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Photochemical Models are Unable to Simulate High Oxidized Mercury Measured in the Western United States

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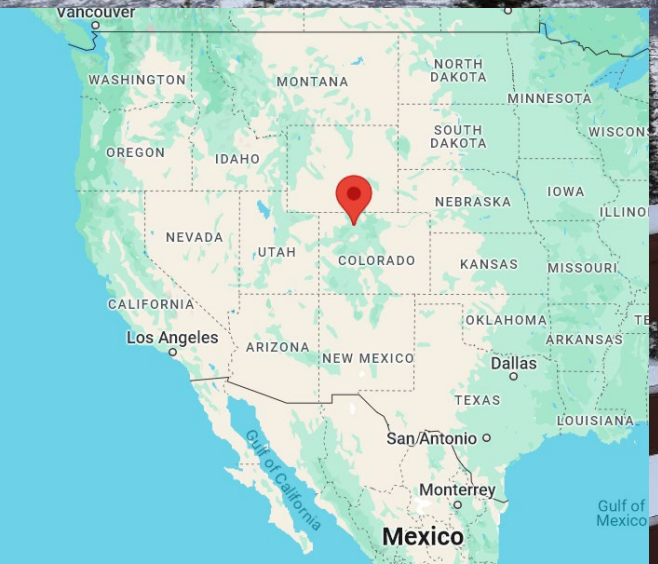
(Tyler Elgiar is now with the U.S. Bureau of Land Management)

Terminology

Hg^0 = gas-phase elemental mercury

Hg^{II} = all atmospheric oxidized mercury, including the gas and particle phases

Measurement Method



Measurement Method

Dual-channel measurement system:

1. Cation-exchange membranes to capture Hg^{II}
2. Thermal converter to transform all atmospheric Hg to Hg^0

- Hg^{II} determined by difference
- 1-hour detection limit of ~10 pg m^{-3}
- 100 ± 8% recovery of $HgBr_2$ in ambient air using a permeation tube-based calibrator
- Calibration was SI-traceable by two different methods

Details in Elgiar et al., 2024

High Hg^{II} Routinely Measured

	Mar 12–Oct 10 2021 (this study)	**Fain et al., 2009 (KCl denuder)**
average Hg^{II} (pg m^{-3})	103 ± 50	20 ± 21
max Hg^{II} (pg m^{-3})	520	137

from Elgiar et al., 2024

Atmospheric Modeling

- GEOS-Chem model with Shah et al., 2021 chemistry
- 2° X 2.5° resolution (comparison against 0.25° X 0.3125° resolution conducted)
- Goddard Earth Observing System-Forward Processing (GEOS-FP) meteorology
- Emissions from Horowitz et al., 2017 and Streets et al., 2019; current biomass burning events used

