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Combating air pollution significantly reduces atmospheric mercury concentrations in China

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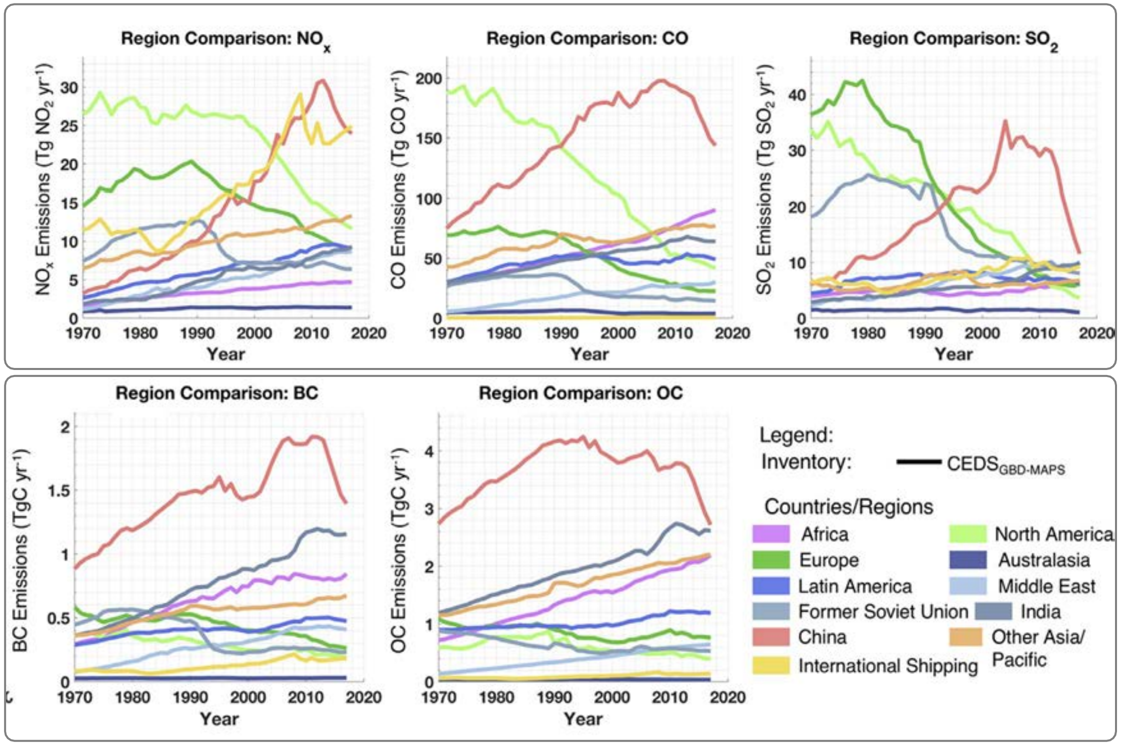
China is the largest emitter of air pollutants worldwide

Global air pollutants emissions by regions



Beijing air pollution in 2013

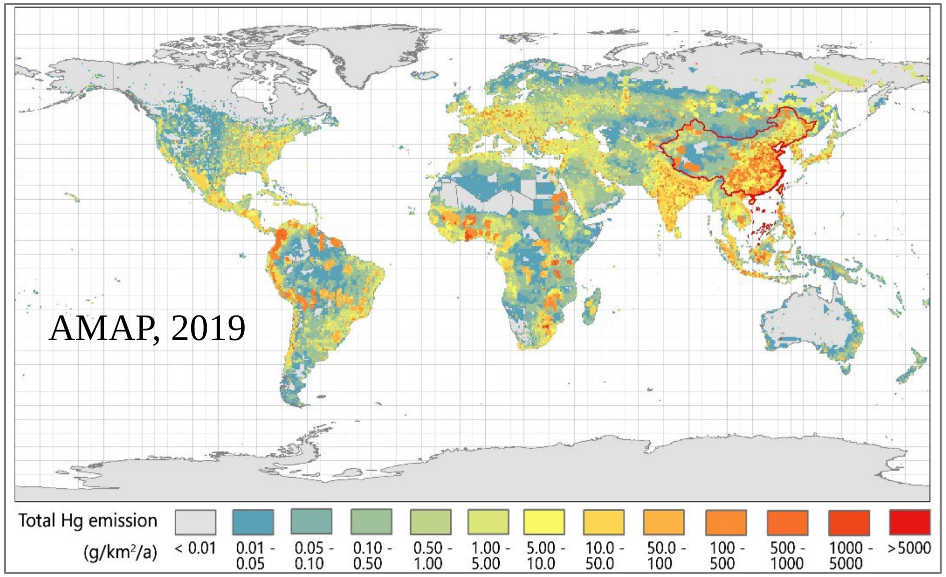
►China is the largest anthropogenic emitter of air pollutants (19-33%), and experienced server and persistent air pollutions in 2013

►China implemented the Clean Air Actions since 2013 and significantly reduced air pollutants emissions and improved air quality, but the effect on atmospheric Hg is not well known

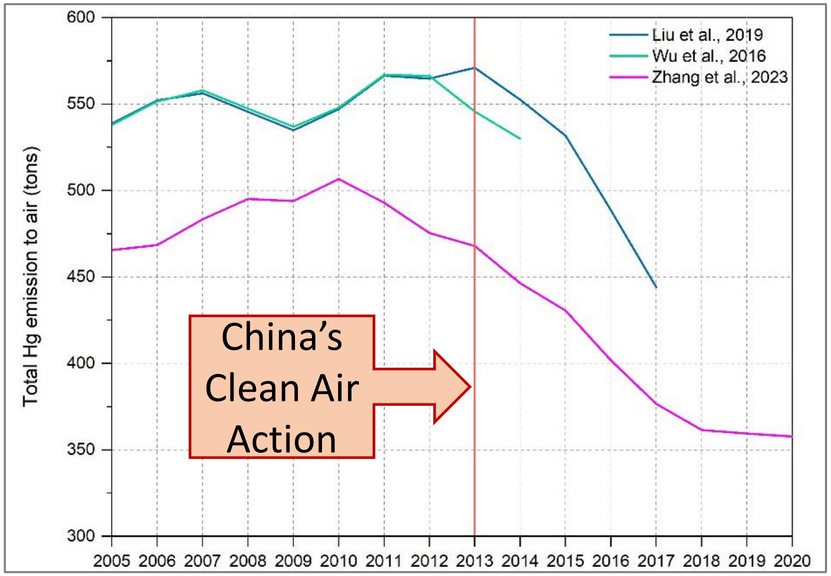
AMAP, 2019; Zheng et al., 2018; McDuffie et al., 2020

China is the largest emitter of air Hg and pollutants

Global anthropogenic Hg emission distribution



Air pollutants emissions by regions



► China is the largest anthropogenic emitter of atmospheric Hg (~30%) worldwide

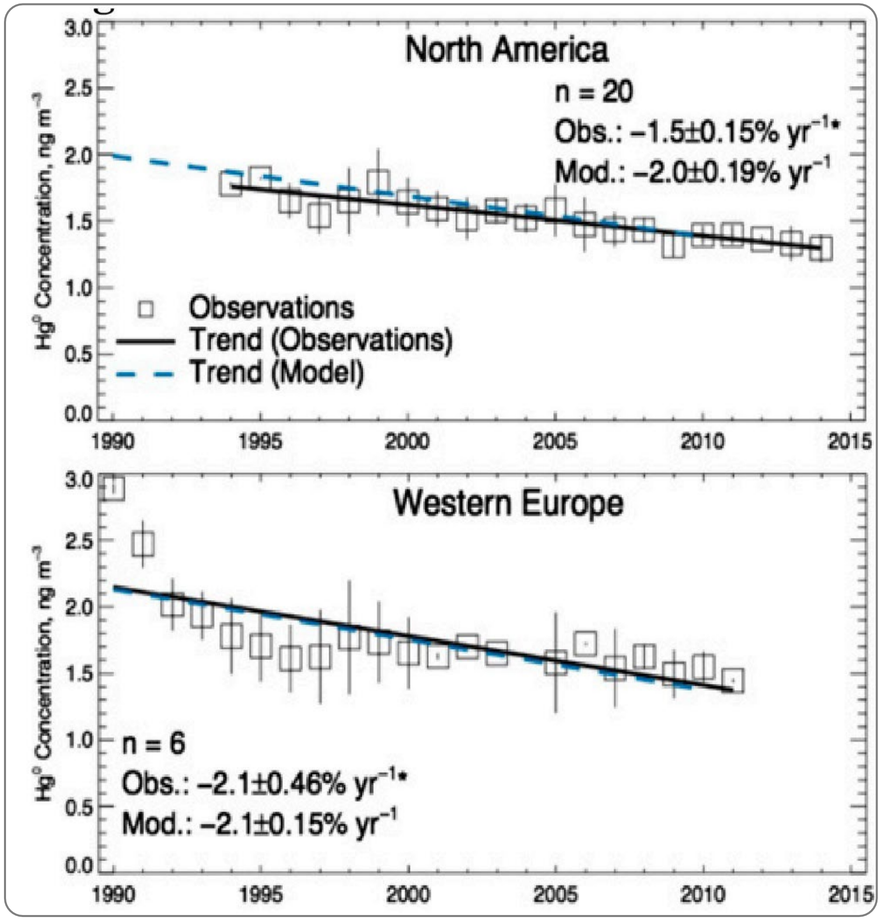
► Previous estimates of China's anthropogenic Hg emissions have many uncertainties

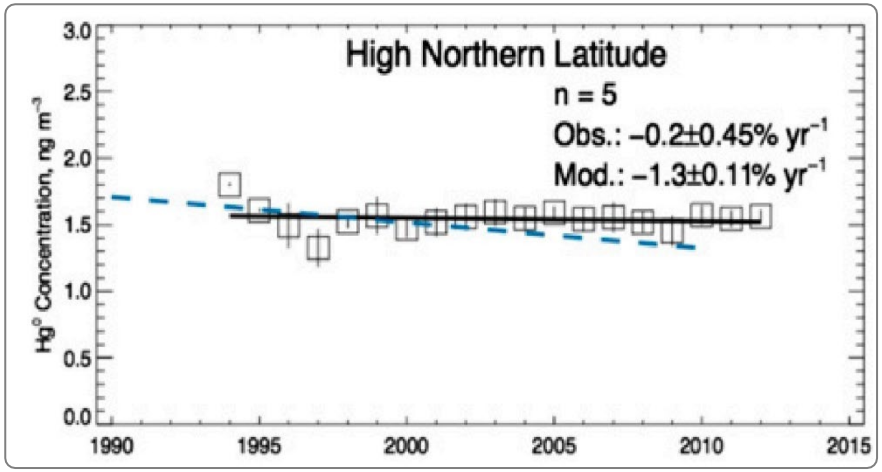
► Long-term continuous observations of atmospheric Hg are very limited in China, making it difficult to constrain the trend in China's anthropogenic Hg emissions and inform the effectiveness of Minamata Convention in China

AMAP, 2019; Wu et al., 2016; Liu et al., 2019; Zhang et al., 2023

Long-term trend analysis of GEM support the effectiveness of the Minamata Convention

Long-term GEM observations in NA, Eu and Polar region



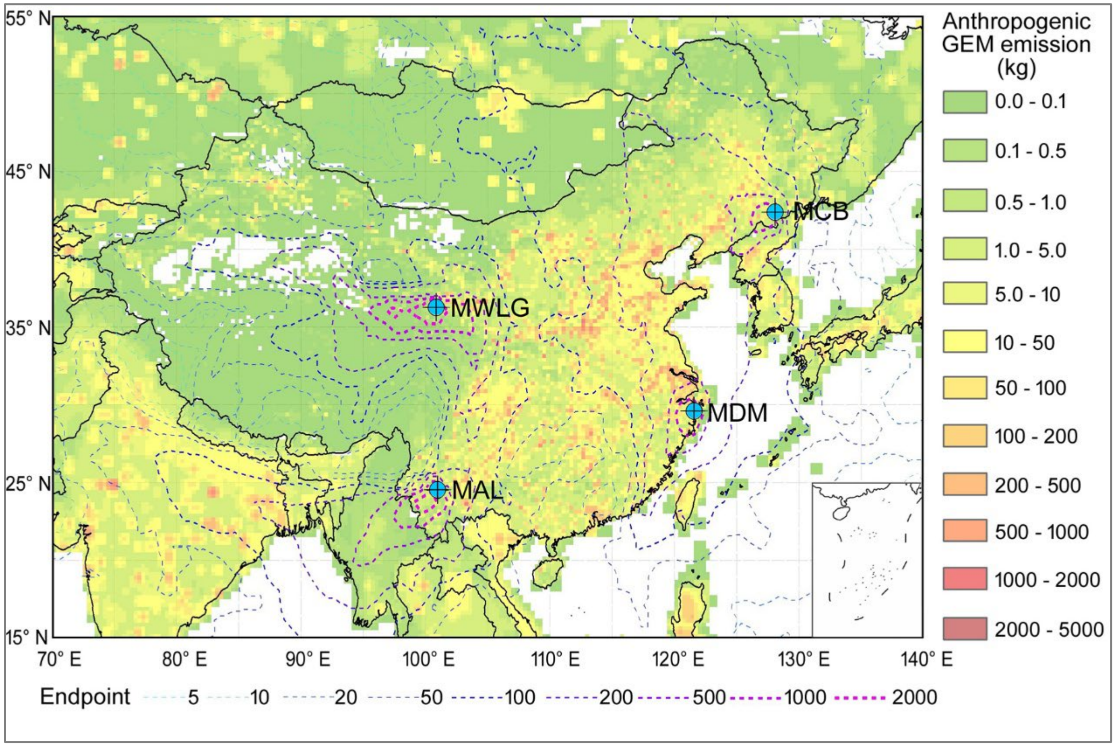


► **Due to the controls of criteria pollutants emissions, GEM concentrations in North America, Europe and Polar region were decreasing since 1990, successfully supported the reductions of anthropogenic GEM emissions in these regions**

