

Article

Nutrients and Heavy Metals Contamination in an Urban Estuary of Northern New Jersey

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Abstract: The Newark Bay Estuary in northern New Jersey contains one of the largest urban wetland complexes in the United States, but the majority of the wetlands and habitats have been lost due to urbanization and industrialization. Field and laboratory research was conducted to understand the impacts of human activities on the biogeochemistry of nutrients and heavy metals in the urban estuary. Concentrations of dissolved nutrients such as nitrate, ammonia, and phosphate were higher in the Hackensack River than in the Passaic River or the Newark Bay, while the Hackensack River was more deficient in dissolved oxygen. Sediment oxygen demand and mobilization of nutrients were higher in sediments with higher organic matter content as a result of microbial decomposition of organic matter. Heavy metals (Cr, Cu, Pb, and Zn) and organic matter were more enriched in finer sediment grains such as silt and clay. There were positive correlations among heavy metals as well as organic matter in sediments. The results suggest that fine grained sediments, which can be readily suspended and transported during tidal cycles, may play a significant role in biogeochemical cycling of nutrients and heavy metals in the urban estuary. It appears that the current sources of nutrients and heavy metals in the water and sediment of the Newark Bay Estuary are mainly domestic effluents from sewage treatment plants during non-storm periods as well as combined sewer overflows during storm events, but further research including more frequent and pervasive water and sediment quality monitoring during dry and wet periods is needed.

Keywords: nutrients; heavy metals; Newark Bay estuary; sewage treatment plants; combined sewer overflows

1. Introduction

Estuaries are zones of biogeochemical and biological mixing and are considered to be one of the most highly productive ecosystems [1]. Estuarine wetlands combine chemical transformation and transport processes in the biogeochemical cycling of nutrients and trace metals [2]. Nitrogen, phosphorus, and other nutrients are essential for growth and reproduction of producers in estuarine wetlands. However, prolonged periods of excess nutrients may lead to unsustainable primary production and eutrophication. In addition, toxic trace metals such as arsenic, copper, and lead may have complex effects on primary and secondary production in estuarine systems [3].

The Newark Bay Estuary including the Newark Bay, the Hackensack River, and the Passaic River in northeastern New Jersey is a part of the Hudson-Raritan Estuary (HRE), which is one of the largest urban estuaries in the United States [4]. The urban estuary is located in the heavily industrialized and densely populated New York–Newark–Jersey City metropolitan area. Historically, the coastal and marine waters of the estuary were known to support diverse populations of fish and shellfish, but the metropolitan region surrounding the urban estuary has been heavily impacted by both industrial and municipal activities [5]. During the 19th and 20th centuries, factories dumped large quantities of untreated waste into the lower portion of the Passaic and Hackensack Rivers to Newark Bay, which has left hotspots of toxic contaminants throughout the estuary's ecosystem [6,7]. Numerous current

and former industrial facilities and commercial landfills including National Priorities List (NPL) sites (e.g., Diamond Alkali Superfund Site and PJP Landfill), as well as combined sewer overflows (CSOs) have discharged various contaminants including dioxins and furans, polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), pesticides, metals, and nutrients to the urban estuary [7–9]. As a result, the estuary has suffered extensive losses in wetland habitat and aquatic vegetation communities over the past two centuries. Although the overall health of the estuary has improved due to enactment of the 1972 Clean Water Act and the decline of industry along the estuary, the Newark Bay Estuary still suffers from substantial degradation of water quality and persistent contaminants in bottom sediments, which pose a significant threat to the health of humans and ecosystems [7,10,11].

Newark Bay is a tidal bay at the confluence of the Passaic and Hackensack Rivers. The Newark Bay receives freshwater contributions mainly from the Passaic and Hackensack Rivers at the northern end of the Bay, while additional freshwater contributions are from a number of other tributaries, combined sewer overflows (CSOs) and storm water outfalls (SWOs) distributed throughout the Bay [12]. The lower Hackensack River spans across Bergen and Hudson counties, New Jersey, runs north to south, and empties into Newark Bay. The lower Hackensack River Estuary beginning from the Oradell Reservoir, drains an urbanized, 218-km² watershed dominated by the Hackensack Meadowlands [13]. Due to large reservoir withdrawals, the long-term average freshwater inflow to the lower Hackensack River is insignificant compared to the mean tidal discharge at the mouth. The Lower Passaic River is a 17-mile tidal stretch from the Dundee Dam to the river's mouth at Newark Bay. The Passaic River drains urbanized watersheds that have experienced some of the greatest population densities and industrial activities in the United States for more than a century [10].

Field and laboratory research was conducted to investigate water and sediment quality of the Newark Bay Estuary in northern New Jersey. While a number of previous studies including the reports for the U.S. Environmental Protection Agency and the Contamination Assessment and Reduction Project (CARP) have extensively investigated dioxins, polycyclic aromatic hydrocarbons (PAHs), pesticides, polychlorinated biphenyls (PCBs), and heavy metals in sediments from the Newark Bay Estuary [7–9,14–17], this study focused primarily on potential impacts of human activities on biogeochemistry of nutrients in water and sediments of the urban estuary. There has been on-going water quality monitoring of dissolved oxygen and nutrients in the New York–New Jersey Harbor by the New Jersey Harbor Dischargers Group (NJHDG), the EPA NY/NJ Harbor and Estuary Program (HEP), and the Meadowlands Environmental Research Institute [18–20]. This study is relevant to urban estuaries of many industrialized and developing countries in the world, where sewage wastes contribute excessive amounts of nutrients into coastal waters, which may lead to water quality degradation such as eutrophication [21–23]. This study also investigated geochemical distribution of heavy metals in surficial sediments of the urban estuary. The contamination of sediments or soils with heavy metals by anthropogenic activities including industrial and domestic wastes discharge poses long-term risk to the health of ecosystems because heavy metals are harmful to humans and wild species, and tend to bioaccumulate in the food chain [24–27]. Due to the disturbance and acceleration of biogeochemical cycling of metals by humans, soils and sediments in urban environments may be enriched with heavy metals such as copper (Cu), zinc (Zn), and lead (Pb) above background values [25]. The objectives of this study were to determine the level of environmental contamination in the urban estuary and to assess the impacts of human activities on biogeochemical cycling of nutrients and heavy metals in water and sediment.

2. Materials and Methods

Water and sediment samples were collected in summer 2015 and 2016 along the Hackensack and Passaic Rivers, as well as the Newark Bay in northeastern New Jersey during non-storm periods with no significant rain event for a week prior to sampling (Figure 1). Sampling locations were selected to cover highly urbanized and industrialized area such as Newark and Jersey City to suburban areas in the northern parts of the estuary. In summer 2016, water samples from the Hackensack River

and the Newark Bay were collected during the transition from high tide to low tide, whereas water samples from the Passaic River were collected during the transition from low tide to high tide (Table 1). In summer 2015, water samples were collected from the Hackensack and Passaic Rivers during the transition from low tide to high tide, while water samples were collected from the Newark Bay during high tide (Table 2). Water samples for water quality monitoring were collected in a 1-L HDPE container at approximately 0.5–1 m depth. Water quality parameters such as pH and electrical conductivity (EC) of water samples were measured using a portable pH-EC meter (HQ40d, Hach Company, Loveland, CO, USA). Dissolved oxygen was determined using CHEMets test kits (CHEMetrics, Inc., Midland, VA, USA) immediately after sampling. The CHEMets test kit for dissolved oxygen employs the indigo carmine method. The concentrations of dissolved oxygen were determined by visually comparing the produced color with a comparator included in the kit. Water samples were collected in 250-mL HDPE bottles without filtration for analyses of nitrate (NO_3^-), ammonia (NH_4^+ or NH_3), and phosphate (PO_4^{3-}) in the field or laboratory on the same day. In summer 2015, additional water samples were collected in 20 mL HDPE vials after filtration through 0.45 μm membrane syringe filters for analysis of sulfate and chloride using Ion Chromatography. Water quality data of pH, EC, DO, and nutrients for the summer 2015 samples are reported elsewhere [28]. Geographic coordinates (latitudes and longitudes) for each sampling location were recorded using a smart phone app in the field.

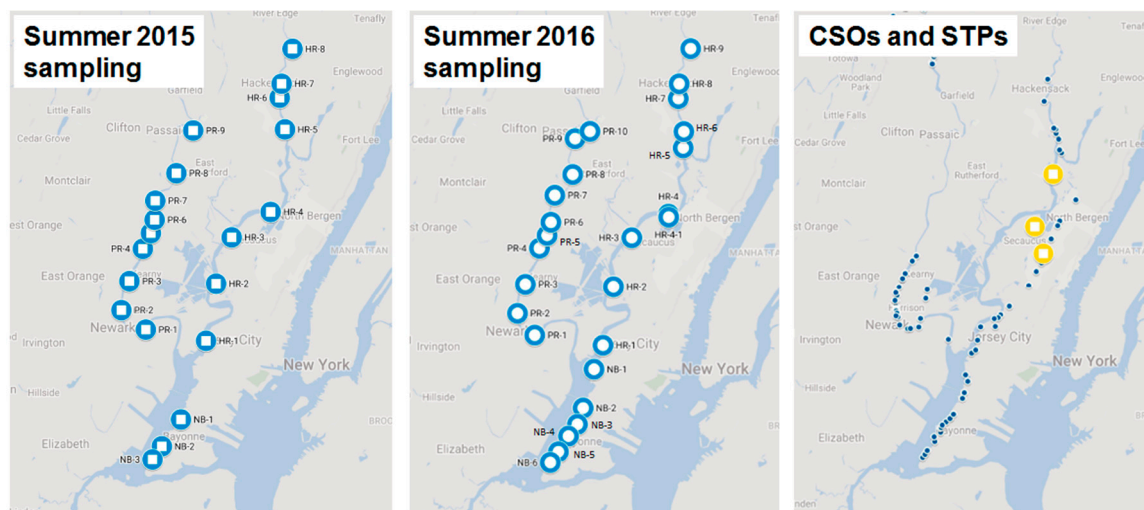


Figure 1. Sampling locations of water and sediment samples from the Newark Bay Estuary (NB: Newark Bay; HR: Hackensack River; and PR: Passaic River) in summer 2015 and 2016, as well as locations of combined sewer overflows (CSOs) outfalls (small blue circles) and sewage treatment plants (STPs; large yellow circles). Note that sediment samples were collected not from all locations (see Tables 4 and 5 for sampling locations for sediments).

