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An Integrated Mercury Monitoring Program for Temperate Estuarine and Marine Ecosystems on the North American Atlantic Coast

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Abstract

During the past century, anthropogenic activities have altered the distribution of mercury (Hg) on the earth's surface. The impacts of such alterations to the natural cycle of Hg can be minimized through coordinated management, policy decisions, and legislative regulations. An ability to quantitatively measure environmental Hg loadings and spatiotemporal trends of their fate in the environment is critical for science-based decision making. Here, we outline a Hg monitoring program for temperate estuarine and marine ecosystems on the Atlantic Coast of North America. This framework follows a similar, previously developed plan for freshwater and terrestrial ecosystems in the United States. Methylmercury (MeHg) is the toxicologically relevant form of Hg, and its ability to bioaccumulate in organisms and biomagnify in food webs depends on numerous biological and physicochemical factors that affect its production, transport, and fate. Therefore, multiple indicators are needed to fully characterize potential changes of Hg loadings in the environment and MeHg

bioaccumulation through the different marine food webs. In addition to a description of how to monitor environmental Hg loads for air, sediment, and water, we outline a species-specific matrix of biotic indicators that include shellfish and other invertebrates, fish, birds and mammals. Such a Hg monitoring template is applicable to coastal areas across the Northern Hemisphere and is transferable to arctic and tropical marine ecosystems. We believe that a comprehensive approach provides an ability to best detect spatiotemporal Hg trends for both human and ecological health, and concurrently identify food webs and species at greatest risk to MeHg toxicity.

Keywords

Mercury; Marine Ecosystems; Estuaries; Monitoring; Birds; Atlantic Ocean

Introduction

Anthropogenic releases of mercury (Hg) to the environment have impacted the biosphere substantially, and the ocean has received a major environmental insult as a result of increased Hg releases (Mason and Sheu, 2002; Sunderland and Mason, 2007). Overall, it appears that Hg in the open ocean has increased substantially during the past 200 years, primarily as a result of greater atmospheric Hg deposition (Mason et al., 1994; Selin et al., 2008). Additional local- and regional-scale contamination of the coastal zone has resulted from human activities and associated runoff from the terrestrial environment (USEPA, 1997; Balcom et al., 2004). Therefore, while the impact of anthropogenic emissions in North America on the open ocean may be difficult to quantify (Sunderland and Mason, 2007), there is growing evidence demonstrating increased levels of Hg in United States (U.S.) estuarine and coastal waters and sediments (Mason and Lawrence, 1999; Varekamp et al., 2003; Conaway et al., 2007; Fitzgerald et al., 2007). Elevated inputs of Hg are a health concern because inorganic Hg can be converted to methylmercury (MeHg) (Clarkson and Magos 2006), a neurotoxin that bioaccumulates and biomagnifies through aquatic food webs, and in some cases adversely impacting high-trophic level aquatic organisms (Wolfe et al., 1998; Scheuhammer et al., 2007; Wolfe et al., 2007). Humans are exposed to MeHg principally by the consumption of contaminated fish (Fitzgerald and Clarkson, 1991; Mahaffey, 1998), and elevated MeHg concentrations in fish have resulted in fish-consumption advisories at state, provincial and federal levels (Rice et al., 2003; Mahaffey et al., 2004; Environment Canada 2008) because of their proven negative effects to humans (Anderson 2008). Aquatic sediments and low-oxygen waters are the main sites for the conversion of inorganic Hg to MeHg (methylation), which is mediated primarily by sulfate-reducing bacteria (Compeau and Bartha, 1985; Gilmour et al., 1992). Of the many potential stressors and contaminants in the coastal environment, the issue of MeHg contamination is one of the most pervasive concerns, as evidenced by the extensive fish-consumption advisories that exist for much of the North American coastline.

The coastal zone plays a crucial role in Hg and MeHg cycles, acting as a site of inorganic Hg entrapment, MeHg formation, and high biological productivity. Sunderland and Mason (2007) estimate that about $8.2 \text{ Mmol of Hg yr}^{-1}$ (range, $5.2\text{--}11.4 \text{ Mmol yr}^{-1}$) is trapped in the estuarine and coastal zone, which is comparable to the estimated point source Hg inputs to the atmosphere of $11.3\text{--}16.9 \text{ Mmol yr}^{-1}$. While the coastal zone is a sink for inorganic Hg, mass balance considerations suggest that estuaries and near-shore marine systems are a net source of MeHg to the coastal ocean (Cossa et al., 1996; Hammerschmidt and Fitzgerald, 2004; Hammerschmidt and Fitzgerald, 2006a).

While a number of field studies have focused on Hg speciation in estuarine and coastal ecosystems (e.g. Baeyens et al., 1998; Benoit et al., 1998; Bloom et al., 1999; Hammerschmidt et al., 2004; Heyes et al., 2004; Kannan et al., 1998; Sunderland et al., 2004), there has been

little coordinated monitoring of Hg and MeHg changes in the coastal zone and the effect of these changes on MeHg concentrations in fish, wildlife, and humans. This is surprising, since the majority of fish consumed by humans are from coastal and open-ocean marine environments (FAO, 2004; Sunderland, 2007) and because there are indications that Hg trends have increased over the past decade in wildlife (Rigét et al., 2007).

