Regional assessment of persistent organic pollutants in resident mussels from New Jersey and New York estuaries following Hurricane Sandy

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ABSTRACT (150 words)

Resident mussels are effective indicators of ecosystem health and have been utilized in national assessment and monitoring studies for over two decades. Mussels were chosen because contaminants concentrations in their tissues respond to changes in ambient environmental levels, accumulation occurs with little metabolic transformation and a substantial amount of historic data were available. Mussels were collected from 10 previously studied locations approximately a year after Hurricane Sandy. Regionally, concentrations of polychlorinated biphenyls (PCBs) and polycyclic aromatic hydrocarbons (PAHs) decreased significantly, while concentrations of organochlorine pesticides (OCPs) remained unchanged, and polybrominated diphenyl ethers (PBDEs) increased compared to historic concentrations. Although concentrations of PCBs, OCPs and PAHs were at or near record low concentrations, long-term trends did not change after Hurricane Sandy. To effectively measure storm-induced impacts it is necessary to understand the factors influencing changes in mussel body burdens, have a long-term monitoring network and an ability to mobilize post event.

Keywords: Persistent organic contaminants, mussel tissue, Hurricane Sandy, estuarine health, chronology

1. Introduction

Bivalves are widely distributed in coastal environments and have long been used as resident sentinels for assessing ecosystem health and productivity (Farrington, 1983). Resident mussels in particular have been used to assess contaminant concentrations in near-shore environments because they are sessile organisms that filter feed and accumulate contaminants directly from the water column (Chase et al., 2001; Sericano et al., 1995). Due to the structure of their digestive systems and lack of a liver-like detoxification function, mussels cannot efficiently metabolize most organic contaminants, making them an ideal integrator of contaminants at the local and regional scale (Baumard et al., 1998; Chase et al., 2001).

The National Oceanic and Atmospheric Administration's (NOAA) Mussel Watch Program began in 1986 and is one of the longest and continuously running coastal monitoring programs in the United States whose mission is to monitor and report the status and trends of contaminants in U.S. coastal waters (Kimbrough et al., 2008). The data generated from the Mussel Watch program has helped characterize the environmental impacts of contaminants (new and legacy) throughout the coastal US (Kimbrough et al., 2008; Kimbrough et al., 2009) and has been useful in the interpretation of potential local and regional impacts from events such as hurricanes, oil spills and other disasters (Soriano et al., 2006; Lauenstein and Kimbrough, 2007; Johnson et al., 2009; Apeti et al., 2011).

Hurricane Sandy caused widespread damage to coastal New Jersey and New York in October, 2012. The resulting disturbances from the storm were considered possible threats to vulnerable coastal ecosystems due to the potential remobilization of contaminants from disturbed bottom sediment as well as inputs from compromised infrastructure (Buxton et al., 2013). Certain anthropogenic hydrophobic contaminants can be stored for extended periods in bottom sediments and resuspension can reintroduce them into the water column, making them more bioavailable for uptake by bottom-dwelling aquatic organisms. Mussels used as sentinel organisms to assess ecosystem health, are filter feeders that can provide an integrated biotic perspective on storm induced impacts of contaminants. For example, resuspension of bottom sediment following an event has the potential to increase the availability of contaminants thus altering the contaminant body burden of resident mussels. Conversely, storms can have a scouring effect on bottom sediments in some estuarine environments, removing adsorbed contaminants from system, and possibly decreasing mussel body burdens. Bivalves have shown a diverse response to contaminant exposure, including decreased immune response (increase susceptibility to parasites), decreased recruitments, and mortality (Bushek et al., 2007; Weis et al., 1994) which could ultimately result in population declines.

In the United States, studies to assess the impacts of storms on contaminants from an ecological health perspective tend to be localized and limited to strong hurricanes such has Hurricanes Katrina and Rita in the Gulf of Mexico (Johnson et al., 2009; Apeti et al., 2011). Although these studies are regionally limited, the results and conclusions are relevant and comparable to other regions and events. To characterize the potential effects of Hurricane Sandy on contaminant redistribution and bioavailability, mussels were chosen to compare body burden residues post-Sandy to trends generated by the Mussel Watch Program pre-Sandy similar to other studies (Johnson et al., 2009; Apeti et al., 2011). Filter feeders such as mussels are ideal because contaminant concentrations in their tissues quickly respond to changes in ambient environmental levels, accumulation occurs with very little metabolic transformation and over 20 years of historical data for the study area are available for many persistent organic pollutants of interest (Kimbrough et al., 2008; Kimbrough et al., 2009).

The objective of the study was to assess the impacts of Hurricane Sandy on the distribution of persistent organic pollutants in mussels. Two mussel species *(Mytilus edulis* and *Geukensia demissa*) were collected throughout the storm-impacted area, the shells were thin-sectioned and aged and the tissues were analyzed for polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), polycyclic aromatic hydrocarbons (PAHs) and organochlorine pesticides (OCPs). Local and regional data after Hurricane Sandy were compared to long-term trends generated for these contaminants over the past 20 years by the Mussel Watch Program. A subset of the mussels collected were aged to determine if the size class available had survived Hurricane Sandy and if the age classes were similar in order to conduct a regional comparison. Understanding the persistence and accumulation of contaminants in mussel tissue will help scientists further assess the impacts to ecosystem health and establish a new baseline for tissue-bound contaminants in the aftermath of a major coastal storm.

2. Methods

2.1 Study area and sample collection

The study area consisted of estuaries adjacent to lands in New Jersey and New York that were inundated by the storm surge from Hurricane Sandy (Fig 1). Sampling locations were selected on the basis of the availability of pre-Hurricane Sandy tissue data (Kimbrough et al., 2008) in order to facilitate a comparison of contaminant data after the storm to a 20 year record. Resident blue mussels (*Mytilus edulis)* and ribbed mussels (*Geukensia demissa*) were collected from 10 NOAA Mussel Watch locations (Fig 1, Table 1) along the New Jersey and New York coastline from December 2013 to April 2014 using previously published methods (Lauenstein and Cantillo, 1993a). At each sampling site, approximately 100 individual mussels were collected from 2 to 3 stations approximately 25 meters (m) apart. Mussels ranging in length from 20 to 80 mm, depending on species and the availability of mussels at each site, were collected by hand, rinsed with seawater to remove any residual debris, and placed in 5-gallon buckets. Only mussels with tightly closed shells were collected. All mussels were collected from rocks, jetties, and marsh sediment near the shoreline. Blue mussels were collected from 8 of the 10 sites, ribbed mussels were collected from 3 of the 10 sites and both species were collected from the Jamaica Bay, NY (HRJB) site (Table 1).

