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Determinants of atmospheric mercury concentrations in Reno, Nevada, U.S.A.

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ABSTRACT

Concentrations of gaseous elemental mercury (GEM), gaseous oxidized mercury (GOM) and particulate-bound mercury (PBM) were measured along with ancillary variables 9 km east of downtown Reno, Nevada, U.S.A. from November 2006 through March 2009. Mean two-year (February 2007 through January 2009) GEM, GOM, and PBM concentrations were 2.0 ± 0.7 ng m^{-3} (± standard deviation), 18 ± 22 pg m^{-3}, and 7 ± 7 pg m^{-3}, respectively. Data collected were compared with observations made at another location just north of the city at 169 m higher elevation. At both locations higher concentrations of GEM and PBM occurred in periods with little atmospheric mixing, indicating that local sources were important for enhancing GEM and PBM concentrations in Reno above that considered continental background. Concentrations of GOM were higher (maximum of 177 pg m^{-3}) during periods with higher temperature and lower dew point. Higher GOM concentrations at the higher elevation site with less urban impact relative to the valley site, along with other data trends, support the hypothesis that in northern Nevada subsiding dry air from the free troposphere is a source of GOM to the surface.

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1. Introduction

The atmosphere has been identified as a major pathway by which mercury reaches and contaminates ecosystems (Fitzgerald et al., 1998). Atmospheric mercury is frequently measured as three fractions: gaseous elemental mercury (GEM), gaseous oxidized mercury (GOM, a.k.a. RGM), and particulate-bound mercury (PBM). Measurements of atmospheric mercury fractions have been made in a variety of environments throughout the world (c.f. Valente et al., 2007), but significant gaps in our understanding of the mechanisms that control mercury concentrations in the atmosphere still exist (Lin et al., 2006; Lindberg et al., 2007).

In urban or other anthropogenically impacted environments concentrations of GEM, GOM, and PBM may depend on releases from specific sources (Liu et al., 2007; Edgerton et al., 2006; Manolopoulos et al., 2007) or on oxidation of GEM by anthropogenic oxidants to produce GOM (Weiss-Penzias et al., 2003; Lynam and Keeler, 2005), though uncertainty exists regarding which oxidants play important roles (Lindberg et al., 2007; Calvert and Lindberg, 2005). Reactions with ozone and OH radical have been investigated (Pal and Ariya, 2004a, 2004b; Calvert and Lindberg, 2005; Sommar et al., 2001), as have reactions with nitrate radical and hydrogen peroxide (Lin and Pehkonen, 1999). Oxidation of GEM by halogen radicals has been shown to account for the dynamic behavior of atmospheric mercury in polar regions (Lindberg et al., 2002; Ebinghaus et al., 2002), and may contribute to formation of GOM in ocean environments (Hedgecock and Pirrone, 2004; Holmes et al., 2009) and the upper atmosphere (Holmes et al., 2006).

Northern Nevada (U.S.A.) is a region with diverse natural and anthropogenic mercury sources (c.f. Engle et al., 2001; Zehner and Gustin, 2002; Lyman and Gustin, 2008). Moreover, before measurements of GOM in northern Nevada had been published, Selin et al. (2007) predicted that high elevation deserts (like Nevada) would have high GOM concentrations due to subsidence of GOM-rich air from the free troposphere. Weiss-Penzias et al. (2009) recently reported high GOM in Nevada and showed that regional variation in GOM concentrations could be explained by air mass transport from a dry, high-altitude area of the atmosphere.

This paper presents an analysis of more than two years of atmospheric mercury measurements collected in the valley hosting Reno, Nevada. It builds on the work of Weiss-Penzias et al. (2009), who report on air mercury data collected at three sites (including the location of this work) in summer 2007, as well as the works of Stamenkovic et al. (2007) and Peterson et al. (2009), who describe observations of air mercury concentrations made at a site 169 m above the valley floor in Reno from 2002 to 2005 and from 2005 to 2007, respectively. Detailed statistical analyses and comparison of data collected at these two closely situated locations provide the basis for discussion of the mechanisms that control atmospheric mercury concentrations in northern Nevada, with implications for the global mercury cycle.

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2. Methods

2.1. Site description

Data were collected from November 2006 through March 2009 at Mercury Deposition Network (http://nadp.sws.uiuc.edu/mdn) site NV98 in Reno, Nevada, U.S.A (39.51° latitude, −119.72° longitude, elevation 1340 m). This site is located on the University of Nevada, Reno Nevada Agricultural Experiment Station approximately 9 km east of downtown Reno and 4 km southeast of downtown Sparks, Nevada (Fig. 1). The Reno–Sparks area is urban and suburban with a population of approximately 400 000, and does not have heavy industrial facilities or any mercury sources listed on the United States Environmental Protection Agency's Toxics Release Inventory (http://www.epa.gov/triexplorer/). The metropolitan area is situated in an arid valley (average annual precipitation of 19 cm) between the Sierra Nevada and Virginia mountain ranges. This region includes some mercury enriched areas, including but not limited to Steamboat Springs (Coolbaugh et al., 2002) and Peavine Peak (Engle and Gustin, 2002).

