

# Top-down $\text{NH}_3$ emissions based on IASI observations and GEOS- Chem simulations, 2008-2018

Zhenqi Luo, visiting student, Atmospheric Environment Research Laboratory, Westlake University  
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# Research Overview

- Observed and Simulated  $\text{NH}_3$  Concentrations
- Optimized Emission Fluxes
- Regional Comparison to GEOS-Chem Emissions

# NH<sub>3</sub> Seasonal Concentrations

Mean ( $10^{15}$  molecules cm $^{-2}$ )

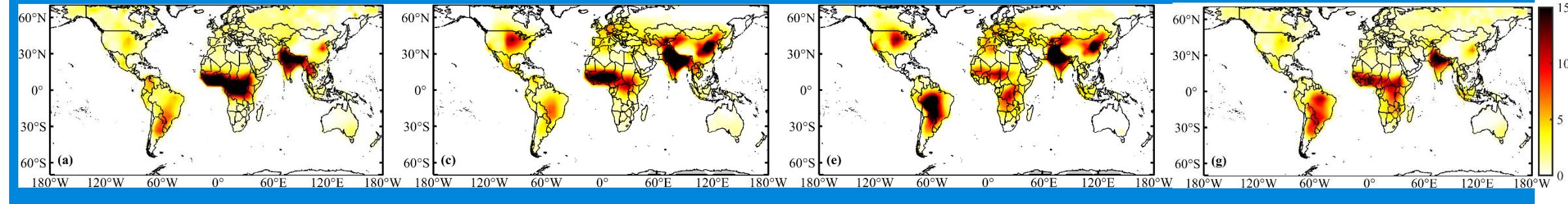
## IASI observations

JFM

AMJ

JAS

OND



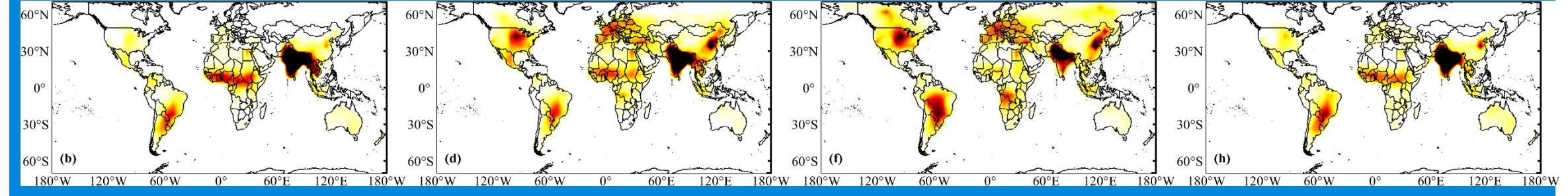
## GEOS-Chem simulations

JFM

AMJ

JAS

OND



# NH<sub>3</sub> Seasonal Concentrations

Trend ( $10^{-6}$  Mol m $^{-2}$  yr $^{-1}$ )

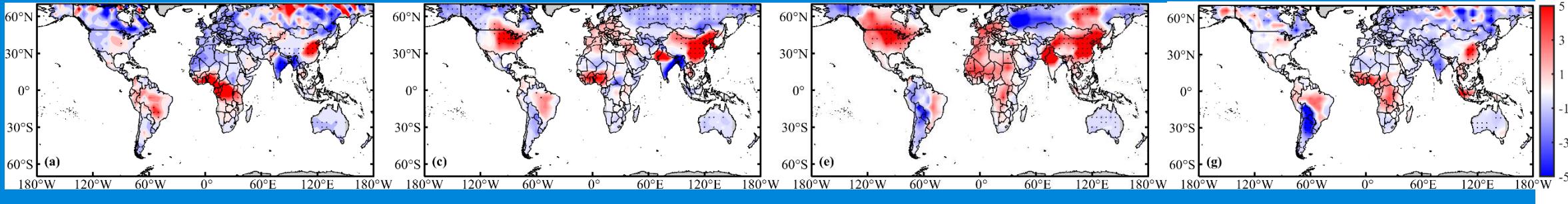
## IASI Observations

JFM

AMJ

JAS

OND



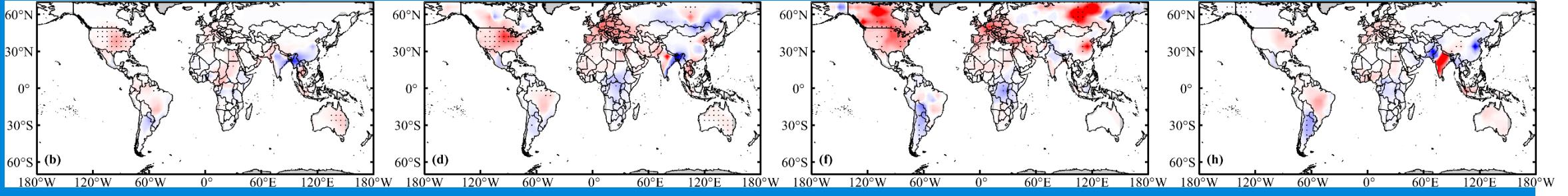
## GEOS-Chem simulations

JFM

AMJ

JAS

OND



# Emission Fluxes

Lifetime ( $\tau$ )

## Quasi-equilibrium

Emission fluxes of ammonia  
are continuous

$$\tau = \frac{M}{F_{out} + L + D}$$

- $M$ :  $\text{NH}_3$  mass
- $F_{out}$ :  $\text{NH}_3$  mass rate of export
- $L$ :  $\text{NH}_3$  mass rate of chemical reaction
- $D$ :  $\text{NH}_3$  mass rate of deposition, including the wet deposition and the dry deposition

## Ammonia-water equilibrium

The  $\text{NH}_4^+$  concentration ( $[\text{NH}_4^+]$ ) is proportional to  $\text{NH}_3$  concentration ( $[\text{NH}_3]$ )

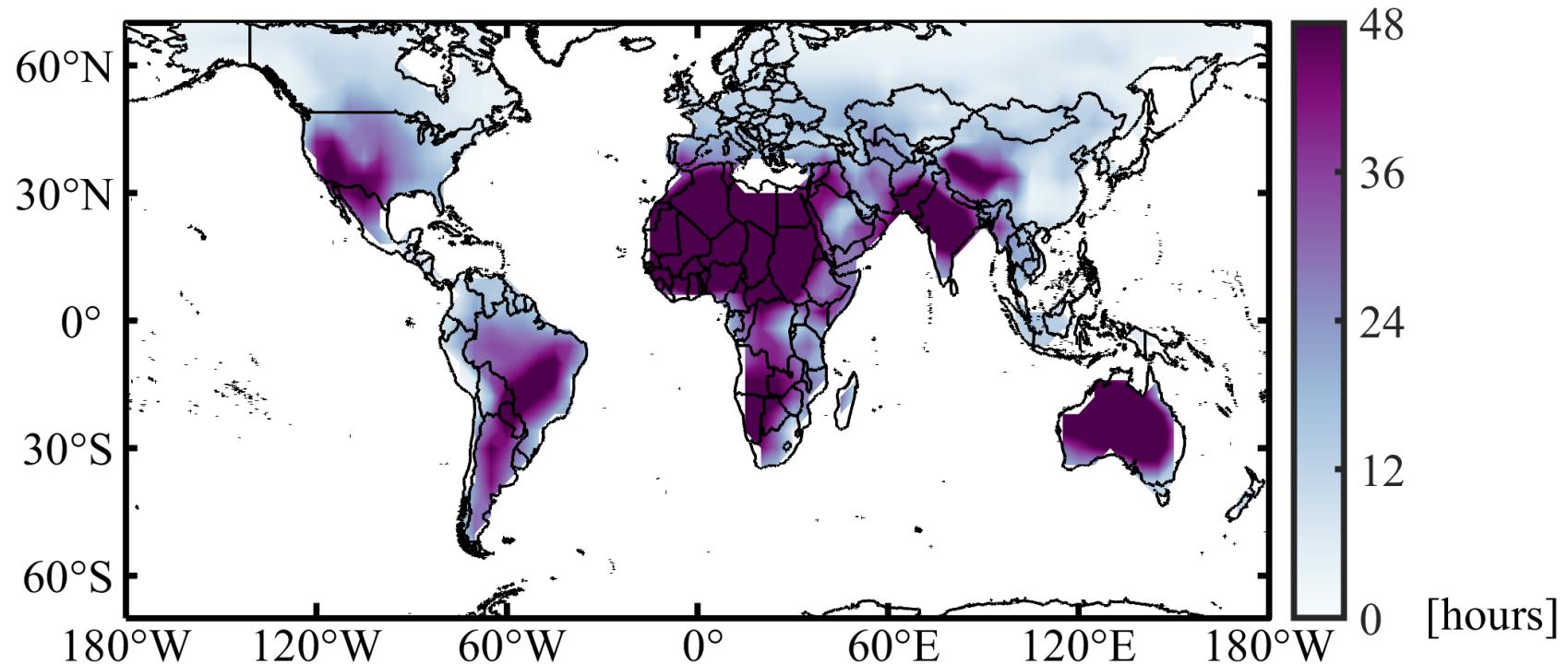
$$[\text{NH}_4^+] = K \times [\text{NH}_3]$$

$$D = D_{\text{NH}_3} + D_{\text{NH}_4^+}$$

## Modeled lifetime

Neglect the  $\text{NH}_3$  export ( $F_{out}$ )

$$\tau_{mod} = \frac{M_{\text{NH}_3}}{D_{\text{NH}_3} + D_{\text{NH}_4^+}}$$



# Emission Fluxes

## Average Lifetime

Apply one-box model that neglect the  $\text{NH}_3$  export and consider the ammonia-water equilibrium.

