Shifts in atmospheric composition since the preindustrial era modified the transport and deposition of mercury

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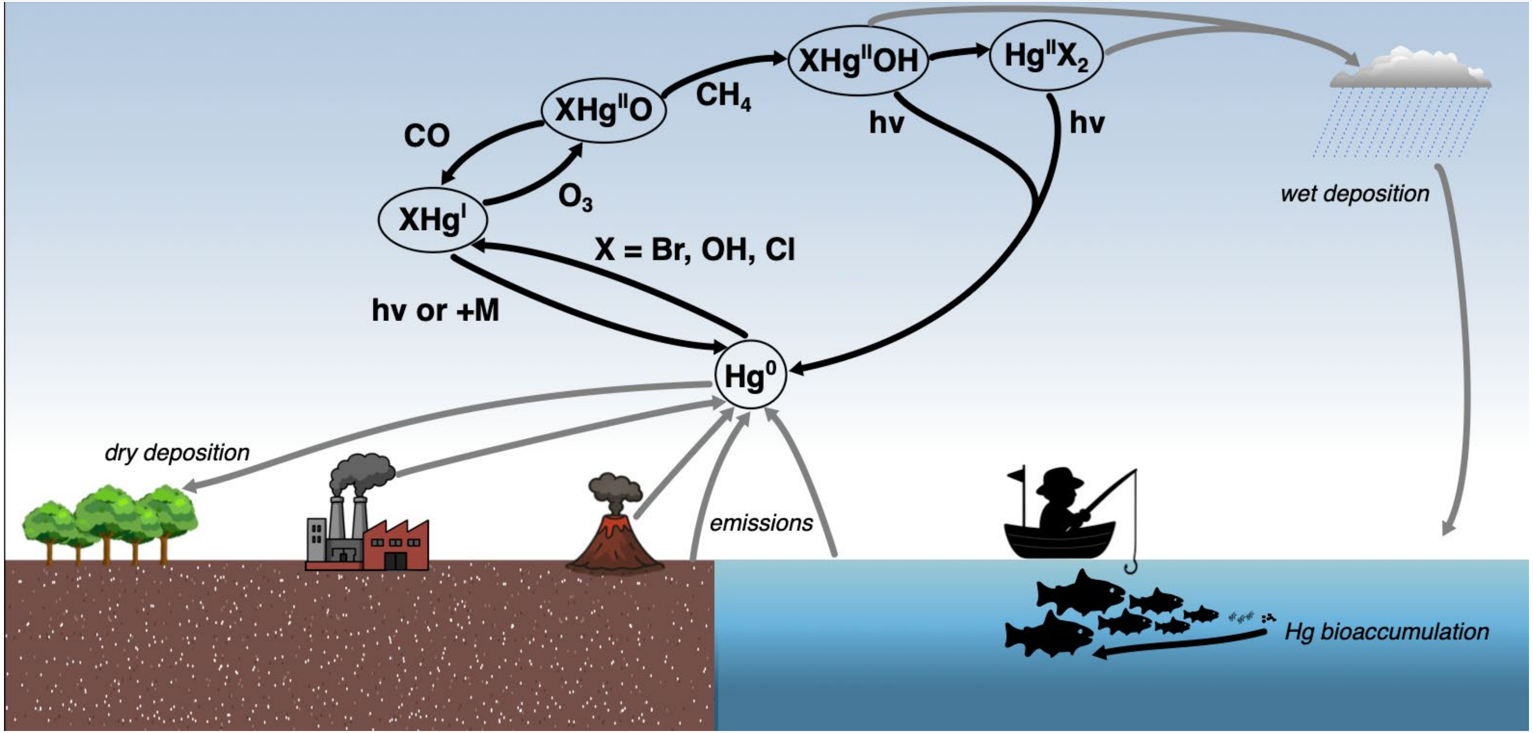
Co-funded by the European Union

Mercury (Hg) is a neurotoxic pollutant, spread in the atmosphere



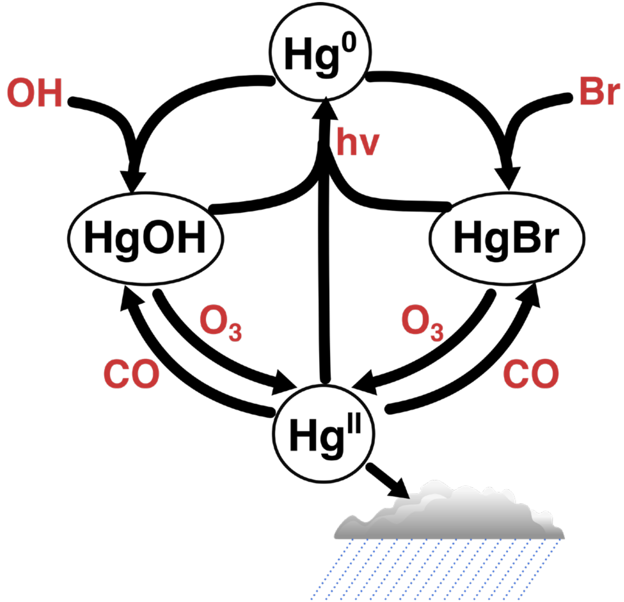
Impact by numbers:

>10 000 victims of Minamata disease in 1950s from chemical factory releases

$117 billion: global annual cost of health impacts (neurological and cardiovascular) associated with methylmercury exposure (Zhang et al., 2021)

Atmospheric Hg cycling is linked to trends in other compounds

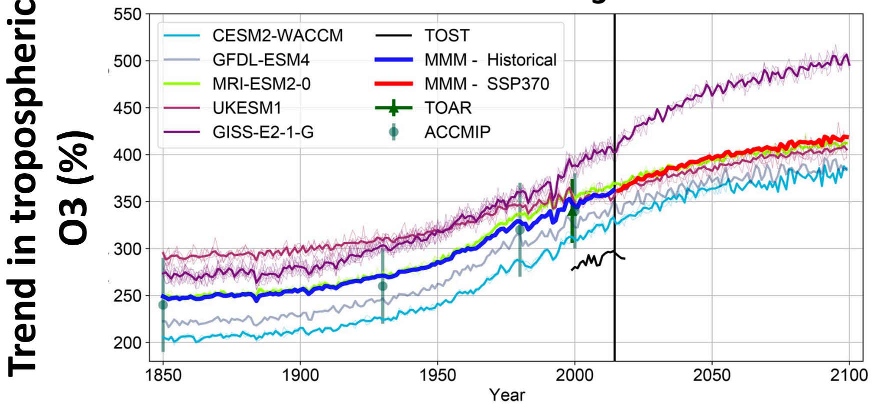
MSCA-PF SUMAC project (2024–2026): TrendS and Uncertainties in Mercury (Hg) Atmospheric Chemistry



