

## Polycyclic Aromatic Hydrocarbon and Petroleum Hydrocarbon Contamination in Sediment from the Newark Bay Estuary, New Jersey

S. L. Huntley<sup>1</sup>, N. L. Bonnevie, R. J. Wenning

ChemRisk®—A Division of McLaren/Hart, Stroudwater Crossing, 1685 Congress Street, Portland, Maine 04102, USA

Received: 2 May 1994/Revised: 7 June 1994

**Abstract.** The presence of numerous industrial and municipal sources such as former creosote wood preserving facilities, petroleum storage and refinery facilities, paint and chemical manufacturers, combined sewer overflows, and sewage treatment facilities along the shores of Newark Bay, New Jersey and its major tributaries suggests the potential for widespread contamination by polycyclic aromatic hydrocarbons (PAHs) and petroleum hydrocarbons. In this study, the concentrations and distributions of 19 PAHs and total extractable petroleum hydrocarbon (TEPH) were determined in 213 sediment samples obtained from 58 sediment cores collected between November 1991 and March 1993 from the Arthur Kill, Elizabeth River, Hackensack River, Kill Van Kull, Newark Bay, Upper New York Bay, and Rahway River. Chronological profiles of PAH and TEPH deposition from pre-1930 to the present were determined in each sediment core using <sup>137</sup>Cs and <sup>210</sup>Pb radioisotope measurements. The concentrations of total PAHs (tPAH) and individual PAHs were compared to National Oceanic and Atmospheric Administration (NOAA) benchmark sediment effects-range median (ER-M) values. The objectives of this study were to: (a) determine the spatial and temporal distributions of PAHs in sediments; (b) identify their possible sources; and (c) assess the potential for sediment toxicity within the estuary. The results indicate elevated concentrations of PAHs and TEPH in surface and buried sediments throughout the estuary, particularly in the Elizabeth River, the Arthur Kill, and in the Passaic River above the Dundee Dam and below the Jackson Street Bridge. Sediments collected from ship berths at Port Newark and Port Elizabeth in Newark Bay were also found to contain elevated levels of PAHs and TEPH. The concentrations of PAHs and TEPH in sediment generally increase with depth throughout the estuary. Comparisons to NOAA ER-M values indicate that the concentrations of many PAHs in surface and buried sediments at several locations in the estuary pose a significant hazard to aquatic organisms. Radiodating of sediment cores reveals that the highest concentrations of PAHs, and the greatest potential hazards to aquatic biota, occur in sediments deposited during the 1950s. Although the major inputs of

PAHs are probably combustion sources and urban runoff entering the estuary through combined sewer overflows and storm drains, the unusually high concentrations of PAHs and TEPH found in some sediments may be best explained by point source inputs or catastrophic oil spills.

The accumulation of high concentrations of polycyclic aromatic hydrocarbons (PAHs) in the sediments of industrialized and densely populated bays and estuaries has been well established (NOAA 1988, 1991). During the past century, these levels have increased due to the escalating use of petroleum products, post-World War II economic expansion, and greater reliance on marine transportation of oil (O'Connor and Hugget 1988). As a result of their hydrophobicity and strong affinity for organic matter, PAHs accumulate in sediments at levels several orders of magnitude higher than those found in source effluents (Herbes and Schwall 1978). Since these compounds are biologically available, PAH-contaminated sediment can adversely affect biota or result in high concentrations in the tissues of indigenous organisms (Pruell *et al.* 1984). It is well known that even moderate concentrations of certain PAHs and petroleum hydrocarbons in sediment can have a detrimental effect on aquatic organisms (Hollister *et al.* 1980; Catallo and Gambrell 1987).

In the Newark Bay Estuary, numerous studies have been conducted to determine the extent of chemical contamination in sediment and biota, but most have focused on other persistent compounds such as dioxins, PCBs, and metals (Wilber and Hunter 1973, 1979; Suszkowski 1978; Williams *et al.* 1978; Meyerson *et al.* 1981; NOAA 1982, 1984a, 1984b; O'Keefe *et al.* 1984; NJDEP 1985; Bopp 1988; Goeller 1989; HMDC 1989; Moser 1990; Bopp *et al.* 1991; Bonnevie *et al.* 1992, 1993; Gillis *et al.* 1993; Wenning *et al.* 1992). It has been shown in whole sediment bioassays that PAHs may act synergistically to elicit toxic responses similar to those demonstrated for dioxins (Brunström *et al.* 1992); therefore, it is important to consider these compounds when evaluating the overall toxicity of sediments.

In general, high PAH concentrations in sediment have been linked to a wide variety of combustion sources and creosote

wood preserving facilities (USEPA 1982; Mueller *et al.* 1989) as well as tanker spills, oil seeps, and other petroleum-related industries (NOAA 1991). Since the early 1800s, a myriad of activities have resulted in the pollution of Newark Bay and its tributaries from municipal and industrial wastewaters, storm-water runoff, accidental spills, direct dumping of wastes, and atmospheric deposition (Olsen *et al.* 1984; HydroQual 1991). The presence of hundreds of industrial and municipal facilities including paint and chemical manufacturing facilities, metal scrapyards and refineries, wood treatment plants, and petroleum terminals provides strong impetus for examining PAH and petroleum hydrocarbon sediment concentrations in the estuary.

In this study, the concentrations of 19 PAHs and TEPH (measured as diesel) were determined in sediments collected from the Newark Bay Estuary, specifically the Arthur Kill, Elizabeth River, Hackensack River, Kill Van Kull, Newark Bay, Passaic River, Upper New York Bay, and Rahway River. The age of the sediment at different depths was established from  $^{137}\text{Cs}$  measurements and confirmed by the distribution of the  $^{210}\text{Pb}$  radioisotope using the dating techniques of Faure (1986). The geochemical data were used to assess historic and current ecological conditions, identify potential sources, and provide information for evaluating future dredging and disposal options in the estuary.

## Materials and Methods

### Sediment Sampling

Two hundred and thirteen sediment samples from 58 sediment cores were collected throughout the estuary during November/December 1991 (stations 1–59), December 1992 (stations 60–72), and March 1993 (stations 73–89). The study area and core sampling stations are shown in Figure 1. With the exception of sediments collected from the Port Newark and Port Elizabeth navigation channels in Newark Bay, sampling stations were located outside of designated ship navigation channels in stable depositional reaches to minimize the impacts associated with dredging activities and ship traffic, and to obtain undisturbed sedimentation profiles. The selection of sampling locations was biased toward areas of apparent high sediment accumulation and current or historical industrial activity. At most locations, cores ranging in length from 1.5 to 4.5 m were collected. However, due to anticipated high rates of sediment accumulation in the lower Passaic River (IT 1986), 6.0 m cores were collected below the Jackson Street Bridge (stations 13–22) during November/December 1991 sampling.