After collection, the mussels were placed in two separate 1-gallon Ziploc bags using gloved hands (50 mussels per bag). The bags were labeled A and B, representing replicate samples which consisted of 50 randomly pooled mussels from the 2–3 stations at each site. All samples were placed in a cooler on ice, transported back to the laboratory, and stored frozen at -20 °C prior to processing and analysis.

In the laboratory, mussels were thawed, and the shells were opened using a methanol rinsed spatula. The tissue was removed with methanol rinsed forceps and placed into a 500-mL clean, baked amber jar. Each 500-mL jar represented a composite tissue sample (~50 individuals) from a single replicate composite sample collected from one location. The tissue samples were frozen at -20 °C and shipped on ice to the NOAA Northeast Fisheries Science Center James J. Howard Marine Sciences Laboratory at Sandy Hook, NJ for analysis. Forty to 80 randomly selected shells of various sizes were washed to remove excess tissue and debris, placed in 1-gallon Ziploc bags and shipped to the LSC Northern Appalachian Research Laboratory (LSC-NARL), PA for chronology.

2.2 Mussel shell chronology

Mussel shells were measured from the umbo through the longest axis of the mussel using digital calipers and were embedded in EpoThin epoxy resin (Buehler, Lake Bluff, Illinois). Embedded shells were sectioned through the umbo, again through the longest axis, using an Allied TechCut4 diamond blade saw (Allied, Rancho Dominguez, California). Blue mussel sections were mounted to standard microscope slides, and ribbed mussel shells were mounted on either standard slides or custom 12- x 4-cm glass slides using Devcon 2-ton epoxy (Devcon, Solon, Ohio). Once mounted, shells were sectioned again to a thickness of approximately 0.25 mm. Mounted sections were sanded using a graded sandpaper (320 grit, 600 grit, 800 grit) and polished with 1-μm polycrystalline diamond suspension (Allied, Rancho Dominguez, Calif.), followed by a 0.04-μm colloidal silica suspension (Allied, Rancho Dominguez, Calif.). Slides were stained in Mutvei's solution (1% acetic acid 25% gluteraldehyde mixed with alcian blue) for 50 min at 37 ºC. Annuli were enumerated by teams of at least three people until consensus was reached using a dissecting microscope at 65x. Digital photographs were taken of each slide and annotated (Lutz, 1976).

2.3 Mussel Tissue Extraction and Analysis

Composite mussel tissue samples were analyzed for 4 classes of select persistent organic pollutants including PCBs, PBDEs, PAHs and OCPs using previously published methods (Deshpande et al., 2013; Deshpande and Dockum, 2013). Analytical methods utilized in this study were similar to those conducted previously by the Mussel Watch Program (Lauenstein and Cantillo, 1993b; Lauenstein and Cantillo, 1998; Kimbrough et al., 2007). Briefly, individual whole freeze-dried tissue composites were pulverized in a blender with diatomaceous earth, extracted with dichloromethane (DCM) using a Soxhlet (18 hours) and reduced under nitrogen gas. Prior to extraction, recovery surrogates DBOFB, Ronnel, PCB 198, d_8-napthalene, d_{10}-acenapthene, d_{12}-benzo[a]pyrene and d_{12}-pyrene, and 6-F-PBDE 47 were added to each tissue sample. The bulk polar interfering compounds of biological origin were removed from the target analytes using florisil/silica/alumina glass column chromatography. Following initial clean-up, twenty percent by volume of the extract was used for the gravimetric lipid determination. Lipids and other interferences were removed using high performance liquid chromatography column (Phenogel 10, 600-mm x 21.20-mm, 100 pore size, 10-µm particle size; Phenomenex, Torrance, California). Prior to lipid removal by HPLC, 1,2,3-trichlorbenzene (TBZ) and PCB 192 were added to the samples. HPLC fractions containing the target analytes were collected, solvent-exchanged to hexane, concentrated to less than 1 ml and each final extract was split into three vials for the analysis of (1) PAHs, (2) PCBs and OCPs and (3) PBDEs.

Target analytes were analyzed using an Agilent 6890 GC coupled to an Agilent 5973 MS operating in SIM mode. PAHs, PCB congeners and OCPs were analyzed using a DB-5 0.25 mm ID x 60-m capillary column. PBDE congeners were analyzed by using a Restek 1614 0.25 mm x 15-m PBDE column. Analyte concentrations are expressed as ng/g on a dry weight basis. Reporting limits (RLs) for PAHs, PCBs, OCPs and PBDEs ranged from 0.5 to 3.2 ng/g dry weight.

2.4 Quality Assurance/quality control

A comprehensive set of quality assurance/quality control (QA/QC) parameters were analyzed with the environmental samples and included laboratory blanks, field replicates and standard reference materials. If compounds were detected in the laboratory blanks above the RLs the environmental samples were censored based on the following criteria: if the environmental concentration was less than 3 times the blank value then that value was changed to a non-detect and not reported, if the environmental concentration was greater than 3 times the blank value then the reported concentration was coded with an 'E' and if the environmental concentration is greater than 10 times the blank value than the actual concentration is reported without censoring. One laboratory blank was analyzed with the batch of mussel samples. No PCBs or OCPs were detected in the laboratory blank at greater than the RLs. Two PBDEs, PBDE 49+71 and 99, were detected in the laboratory blank at greater than the RLs. Recovery surrogates were added to each sample to measure method performance. For PCBs and OCPs, mean recoveries with standard deviations of 1,2,3-TCB, DBOFB, Ronnel, PCB 192 and PCB 198 were 97 ± 25%, 70 ± 11%, 75 ± 22%, 117 ± 20% and 90 ± 11%, respectively. For PAHs, mean recoveries with standard deviations of d_8-napthalene, d_{10}-acenapthene, d_{12}-benzo[a]pyrene and d_{12}-pyrene were 44 ± 19%, 61 ± 8%, 58 ± 15% and 68 ± 7%, respectively. For PBDEs, mean recovery with standard deviation of 6-F-PBDE47 was 101 ± 18%. The percent recoveries based on NIST certified values for PAHs, PCBs, PBDEs and OCPs in SRM 1947c (Organics in Mussel Tissue) ranged from 50 to 132% with a median of 79%, from 52 to 139% with a median of 84%, from 71 to 115% with a median of 105% and from 63 to 118% with a median of 77%. All environmental data was considered of acceptable quality based on project QA/QC data as well as the QA/QC criteria supported by the Mussel Watch Program (Kimbrough et al., 2007).