Zhang et al., 2016

Long-term sampling sites in China

Site locations and air masses origins



Mt. Waliguan
2007-

Mt. Changbai
2008-

Mt. Ailao
2011-

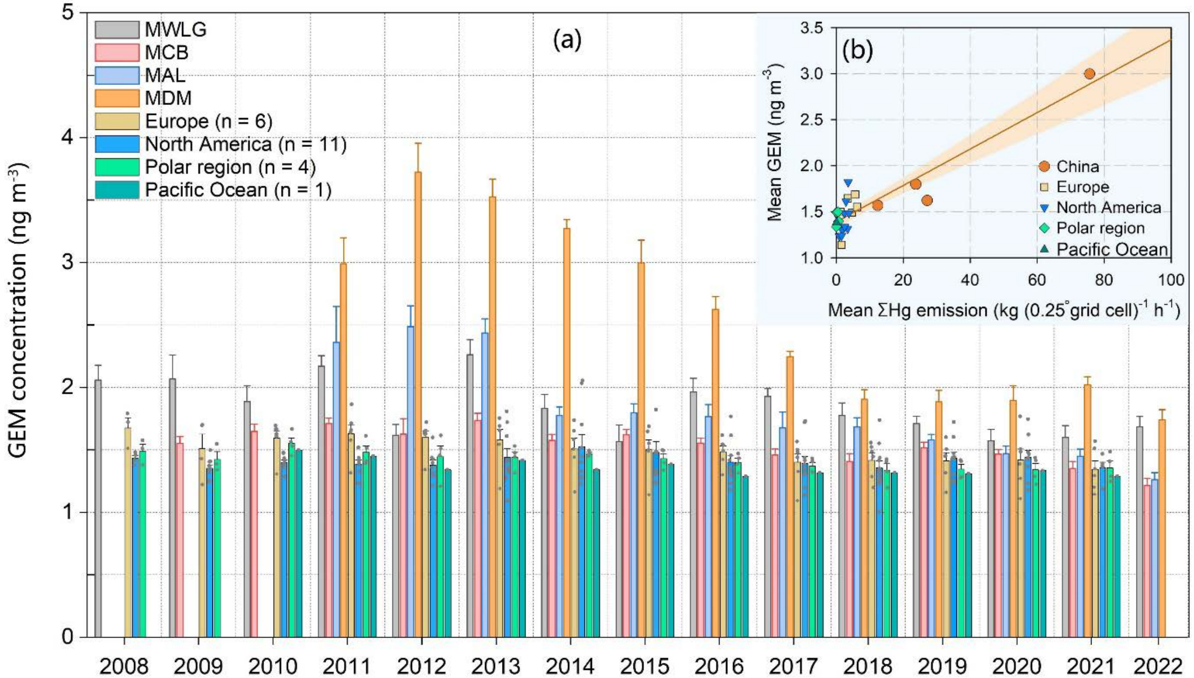
Mt. Damei
2011-

► **The sites are isolated from local anthropogenic emissions and receive air masses that passed over nearly all the geographical regions of China**

► **Monitoring data at these sites could be used to study the trends in atmospheric GEM concentrations and inform the changes of anthropogenic Hg emissions in China**

Mean levels during the whole study period

Yearly mean GEM concentrations in China, Europe, North America, Pacific Ocean and Polar region



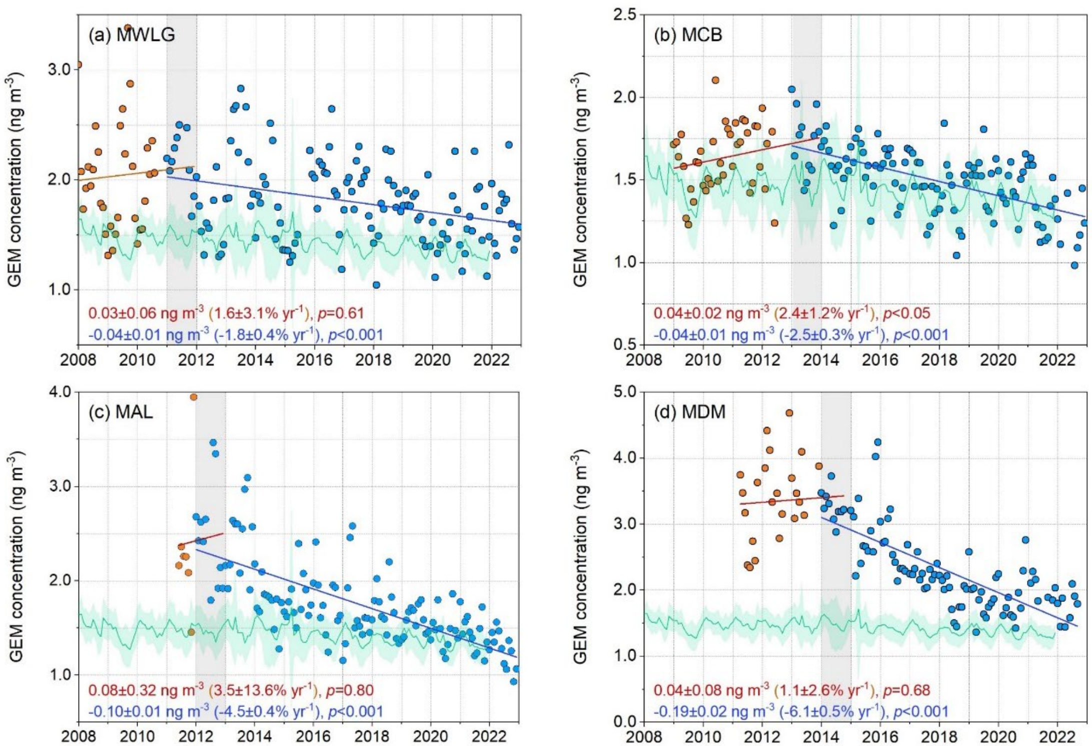
- ▶ **Mean GEM concentrations varied spatially in China**
- ▶ **GEM was highest at MDM in eastern China; and was lowest at MCB, where had low regional Hg emissions and strong vegetation uptake**
- ▶ **Mean GEM in China was 1.4 times higher than elsewhere in the Northern Hemisphere, and 1.9 times higher than the Southern Hemisphere**

Site	GEM
MWLG	1.85 ± 0.23
MCB	1.53 ± 0.14
MAL	1.81 ± 0.40
MDM	2.57 ± 0.71
Europe	1.49 ± 0.16
North America	1.42 ± 0.13
Pacific Ocean	1.36 ± 0.07
Polar region	1.42 ± 0.08
Cape Point	1.01 ± 0.05
Amsterdam Island	1.04 ± 0.02

Data are from this study, EMEP, AMNet, CAPMoN Networks and Slemr et al., 2020

Trend in GEM in China and other regions worldwide

Monthly mean and trends in GEM concentrations in China and other regions in the Northern Hemisphere

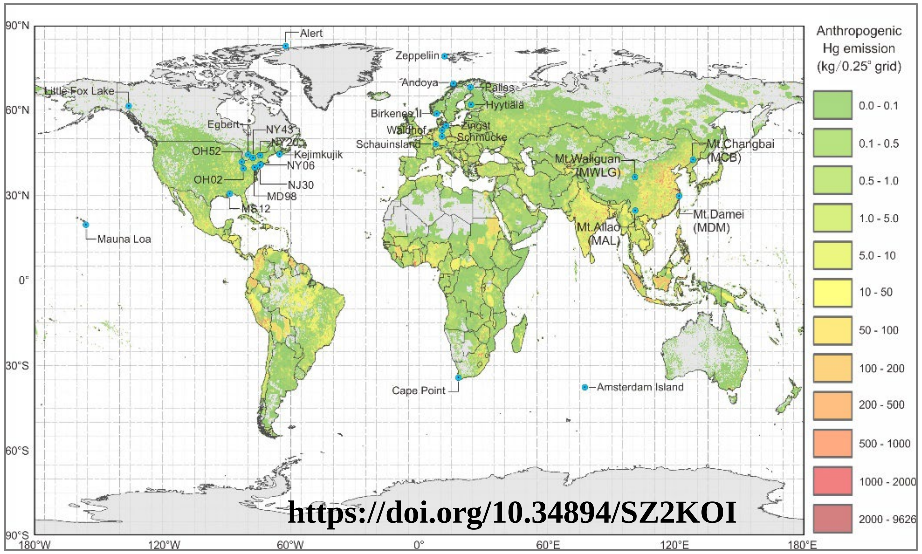


- ► **GEM concentrations at MWLG, MCB, MAL and MDM have been reversed from increase towards rapid declines in 2011, 2013, 2012, and 2014, respectively**
- ► **Recent decreasing rates ranged from 1.8 to 6.1% yr^{-1}. Highest decline was in eastern China, followed by southwestern, northeastern, and northwestern China.**
- ► **Recent declines in China were relative higher than elsewhere in the Northern Hemisphere (0.5-2.6% yr^{-1}) during the past two decades. GEM concentrations in the Southern Hemisphere were stable during the past decade**

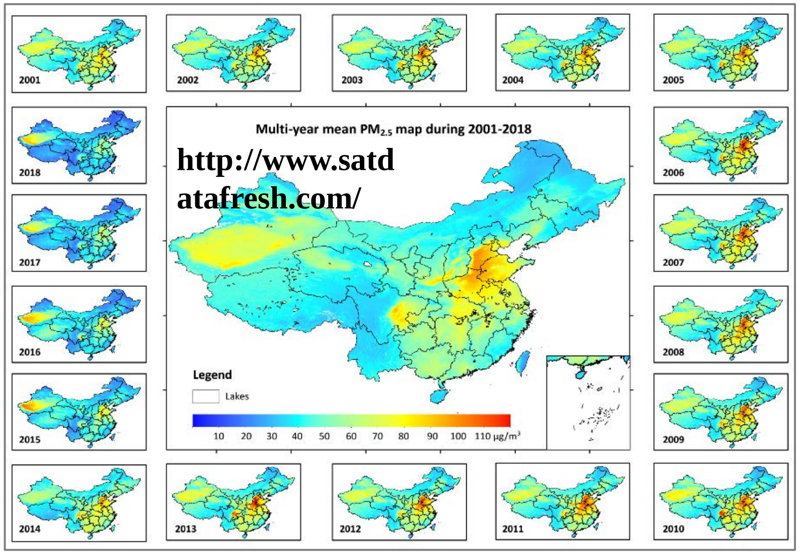
Zhou et al., 2017; Cole, et al., 2013; Custódio et al., 2022; Zhang et al., 2016; Slemr et al., 2011; Soerensen et al., 2012

Ancillary global and national parameters

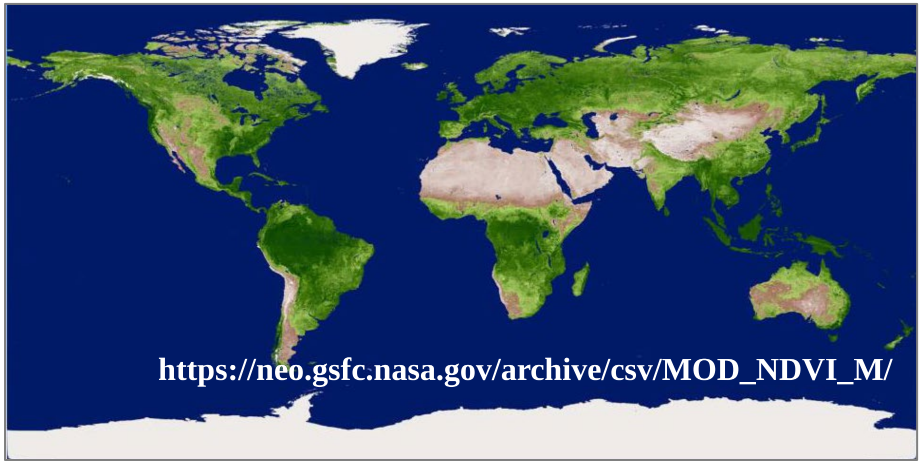
EMEP, AMNet, and CAPMoN GEM networks



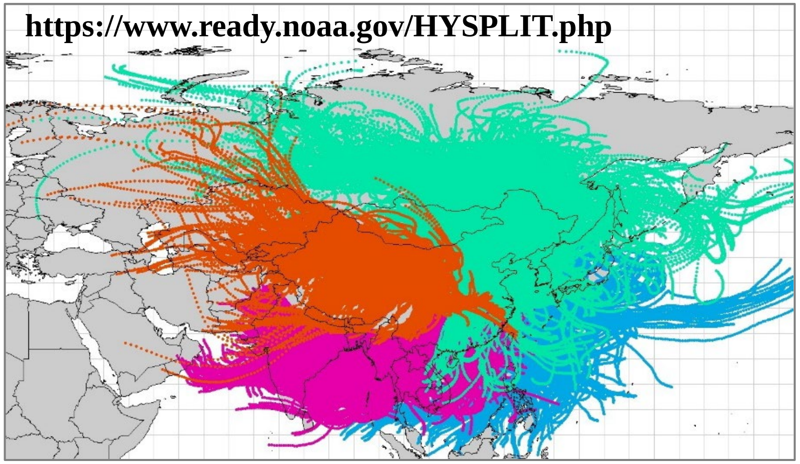
SDTE national criteria air pollutants



Global NDVI



NOAA hysplit backward trajectory



Source regions identification and effect of anthropogenic GEM emissions on GEM variations

► **Source regions identification: a multiple-site concentration-weighted trajectories (CWTs) receptor model**

$$CWT_{ij} = \frac{\sum_{l=1}^{L} C_l \tau_{ijl}}{\sum_{l=1}^{L} \tau_{ijl}} \quad (1)$$

where CWT_{ij} is the GEM CWT value in grid cell (i, j); C_l is the 4-h mean GEM concentration associated with trajectory l; τ_{ijl} is the number of trajectory segment endpoints in grid cell (i, j) for backward trajectory l; and L is the total number of trajectories calculated over the study period.

