Drivers of mercury variability in the Arctic atmosphere and surface ocean: impacts of changing climate and emissions

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SUMMARY

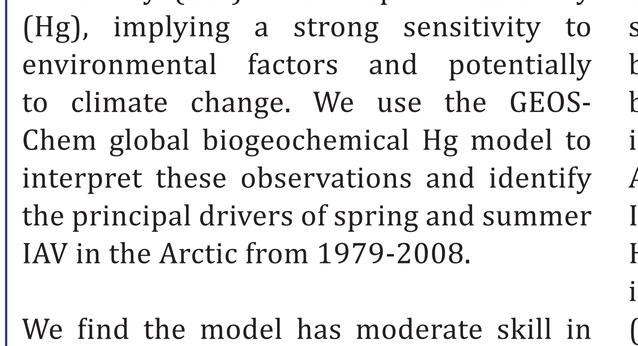
Long-term observations at Arctic sites (Alert and Zeppelin) show large interannual variability (IAV) in atmospheric mercury (Hg), implying a strong sensitivity to environmental factors and potentially to climate change. We use the GEOS-Chem global biogeochemical Hg model to interpret these observations and identify the principal drivers of spring and summer IAV in the Arctic from 1979-2008.

We find the model has moderate skill in simulating the observed atmospheric IAV at the two sites (r ~ 0.4) and successfully reproduces a long-term shift at Alert in the timing of the spring minimum from May to April (r = 0.7).

Principal component analysis indicates that IAV in the model can be explained by a climate mode with high temperatures, low sea ice fraction, low cloudiness, and shallow boundary layer. This mode drives decreased bromine-driven deposition in spring and increased ocean evasion in summer. In the Arctic surface ocean, we find that modeled IAV is dominated by the meltwater flux of Hg previously deposited to sea ice, which is largest in years with high solar radiation (low cloudiness) and a large transition in air temperature from a cold spring to a warm summer. Projected future increased cloudiness and stronger warming in spring than summer may thus lead to decreased Hg inputs to the Arctic Ocean.

30-year Arctic Hg simulation

The GEOS-Chem global biogeochemical Hg model (v9-01-02) was run from 1979-2008, driven by meteorology from the MERRA retrospective assimilated dataset at 4x5° horizontal resolution. Anthropogenic emissions are based on the *Streets et al. (2011)* inventory. The Arctic simulation is based on *Fisher et al. (2012)* with minor updates to bromine release, net primary productivity, UV-driven oceanic photoreduction, and freshwater discharges.

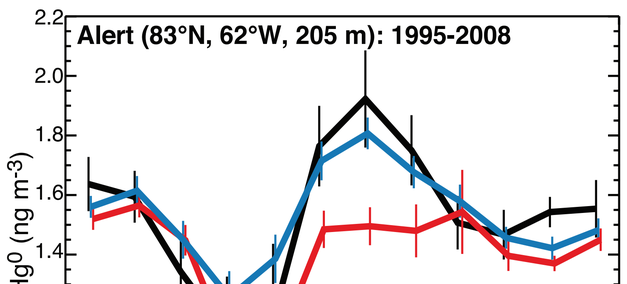


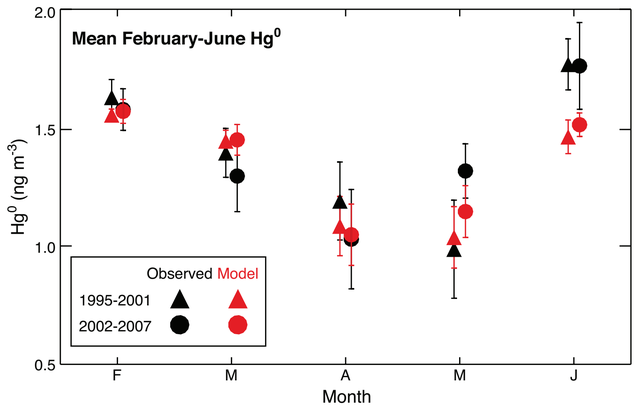
The figure shows mean simulated Hg^0 in Arctic surface air averaged over the period of observational record (1995-2008).