Structure of Monitoring Program

There is a substantial effort underway to develop a Hg monitoring program across the United States (Mason et al., 2005; Harris et al., 2007), which, while not specifically excluding the coastal zone, has a strong focus on terrestrial and freshwater environs. To augment this effort, the design of a marine monitoring program that quantifies inputs of Hg to coastal and estuarine systems and associated exposure and impact of MeHg on organisms is needed. In 2006, the workshop *Fate and Bioavailability of Mercury in Aquatic Ecosystems and Effects on Human Exposure* was convened by the Dartmouth Toxic Metals Research Program, where marine Hg scientists and human health experts were gathered to articulate research and monitoring needs (Chen et al., 2008a). From that workshop we determined that a coordinated and expanded monitoring program is needed to evaluate 1) spatial and temporal patterns of Hg deposition and transport, 2) MeHg formation and bioaccumulation, 3) wildlife populations at risk from MeHg exposure, and 4) human exposure. These priorities are also embraced by the proposed National Mercury Monitoring Program (Harris et al., 2007). That program identified several monitoring design elements including a national distribution of 10–20 monitoring stations that would include intensive sites bounded by multiple cluster or extensive sites. Intensive sites would establish cause and effect relationships between Hg deposition and environmental change based on a comprehensive range of measurements, while cluster sites would use fewer measurements to better characterize environmental responses in varying ecosystem types. We suggest following this monitoring design for estuarine and marine ecosystems.

Our region of interest includes the estuarine and coastal ecosystems bounded by Chesapeake Bay north to the Gulf of Maine (Figure 1). We organized our Hg monitoring program into four major habitat types: estuarine, coastal, semi-pelagic, and pelagic. We define these habitat types using the following criteria: (1) estuarine areas are semi-closed coastal waters that have free access to the ocean and variable but generally shallow water depths; (2) coastal areas extend out to a depth of 100 m (and are often within three nautical miles [5.6 km] from shore); (3) semi-pelagic areas represent the middle to outer continental shelf and range in water depth between 100 and 200 m (and are often located from three to 200 nautical miles [5.6 to 370.0 km] from shore but occasionally extend further); and (4) pelagic areas are open ocean waters that are >200 m in depth (and are usually >200 nautical miles [>370 km] from shore). Within these habitat types, we recognize different and relatively independent marine food webs exist that should be monitored separately for their ability to transfer MeHg to upper trophic levels. For example, benthic and demersal food webs at the bottom of the ocean need to be monitored separately from pelagic food webs higher in the water column.

We have identified five categories of indicators: abiotic measurements, invertebrates, fish, birds, and mammals. Within the biotic categories, we based taxonomic selection on multiple criteria (Table 1). A description of the target taxa within each of the four major habitat types is followed by sampling strategies that identify best tissues and ancillary data that increase interpretive power, including selenium (Se) concentrations and stable isotopes. The best indicators for evaluating toxicity and spatiotemporal trends of MeHg for protection of human health can differ from those for ecological health. In order to encompass both for fish we created low and high trophic level categories (Table 2) that reflect Hg concentrations for ecological and human health concerns, respectively.

There are several existing, long-term marine Hg monitoring programs. They include the National Atmospheric Deposition Program's Mercury Deposition Network for air (Lamb and Bowersox 2000), the National Oceanographic and Atmospheric Administration's (NOAA) Mussel Watch Program for shellfish (Chase et al. 2001), the U.S. Environmental Protection Agency's (USEPA) National Coastal Assessment Program for shellfish and marine fish (Cunningham et al. 2003), the U.S. Geological Survey's Contaminant Exposure and Effects – Terrestrial Vertebrates program for terrestrial wildlife (Cohen et al. 2003), and the Atlantic, Arctic and Pacific Seabird Egg Contaminant Monitoring Programs of Environment Canada (Pearce et al., 1979; Harris et al., 2005; Braune, 2007). We use these existing programs as a basis for identifying indicators and recommending Hg monitoring locations. Broad justifications and specific methodologies not detailed in this paper appear in Harris et al. (2007). Tissue Hg concentrations are provided on a wet weight (ww) basis, unless otherwise noted. Scientific names for the program's indicator organisms are provided in Table 3.

Indicator Compartments

Abiotic Indicators

Major sources of inorganic Hg to the coastal zone include direct atmospheric deposition, rivers (including runoff of atmospherically deposited Hg), and water pollution control facilities, with most of the MeHg derived from *in situ* transformation of inorganic Hg (Mason et al., 1999; Balcom et al., 2004, 2008). Given these sources, it is important to measure both direct atmospheric inputs and to quantify watershed contributions of inorganic Hg to the coastal zone. Collection of sediment cores and determination of total Hg and MeHg in both surficial sediment and water at intensive sites is recommended (Table 2).

At intensive sites, event-based wet deposition collection (Driscoll et al., 2007) and Hg atmospheric speciation and deposition should be measured. While the atmospheric concentration of Hg⁰ is reflective of the global atmospheric Hg pool and is not a sensitive local indicator of short-term regional change (Slemr et al., 2003), levels of ionic gaseous and particulate Hg show a higher regional variability that may reflect regional emissions (Schroeder and Munthe, 1998; Ryaboshapko et al., 2007a, b). Ionic gaseous and particulate Hg species are, however, relatively difficult to measure (Landis et al., 2002) and although there is large uncertainty in the current estimates of Hg dry deposition, new approaches are being developed (Skov et al., 2006). Atmospheric Hg speciation measurements at intensive sites should be coupled with flux estimates from other sources into the coastal zone. Specifically, measurements of groundwater and surface river water fluxes and Hg speciation provide an ability to examine the role of Hg and MeHg transport from associated watersheds to the coastal zone.

Precipitation and other climatic variables also influence export to coastal ecosystems, especially with respect to sporadic and extreme events. Measurements of Hg in wet deposition are accomplished relatively easily and are monitored currently at the national level by the National Atmospheric Deposition Program's Mercury Deposition Network (MDN), which includes 11 sites within our study area (MDN, 2008; Figure 2).

