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Investigating the impact of local urban sources on total atmospheric mercury wet deposition in Cleveland, Ohio, USA



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HIGHLIGHTS

- Total Hg event wet deposition was sampled at a rural and urban site in Cleveland, OH.
- In the 18-month period the urban site received 37% more Total Hg wet deposition.
- Concurrent events delivered 2.4 times more Total Hg wet deposition to the urban site.
- The increased 2010 urban site deposition may reflect local post-recession emissions.

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ABSTRACT

Event-based precipitation samples were collected at a downtown industrial and a predominantly up-wind rural location in the Cleveland, Ohio metropolitan area from July 2009 through December 2010 to investigate the potential local total mercury (Hg) wet deposition enhancement in a region with a high concentration of coal combustion sources. Total Hg wet deposition for the 18-month period was $6.8 \mu\text{g m}^{-2}$ ($n = 81$) at the rural site and $10.7 \mu\text{g m}^{-2}$ ($n = 98$) at the urban site demonstrating a significant ($p = 0.046$) 37% enhancement in deposition between the two sites. Large deposition events ($>0.2 \mu\text{g m}^{-2}$) occurred predominantly from May through October ($n = 16$ (urban), $n = 10$ (rural)) and represented from 2 to 8% of total 18-month deposition per event. At the downtown urban site, the average Hg precipitation concentration was 53% higher for these large deposition events. Concurrently measured precipitation events delivered in aggregate 2.4 times more total Hg wet deposition to the urban site compared to the rural site. Hg rainfall concentrations for concurrent events with similar precipitation depth were 2–4 times higher at the urban site and suggest scavenging of local Hg emissions. Further evaluation of these events revealed 83% more total Hg deposition at the urban site from January to December 2010 compared to July to December 2009, while there was 26% more at the rural site during these same time periods. The larger increase in deposition at the urban site in 2010 may be evidence of increased local emissions from sources that were known to be offline during this study period because of an economic recession.

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1. Introduction

Bulk atmospheric wet deposition of total mercury (Hg) in the US is currently monitored by the U.S. National Atmospheric Deposition

Program Mercury Deposition Network, MDN (NADP, 2015a). This network is comprised of sites where Hg wet deposition is primarily collected on a weekly basis in rural areas that are considered to be regionally representative. Data obtained from this network is utilized to provide national scale spatially interpolated Hg deposition maps which provide important information to stakeholders in thirty seven states and three Canadian provinces (NADP, 2015b). However, results from several long-term Hg event wet deposition studies, especially in the Great Lakes region reveal that gradients in deposition exist with enhancement in Hg concentration between

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urban/industrial and rural sites (Hoyer et al., 1995; Landis et al., 2002; White, 2009; White et al., 2009, 2013). This highlights the importance of investigating atmospheric deposition from local sources in urban and industrial regions, especially when in close proximity to sensitive aquatic ecosystems.

Cuyahoga County, Ohio which constitutes the majority of the Cleveland metropolitan area is currently designated by the U.S. Environmental Protection Agency (USEPA) as nonattainment for the national ambient air quality standards (NAAQS) annual fine particulate matter (PM_{2.5}) standard as a result of local and regional air pollution contributions (U.S. EPA, 2015). Cleveland is also located in the Lake Erie watershed which contains a high concentration of coal-fired power plants and is the 3rd largest atmospheric Hg emissions source area in Ohio. (USEPA, NEI). In 2010 the USEPA estimated that the three largest emitters of Hg in the Great Lakes region were the states of Ohio, Pennsylvania and Indiana (NDRC, 2012). Thus, emissions from large regional sources will also contribute to Hg deposition in Ohio.

The objective of this study was to evaluate the existence and magnitude of any Hg wet deposition enhancement at a downtown Cleveland urban/industrial location versus a predominantly upwind background location 52 km to the southwest during an 18-month study. Wet deposition samples were collected using an Automated Sequential Precipitation Sampler, ASPS, which facilitates collection of unattended wet-only event-precipitation samples (White et al., 2013). Collection of event samples rather than aggregated samples (e.g. weekly, monthly) provides higher resolution data thereby capturing temporal and spatial variability that is potentially lost in integrated samples (Landis and Keeler, 2002; White et al., 2009).

