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Potential impact of rainfall on the air-surface exchange of total gaseous mercury from two common urban ground surfaces

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ABSTRACT

The impact of rainfall on total gaseous mercury (TGM) flux from pavement and street dirt surfaces was investigated in an effort to determine the influence of wet weather events on mercury transport in urban watersheds. Street dirt and pavement are common urban ground surfaces that concentrate many substances (eroded soil, leaf and vegetation litter, automobile debris, industrial atmospheric fallout) which can contain elevated mercury concentrations. In this study, the primary analyses included (i) observing the time series flux of TGM from pavement and street dirt following surface wetting and (ii) determining if wet deposition provides a fresh source of mercury that is available for release (emission) when applied to these surfaces. Application of de-ionized water (DI) and rainwater both induced an immediate 65% increase in TGM emission from pavement (from 0.5 to 1.4 ng m^{-2} h^{-1} [based on averages]). For street dirt, an immediate 70% increase in emission was induced following DI water application (from 3.0 to 9.0 ng m^{-2} h^{-1} [based on averages]) and an immediate 30% increase in emission following rainwater application (from 4.5 to 6.5 ng m^{-2} h^{-1} [based on averages]). Both surfaces showed continuous elevated release of TGM following the initial water application stage. There was a decrease in emission as the pavement surface dried. Despite the difference in immediate TGM emission from street dirt using both solutions, statistical evaluation indicated there was no prolonged difference. This suggests that mercury in rainwater was not available for re-emission when applied to these surfaces, at least for the time frame studied (2 h after water application). Therefore, it is likely that the elevated TGM emission following water application resulted primarily from pre-existing mercury. Removal of pre-existing mercury by water application followed a zero order process for both surfaces; however, removal rates were much different for each surface ($k = 0.26$ ng m^{-2} min^{-1} for street dirt; $k = 0.03$ ng m^{-2} min^{-1} for pavement). Results from laboratory surface washing experiments revealed only 0.1% of all available surface-bound mercury on pavement was removed by surface emission 90 min after a simulated light rainfall event (0.13 cm of rainfall).

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1. Introduction

Street dirt and pavement are common ground surface characteristics in urban areas that can add to the complexity of gaseous mercury re-emission to the atmosphere. Street dirt is a frequently used term in storm water management practice to describe particulates on streets and roadways. Street dirt typically collects near curbs and low depression areas and is commonly removed by street cleaning operations and storm flow. A myriad of substances comprise street dirt, such as, eroded soil, leaf and vegetation litter, road salts and automobile debris (e.g. tire break-off, grease and lubricants, brake pad debris) (Burton and Pitt, 2002). Much of this debris contains elevated concentrations of metals e.g. lead, copper, zinc, cadmium, chromium, nickel, mercury; bacterial substances such as fecal coliform and strepotocci; and organic pollutants such as PCB's (Burton and Pitt, 2002; Fulkerson et al., 2007). Pavement and impervious surfaces are prominent features in urban areas. In the contiguous 48 United States, impervious surfaces cover 110,000 km^2 (Groffman et al., 2006; Nowak, 2006), an area equal to the state of Ohio. Yearly development adds approximately 1000 km^2 yr^{-1}. Typically, one-third of total impervious cover is building roofs and two-thirds represents various pavement types (streets, sidewalks, parking lots, driveways, etc.).

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Primary sources for mercury in urban areas that contribute to mercury build up in street dirt and on the surface of pavement are the following: (1) the upper few centimeters of soil and leaf litter (Schuster, 1991; Grigal, 2002; Friedli et al., 2003; Gabriel and Williamson, 2004) (2) wet and dry deposition (Bigham and Vandal, 1996; Atasi et al., 2002; Eckley et al., 2008) and (3) automobile products e.g. engine oil and fuel (Wilhelm and Bloom, 2000; Wilhelm, 2001; Conaway et al., 2005). The spatial coverage of street dirt is typically small in urban areas (<5% of total surface area) but may be an area of elevated Hg emission due to higher surface mercury concentration.

In this research we assessed the impact of rainfall on total gaseous mercury (TGM) (~95% elemental mercury [Hg^0]) flux from pavement and street dirt using wetting experiments. Investigations took place to address the following questions: (i) How does wetting pavement and street dirt impact time series flux of TGM? (ii) If surface wetting increases TGM emission, does wet deposition (rain water) provide a fresh pool of mercury that is available for release when applied? From the adopted methodology, we evaluated the impact of short, intermittent periods of rainfall on terrestrial TGM flux that occurs during the spring and summer seasons in temperate and, particularly, humid climates. It is widely known that sunlight, surface moisture, and surface temperature impact TGM flux from many surfaces (Poissant et al., 1999; Lindberg et al., 1999; Zhang and Lindberg, 1999; Gillis and Miller, 2000; Zhang et al., 2001; Scholtz et al., 2003; Gustin et al., 2002, 2006; Choi and Holsen, 2009). With pavement temperatures reaching well over 37 °C in spring and summer months, short intense periods of precipitation followed by high sunlight intensity may result in large TGM emission.

