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

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ABSTRACT

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 contaminant 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 and have a long-term monitoring network and an ability to mobilize post event.

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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 scales (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 trend 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 & 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

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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 as 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 (*M. edulis*) and ribbed mussels (*G. 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 et al., 1993; Lauenstein & Cantillo, 1993; Lauenstein & Cantillo, 1998). 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 $12 \times 4\text{-cm}$ 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 $1\text{-}\mu\text{m}$ polycrystalline diamond suspension (Allied, Rancho Dominguez, Calif.), followed by a $0.04\text{-}\mu\text{m}$ colloidal silica suspension (Allied, Rancho Dominguez, Calif.). Slides were stained in Mutvei's solution (1% acetic acid, 25% glutaraldehyde mixed with alcian blue) for 50 min at 37°C . Annuli were enumerated by teams of at least three people until consensus was reached using a dissecting microscope at $65\times$. 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 & Dockum, 2013). Analytical methods utilized in this study were similar to those conducted previously by the Mussel Watch Program (Lauenstein & Cantillo, 1993; Lauenstein & 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 h) and reduced under nitrogen gas. Prior to extraction, recovery surrogates DBOFB, Ronnel, PCB 198, $d_8\text{-naphthalene}$, $d_{10}\text{-acenaphthene}$, $d_{12}\text{-benzo[a]pyrene}$ and $d_{12}\text{-pyrene}$, 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, 20% 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 \times 21.20-mm, 100 pore size, $10\text{-}\mu\text{m}$ particle size; Phenomenex, Torrance, California). Prior to lipid removal by HPLC, 1,2,3-trichlorobenzene (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 $0.25\text{ mm ID} \times 60\text{-m}$ capillary column. PBDE congeners were analyzed by using a Restek 1614 $0.25\text{ mm} \times 15\text{-m}$ 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.

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