Long-Term Changes in Contributions of Anthropogenic and Natural Perturbations to Atmospheric Mercury in the United States

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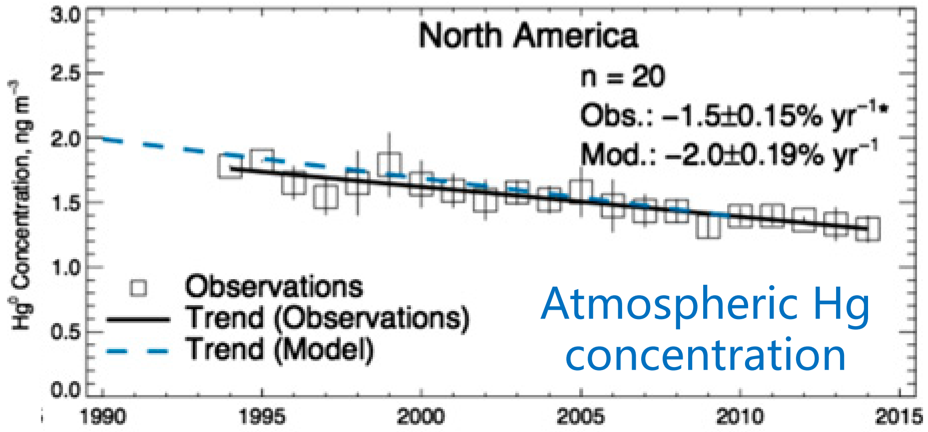
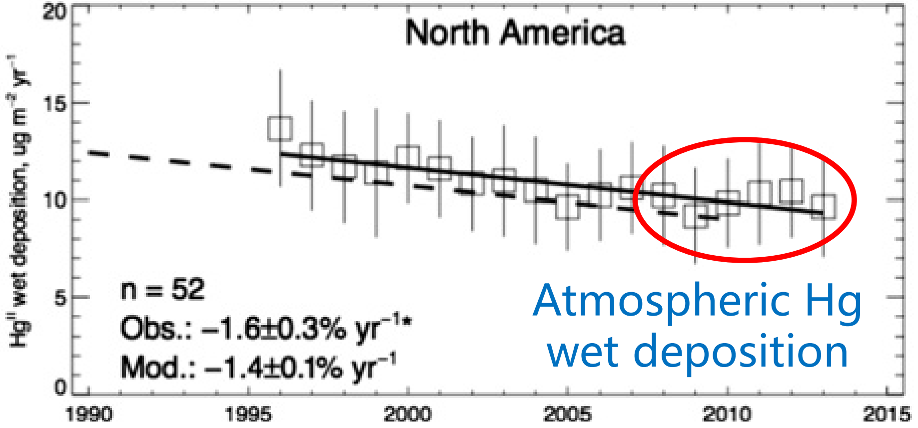
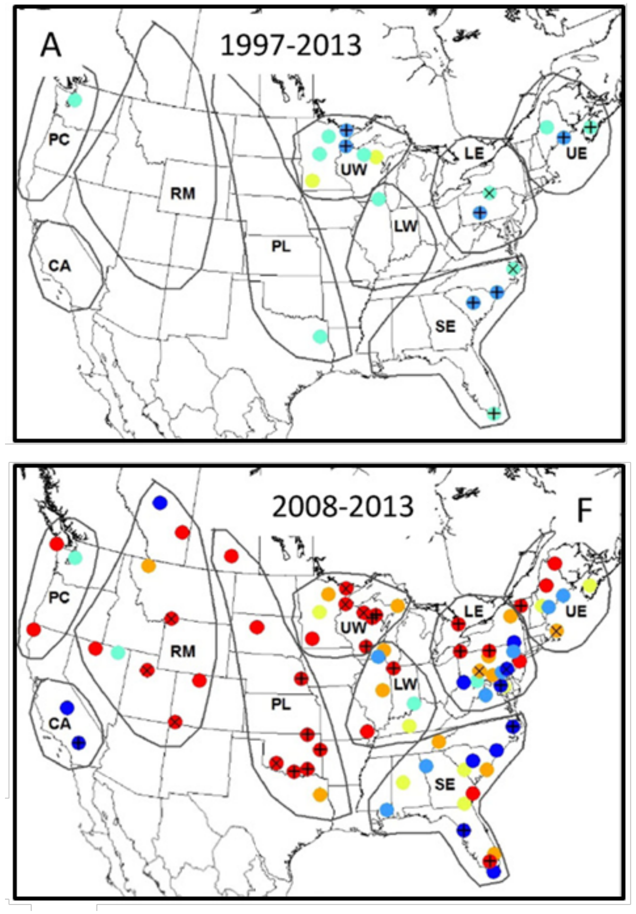
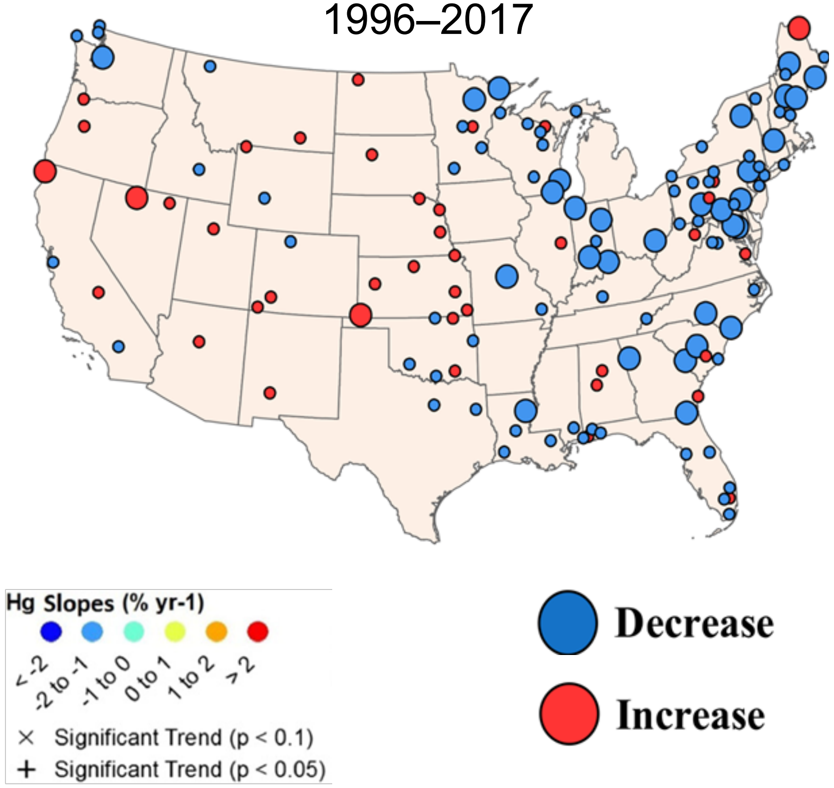
Long-term change of atmospheric Hg pollution in the US