2.5 Historical data retrieval and statistical analysis

Data from the Mussel Watch Program in the study area prior to Hurricane Sandy were compiled to examine the ecological impacts of the storm in a historical context (Smalling et al., 2015). Over 20 years of Mussel Watch Program data for PCB, PAHs and OCPs were available from 1985-2012. For PBDEs the only data available were from 2004 and 2008. Mussel tissues in the current study were analyzed for a suite of organic contaminants including 53 PAHs and alkylated PAHs, 34 PCBs, 24 OCPs and 28 PBDEs (Smalling et al., 2015). However, to compare trends in contaminant concentrations to previous studies (Kimbrough et al., 2008; Johnson et al., 2009) a subset of compounds was used for all statistical analyses (Table 2).

Nonparametric methods were used to evaluate trends in contaminant concentrations in composite mussel-tissue samples from the 10 sites. Nonparametric methods are commonly applied when analyzing biological and other environmental data, as it eliminates the need to assume that specific distributions, such as normal or log-normal apply to the data (Helsel and Hirsch, 2002). The two-sample Wilcoxon test was used to compare pre- to post-Sandy total concentrations of contaminant groups (PCBs, DDTs, chlordanes, dieldrins, PBDEs, low-molecular-weight PAHs, high-molecular-weight PAHs, and total PAHs). Where the p-value (probabability of no difference between the distributions of pre-and post-Sandy samples) was ≤ 0.05, a statistically significant change in median concentrations was indicated. A negative Z-statistic value indicated a decrease in the median concentration after Sandy, and a positive value indicated an increase. Spearman's rank correlation coefficient (rho) was used to test for trends in concentrations of contaminant groups over time at each site. Significant increasing or decreasing trends are indicated by p-values ≤ 0.05, and the value of rho indicates the slope (rate) of increase or decrease in concentration over time. The application S-Plus (TIBCO Software Inc., Palo Alto, CA) was used for all statistical analysis.

A von-Bertalanffy growth model was fitted to the length at age data using the 'fishmethods' package in R (R core team, 2015). Due to a targeted size range for field sampling, the program could not converge on a model solution. Instead, a quadratic equation was fitted to length-at-age data using SimgaPlot v. 12.5. Deviations from the model were compared among sites using a nonparametric Kruskal-Wallis test with IBM SPSS Statistics v. 20.0.0.2 software. Pairwise post hoc tests were also completed with SPSS to assess among site differences in growth rates.

3. Results and Discussion

3.1 Age of resident mussels

In an attempt to understand the ecological impacts of Hurricane Sandy, 10 Mussel Watch sampling locations along the New Jersey and New York coastline were reoccupied about 15-17 months after the storm. The standard Mussel Watch Program sample protocol suggests a size range of 50-80 mm to avoid large discrepancies in age for site comparisons and a more robust trend analysis (Lauenstein and Cantillo, 1993a). However, following Hurricane Sandy populations consisted of much smaller mussels (< 50 mm) at many of the sites making it impossible to collect enough in the recommended size range. Therefore, the individual age of a subset of mussels at all sites were evaluated by thin-sectioning of the shells to determine if the mussels collected had survived Hurricane Sandy and if there were regional differences in populations resulting from the storm. Mussel size and age varied by species and collection site (Table 3). Blue mussels were generally smaller (range 23.2 to 63.0 mm) than ribbed mussels (range 28.9 to 86.3 mm). Age ranges were similar with blue mussels ranging from 4 to 13 years and ribbed mussels ranging from 3 to 12 years, indicating that ribbed mussels have higher growth rates than blue mussels. The age and size of the mussels sampled during the current study are consistent with life histories of these species along the Atlantic Coast. Adult ribbed mussels can live for more than 15 years, grow to nearly 10 cm in length and can be found in numbers over 1,500 per m^2 (Coen et al. 1997; Brousseau, 1984). Ribbed mussels have the ability to reattach if dislodged, providing this species with more opportunities to respond to disturbance. The longevity of blue mussels is more dependent on locality and habitat. In the lower intertidal zone, few individuals are likely to survive more than 2-3 years due to intense predation and disturbances, whereas high intertidal zone populations are composed of numerous year classes (Seed, 1969) and have been reported to reach 18-24 years of age (Thiesen, 1973).

Among-site variability in growth rates of ribbed mussels have been noted by other studies and these differences have been attributed to shore level, temperature, salinity, tidal/current exposure, quantity and quality of food sources and stress from contaminants (Seed 1980; Bertness and Grosholz 1985; Franz 1993; Franz and Tanacredi 1993). Typical age-growth relationships for blue mussels are asymptotic, resulting in decreases in absolute and relative growth rates as the mussel ages (Sukhotin and Portner, 2001). In the current study, there was a significant quadratic relationship between length and age for both blue mussels ($r^2=0.19$, $p<0.0001$) and ribbed mussels ($r^2=0.42$, $p=0.0002$), supporting these previously described patterns. Among-site differences in their deviations from the global growth model were also detected for both species (blue mussel: $H=42.34$, $p<0.001$; ribbed mussel: $H=15.62$, $p<0.001$). Specifically, blue mussels collected from the BIBL site had significantly higher deviations from the growth model than all of the other sites (i.e., mussels here were younger than expected based on their size). A similar pattern was observed in ribbed mussels for the HRJB site, which also experienced greater positive deviations from the growth model relative to the other sites. There was no relationship between latitude (site) and blue mussel length ($r^2=0.25$, $p=0.10$) or age ($r^2=0.32$, $p=0.07$), likely because of the narrow spatial scale of this study. Despite some variations in growth rates among sites, primarily limited to two locations, the size class sampled during the previous study allows for regional comparison of contaminant tissue concentrations after Hurricane Sandy.

3.2 Persistent organic pollutants

At each site, trends prior to Hurricane Sandy (1985-2012) for all compound classes listed in Table 2 were compared to new trends after the storm (Table A1). Regionally, historic concentrations pre- and post-Hurricane Sandy were compared for all compound classes to understand the ecological impacts of the storm (Table 2). Although 2 different species were collected throughout the study area, their tissue concentrations are comparable and contaminant profiles and bioaccumulation potential are similar between blue and ribbed mussels. Other studies have noted after 28 days of exposure a steady-state condition was achieved for both species and PCB concentrations and congener profiles were statistically comparable between the species (Bergen et al., 1993; Nelson et al., 1995).

