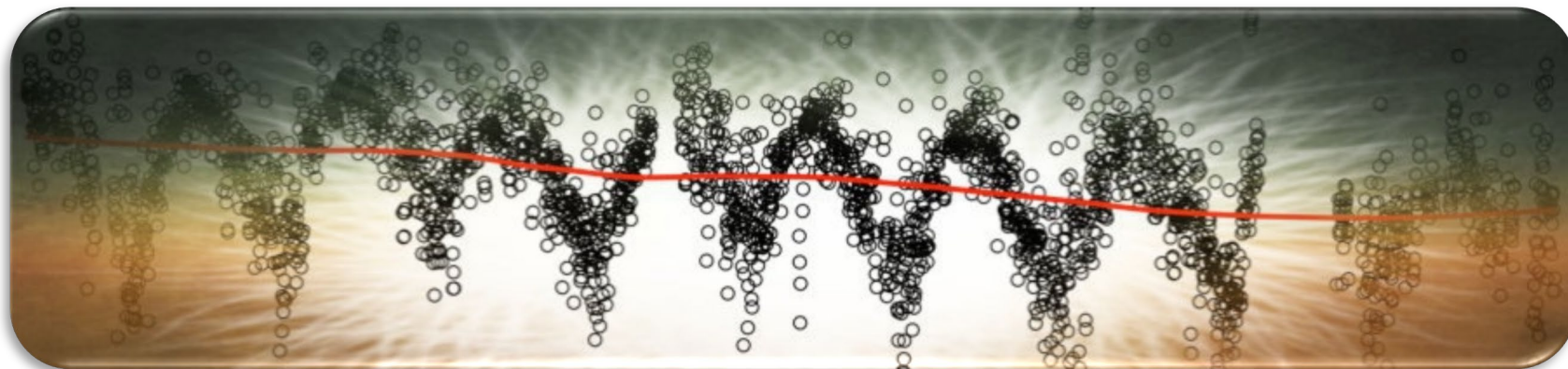


# Increasing anthropogenic emissions are inconsistent with the declining atmospheric mercury concentrations observed in the Northern Hemisphere



Ari Feinberg

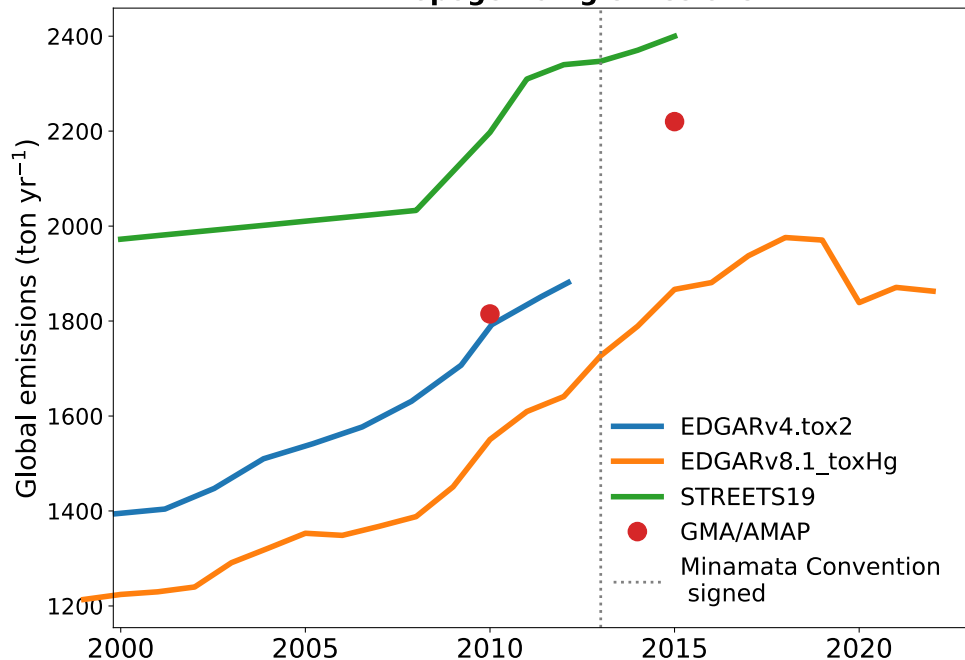
*Institute of Physical Chemistry (IQF-CSIC) & MIT*