2. Materials and methods

2.1. Study sites

Wet deposition of Hg was monitored on an event basis for 18 months (July 2009 to December 2010) at the George T. Craig site (GTC; 41° 29' 31.35" N, 81° 40' 42.64" W) in urban Cleveland, OH (Cuyahoga County) and at the Chippewa Lake, OH (LCM; 41° 3' 37.35" N, 81° 55' 26.17" W) rural site (Medina County) during the USEPA Cleveland Multiple Air Pollutant Study (CMAPS; Norris et al., 2009). The GTC site is located at the corner of 14th Street and Orange Avenue just south of downtown Cleveland, near the junction of two main interstate freeways (I-90 and I-77). It is immediately downwind of the industrial valley location, in an area between the outer boundary of the city and the Industrial valley (Ohio Environmental Protection Agency, 2015). The GTC site was an existing City of Cleveland Department of Air Quality ambient air pollution monitoring location that was part of the USEPA State or Local Air Monitoring Stations Network (EPA SLAMS), the National Core Multipollutant Monitoring Network (USEPA NCore), and the Chemical Speciation Network (USEPA CSN). The LCM site was located to the southwest of the greater Cleveland metropolitan area and was established by USEPA for CMAPS to represent an upwind regional background site and was subsequently integrated into the EPA SLAMS network. Fig. 1 depicts the GTC and LCM site locations as well as the major surrounding Hg emission sources.

2.2. Wet deposition sampling and analysis techniques

Wet deposition samples were collected using Automated Sequential Precipitation Samplers (ASPS), which are modified computer-controlled versions of our standard MIC-B wet-only precipitation collectors (Landis and Keeler, 1997). The ASPS are designed to facilitate automated unattended wet-only event

precipitation collection by the sequencing of samples into multiple collection bottles (White et al., 2013). The ASPS were configured for this study with a Hg sampling train consisting of a borosilicate glass funnel coupled to a rack of eight 1 L fluorinated ethylene propylene, (FEP) Teflon® collection bottles with 1 cm O.D. C-Flex™ tubing. The internal cabinet temperatures of the samplers were automatically controlled to provide heat in the winter to melt snow and ventilated in the summer to assist in keeping samples cool. The ASPS were programmed to collect 8 daily event precipitation samples without operator intervention. Precipitation volumes and temperatures were recorded continuously.

Field and analytical supplies for use in sampling of Hg wet deposition were pre-cleaned using a rigorous 11-day acid-cleaning procedure prior to their use in the field (Landis and Keeler, 1997). The FEP sample bottle cleaning procedure was slightly modified to lower bottle blanks and consisted of an additional 24-h bromine monochloride (BrCl; 1% v/v) soak at room temperature. The bottles were then emptied of BrCl and rinsed 5 times with ASTM Type I laboratory deionized water (18.2 MΩ cm; hereinafter milliQ). Twenty mLs of 0.8% (v/v) hydrochloric acid (HCl), preservative was added to each sample bottle (Landis and Keeler, 1997). Bottles containing acid preservative were capped and sealed with Teflon tape and triple bagged in preparation for transportation to the field.

Field site personnel utilized clean handling techniques at all times when deploying and retrieving sample bottles and funnels such as wearing powder-free vinyl gloves and standing downwind to minimize contamination (Landis and Keeler, 1997). Samples were retrieved by field operators, capped, sealed with Teflon tape, and triple bagged to prevent contamination during handling and transportation. Used funnels and tubing were replaced with clean supplies and shipped with samples to the University of Michigan Air Quality Laboratory (UMAQL). Field operators documented observations including precipitation event type (rain, mix, snow), times, and dates on tracking forms which were returned with the samples.

Upon receipt at the UMAQL Hg samples were immediately oxidized in a class 100 clean room with concentrated BrCl to a 1% v/v solution, and were stored in a dark cold room (at 4 °C) for at least 24 h prior to analysis. Samples were analyzed within 7 days for total Hg using a Brooks Rand Instruments (Seattle, WA) Model 2 cold vapor atomic fluorescence spectroscopy (CVAFS) system (Landis et al., 2002). The method detection limit (MDL) was determined to be 0.3 ng L⁻¹ using EPA method 200.8 (U.S. EPA, 1994). The frequency of replicate sample analysis was 20% and mean precision (in absolute % deviation) for these samples was 5.5 ± 4.5%. The mean absolute deviation for control standards was 3.0 ± 7.3%. The volume of each precipitation sample was determined gravimetrically and precipitation depth was determined by dividing the volume of precipitation by the collection area of the funnel.

2.3. Statistical analysis

Data processing and all statistical analyses were performed using SAS v.9.4 (SAS Institute, Cary, NC). Parametric statistics used in this analysis include: a t-test for independent samples, and one-way analysis of variance (ANOVA). The assumptions of the parametric procedures were examined using residual plots, skewness and kurtosis coefficients, and the Shapiro–Wilk test. If assumptions of the parametric procedures were violated, then the Wilcoxon and Kruskal–Wallis non-parametric procedures were used, respectively. One-sided tests and a level of significance of $\alpha = 0.05$ was used for all statistical procedures unless otherwise stated.

Spatial interpolation of MDN Hg deposition and concentration data to the LCM and GTC sites was done using ArcMap v.10.3.1 (Esri, Redlands, CA, USA) using inverse distance weighting in a manner

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