

# Top-down ammonia emissions 19

IASI data and GEOS-Chem simulation

2021.8

- Method:
  - 1. IASI observations
  - 2. GEOS-Chem simulations
  - 3. NH<sub>3</sub> emission flux estimations
  - 4. Uncertainty and sensitivity analysis
- Results and discussion:
  - Observed and simulated NH<sub>3</sub> concentrations
  - Comparison of top-down NH<sub>3</sub> emission estimations
    - compare lifetime of NH<sub>3</sub> and NH<sub>x</sub> and their emissions
    - compare concentration based on updated emission over 2008 and 2018
  - Global NH<sub>3</sub> emissions distributions
  - Regional NH<sub>3</sub> emissions trends
    - Find evidence of biomass burning in northern high latitudes
  - Relation to methane emissions

	Parameter perturbed	Average emission (Tg a <sup>-1</sup> )
0	None <sup>a</sup>	92
1	Half lifetime <sup>b</sup>	115
2	Double lifetime <sup>c</sup>	80
3	Upper IASI column error	107
4	Lower IASI column error	77
5	Number of retrievals > 400 <sup>d</sup>	95
6	Number of retrievals > 1200 <sup>e</sup>	87
7	Transport/Emission < 0.2 <sup>f</sup>	87
8	Transport/Emission < 5 <sup>g</sup>	102

<sup>a</sup>The number of retrievals larger than 800, and the transport dominates over emissions or depositions when exclude grids.

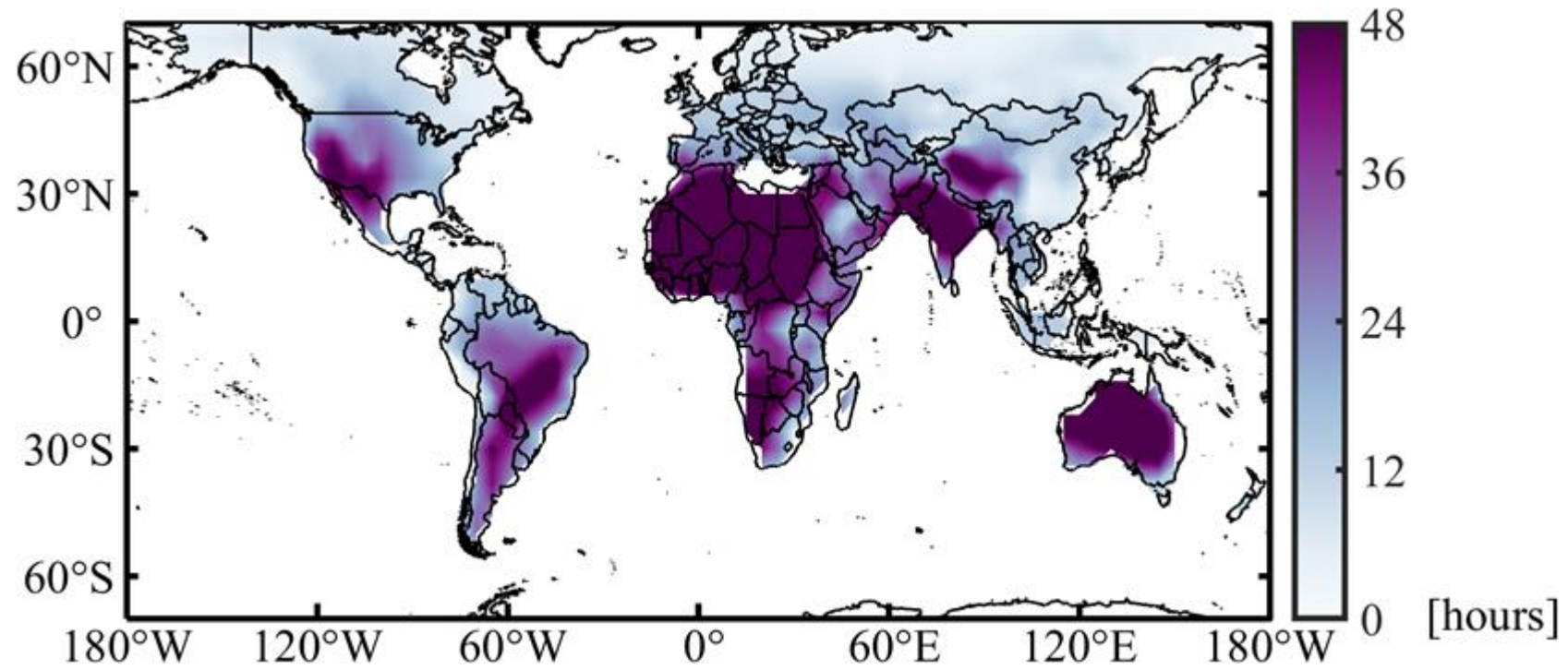
<sup>b-c</sup>The lifetime is 50% and 200% in Eq. (1), respectively.

<sup>d-e</sup>The number of retrievals larger than 400 and 800 when exclude grids, respectively.

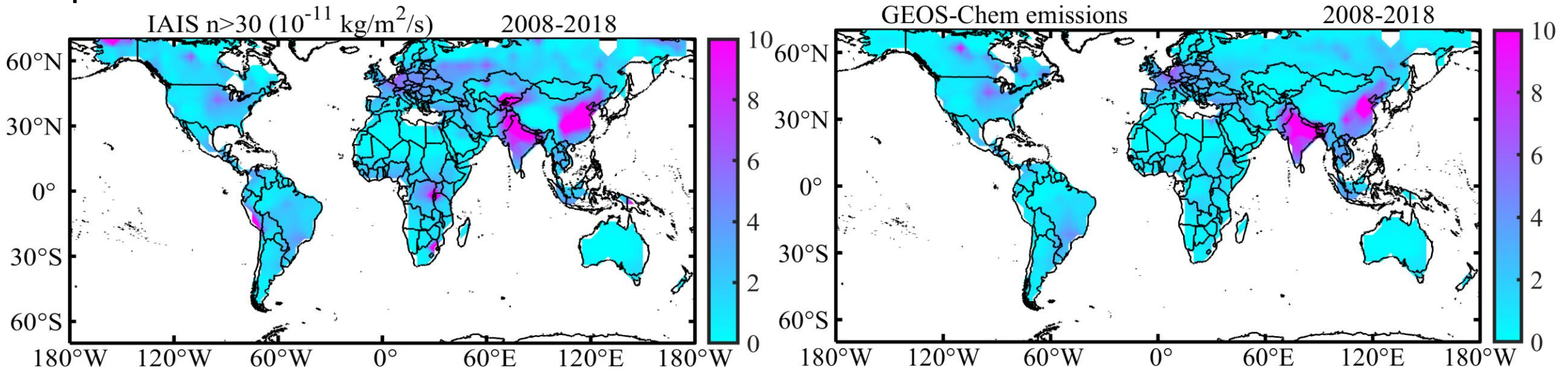
<sup>f-g</sup>The threshold used in the budget analysis is 0.2 and 5, respectively.



