



Regional variability in bed-sediment concentrations of wastewater compounds, hormones and PAHs for portions of coastal New York and New Jersey impacted by hurricane Sandy



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ARTICLE INFO

Article history:

Received 29 September 2015

Received in revised form 9 April 2016

Accepted 25 April 2016

Available online 11 May 2016

Keywords:

Bed sediment

Hormones

Personal care/domestic use

Urban

Wastewater

Hurricane

ABSTRACT

Bed sediment samples from 79 coastal New York and New Jersey, USA sites were analyzed for 75 compounds including wastewater associated contaminants, PAHs, and other organic compounds to assess the post-Hurricane Sandy distribution of organic contaminants among six regions. These results provide the first assessment of wastewater compounds, hormones, and PAHs in bed sediment for this region. Concentrations of most wastewater contaminants and PAHs were highest in the most developed region (Upper Harbor/Newark Bay, UHNB) and reflected the wastewater inputs to this area. Although the lack of pre-Hurricane Sandy data for most of these compounds make it impossible to assess the effect of the storm on wastewater contaminant concentrations, PAH concentrations in the UHNB region reflect pre-Hurricane Sandy conditions in this region. Lower hormone concentrations than predicted by the total organic carbon relation occurred in UHNB samples, suggesting that hormones are being degraded in the UHNB region.

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1. Introduction

Tidal surge associated with Hurricane Sandy inundated low-elevation coastal areas in New Jersey and New York in late October 2012. Most, if not all, bays along the New Jersey shore, New York/New Jersey Harbor, and the southern shore of Long Island were impacted by tidal surge and/or, to a lesser extent, river flood-waters (Schubert et al., 2015; Hall and Sobel, 2013). Residential structures, industrial manufacturing and storage facilities, and wastewater-treatment facilities were affected by this flooding. Storm-related damage to buildings and infrastructure had the potential to release a variety of contaminants that could subsequently be transported to local rivers and bays. In some cases, this damage persisted for several weeks, with untreated or partially treated wastewater released from wastewater-treatment facilities that failed as a result of disruptions in electrical service and flooding of treatment works. Sewage overflows totaled 19.7 billion liters (L) in New York and 19.3 billion in New Jersey (Kenward et al., 2013). Public-health agencies responded to address acute effects (such as damaged homes and spills) of Hurricane Sandy, but the potential long-term human and ecological effects caused by the introduction of

contaminants from compromised infrastructure, weathering debris, and redistribution of previously contaminated sediments are unknown.

Persistent organic pollutants, including polychlorinated biphenyls (PCBs), dibenzo-*p*-dioxins/furans (PCDD/Fs), and polycyclic aromatic hydrocarbons (PAHs), are widespread in bed sediments in New York Harbor and Newark Bay bed sediments. These contaminants are primarily from historical sources, have been decreasing through time, and have been monitored long-term by several programs (Adams, 2003; Lodge et al., 2015).

Additional studies have evaluated the impact of short-term inputs of organic pollutants into New York Harbor. For example, the impact of the September 11, 2001 (9/11) destruction of the World Trade Center on the occurrence and distribution of persistent organic pollutants has also been assessed by Litten et al., 2003 and Lauenstein and Kimbrough, 2007. Litten et al. (2003) did not find a substantial impact on the presence of PCBs, PCDD/Fs and other persistent organic pollutants in sediments in the New York Harbor after 9/11, with only 5% of the PCBs in sediment in the vicinity of Manhattan attributable to the 9/11 destruction; Lauenstein and Kimbrough (2007) similarly did not find any substantial changes in PCBs or PAHs in sediments after 9/11.

Other extreme events, including large storms, can affect estuarine conditions that control the distribution of contaminants in sediments. Estuaries trap and retain much of the sediment material inputs to them by the surrounding watershed (Meade, 1969; Chapman and Wang, 2001). Local maximums of sediment deposition rates typically occur at or near the brackish-freshwater interface where particle settling is enhanced (Meade, 1969). Thus, sediment deposition dynamics can depend in part on salt-water intrusion. Storms can increase bed stress and cause re-suspension of sediments in many estuaries including New York Harbor. For example, Tropical Storms Irene and Lee significantly increased discharge in the Upper Hudson River, but turbidity maximums and sediment dynamics in the estuary were the result of sediment re-suspension rather than new fluvial sediment delivery (Ralston et al., 2013). Sediment re-suspension can alter contaminant distributions because concentrations of some contaminants, including trace metals, PCB, and PAHs have been declining with time and thus can be higher in areas with deeper sediments (Bopp et al., 1993). However, recent work in Jamaica Bay, NY has shown that concentrations of some emerging, sewage-associated contaminants have been increasing over the past five decades and the highest concentrations are in surface sediments (Lara-Martín et al., 2014). Thus, it is difficult to predict how storm surge, sediment re-suspension, and significant sewage outflows resulting from failed infrastructure might affect the concentration and distribution of historical (e.g. PCBs, PAHs) and emerging (e.g. hormones, personal care and domestic use products) contaminants.

Emerging contaminants (including but not limited to hormones, antimicrobials, surfactants, and fragrances) associated with wastewater discharges have not been widely assessed in the sediments in the New York Harbor or surrounding areas. To our knowledge, no regional assessment of hormones and other emerging contaminants has been undertaken in urbanized estuaries in the United States. By contrast, recent research in large urban areas in China (Zhang et al., 2014; Chang et al., 2011), and Europe (Matic et al., 2014; López de Alda et al., 2002) have demonstrated the presence of these contaminants in bed sediments in estuaries adjacent to large urban areas. Recent investigations of wastewater associated organic contaminants including estrogens, androgens, plant and animal biochemicals (PABs), fragrances, and antimicrobial compounds indicate that sewage inputs can be an important source of contaminants (Phillips et al., 2012; Cantwell et al., 2010; Nilsen et al., 2014; Gorga et al., 2015). A few studies have included emerging contaminants in sediments in the New York Harbor area (Reddy and Brownawell, 2005; Benotti and Brownawell, 2009; Ferguson et al., 2003; Lei et al., 2009; Lara-Martín et al., 2014) but these studies have 1) largely focused on surfactants, and have generally not included analyses for hormones (especially androgens), and 2) mostly focused on the Jamaica Bay portion of the study area and have not included sampling south in New Jersey or in eastern New York (i.e. Long Island).

