides including DDT and its metabolites, polychlorinated biphenyls (PCBs), and 16 trace and heavy metals (Robertson et al. 1993). Standard National Status and Trends Program quality assurance was applied throughout. Briefly, PAHs, PCBs, and chlorinated pesticides were analyzed by electron-capture gas chromatography or selective ion Gas Chromatography-Mass Spectrometry (Peven and Uhler 1993).

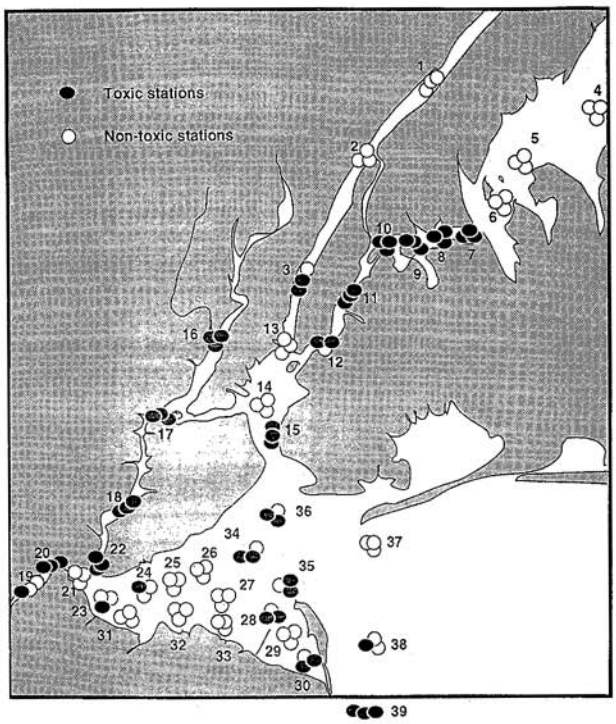

Fig. 2. Sampling stations where sediments were significantly toxic (n = 5; $\alpha \leq 0.05$) to amphipods (*Ampelisca abdita*) in a 10-d static test with whole sediments.

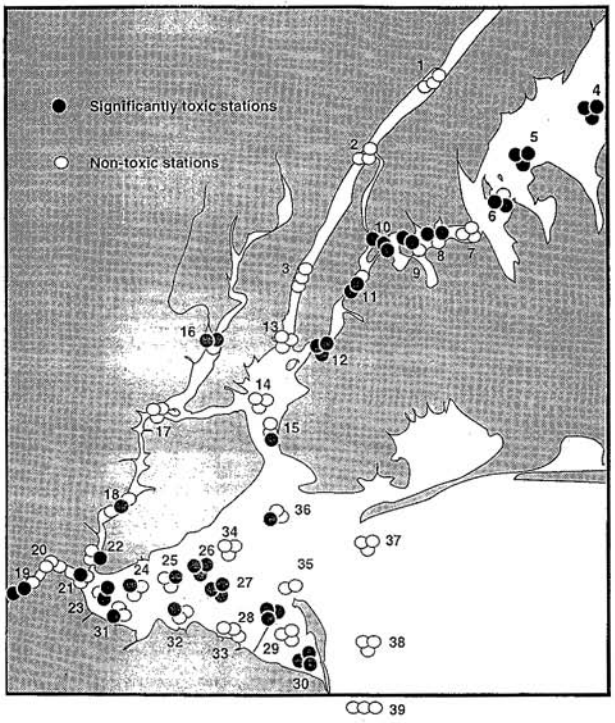

Fig. 3. Sampling stations where sediments were significantly toxic (n = 5; $\alpha \leq 0.05$) to bacteria (*Photobacterium phosphoreum*) in the Microtox^R test with organic extracts.

Concentrations of different metals were determined either by cold vapor atomic absorption, hydride generation atomic absorption, graphite furnace atomic absorption, or inductively coupled plasma/mass spectrometry (Crecelius et al. 1993).

After carefully scraping off any visibly oxidized surficial layer in the stored samples (personal communications: Dominic Ditoro, Manhattan College, Queens, New York; Eric Crecelius, Battelle Pacific Northwest Laboratory, Sequim, Washington), analyses for Acid-Volatile Sulfide (AVS) were performed using selective generation of hydrogen sulfide, cryogenic trapping, gas chromatographic separation, and photoionization detection (Cutter and Oates 1987; Allen et al. 1991). Following AVS analysis, the HCl digestate was filtered, and simultaneously extracted metals (SEM: cadmium, copper, lead, mercury, nickel, and zinc) were determined in the filtrates. Total organic carbon content was determined using a LECO carbon analyzer after first removing inorganic carbon with 6N HCl. Grain size was determined using a standard sieve and pipette method (Padell and Hillman 1993).

