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Mercury Contamination in Benthic Biota and Sediments within the New York Bight Wind Energy Area

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Abstract: Aquatic ecosystems are showing increasing evidence of contamination by persistent, toxic substances, including metals such as mercury. Mercury (Hg) is truly an unusual element, having no essential biological function. Its unique physical properties have been utilized for various industrial and commercial purposes. This has led to serious exposure to this known neurotoxin. Additionally, the deposition and effluents of mercury in air, water, and soil have impacted food chain dynamics. The potential of bioaccumulation and biomagnification of Hg within aquatic ecosystems can have serious negative implication on ecosystem functions and services. Furthermore, understanding the difference between those pathways can provide a fundamental role in heavy metal cycling within aquatic food webs. The primary objective of this research was to establish a baseline for mercury contamination of benthic biota and sediments in the New York Wind Energy Area (NYWEA), which could be useful to the US Department of Energy for their site assessment and planning and installation of wind farms within the NYWEA. Analysis of sediment samples from 18 sampling sites was conducted to measure total Hg concentration. Station 41 (14.08 µg/kg) and Station B73 (5.51 µg/kg) exhibited the highest total mean Hg concentration whereas Station 27 (1.883 µg/kg), Station 21 (1.821 µg/kg), and Station 33 (1.7496 µg/kg) exhibited the lowest total mean Hg concentration. Analysis of biota from 19 sampling sites within the NYWEA was conducted to assess total Hg concentration. The long-clawed hermit crab (*Pagurus longicarpus*), sand shrimp (*Crangon septemspinosa*), gulf stream flounder (*Citharichthys arctifrons*), dog whelk (*Citharichthys arctifrons*), and rock crab (*Cancer irroratus*) all exhibited significant differences in mean total Hg concentration among sampling sites. While diversity and species richness are considered good indicators of stress of contaminated systems, Hg contaminant loads observed here did not seem to influence community structure or individual species. The results of this study show that Hg contamination in both sediments and biota is present at low levels in the NYWEA, but below US Environmental Protection Agency limits. This suggests that limited Hg contamination in this region is a positive evaluation for the region and food webs in the New York Bight.

MONTCLAIR STATE UNIVERSITY

Mercury contamination in Benthic Biota and Sediments within the New York Bight Wind
Energy Area

by

Jordan F. Gilruth

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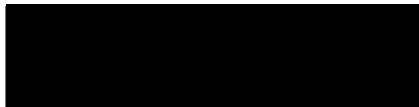
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Montclair, NJ

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Introduction

Background

Heavy metals and other containments enter our waterways from a variety of anthropogenic activities, including combustible coal burning, runoff, mining operations, and waste disposal. These principal vectors of entry are considered the root cause for mercury (Hg) enrichment in the biosphere and hydrosphere in the modern era. Hg is a known toxic element and has been deemed a major global issue. Furthermore, US Food and Drug Administration recommend that fish and shellfish may not contain methyl mercury levels in excess of 1.0 µg/g or ppm (wet wt.) (Yess, 1993). This criterion was established in Section 304 of the Clean Water Act as a guide to human unrestricted consumption of fish (USEPA, 2004). Furthermore, the EPA reported in 2011 that each year in the United States approximately 630,000 newborns are born with unsafe levels of Hg in their blood (USEPA, 2011). This known pollutant is recognized as a neurotoxin with capabilities to induce neurological and cardiovascular disorders, immune deficiencies, and reproductive defects (Taylor and Williamson, 2017). The US Center for Disease Control and Prevention (USCDC) reports that the lethal oral dose in humans is estimated to be around 200 mg/kg of methyl mercury and is analogous to workers being exposed to about 100 mg Hg/m^3 for approximately 30 minutes long (Center for Disease Control and Prevention, 2014). Based on those numbers, the United States Food and Drug Administration (FDA) and the EPA set a limit of $0.3 µg/g^{-1}$ (wet weight) in any fish or shellfish tissue (USEPA, 2009). Due to its negative impacts on living organisms, Hg is deemed a primary global pollutant and is a priority pollutant by international agencies. Furthermore, experts predict that global pool of atmospheric Hg is likely to intensify with

continued industrial development and subsequent emissions throughout the world (Streets et al., 2009). Nevertheless, the underlying foundation for this project stems from a public health consideration concerning the ingesting of contaminated food products resulting from biomagnification and the potential environmental perturbations of Hg.

Mercury Complexes: Inorganic and Organic Mercury

Mercury is a naturally occurring element within the lithosphere, averaging about 80 ppb or less. Additionally, fossil fuels and lignite are considered substantial sources of Hg containing concentrations up to 100 ppb. Hg typically enters the atmosphere as volatile elemental mercury (elemental form or Hg^0 and divalent mercury or $Hg^{(II)}$) from volcanoes, geothermal activities, wild fires, or more commonly through anthropogenic activities. Both forms of Hg occur in the atmosphere, but vary in their physical properties. Inert Hg or Hg^0 is relatively passive with a low solubility (H= 0.11 M atm^{-1} at 20 ˚C [Morel and Hering, 1993]), low reactivity, and low deposition velocity. Ionic Hg or inorganic divalent $Hg^{(II)}$ exists as a variety of complexes and overall this fraction has been termed reactive gaseous mercury (RGHg) (Morel and Hering, 1993). RGHg is highly soluble (H = 2.78^{x}106 M atm^{-1} at 20° C [Schroeder et al., 1991]) and very reactive, this yields $Hg^{(II)}$ to exhibit extremely quick deposition velocity resulting in brief atmospheric residence times. $Hg^{(II)}$ can also bind to other elements (primarily halogens and ligands) forming various compounds while in the atmosphere, the most abundant is $HgCl_2$ and $HgBr_2$ (Ulrich et al., 2001). There are three vectors in RGHg deposition: direct deposition into surface waters, atmospheric water absorption, or atmospheric particles (aerosols) absorption. $Hg^{(II)}$ removal from the atmosphere can occur through

precipitation events known as wet deposition. $Hg^{(II)}$ can also be removed by settling

aerosol particles known as dry deposition. The combination of these different Hg species

can result in the prominent global reach and regional impact of Hg emissions. Once

deposited, volatile organomercury compounds, e.g. dimethylmercury $(CH_3)_2Hg$, can form

through various biochemical pathways and can become bioavailable. Furthermore, the

application of organomercury compounds in laboratories, batteries, fungicides,

bactericides and pharmaceutical products have substantial contributed to environmental

Hg emissions.

However, coal combustion, waste incineration, metal mining, and chlorine-alkali

production primarily contribute to an overwhelming amount of Hg emissions. Schuster at

al. (2002) reported a high-resolution record of total atmospheric Hg deposition (ca. 1720 –

1993) through ice cores collected from, the Upper Fremont Glacier, Wyoming. The ice

core revealed the source of Hg deposition: 52% anthropogenic input, 6% volcanic activity,

and 42% background sources (Schuster et al., 2002). Scientists current estimates report that

around 2190 tons of global Hg emissions are anthropogenic in nature, with 2/3 of the 2190

tons resulting from fossil fuels beginning of the 21st century and concluding that total Hg

deposition rates have grown significantly by 1.5-3 times during the modern era (Mason

and Sheu, 2002).

Mercury Biochemical Pathways

The atmosphere is as a significant reservoir of Hg and contributor to Hg deposition

into natural waters (Hurley et al. 1998; Fitzgerald et al. 2007). Increases in Hg

concentrations in the oceanic surface waters have predominantly resulted from the