2.2. Atmospheric mercury

Concentrations of GEM, GOM, and PBM (fine mode) were measured with a Tekran 2537A/1130/1135 system as described by Landis et al. (2002). The flow rate through the denuder and particulate filter assembly was 7 standard L min^{-1} (standard conditions of 0 °C and 100 kPa), and the flow rate through the Model 2537A was 1 standard L min^{-1}. The volumetric flow at the elevation and range of temperatures in this study was ~10 L min^{-1} and as such the particulate size cutoff point of the system was in the range of 2.5 to 2.8 µm (cf. Lyman and Gustin, 2008). The system alternated between 2 h sampling periods and 1 h desorption periods, and the Model 2537A was automatically calibrated using its internal permeation source every eight sampling cycles. The denuder and the 0.1 µm filter on the particulate assembly were replaced every three weeks. The particulate assembly, sample train glassware, and sample lines were removed and cleaned every six months. The accuracy of the permeation rate of the internal permeation source was checked every three months by repeated injections of a known amount of saturated mercury vapor into mercury-free air being sampled by the instrument. Calibration checks were performed weekly by injecting a known amount of saturated mercury vapor into ambient air being sampled by the instrument. Data were only included for analysis if bracketed by calibration checks that fell within the range of 100 ± 10%.

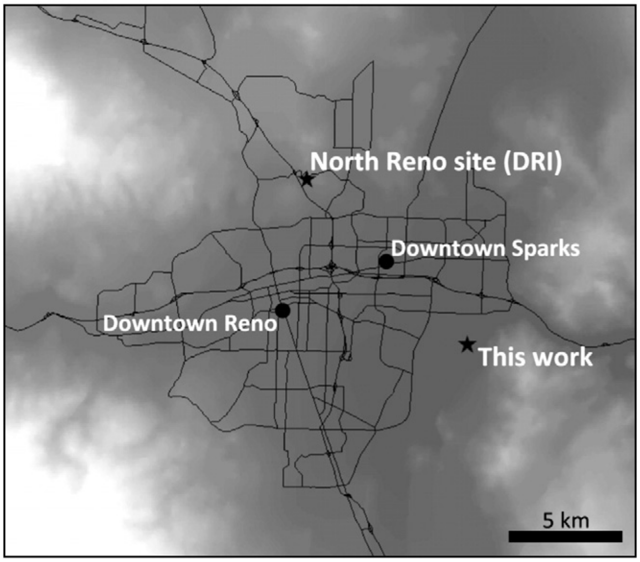

Fig. 1. Map of the Reno–Sparks area, including the study site from this work and the site in north Reno (Peterson et al., 2009). Lighter shading indicates higher elevation.

Also, the Tekran system from this study and that from the higher elevation site in Reno reported on by Peterson et al. (2009) were compared for 10 days in February 2008. The systems were mounted side by side at the same height and were programmed and calibrated identically. The difference between GEM concentrations reported by the two systems was 11 ± 4%, while the differences between GOM and PBM concentrations were 8 ± 40% and −71 ± 41%, respectively (positive values indicate the system from this study was higher).

2.3. Ozone

Ozone concentrations were measured using a Teledyne-API Model 400E ozone analyzer. Calibration checks of the Model 400E were performed weekly using a Teledyne-API Model 700E Dynamic Dilution Calibrator equipped with an internal ozone generator. The Model 700E was checked against a NIST-traceable ozone standard every 6 months. Data were only included for analysis if bracketed by span calibration checks that fell within the range of 100 ± 10% and zero calibration checks that were within the range of 0 ± 2 ppb.

2.4. Meteorology

Wind speed and direction (Young Model 05305), humidity and temperature (Vaisala Model HMP45AC), and solar radiation (LI-COR Model LI200X) were measured and values were logged with a Campbell Scientific CR1000 data logger.

2.5. Particulate matter

Two DustTrak Model 8520 Aerosol Monitors were used, one with a 1 µm impactor inlet and one with a 10 µm cyclone inlet, and they sampled outside air through a 1.5 m grounded copper tube. DustTrak analyzers measure particle scattering and are factory calibrated for mass concentrations using a road dust standard. These instruments were zero checked each week, and data were only included for analysis if zero checks were within the range of 0 ± 1 µg m^{-3}. Flow rates and instrument filters and inlets were checked every three weeks, and filters and inlets were replaced or cleaned as needed.

Also, measurements of PM2.5 were obtained from the Washoe County Health District Air Quality Management Division for June–August 2007 and June–August 2008. These measurements were collected in downtown Reno on filters deployed for 24 h every third day.

2.6. Dataset structure

The Tekran system returned one 2 h average measurement of GOM and PBM every 3 h, but GEM, ozone, and meteorological parameters were collected as 5 min averages, so 2 h composite averages of GEM and ozone concentrations and meteorological parameters were calculated for each GOM and PBM measurement period, and these composite averages were used for analysis. Composite 24 h averages were also computed for all parameters and used to elucidate and compare trends at a coarser temporal resolution.

2.7. Data analyses

Data were processed using Microsoft Excel 2003 and SPSS 15.0. Multiple linear regression analyses and Pearson correlation coefficients were developed using SPSS. While Pearson correlation

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