Results

Distribution of Toxicity

The toxicity data are summarized in Figs. 2 and 3; complete numerical data will be presented elsewhere (Long et al. 1995c). Of the 117 individual sediment samples tested, 54 were significantly toxic (one-tailed, unpaired, *t*-test on arcsine-square-root-transformed data; n = 5; alpha ≤ 0.05) relative to controls in the amphipod test (Fig. 2); 47 were significantly toxic in the Microtox^R test (Fig. 3); and 23 and 21 were toxic to bivalve larval survival and normal development, respectively (not illustrated). Figure 4 summarizes the collective toxicity responses by site means for all four tests; that is, the mean toxicities for the three stations at each site were averaged, and the site mean was then compared (one-tailed *t*-test on untransformed data; n = 3; alpha ≤ 0.05) to the mean toxicity (three replicates) at the control site. This comparison showed only one site (near Throgs Neck) to be significantly toxic in all four tests, while five sites (in the East River, Kill Van Kull, Arthur Kill, Verazzano Narrows, and Sandy Hook) were toxic in three tests. Most of the stations (Figs. 2 and 3) and sites (Fig. 4) in the East River, the Raritan River and Raritan Bay navigation channel, and Sandy Hook Bay were toxic in at least one test.

The mean response, by site, for each toxicity test was significantly correlated ($p \leq 0.05$; Spearman rank) with the site means for all other tests (Table 1). The strongest correlations were between the bivalve survival and development endpoints, and

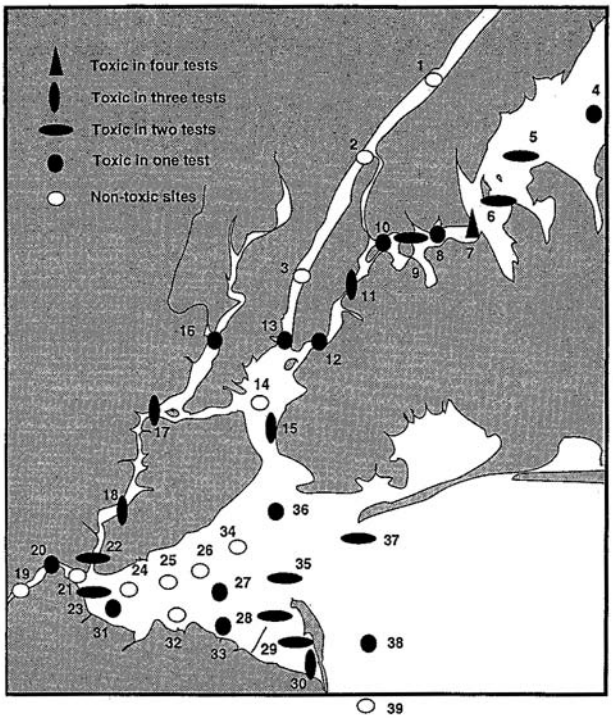

Fig. 4. Sampling sites in the Hudson-Raritan Estuary where sediments were determined to be nontoxic with any test, or where site means were significantly toxic relative to the control site mean in one, two, three, or four of the toxicity tests (amphipod survival, bivalve larval survival, bivalve larval development, and Microtox^R).

between Microtox^R and bivalve development, respectively. Though amphipod survival and Microtox^R were almost equally sensitive in terms of numbers of samples and sites exhibiting significant toxicity, these two endpoints were only moderately correlated with each other (Table 1). Of the 39 sites, 20 were significantly toxic with Microtox^R, but only 11 of those sites were included within the 16 that were toxic to amphipods.

The areas within each of the sampling zones (Fig. 1) were subdivided into three sectors corresponding to the individual sampling sites to permit a first approximation of the areal extent of toxicity throughout the Hudson-Raritan Estuary, based either on site means that were less than 80% of control response or, much more stringently, on those that were less than 20% of control response (Table 2). Although the sampling was not random in this survey, the large number of samples broadly distributed throughout the estuary should provide a reasonable first approximation of areal extent of toxicity of depositional sediments. While amphipod survival and Microtox^R response resulted in about equal estimates of degraded area (133 km^2 and 136 km^2, respectively), these two endpoints were not highly concordant with each other. Only

TABLE 1. Spearman Rank Correlations (Rho) among sediment toxicity test results from the Hudson-Raritan Estuary, based on control-corrected site means (n = 39).

	Amphipod Survival	Bivalve Survival	Bivalve Development
Bivalve survival	0.229*		
Bivalve development	0.226*	0.644***	
Microtox^R bioluminescence	0.285*	0.270*	0.353**

* $p \leq 0.05$. ** $p \leq 0.01$. *** $p \leq 0.001$.