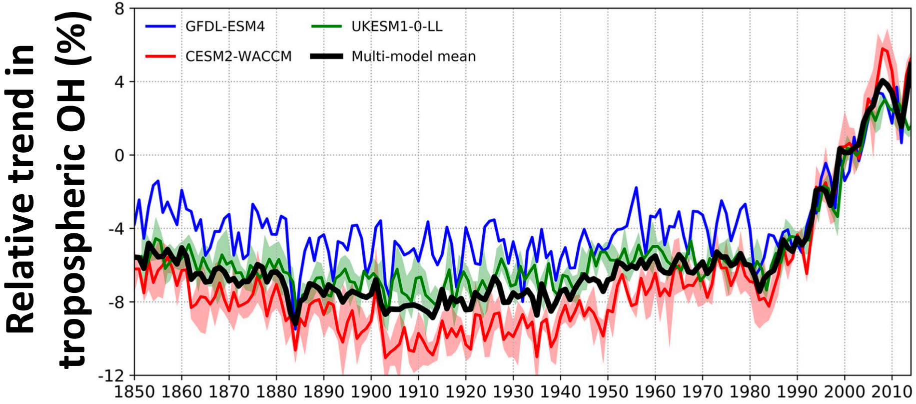
Saiz-Lopez et al. (2018, 2019, 2020, 2022, 2025); Shah et al. (2021); Gómez Martín et al. (2022); Castro et al. (2022);

Griffiths et al. (2021); Stevenson et al. (2020)

Ozone (O_3)



Hydroxyl radical (OH)



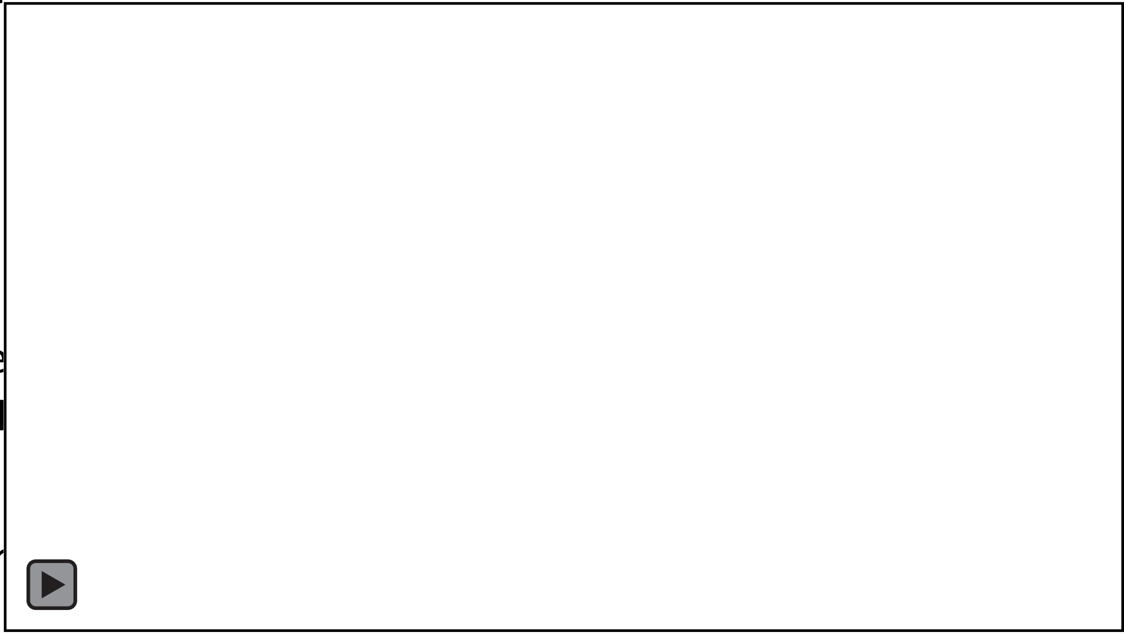
The chemistry-climate model WACCM: a unique tool for exploring Hg cycle

- Mercury chemistry now in WACCM model: whole atmosphere component of Community Earth System Model (CESM2) (Saiz-Lopez al., *Sci. Adv.*, 2025)

- First Hg model in a fully online chemistry-climate model with state of-the-art O_3-NO_x-SO_x-Org-Cl-Br-I-H chemistry

- Includes 62 reactions of atmospher Hg chemistry

- Simulations at 1.9° × 2.5° resolution, 88 vertical levels to 144 km altitude

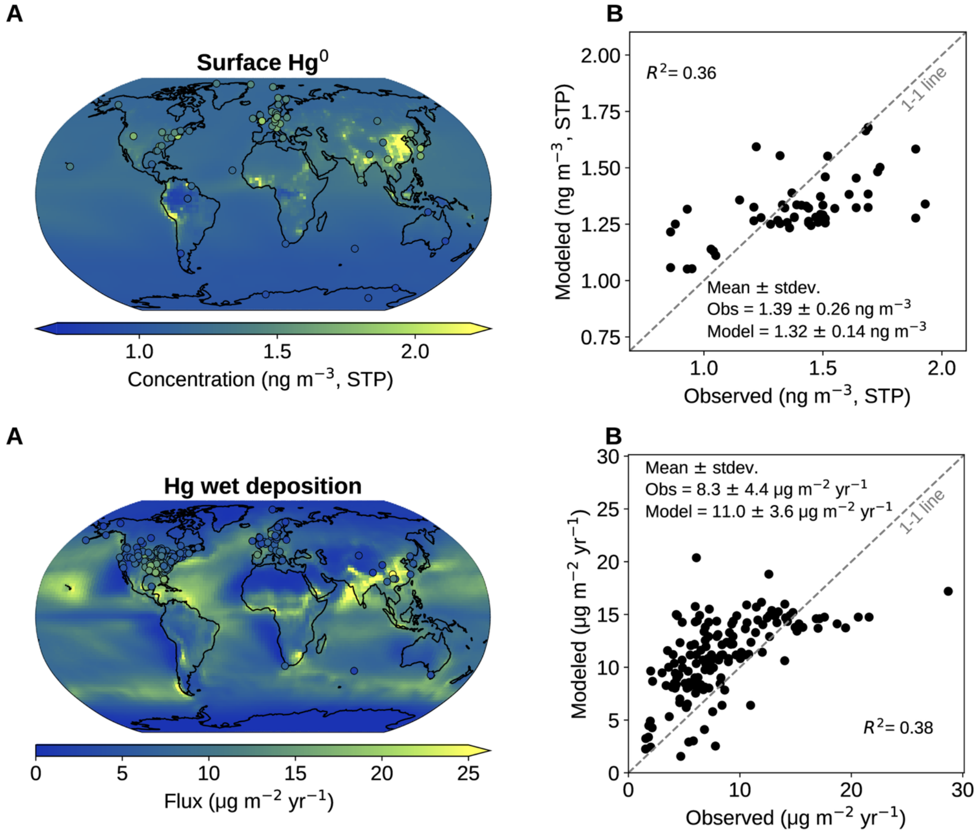


How has Hg redox chemistry changed since preindustrial?