Emerging contaminants that can sorb to sediments can affect fish health, sexual characteristics of fish, fish reproduction, and potentially affect the health of benthic invertebrates (Jobling et al., 1996; Sumpter and Johnson, 2005; Richardson and Kimura, 2016). Given the widespread damage of wastewater infrastructure, release of large amounts of untreated and partially treated sewage during Hurricane Sandy (Kenward et al., 2013), and the propensity of these contaminants to be associated with wastewater, the assessment of emerging contaminants post-Sandy is particularly relevant. However, emerging contaminants have not been sampled in this area prior to Hurricane Sandy; therefore, it is impossible to provide a regional assessment of the impact of Hurricane Sandy on these contaminants. Given these constraints, the purpose of this study was to 1) document post-Sandy occurrence of emerging contaminants, including hormones, plant and animal biochemicals, and fragrances in coastal New Jersey and New York area; 2) use ratios of analytes to determine potential sources of contaminants; and 3) analyze relationships between hormones and other compounds as a means to explore the distribution of hormones in coastal New York and New Jersey.

2. Methods

2.1. Sample network, land use analysis, and data availability

Sediment samples were collected from 79 sites distributed among six coastal regions from southern New Jersey to eastern New York (Table S01, Fig. S01). The study area was divided into 6 study regions on the basis of hydrologic divides and similar patterns of land use to evaluate differential impacts associated with Hurricane Sandy. The 6 regions are based on aggregation of 13 regions described by Fischer et al., 2015 (Table S02 gives the equivalent regions used in this paper and Fischer et al., 2015). Land use estimates for the regions are based on the 2011 National Land Cover database for New York and New Jersey obtained from the USDA NRCS Geospatial Data Gateway (<http://datagateway.nrcs.usda.gov/>), and mosaicked to provide continuous coverage of the study area. The areas inundated during the storm surge are based on models incorporating field-based observations of high-water marks and monitoring data from stream, tide and surge monitors (Federal Emergency Management Agency [FEMA], 2013). Land use was summarized to Level I Anderson classes. Additional details on sample network and land use analysis are provided by Fischer et al., 2015. The proportion of each region and inundated portion of the region in high-intensity developed land use is given in Fig. S02.

All the data used for the analysis in this paper are available in Fischer et al., 2015, where data tables are provided in a spreadsheet form.

2.2. Sample collection and handling

Bed-sediment samples were collected from boats using grab samplers during June–October 2013; all sites were sampled once. Samples were collected using standard US Geological Survey (USGS) or US Environmental Protection Agency methods described in Hladik et al., 2009; Radtke, 2005; Lauenstein and Cantillo, 1993, and US Environmental Protection Agency, 2001. Sediment samples were collected using either an Ekman sampler, Petite Ponar sampler, or a modified Van Veen type sampler depending on sampling conditions (for example, sediment texture, or presence of shells or vegetation). The upper 2 cm of sediment was retained for analysis to standardize sample collection among sites in an attempt to obtain samples representative of sediment-quality conditions after Hurricane Sandy. Additional details of sediment sampling and handling are given in Supplemental Materials and in Fischer et al. (2015).

2.3. Analytical methods

Estuarine sediments collected at each site were analyzed for more than 75 organic chemical constituents. Analytical results for these samples are given in Fischer et al., 2015. Two USGS methods were used to analyze compounds, and are referred to as 1) the wastewater method and 2) the hormone method. A summary of these methods is provided below, with more detail available in Supplemental Material and Fischer et al. (2015).

The wastewater method includes analysis for 57 compounds that include surfactants, fragrances, antioxidants, disinfectants, food additives, plastic components, industrial solvents, PAHs, PABs, phosphate flame retardants, and high-use domestic pesticides using a method described by Burkhardt et al. (2006). Because this method largely focuses on compounds often associated with wastewater, it is referred to here as the wastewater method. Analytes were extracted from sediment by pressurized solvent extraction, further isolated by solid-phase extraction, and determined by gas chromatography/mass spectrometry. Reporting levels ranged from 50 to 1000 µg/kg (micrograms per kilogram) for a 10 g (gram) sample, and were adjusted based on the mass of the sample.

The second method, referred to as the hormone method, was used to analyze for 20 compounds, including 9 estrogens, 6 androgens, two

progesterins, two plant and animal biochemicals, and bisphenol A (BPA). Cholesterol (CHO) and 3 β -coprostanol (COP) are included in both the wastewater and hormone method; however, we only use data determined using the hormone method in interpretation for this article.

Sample extraction and analyte isolation procedures were comparable to that described for the wastewater method except that pressurized solvent extraction was conducted at 165 °C only. Method compounds were derivatized using 500 μ l of activated *N*-methyl-*N*-(trimethylsilyl)trifluoroacetamide, analyzed by gas chromatography/tandem mass spectrometry, and quantified by using an isotope dilution quantification procedure as described by Foreman et al. (2012) for hormones in water samples. Reporting limits ranged from 0.1 μ g/kg for most hormone compounds to 25 μ g/kg for cholesterol CHO and COP for a 10 g sample. Reporting limits were varied for samples based on the mass of the sample. More details are provided in the Supplemental Materials text.

The method used to determine TOC (total organic carbon) is described in Fischer et al. (2015). TOC was measured using the EPA method 415 (US Environmental Protection Agency, 1983) using flame ionization, with a reporting limit of 1000 mg per kg.

2.4. Quality assurance/quality control

Quality assurance and quality control (QA/QC) data available for the wastewater and hormone methods include analysis results for field replicates, lab replicates, lab blanks and lab spiked samples. Laboratory QA/QC for the wastewater method and the hormone method are detailed in Supplemental Materials.

Six field replicate samples were analyzed using the wastewater method during the study. This analysis yielded 77 paired concentration comparisons (with detections in both field and replicate samples), and 22 unpaired comparisons (with a detection in only one of the two samples). The median replicate relative percent difference (RPD) for these replicates was 27%. Analysis of the six field replicates using the hormone method yielded 23 paired and seven unpaired concentration comparisons. The median RPD for these analyses was 31%. Laboratory replicate results are discussed in Supplemental Materials.

2.5. Data analysis

Because of the large number of analytes included in this study, it is necessary to assess concentrations of analytes by compound type. The primary data analysis focuses on a discussion of variability in concentrations among the six regions for four different categories (types or classes) of compounds, with additional select comparisons using individual compounds.