28% of the 136 km^2 estimated by Microtox^R was also indicated as toxic by amphipod survival (Table 2). The total area indicated as toxic by at least one test was therefore over 230 km^2, or at least two-thirds of the total system. Of that total, however, about 72 km^2 (~20% of total area) gave positive responses in three tests, while less than 20 km^2 (<6% of total area) were indicated as severely toxic, either in terms of significant response to all four tests or a response of less than 20% of control survival or normal development in either the amphipod or bivalve test (Table 2).

Correlations Between Toxicity and Contamination

Results of chemical analyses are summarized for most contaminants in Table 3. Concentrations of lindane, aldrin, heptachlorepoxide, endrin, and mirex were at or below limits of detection (0.10 ppb, 0.21 ppb, 0.23 ppb, 0.57 ppb, 0.25 ppb, respectively) at all or nearly all of the stations; summary data for these chemicals are not included in the table. The summary data for dieldrin includes 10 values at the detection limit of 0.2 ppb. Complete results of the individual chemical analyses will be reported separately (Long et al. 1995c).

Table 4 shows the mean contaminant concentrations for four groupings of samples: those that were not statistically significantly toxic, and those that were "highly toxic", that is, both statistically significantly toxic *and* less than 80% of the control values, for the amphipod and the Microtox^R tests. In the amphipod tests, the nontoxic samples (n = 17) showed a mean survival of 98.4 ± 11.1%, while

TABLE 2. Spatial extent (km^2) of sediment toxicity in the Hudson-Raritan Estuary, where total area sampled was 350 km^2.

Test	<20% of Control	<80% of Control	Percent Concordance[a]
Amphipod survival	12.0	133	
Bivalve survival	0	87.4	74%
Bivalve development	16.1	104	65%
Microtox^R luminescence	0	136	28%
3 of the 4 tests		71.9	
All 4 tests		19.9	

[a] Percent of area included also in amphipod-positive area.

TABLE 3. Mean concentrations, ranges, and the three most contaminated stations for various contaminants measured at 38 stations in the Hudson-Raritan Estuary. Except as noted, concentrations of metals and PAHs are in ppm, while those of other organic contaminants are in ppb.

Contaminant	Mean	Std Dev	Max	Min	Three Highest Stations
HCB	4.8	5.3	20.2	0.14	22C, 12B, 23A
Cis-Chlordane	4.8	4.5	19.0	0.17	18C, 17C, 17B
Trans-Nonachlor	3.8	3.7	14.8	0.14	18C, 12A, 17C
Dieldrin	3.1	3.2	13.6	0.20	12B, 12A, 23A
Sum Indeno pesticide	9.2	8.5	34.3	0.81	18C, 12A, 17C
Sum non-DDT pesticide	19.4	14.4	63.2	2.4	18C, 12A, 12B
4,4-DDE	27.3	41.7	182	0.13	17B, 17C, 18C
4,4-DDT	79.3	272	1,610	0.094	17B, 18A, 17C
Sum DDT	140	241	921	1.3	18A, 17B, 17C
Sum PCB	281	333	1,970	5.0	12A, 18C, 17B
acenaphthene	1.60	9.0	56.3	0.1	9B, 10B, 7B
phenanthrene	5.88	31.0	194	0.39	9B, 7B, 10B
fluoranthene	4.77	17.3	108	0.31	9B, 7B, 23A
Petroleum PAH	7.37	33.6	210	2.7	9B, 5B, 7B
Combustion PAH	24.2	86.5	543	3.3	9B, 7B, 8C
Sum PAHs	42.9	179	1,120	7.9	9B, 7B, 10B
Sum LMWPAH	17.8	91.2	572	4.2	9B, 7B, 10B
SumHMWPAH	27.3	87.6	552	3.7	9B, 7B, 10B
Aluminum (%)	5.40	1.69	8.1	1.2	33B, 23A, 25A
Antimony	2.0	1.7	7.8	0.13	18C, 9B, 12A
Arsenic	20.0	11.0	41.0	1.7	10B, 22C, 30C
Cadmium	1.72	1.36	6.4	0.03	18C, 12A, 22C
Copper	142	112	520	3.3	18C, 12A, 9B
Chromium	122	67	420	12	12A, 18C, 33B
Iron (%)	3.53	1.24	5.5	0.63	30C, 30A, 33B
Lead	160	103	510	16	12A, 9B, 18C
Mercury	2.29	2.46	15	0	18C, 9B, 7B
Nickel	37.8	20.2	130	4.5	12A, 11B, 17C
Selenium	1.1	0.78	4.3	0.16	18C, 22C, 23A
Silver	1.97	0.81	3.4	0.01	29A, 2A, 30C
Tin	21.7	17.4	100	1.2	18C, 9B, 17C
Zinc	299	217	1,400	38	12A, 18C, 9B
AVS[a] μmol g^{-1}	28.5	21.1	79.7	0.036	5B, 9B, 18C
Total SEM[b], μmole g^{-1}	3.36	1.63	7.0	0.27	18C, 12A, 33B
SEM/AVS	0.63	1.7	9.3	0.04	37B, 38B, 2A
Organic Carbon (%)	2.62	1.25	5.02	0.07	9B, 12A, 7B
% fines	39.3	21.4	76.7	0	17B, 6C, 2A