3.2.1 Comparison to pre-hurricane Sandy trends

Concentrations of PCBs, OCPs and PAHs in mussels collected about a year after Hurricane Sandy were compared to long-term site-specific, regional and national trends generated by the Mussel Watch Program (Kimbrough et al., 2008). At the current time, not enough data for PBDEs are available for a trend analysis. For PCBs, 80% of the sites had total PCB concentrations that were below the lowest value reported by the Mussel Watch Program for the same site, and some were an order of magnitude lower in 2014 compared to the available pre-Sandy data (Fig 2). PCBs at 2 sites, Fire Island (LIFI) and Gardiners Bay (LIGB) were detected above the lowest value, but below the site-specific 20 year median values. Overall, the trends observed at each site for total PCBs did not change following Hurricane Sandy. A decreasing trend in total PCBs before and after the storm was observed at 7 of the 10 sites sampled (Table A1). A similar decreasing trend in PCB concentrations nationally has also been reported by the Mussel Watch Program including significant decreases throughout the Northeast over the past 20 years (Kimbrough et al., 2008). No change in PCB concentrations were observed in the Hudson/Raritan Estuary (HRRB), Fire Island Inlet (LIFI) and Gardiners Bay (LIGB). The Hudson/Raritan Estuary remains heavily contaminated with PCBs attributed to previous industrial activity is one of the most contaminated estuaries in the country (Ayers and Rod, 1986) and sediments in the estuary will continue to be a source of PCBs and other contaminants into the future. However, any resuspension of contaminated sediment from the Hudson/Raritan Estuary was not evident in mussels collected a year after the storm (Fig 2).

As stated previously, 24 OCPs were analyzed in the current study but many of these compounds were detected infrequently and significant historic data are not available. For these reasons, total OCP concentrations reported in the current study consisted of the sum of total DDTs, total chlordanes and total dieldrins similar to those OCPs reported by the Mussel Watch Program during their trend analysis (Kimbrough et al., 2008). Concentrations of total OCPs varied by site, however, OCPs at 50% of the sites were below the lowest previously observed value, 30% were below the median reported values and concentrations at 20% of the sites were above the historical median value (Fig 3A). OCP concentrations at Fire Island (LIFI) were close to the maximum reported value and concentrations of total DDTs and chlordanes increased in 2014 by a factor of 2 compared to the 20 years prior to Hurricane Sandy (Table A1). Post Hurricane Sandy total DDT concentrations were less than the site-specific median values at 70% of the sites (30% of the sites were below the lowest recorded value) while DDT concentrations in

Fire Island (LIFI) were greater in 2014 than the highest reported historic value (Fig 3B). Similarly, total chlordane values in 2014 were generally low (80% of the sites were less than the site specific median values), however concentrations of total chlordanes were elevated at 2 sites in Long Island, Fire Island (LIFI) and Gardiners Bay (LIGB) (Fig 3C) while dieldrins (data not shown) were only detected at 2 sites in the New York Bight, Shark River (NYSR) and Long Branch (NYLB). A greater amount of variability was observed in the trend data for OCPs following Hurricane Sandy compared to total PCBs. This variability in OCPs and increasing concentrations at certain sites could suggest local sources. The increase in DDTs and chlordanes after Hurricane Sandy in Long Island (LIFI and LIGB) is unexpected since pre-Sandy concentrations were lower than those reported at other sites in the study area. Even though total concentration increased at LIFI and LIGB in 2014, no change in trends was observed at these sites. Decreasing trends in chlordanes, DDTs and dieldrins were observed in 6, 5 and 3 sites, respectively both before and after Hurricane Sandy (Table A1). At several sites, after Hurricane Sandy a shift to a decreasing trend was observed in samples from LIJI for chlordanes and DDTs and dieldrins at HRRB. Nationally as well as throughout the Northeast, the concentrations of chlordanes, DDTs and dieldrins are declining (Kimbrough et al., 2008), though concentrations at some sites still remain elevated based on inputs from previous industrial and urban sources throughout the region.

PAHs were divided into low molecular weight (LMW) compounds and high molecular weight (HMW) compounds for site and regional comparisons (Table 2). Separation of PAHs by molecular weight helps differentiate between petroleum (LMW) and combustion or pyrogenic (HMW) sources, respectively. None of the sites sampled after Hurricane Sandy had elevated concentrations of low or high molecular weight PAHs compared to the preceding 20 year record.

A year after Hurricane Sandy, total PAHs (sum of LMW and HMW PAHs) at 50% of the sites were below the lowest recorded value (Fig 4A) while 40% of the LMW PAHs and 50% of the HMW PAHs were below the lowest recorded value (Figs 4B and 4C). Based on the low concentrations of LMW PAHs observed in mussel tissue a year after the storm throughout the region, any oil spills that occurred as a result of Hurricane Sandy were not persistent or long-lasting. No change in the total PAH trend was observed in 90% of the locations both before and after Hurricane Sandy (Table A1) which is consistent with previous results (Kimbrough et al., 2008).

Organic contaminant concentrations in mussels were low, but varied by location and compound class and in most cases the overall trend did not change after the storm. Johnson et al. (2009) reported similar results in which organic contaminant levels in oysters following Hurricanes Katrina and Rita were near record lows but comparisons to regional trends were not discussed. However, when comparing multiple storms over a 7 year period, Apeti et al. (2011) reported apparent fluctuations for some contaminants but most were within their normal site-specific concentrations ranges after the storms indicating no apparent shift in concentrations or trends following major coastal storms.

3.2.2 Regional differences

Regional differences in concentrations for PCBs, OCPs, PAHs and PBDEs were observed a year after Hurricane Sandy (Table 4). Total PCBs and total dieldrins were significantly lower after the storm, while total chlordanes and DDTs were not different from the 20 year record. Concentrations of total chlordanes, DDTs and dieldrins have been decreasing at 50-75% of the sites in the Northeast based on previous trend analysis (Kimbrough et al., 2008).