exchange of gaseous mercury (Hg^0) (Corbitt et al., 2011). Hg^0 has a relatively long residence time (0.5 to 2 years [Schroeder and Munthe, 1998]) and it can lead to long distance transportation of Hg in the atmosphere resulting in Hg deposition at great distances (as far as 1000km) from its original source (Johansson et al., 2001). Once Hg is present in surface waters, the movement and distribution of mercury within aquatic ecosystems becomes greatly influenced by the lateral and vertical water circulations and the settling of particulate matter to the benthos (Mason et al., 2012). Hg persists in aquatic environments and eventually accumulates in particulate matter and biota. The physical, chemical, and biological dynamics of the benthic environment have been linked to the biochemical pathway of Hg in upper ocean waters. Estuarine and coastal sediments are repositories for high amounts of Hg. Anaerobic sulfate-reducing bacteria (SRB) (*Desulfobulbus propionicus* ND 132 [Aiken et al., 2011]*)* and iron-reducing bacteria (FeRB) are reported to convert Hg to methylmercury (MeHg; CH_3Hg^+) through the acetyl-CoA pathway (Ekstrom et al., 2003); indicating a tight coupling between the presences of Hg methylation and MeHg export out of the bacteria cell wall in an anaerobic condition (Morel et al., 1998). Methylation occurs through the acetyl-CoA pathway, which converts acetate into carbon dioxide. Acetate breaks down into CO and a methyl moiety by the enzyme carbon monoxide dehydrogenase (CODH), and followed by the oxidation of both products, yields CO_2 (Ekstrom et al., 2003). Furthermore, a corrinoid-containing protein known as methyltransferase (MeTr) is responsible during the acetyl-CoA pathway for donating a methyl group to Hg; hence completing Hg metabolism in SRB, specifically in *D. desulfuricans* LS (Ekstrom et al., 2003) (Fig. 1).
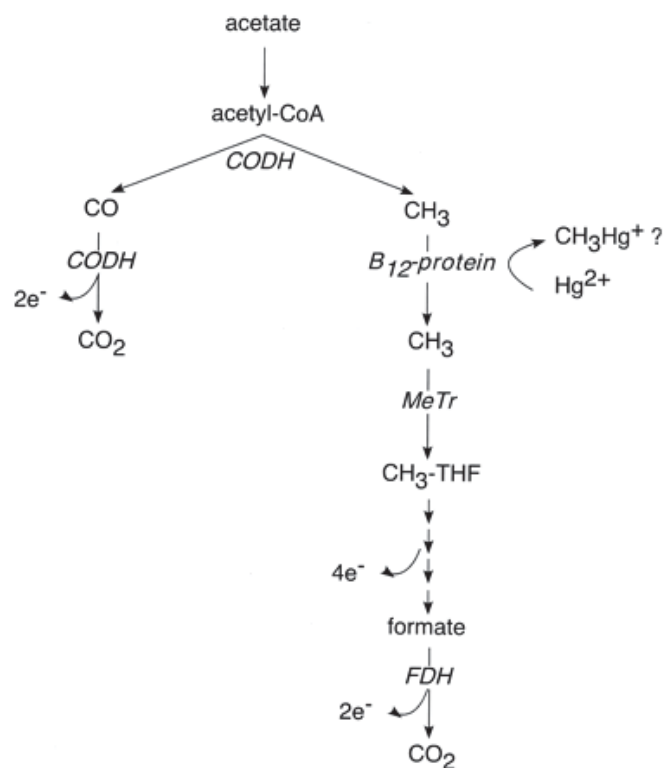
Figure 1. Acetyl-CoA pathway: The biochemical pathway of converting inorganic Hg to organic Hg (Ekstrom et al., 2003).

MeHg is the most toxic form of Hg and it dominates biota in aquatic systems (Ullrich et al., 2001). Therefore, as there is an increase in deposited atmospheric Hg and total MeHg production; there is a coupled relationship with elevated sulfate and iron loading within aquatic ecosystems (Bailey, 2015). Nevertheless, studies have shown that both inorganic (Hg^{II}) and methylated (CH_3Hg & $(CH_3)2Hg$) forms of Hg have large-scale impacts on the aquatic systems (Mason et al., 2012). Coastal ecosystems are considered a major source for elevated MeHg production via microbial sedimentary process (Hammerschmidt et al., 2004). There are variations in MeHg uptake in primary and secondary trophic levels which can be attributed to two major processes. Firstly, MeHg uptake is directly influenced by the potential of methylation based on microbial activity.

Secondly, the variations in Hg speciation, organic matter speciation, sulfur content, and redox potential in porewater can influence sediment fluxes (Chen et al., 2009; Ullrich et al., 2001). Additionally, the degree of bioturbation and hydrodynamics at the sediment water interface influences the availability of MeHg (Chen et al., 2009). Once MeHg is transported into an organism, the chemical residue has several biochemical fates: it may accumulate within the specimen and be stored in the organism's tissues, specifically binding to the thiol moieties of proteins in muscle tissue (Kuwabara et al., 2007); or it may be actively or passively removed from the organism. The potential of the first pathway, accumulation, can lead to the organometal being able to bioaccumulate and biomagnify within an aquatic ecosystem through the active transferring of contaminated food sources from one trophic level to the next. This active accumulation and magnification in Hg concentration levels within organisms can exceed the ambient concentrations in the environment resulting in an imminent threat to freshwater and marine ecosystems and human populations. These biomagnification events occur when each trophic level in the food web take in more Hg than is excreted, causing excess accumulation (Marshall et al., 2016). This leads to elevated levels of Hg, where the accumulation of MeHg comprises up to 85% of accumulated mercury in marine vertebrate and invertebrate tissue (Hsu-Kim et al., 2013). The presence of MeHg within biota, and its toxicity potential within aquatic matrices, is greatly influenced by salinity, temperature, dissolved oxygen, and water hardness (Boening et al., 2000).

It is vital to understand and quantify Hg and MeHg in lower trophic levels to determine the transport potential to upper levels of the food web; ultimately determining the exposure levels for humans (Chen et al., 2009). The potential movement of MeHg

bioaccumulation within lower trophic levels of benthic food webs is largely unknown and a primary focus for this study is to determine Hg levels in lower trophic levels.

Even though the literature on the mechanisms of MeHg uptake and bioavailability is limited for marine food webs, the uptake and bioaccumulation does in fact occur at every level of the food web beginning with phytoplankton (Wiener et al., 2003). MeHg derived from chemical fluctuations in surface sediments in turn effectively transfers MeHg through the bioconcentration of the contaminants in phytoplankton to both the pelagic and benthic food webs. It is thought that MeHg uptake in phytoplankton occurs passively via diffusion across the cell membrane. Once MeHg bioconcentrates, it has been observed to biomagnify across successive trophic levels (Marshall et al., 2016). Although, benthic sediments are the main depository for Hg and MeHg, and the potential source of dissolved (and particulate) MeHg to the water column; the benthic transport of MeHg contamination in biota does not directly relate to sediment MeHg content. Therefore, Hg sediment loads are not an accurate predictor of MeHg bioaccumulation in benthic species (Benoit et al., 2006; Chen et al., 2009). Nevertheless, Hg and MeHg bioaccumulation is greatly influenced by the aqueous supply of MeHg to the base of the pelagic food web, where the transport of MeHg from benthic sediments into the water column is key mechanism MeHg availability for both phytoplankton or particulate uptake and then subsequently the ingestion by invertebrates and fish (Chen et al., 2009). This concept is the byproduct of Hg speciation within benthic sediments and the influence of organic carbon on Hg bioavailability in surface sediments (Chen et al., 2009). Organic matter has an important role in controlling the biogeochemistry of MeHg in sediments and in the water column. Dissolved organic matter (DOM) or dissolved organic carbon (DOC) is highly influential with MeHg

complexation and availability (Ulrich et al., 2001; Benoit et al., 2003). This positive relationship between Hg and DOC is strictly dependent on redox conditions, biological activity, reduction and volatilization of elemental Hg, as well as pH. These relationships appear to be the major factors influencing bioavailability of Hg at the base of food web. DOC, a very fine colloidal suspension, is the decay products of phytoplankton and plays a fundamental role with the formation of MeHg complexation and availability (Ulrich et al., 2001; Benoit et al., 2003). DOC is a vital part of MeHg complexation and availability as it acts as a strong chelating agent for metals, thus affecting their solubility, transport, and toxicity (Nebbioso and Piccolo, 2013). Therefore, DOC is fundamentally involved in the transportation of Hg and stimulating MeHg production in sediments (Haitzer et al., 2002). This relationship contributes to the bioavailability of Hg at the base of food web. Studies have shown that once MeHg is present in a given marine environment, and where exportation rates of ambient methylmercury occur at a rate of 0.2–0.4 $\mu g \cdot m^{-2} \cdot year^{-1}$, then the detection within the sediments, zooplankton, and epifaunal species can take place within a period of 1 month (Harris et al., 2007). Under similar conditions, mercury bioaccumulation can take place within a period of 2 months in most fish species (Harris et al., 2007). Regardless of consumption patterns by pelagic and benthic species their MeHg concentration levels are related to water column particulate MeHg concentrations (Chen et al., 2009).

In invertebrates, the larval and juvenile stages are the most vulnerable time for invertebrate life history to experience mercury exposure. Research has shown that MeHg typically accumulates in soft (edible) tissues and when invertebrates (e.g., *Mytilus edulis* [Linnaeus, 1758]) experience acute and chronic exposure, individuals will endure acute

toxicity which greatly impacts development (Gagnon and Fisher, 1997). Additionally, DeFreitas et al. (1981) found a net assimilation of 70-80% MeHg when shrimp, *Hyalella azteca* (Saussure, 1858), were fed a diet exposed to Hg. The potential of Hg poisoning and MeHg bioaccumulation is a significant environmental problem on a global-scale. Hg concentration is reported to be positively correlated to body size and age when dietary rates outweigh depuration rates of the contaminate (Trudel and Rasmussen, 1997). In addition, prey preference and foraging ecology plays an essential role on MeHg dynamics within an aquatic system. The effects of acute and chronic MeHg exposure have been reported to have serious effects on both pelagic and benthic species including neurological disease, decrease in overall activity, decrease in population recruitment, and death. Understanding the negative effects of both acute and chronic Hg exposure to aquatic ecosystem dynamics is vital. However, a majority of studies on Hg exposure have primarily been focused on bony fish, due to the linkage with humans as a major food resource. Given these circumstances, not only does MeHg poisoning possess a threat to top-predators in pelagic and benthic food webs, but the potential for MeHg poisoning exists for all species within the food web since they are all dynamically connected. This study reveals mercury contamination levels among important foraging species.