[a] AVS = Acid-volatile sulfide.
[b] SEM = Simultaneously extracted metals, the sum of Cd, Cu, Hg, Ni, Pb, and Zn.

the "highly toxic" samples (n = 19) had a mean survival of 30.1 ± 26.3%. In the Microtox^R tests, the nontoxic samples (n = 22) had a mean EC50 of 6.3 ± 8.8 mg sediment ml^{-1}, while the "highly toxic" samples (n = 10) had a mean EC50 of 1.3 ± 0.4 mg ml^{-1}. In nearly all cases, the mean concentration of any given contaminant was greater in the toxic group than in the nontoxic group, indicating a general association of toxicity with chemical contamination. For many of the chemicals, however, the mean concentrations are not greatly different in the toxic and nontoxic groups, suggesting a small overall gradient for that particular contaminant and discounting the probability that it is related causally to toxicity. Because of the strong covariance and likely interactions among contaminants, the specific cause(s) of chemical toxicity cannot be determined with certainty from these compositional data alone. However, those chemicals that were most strongly associated with measures of toxicity can be identified through correlation analyses and comparison with existing sediment quality guidelines.

Spearman rank coefficients (Rho) for correlations between the concentrations of various sediment contaminants and the amphipod toxicity or Microtox^R response are also shown in Table 4. The bivalve larval survival and development endpoints were not significantly correlated with the concentrations of any of the contaminants measured, despite the previously noted (Table 1) correlation with both the amphipod and Microtox^R results. Contaminants analyzed, but not listed in Table 4, exhibited weaker correlations, usually not significant, with the toxicity measures than those listed.

Amphipod survival was significantly inversely correlated with the concentrations of tin and mercury (Fig. 5) in sediments but not with any other

TABLE 4. Spearman rank correlations (Rho) between sediment toxicity and contaminant concentrations in sediments from the Hudson-Raritan Estuary (n = 35) for the amphipod survival and Microtox endpoints, and mean concentrations of chemical contaminants in samples that were significantly toxic or nontoxic in these two tests. Except as noted, concentrations of metals and PAHs are in ppm, while those of other organics are in ppb.