Precipitation Hg concentration

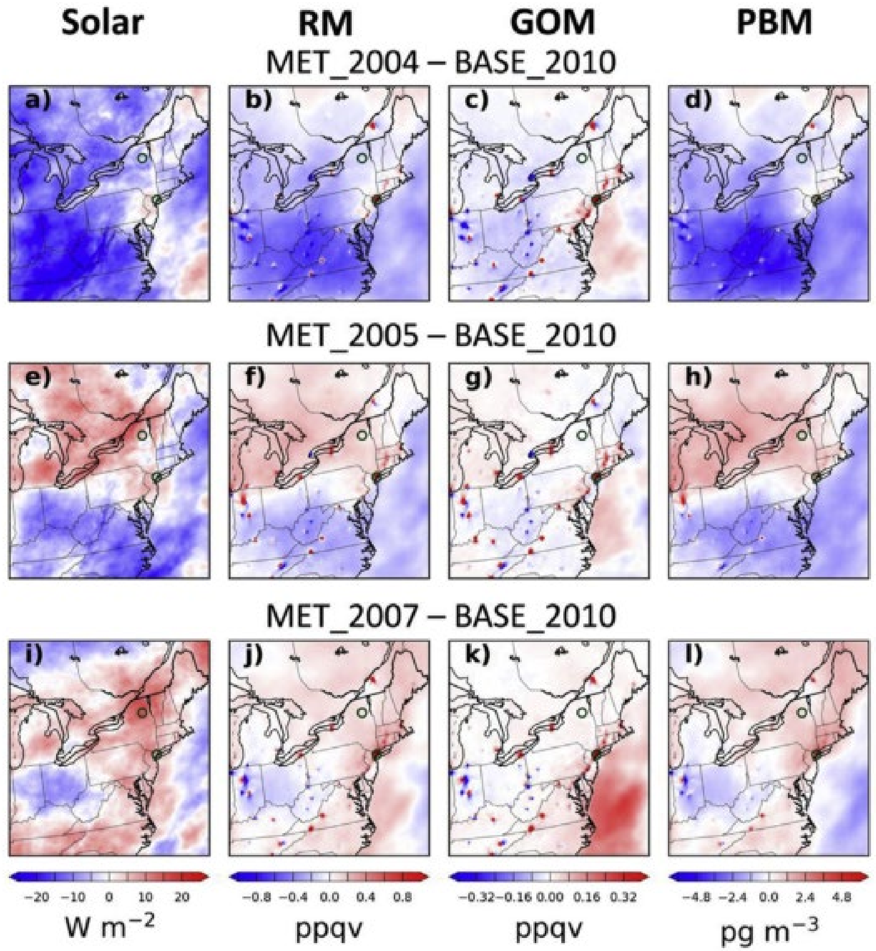


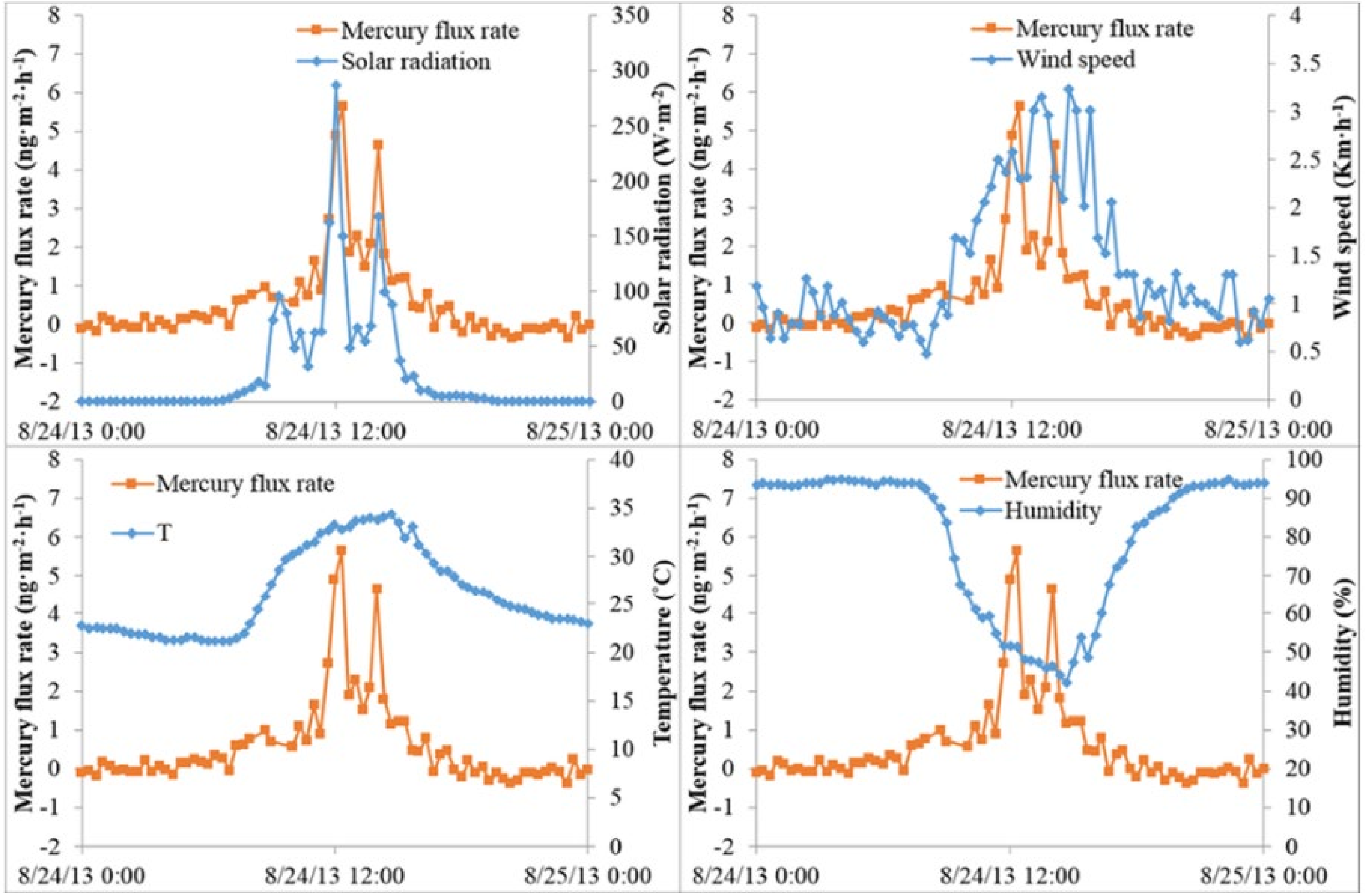
Long-term decrease in North America during 1995–2013

Overall decreasing trend in the eastern US during 1995–2017, but significant increasing trend in the western US after ~2008

Zhang et al., ***Proc. Natl. Acad. Sci.***, 2016; Weiss-Penzias et al., ***Sci. Total Environ.***, 2016; Olson et al., ***Environ. Sci. Technol. Lett.***, 2020