Marine Pollution in the NY Bight

The NY Bight (including the coastal boundaries of NY and NJ) is an economically and ecologically important region with many sectors reliant on its ecosystem services. Assessing the monetary impacts of marine pollution and toxicants on the recreational and industrial sectors is extremely important in a region so contingent on

its waterways. Since the 1980s, documenting and investigating the harmful effects of marine pollutants and toxicants has come into the forefront of marine pollution literature in this region. Scientists are continually attempting to evaluate the impacts of marine contaminants on marine life, marine ecosystems, and the public with the goal to achieve environmental restoration, protection and conservation. The National Oceanic & Atmospheric Administration (NOAA), the Bureau of Ocean Energy Management (BOEM), the US Fish & Wildlife Service (USFWS), and the US Navy began to bring those ambitions and goals to reality. In the Climate Action Plan implemented by President Obama in 2013, various federal departments began the initiative to developing domestic, clean energy resources off the coast of several North, Mid, and South Atlantic States in federal waters, referred to as the Wind Energy Area (WEA). In order to determine the environmental impact a WEA would have in a set region, habitat characterization were performed to describe the habitat with respect to the bottom type and topography of the shelf, physical oceanography, and the distribution of infaunal and epifaunal biota to model and evaluate the benthic fisheries habitats at the potential WEA. The main purpose of the habitat characterization is to focus on understanding the spatial variability of the seabed and to assess the impacts on benthic habitats at the potential WEA, due to construction and operations of the farms. Understanding the potential change of topography and having accurate surveying depicting potential consequences on the on the soft sediment benthos habitats and infauna richness is a primary objective for wind farm management.

Although mercury fluxes are limited on the Atlantic continental shelf and the slope sediments, Hammerschmidt et al. (2004) & Hammerschmidt and Fitzgerald (2008)

have reported relatively high benthic fluxes of methylmercury in coastal sediments near New York Harbor, Baltimore Harbor, and Long Island Sound; all possessing a potential negative impact on benthic biota. The presence of mercury within the food web in these waters raises concerns for organismal health, specifically toxicological, ecological, and breeding impacts (Peycheva et al., 2014). Therefore, research is needed to determine the areas of elevated mercury contamination. This research investigates this concern for benthic habitat locations within the New York Wind Energy Area (NYWEA). The primary objective of this work is to establish a baseline for mercury contamination of benthic biota and sediments in the NYWEA, which could be useful to the Department of Energy (DOE) for their site assessment and planning and installation of wind farms within the NYWEA. Moreover, knowing the prolonged occurrence of marine pollution in this region and economic value the NY Bight has towards fisheries, recreation, and tourism; this project can provide valuable information to managers and modelers concerned with Hg bioaccumulation and biomagnification.

Research Objectives

The objectives of this project are as follows:

1. Quantification of Hg levels in epifaunal species and sediments of the NYWEA.

2. Comparisons of Hg concentrations in sediments and biota.

3. Determination of whether sediment Hg loads influenced species diversity and richness.

Materials and Methods

Study Location

A nine-day cruise was conducted from September 21st through September 29th, 2016 on NOAA RV Pisces (PC-16-06): 9 Days-at-Sea (DAS) with designated stations in the New York/New Jersey continental shelf within the U.S. Department of Interior, Bureau of Ocean Energy Management (BOEM) Wind Energy Areas (WEA). The intention of this cruise was to characterize the benthic and demersal habitats in U.S. Department of Interior, BOEM New York Wind Energy Area (NYWEA) which resides in the New York Bight (40.2164° N, 73.2765° W; Fig. 2). The specific location was chosen based on a series of assessments by BOEM, USFWS, and NOAA to determine a region within the Bight that experienced the least amount of anthropogenic activity (e.g. shipping routes, military activities, etc…). Benthic sampling sites were laid out in a grid of approximately 1.5 x 1.5 nautical miles (Fig. 2) in a stratified sampling approach. 38 sampling stations were designated and sampled for sediments and fauna (Fig. 3a).
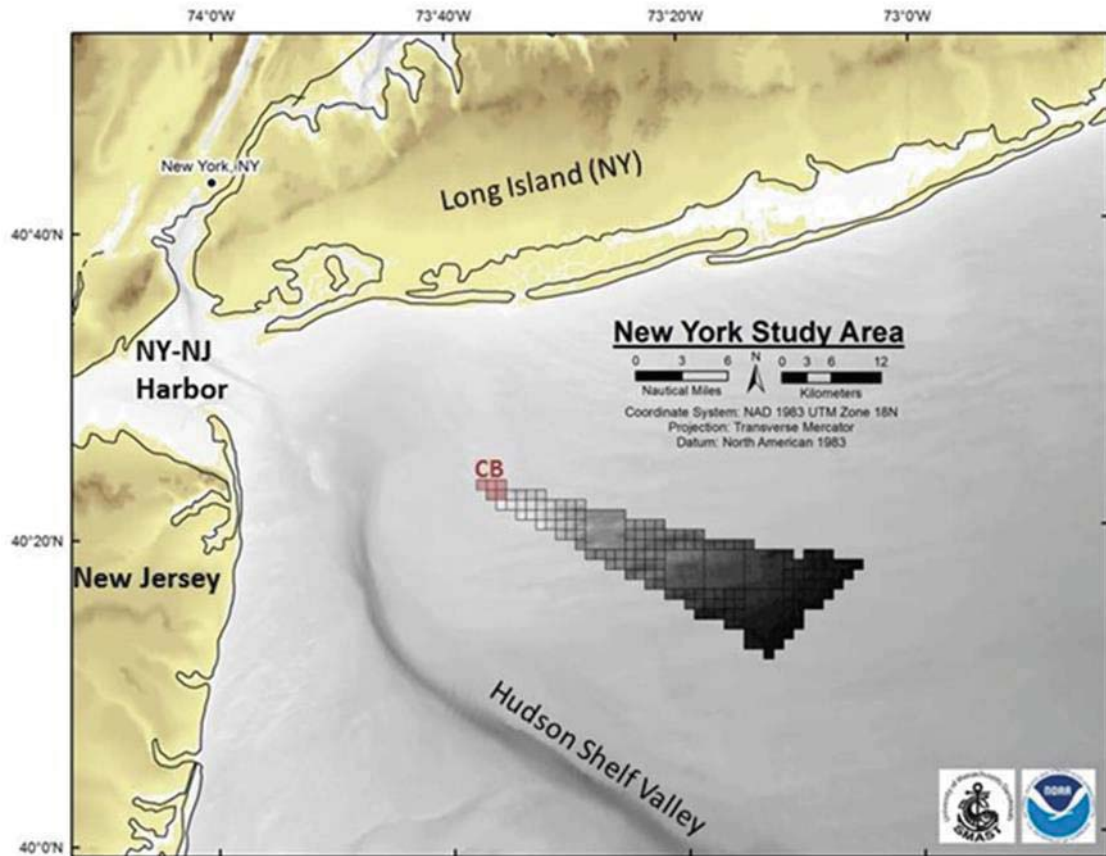
Figure 2. New York Study Area: Cruise starting in Newport, RI to New York Wind Energy Area (NY WEA) high (Guida, 2017). Brown sub-blocks labeled CB represent the Cholera Bank sensitive habitat.

NOAA Pisces Benthic Habitat Cruise

All samples were collected on the NOAA RV Pisces Fisheries Survey Ship by NOAA Scientists under the directions of Chief Scientist Vince Guida. The overlying mission for this cruise was the assessment and characterizing of the existing biotic and abiotic benthic habitat with in the BOEM Atlantic WEAs (Guida, 2016). This assessment was undertaken by the Ecosystem and Aquaculture Division as part of a larger scheme to model and assess fisheries habitats throughout the Greater Atlantic Region. Beam trawls

were used to enumerate benthic epifauna at 38 sampling stations. Benthic biota samples were archived for mercury analyses at 18 of these stations. At the end of the cruise, the samples were returned to NOAA-NMFS-NEFSC, James J. Howard Marine Sciences Laboratory at Sandy Hook, New Jersey, US, and stored at -20°C until analyzed.

Sediment Sampling

Triplicate grab samples were performed using a 10 m² Young-modified Van Veen grab sampler at 38 stations (Fig. 3b). Grab sample replicates were photographed in the sampler and recorded upon retrieval and a 35-mm diameter core was taken for grain size analysis. The remaining sediment sample was then placed into zip lock plastic bags, placed on ice and then archived at -20°C to be processed later for mercury concentration. Of the 38 sampling stations for beam trawl sampling, only at 33 stations had samples that were preserved for mercury analysis (Table 1). However, only 18 sediment sampling sites were included for data analysis to correspond with biota sampling sites. All preserved sediment samples were later processed and characterized by averaging the grain size using Wentworth classification. All grain size classification was processed by NOAA Scientist DeMond Timmons.

Beam Trawl Sampling for Benthic Epifauna Biota

A two-meter beam trawl net was towed at 2 kt. for 15 min. at each of the 38 stations (Fig. 3b) following the sediment sampling and a water quality Conductivity, Temperature, Depth cast. Catches were sorted and identified to the lowest practicable taxon (LPT), which for most samples was species. Each different taxon was then counted and weighed by LPT.