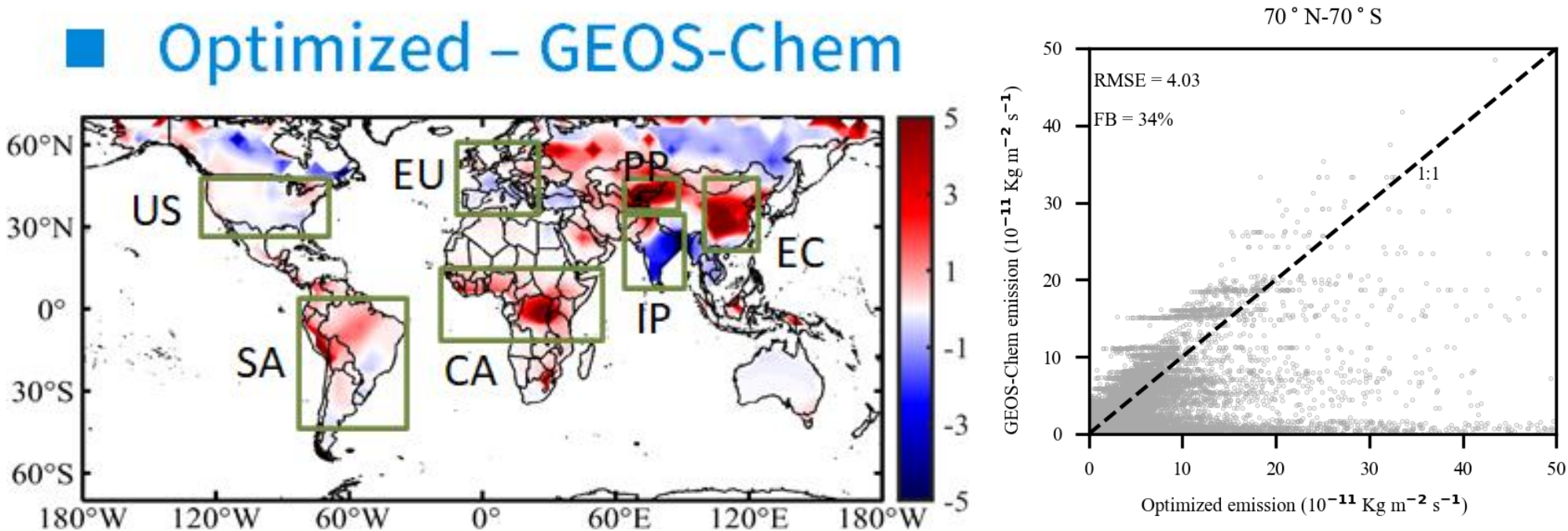
# lifetime



# Optimized versus GEOS-Chem

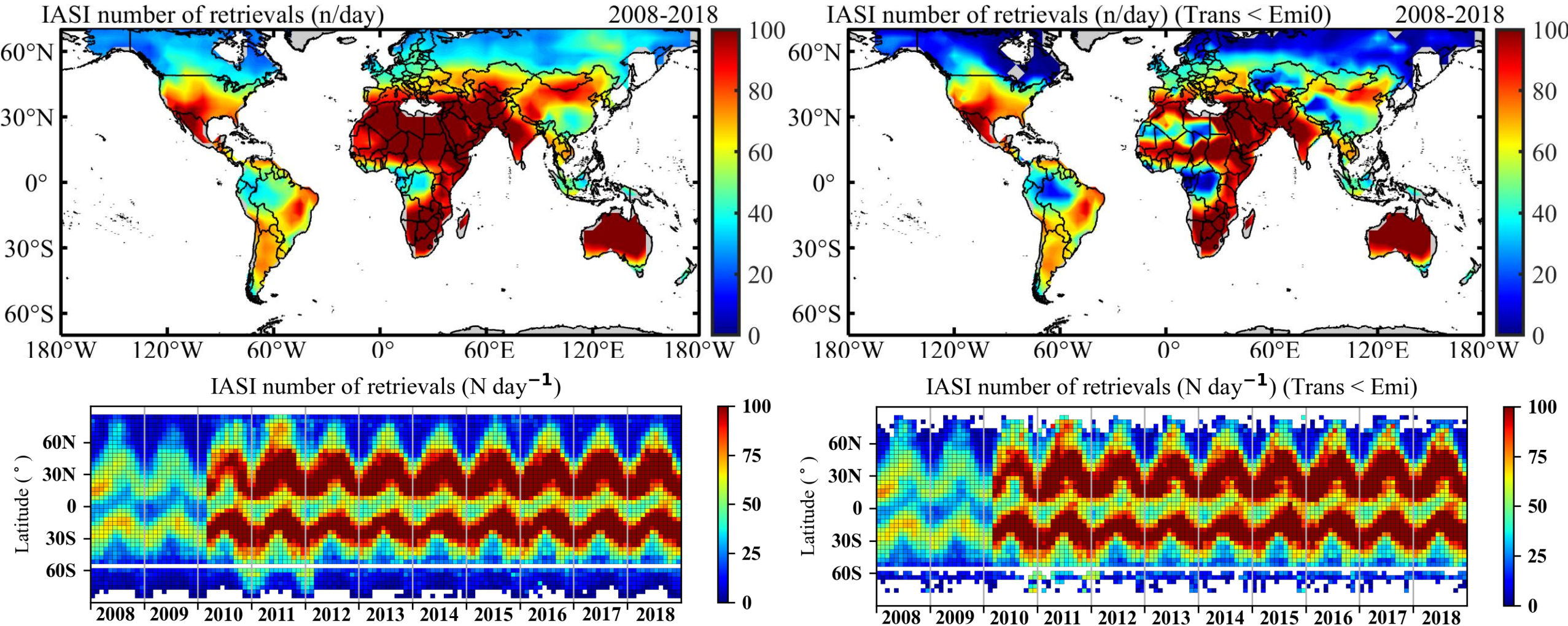


## ■ Optimized – GEOS-Chem

