



Seasonal and spatial patterns of mercury wet deposition in the United States: Constraints on the contribution from North American anthropogenic sources

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Abstract

Observed wet deposition fluxes of mercury in the United States show a maximum in the Southeast, and a consistent seasonal variation (maximum in summer, minimum in winter) that increases in amplitude from north to south. We simulate these patterns successfully with a global 3-D chemical transport model (GEOS-Chem) including our best estimates of sources and processes. We attribute the high wet deposition over the Southeast in summer to scavenging of upper-altitude Hg(II) by deep convection. Seasonal variation at higher latitudes is attributed to a combination of enhanced summertime oxidation of Hg(0) and inefficient scavenging of Hg(II) by snow. Scavenging of Hg(II) from above the boundary layer contributes over half of wet deposition to the US in the model. Even within the boundary layer, we find that most of Hg(II) originates from the global mercury pool. Wet deposition in the model accounts for only 30% of total mercury deposition in the US, the remainder being from dry deposition, including 42% from Hg(0) uptake. North American anthropogenic emissions contribute 20% of total mercury deposition in the US (up to 50% in the industrial Midwest and Northeast).

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1. Introduction

Regulation of mercury emissions in the United States aims to reduce the deposition of mercury to domestic ecosystems. Global modeling studies indicate that most of this deposition originates in

fact from emissions outside North America (Seigneur et al., 2004; Selin et al., 2007; Strode et al., 2007, 2008), but there are large uncertainties in model representations of mercury chemistry and deposition processes. We use here data from the national mercury deposition network (MDN) (National Atmospheric Deposition Program, 2007) to test the ability of a global 3-D model (GEOS-Chem) to reproduce observed seasonal as well as spatial wet deposition patterns, and from there to better quantify the sources of mercury deposition in the United States.

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Mercury is emitted to the atmosphere in gaseous elemental form Hg(0), in semivolatile oxidized form Hg(II), and in nonvolatile particulate form Hg(P). Hg(0) has a long (0.5–2 yr) atmospheric lifetime and represents a globally well-mixed mercury pool; it is eventually oxidized to Hg(II), which is highly water-soluble and readily deposited. Deposition of emitted Hg(II) and Hg(P) can directly affect the region of emission, although Hg(II) can also be reduced to Hg(0) and enter the global pool. Anthropogenic emission of mercury from North America is mostly from coal combustion; about half is as Hg(0) and half is as Hg(II)+Hg(P) (Pacyna et al., 2006). Considering that North America accounts for only 7% of global anthropogenic emission of mercury (2000 statistics) (Pacyna et al., 2006), any diagnosis of regional vs. global contributions to mercury deposition must focus on the fate of the emitted Hg(II)+Hg(P) and on the supply of Hg(II) by oxidation of Hg(0) from the global pool.

Previous analyses of wet deposition data have reached conflicting conclusions regarding the relative contributions of domestic vs. global contributions to mercury deposition in different US regions (Dvonch et al., 1998, 2005; Guentzel et al., 2001; Keeler et al., 2006b; Vanarsdale et al., 2005). We show here that the observed seasonal variation of mercury deposition and its latitudinal gradient provide important constraints on this problem when interpreted with a global 3-D model. We focus our analysis on MDN data for 2004–2005, the two most recent years of data available and with the best coverage.

2. Model description

The GEOS-Chem atmosphere–land–ocean mercury simulation is described by Selin et al. (2008). We use here GEOS-Chem version 7.04 (<http://www.as.harvard.edu/chemistry/trop/geos>) (Bey et al., 2001) at $4^\circ \times 5^\circ$ resolution with assimilated meteorological data for 2004–2005 from the NASA Goddard Earth Observing System (GEOS-4). Three mercury species (Hg(0), Hg(II), and Hg(P)) are transported in the atmosphere. Anthropogenic emissions are from the GEIA inventory for the year 2000 (Pacyna et al., 2006), modified as described in Selin et al. (2008) to satisfy global observational constraints. These modifications include a 50% increase in Hg(0) in Asia (now 1939 Mg yr^{-1} total Hg), a 30% increase in the rest of the world (now 1011 Mg yr^{-1}), and addition of

emissions from biomass burning (600 Mg yr^{-1}) and artisanal mining (450 Mg yr^{-1}). The total emissions from anthropogenic sources and biomass burning are thus 4000 Mg yr^{-1} . Atmosphere–ocean coupling is treated with a slab model for the ocean including cycling between Hg(0), Hg(II), and nonreactive mercury in the oceanic mixed layer (Strode et al., 2007). Atmosphere–land coupling includes partial recycling of deposited Hg(II) and mobilization of long-lived soil mercury through volatilization and evapotranspiration (Selin et al., 2008). Mercury is volatilized from the land and oceans exclusively as Hg(0); direct emission of Hg(II) and Hg(P) is solely anthropogenic.

Atmospheric oxidation of Hg(0) to Hg(II) in the model takes place by OH ($k = 9 \times 10^{-14} \text{ cm}^3 \text{ s}^{-1}$, Sommar et al., 2001; Pal and Ariya, 2004) and O_3 ($k = 3 \times 10^{-20} \text{ cm}^3 \text{ s}^{-1}$, Hall, 1995). In-cloud (aqueous) photochemical reduction of Hg(II) to Hg(0) is included to accommodate observational constraints on global mercury atmospheric concentrations and seasonal variation at northern mid-latitudes (Selin et al., 2007). Hg(P) is considered chemically inert and is removed by deposition. Fast Hg(II) reduction in power plant plumes (Lohman et al., 2006) remains hypothetical and is not included in the model.

Fig. 1 shows the spatial distribution of Hg(0), Hg(II) and Hg(P) anthropogenic emissions in North America in GEOS-Chem, at the original $1^\circ \times 1^\circ$ resolution of Pacyna et al. (2006). Hg(II) and Hg(P) represent, respectively, 33% and 14% of the total mercury emission of 169 Mg yr^{-1} for the domain of Fig. 1. Emissions are highest in the industrial Midwest (Illinois, Indiana, Ohio, Kentucky, Pennsylvania, and West Virginia) due to coal combustion, but high values are generally found in population centers due to additional sources from waste incineration and industrial processes. Some high values in the West (notably in northern Nevada) are from gold mining.

Mercury deposition processes are of particular interest here. GEOS-Chem includes wet and dry deposition of Hg(II) and Hg(P), as well as dry deposition of Hg(0). Wet deposition of Hg(II) and Hg(P) includes rainout and washout from large-scale and convective precipitation, and scavenging in convective updrafts (Liu et al., 2001; Selin et al., 2008). Hg(P) is scavenged as a water-soluble aerosol (Liu et al., 2001), while Hg(II) is scavenged as a highly water-soluble gas. Hg(II) is released to the gas phase when water freezes (zero retention

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