



Sources of mercury in precipitation to Underhill, VT

Lynne E. Gratz*, Gerald J. Keeler

University of Michigan Air Quality Laboratory, 1415 Washtenaw Heights, SPH 1-Tower, Ann Arbor, MI 48109-2029, USA

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ABSTRACT

Daily event precipitation samples were collected from 1995 to 2006 in Underhill, VT, USA and analyzed for mercury (Hg) and trace elements. Over the 12-year period, annual Hg deposition levels at Underhill did not decline significantly despite regulatory efforts to reduce Hg emissions in the United States. Mercury and trace element wet deposition data were examined using the multivariate receptor model EPA PMF 3.0 to identify the sources contributing to Hg deposition at the Underhill site. Results indicate that coal combustion, a mixture of incineration and non-ferrous metal smelting, and a phosphorus source contributed to the observed Hg wet deposition. The model consistently indicated that coal combustion contributed ~60% of the total Hg measured in wet deposition and the contribution from this source did not change significantly over time. Mercury and trace element deposition were further analyzed using a hybrid-receptor model, quantitative transport bias analysis (QTBA), to identify the likely source locations contributing to Hg deposition. Results from QTBA indicate that the majority of Hg deposition at Underhill is due to transport from the Midwestern United States, where the density of coal-fired utility boilers in the U.S. is largest.

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1. Introduction

Mercury (Hg) is a persistent hazardous air pollutant and bioaccumulative neurotoxin. Atmospheric deposition is widely recognized as a dominant mechanism by which Hg enters terrestrial and aquatic ecosystems (Landis and Keeler, 2002; Hammerschmidt and Fitzgerald, 2006) where, upon conversion to the organic form methylmercury, it can bioaccumulate within the food chain (Schroeder and Munthe, 1998). Investigating atmospheric Hg emissions and transport pathways is critical for mitigating its environmental impact.

Mercury exists in the atmosphere predominantly as gaseous elemental Hg (Hg^0), fine particulate bound Hg (Hg_p), and divalent reactive gaseous Hg (RGM). Elemental mercury is relatively stable and able to travel long distances before being converted to the more soluble divalent forms (Hg_p and RGM; or $\text{Hg}(\text{II})$) that deposit readily (Schroeder and Munthe, 1998; Lin and Pehkonen, 1999). Consequently, Hg can be transported locally, regionally, and globally and Hg deposition at any location may represent a complex mixture of source emissions and atmospheric processes.

Underhill, VT, USA is a remote location where elevated levels of Hg in the region's fish and wildlife remain a critical issue

(Hammerschmidt and Fitzgerald, 2006; Evers et al., 2007) despite efforts to reduce regional Hg emissions. Previously, municipal and medical waste incinerators were considered one of the largest anthropogenic sources of atmospheric Hg; however, following the 1990 Clean Air Act Amendments the U.S. EPA imposed nationwide restrictions on waste incinerator stack emissions (U.S. EPA, 1997; NESCAUM, 2005). By 1999, the targeted 95% reduction in waste incinerator Hg emissions had largely been met (Cohen et al., 2007). In the northeast, where municipal waste incineration once accounted for ~55% of the estimated Northeast Regional Mercury Emissions Inventory (NEIWPPC, 2007), emissions from municipal and medical waste incinerators declined by ~70% between 1998 and 2002 (NESCAUM, 2005). Yet despite these regulatory successes, annual Hg wet deposition at Underhill did not decline significantly between 1995 and 2006 as transport from the industrialized Midwest and urban East Coast corridor consistently contributed to the largest observed Hg wet deposition amounts (Gratz et al., 2009). These findings suggest that additional regional emission sources contributed to Hg deposition at Underhill.

In fact, fossil fuel combustion remains the largest anthropogenic Hg source in the U.S. because emissions, such as those from coal-fired utility boilers (CFUBs), have not declined substantially over time (Cohen et al., 2007; Butler et al., 2008). CFUB emission reductions were anticipated under U.S. EPA's cap-and-trade approach proposed in the 2005 Clean Air Mercury Rule (Cohen et al., 2007). However, this rule was overturned in 2008 when

* Corresponding author. Present address: CNR-Institute of Atmospheric Pollution Research, c/o UNICAL-Polifunzionale, 87036 Rende, Italy. Tel.: +39 0984 493250.
E-mail addresses: lgratz@umich.edu, lgratz@iia.cnr.it (L.E. Gratz).

states and other parties encouraged the use of more stringent maximum achievable control technologies (MACT), as stipulated in the 1990 Clean Air Act Amendments. In light of these efforts to regulate Hg emissions and the observed temporal patterns in Hg wet deposition, the Underhill dataset presents a unique opportunity to study source contributions to Hg deposition over time.

Multivariate statistical receptor models are widely used in air quality studies to determine the sources of atmospheric constituents. Previous applications of positive matrix factorization (PMF) to aerosol concentrations at Underhill indicated that fossil fuel combustion, local wood smoke, and secondary sulfate production were the dominant sources of PM_{2.5} mass to the site (Polissar et al., 2001; Poirot et al., 2001; Gao et al., 2006). Municipal waste incineration was identified as a major source from 1988 to 1995 (Polissar et al., 2001), but could not be identified as a stand-alone source in 2001–2003, likely due to emission reductions (Gao et al., 2006). The application of receptor models to aerosol measurements at other U.S. Midwest and East Coast locations also demonstrated the regional influence of industrial sources (e.g. coal- and oil-fired utility boilers, iron–steel manufacturing) (Olmez et al., 1998; Liu et al., 2003; Morishita et al., 2006; Hammond et al., 2008). Given the pattern of Hg transport from the Midwest and East Coast (Gratz et al., 2009), wet deposition at Underhill is likely influenced by these regional sources.

