GLOBAL CYCLING AND DEPOSITION OF MERCURY:
CONSTRAINTS ON THE CONTRIBUTION FROM NORTH AMERICAN ANTHROPOGENIC SOURCES

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Introduction

We develop the first global 3D atmospheric model for mercury that accounts for cycling with the surface ocean and land reservoirs. We use this model to construct and interpret the pre-industrial and present-day global biogeochemical budgets of mercury. We then compare our model to measured seasonal and latitudinal patterns of wet deposition fluxes of mercury in the United States, to test our understanding of the factors controlling mercury deposition and the contribution from North American anthropogenic emissions. We estimate that North American anthropogenic emissions contribute 20% of total mercury deposition in the US (up to 50% in the industrial Midwest and Northeast).

GEOS-Chem Mercury Simulation

Model: GEOS-Chem version 7.04 (http://www.as.harvard.edu/ctm/geos/) at 4°x5° [1], using assimilated meteorology from NASA Goddard Earth Observing System (GEOS-4). Includes elemental (Hg(0)), divalent (Hg(II)), and primary particulate (Hg(P)) mercury.
Sources: GEIA emissions inventory for 2000 [2]; incorporates coupled land-ocean-atmosphere model [3,4,5].
Chemistry: Hg(0) oxidation by OH ($k = 9 \times 10^{-14}$ cm^3 s^{-1} [6,7]) and ozone ($k = 3 \times 10^{-20}$ cm^3 s^{-1} [8]); Hg(II) aqueous-phase, photochemical reduction.
Deposition: Wet deposition of Hg(II) and Hg(P) includes rainout and washout from large-scale and convective precipitation, and scavenging in convective updrafts [9]. Dry deposition of Hg(0) to land, of Hg(II), and of Hg(P) are simulated with a standard resistance-in-series scheme [10,11]. Dry deposition of Hg(0) to the ocean is simulated as part of a bi-directional exchange model [3].
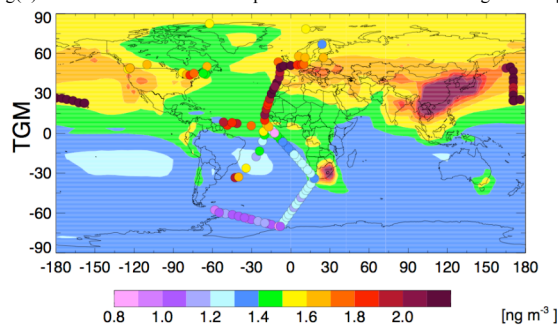

Annual average total gaseous mercury (TGM, Hg(0)+Hg(II)) concentrations in surface air. Model results (background, 2004) are compared to observations (circles) from long-term surface sites [12-18]

References

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(5) Selin NE, et al. (2008) Global Biogeochemical Cycles 22(22):GB2011. **(6)** Pal B & Ariya PA (2004) Environmental Science & Technology 21:5555-5566. **(7)** Sommar J, et al. (2001) Atmospheric Environment 35:3049-3054. **(8)** Hall B (1995) Water, Air, and Soil Pollution 80:301-315. **(9)** Liu H, et al. (2001) Journal of Geophysical Research 106(D11):12,109-112,128. **(10)** Wang Y, Jacob DJ, & Logan JA (1998) Journal of Geophysical Research 103(D9):10,713-710,726. **(11)** Wesely ML (1989) Atmospheric Environment 23(6):1293-1304. **(12)** Baker PGL, et al. (2002) Atmospheric Environment 36(14):2459-2465. **(13)** Ebinghaus R, et al. (2002) Atmospheric Environment 36(34):5267-5276. **(14)** EMEP (2005), EMEP measurement data, accessed via internet. **(15)** Environment Canada (2003) Canadian National Atmospheric Chemistry (NAtChem) Database (Meterological Service of Canada, Toronto). **(16)** Kellerhals M, et al. (2003) Atmospheric Environment 37(1003-1011). **(17)** Poissant L, et al. (2005) Atmospheric Environment 39(7):1275-1287. **(18)** Weiss-Penzias P, et al. (2003) Environmental Science and Technology 37:3755-3763. **(19)** Mercury Deposition Network data, http://nadp.sws.uiuc.edu/mdn/