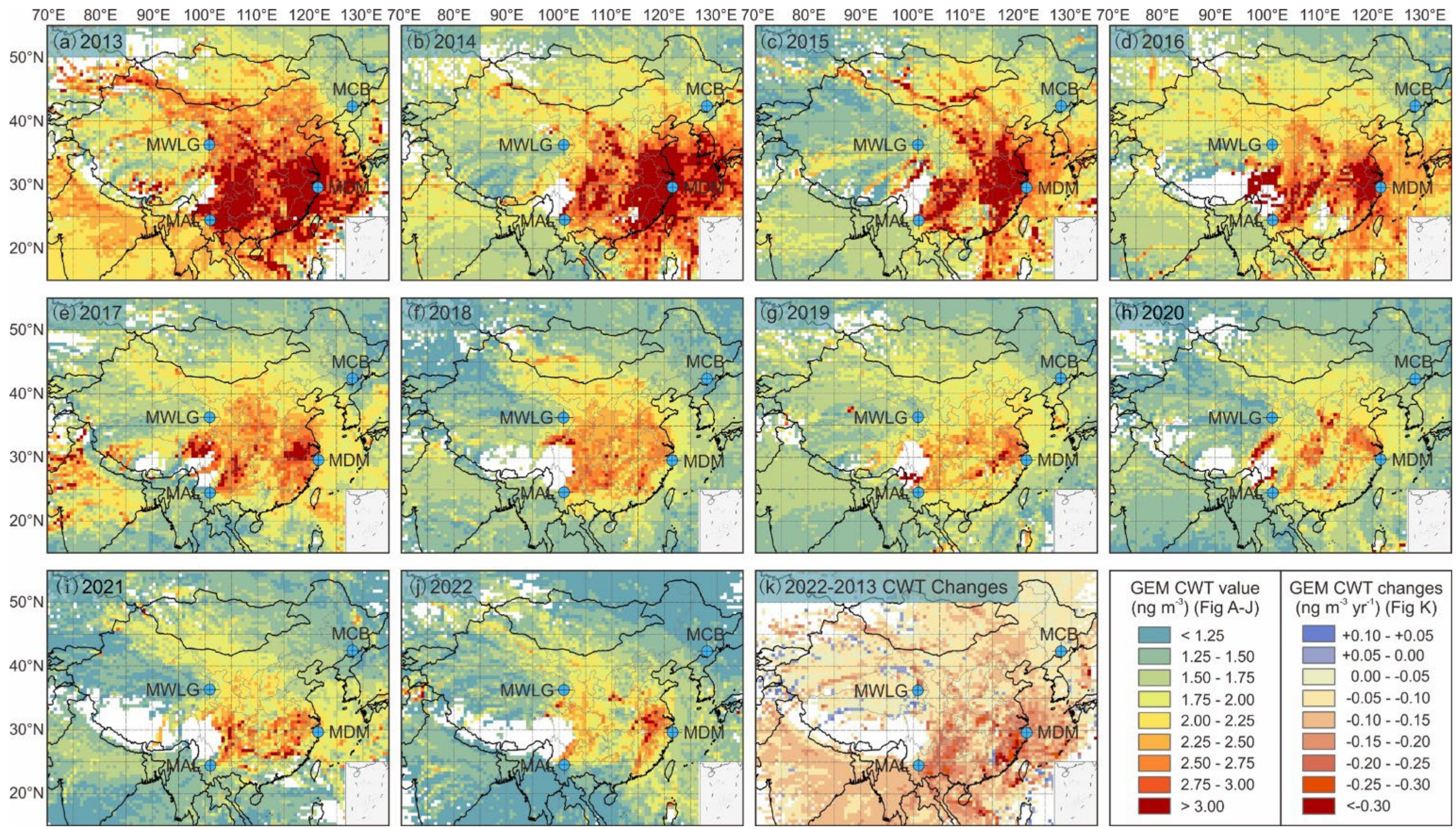
► **ΣGEM emissions: The prior exposure of air masses to anthropogenic GEM emissions based on backward trajectory to explore the links between GEM concentrations and anthropogenic emissions**

$$\sum \text{GEM emissions} = \sum_{t=0}^{120} (\tau_{ij} \times \text{Emis}_{ij})/121 \quad (2)$$

where τ_{ij} and Emis_{ij} are the number of segment endpoints and anthropogenic emission (kg) in grid cell (i, j); 121 is the total segment endpoints for each backward trajectory.

Source regions of GEM in China

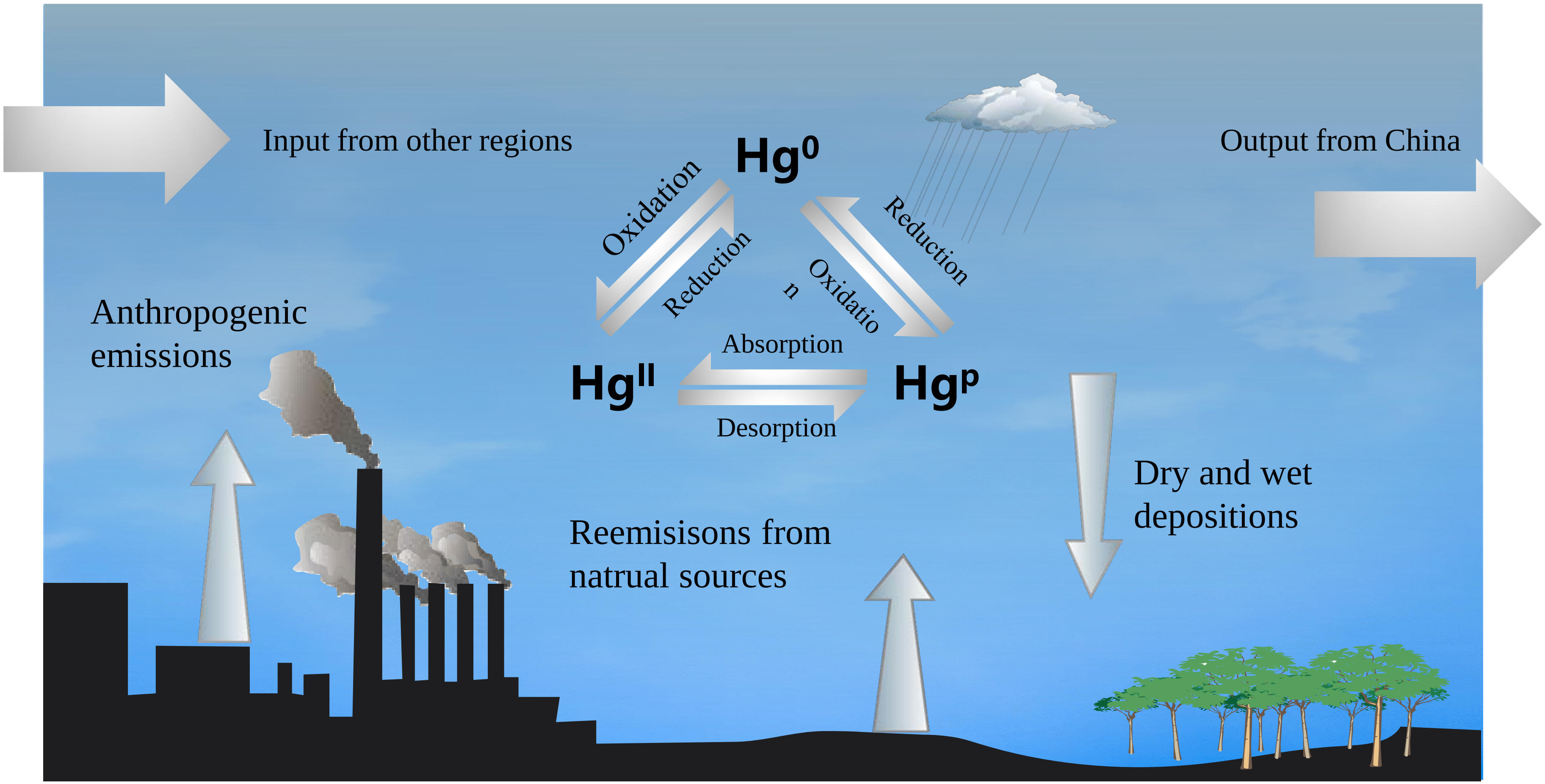
Source regions of GEM at Chinese remote sites identified using Concentration-Weighted Trajectory (CWT) model



► **GEM source regions were consistently located in eastern and southwestern China, matching well with the distributions of anthropogenic GEM emissions**

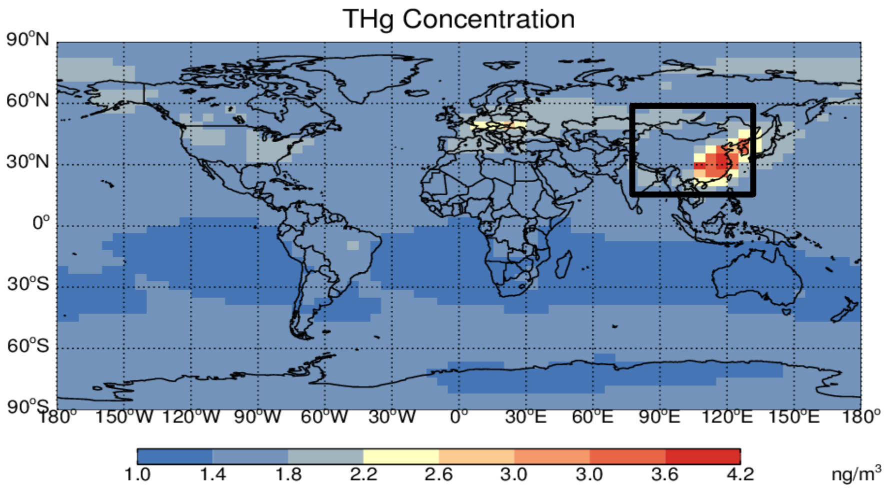
► **Similarly, source intensity decreased fast in eastern and southwestern China from 2013 to 2022, indictive of efficient reductions of anthropogenic GEM emission in these regions**

Atmospheric Hg Mass Balance in China



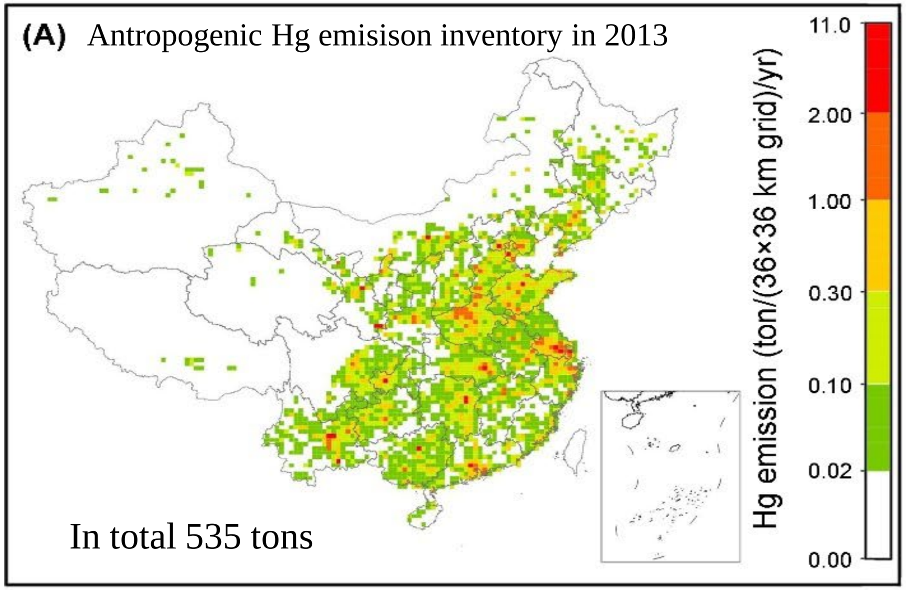
CMAQ model was used to estimate atmospheric Hg mass balance in China