Impacts of meteorology on atmospheric Hg pollution

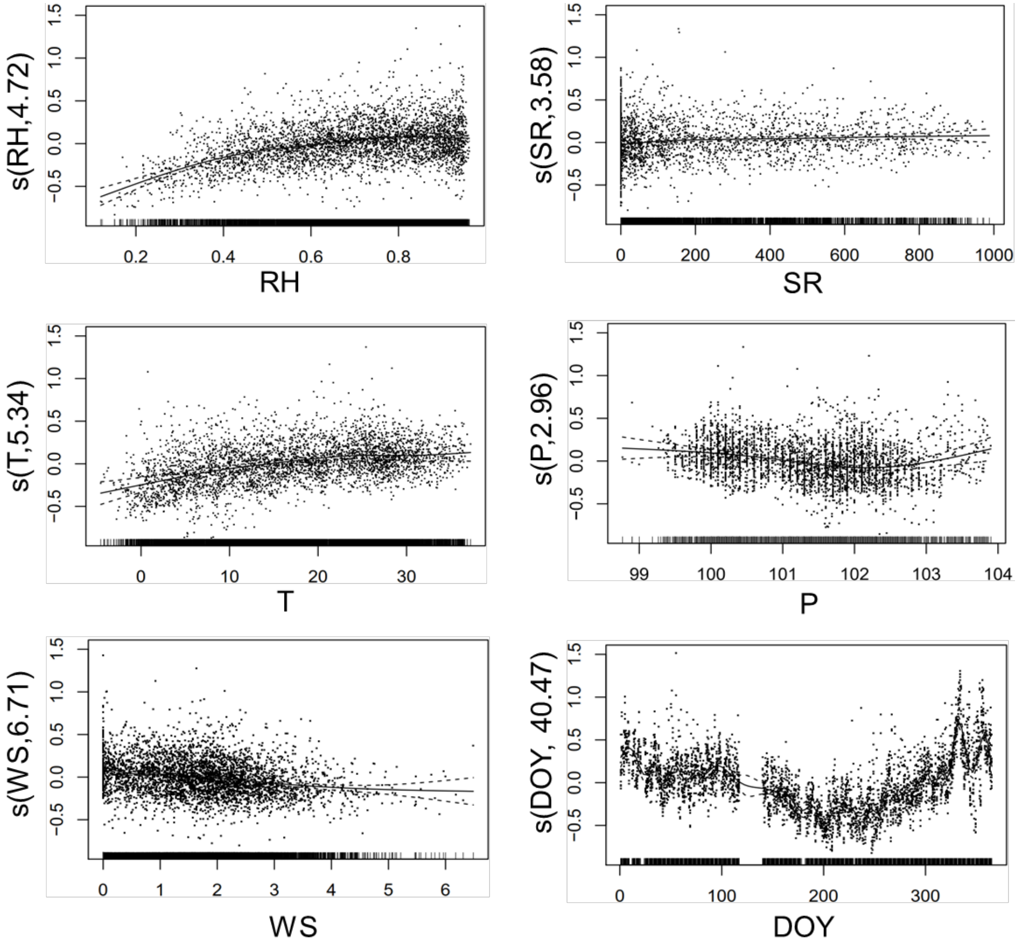


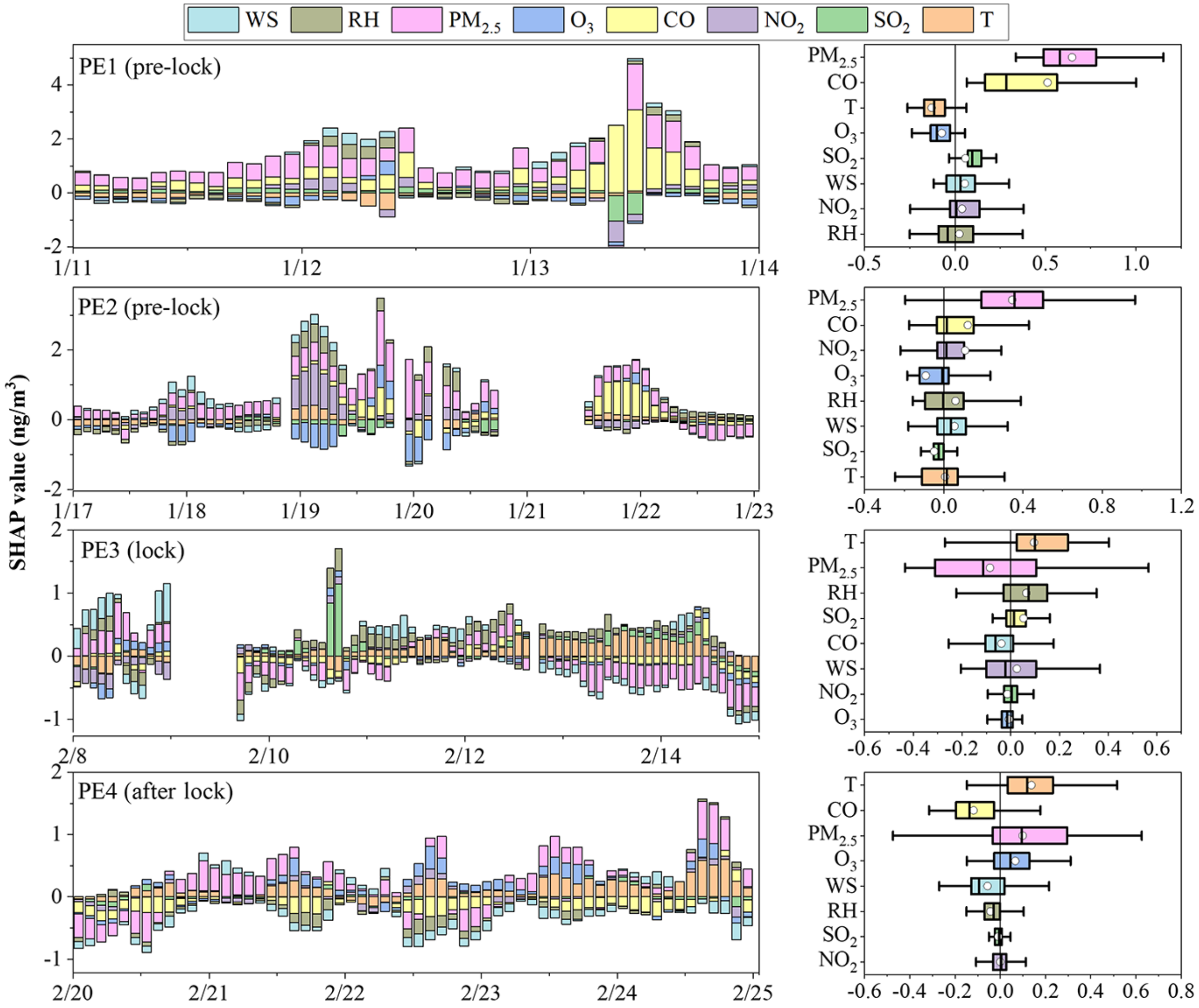


Solar radiation, **wind speed**, **temperature** and **humidity** are key to atmospheric Hg concentration and deposition

Ye et al., ***Atmos. Environ.***, 2019; Cizdziel et al., ***Atmosphere***, 2019

Quantification of anthropogenic and natural contributions





Advanced statistical and machine learning models have been used to quantify impacts of key factors

Zhang et al., ***Atmos. Environ.***, 2021; Qin et al., ***Atmos. Chem. Phys.***, 2022

Scientific questions in this study

- How did long-term anthropogenic and natural perturbations change regional atmospheric Hg pollution patterns?
- How different are the changes of atmospheric Hg concentration and wet deposition flux in response to the perturbations?
- What will be the influence of global climate change in future on the regional atmospheric Hg pollution patterns?

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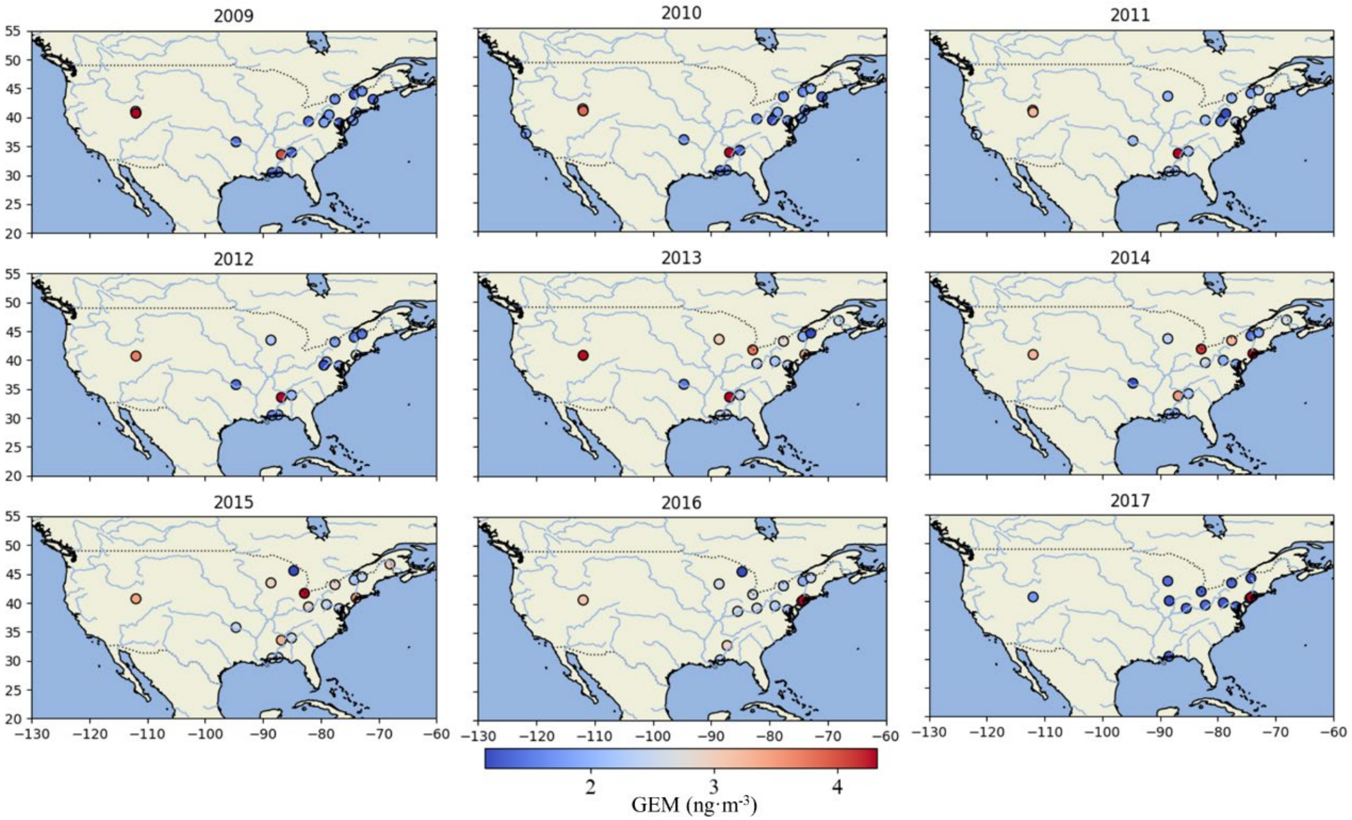
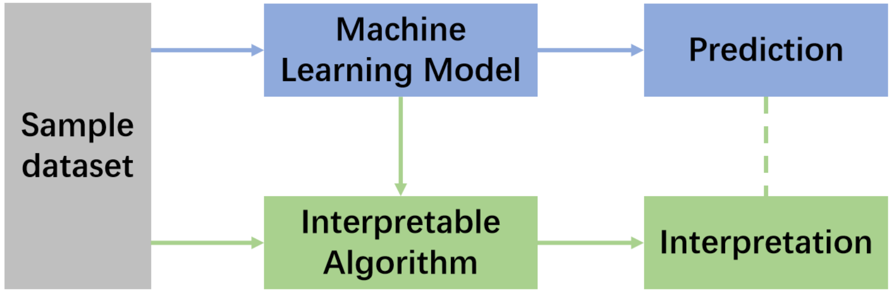
A machine learning model for atmospheric Hg concentration