Once total catch was tallied and weighted, specimens from each taxa were preserved by being placed in aluminum foil, placed on ice and then archived at -20ºC to be processed later for total mercury concentration. Of the 38 sampling stations for beam trawl sampling, only samples from 19 stations were preserved for mercury analysis (Table 1). Sampling for Hg contamination did not start until the 12th station (ST21) on 9/25/2016.

Epifauna Biota

The major sampled taxa for mercury analyses were sand shrimp, *Crangon septemspinosa* (Say, 1818), hermit crab, *Pagurus longicarpus* (Say, 1817), Gulf Stream flounder, *Citharichthys arctifrons* (Goode, 1880), Dog Whelk, *Nassarius obsoletus* (Say, 1822), and rock crabs, *Cancer irrotous* (Poeppig, 1836). All of these species play a fundamental role in predator – prey interaction and make up a crucial component to predatory fish's diets. Additionally, several other benthic species were collected and analyzed during research activities including: the Flat-clawed Hermit Crab, *Pagurus pollicaris* (Say 1817), Greedy Dove Snail, *Anachis avara* (Say, 1822), Common Slipper Shell, *Crepidula fornicate* (Linnaeus, 1758), Common Spider Crab, *Libinia emarginata* (Leach, 1815), Asteriid Sea Star, *Asterias forbesi* (Desor, 1848), Longfin Squid, *Loligo pealei (Lesueur, 1812)*, Surf Clam, *Spisula solidissima* (Dillwyn, 1817) and skate eggs (full egg cases only).

Table 1. The coordinates for each station in the NYWEA: All coordinates and depth measurements were logged by NMFS CRUISE PC16-06. "x" indicates that sampling occurred at station. Pink type indicates estimated GPS locations.

| Station Name (ST)	Start Latitude (ºN)	Start Longitude (ºW)	End Latitude (ºN)	End Longitude (ºW)	Depth (m)	Grab	Trawl	CTD
1	40.2285	-73.2323	40.2351	-73.2393	43	X		
5	40.2750	-73.1644	no data	no data	40	X		
7	40.2537	-73.2394	40.2602	-73.2364	40	X		
9	40.3171	-73.2471	40.3251	-73.2437	37	X	x	
10	40.2734	-73.2138	40.2821	-73.2048	40	X		
14	40.2750	-73.1644	no data	no data	41	X		
21	40.2877	-73.3300	40.2815	-73.3179	38	X	x	
27	40.3081	-73.2672	40.3073	-73.262	37	X	x	
29	40.3141	-73.3362	40.3151	-73.2651	34	X	x	x
32	40.3244	-73.3419	40.3162	-73.3372	36	X	x	
33	40.3072	-73.3677	40.3041	-73.3622	36	X	x	
36	40.2848	-73.4002	40.2841	-73.3871	34	X	x	x
38	40.3451	-73.4166	40.3353	-73.4475	31	X		
40	40.3331	-73.4481	40.3155	-73.4590	32	X		
41	40.3596	-73.4409	40.3514	-73.4354	34	X	x	x
43	40.3150	-73.4598	40.3251	-73.4902	33	X	x	x
47	40.3251	-73.4902	40.3366	-73.4807	32	X	x	
49	no data	no data	no data	no data	no data		x	
51	40.3735	-73.5647	40.3752	-73.5425	27	X		
52	40.3752	-73.5915	40.3634	-73.5779	25	X	x	
53	40.3698	-73.5728	40.3844	-73.5975	25	X	x	
54	40.3834	-73.5894	40.3765	-73.5917	25	X	x	x
B73	40.3634	-73.5778	40.3734	-73.564	no data	X	x	
B74	40.3751	-73.5424	40.3772	-73.5326	no data	X		
B75	40.3451	-73.4773	40.3399	-73.4687	32	X	x	x
B76	40.3117	-73.1329	40.2978	-73.4199	32	X	x	
B77	40.3385	-73.4076	40.3324	-73.3969	32	X	x	
B78	40.283	-73.3334	40.2883	-73.3308	38	X		
B79	40.2896	-73.290	40.2978	-73.2929	39	X	x	x
B80	40.2461	-73.2600	40.2530	-73.2395	42	X		
B81	40.2404	-73.2067	40.2301	-73.2312	42	X		
B82	40.2904	-73.1979	40.3165	-73.2129	38	X		
B83	40.2917	-73.1406	no data	no data	42	X		
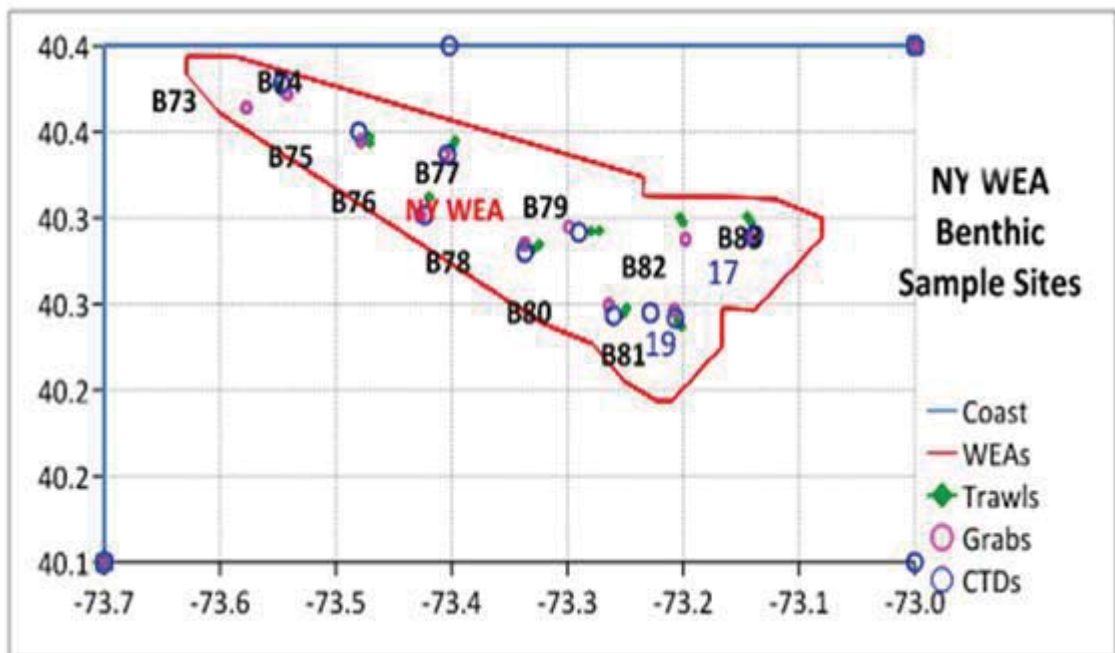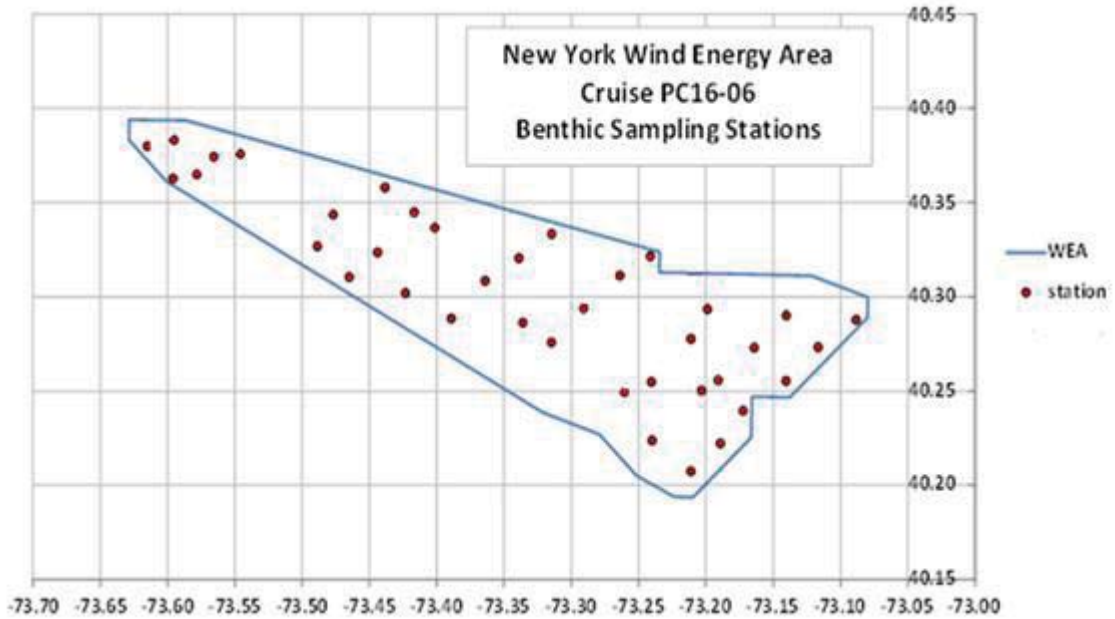
Figure 3. a. Final grid of Benthic Sampling Stations in the New York Wind Energy Area.

b. Labeled grid of sites and sampling locations including trawls, grabs, and CTD.