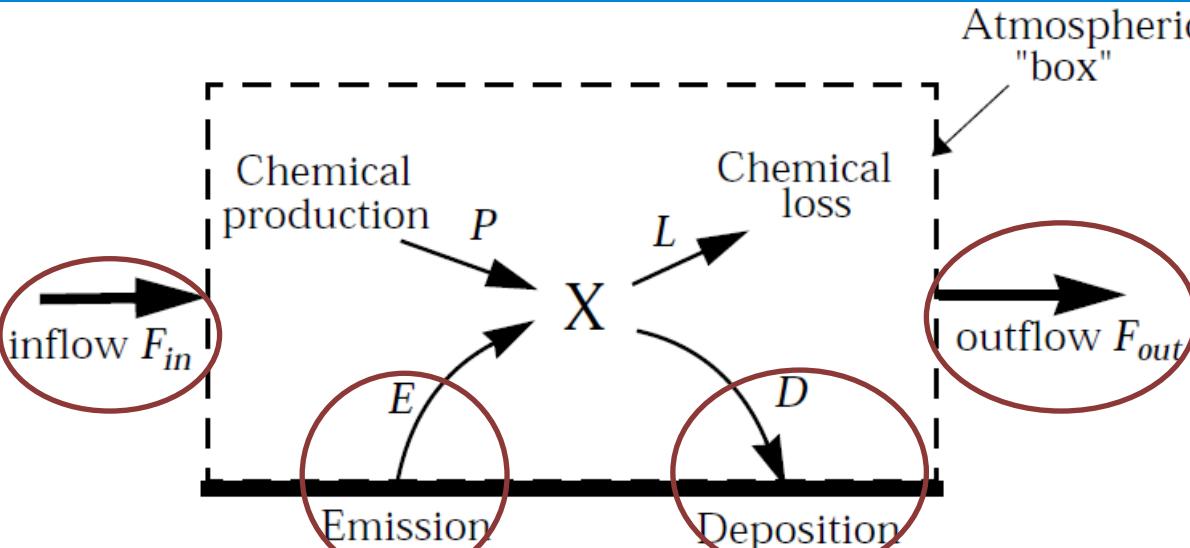


Figure 3-1 One-box model for an atmospheric species X

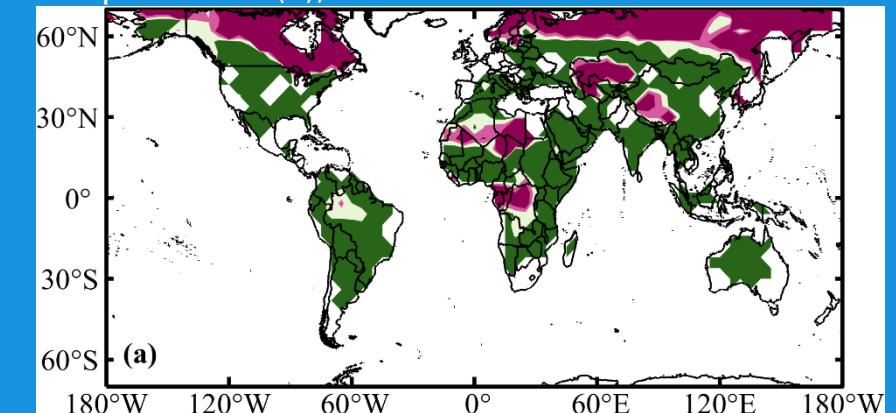
(Jacob, 1999)

## Emission Fluxes

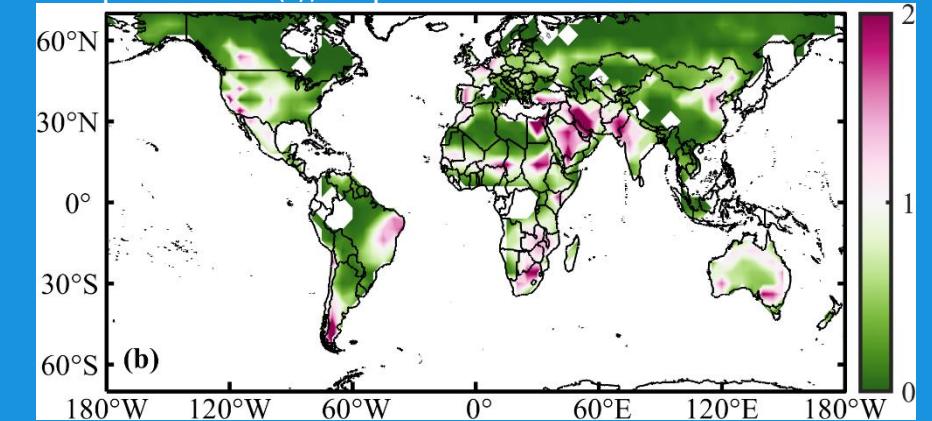
$$\text{Transportation} = F_{\text{in}} - F_{\text{out}}$$

Note Two Situations!

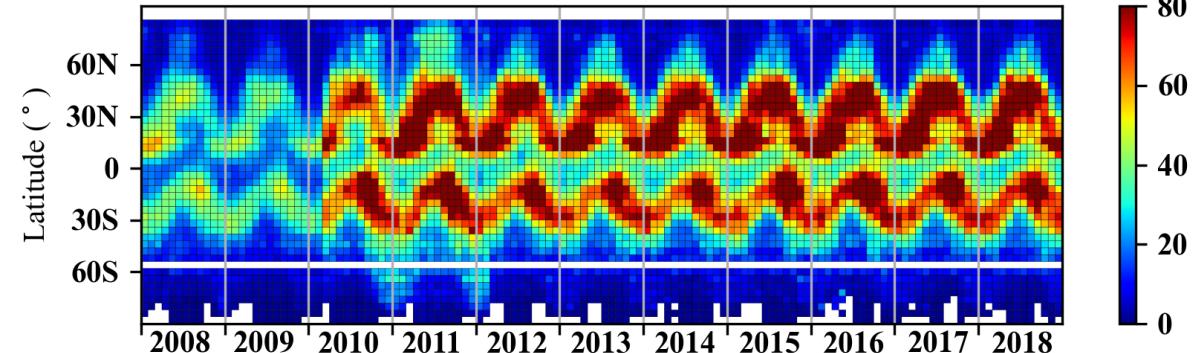
- Transportation  $> 0, F_{\text{in}} > F_{\text{out}}$
- Emissions may be unreliable if the transportation of  $\text{NH}_3$  and  $\text{NH}_4^+$  are larger than emissions
- Transportation (+)/Emissions:



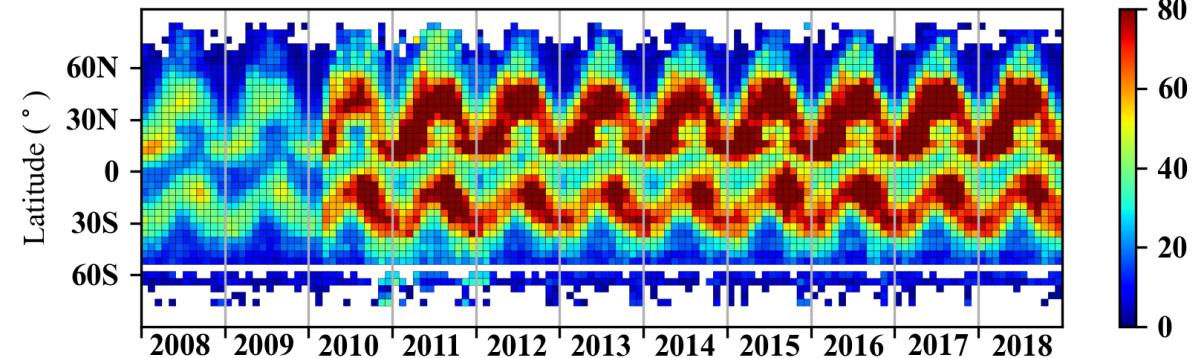
- Transportation  $< 0, F_{\text{in}} < F_{\text{out}}$
- Emissions may be unreliable if the transportation of  $\text{NH}_3$  and  $\text{NH}_4^+$  are larger than deposition
- Transportation (-)/Depositions:



First filter



Second filter

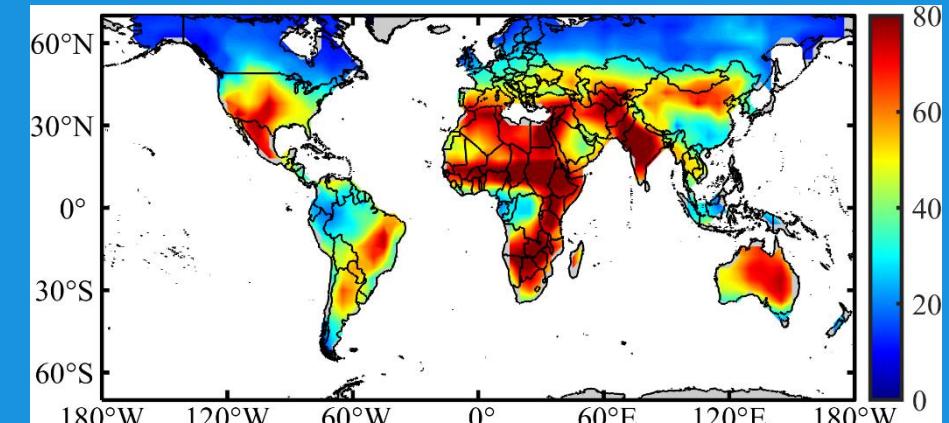


## Emission Fluxes

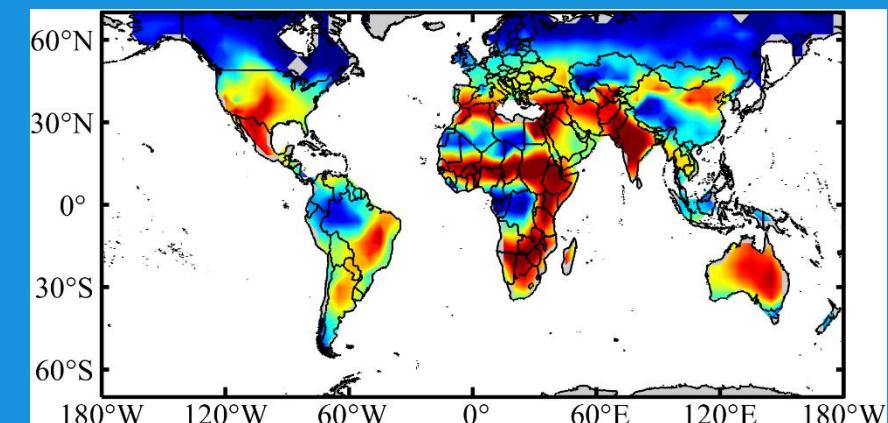
Number of Retrievals

Note Data Validation!

- First filter—based on IASI datasets (Damme et al., 2014)
  - Cloud coverage: [0, 10%]
  - Skin temperature:  $> 263.15 \text{ K}$
- Average number of retrievals ( $\text{N grid}^{-1} \text{ day}^{-1}$ )



- Second filter—based on GEOS-Chem output
  - Transportation < Emission

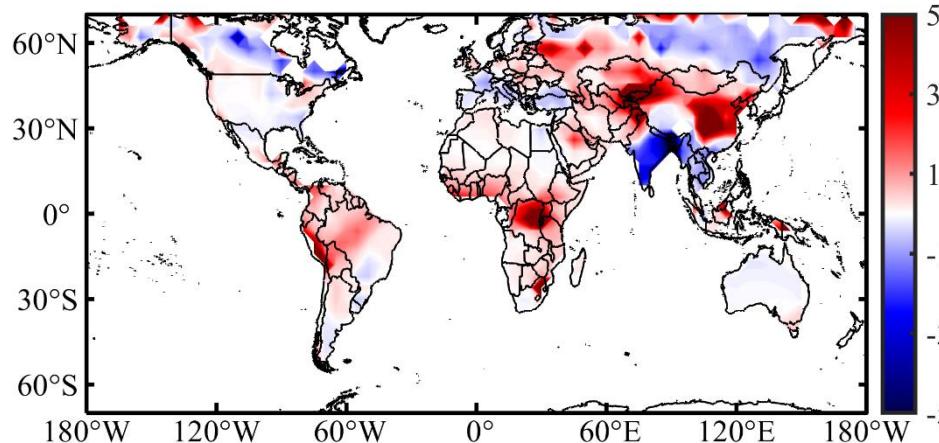


# Emission Fluxes ( $\hat{E}$ )

$$\hat{E} = \frac{\Delta M_{NH_3}}{\tau_{mod}} + E_{mod}$$

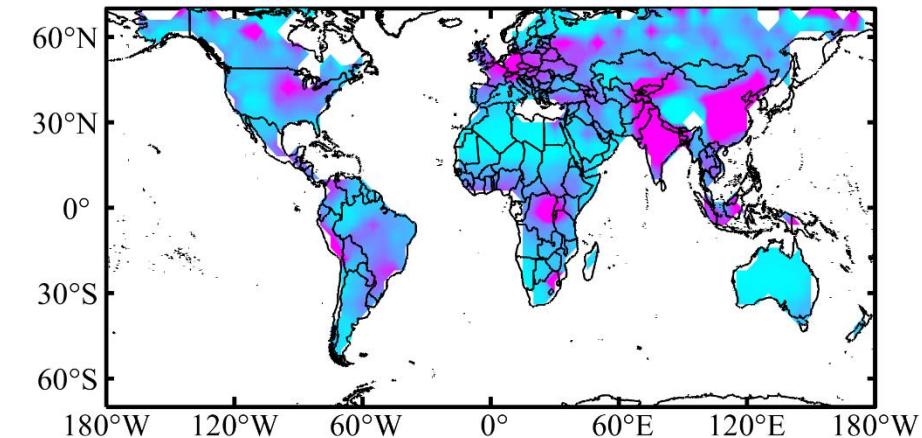
- $\Delta M_{NH_3}$ : the  $NH_3$  mass difference between observation and simulation in each atmospheric box
- $E_{mod}$ : GEOS-Chem emission

## ■ Optimized – GEOS-Chem

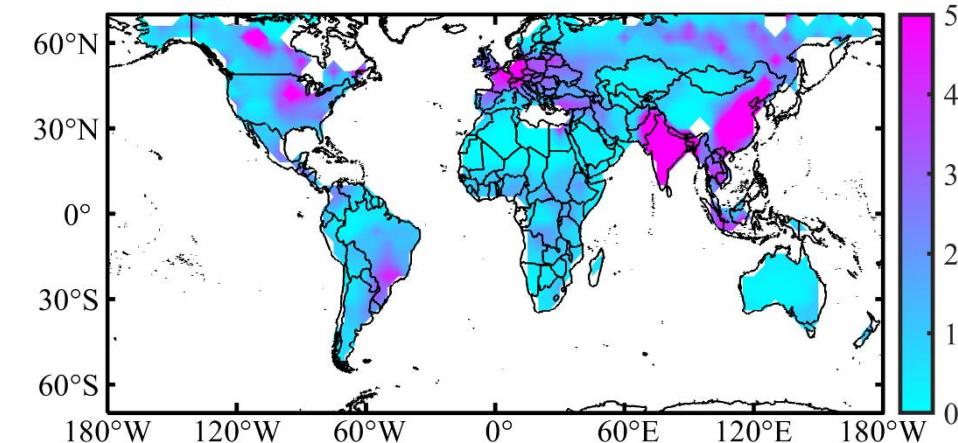


Mean ( $10^{-11} \text{ kg m}^{-2} \text{ s}^{-1}$ )

■ Optimized ( $N > 30 \text{ grid}^{-1} \text{ day}^{-1}$ )



