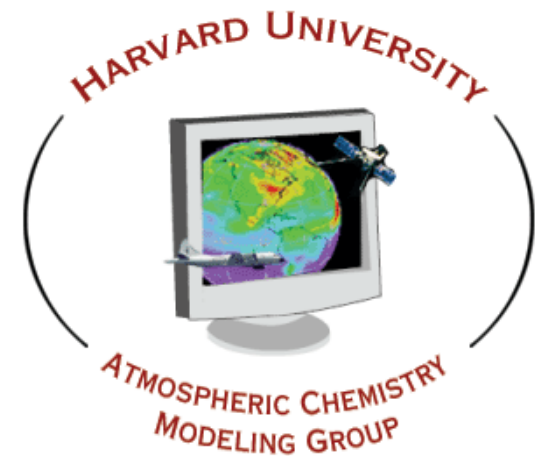


# Hg(II) Gas-Particle Partitioning and Its Effect on Global Mercury Deposition

10<sup>th</sup> ICMGP – Halifax 2011

**Helen Amos (amos@fas.harvard.edu)**

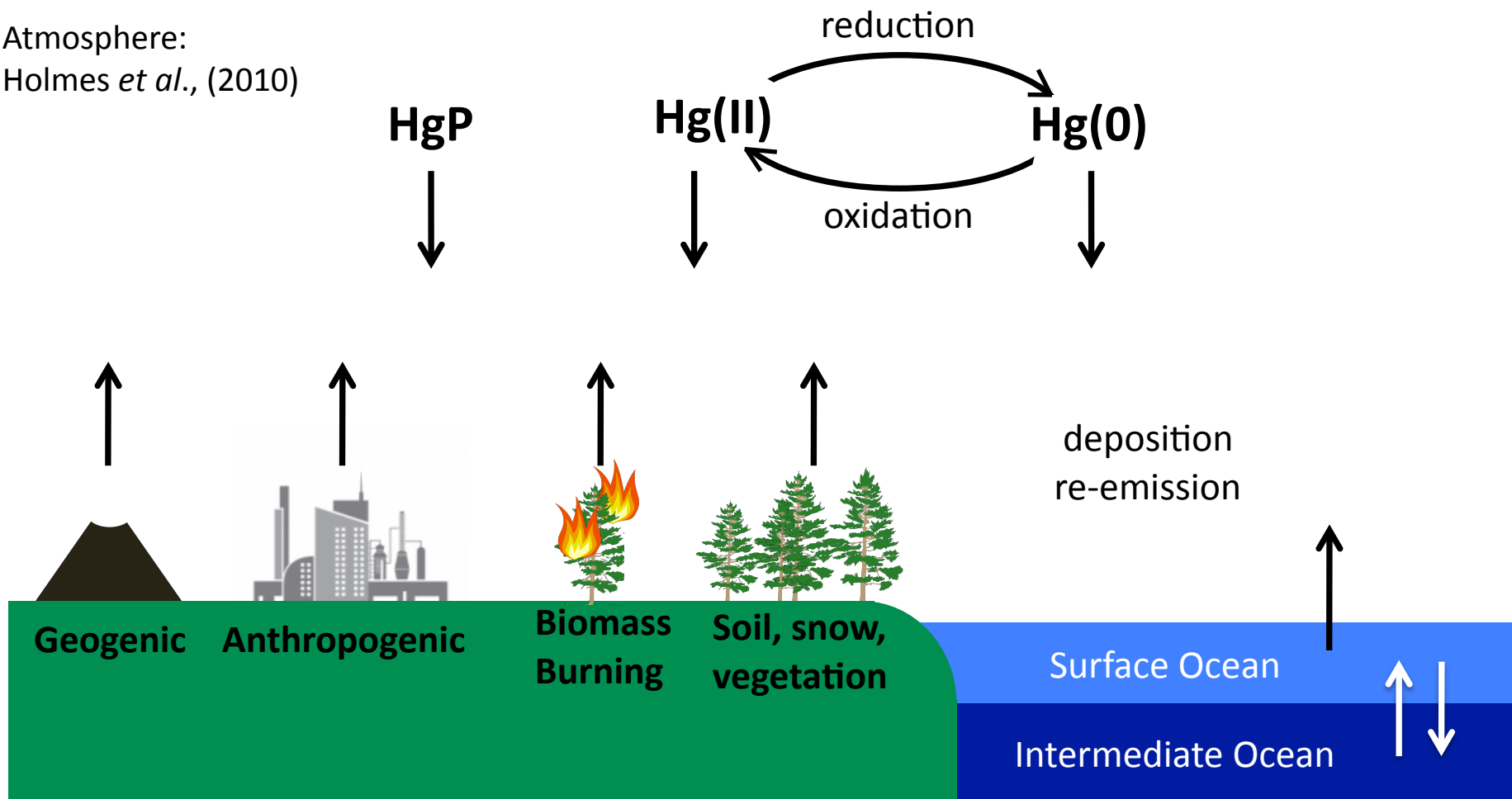
D. Jacob, C. Holmes, E. Corbitt, R. Yantosca, J. Fisher, A. Steffen, E. Galarneau, J. Graydon, V. St Louis, M. Gustin, E. Edgerton, R. Talbot, L. Murray, Y. Zhang, E. Sunderland