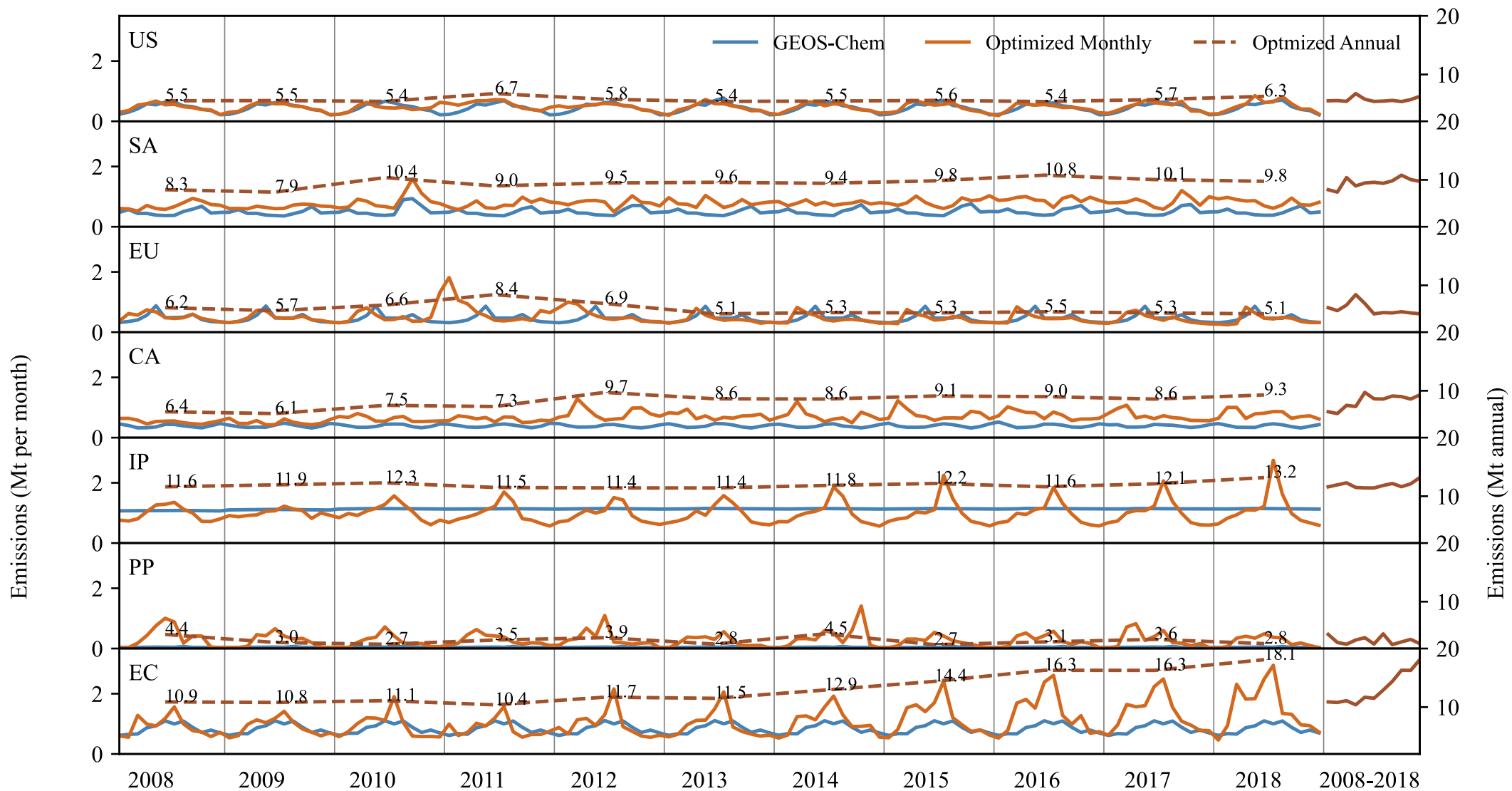




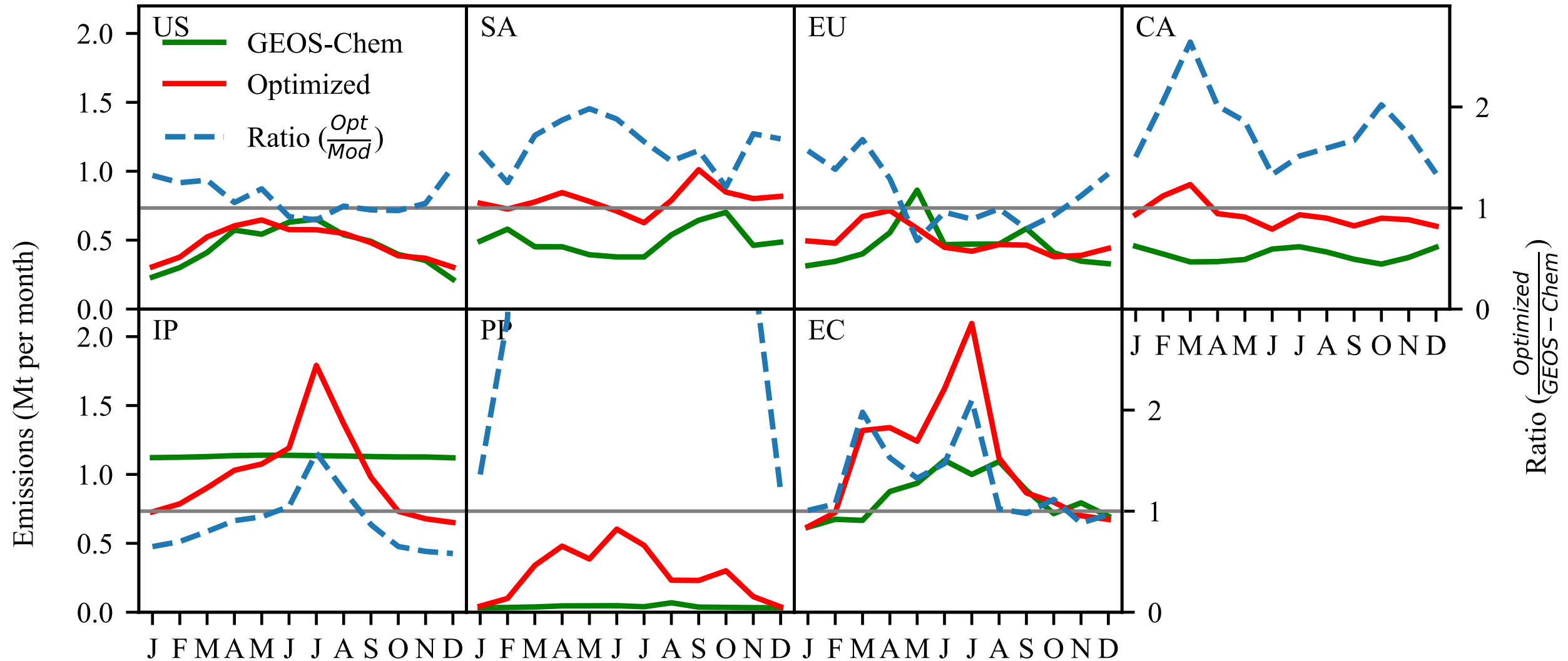
# Number of retrievals



# Optimized emissions timeseries