Methods

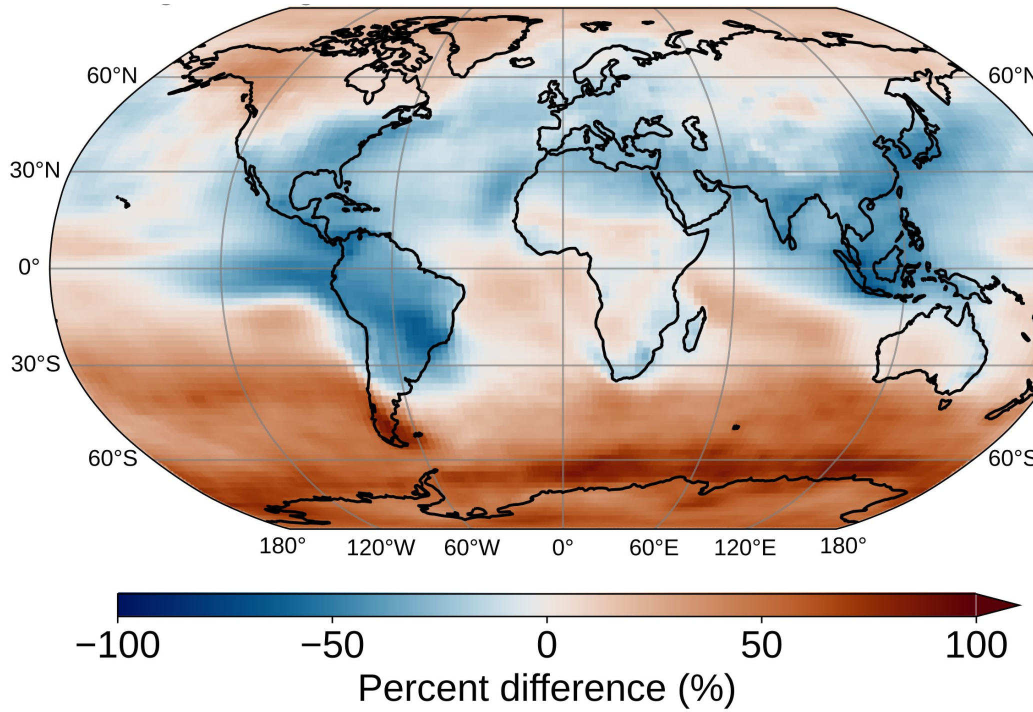
- WACCMv6 in CESM2
- Simulations for preindustrial (PI, 1850) and present day period (PD, 2010–2019)
- Emissions follow IPCC assessment CMIP6 setup (Hoesly et al., 2018), short-lived halogens follow Fernandez et al. (2025)
- Mercury emissions for preindustrial and present day come from available data (UNEP et al., 2019; Streets et al., 2019; Dastoor et al., 2025)

Evaluation



Response of Hg chemistry varies by region, hemispheric asymmetry

Change in tropospheric Hg^0 chemical lifetime between preindustrial and present-day



$$\text{chemical lifetime} = \frac{\text{Hg}^0\ \text{burden}}{\text{Hg}^0\ \text{net oxidation flux}}$$

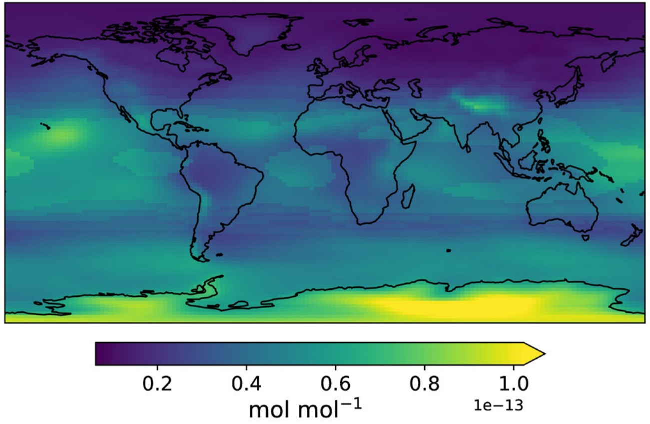
Blue: Hg^0 oxidation is faster in present-day

Red: Hg^0 oxidation is slower in present-day

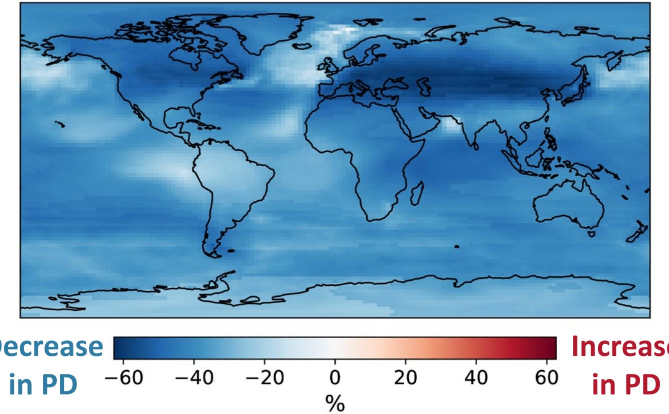
Global lifetime not substantially different
NH: 16% faster in present day
SH: 20% slower in present day

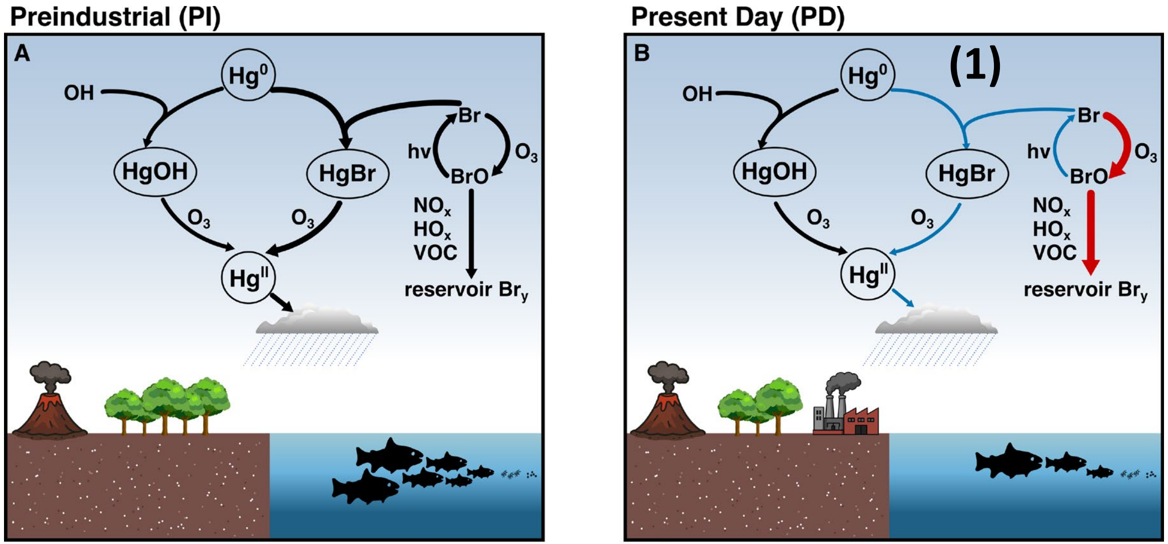
Why does Hg^0 oxidation decelerate in the Southern Hemisphere?

