



Atmospheric mercury and fine particulate matter in coastal New England: Implications for mercury and trace element sources in the northeastern United States



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HIGHLIGHTS

- Contribution of natural and anthropogenic PM_{2.5} sources is quantified.
- Back-trajectories for marine and crustal sources each show input from offshore.
- Results for crustal source are explained by long-range transport of Saharan dust.
- Results for Hg species, SO₂ and NO_x show influence of combustion-related plumes.
- Results for RGM show diurnal cycle of production characteristic of marine influence.

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ABSTRACT

Intensive sampling of ambient atmospheric fine particulate matter was conducted at Woods Hole, Massachusetts over a four-month period from 3 April to 29 July, 2008, in conjunction with year-long deployment of the USGS Mobile Mercury Lab. Results were obtained for trace elements in fine particulate matter concurrently with determination of ambient atmospheric mercury speciation and concentrations of ancillary gasses (SO₂, NO_x, and O₃). For particulate matter, trace element enrichment factors greater than 10 relative to crustal background values were found for As, Bi, Cd, Cu, Hg, Pb, Sb, V, and Zn, indicating contribution of these elements by anthropogenic sources. For other elements, enrichments are consistent with natural marine (Na, Ca, Mg, Sr) or crustal (Ba, Ce, Co, Cs, Fe, Ga, La, Rb, Sc, Th, Ti, U, Y) sources, respectively. Positive matrix factorization was used together with concentration weighted air-mass back trajectories to better define element sources and their locations. Our analysis, based on events exhibiting the 10% highest PM_{2.5} contributions for each source category, identifies coal-fired power stations concentrated in the U.S. Ohio Valley, metal smelting in eastern Canada, and marine and crustal sources showing surprisingly similar back trajectories, at times each sampling Atlantic coastal airsheds. This pattern is consistent with contribution of Saharan dust by a summer maximum at the latitude of Florida and northward transport up the Atlantic Coast by clockwise circulation of the summer Bermuda High. Results for mercury speciation show diurnal production of RGM by photochemical oxidation of Hg⁰ in a marine environment, and periodic traverse of the study area by correlated RGM-SO₂(NO_x) plumes, indicative of coal combustion sources.

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1. Introduction

Proximity of urbanized areas to coastal ecosystems provides a ready source for atmospheric transport of mercury (Hg) and other contaminants. Where element enrichments in atmospheric aerosols greatly exceed proportions expected in natural crustal or

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marine background inputs, anthropogenic sources are indicated (Duce et al., 1975; Pacyna and Pacyna, 2001; Song et al., 2001). In the absence of anthropogenic inputs, coastal areas are primarily influenced by marine aerosols enriched in constituents that are concentrated in seawater, such as Na, Mg, and Sr, and terrigenous aerosols characterized by lithophile elements that are enriched in the Earth's crust, such as Al, Zr and the rare earth elements (REE).

In the present study, we collected a nearly four month (03 April to 29 July, 2008) sequence of atmospheric fine particulate matter (PM_{2.5}) at a coastal site on the Quissett Campus of Woods Hole Oceanographic Institution (WHOI), at the southwestern extent of Cape Cod, Massachusetts, USA (Fig. 1). We use positive matrix factorization (PMF), a data analysis approach, together with concentration weighted trajectory analysis (CWT), an air-mass trajectory approach, to understand the sources controlling PM_{2.5} chemistry and their geographic distribution. The study area is residential to rural, with no large emission sources within 50 km of the site (2008 Hg emissions only 6.4 kg yr⁻¹), and none within 10 km (Engle et al., 2010a). However, the area is located within 100 km of the urban centers of Boston, Massachusetts and Providence, Rhode Island, and within about 275 km of the New York City metropolitan area (Fig. 1). The proximity of these northeastern urban centers influences air quality on Cape Cod, including ozone levels, especially during the summer months (Massachusetts Department of the Environment, 2012; U.S. EPA, 2012b; Cooper et al., 2012).

Previous sampling for PM_{2.5} chemistry at coastal sites in the eastern U.S. includes the New York–New Jersey Harbor estuary (Gao et al., 2002), and Brigantine, New Jersey, along the southern New Jersey coast (Song et al., 2001) together with two inland sites. These studies identify source categories including fossil energy, marine, soil/crustal, and metal smelting, as well as waste incineration, whose impact has been reduced by subsequent EPA regulations. Golomb et al. (1997) compared trace element concentrations in wet and dry deposition over a one-year interval at Nahant, Massachusetts, near Boston, with that at Truro, a more rural site near the northern extent of Cape Cod. These authors found that

although metal concentrations in dry deposition were generally higher at Nahant, annual wet deposition for elements such as Se, Co, Sb, Zn, Pb, etc. was higher at Truro. Golomb et al. considered this to be due to rainout of metals from air masses that had traversed the urban corridor to the southwest that includes New York City.

