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Dry deposition of gaseous oxidized mercury in Western Maryland

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ABSTRACT

The purpose of this study was to directly measure the dry deposition of gaseous oxidized mercury (GOM) in western Maryland. Annual estimates were made using passive ion-exchange surrogate surfaces and a resistance model. Surrogate surfaces were deployed for seventeen weekly sampling periods between September 2009 and October 2010. Dry deposition rates from surrogate surfaces ranged from 80 to 1512 pg m^{-2} h^{-1}. GOM dry deposition rates were strongly correlated ($r^2 = 0.75$) with the weekly average atmospheric GOM concentrations, which ranged from 2.3 to 34.1 pg m^{-3}. Dry deposition of GOM could be predicted from the ambient air concentrations of GOM using this equation: GOM dry deposition (pg m^{-2} h^{-1}) = 43.2 × GOM concentration − 80.3. Dry deposition velocities computed using GOM concentrations and surrogate surface GOM dry deposition rates, ranged from 0.2 to 1.7 cm s^{-1}. Modeled dry deposition rates were highly correlated ($r^2 = 0.80$) with surrogate surface dry deposition rates. Using the overall weekly average surrogate surface dry deposition rate (369 ± 340 pg m^{-2} h^{-1}), we estimated an annual GOM dry deposition rate of 3.2 μg m^{-2} $year^{-1}$. Using the resistance model, we estimated an annual GOM dry deposition rate of 3.5 μg m^{-2} $year^{-1}$. Our annual GOM dry deposition rates were similar to the dry deposition (3.3 μg m^{-2} h^{-1}) of gaseous elemental mercury (GEM) at our site. In addition, annual GOM dry deposition was approximately 1/2 of the average annual wet deposition of total mercury (7.7 ± 1.9 μg m^{-2} $year^{-1}$) at our site. Total annual mercury deposition from dry deposition of GOM and GEM and wet deposition was approximately 14.4 μg m^{-2} $year^{-1}$, which was similar to the average annual litterfall deposition (15 ± 2.1 μg m^{-2} $year^{-1}$) of mercury, which was also measured at our site.

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1. Introduction

Mercury is a neurotoxin that can affect human health. Human exposure to mercury occurs primarily through the consumption of contaminated fish. All fifty states in the United States had fish consumption advisories for mercury in 2008 (EPA, 2009). The mercury in these fish enters aquatic ecosystems primarily from wet and dry deposition. Wet deposition transfers soluble forms of mercury from the atmosphere to ecosystems. Dry deposition can remove particulate and gaseous forms of mercury from the atmosphere. The relative importance of wet and dry deposition is related to the atmospheric mercury species, characteristics of the deposition site, and proximity to anthropogenic sources.

Atmospheric mercury species include gaseous elemental mercury (GEM), gaseous oxidized mercury (GOM), and particulate bound mercury (PBM). GOM represents a variety of oxidized mercury compounds (e.g. $HgCl_2$, $HgBr_2$), which, away from active volcanoes and forest fires, are produced primarily by human activities, such as coal combustion (Lohman et al., 2006; Gustin and Jaffe, 2010). In addition, GOM in the troposphere may also be produced from the oxidization of GEM and the atmospheric transport of GOM from the free troposphere (Swartzendruber et al., 2006; Weiss-Penzias et al., 2009). Typically, ambient air concentrations of GOM in the troposphere are around 10 pg m^{-3}, but can be greater than 200 pg m^{-3} downwind of sources (Gabriel et al., 2005; Velente et al., 2007; Engle et al., 2008, 2010; Lyman and Gustin, 2008).

GOM is highly reactive, water soluble, and, can be removed from the atmosphere relatively quickly by both wet and dry deposition. Dry deposition of GOM is rapid compared to the dry deposition of other atmospheric mercury species. For example, dry deposition velocities for GOM (0.5 to up 6 cm s^{-1}) are faster than dry deposition velocities for GEM (0 to 0.4 cm s^{-1}) and particulate bound mercury with diameters less than 2.5 μm ($PBM_{2.5}$) (0.02 to 2 cm s^{-1}) (Zhang et al., 2009). In addition, once atmospheric GOM enters terrestrial and aquatic ecosystems, it may be more easily converted to methyl mercury than GEM and $PBM_{2.5}$ (Grigal, 2002). This is important

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because methyl mercury can bioaccumulate in aquatic and terrestrial food chains.

Recently, resistance and transport models have been used to predict dry deposition of atmospheric mercury species (Engle et al., 2010; Zhang et al., 2011). For example, Engle et al. (2010) used atmospheric concentrations of GOM and $PBM_{2.5}$ and a resistance model to predict GOM and $PBM_{2.5}$ dry deposition. Their results suggest that dry deposition of GOM accounted for more than 90% of the total dry deposition of these two mercury species. In addition, dry deposition of GOM was strongly influenced by land-use and meteorology. For example, water surfaces had the lowest dry deposition velocities (0.5 to 1.8 cm s^{-1}) and urban areas had the highest dry deposition velocities (0.9 to 5 cm s^{-1}). Also, study sites with the highest wind speeds had higher GOM dry deposition velocities.