	Correlation Coefficients		Amphipod		Microtox	
	Amphipod	Microtox	Nontoxic	Toxic	Nontoxic	Toxic
Cadmium	−0.264ns	−0.472*	1.2 ± 0.6	2.2 ± 1.7	1.6 ± 1.5	2.1 ± 1.3
Copper	−0.255ns	−0.449*	111 ± 63	174 ± 139	124 ± 113	191 ± 124
Lead	−0.295ns	−0.478*	191 ± 124	131 ± 68	132 ± 80.5	224 ± 128
Mercury	−0.437*	−0.377*	1.4 ± 1.0	3.2 ± 3.1	2.4 ± 3.1	2.2 ± 1.2
Nickel	−0.095ns	−0.451*	34.9 ± 14.2	41.0 ± 24.7	32.3 ± 15.4	51.3 ± 27.4
Tin	−0.342*	−0.427*	16.3 ± 9.3	27.2 ± 21.5	18.6 ± 13.1	30.1 ± 25.2
Zinc	−0.134ns	−0.433*	265 ± 123	331 ± 279	241 ± 132	442 ± 327
Total SEM μmole g^{-1}	−0.130ns	−0.417*	3.30 ± 1.68	3.38 ± 1.64	3.1 ± 1.8	4.3 ± 1.0
Total AVS μmole g^{-1}	−0.150ns	−0.544**	25.4 ± 22.6	31.6 ± 20.4	22.1 ± 18.4	39.7 ± 16.6
SEM:AVS ratio	+0.197ns	+0.454*	1.21 ± 2.41	0.16 ± 0.15	1.0 ± 2.2	0.1 ± 0.1
TOC (%)	−0.151ns	−0.581**	2.4 ± 1.1	2.8 ± 1.4	2.2 ± 1.2	3.6 ± 1.0
Fines (%)	+0.196ns	−0.347*	41.4 ± 24.2	37.2 ± 18.8	34.3 ± 24.0	45.2 ± 15.0
cis-Chlordane	−0.204ns	−0.384*	3.3 ± 2.0	6.4 ± 5.6	4.9 ± 5.2	5.7 ± 3.4
trans-nonachlor	−0.150ns	−0.402*	2.5 ± 1.5	5.0 ± 4.7	3.7 ± 4.1	4.6 ± 3.4
4,4′ DDE	−0.287ns	−0.402*	10.8 ± 8.9	43.7 ± 53.5	34.3 ± 50.9	24.2 ± 22.6
4,4′ DDT	−0.476*	−0.341*	7.4 ± 19	151 ± 371	130 ± 349	11.8 ± 12.8
Total DDT	−0.311ns	−0.271ns	48.4 ± 43.6	232 ± 313	192 ± 300	91.7 ± 84.6
Indeno pesticides	−0.197ns	−0.395*	6.3 ± 3.5	12.2 ± 10.7	9.2 ± 9.3	11.3 ± 8.3
Total other ChlPest	−0.146ns	−0.250ns	16.0 ± 9.3	22.8 ± 17.7	19.4 ± 14.8	24.3 ± 15.1
Sum PCB	−0.124ns	−0.306ns	196 ± 132	370 ± 436	516 ± 432	865 ± 1,060
Sum LMW[b] PAH	−0.592**	−0.650***	0.922 ± 0.583	34.7 ± 127	2.34 ± 4.39	59.8 ± 171
Sum HMW PAH	−0.471*	−0.512*	6.81 ± 8.28	48.1 ± 120	10.3 ± 17.1	70.1 ± 161
Petroleum PAH	−0.468*	−0.625***	2.06 ± 6.66	12.9 ± 46.5	0.845 ± 1.38	22.1 ± 62.6
Combustion PAH	−0.576**	−0.602**	3.53 ± 2.26	44.7 ± 119	9.33 ± 16.7	62.9 ± 160
Total PAH	−0.495*	−0.603**	5.43 ± 3.84	80.3 ± 247	11.9 ± 21.4	124 ± 333
Fluoranthene/TOC	−0.559**	−0.418*	0.474 ± 0.956	2.16 ± 4.67	0.672 ± 0.927	2.92 ± 6.32
Acenaphthene/TOC	−0.641***	−0.437*	0.011 ± 0.007	0.668 ± 2.49	0.034 ± 0.048	1.15 ± 3.36
Phenanthrene/TOC	−0.571**	−0.398*	0.123 ± 0.125	2.49 ± 8.55	0.294 ± 0.386	4.07 ± 11.5

Spearman rank coefficients: ns = not significant, *p < 0.05, **p < 0.01, ***p < 0.001.

[a] SEM/AVS = simultaneously extracted metals/acid-volatile sulfide.

[b] LMW = sum of low-molecular-weight PAHs, up to and including three condensed rings; HMW = sum of high-molecular-weight PAHs, 4–6 condensed rings; Petroleum PAH = sum of petroleum (substituted) PAHs; combustion PAH = sum of unsubstituted (parent) PAHs.

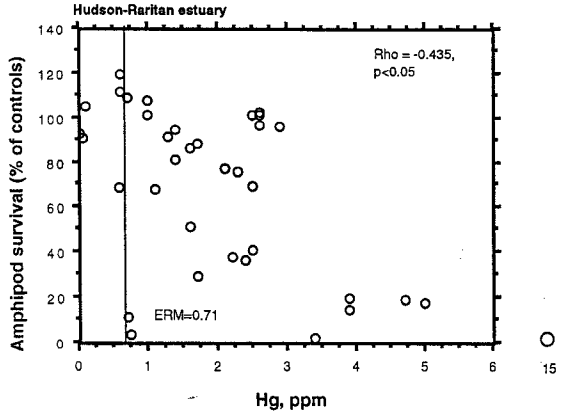

Fig. 5. Relation between amphipod survival and the concentration (μg g^{-1} dry weight) of mercury in sediments from the Hudson-Raritan Estuary.

of the metals analyzed (Table 4). Microtox[R] was significantly inversely correlated with all of the metals shown, and with TOC and % fine sediments as well. Unlike the other three endpoints, the Microtox[R] response was also significantly inversely correlated with total SEM and with total AVS in the sediments. However, the correlation with AVS was more strongly negative than that with SEM, and when analyzed for the SEM:AVS ratio (i.e., the cumulative molar ratio of Pb, Hg, Cd, Cu, Ni, and Zn to AVS), the correlation with Microtox[R] response became significantly positive (Table 4). Only three of the analyzed samples exhibited an SEM:AVS ratio greater than 1, and all three of these were very sandy samples that had extremely low AVS concentrations and were nontoxic to the amphipods.