Mercury Analysis

At the end of the cruise, a Direct Mercury Analyzer (DMA-80) (Milestone. Inc. Shelton, Connecticut, USA) was used to complete analyses of total mercury in the biota and sediment samples. The DMA-80 instrument complies with the US EPA method 7473 (Mercury in solids and solutions by thermal decomposition, amalgamation, and automated combustion atomic absorption spectrophotometry [AC-AAS]). Furthermore, it is compliant with ASTM method D-6722-01 (Total mercury in coal and coal combustion residues) and ASTM method D-7623-10 (Total mercury in crude oil). The DMA-80 tri-cell detection range is 0.01 to 1,500 ng of Hg, with a detection limit of 0.001 ng Hg. The DMA-80 was calibrated using certified reference material (CRMs) of known Hg concentrations and included solid standards (DORM-4; dogfish muscle [0.412 ± 0.036 ppm], PACS-3; marine sediment [2.98 ± 0.36 ppm], MESS-4; marine sediment [0.08 ± 0.06 ppm]) prepared by the National Research Council Canada, Institute of Environmental Chemistry (Ottawa, Canada). For additional quality control, blanks of the catalyst tube and the amalgamator were performed at the beginning and ending of sampling. Moreover, further conditioning of the catalyst tube and the amalgamator was carried out including blanks with nickel boats, flour blanks (0.300 g of flour and 50 uL of DI water), and blanks without a nickel boat to ultimately assess instrument accuracy and potential drift. Measurement results yielded a blank total mercury of < 0.003 ng. Calibration curves were linear (mean $R^2 = 0.99$; range $R^2 = 0.998$-0.99; $p < 0.0001$), and the recovery of the independently CRM samples analyzed ranged from 74.7% - 125.6% (mean = 103%).

All samples were removed from the -20°C freezer to thaw. Once fully thawed, samples less than 0.30g were weighed on a nickel sample boat and loaded on the auto-

sampler tray. If a sample weight exceeded the 0.30 g threshold, subsampling of the sample took place. Samples were then inserted into the DMA-80 for analysis and total Hg concentration was measured in mg/kg (ppm).

Data analysis

Species and sediment differences in mean total Hg concentration were analyzed among all taxa and sediment samples using a one-way analysis of variance (ANOVA) models with site as the independent variable and Hg concentration as the dependent variable. Post hoc separation of mean differences in Hg concentration across 5 different taxa and 18 sediment sampling stations were contrasted using the Ryan-Einot-Gabriel-Welsch Multiple Range Test (Ryan's Q). Note there is one extra biota sampling station (19) than sediment sampling station in the data analysis.

Results

Mercury concentrations in sediments

All sampled sites were under the EPA's set threshold of <0.29 µg/g [ppm (wet wt.)] or 229.0 µg/kg [ppb (wet et.) but, there was a detection of Hg in all samples. Sediment mean total Hg concentrations varied significantly among sediment samples within the 18 sampling stations (ANOVA; $F_{17,36}$=16.58; p=<.0001). ST 41 had the highest mean Hg concentration and was significantly greater than all other sites. The remaining sites showed very low relative concentrations with only some variations among stations (Fig. 4).
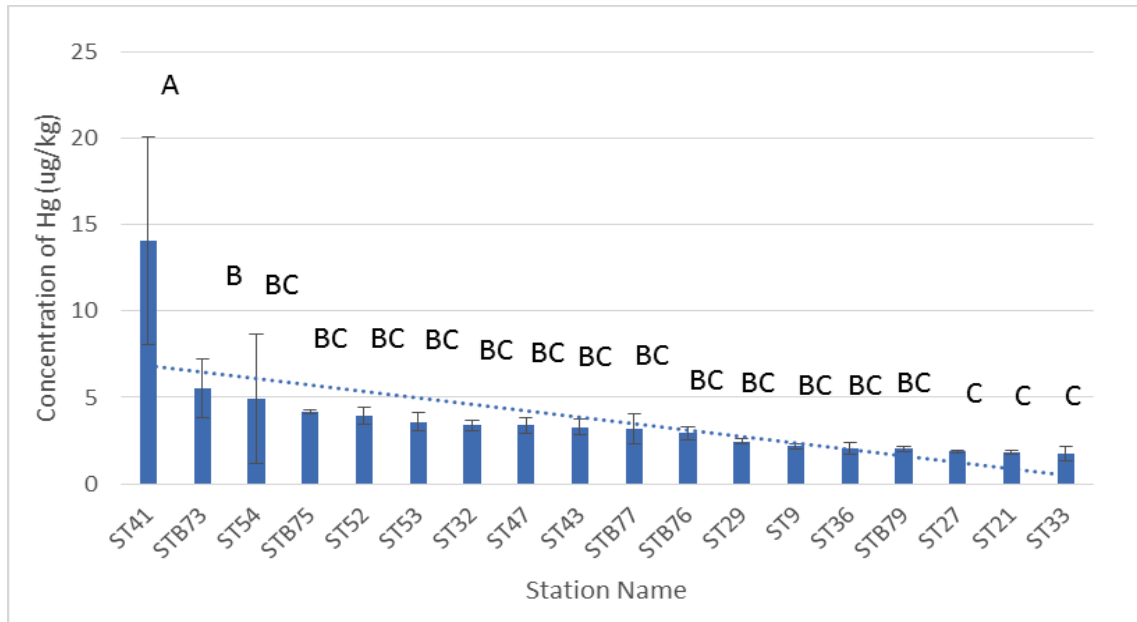
Figure 4. Average mercury concentrations for sediment samples. 54 sediment samples were analyzed for total mercury concentration (µg/kg). Means with the same letter are not significantly different.

Epifaunal Collections

Beam trawl sampling resulted in the collection of 569 individuals of 13 different taxa used in the Hg analyses (Table S1). Catches in the NYWEA did include other recognizable taxa (24 fishes, 35 invertebrates), in which the common sand dollar (*Echinarachnius parma*) was the overwhelming dominant in terms of both numbers and weight representing 99% of the catch. However, the other prominent taxa caught and analyzed included the sand shrimp (*C. septemspinosa:* 75% of the non-sand dollar catch), rock crabs (*C. irroratus*, almost exclusively newly-settled juveniles), hermit crabs (*P. longicarpus*) and Gulf Stream flounder (*C. arctifrons*) which comprised of the other 1%. In general, species richness was relatively low (13 different taxa, Table S1), but enough individuals were collected from among these taxa to conduct Hg concentration analyses.

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Table 2. The scientific name, common name, mean Hg, and range for each analyzed taxon.

Scientific name	Common Name	Mean Hg (μg/kg)	Range (μg/kg)
Pagurus longicarpus	Long-clawed Hermit Crab	24.21= ± 7.11	17.074 - 40.018
Crangon septemspinosa	Sand Shrimp	20.76± 11.27	0.12 - 45.323
Citharichthys arctifrons	Gulf Stream Flounder	17.21± 10.91	1.06 - 37.517
Nassarius obsoletus	Dog Whelk Snail	15.28 ± 8.83	0.018 - 37.824
Cancer irroratus	Rock Crab	12.05± 6.57	1.4^{-5} - 23.592

Mercury concentrations in epifaunal species

Mean total Hg concentrations varied significantly among epifaunal species within the 19 sampling stations. *Pagurus longicarpus* had the highest mean Hg concentration followed by *Crangon septemspinosa*, *Citharichthys arctifrons*, *Nassarius obsoletus*, and *Cancer irroratus* (Table 2).

Pagurus longicarpus

Hg concentration amongst the different populations ranged from 17.074 μg/kg to 40.018 μg/kg (Table 2). ANOVA results of *Pagurus longicarpus* showed significant differences among populations (ANOVA; $F_{8,66}$ =23.66; p=<0.0001). Results showed that ST 43 had significantly greater concentrations compared to all other sites (Fig. 5).
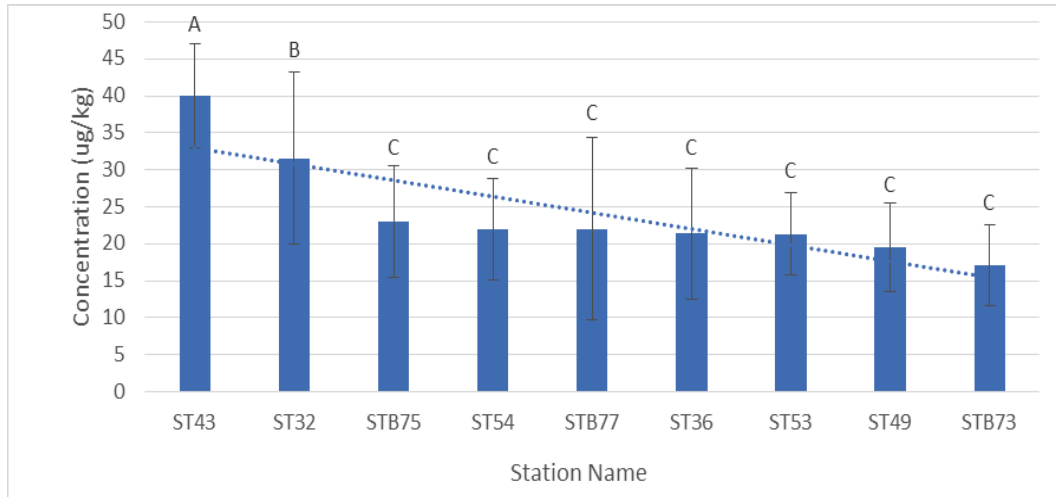
Figure 5. Average mercury concentration for *Pagurus longicarpus*. 75 Long-claw hermit crab samples were analyzed for total mercury concentration (µg/kg). Means with the same letter are not significantly different.