At all stations, except 1, 2, 5, and 49, a Model 1400 Vibracore unit equipped with a 1.5, 3.0, 4.5, or 6.0 m stainless steel core barrel and Lexan liner was used to obtain sediment cores from water depths averaging less than 3.0 m to as much as 8.0 m. A portable Vibracore unit with a 3.0 m, unlined, aluminum core tube was used at stations 1, 2, 5, and 49. All field sampling and sample processing equipment used in the November/December 1991 sampling (core barrels and Lexan liners) was rinsed with acetone and hexane (50:50), analyte-free trichloroethylene, and analyte-free deionized water prior to the collection of sediment cores. In the 1992 and 1993 sampling events, decontamination of sampling equipment consisted of a hexane rinse, followed by isopropanol and deionized water. Field equipment rinse samples were utilized to verify the levels, if any, of residual contamination. Sediment samples were collected from the intact Lexan liner at up to eight depths for chemical (5.0 cm subsection) and radioisotope (2.5 cm subsection) analyses. The outermost layer of sediment was discarded to minimize contamination from vertical mixing of sediments during core collection. In accordance with USEPA Contract

Fig. 1. Sediment sampling locations in the Newark Bay Estuary, New Jersey

Laboratory Program (CLP) requirements, samples were transferred to precleaned amber glass containers, sealed, and maintained at 4°C prior to chemical analysis.

### Chemical Analysis

Sediments were assayed for base/neutral and acid extractable (BNA) organic chemicals by S-Cubed Laboratory (San Diego, CA) using gas chromatography/mass spectroscopy (GC/MS) according to USEPA Method 8310 (USEPA 1991), and for total extractable petroleum hydrocarbons (TEPH) using a modification of SW-848 USEPA Method 8015. For TEPH analyses, sediments were extracted with methylene chloride by sonication and analyzed on a GC equipped with a flame ionization detector. Diesel was used as the calibration standard.

### $^{210}\text{Pb}$ and $^{137}\text{Cs}$ Analyses

Sediments were assayed for  $^{210}\text{Pb}$  and  $^{137}\text{Cs}$  activity by Teledyne Isotopes (Westwood, NJ).  $^{210}\text{Pb}$  activity was determined by radiochemical separation and assay of the  $^{210}\text{Bi}$  daughter product. Sediments were leached with 6M hydrochloric acid together with stable lead and bismuth carriers. Sample filtrate was evaporated, oxidized with nitric acid, and dissolved in 1.8M hydrochloric acid. Using an anion exchange column, lead was eluted with 0.05M hydrochloric acid followed by 9M hydrochloric acid and deionized water. Bismuth was then eluted with 2M sulfuric acid. The bismuth was precipitated as the oxychloride and collected on a 2.5 cm glass fiber filter. Bismuth yield was determined gravimetrically. Beta activity of the bismuth precipitate was measured using a low level, gas-flow proportional counter.  $^{137}\text{Cesium}$  activity was determined by gamma spectral analysis of sediment for at least five hours using lithium-drifted germanium and high purity (>55 cc) germanium detectors. The activity of  $^{137}\text{Cs}$  was measured by integrating its gamma emission at 661.6 KeV.

### Sediment Accumulation Rates

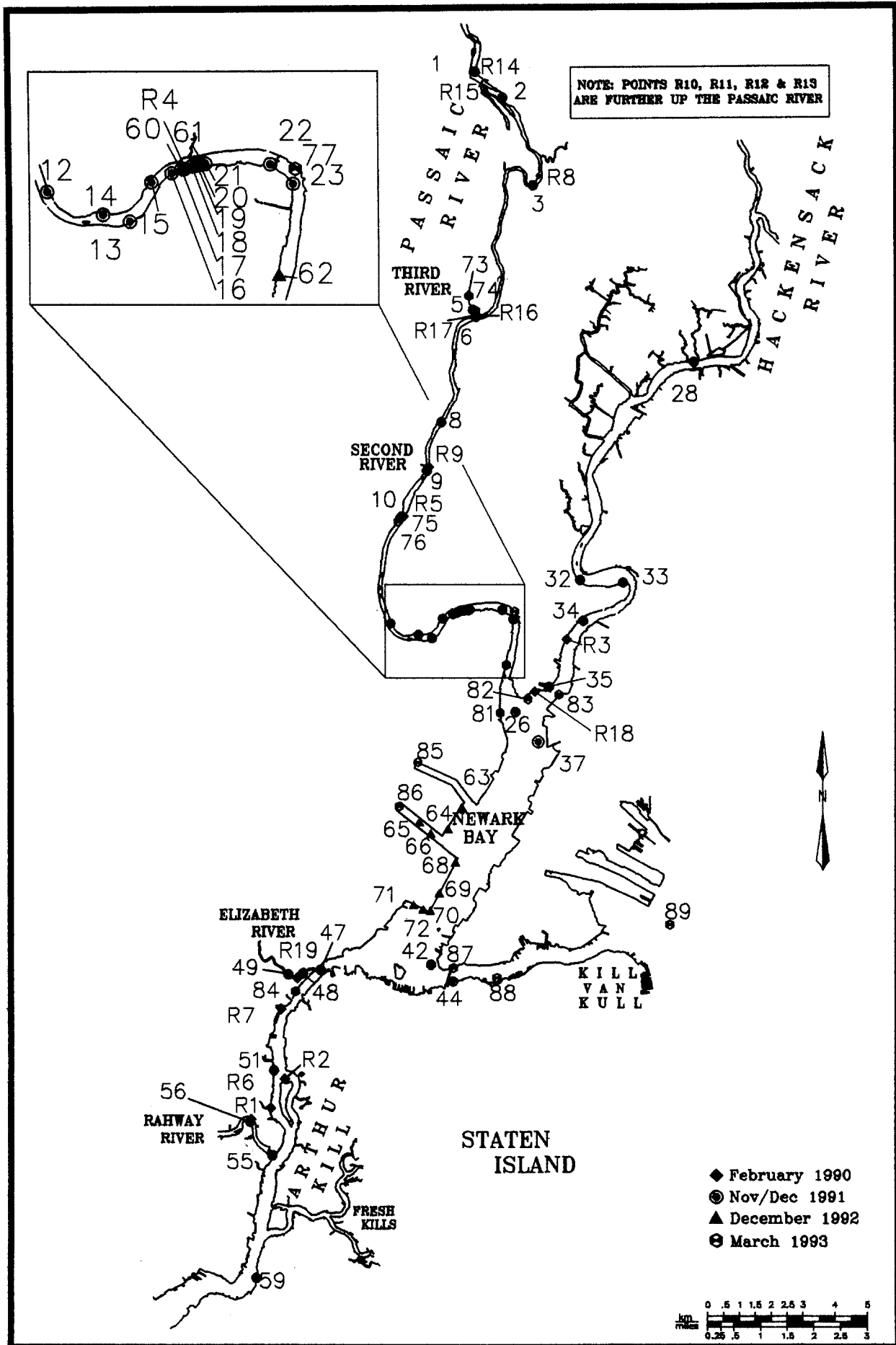
$^{210}\text{Pb}$  analyses were conducted prior to  $^{137}\text{Cs}$  analyses to more accurately predict the depth interval of the  $^{137}\text{Cs}$  1954 horizon in each sediment core. The  $^{210}\text{Pb}$ -derived sediment accumulation rates were estimated using Eqs. (1) and (2) as described by Faure (1986). For  $^{210}\text{Pb}$  activities, the time (t) that correlates to a specific depth (h) is described by