Table 1. Water quality data for samples collected from the Hackensack River (HR), the Passaic River (PR), and the Newark Bay (NB) in summer 2016. Distance (km) indicates the distance from NB-6 located near the mouth of the Newark Bay. Water level data (tidal datum: mean lower low water) at Bergen Point West Reach, NY, are from National Oceanic and Atmospheric Administration (<https://tidesandcurrents.noaa.gov>).

Sample ID	LAT	LONG	Distance (km)	pH	EC (mS/cm)	DO (mg/L)	Nitrate (mg/L)	Ammonia (mg/L)	Phosphate (mg/L)	Water Level (m)
HR-1	40.727	74.096	8.8	7.91	27.20	7	9.7	0.24	0.67	1.92
HR-2	40.763	74.088	15.6	7.79	24.00	5	12.4	0.39	0.73	1.86
HR-3	40.792	74.073	19.6	7.79	13.81	5	14.2	1.80	1.30	1.62
HR-4	40.806	74.044	22.8	7.68	17.85	3	13.3	0.90	1.18	1.37
HR-4-1	40.805	74.043	22.9	7.67	16.24	3	13.3	0.75	0.93	1.34
HR-5	40.847	74.032	22.9	7.81	9.72	3	12.4	2.83	1.36	1.08
HR-6	40.856	74.031	28.1	7.78	8.33	4	14.2	2.76	1.20	0.93
HR-7	40.876	74.036	29.2	7.76	5.60	4	14.2	2.76	1.01	0.74
HR-8	40.885	74.035	32.5	7.74	4.30	5	11.5	1.41	0.78	0.60
HR-9	40.907	74.026	33.6	8.07	1.23	6	8.0	0.15	0.43	0.45
Min				7.67	1.23	3	8.0	0.15	0.43	
Max				8.07	27.20	7	14.2	2.83	1.36	
AVG				7.80	12.83	5	12.3	1.40	0.96	
PR-1	40.733	74.151	12.9	6.74	8.15	4	6.5	<0.02	0.86	0.37
PR-2	40.746	74.165	15.0	7.21	3.36	5	7.7	0.03	0.87	0.49
PR-3	40.764	74.159	17.1	7.57	1.28	5	10.0	0.04	0.72	0.66
PR-4	40.786	74.147	19.8	7.64	0.64	7	10.0	<0.02	0.57	0.78
PR-5	40.794	74.141	20.8	7.61	0.62	5	9.4	<0.02	0.67	0.85
PR-6	40.802	74.138	21.7	7.62	0.60	7	8.3	<0.02	0.61	0.94
PR-7	40.818	74.135	23.7	7.65	0.62	8	9.4	<0.02	0.89	1.18
PR-8	40.830	74.121	25.8	7.73	0.63	9	9.4	<0.02	0.83	1.27
PR-9	40.852	74.119	28.3	7.80	0.62	8	10.6	0.04	0.99	1.40
PR-10	40.857	74.107	30.6	7.86	0.65	9	8.3	<0.02	0.85	1.48
Min				6.74	0.60	4	6.5	0.03	0.57	
Max				7.86	8.15	9	10.6	0.04	0.99	
AVG				7.54	1.72	7	9.0	0.02	0.79	
NB-1	40.712	74.104	7.0	7.66	29.7	5	10.6	0.04	0.46	1.31
NB-2	40.689	74.112	4.3	7.73	35.7	8	9.7	0.10	0.39	1.14
NB-3	40.679	74.117	3.2	7.78	35.2	6	9.7	0.06	0.29	0.99
NB-4	40.672	74.124	2.2	7.83	35.8	7	11.5	<0.02	0.35	0.74
NB-5	40.662	74.132	0.9	7.96	36.0	8	8.9	0.05	0.2	0.66
NB-6	40.656	74.139	0.0	8.04	35.9	5	8.9	0.03	0.28	0.55
Min				7.66	29.7	5	8.9	0.03	0.20	
Max				8.04	36.0	8	11.5	0.10	0.46	
AVG				7.83	34.7	7	9.9	0.05	0.33	

Dissolved nitrate, ammonia, and phosphate were measured using a Hach DR/890 portable colorimeter (Hach Company, Loveland, CO, USA), which is a microprocessor-controlled, light emitting diodes-sourced filter photometer suitable for colorimetric testing in the laboratory or the field. Nitrate concentrations were determined by the cadmium reduction method, while ammonia concentrations were determined by the salicylate method. Phosphate concentrations were determined by the molybdenum blue method. Standard solutions from Hach Company (phosphate standard solution, 50 mg/L as PO₄ and nitrogen-ammonia standard solution, 10 mg/L as NH₃-N) were used to check the accuracy of colorimetric measurements, which showed less than $\pm 10\%$ error in measurements for ammonia and phosphate. Because of interference in nitrate measurements by high chloride in water samples, nitrate concentration was determined by the standard addition method using nitrogen-nitrate standard solution (100 mg/L as NO₃-N) from Hach Company. Dissolved sulfate and chloride concentrations in water samples collected in summer 2015 were determined using Ion Chromatography (Thermo Scientific Dionex ICS-5000+, Waltham, MA, USA) following a standard EPA protocol in the NJCU Environmental Geochemistry Laboratory. Errors in measurements for sulfate and chloride were less than $\pm 5\%$.

Table 2. Concentrations of chloride and sulfate in water samples collected from the Hackensack River, the Passaic River, and the Newark Bay in summer 2015. Distance (km) indicates the distance from NB-6 located near the mouth of the Newark Bay. Water level data (tidal datum: mean lower low water) at Bergen Point West Reach, NY, are from National Oceanic and Atmospheric Administration (<https://tidesandcurrents.noaa.gov>).

Water Body	Sample ID	LAT	LONG	Distance	Chloride	Sulfate	Water Level
				(km)	mg/L		m
Hackensack River	HR-1	40.7273	74.0963	8.8	7674	1014	0.33
	HR-2	40.7625	74.0881	15.6	3868	531	0.41
	HR-4	40.8065	74.0436	22.8	2666	317	0.95
	HR-5	40.8572	74.0319	28.1	1014	117	1.27
	HR-6	40.8766	74.0361	29.2	605	69	1.44
	HR-7	40.8853	74.0347	32.5	554	60	1.49
	HR-8	40.9067	74.0257	33.6	276	29	1.54
	PR-1	40.7342	74.1453	12.9	402	56	0.13
Passaic River	PR-2	40.7462	74.1652	15.0	157	28	0.05
	PR-3	40.7640	74.1588	17.1	161	24	0.06
	PR-4	40.7839	74.1478	19.8	141	23	0.12
	PR-5	40.7935	74.1412	20.8	140	23	0.29
	PR-6	40.8017	74.1381	21.7	132	21	0.40
	PR-7	40.8134	74.1377	23.7	123	22	0.58
	PR-8	40.8303	74.1205	25.8	124	22	0.83
	PR-9	40.8565	74.1067	28.3	86	17	1.10
Newark Bay	NB-1	40.6789	74.1167	3.2	9460	1274	1.64
	NB-2	40.6624	74.1323	0.9	9898	1268	1.70
	NB-3	40.6543	74.1398	0.0	10,261	1329	1.73

Surficial sediment samples were collected along the Hackensack and Passaic Rivers, as well as the Newark Bay. Sediments were collected within a meter of the shoreline in water less than 1 m depth using a shovel and were stored in Ziploc bags. Sediment samples were collected from the same sites as water sampling, except for several locations with gravelly sediments. In the laboratory, sediments were air-dried for a week, and then sediment grain size analysis was performed using a stainless steel sieve after removing gravels from sediments and homogenizing sediments. For the sediment samples collected in summer 2015, heavy metal concentrations in different grain size fractions (<2 mm grain size) were determined using a portable X-ray fluorescence (Niton™ XL3t XRF Analyzer; Thermo Scientific, Waltham, MA, USA). The XRF was calibrated using NIST standard reference materials 1646a (Estuarine Sediment) and 2709a (San Joaquin Soil). For each sample, five replicate analyses were performed. The variation in XRF measurements of replicates was typically less than $\pm 20\%$. The recoveries from the standard reference materials generally ranged from 80% to 120%. If the recovery was outside the range (80–120%), the calibration was repeated.

