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Distribution and Possible Sources of Total Mercury in Sediments from the Newark Bay Estuary, New Jersey

T. J. Iannuzzi,[1] R. J. Wenning[2]

[1]ChemRisk®, A Division of McLaren/Hart Environmental Engineering Corporation, 1685 Congress Street, Portland, Maine 04102 USA,
[2]ChemRisk®, A Division of McLaren/Hart Environmental Engineering Corporation, 54 Waterloo Road, North Ryde, NSW 2113, Australia

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A screening survey conducted by investigators of total mercury (Hg) concentrations in freshwater fishes from New Jersey rivers and lakes revealed elevated concentrations (> 0.5 mg/kg wet weight) in the edible tissues of several species (ANSP 1994). The highest Hg concentrations were measured in largemouth bass (*Micropterus salmoides*) and chain pickerel (*Esox niger*) from waterbodies in the northeastern portion of the state that are heavily affected by agriculture, industry, and urbanization, and in waterbodies throughout the state with low pH (< 5.0). Similar results have been reported in saltwater fishes collected from the Raritan River, Passaic River, and Newark Bay (Jacangelo 1977; Ellis et al. 1980; Santoro and Koepp 1986).

Mercury is among the most toxic of metals and one of the most persistent in sediments and organisms. Its presence in various industrial wastes, its liberation in relatively acidic environments, and transformation to various organic and inorganic compounds with differing physiological behaviors and toxicities, make a clearer understanding of its presence in the aquatic environment necessary. At present, sources of Hg contamination in the Newark Bay Estuary are poorly understood, although industrial and municipal point and non-point sources (i.e., sewage sludge, landfill leachates, waste incineration, industrial effluents, and urban runoff) and atmospheric deposition from industrial sources in the midwest and the Delaware Valley are likely contributors (ANSP 1994).

This paper reports the results of total Hg analysis conducted on samples of surface and buried sediments from 72 cores collected from Newark Bay and its major tributaries between 1991 and 1993. The Newark Bay Estuary is surrounded by heavy industry and the New York City - Newark Metropolitan area. The data presented in this study may provide insight on the correlation between the concentrations of Hg reported in fishes and inputs from suspected point- and non-point sources of Hg contamination as revealed in recently deposited and buried sediments. From this information, it may be possible to identify possible current and historical sources of Hg contamination and areas where toxicological testing should be conducted to evaluate possible ecological and human health hazards.

MATERIALS AND METHODS

Seventy-two 1.5-, 3-, and 6-m sediment cores (one core per sampling location) were collected between November 1991 and July 1993 from the Arthur Kill and its

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Correspondence to: T. J. Iannuzzi

tributaries (7 cores, including 4 from the upper Arthur Kill/Elizabeth River, and 3 from the lower Arthur Kill/Rahway River), the Hackensack River (6 cores), the Kill van Kull (5 cores), Newark Bay (15 cores), and the Passaic River (39 cores, including 9 in the upper Passaic River, above its confluence with the Second River, and 30 in the lower Passaic River) (Fig. 1). With the exception of Port Newark and Port Elizabeth, sediment cores were collected outside of federal navigation channels in stable depositional reaches to minimize the impacts associated with dredging activities and ship traffic and to obtain undisturbed sedimentation profiles. Sampling locations were biased towards areas adjacent to active or historical industrial and municipal outfalls, storm drains, and combined sewer overflows.

Sediment cores were collected using a Model 1400 Vibracore unit equipped with either a 3- or 6-m stainless steel core barrel and Lexan liner or a smaller Vibracore unit equipped with a 1.5 m, unlined, aluminum core barrel. Sediment samples were collected from the intact core at up to eight depths (5 cm subsection) for chemical analysis and up to ten depths (2.5 cm subsection) for radioisotope analysis. The outermost layer of sediment was discarded to minimize contamination from vertical mixing of sediments during core collection. Samples were transferred into precleaned amber glass containers, sealed, and maintained at 4°C prior to chemical analysis, in accordance with EPA Contract laboratory Program (CLP) requirements. As indicated in Table 1, surface sediment samples were collected from each core, while a variable number of discrete samples were collected at depth from some or all of the cores. Sample analytical results were averaged for all cores collected from each waterway for the surface and three general depth zones: 20 - 80 cm, 90 - 140 cm, and > 150 cm (Table 1).