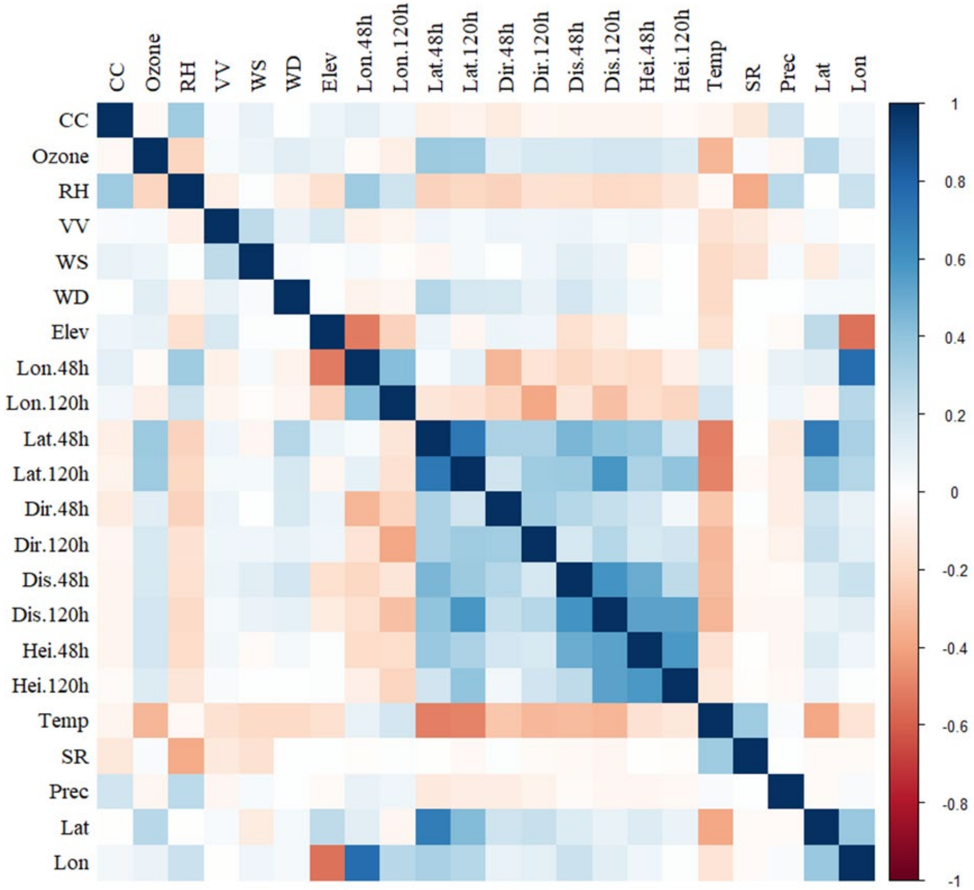
XGBoost

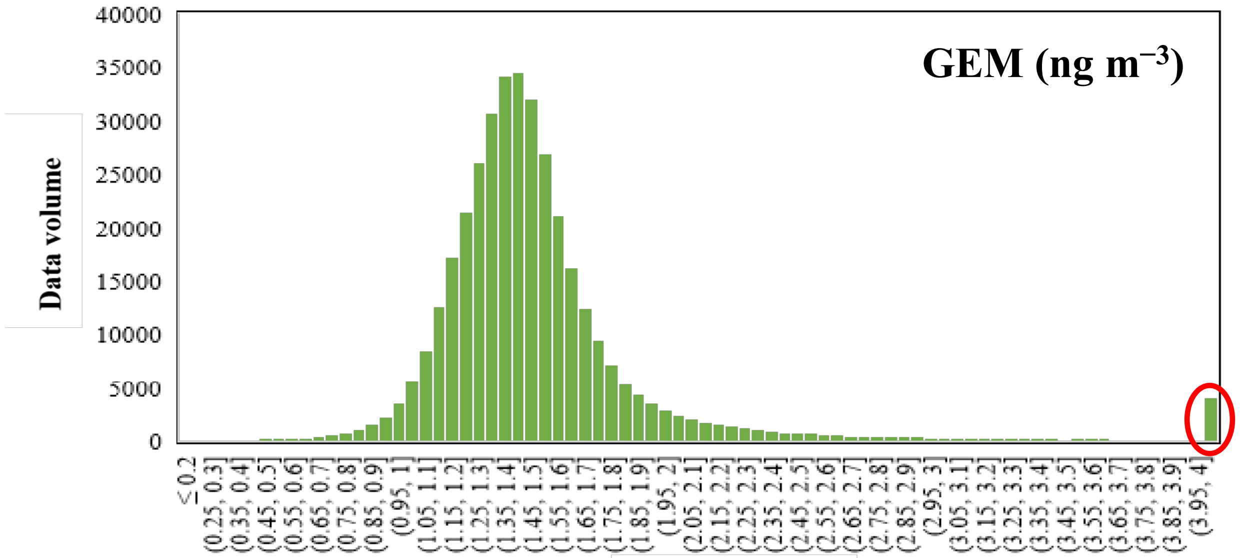
+

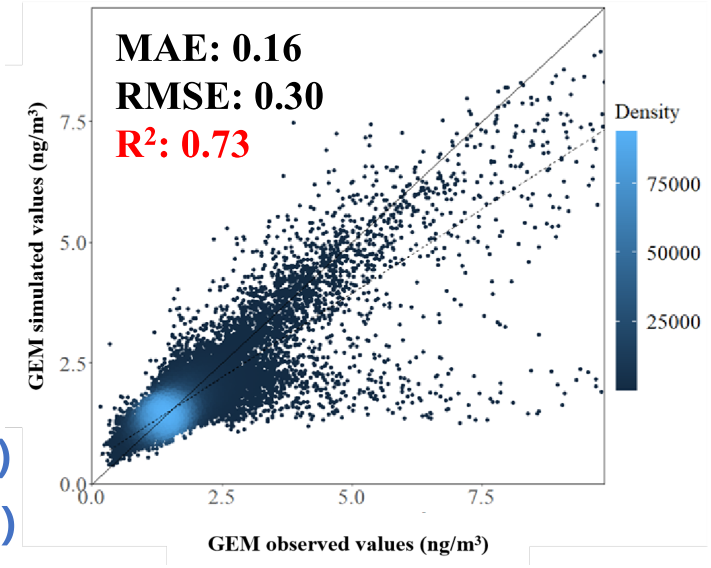
SHAP

Database	Variables
AMNet	✓ GEM (**hourly data**, ng m^{-3}) ✓ Elevation (Elev, m) ✓ Longitude (Lon, °) ✓ Latitude (Lat, °)
HYSPLIT	✓ 48-h backward trajectory ✓ 120-h backward trajectory
ECMWF (ERA5)	✓ Relative humidity (RH, %) ✓ Cloud cover (CC, %) ✓ Solar radiation (SR, J m^{-2}) ✓ Total column ozone (Ozone, kg m^{-2}) ✓ Wind speed (WS, m s^{-1}) ✓ Wind direction (WD, °) ✓ Temperature (Temp, K) ✓ Precipitation (Prec, mm) ✓ Vertical velocity (VV, Pa s^{-1})

XGBoost model configuration and validation







No.	R^2	RMSE	MAE
1	0.724	0.295	0.157
2	0.715	0.304	0.158
3	0.736	0.295	0.157
4	0.734	0.287	0.157
5	0.738	0.294	0.156
6	0.739	0.296	0.158
7	0.731	0.301	0.159
8	0.722	0.304	0.159
9	0.733	0.299	0.157
10	0.734	0.288	0.157

- Limited correlations between any two predictors
- Outliers removed for the dependent variable (GEM)
- Good model performance (10-fold cross-validation)