Noelle Selin (MIT), Christine Braban, Kai-Lan Chang, Danilo Custódio, Daniel Jaffe, Katriina Kyllönen, Matthew Landis, Sarah Leeson, Winston Luke, Koketso Molepo, Marijana Murovec, Michelle Nerentorp Mastromonaco, Katrine Aspmo Pfaffhuber, Julian Rüdiger, Guey-Rong Sheu, and Vincent L. St. Louis



# Dispute: inventories show emissions increase but observed trends decline

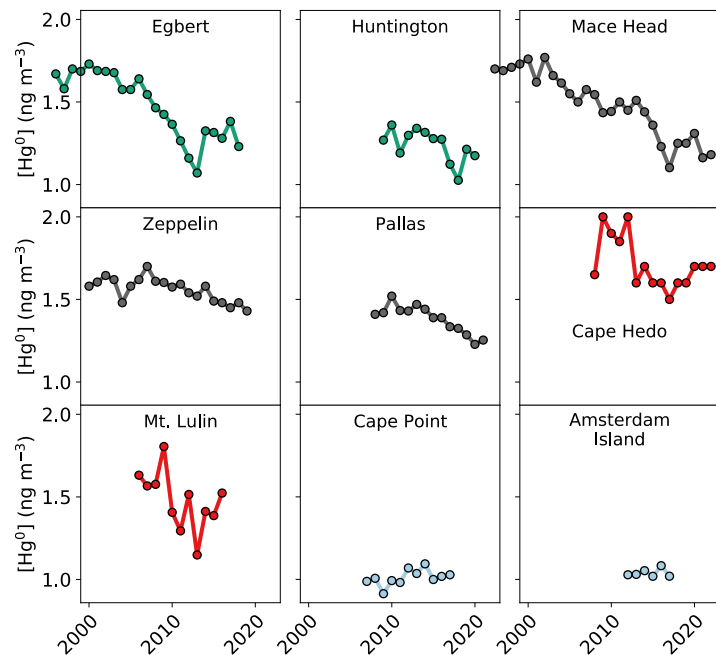
### Anthropogenic Hg emissions



### Observed Trends in Atmospheric Mercury (Hg<sup>0</sup>)

N. America Europe E. Asia South Hemisphere

vs.



**Are these anthropogenic emissions trends compatible with the observed trends?**

Observations from: ECCC, AMNet, EMEP, GMOS, MoE Japan, Nguyen et al. (2019), Slemr et al. (2020)

Emissions inventories from: Streets et al., *Atmos. Environ.* (2019); Muntean et al., *Atmos. Environ.* (2018); Global Mercury Assessment (2019);

Muntean et al., in. prep.

# Statistical and mechanistic modelling to analyze NH Hg<sup>0</sup> trends

Paper is under review, available as preprint on EarthArXiv: Feinberg et al., 2024, doi: [10.31223/X5B38K](https://doi.org/10.31223/X5B38K)

## Observed data

Use Hg<sup>0</sup> data from 51 Northern Hemisphere (NH) stations between 1992 and 2022

## Statistical method

Linear mixed effects modelling

(Chang et al., 2023 for Tropospheric Ozone Assessment Report)

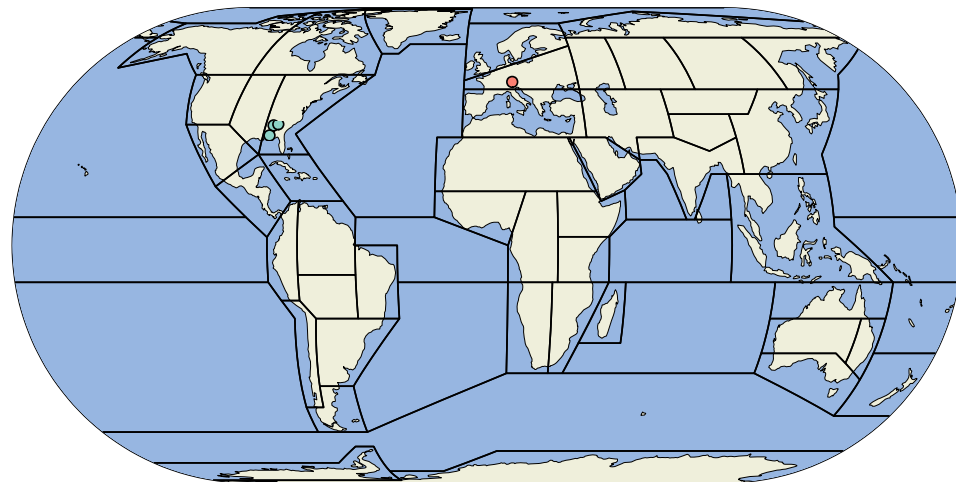
Regional trends from multisite data, accounts for: seasonality, gaps, autocorrelation, site offsets