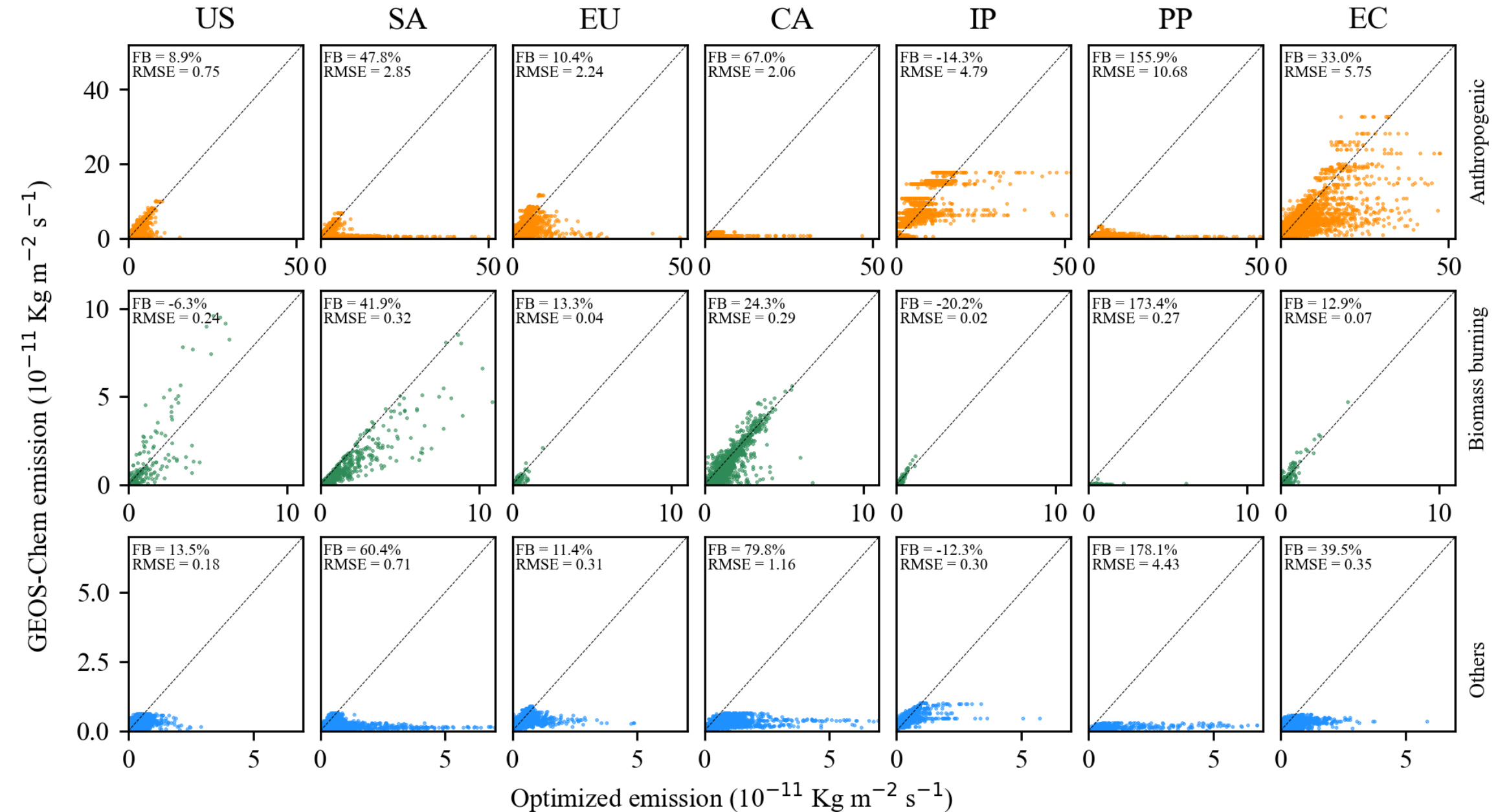


# Optimized emissions monthly variations

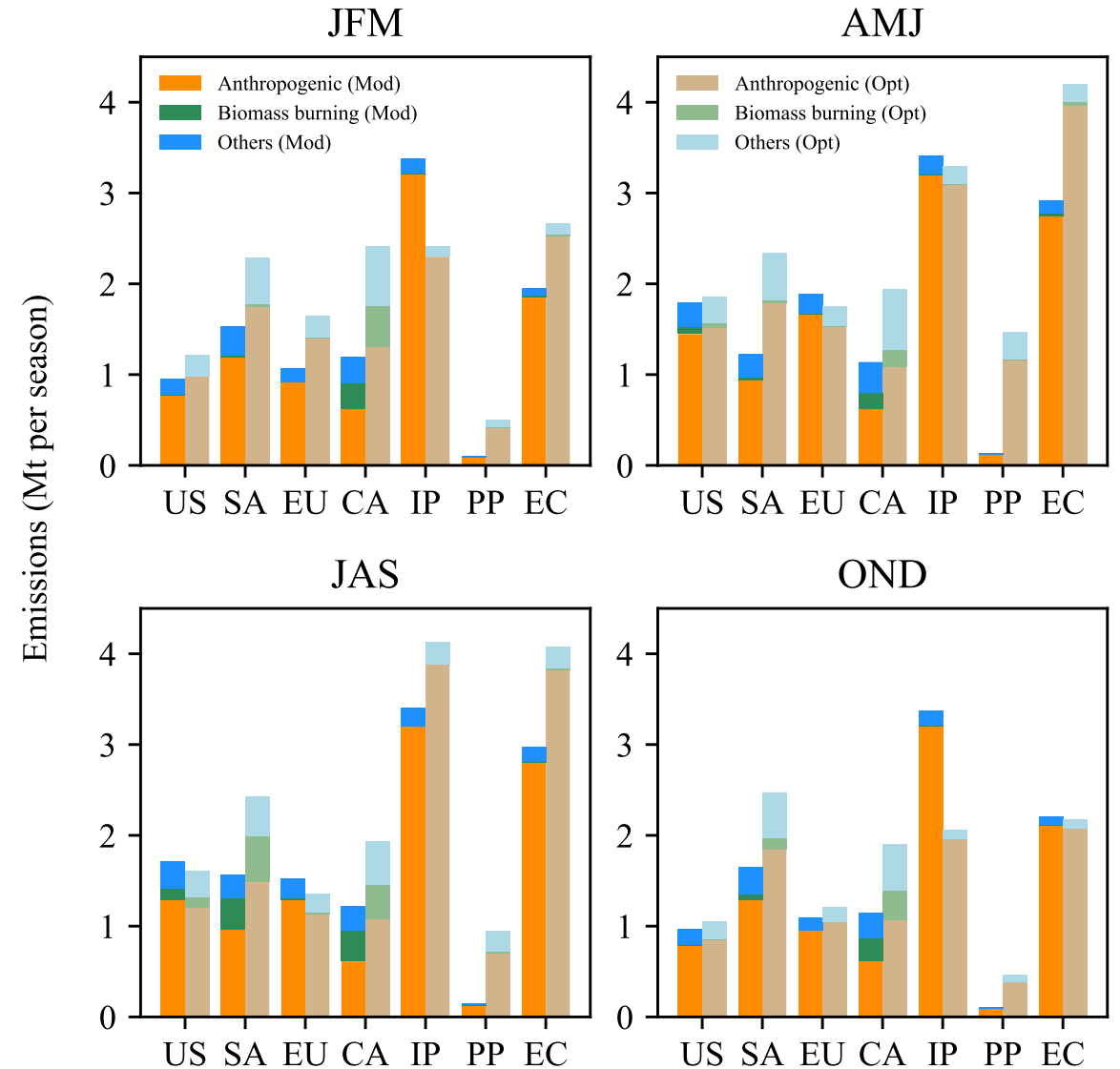
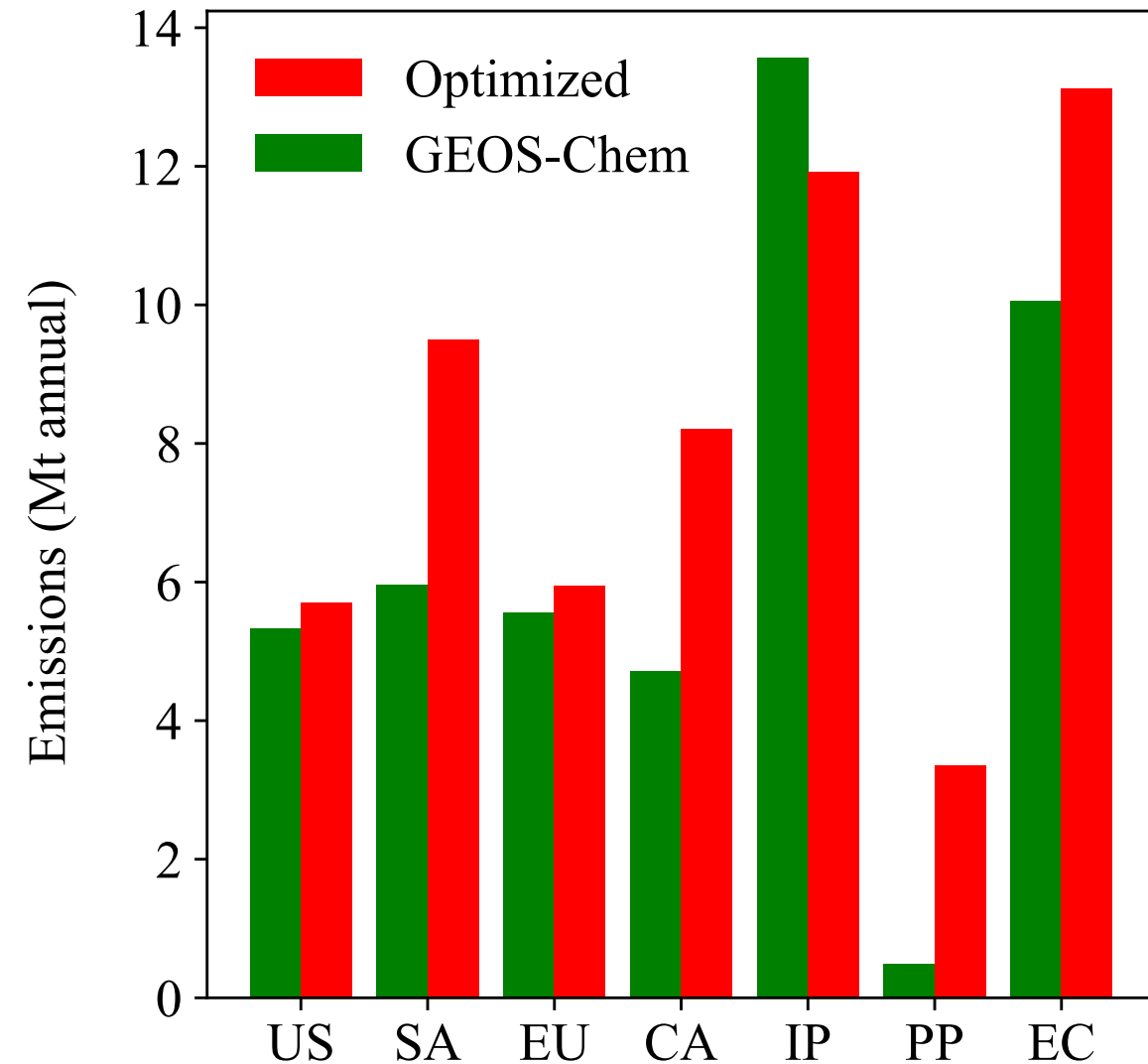