Regionally, LMW PAHs, HMW PAHs and total PAHs were significantly lower a year after Hurricane Sandy compared to the 20 year historic record (Table 4). A similar comparison was conducted following Hurricanes Katrina and Rita where a decreasing regional trend was reported for all organic contaminants except total PAHs and HMW PAHs (Johnson et al., 2009). Following Hurricane Katrina, there were 10 major spills in Louisiana resulting in the release of 8 million gallons of oil throughout the Gulf of Mexico, but concentrations of LMW PAHs were low, similar to the current study, indicating effects from the oil spills were not persistent. However, in the Gulf of Mexico following Hurricane Katrina, pyrogenic PAHs were the dominant form in sediment (Van Metre et al., 2006) and mussels (Johnson et al., 2009) which explains the regional increase in total PAHs after Katrina. An extensive comparison of organic contaminant concentrations after 7 major storms in the Gulf of Mexico also indicated that PCBs, PAHs, HMW PAHs, DDTs and dieldrins tended to decrease while chlordanes and LMW PAHs were unchanged (Apeti et al., 2011). Results from the current study are in general agreement with Apeti et al. (2011) and indicate that despite local and regional differences in sources, the impacts of severe storms could have characteristic and predictive patterns in the distribution of organic contaminant body burdens in mussels.

Because of the limited historic PBDE dataset available for the study area prior to Hurricane Sandy no trend analyses could be conducted. The three most ubiquitous congeners, 47, 99 and 100 were detected in 100% of the samples (Smalling et al., 2015) and were used to compare tissue concentrations throughout the region. Total PBDE concentrations after Hurricane Sandy ranged from 16.5 to 285 ng/g dry weight (Smalling et al. 2015). Unlike the general decrease observed for PAHs, PCBs and OCPs, total PBDE concentrations at all sites after Hurricane Sandy were higher than those observed between 2004 and 2008 (Fig 5). In bluefish sampled in the summer following Hurricane Sandy, PBDE concentrations were significantly lower in the study area compared to previous studies with young of year bluefish (Smalling et al. 2016, this volume). The conflicting results between mussels and bluefish could be a direct result of differences in sampling locations, life history and/or PBDE bioavailability. Along the coast of New Jersey, PBDE concentrations in mussel tissue were only slightly higher than pre-Sandy concentrations. However, total PBDEs in the Hudson/Raritan Estuary (HRRB, HRJB), Long Island Sound (LIFI, LIGB) and Moriches Bay (MBTH) were 5 to 100 times higher in 2014 compared to results reported previously (Fig 5). Regionally, total PBDE concentrations were significantly higher after Hurricane Sandy compared to data generated between 2004 and 2008 (Table 4). Increasing trends in PBDE concentrations have been observed at selected sites nationally between the late 1990s and mid-2000s (Kimbrough et al., 2009). PBDEs are distributed widely in the marine environment and potential sources include atmospheric transport (Strandberg et al., 2001), urban runoff, industrial point sources and sewage outfalls (Litten et al., 2003; de Wit, 2002; Song et al., 2006). PBDEs have the potential to biomagnify, and concentrations in marine food webs have increased substantially since the 1970s (Kimbrough et al., 2009). In the aftermath of Hurricane Sandy, over ten billion gallons of untreated or partially treated wastewater were released into the coastal environment from numerous waste water treatment plants (WWTP) failures (Kenward et al., 2013). Elevated concentrations of PBDEs are found in municipal sewage which is evidence of an increased threat to the environment from land based sources (Song et al., 2006; de Wit, 2002). Many of the mussel sites experiencing significantly higher PBDE concentrations post-Sandy were not in areas impacted by the largest WWTP failures and thus it is difficult to determine based on the limited pre-storm data if the increase is related to WWTP failures or a representation of an increasing trend in total PBDE concentrations regionally. Regardless, PBDE concentrations along the New Jersey and New York coastlines have increased over the last 5 years and continued monitoring is necessary to understand the ecosystem effects of these ubiquitous contaminants on marine biota. The data generated by the current study cannot distinguish between a regional increase and a direct, long-lasting impact from WWTP failures due to Hurricane Sandy and more information is needed to understand changes in PBDE trends in mussel tissues.

3.3 Implications to measure storm-induced impacts

In the current study, mussels were collected throughout the storm-impacted area 15-17 months after Hurricane Sandy and the concentrations measured represent a new contaminant baseline following a significant coastal disturbance. Timing between episodic events such as hurricanes and mussel collection is one factor that will affect the detection of specific storm-induced impacts on contaminant body burdens. For example, if a mussel population was exposed to a plume of contaminated water or sediment as a result of a storm it can take up to two months after the disturbance for the tissue to reestablish equilibrium with its surrounding environment (Bergen et al., 1993; Nelson et al., 1995; Apeti et al., 2011), after which the body burdens returns to steady state. In the current study adult mussel populations in the region also completed a spawning event in spring/summer (Borrero, 1987) following the hurricane which would result in a decrease of hydrophobic contaminants in the mussel tissue. Capuzzo et al. (1989) noted that a decline in PCB congener concentrations in blue mussels in Buzzards Bay was correlated with spawning activity and concentrations fluctuated during the late spring and early summer with a marked decline occurring during the autumn. Another study noted that PCB concentration in mussels tended to increase in the summer, autumn and winter and decreased strongly during spring which was related directly to the spawning of gametes (Hummel et al., 1990). To understand the implications of a storm or other disturbance, tissue samples should be collected within 1-2 months of the disturbance to maximize the potential of recording a short-lived storm induced impact. Significant routine monitoring data are also necessary to tease out the direct effects of a storm in relation to normal spatial and temporal variability in contaminant body burdens.

Regionally, concentrations of PCBs, OCPs and PAHs were significantly lower after the storm; however, for most of the compound classes these concentrations during the single sampling event of 2014 did not change the overall 20 year trend at each site. Johnson et al. (2009) noted that the low organic contaminant concentrations in oysters following Katrina and Rita could be the direct result of a reduction in bioavailability due to increased suspended sediment concentrations in conjunction with a decrease in metabolic activity during the winter. With the exception of PBDEs, results from the current study suggest that the low concentrations in PCBs, PAHs and OCPs and no change in the overall trend observed a year after the storm are more likely a result of normal temporal oscillations in contaminant body burdens which is consistent with results reported by Apeti et al. (2011). A marked increase in PBDE concentrations was observed particularly in locations throughout the Hudson/Raritan Estuary and Long Island Sound. There are no studies to date assessing the impacts of storms on PBDE concentrations and without recent monitoring data it is difficult to conclude whether this increase was related to long-lasting impacts of the storm or the direct result of a steady increase in PBDE concentrations in the coastal environment.