Sensitivity Tests

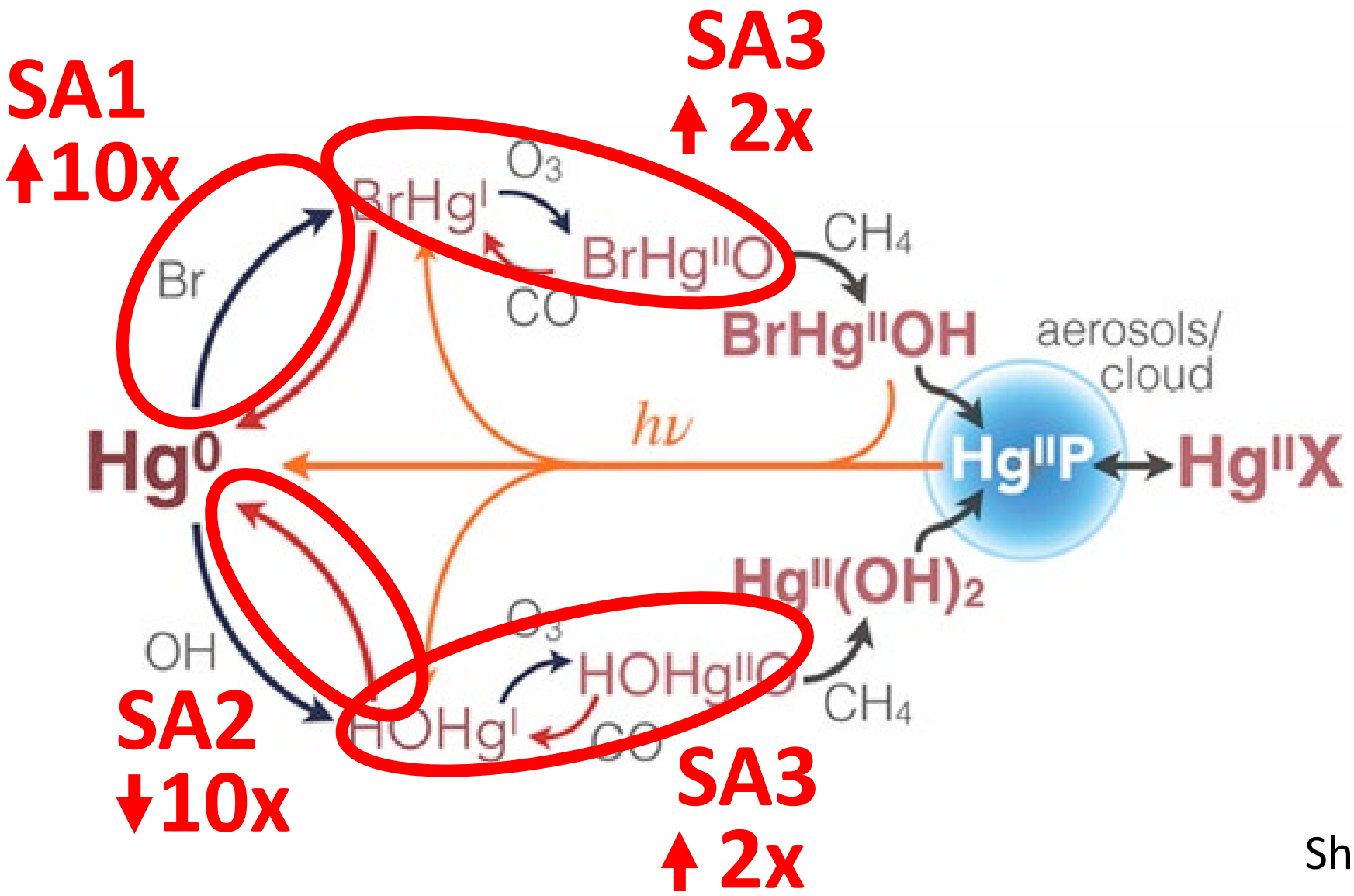