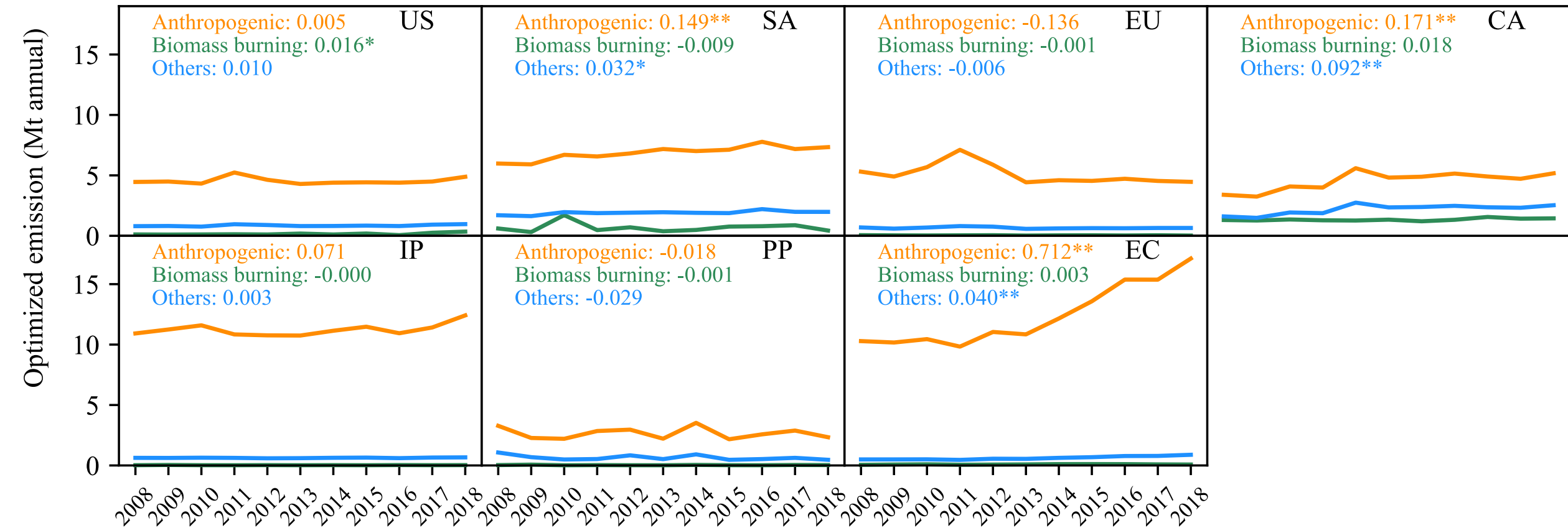
# Optimized versus GEOS-Chem by sectors



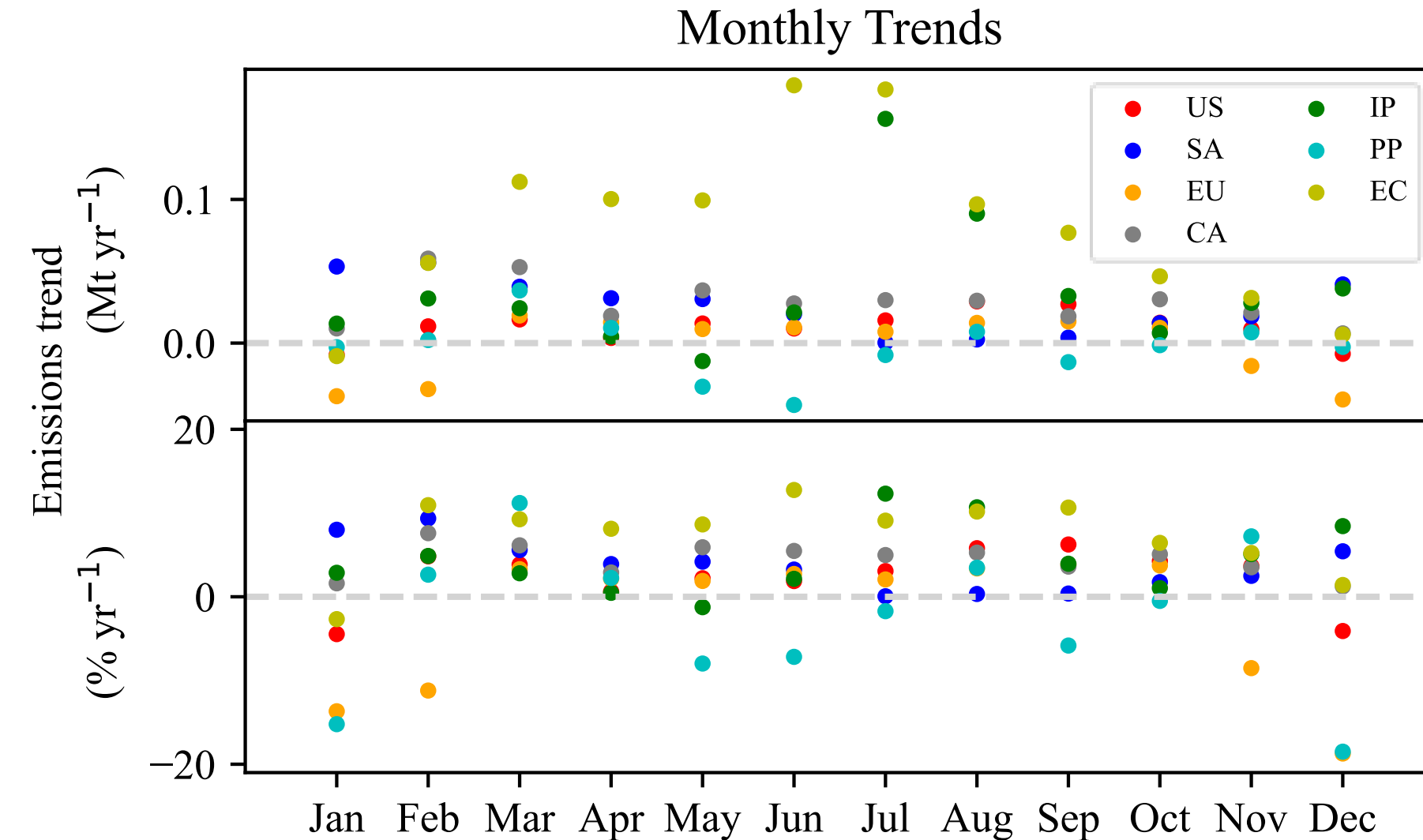
# Optimized emissions by sectors



# Optimized emissions trend by sectors



# Optimized emissions monthly trend



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

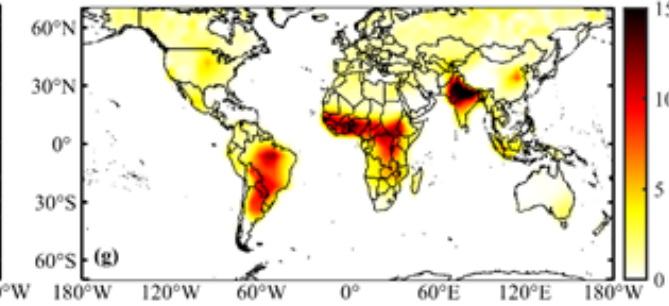
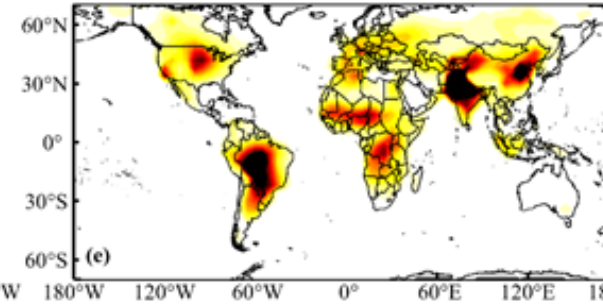
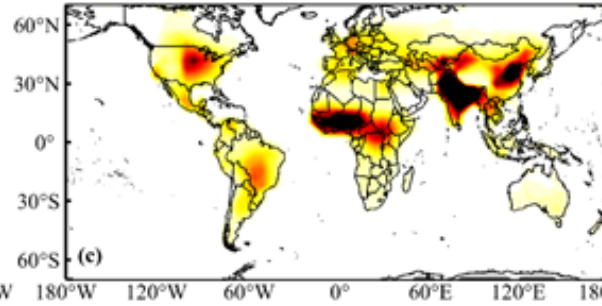
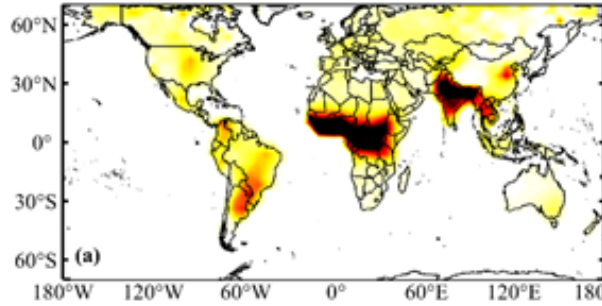
Mean ( $10^{15}$  molecules cm<sup>-2</sup>)

JFM

AMJ

JAS

OND

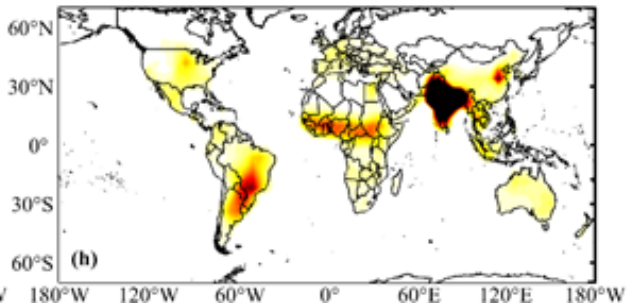
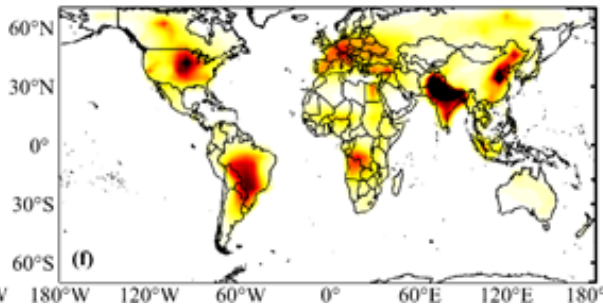
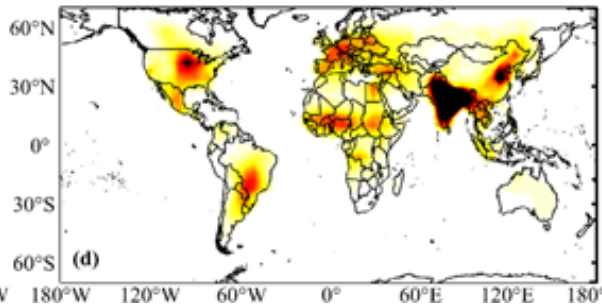
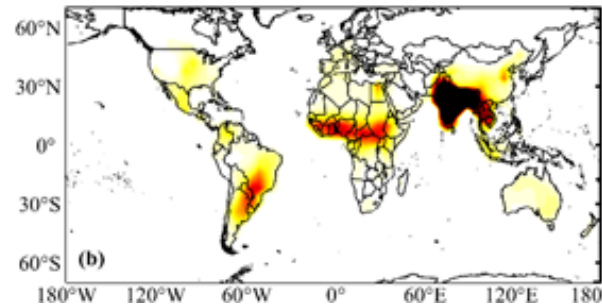


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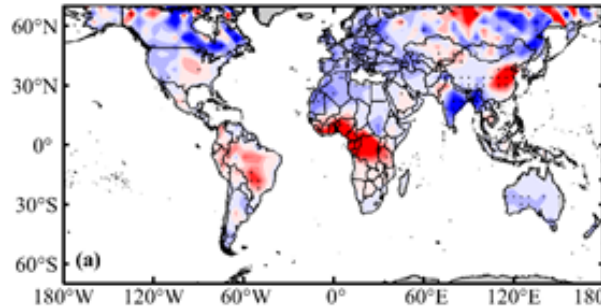




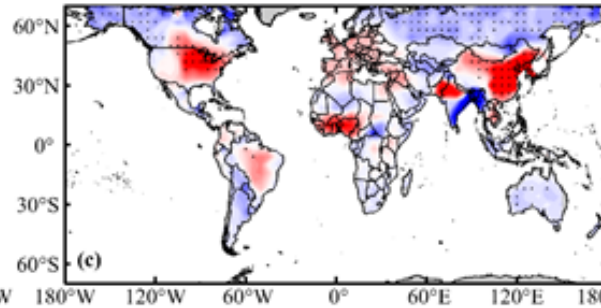
# NH<sub>3</sub> Seasonal Concentrations

Trend ( $10^{-6}$  Mol m<sup>-2</sup> yr<sup>-1</sup>)

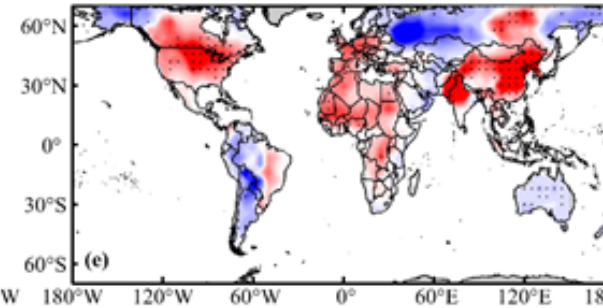
JFM



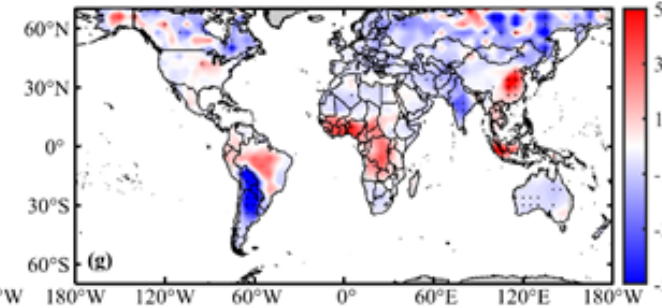
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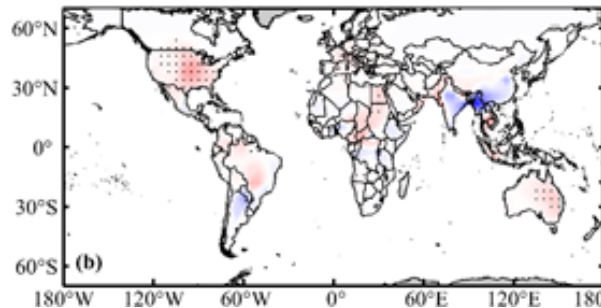
JAS