Sediment samples were assayed for total Hg by cold vapor atomic absorption spectroscopy (AAS), according to the USEPA Contract Laboratory Program (CLP) Statement of Work for Inorganic Analysis (ILM02.0), Method 7471. A 0.2 g portion of sediment from each sample was digested in a solution of concentrated H_2SO_4 (5 ml) and HNO_3 (2 ml), to which 5 ml of $KMnO_4$ was added to eliminate interferences from sulfides. The solution was autoclaved for 15 minutes at 121°C and 15 lb, cooled, and diluted to 100 ml with water, to which 6 ml of sodium chloride-hydroxylamine sulfate solution was added to reduce the excess permanganate. The digested samples and appropriate standards were aerated from solution by a closed system circulator pump; absorbance at the 253.7 nm wavelength by the Hg vapor was analyzed by AAS. Recoveries were determined from matrix spikes and ranged from 75 percent to 110 percent. The limits of detection for total Hg in sediment ranged from 0.1 to 0.5 mg/kg.

Radioisotope analyses were conducted using the methods described in Wenning et al. 1994. In general, Pb^{210} was assayed by radiochemical separation and beta assay for the ^{210}Bi product, and Cs^{137} activity was determined using gamma spectral analysis. A regression was developed to calculate sediment accretion rates (cm/yr) for each core based on Pb^{210} and Cs^{137} activity. The chemistry samples were then dated based on the calculated accretion rates and depth at which they were collected. Results of Hg analyses from all cores collected throughout the Estuary were averaged by decade to evaluate historical mean concentrations of Hg in sediments, and to compare the historical concentrations to available benchmark sediment quality guidelines developed by Long et al. (1995).