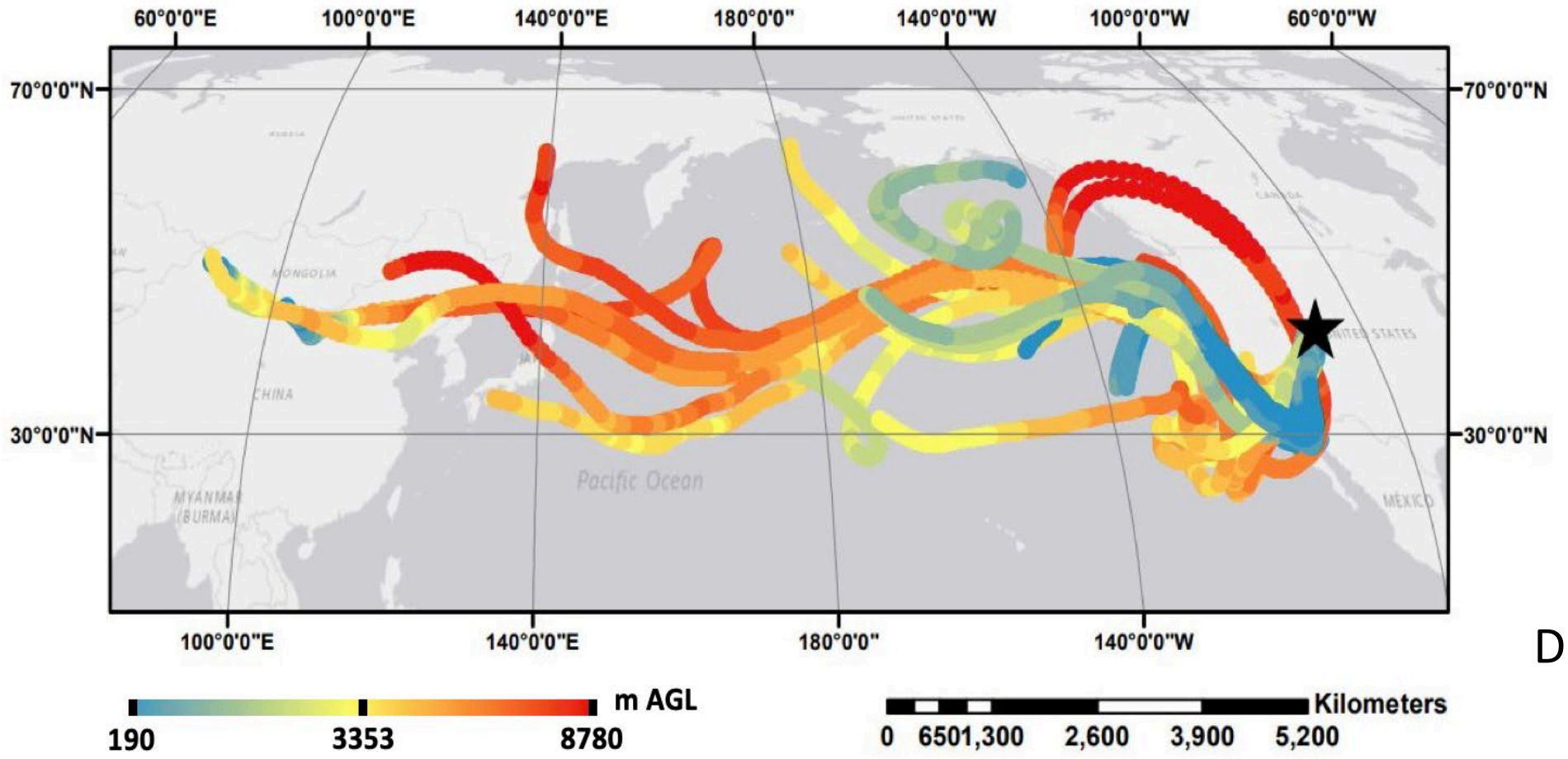


