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Mercury sources and fate in the Gulf of Maine ☆

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ABSTRACT

Most human exposure to mercury (Hg) in the United States is from consuming marine fish and shellfish. The Gulf of Maine is a complex marine ecosystem comprising twelve physioregions, including the Bay of Fundy, coastal shelf areas and deeper basins that contain highly productive fishing grounds. Here we review available data on spatial and temporal Hg trends to better understand the drivers of human and biological exposures. Atmospheric Hg deposition from U.S. and Canadian sources has declined since the mid-1990s in concert with emissions reductions and deposition from global sources has increased. Oceanographic circulation is the dominant source of total Hg inputs to the entire Gulf of Maine region (59%), followed by atmospheric deposition (28%), wastewater/industrial sources (8%) and rivers (5%). Resuspension of sediments increases MeHg inputs to overlying waters, raising concerns about benthic trawling activities in shelf regions. In the near coastal areas, elevated sediment and mussel Hg levels are co-located in urban embayments and near large historical point sources. Temporal patterns in sentinel species (mussels and birds) have in some cases declined in response to localized point source mercury reductions but overall Hg trends do not show consistent declines. For example, levels of Hg have either declined or remained stable in eggs from four seabird species collected in the Bay of Fundy since 1972. Quantitatively linking Hg exposures from fish harvested from the Gulf of Maine to human health risks is challenging at this time because no data are available on the geographic origin of seafood consumed by coastal residents. In addition, there is virtually no information on Hg levels in commercial species for offshore regions of the Gulf of Maine where some of the most productive fisheries are located. Both of these data gaps should be priorities for future research.

1. Introduction

High levels of methylmercury (MeHg) exposure through fish consumption causes a variety of adverse neurological and reproductive effects in humans and wildlife (Clarkson and Magos, 2006; Mahaffey et al., 2011; Scheuhammer et al., 2007). Coastal ecosystems provide foraging habitat for a variety of commercial and recreationally harvested fish species and through this pathway contribute substantially to human exposures to mercury (Hg). The Bay of Fundy/Gulf of Maine region, located on the east coasts of Canada and the United States, supports what has historically been some of the world's most productive fisheries and provides important habitat for abundant whales, porpoises, seals and many bird species (Hildebrand et al., 1997; Sinclair et al., 1991; Thompson, 2010). Like many coastal regions of the United States and Canada, high Hg levels have been documented in fish and wildlife from this area since measurements began in the 1970s (Gaskin et al., 1972; 1973; 1979). Although anthropogenic Hg emissions within North America have declined substantially and many large point sources eliminated, the effectiveness of these and other management actions at reducing risks to human and ecological health is still poorly understood.

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Here we use data from the Bay of Fundy/Gulf of Maine ecosystem to examine controls on Hg concentrations across a range of physical environments, industrial development and population densities. We use our synthesis to comment on the apparent effectiveness of various past and future management efforts for Hg and coastal water quality and to highlight gaps in monitoring data and research.

A key question often asked by environmental managers is how long will it take for an ecosystem to respond to changes in Hg inputs from human sources. Hg emissions have declined by over 90% in Canada since the 1970s and by over 60% in the United States since 1990 (Sunderland and Chmura, 2000a; 2000b; U.S. EPA, 2005). Coastal zones represent the transition between terrestrial environments and the ocean, and are often heavily influenced by watershed processes and human activities. Prior studies of freshwater ecosystems have shown that fish Hg levels often exhibit a rapid decline in response to decreased atmospheric deposition followed by a second, more gradual decline in response to reduced inputs from watershed sources (Harris et al., 2007; Knightes et al., 2009). Understanding the relative importance of direct atmospheric deposition, watershed inputs and other Hg sources to coastal ecosystems therefore provides insights on the expected temporal dynamics of Hg concentrations.

Here we review available information on total Hg and MeHg inputs to the Gulf of Maine from direct atmospheric deposition, wastewater and industry, rivers and oceanographic influences. Comparing input sources to measured concentrations in water, sediments and biota of different physioregions reveals observable associations between concentrations and inputs, and helps to identify the likely drivers of human and biological Hg exposures in the different regions of the Gulf. We use this analysis to highlight key uncertainties for future research initiatives and conclude with recommendations for future management aimed at further reducing Hg exposures of both humans and the marine ecosystem.

2. Major physioregions of the Gulf of Maine

The Gulf of Maine includes twelve major physioregions bounded inland by the Canadian provinces Nova Scotia and New Brunswick and the states of Maine, New Hampshire and Massachusetts, USA. It is frequently referred to as a “sea within a sea” because the only exchange with the North Atlantic is through the Northeast Channel and the mid-shelf channel entering the Scotian Shelf (Fig. 1). Around 60% of the surface area and volume of the Gulf of Maine is found offshore in the central Gulf of Maine and Georges Bank (Table 1). Offshore regions do not receive any direct freshwater or point source Hg inputs from the watershed although freshwater inflows do exert a major influence on near surface waters and circulation (Geyer et al., 2004; Hetland and Signell, 2005). During spring freshet in particular, the St. John River and rivers along the Northern Coastal Shelf (Androscoggin, Penobscot, Merrimack and Kennebec Rivers) contribute substantially to the upper 40 m of the water column and maintain the counterclockwise circulation of the Gulf (Xue et al., 2000).