The application of receptor models to precipitation data is complex given that the wet removal of atmospheric constituents may vary with aerosol size fraction, distance between source and receptor, meteorological conditions and atmospheric chemistry during transport. However, the chemical compositions and elemental ratios of many pollutants are reasonably well preserved during transport (Keeler and Samson, 1989; Rahn and Lowenthal, 1984) suggesting that statistical models can be applied to precipitation data as long as the atmospheric behavior

of measured species are considered. PMF has been successfully applied to precipitation data to determine the sources of Hg and other trace elements (Juntto and Paatero, 1994; Anttila et al., 1995; Keeler et al., 2006).

In this study we applied the multivariate statistical receptor model EPA PMF 3.0 to 12 years of Hg and trace element precipitation measurements to identify the sources contributing to Hg wet deposition at Underhill. Additionally we utilized a trajectory-based hybrid-receptor model, quantitative transport bias analysis (QTBA), to determine the locations of the identified source emissions.

2. Methodology

2.1. Sample collection and analysis

The Underhill site is ~25 km east of Lake Champlain on the west slope of Mount Mansfield (elevation 399 m) (Fig. 1). Daily event wet-only precipitation samples were collected for Hg and trace element analysis using a modified MIC-B (MIC, Thornhill, Ontario) automatic precipitation collector (Landis and Keeler, 1997). Separate sampling trains for Hg and trace elements (Keeler et al., 2005; Gratz et al., 2009) were prepared at the University of Michigan Air Quality Laboratory (UMAQL) (Landis and Keeler, 1997) to ensure that they were Hg- and trace element-free prior to use. Site operators replaced sampling trains after daily precipitation events. Precipitation events > 0.10 cm were used in the subsequent data analysis. The volume of each precipitation sample was determined gravimetrically. Precipitation depths were determined using the Hg funnel area (191 cm²).

Following collection, Hg samples were oxidized with concentrated BrCl to a 1% solution (v/v) and stored for at least 24 h in a dark cold room prior to Hg concentration analysis by cold-vapor atomic fluorescence spectrometry (CVAFS) (Landis and Keeler,

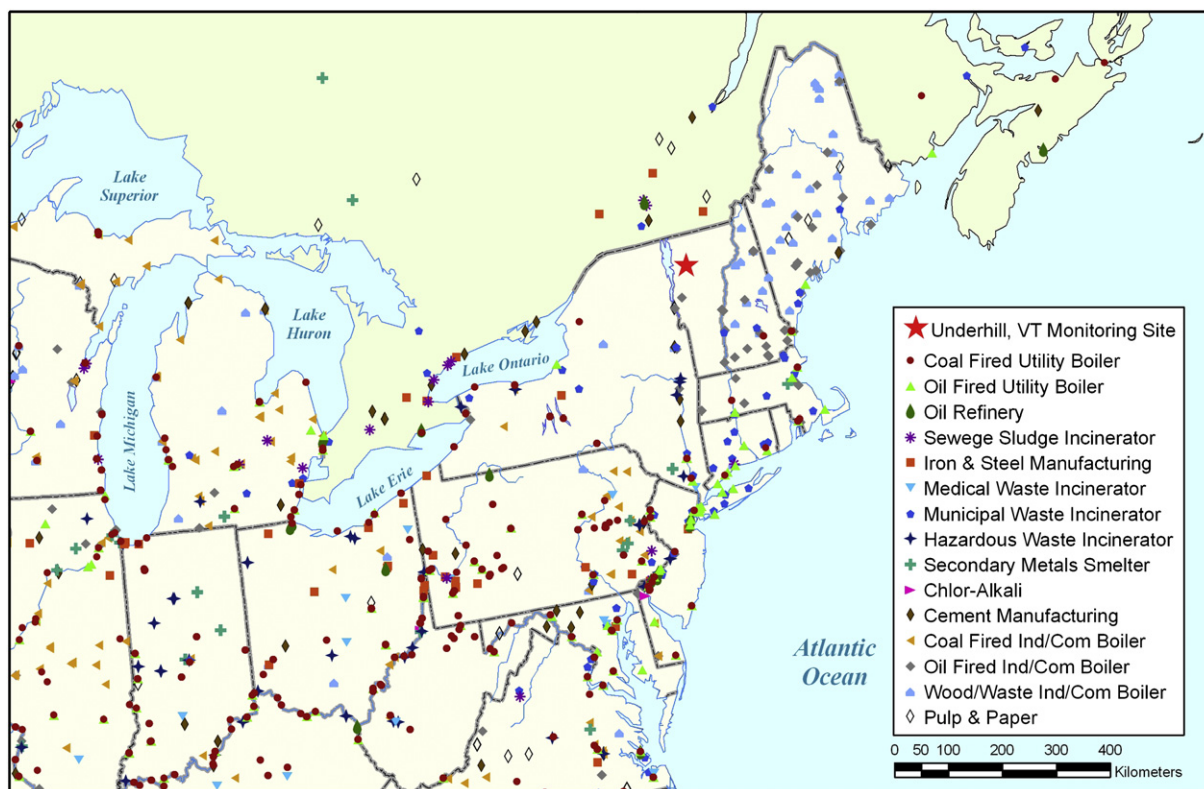


Fig. 1. Location of the Underhill, VT monitoring site and major mercury point sources emitting ≥ 0.1 kg yr⁻¹ (U.S. EPA, 2005; Environment Canada, 2007). U.S. sources correspond to the MACT source category.

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