# 3D Global Mercury Simulation in GEOS-Chem

Atmosphere:

Holmes *et al.*, (2010)



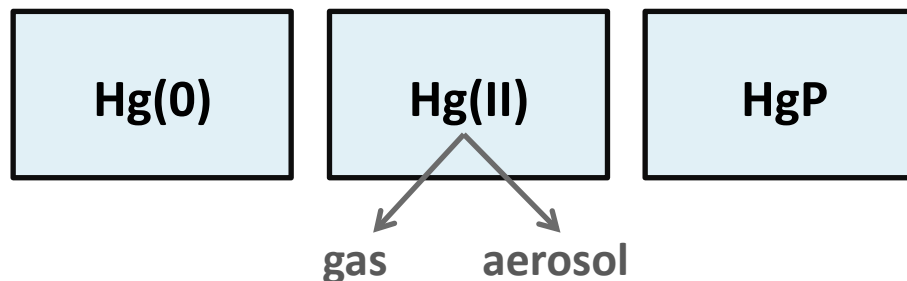
Land: Selin *et al.*, 2008; Holmes *et al.*, 2010

Ocean: Soerensen *et al.*, 2010

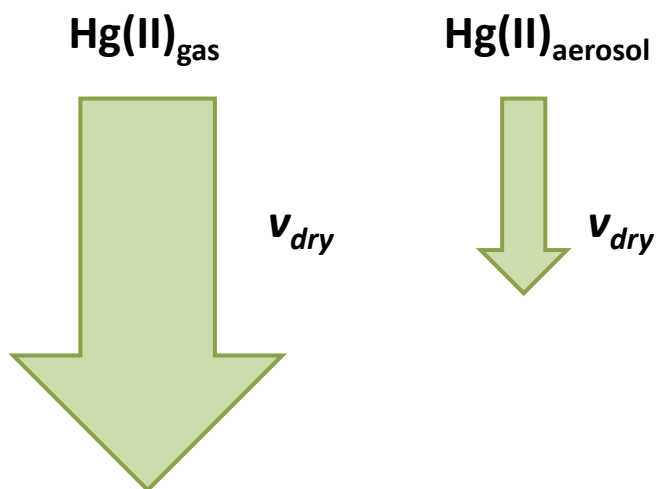
**This work: Implementing mechanistic  $\text{Hg(II)}$  gas-particle partitioning and other updates to the atmosphere.**

# Improving Simulated Hg(II): Phase Partitioning

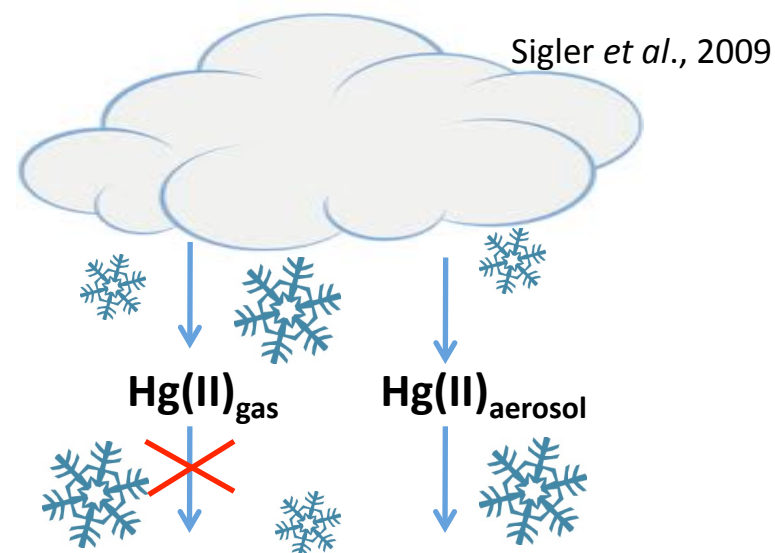
GEOS-Chem Hg Simulation  
--  
3 Atmospheric Tracers