## Biogeochemical two-hemisphere box model

10<sup>5</sup> scenarios/parameter options for 1510–2020

## GEOS-Chem global atmospheric chemistry model

5 simulations for 2005–2020



- Station included in this study
  - Station not included in this study
- IPCC regions

*Selected long-term (>6 years measurements) stations*

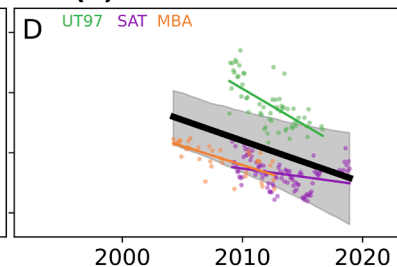
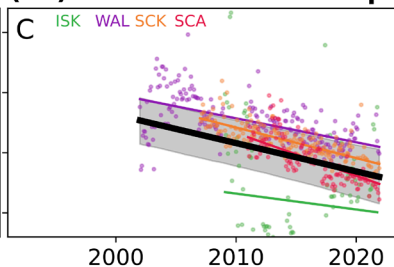
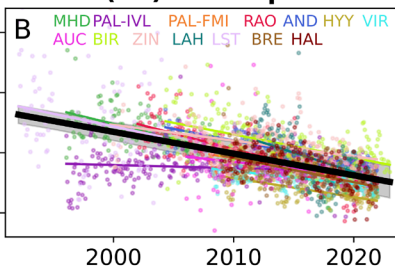
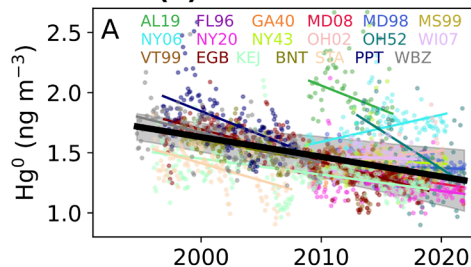
# Calculate regional trends by combining information from different sites

(5) East N America

(16) N Europe

(17) West & Central Europe

(3) West N America

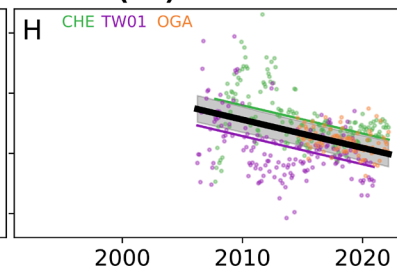
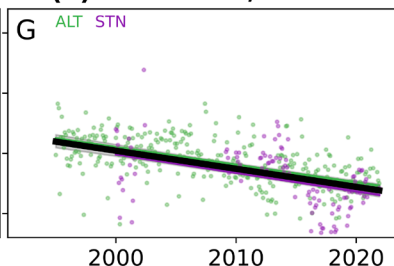
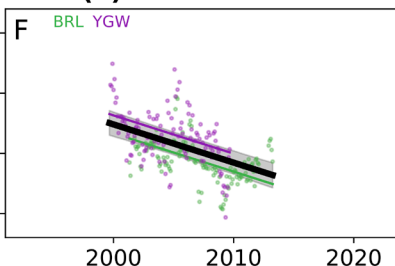
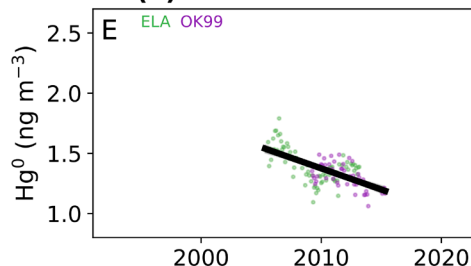