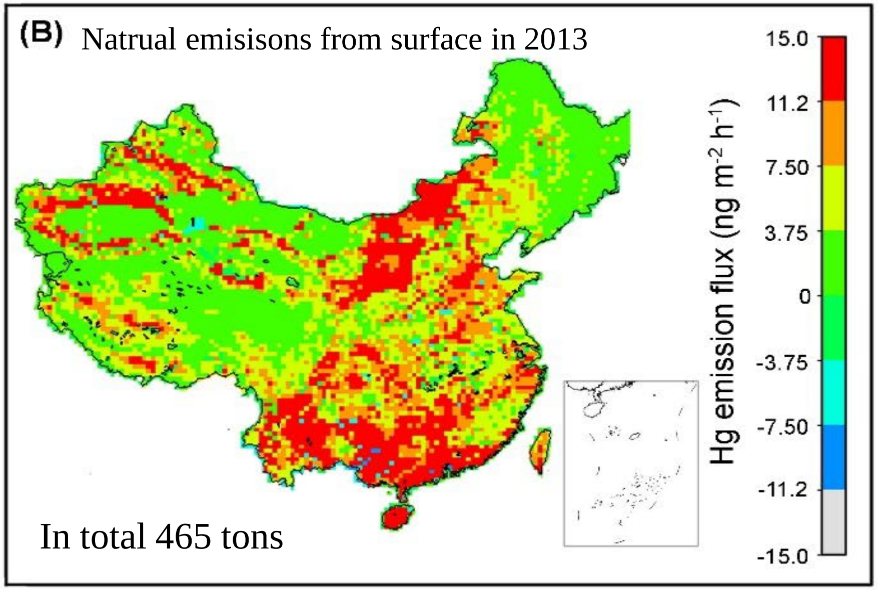
Regional CMAQ Model



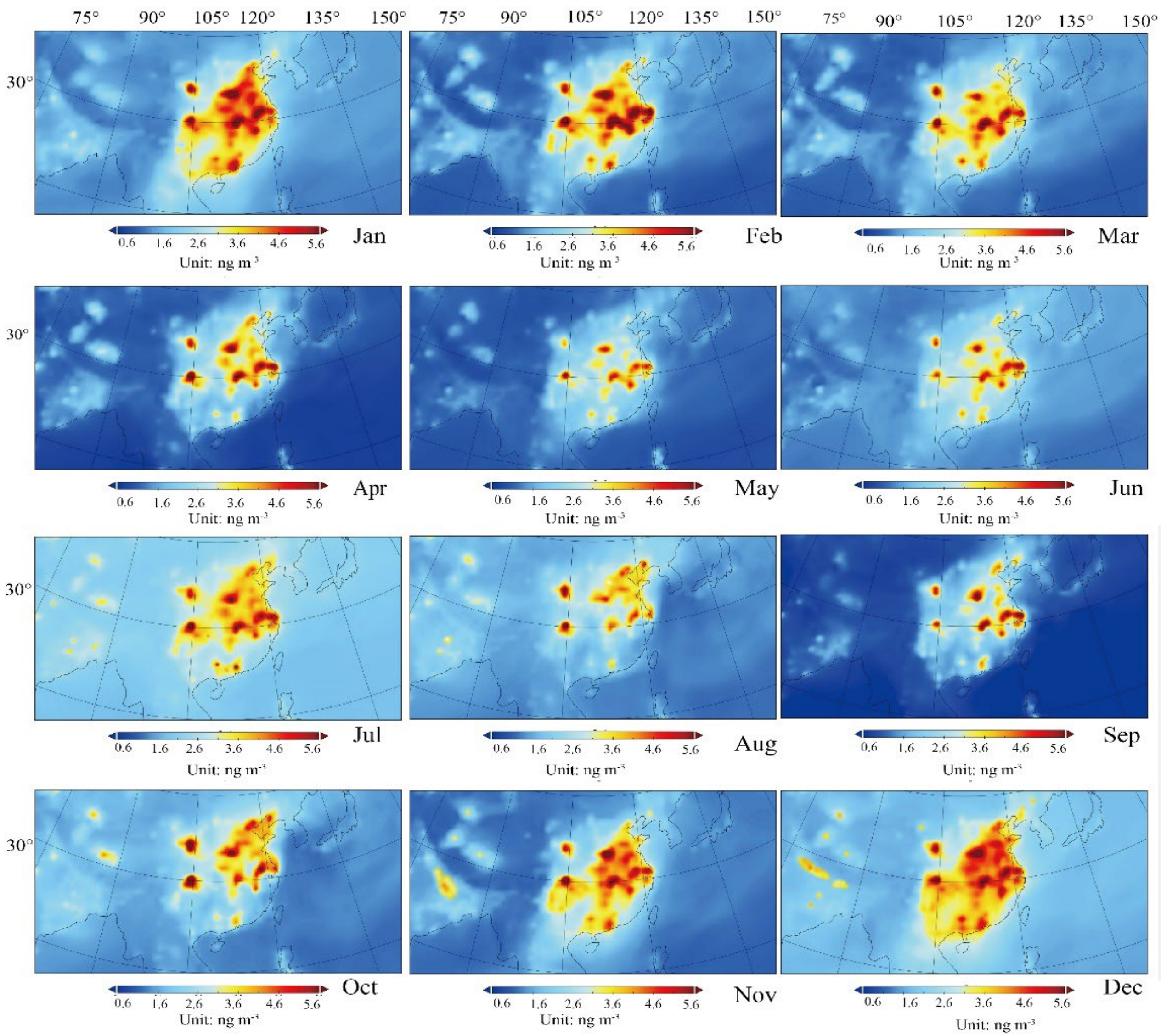
Global TGM concentrations in 2010（4° × 5°）

- Running GEOS-Chem model, we obtained the boundary and background values of atmopseric Hg concnetrations in China and East Asia
- By inputing the anthrpogenic emission inventory and natrual reemission estimates to CMAQ regional model, we calaculated the atmospheric mercury mass balance in China in 2013





Modeled spatial distribution of GEM concentrations in air

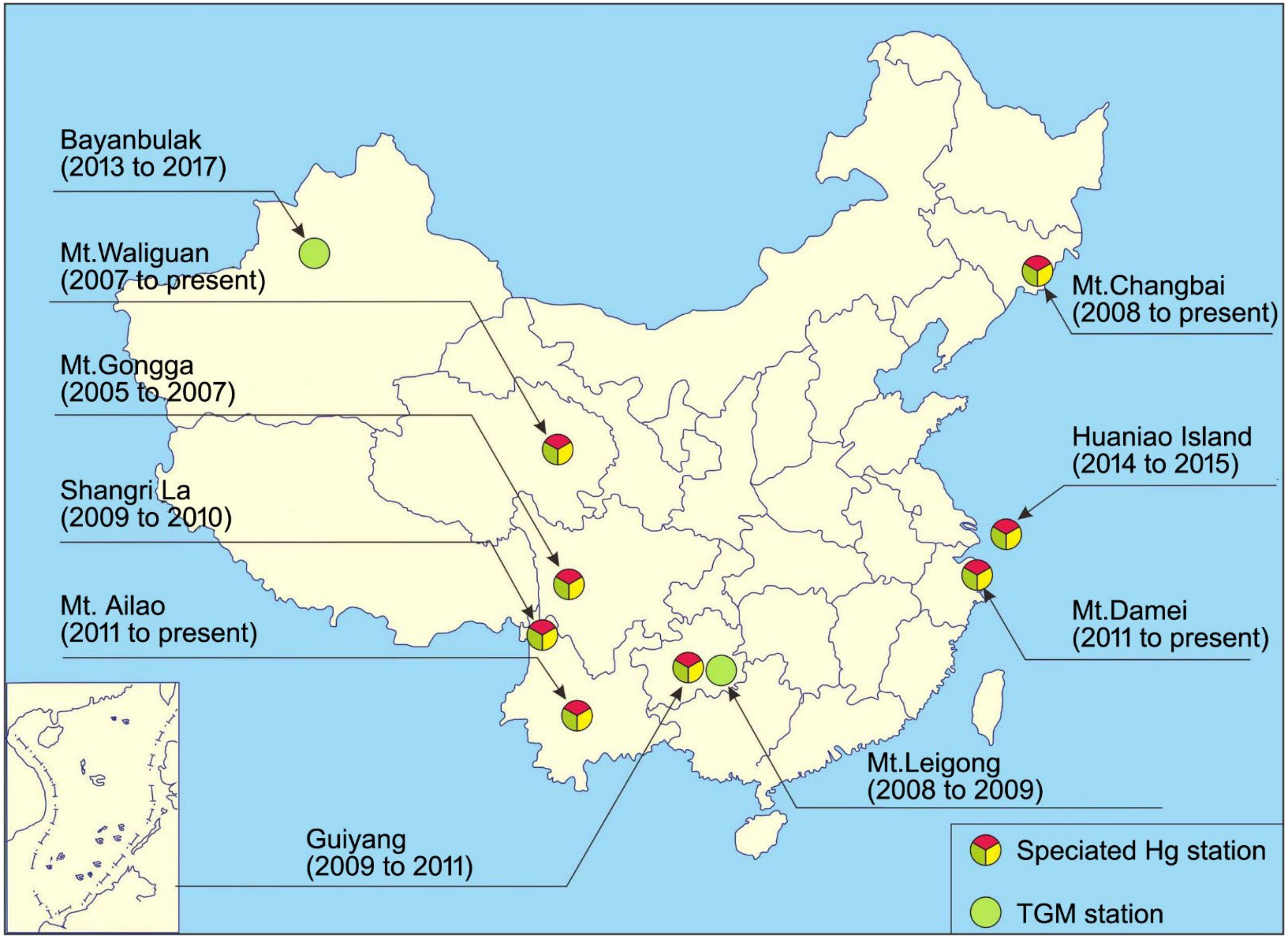


Wang et al., JGR-atmos （2018）

The annual average GEM concnetration is 2.9 ng m^{-3}

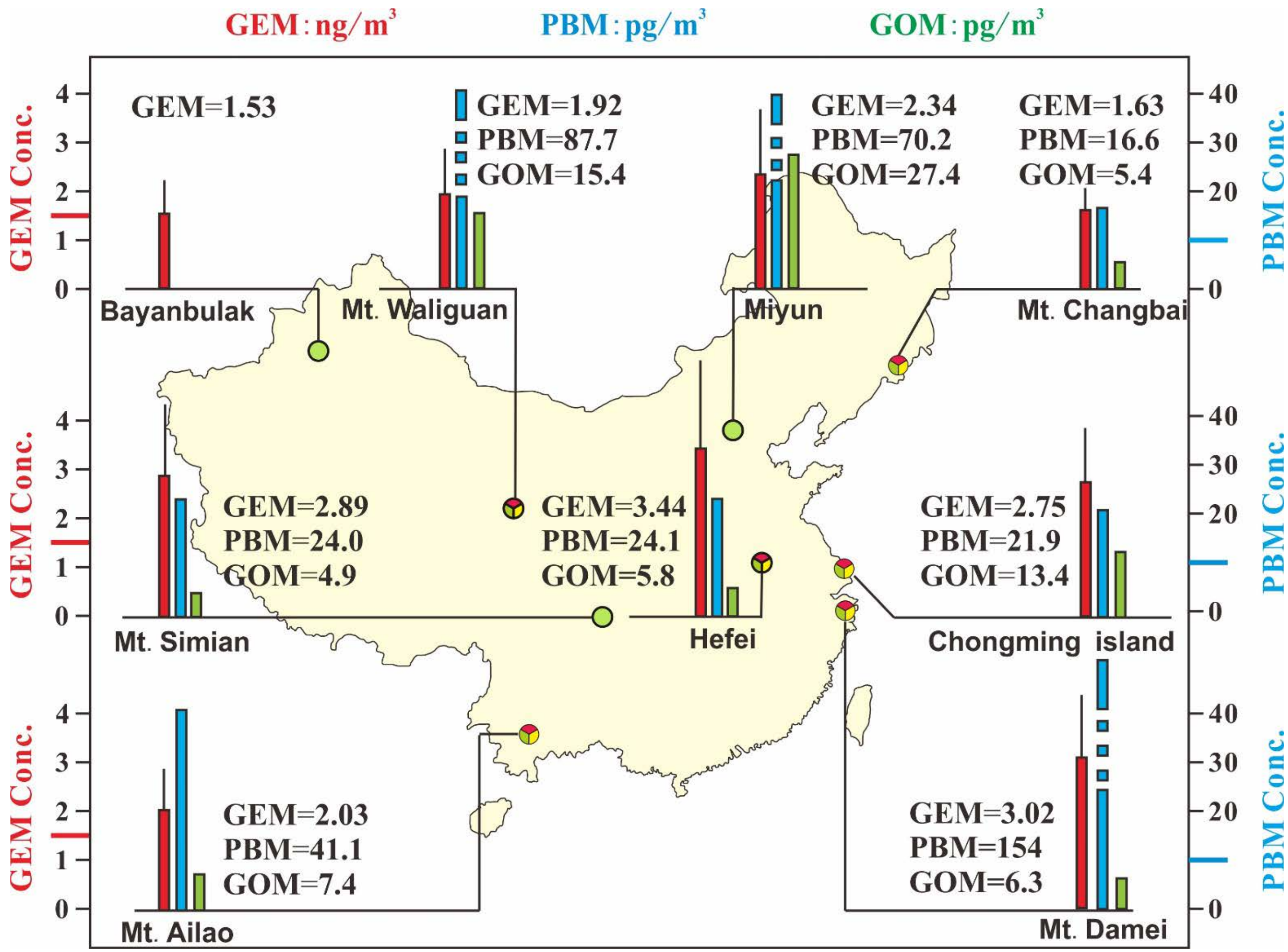
GEM concentrations were controlled by emission sources, monsoon circulation and haze pollution