Contaminant body burden concentrations following storm events can be influenced by storm characteristics (storm strength, surge, flooding), contaminant sources and bioavailability, resuspension of bottom sediments, mussel metabolism (contaminant uptake and depuration potential), as well as the timing between the storm and the sample collection. Body burdens in mussels and other bivalves are influenced by contaminant bioavailability (Roesijadi 1996) which is controlled by local sources (natural versus anthropogenic) as well as the strength and path of the storm. Storm-induced transport of contaminants to the coastal environment occurs through resuspension of bottom sediments, flooding and runoff from anthropogenic sources (DiGiacomo et al. 2004; Farris et al. 2007). For a storm to cause a detectable change in contaminant body burden, the event must elicit a disturbance that directly increases the bioavailable concentration of the contaminants (Apeti et al., 2011). Because such an effect may be short lived, mussels must be collected at the appropriate time to maximize the effectiveness of mussels as an indicator of storm induced impacts. Even though direct, short-lived storm induced impacts could not be determined based on the timing of collection, the information generated from the current study provides a new baseline for organic contaminants in mussel tissue and highlights some interesting trends, particularly with PBDEs that could be investigated in future studies.

4. Conclusions

Mussels are an ideal indicator of ecosystem's health because contaminant concentrations in their tissues respond to changes in ambient environmental levels, accumulation occurs with very little metabolic transformation, and nationally, over 20 years of historical data are available for many of the persistent organic pollutants of interest. Mussels have been used worldwide to monitor changes in contaminants in the coastal environment but results from this and other studies suggest that they also have the potential to detect short and long-term impacts of episodic events as well as long-term impacts of incremental (sea level rise and land-use change) changes.

The substantial 20 year record allows for a robust comparison of pre- and post-Sandy mussel contaminant body burdens. Regionally, concentrations of total PCBs, dieldrins, and PAHs (LMW and HMW) decreased, while total PBDE concentrations increased. Overall, the local and regional trends for PCBs, PAHs and OCPs did not change as a result of the hurricane and any decreases observed are potentially associated with normal temporal and spatial variations in body burden concentrations. However, at some locations concentrations were an order of magnitude lower than the pre-Sandy concentrations, indicating that the current study indeed provides a new baseline in organic contaminants after a major coastal storm. A subset of shells from each site was aged by thin-sectioning to determine if there were any site-specific or local differences in populations. There was no relationship between length or age and latitude indicating a similar distribution of ages across the sampled mussels. However, to determine if Hurricane Sandy impacted mussel populations regionally, a larger cross section of available mussels would need to be collected. A combination of tissue analysis, chronology and histology immediately following a major event would be valuable and necessary to establish a link between contaminants and the resulting ecological effects.

Role of the Funding Source

This study was funded through the Disaster Relief Appropriations Act of 2013 (PL 113-2). The managers of the funding sources did not participate in the design of the study, nor the interpretation or writing of the manuscript. All such decisions were solely made by the authors.

Acknowledgments

The authors would like to thank Jeffrey Pessutti and Daniel Wieczorek (NOAA Fisheries Sandy Hook Laboratory, New Jersey) as well as Irene Fisher, Kaitlin Colella (USGS) who all made significant contributions to the sample collection efforts. We thank Peter Straub (Stockton College) for assistance with freeze drying of mussel tissue samples. We also thank Carrie Blakeslee, Jeffrey Cole, Lisa Sumner and Sophie Weaver (USGS) for assistance with mussel thin-section preparation. Any use of trade, firm, or product names is for descriptive purposes only and does not imply endorsement by the U.S. Government.

Appendix A. Supplementary material

Supplementary material associated with this article can be found, in the online version, at PANGAEA

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Figure Captions

Figure 1. Location of mussel sampling sites in the Hurricane Sandy impacted area along the New Jersey and New York coastlines. Ten historic National Oceanic and Atmospheric Administration Mussel Watch sites were re-occupied approximately 1 year after the hurricane.

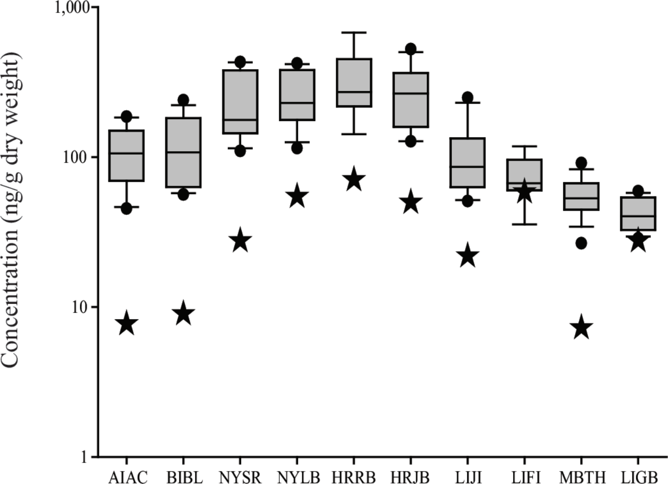
Figure 2. Total polychlorinated biphenyl (PCB) concentrations (ng/g dry weight) measured in mussel tissue after Hurricane Sandy by site. *Stars* indicate concentrations after Hurricane Sandy; *boxes* summarize the historic Mussel Watch data (1985-2012). The top and bottom of each box represent the interquartile range (25th and 75th percentile), the black lines represent the maximum and minimum values and the solid circles are considered outliers. For site location information refer to Table 1 and Fig. 1.

Figure 3. Concentrations (ng/g dry weight) of A) total organochlorine pesticides, B) total DDTs and C) total chlordanes in mussel tissue collected after Hurricane Sandy by site *Stars* indicate concentrations after Hurricane Sandy; *boxes* summarize the historic Mussel Watch data (1985-2012). The top and bottom of each box represent the interquartile range (25th and 75th percentile), the black lines represent the maximum and minimum values and the solid circles are considered outliers. ND, not detected. For site location information refer to Table 1 and Fig. 1.

Figure 4. Concentrations (ng/g dry weight) of A) total polycyclic aromatic hydrocarbons (PAHs), B) low molecular weight PAHs and C) high molecular weight PAHs in mussel tissue collected after Hurricane Sandy by site *Stars* indicate concentrations after Hurricane Sandy; *boxes* summarize the historic Mussel Watch data (1988-2012). The top and bottom of each box represent the interquartile range (25th and 75th percentile), the black lines represent the maximum and minimum values and the solid circles are considered outliers. For site location information refer to Table 1 and Fig. 1.