Preindustrial (PI) tropospheric mean vmr Br



Change between preindustrial and present day

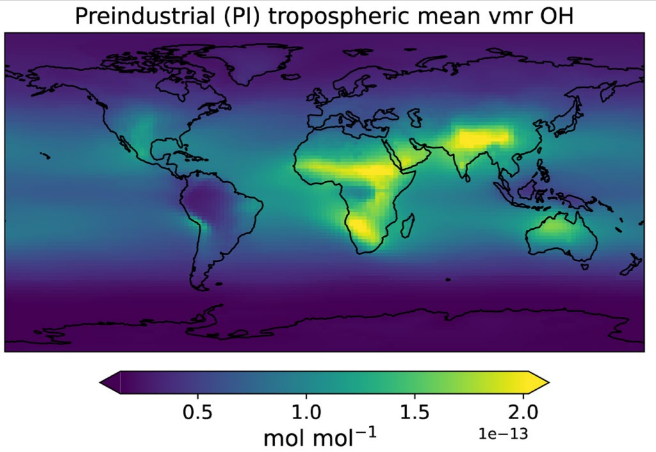


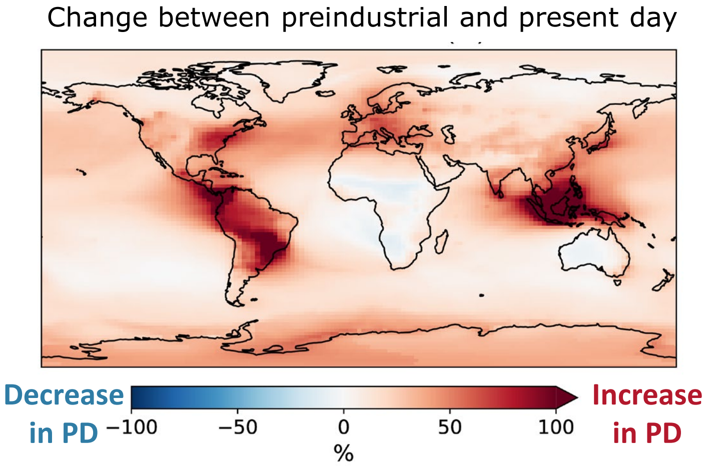


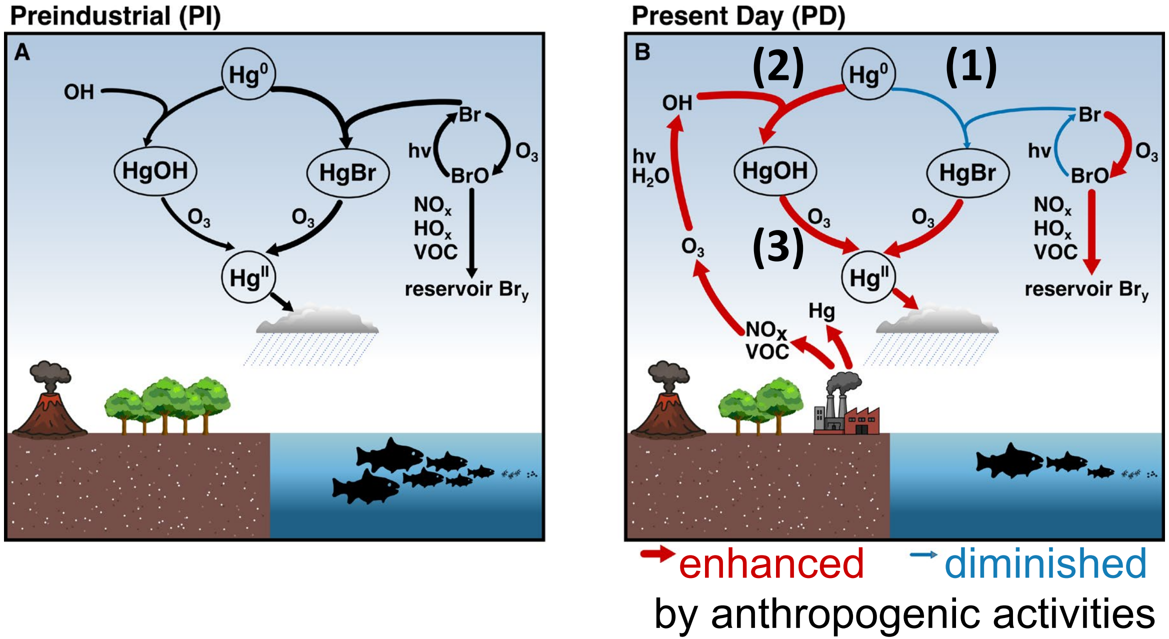
enhanced diminished
by anthropogenic activities

(1) In present day, more atomic Br gets converted to unreactive BrO and reservoir species (Br_y = HBr + HOBr + $BrONO_2$ +…), slowing Hg^0 oxidation

Why does Hg^0 oxidation accelerate in NH tropics and subtropics?