Changes in atmospheric Hg deposition are also recorded by sediments, and historical records of input have been inferred from the analysis of estuarine and wetland sediment cores (Conaway et al., 2007; Varekamp et al., 2003). There is a large body of experimental and observational evidence for their reliability, and well-established protocols for the collection, processing, and interpretation of these records (Porcella, 1996). However, there are confounding effects of watershed input, sediment mixing (physical and bioturbation) and other factors that impact the level of temporal resolution.

While the relationship between biota and sediment Hg and MeHg concentrations is difficult to construct (Mason, 2000), measurements of sediment MeHg provide an integrative measure of the impact of changes in Hg input and other factors on net production of MeHg (Benoit et al., 2003). MeHg in sediment and interstitial waters is available for uptake by benthic organisms and may be an important source to overlying water, either by upward diffusion or bioadvection. It has been shown for numerous freshwater (Benoit et al., 2003) and some marine (Heyes et al., 2006; Kim et al., 2006) ecosystems that there is a relationship between short-term Hg methylation rate measured using assays and *in situ* MeHg concentration or %MeHg in sediments; however, these correlations do not necessarily extend to all coastal marine deposits (Hammerschmidt and Fitzgerald, 2004, 2006a; Ogrinc et al., 2007; Hammerschmidt et al., 2008). Assays of Hg methylation are not recommended for the extensive cluster sites. Nonetheless, sediment MeHg concentration and %MeHg may be useful proxies of the rate of change in bulk MeHg concentration and relative production. Measurements of %MeHg in sediment will allow assessment of whether changes of sedimentary MeHg content is related directly or indirectly to changes in atmospheric Hg input, as well as provide an abiotic indicator of potential biological exposure.

Total Hg and MeHg measurements in water, both in bulk and in the dissolved and particulate fractions, have been made in many coastal ecosystems (Kannan et al., 1998; Mason et al., 1999; Whalin et al., 2007; Balcom et al., 2008). Concentrations in water can be influenced by factors unrelated to Hg inputs, such as the variation in particulate matter and dissolved (DOC) and particulate organic carbon (POC), which are needed to improve interpretation (Table 2). Despite spatial variation in these factors, some studies have shown a reasonable correlation between MeHg in water and MeHg in freshwater fish, reflecting the influences of bioaccumulation at the base of the pelagic food chain (Brumbaugh, 2001). As levels of Hg species in water vary seasonally and with depth within a particular water body, these measurements must be assessed with consideration of anticipated spatial variability. Generally, developing a relationship between Hg or MeHg concentrations in water and estuarine or marine organisms is challenging and remains poorly described in the literature.

Invertebrate indicators

Selection of indicator species at the base of pelagic and benthic food webs in marine systems should include invertebrate species representing different functional feeding groups, such as benthic infauna, benthic epifauna, epipelagic, and mesopelagic biota (Table 1, 3). Inclusion of both benthic and pelagic food webs facilitates an understanding of differing pathways of MeHg transfer into higher trophic levels. Studies in freshwater ecosystems suggest that pelagic feeding organisms have the ability to bioaccumulate greater concentrations of MeHg than benthic feeding fauna (Gorski et al., 2002; Power et al., 2002). However, little is known for marine systems. Specifically, some marine food webs may differ from freshwater systems in the manner MeHg is transferred to higher trophic positions. For example, nurse sharks (*Ginglymostoma cirratum*) can have greater muscle Hg concentrations than associated pelagic species, such as the blacktip shark (*Carcharhinus limbatus*) (BioDiversity Research Institute, unpubl. data).

Benthic infauna that live in the sediments of estuarine and coastal areas are useful taxa for monitoring MeHg bioaccumulation directly from ingestion or absorption from sediment. Benthic amphipods are regular inhabitants of sediments and are used frequently in sediment toxicity testing (e.g., *Leptochirus*, *Corophium*). They obtain their food, and likely their MeHg, from ingesting detrital material and sediments. The most abundant sediment infauna are polychaetes.

In estuarine and coastal waters several non-native, epifaunal species are common. The periwinkle is a primary consumer that grazes on biofilms and periphyton on sediment and rock

surfaces. The green crab also is non-native to the U.S. but now inhabits coastal waters across the study area. It is a secondary consumer that feeds on small fish, benthic invertebrates, and detrital material. MeHg concentrations in these two species range greatly across sites and appear to be related modestly with sediment Hg concentrations (Chen et al., unpublished data).

Benthic invertebrate indicators for human health risk that inhabit estuary, coastal, and semipelagic areas include a wide variety of shellfish such as crabs, mussels, clams, oysters, and lobsters. Being important commercially harvested species, the monitoring of these taxa for certain contaminants is conducted by governmental regulatory agencies. For example, the NOAA Mussel Watch and Gulfwatch Programs have historically collected contaminant data for blue mussels, eastern oyster, and other bivalves across broad spatial and temporal scales (Jones et al., *This issue*). Several studies have documented elevated muscle Hg concentrations in Atlantic lobster (Greig et al., 1975; Vassiliev et al., 2005; Hammerschmidt and Fitzgerald, 2006b), including monitoring efforts by the USEPA (Cunningham et al. 2003). Differences in total Hg concentrations vary less than other metals for synoptically collected, co-located species (Jones et al. *This issue*), yet there is evidence to suggest that concentrations of MeHg vary in bivalves according to feeding strategy.

Pelagic cephalopods, such as squid, are important ecologically and for human health. In the northeastern Atlantic Ocean, benthic cephalopods had significantly higher Hg concentrations than pelagic species (Bustamante et al. 2006). In the Mediterranean Sea, cephalopods Hg concentrations can be substantially higher than small fishes of similar trophic position, and appear to account for the highly elevated Hg liver body burdens in marine mammals (Frodello et al. 2000). Euphausiids are also typically relevant epipelagic and mesopelagic organisms for monitoring MeHg availability (Braune et al., 1987a; Monteiro et al., 1996).