## ■ GEOS-chem



# Emission Fluxes ( $\hat{E}$ )

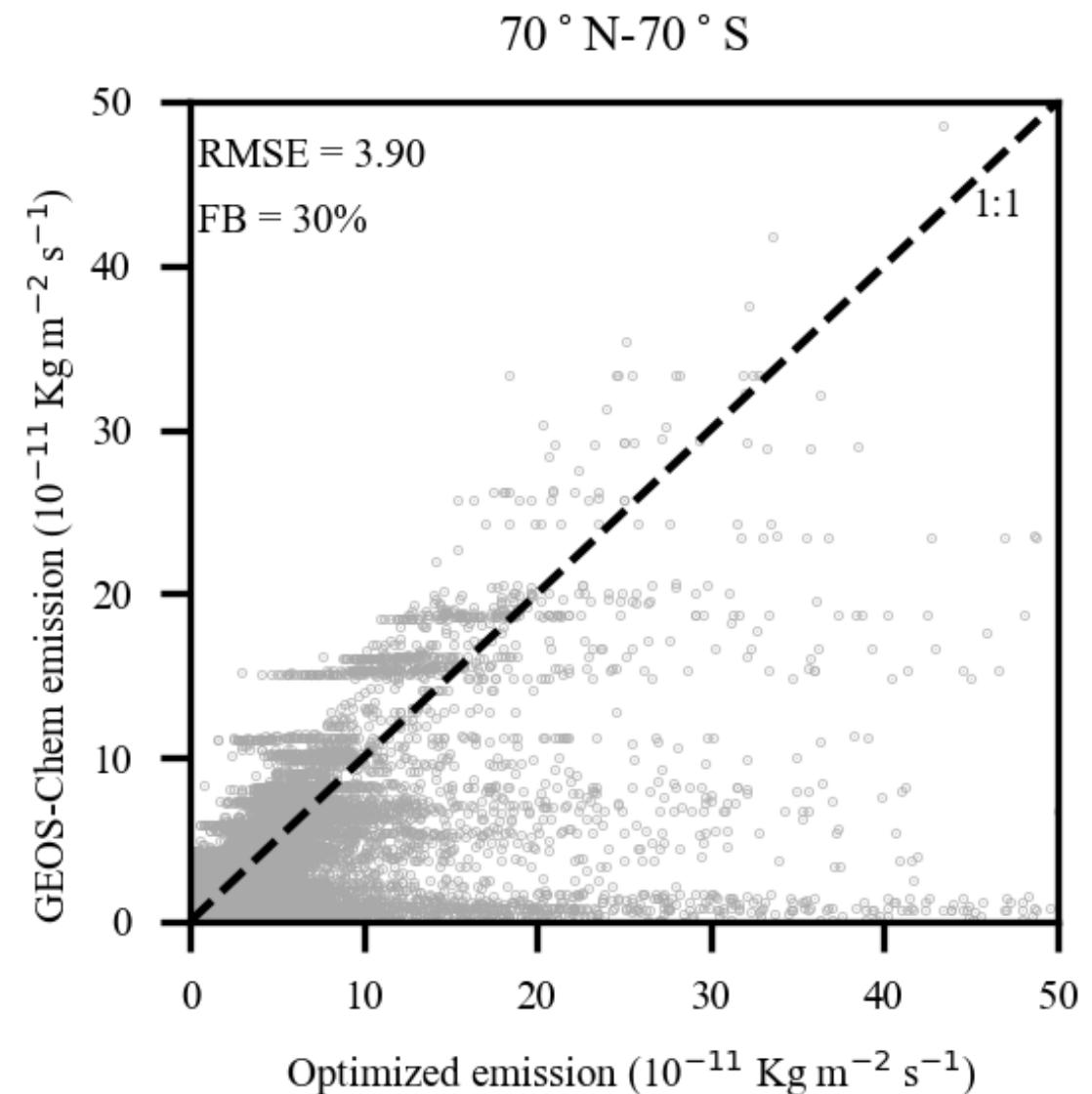
## ■ Optimized versus GEOS-Chem

- Root Mean Square Error (RMSE)

$$RMSE = \sqrt{\frac{1}{N} \sum_{i=1}^N (E_{Opt,i} - E_{Mod,i})^2}$$

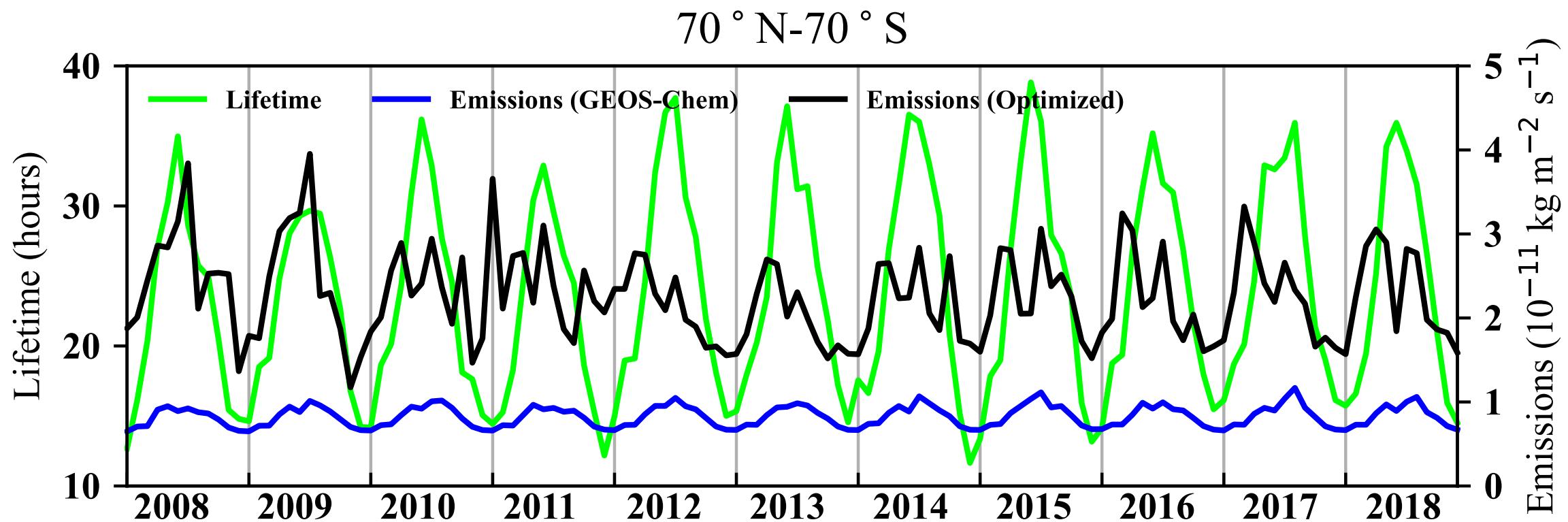
- Fractional Bias (FB)

$$FB = \frac{\sum_i^N (E_{Opt,i} - E_{Mod,i})}{\sum_i^N (E_{Opt,i} + E_{Mod,i}) / 2} \times 100\%$$



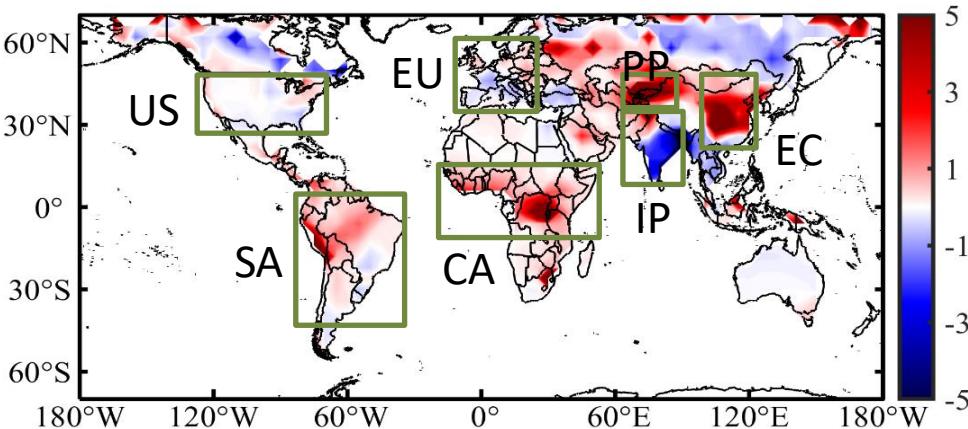
# Emission Fluxes ( $\hat{E}$ )