Image from
Shah et al., 2021

Modeled episode: 7-11 June 2021

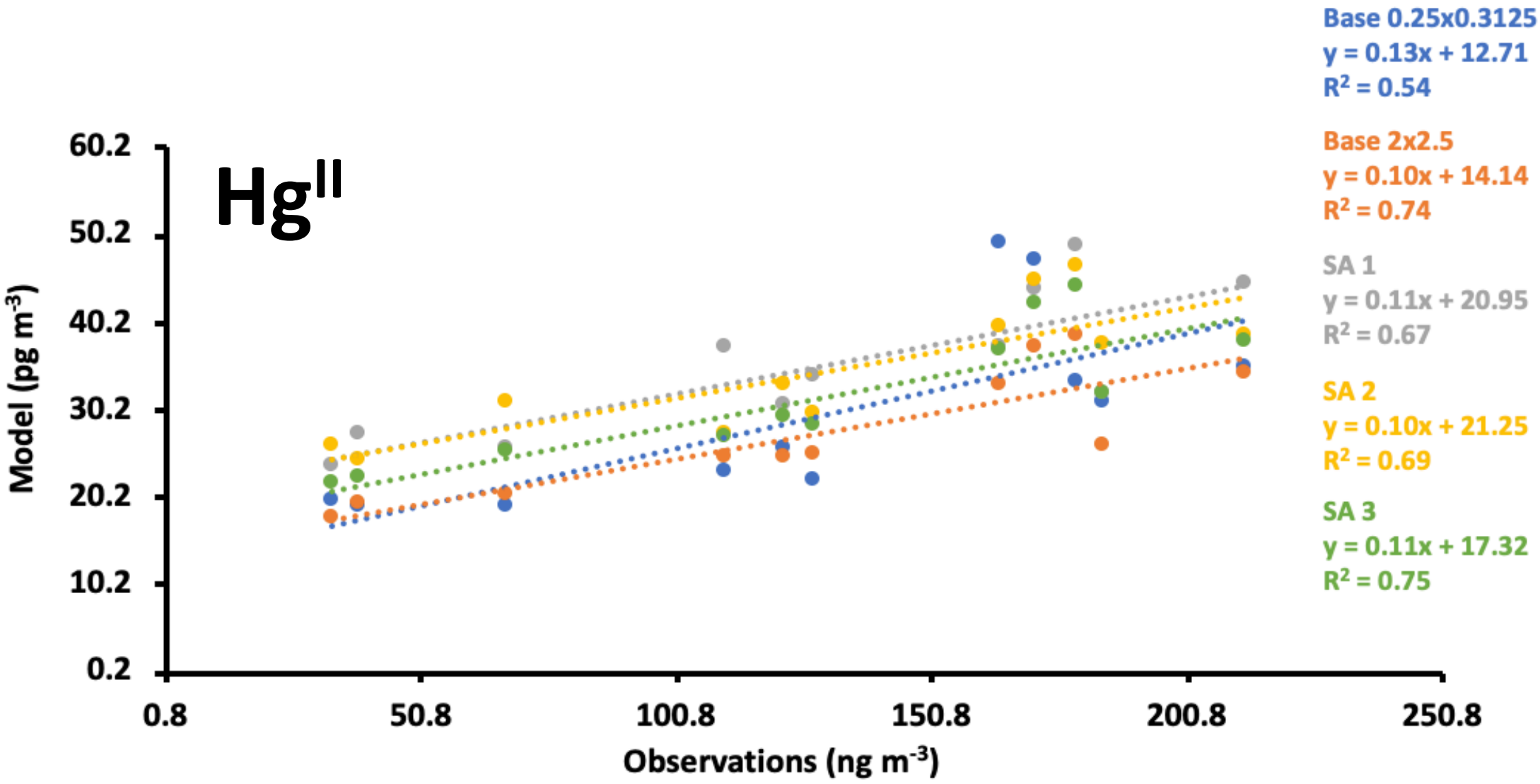
Hg^{II}: 150.78 ± 74.10 pg m^{-3}

Hg^{0}: 1.08 ± 0.19 ng m^{-3}

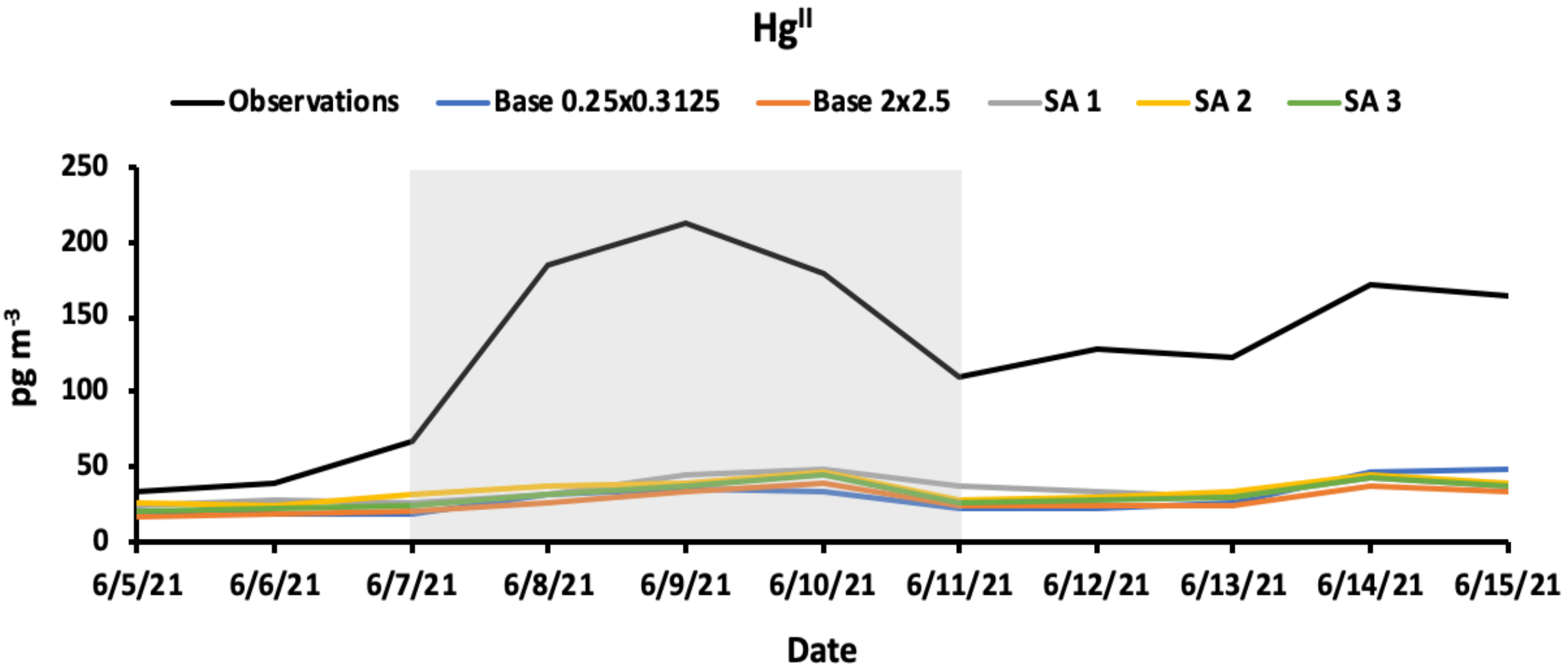


Details in Gratz et al. (under review)

Measurement-model correlations are strong



GEOS-Chem underestimates peak Hg^{II}



Measured Hg^{II}:Hg^{0} slope is not low enough

