Estimating Global Source Influence on U.S. Mercury Deposition using GEOS-Chem
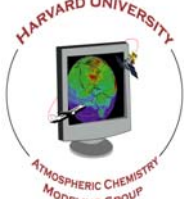
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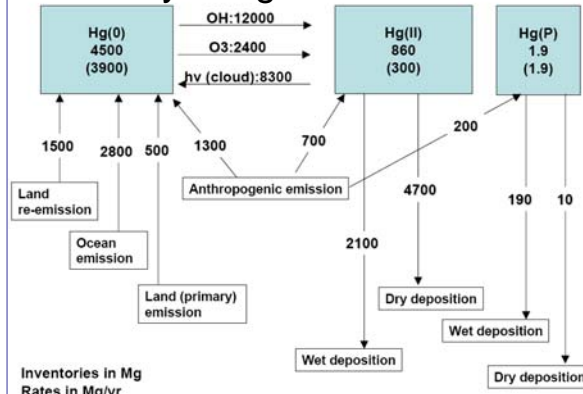1. Harvard University, 2. University of Washington

Abstract: The GEOS-Chem global tropospheric chemistry model is being applied to provide boundary conditions for regional modeling in the United States with regulatory applications in the context of the U.S. Clean Air Mercury Rule. This allows regulators to take into account the contributions of global mercury sources in a consistent way with other pollutants of concern. We use the GEOS-Chem mercury simulation (which includes the species Hg(0), Hg(II) and Hg(P)) to show the contributions of sources other than domestic anthropogenic emissions to wet and dry deposition to the U.S. We compare these model results to the wet deposition measurements taken by the U.S. Mercury Deposition Network (MDN). Simulated wet deposition shows a latitudinal gradient, with higher deposition at lower latitudes reflecting the influence of oxidation of Hg(0) to Hg(II) by OH and its subsequent deposition. A regional enhancement in total (wet+dry) deposition is seen in the Midwest in the model, which is a result of proximity to sources. Measurements show a similar regional enhancement. The simulated contribution of North American anthropogenic sources to total deposition over the U.S. is extremely variable, contributing on average 20%.
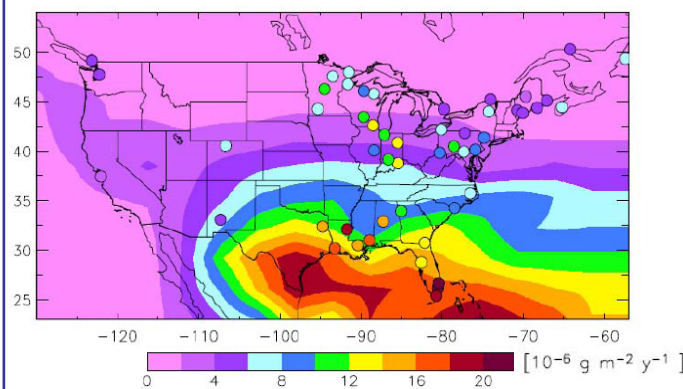
1. GEOS-Chem mercury model description

• GEOS-Chem CTM version 7.04 (http://www-as.harvard.edu/chemistry/trop/geos/) [*Bey et al.*, 2001], using assimilated meteorological data from the NASA Goddard Earth Observing System (GEOS-4). Resolution of model is 4°x 5°.

• Three species of atmospheric mercury: elemental (Hg(0)), divalent (Hg(II)), and primary particulate (Hg(P)).

• Uses GEIA emissions inventory for 2000 [*Pacyna and Pacyna*, 2005; *Wilson and Steenhuisen*, 2005]; includes sources from land re-emission and primary emission; incorporates coupled ocean-atmosphere model [*Strode et al.* 2006]

• Chemical reactions: Hg(0) oxidation by OH ($k = 9 \times 10^{-14}$ cm^3 s^{-1} [*Pal and Ariya*, 2004; *Sommar et al.*, 2001]) and ozone ($k = 3 \times 10^{-20}$ cm^3 s^{-1} [*Hall*, 1995]); Hg(II) aqueous-phase, photochemical reduction.

• 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]

• Dry deposition of Hg(II) and Hg(P) simulated with a standard resistance-in-series scheme based on local surface type and turbulence [*Wang et al.*, 1998; *Wesely*, 1989], assuming zero surface resistance for Hg(II). Dry deposition of Hg(0) to the ocean is simulated as part of the bi-directional exchange model of *Strode et al.* [2006].

For further information about the GEOS-Chem mercury simulation, see:
*Selin et al.* [2006], *Journal of Geophysical Research*, accepted
http://www-as.harvard.edu/chemistry/trop/publications/selin2006a.pdf

Mercury budget in GEOS-Chem

Global atmospheric mercury budget in GEOS-Chem. Tropospheric inventories are given in parentheses (most of atmospheric Hg(II) is in the stratosphere).

2. Comparison with wet deposition measurements over the U.S.

Annual mercury wet deposition fluxes over the United States for 2003-2004. Observations from the Mercury Deposition Network (circles, NADP (2003)) are compared to model results (background).
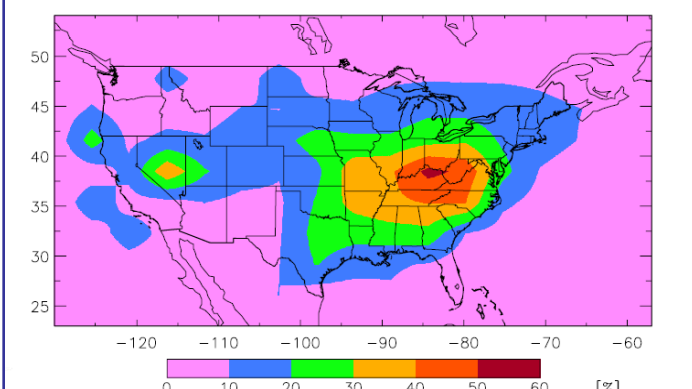
Percentage contributions from North American primary anthropogenic sources to total (wet + dry) annual mercury deposition simulated in the model for 2003 (not including re-emission), as determined from a sensitivity simulation with these sources shut off in the geographical domain covered by the figure.

Two patterns of mercury deposition: latitudinal gradient (driven by OH oxidation in sunny, wet areas) and regional enhancement (Midwest U.S.). We underestimate this enhancement in GEOS-Chem and simulate mostly dry deposition and Hg(P) wet deposition in this area.

Average simulated contribution of anthropogenic mercury sources in North America to deposition over the United States is 20%. The largest contributions from North American sources are in the Midwest U.S.

3. References

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