The four different categories include total concentrations of personal care/domestic use compounds (PCDU), total PAHs, total androgens and total estrogens. Table S03 details the compounds included in each of these categories, provides percent detections for each individual compound included in the category, and provides concentrations that correspond to the 50th, 75th, and 90th percentiles. (Table S04 provides similar information on the concentrations of total androgens, total estrogens, total PAHs and total PCDU).

Detected concentrations of 11 different compounds were used to compute the PCDU compounds, including several fragrances (acetophenone, galaxolide [GAL], and tonalide [TON]), surfactants (4-*tert*-octylphenol [4tO], nonylphenol monoethoxylate [NP1EO] and *p*-nonylphenol [*p*NON]), and other compounds associated with consumer use. Total androgens includes the five androgens that were detected (and are based largely on concentrations of the two androgens detected most frequently: 4-androstene-3,17-dione [ADSD] and *cis*-androsterone [CAND]). Total estrogens includes the sum of the four detected estrogens, but is largely based on the two estrogens detected in half or more of the samples (17 β -estradiol [E2] and estrone [E1]). Total PAHs includes

concentrations of the 10 PAHs detected in samples. Several PAHs (2,6-dimethylnaphthalene, anthracene [ANT], benzo[*a*]pyrene [BAP], fluoranthene [FLU], and pyrene [PYR]) were detected in more than 80% of the samples, and constitute the majority of the total PAH concentrations.

Because much of the data are not normally distributed, non-parametric statistical methods were used to identify relations among the data. Non-parametric methods are generally used with water-quality data (Helsel and Hirsch, 2002) and are more robust than parametric approaches for these types of data. Statistical analyses included the analysis of variance using the Kruskal–Wallis test to compare concentrations among regions, and Spearman correlations and lowess smooth lines to assess relations among the variables. Detailed discussion of the approaches used for data that near reporting levels is given in Supplemental Materials.

3. Results and discussion

3.1. Regional variability in wastewater and hormone concentrations

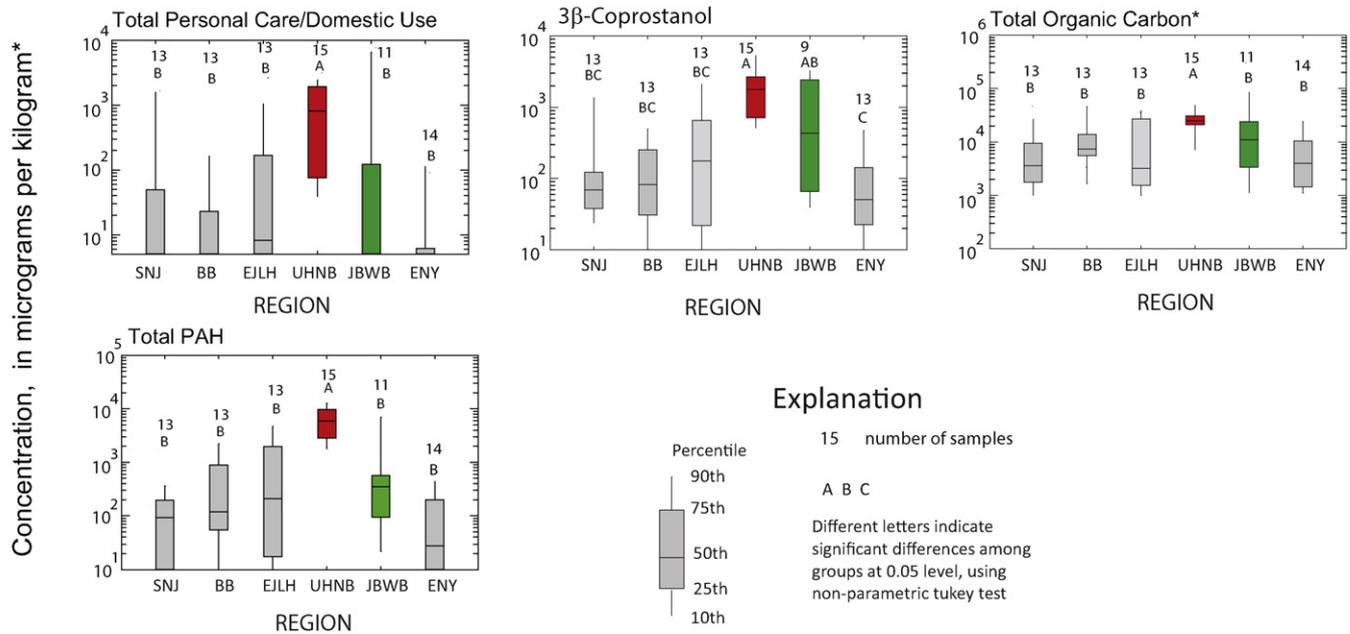
Contaminant distribution and concentrations varied by region and contaminant category. Most of the different compound categories (e.g., PCDUs, TOC, PABs, and PAHs) showed a similar pattern of significantly higher concentrations in the Upper Harbor/Newark Bay (UHNH) region compared to the other 5 regions. However, concentrations of hormones (i.e. total androgens and total estrogens) and CHO did not vary by region, despite their expected association with wastewater inputs. We discuss our findings below with 1) the compound classes that have maximum concentrations in UHNH and 2) the compound classes with similar concentrations across all regions. Because samples were collected from sites after Hurricane Sandy occurred, and were not available for pre-Hurricane Sandy conditions, it is not possible to directly relate the occurrence of these compounds to the effects of Sandy. However, these data provide a useful baseline for assessing future changes in concentrations of a wide range of compounds that to date have not been assessed in bed sediment in the study area.

3.1.1. Wastewater and plant and animal biochemical compounds with highest concentrations in UHNH region

In urban environments, wastewater, stormwater, and combined sewer overflows have been shown to be significant sources of a variety of wastewater contaminants including PCDUs, hormones, and PAHs (Ellis, 2006; Gasperi et al., 2014; Phillips et al., 2012; Nilsen et al., 2014; Madoux-Humery et al., 2015). We found similar results in this study; concentrations of total PCDU compounds, TOC, COP, and other PAB compounds were highest in the UHNH region compared to the other regions (Fig. 1). Over 70% of the land-use in the UHNH and Jamaica Bay/Western Bay (JBWB) regions is classified as developed, and over 60% of the inundated area in the UHNH region was developed land (Fig. S02). Thus, the high concentrations of PCDU and PAB compounds in the UHNH region likely reflect large inputs of treated and untreated wastewater to these sites, as well as other factors identified below.