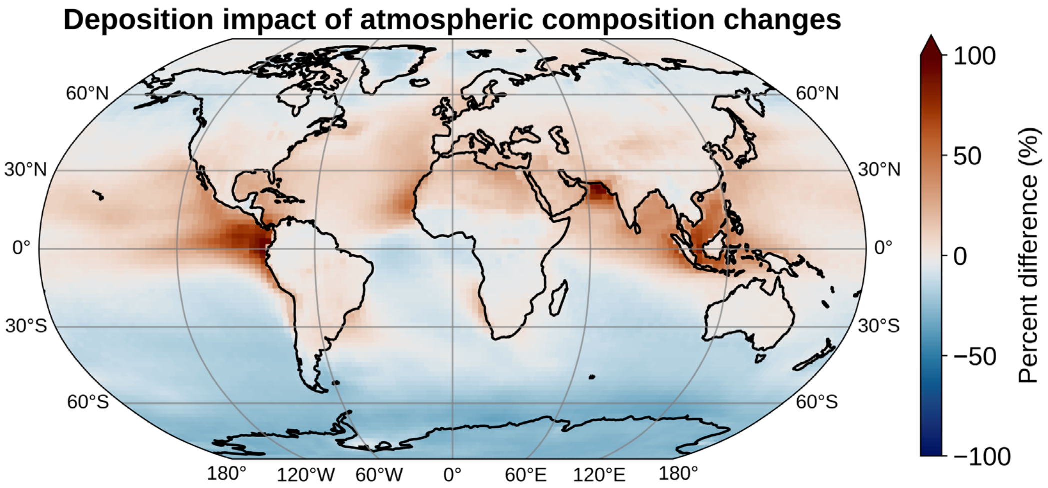




In present day, increases in OH (2) and O_3 (3) enhance Hg^0 oxidation

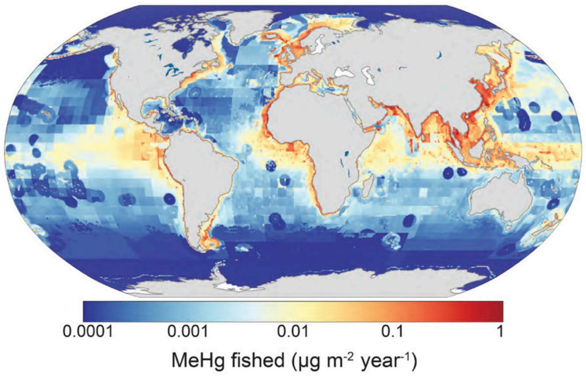
Buffering due to ozone having accelerating effects (2 & 3) and decelerating indirect effect through Br (1)

Oxidation changes impact the pattern of Hg deposition



Red: present-day chemistry enhances Hg deposition

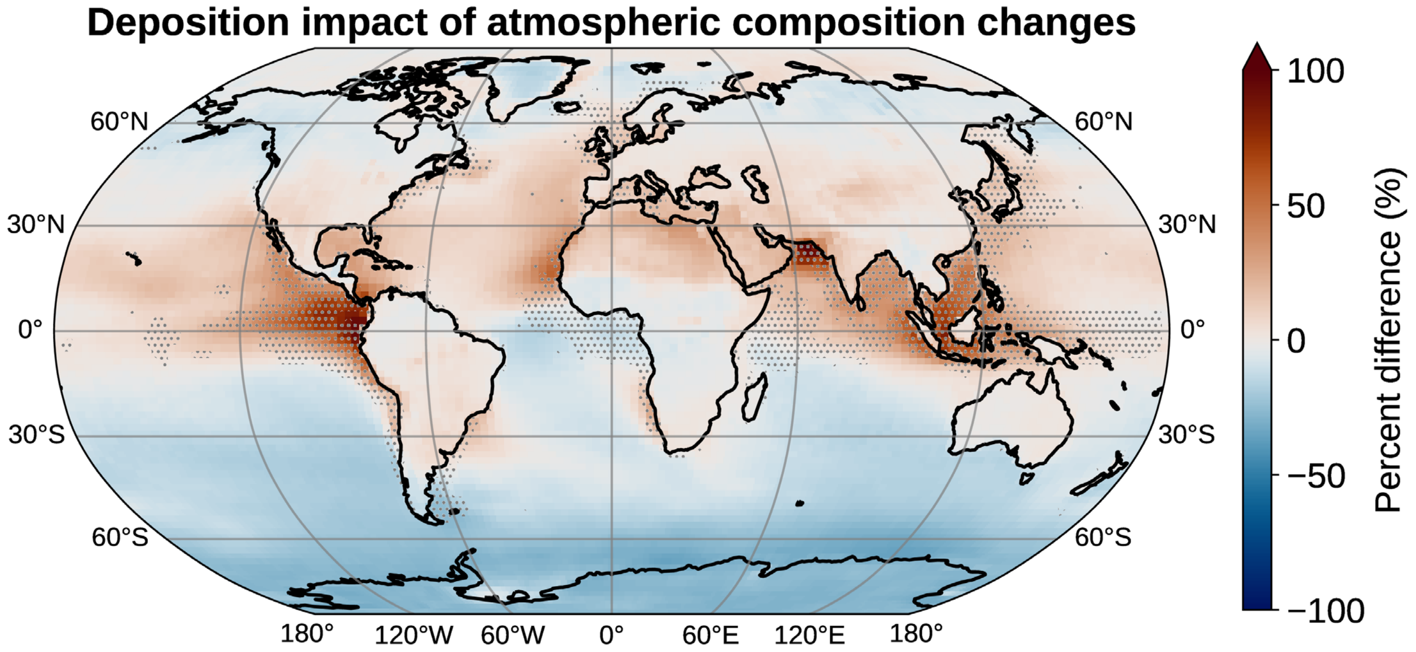
Blue: present-day chemistry decreases Hg deposition



Li et al., *PNAS* (2024): methylmercury (MeHg) that we fish mainly comes from tropics and subtropics – where have methylation hotspots and tuna fisheries

Chemical composition changes aggravate risk of human Hg exposure

Stippling shows key exposure areas (where 91% of fished MeHg originates)
In these areas, deposition due to chemistry changes increases by 15% (up to 30% in certain regions)

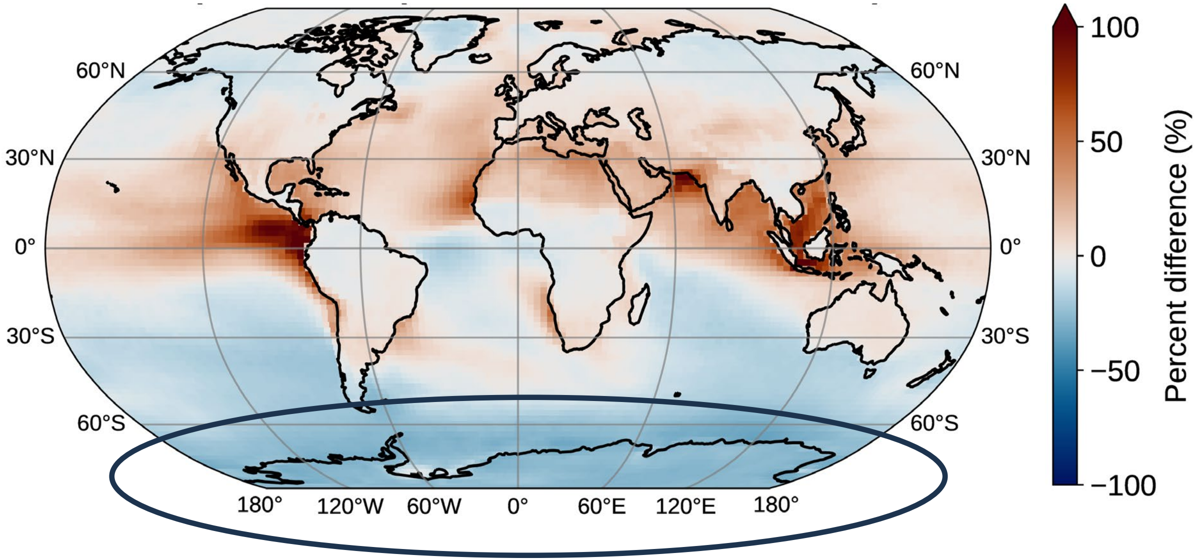


Apart from anthropogenic Hg emissions, atmospheric chemistry changes increase Hg deposition to tropical and subtropical oceans: where our highest Hg exposure comes from

Impact on interpreting natural archives (e.g., Antarctic ice cores)

Previous modelling studies have assumed no change in Hg oxidants in past periods, when analyzing deposition records (ice cores, peat cores, lake sediments, etc.)