Atmospheric observation network in China



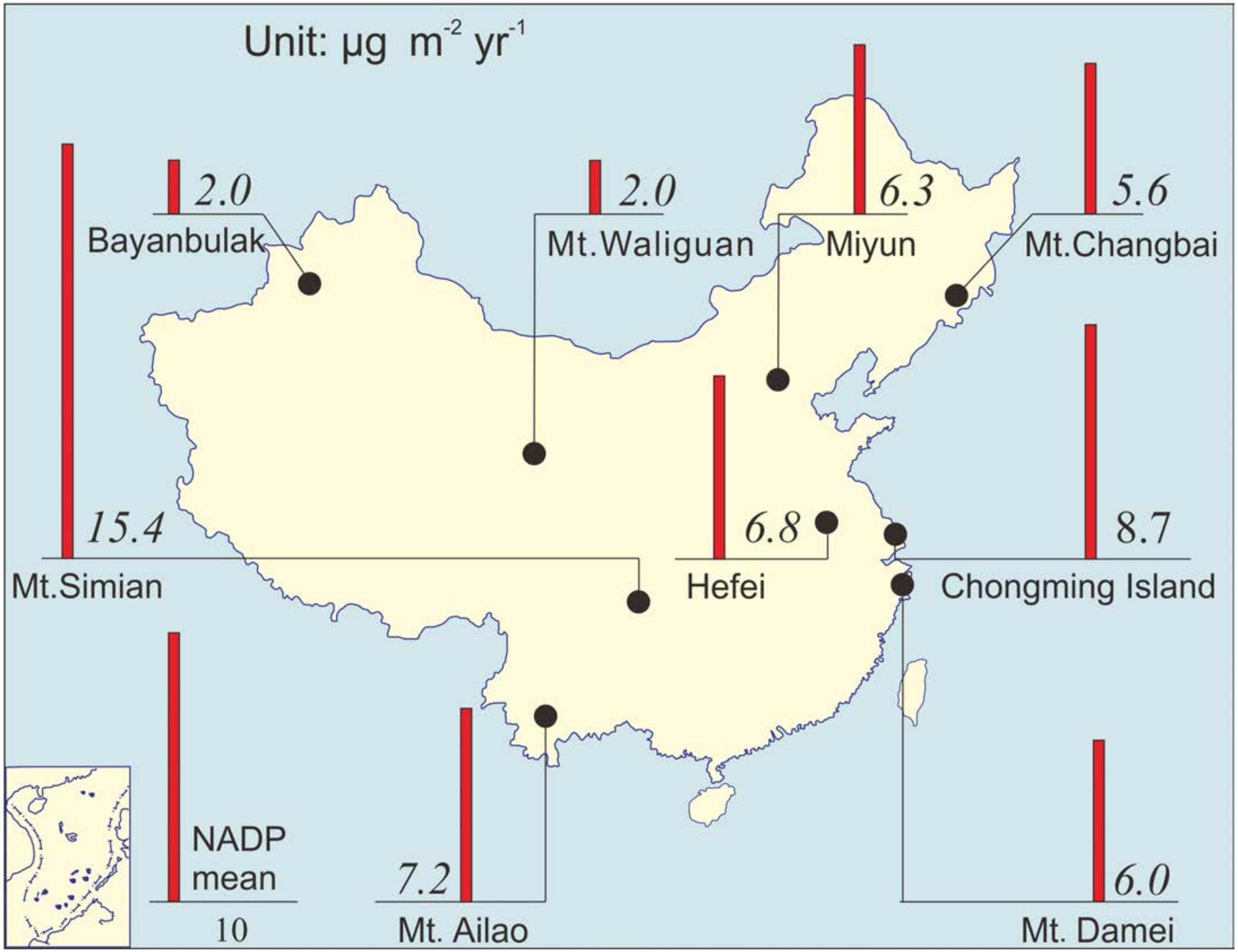
Fu et al., Atmos. Chem. Phys., 2010, 2012a,b, 2015a,b
Fu et al., Atmos. Environ., 2008, 2011; Fu et al., Sci. Total Environ., 2009
Fu et al., Environ. Pollut., 2010; Fu et al., Appl. Geochem., 2008

Concnetrations of different mercury speices in air in China



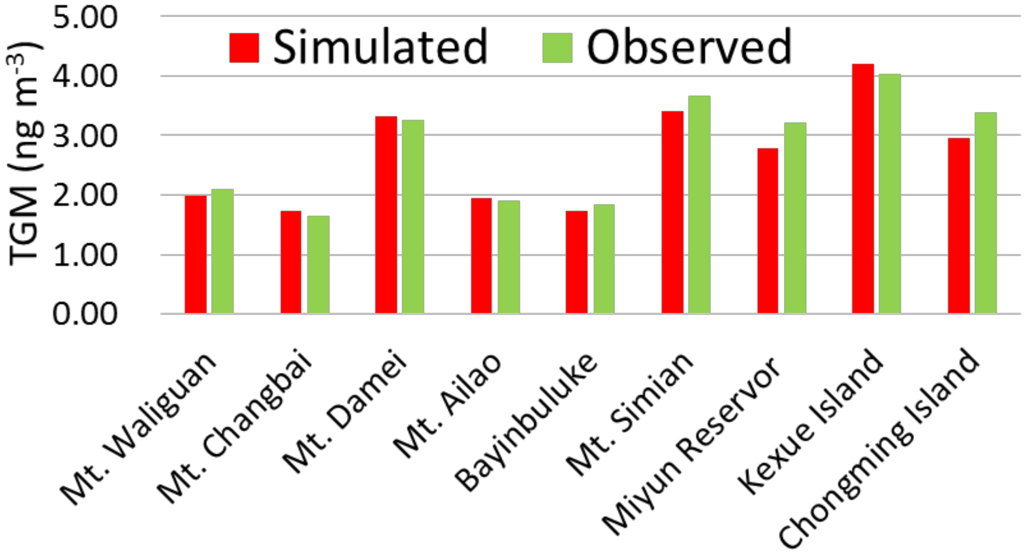
Fu et al., ACP, 2015a
Yu et al., JGR, 2015
Hong et al., ACP, 2016
Li et al., Environ. Sci. 2016, (in Chinese)

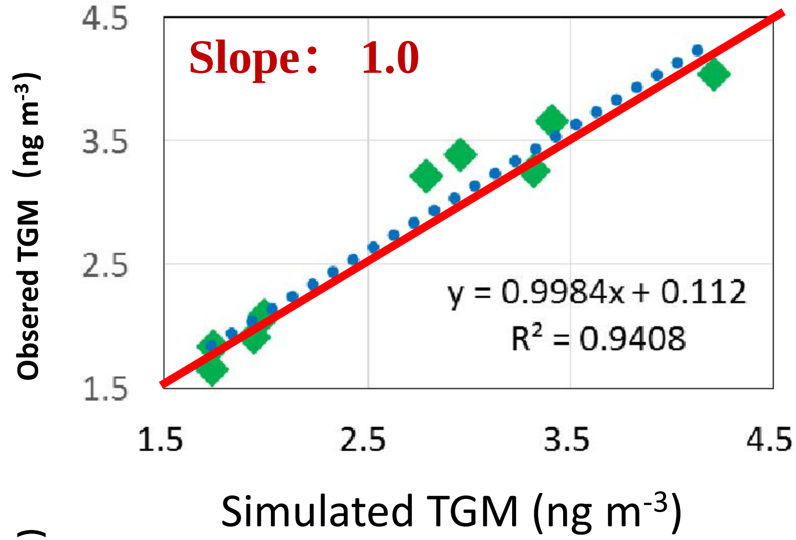
Mercury wet deposition fluxes

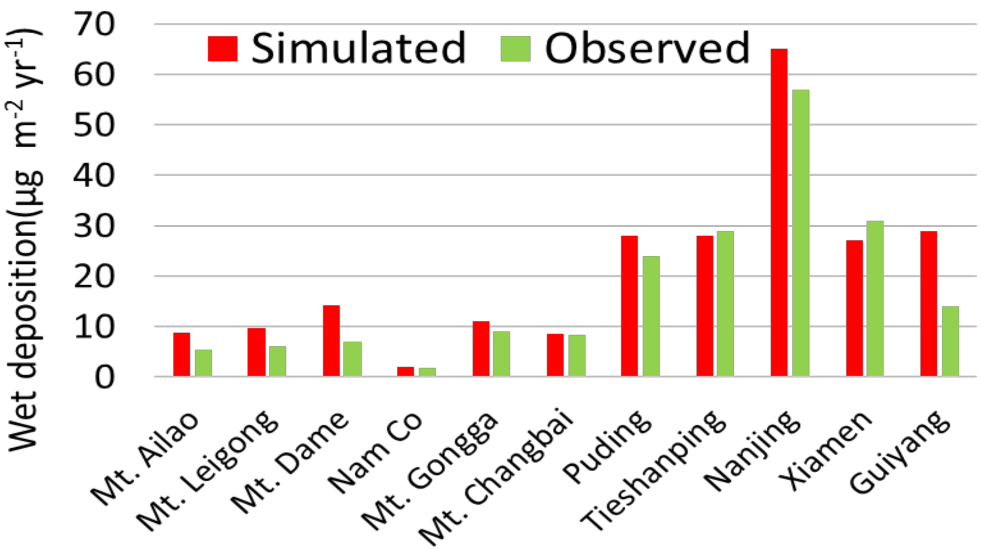


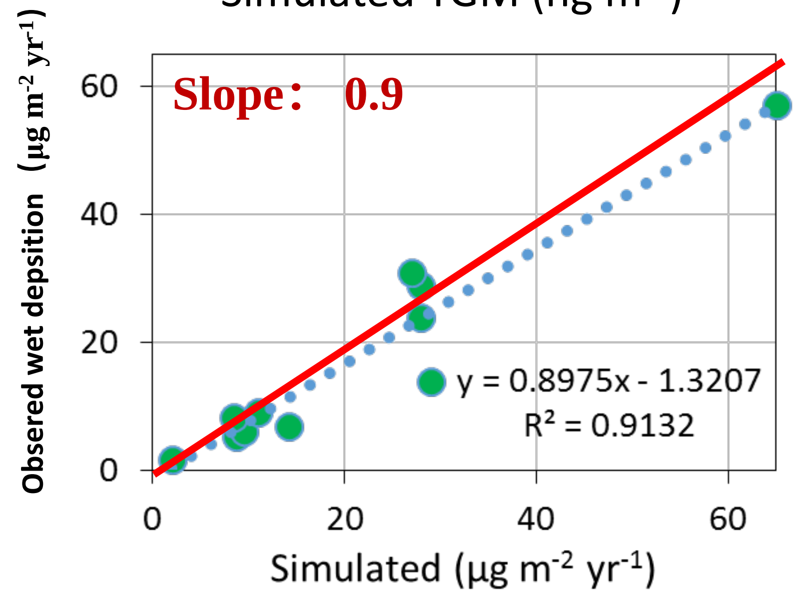
Fu et al., ACP, 2016a
Ma et al., ACP, 2016