Microtox[R] response and amphipod survival exhibited consistently negative correlations with all of the chlorinated pesticides (Table 4). Microtox[R] response was significantly correlated with cis-chlordane, trans-nonachlor, 4,4′ DDT and DDE, and indeno pesticides, whereas amphipod survival was

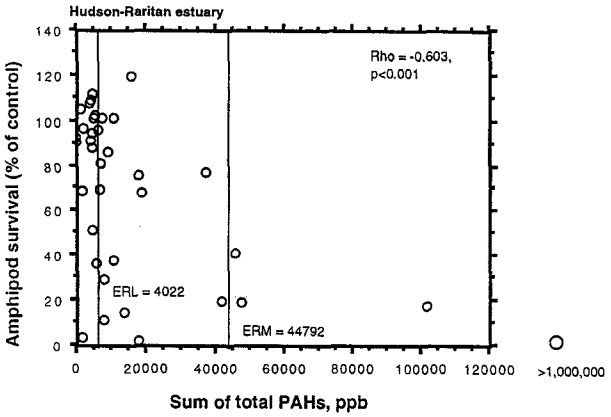

Fig. 6. Relation between amphipod survival and the concentration (ng g^{-1} dry weight) of total polynuclear aromatic hydrocarbons in sediments from the Hudson-Raritan Estuary.

significantly correlated only with 4,4′ DDT. Neither of the bivalve endpoints showed consistent negative correlations with chlorinated compounds.

Both Microtox^R response and amphipod survival were significantly correlated, (sometimes at the $p \leq 0.001$ level) with various combinations of PAH compounds (Table 4). In addition, both larval bivalve endpoints showed consistently negative, though not significant, correlations with PAH fractions. The amphipod survival, as percent of control value, is shown in Fig. 6 as a function of total PAH concentration in the sediment samples. All four of the samples that exceeded the ERM (~45,000 ng g^{-1}; Long et al. 1995b) were significantly toxic to amphipods; 13 of the 22 samples with PAHs between the Effects Range-Low (ERL, ~4,000 ng g^{-1} for tPAH) and the ERM were toxic; while only 2 of the 10 samples with PAHs lower than the ERL were toxic (Fig. 6).

Comparisons With Sediment Quality Guidelines

Table 5 lists the ratios (toxic to nontoxic) of the mean chemical contaminant concentrations enumerated in Table 4. The ratios of those same toxic group means to the respective sediment quality guidelines (Effects Range-Median, or ERMs of Long et al. 1995b, or Long and Morgan 1990) or the proposed National sediment quality criteria (SQC) of the United States Environmental Protection Agency (1994) are given. Also listed are the number of samples (out of 38) that exceeded the respective guideline for each chemical.

For both toxicity tests, the mean concentrations of trace metallic contaminants were generally only slightly higher in the toxic samples than in the nontoxic samples, and for most metals the mean concentrations in the toxic samples were lower than the respective ERM values of Long et al. (1995b). The mean concentrations of lead and zinc in the samples toxic in one or both tests equalled the applicable ERM values, whereas the toxic means for mercury exceeded the ERM value by 3-fold to 4.5-fold, and 30 of the 38 samples equalled or exceeded the ERM value for mercury.

Four chlorinated organic compound groups were significantly correlated with the Microtox^R response and one of these (4,4′ DDT) was also significantly correlated with amphipod toxicity (Table 4). The mean concentration gradient between toxic and nontoxic samples was greater, however, for the amphipod test than for Microtox^R for all (cis-chlordane, trans-nonachlor, 4,4′-DDE, and 4,4′-DDT) of these compounds, and the mean concentration of 4,4′-DDT in the samples toxic to amphipods was 21.6× the ERM value, compared with

TABLE 5. Ratios of mean contaminant concentrations in groups of samples that were significantly toxic or nontoxic in the amphipod and Microtox^R tests, and comparison of the toxic sample means with sediment quality guidelines (SQG)[a].