(4) Central N America

(2) NE N America

(0) Greenland/Iceland

(35) EastAsia

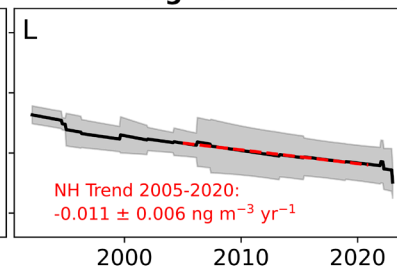
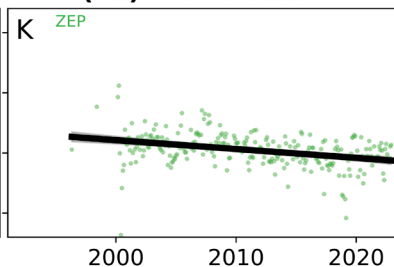
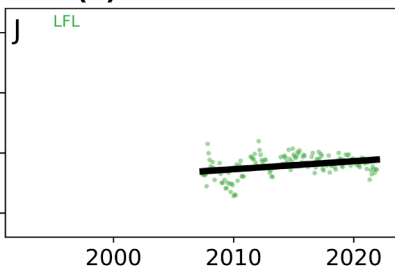
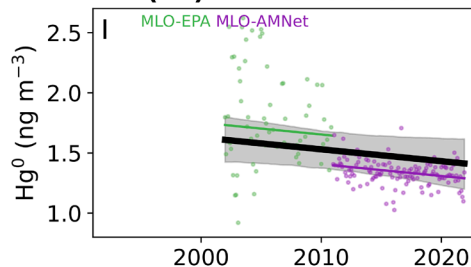


(47) N Pacific Ocean

(1) NW N America

(46) Arctic Ocean

Area-weighted NH trend



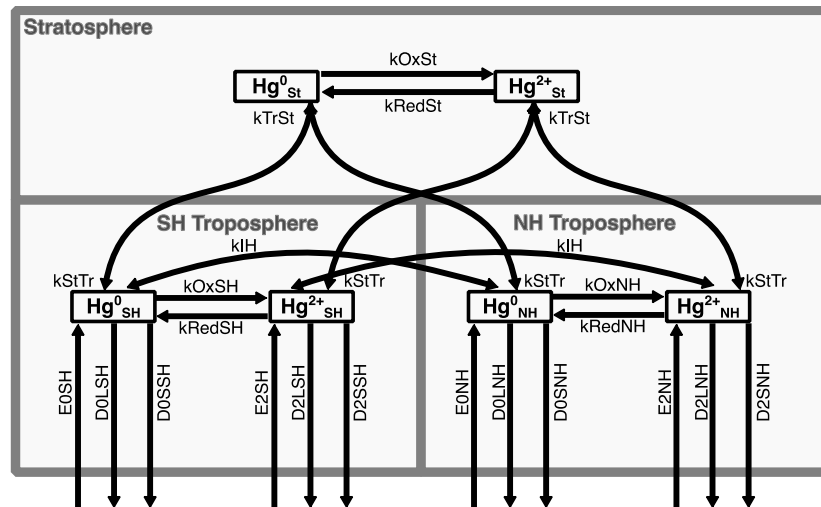
# Box model to derive the emissions trends compatible with observations

**What anthropogenic emissions trends are compatible with NH trend  $-0.011 \pm 0.06 \text{ ng m}^{-3} \text{ yr}^{-1}$  ?**

- Test  $10^5$  scenarios in box model
- Account for ranges in anthropogenic emissions & releases trends and parameter uncertainties
- Use Streets et al. (2019) for 1510–2005 anthropogenic emissions

Parameter	Min	Max	Units
NH anthro. emis. trend 2005–2020	-70	70	$\text{Mg yr}^{-2}$
SH anthro. emis. trend 2005–2020	-10	10	$\text{Mg yr}^{-2}$
... (19 total parameters)	...	...	...