Comparison between simulated and observed values

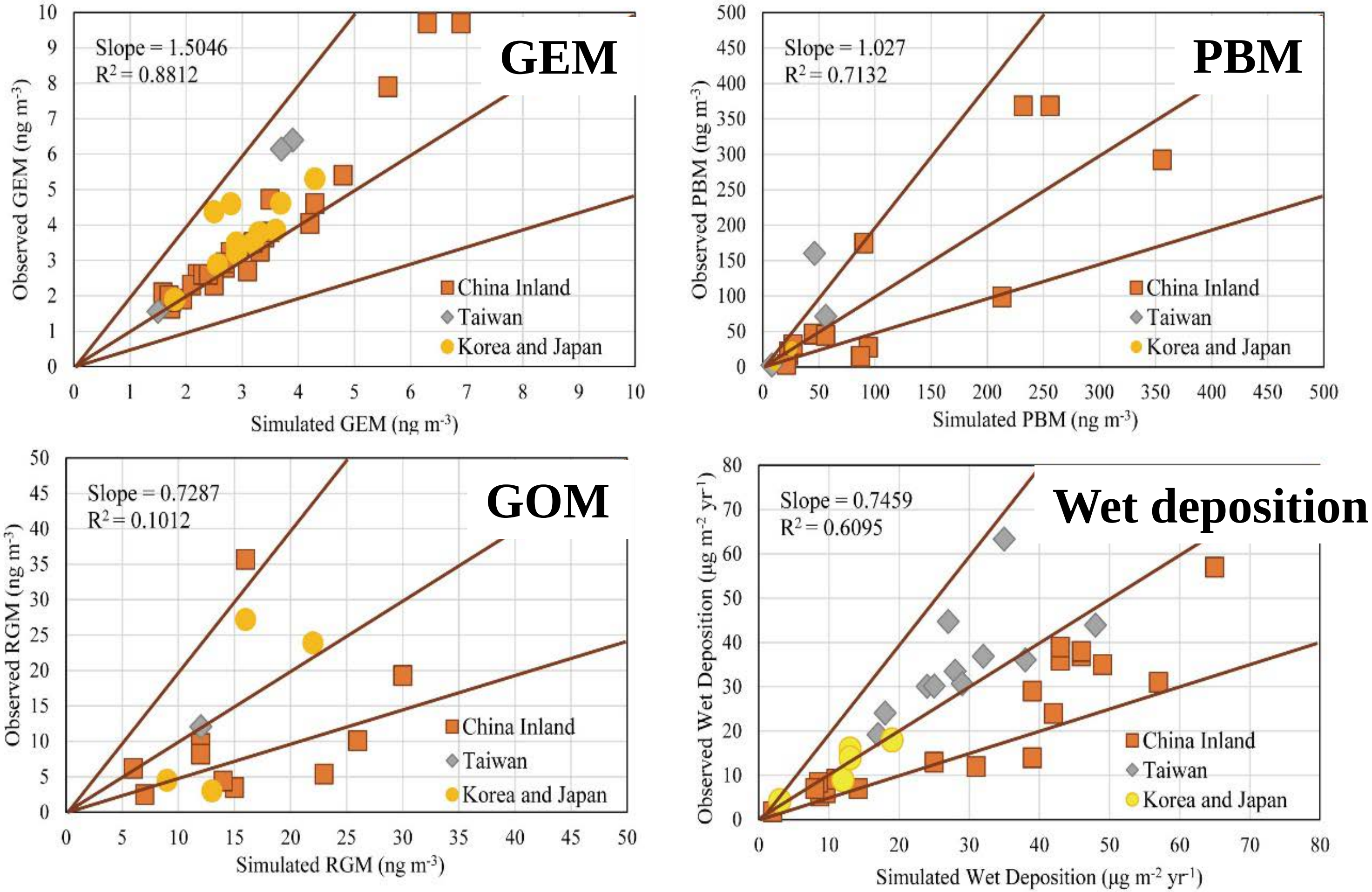








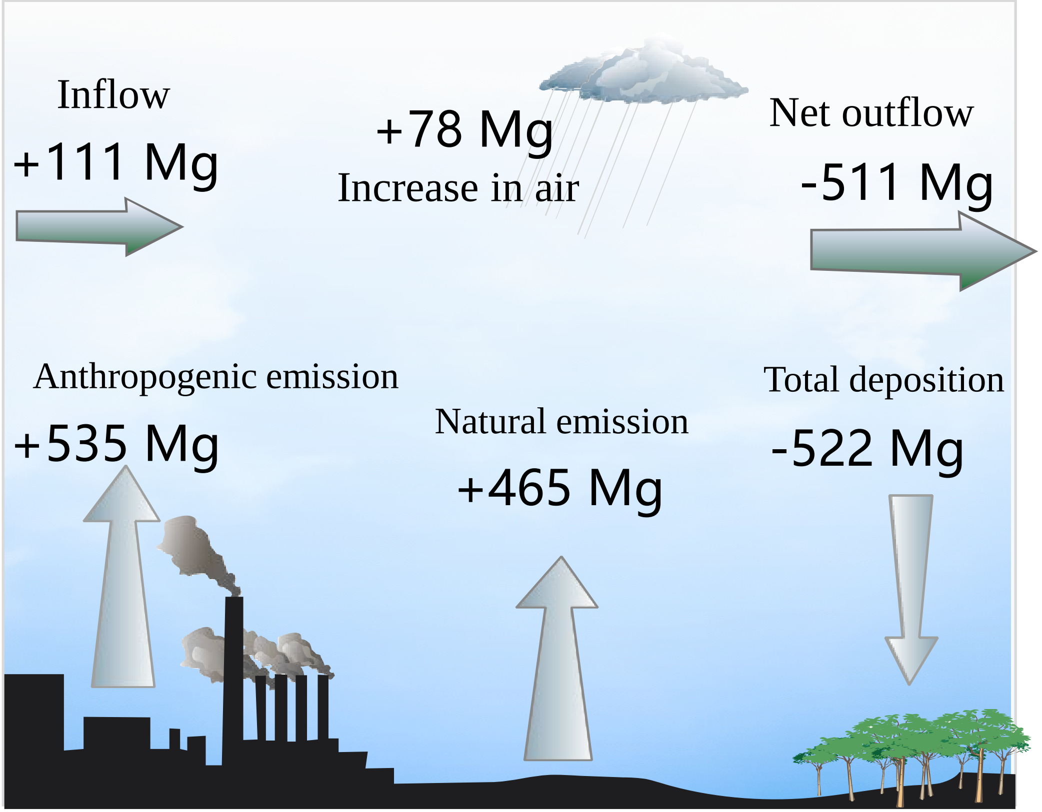
Obsered data vs simulated data in Asia



Slopes： GEM=1.5； PBM=1.0； GOM=0.73； Wet deposition =0.74

Atmospheric Hg mass balance in 2013 in China

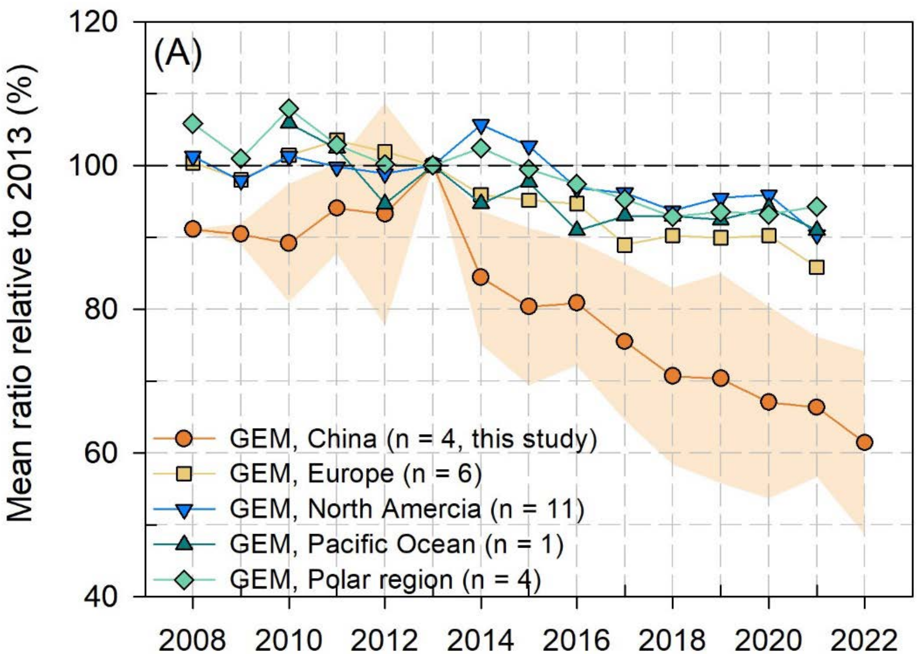
- The net outflow from China was **511 Mg**
- **65%** from re-emission from natural surfaces
- **70%** of anthropogenic emissions deposited within domain
- 522 Mg of Hg was deposited in China



Wang et al., JGR-Atmos, 2015, 120, 9758-9771; Wang et al., JGR-Atmos, 2018, 123: 9868-9890

Comparison of GEM trends in the Northern Hemisphere

GEM trends in northern hemisphere regions



GEM declines since 2013

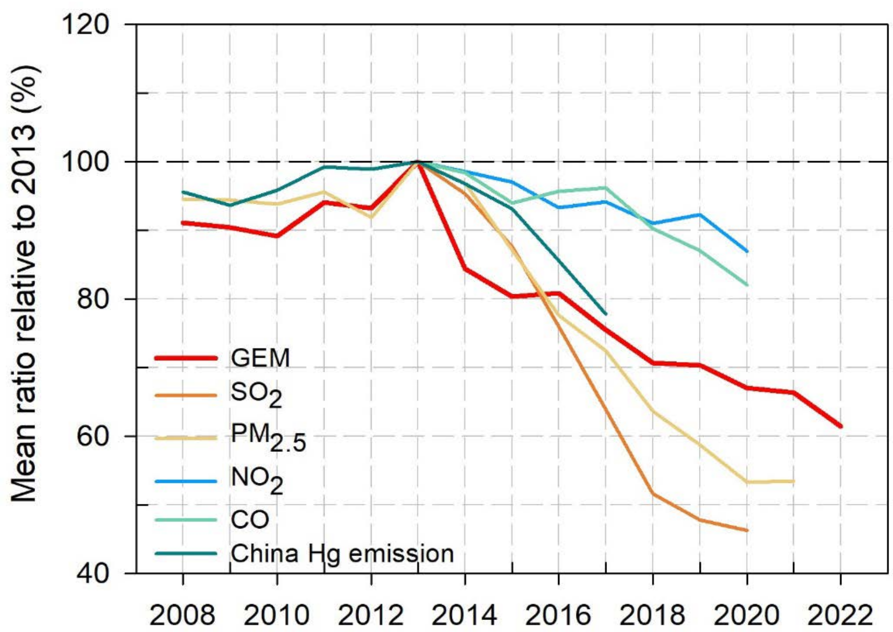
Regions	Declines
China	38.6
Europe	14.2
North America	9.6
Polar region	5.7
Pacific Ocean	9.0

- ► **From 2008 to 2013, GEM concentrations in China increased on average by 8.9%, whereas they were constant in Europe, North America, Pacific Ocean and Polar regions**
- ► **Observations in the Northern Hemisphere showed a widely-distributed declines over the past decade**
- ► **Since 2013, GEM in China on average decreased by 38.6%, approximately 4 times of the means in other regions in the Northern Hemisphere**

Data are from this study, EMEP, AMNet, CAPMoN Networks

Links between the GEM and air pollutants trend in China

Trends in GEM and air pollutants in China



Declines since 2013

Pollutants	Declines
GEM	38.6
SO_2 - to 2020	53.7
$PM_{2.5}$ - to 2021	46.6
NO_2 - to 2020	13.1
CO - to 2020	18.0

► **GEM decline in China matched well with the trends in air pollutants and reduced anthropogenic Hg emissions**

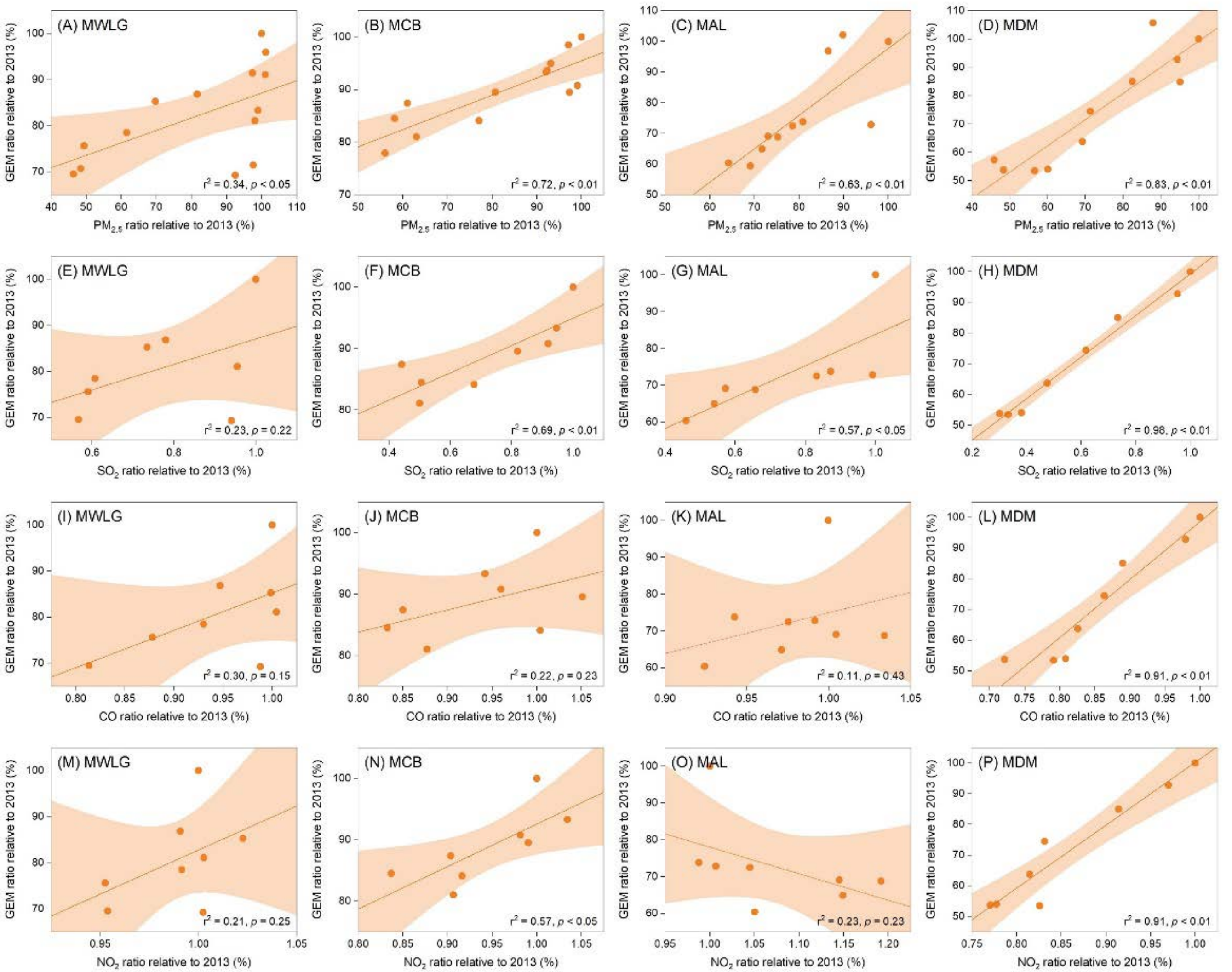
► **PM2.5 and SO2 concentrations decreased by 53.7 and 46.6% since 2013, respectively, higher the GEM declines**