PCDU compounds were most frequently detected in the UHNH region; eleven PCDU compounds were detected in bed sediment samples (Table S03) and most of these were detected in the UHNH region. For example, the disinfectant 1,4DB was detected in every sample collected in the UHNH region (in concentrations ranging from 12 to 150 μ g/kg; see Fig. S03); by contrast, 1,4DB was detected at concentrations greater than 10 μ g/kg in less than 10% of the samples collected in other regions. BPA, the fragrances GAL and TON, and detergent degradates showed similar patterns (Fig. S03). Similarly, one or more of the three surfactants (4tO, *p*NON, and NP1EO) were detected in >90% of the samples collected in the UHNH region, with concentrations of these compounds generally >300 μ g/kg and as high as 1900 μ g/kg. Several of the PCDU

A. Compounds with highest concentrations in Upper Harbor/Newark Bay Region.



B. Compounds with concentrations that do not differ among regions.

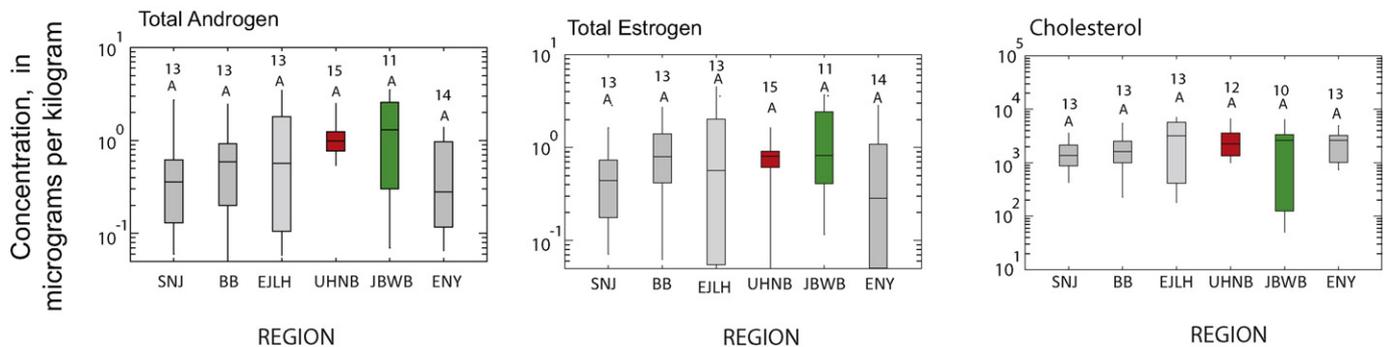


Fig. 1. Concentrations of select compounds in estuarine bed sediment samples collected in New York and New Jersey, 2013. A. Compounds with concentrations that are highest in samples from Upper Harbor/Newark Bay. B. Compounds with concentrations that do not differ by region. SNJ = South New Jersey, BB = Barneget Bay, EJLH = Eastern New Jersey and Lower Harbor, UHNB = Upper Harbor and Newark Bay, JBWB = Jamaica Bay and Western Bay, ENY = Eastern New York. *Units for TOC are milligram per kilogram.

compounds have been found in estuaries impacted by sewage inputs (Lara-Martín et al., 2014; Zhang et al., 2014; Koyama et al., 2013; Chen et al., 2006).

PCDU compounds were less frequently detected in the JBWB and EJLH (Eastern New Jersey and Lower Harbor) regions compared to the UHNB region, and were rarely detected in the other three regions. For example, one or more surfactants were detected in 31% of the samples from the EJLH region, and in 27% of the samples from the JBWB region. With the exception of the SNJ (Southern New Jersey) region (17% detection for surfactants), surfactants were not detected in the remaining regions. These detections reflect differences in land use among the regions, as JBWB and EJLH regions are less developed than the UHNB region, but more developed than regions to the south or east (Fig. S02).

Many of the PCDUs found in the sediments in this study have been identified in past studies as resulting from discharge of poorly treated or untreated wastewater by combined sewer overflows (CSOs) (Phillips et al., 2012). The average number of CSO outfalls within 2 km (kilometers) of the 15 sample sites from the UHNB region is 9, and only one of these sites does not have a CSO outfall within 2 km (see Fischer et al., 2015, Table 10-2). By contrast, only two of the sites

sampled in the EJLH region and in the JBWB regions had a CSO outfall within 2 km, and only one site in the Eastern New York (ENY) region had a CSO outfall within 2 km. Elevated bed sediment concentrations of PCDUs have been found in estuaries impacted by sewage inputs (Lara-Martín et al., 2014; Zhang et al., 2014; Koyama et al., 2013; Chen et al., 2006). The extent to which elevated concentrations of PCDUs in samples from the UHNB, JBWB, and EJLH regions reflect releases of sewage from CSOs during Sandy (and other storms) versus treated wastewater discharges is not clear, as there are no data available for comparison with pre-Sandy conditions.

Concentrations of TOC and PABs were generally higher in the UHNB region than in other regions and likely reflect increased wastewater contributions in the region. Median TOC and COP concentrations for UHNB (25,000 mg/kg and 1800 µg/kg, respectively) were higher than the respective median concentrations from all other regions (Fig. 1). Median TOC and COP concentrations were also elevated in the JBWB region compared to other regions (median JBWB TOC > 10,000 g/kg, other region's median < 5000 mg/kg; median JBWB COP > 400 µg/kg, other region's median < 200 µg/kg). High TOC concentrations in the UHNB region has been noted (Adams, 2003), and these findings are consistent

with studies that found elevated TOC in sediment samples downstream of cities and demonstrating the link between wastewater inputs and TOC on a regional scale (Abril et al., 2002; Zhang et al., 2014).

In addition to COP, several other PABs exhibited similar regional differences. Skatole (SKAT), indole (INDO), and β -stigmastanol were all higher in the UHNB and JBWB samples than the other regions (Fig. S03). SKAT and INDO are major components of sewage stench (Rudolfs and Chamberlin, 1932) and their elevated concentrations reflect the importance of wastewater impacts on the UHNB region, and to a lesser extent the JBWB region.