A variety of aqueous and solid surfaces have also been evaluated for their effectiveness to measure dry deposition of atmospheric mercury species (Lyman et al., 2009; Lai et al., 2011). For example, Lyman et al. (2009) described a solid surrogate surface approach to estimate dry deposition of GOM. This method uses ion exchange membranes attached to aerodynamic filter holders, which passively collect GOM in ambient air. After field exposure for up to one week, these surrogate surfaces were analyzed using cold vapor atomic fluorescence spectroscopy (CVAFS). To date, however, this approach has been used at only a few locations (Lyman et al., 2007, 2009). In addition, dry deposition to these surrogate surfaces may not accurately reflect dry deposition to heterogeneous natural surfaces. However, this approach may accurately describe temporal and spatial variations of GOM dry deposition.

In June 2005, we started measuring ambient air concentrations of GEM, GOM and $PBM_{2.5}$ in Garrett County, Maryland. These data have been used to estimate dry deposition using several different models and approaches. Many of our estimates suggest that the dry deposition of GOM may be several times greater than wet deposition of mercury. Note, however, that model predictions help us understand atmospheric deposition processes, but their accuracy needs to be verified by direct field measurements of GOM dry deposition. Thus, the purpose of this study was to directly measure GOM dry deposition for comparison with model estimates of GOM dry deposition.

2. Methods

2.1. Study site

This project was conducted at the Piney Creek Reservoir (39° 42′ 21″ N, 79° 00′ 43″ W) in Garrett County, Maryland (Fig. 1). This site is in several National Atmospheric Monitoring Programs: the National Atmospheric Deposition Program's (NADP) National Trends Network (NTN), Mercury Deposition Network (MDN), Atmospheric Mercury Network (AMNet), the United States Forest Service Interagency Monitoring of Protected Visual Environments (IMPROVE) network and the Northeastern States for Coordinated Air Use Management's camera network (CAMNET) program.

Our study site is on a flat ridge top (elevation 869 m) that was selectively harvested in the winter of 2004. This site is surrounded by agricultural and forest lands. The shortest distance to the adjacent forest is about 250 m east of our monitoring station. During each growing season, all vegetation is cut to ground level within 100 m of our monitoring equipment. The rest of the vegetation up to the forest line is cut in the winter. The dominant ground cover consists of perennial vegetation, such as orchard grass (*Dactylis glomerata*) with a mix of thorns (*Rubus spp.*), flowers (*Aster spp.*), and Virginia creeper (*Parthenocissus quinquefolin).* There are also a few scattered black locust (*Robinia pseudoacacia*), black cherry (*Prunis serotina*) and sugar maple (*Acer saccharum*) trees.

The predominant wind directions are from the west and northwest. In the summer, however, winds also arrive from the south and southeast. Some of the largest mercury sources in the United States are located west and northwest of our site (Fig. 1). For example, the Keystone power plant in Armstrong County, Pennsylvania released 455 kg of mercury into the atmosphere in 2008. This power plant

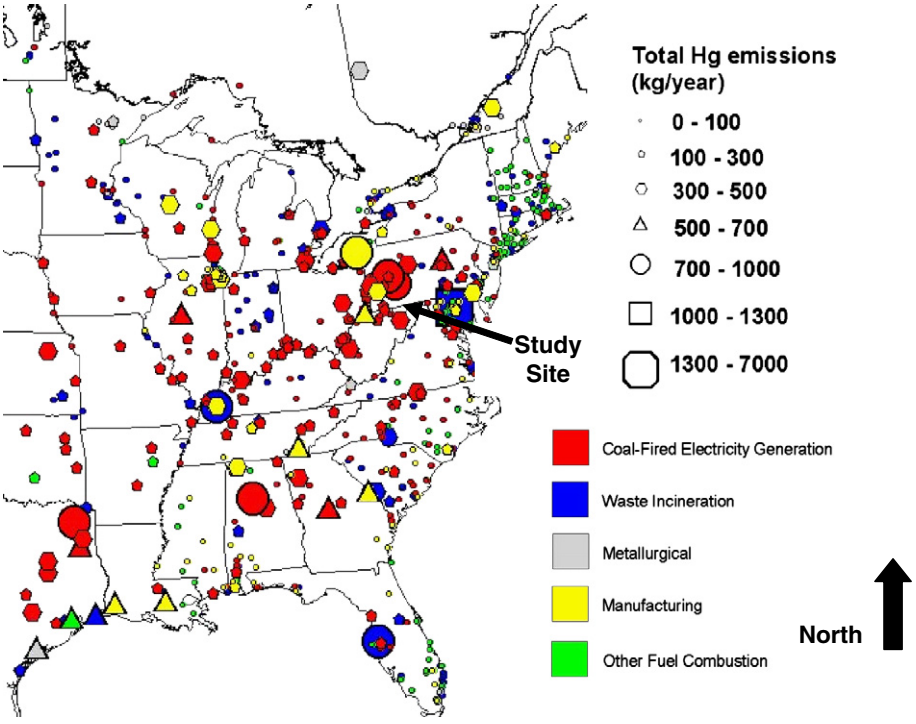

Fig. 1. Mercury emissions sources in the eastern United States.
This figure was provided by Dr. Mark Cohen.

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