$$t_h = \frac{1}{\lambda} \log\left(\frac{{}^{210}\text{Pb}_A^0}{{}^{210}\text{Pb}_A}\right) \quad (1)$$

where  ${}^{210}\text{Pb}_A^0$  is the unsupported activity (pCi/g) in surface sediments,  ${}^{210}\text{Pb}_A$  is the unsupported activity (pCi/g) at a specific sediment depth,  $\lambda$  is the  $^{210}\text{Pb}$  decay constant ( $3.11 \times 10^{-2} \text{ yr}^{-1}$ ), and  $t_h$  is the estimated age (years) of the sediment at a given depth. The  $^{210}\text{Pb}$ -derived sediment accumulation rate ( $R_{\text{Pb}}$ ; cm/yr) is based on the slope (m) of the regression line of  $\log({}^{210}\text{Pb}_A - {}^{210}\text{Pb}_{\text{As}})$  versus depth as described in

$$R_{\text{Pb}} = \frac{-\lambda}{m} \quad (2)$$

Based on the calculated sediment accumulation rates, approximate dates of deposition were determined for each sample. These approxi-



mate dates were used to determine depth intervals for  $^{137}\text{Cs}$  analyses in each sediment core. The precise depth interval of the 1954 horizon was confirmed by determining the depth of diminished  $^{137}\text{Cs}$  activity along the length of the sediment core. Point estimates of the  $^{137}\text{Cs}$  sediment accumulation rates ( $R_{\text{Cs}}$ ; cm/yr) in each sediment core were determined using

$$R_{\text{Cs}} = \frac{d}{\Delta t} \quad (3)$$

where  $d$  is the depth (cm) of the 1954 horizon and  $\Delta t$  is the period of time from 1954 to the date when the sediment core was collected. The depth of the 1954 horizon ( $d$ ) was assumed to be the midpoint of the depth interval bounded by the deepest depth at which  $^{137}\text{Cs}$  activity was detected and the shallowest depth at which  $^{137}\text{Cs}$  activity was not detected in sediment.

### Data Analysis

Summary statistics (arithmetic mean, standard deviation, median, and upper 90% confidence limit on the mean) were calculated for individual PAHs, total PAHs (tPAH), and TEPH for the Arthur Kill, Hackensack River, Kill Van Kull, Newark Bay, and Passaic River. Statistical summaries for the Newark Bay consisted of measurements from six areas, including Port Elizabeth Channel, Port Elizabeth Pierhead, Port Elizabeth South Channel, Port Newark Channel, the Northern Reach adjacent to Kearny Point, and the Southern Reach adjacent to Shooter's Island. Similarly, summary statistics for the Arthur Kill include Elizabeth River and Rahway River data. Total PAHs were calculated as the sum of the 19 individual PAHs. The upper 90% confidence limit on the mean was determined for individual PAHs, tPAH, and TEPH and compared to specific concentrations found at each depth and location. Chemical concentrations exceeding the 90% upper confidence limit were considered significantly elevated above the mean concentration for the purposes of evaluating inputs from point and non-point sources. Historical trends were evaluated by grouping PAH and TEPH results into 10-year intervals, calculating the mean PAH and TEPH concentrations for each decade, and plotting concentration against time. All statistical analyses were performed using one-half the analytical detection limit for non-detected values, as described by Haas and Scheff (1990).

### Assessment of Aquatic Hazards

As part of ongoing studies associated with the National Oceanic and Atmospheric Administration's (NOAA) National Status and Trends Program, NOAA has derived effects-range low (ER-L) and effects-range median (ER-M) benchmark sediment toxicity values for predicting the potential for adverse effects of persistent chemicals (Long and Morgan 1991). For 18 PAHs, NOAA determined the distribution of sediment concentrations that were associated with any degree of toxicity to freshwater, marine, and estuarine organisms. The 50th percentile of the distribution was identified as the effects range-median (ER-M) value and the 10th percentile of the distribution was identified as the effects range-low (ER-L) value. Although these values are not intended for use as regulatory guidelines, they provide useful benchmarks for assessing the potential toxicity of contaminated sediments. The NOAA ER-M and ER-L values used in this analysis are presented in Table 1.

Using NOAA ER-M values, hazard ratios were calculated for individual PAHs and tPAH for both mean surface sediment concentrations and for mean sediment concentrations across the entire core depth. A hazard ratio of greater than 1 indicates that chemical concentrations may pose a potential hazard to aquatic organisms. Surface sediments were evaluated separately to assess current potential hazards to aquatic organisms. Hazard ratios were calculated for buried sediments to assess the possible impacts associated with dredging and other distur-

**Table 1.** Benchmark sediment quality values (mg/kg) proposed by NOAA for polycyclic aromatic hydrocarbons (PAHs)

Chemical <sup>a</sup>	United States NOAA	
	ER-L values	ER-M values
Acenaphthene	0.15	0.65
Anthracene	0.085	0.96
Benzo(a)anthracene	0.23	1.6
Benzo(a)pyrene	0.40	2.5
Chrysene	0.40	2.8
Dibenzo(a,h)anthracene	0.06	0.26
Fluoranthene	0.60	3.6
Fluorene	0.035	0.64
2-Methylnaphthalene	0.065	0.67
Naphthalene	0.34	2.1
Phenanthrene	0.225	1.38
Pyrene	0.35	2.2
Total PAHs	4	35

<sup>a</sup>NOAA has not derived toxicity values for six of the PAHs evaluated in this study

bances of these sediments. Generally, when dredging has occurred in the Newark Bay Estuary, the upper Passaic River channel has been dredged to approximately 3 m, the middle Passaic River channel to approximately 5 m, and the lower Passaic River channel to a depth of 6–9 m. The lower Hackensack River channel, below the turning basin, has typically been dredged to a depth of 9 m (ACOE 1988). Therefore, all sediment sample depths evaluated in the present investigation were included in the evaluation of buried sediments and the effects of dredging.

