Historical accumulation of anthropogenic mercury in soils
Simulated with the GEOS-Chem coupled atmosphere-ocean-land model

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GEOS-Chem global mercury model

3D Atmosphere
Vertical & Horizontal Transport

Deposition to oceans
Wet scavenging $\text{Hg}^\text{II} & \text{Hg}^\text{P}$
Sea salt uptake of $\text{Hg}^\text{II}$
Surface ocean uptake of $\text{Hg}^0$

Surface ocean evasion

Geogenic
Anthropogenic

Biomass burning
Respiration

Prompt Reemissions

Deposition to land
Wet scavenging $\text{Hg}^\text{II} & \text{Hg}^\text{P}$
Dry deposition of all species

Converting $\text{Hg}^0$ to $\text{Hg}^\text{II}$ in clouds

Oxidation of $\text{Hg}^0$ by Br atoms

2D slab surface ocean
3300 Mg

2D surface organic soils

Coupling to Global Terrestrial Mercury Model
160,000 Mg

Particle removal
Net vertical transport

Basin-scale fixed subsurface Hg concentrations

Surface reservoirs in Mg
Fluxes in Mg a$^{-1}$

$\text{Hg}^0$ fluxes in red
$\text{Hg}^\text{II}+\text{Hg}^\text{P}$ fluxes in blue
Global Terrestrial Mercury Model

- Terrestrial mercury model based on the CASA biogeochemical carbon model
- GEOS-Chem atmosphere model run with preindustrial and historical industrial & mining emissions
- Monthly deposition fluxes input to surface pools to drive GTMM
- Deposited mercury that is not quickly revolatilized is partitioned to soil pools based on carbon mass
- Fraction of mercury released to atmosphere during respiration

modified from Smith-Downey, et al.
Preindustrial soil Hg distribution

In steady-state, preindustrial conditions, mercury accumulates in the most long-lived, recalcitrant pools.

Shift in oxidation of Hg$^0$ → deposition of Hg$^{II}$ → soil Hg accumulation to higher latitudes in the simulation with oxidation by Br.
Total Gross Mercury Deposition to Land 1840-2000

Regionally Resolved Historical Emissions & Hg$^0$ Oxidation by Br
Enhancement of soil Hg due to anthropogenic emissions since 1840

- Soil accumulation of anthropogenic mercury 1840-2000: 16,000 Mg (7% of emitted Hg)
- Sensitive to magnitude and spatial distribution of deposition
• Greatest magnitude of anthropogenic mercury accumulation in slow & armored pools, while greatest relative enhancement in fast pool.

• Most emissions over time period from Europe and North America, while deposition and accumulation is more distributed.
Mercury accumulation relative to carbon stocks

Where mercury deposition is moderate or high but carbon turnover is slow, mercury accumulates.

If the fraction of mercury released during decomposition were greater, the distribution of mercury would more closely resemble that of carbon. (Smith-Downey et al. 2010)
Terrestrial emissions

- **Hg° Revolatilization**
  - Preindustrial: 260 Mg a\(^{-1}\)
  - Present: 860 Mg a\(^{-1}\)

- **Hg\(^{II}\) Photoreduction**
  - Preindustrial: 180 Mg a\(^{-1}\)
  - Present: 360 Mg a\(^{-1}\)

- **Respiration**
  - Preindustrial: 200 Mg a\(^{-1}\)
  - Present: 440 Mg a\(^{-1}\)

The images show maps of the world with color gradients indicating the concentration of mercury emissions in units of μg m\(^{-2}\) a\(^{-1}\).
Coupled atmosphere-terrestrial model

- Capacity for long-term coupled runs to study historical accumulation of anthropogenic mercury in soils, response to climate change

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**Modeled TGM at Northern Mid-latitude Stations**

*version with OH/O$_3$ oxidation & historical emissions scaled to North American industry*

- GTMM fluxes: calculated monthly
- GEOS-Chem fluxes: calculated hourly

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**Atmospheric pools**

- Bidirectional Exchange of Hg$^0$
- Photoreduction of Hg$^II$
- Wet Deposition
- Respiration

**Soil pools**

- Leaf & Surface Pools
- Leaf Senescence

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**Bidirectional Exchange of Hg$^0$**

- Dry Deposition
- Wet Deposition

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**Leaf & Surface Pools**

- Leaf Senescence

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**Soil pools**

- GTMM fluxes: calculated monthly
- GEOS-Chem fluxes: calculated hourly
Timescales of anthropogenic mercury accumulation

- Fast terrestrial, atmosphere, and surface ocean respond most quickly to changes in emissions
- Longer-lived reservoirs record cumulative emissions
- Biggest long-term sink for anthropogenic mercury in the deep ocean
- Armored soil pool of comparable magnitude to deep ocean mercury reservoir, but less relative increase in mercury storage

Streets et al. in prep; courtesy of Elsie Sunderland