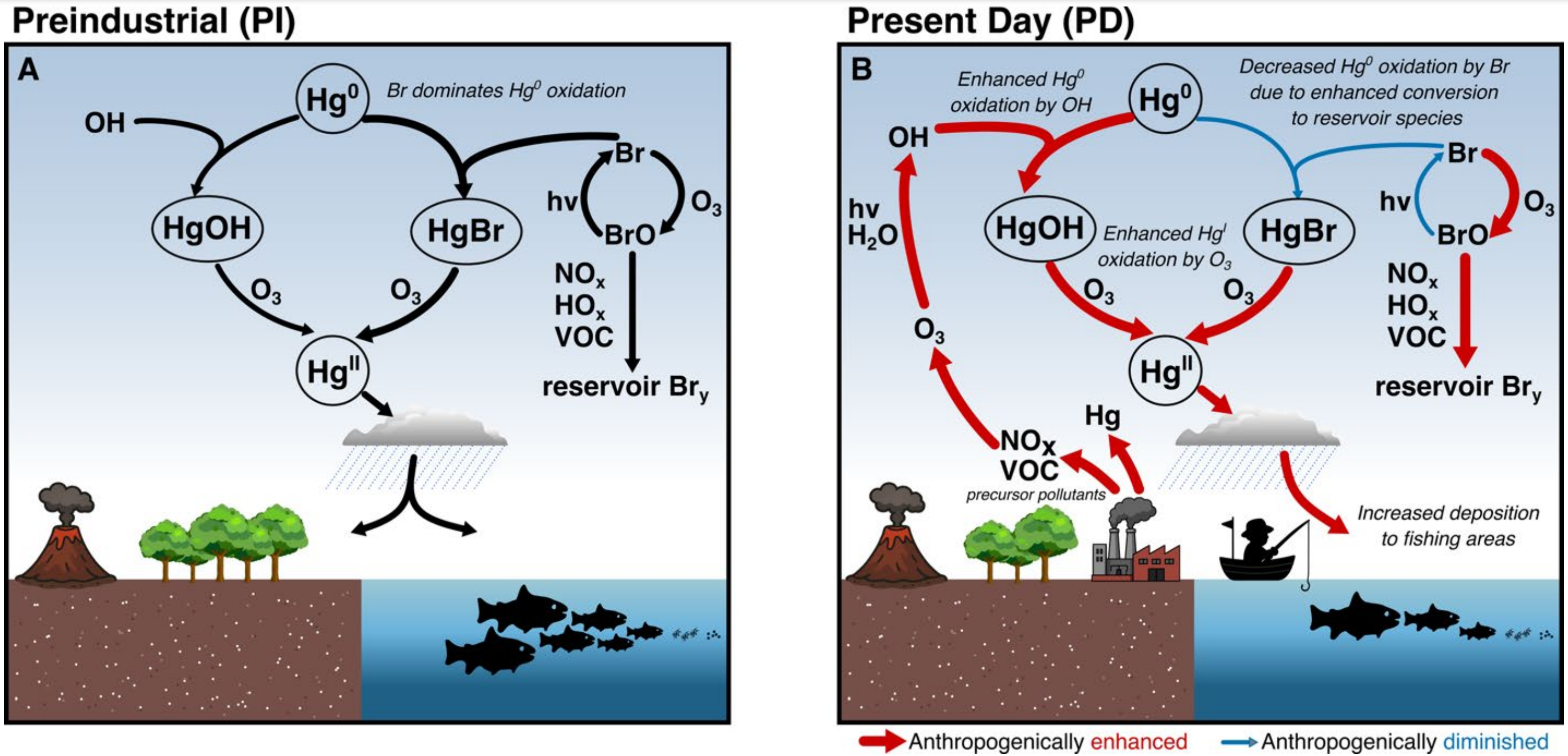
Deposition impact of atmospheric composition changes



Less transport of Hg to the Southern Hemisphere due to faster oxidation of Hg in tropics

Assuming present day oxidants can bias preindustrial emissions estimates derived from Antarctic ice cores by +40%

Summary: atmospheric chemistry as a driver of Hg cycle trends

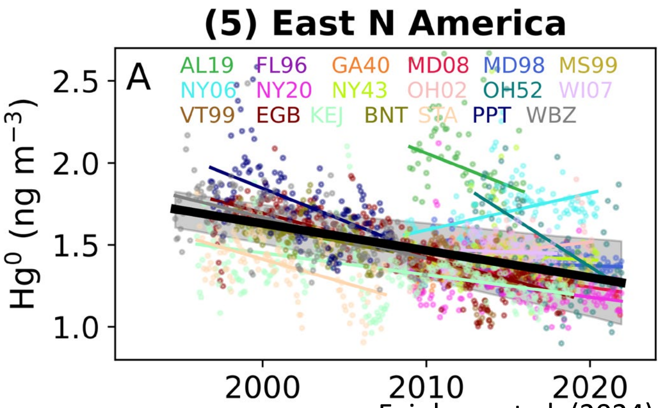


Between PI and PD have shifts in: oxidation pathway, Hg^0 lifetime, and deposition patterns

Just at the start: how this impacts measurements (e.g., Hg isotopes) and other periods?

Atmospheric chemistry impact on Hg cycling: past

What is the effect of chemical changes on recent Hg trends? Does it change by region?



Feinberg et al. (2024)

Hg is used as a proxy for volcanic emissions in paleoscience — how are interpretations affected by chemical lifetime changes in different periods?