Sediment organic matter content was determined for bulk sediment (<2 mm grain size) by the loss on ignition method [29]. After wet sediment samples were air-dried for a week, approximately 10 g of sediment samples were placed in crucibles and were dried at 105 °C in an oven (Thermo Scientific, Waltham, MA, USA) overnight. After cooling the samples, the weight of samples was measured using a balance. Then, samples were transferred to a muffle furnace (Thermo Scientific) and were heated at 550 °C for 4 h. After cooling the samples, the samples were weighed again. The weight loss between the heating at 105 °C and 550 °C as a percentage of the total original dry sample weight is the sediment organic matter content.

Sediment oxygen demand experiments were conducted for the summer 2016 samples to study dissolved oxygen consumption by microbial decomposition of sediment organic matter and subsequent mobilization of nutrients from the sediment. Approximately 10 g of air-dried sediment (<2 mm grain size) was placed in a glass flask and 30 mL of deionized water was added. The glass flasks were tightly closed with rubber stoppers and were reacted on a shaker over a week. Dissolved oxygen of deionized water was measured before and after the reaction using a CHEMet dissolved oxygen test kit.

Nitrate, ammonia, and phosphate mobilized from sediments were measured using a Hach DR/890 portable colorimeter.

The spatial distribution of water quality parameters (e.g., pH, EC, DO, and nutrients) in the urban estuary was analyzed. The correlations between sediment grain size and organic matter content as well as heavy metal concentrations in sediments were examined. The relationships between sediment organic matter content and the consumption of dissolved oxygen as well as the mobilization of nutrients from sediments were investigated.

3. Results and Discussion

3.1. Spatial Variations of Water Quality in the Newark Bay Estuary

In summer 2016, the Hackensack River ($n = 10$), Passaic River ($n = 10$) and the Newark Bay ($n = 6$) were all characterized with slightly alkaline pH with average pH ranging from 7.54 ± 0.33 to 7.83 ± 0.14 (Table 1, Figure 1). The electrical conductivity (EC) was overall higher in the Hackensack River than in the Passaic River. The EC decreased gradually from 27.20 mS/cm at HR-1 (8.8 km upstream from NB-6) to 1.23 mS/cm at HR-9 (33.6 km from NB-6) in the Hackensack River, while it decreased from 8.15 mS/cm at PR-1 (12.9 km upstream from NB-6) to 0.65 mS/cm at PR-10 (30.6 km upstream from NB-6) in the Passaic River (Table 1, Figure 2).

It should be noted that water samples were collected from the Hackensack River during the transition from high tide to low tide, whereas water samples were collected from the Passaic River during the transition from low tide to high tide (Table 1). Although different tidal levels could have affected the electrical conductivity of two tidal rivers in summer 2016, the ion chromatography data for water samples collected from both rivers during the transition from low tide to high tide in summer 2015 demonstrate that the Hackensack River is more saline than the Passaic River (Table 2). In summer 2015, chloride and sulfate concentrations ranged from 276 mg/L (33.6 km from NB-6) to 7674 mg/L (8.8 km upstream from NB-6) and from 29 mg/L to 1014 mg/L, respectively in the Hackensack River, while chloride and sulfate concentrations varied from 86 mg/L (28.3 km upstream from NB-6) to 402 mg/L (12.9 km upstream from NB-6) and from 17 mg/L to 56 mg/L, respectively in the Passaic River (Table 2). In the Newark Bay, chloride ranged from 9460 mg/L to 10,261 mg/L, while sulfate ranged from 1274 mg/L to 1329 mg/L in summer 2015, consistent with high EC of 29.7–36.0 mS/cm in summer 2016.

The higher EC and chloride concentrations in the Hackensack River than the Passaic River indicate that tidal water intrudes further inland through the Hackensack River than the Passaic River. The Hackensack River is more brackish due to the reduction of freshwater discharge below the Oradell Dam [11], which caused freshwater lowland to transform into a tidal estuary and resulted in the destruction of freshwater aquatic plant and animal species [30]. The construction of the Oradell Dam, as well as several reservoirs constructed to serve the growing suburban/urban populations resulted in profound changes to the Hackensack River Watershed, including the river's hydrology, geomorphology, and water chemistry [31].

Dissolved oxygen (DO) was generally lower in the Hackensack River than in the Passaic River or the Newark Bay. DO ranged from 3 to 7 mg/L (average DO = 4 ± 1 mg/L) in the Hackensack River, while it ranged from 4 to 9 mg/L (average DO = 7 ± 1 mg/L) in the Passaic River (Table 1, Figure 2). DO in the Newark Bay was similar to that in the Passaic River, ranging from 5 to 8 mg/L (average DO = 7 ± 2 mg/L). In the Hackensack River, the lowest level of DO (3–4 mg/L) was observed from HR-4 to HR-7 samples (Table 1, Figure 1). It appears that the lowest DO levels were associated with sewage treatment plants that are located near HR-4 and HR-5 (Figure 1), including Bergen County Utilities Authority (approximate discharge rate: 69 mgd, million gallons per day), North Bergen Municipal Utilities Authority (approximate discharge rate: 7 mgd), and Secaucus Municipal Utilities Authority (approximate discharge rate: 3 mgd) [32].

Depressed levels of dissolved oxygen, which reduce species abundance and diversity, have been known to be a chronic problem in Newark Bay and its tributaries since the early 1900s [7,33]. While it has been reported that oxygen concentrations in the Hackensack River regularly fall below the New Jersey Department of Environmental Protection's water quality criterion (4.0 mg/L) and U.S. Environmental Protection Agency's protective minimum oxygen concentration (2.3 mg/L) for adult fishes [11], DO of water samples collected from the Newark Bay Estuary in summer 2015 and 2016 ranged approximately from 3 mg/L to 9 mg/L, which is above hypoxia (Table 1). The water quality data are consistent with improved water quality in the Hudson Raritan Estuary (HRE) since the enactment of the 1972 Clean Water Act [5,34]. However, some aquatic species can still be negatively affected by DO level as low as 3–4 ppm [35].

Dissolved nitrate was slightly higher in the Hackensack River than in the Passaic River or the Newark Bay (Table 1, Figure 2). Average nitrate concentration was 12.3 ± 2.1 mg/L in the Hackensack River, while it was 9.0 ± 1.3 mg/L in the Passaic River and 9.9 ± 1.0 mg/L in the Newark Bay. Dissolved ammonia was remarkably higher in the Hackensack River than in the Passaic River or the Newark Bay (Table 1, Figure 2). The average concentration of ammonia (as NH_4^+) was 1.40 ± 1.08 mg/L in the Hackensack River, while it was near the detection limit (0.02 mg/L) in the Passaic River (0.02 ± 0.01 mg/L) or in the Newark Bay (0.05 ± 0.03 mg/L). The highest level of ammonia (approximately 2.8 mg/L) was found from HR-5 to HR-7 samples, which are located within 5 km upstream from the Bergen County Utilities Authority (BCUA)'s wastewater treatment plant (Figures 1 and 2). The ratio of nitrate-N to ammonia-N was also significantly lower in the Hackensack River than in the Passaic River or the Newark Bay. The average ratio was 5 ± 5 for the Hackensack River, while it was 174 ± 71 for the Passaic River and 260 ± 30 for the Newark Bay. This suggests that the Hackensack River is under significantly less oxidic condition than the Passaic River or the Newark Bay. Dissolved phosphate was also higher in the Hackensack River than in the Passaic River or the Newark Bay. The average concentration of phosphate was 0.96 ± 0.30 mg/L for the Hackensack River, 0.79 ± 0.14 mg/L for the Passaic River, and 0.33 ± 0.09 mg/L for the Newark Bay (Table 1). The highest level of phosphate was 1.36 mg/L at HR-5, which was located adjacent to the BCUA's wastewater treatment plant (Figures 1 and 2). It is also worth mentioning that six CSOs are densely distributed between HR-5 and HR-7 along the Hackensack River (Figure 1). In addition to sewage treatment plants, CSOs can discharge nutrients and organics into the river during storm events.

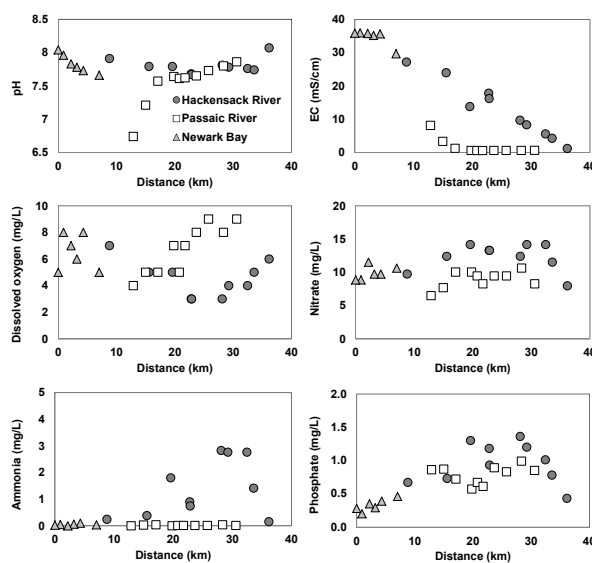


Figure 2. Variations of water quality data (pH, EC, DO, and nutrients) in summer 2016 for the Newark Bay Estuary (Hackensack River, Passaic River, and Newark Bay) as a function of the distance from the mouth of Newark Bay (NB-6).