### Results

The range of PAH and TEPH concentrations varied considerably, both vertically and spatially, throughout the estuary as shown in Table 2 (Arthur Kill and Hackensack River), Table 3 (Kill Van Kull and Newark Bay), and Table 4 (Passaic River). The very high standard deviations for many of the chemicals and waterways is indicative of the large degree of variation in the data. For example, the concentration of phenanthrene in the Arthur Kill varied by over 2 orders of magnitude, ranging from 0.36 to 89 mg/kg (Table 5). Similarly, the concentrations of tPAH in the Arthur Kill ranged from 0.31 to 420 mg/kg with a mean concentration of 37 mg/kg (s.d., 81.7 mg/kg). In contrast to the relatively high tPAH concentrations found in Arthur Kill sediments, the concentration of tPAH in Hackensack River sediments ranged from 2.2 to 71 mg/kg. Kill Van Kull sediments were found to be contaminated with relatively lower concentrations of tPAH ranging from 1.9 to 36 mg/kg. The levels of tPAH in Newark Bay (0.22–400 mg/kg) were comparable to Arthur Kill concentrations. The Passaic River was found to be the most highly contaminated of the five waterways examined. Total PAH concentrations in sediment ranged from 0.22 to 8,000 mg/kg (Table 5) with a mean concentration of 145 mg/kg (s.d., 739 mg/kg; Table 4).

The frequency of detection for many of the compounds was highly variable due, in part, to a high degree of variability in detection and quantitation limits. The relatively high detection limits reported for the various PAHs were attributed to matrix

**Table 2.** Polycyclic aromatic hydrocarbons (PAHs) and petroleum hydrocarbons (mg/kg, d.w.) in Arthur Kill and Hackensack River sediments

Chemical	Arthur Kill (n = 24)			Hackensack River (n = 12)		
	Median	Mean $\pm$ s.d.	90% UCL	Median	Mean $\pm$ s.d.	90% UCL
Acenaphthene	0.75	2.4 $\pm$ 6.88	4.3	0.41	0.74 $\pm$ 0.78	1.0
Acenaphthylene	0.65	0.73 $\pm$ 0.626	0.90	0.40	0.54 $\pm$ 0.32	0.66
Anthracene	0.83	2.3 $\pm$ 6.12	3.9	0.96	1.3 $\pm$ 1.22	1.8
Benzo(a)anthracene	0.83	1.6 $\pm$ 2.46	2.3	1.3	1.8 $\pm$ 1.55	2.4
Benzo(a)pyrene	0.80	1.4 $\pm$ 2.19	2.0	1.3	1.8 $\pm$ 1.51	2.4
Benzo(b)fluoranthene	0.76	1.3 $\pm$ 1.77	1.8	0.82	1.6 $\pm$ 1.53	2.2
Benzo(g,h,i)perylene	0.62	0.72 $\pm$ 0.528	0.86	0.96	1.1 $\pm$ 0.613	1.3
Benzo(k)fluoranthene	0.77	1.3 $\pm$ 1.84	1.8	0.90	1.3 $\pm$ 0.827	1.6
Carbazole	0.70	0.78 $\pm$ 0.530	0.93	0.65	0.60 $\pm$ 0.258	0.70
Chrysene	0.91	1.9 $\pm$ 2.79	2.6	1.7	2.1 $\pm$ 1.89	2.9
Dibenzo(a,h)anthracene	0.70	0.68 $\pm$ 0.274	0.8	0.58	0.56 $\pm$ 0.287	0.68
Dibenzofuran	0.65	0.88 $\pm$ 1.12	1.2	0.58	0.57 $\pm$ 0.280	0.68
Fluoranthene	1.6	5.2 $\pm$ 13.2	8.7	3.1	3.7 $\pm$ 3.26	5.0
Fluorene	0.75	1.4 $\pm$ 3.34	2.3	0.52	0.72 $\pm$ 0.56	0.93
Indeno(1,2,3-cd)pyrene	0.56	0.74 $\pm$ 0.625	0.91	0.91	1.0 $\pm$ 0.648	1.3
2-Methylnaphthalene	0.70	1.6 $\pm$ 4.36	2.8	0.59	0.63 $\pm$ 0.42	0.80
Naphthalene	0.75	3.0 $\pm$ 10.9	5.9	0.60	0.77 $\pm$ 0.77	1.1
Phenanthrene	0.80	5.8 $\pm$ 18.0	11	1.5	2.7 $\pm$ 3.33	4.0
Pyrene	1.6	3.6 $\pm$ 5.59	5.1	2.6	3.0 $\pm$ 2.58	4.1
tPAH <sup>a</sup>	17	37 $\pm$ 81.7	59	23	27 $\pm$ 19.3	34
TEPH <sup>b</sup>	205	703 $\pm$ 1184	1020	106	119 $\pm$ 86.89	153

<sup>a</sup>PAH = polycyclic aromatic hydrocarbon<sup>b</sup>TEPH = total extractable petroleum hydrocarbon**Table 3.** Polycyclic aromatic hydrocarbons (PAHs) and petroleum hydrocarbons (mg/kg, d.w.) in Kill Van Kull and Newark Bay sediments

Chemical	Kill Van Kull (n = 12)			Newark Bay (n = 45)		
	Median	Mean $\pm$ s.d.	90% UCL	Median	Mean $\pm$ s.d.	90% UCL
Acenaphthene	0.40	0.41 $\pm$ 0.204	0.49	0.60	2.3 $\pm$ 5.45	3.4
Acenaphthylene	0.40	0.45 $\pm$ 0.233	0.55	0.55	1.5 $\pm$ 4.14	2.3
Anthracene	0.40	1.3 $\pm$ 2.58	2.3	0.55	2.4 $\pm$ 5.57	3.5
Benzo(a)anthracene	0.57	0.94 $\pm$ 0.832	1.3	0.62	2.2 $\pm$ 4.74	3.1
Benzo(a)pyrene	0.52	1.0 $\pm$ 0.849	1.3	0.56	1.7 $\pm$ 4.22	2.5
Benzo(b)fluoranthene	0.44	1.0 $\pm$ 0.864	1.3	0.61	1.8 $\pm$ 4.26	2.7
Benzo(g,h,i)perylene	0.36	0.52 $\pm$ 0.426	0.69	0.46	1.5 $\pm$ 4.16	2.3
Benzo(k)fluoranthene	0.41	0.92 $\pm$ 0.989	1.3	0.55	1.7 $\pm$ 4.20	2.5
Carbazole	0.40	0.58 $\pm$ 0.505	0.77	0.55	1.7 $\pm$ 4.23	2.5
Chrysene	0.52	1.0 $\pm$ 0.862	1.3	0.65	2.2 $\pm$ 4.90	3.2
Dibenzo(a,h)anthracene	0.40	0.44 $\pm$ 0.233	0.54	0.55	1.5 $\pm$ 4.14	2.3
Dibenzofuran	0.40	0.44 $\pm$ 0.233	0.54	0.60	2.0 $\pm$ 4.60	2.8
Fluoranthene	0.74	1.6 $\pm$ 1.75	2.3	1.0	5.3 $\pm$ 15.0	8.2
Fluorene	0.39	0.44 $\pm$ 0.228	0.53	0.60	2.1 $\pm$ 5.60	3.1
Indeno(1,2,3-cd)pyrene	0.41	0.53 $\pm$ 0.381	0.68	0.49	1.8 $\pm$ 4.31	2.7
2-Methylnaphthalene	0.36	0.44 $\pm$ 0.235	0.53	0.60	1.5 $\pm$ 4.15	2.3
Naphthalene	0.40	0.44 $\pm$ 0.233	0.54	0.55	1.8 $\pm$ 4.44	2.7
Phenanthrene	0.45	0.85 $\pm$ 0.796	1.2	0.55	4.7 $\pm$ 15.8	7.7
Pyrene	0.97	1.5 $\pm$ 1.33	2.0	0.91	3.9 $\pm$ 9.76	5.8
tPAH <sup>a</sup>	7.9	15 $\pm$ 11.9	19	14	44 $\pm$ 98.5	63
TEPH <sup>b</sup>	77.5	370 $\pm$ 797.0	684	149	339 $\pm$ 534.8	441