## ■ Monthly timeseries



# Regional Comparison

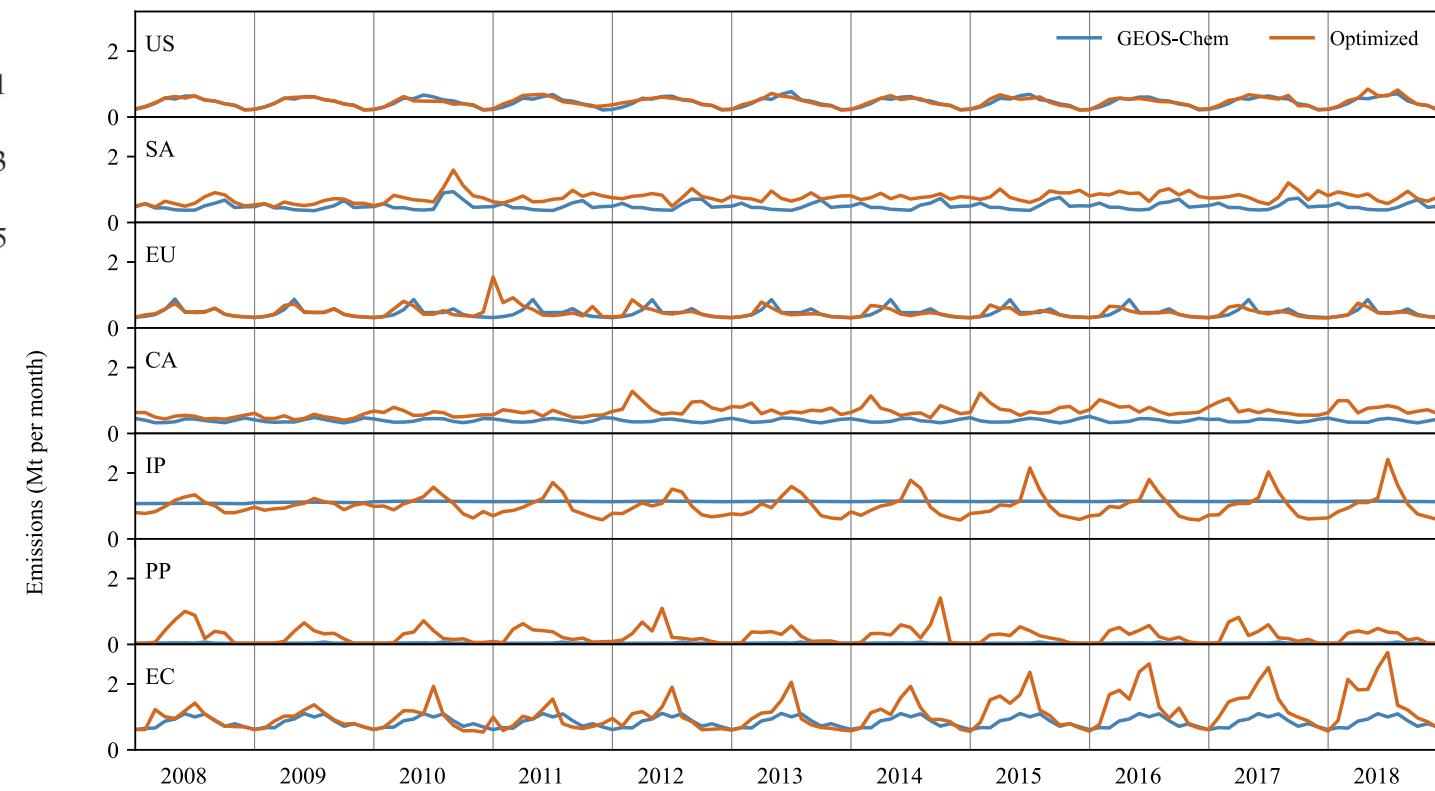
## ■ Optimized – GEOS-Chem



### 7 Regions:

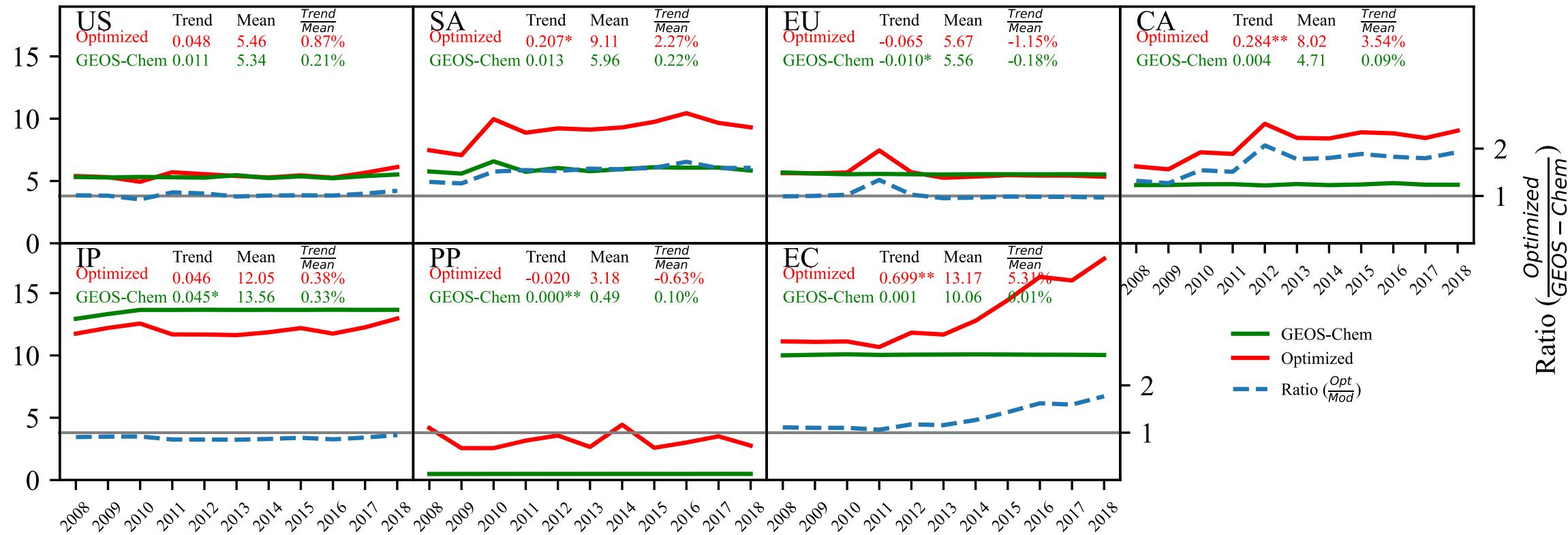
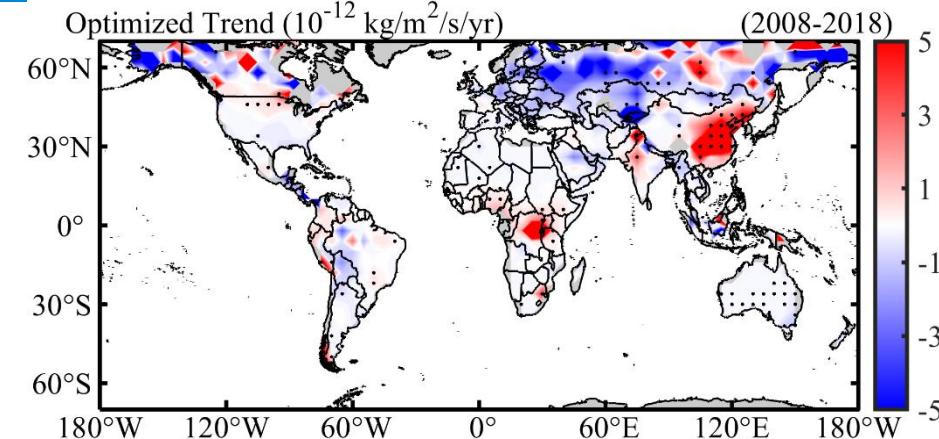
- US: Contiguous United States
- SA: south America
- EU: Europe
- CA: central Africa
- PP: Pamirs Plateau
- IP: India Peninsula
- EC: eastern China