Earth-Science Reviews 196 (2019) 102880

Contents lists available at ScienceDirect

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journal homepage: www.elsevier.com/locate/earscirev

Mercury as a proxy for volcanic emissions in the geologic record

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nature communications

Article

https://doi.org/10.1038/s41467-022-35272-8

Mercury evidence from southern Pangea terrestrial sections for end-Permian global volcanic effects

Received: 21 August 2022

Accepted: 25 November 2022

Published online: 03 January 2023

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PNAS

Mercury evidence for pulsed volcanism during the end-Triassic mass extinction

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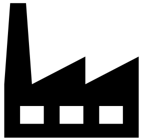
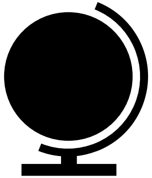
Edited by Edward A. Boyle, Massachusetts Institute of Technology, Cambridge, MA, and approved May 19, 2017 (received for review April 3, 2017)

Takeaway message

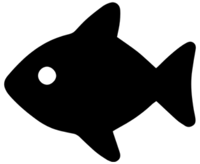
In the Northern Hemisphere Hg^0 oxidation sped up between preindustrial and present day due to OH and O_3 increases



In the Southern Hemisphere Hg^0 oxidation slowed down due to less atomic Br, more partitioning to reservoir Br_y



Impacts: The role of atmospheric chemistry changes in aggravating human Hg exposure risks via altering deposition patterns has been previously overlooked



Recently accepted in *AGU Advances*, doi: 10.1029/2025AV002158

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Acknowledgements

- Funding for this project: Horizon Europe Marie Skłodowska Curie Postdoc Fellowship (#101103544)
- HPC support from the Derecho system (doi:10.5065/qx9a-pg09) provided by the NSF National Center for Atmospheric Research (NCAR), sponsored by the National Science Foundation
- Thanks to all researchers involved in collection of public atmospheric Hg measurement datasets
- Thanks to all colleagues at IQF-CSIC, and collaborators that made this work possible!

AC2 group at IQF

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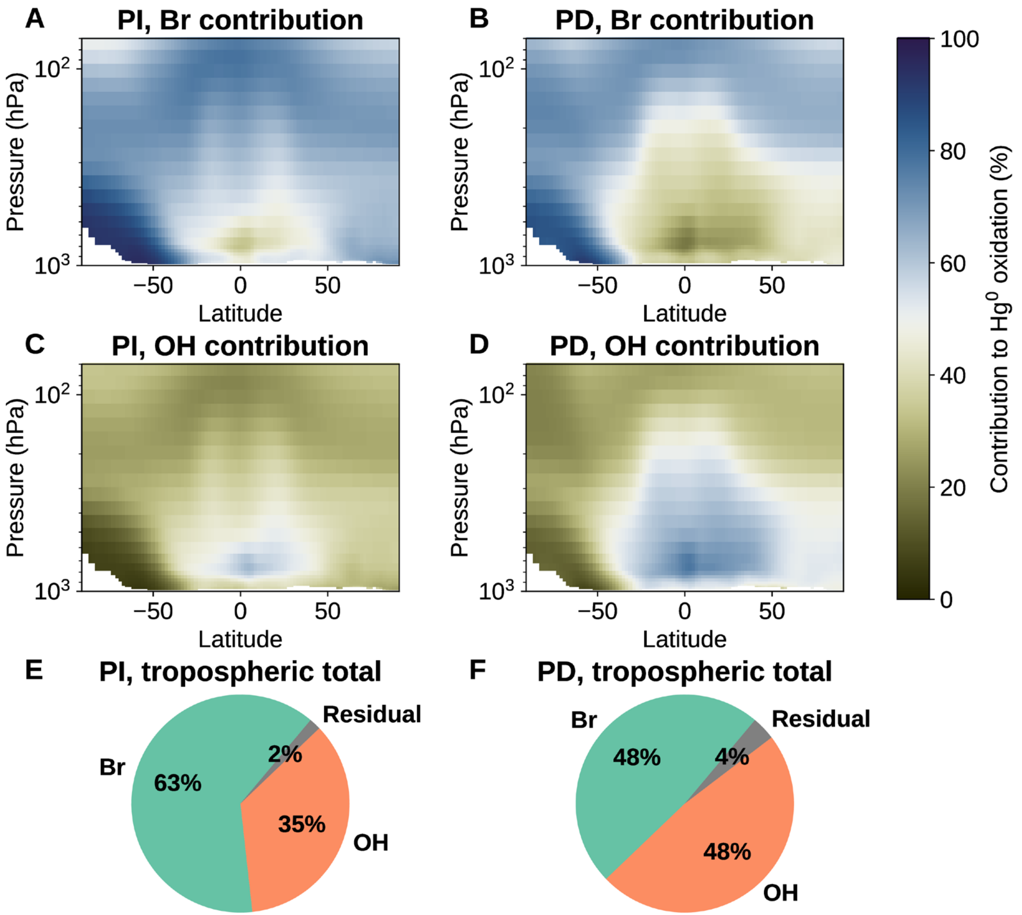
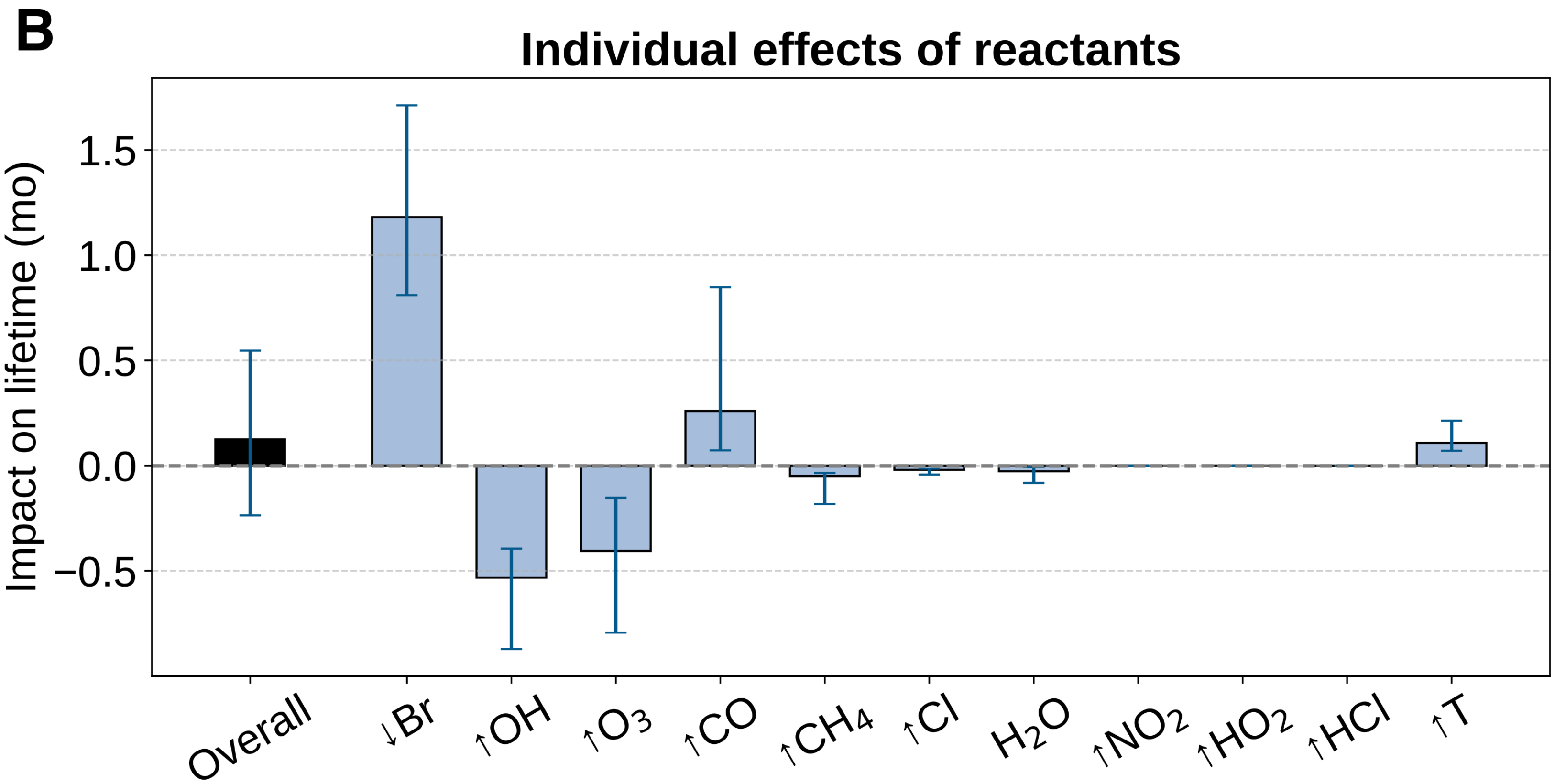
Co-funded by the European Union