The highest concentrations of PAHs occurred in samples from the UHNB site; median total PAH concentrations for these samples were 6000 $\mu\text{g}/\text{kg}$, whereas median total PAH concentrations ranged from 350 to 510 $\mu\text{g}/\text{kg}$ for samples from the JBWB and EJLH regions, and less than 100 $\mu\text{g}/\text{kg}$ for the other regions (Fig. 1). We expected to find elevated concentrations of PAHs in the UHNB region because some of the highest PAH concentrations in the United States have been documented in the UHNB region (Adams, 2003; Ayres and Rod, 1986; Lodge et al., 2015). Stormwater runoff is the major contributor of PAHs to the New York Harbor waters, with CSOs as a lesser source (Rodenburg et al., 2010). The persistence of PAHs and other organic pollutants (including PCBs, DDT, and other persistent organochlorines) in UHNB has been attributed to 'estuarine trapping' where re-suspended sediments are carried upstream due to the interaction of fresh and saltwater (Lodge et al., 2015). The lack of PAH data for sediments in the period (<1 year) just before Hurricane Sandy occurred makes it impossible to assess the impact of Hurricane Sandy on PAH concentrations. However, the patterns of PAH data across these regions did not differ from patterns present well before Sandy (>1 year) suggesting that the PAHs are primarily the result of historical PAH inputs rather than inputs of fuel oils and other petroleum products discharged into the estuaries during Hurricane Sandy. Moreover, PAH ratios (see below) support the conclusion that the observed PAHs at these sites were not the result of fuel spills associated with inundation.

Comparison of concentrations of PAHs in this study with standards used to indicate the potential for adverse biological effects show similar regional patterns to those observed in past studies. Many of the PAHs detected in this study have available concentration ranges that have been identified in past studies as having biologic effects. ERL (Effects Range – Low) and ERM (Effects Range-Median) concentrations identified by Long and Morgan (1991) and Long et al. (1995) have been used in previous assessments of sediment quality in New York Harbor (Adams, 2003; see Table S05 for these levels) and so are used in this study to indicate samples with PAH contamination. Comparison between ERL and ERM concentrations for the samples from different regions show that all the samples collected in the UHNB region exceed ERL levels for at least one PAH, and 33% exceed ERM levels (Fig. S04). By contrast, all other regions generally have less than 40% of the sites with one or more PAHs exceeding an ERL level, and none of the other regions have sites with a sample exceeding a ERM level. This is similar to findings by Adams, 2003 who found the highest exceedance of PAH ERL and ERM levels in the Upper Harbor and Newark Bay portions of the New York Harbor. As with our study, Adams (2003) found less frequent exceedance of ERL levels in the Lower Harbor and Jamaica Bay portions of New York Harbor (which correspond to portions of the EJLH and JBWB regions, respectively).

3.1.2. Compounds with concentrations that do not differ among the regions

Concentrations of total androgens, total estrogens and CHO, unlike most other compounds included in this study, were not higher in samples collected from the UHNB region than other regions (Fig. 1). This pattern is unexpected considering 1) the high sewage inputs to the UHNB region relative to other regions, and 2) the documented association between CSO inputs and hormone loads found by Phillips et al., 2012. These two factors suggest that the distribution of these compounds would be similar to the other sewage-associated compounds, including PCDUs, COP and indole (Figs. 1, S03).

Hormones were commonly detected in samples, with 90% of the samples containing one or more androgens and 85% containing one or more estrogens. Total androgens ranged from a median of 0.36 to 1.3 $\mu\text{g}/\text{kg}$ among all regions (Fig. 1). Although the highest median concentration for androgens (>1 $\mu\text{g}/\text{kg}$) occurred in samples from the UHNB and JBWB regions, the differences among the regions were not significant. ADSD comprised most of the total androgen concentration (generally >85%), and when detected, was around 0.6 $\mu\text{g}/\text{kg}$ (Fig. S05). CAND and epitestosterone were less frequently detected, and when detected were generally less than 0.25 $\mu\text{g}/\text{kg}$. Median total estrogen concentrations ranged from 0.3 to 0.9 $\mu\text{g}/\text{kg}$ among the regions, with the highest median concentrations occurring in the Barneget Bay (BB), UHNB, and JBWB regions (Fig. 1); again, the differences among the regions were not statistically significant. E1 was the predominant estrogen, as it comprised more than 95% of the total estrogens, and when detected was around 0.7 $\mu\text{g}/\text{kg}$ (Fig. S05). E2 was also detected in many (around half) of the samples, but when detected was generally present in concentrations <0.25 $\mu\text{g}/\text{kg}$ (Fig. S05).

The predominant androgens and estrogens detected at the highest concentrations in bed sediment in this study generally correspond to those most commonly detected in wastewater effluent or in CSO discharges. For example, Phillips et al. (2012) found that ADSD and CAND represent the bulk of androgens in treated and untreated wastewater, and that E1 has higher concentrations in treated and untreated effluent compared to Estriol (E3) and E2. E1 was the predominant estrogen present in bed sediment samples in the highly urbanized Snongua River, China (Zhang et al., 2014), and concentrations peaked downstream of large cities and were similar (0.1 to 3 $\mu\text{g}/\text{kg}$) to what we observed in this study, as were concentrations reported in the JBWB region in a previous study (Reddy and Brownawell, 2005). Although androgens are less frequently included in assessment of hormones in the environment, their concentrations likely reflect sewage inputs and they recently have been found to affect fathead minnow reproductive physiology (DeQuattro et al., 2015). Given that other studies have shown that both CSOs and treated effluents are important sources of estrogens and other wastewater associated compounds including PCDUs (Zhang et al., 2014; Isobe et al., 2006; Petrovic et al., 2002), and the distribution of other wastewater compounds, we expected total androgen and estrogen concentrations to be highest in the UHNB region. Thus, these results indicate that some other factors may be affecting concentrations of the hormones and potentially CHO in the UHNB region; possible factors causing this are addressed in a subsequent section on interrelations among concentrations of hormones and other compounds.