- **Hg^{II}:Hg^{0} slope in measurements**
  - **-0.37**; i.e., 63% of Hg^{II} was lost from air mass to deposition:
    - After formation from Hg^{0} and
    - Prior to measurement

- **Hg^{II}:Hg^{0} slope in model runs**
  - **-0.19 to -0.26**

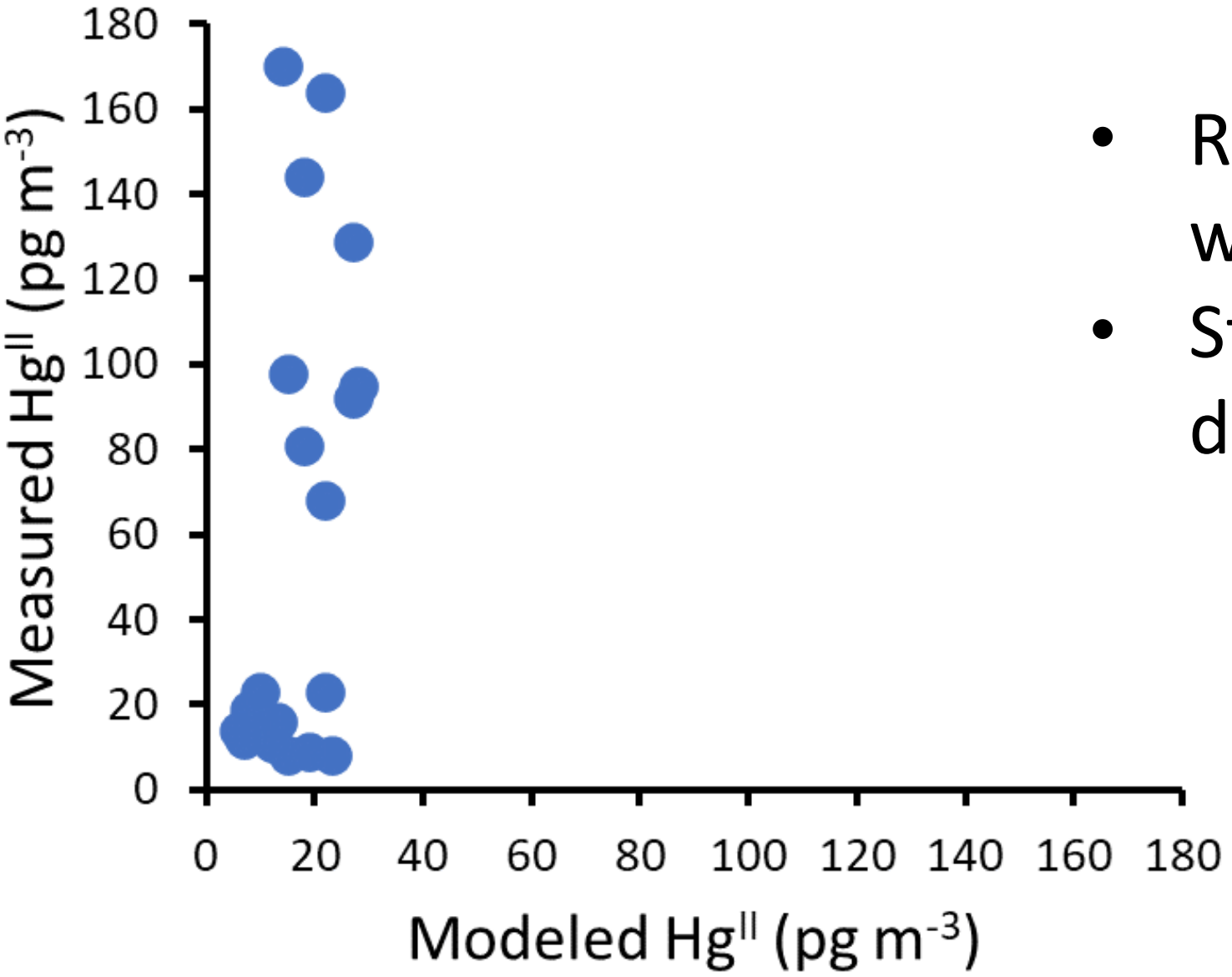
GEOS-Chem appears to lose too much Hg^{II} to deposition