► **The declines of NO2 and CO concentrations were 13.1 and 18.0%, respectively, lower than the GEM declines in China**

Data are from this study, SDTE, and Liu et al., 2019

Anthropogenic drivers of GEM trends in China

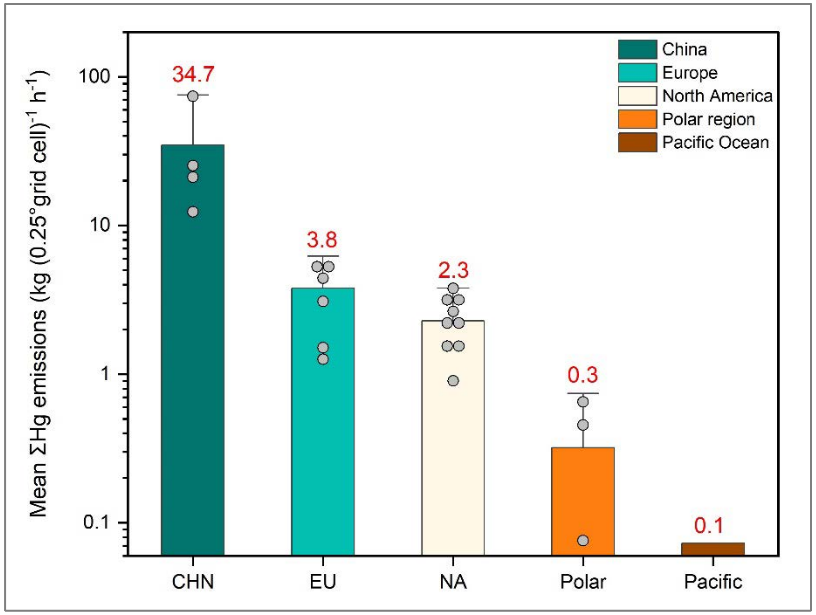
Relationship between GEM and air pollutants trends



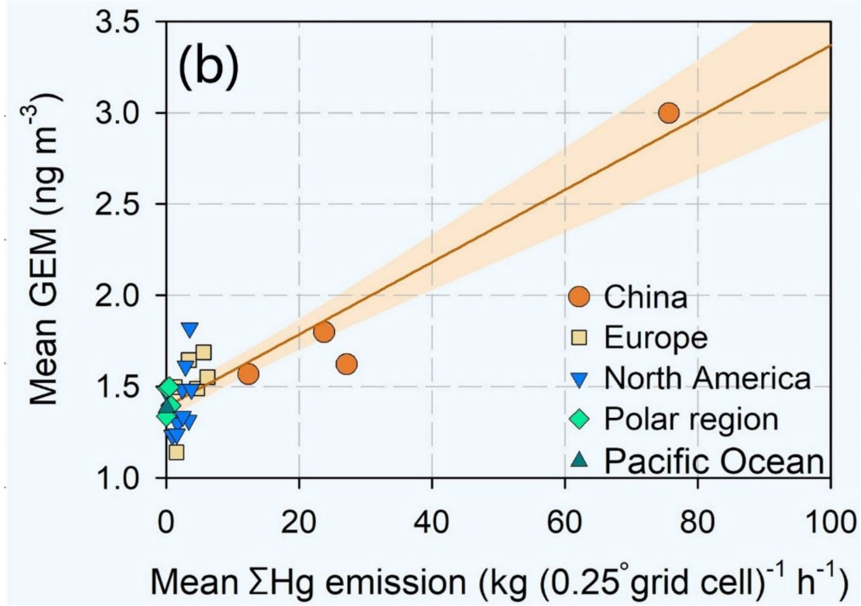
- ► **GEM in China is closely correlated with $PM_{2.5}$ and SO_2, levels.**
- ► **Emission control measures for $PM_{2.5}$ and SO_2 led to the reduction of anthropogenic GEM emissions.**
- ► **GEM declines in China accordingly.**

Anthropogenic drivers of the spatial GEM distribution

Regional cumulative anthropogenic Hg emissions in the Northern Hemisphere



Cumulative anthropogenic Hg emissions versus GEM concentrations



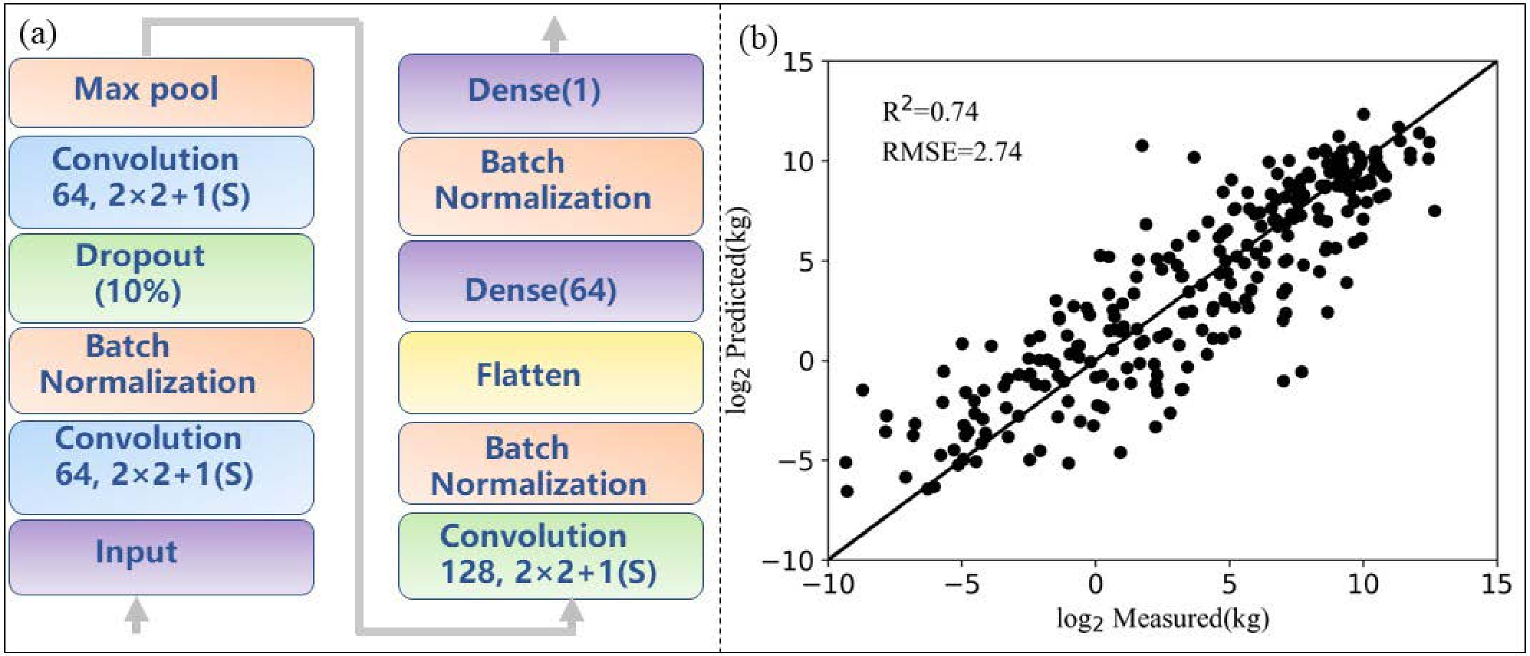
►**Rural sites in China were generally exposed to more anthropogenic Hg emissions (more than ten times) than elsewhere in the Northern Hemisphere, which is the major driver of the spatial distributions in the GEM concentrations in the Northern Hemisphere**

Data are from this study, EMEP, AMNet, CAPMoN Networks and Slemr et al., 2020

Estimate anthropogenic GEM emissions-CNN model

► **A machine learning (i.e., the Convolutional Neural Network model; CNN) approach is used to estimate the anthropogenic GEM emissions in China during 2013-2022, which considered the contributions of anthropogenic and natural emissions as well as the changes of vegetation activity and atmospheric background (e.q. (3))**

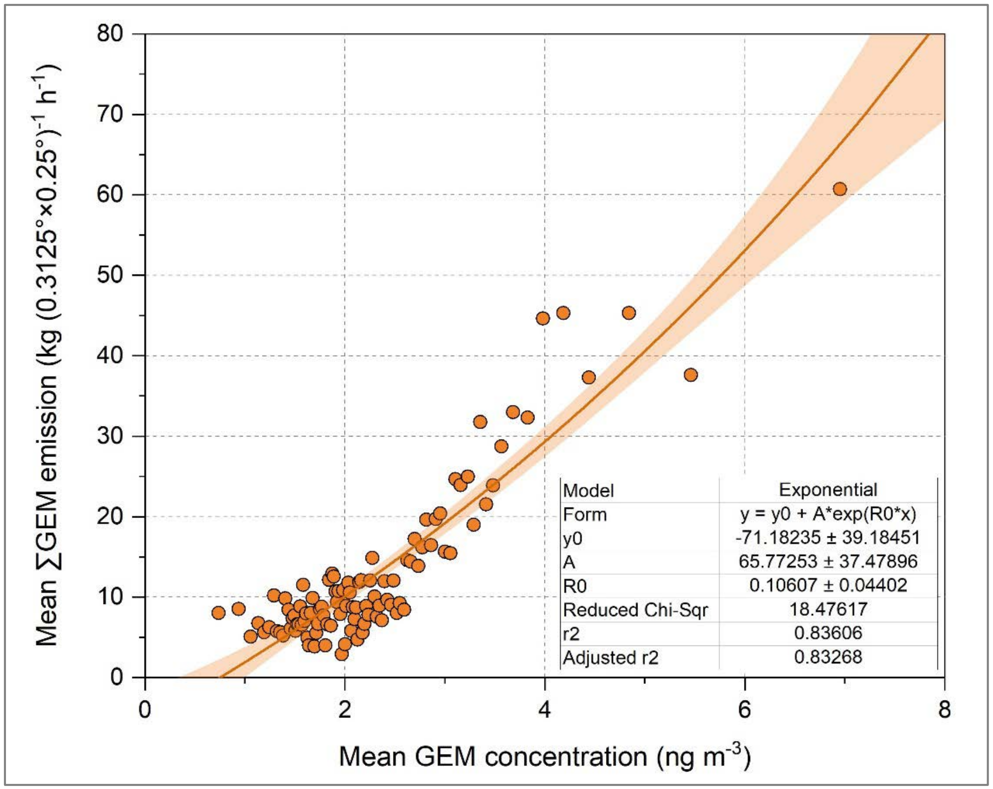
$$F^{ant} = \text{function}\ (y^{GEM}, y^{nat}, y^{bac}) \quad (3)$$



Machine learning model setup. (a) is the flowchart of CNN, and (b) evaluation of CNN modeling result in pretrained 40,000 models.