Also shown are observed long-term means from
Alert: 1995-2008 (*Cole et al., 2013*)
Zeppelin: 2000-2008 (*Berg et al., 2013*)
Pallas: 1996-2008 (*Aas & Brevik, 2012*)

The black line delineates the boundary of the "Arctic Ocean air mass", defined as all model grid squares in 68-90°N with at least 20% ocean area.

- The observed Hg^0 seasonal cycle is characterised by a **spring minimum** caused by Atmospheric Mercury Depletion Events (AMDEs) and **summer maximum** caused by ocean evasion.
- The seasonal cycle is damped at Zeppelin due to the high altitude of the measurement site.
- **GEOS-Chem** Hg^0 averaged over the Arctic Ocean air mass **captures these key features**.
- GEOS-Chem sampled at Alert underestimates the summer peak, likely due to an overestimate in MERRA sea ice cover in the region.
- Model low bias at Zeppelin in fall appears to be caused by an overly strong simulated vertical gradient.





- GEOS-Chem reproduces the **shift at Alert** in the timing of the spring minimum from May to April observed by *Cole & Steffen (2010)*. This can be attributed to local warming in May and cooling in April.
- The model indicates that this **shift is not representative of the Arctic as a whole** for either the 1995-2007 period, or for the entire 30-year simulation.

Acknowledgements:

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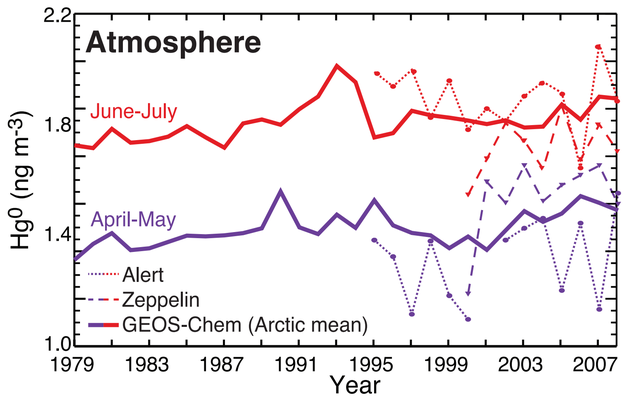
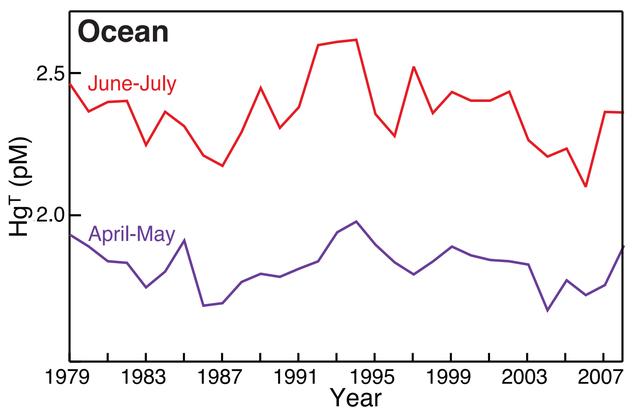
Trends and interannual variability



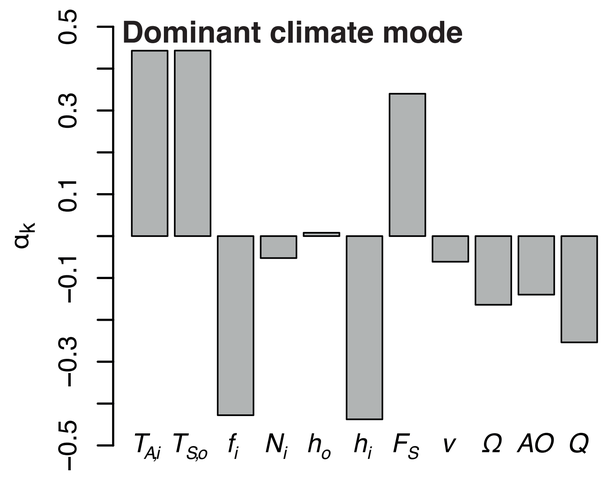
- **Trends** in atmospheric Hg^0 are **negligible or absent** in observations (*Cole et al., 2013; Berg et al., 2013*) and in GEOS-Chem.
- Observed **IAV is large** and not coherent between sites.
- GEOS-Chem Arctic-mean Hg^0 falls between the observations, and can reproduce to some extent the observed IAV ($r \sim 0.4$).