This investigation presents some of the first data addressing the impact of rainfall on mercury air-surface exchange from urban ground surfaces. Similar urban-related investigations have been conducted by Eckley and Branfireun (2008) and Eckley et al. (2008). The specific topics evaluated in this research (aforementioned i and ii) have been investigated before (e.g. Lindberg et al., 1999; Gillis and Miller, 2000; Song and Van Heyst, 2005), but for background surface soils. Since irrigation impacts on TGM flux are suggested to be a function of surface geochemistry and physical characteristics (e.g. surface permeability) (Lindberg et al., 1999), pavement and street dirt surfaces may show highly varying results in comparison to soil. Molecular diffusion, drying and heating, and particle adsorption for pavement are significantly different than natural surfaces. Street dirt is generally much less compact and more permeable than background soil thereby potentially promoting higher TGM emission. Overall, the results from this study will help to understand the air-surface of mercury from urban areas and aid in the development of mercury emission inventory estimates.

2. Methods and materials

2.1. Site description

Flux monitoring was conducted in the city of Tuscaloosa, AL over a total of twelve days during the spring and summer seasons of 2004. Water application and associated measurement of TGM flux over pavement and street dirt was performed at a fixed location within a mixed landuse area of Tuscaloosa. Selection of this site was based on its security, accessibility and its close representation of Tuscaloosa in terms of surface cover characteristics and proximity to local mercury sources (Gabriel et al., 2006).

TGM flux measurements were conducted over two different surfaces: street dirt and pavement. The street dirt analyzed in this research was produced from locally eroded soil ("Georgia red clay" [mainly kaolinite]), decomposing turf grass, tree and leaf litter and weathered pavement (gravel [up to ~10 mm in diameter]) from the catchment area (2406 m^2). Its average bulk density was 1.7 g cm^{-3} and was ~1 cm depth for both sampling seasons. Small fractions of street dirt debris were coated with engine oil as a result of automobile traffic. Thirty percent of the gravel had diameters >1 mm. The street dirt collected along a curb as a result of previous wet weather events. The surface area of the street dirt was large enough to place the flux chamber over fresh locations each time an experiment was run. The pavement surface that was investigated was adjacent to the street dirt. The pavement was set in place approximately 10 to 15 years prior, was composted of a mixture of asphalt and limestone aggregates and had moderate wear from weathering and automobile traffic.

2.2. TGM flux calculation

TGM surface fluxes were measured using the following equipment: (1) Tekran 2537A TGM analyzer (0.1 ng m^{-3}detection limit) (2) Tekran Automated Dual Sampling Unit (TADS) and (3) a polycarbonate chamber (dynamic flux chamber). The 2537A analyzer uses cold vapor atomic fluorescence spectrometry (CVAFS) for mercury detection. TGM flux was calculated using Equation (1). The terms in Equation (1) are as follows: F (ng m^{-2} h^{-1}) is the TGM flux; C_i (ng m^{-3}) is the inlet concentration (ambient concentration); and C_o (ng m^{-3}) is the outlet concentration (chamber concentration). Q (L m^{-1}) is the Tekran sampling flow rate and A is the chamber footprint (669 cm^2). The chamber has a volume of 9.3 L, a height of 14 cm and a diameter of 29.2 cm. Lindberg et al. (2002) and Zhang et al. (2002) provide a detailed description of this chamber including its performance properties.

$$F = \frac{(C_o - C_i)Q}{A} \quad (1)$$

The Tekran sampling flow rate (Q) was set at 1.5 L min^{-1} for the entire study and the chamber flushing flow rate was set at 2.0 L min^{-1}. Low chamber flushing flow rates were chosen due to the low TGM flux magnitudes and THg concentrations for these surfaces (Poissant and Casimir, 1998; Zhang et al., 2001, 2002; Lindberg et al., 2002; Gabriel et al., 2006). Four 9.7 mm diameter holes were drilled near the base of the chamber. These holes were used to create an interaction between the internal chamber and the ambient boundary layer condition outside the chamber. TGM concentrations were quantified every five minutes. One inlet (C_i) then one outlet (C_o) concentration were sequentially quantified through time. These concentrations were used to calculate one flux every ten minutes.

The chamber used in this study has a similar design to chambers used in other studies (Lindberg et al., 1999, 2002; Eckely et al., 2010). Chamber design (e.g. placement of air-exchange holes, volume, material, surface area) can have major impact on TGM fluxes (Carpi et al., 2007; Eckely et al., 2010). Limitations associated with polycarbonate chambers are weak transmission of ultra-violet light and underestimation of surface fluxes (Carpi et al., 2007; Eckely et al., 2010).

2.3. Meteorological data collection

Meteorological data was collected by a NIST (National Institute of Standards and Technology) certified and calibrated Davis Meteorological Station. This station was used to collect the following parameters: surface and air temperature (°C), surface moisture (centibar), relative humidity (%), and solar radiation (W m^{-2}). Sensors for the surface temperature unit were buried to a 1 cm depth for street dirt and directly placed on the pavement surface.

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