

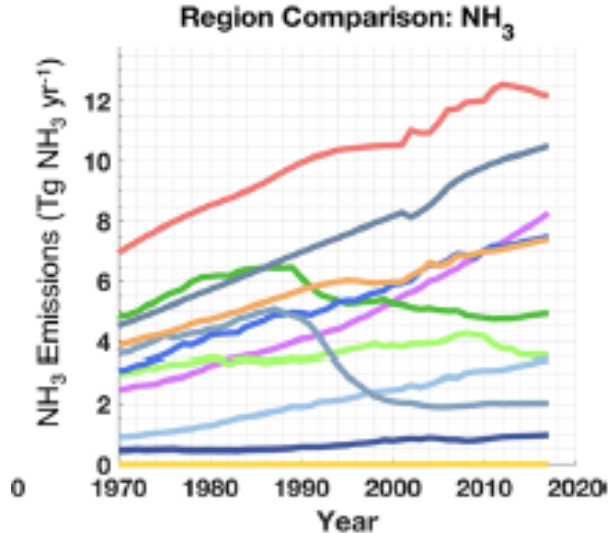
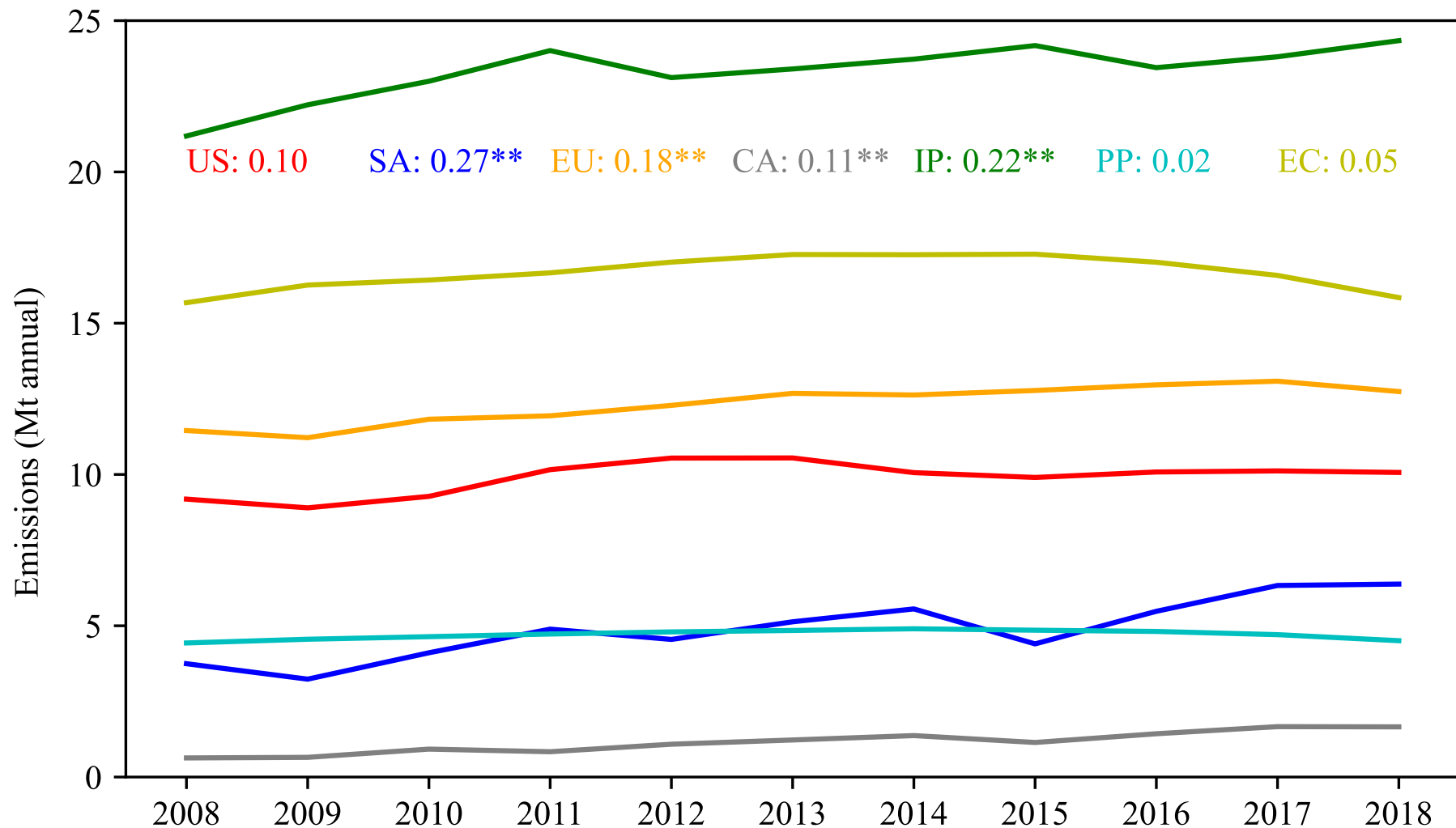
# Optimized ammonia emissions 18