3.2. Ratios of concentrations of select analytes as source indicators

The ratio of COP to CHO concentrations (COP/CHO) indicates substantial differences in wastewater contribution to sediment chemistry across the region and is consistent with highest sewage impacts in UHNB region samples (Fig. 2). COP/CHO ratios have been widely used to indicate human wastewater discharges (Tse et al., 2014; Furtula et al., 2012; Saim et al., 2009; Vane et al., 2010) with a ratio of >0.2 used as an indicator of human fecal sources. The two most intensively developed regions with the closest proximity to both treated and untreated wastewater discharges, UHNB and JBWB, had COP/CHO ratios significantly greater than those observed at other regions (Fig. 2). Median COP/CHO ratio in the UHNB region was 0.7, indicating strong likelihood of human fecal sources for most of the samples from this region. Moreover, the COP/CHO ratios were consistently >0.2 across all 15 samples from the UHNB region, indicating that all samples in this region were affected by wastewater. Median COP/CHO ratios were also elevated in the JBWB (median of 0.2) compared to the other regions with median COP/CHO ratios ranging from 0.03 to 0.07 (Fig. 2). COP concentrations >500 $\mu\text{g}/\text{kg}$ are also considered indicative of human wastewater contamination (Readman et al., 2005). Of the 24 samples with COP/CHO

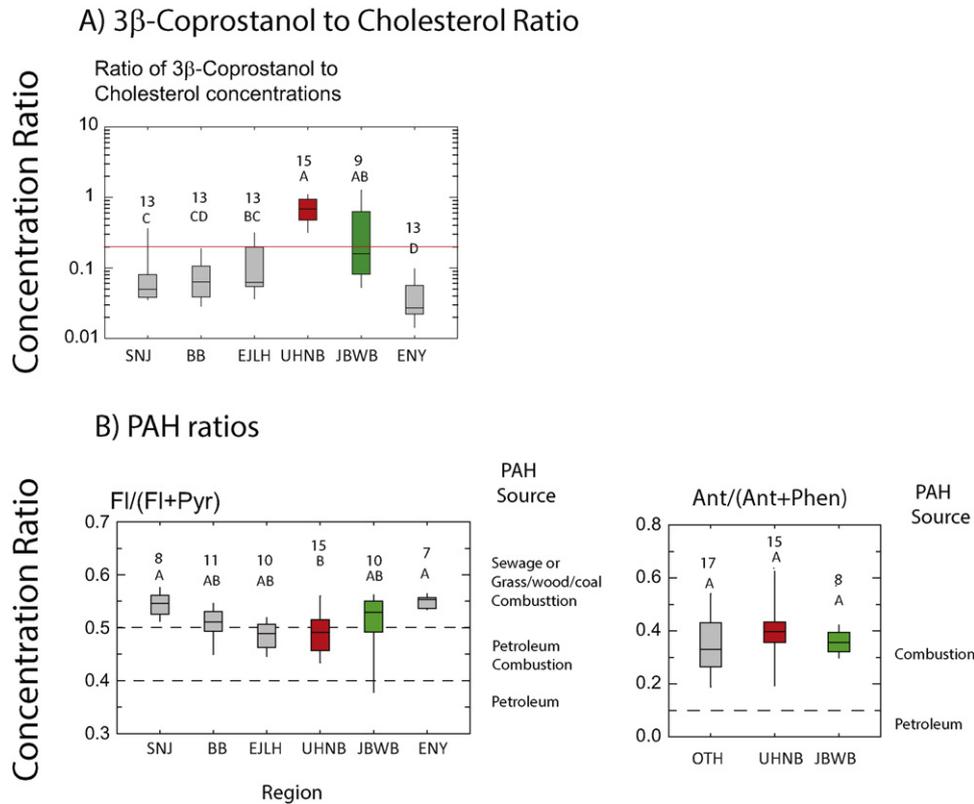


Fig. 2. Plots of A) ratio of 3β -coprostanol to cholesterol and B) ratios of select PAHs for estuarine bed sediment samples collected in New York and New Jersey, 2013. Data for regions other than UHNB and JBWB are categorized as OTH for graphs and analysis for the Anthracene to Anthracene plus Phenanthrene due to low numbers of detection for these compounds in most regions. Descriptions of sources by ratios come from [Kanzari et al., 2014](#). Ant = anthracene; Phen = phenanthrene; FI = fluoranthene; Pyr = pyrene.

ratio >0.2 , all but 3 had COP concentrations greater than $500 \mu\text{g}/\text{kg}$; most of the samples with COP concentrations >500 and COP/CHO ratio >0.2 are in the UHNB region.

Ratios of PAHs indicate that bed sediment PAHs were consistently from pyrogenic sources across all regions (Fig. 2). Several studies have used PAH ratios to differentiate between pyrogenic and petrogenic sources ([Kanzari et al., 2014](#); [Araghi et al., 2014](#); [Yunker et al., 2002](#)) and we utilized two ratios for this purpose: fluoranthene to fluoranthene plus pyrene (FI/(FI + Pyr)) and anthracene to anthracene plus phenanthrene ratio (Ant/(Ant + Phen)). FI/(FI + Pyr) ratios could be calculated for most samples ($n = 61$), so that statistical comparisons were possible among the 6 regions; Ant/(Ant + Phen) ratios were available for around half the samples ($n = 40$) so comparisons were only widely available for the UHNB and JBWB regions; values for the other regions were pooled for analysis.

Both PAH ratios indicated that the predominant source of PAHs for the samples collected in this study are generally from pyrogenic sources and not petrogenic sources, suggesting that fuel spills, although potentially important in certain settings, were not a major regional source of PAHs for these samples. All but one of the samples collected in the study area with sufficient data for calculation of FI/(FI + Pyr) ratios had a ratio >0.4 , indicating a pyrogenic source ([Kanzari et al., 2014](#)). Similarly, all but one of the 40 samples for which Ant/(Ant + Phen) ratios were available had a ratio >0.1 (Fig. 2), again suggesting that the primary source of PAHs in these samples were formed from combustion of organic material, and not unburned petroleum sources ([Kanzari et al., 2014](#)). The ratio of FI/(FI + Pyr) varied across the regions, with the lowest ratios occurring in the highly developed UHNB region, and the highest in the least developed regions (Southern New Jersey and Eastern New York). These higher ratios may indicate that in the less developed areas, sources besides petroleum combustion (such as sewage, grass, wood, or coal combustion) may represent a more important source of PAHs than in more developed areas.

Although flooding during Hurricane Sandy caused fuel spills in portions of the study area, particularly the UHNB and JBWB regions, the ratios of PAHs indicate that these fuel spills were not a major source of PAHs to the bed sediments in this sampling network. This study did not target locations of fuel spills, and more detailed sampling adjacent to fuel spills that occurred during Sandy could produce a different result. Across the entire study area, however, these data indicate that fuel spills were not an important source of sediment PAHs.

