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Temporal patterns of atmospheric mercury species in northern Mississippi during 2011–2012: Influence of sudden population swings

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HIGHLIGHTS

- Airborne mercury species were monitored for a year in the mid-south USA.
- Temporal trends and the influence of meteorological conditions were evaluated.
- GOM and PBM increased with a relatively large and sudden increase in population.
- Modeling shows plume events often occur with air masses from the northern USA.

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ABSTRACT

Gaseous elemental mercury (GEM), gaseous oxidized mercury (GOM) and particulate bound mercury (PBM) were measured on the University of Mississippi campus from July 2011 to June 2012. It is believed to be the first time that concentrations of atmospheric mercury species have been documented in northern Mississippi, and at a location with relatively large and sudden swings in population. The mean concentration (±1SD) of GEM was 1.54 ± 0.32 ng m^{-3}; levels were lower and generally more stable during the winter (1.48 ± 0.22) and spring (1.46 ± 0.27) compared with the summer (1.56 ± 0.32) and fall (1.63 ± 0.42). Mean concentrations for GOM and PBM were 3.87 pg m^{-3} and 4.58 pg m^{-3}, respectively; levels tended to be highest in the afternoon and lowest in the early morning hours. During the fall and spring academic semesters concentrations and variability of GOM and PBM both increased, possibly from vehicle exhaust. There were moderate negative correlations with wind speed (all species) and humidity (GOM and PBM). Backward air mass trajectory modeling for the ten highest peaks for each mercury species revealed that the majority of these events occurred from air masses that passed through the northern continental US region. Overall, this study illustrates the complexity of temporal fluctuations of airborne mercury species, even in a small town environment.

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1. Introduction

Mercury is a toxic environmental pollutant capable of long-range (global) transport through the atmosphere (Lin et al., 2006). Airborne mercury comes from both natural sources (e.g., evasion from soils, vegetation, volcanoes, forest fires, and mineral deposits) and anthropogenic sources (e.g., coal combustion, waste incineration, and certain industrial processes), and these sources have been estimated at 5207 Mg/year and 2320 Mg/year, respectively (Pirrone et al., 2010). Anthropogenic emissions appear to be leading to a general increase in mercury on local, regional and global scales (Pirrone et al., 2010). Mercury deposits to terrestrial and aquatic systems through wet and dry mechanisms where it can undergo biotic and abiotic transformation to methylmercury, which in-turn readily bioaccumulates and concentrates in food webs (Watras et al., 1998). This has motivated intensive research on mercury as a pollutant.

Airborne mercury is commonly classified as existing in three primary forms (species), gaseous elemental mercury (GEM), gaseous oxidized mercury (GOM) and particulate bound mercury (PBM), each with distinctive chemical and physical properties and environmental behavior. GEM is the predominant form of atmospheric mercury (often >95% of the total). Because it is relatively inert, sparingly soluble in water, and has low dry deposition velocity (i.e., 0.02–0.2 cm s^{-1}) (Seigneur et al., 2004), it has a long residence time (months to years), allowing it to circulate globally (Schroeder and Munthe, 1998; Weiss-Penzias et al., 2003). The northern hemispheric background level of GEM is approximately 1.6 ng m^{-3} (Lindberg et al., 2007). GEM is slowly converted to soluble GOM through various photochemical reactions, including reactions which may involve ozone (O_3), hydroxyl radical (OH), ni-

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trate (NO_3), hydrogen peroxide (H_2O_2), and/or atomic bromine (Br) (Holmes et al., 2010). GOM and PBM have much shorter residence times (hours to weeks) in the atmosphere, and are readily removed through wet and dry deposition mechanisms (Lyman et al., 2007). PBM is any form of mercury contained in- or associated with-aerosol particles, whose transport depends greatly on particle size and meteorological conditions (Keeler et al., 1995). Natural emissions are primarily in the gaseous elemental form, whereas anthropogenic emissions often include all three species (GEM, GOM and PBM) (Schroeder and Munthe, 1998). Several studies have demonstrated the importance of determining mercury speciation, particularly for GOM which is often responsible for contamination near point sources (Peterson et al., 1995; Dvonch et al., 1998; Landis et al., 2002).

Most studies of atmospheric mercury species in the United States have been conducted in the northeast (Sillman et al., 2007), in the south in Florida or along the Gulf of Mexico (Rolison et al., 2013), and in the far west in Nevada or California (Stamenkovic et al., 2007). Our work has been motivated in part by the lack of data for mercury in the mid-south environment, despite the region having several large-scale coal-fired power plants (CFPPs), conditions that favor methylation of Hg^{+2} (e.g. wetlands and backwaters with high levels of natural organic matter, sulfate-reducing bacteria, and long-hot summers), and lakes with fish consumption advisories due to high-levels of mercury (Ullrich et al., 2001).