For many benthic invertebrates, sampling often simply includes hand collection. Trap and baiting techniques are used for crustaceans, while nets are employed for pelagic euphausiids and cephalopods. Whole body analysis of soft tissue or solely muscle tissue are regularly used and presented on a wet weight basis. Because the fraction of total Hg as MeHg can vary widely in invertebrates, measurements of MeHg concentrations are important for assessing risk and developing food web models (Tremblay et al., 1996a, b; Gorski et al., 2002; Mason and Benoit, 2003).

Fish Indicators

There are numerous fish species that, when routinely sampled for Hg, are useful indicators of human and ecological health (Table 1, 3). For assessing and monitoring trends in ecological health risk, we also emphasize species that are target prey for piscivorous fish and wildlife and species that maintain sustainable and robust populations. Because there is a growing body of evidence of Hg-related effects to freshwater (Drevnick et al., 2008; LaRose et al., 2008) and estuarine fish species (del Carmen Alvarez et al., 2006) the sustainability of healthy populations of high trophic level marine fish needs to be considered for purposes of conservation and management. For assessing and monitoring trends in human health risk, we emphasize commercially and/or recreationally valuable fish that have documented patterns of Hg bioaccumulation.

In estuaries, mummichogs are ubiquitous because they can withstand broad environmental conditions. They are often used for examining environmental Hg loadings (Khan and Weis, 1993) and their potential ecological effects (Zhou et al., 1998). Some studies indicate they can build a tolerance to contaminants, including MeHg (Weis, 2002), although individuals with elevated MeHg concentrations are more prone to being preyed on because of impairment to predator-avoidance behaviors (Smith and Weis, 1997). While the mummichog inhabits quiet backwaters, the American sand lance prefers shallow, sandy brackish waters and is an

important prey item for estuarine, coastal, and even semi-pelagic birds. The striped bass is a common recreationally-fished species of estuaries and has been shown to accumulate $> 0.5 \mu\text{g/g}$ (ww) of Hg (Davis et al., 2002; Mason et al., 2006). However, because it has multiple feeding strategies, migrates amongst areas of differing Hg sensitivity, and exhibits poor growth-Hg relationships, its use as an indicator species is limited. A species of less importance to the sport fishery industry, but one that has commercial and subsistence interests is the scup or porgy. It is found in estuaries and nearshore areas and is a primary indicator species used by the USEPA National Contaminant Assessment Program (NCAP) and rarely exceeds $0.10 \mu\text{g/g}$ (ww) (Cunningham et al. 2003).

In coastal waters, the striped anchovy and butterfish are common and widespread forage fish for piscivorous wildlife. Bluefish are a recreationally important, coastal piscivore that have 1) well-defined age-growth and growth-Hg relationships, 2) are widely distributed, and 3) been characterized for MeHg contamination in a variety of coastal waters (Ashley and Horwitz, 2000; Burger et al., 2005; Hammerschmidt and Fitzgerald, 2006b). The winter flounder is a highly valued species for Hg monitoring because of its recreational and commercial interests, extensive use by the NCAP, and prevalence of tumors related to contaminant exposure (Moore et al. 2004) – making it a more sensitive indicator species than the closely related summer flounder (*Paralichthys dentatus*). Both flounder species are routinely sampled by the NCAP and generally have fillets $< 0.10 \mu\text{g/g}$ (ww) of Hg.

The source of MeHg in semipelagic and pelagic habitats is unknown. MeHg bioaccumulation in these regions may result from deep ocean waters, sediments, or hydrothermal vents (Kraepiel et al., 2003; Lamborg et al., 2006; Chen et al., 2008b), hydrologic advection (Hammerschmidt and Fitzgerald, 2006a) and bioadvection from the coastal zone (Fitzgerald et al., 2007), or methylation in waters of high-biological productivity (Topping and Davies, 1981; Mason and Sullivan, 1999; Chen et al., 2008b). Different fish species emerge as suitable indicators of MeHg availability in these offshore habitats. Ecological risk in semipelagic waters can be well documented using Atlantic herring (Braune 1987a). Measurements of Hg in sedentary, demersal species such as the Atlantic cod provide an opportunity to monitor MeHg bioaccumulation in specific locations over time (e.g., Staveland et al. 2005). Other high trophic-level species with commercial value and known Hg concentrations include the yellowfin tuna (Kraepiel et al., 2003) and related species.

The North Atlantic saury is one of the most abundant epipelagic species (< 200 m depths) in our study area and serves as prey for many high trophic level species. A more novel fish group to monitor are lanternfish that are found in mesopelagic environments (> 300 m in depth) and migrate diurnally over hundreds of meters in depth. They regularly form the prey base for pelagic seabirds such as the Leach's storm-petrel, which forage nocturnally on lanternfish at the ocean surface (Montevecchi et al. 1992). Concentrations of Hg in lanternfish have been used to support the current understanding of deepwater MeHg production in the open ocean, and to identify temporal and spatial patterns in oceanic Hg bioavailability (Montiero et al., 1996; Martins et al., 2006).

While high-trophic level species such as the blue shark and swordfish are of commercial and recreational interest, they can bioaccumulate concentrations of Hg that are harmful for human consumption (often $> 1.0 \mu\text{g/g}$, ww; unpubl. data from the U.S. Food and Drug Administration [FDA] and USEPA; Branco et al., 2007). However, their low densities and wide ranging abilities make their use as indicators of specific areas challenging. Although average shark Hg concentrations have been generically described by the FDA, some species can attain exceedingly high muscle Hg concentrations depending on their size, prey base and geographic origin. For example, Garica-Hernandez et al. (2007) found smooth hammerhead sharks (*Sphyrna zygaena*) in the Gulf of California surpassing $21.0 \mu\text{g/g}$ (ww) in their muscle tissue.