3.3. Interrelations among hormone concentrations and concentrations of other compounds

The lack of higher estrogen and androgen concentrations in bed sediment in the UHNB region is surprising, given the findings in other studies that show a link among wastewater inputs in urbanized estuaries and hormone concentrations. As indicated previously, researchers have frequently found highest bed sediment concentrations of hormones associated with proximity to wastewater discharges ([Isobe et al., 2006](#); [Lei et al., 2009](#)). Relations between hormone concentrations and other compounds included in this study give insights into potential causes for the observed lack of elevated concentrations of hormones in the UHNB region.

Hormones, PCDUs, and other wastewater source compounds were strongly positively correlated with TOC concentrations (Spearman-rho values ranged between 0.7 and 0.87) (Figs. 3, S06). However, for total androgens, total estrogens, and cholesterol, the nature of the relationship shifted at high TOC concentrations (Figs. 3, S06). Sewage inputs have been identified as important sources of total organic carbon in other urbanized estuaries ([Abril et al., 2002](#); [Zhang et al., 2014](#)). TOC is also known to increase sorption of hormones, PCBs, alkylphenols, and other hydrophobic chemicals and is frequently correlated with organic contaminants ([Lai et al., 2000](#); [Nowell et al., 2013](#)). However, examination of lowess lines for the relations of TOC with total androgen, total

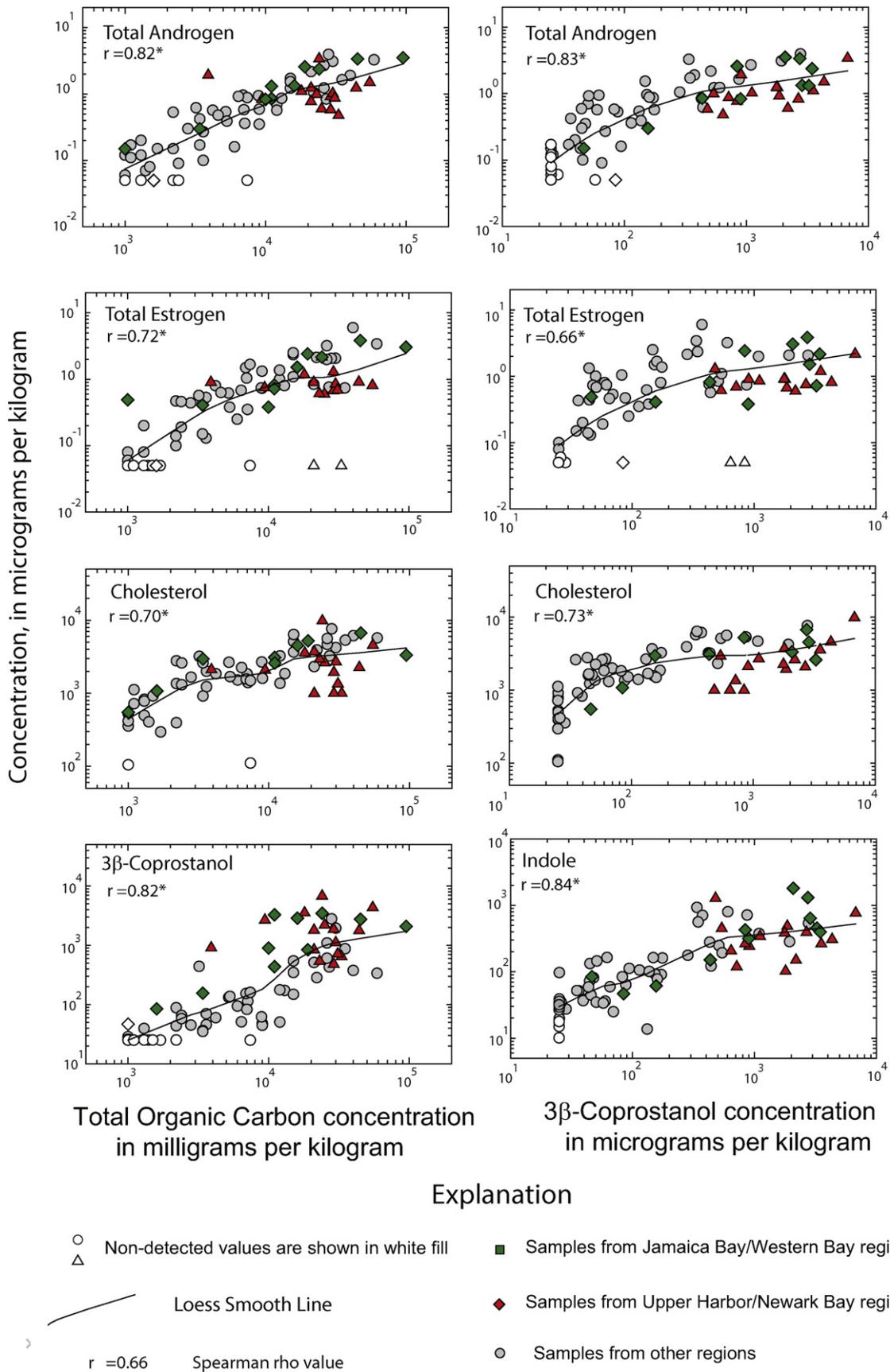


Fig. 3. Plots of total organic carbon and 3β-coprostanol with select compounds for estuarine bed sediment samples collected in New York and New Jersey, 2013 with lowess line indicating trend.

estrogen, and CHO indicate that many of the samples collected in the UHNB region have lower concentrations of these analytes than predicted by the overall trend (Figs. 3, S06). The total androgen and total estrogen lowess lines also tend to have a decrease in slope for TOC concentrations at about 10^4 mg/kg, which is the lower end of TOC concentrations for most of the samples collected in the UHNB region.