## Dry Deposition



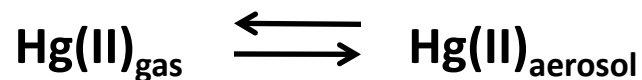
## Snow Scavenging



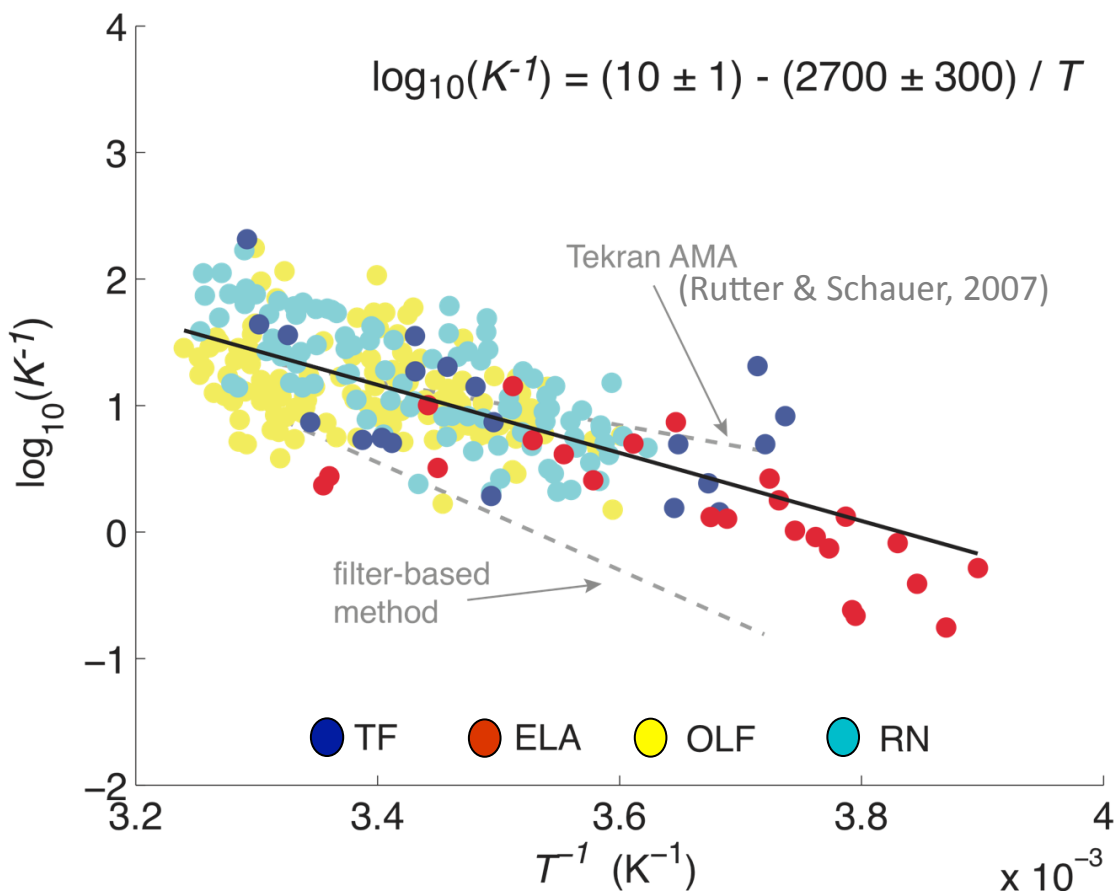
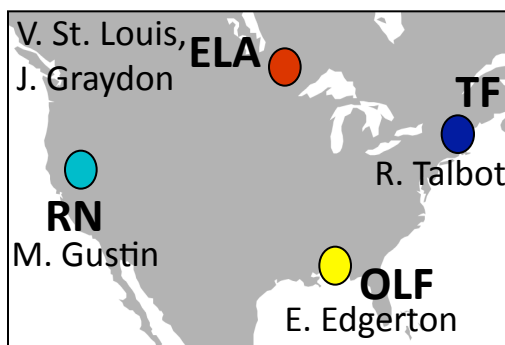
Gases and aerosols → different removal processes

# Thermodynamic Representation of Hg(II) Partitioning

Amos *et al.*, (in prep)