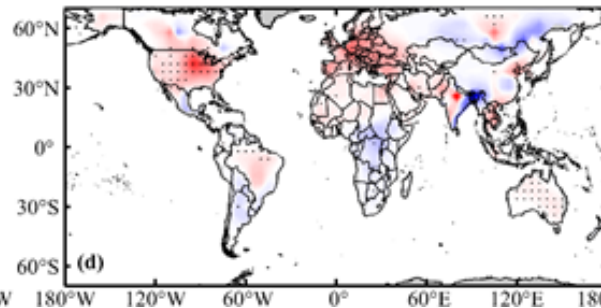
OND



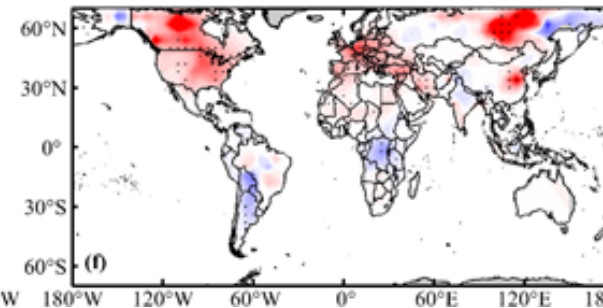
JFM



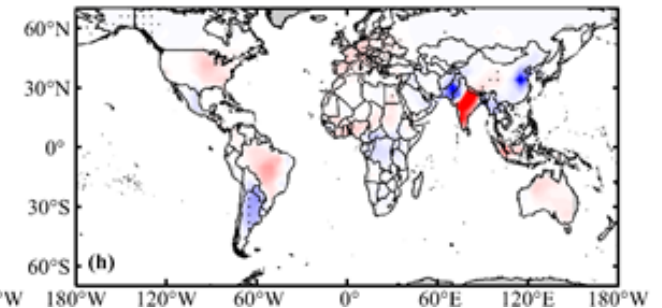
AMJ



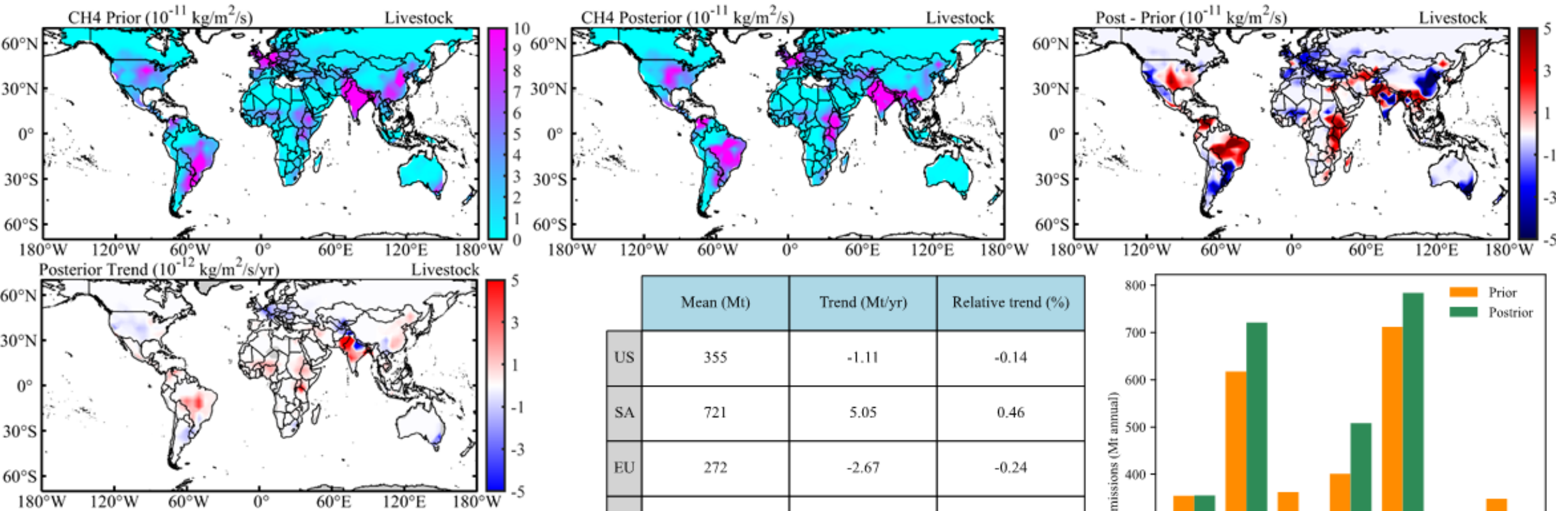
JAS



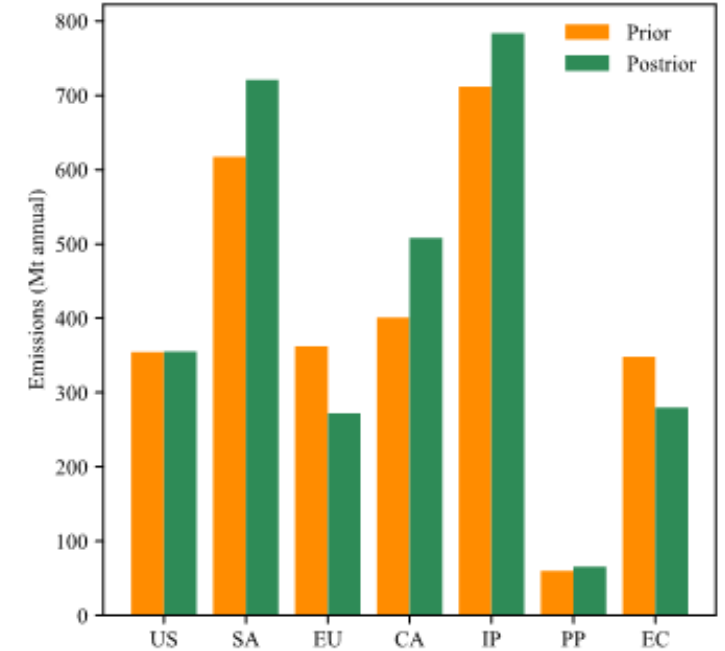
OND



# CH<sub>4</sub> livestock emissions

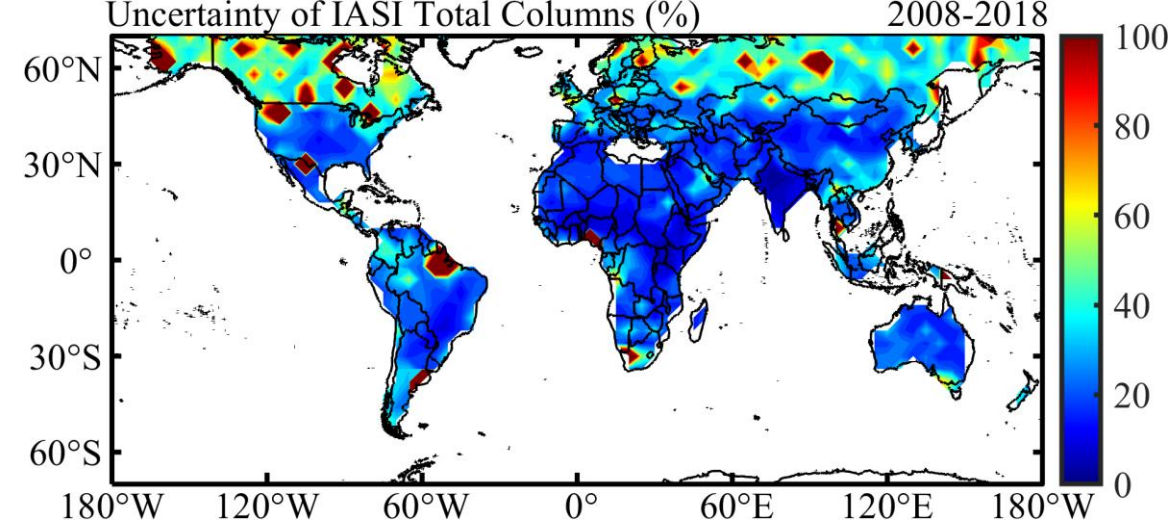


	Mean (Mt)	Trend (Mt/yr)	Relative trend (%)
US	355	-1.11	-0.14
SA	721	5.05	0.46
EU	272	-2.67	-0.24
CA	508	8.02	0.86
IP	783	10.68	0.71
PP	66	-1.65	-1.15
EC	280	1.37	0.49



# Uncertainty

- IASI total columns:
- Lifetime: ~40%
  - Deposition
  - Transportation (-)