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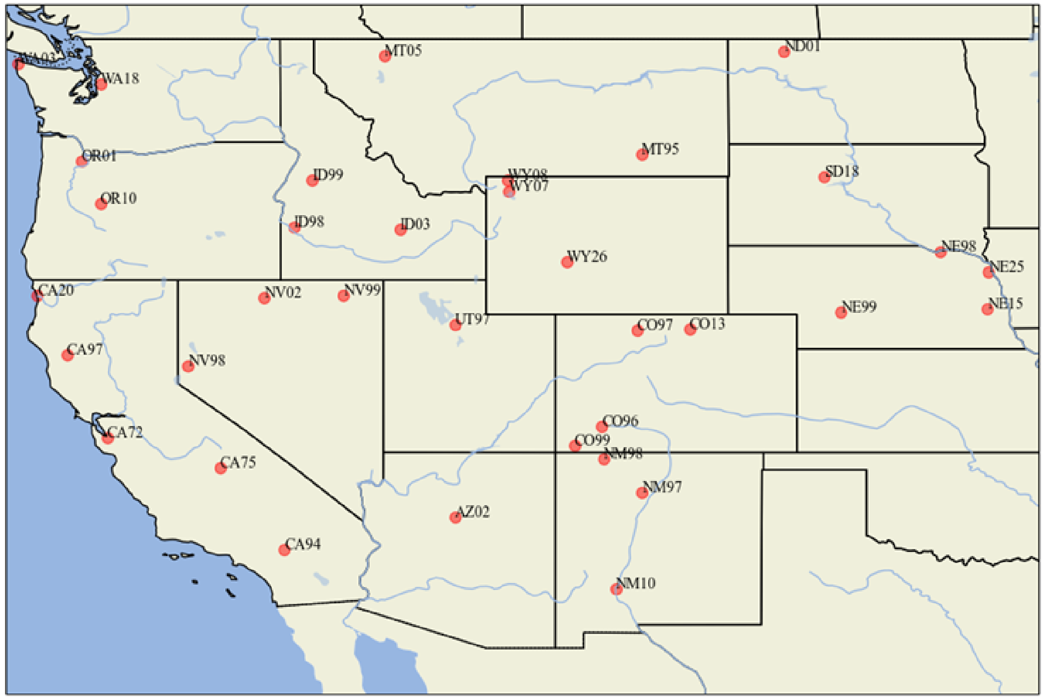
- Introduction
- **Methodology**
  - The machine learning model (XGBoost)
  - **The generalized additive model (GAM)**
- Impacts on Atmospheric Mercury Concentration
- Impacts on Atmospheric Mercury Wet Deposition
- Take-Home Messages

An generalized additive model for Hg wet deposition flux



Target region: Western US

Generalized Additive Model (GAM)

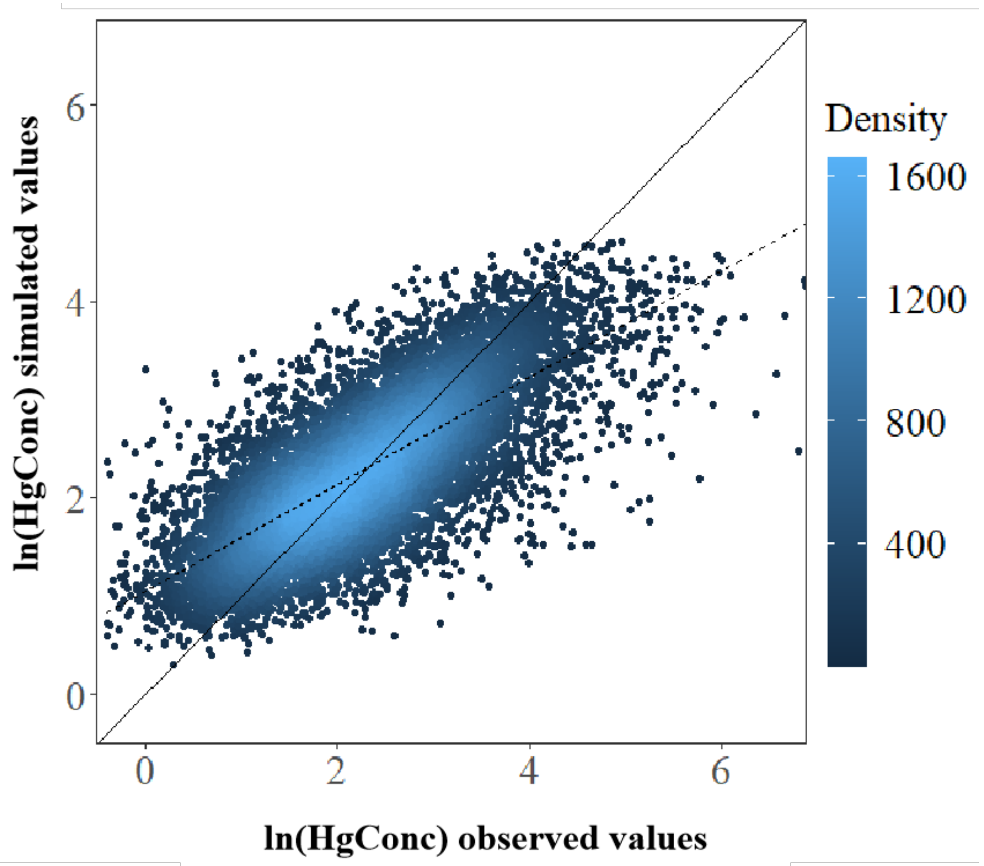
$$g(\mu) = a_0 + s_1(x_1) + s_2(x_2) + \cdots + s_k(x_k)$$

- Observation data: Mercury Deposition Network (MDN)
- Historical meteorological data: European Centre for Medium-range Weather Forecast (ECMWF)
- Future meteorological data: Coupled Model Intercomparison Project Phase 6 (CMIP6)

Database	Variables	
MDN	✓ Precipitation Hg concentration (HgConc, **weekly data**, ng L^{-1})	✓ Precipitation (Prec, mm) ✓ Elevation (Elev, m)
ECMWF (ERA5)	✓ Relative humidity (RH, %) ✓ Total cloud cover (TCC, %) ✓ Solar radiation (SR, W m^{-2})	✓ Wind speed (WS, m s^{-1}) ✓ Temperature (Temp, K)
ECMWF (CMIP6)	✓ Relative humidity (RH, %) ✓ Total cloud cover (TCC, %) ✓ Solar radiation (SR, W m^{-2})	✓ Wind speed (WS, m s^{-1}) ✓ Temperature (Temp, K) ✓ Precipitation (Prec, mm)

Performance of the generalized additive model

$$\ln(HgConc) \sim s(Prec) + s(Temp) + s(TCC) + s(WS) + s(RH) + s(Elev) + s(SR)$$



No.	Random cross-validation		Temporal cross-validation		Spatial cross-validation	
	R^2	RMSE	R^2	RMSE	R^2	RMSE
1	0.589	0.687	0.462	0.762	0.557	0.744
2	0.548	0.725	0.526	0.701	0.505	0.745
3	0.504	0.771	0.544	0.715	0.534	0.634
4	0.543	0.699	0.481	0.785	0.452	0.711
5	0.510	0.725	0.548	0.741	0.515	0.831
6	0.523	0.742	0.555	0.759	0.489	0.766
7	0.571	0.710	0.527	0.728	0.539	0.791
8	0.526	0.732	0.588	0.682	0.448	0.723
9	0.542	0.715	0.556	0.725	0.342	0.751
10	0.502	0.753	0.536	0.695	0.505	0.699

- The dependent variable was selected to be the **logarithm of Hg concentration in precipitation**
- Both **temporal and spatial** 10-fold **cross-validations** show **good** model performance for GAM