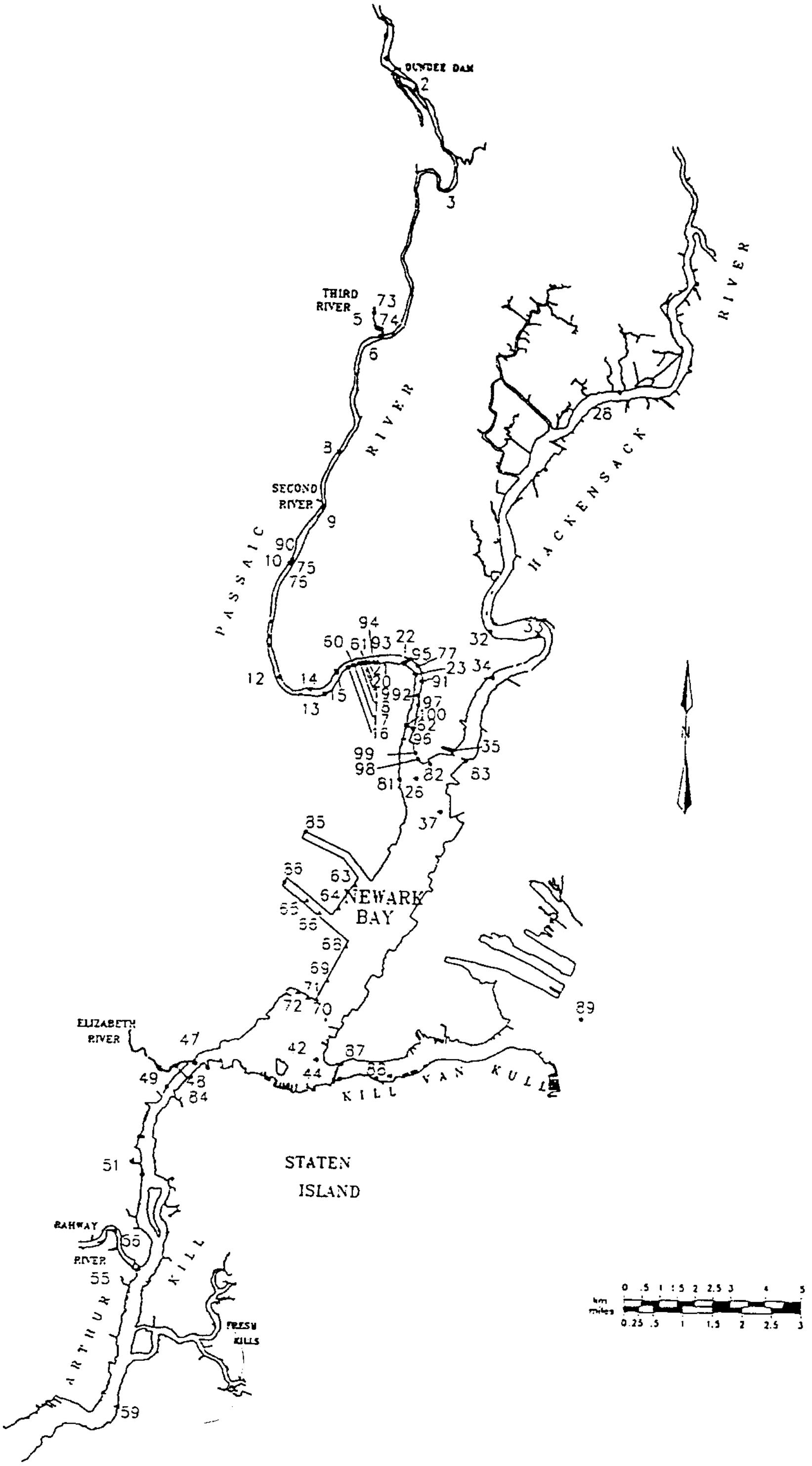

Figure 1. 1991-93 sediment sampling locations in the Newark Bay Estuary

Table 1. Concentrations (mg/kg, dry weight) of total mercury in surface and buried sediments collected from the Newark Bay Estuary between 1991 and 1993 (a)

Waterway	Depth	n	Mean	±	S.D.
Upper Passaic River	Surface	9	2.6	±	3.8
	20-80 cm	12	2.8	±	4.2
	90-140 cm	5	3.5	±	4.3
Lower Passaic River	Surface	30	3.5	±	2.0
	20-80 cm	26	7.5	±	5.8
	90-140 cm	30	7.2	±	6.9
	>150 cm	55	5.2	±	7.7
Hackensack River	Surface	6	6.0	±	2.8
	20-80 cm	6	4.1	±	3.3
	90-140 cm	6	3.3	±	3.6
	>150 cm	1	8.0	±	-
Newark Bay	Surface	15	2.6	±	1.6
	20-80 cm	15	2.1	±	1.7
	90-140 cm	15	2.5	±	3.1
Upper Arthur Kill / Elizabeth River	Surface	4	2.8	±	1.4
	20-80 cm	6	3.4	±	5.6
	90-140 cm	2	2.2	±	2.7
Lower Arthur Kill / Rahway River	Surface	3	5.9	±	3.4
	20-80 cm	3	12.7	±	6.1
	90-140 cm	3	11.6	±	10.9
Kill Van Kull	Surface	5	3.2	±	2.3
	20-80 cm	6	4.4	±	5.1
	90-140 cm	4	0.4	±	0.3

a. Arithmetic mean and standard deviation were calculated assuming nondetect (ND) measurements were equal to the limit of detection; detection limits for individual samples ranged from 0.1 - 0.5 mg/kg.