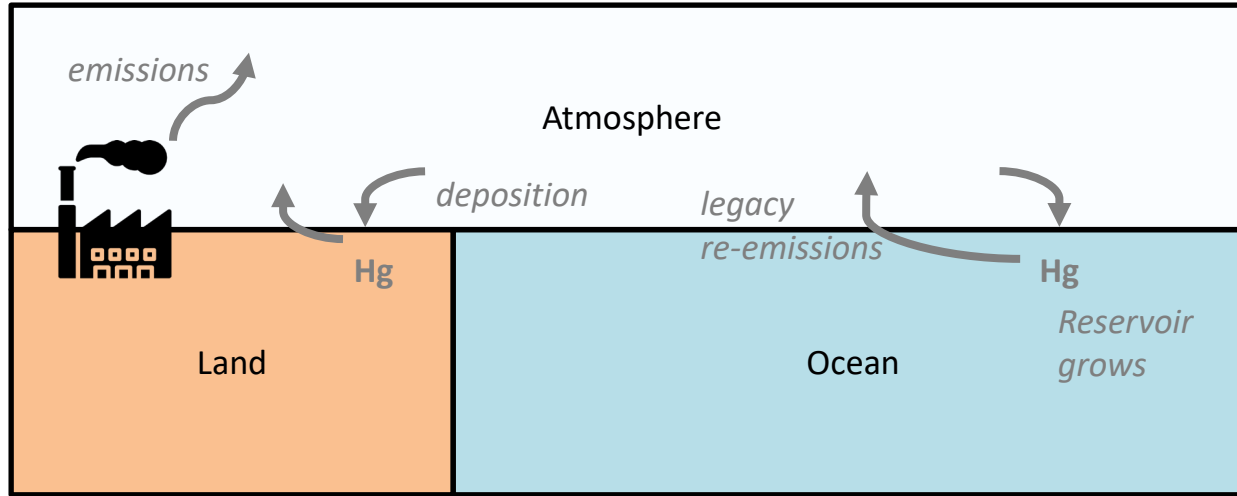
Analyze box model for 2005–2020 trends



**How will legacy re-emissions vary given trends in anthropogenic emissions?**

# The link between past anthropogenic and legacy emissions

To derive trends in anthropogenic emissions based on  $\text{Hg}^0$  measurements, must consider anthro-legacy link:



**Even if anthropogenic emissions stay constant over time, legacy emissions will grow**

(Angot et al., ES&T, 2018; Selin, Science, 2018)

Legacy emissions =  $f(\text{past anthropogenic emissions, legacy lifetime}_{1,2}, \text{coefficient}_{1,2})$

(derived from Amos et al., 2013, box model)

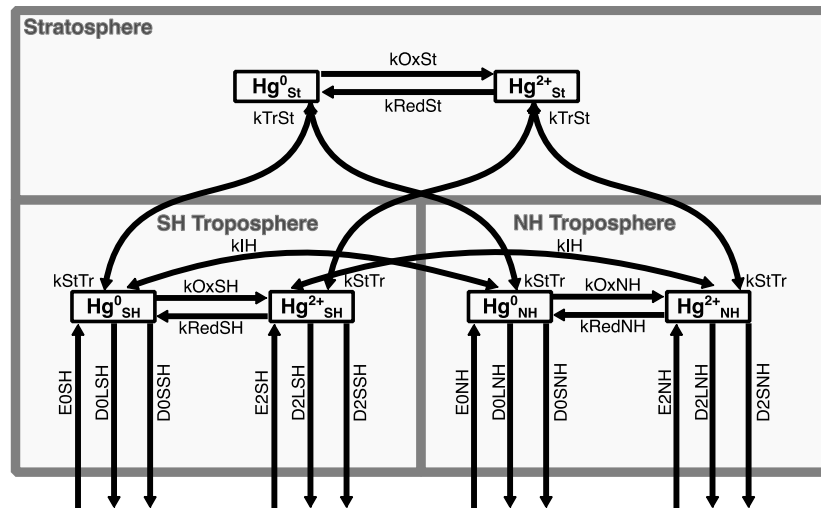
# Box model: disentangling role of anthropogenic vs. legacy emissions

**What anthropogenic emissions trends are compatible with NH trend  $-0.011 \pm 0.06 \text{ ng m}^{-3} \text{ yr}^{-1}$  ?**