Crangon septemspinosa

Hg concentration amongst the different populations ranged from 0.120 µg/kg to 45.323 µg/kg (Table 2). ANOVA was performed on the average mercury concentration of *Crangon septemspinosa* with significant differences among stations ($F_{13,121}$ = 23.66; p = <0.0001). ST 53 had significantly higher concentrations compared to other sites (Fig. 6). The next 11 stations had relatively similar concentration of Hg, but stations ST29 and ST9 had significantly lower Hg concentrations, almost below the level of detection (Fig. 6).
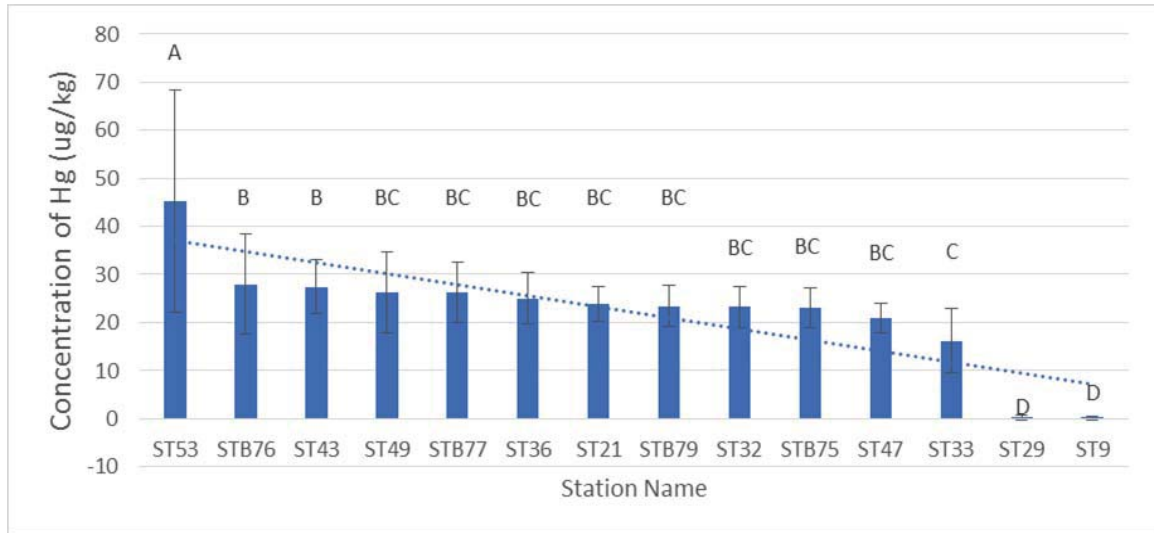
Figure 6. Average mercury concentration for *Crangon septemspinosa*. 134 Sand Shrimp samples were analyzed for total mercury concentration (µg/kg). Means with the same letter are not significantly different.

Citharichthys arctifrons

Hg concentration amongst the different populations ranged from 1.06 µg/kg - 37.517 µg/kg (Table 2). ANOVA was performed on the average mercury concentration of *Citharichtys arctifrons* with significant differences between stations (ANOVA; $F_{11,88}$ =12.33; p = <0.0001). ST 43 and St 47 had significantly higher concentrations compared to other sites (Fig. 7). The next 9 stations had relatively similar concentration of Hg, but station ST29 had significantly lower Hg concentrations, almost below the level of detection (Fig. 7).

Figure 7. Average mercury concentration for *Citharichtys arctifrons*: 100 Gulf Stream Flounder samples were analyzed for total mercury concentration (μg/kg). Means with the same letter are not significantly different.

Nassarius obsoletus

Hg concentration amongst the different populations ranged from 0.018 μg/kg to 37.675 μg/kg (Table 2). ANOVA was performed on the average mercury concentration of with significant differences between stations (ANOVA; $F_{11,74} = 12.33$; $p = <0.0001$). ST 43 and St 47 had significantly higher concentrations compared to other sites (Fig. 8). The next 9 stations had relatively similar concentration of Hg, but station ST29 had significantly lower Hg concentrations, almost below the level of detection (Fig. 8).
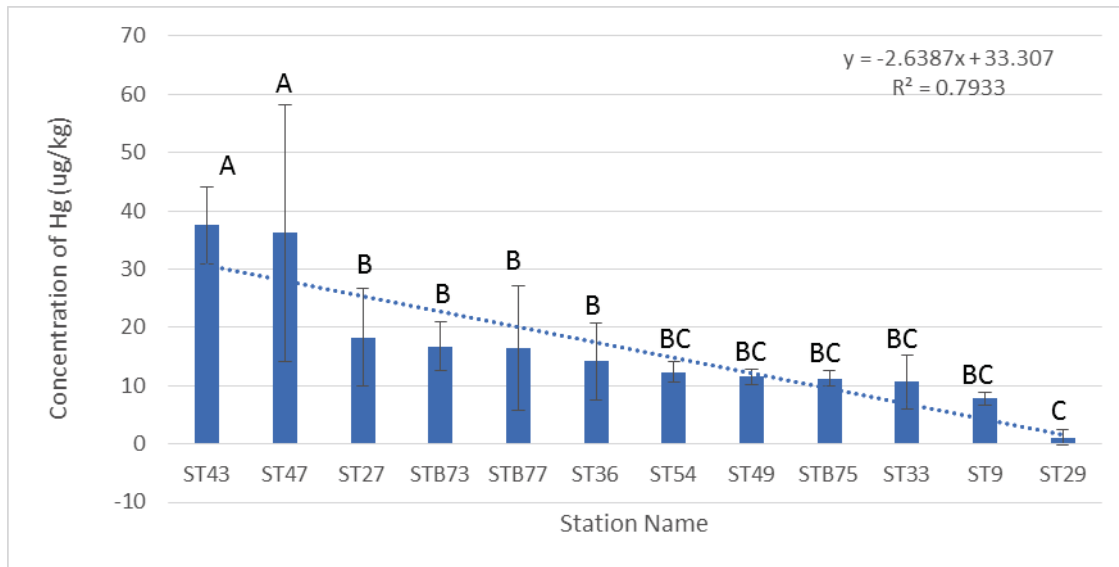
Figure 8. Average mercury concentration for *Nassarius obsoletus*: 86 Dog Whelk Snail samples were analyzed for total mercury concentration (μg/kg). Means with the same letter.

