



# Mercury wet deposition and speciated mercury air concentrations at rural and urban sites across New York state: Temporal patterns, sources and scavenging coefficients

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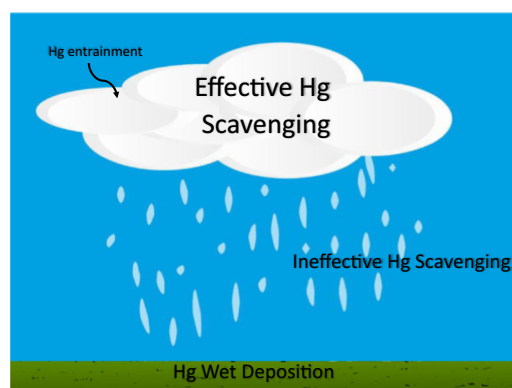
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## HIGHLIGHTS

- Speciated Hg depends on emissions, photochemical processes, and meteorology.
- Scavenging of oxidized Hg was higher than particle-bound Hg at all 3 sites.
- Similar GOM and PBM values pre and post-precipitation show cloud scavenging prevails.
- Regional sources were important for urban Hg; local sources for the rural site.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Measurements of ambient speciated mercury (Hg) concentrations and Hg wet deposition were made at two urban sites (Bronx, NY and Rochester, NY) and one rural site (Potsdam, NY) in New York State in 2013 and 2014 to: 1) determine the factors influencing Hg wet deposition concentrations, 2) identify the contribution of gaseous oxidized Hg (GOM) and particulate bound Hg (PBM) scavenging to Hg wet deposition concentrations, and 3) identify potential source areas associated with high concentration events. The Bronx had the highest mean gaseous elemental Hg (GEM) and GOM concentrations, Rochester had the highest mean PBM and the lowest GOM concentrations, and Potsdam had the lowest mean GEM and PBM concentrations. The annual volume weighted mean (VWM) Hg concentrations and Hg wet deposition fluxes in the Bronx, Rochester, and Potsdam were significantly different with mean values of  $10.3 \pm 8.16$ ,  $10.2 \pm 9.06$ , and  $5.07 \pm 1.79$   $\text{ngL}^{-1}$  and  $8.45 \pm 0.64$ ,  $6.65 \pm 0.21$ , and  $5.25 \pm 0.49$   $\mu\text{g}/\text{m}^2 \text{ year}^{-1}$ , respectively. Hg wet deposition flux and precipitation depth were positively correlated at all three sites as were Hg concentration in precipitation and weekly GOM concentrations at the Bronx and Potsdam sites. Scavenging coefficients (SC) of 680, 630, 850 for GOM and 410, 320, and 410 for PBM at Bronx, Rochester, and Potsdam, respectively, suggest GOM is responsible for most of the scavenged Hg. Measured GOM and PBM concentrations were relatively constant before precipitation events and Hg concentrations in precipitation did not vary significantly during precipitation events implying the scavenging process mainly occurred in clouds. VWM Hg concentrations, monthly accumulated Hg flux, and SCs for GOM

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and PBM were higher at the urban sites and significantly different for non-snow and snow events. Local sources appeared more important at the rural site while regional sources affected high urban concentrations.

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## 1. Introduction

Mercury (Hg) is an important global pollutant and was classified as a hazardous chemical by the U.S. EPA in 1997 because it is persistent, bio-accumulative, and toxic (Keating et al., 1997). Concerns about Hg are primarily due to adverse human health effects caused from the consumption of fish which contain Hg (UNEP, 2013). Hg deposition is a major source of Hg to aquatic ecosystems where it can be transformed to methyl Hg, the form of Hg that readily bioaccumulates (Lindberg et al., 2007). Therefore, transport of Hg in the atmosphere has been the focus of considerable research in recent years (Ariya et al., 2015; Lin et al., 2006; Pirrone and Mason, 2009).

Natural and anthropogenic sources exist for atmospheric Hg (Lamborg et al., 2002; Seigneur et al., 2004). Natural Hg sources include volcanic eruptions, geothermal vents, wildfires, and emissions from historically deposited Hg from land and sea surfaces (Pirrone et al., 2009). The major sources of anthropogenic Hg are fossil-fuel power plants, metal manufacturing facilities, chemical plants, and waste incinerators (Pirrone et al., 2010). Song et al. (2015) suggested that primary anthropogenic Hg contributes up to 23% of present-day atmospheric deposition. Hg control policies, both legally binding and voluntary actions, have been adopted on regional, national, and global scales resulting in decreases in deposition and exposure of humans to Hg in fish in some locations (Driscoll et al., 2013). Emissions of Hg in the United States have decreased recently due to a series of Hg control policies and economic factors. However, emissions in other regions such as Asia have increased primarily because of their increasing fossil fuel combustion (Pacyna et al., 2003; Li et al., 2017).