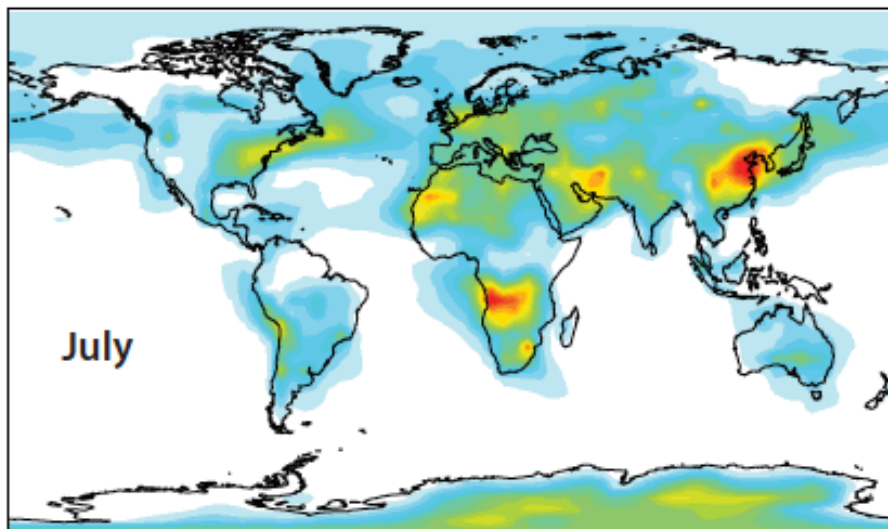
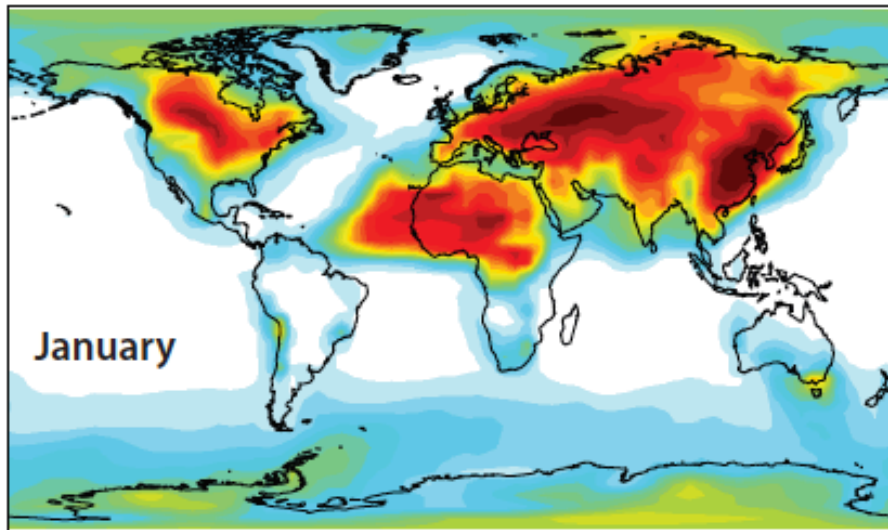
$$K^{-1} = \frac{[\text{RGM}]}{[\text{PBM}] / \text{PM}_{2.5}}$$



**Hg(II) gas-aerosol partitioning at all sites can be successfully described by a standard thermodynamic relationship for semi-volatile species.**

# Hg(II) Partitioning Parameterization in GEOS-Chem

Fraction of Hg(II) Partitioned to Aerosol Phase  
GEOS-5 2007-2009 Mean, Surface Level



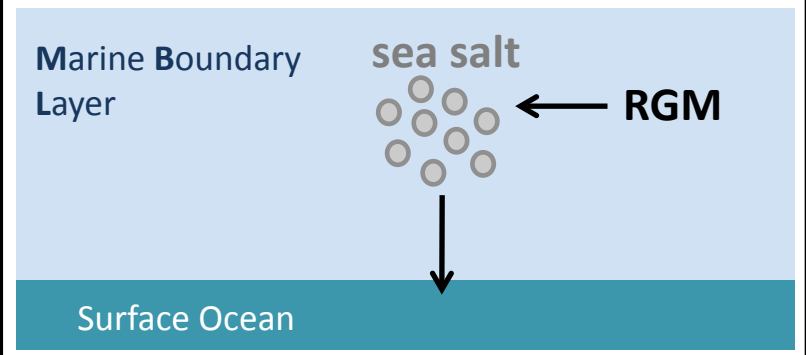
Partitioning depends on:

- temperature
- aerosol burden (excludes sea salt)

**New model capability to have seasonality in Hg(II) partitioning.**

**Side Note:**

RGM is taken up onto seasalt.  
(Holmes *et al.*, 2010; 2009)



# Global Hg Deposition: Sensitivity to Hg(II) Phase

2007-2009 Mean

## Simulation A:

Mechanistic Hg(II) gas-particle partitioning.