Item	Bias	Period	Study area	Observation	Paper
NH4 wet deposition	1.2% (-9.8-11%)	2006	US	NADP/NTN	<a href="#">Zhang et al., 2012</a>
NH4 wet deposition	-23-25%	2006-2009	US	NADP/NTN	<a href="#">Zhu et al., 2013</a>
NH4 wet deposition	-1% (-25-12%)	2008-2012	China	EANET	<a href="#">Zhao et al., 2017</a>
dissolved inorganic N deposition	9% (-4-52%)	2000-2014	Southern China	Literature review	<a href="#">Xu et al., 2018</a>

# Uncertainty (Mt)

- Lifetime
- Transportation (+)/emission ratio
- Number of retrievals
- IASI column

$$\sigma_C = \sqrt{\frac{\sum(\sigma_i \times \Omega_i)^2}{n-1}}$$

- $\sigma_C$ : the total column error in each grid, [mol m<sup>-2</sup>]
- $\sigma_i$ : the ith retrieval relative error, [mol m<sup>-2</sup>]
- $\Omega_i$ : the ith retrieval total column, [%]

$$\sigma_{IASI} = \frac{\sigma_C \times M}{\tau_{mod}}$$

- $\sigma_{IASI}$ : the emission error in each grid associated with IASI total column error, [kg m<sup>-2</sup> s<sup>-1</sup>]
- $M$ : relative molecular mass, 17 [kg mol<sup>-1</sup>]

$$\overline{\sigma_{IASI}} = \sqrt{\sum(\sigma_{IASI_j} \times A_j \times t)^2}$$

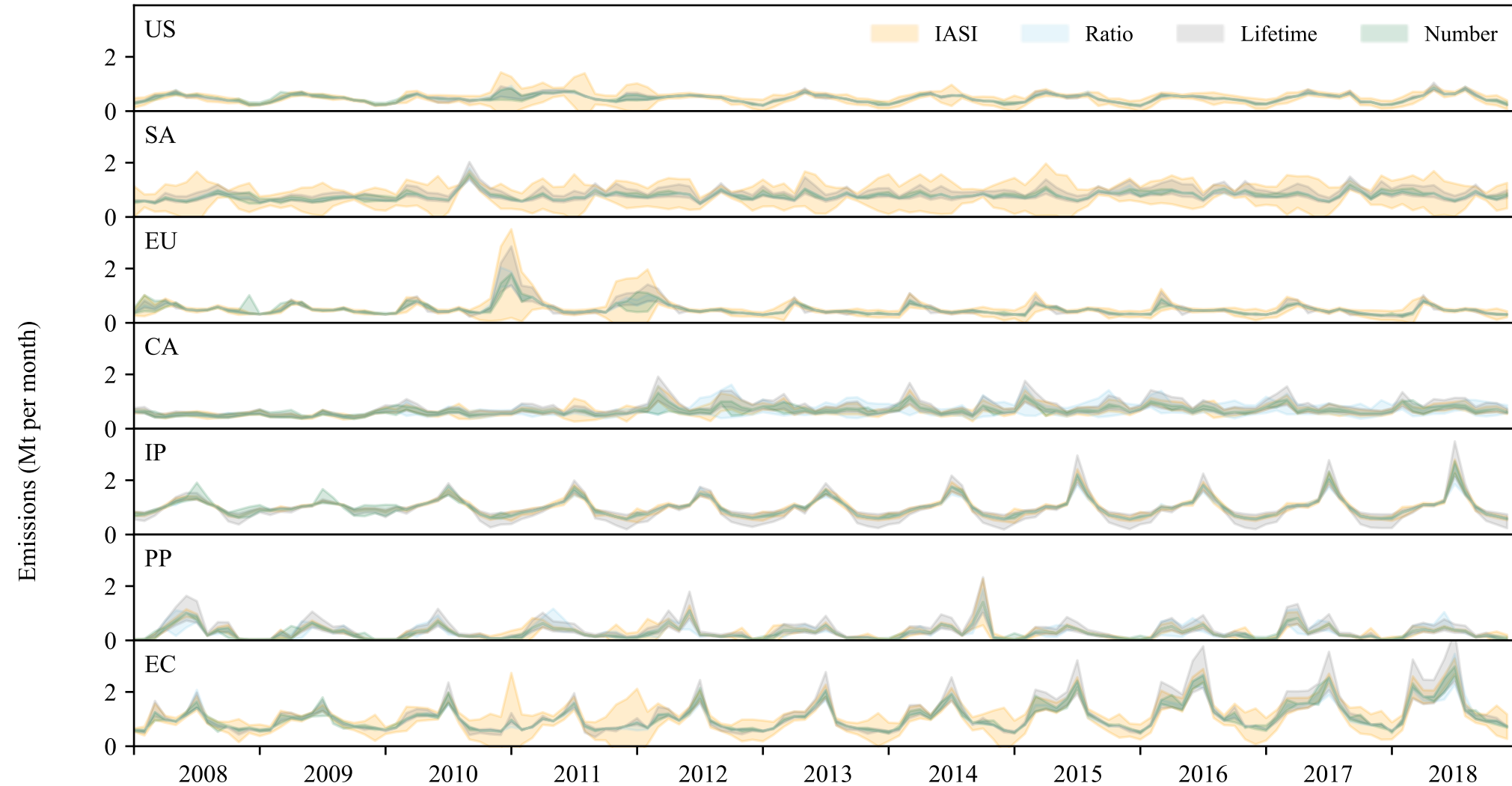
- $\overline{\sigma_{IASI}}$ : the domain mean error, associated with IASI total column error, [kg]
- $\sigma_{IASI_j}$ : the emission error in jth grid, [kg m<sup>-2</sup> s<sup>-1</sup>]
- $A_j$ : the area of jth grid, [m<sup>2</sup>]
- $t$ : the defined time period, [s]

Parameter perturbed	Averaged emission	Standard deviation
Initial: ratio < 1, n > 800	92	7.4
Transportation (-)	101	9.1
Transportation(+)/emission ratio < 0.2	87	5.2
Transportation(+)/emission < 5	102	17.4
Number of retrievals > 400	95	11.2
Number of retrievals > 1200	87	13.9
Lifetime -40%	107	11.0
Lifetime +40%	85	5.6
IASI column uncertainty	±15	±16.9



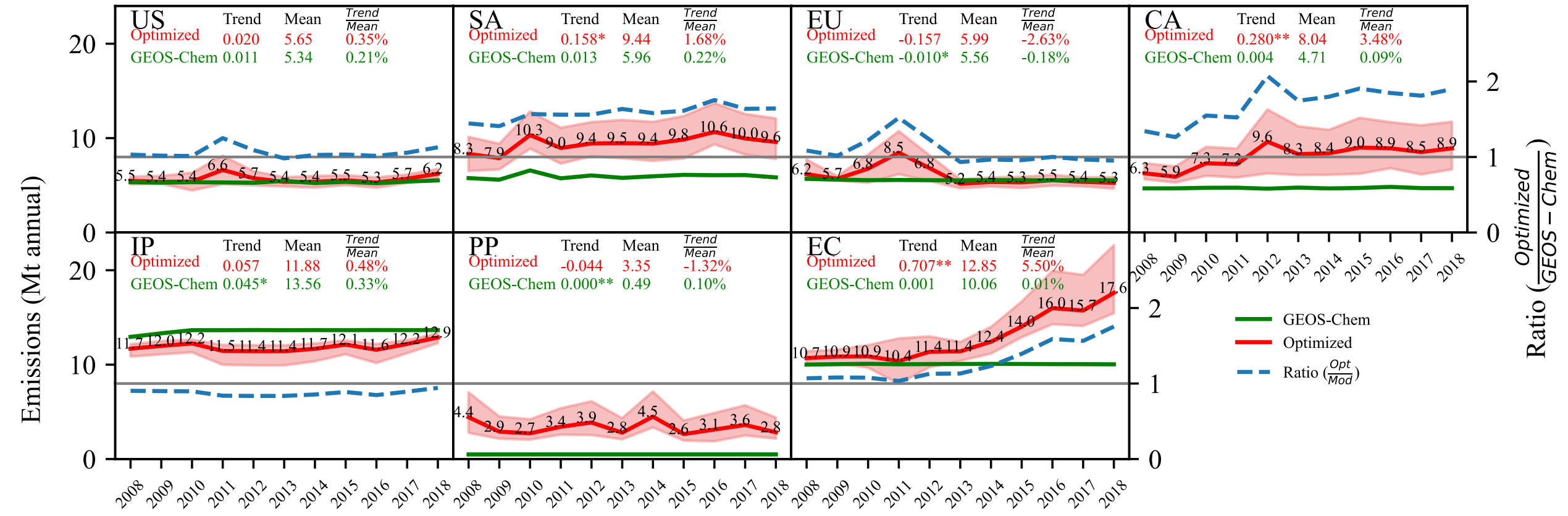
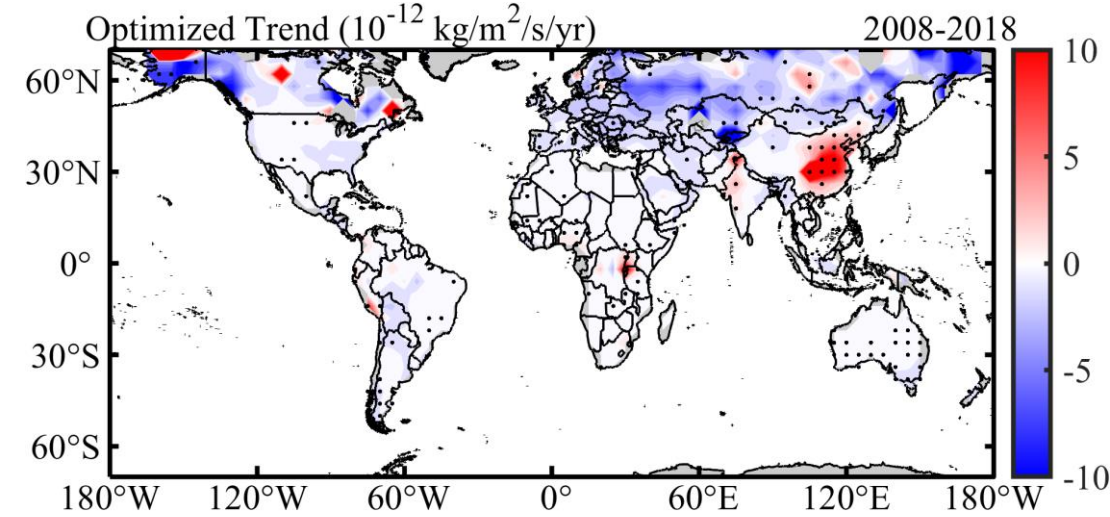
# Uncertainty