Discharges from CSOs represent a major anthropogenic source of nutrients to receiving waters in an urban watershed [36]. During rain events when the combined volume of sewage and stormwater flows is too high for the treatment plant to handle, the system is designed to discharge combined sewage directly into nearby water bodies without treatment, causing significant contamination of waterways and aquatic systems. New Jersey's combined sewer systems discharge an estimated seven billion gallons per year (from 16 cities) and, on average, CSO drainage areas cover 53% of municipal land area [37]. It is estimated that CSOs are responsible for approximately 20% of the biological oxygen demand (BOD) discharged to the Hudson/Raritan Estuary [7,38]. In addition to CSOs, wastewater treatment plants discharge nutrients and organics into the estuary because the treated wastewater still contains large amounts of nutrients and organic matter. Since the construction of the Oradell Reservoir in 1922, the effluent from sewage treatment plants (STPs) comprises more than 75% of the freshwater input to the Hackensack River under non-storm conditions [11]. The reduced flow of the Hackensack River, as well as the high nutrient concentrations in sewage effluents that now dominate the freshwater inputs, can cause the hypoxia during the summer growing season and create a stressed system for aquatic organisms. Because of its limited freshwater inflow, the lower Hackensack River is susceptible to local pollution sources, which have collectively degraded water quality and sediment quality [16]. In contrast, the concentrations of nutrients were relatively lower in the Passaic River than in the Hackensack River, probably because of faster draining of combined sewer overflows into the Newark Bay due to higher freshwater discharge in the Passaic River than in the Hackensack River [12].

Additional water samples ($n = 2$) collected in summer 2015 from the headwater areas (Mendham, NJ, USA) of the Passaic River without CSOs or STPs showed on average pH of 6.95 ± 0.10 , EC of 0.64 ± 0.05 mS/cm, DO of 9.5 ± 0.7 mg/L, nitrate of 7.8 ± 2.8 mg/L, ammonia of <0.02 mg/L, and phosphate of 0.17 ± 0.06 mg/L. The water quality was similar to that of the lower Passaic River (Tables 1 and 2), except for phosphate, which was significantly higher in the lower Passaic River than in the headwater. This suggests that the discharge of sewage effluents to the Passaic River is limited during non-storm periods without the operation of CSOs, but additional water quality monitoring during storm events is needed to evaluate the impacts of sewage discharge from CSOs on water quality in the urban estuary.

3.2. Effects of Tidal Cycle on Water Quality in the Newark Bay Estuary

Tidal advection periodically displaces dissolved and suspended matter in estuaries, altering water quality over tidal periods [13]. Tidal shears can disperse pollutants and reduce peak concentrations near pollutant sources [39]. The lower Hackensack and Passaic Rivers as well as the Newark Bay are affected by tides, and the tidal flows distribute dissolved and suspended materials, including contaminants, from Newark Bay to the Hackensack River and the Passaic River [11].

The variation of water quality over a tidal cycle from high tide to low tide was monitored in the estuary on the same day (Table 3). The water levels (tidal datum: mean lower low water) at the Bergen Point West Reach station, New York, were used because it was the closest station to the Newark Bay Estuary. The water level at the station decreased from 1.17–1.58 m to 0.40–0.89 m, from 0.48–0.67 m to 0.12–0.17 m, and from 0.99–1.31 m to 0.21–0.42 m during the sampling along the Hackensack River, the Passaic River, and the Newark Bay, respectively. It should be noted that the water levels were lower during the sampling along the Passaic River than during the sampling along the Hackensack River or the Newark Bay. In addition, the difference between high water level and low water level was approximately 0.8 m during the water sampling along the Hackensack River and the Newark Bay, while it was approximately 0.4 m during the water sampling along the Passaic River. Therefore, the water quality data should be compared between water bodies in the estuary with caution. During the interval, the pH increased consistently by 0.15–0.56 for the Hackensack River, by 0.42–0.55 for the Passaic River, and by 0.18–0.34 for the Newark Bay. The increase of pH during the transition from high tide to low tide is attributed to the flow of upstream freshwater with higher pH toward the bay (Tables 1 and 2). The EC varied slightly by 0.1–2.55 mS/cm for the Hackensack River, while the

variation of EC was more evident for the Passaic River, ranging from 3.90 to 7.57 mS/cm during the tidal cycle. For the Newark Bay, the variation of EC was negligible for NB-2 and NB-3, whereas NB-1 located near the mouth of the Hackensack River showed a large change of EC by 6.20 mS/cm. Despite the larger changes in water levels for the Hackensack River than for the Passaic River, less significant change in EC for the Hackensack River than for the Passaic River during the tidal cycle is attributed to significantly lower volume of freshwater flow in the Hackensack River than in the Passaic River, which caused the entire lower portion of Hackensack River brackish over a tidal cycle.

In the Hackensack River, dissolved oxygen for HR-5 to HR-7 was as low as 2.5 to 3.5 mg/L during high tide, but increased to 4 to 8 mg/L during low tide (Table 3). The significant increase in DO during low tide may be associated with the downstream flow of oxygenated freshwater. During high tide, the effluent from the BCUA's wastewater treatment plant flows upstream, while more oxygenated river water flows downstream during low tide in the Hackensack River. In the tidal section of Passaic River, dissolved oxygen also increased slightly from 4–6 mg/L to 5–9 mg/L during the transition from high tide to low tide perhaps because of the downstream flow of more oxygenated river water, given that the Passaic River showed relatively higher DO in the upstream sites (8–9 mg/L for PR-7 to PR-10; Table 1). In the Newark Bay, DO level was fairly constant and maintained relatively high, with 5–8 mg/L DO during high tide and 6–9 mg/L DO during low tide. It should be noted that there is a lag time for the tidal cycle in the estuary between the Newark Bay and the Hackensack and Passaic Rivers [13]. Therefore, the direction of water flow in the lower and upper portions of the estuary could be different occasionally over a tidal cycle.

Table 3. Water quality data for the Hackensack River, the Passaic River, and the Newark Bay collected during a tidal cycle (no shade: high water level, gray shade: low water level) in summer 2016. Distance (km) indicates the distance from NB-6 located near the mouth of the Newark Bay. Water level data (tidal datum: mean lower low water) at Bergen Point West Reach, NY, are from National Oceanic and Atmospheric Administration (<https://tidesandcurrents.noaa.gov>).

Water Body	Sample ID	Distance (km)	pH	EC (mS/cm)	DO (mg/L)	Nitrate (mg/L)	Ammonia (mg/L)	Phosphate (mg/L)	Water Level (m)
Hackensack River	HR-2	15.6	6.76	26.70	5	11.5	0.05	0.65	1.58
	HR-4	22.8	7.30	15.20	4	22.1	1.54	1.53	1.55
	HR-5	22.9	7.39	10.85	3.5	17.7	3.73	1.66	1.44
	HR-6	28.1	7.50	10.13	3.5	16.8	4.24	1.76	1.25
	HR-7	29.2	7.63	8.30	2.5	18.6	3.99	1.68	1.17
	HR-2-1	15.6	7.32	25.40	4.5	15.1	0.14	0.70	0.89
	HR-4-1	22.8	7.56	15.30	3	19.5	1.54	1.38	0.67
	HR-5-1	22.9	7.69	11.24	8	17.7	3.34	1.24	0.56
	HR-6-1	28.1	7.76	8.64	4	18.6	4.24	1.17	0.48
	HR-7-1	29.2	7.78	5.75	5	14.2	1.80	0.99	0.40
Passaic River	PR-1	12.9	7.00	18.69	4	7.7	0.03	0.68	0.67
	PR-2	15.0	7.35	10.91	5	5.9	<0.02	0.74	0.58
	PR-3	17.1	7.59	5.92	6	7.7	0.03	0.59	0.48
	PR-1-1	12.9	7.55	11.12	5	6.5	<0.02	0.75	0.17
	PR-2-1	15.0	7.79	5.74	6	7.7	<0.02	0.88	0.15
	PR-3-1	17.1	8.01	2.02	9	8.3	<0.02	0.68	0.12
Newark Bay	NB-1	7.0	7.66	29.7	5	10.6	0.04	0.46	1.31
	NB-2	4.3	7.73	35.7	8	9.7	0.10	0.39	1.14
	NB-3	3.2	7.78	35.2	6	9.7	0.06	0.29	0.99
	NB-1-1	7.0	8.00	35.9	6	8.9	0.04	0.30	0.21
	NB-2-1	4.3	7.99	35.5	6	9.7	<0.02	0.48	0.34
	NB-3-1	3.2	7.96	35.9	9	9.7	<0.02	0.24	0.42

During the transition from high tide to low tide, the concentration of nutrients including nitrate, ammonia, and phosphate varied inconsistently, but the concentrations of all nutrients were constantly higher in the Hackensack River than in the Passaic River and the Newark Bay during the tidal cycle (Table 3). Average concentrations of nitrate were 17.4 ± 3.8 and 17.0 ± 2.3 mg/L for the Hackensack River, 7.1 ± 1.0 and 7.5 ± 0.9 mg/L for the Passaic River, and 10.0 ± 0.5 and 9.4 ± 0.5 mg/L for the Newark Bay during high tide and low tide, respectively. Average concentrations of ammonia (as NH_4^+) were 2.71 ± 1.83 and 2.21 ± 1.61 mg/L for the Hackensack River during high tide and low

tide, respectively, while ammonia concentrations were near or below the detection limit (0.02 mg/L) for the Passaic River and the Newark Bay over a tidal cycle. Average concentrations of phosphate were 1.46 ± 0.46 and 1.10 ± 0.26 mg/L for the Hackensack River, 0.67 ± 0.08 and 0.77 ± 0.10 mg/L for the Passaic River, 0.38 ± 0.09 and 0.34 ± 0.12 mg/L for the Newark Bay during high tide and low tide, respectively.