- No long-term observations exist for Hg in the Arctic surface ocean. GEOS-Chem is consistent with present-day observations.
- The simulation shows **no trends but large IAV in surface ocean Hg**. Both concentrations and IAV peak in summer.

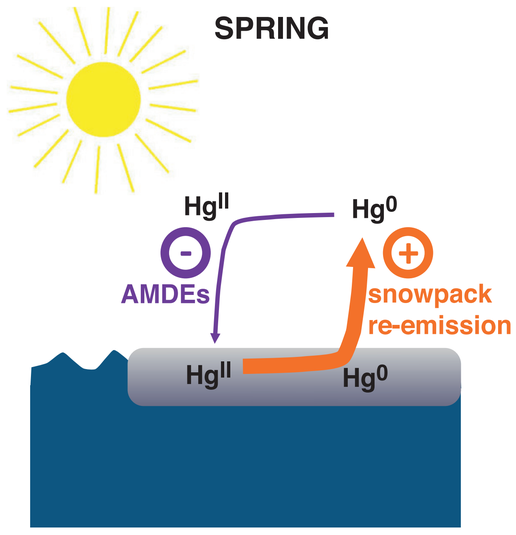


Factors driving interannual variability

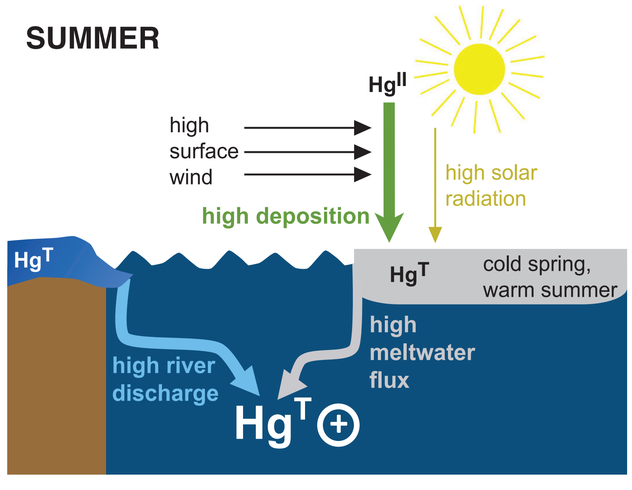
We performed a principal component analysis and regression to identify the environmental factors driving IAV of atmospheric Hg^0. Environmental variables considered were: temperature ($T_{A,i}$, $T_{S,o}$), sea ice fraction (f_i, N_i), planetary boundary layer height (h_o, h_i), solar radiation (F_S), wind speed(v), ozone column(Ω), freshwater discharge(Q), and Arctic Oscillation Index (AO).



- The first principal component (climate mode) was the same in spring and summer. **Positive phases indicate "warm" years**.
- This mode accounts for **80% of the IAV** in simulated Hg^0 **in spring** and **40% in summer.** It is also moderately correlated with IAV in the observations ($r \sim 0.4$).
- **Positive ("warm") phases increase atmospheric** Hg^0 in both spring and summer, although the mechanisms differ.



- Simulated IAV in surface ocean Hg is **dominated by IAV in the meltwater flux**.
- The **meltwater** flux is **largest** in years with high solar radiation (**low cloudiness**) and when a **cold spring** (frequent AMDEs) is followed by a **warm summer** (rapid melt limiting re-emission from snow).
- These variables combined with May river discharge and surface wind speed can explain 53% of the IAV in simulated surface ocean Hg in summer.



Current and projected future changes associated with climate warming (high air temperatures, low sea ice area, stronger warming in spring than summer, and high cloudiness) may decrease Hg levels in the Arctic Ocean.

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