<sup>a</sup>PAH = polycyclic aromatic hydrocarbon<sup>b</sup>TEPH = total extractable petroleum hydrocarbon

effects associated with significant organic chemical and petroleum contamination. The individual PAHs that were consistently measured at the highest concentrations in the Newark Bay Estuary were acenaphthene, benzo(a)anthracene, benzo(a)pyrene, benzo(b)fluoranthene, benzo(k)fluoranthene, chrysene, fluoranthene, phenanthrene, and pyrene.

Nearly half of the 58 sediment cores collected from the Newark Bay Estuary contained PAH concentrations that were statistically ( $p < 0.05$ ) elevated above the mean concentration for their respective waterways for at least one individual PAH or for tPAH (Table 5). Geographically, these 27 sampling locations were grouped into seven different areas: the Passaic

**Table 4.** Polycyclic aromatic hydrocarbons (PAHs) and petroleum hydrocarbons (mg/kg, d.w.) in the Passaic River

Chemical	Passaic River		
	Median	Mean $\pm$ s.d.	90% UCL
Acenaphthene	0.68	13 $\pm$ 93.0	24
Acenaphthylene	0.65	2.9 $\pm$ 8.86	3.9
Anthracene	0.75	8.3 $\pm$ 47.0	14
Benzo(a)anthracene	1.4	7.4 $\pm$ 30.5	11
Benzo(a)pyrene	1.5	5.5 $\pm$ 20.1	7.9
Benzo(b)fluoranthene	1.4	4.2 $\pm$ 11.2	5.5
Benzo(g,h,i)perylene	0.90	2.8 $\pm$ 6.12	3.5
Benzo(k)fluoranthene	1.2	3.9 $\pm$ 11.2	5.3
Carbazole	0.70	2.3 $\pm$ 6.41	3.1
Chrysene	1.9	7.9 $\pm$ 32.2	12
Dibenzo(a,h)anthracene	0.65	2.2 $\pm$ 5.21	2.8
Dibenzofuran	0.65	2.6 $\pm$ 8.04	3.6
Fluoranthene	3.2	10 $\pm$ 40.2	15
Fluorene	0.65	7.7 $\pm$ 49.3	14
Indeno(1,2,3-cd)pyrene	0.88	3.0 $\pm$ 6.96	3.9
2-Methylnaphthalene	0.70	11 $\pm$ 80.0	21
Naphthalene	0.70	16 $\pm$ 123	31
Phenanthrene	1.8	19 $\pm$ 113	33
Pyrene	3.4	14 $\pm$ 61.9	22
tPAH <sup>a</sup>	1960	145 $\pm$ 739	232
TEPH <sup>b</sup>	294	1520 $\pm$ 5,970	2,230

<sup>a</sup>PAH = polycyclic aromatic hydrocarbon

<sup>b</sup>TEPH = total extractable petroleum hydrocarbon

River, above the Dundee Dam and below Second River; the Hackensack River, adjacent to the Koppers Coke hazardous waste site and the former federal ship terminal at the confluence of Newark Bay; the Kill Van Kull near Bergen Point; the upper portion of the Arthur Kill and in the Elizabeth River adjacent to the Chemical Control Superfund site; and at the end of the Port Newark navigation channel. TEPH concentrations were statistically elevated above mean concentrations for their respective waterways at 20 sampling locations throughout the estuary, which generally correspond to areas with elevated PAH concentrations. Significantly elevated TEPH levels were also found in sediments at Kearny Point, as well as in southern reaches of the Arthur Kill adjacent to Tremley Point and the Fresh Kills Landfill (Table 5, Figure 1). The concentrations of tPAH and TEPH were generally higher in buried sediments than in surface sediments at most locations (Table 5).

Sediment accumulation rates derived from <sup>210</sup>Pb and <sup>137</sup>Cs measurements for 52 of the 58 sediment cores are presented in Table 6. The six sediment cores collected in the Port Newark and Port Elizabeth navigation channels were not assayed for <sup>210</sup>Pb and <sup>137</sup>Cs because sediments in these areas represent only accumulation after the completion of the last dredging project in 1989. Sediment accumulation rates throughout the estuary ranged from 0.41 to 8.9 cm/year. Sediment cores from the lower Passaic River were found to have the highest depositional rates (3.8–8.9 cm/year), confirming those reported in a previous study (IT 1986). The lower Passaic River, between the Pulaski Skyway Bridge and just above the Clay Street Bridge, has not been dredged by U.S. Army Corps of Engineers since 1949 (IT 1986); the high sedimentation rates estimated in the present study indicate that as much as 240 cm or more of sediment has accumulated in the Brown Street storm sewer

outfall area (adjacent to station 17) since that time. The varied distributions of <sup>210</sup>Pb activity in several cores that could not be dated suggest that sediments at these locations are well mixed, precluding a retrospective evaluation of chemical deposition and sediment accumulation.