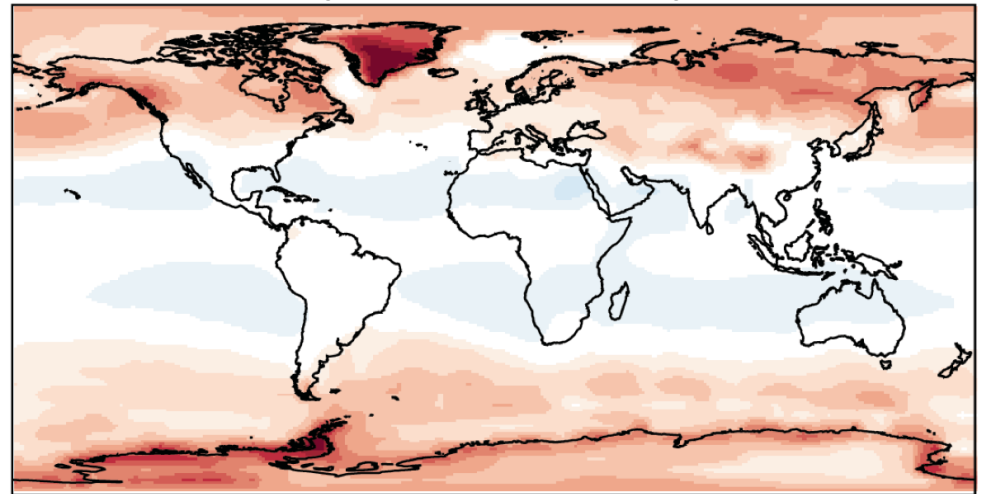
## Simulation B:

Assume Hg(II) is 100% gas.

## Gross Hg(II)+HgP deposition includes:

- wet deposition
- dry deposition
- loss via sea salt

Difference in Annual Gross Hg(II)+HgP Deposition  
( % =  $100 * A-B/B$  )