The results indicate that the Hackensack River is the most oxygen deficient and nutrient rich water body in the Newark Bay Estuary over a tidal cycle during non-storm periods. In water samples ($n = 20$) collected from the Hackensack River under various tidal conditions in summer 2016 (Tables 1 and 3), dissolved nutrients and oxygen showed moderate to strong correlations with the distance from the BCUA's sewage treatment plant (STP) (Figure 3). The concentrations of nitrate, ammonia, and phosphate were higher with decreasing distance from the STP, while dissolved oxygen concentrations were generally lower with decreasing distance from the STP (Figure 3). The spatial pattern of nutrients in summer 2016 is consistent with the summer 2015 data, which showed the highest levels of nitrate, ammonia, and phosphate at HR-4 or HR-5 adjacent to the Bergen County Utilities Authority's STP [28]. Therefore it appears that the continuous discharge of high concentrations of nutrients in the sewage effluent from the Bergen County Utilities Authority's STP, which is the single largest source of freshwater into the Hackensack River, results in high nutrients concentrations and consequent oxygen deficiency in the vicinity of the discharges and tides may transport dissolved nutrients to upstream and downstream portions of the river [13]. Because the lower Hackensack River receives considerable tidal flows of brackish water from Newark Bay [13], the tidal flows can distribute dissolved and suspended materials, including nutrients and heavy metals, from Newark Bay throughout the Hackensack River as far as the Oradell Dam during tidal cycles [11]. Pollutants entering Newark Bay from the lower Passaic River may be transported into the Hackensack River by tides, whereas pollutants from the Newark Bay or the Hackensack River are unlikely to be transported far into the Passaic River due to its high freshwater discharge into the Newark Bay [30].

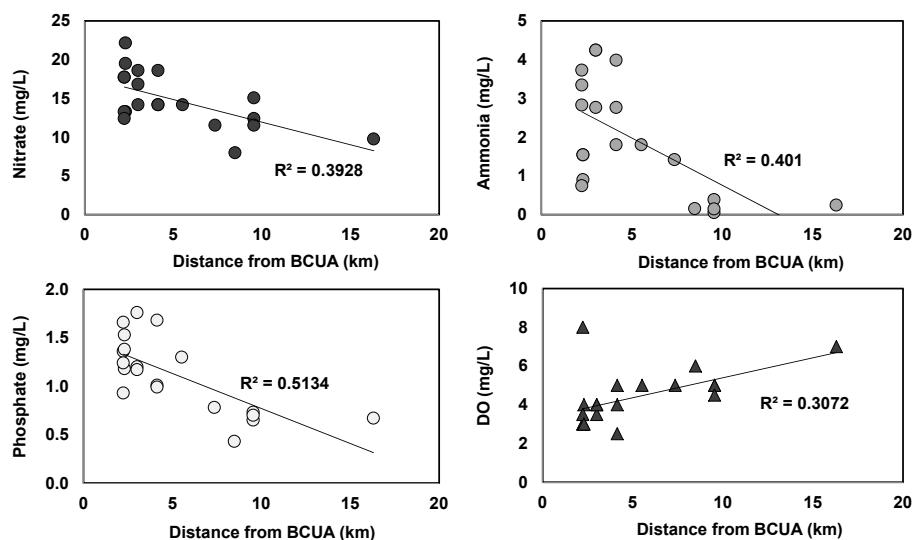


Figure 3. Correlation between the distance from the Bergen County Utilities Authority (BCUA)'s wastewater treatment plant and dissolved nutrients and oxygen in water samples ($n = 20$; Tables 1 and 3) collected from the Hackensack River in summer 2016.

3.3. Spatial Variations of Sediment Quality in the Newark Bay Estuary

Sieve analyses indicate that surficial sediments collected from the Newark Bay Estuary in 2015 and 2016 consisted dominantly of sand. For the 2015 samples (<2 mm grain size), the Hackensack River sediments ($n = 8$) consisted on average of $34 \pm 13\%$ coarse sand (0.5–2 mm), $18 \pm 7\%$ medium sand (0.25–0.5 mm), $33 \pm 18\%$ fine sand (0.063–0.25 mm), and $15 \pm 5\%$ silt and clay (<0.63 mm), while

the Passaic River sediments ($n = 7$) consisted on average of $39 \pm 10\%$ coarse sand, $23 \pm 10\%$ medium sand, $21 \pm 6\%$ fine sand, and $17 \pm 12\%$ silt and clay (Table 4).

Table 4. Grain size distribution (wt %) for sediments (<2 mm grain size) collected from the Hackensack River (HR) and the Passaic River (PR) in summer 2015. Distance (km) indicates the distance from NB-6 located near the mouth of the Newark Bay.

Sample ID	Distance	Coarse Sand		Medium Sand	Fine Sand		Silt + Clay
	km	1–2 mm	0.5–1 mm	0.25–0.5 mm	125–250 μm	63–125 μm	<63 μm
HR-1	9	1	3	9	60	16	11
HR-2	15	17	13	33	19	9	9
HR-3	20	21	15	15	16	11	21
HR-4	23	17	16	17	14	13	23
HR-5	29	27	19	16	11	12	15
HR-6	32	25	19	15	11	13	18
HR-7	33	17	19	24	18	9	12
HR-8	36	20	18	19	18	11	15
AVG		18	15	18	21	12	15
PR-1	13	14	19	15	11	11	31
PR-2	15	22	19	12	10	11	26
PR-3	18	32	25	17	8	5	13
PR-4	20	12	17	15	13	11	32
PR-6	21	13	18	36	19	7	7
PR-7	22	16	29	33	9	4	8
PR-8	24	12	22	32	22	7	5
AVG		17	21	23	13	8	17

For the 2016 samples (<2 mm grain size), coarse sand, medium sand, fine sand, and silt + clay fractions were $53 \pm 19\%$, $22 \pm 10\%$, $18 \pm 11\%$, and $6 \pm 2\%$ for the Hackensack River; $43 \pm 17\%$, $22 \pm 4\%$, $28 \pm 15\%$, and $7 \pm 2\%$ for the Passaic River; and $38 \pm 17\%$, $38 \pm 16\%$, $21 \pm 20\%$, and $3 \pm 4\%$ for the Newark Bay, respectively (Table 5). The relatively lower silt and clay fractions in the 2016 sediment samples than in the 2015 sediment samples are attributed to heterogeneous nature of estuarine sediments. It is also possible that top layers of surficial sediment largely consisting of fine silt and clay grains could have been disturbed and partially lost during sediment sampling using a shovel. It may be necessary to use a coring device to preserve all sediment layers with minimum disturbance.

Table 5. Grain size distribution (wt %) for sediments (<2 mm grain size) collected from the Hackensack River (HR), the Passaic River (PR), and the Newark Bay (NB) in summer 2016. Distance (km) indicates the distance from NB-6 located near the mouth of the Newark Bay.

Sample ID	Distance	Coarse Sand		Medium Sand	Fine Sand		Silt + Clay
	km	1–2 mm	0.5–1 mm	0.25–0.5 mm	125–250 μm	63–125 μm	<63 μm
HR-1	9	4	16	38	29	9	5
HR-2	16	36	31	17	8	4	5
HR-4	23	28	32	24	8	4	5
HR-5	28	12	23	32	21	8	4
HR-6	29	38	23	14	9	5	10
HR-8	34	35	26	16	9	5	9
HR-9	36	45	26	13	7	3	5
AVG		28	25	22	13	5	6
PR-1	13	28	26	22	12	6	7
PR-3	17	25	30	23	10	5	7
PR-5	21	23	29	27	13	5	4
PR-6	22	24	21	19	16	9	11
PR-7	24	5	10	23	37	18	7
PR-8	26	8	17	31	24	11	9
AVG		21	22	22	18	10	7
NB-1	7	8	11	26	39	9	8
NB-2	4	26	32	30	13	0	0
NB-3	3	15	35	43	6	0	0
NB-4	2	22	32	39	6	0	0
NB-5	1	5	19	66	9	1	1
NB-6	0	10	18	21	29	16	6
AVG		14	24	38	17	4	3

Sediment organic matter content was determined using the loss on ignition method (Figure 4). For 2015 samples ($n = 16$), sediment organic matter content was on average 7.0 ± 3.7 wt % (2.1–13.0 wt %) for the Hackensack River, while it was on average 6.3 ± 3.9 wt % (0.8–11.7 wt %) for the Passaic River. For 2016 samples ($n = 19$), sediment organic matter content was higher in sediments from the Hackensack River (6.3 ± 3.8 wt % organic matter) and the Passaic River (7.0 ± 2.3 wt % organic matter) than in sediments from the Newark Bay (3.2 ± 2.0 wt % organic matter). Sediment organic matter content ranged from 1.7 to 10.7 wt % for the Hackensack River, from 4.1 to 10.3 wt % for the Passaic River, and from 0.9 to 6.4 wt % for the Newark Bay. For both 2015 and 2016 sediment samples, organic matter content was correlated with sediment grain size distribution. As the proportion of silt and clay fractions increased, sediment organic matter content increased, showing R^2 of 0.7521 for the 2015 samples and R^2 of 0.5581 for the 2016 samples (Figure 4). This suggests that organic matter in sediments of the Newark Bay Estuary is predominantly associated with silt and clay grains.