## ■ Monthly timeseries (replace the missing value by GEOS-Chem emissions)



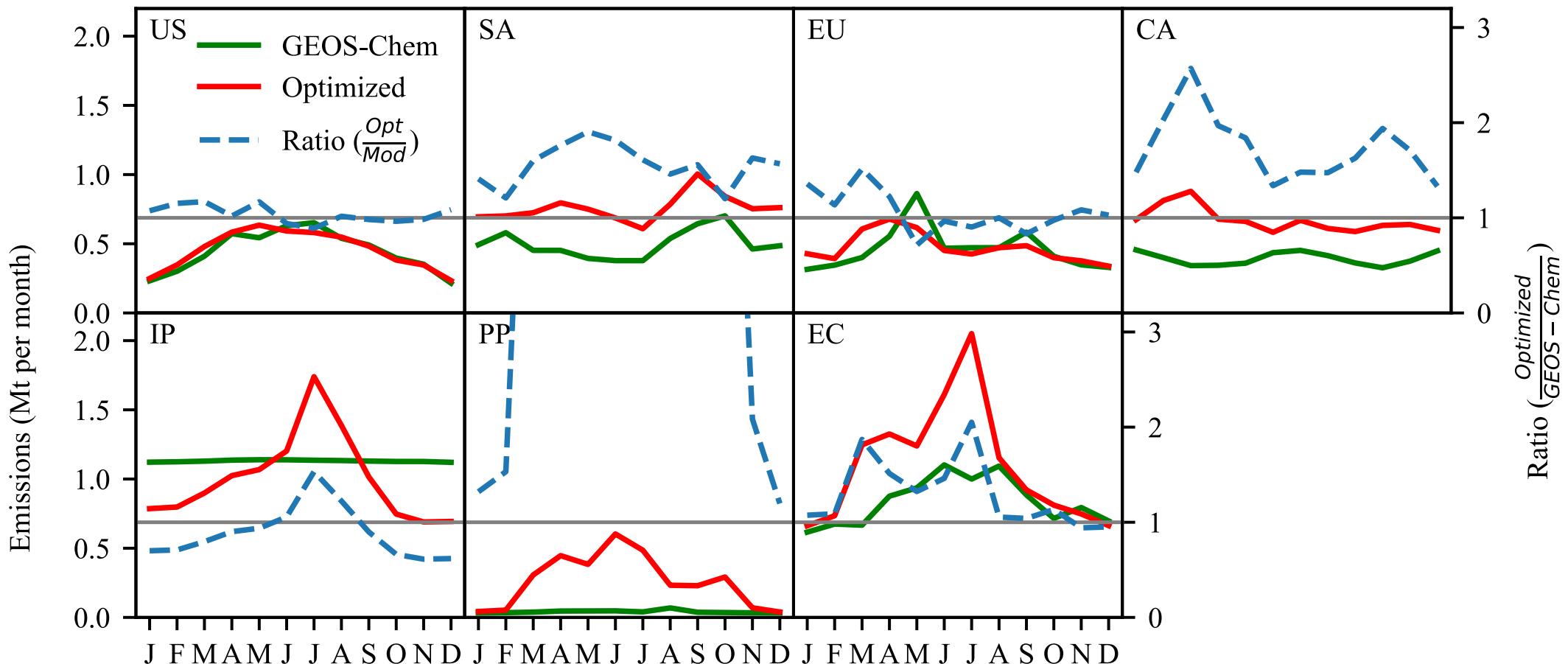
# Regional Comparison

## ■ Annual timeseries



# Regional Comparison

## ■ Monthly variation

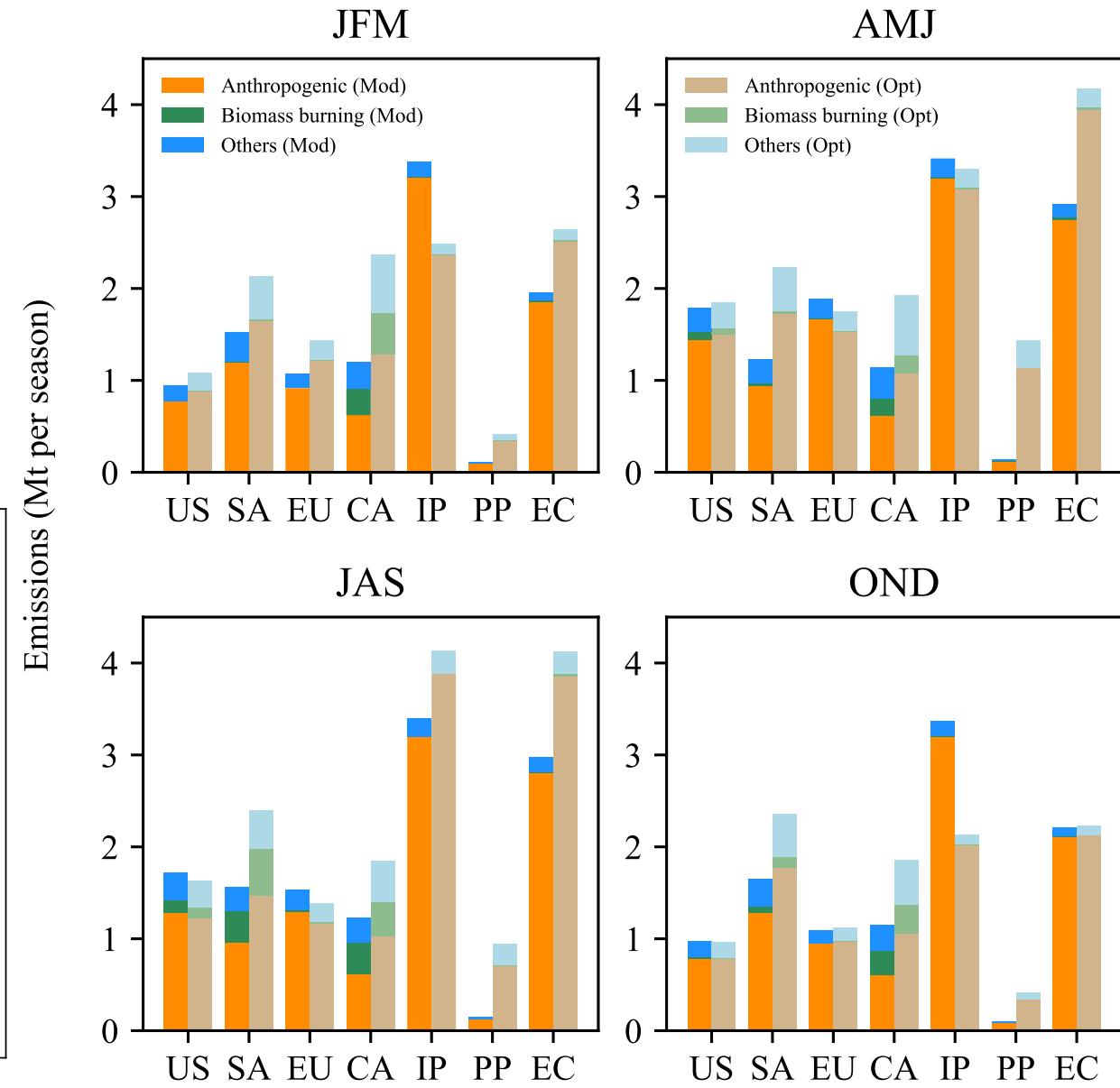
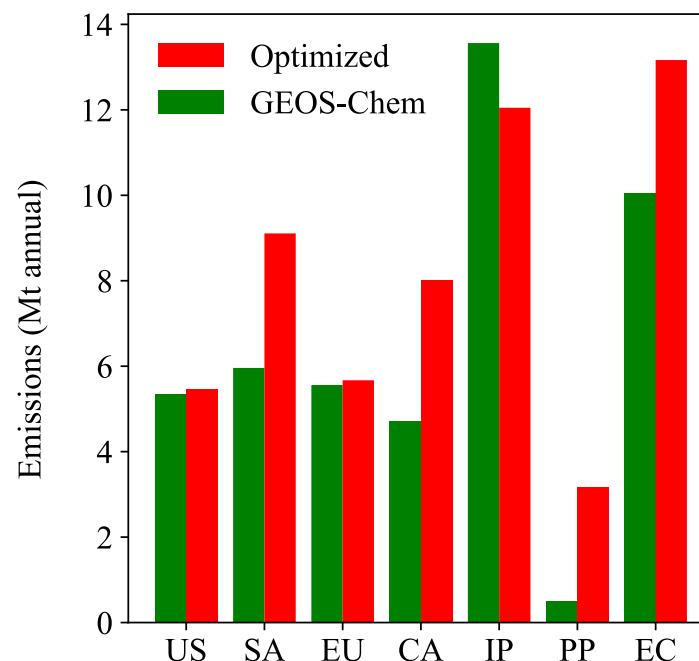


# Regional Comparison

## ■ Proportions of emission sectors

From GEOS-Chem (monthly) :

