Hg/POPs Working Group Updates

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on behalf of all co-chairs
08/24/2021

Contents

- New Hg chemistry
- New Hg isotope model
- Updated Hg^0 vegetation uptake

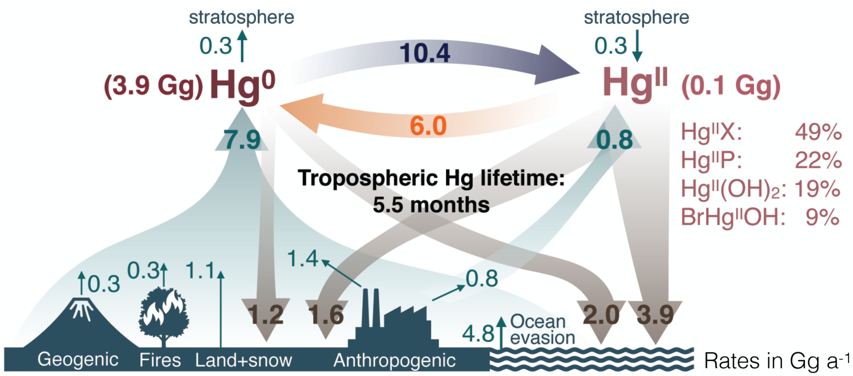
GEOS-Chem Hg chemistry updates

- Hg chemistry updated to include computational and lab results since 2017. Updates include:
  - Hg(0)+OH reaction; Hg(I)+ozone reactions
  - Hg(II) gas-phase photolysis
  - Improved Hg(II) gas-particle partitioning (kinetic process; organic+inorg HgP species)
- Structural updates to mercury_mod
  - Many more tracers than before (several Hg(II) species)
  - Online photolysis with FAST-JX
  - Chemistry solved in KPP
  - Oxidant & aerosol fields from version 12.9 (halogens from Wang et al. 2021)
- Tagged Hg simulation no longer works (chem involves many species now)
- Updated code being ported to version 13 by GCST & Mike Long

V. Shah (Harvard) et al. ES&T 2021

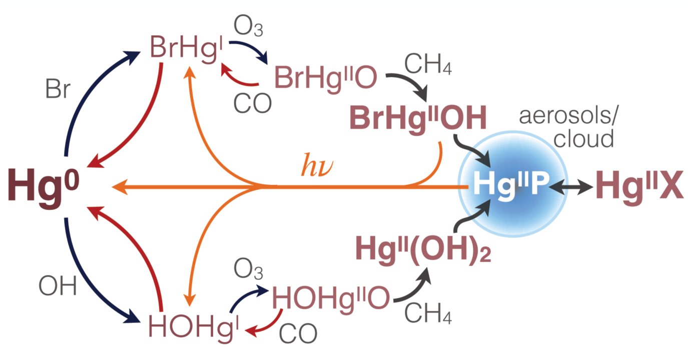
Key results of the Hg chemistry update

Global tropospheric Hg budget



- Atmospheric Hg lifetime consistent with Horowitz et al. (2017).
- Small changes in deposition spatial patterns.

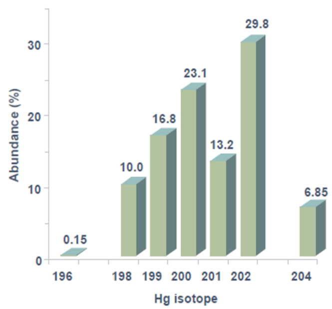
Major Hg redox pathways



- Net Hg^0->Hg^{II} oxidation occurs via Br and OH almost equally
- Ozone is the main second-step oxidant
- Hg^{II} reduction in aqueous phase is faster than in gas phase
- $Hg^{II}X$ is modeled as $HgCl_2$

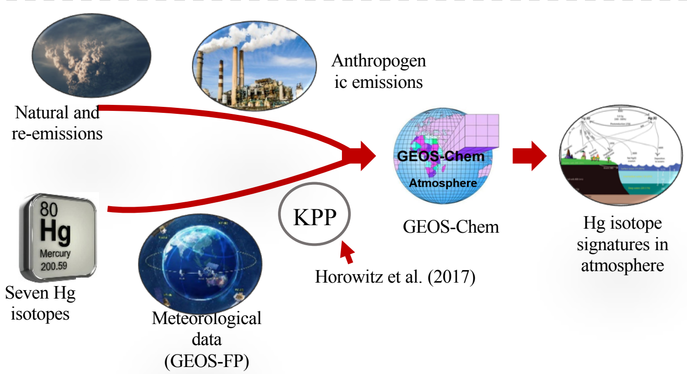
V. Shah (Harvard) et al. ES&T 2021

Mercury Isotope model



- Hg has seven stable isotopes, which has been widely used for source apportionment and processes determination.
- It is challenging to link the observed isotope fractionation data to global Hg cycling.