Estimate anthropogenic GEM emissions-Empirical model

Exponential relationship between grouped mean GEM concentrations and corresponding mean ∑GEM emission in China in 2013



Empirical relationship model:

► **Observational principle: ∑GEM emissions in China were significantly correlated the observed GEM concentrations in 2013, which is in the form of:**

$y = -71.182 + 65.772 * e^{0.106*x}$ ($r^2 = 0.84$, $p < 0.001$)

► **Calclulations:**

$$\Sigma \text{GEM emissions}_{G_i} = 65.772 \times e^{0.106 \times GEM_{obs-G_i}} - 71.182$$

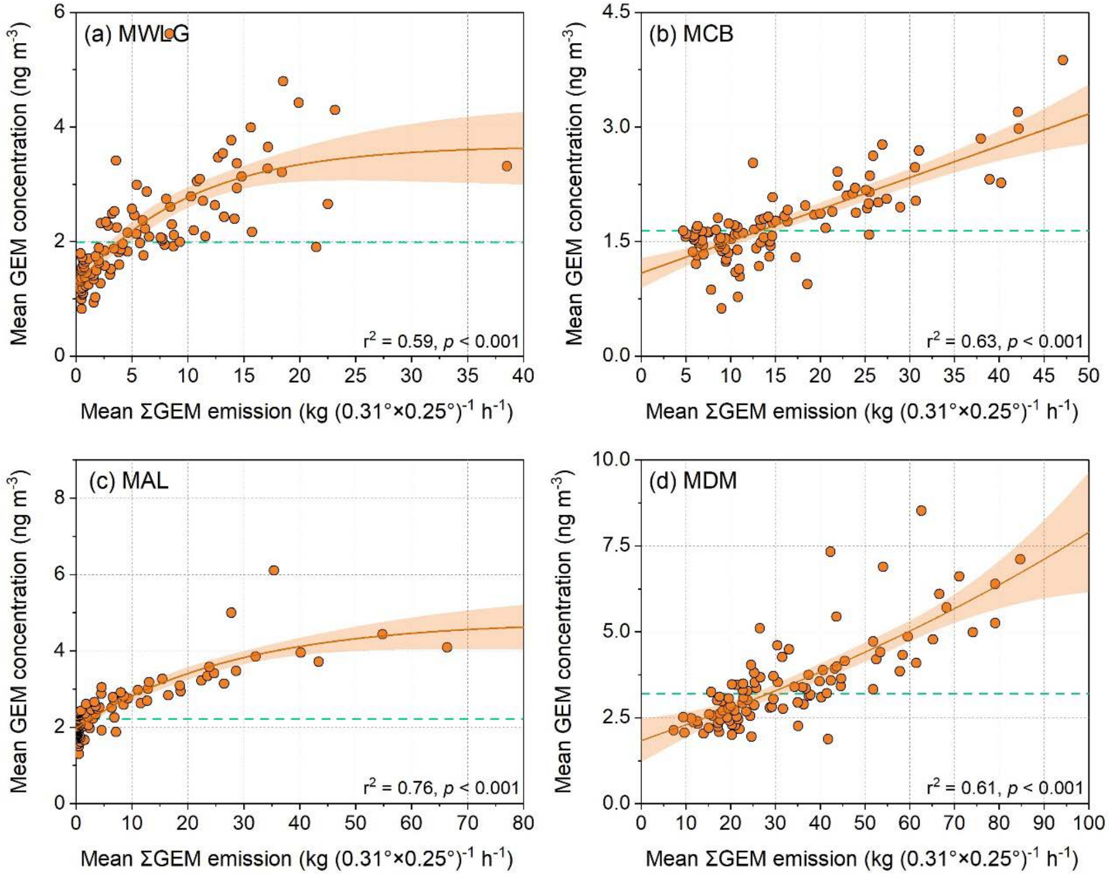
where $\sum \text{GEM emission}_{\mathrm{G_i}}$ is the mean exposure of air masses to Chinese anthropogenic GEM emissions for the mean GEM concentration within group G_i ($\text{GEM}_{\text{Obs}-\mathrm{G_i}}$) in year *yyyy*.

► **Emission ratios relative to 2013:**

$$\text{Ratio}_{yyyy} = \Sigma_1^{100}\left[\left(\sum \text{GEM emission}_{G_i} / \sum \text{GEM emission}_{\mathrm{G_i}-2013}\right) \times \left(\sum \text{GEM emission}_{\mathrm{G_i}-2013} / \Sigma_1^{100} \sum \text{GEM emission}_{\mathrm{G_i}-2013}\right)\right]$$

Anthropogenic drivers of GEM trends in China

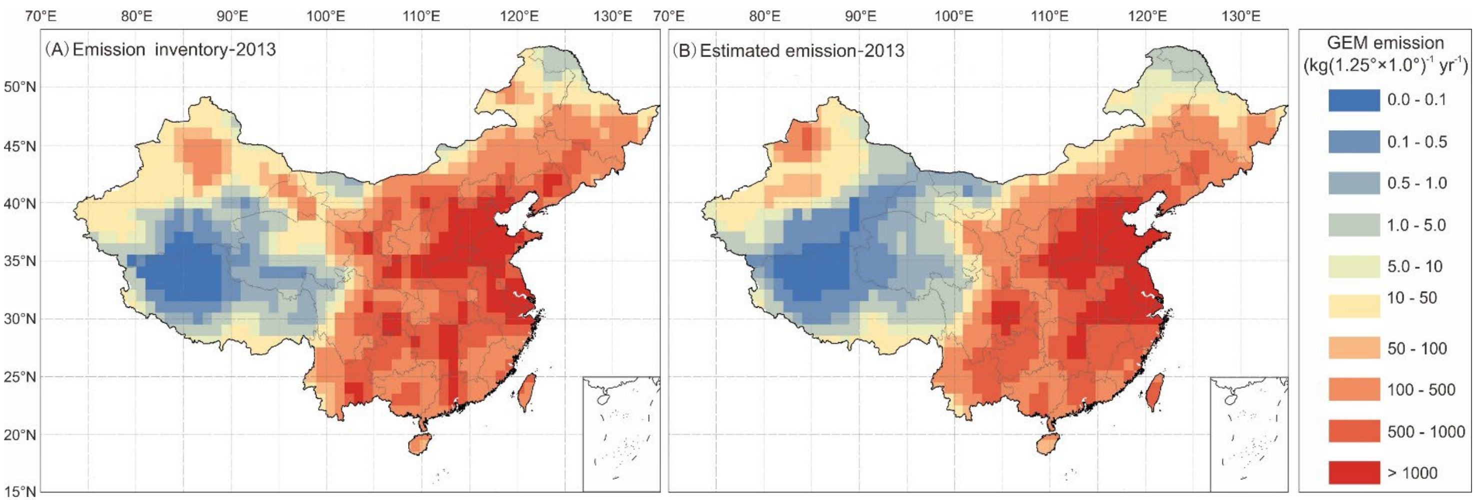
Relationship between GEM concentrations and anthropogenic GEM emissions in 2013



►**GEM concentrations in 2013 were positively correlated with ΣGEM emissions in China**

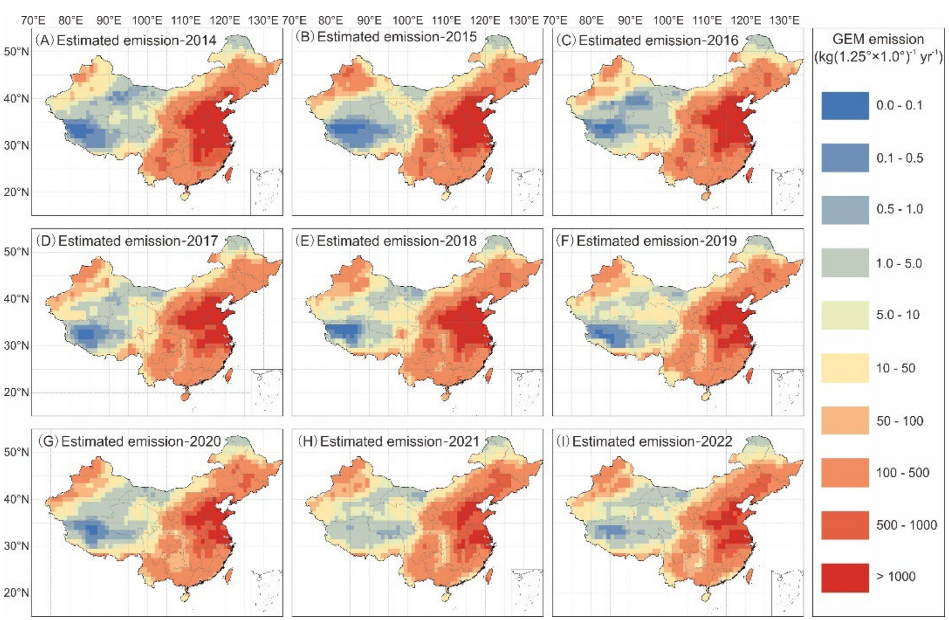
►**Variations in GEM concentrations in China were controlled by the national anthropogenic emissions**

Estimated anthropogenic GEM emissions during 2013-2022



CNN machine leaning well reproduced gridded anthropogenic GEM emissions

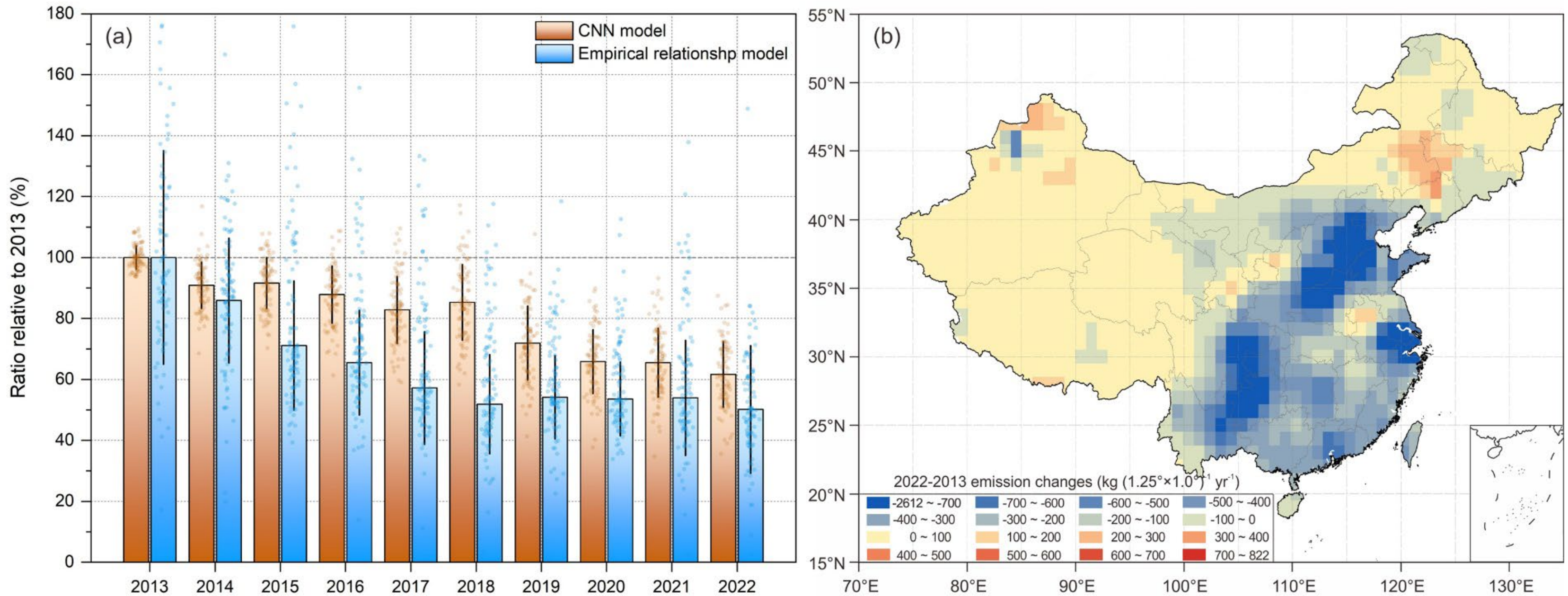
Estimated 2014-2022 anthropogenic GEM emissions



► **Major anthropogenic GEM emission regions in China were similar during 2013-2022, which were mostly located in the east domain of mainland China**

Trend in anthropogenic GEM emissions in China

Temporal and spatial changes in China's anthropogenic GEM emissions during the past decade



- ►**Chinese anthropogenic GEM emissions decreased by 38~50% since 2013, i.e., 152~187 tons since 2013**
- ►**The decreasing rates in Chinese anthropogenic GEM emissions since 2013 ranged from 4.2 to 5.5% yr^{-1}, which is overall consistent with bottom-up inventories (3.4~5.6% yr^{-1}).**
- ►**Large reductions occurred in northern, eastern, and southwestern China**
- ►**The decreasing of anthropogenic GEM emissions in China become slow since 2019**
- ►**Reductions in China contribute a 7~10% reductions of global anthropogenic GEM emissions**

Wu et al., 2016; Liu et al., 2019; Zhang et al., 2022

Conclusions

- **Both human activities and natural surfaces are important atmospheric Hg emission sources in China.**
- **Observed Hg concentration & deposition reflect regional emission influence, particularly from anthropogenic sources.**
- **Observed GEM concentrations have declined significantly at remote Chinese sites since the early 2010s.**
- **The co-benefits of China's Clean Air Action reduce anthropogenic Hg emissions in China.**
- **We estimate that anthropogenic GEM emissions have been reduced by 116-151 tons from 2013 to 2022, representing a 7~10% reduction of the global GEM emissions**

Acknowledgements

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- the National 973 Program of China (2013CB430003)
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