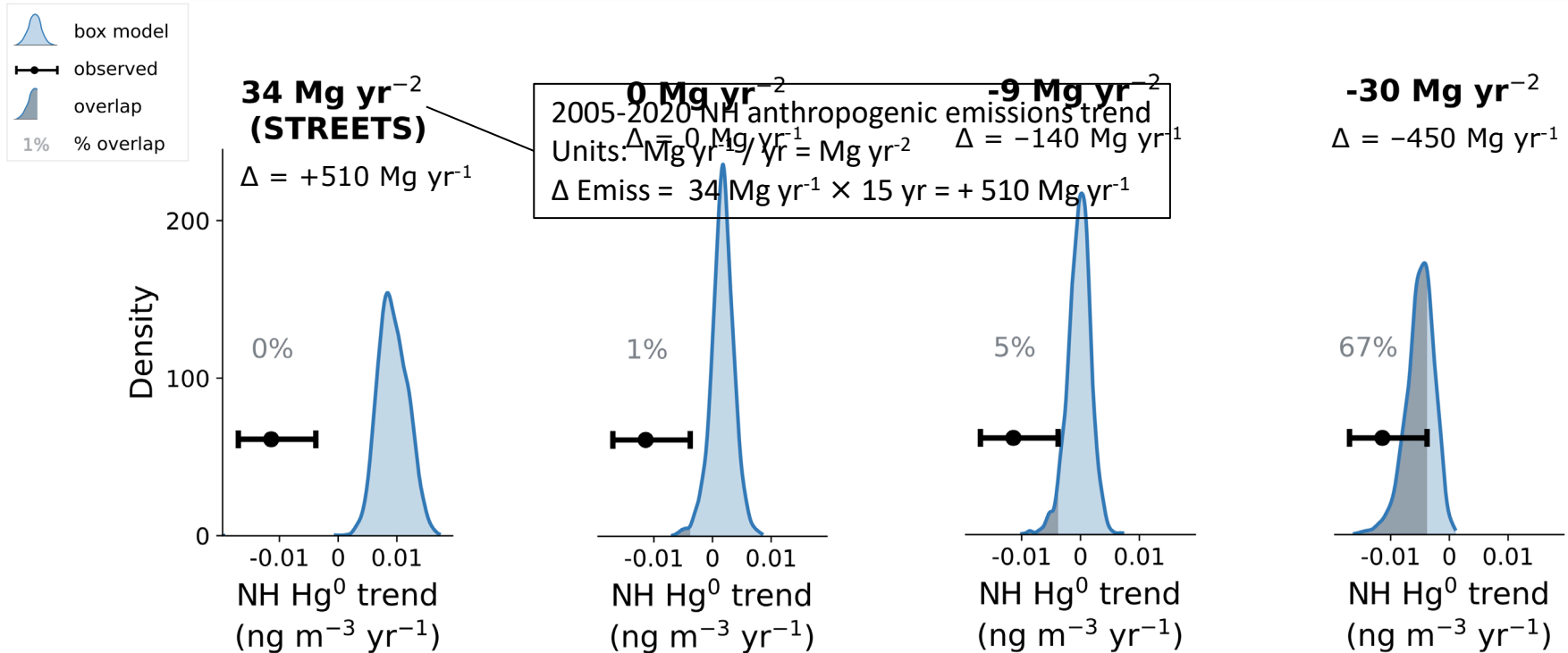
- Test  $10^5$  scenarios in box model
- Account for ranges in anthropogenic emissions & releases trends and parameter uncertainties
- Use Streets et al. (2019) for 1510–2005 emissions

Parameter	Min	Max	Units
NH anthro. emis. trend 2005–2020	-70	70	$\text{Mg yr}^{-2}$
SH anthro. emis. trend 2005–2020	-10	10	$\text{Mg yr}^{-2}$
Legacy lifetime (short)	6	15	months
Legacy lifetime (long)	29	97	years
Error in year 1970, 1980, 1990, 2000, emissions	-20	40	%
... (19 total parameters)	...	...	...