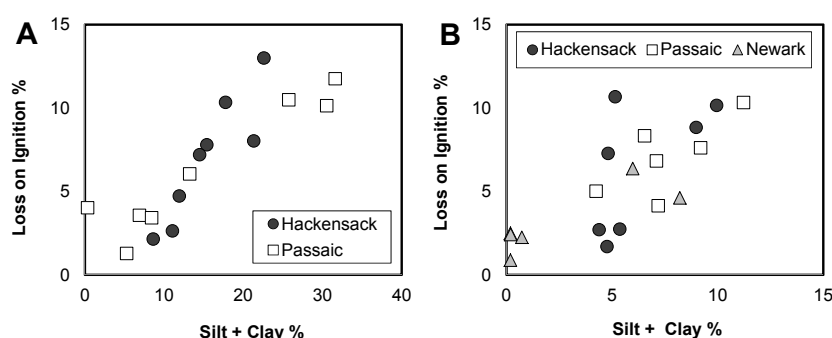


Figure 4. Correlation between sediment organic matter content (Loss on Ignition %) and the proportion of silt and clay fractions for sediments collected from the Hackensack River, the Passaic River, and the Newark Bay in summer: 2015 (A); and 2016 (B).

The consumption of dissolved oxygen for a week was related to organic matter content (Loss on Ignition %) in the sediment ($R^2 = 0.7076$; $n = 19$) (Figure 5), which suggests that the consumption of dissolved oxygen was caused by microbial decomposition of organic matter. Consumption of dissolved oxygen increased from 0 to 6.5 mg/L as sediment organic matter content increased from 0.9 to 10.7 wt %. Average DO consumption was approximately 3 ± 3 mg/L and 3 ± 2 mg/L for sediments from the Hackensack River (0–6.5 mg/L DO consumption) and Passaic River (1–6 mg/L DO consumption), respectively, while it was 1 ± 2 mg/L for sediments from the Newark Bay (0–4 mg/L DO consumption).

The extent of nutrient mobilization from sediments was normally associated with sediment organic matter content ($R^2 = 0.4396$ for nitrate, $R^2 = 0.5068$ for ammonia, and $R^2 = 0.5205$ for phosphate; $n = 19$) (Figure 5). In general, the concentration of nitrate mobilized from sediments increased from 9 mg/kg to 186 mg/kg with increasing organic matter content in sediments. The concentration of ammonia and phosphate released from sediments also increased from 2 mg/kg to 138 mg/kg and from 1 mg/kg to 56 mg/kg with increasing sediment organic matter content, respectively. The results suggest that organic-rich sediment contains a higher concentration of bioavailable nutrients and the estuarine sediment could act as both sink and source of nutrients in the estuary through biogeochemical cycling.

The mobilization of nutrients was significantly higher in sediments from the Passaic and Hackensack Rivers than in sediments from the Newark Bay (Figure 5). Average concentrations of nitrate, ammonia, and phosphate mobilized from the Hackensack River sediment were 70 ± 32 , 46 ± 58 , and 24 ± 19 mg/kg, respectively. Average concentrations of nitrate, ammonia, and phosphate mobilized from the Passaic River sediment were 142 ± 43 , 60 ± 47 , and 20 ± 14 mg/kg, respectively. Average concentrations of nitrate, ammonia, and phosphate mobilized from the Newark Bay sediment were 19 ± 10 , 16 ± 15 , and 3 ± 2 mg/kg, respectively. The significantly higher organic matter and nutrients in the Passaic and Hackensack River sediments than in the Newark Bay sediments suggest

more extensive historic or current pollutions of sediments by anthropogenic sources (e.g., CSOs, sewage treatment plants) or natural sources (e.g., the Meadowlands) along the urban rivers than the urban bay.

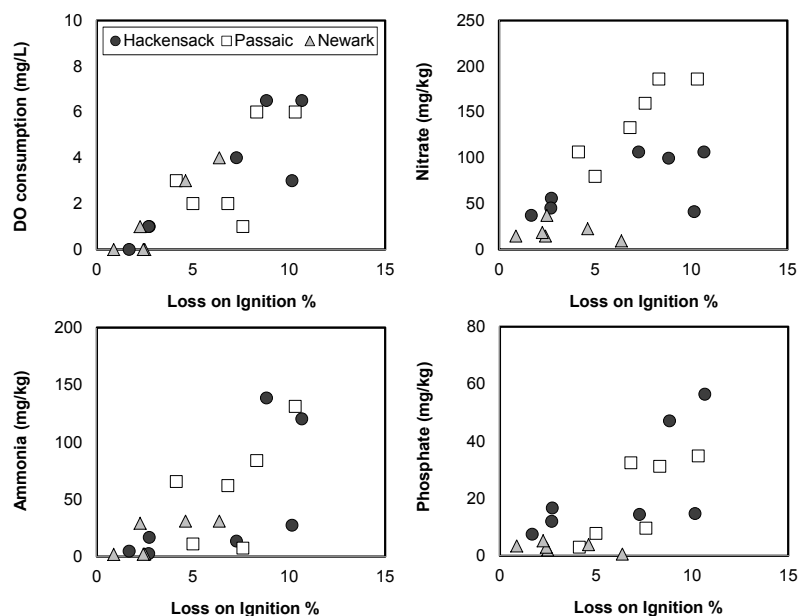


Figure 5. Correlation between organic matter content (Loss on Ignition %) and dissolved oxygen consumption as well as concentrations of mobilized nutrients in sediments collected from the Hackensack and Passaic Rivers as well as the Newark Bay in summer 2016.

Despite the improvement of water quality in the Newark Bay Estuary since the 1970s, elevated levels of contaminants may persist in sediments, acting as a source of contaminants that can cycle through the ecosystems [34]. Increasing water demand to support the region's increasing human population and withdrawal from reservoirs in the Hackensack River Watershed increased the volume of withdrawn water returning to the river in the form of sewage effluent from sewage treatment plants and CSOs, resulting in considerable quantities of nutrients likely accumulating in the sediment of the estuary. Oxygen concentrations in bottom waters are heavily influenced by oxygen consumption or uptake in the bottom sediments. Decay of organic matter by bacteria, ammonia oxidation by nitrifiers, algal respiration and flux of oxygen into the sediment all increase oxygen demand [40]. Hypoxic or anoxic conditions can be fatal especially for shellfish and other sessile organisms since these organisms cannot relocate themselves to a more oxygenated area. The sediment oxygen demand experiments showed that dissolved oxygen level dropped from 7 mg/L to ~1 mg/L in some sediment samples with high organic matter content after a week of reaction. This suggests that DO levels of bottom water in the Hackensack and Passaic Rivers may locally decrease to the hypoxic condition, threatening aquatic species in the estuary. The results also show that significant amount of nutrients accumulated in surficial sediments could be remobilized into water column due to physical and chemical changes, which can lead to eutrophication.

3.4. Geochemical Distribution of Heavy Metals in Sediments

The Newark Bay Estuary has been contaminated by various metal contaminant sources including industrial and municipal point source and non-point source discharges, urban storm water runoff, atmospheric deposition, and CSOs [41–43].

XRF data for sediments collected from the Hackensack ($n = 8$) and Passaic ($n = 7$) Rivers in summer 2015 show elevated levels of toxic heavy metals including As, Cr, Cu, Pb, and Zn, as well as Fe and Mn (Table 6). Concentrations of heavy metals were determined in different grain size fractions,

including very coarse sand (1–2 mm), coarse sand (0.5–1 mm), medium sand (0.25–0.5 mm), fine sand (125–250 μm), very fine sand (63–125 μm), as well as silt and clay (<63 μm).

Concentrations of heavy metals in bulk sediment (<2 mm grain size) were calculated based on the grain size distribution determined by sieve analysis and XRF data for each grain size fraction. Sediments from the Hackensack River ($n = 8$) contained on average: As of 12 ± 3 mg/kg, Cr of 195 ± 64 mg/kg, Cu of 126 ± 46 mg/kg, Fe of $39,467 \pm 15,923$ mg/kg, Mn of 756 ± 392 mg/kg, Pb of 96 ± 41 mg/kg, and Zn of 286 ± 104 mg/kg (Table 6). Sediments from the Passaic River ($n = 7$) showed on average: As of 11 ± 5 mg/kg, Cr of 137 ± 40 mg/kg, Cu of 137 ± 55 mg/kg, Fe of $32,433 \pm 10,684$ mg/kg, Mn of 555 ± 158 mg/kg, Pb of 155 ± 69 mg/kg, and Zn of 346 ± 182 mg/kg (Table 6).

The concentrations of heavy metals in sediments of the urban estuary were compared with sediment quality guidelines, the effects range-low and effects range-median (ERL and ERM), which are concentrations corresponding to the 10th and 50th percentile of the distribution observed in toxic sediment samples, respectively [44]. The guidelines have been used as interpretive tools in many sediment assessments throughout North America and elsewhere. In both sediments from the Hackensack and Passaic Rivers, the average concentrations of As, Cr, Cu, Pb, and Zn were consistently higher than ERL but lower than ERM (Table 6), suggesting that sediment of the urban estuary could occasionally cause toxic effects. However, three sediment samples (476, 492, and 582 mg/kg) from the Passaic River and a sediment sample (421 mg/kg) from the Hackensack River showed Zn concentration higher than ERM (410 mg/kg). The concentrations equivalent to and above the ERM value represent a probable-effects range within which toxic effects would frequently occur [44].