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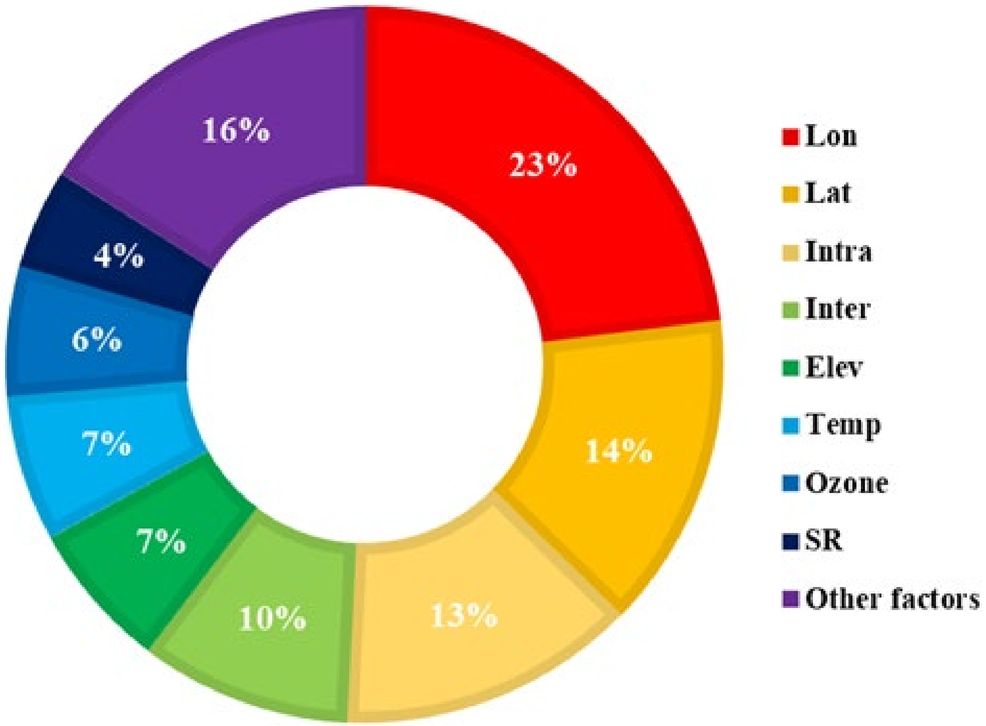
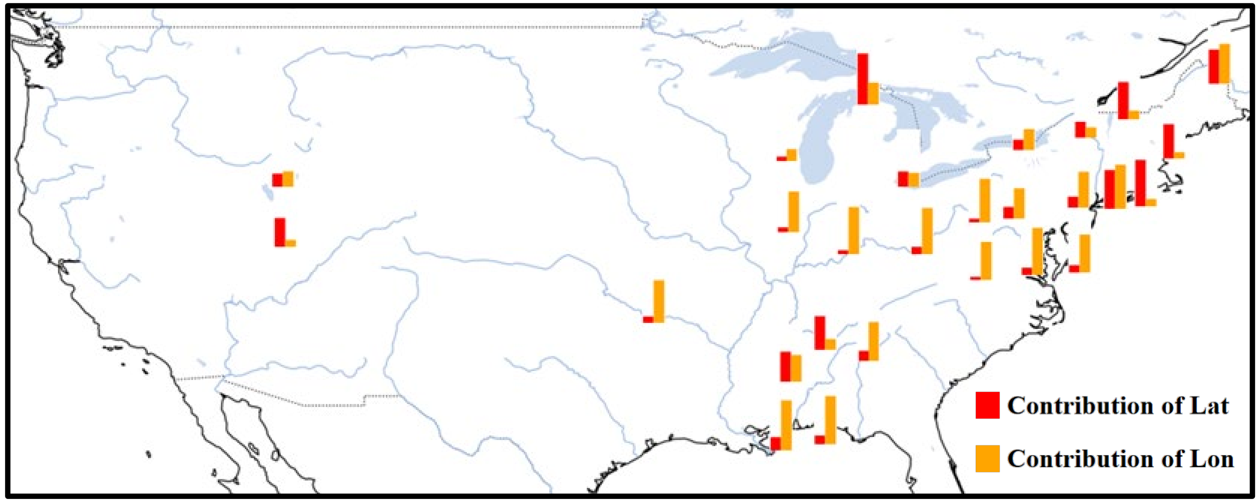
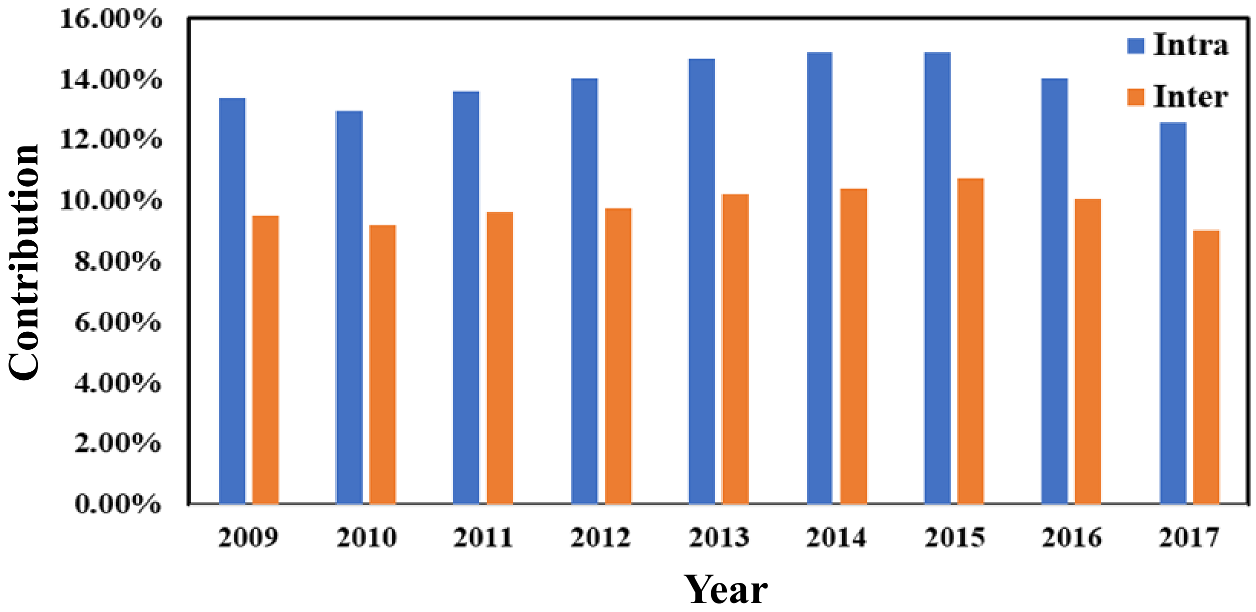
Contributions of key drivers to GEM concentration variation