Present-day vs. Pre-industrial Mercury Cycles

*Inventories in Mg, rates in Mg y^{-1}*
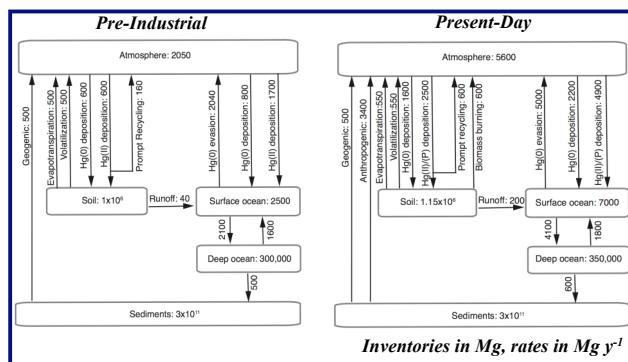
Mercury in the model has a relatively short lifetime in the atmosphere (0.55 y) and in the surface ocean (0.60 y) while its lifetime in soils is much longer (1000 y). Emitted Hg(0) is transferred to the soil and deep ocean reservoirs in equal amounts, even though deposition to the ocean is twice as large.
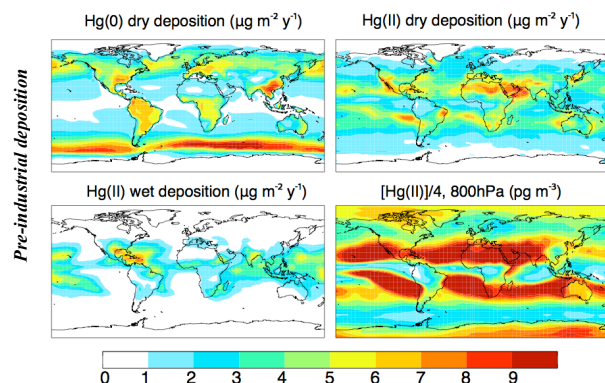

Dry deposition is highest in the subtropics, reflecting high boundary layer concentrations of Hg(II) brought down by subsidence in the general circulation.

Wet deposition is highest where this subtropical downwelling interacts with regional circulation regimes promoting precipitation, such as in the southeast U.S.

Deposition from North American Sources
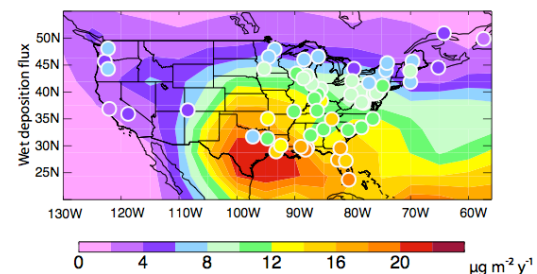
GEOS-Chem reproduces the large-scale spatial patterns in wet deposition data from the Mercury Deposition Network (MDN) for 2004-2005 [19] (r^2=0.73, mean bias 1.7%). The latitudinal gradient explains 60% of variation in the MDN data (r^2=0.60).
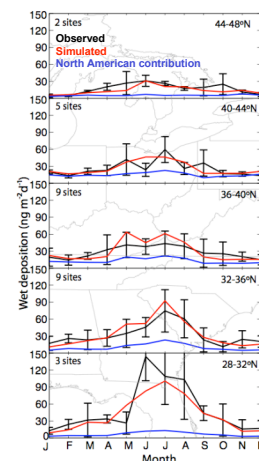
We attribute seasonal variation at northern latitudes to a greater scavenging efficiency of Hg(II) in summer.

The amplitude of seasonal variation in wet deposition decreases with increasing latitude.

We attribute seasonal variation at subtropical latitudes to a seasonal precipitation associated with deep convection, scavenging Hg(II) from the free troposphere.

The overall contribution of North American sources to deposition in the U.S. is 20%, but it is up to 50% in the industrial Midwest/Northeast.