Figure 5. Total polybrominated diphenyl ether (PBDE) concentrations (ng/g dry weight) in mussel tissue after Hurricane Sandy by site. *Stars* indicate concentrations after Hurricane Sandy and open circles represent historic Mussel Watch data (1996, 2003-2008). Two stars in 2014 are representative of replicate samples analyzed at the site. For site location information refer to Table 1 and Fig. 1.

Base from U.S. Geological Survey 1:24,000 scale digital data,
Universal Transverse Mercator Zone 18N,
North American Datum of 1983 (NAD83)


Concentration (ng/g dry weight)
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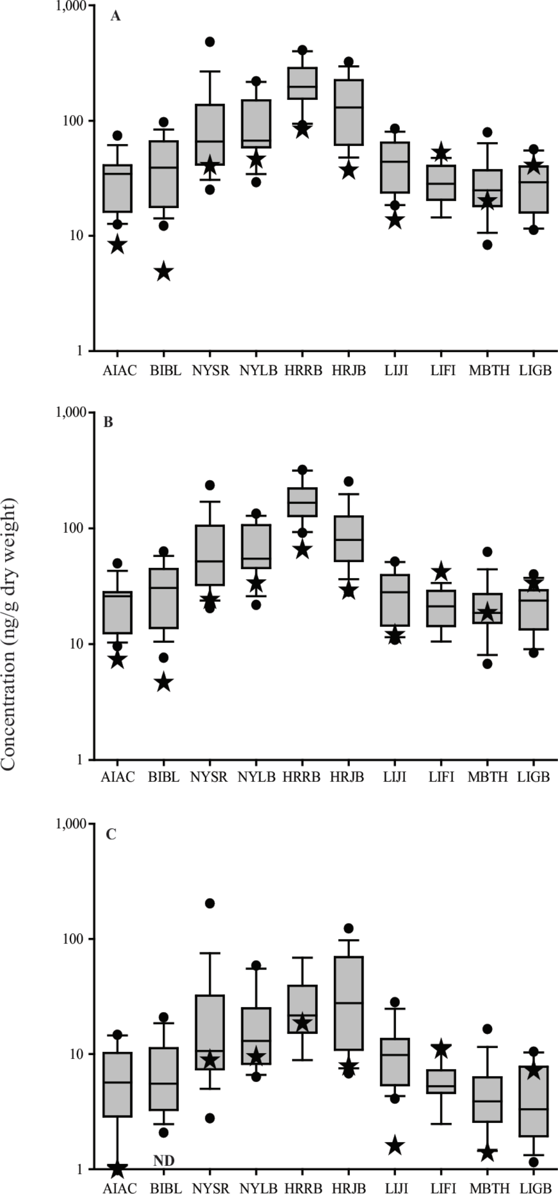
100
10
1
AIAC
BIBL
NYSR
NYLB
HRRB
HRJB
LIJI
LIFI
MBTH
LIGB


Concentration (ng/g dry weight)
A
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1,000
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AIAC
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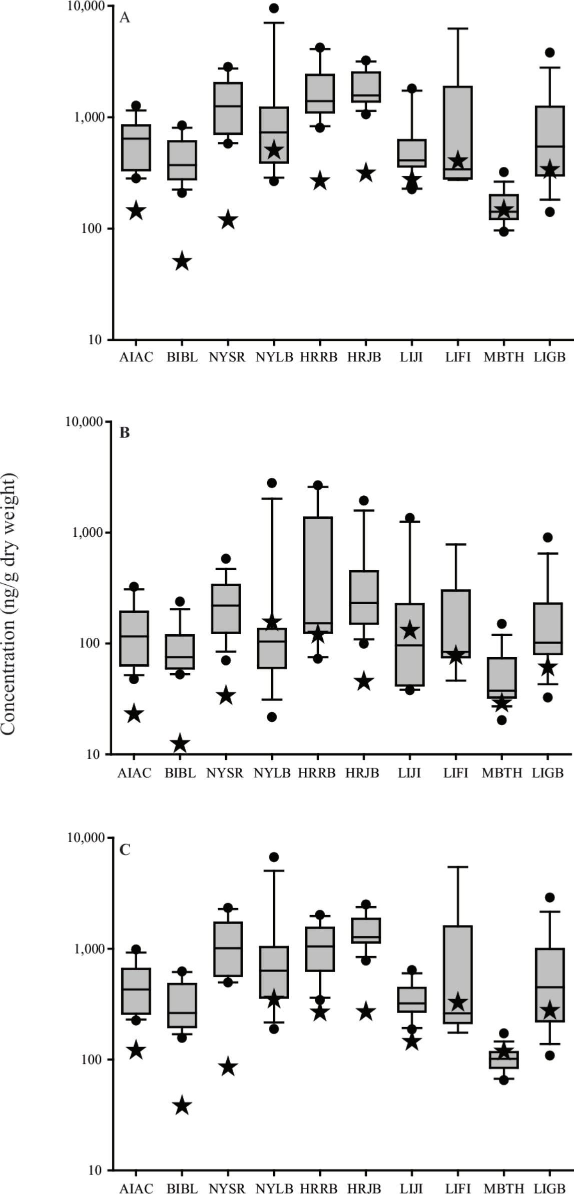
LIFI
MBTH
LIGB
ND


Concentration (ng/g dry weight)
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10,000
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AIAC
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HRJB
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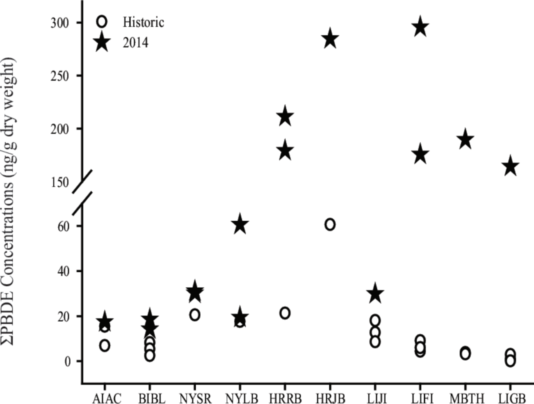
MBTH
LIGB



Historic
2014
ΣPBDE Concentrations (ng/g dry weight)
300
250
200
150
60
40
20
0
AIAC
BIBL
NYSR
NYLB
HRRB
HRJB
LIJI
LIFI
MBTH
LIGB

Table 1. Locations visited after Hurricane Sandy, including the species collected and the years of historic data available for PAHs, PCBs, and OCPs through the National Oceanic and Atmospheric Adminstrations Mussel Watch Program.