- Total
  - Biomass burning
  - Anthropogenic
  - Others



# Regional Comparison

■ Optimized versus GEOS-Chem

## Optimize < GEOS-Chem

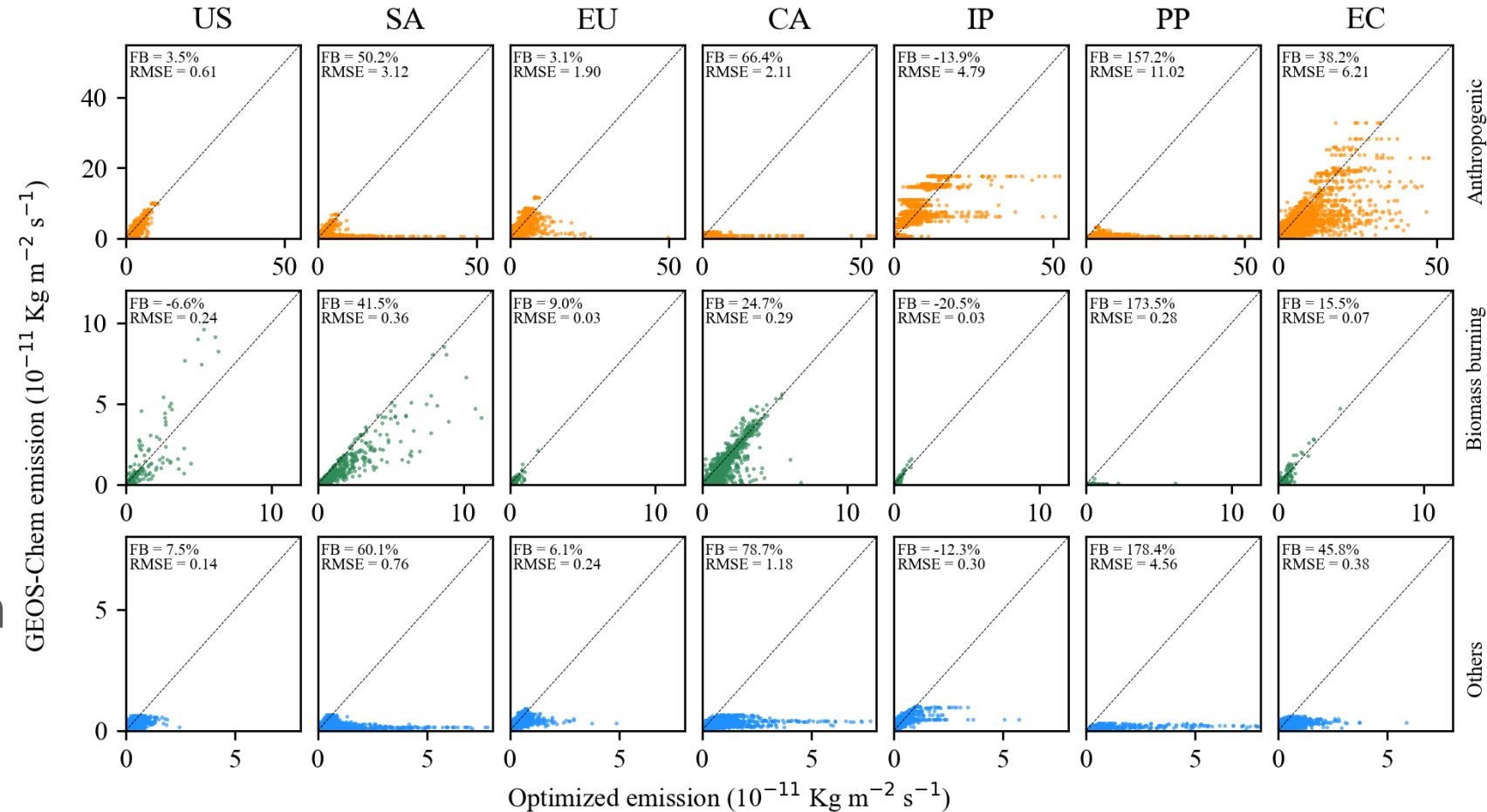
- IP

## Optimize > GEOS-Chem

- SA
- CA
- PP
- EC

## Optimize ≈ GEOS-Chem

- US
- EU

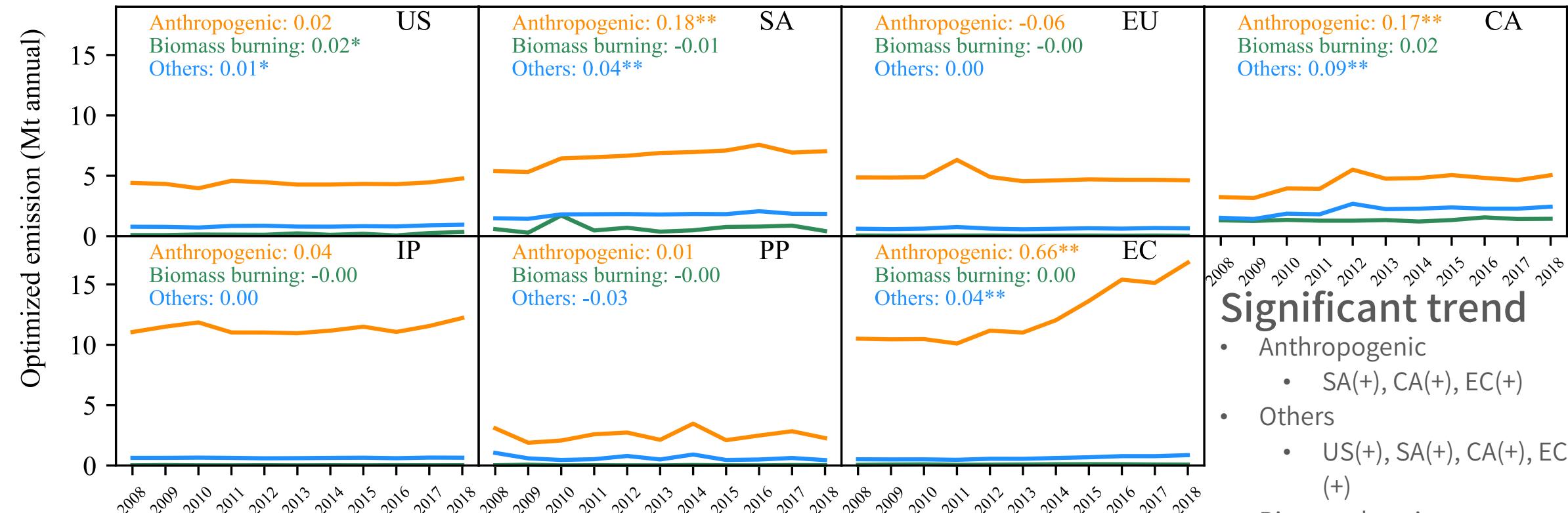


# Regional Comparison

## ■ Optimized emission trends

### Significant confidence levels

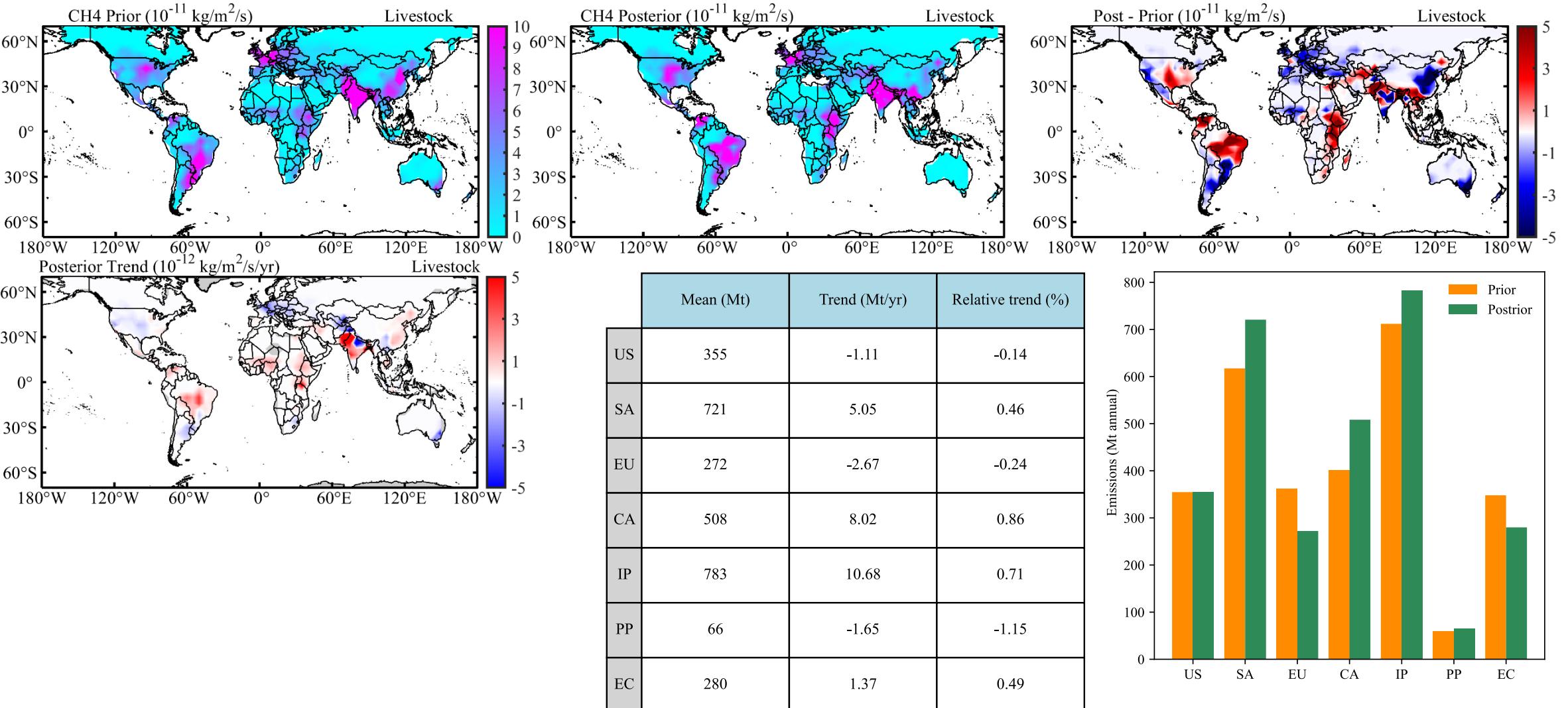
- >95%: \*
- >99%: \*\*



### Significant trend

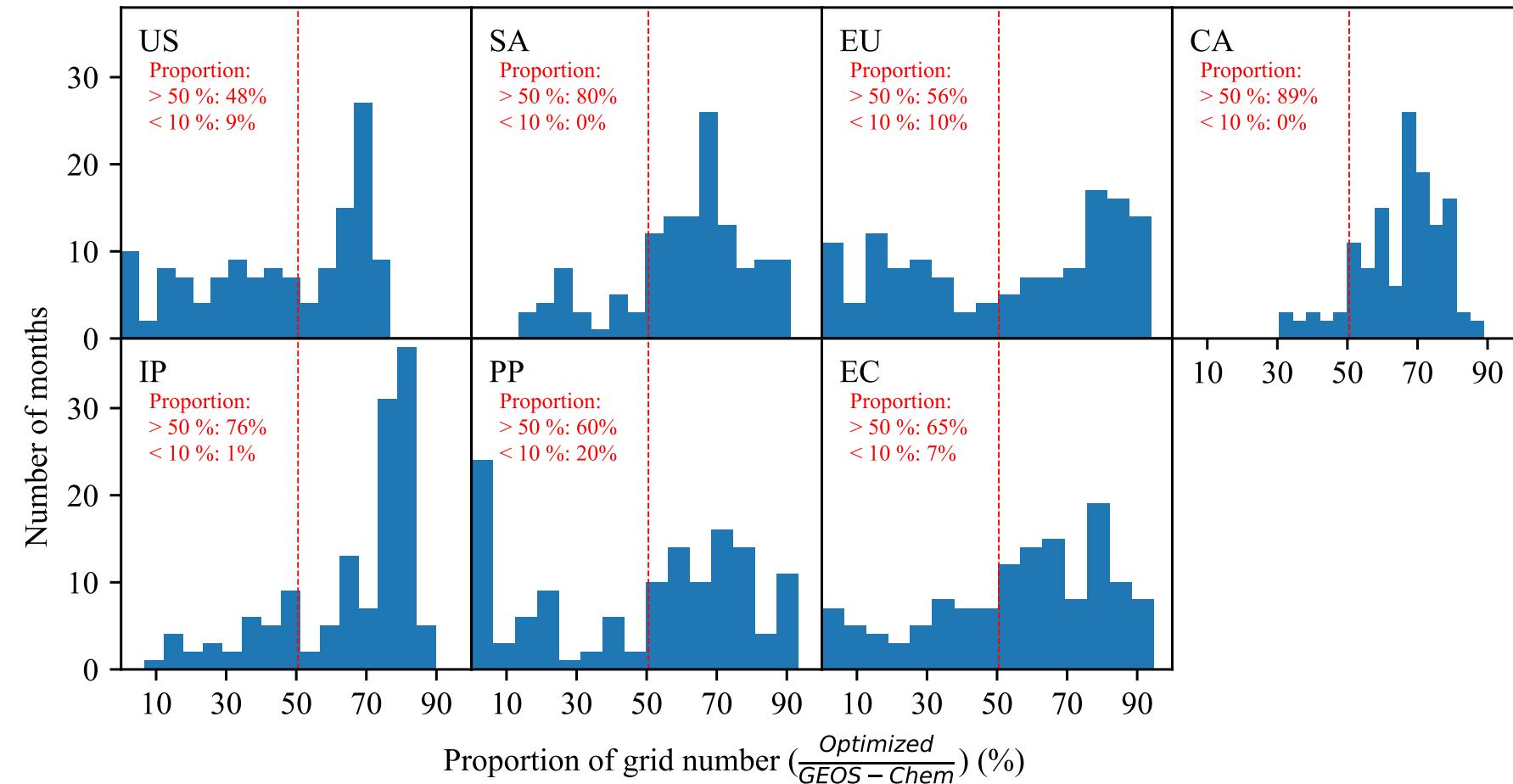
- Anthropogenic
  - SA(+), CA(+), EC(+)
- Others
  - US(+), SA(+), CA(+), EC(+)
- Biomass burning
  - US(+)

# CH<sub>4</sub> Livestock Emission Fluxes



# Regional Comparison

## ■ Optimized grid number proportion

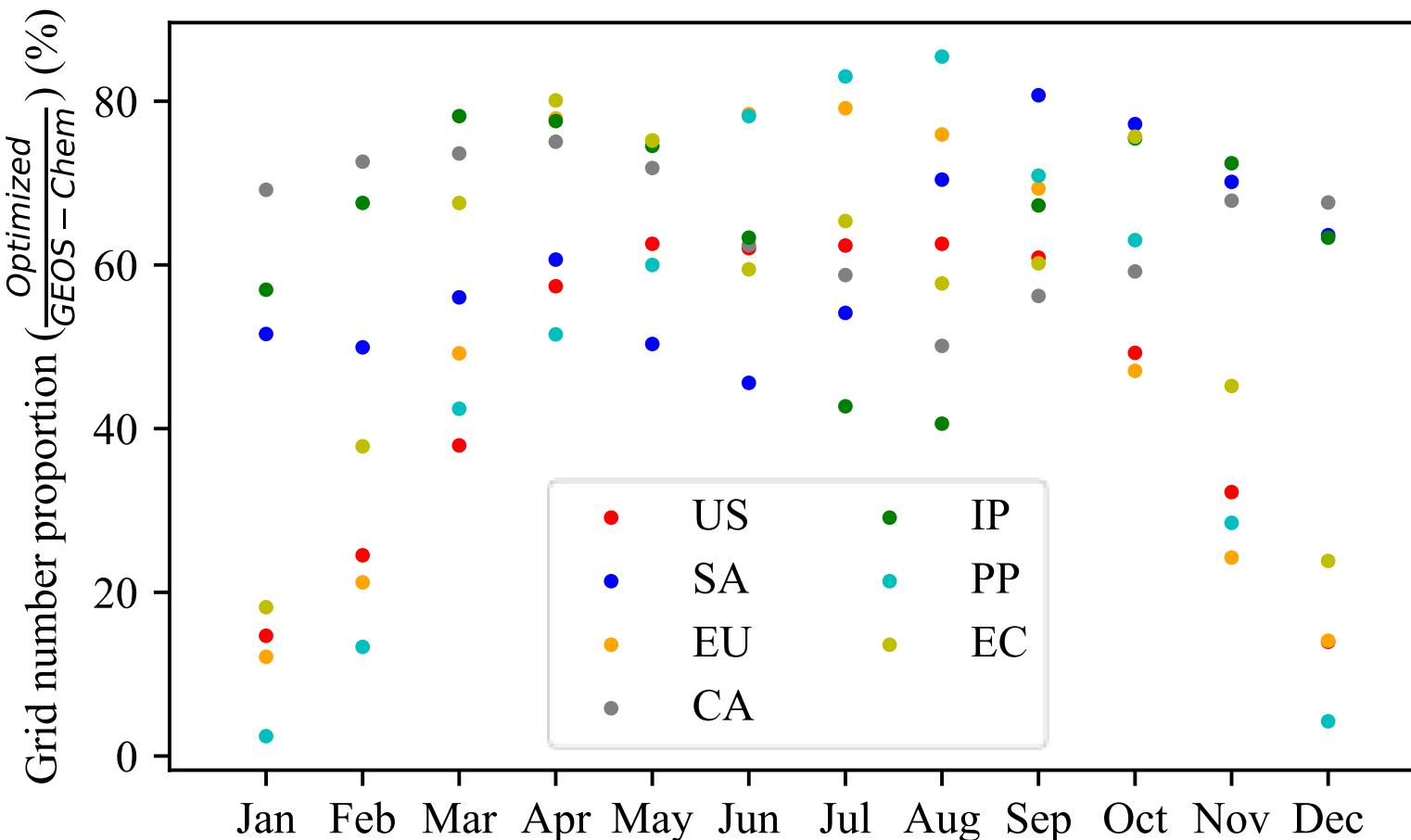


### Data quantity

- Good:
  - SA, CA, IP
- Not bad:
  - US, EC, EU
- Bad:
  - PP

# Regional Comparison

## ■ Optimized grid number proportion



# Reference

- Jacob, Daniel J. Introduction to atmospheric chemistry. Princeton University Press, 1999.
- Van Damme M, Clarisse L, Whitburn S, et al. Industrial and agricultural ammonia point sources exposed[J]. Nature, 2018, 564(7734): 99-103.
- Warner J X, Dickerson R R, Wei Z, et al. Increased atmospheric ammonia over the world's major agricultural areas detected from space[J]. Geophysical Research Letters, 2017, 44(6): 2875-2884.
- Evangelou N, Balkanski Y, Eckhardt S, et al. 10–year satellite-constrained fluxes of ammonia improve performance of chemistry transport models[J]. Atmospheric Chemistry and Physics Discussions, 2020: 1-41.
- Van Damme M, Clarisse L, Heald C L, et al. Global distributions, time series and error characterization of atmospheric ammonia ( $\text{NH}_3$ ) from IASI satellite observations[J]. Atmospheric chemistry and physics, 2014, 14(6): 2905-2922.

# THANK YOU!