Contaminant	Toxic:nontoxic Ratios		Toxic:SQG Ratios		No. Samples Exceeding SQG
	Amphipod	Microtox^R	Amphipod	Microtox^R	
Cadmium	1.9	1.3	<1	<1	0
Copper	1.6	1.5	<1	<1	2
Lead	1.5	1.7	<1	1.0	8
Mercury	2.2	0.9	4.5	3.1	30
Nickel	1.2	1.6	<1	<1	3
Tin	1.7	1.6	na	na	na
Zinc	1.2	1.8	<1	1.1	5
Total SEM[b]	1.0	1.4	na	na	na
Total AVS[b]	1.2	1.8	na	na	na
SEM:AVS[b] ratio	0.1	0.1	na	na	na
TOC	1.2	1.6	na	na	na
% fines	0.9	1.3	na	na	na
cis-Chlordane	2.0	1.1	na	na	na
trans-nonachlor	2.0	1.2	na	na	na
4,4′ DDE	4.0	0.7	1.6	<1	12
4,4′ DDT	20.3	0.1	21.6	1.7	14
Total DDT	4.8	0.5	na	na	na
Indeno pesticides	−1.9	1.2	na	na	na
Sum PCB	1.9	1.7	na	na	na
Total other ChlPest	−1.4	1.3	na	na	na
Sum LMW[c] PAH	37.6	18.9	11.0	18.9	9
Sum HMW[c] PAH	7.1	6.8	5.0	7.3	14
Petroleum[c] PAH	6.3	26.2	na	na	na
Combustion[c] PAH	12.7	6.7	na	na	na
Total PAH	14.8	10.4	1.8	2.8	4
Fluoranthene/TOC	−4.6	4.3	7.2	9.7	20
Acenaphthene/TOC	−58.7	34.2	2.8	4.8	2
Phenanthrene/TOC	−20.2	13.9	10.4	17.0	14

[a] na = no sediment quality guideline available.

[b] SEM/AVS = simultaneously extracted metals/acid-volatile sulfide.

[c] LMW = sum of low-molecular weight PAHs, up to and including three condensed rings; HMW = sum of high-molecular weight PAHs, 4–6 condensed rings; Petroleum PAH = sum of petroleum (substituted) PAHs; combustion product PAH = sum of unsubstituted (parent) PAHs.

only 1.7× for the samples toxic to Microtox^R (Table 5). All of the classes and compounds of PAHs exhibited strong concentration gradients between the toxic and nontoxic samples, and the toxic means for both toxicity tests also consistently exceeded the guideline or criteria values (Table 5). The concentrations of low molecular weight PAHs, in particular, corresponded well to the toxicity results in both tests.

Discussion

The four toxicity endpoints provided overlapping, but different, estimates of the severity and spatial extent of toxicity in the Hudson-Raritan Estuary. The 10-d, whole sediment, static test with amphipods (*Ampelisca abdita*) was the most sensitive test employed here, indicating toxicity in the greatest number of individual samples. However, the Microtox^R test indicated a greater number of significantly toxic site means, and produced estimates for a degraded area similar to that from the amphipod test. Although the bivalve larval survival test was much less sensitive than the amphipod test, the concordance between the two was fairly strong: that is, most of the positive tests with *Mulinia* were included in the area that tested positive with amphipods. For Microtox^R however, this measure of concordance was fairly low, with only 28% of the Microtox^R-positive area included in the *Ampelisca*-positive area. These results are not necessarily unexpected from a selection of toxicity assays, because the various test organisms are responding to different exposure modes, exposure durations, and contaminants, with different sensitivities. However, the Microtox^R assay using organic extracts, as employed in this and other National Oceanic and Atmospheric Administration studies, probably estimates the total toxicity potential from bulk organic contaminants in the sediments, without reflecting the natural mediating effects of total organic carbon on exposure and bioaccumulation (Wolfe et al. 1994). The potential for variability of responses among different tests is nonetheless sound justification for using a variety of sensitive tests in sediment quality assessment (Wolfe 1992).

The area indicated by all four assays to be toxic (estimated at about 20 km^2) encompassed the extreme western portion of Long Island Sound near Throg's Neck, the East River, and a portion of Sandy Hook Bay. Parts of Arthur Kill and Kill Van Kull were included in the area that met the three-test criterion. Portions of these same areas also exhibited the strongest responses (i.e., <20% survival or <20% normal development) in the amphipod and bivalve tests, respectively. The areas of least toxicity in the Hudson-Raritan Estuary included the Hudson River west and north of Manhattan, upper New York Harbor, and the lower harbor southeast and east of Staten Island. After this survey was completed, a more intensive survey of sediment toxicity and chemistry was carried out in Newark Bay where widespread toxicity was indicated (Long et al. 1995c).

The incidences of toxicity found with the three tests reported here are considerably greater than those that have been obtained with traditional endpoints (survival of *Palaemonetes pugio*, *Nereis* worms, and *Mercenaria mercenaria*) used in predredging studies of sediments from the Hudson-Raritan Estuary. In 70 public notices issued between 1985 and 1993, the United States Army Corps of Engineers has reported 12 positive results of toxicity in 252 tests. Part of the apparent difference in sensitivity compared to the toxicity results reported here could have arisen merely from the specific sources of sediment, or from chemical differences between the surficial sediments tested here and the deeper, homogenized sediments tested by the United States Army Corps of Engineers. The apparent disparity in these results suggests that multiple tests (drawn, for example, from those recommended by the United States Environmental Protection Agency/United States Army Corps of Engineers 1991) using sensitive test organisms and representing a wide range of exposure conditions and endpoints are desirable for a complete assessment of sediment quality.