RESULTS AND DISCUSSION

The concentrations of total Hg in surface (0-5 cm) and buried sediments throughout the Estuary are summarized in Table 1. The number of samples (n) that were analyzed from each core vary between depth interval and waterway, as previously described. The highest concentrations of total Hg in surface sediment were measured in the Hackensack River (mean of 6.0 mg/kg), and the lower Arthur Kill (mean of 5.9 mg/kg). Total Hg concentrations elsewhere in the Estuary were generally less than 3.5 mg/kg.

Total organic carbon (TOC) content of sediment samples collected from various stations and depths throughout the Estuary was consistently low, ranging from 0.01 to 4.9 % with a mean value of 0.8 (±0.8) %. Similarly, Meyerson et al. (1981) determined that the grain size of sediments is similar in major tributaries of Newark Bay. Because of the relative consistencies in TOC and grain size, it is likely that differences in the Hg content of sediments are primarily due to differences in source contributions between waterways.

Elevated concentrations in the upper Arthur Kill and Elizabeth River may be associated with effluents and emissions from several petroleum refining facilities located in the Exxon Bayway industrial complex (Fig. 1). Petroleum refineries and burning of fossil fuels have been shown to be sources of Hg in the aquatic environment (Zoller et al. 1983). The Arthur Kill and Morses Creek are reported to be major sources of metals to southern Newark Bay (Meyerson et al. 1981). Metals in sediments from the lower Arthur Kill and Raritan River may be attributable to these sources, as well as leachate from the Fresh Kills landfill, one of the largest landfills in the world, and ore smelters. The presence of Hg in the Hackensack River has been attributed to a chloralkali facility located on Berry's Creek (Weis et al. 1986). The highest concentration of total Hg in surface sediment (9.3 mg/kg) was measured nearby at station 28. In the lower Passaic River, total Hg concentrations were highest between stations 12 and 23 (Fig. 1). This area has historically been impacted by discharges from combined sewer overflows (CSOs) at Brown Street and Blanchard Street and by discharges from tanneries, scrap-metal yards, and several paint manufacturers (PVSC 1970). Urban runoff, wastewater and treatment plant effluents have been found to be major sources of Hg and other metal to the Newark Bay Estuary (Whipple et al. 1976; Ellis et al. 1980; Mytelka et al. 1993). Recent surveys of metal loading to the NY / NJ Harbor Estuary indicate that municipal discharges may contribute as much as 40 % of the Hg input to the Estuary (HydroQual 1991).

In general, total Hg concentrations found at depth were higher than at the surface (Table 1). The concentrations of total Hg in buried sediments were evaluated using radioisotope dating to investigate historical changes in anthropogenic contributions to the aquatic environment. Temporal profiles (by decade) of total Hg in sediment cores from the Estuary were generated using average sediment accretion rates calculated from ^{210}Pb and ^{137}Cs measurements. The results are shown in Figure 2. Elevated concentrations of Hg were evident in sediments deposited prior to 1940, reached their highest levels during the 1940s, and declined to current levels from the 1960s. These results suggest that more recent inputs have declined relative to historical discharges.

These profiles parallel the industrial evolution of the New York-Newark metropolitan area (Crawford et al. 1994). Although the region has been heavily populated for nearly 200 years, intense industrialization occurred after World War II (Crawford et al. 1994). Mercury enters the environment from various sources including paper factories which use phenyl mercuric acetate, chlorine factories using Hg electrodes, electrical industries, combustion of coal containing mercury, Hg-containing catalysts for manufacturing vinyl chloride, Hg-containing fungicides and pesticides, and Hg coatings for seeds (Cassidy 1978; Zoller et al. 1983; Steinnes 1990); many of these uses occurred in New Jersey (e.g., in the Passaic-Hudson River drainages). In addition to several point sources, suspected sources of Hg in sediments and biota from the Newark Bay Estuary include emissions from