Analyze box model for 2005–2020 trends



# Box model: anthropogenic declines are needed to explain $\text{Hg}^0$ trend



**Northern Hemisphere (NH) anthropogenic emissions likely decreased by at least  $140 \text{ Mg yr}^{-1}$  between 2005 and 2020**

# Could other factors reconcile emissions inventories with observed declines?

Many climate change factors that could lead to **increasing**  $\text{Hg}^0$  trends:

- Release of Hg from melting permafrost
- Increased  $\text{Hg}^0$  ocean evasion due to warming
- Decreased sea ice coverage allowing more  $\text{Hg}^0$  evasion from ocean
- Enhanced wildfire emissions



We studied two factors that might lead to **decreasing**  $\text{Hg}^0$  trends:

- Enhanced vegetation growth leading to enhanced  $\text{NH Hg}^0$  uptake
  - Unlikely to outweigh emissions increase, observed vegetation change is small
- Oxidation of  $\text{Hg}^0$  speeds up due to changing atmospheric chemistry
  - Unlikely based on other compounds & would expect increases in Hg wet deposition

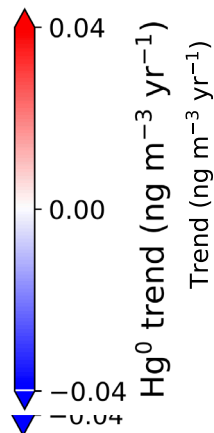
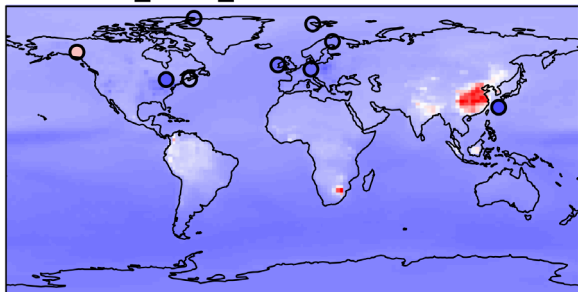
**Anthropogenic emissions declines are the most likely cause!**

# GEOS-Chem simulations corroborate box model results

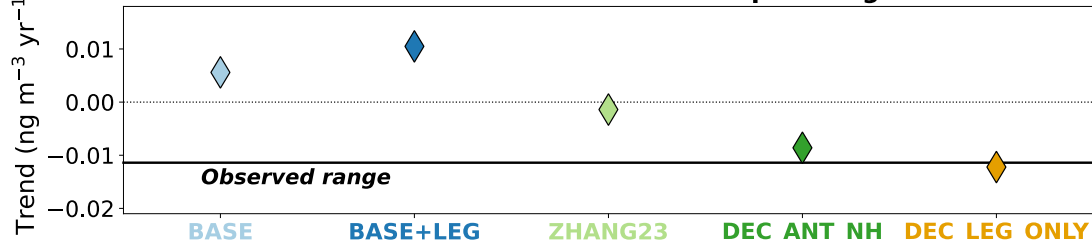
## Simulated median trends

F

### DEC\_LEG\_ONLY: P50 trend



### Mean trend in Northern Hemisphere Hg<sup>0</sup>



**BASE:** Run 2005–2020 GEOS-Chem simulations with Streets et al. (2019) emissions

**BASE+LEG:** Consider legacy re-emissions feedback to anthropogenic emissions trend

**ZHANG23:** Substitute Chinese emissions from regional inventory (Zhang et al., ES&T, 2023)

**DEC\_ANT\_NH:** Additional 23 Mg yr<sup>-2</sup> decline in anthro emissions spread across NH

**DEC\_LEG\_ONLY:** -50 Mg yr<sup>-2</sup> decline imposed on the legacy emissions from NH oceans

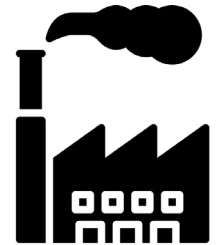
- **Unlikely scenario** due to East Asian observations, quantile regression, box model results

# Takeaway messages and outlook

Atmospheric mercury concentrations have generally declined across the Northern Hemisphere from 2005



The most likely cause of the decline is anthropogenic emissions declining by at least 140 Mg yr<sup>-1</sup>, but this contradicts bottom-up emissions inventories



**Outlook:** Understanding this gap between top-down and bottom-up emissions trends is critical for predicting future Hg levels and evaluating policy effectiveness



# Acknowledgements

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- Current funding: Horizon Europe Marie Skłodowska Curie Postdoc Fellowship (#101103544)
- Thanks to all researchers involved in collection of public atmospheric Hg measurement datasets (Environment and Climate Change Canada, NADP Atmospheric Mercury Network, New York State Department of Environmental Conservation, EMEP, GMOS, Ministry of Environment Japan, Ministry of Environment Taiwan, IISD Experimental Lakes Area, Defra, UKEAP, NERC, NOAA, EPA)
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