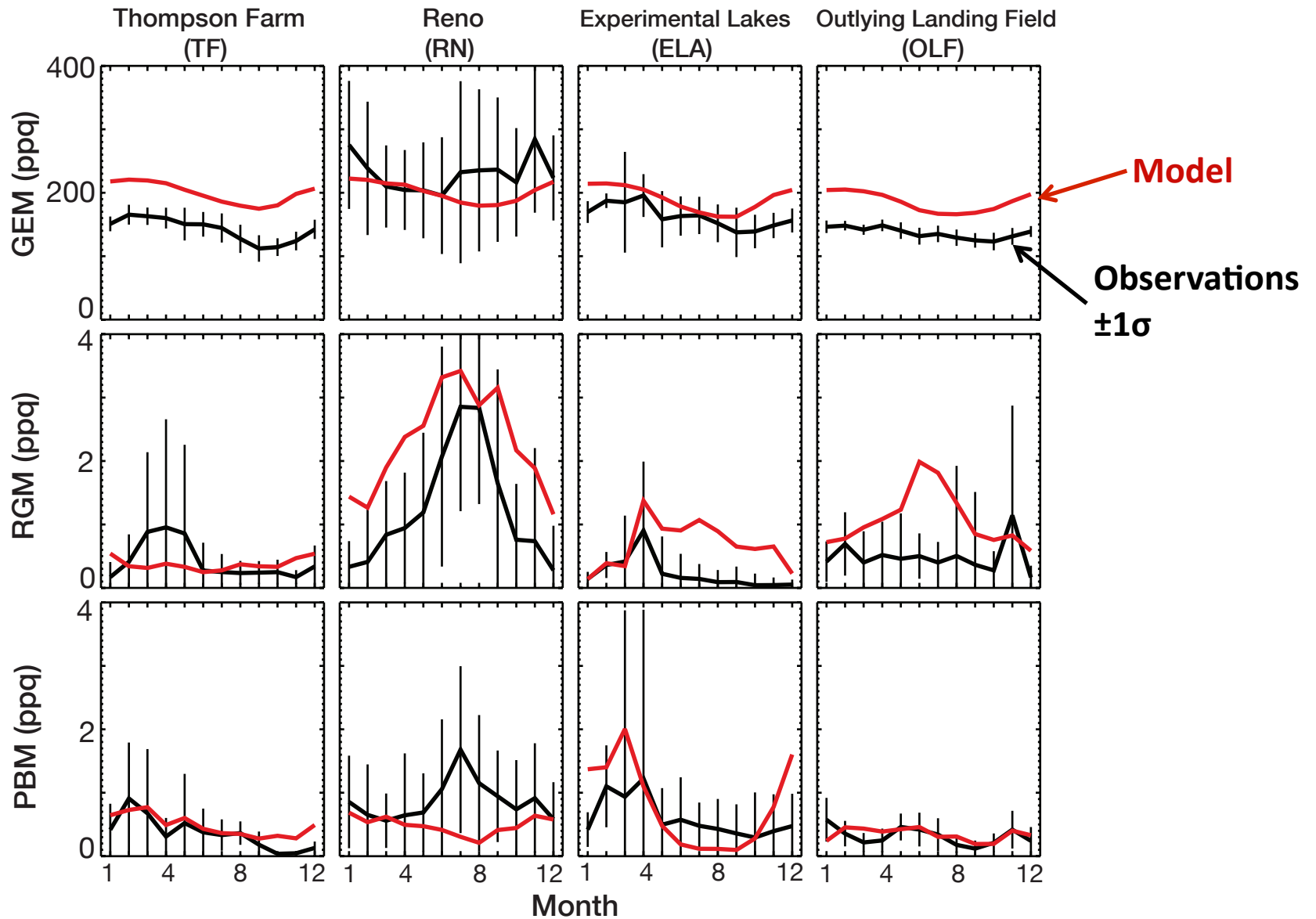


-75 0 75 %

**Large difference over mid-latitudes and poles due to snow scavenging. Falling snow scavenges PBM, but not RGM (Sigler *et al.*, 2009; Lombard *et al.*, 2011 – ACPD).**

**Small difference over mid-tropical open ocean because non-sea salt aerosol burdens are very low.**

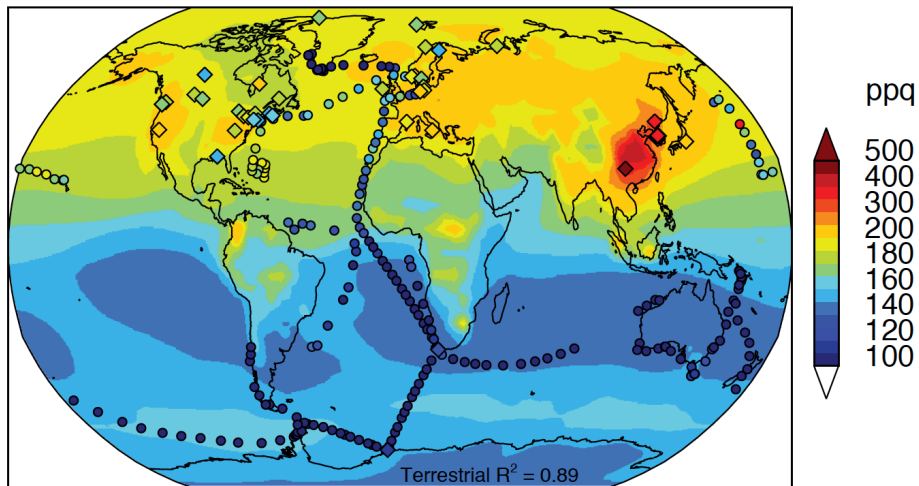
# Hg Seasonality: Observed and Simulated



Seasonality of RGM and PBM is controlled by more than just Hg(II) partitioning.

# Annual Mean Hg: Simulated and Observed

## Annual Mean Surface TGM

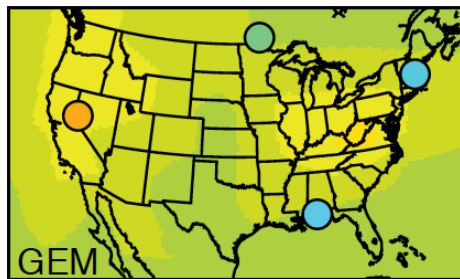


### Global TGM

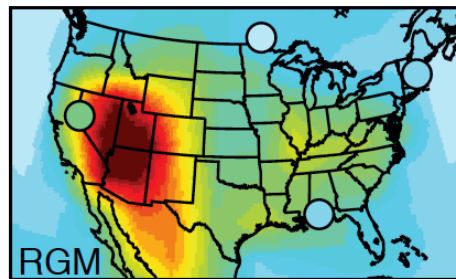
Observed	209 ± 112 ppq
Model	206 ± 47 ppq

**TGM is higher than in *Holmes et al. (2010)* – it's ocean evasion.**

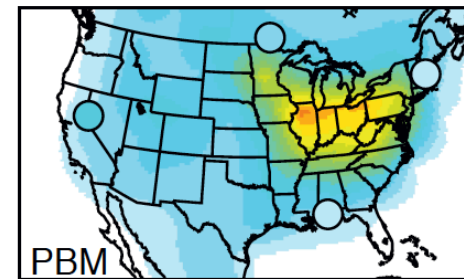
Ensemble of TGM observations from *Holmes et al. (2010)*, *Selin et al. (2007)*