For Hg speciation, recent work provides multiyear data for two sites in southern New Hampshire that should be comparable to the WHOI site. These include a coastal site at Thompson Farm, and an offshore site at Appledore Island (Mao and Talbot, 2012; Mao et al., 2012; Sigler et al., 2009a,b). These studies show differences in Hg speciation between the two coastal/offshore sites and an upland site inland. The two marine-influenced sites show a diurnal cycle of reactive gaseous mercury (RGM) production characteristic of sites having a marine influence (Laurier and Mason, 2007; Engle et al., 2008).

The present study is one of a series of coastal studies conducted by the U.S. Geological Survey between 2005 and 2010, aimed at understanding controls on atmospheric Hg deposition in coastal environments (Kolker et al., 2007; Engle et al., 2008, 2010a), and ultimately, Hg uptake and availability in coastal ecosystems (Engle et al., 2010b, Geboy et al., 2011; Fig. 1). A summary of seasonal variation in Hg speciation at the WHOI site is given by Engle et al. (2010a), in comparison with eight other sites in central and eastern North America. Among coastal sites studied by the USGS, the WHOI site is the first of two more northerly sites (Acadia National Park, Maine is the other), sampled to determine the effect of climate on Hg speciation, in comparison with warmer water coastal sites at Weeks Bay National Estuarine Reserve, Alabama, Cape Romain, South Carolina, and Luquillo National Forest, Puerto Rico (Engle et al., 2008, 2010a).

2. Methods

2.1. Sampling protocols

Atmospheric aerosol samplers were deployed above tree-top level on a permanent observation tower with open exposure to the ocean less than 200 m from the shore. The bulk of this study is

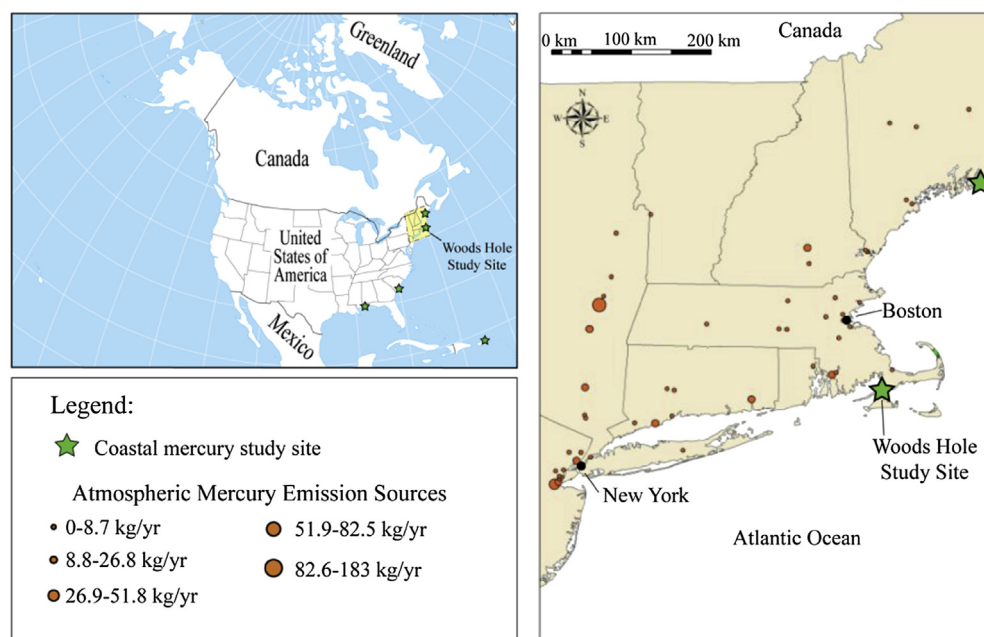


Fig. 1. Study location map showing locations of major emission sources for atmospheric mercury as of 2008 and location of coastal sites investigated for atmospheric mercury and particulate matter in a multi-year collaborative USGS study. Atmospheric mercury emission data is taken from the U.S. EPA (2012a) Toxic Release Inventory (<http://www.epa.gov/triexplorer/>).

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