The most productive finfish fisheries and biological foraging grounds of the Gulf of Maine are found in offshore environments (Thompson, 2010), although the lucrative lobster fisheries are predominantly inshore in Maine and Nova Scotia. Both surface and deeper water in the offshore regions are heavily influenced by surface inflows from the Scotian Shelf and deeper, nutrient rich flows through the Northeast Channel (Sutcliffe et al., 1976; Townsend et al., 2010; Xue et al., 2000). The Scotian Shelf and Northeast Channel inflows are, in turn, influenced by a combination of relatively fresh Labrador Sea Water and freshwater

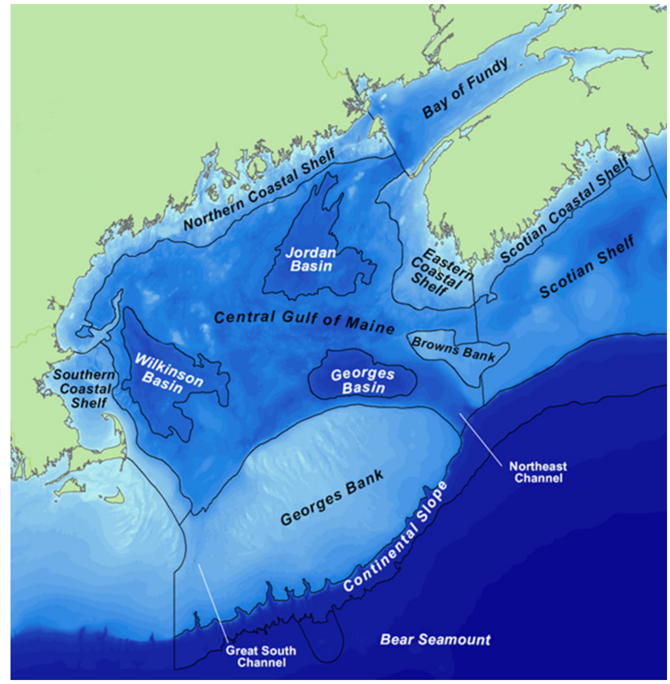

Fig. 1. Map of study region located on the east coast of Canada and the United States (from: http://www.gommea.org/about-the-gulf/physical-characteristics/physioregions/). Note that the Bay of Fundy is part of the Gulf of Maine ecosystem.

discharges from the St. Lawrence River Basin (Sutcliffe et al., 1976; Townsend et al., 2010). Primary productivity is elevated in the offshore Georges Bank region due to upwelling of nutrient rich waters reaching 470 g C m^{-2} $year^{-1}$ compared to an average of 290 g C m^{-2} $year^{-1}$ for the rest of the region (Hameedi et al., 2002; Smith et al., 1984).

The nearshore coastal regions of the Gulf of Maine include the Bay of Fundy and the Eastern, Northern and Southern Coastal Shelves (Fig. 1). Water circulation patterns in these regions are highly influenced by freshwater discharges from Gulf of Maine watershed (Geyer et al., 2004; Xue et al., 2000). For example, large freshwater discharges into the Bay of Fundy from the St. John River travel south in a coastal current in the eastern Gulf of Maine, which is eventually deflected offshore near the entrance to Penobscot Bay (Geyer et al., 2004). The dominant coastal current (Western Maine Coastal Current) flows southwestward around the perimeter of the Gulf of Maine (Geyer et al., 2004).

In the nearshore coastal environment, Hg is input directly through sewage and industrial effluent outfalls along the coast as well as from point watershed sources along rivers and streams. These terrestrial sources impact chemical dynamics of the inshore more than the offshore areas. A large gradient in human population density across these coastal areas also influences the extent of anthropogenic Hg inputs. Most urban development is concentrated around the city of Boston in the Southern Coastal Shelf region (Thompson, 2010).

Hg dynamics are also influenced by large-scale patterns in sediment transport and deposition. Fader et al. (1977) estimated that significant sediment accumulation occurs in less than 30% of the surface area of the Gulf of Maine. Deposition regions with elevated clay-silt content and higher levels of organic matter tend to have higher Hg concentrations (Loring, 1979, 1982). Surface sediments of offshore regions are most commonly defined as gravel, sand and mud (Kelley and Belknap, 1991). Concentrations of Hg in sandy sediments of these dynamic erosional areas of the Bay of Fundy tend to be near background concentrations in geological materials ($\sim$15–20 ppb) (Loring, 1979, 1982).