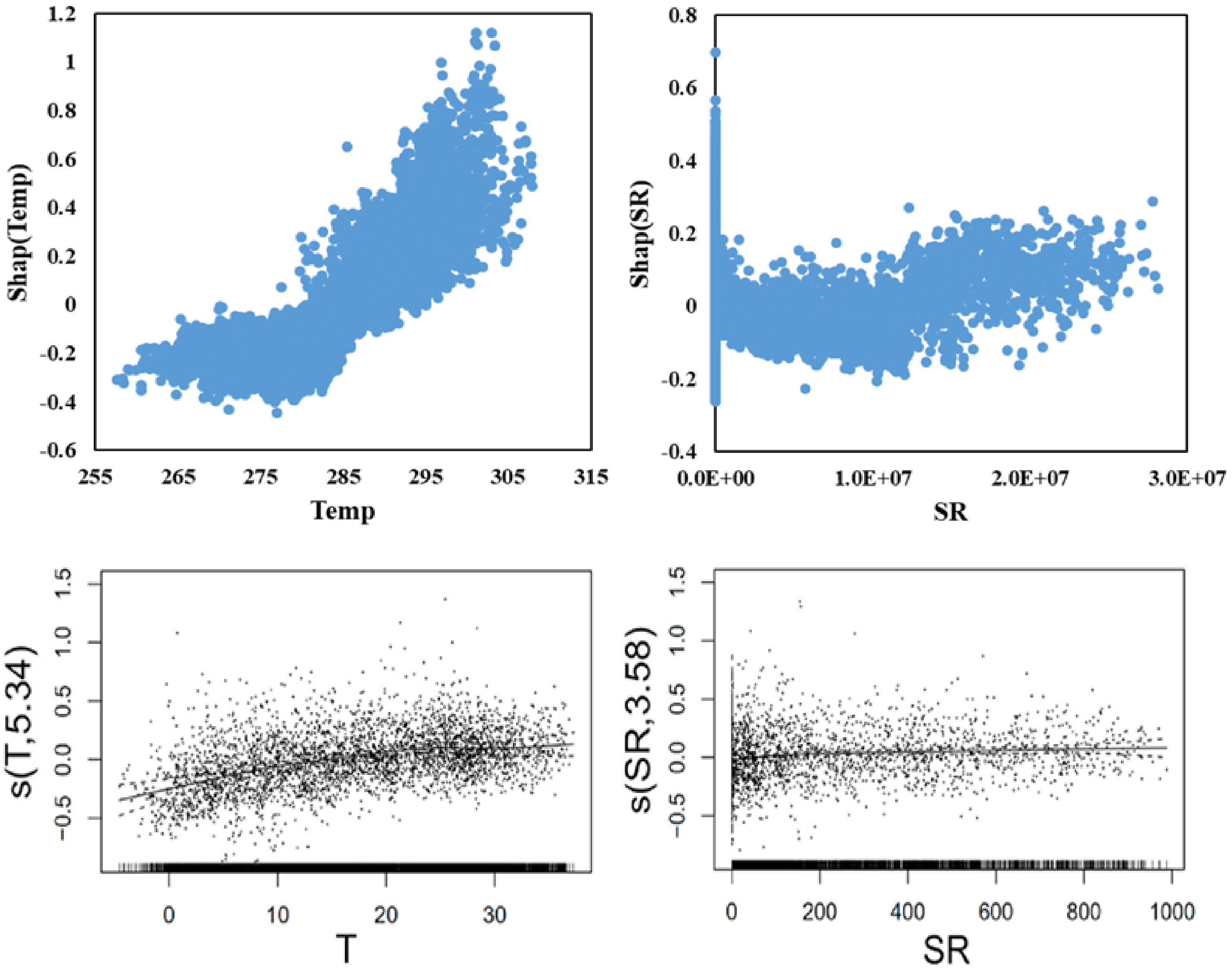


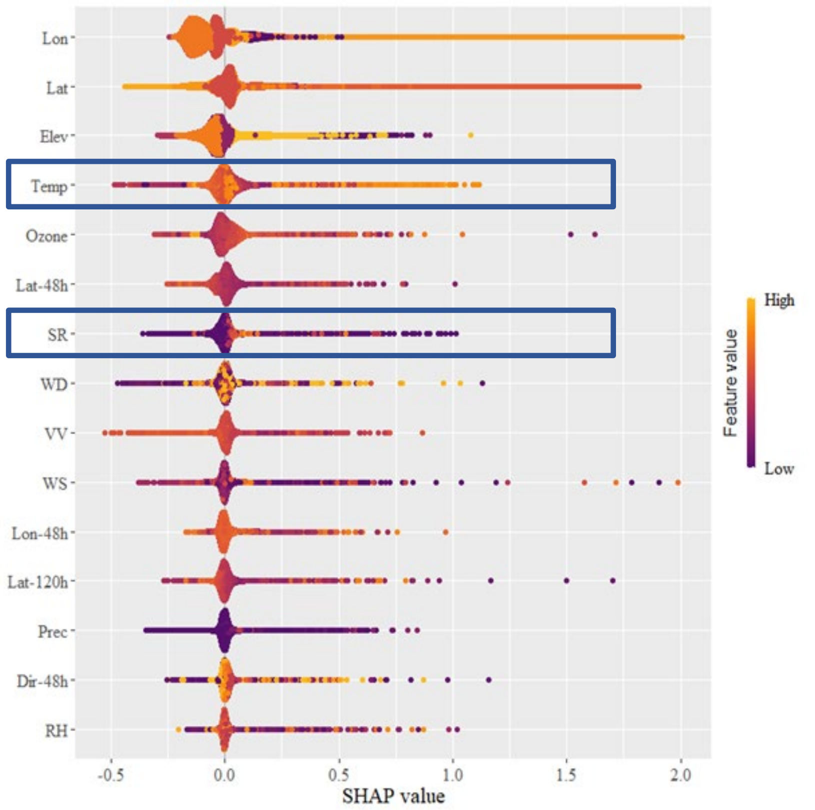
- Geographic features, meteorological factors, and Hg transport contributed 44%, 27%, and 23% to GEM variation, respectively
- The contribution of intra- and inter-regional Hg transport to GEM variation experienced an increase followed by a decrease

Impacts of meteorological factors on GEM concentration

Continental US

Eastern China





- **Temperature and solar radiation are key to GEM concentration**
- **Natural and legacy Hg emissions are important via high temperature**

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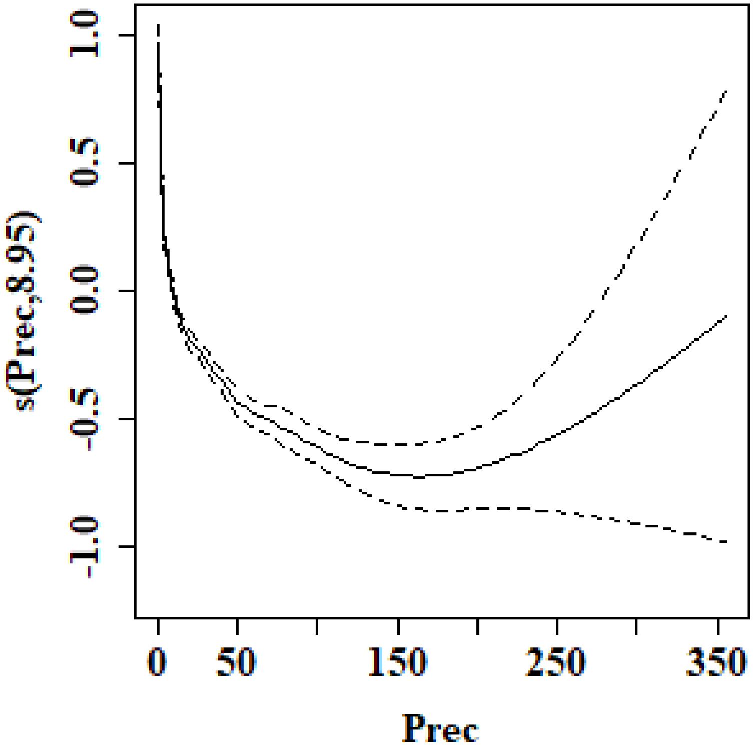
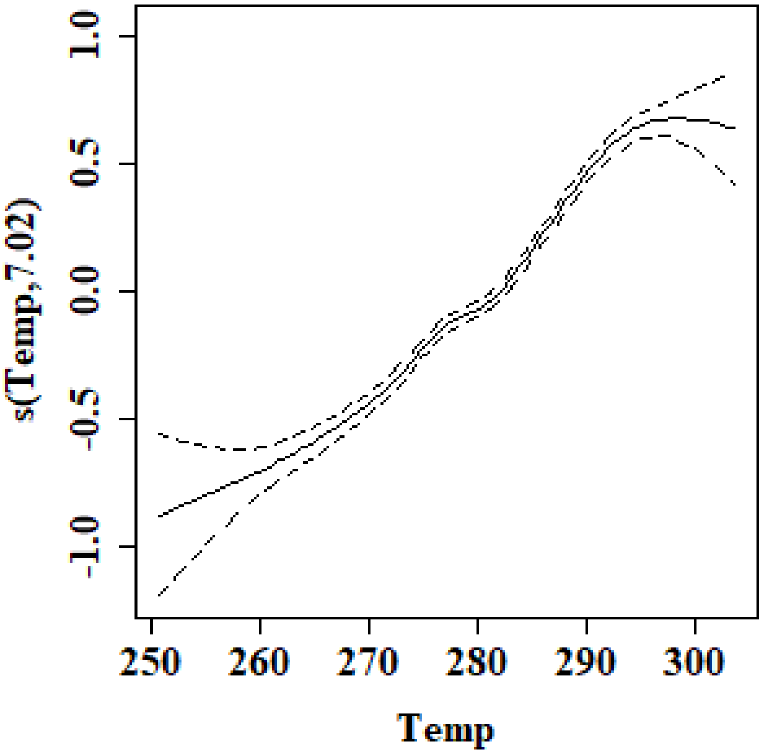
Impacts of meteorology on precipitation Hg concentration