Based on $\delta^{15}\text{N}$ values, less variable prey bases in some shark species, such as the blue shark, make it a more preferable species to monitor Hg concentrations over spatiotemporal scales of interest than species with highly variable prey bases, such as the shortfin mako shark (*Isurus oxyrinchus*) (Estrada et al., 2003).

Sampling strategies can vary widely and depend on the target habitat and species. Fish are typically analyzed on a whole body basis for ecological health monitoring, while muscle tissue is removed and the fillet is analyzed for assessment of human exposure. Both approaches tend to analyze total Hg on a wet weight basis. Most of the Hg in fish muscle is in the methyl form (Bloom 1992). Lower trophic level fish also provide an ability to predict MeHg transfer rates to higher trophic levels. For fish species of commercial and recreational interest, acquisition of tissue samples for Hg analysis using biopsy plugs of muscle directly at the dock is a viable and cost-effective strategy (Bank et al., 2007a). Biopsies and other non-lethal sampling methods are recommended for sharks, which have experienced tremendous declines because of overfishing (Myers et al., 2007).

Since variation in fish Hg concentrations is commonly influenced by the growth characteristics of length, age, or weight, those fish species with well-defined, growth-Hg relationships are the best candidates as indicator species. Monitoring Hg programs using fish should always acquire total length, weight, and ideally other important metrics such as age. There are other associated data that increase interpretation powers, such as Se levels and stable isotope information. The general theme of the current literature indicates that Se should be measured in fish tissue to better understand risks to consumers. The role of bioavailable Se is of growing interest for human exposure (Ralston et al., 2007). Some variations in Hg levels can be explained by trophic position as determined by $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$. Bank et al. (2007a) found that red and gray snapper (*Lutjanus campechanus* and *L. griseus*, respectively) exhibit modest but consistently predictable differences in Hg concentrations and these were related to differences in trophic position.

Bird Indicators

Suggested bird species for monitoring Hg trends vary according to foraging guild (i.e. piscivore vs. invertivore) and habitat type (Table 1, 3). Our selection criteria for birds emphasizes breeding individuals because they are generally territorial (or have small home ranges), are likely consuming prey items that have higher MeHg concentrations than in the winter (Leermakers et al., 1995; Ramlal et al., 1993), and are more reflective of local conditions compared to migrants. Summer time Hg concentrations can also be better linked to meaningful endpoints of adverse effects, such as reproductive success (Burgess and Meyer, 2008; Evers et al., 2008). Use of multiple bird species for monitoring Hg provides the most comprehensive coverage for detecting changes in various habitat types and food webs that may not be predictive from one another (Pearce et al. 1989).

In estuaries, there are often species of high conservation concern (Ackerman et al., 2007) with apparently high sensitivity to Hg input (Heinz et al., 2008), and recently documented adverse reproductive effects from Hg (Schwarzbach et al., 2006). *Ammodramus* sparrows are recommended as indicators for estuarine invertivore food webs. One species, the saltmarsh sharp-tailed sparrow has Hg body burdens that tend to exceed those in associated songbirds (Shriver et al., 2006), and in some estuaries, lowered reproductive success is related to elevated blood Hg concentrations (Lane et al., 2008). Indicators of the estuarine piscivore food web include a choice of over 10 species of wading birds in our study area. Unfledged wading birds, particularly the black-crowned night-heron, is often-used as the indicator age group (Rattner et al., 2000; Henny et al., 2002). Areas without wading bird sampling opportunities can be evaluated for MeHg availability in piscivores through sampling of the belted kingfisher.

Kingfishers are a relatively unique indicator, as they are one of the few birds that can be used to compare MeHg availability across marine and freshwater habitats (Evers et al., 2005).

In coastal waters of the Gulf of Maine, the common eider regularly forages on the blue mussel (Wayland et al., 2001), while the piscivorous black guillemot depends on benthic fish (Butler and Buckley, 2002). Mean Hg concentrations in eggs of the black guillemot are significantly greater than those in associated seabirds (Goodale et al., *This issue*), and because their prey occupy relatively small home ranges, guillemots are valuable indicators for characterizing distinct areas of interest. The osprey breeds along our entire study area and is an important indicator species since it is an obligate piscivore, is found across the northern hemisphere, and is commonly monitored for Hg in both freshwater (Hughes et al., 1997) and estuarine and marine ecosystems (Golden and Rattner, 2003; Henny et al., 2008; Rattner et al., 2008).

The common tern is another ubiquitous piscivore that regularly forages in coastal and offshore areas and has well-described Hg body burdens (Braune, 1987b; Burger et al., 1994; Nisbet et al., 2002). The common tern has Hg body burdens that are comparable to the larger double-crested cormorant (Goodale et al., *This issue*). Mercury monitoring efforts with cormorant eggs are an efficient approach because of the cormorant's common status and colonial nesting tendencies.

Describing the availability of MeHg in pelagic waters of our study area using birds is challenging. The Leach's storm-petrel is best. Although this seabird nests on outer coastal islands in the Gulf of Maine, it forages along the continental shelf on mesopelagic organisms such as myctophids, amphipods, and euphausiids (Montevecchi et al., 1992). Atmospheric deposition of Hg on the ocean surface increasingly reflects global airshed Hg concentrations as the distance from mainland increases (Gill and Fitzgerald, 1987). Blood Hg concentrations in the storm-petrel may provide a relatively accessible technique for monitoring changes in food web MeHg availability that is related to either global Hg pools or MeHg sources relatively independent of continental influences.