100 200 300 ppq



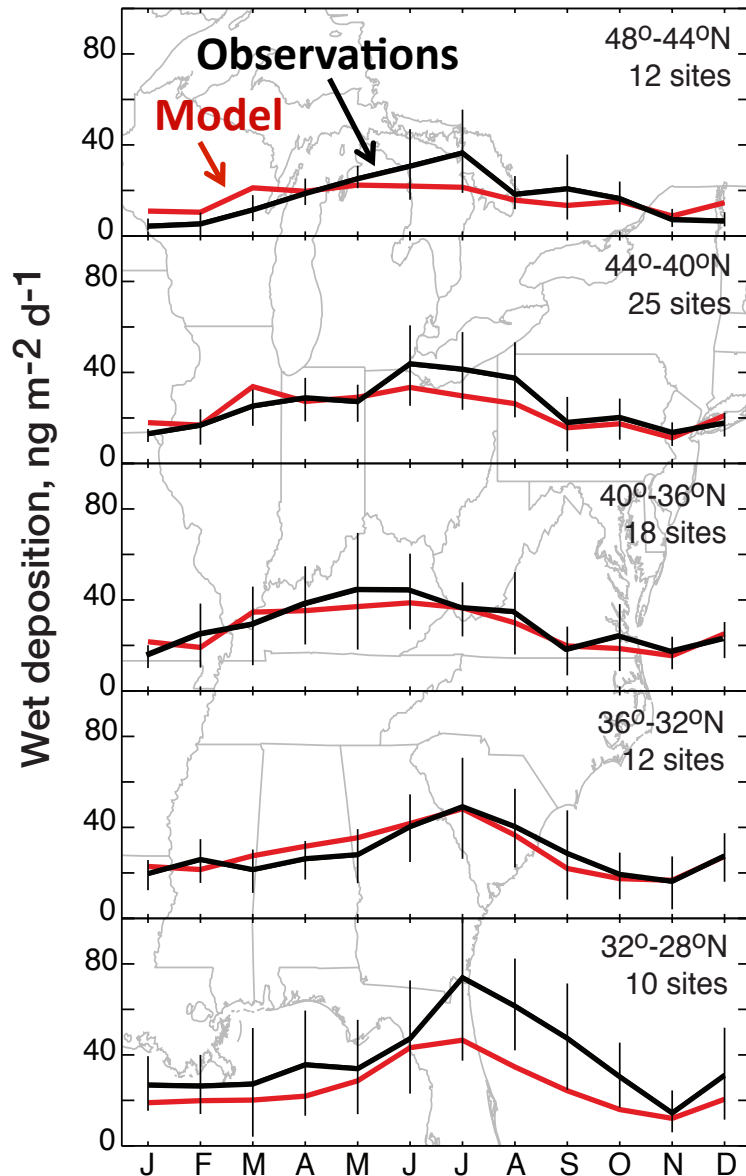
0.00 2.00 4.00 ppq



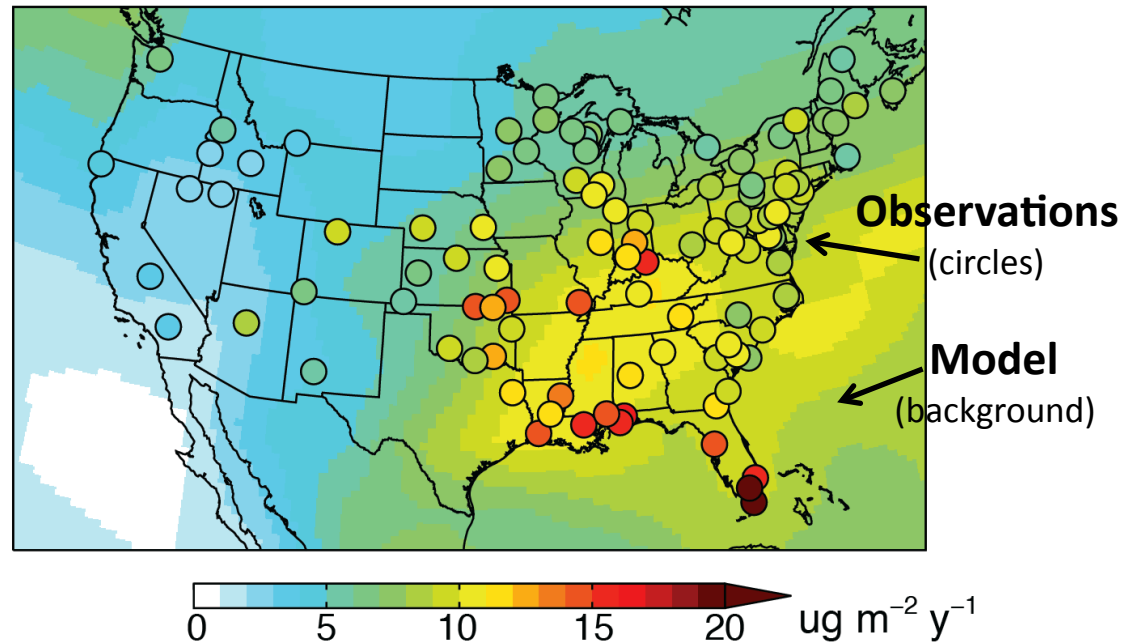
0.00 2.00 4.00 ppq

**High RGM in Southwest is from subsidence. High PBM is collocated with high anthropogenic emissions and high aerosol burdens.**