Site code	**General location**	**Specific location**	**Lattitude**	**Longitude**	**Sampling date**	**Species sampled**	**Historical data available**[1]
AIAC	Absecon Inlet	Atlantic City	39.3672	-74.4112	01/10/2014	*Mytilus edulis*	1988-2012 (N=14)
BIBL	Barnegat Inlet	Barnegat Light	39.7617	-74.0950	12/19/2013	*Mytilus edulis*	1988-2007 (N=14)
NYSR	New York Bight	Shark River	40.1870	-74.0090	04/10/2014	*Mytilus edulis*	1986-2008[2] (N=17)
NYLB	New York Bight	Long Branch	40.2948	-73.9787	04/10/2014	*Mytilus edulis*	1986-2008[2] (N=15)
HRRB	Hudson/Raritan	Raritan Bay	40.5190	-74.1845	01/15/2014	*Geukensia demissa*	1989-2008 (N=10)
HRJB	Hudson/Raritan	Jamaica Bay	40.5667	-73.8953	01/13/2014	*Mytilus edulis and Geukensia demissa*	1986-2008[2] (N=16)
LIJI	Long Island Sound	Jones Inlet	40.6252	-73.2795	01/16/2014	*Geukensia demissa*	1989-2011 (N=12)
LIFI	Long Island Sound	Fire Island Inlet	40.5955	-73.5867	01/16/2014	*Mytilus edulis*	1986-2011 (N=9)
MBTH	Moriches Bay	Tuthill Point	40.7824	-72.7670	01/17/2014	*Geukensia demissa*	1985-2011(N=18)
LIGB	Long Island	Gardiners Bay	40.9982	-72.1162	01/27/2014	*Mytilus edulis*	1988-2011 (N=13)

PAH, polycyclic aromatic hydrocarbon; PCB, polychlorinated biphenyl; PBDE, polybrominated diphenyl ether; OCP, organochlorine pesticide

[1] http://ccma.nos.noaa.gov/about/coast/nsandt/musselwatch.aspx.

[2] data for PAHs starting in 1988

Table 2. List of persistent organic contaminant classes used for data analysis and historical data comparisons. The list of analytes is similar to those used by the NOAA Mussel Watch Program for trend analysis (Kimbrough et al., 2008)

Compound Class	**Individual congeners/compounds**
Σ PCB (sum of 18 PCB congeners)	PCB8/5, PCB18, PCB28, PCB44, PCB52, PCB66, PCB101/90, PCB105, PCB118, PCB128, PCB138, PCB153/132/168, PCB170/190, PCB180, PCB187, PCB195/208, PCB206, PCB209
Σ PBDEs (sum of 3 PBDE congeners)	PBDE47, PBDE99, PBDE100
Σ DDTs (sum of 6 compounds)	2,4’-DDD, 2,4’-DDE, 2,4’-DDT, 4,4’-DDD, 4,4’-DDE, 4,4’-DDT
Σ Chlordanes (sum of 4 compounds)	Alpha-Chlordane, heptachlor, heptachlor-Epoxide, trans-Nonachlor
Σ Dieldrins (sum of 2 compounds)	Aldrin, dieldrin
Σ OCPs (sum of 12 compounds)	Sum of 6 DDTs, 4 chlordanes and 2 dieldrins
Σ LMW PAH (sum of 7 low molecular weight PAHs with 2 or 3 rings)	Naphthalene, biphenyl, acenaphthene, acenaphthylene, fluorene, phenanthrene, anthracene
Σ HMW PAH (sum of 12 high molecular weight PAHs with 4 or more rings)	Fluoranthene, pyrene, benz[a]anthracene, chrysene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[e]pyrene, benzo[a]pyrene, perylene, dibenz[a,h]anthracene, indeno[1,2,3-cd]pyrene, benzo[ghi]perylene
Σ PAH (sum of 19 PAH compounds)	Sum of 7 LMW PAHs with 2 or 3 rings plus the sum of 12 HMW PAHs with 4 or more rings

PAH, polycyclic aromatic hydrocarbon; PCB, polychlorinated biphenyl; PBDE, polybrominated diphenyl ether; OCP, organochlorine pesticide

Table 3. Mean length (mm) and age (Inumber of annuli) for *Mytilus edulis* (blue mussels) and *Geukensia demissa* (ribbed mussels) collected after Hurricane Sandy. Standard deviation is also included in parentheses.

Species	**Site**	**Mean Length (SD)**	**Annuli (SD)**	**Sample Size**
Mytilus edulis	AIAC	34.91 (3.23)	6.3 (1.1)	20
	BIBL	53.38 (4.01)	8.0 (2.0)	20
	NYSR	44.56 (6.12)	7.8 (2.5)	18
	NYLB	39.61 (5.55)	7.1 (1.7)	20
	HRJB	33.98 (5.09)	6.0 (1.4)	20
	LIFI	32.20 (3.79)	5.3 (1.1)	20
	MTBH	33.47 (2.75)	5.4 (0.8)	20
	LIGB	30.05 (2.63)	5.4 (1.0)	20
Geukensia demissa	HRRB	70.78 (9.8)	7.0 (2.4)	19
	HRJB	48.50 (11.8)	5.1 (2.6)	15
	LIJI	62.51 (10.4)	4.3 (1.6)	19

SD, standard deviation

Table 4. Regional comparison of mussel tissue concentrations before and after Hurricane Sandy using a two sample Wilcoxon Rank Test. Median concentraions of contaminants in mussels (ng/g dry weight) are also reported.

Compound Class	**Z**	**p-value**	**2014 (Median)**	**Historic (Median)**	**Change**
Σ PCB	-4.240	<0.001	27.8	115	Decrease
Σ DDTs	-1.631	0.0514	32.8	24.3	None
Σ Chlordanes	-1.854	0.0566	8.0	7.5	None
Σ Dieldrins	-3.559	<0.001	0.1	1.3	Decrease
Σ PBDEs	3.893	<0.001	102	7.8	Increase
Σ LMW PAH	-2.578	0.005	27.5	72.4	Decrease
Σ HMW PAH	-3.086	0.001	207	470	Decrease
Σ PAH	-3.679	0.001	623	272	Decrease

HMW, high molecular weight; LMW, low molecular weight; PAH, polycyclic aromatic hydrocarbon; PCB, polychlorinated biphenyl; PBDE, polybrominated diphenyl ether; OCP, organochlorine pesticide