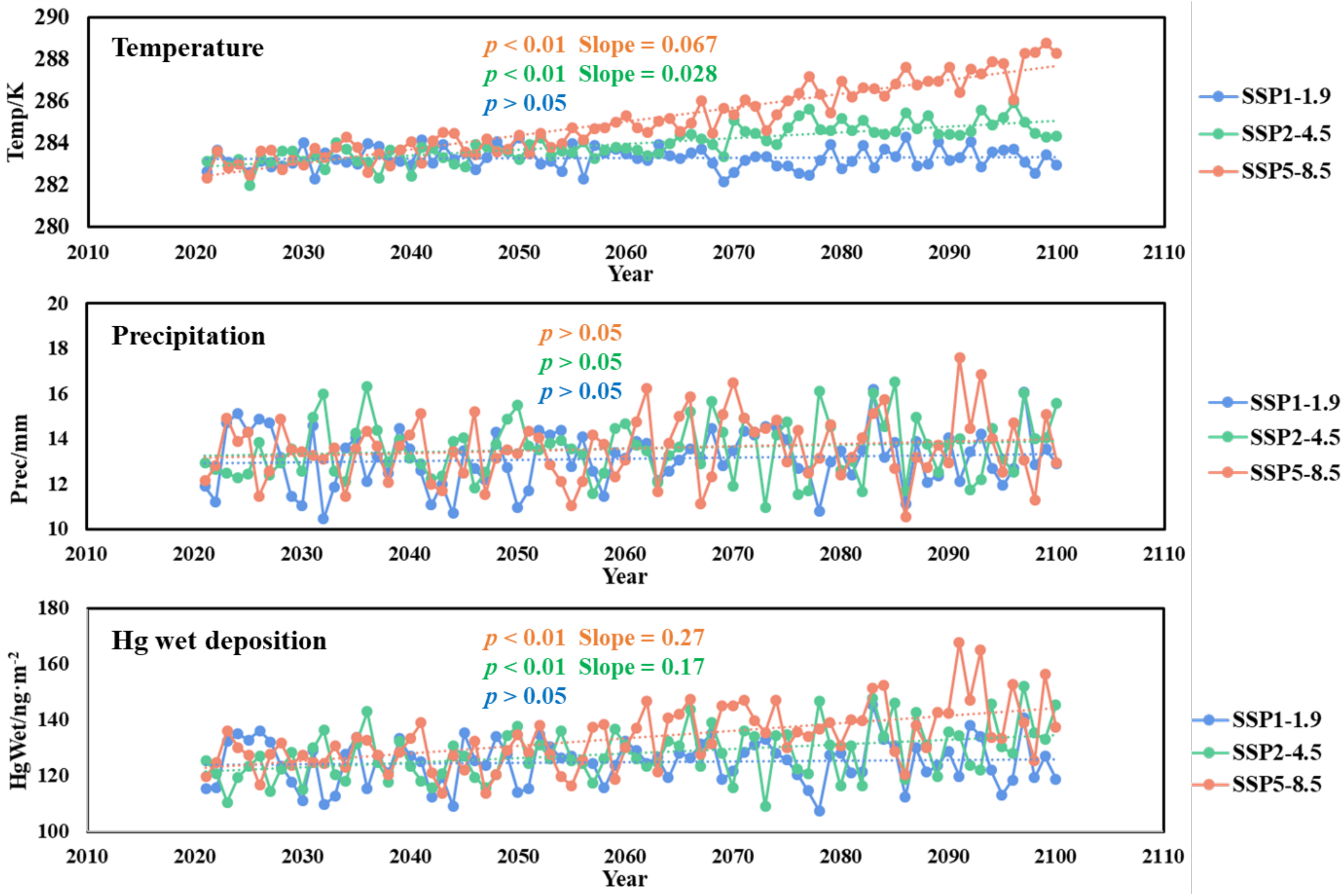




Variable	*F* value	Share	*p* value
Prec	223.96	36.71%	$< 2\times10^{-16}$
Temp	123.58	20.26%	$< 2\times10^{-16}$
SR	97.12	15.92%	$< 2\times10^{-16}$
TCC	58.52	9.59%	$< 2\times10^{-16}$
Elev	43.62	7.15%	$< 2\times10^{-16}$
RH	33.56	5.50%	$< 2\times10^{-16}$
WS	29.70	4.87%	$< 2\times10^{-16}$

- **Precipitation** directly contributes to Hg wet deposition, while Hg concentration in precipitation **decreases dramatically** with increasing precipitation when it is low due to **the dilution effect**
- **Temperature** is **positively** correlated with precipitation Hg concentration, with a similar pattern as on GEM, but in a **milder** way at high level

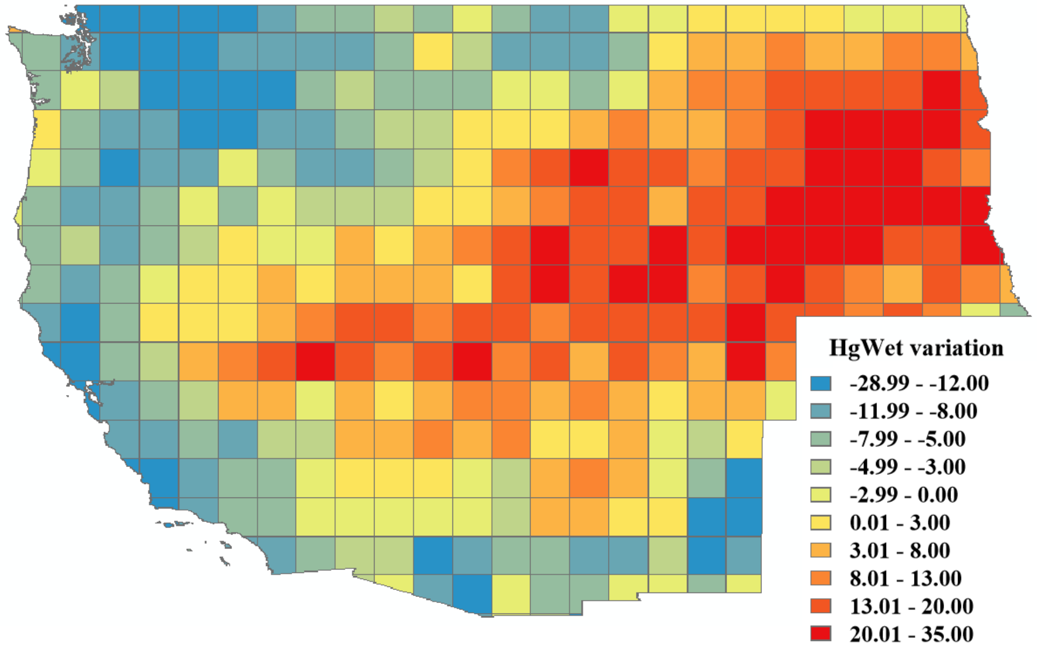
Trends of future meteorology and Hg wet deposition



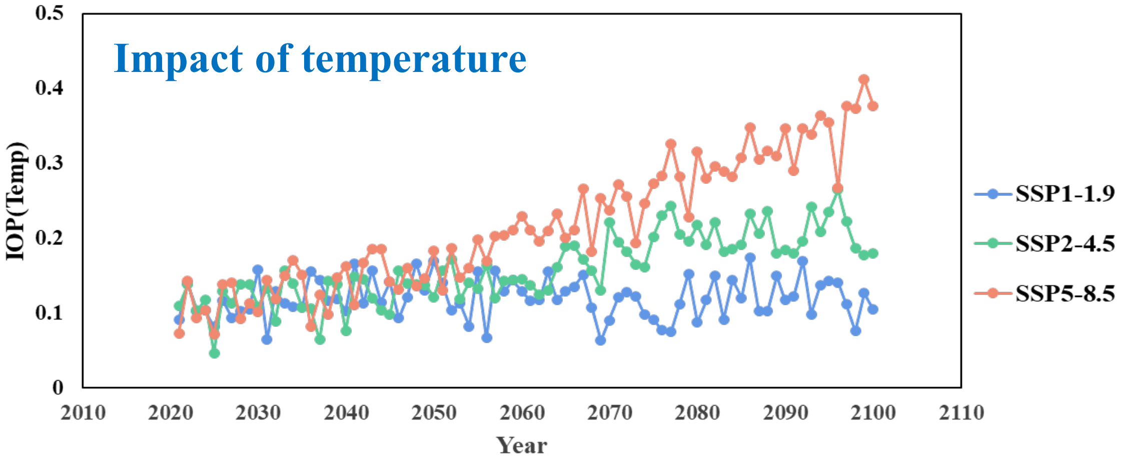
- Hg wet deposition flux will **increase** with more significant **fluctuations** in **SSP2-4.5** or **SSP5-8.5** scenarios
- The **increase** is going to be driven by **increasing temperature**
- The **larger fluctuation** is going to be driven by **extreme precipitation**

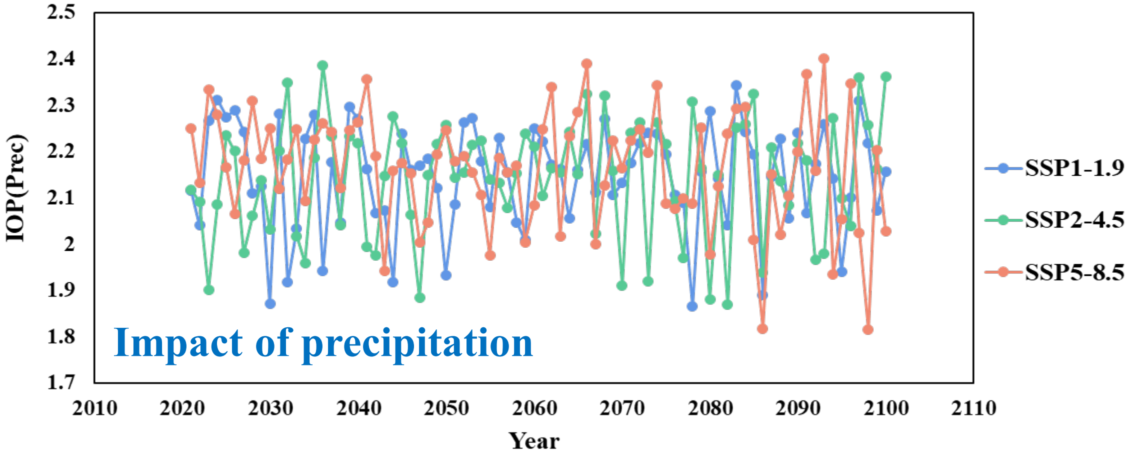
Impacts of future climate change on Hg wet deposition

Changes in Hg wet deposition in the western US from 2030 to 2060



Impacts of temperature and precipitation on Hg wet deposition flux in the western US (2020–2100)





- Hg wet deposition will reduce along the west coast of the US and increase significantly in inland area
- Global warming will boost Hg wet deposition flux, while extreme precipitation will intensify variability

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Take-home messages

- Advanced statistical and machine learning models are effective tools for identifying the key drivers for atmospheric Hg pollution.
- The declining of local anthropogenic emissions and intra- and inter-regional transport drives the decrease of GEM in the eastern US.
- Precipitation and temperature are key factors for Hg wet deposition which will be controlled by future extreme weather conditions.

Thank you very much for your attention

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