Bird tissues regularly used for monitoring environmental Hg loads for short-term exposure are blood and eggs (usually as ww) and for longer-term exposure are feathers (as fresh weight) (Evers et al. 2005). Feathers provide a good measure for examining long-term Hg trends (Thompson et al., 1992; Monteiro and Furness, 1997). These three tissues are generally analyzed for total Hg because they are mostly representative of MeHg (Wolfe et al. 2007). Similar to fish, other measurements are typically required to best describe Hg body burdens. While bird age past the first 4–5 years is generally unknown (unless uniquely marked), the size, molt status, sex and age class (juvenile vs. adult) are important for interpreting Hg concentrations (Evers et al. 2005). An understanding of Se levels and stable isotopes are also useful. The potential protective role of bioavailable Se in birds appears to be complex (Scheuhammer et al. 2008). Unlike marine mammals where the interaction between Se and Hg is often a predictable 1:1 ratio in organs such as the liver (Ikemoto et al. 2004), such a pattern is not observed in birds (Ohlendorf et al. 1986, 1991). There may even be an additive effect on the frequency of teratogenic effects in waterfowl when highly elevated dietary levels of Se and Hg (10.0 µg/g) are ingested (Heinz and Hoffman 1998). The benefits of measuring stable isotopes (e.g. ¹⁵N, ¹³C) along with Hg in birds include (1) better understanding intraspecific variability within colonies where dietary specialization by different individuals can influence blood Hg levels (Bearhop et al. 2000), (2) distinguishing local uptake of dietary MeHg from off-site uptake (Nisbet et al. 2002), and (3) selecting the best tissue for monitoring local Hg levels (Thompson et al. 1998).

Mammal Indicators

Our suite of mammalian indicators includes a broad mix of terrestrial and marine taxa (Table 1, 3). Although mustelids are used widely as indicators of MeHg availability in freshwater ecosystems (Strom, 2008; Klenavic et al., 2008), two species, the American mink and North American river otter, also forage regularly in estuarine and marine ecosystems where fish are a dominant prey (Yates et al., 2005; Lake et al., 2007).

Pinnipeds are the most accessible marine mammal to be utilized as indicators for coastal habitats. Seals depend on ledges or beaches for pupping and resting, which provides a feasible method for tracking individual MeHg body burdens. Harbor seals are the most common pinnipeds in our study area, have the smallest home range compared to associated seal species, and are most accessible for sampling purposes. Their importance as a global sentinel species is also well recognized (Ross, 2000). Harbor seals selectively forage on small, schooling fish and squid and their diet changes seasonally (Payne and Selzer, 1989).

In semi-pelagic waters, toothed cetaceans forage on prey items such as squid and fish that are higher on the food chain than krill and other invertebrates. Their Hg body burdens therefore average higher than baleen cetaceans (Hansen et al. 1990). The harbor porpoise is one of the more common and ubiquitous toothed cetacean in coastal and semi-pelagic waters, feeding on cephalopods and Atlantic herring (Fontaine et al., 1994). In more southern waters of our study area, the bottlenosed dolphin is often used as an indicator of contaminants, including Hg (Kuehl and Haebler, 1995; Frodello et al., 2000).

In pelagic and more coastal waters of the northern Atlantic Ocean, pilot whales are useful indicator species because of their dietary importance to many native cultures (Andersen et al., 1987), where consumption has been regulated due to elevated Hg concentrations in muscle tissue (3.3 µg/g, ww) (Weihe et al., 1996). The diet of pilot whales includes fish and cephalopods (Katona et al., 1993). Beaked whales are rarely accessible for sampling purposes unless they are stranded. Their novel value as indicators of MeHg availability in open ocean habitats is their longevity and high trophic-level position, which can result in highly elevated liver Hg concentrations (Bustamante et al. 2003).

Marine mammals are known to bioaccumulate varying levels of MeHg depending on species, diet, age, sex, reproductive status, geographic distribution and range of ocean habitat (Nagakura et al., 1974; Gaskin et al., 1979; Dietz et al., 1996; Wagemann et al., 1998; Das et al., 2003). Multiple tissues are regularly used to characterize Hg body burdens, including skin, blubber, muscle, kidney, and liver. Muscle is more likely to contain a higher proportion of MeHg (50–100%), while liver contains a lower percentage (O'Hara et al. 2003). While sampling live marine mammals is challenging, samples taken from individuals stranded or by-catch from fish nets does provide a routinely available approach for acquiring tissues. As in fish and birds, the protective or toxic role of associated Se levels should be determined as well. Marine mammals have adapted to elevated levels of dietary MeHg by sequestering it as a nontoxic, inorganic form in the liver, often with a 1:1 ratio with Se (Koeman et al., 1973; Itano et al., 1984; Ikemoto et al., 2004). Stable isotope analysis can demonstrate prey choice (Das et al., 2000) and food web relationships (Campbell et al., 2005).

Conclusions

A high resolution and comprehensive program for monitoring environmental Hg loads in air, sediment, and water of estuarine and marine environments and the subsequent ecological response in invertebrates, fish, birds, and mammals in terms of both human and ecological health concerns is described above. Our intention for developing such a detailed list of indicator compartments is to provide multiple selection options to accommodate existing monitoring

efforts, and different site-specific objectives, expertise, and funding. To best detect temporal changes in environmental loading, we recommend a minimum effort to include measurements of wet Hg deposition, MeHg and percent MeHg in estuarine/marine sediment, MeHg in young fish occupying small home ranges, high trophic level fish of greatest local interest for human consumption, and relevant breeding birds.