Using sediment accumulation rates calculated from <sup>137</sup>Cs measurements (Table 6), profiles of mean concentrations of PAHs and TEPH in sediment were determined according to the decade of deposition from pre-1930 to the present in the Passaic River and the Newark Bay Estuary. The results are presented in Figure 2. In both the Passaic River and the estuary, the mean concentrations of tPAH associated with sediments deposited prior to 1940 ranged from 75 to 95 mg/kg. Mean tPAH concentrations declined during the 1940s (45–55 mg/kg), reached their maximum in the 1950s, and then declined during the 1960s and 1970s to pre-1940 levels. Mean concentrations of tPAHs have further declined since the 1980s to levels ranging from 27 to 40 mg/kg. The mean concentration of TEPH in sediments deposited prior to 1960 ranged from 1,000 to 1,300 mg/kg in the Newark Bay Estuary and the Passaic River and peaked during the 1960s (2,300–3,100 mg/kg). Mean TEPH concentrations have declined since the 1970s in both the Passaic River and elsewhere in the estuary to levels ranging from 240 to 280 mg/kg, respectively.

The hazard ratios for individual PAHs present in Newark Bay Estuary sediments are presented in Figure 3. In surface sediments, all 12 PAHs evaluated, except benzo(a)pyrene and chrysene, pose a potential hazard to aquatic organisms, as indicated by the hazard ratios exceeding a value of 1. In general, buried Newark Bay Estuary sediments posed nearly three times more of a toxic hazard than surface sediments. In the Passaic River, buried sediments pose, on average, approximately six times more of a toxic hazard than surface sediments (Figure 3). The high hazard associated with buried sediments in the estuary is attributed primarily to acenaphthene, anthracene, dibenzo(a,h)anthracene, fluorene, 2-methylnaphthalene, naphthalene, and phenanthrene. These same compounds also appear to be responsible for most of the potential toxicity due to PAHs in buried Passaic River sediments. Further assessment of the relative toxicity of sediments in the lower reach of the Passaic River indicates that hazard ratios for tPAH in surface and buried sediments are higher in the lower reach of the river than elsewhere in the Passaic River and the Newark Bay Estuary.

## Discussion

The results of this investigation indicate that sediments throughout the Newark Bay Estuary contain elevated levels of PAHs and TEPH. These findings are consistent with a previous study of surface sediments in the Arthur Kill, Hackensack River, Newark Bay, and the Passaic River (USEPA 1981). In that study, tPAH concentrations exceeded 100 mg/kg in the lower reaches of the Passaic River and above the Dundee Dam (USEPA 1981). The concentrations of PAHs and TEPH exceeding the 90% UCL in some areas of the Arthur Kill and the Passaic River strongly suggest that current and historical levels are the result of point source discharges, rather than nonpoint sources.

Numerous point sources of PAH compounds have been identified in the environment. High levels of PAHs have been







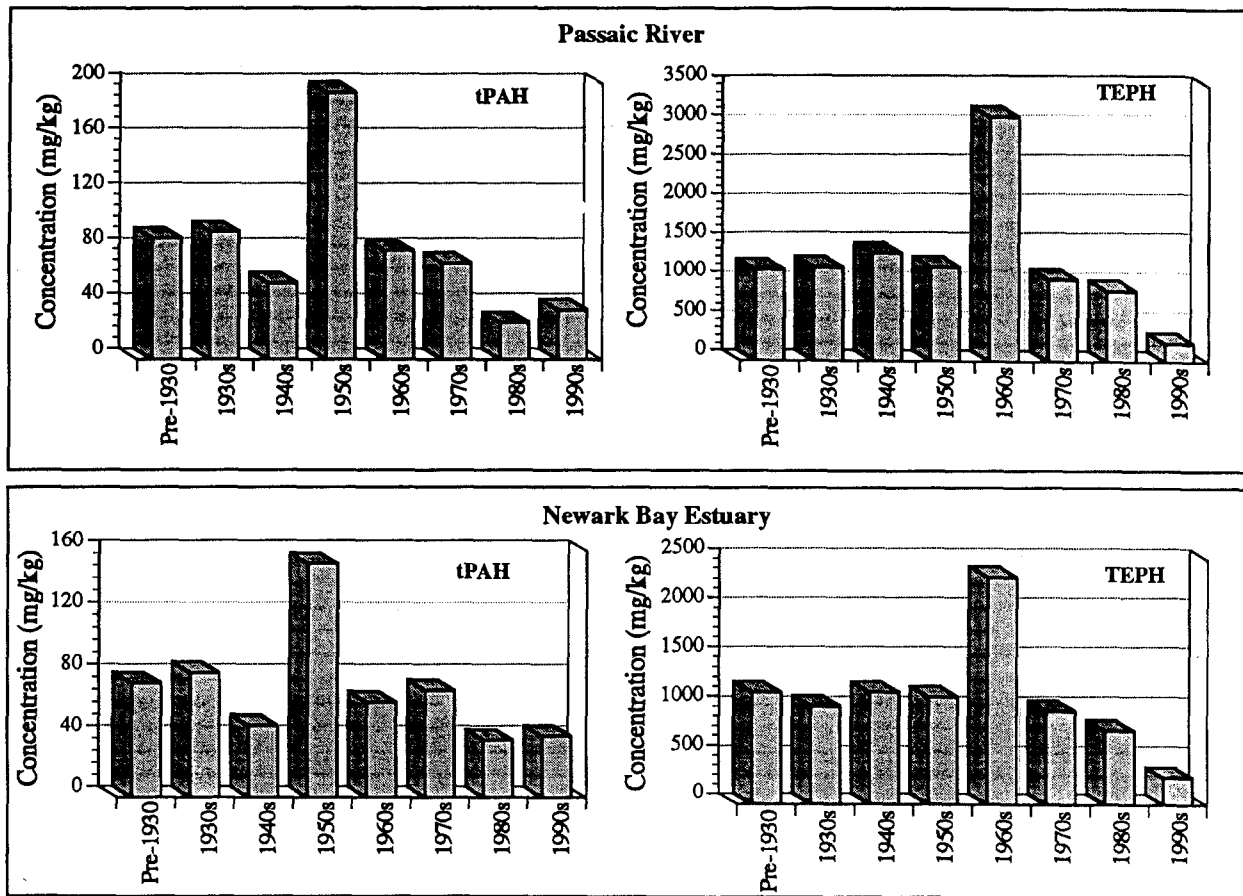




**Table 6.** Sediment accumulation rates derived from <sup>137</sup>Cs and <sup>210</sup>Pb assay results

Station	Rate (cm/yr)	Station	Rate (cm/yr)	Station	Rate (cm/yr)	Station	Rate (cm/yr)
1	0.61	17	8.9	37	<sup>a</sup>	74	1.2
2	1.3	18	3.8	42	<sup>a</sup>	75	3.6
3	6.6	19	5.3	44	>4.8	76	<sup>a</sup>
5	>1.2	20	<sup>a</sup>	47	<sup>a</sup>	77	>3.0
6	4.6	21	6.1	48	2.0	81	>3.0
8	<sup>a</sup>	22	6.1	49	0.66	82	<sup>a</sup>
9	>4.8	23	>7.4	51	<sup>a</sup>	83	2.0
10	3.0	26	0.61	55	0.58	84	2.8
12	0.66	28	0.99	56	6.4	85	5.1
14	0.99	32	<sup>a</sup>	59	3.3	86	10.7
13	0.66	33	<sup>a</sup>	60	3.8	87	<sup>a</sup>
15	0.41	34	<sup>a</sup>	61	>3.3	88	<sup>a</sup>
16	8.6	35	>6.9	62	>3.3	89	<sup>a</sup>