Comparison from Gustin et al., 2023

- Direct Hg^{II} capture and analysis with cation-exchange membranes
- 2-week average samples

Details in Gustin et al., 2023

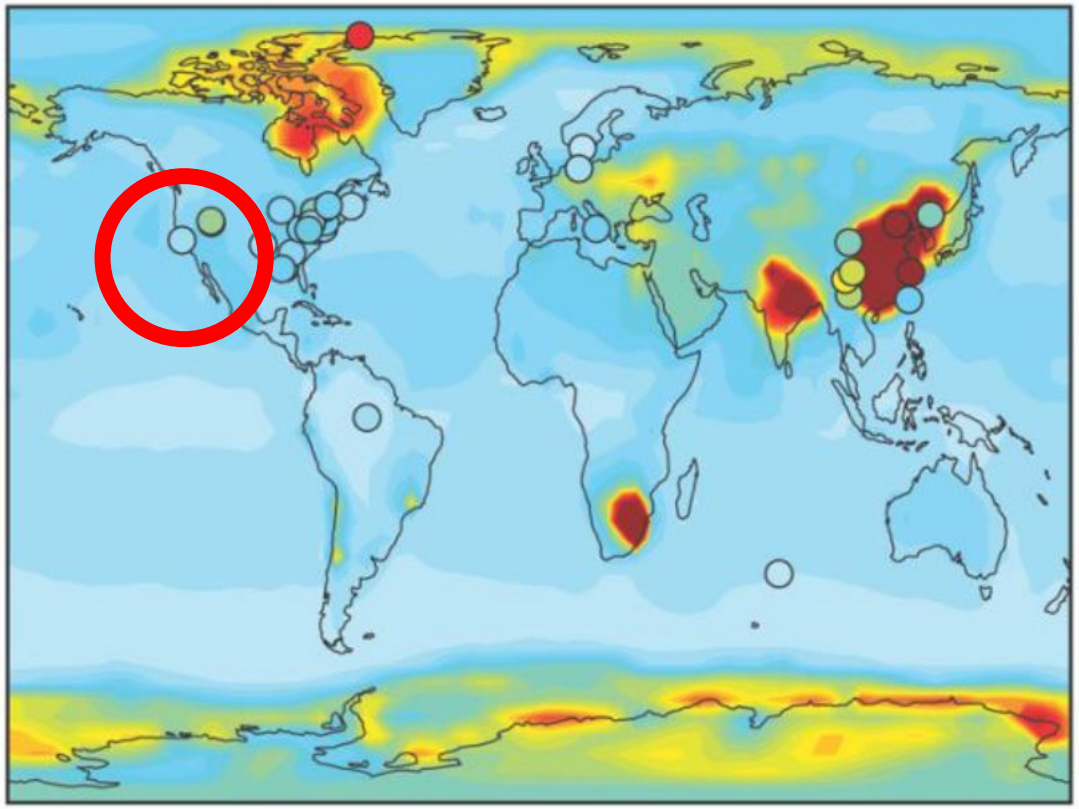
GEOS-Chem strongly underestimates peak Hg^{II}