Mercury data have been amassed for several coastal areas and represent multiple compartments measured in parallel with process-level investigations, including the Chesapeake Bay (Mason et al., 1999; Lawson et al., 2001); Long Island Sound (Rolfhus and Fitzgerald 2001; Balcom et al. 2004; Hammerschmidt et al. 2004; Hammerschmidt and Fitzgerald, 2006b); Acadia National Park (Bank et al., 2007b; Kahl et al., 2007); and outer Bay of Fundy (Gaskin et al., 1979; Pearce et al., 1979; Braune 1987a,b; Elliot et al., 1992; Harding et al., 2005; Ritchie et al., 2006; Sunderland et al., 2004, 2008) (Figure 1). These locations are strong candidates for the basis of a comprehensive, standardized monitoring program that tracks estuarine and marine environmental Hg loadings as well as watershed contributions. Within these and other areas, site considerations should encompass protected areas, which commonly serve as important shellfish beds, nursery areas for fish, and areas of special concern for wildlife conservation. Examples of such areas include National Wildlife Refuges, National Parks, National Estuarine Reserves, and The Nature Conservancy preserves.

Establishing a standardized, long-term Hg monitoring network in coastal areas is essential for monitoring and demonstrating the impact of domestically-produced Hg emissions, as well as the increasing global Hg loads stemming from rapidly growing sources in Asia that, based on current models and estimates, contribute an estimated 54% of the global anthropogenic Hg emissions in 2000 (Pacyna et al., 2006). Today, those sources are projected to contribute at even higher rates, potentially offsetting emission declines in other regions of the northern hemisphere (e.g., northeastern United States). The subsequent uncertainty for increases in the global Hg pool from Asia and elsewhere, combined with effects from climate change (e.g., Faroe Islands, Booth and Zeller, 2005) and ocean acidification (Caldeira and Wickett, 2003), both of which may exacerbate MeHg availability, are especially serious threats to marine-based human and ecological health. While our taxonomic template for monitoring environmental Hg loading is most applicable to marine ecoregions identified by Spalding et al. (2007)) in the temperate North Atlantic realm, it can be readily modified for other biogeographic areas that need to be characterized (Figure 2). While arctic realms are of increasing concern for the magnitude of Hg loadings (Lindberg et al. 2002) and related adverse implications to wildlife (Braune et al., 2006) and human health (Jewett et al., 2003), there is also evidence that marine ecoregions encompassing equatorial realms support high trophic marine inhabitants with elevated Hg concentrations that may reflect habitat sensitivity to environmental Hg loading (Evers et al., *In Press*). The threats posed by anthropogenically-redistributed Hg on marine habitats require strong scientific underpinnings to understand the considerable complexities in Hg biogeochemistry and MeHg production. This understanding is critical to properly regulating and managing Hg at local, continental and global scales. Standardized Hg monitoring programs can ultimately provide the necessary scientific foundation for comprehensive and protective policies (Chen et al., 2008a).

New policies to reduce environmental Hg loading are urgently needed as evidenced by recent scientific findings documenting human health concerns (Mergler et al., 2007; Mahaffey et al., 2008). Although long-term studies of MeHg uptake through marine food on the Seychelles Islands are not definitive in demonstrating significant adverse effects to children (Myers et al., 1997, 2003), a parallel study on the Faroe Islands has shown a relationship with high MeHg uptake and cognitive deficits in children (Grandjean et al., 1997). While these effects are potentially confounded by elevated polychlorinated biphenyls (Grandjean et al., 2001), numerous recent studies now definitively establish elevated Hg body burdens with

neurotoxicity that results in adverse changes in developmental, behavioral and physiological endpoints in humans (Mergler et al. 2007; Anderson 2008). Greater scientific evidence in documented effects and toxicity threshold levels for human health increases the urgency for assessing the impacts of environmental loading and subsequent MeHg uptake. Based on blood and hair Hg concentrations, from 5–10% of American women are estimated to be at risk to MeHg toxicity (McDowell et al., 2004; Mahaffey et al., 2008), which may relate to average annual costs of >\$8 billion of diminished U.S. economic productivity (Trasande et al., 2005). People in coastal communities are at greatest risk to Hg-contaminated food (Yasutake et al., 2004; Hightower et al., 2006; Mahaffey et al., 2008). To better assess ingestion of MeHg from marine food items, Chen et al. (2008a) suggested a tracking system to inform human consumers about catch location, type of fish, and brand of fish product.

To meet the increasingly-evident demands for policy-makers to set rules and regulate the anthropogenic release of Hg into the environment (UNEP, 2002; USEPA, 2005; Swain et al., 2007), a standardized monitoring network would meet multiple tangible needs to (1) assess current and future MeHg toxicity for human and ecological health, (2) provide an ability to measure and model risk of human health, (3) track changes in environmental Hg loads with changes in Hg emissions and effluents, (4) identify biological Hg hotspots, (5) provide a basis for merging existing Hg datasets, and (6) establish a collaborative of governmental, academic, non-profit organizations, and industry representatives that will collect and share Hg data. Such a network is now planned for 10–20 stations across freshwater and marine ecosystems in the United States (Harris et al., 2007). Transboundary extensions would greatly enhance network effectiveness and provide a strong basis for global-scale decision-making.

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Figure 1.

Area for consideration of a standardized Hg monitoring network, including sites of potential Hg monitoring stations and their association with existing Hg deposition network stations¹.