<sup>a</sup>Derivation of rates was not possible due to inability to identify the <sup>137</sup>Cs 1954 horizon, unacceptable scatter of <sup>210</sup>Pb activities, and/or <sup>210</sup>Pb activities near the supported level



**Fig. 2.** Temporal distributions (mean concentrations, mg/kg) of TEPH and tPAH in the Passaic River and Newark Bay Estuary

detected near wood preserving facilities (Zitco 1978; Neff 1980). Creosote used as a wood preservative contains approximately 80–85% PAHs (Ehrlich *et al.* 1982; USEPA 1988). Mueller *et al.* (1989) reported that coal tar creosote consists of a variety of PAHs, predominately naphthalene, 2-methylnaphthalene, phenanthrene, and anthracene as well as 1-methylnaphthalene, fluorene, fluoranthene, chrysene, pyrene, and benzo(a)pyrene. Because of its predominance in coal tar creosote,

naphthalene has been used as a chemical marker for distinguishing inputs from wood preserving industry effluent (USEPA 1982). Catallo and Gambrell (1987) characterized PAH contamination of surface sediments associated with the Bayou Bonfouca Superfund site in Louisiana and found that the most extremely contaminated area had levels ranging from 610 mg/kg for benzo(a)pyrene to as high as 2,931 mg/kg for phenanthrene. The authors estimated that this sample contained

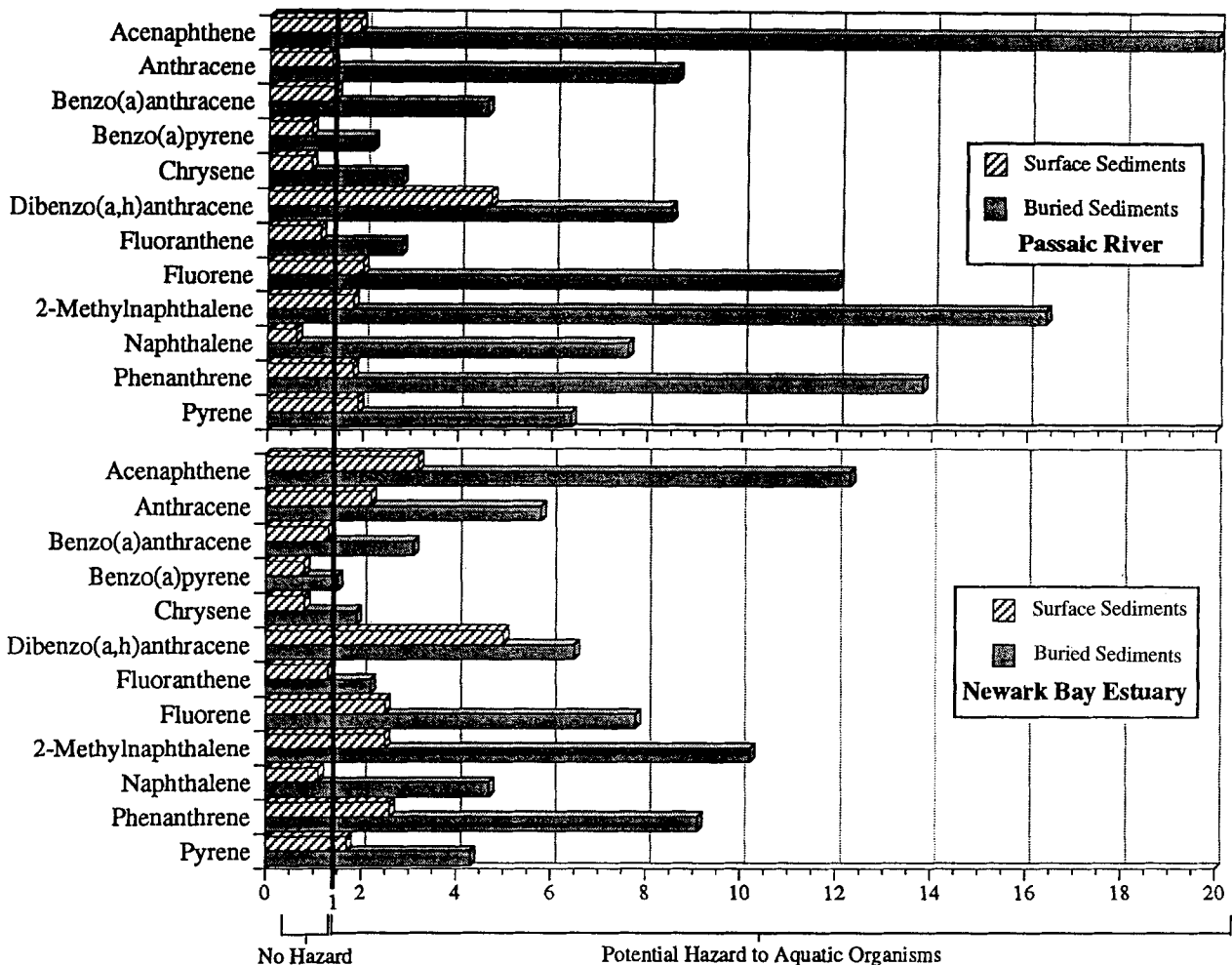


Fig. 3. Comparison of PAH hazard ratios in Passaic River and Newark Bay Estuary buried and surface sediments

approximately 25% creosote based on a comparison of phenanthrene concentrations in the sample and site creosote.

The highest levels of PAHs and TEPH were found in the lower Passaic River in the depositional reach between the Jackson Street bridge and New Jersey Turnpike. The highest concentrations were found near Frank Creek at station 20; at 46 cm, all PAHs except carbazole, were significantly elevated above mean Passaic River concentrations. Very high hazard ratios ranging up to 320 and 330 for 2-methylnaphthalene and acenaphthene were estimated for this location. Unfortunately, radionuclide activities at this station were not sufficient to allow an accurate estimate of sediment accumulation rates. Sediment accumulation at the nearest upstream and downstream sampling locations (station 19, 5.3 cm/yr; station 21, 6.1 cm/yr) suggest that the rate of accumulation at station 20 is approximately 5.8 cm/yr. Based on this estimate, the 46 cm sediments at station 20 were likely deposited in the early 1980s. Frank Creek has been known as a source of oil pollution in the Passaic River since the early 1970s (PVSC 1970); the 6.1% TEPH measured in these sediments (Table 5) strongly suggests that petroleum contamination may be the source of PAHs in this reach of the river.