Lower than expected concentrations for hormones and cholesterol based on TOC concentrations in the UHNB region is unique among all the compounds. Samples from the UHNB region tend to plot above the lowest trend line for relations between TOC and COP and total PAH, and along the line for indole (Fig. 3, Fig. S06). Moreover, plots of CHO with total androgen and total estrogen show a similar pattern of samples from the UHNB region falling below the lowess line, while indole falls along the lowess line (Fig. 3, Fig. S06), suggesting a preferential decrease in hormones in the UHNB region. The elevated concentrations of COP and total PAH for UHNB samples with TOC compared to the lowest trend line are attributable to the large inputs of wastewater (for COP) and known elevated inputs of PAHs in the UHNB region. By contrast, the lower androgen and estrogen concentrations for samples from the UHNB region with respect to both TOC and COP (Figs. 3, S06, S07) offer further evidence that some process or processes are diminishing the hormone concentrations in this region.

One possible reason for the lower than expected concentrations is that conditions at these sites may have promoted hormone and cholesterol degradation. Tan et al., 2015 showed that E1 degradation was promoted by 8 day old wastewater, but not by fresher wastewater. These researchers concluded that incubation of fresh wastewater resulted in the generation of microbial degradation products that enabled E1 degradation, with the aging of the wastewater resulting in selection of bacteria that degraded E1. Data from the present study indicate that a similar process could be promoting the degradation of total androgens at some sites (particularly those in the UHNB region) resulting in the observed lower than expected concentrations of these compounds. Concentrations of CHO for samples from the UHNB region are similarly lower than predicted by the lowess line for TOC and COP (Fig. 3, S06–S08), indicating that CHO concentrations may be decreased by the same processes in the UHNB region.

4. Conclusions

Bed sediment samples collected from 79 estuarine sites in the area affected by Hurricane Sandy in New York and New Jersey and analyzed for a variety of wastewater associated compounds, hormones, and PAHs, generally show a regional trend for most measured contaminants. These samples represent the first large scale assessment of wastewater associated compounds (including fragrances, detergent degradates) and hormones in this area. Concentrations of total PCDD compounds and PAHs were highest in samples collected from the Upper Harbor/Newark Bay region and lower in other areas to the east and south. For example, one or more of the three surfactants (4rO, pNon, and NP1EO) were detected in >90% of the samples collected in the UHNB region, with concentrations generally >300 µg/kg and as high as 1900 µg/kg. By contrast, one or more of these compounds were detected in few (<20%) of the samples from other regions, and when detected were generally <250 µg/kg. Although the lack of long-term (pre-Hurricane Sandy) data make it impossible to assess the impact of Hurricane Sandy on wastewater contaminants, these elevated concentrations reflect the larger wastewater inputs associated with the high amounts of development in this area. By contrast, the elevated PAH concentrations in the UHNB region reflect known patterns of contamination in the area that are in part attributable to long-term (pre-Hurricane Sandy) sediment deposition in this area.

The importance of wastewater inputs in the UHNB region are further illustrated by the COP/CHO ratios (generally >0.1), which are indicative of sewage sources. Ratios of select PAH compounds indicate that the main source of PAHs in all regions is pyrogenic PAHs, and are not

attributable to petrogenic PAHs. These results show that PAH concentrations in post-Sandy sediments do not reflect petroleum spills.

Although the UHNB region generally has the highest concentration of wastewater derived compounds such as surfactants and fragrances, hormone concentrations in bed sediment samples from this region are not significantly higher than those present in other regions. Concentrations of total androgens generally ranged from 0.4 to 1.3 µg/kg, with ADSD the most commonly detected androgen. Total estrogens generally ranged from 0.3 to 0.9 µg/kg, with E1 the predominant form of estrogen in bed sediments.

The reason for the lower than expected hormone concentrations in the UHNB region is unclear, but may be related to biodegradation in the samples collected from this region. Examination of lowess smooth line plots for TOC with total androgen, total estrogen, and cholesterol indicate that many of the samples collected in the UHNB region have lower concentrations of these compounds than predicted by the regional trend. Lower than expected concentrations for hormones and cholesterol in the UHNB region is unique among all the compounds. Thus, the lower androgen and estrogen concentrations for samples from the UHNB region with respect to TOC and COP are a further indication that an unknown process could be decreasing the hormone concentrations in the study area. Data from the present study indicate that biodegradation in aged wastewater, which has been observed in other studies, may be occurring in the UHNB region, and so resulting in the lower than expected hormone concentrations. These data provide the first large scale assessment of a large variety of wastewater sourced compounds in the New York and New Jersey estuarine area impacted by Hurricane Sandy, and also provide a new baseline for post-Sandy contaminant conditions.

Contributions

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Network design and sampling: Phillips, S Fisher, I Fisher, Reilly, Smalling, Focazio
GIS and Land Use Analysis, preparation of figures: Phillips, Jones
Data Management: Romanok, I Fisher
Chemical Analysis: Foreman, ReVello

Acknowledgments

This study was funded through the Disaster Relief Appropriations Act of 2013 (PL 113-2).: Several USGS employees contributed to sampling: Lisa Carper, Anthony D. Cerruti, Kaitlyn Colella, Jonathan Cohl, Anna Deetz, Heather A. Heckathorn, and Molly L. Schreiner. Special thanks are due to Eric W. Best, Patrick W. Bowen, Karl M. Braun, Heidi L. Hoppe, Michal J. Niemoczynski, Jason C. Shvanda, Daniel D. Skulski, and John J. Trainor. The US Environmental Protection Agency (EPA) provided sampling and analytical support at the direction of Darvene Adams, Regional Monitoring Coordinator.

Agencies and researchers consulted on study design including Lorraine Holdridge, New York State Department of Environmental Conservation; Gary Buchanan, Steve Anderson, Sherry Driber, Angela Witcher, and Michael DiGiore, New Jersey Department of Environmental Protection; Jim Browne and Ron Masters, Town of Hempstead, New York; Michael Jensen, Mark Long, Shawn Droskoski, and Alison Branco, Suffolk County Department of Health Services, New York; Bruce Brownawell and Park Ng, Stony Brook University, Stony Brook, New York; and Caitlyn Nichols, New England Interstate Water Pollution Control Commission.

Tia-Marie Scott, USGS, provided comments on an early draft version.

Appendix A. Supplementary data

Supplemental Materials associated with this article can be found in the online version <http://dx.doi.org/10.1016/j.marpolbul.2016.04.050>.

These materials include tables and text discussing methods used, sites sampled, and compounds used in statistical analysis presented in the paper. Supplemental Materials also includes eight figures, Figs. S01–S08 (found in the Supplemental Materials text) and five tables, Tables S01–S05. (provided as an excel spread sheet).

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