¹Using the National Atmospheric Deposition Program's (NADP) wet deposition of chloride pattern as a proxy for marine influence on precipitation chemistry, a sharp change in influence is clearly present (David Gay, pers. comm.). Marine air greatly influences precipitation chemistry within the first 400 to 500 kilometers along the Gulf Coast, and less than 250 kilometers along the Mid-Atlantic and Northeast coasts of the United States and Canada. See the NADP chloride deposition measurements (<http://nadp.sws.uiuc.edu/isopleths/maps2006/cldep.gif>).

Figure 2.

Simplified universal food web components recommended for mercury sampling¹.

¹Recommended sampling design for intensive sites includes all components. For cluster sites include at least primary components (p) and secondary components (s) when possible. The influence of squid and other cephalopods as a transfer mechanism for MeHg in the food web may be important for higher trophic levels.

Table 1

Criteria used for each major biotic category of indicators.

Criteria type	Invertebrates	Fish	Birds	Mammals
Ubiquitous and abundant in study area	X	X	X	X
Ease of sampling	X	X	X	X
Comparability with existing Hg data	X	X	X	X
Understanding of MeHg transfer in food web	X	X	X	X
A distinct point of time is in a major habitat type	X	X	X	X
Part of existing Hg monitoring programs	X ¹	X ²	X ³	
Used for regular human consumption	X	X		
Reflect highest MeHg exposure		X	X	X
Existing information on effects		X	X	X

¹ National Oceanographic and Atmospheric Administration Mussel Watch (Chase et al., 2001)

^{1,2} U.S. Environmental Protection Agency National Coastal Assessment Program (Cunningham et al., 2003)

³ Environment Canada Seabird Egg Contaminant Monitoring Program (Harris et al., 2005; Braune, 2007)

Table 2

Suggested abiotic compartments for monitoring Hg from Chesapeake Bay north to the Gulf of Maine.

Matrix measured for Hg	Associated measurements
Air (total in wet and dry deposition)	Precipitation
Water (total and methyl)	Dissolved organic carbon
	Particulate organic carbon
	Particulate matter
Sediment (total and methyl)	Organic carbon

Table 3Suggested indicator taxa for monitoring Hg from Chesapeake Bay north to the Gulf of Maine¹

Taxa ²	Estuary	Coastal	Semi-pelagic	Pelagic
Invertebrates - Benthic infauna	Amphipods	Polychaetes	-	-
Invertebrates – Benthic epifauna and Epipelagic/ Mesopelagic fauna	Periwinkle ^A (<i>Littorina littorea</i>) Green crab ^A (<i>Carcinus maenas</i>)	Blue mussel ^A (<i>Mytilus edulis</i>) Eastern oyster ^S (<i>Crassostrea virginica</i>)	Atlantic lobster ^N , ³ (<i>Homarus americanus</i>)	Squid ^A (<i>Loligo</i> spp.) Euphausiids ^A
Fish – low trophic level	Mummichog ^A (<i>Fundulus heteroclitus</i>) American Sand lance ^A (<i>Ammodytes americanus</i>)	Striped anchovy ^A (<i>Anchoa hepsetus</i>) Butterfish ^A (<i>Peprilus triacanthus</i>)	Atlantic herring ^A (<i>Clupea harengus</i>)	North Atlantic saury ^A (<i>Scomberesox saurus saurus</i>) ^N Lantern fish ^N (Myctophid spp.)
Fish – high trophic level	Striped bass ^A (<i>Morone saxatilis</i>) Scup ^A , ³ (<i>Stenotomus chrysops</i>)	Bluefish ^A (<i>Pomatomus saltatrix</i>) Winter Flounder ^A , ³ (<i>Pseudopleuronectes americanus</i>)	Atlantic cod ^N , ³ (<i>Gadus morhua</i>) Yellowfin tuna ^A (<i>Thunnus albacaras</i>)	Swordfish ^A (<i>Xiphias gladius</i>) Blue shark ^A (<i>Prionace glauca</i>)
Birds – invertivores	Nelson's ^N and Saltmarsh ^A sharp-tailed and Seaside ^S sparrows (<i>Ammodramus nelsoni</i> , <i>A. caudacutus</i> , <i>A. maritimus</i>)	Common eider ^N (<i>Somateria mollissima</i>)	-	-
Birds – piscivores	Black-crowned night heron ^A (<i>Nycticorax nycticorax</i>) Belted kingfisher ^A (<i>Megasceryle alcyon</i>)	Black guillemot ^N (<i>Cepphus grylle</i>) Osprey ^A (<i>Pandion haliaetus</i>)	Double-crested cormorant ^A (<i>Phalacrocorax auritus</i>) Common tern ^A (<i>Sterna hirundo</i>)	Leach's' storm-petrel ^N (<i>Oceanodroma leucorhoa</i>)
Mammals	American mink ^A (<i>Mustela vison</i>)	Harbor seal ^N (<i>Phoca vitulina</i>) North American river otter ^A (<i>Lontra canadensis</i>)	Harbor porpoise ^A (<i>Phocoena phocoena</i>) Bottlenosed dolphin ^S (<i>Tursiops truncatus</i>)	Pilot whale spp. ^A (<i>Globicephala</i> spp.) Beaked whale spp. ^A (<i>Mesoplodon</i> & <i>Ziphius</i> spp.)

¹ Areas where species are principally found are identified as: S = South (Chesapeake Bay to Hudson River mouth), N = North (Long Island Sound to Gulf of Maine), and A = All (Chesapeake Bay to Gulf of Maine).

² Species-specific taxa are organized by major habitat type to indicate primary area of residence. Most species are often found in multiple habitat-types, which provides a horizontal trophic relay of MeHg, or bioadvection, across habitat types because of species movement patterns and subsequent predator-prey interactions.

³ Part of the USEPA's National Coastal Assessment Program for the Northeast Region (Acadian and Virginian biogeographic regions).