Extra: contribution of Br and OH



Extra: one-at-a-time effects



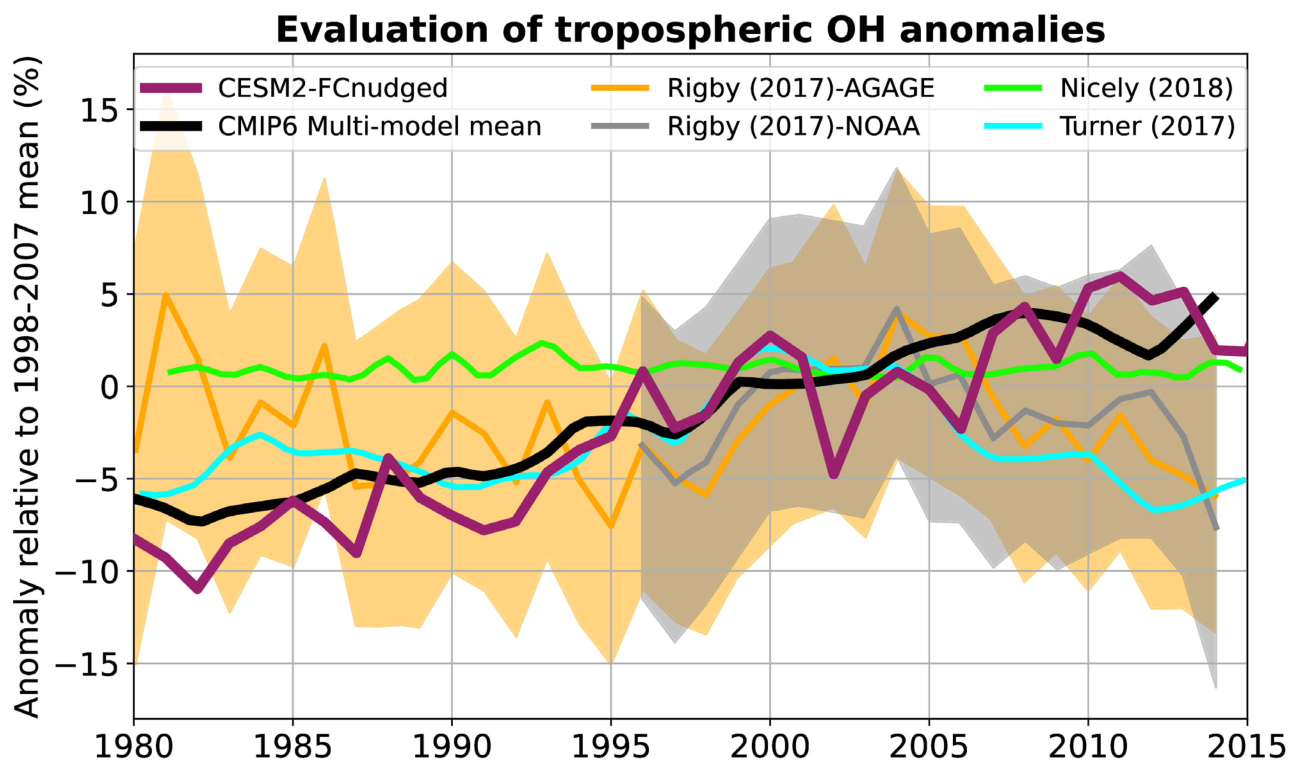
Extra: PI to PD changes in CESM2 chemistry

Table S1. Comparison of modelled changes in major Hg oxidants from PI to PD with respect to literature studies.

Type	(PD – PI) / PI × 100	Periods	Reference
OH			
Model	25 ± 2%	1850 (PI); 2010–2019 (PD)	This study*
Model	8 to 10%	1850–1859 (PI); 2005–2014 (PD)	AERChemMIP models[29]
Model	–12.7 to 14.6%	1850 (PI); 2000 (PD)	ACCMIP models[30]
Model	2.2%	1850 (PI); 2000 (PD)	CESM1-CAM-Chem-SLH[31]
O_3			
Model	64 ± 3%	1850 (PI); 2010–2019 (PD)	This study*
Model	20 to 58%	1850 (PI); 2005–2014 (PD)	AERChemMIP models[32]
Model	20 to 57%	1850 (PI); 2000 (PD)	ACCMIP models[33]
Model	41%	1850 (PI); 2000 (PD)	CESM1-CAM-Chem-SLH[31]
Model	48%	Unspecified (PI); 2007 (PD)	GEOS-Chem[34]
Measurement	<40%	1590–1958 (PI); 1992–2016 (PD)	Isotopes in natural archives[35]
Br			
Model	–37 ± 3%	1850 (PI); 2010–2019 (PD)	This study*
Model	–39%	1850 (PI); 2000 (PD)	CESM1-CAM-Chem-SLH[36]
Model	–28%	Unspecified (PI); 2007 (PD)	GEOS-Chem[34]

* For the values calculated in this study, we provide the mean change with the standard deviation calculated from interannual variability.

Extra: OH changes



Minimal differences between this study and CMIP6 version of WACCM for observed period (1980–2015)
Larger differences for previous period (1850–1980); not due to halogens or emissions

Extra: O3 changes