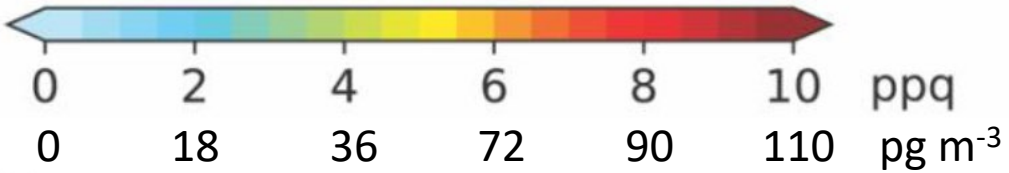


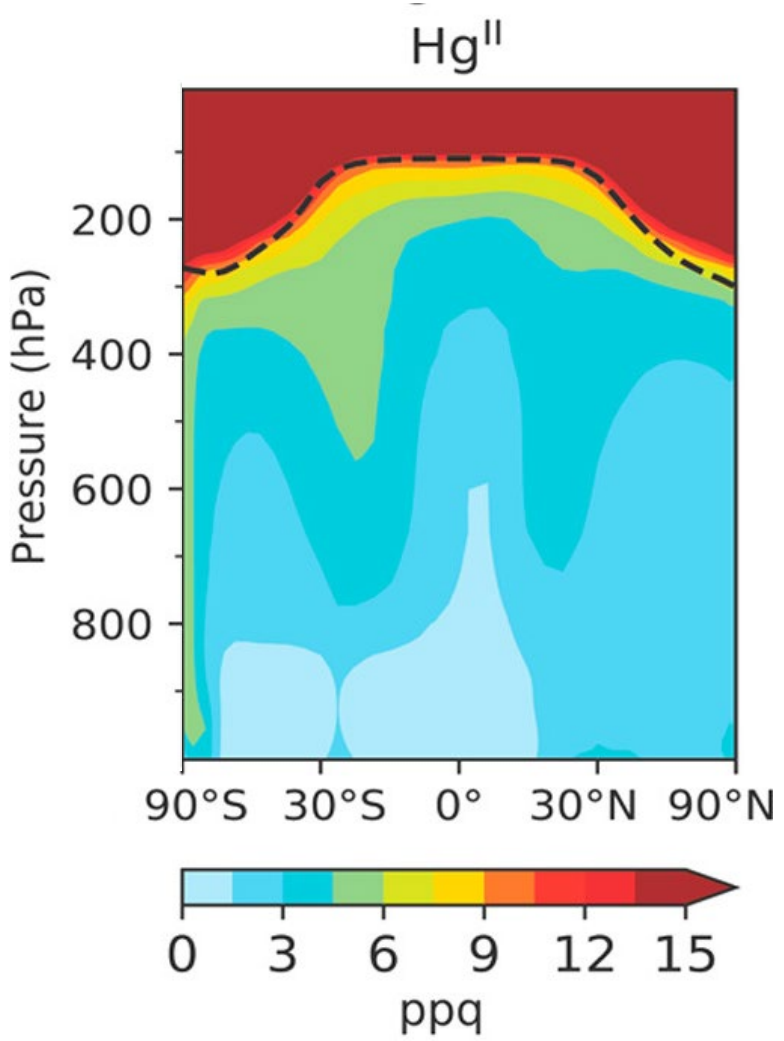
- Reasonable agreement when Hg^{II} is low
- Strong underestimation during high Hg^{II} in
  - Nevada
  - West Texas

Are model averages off?