There are three major Hg species in the atmosphere: gaseous elemental Hg (GEM), gaseous oxidized Hg (GOM), and particle-bound Hg (PBM) (Schroeder and Munthe, 1998). GEM is the dominant Hg species representing around 95% of total atmospheric Hg. It has a long atmospheric lifetime (months) and can be transported over long distances (Choi et al., 2008a, b; Amos et al., 2012; Amouroux et al., 1999). It has low reactivity and low water solubility, so it is not efficiently incorporated into wet deposition. A significant portion of Hg in wet deposition originates from the global transport of elemental Hg followed by chemical conversion to its divalent form, GOM (Dastoor and Larocque, 2004). GOM is water soluble with relatively strong surface adhesion properties (Han et al., 2005). It can be scavenged by rain within and below precipitating clouds (Lin and Pehkonen, 1999). PBM can also be wet deposited relatively efficiently if it is in or below precipitating clouds (Cohen et al., 2016). GOM and PBM account for only a small percentage of ambient Hg in the atmosphere (Landis et al., 2004), but are the predominant forms of Hg removed from the atmosphere in wet deposition (Seo et al., 2012).

Wet deposition is the main Hg pathway to aquatic systems, either as direct deposition to the water surface or as indirect deposition in runoff from the watershed (Mason, 2009; Pirrone et al., 2010; Risch et al., 2012). Hg in wet deposition are measured at Hg Deposition Network (MDN) sites across North America. At these sites, total Hg wet deposition fluxes range from 2.72 to 21.1  $\mu\text{g m}^{-2} \text{yr}^{-1}$  (NADP, 2014). Weiss-Penzias et al. (2016) analyzed MDN data and found that from 1997 to 2013, most of the 19 sites for which data were available for this entire period had significant negative trends in Hg concentration in wet deposition, especially in the northeastern U.S. Mao et al. (2017) also investigated Hg wet deposition and VWM concentrations in rural Adirondack region sites during the past decade and also found declining trends.

Many studies have explored the mechanisms involved in Hg wet deposition in different regions (Keeler et al., 2006; Lai et al., 2007; Mason et al., 2000; Choi et al., 2008a, b; Marumoto and Matsuyama, 2014;

Civerolo et al., 2014). For example, Selin and Jacob (2008) reported that seasonal variations in scavenging coefficients (SCs) can be attributed to a combination of enhanced summertime oxidation of GEM and inefficient scavenging of GOM by snow. Both Hg concentrations in precipitation and wet deposition fluxes are higher in summer than in winter due to differences in precipitation type and atmospheric concentrations (Choi et al., 2008a, b; Prestbo and Gay, 2009). Hg wet deposition rates vary substantially and change drastically upon transformation (Ariya et al., 2015). Actual field measurements of GOM wet deposition velocity were reported from 1 to 4  $\text{cm s}^{-1}$ , but PBM has a much smaller deposition velocity (usually  $<0.1 \text{ cm s}^{-1}$ ) (Lin et al., 2006).

The scavenging coefficient (SC) is defined as a ratio of Hg concentration in precipitation (mass/volume water) and Hg in air (mass/volume air) as Eq. (1).

$$SC_{\text{Hg}} = \frac{C_{\text{Hg in precipitation}}}{C_{\text{Hg in air}}} \quad (1)$$

It can be used to investigate the relationship between species concentration in precipitation and water-soluble species or particle-bound species concentrations (McMahon and Tsoukalas, 1978). Research on Hg SC is limited, but the available data suggest that Hg SCs vary with locations and measurement strategy (event based, weekly, or bi-weekly). SC values for Hg have been reported to range from 100 to 5500 depending on the species (Harrison and Allen, 1991). For reactive Hg, SC values ranged from 1700 to 4500 in the eastern United States (Huang et al., 2013). Seo et al. (2012) performed a multiple linear regression analysis on GOM/PBM and Hg concentration in precipitation and found that GOM was more effectively scavenged by wet deposition than PBM ( $SC_{\text{GOM}} = 750$  and  $SC_{\text{PBM}} = 380$ ). Differences in SCs were likely due to differences in meteorological conditions, the forms of Hg in the atmosphere, and vertical concentration variations.

The objectives of this study were to compare temporal patterns, sources, and scavenging coefficients of Hg wet deposition at rural and urban sites in New York State and identify the factors that affect Hg wet deposition at these sites including the contribution of GOM and PBM to Hg wet deposition.

## 2. Methods

### 2.1. Site description

Data from two urban MDN sites: Bronx, NY06 (40.87N,  $-73.87\text{W}$ ), Rochester, NY43 (43.15N,  $-77.55\text{W}$ ), and one rural site: Potsdam (POT) (44.66N,  $-75.00\text{W}$ ) (rural) in New York State as shown in Fig. S1, were analyzed for this study. The Bronx site is located on the roof of a building in the north Bronx, surrounded by highways and mixed residential and commercial areas in the New York City metropolitan region which has a long manufacturing, petrochemical, and industrial legacy (Civerolo et al., 2014). The Rochester site is in western New York State, downwind of coal-fired electricity generation facilities in Dunkirk and Tonawanda, New York, and the Midwestern USA. Two interstate and one state highways are within 1 km of the site (Wang et al., 2013) as are the mainline tracks of a railroad. The New York State Department of Environment Conservation (NYSDEC) currently monitors hourly averaged ozone ( $\text{O}_3$ ), wind speed, wind direction, and ambient temperature at both sites (Choi et al., 2013; Civerolo et al., 2014). Because the Bronx and Rochester sites are in the MDN and the Atmospheric Hg Network (AMnet), weekly wet deposition measurements are available in addition to the continuous ambient Hg concentration measurements

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