Cancer irroratus

Hg concentration amongst the different populations ranged from 1.4^{-5} μg/kg to 23.592 μg/kg (Table 2). ANOVA was performed on the average mercury concentration of with significant differences between stations (ANOVA; $F_{15,114}$ =22.17; p = <0.0001). ST 43 had significantly higher concentrations compared to other sites (Fig. 9). The next 6 stations had relatively similar concentration of Hg, but stations ST 29, and ST09 had significantly lower Hg concentrations, almost below the level of detection (Fig. 9)
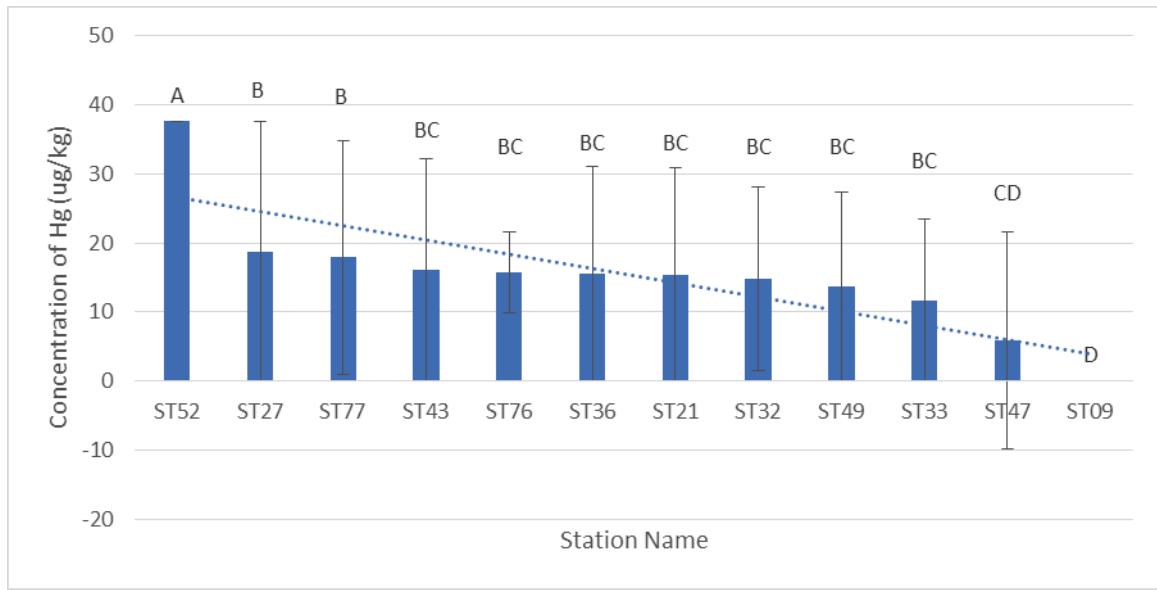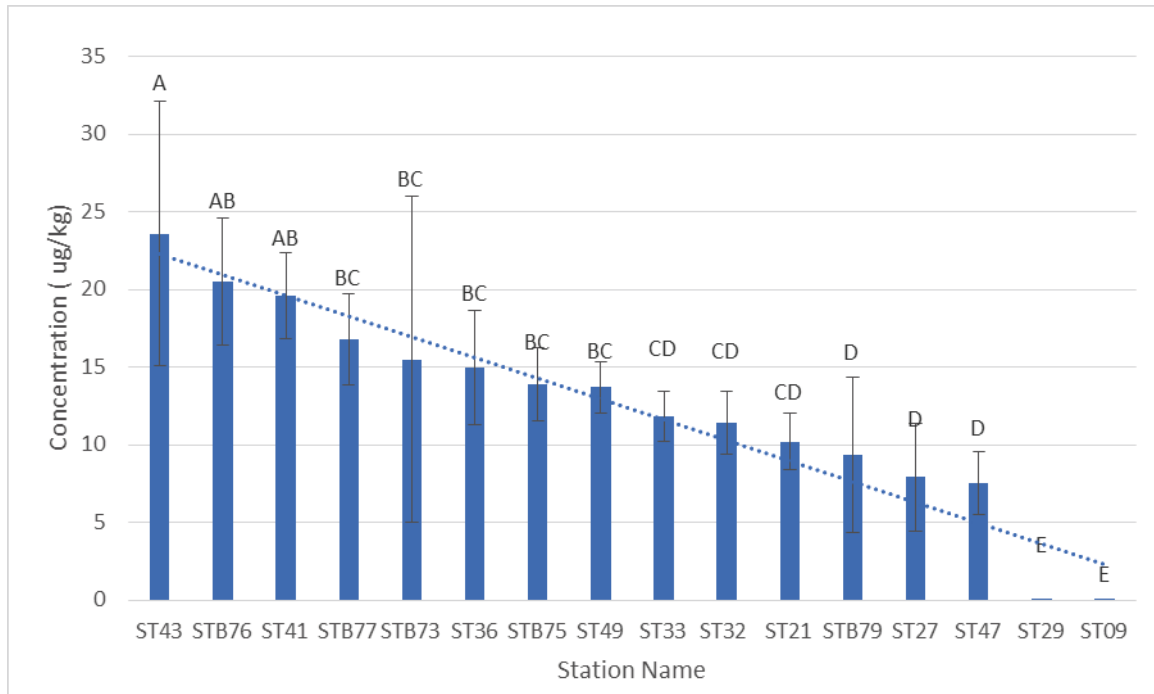
Figure 9. Average mercury concentrations for *Cancer irroratus*: 130 Rock crab samples were analyzed for total mercury concentration (µg/kg). Means with the same letter are not significantly different.

Discussion

An important question that arises when dealing with trace metal contamination is what are the toxicological and ecological impacts to food chain dynamics? More specifically, what is the potential of bioaccumulation and biomagnification at the base of the food web? It is evident that society is reliant on aquatic ecosystems for its natural ecosystem services. Thus, it is imperative to understand and compare abiotic and biotic Hg levels in various aquatic habitats. When examining the speciation of Hg and its potential perturbations on aquatic system dynamics, MeHg is primarily the main speciation of Hg that promotes Hg poisoning in bony and cartilaginous fish and other aquatic biota (Chakraborty et al., 2017). However, little is known about the impacts of Hg poisoning and

bioaccumulation at the foundation of the benthic food web. These species play a fundamental role within benthic ecosystems. More specifically, these species support crucial trophic interactions serving as key prey species and play a vital role on importing and exporting nutrients and energy at the base of the food web (Barkai and McQuaid, 1988). Therefore, quantifying Hg levels in such a traditionally heavily impacted marine ecosystem, such as the New York Bight, can provide further insight on Hg cycling within benthic habitats and the interaction between benthic sediments and biota. This data can provide important information for Federal and State agencies that provide the public with dietary advice and public health recommendation for fish consumption. The observed spatial-temporal differences in mean Hg contamination in benthic food webs can be attributed to geographic variability and historical changes in contaminant inputs to coastal habitats (Benoit et al., 2003). Moreover, geochemical and physiochemical processes in a marine coastal habitat can vary over relatively small spatial and temporal scales, thus directly affecting Hg mobilization and its eventual incorporation and transfer trophic pathways (Chen et al., 2009).

Within the NYWEA, my results showed that overall levels of Hg were relatively low and below the US-EPA threshold levels (Table 2). This may be a result of low levels of organic matter deposition in sediments heavily dominated by sand throughout the entire NYWEA (Supplemental Figure S1). Moreau et al. (2015) reported that sedimentary organic matter (POM) levels are coupled with Hg concentrations. Their research concluded that there is a strong association with POM & DOM in the conversion of ReHg to MeHg in both the natural environment and laboratory. Furthermore, Moreau et al. (2015) state that the degree of POM hydrophobicity (mainly imparted from aromatic functional

groups) strongly influences the uptake and/or methylation of mercury, hence impacting the levels of MeHg in a certain environment.

During the NYWEA sampling, 13 different taxa were identified, but only the five most abundant species were analyzed for Hg. Hg concentration varied among the five taxa, with averages ranging from 12.05 µg/kg to 24.21 µg/kg (Table 2). Each taxon can be a critical link in benthic foods webs and have the potential to bioaccumulate and biomagnify Hg in their tissues through the consumption of contaminated organic matter particulate or food substrate. Although there is an observed Hg contamination in each taxon, with *P. longicarpus* having the highest observed mean Hg concentration at 24.21 µg/kg (ppb), it is still substantially lower than the recommended US-EPA criterion of <300 µg/kg or ppb (wet wt.). However, the potential for biomagnification through consumption of individuals poses a potential risk within the benthic food web and pelagic food web since MeHg concentration levels are related to water column particulate MeHg concentrations (Chen et al., 2009).

The observed results for all taxon and sediments having a low level of mean Hg concentration does not align with the expected results, that being the NY Bight has been historically plagued with modern marine pollutants and toxicants. Hammerschmidt et al. (2004) reported that the NY Bight, specifically the NY/NJ harbor, possess larger amounts of allochthonous organic matter (sewage and terrestrial) which was observed to directly influence seasonal methylation rates. However, observed results for Hg contamination in benthic biota and sediments at the NYWEA did not exhibit high levels of Hg contamination. Nevertheless, benthic biota and species did exhibit contamination and the reasoning behind the contamination directly falls on the deposition and influence of organic

matter on increasing the bioavailability of Hg (Chen et al., 2009). These results infer that if there is an Hg contaminate load of any level, regardless of spatial and temporal patterns, that MeHg possess the potential to first bioaccumulate in organisms within the base of the food web, and then biomagnify to higher trophic levels through trophic interactions.

The top three contaminated sites for each taxon were compared to sediment samples to determine if there were any patterns and trends present. Station 43 appeared to be a hot spot for Hg contamination. Gulf Stream flounder (37.517 µg/kg) (Fig. 7), sand shrimp (27.432 µg/kg) (Fig. 6), rock crab (23.592 µg/kg) (Fig. 9) and long-clawed hermit crab (40.018 µg/kg) (Fig 5.) were all observed to have higher levels of mean Hg contamination. However, the sediment at this site had a relatively low concentration (3.289 µg/kg) (Fig. 4). Other hot spots appeared at Station 27 and Station B76. Dog whelk (18.824 µg/kg) (Fig. 8) and Gulf Stream flounder (18.290 µg/kg) (Fig. 7) at Station 27 both exhibited high levels of Hg, whereas Station 27 sediment sample exhibited low level of Hg contamination (1.821 µg/kg) (Fig. 4). Similarly, sand shrimp (20.514 µg/kg) (Fig. 6) and rock crab (20.514 µg/kg) (Fig. 9) at Station B76 both exhibited high levels of Hg, but in contrast, Station B76 sediment exhibited a low level of Hg contamination (2.922 µg/kg) (Fig. 4). These results confirm the findings by Chen et al. (2009) that although benthic sediments are the main depository for Hg and MeHg, and a potential source of dissolved (and particulate) MeHg to the water column; sediment loads can't be used as a predictor for MeHg bioaccumulation in benthic species (Chen et al., 2009).