Annual mean Hg^{II} from Shah et al., 2021



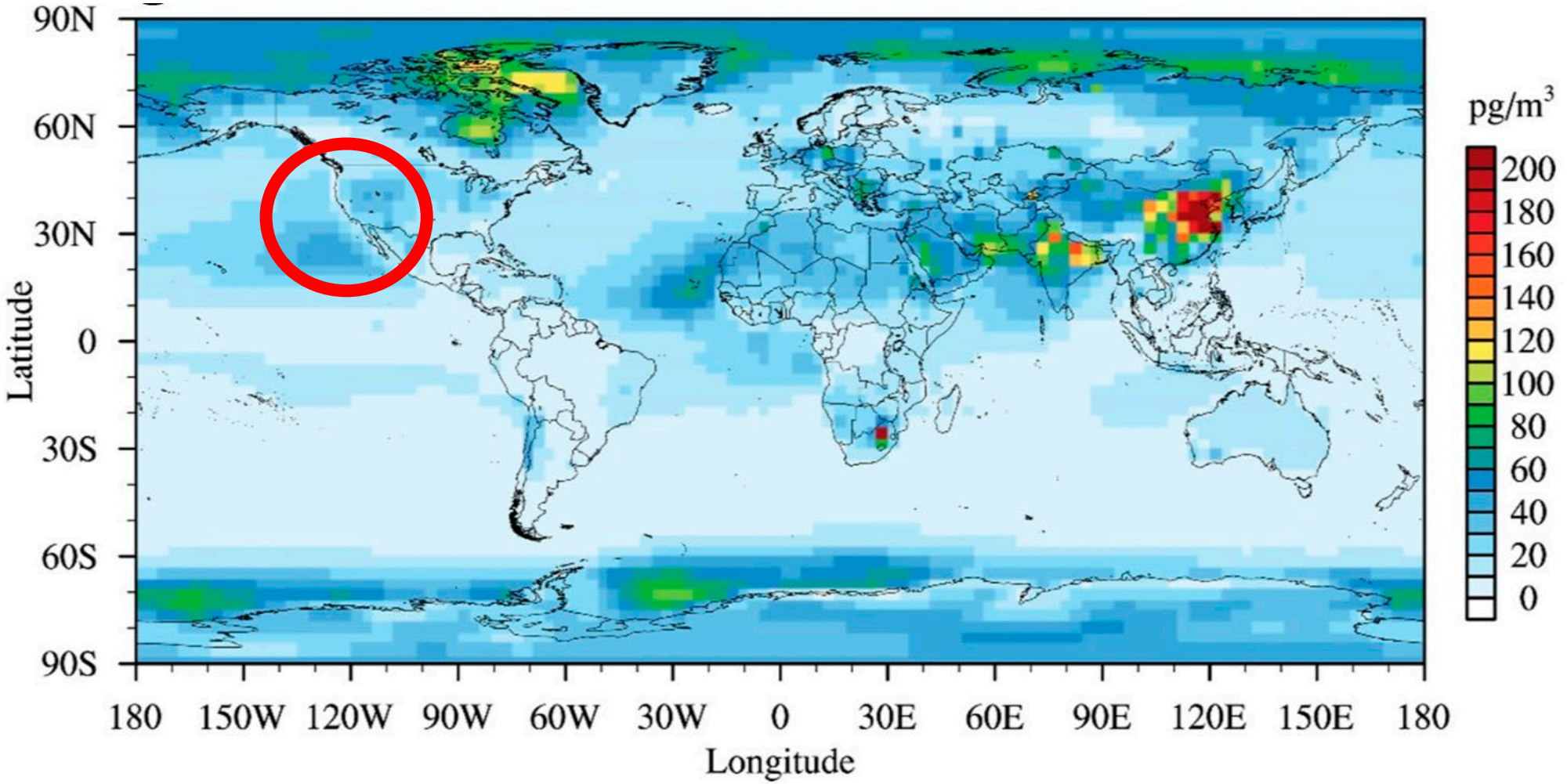




Are model averages off?

Annual mean surface Hg^{II} from Fu et al., 2024

(without anthropogenic halogen emissions added)



Models miss magnitude and variability of Hg^{II}

Measurement-model comparisons from Fu et al., 2024