# Hg Wet Deposition Over the USA (2007-2009)



## Hg Total Annual Wet Deposition

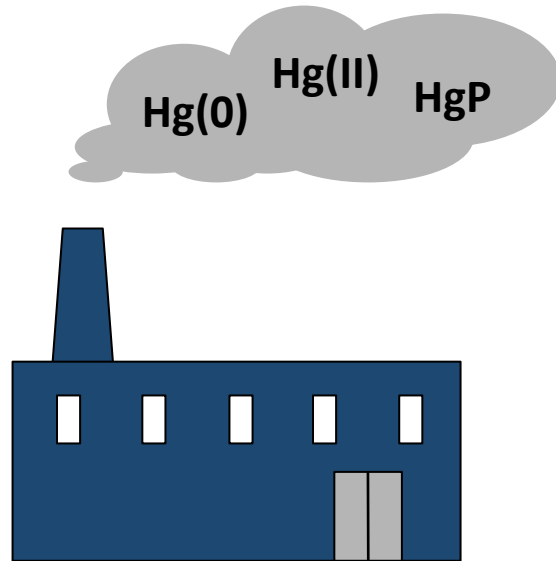


**Mechanistic Hg(II) partitioning improves simulated wet deposition.**

**Omitting cold temperature RGM scavenging in clouds is necessary to match wintertime observations.**

**Low bias in Gulf States persists across model versions (Selin & Jacob, 2008; Holmes *et al.*, 2010).**

# Uncertainty in Emission Inventory HgP



Primary Anthropogenic Emissions

What is primary HgP?

Refractory or semi-volatile?

Fine or coarse?

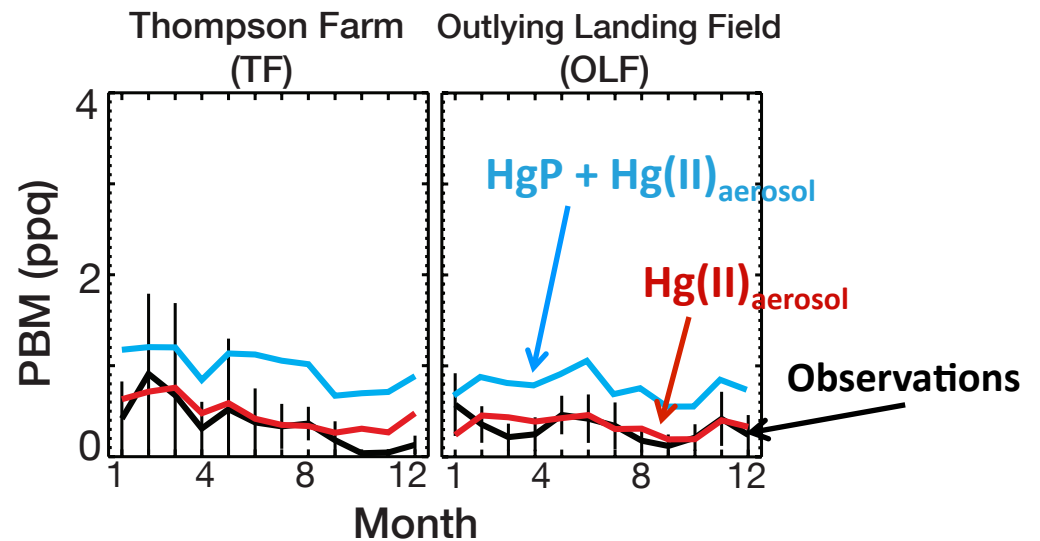
Measured by a Tekran unit?

## Total HgP near Detroit, Michigan

Urban: 37% coarse

Rural: 20% coarse

Gildemeister *et al.*, 2005



# Summary

- **Hg(II) gas-particle partitioning across sites can be described by a single thermodynamic relationship.**
- **Treating Hg(II) partitioning increases Hg(II) deposition to mid-latitudes and poles.**
- **US: maximum PBM from anthropogenic emissions and maximum RGM from subsidence.**
- **Excellent agreement with Hg deposition observations from Mercury Deposition Network (MDN). Low bias in Gulf States still persists.**

## Critical uncertainties:

- **Is primary HgP refractory or semi-volatile? Fine or coarse?**
- **Is primary HgP quantified by a Tekran?**