Species diversity and species richness are two key indicators of stress in contaminated systems. A marked decrease in species diversity is considered an indicator of contaminate presence within a set location (Clements and Newman, 2003; Gagneten and

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Paggi, 2009). In this study, diversity varied between 0 and 0.86 at each station and individual abundance varied between 1 and 61 individuals (S2). However, no conclusive evidence was observed to infer that sediment contaminate loads influenced community dynamics inter-station dynamics. Yet there is one station that did exhibit an elevated contaminated load that may have influenced the community abundance at that site. Station 41 exhibited the highest mean Hg contamination among all analyzed stations in the NYWEA (Fig. 4). Additionally, Station 41 exhibited low individual abundance and species diversity with only 7 sampled individuals and 1 sampled taxon (rock crab). Station 41 mean Hg concentration for rock crab was observed to be 19.578 μg/kg, which is the third highest mean Hg load among this taxon (Fig. 9). A logical causation for this observation would be that this elevated Hg load at Station 41 could possibly inhibit species abundance or presence at this site causing low levels of diversity and abundance. Furthermore, the individuals that were present at this station did exhibited higher Hg contamination for rock crabs, which could infer that the remaining individuals may have the necessary mechanism to resist contaminate conditions or these individuals were not at this site for an extended period of time, since rock crabs are reported to be a highly active and mobile species (Gosner, 2014). However, since the Hg load at this site does not exceed the EPA's recommended criterion for Hg loads at <0.29 μg/g (ppm) or 229 μg/kg (ppb); pollutant load may not be playing a large role in these communities. Additionally, observed background levels for Hg contamination in sediment samples are observed to vary between from 10.0 to 240 μg/kg or ppb of Hg (Syres et al.,1972). All levels of Hg contamination in sediment samples fell within this range of background levels, so in conclusion sediment samples within this site essentially possess no levels of Hg contamination.

Station 52 also exhibited low levels of species diversity and richness with only one samples individual and taxon (Dog whelk). Station 52 Hg concentration was 37.67 µg/kg however, Hg contamination is only 3.92 µg/kg. At Station 52, Hg loading was meniscal and in theory should not influence species richness. Nevertheless, the prediction that higher concentrations of Hg would prevent the establishment and development of benthic populations was not observed. On the contrary, the concentration of Hg had little to no effect on benthic populations.

Benthic biota and benthic sediments have a dynamic relationship. MeHg genesis in benthic sediments is directly controlled by many factors including concentrations of total Hg, organic matter speciation and input, sulfide loading, and redox potentials. However, benthic sediment MeHg does not leave species vulnerable to acute and chronic exposure to MeHg, whereas MeHg concentration levels in benthic biota are a result of the exposure and uptake of water column particulate MeHg (Chen et al., 2009). The predominant sources of MeHg to the water column pathway of MeHg is poorly understood but yet it is the basis of bioaccumulation, where accumulation occurs at higher trophic levels due to elevated levels of MeHg consumption and low levels of MeHg excretion.

In summary, there was low levels total Hg contamination in both benthic biota and sediments. Due to benthic sediment substrate, predominately being sand; Hg contamination levels were very low, well within the background range (10 µg/kg – 240 µg/kg) for benthic sediments. Additionally, each of the five analyzed taxon, long-clawed hermit crab, sand shrimp, dog whelk snail, gulf stream flounder, and rock crab; exhibited low levels of Hg contamination. This could be a result of minimal levels of acute MeHg exposure in a location that has low levels of allochthonous organic matter deposition.

Conclusion

The results of this study show that there is low level presence of Hg contamination in both benthic sediment and biota in the NYWEA. These results indicate that the benthic community is not stressed by Hg contamination. Furthermore, Hg contamination did not appear to have an effect on species richness and diversity at these sites, but both were extremely low and may reflect the sampling protocol. However, Station 41 which exhibited a high Hg load, did in fact favor fewer tolerant species (rock crab) and a showed a decrease in total number of individuals. My results confirm Chen et al. (2009) findings in relation to the counterintuitive relationship of MeHg genesis in surface sediments and Hg contamination levels of biota. Considering the results obtained in this study, it can be concluded that contamination of this system is minimal, but present. Any Hg contaminate load can have a gradual impact on benthic biota and a cascading effect through trophic interactions with the potential for biomagnification. Biological and chemical monitoring at these sites with continual sampling can provide a clearer assessment of the pollutants and contaminates in the NYWEA. Regardless, the results from this study demonstrating Hg presence in sediments and biota suggests that Hg in the NYWEA should still be considered a potential threat to the NY Bight ecosystems and the human populations that are associated with it.

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Supplementary Material

See supplementary material for the table on Community dynamics and diversity index

and figures on predicted average sediment type (Wentworth Classification) of sediments

and surficial sediments (presence-absence) observations.

Supplementary Materials:

Table S1. RC Rock Crab, SS sand shrimp, LHC long-clawed hermit crab, FHC flat-clawed hermit crab, CSC common spider crab, DW dog whelk, CSS common slipper shell, LFS long-finned squid, SS Asteriid sea star, SC surf clam, SE skate egg, GSF gulf stream flounder, and GDS greedy dove snail. Any cell that does not have a number in it (-), no samples were observed within the station. Simpson's Diversity Index is indicated by SDI. Total mean Hg concentration is in µg/kg or ppb.

Species	C. irroratus	C. septemspinosa	P. longicarpus	P. pollicaris	L. emarginata	N. obsoletus	C. fornicata	L. pealei	A. forbesi	S. solidissima	Rajidae	C. arctifrons	A. avara	SDI
Common Name	RC	SS	LHC	FHC	CSC	DW	CSS	LFS	SS	SC	SE	GSF	GDS	-
49	8	10	9	-	-	12	-	-	-	-	3	10	9	0.86
B75	9	10	10	-	-	-	-	-	--	-	-	10	-	0.77
52	-	-	-	-	-	1	-	-	-	-	-	-	-	0.00
54	-	-	10	3	2	-	3	-	-	-	-	10	9	0.80
32	-	10	2	-	-	-	-	2	-	-	-	-	-	0.48
33	9	10		-	-	5	-	-	1	-	-	4	-	0.76
36	10	10	15	-	-	10	-	1	-	-	-	12	-	0.81
21	10	10	-	-	-	10	-	-	-	-	-	-	-	0.69
41	7	-	-	-	-	-	-	-	-	-	-	-	-	0.00
43	10	10	9	-	-	10	-	-	-	-	-	5	-	0.81
9	11	11	-	-	-	7	-	-	-	-	-	11	-	0.76
29	5	11	-	-	-	-	-	-	-	-	-	6	-	0.65
B73	4	-	10	-	-	-	-	-	-	7	-	9	-	0.75
B77	10	10	2	-	-	10	-	4	1	-	3	15	-	0.83
53	-	6	8	-	-	-	-	-	-	-	-	-	-	0.53
47	5	10	-	-	-	6	-	-	-	-	-	5	-	0.75
B79	12	10	-	-	-	-	-	-	-	-	-	-	-	0.52
27	10	-	-	-	-	6	-	-	-	-	1	3	-	0.67
B76	5	6	-	-	-	6	-	-	-	-	-	3	-	0.77
TOTAL	125	134	75	3	2	83	3	7	2	7	7	103	18	-
Mean Hg	12.30	22.07	24.21	13.64	8.26	15.07	22.03	14.01	28.33	13.33	29.85	17.22	21.90	-
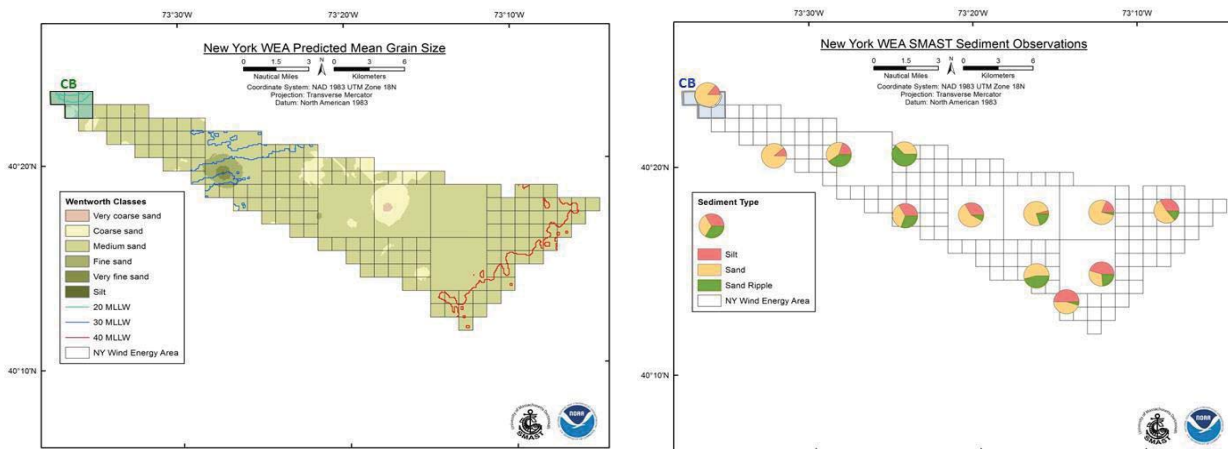
S1 (a) Predicted average sediment type (Wentworth Classification) of sediments based on mean grain size for the NY WEA physical samples: Figure displays interpolated average grain size distribution in NY WEA. The area marked CB is the Cholera Bank. Data Source – Data Source – sediment data (Guida, 2017). (**b**) Surficial Sediments (presence-absence) observations: Observations proportions in pie charts were compiled to show ratio of sediment observations from multiple images taken at each station. Source data: NOAA (Guida, 2017).