The aim of this study was to fill a data-gap for atmospheric mercury in the mid-south USA to aid regional mercury models and deposition studies. The study included monitoring airborne mercury species in northern Mississippi for a full year. Here, we report on their concentration, temporal variations, and sensitivity to meteorological parameters. We include backward trajectory air mass modeling to investigate possible sources for mercury. The study also allowed us to evaluate the impact of a sudden doubling of a population center on airborne mercury, because it took place in a "college town" (Oxford, Mississippi), where the population about doubles from approximately 25 000–50 000 while school is in session.

2. Material and methods

2.1. Sampling site

Oxford is located in northern Mississippi at an elevation of ~150 m above sea level. This relatively small urban college town has a population that about doubles during the traditional academic year, starting in late August. The atmospheric mercury speciation system was situated about 20 m above the ground on a platform raised several meters above the roof of a building (34°21′50″N, 89°32′60″W) in the center of the University of Mississippi (Ole Miss) campus. The location on the top of the building is high enough to receive air masses unimpeded by trees and vegetation which can influence speciation via absorption and volatilization.

Major sources of anthropogenic airborne mercury in the region are shown in Fig. 1. There are no major sources in the immediate study area. However, there are several within a 125 km radius, including two CFPPs. The Allen Fossil Plant is a 552 MW facility located near Memphis, Tennessee, a city with a population of about 650 000; it reported releases of 86.2 kg of mercury in 2010 (USEPA, 2010). The other CFPP is the 514 MW Red Hills generation facility located ~110 km to the south with 186 kg of mercury reported released in 2010 (USEPA, 2010). Further to the north and west are two additional CFPPs (Fig. 1), which together released 526 kg of mercury to the air in 2010, and two industrial steel facilities (347 kg), and a manufacturing facility (143 kg). In the surrounding region as a whole (defined here as Mississippi, Tennessee, Arkansas, Louisiana, Alabama, Georgia, Kentucky, Indiana, Illinois, and Missouri) anthropogenic sources were estimated to release a total of 13 085 kg to the air in 2010 (USEPA, 2010). Among the sources, CFPPs accounted for 70.1% of the emissions; steel and chemical companies yielded a total of 18.5%, waste incinerators 1.4%, and the remaining (other) 10%. The emission from waste incinerators is notable because it represents a significant decline from earlier emission inventories.

Emissions from natural sources in the region are complex and include evasion from agricultural fields, forests, waterbodies. Moreover, evasion of mercury from surfaces includes re-emission of previously deposited mercury originating from both natural and anthropogenic sources. We are not aware of any studies evaluating the natural mercury emission inventory for the region.

2.2. Mercury speciation system

We measured mercury species in the ambient air from July 2011 to June 2012 using an automated system from Tekran Inc. (Toronto, Canada). The system uses a model 2537B mercury analyzer for GEM measurements based on cold vapor atomic fluorescence spectrometry (CVAFS), and is combined with a model 1130 GOM and model 1135 PBM speciation units, to sample and detect the three mercury species. The unit was operated with a total flow rate of 10 L min^{-1} and 1 L min^{-1} for analytical sampling, and an inlet with an impactor is used to remove coarse particle ($\geqslant$2.5 μm) from the ambient air. In short, ambient air is sequentially passed through a KCl-coated quartz annular denuder which captures GOM, through a quartz regenerable particulate filter to sample PBM, and through one of two gold traps to collect GEM. The system uses two parallel gold traps to alternately adsorb and desorb (thermally) GEM every 5 min. High-purity argon carries the desorbed mercury atoms through the fluorescence cell of the analyzer. PBM and GOM are measured on a 2 h basis after being thermally desorbed from the filter and denuder, respectively, a process that assures these operationally defined mercury species are converted to GEM for measurement with the 2537B. The speciation system reports twelve PBM and twelve GOM values for each 24 h period.

For quality assurance, the system was set to perform an automatic calibration using an internal permeation source every 24 h. In addition, an external calibration check was performed regularly (~monthly) using a Tekran 2505 mercury vapor calibration unit and a digital gas-tight syringe. Injection recoveries were within 92–105%. During one period results for adjacent gold traps started to deviate from each other by >10%, possibly due to passivation of the gold surface. The gold traps were replaced and the deviation was eliminated. The suspect data (about 2 weeks' worth) associated with this event was removed from the data set. Bi-weekly maintenance included replacing the frit in the impactor, sodalime trap, sample filter and zero air filters, and re-coating the KCl-denuder. The regenerable particulate filter was replaced monthly.

2.3. Modeling and data analysis

To investigate potential source areas of mercury observed in Oxford, MS, the NOAA hybrid single-particle Lagrangian integrated trajectory model (HYSPLIT) using EDAS 40 km input data, was used to calculate backward air mass trajectories (Draxler and Hess, 1997). The HYSPLIT model was initially designed to help in case of atmospheric emergencies. It computes advection, dispersion and deposition using either puff or particle approaches. The dispersion rate is calculated by considering vertical diffusivity profile, wind shear, and horizontal deformation of the wind field. The concentrations are calculated at a specific grid point for puffs and as

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