Clearly, many sources may have contributed to the excessive PAH concentrations in the lower Passaic River and elsewhere in the Newark Bay Estuary. The elevated PAH concentrations measured in the lower reach are likely influenced by numerous

stormwater and sewer outfalls and industrial sites located upstream (Brydon 1974). This is consistent with other heavily used estuaries such as the Southern California Bight, for example, where several sources of PAHs, including treated sewage (Eganhouse *et al.* 1981; SCCWRP 1988), stormwater run-off (Eganhouse and Kaplan 1982; SCCWRP 1988), and oil spills (Hyland *et al.* 1989), provide considerable inputs to the aquatic environment. The Roanoke Street combined sewer outfall (CSO) located adjacent to station 23 and the Brown Street CSO located adjacent to station 17 have been identified as two of Newark's most contaminated discharge sewers (PVSC 1970). This area of the river is also of particular interest because of historical chemical and paint manufacturing dating back to the 1930s. However, given the high levels of TEPH found in PAH-contaminated sediments, petroleum-related sources provide the most plausible explanation for the significantly elevated levels found in the lower Passaic River sediments. Many petroleum terminals are located along the Passaic River and have operated since the first half of the century (RPI 1989). Furthermore, numerous oil spills have been documented throughout the Newark Bay Estuary (NOAA 1982; Gunster *et al.* 1993a, 1993b.). The percentage of PAHs in crude oil, Bunker C oil, and No. 2 diesel oil are about 1%, 4%, and 9% by weight, respectively (NRC 1985), while refinery sludges contain approximately 1 to 3% PAHs (API 1984).

The highest PAH concentrations in Arthur Kill sediments were measured in a core collected at the mouth of the Elizabeth River. Nineteen PAH compounds were significantly elevated ( $p < 0.05$ ) above mean concentrations in surficial sediments at station 47, downstream of the Chemical Control Superfund Site. Fourteen PAHs were detected in surficial sediments at station 48, located in the Elizabeth River upstream from the Chemical Control site, at concentrations about 1 order of magnitude lower than at the confluence. The Chemical Control Superfund Site is a former waste treatment and storage facility that received hazardous wastes from chemical manufacturers, metal finishers and processors, waste haulers, paper companies, home care product manufacturers, oil refineries, and paint and varnish manufacturers (USEPA 1987; NJDEP 1989). The facility operated from 1970 to 1979. During cleanup operations in 1980, an explosion and fire at the site sent drums of hazardous waste into the Elizabeth River. In addition, there had been reports that Chemical Control had dumped wastes directly into the river, and that the sewage system discharged directly into the Arthur Kill (USEPA 1987). Several other industries are also located proximate to that portion of the river, including petroleum refineries and metal processing facilities. For example, a chemical manufacturer and an oil refining company were located directly across the Elizabeth River from the Chemical Control facility during this period (ACOE 1965; USEPA 1987). Although sediments at station 47 could not be dated, sediments from the surface and 20 and 46 cm at station 48 correlate approximately to 1990, 1980, and 1967, respectively.

Hackensack River sediments appear to be moderately contaminated with PAHs and TEPH compared to either the Arthur Kill or the Passaic River. Sediment samples collected from the northern-most sampling station on the Hackensack River (station 28) were relatively uncontaminated compared to Arthur Kill samples and samples collected in southern reaches of the Hackensack River. Sediments collected at station 32, located just downstream of the Standard Chlorine and Koppers Coke hazardous waste sites, had the highest PAH concentrations. Nine PAHs in surface sediments and 11 PAHs and TEPH at 46 cm had concentrations that were significantly elevated ( $p < 0.05$ ) above mean concentrations. The relatively high concentrations of naphthalene found in surface sediments at this location warrant further investigation of discharges from the nearby abandoned Koppers Coke site. The site was a naphthalene refining facility during the 1950s (NJDEP 1989). Radiodating results could not be interpreted for sediments collected at this station.

Among the more interesting findings of this investigation were the high levels of PAHs and TEPH in the 97 cm deep sediments collected above the Dundee Dam on the Passaic River. These sediments, which date back to the mid-1800s, had detectable PAH concentrations ranging from 0.58 to 91 mg/kg and a TEPH concentration of 11,330 mg/kg. Ten PAHs were significantly elevated above mean Passaic River concentrations. Although hazard ratios indicate only a slight potential hazard associated with surface sediments above the Dundee Dam, the hazards associated with the mean concentration in buried sediments is appreciable, particularly with respect to phenanthrene, fluoranthene, acenaphthene, and anthracene. Early industrial activities on the Passaic River are known to have had a significant impact on the aquatic ecosystem. The dam was constructed in 1859 (Brydon 1974), and by the late 1880s, numerous silk-dyeing facilities were discharging millions

of gallons of colored dyestuffs into the river at Paterson, several miles upstream of the Dundee Dam (Brydon 1974; Galishoff 1988).

Although grain size and total organic carbon are known to influence the concentrations of organic chemicals in sediments (McCain *et al.* 1992), the data presented in this study strongly suggest the presence of multiple point sources. In general, the very highest levels of PAH and petroleum hydrocarbon contamination appear in sediments dated to the years ranging from about the 1950s to 1960s, corresponding to periods of heavy industrialization in the region. PAH concentrations in the lower Passaic River were elevated relative to other areas of the estuary even when these data were normalized to total organic carbon.

## Conclusions

Polycyclic aromatic hydrocarbon and petroleum hydrocarbon contamination is widespread in the Newark Bay Estuary. Severely high concentrations in sediments occur in several areas, including the upper Arthur Kill near its confluence with the Elizabeth River, in the Passaic River above the Dundee Dam, and in the lower Passaic River, the lower Hackensack River, and the Port Newark navigation channel in Newark Bay. These chemical hot spots are proximate to suspected point sources and to combined sewer outfalls. PAH contamination is also evident in the Third River and at the confluence of the Rahway River and the Arthur Kill. The historical distribution of PAHs in sediments throughout the estuary is consistent with post World War I and World War II economic expansion and is consistent with the standard waste disposal practices at that time. The results indicate that PAH concentrations in the majority of surface and buried sediments in the Newark Bay Estuary are potentially toxic to aquatic biota. The concentrations of most PAHs in surface sediments pose less of a potential hazard. Although PAHs in lower Passaic River surface sediments pose a low hazard to aquatic biota, severely elevated concentrations in buried sediments raise concerns regarding their potential hazard to aquatic organisms if these sediments are disturbed by dredging or other activities. These data provide a strong impetus for conducting aquatic bioassays on these sediments to determine the relative toxic contribution of each class of chemicals.

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