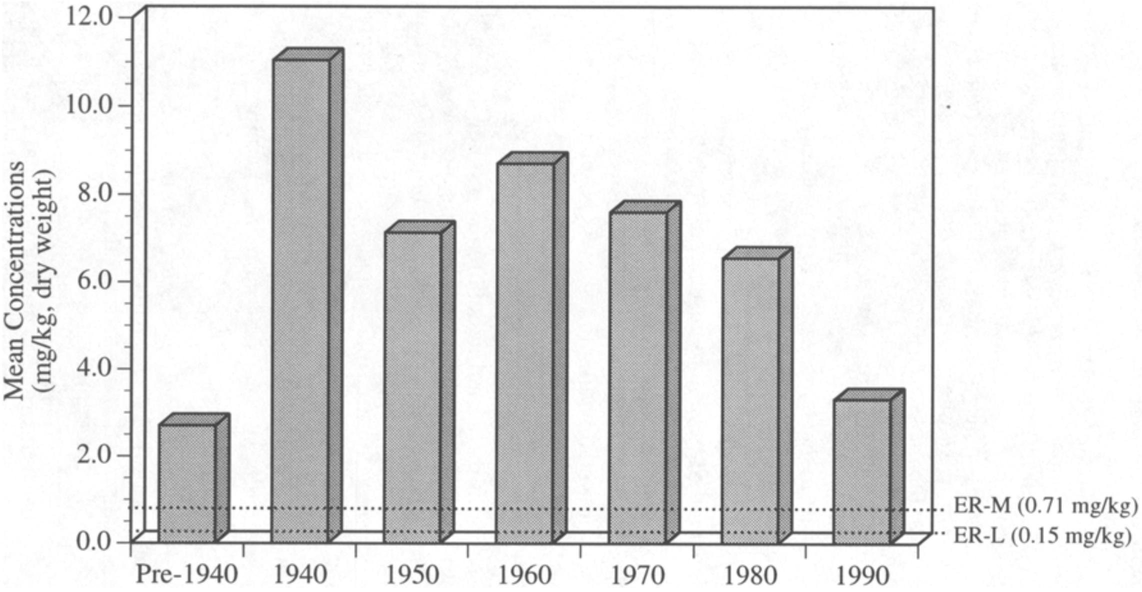

Figure 2. Historical distributions of mean total mercury in surface and buried sediments from the Newark Bay Estuary. Results are compared to effects-range low (ER-L) and median (ER-M) benchmark sediment toxicity values proposed by Long et al. (1995)

CSOs, sewage sludge, and municipal wastewater treatment plant effluents (Mytelka et al. 1973; Whipple et al. 1976; Meyerson et al. 1981). Industries related to the war effort, including paint and pigment formulators, metal plating, and ship building, are likely to have contributed significant amounts of trace metals to the Estuary during the period between 1940 and 1960. The gradual decline of Hg concentrations in sediments deposited after the 1960s may reflect increased regulations of waste discharges, as well as improvements in the functioning of CSOs and waste treatment facilities (Crawford et al. 1994).

Fish accumulate Hg mainly as methylmercury (D'Itri 1990). Since methylmercury is formed from Hg within aquatic systems, the rate of methylation may have a greater effect on the amount of bioaccumulation than the input rate of total Hg. While point source discharges of mercury have decreased, some direct sources may exist and sediments in the biologically active zone (e.g., to about 10 cm in depth) may continue to contain Hg from earlier deposition and act as sources of Hg to fish and other aquatic organisms (Rada et al. 1979).

The results presented here demonstrate that Hg is present in surface sediments throughout the Newark Bay Estuary, typically at elevated concentrations above both the Long et al. (1995) effects-range low (ER-L) and median (ER-M) benchmark sediment toxicity values (0.15 and 0.71 mg/kg, respectively) (Fig. 2). This raises a concern that sediments may be causing adverse effects to aquatic organisms in the Estuary. Although inputs appear to have declined relative to historical sources, elevated Hg concentrations in surface sediments throughout the Estuary suggest ongoing inputs. Additional research will be necessary to identify potential sources and to evaluate their contribution to Hg contamination in these waterways.

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