Method

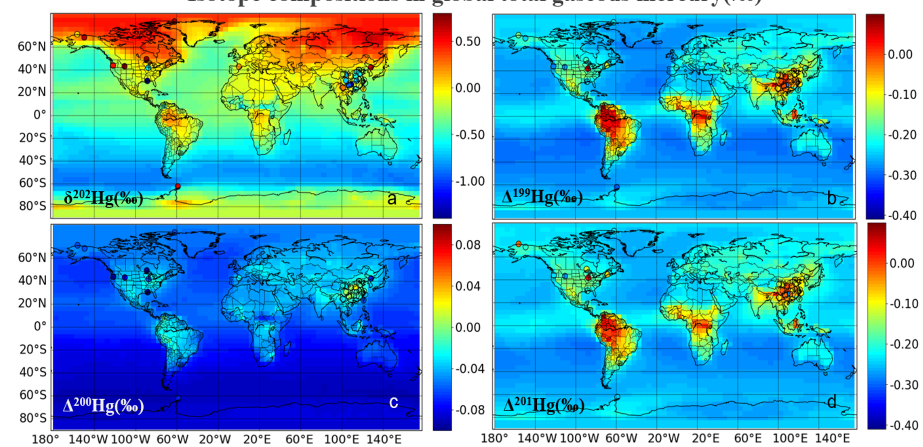


- It is the first 3-D model for atmospheric Hg isotopes.
- Hg isotopic inventory of natural, re-emission, and anthropogenic sources are developed.
- We consider all the seven stable isotopes of Hg for Hg(0), Hg(Ⅱ)$_g$ and Hg(Ⅱ)$_P$, which results in a total of 21 advected tracers.

- Process-based isotope fractionations are collected and integrated into the GEOS-Chem model platform.
- The chemical mechanism is implemented from Horowitz et al. (2017) using the Kinetic PreProcessor (KPP).

Results

Isotope compositions in global total gaseous mercury(‰)

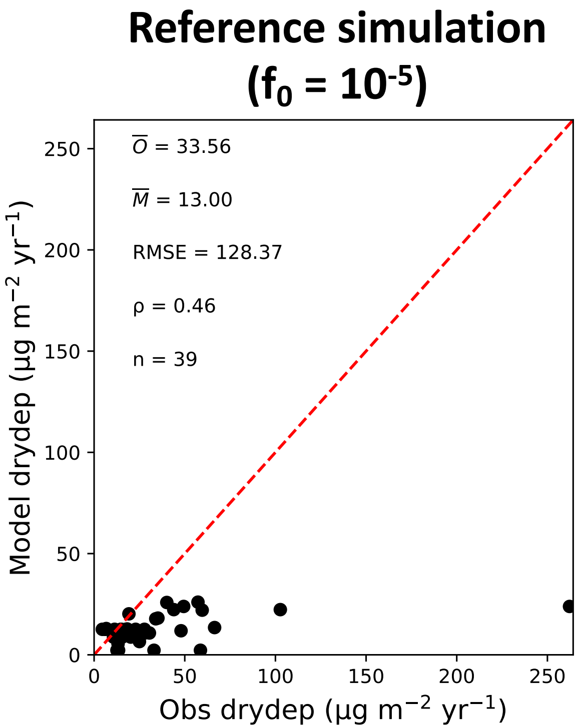


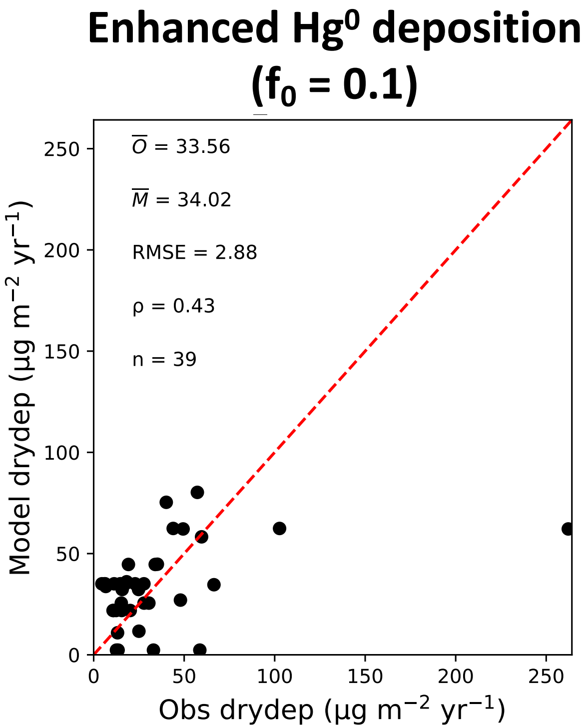
- Our simulated isotope compositions of TGM are broadly comparable with available observations across different global regions.
- Source emissions have great impact on regional isotope composition of TGM.
- Hg(0) uptake by terrestrial surface (e.g. vegetation) and Hg(Ⅱ) reduction play a key role in changing isotope composition of global TGM.
- Codes available in next couple of versions

Z. Song, Y. Zhang (NJU) ES&T submit soon

Constraining Hg vegetation uptake in GEOS-Chem using available measurements

- Reference GEOS-Chem (v12.8.1) simulation underestimates Hg dry deposition measured by litterfall and throughfall fluxes
- Tests are underway with enhanced Hg^0 reactivity (f_0) in the dry deposition scheme, which better agrees with measurements and recent studies (e.g. Obrist et al., *PNAS*, 2021; Fu et al, *ES&T*, 2021)
- Adjustments to Hg reduction rate will be needed to compensate for too low Hg^0 concentration and wet deposition
- Expected submission of paper: December 2021





Observations extended from previous reviews: Wright et al., (2016); Zhou et al. (2020)

A. Feinberg, T. Dlamini, N. Selin (MIT), V. Shah (Harvard)