Table 6. Average concentrations (mg/kg) of heavy metals in bulk sediments (<2 mm grain size) and different size fractions of sediments collected from the Hackensack ($n = 8$) and Passaic ($n = 7$) Rivers in summer 2015. ERL and ERM (the effects range-low and effects range-median, respectively) are sediment quality guidelines (mg/kg).

Element	River	Sediment Grain Size						Bulk <2 mm	ERL	ERM
		1–2 mm	0.5–1 mm	0.25–0.5 mm	125–250 μm	63–125 μm	<63 μm			
As	Hackensack	13	14	12	12	14	14	12	8	70
	Passaic	14	11	12	10	10	6	11		
Cr	Hackensack	183	182	201	202	214	231	195	81	370
	Passaic	130	121	120	151	181	173	137		
Cu	Hackensack	127	132	124	118	146	146	126	34	270
	Passaic	114	120	141	160	191	185	137		
Fe	Hackensack	47605	41690	38258	37914	38810	38743	39467		
	Passaic	34940	31298	29249	35083	44363	39849	32433		
Mn	Hackensack	729	817	779	901	773	703	756		
	Passaic	579	504	508	600	754	725	555		
Pb	Hackensack	92	99	93	90	123	125	96	47	218
	Passaic	126	121	122	163	272	333	155		
Zn	Hackensack	284	312	292	273	314	318	286	150	410
	Passaic	315	312	304	364	474	485	346		

Pb and Zn concentrations were generally higher in the Passaic River sediments than in the Hackensack River sediment, while Cr concentrations were higher in the Hackensack River sediments than in the Passaic River sediments on average (Table 6). Concentrations of Cr, Cu, Pb, and Zn increased with decreasing grain size in the Hackensack and Passaic River sediments (Figure 6). The pattern was more obvious for the Hackensack River sediments than for the Passaic River sediments. It has been reported that trace element concentrations are strongly dependent on grain size, showing higher concentrations with smaller grain sizes, which typically have larger surface areas [45,46].

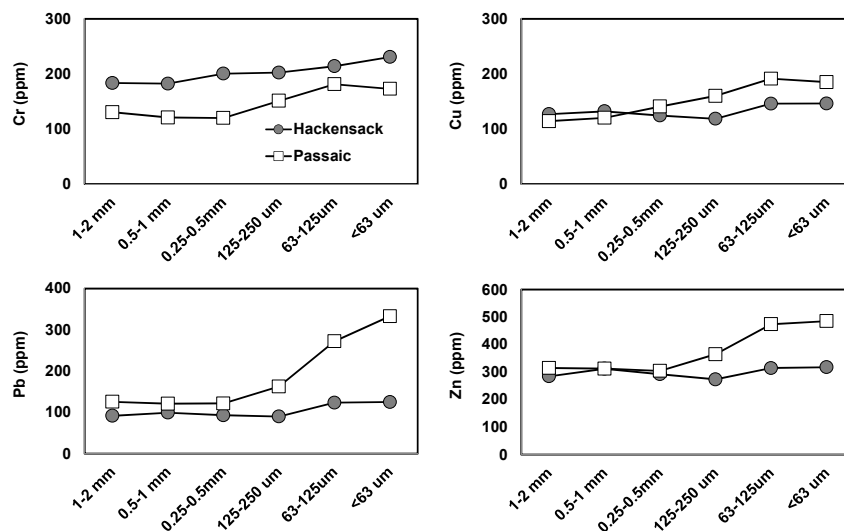


Figure 6. Average concentrations of heavy metals in sediments collected from the Hackensack ($n = 8$) and Passaic ($n = 7$) Rivers in summer 2015 as a function of sediment grain sizes (<2 mm grain sizes).

Rice et al. (1999) reported trace element concentrations in 541 streambed-sediment samples (<63 μm size fraction; silt and clay) collected from 20 study areas across the contiguous United States [46]. For the same grain size fractions (<63 μm), the average concentrations of As (14 ± 2 and 6 ± 5 ppm) in the Hackensack and Passaic River sediments were similar to the medians of As (5–21 ppm) in the streambed sediments from nonurban areas that are least affected by human activities. Average Cr concentrations in silt and clay fractions were 231 ± 66 and 173 ± 24 ppm for the Hackensack River sediments and the Passaic River sediments, respectively, which were significantly higher than the medians of Cr in the nonurban streambed sediments (46–105 ppm). Average Cu concentrations in the silt and clay fractions were 146 ± 60 and 185 ± 61 ppm for the Hackensack River sediments and the Passaic River sediments, respectively, while the medians of Cu in the nonurban streambed sediments were 13–47 ppm. Average Pb concentrations in silt and clay fractions were also remarkably higher in the Hackensack River sediments (125 ± 55 ppm) and the Passaic River sediments (333 ± 168 ppm) than the medians of Pb in nonurban streambed sediments (11–57 ppm). Average Zn concentration in silt and clay fractions were 318 ± 133 and 485 ± 183 ppm in the Hackensack River sediments and the Passaic River sediments, respectively, which were also higher than the medians of Zn in nonurban streambed sediments (69–240 ppm). The sources of heavy metals in the surficial sediments of the urban estuary could be historic industrial wastes dumped in 19th and 20th centuries or on-going discharge of domestic wastes from CSOs or sewage treatment plants.

Heavy metals in both Hackensack and Passaic River sediments were positively correlated (Figure 7). As the concentrations of sediment Pb increased, the concentrations of As, Cr, Cu, and Zn in sediments increased, showing R^2 of 0.5729 to 0.9093. Heavy metal concentrations in sediments also exhibited overall moderate to strong relationships with sediment organic matter content ($R^2 = 0.2516$ to 0.9249) (Figure 8). Organic matter in sediments may increase the adsorption capacity of sediments for heavy metals such as Zn, Pb, Cu, and Cr [47]. The positive correlations among heavy metals and sediment organic matter suggest that the sediments of the urban estuary have been contaminated by anthropogenic pollution sources containing both elevated levels of heavy metals and organic substances, which could be CSOs or STPs. Sampling of publicly owned treatment works (POTWs), combined sewer overflows (CSOs) and storm water outfalls (SWOs) in the NY–NJ Harbor from 2000 to 2004 reported the presence of toxic heavy metals such as Cd, Pb, and Hg in the effluents, with typically the largest loads of the contaminants found in the effluents from the POTWs because of the volume of their discharge [32]. The elevated concentrations of heavy metals (arsenic, cadmium, copper, chromium, lead, mercury, nickel and zinc) in the Newark Bay Estuary has been reported, which currently pose a

potential threat to the aquatic community [5,10,11]. For example, analysis of sediment cores taken from the tidal portion of the Passaic River during the early 1990s detected average metal concentrations ranging from 78 to 593 mg/kg for chromium, 134 to 549 mg/kg for copper, 244 to 633 mg/kg for lead, and 387 to 1146 mg/kg for zinc [48]. Bonnevie et al. (1994) collected and analyzed surficial sediments from three major tributaries to Newark Bay, including the Passaic River and the Hackensack River. The mean concentrations of As (17 ± 16 mg/kg), Cu (237 ± 222 mg/kg), Pb (421 ± 571 mg/kg) and Zn (395 ± 403 mg/kg) measured in the estuary exceeded several benchmark sediment quality values [14]. Chromium is a common pollutant in Hackensack River sediments because Hudson County, NJ was a center for chromate and bichromate chemical manufacturing in the 20th century [49]. It was reported that sediments along the eastern shore of the Hackensack River near its confluence with the Passaic River and Upper Newark Bay contained total chromium ranging between 5 and 9190 mg/kg, while the arithmetic mean and median concentrations of total chromium were 499 and 188 mg/kg, respectively [50]. The previously reported heavy metal concentrations in sediment of the Newark Bay Estuary are comparable to the heavy metal concentration in the Hackensack and Passaic Rivers sediments analyzed by XRF in this study (Table 6).

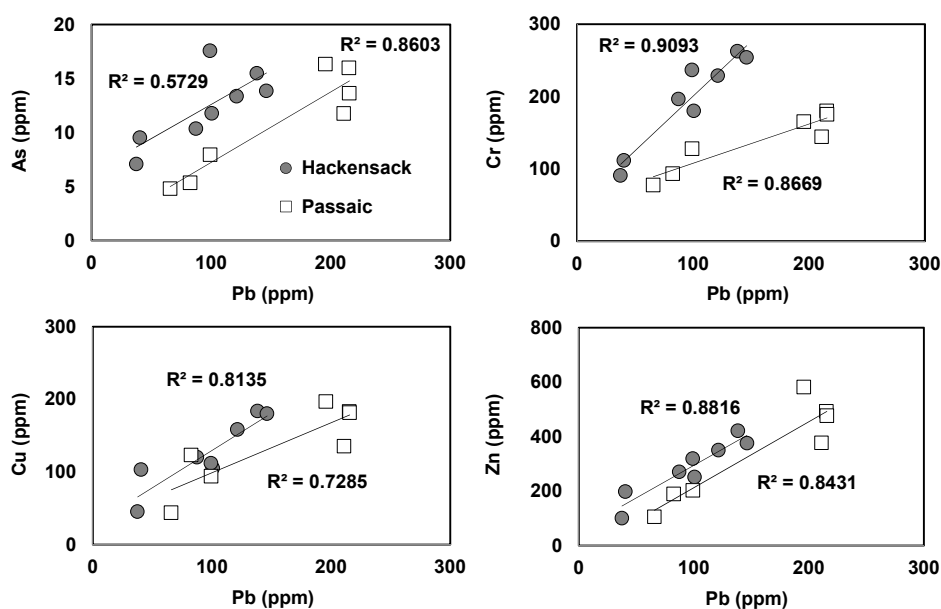


Figure 7. Correlations of heavy metals in sediments (bulk concentration for <2 mm grain size) collected from the Hackensack and Passaic Rivers in summer 2015.