The prevalence of sediment toxicity in the Hudson-Raritan Estuary is similar to that observed in analogous studies in San Francisco Bay (Long et al. 1990), Los Angeles and Long Beach harbors in San Pedro Bay (E.R. Long, National Oceanic and Atmospheric Administration, unpublished data), and in the confined bays (primarily in Connecticut and New York) along the coast of Long Island Sound (Wolfe et al. 1994) but is considerably higher than in Tampa Bay (Long et al. 1994) or Pensacola Bay (E. R. Long, National Oceanic and Atmospheric Administration, unpublished data).

The correlative approaches used here cannot positively identify the chemical causes of the observed toxicity. The test organisms are exposed to the full suite of chemical contaminants in the sediments, and toxicity is probably elicited by various contaminants and other stressors in combination with each other.

Unionized ammonia has been identified as a probable cause of toxicity in bioassays of some sediments (Jones and Lee 1988; Ankley et al. 1990). We did not measure ammonia nitrogen and calculate unionized ammonia in the tests reported here. However in more recent studies in other estuarine systems (e.g., Long et al. 1994, 1995a), we have performed these analyses, with the consistent

result that very few samples have exceeded either the reported LC-50 (0.83 ppm) for *Ampelisca abdita* or the No-Observable-Effects Level of about 0.4 ppm. A similar result has recently been obtained with a suite of 57 samples from Newark Bay (Long et al. 1995c), leading us to conclude that NH_3 probably did not account for a significant portion of the observed toxicity in the samples from the Hudson-Raritan Estuary.

We found consistently stronger Spearman rank correlations with concentrations of most contaminants in the sediments for the Microtox^R endpoint than for other toxicity endpoints. While this result was expected with the organic constituents, it was unexpected with the metals because the organic extracts tested with Microtox^R would tend to select against the metals. The Microtox^R endpoint, however, was also very strongly correlated with TOC and also with grain size, and we believe that the observed correlations with metals are the result of simple covariance of metals with grain size, TOC, and organic contaminants in the sediments.

The likelihood of a significant metals effect was also discounted by our SEM:AVS data: only three of the analyzed samples exhibited an SEM:AVS ratio greater than 1, and all three of these were sandy samples with very low AVS and also with very low toxicity. The chemical data for many metals were not concordant with toxicity to either *Ampelisca* or *Mulinia*; the nontoxic samples tended to have similar (and occasionally higher) concentrations of many metals than did the toxic samples, suggesting that the metals were not strongly related to our observed toxicity. Among the metals analyzed, mercury was the most likely contributor to the observed toxicity. Mercury showed the strongest correlation with amphipod toxicity (Table 4), as well as the strongest concentration gradient between toxic and nontoxic samples. Compared with the other contaminants, it exceeded the ERM guideline values (Table 5) to the greatest degree. Based on the quality of the underlying database, however, Long et al. (1995b) reported only a moderate level of confidence in the ERM value for mercury, compared to a relatively high degree of confidence in the values for lead and zinc.

Except for the correlation of amphipod toxicity with 4,4′ DDT (Table 3), only the Microtox^R response showed significant Spearman correlations with pesticides and other categories of chlorinated hydrocarbons, and even then the significance levels were generally low. Among the chlorinated compounds analyzed, DDT and its metabolites exhibited the strongest correlations with amphipod toxicity (Table 4), as well as the greatest concentration differences between toxic and nontoxic samples, and ERM exceedances (Table 5). Correlations between PCBs and toxicity were not significant even for Microtox^R (Table 4). Only six samples exceeded the Long and Morgan (1990) ERM values for PCBs, and although five of these were significantly toxic, only one exceeded the ERM by a substantial amount.

By far the most consistent pattern of correlation with toxicity was seen for the PAHs in these samples. A broad suite of individual PAHs was significantly correlated with both amphipod survival and with Microtox^R response, as were a variety of PAH sums, including total PAHs. Correlations between PAHs and the bivalve endpoints, although not significant, were at least consistently negative. Most of the samples containing PAHs at concentrations higher than Long and Morgan's (1990) ERL value for total PAH were significantly toxic to amphipods, as were all those exceeding the ERM. This same relationship pertained also when PAH concentrations were normalized to TOC concentrations, and compared to the proposed (United States Environmental Protection Agency 1994) National Sediment Quality Criteria for phenanthrene, fluoranthene, and acenaphthene, taken individually (Table 5). Although not conclusive, these patterns suggest that PAHs may be a substantial contributor to the observed toxicity in the Hudson-Raritan Estuary.

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