Uncertainty: Lifetime = 60-140%; Number = 400-1200; Tran/Emi = 0.2-5





# Optimized emissions trend ( $p < 0.05$ )



# Optimized versus GEOS-Chem

- Root Mean Square Error (RMSE)

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

- Mean Fractional Bias (MFB)

$$MFB = \frac{2}{N} \sum_{i=1}^N \frac{E_{Opt,i} - E_{Mod,i}}{E_{Opt,i} + E_{Mod,i}} \times 100\%$$

# IASI daily data

- Missing date (37 days):
  - 2008 (13 days): 1.17-18, 3.20-3.26, 12.10-11, 12.30-31
  - 2009 (3 days): 1.1, 1.23, 10.1
  - 2010 (5 days): 5.18, 8.31, 9.1-9.3
  - 2011 (2 days): 10.23-24
  - 2012 (0)
  - 2013 (2 days): 11.6-7
  - 2014 (7 days): 2.19-2.20, 9.9-9.13
  - 2015 (3 days): 4.10-4.12
  - 2016 (0)
  - 2017 (1 day): 6.7
  - 2018 (1 day): 12.31
- Filter
  - Cloud coverage: [0, 10%]
  - Skin temperature: > 263.15 K

# IASI emission flux calculations——fixed $\tau$

- $E = M/\tau$

- $E$ : emission fluxes, assumes stationarity and constant first-order loss terms
- $M$ : the total mass contained within the assumed box
- $\tau$ : The effective lifetime or residence time of  $\text{NH}_3$  within a given box

- $\tau_{mod} = \frac{(K_{\text{NH}_4^+/\text{NH}_3}^{mod} + 1)M_{mod}}{-\Delta M_{\text{NH}_3, \text{NH}_4^+}^{drydep, wetdep}}$

- $\tau'_{mod} = \frac{\tau_{mod}}{K_{\text{NH}_4^+/\text{NH}_3}^{mod} + 1} = \frac{M_{\text{NH}_3}}{-\Delta M_{\text{NH}_3, \text{NH}_4^+}^{drydep, wetdep}}$

- $\hat{E}_{obs} = \frac{(M_{obs} - M_{mod})}{\tau'_{mod}} + E_{mod}$

**Table SI1:  $\text{NH}_3$  lifetime estimates reported in the literature.**

REFERENCE	LIFETIME	COMMENT
Norman and Leck, 2005	Few hours Several days	Clean remote ocean Dust/Biomass plumes over ocean
Quinn et al., 1990	Order of hours	Central Pacific Ocean
Flechard and Fowler, 1998	1-2 hours	Scottish moorland site
Sutton, 1990	10 hours	Using dry deposition velocity by Duyzer et al. (1987)
Möller and Schieferdecker 1985	19 hours	Using dry deposition rates of Mészáros and Horváth (1984)
Hertel et al., 2012	24 hours	Simulations over Europe
Dentener and Crutzen, 1994	Order of hours	
Whitburn et al., 2016	17-23 hours	Fire plume
Hauglustaine et al., 2014	15 hours	Average global model

# total column concentration

- $\Omega = \sum_{i=1}^{47} c_i \times rho_i \times h_i \times k$ 
  - $\Omega$ : total column concentration, [mol/m<sup>2</sup>]
  - $c_i$ : 'IJ-AVG-\$\_NH3', mixing ratio for each level, [ppbv] to [v/v] (\*1E-9)
  - $rho_i$ : 'TIME-SER\_AIRDEN', air density for each level, [molecules/cm<sup>3</sup>]
  - $h_i$ : 'BXHGHT-\$\_BXHEIGHT', grid box height for each level, [m] to [cm] (\*100)
  - $k$ : 1/6.02214179E19, multiplication factor to convert [molecules/cm<sup>2</sup>] to [mol/m<sup>2</sup>]



# Regrid 180x360 to 46x72

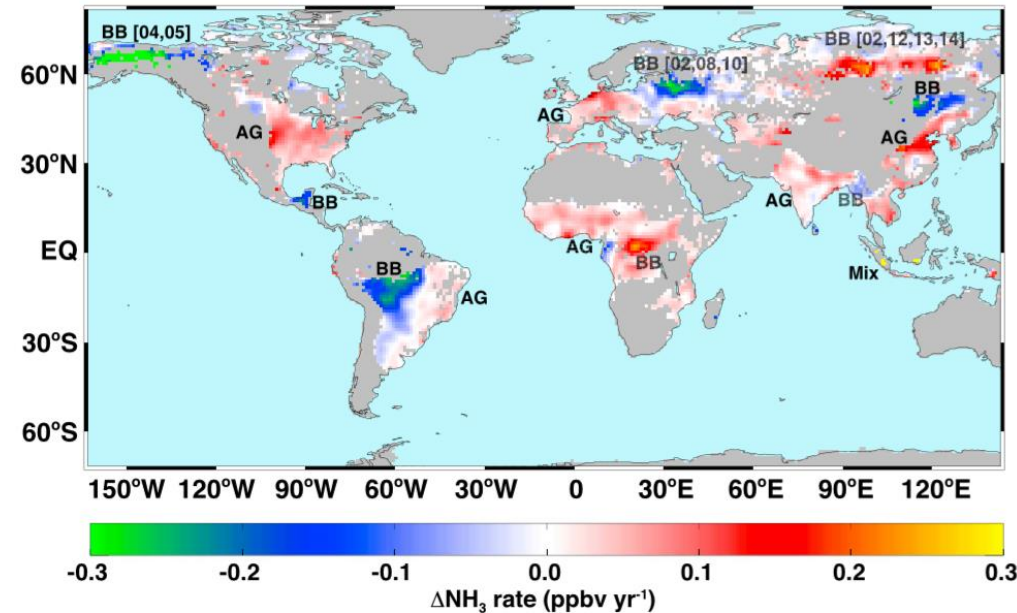
- Latitude: 46 degrees
  - 88°-90°: 2x5 to 1x1, 2 degrees
  - 0-88°: 4x5 to 1x1, 44 degrees
- Method:
  - Step1: mask ocean, set as NaN
  - Step2: calculate mean value in each upscaling grid

# emissions

- Anthropogenic
  - APEI: Historical Canadian emissions (1990-2014)
  - NEI2011\_MONMEAN: US emissions
  - MIX: Asian anthropogenic emissions
  - DICE\_Africa: emissions from inefficient combustion over Africa
  - **CEDS: Global anthropogenic emissions**
  - POET\_EOH: aldehydes and alcohols
  - TZOMPASOSA: global fossil fuel and biofuel emissions of C<sub>2</sub>H<sub>6</sub> for 2010
  - XIAO\_C<sub>3</sub>H<sub>8</sub>: C<sub>2</sub>H<sub>6</sub> and C<sub>3</sub>H<sub>8</sub>
  - AFCID: PM<sub>2.5</sub> dust emission
- Natural
  - GEIA\_NH<sub>3</sub>: 1990 (obsolete now)
  - SEABIRD\_DECAYING\_PLANTS: the oceanic emissions of acetaldehyde
  - NH<sub>3</sub>: the Arctic seabird
  - MEGAN: biogenic emissions
- Biomass burning
  - GFED4: biomass burning emissions
- Ship
  - CEDS\_SHIP
  - SHIP

# Increased atmospheric ammonia over the world's major agricultural areas detected from space

- provides evidence of substantial increases in atmospheric ammonia ( $\text{NH}_3$ ) concentrations (14year) over several of the world's major agricultural regions
- The rate of change of  $\text{NH}_3$  volume mixing ratio (VMR) in parts-per-billion by volume (ppbv) per year computed
  - BB: biomass burning
  - AG: agricultural



(Warner et al, 2017)

End