Average concentrations of heavy metals in the bulk sediments from the Hackensack River (As: 12 ± 3 ppm, Cr: 195 ± 64 ppm, Cu: 126 ± 46 ppm, Pb: 96 ± 41 ppm, and Zn: 286 ± 104 ppm) and Passaic River (As: 11 ± 5 ppm, Cr: 137 ± 40 ppm, Cu: 137 ± 55 ppm, Pb: 155 ± 69 ppm, and Zn: 346 ± 182 ppm) are significantly higher than background metal concentrations for lithosphere (As: 1.5 ppm, Cr: 100 ppm, Cu: 70 ppm, Pb: 14 ppm, and Zn: 80 ppm) and soils (As: 5 ppm, Cr: 65 ppm, Cu: 18 ppm, Pb: 32 ppm, and Zn: 100 ppm) [51] by a factor of approximately 2–8. In contrast, concentrations of heavy metals in the sediments of the Hackensack and Passaic Rivers were similar or slightly higher than the average sediment metal concentrations in adjacent urban estuaries such as the lower Hudson River or the NY–NJ Harbor. Heavy metal concentrations in sediments collected over 100 km along the lower Hudson River during 1994–1996 ranged from 18 to 149 ppm for Cu, from 24 to 177 ppm for Pb, and from 101 to 257 ppm for Zn [52]. Mean metal concentrations of sediments collected from six sub-basins (Newark Bay, Lower Harbor, Upper Harbor, Jamaica Bay, western Long Island Sound, and the Bight Apex) of the NY–NJ Harbor in 1993–1994 were 10 ± 2 ppm for As, 78 ± 10 ppm for Cr, 73 ± 17 ppm for Cu, 79 ± 13 ppm for Pb, 170 ± 26 ppm for Zn [53].

The XRF analyses for different grain sizes also showed that finer grains contained higher concentrations of heavy metals (Figure 6). Contaminants associated with fine-grained sediments have a potential to release into the aquatic system by diffusion and resuspension, causing hazardous effects to aquatic species [17,41,43,54]. Surficial sediment is more subject to daily tidal actions and occasional scouring during high river flows, and thus the river movements can cause the contaminated fine surface sediment to be resuspended and redeposited. Heavy metals bound to fine grained sediments may accumulate in turbidity maximum zones where suspended sediments settle down [55,56]. The fine grained sediments containing high levels of heavy metals and nutrients can be tidally transported from the Hackensack River or the Passaic River to the Newark Bay [11]. A large increase in Cd, Pb, and Hg in the Hackensack River was found to correspond generally to higher suspended sediment concentrations during the last stages of the ebb [57]. In addition, organic matter in sediments of the Newark Bay Estuary is largely associated with fine silt and clay grains (Figure 4). Therefore, fine grained surface sediments in the Newark Bay Estuary require special management during environmental cleanup (e.g., sediment dredging) or urban development and construction along the estuary to minimize the disturbance of fine grained sediments because the levels of heavy metals, organic matter, and nutrients in water and sediment of the urban estuary is likely to be significantly affected by the suspension and transport of fine sediment grains such as silt and clay.

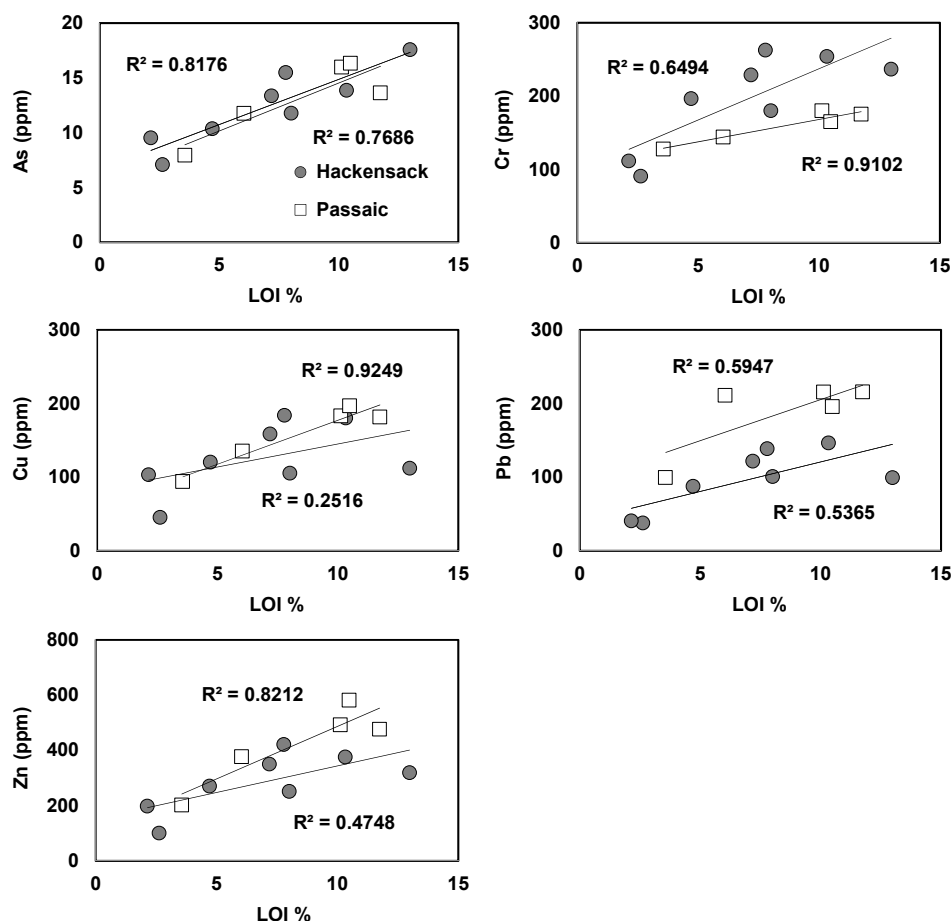


Figure 8. Correlations between LOI % (Loss on Ignition %; sediment organic matter content) and concentrations of heavy metals in sediments (<2 mm grain size) collected from the Hackensack and Passaic Rivers in summer 2015.

4. Conclusions

The Newark Bay Estuary, one of the most urban estuaries in the nation, suffered unregulated industrial and municipal waste discharges due to intense industrialization and urbanization since the late 19th century. As a result, biological diversity and abundance of aquatic species within the estuary have been substantially reduced. Water and surficial sediment samples were collected along the Hackensack and Passaic Rivers as well as the Newark Bay during non-storm periods in summer 2015 and 2016. The Hackensack River was more brackish than the Passaic River due to reduced freshwater discharge below the Oradell Dam. While dissolved oxygen level was lower in the Hackensack River than in the Passaic River or the Newark Bay, concentrations of dissolved nutrients such as nitrate, ammonia, and phosphate were significantly higher in the Hackensack River than in the Passaic River or the Newark Bay. While water quality of the estuary varied to some extent during a tidal cycle, elevated levels of dissolved nutrients and low levels of dissolved oxygen in the Hackensack River were spatially associated with the BCUA's sewage treatment plant over a tidal cycle. Laboratory measurements showed that sediment oxygen demand and mobilization of nutrients from sediments for a week increased with higher sediment organic matter content. The amount of nutrients mobilized from sediments was significantly higher for the Hackensack and Passaic Rivers than for the Newark Bay.

XRF analyses showed that average concentrations of heavy metals such as As, Cr, Cu, Pb, and Zn in bulk sediments (<2 mm grain size) from the Hackensack and Passaic Rivers were all above ERL (Effects Range–Low) but below ERM (Effects Range–Median). Heavy metals in sediments were positively correlated with each other, and were also associated with sediment organic matter. The concentrations of Cr, Cu, Pb, and Zn in the sediments were generally higher in the finer grain size fractions such as silt and clay. Higher content of sediment organic matter was also associated with finer grain sizes. The results suggest that surficial sediments in the urban estuary could act as both sink and source of nutrients and heavy metals. Fine grained sediments that are typically enriched with heavy metals and organics can be readily suspended and transported upstream and downstream during tidal cycles in the estuary, influencing the biogeochemical cycling of nutrients and heavy metals in coastal wetland ecosystems. It appears that the major sources of nutrients and heavy metals in water and sediment of the Newark Bay Estuary are currently the discharge of domestic wastes from sewage treatment plants (STPs) during non-storm periods as well as combined sewer overflows (CSOs) during storm events. Further research with more frequent and pervasive water and sediment quality monitoring over a complete tidal cycle during dry and wet periods is needed to improve the understanding of the biogeochemical cycling of nutrients and heavy metals in the urban estuary receiving sewage effluents from STPs and CSOs.

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