IASI data and GEOS-Chem simulation

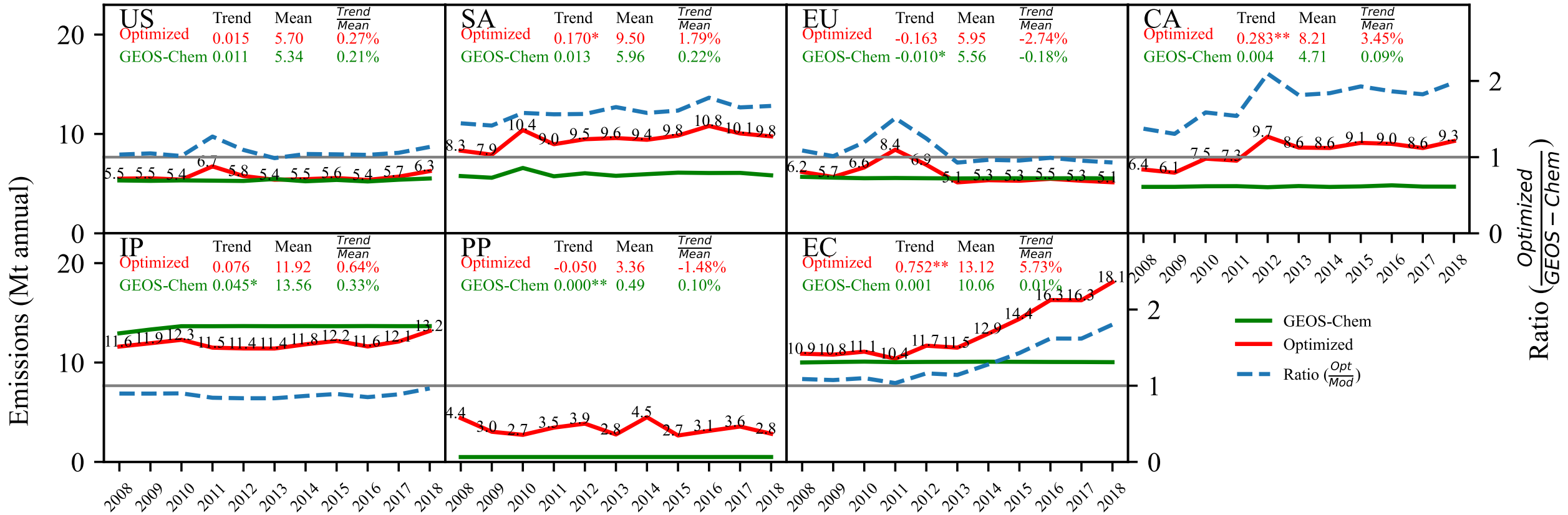
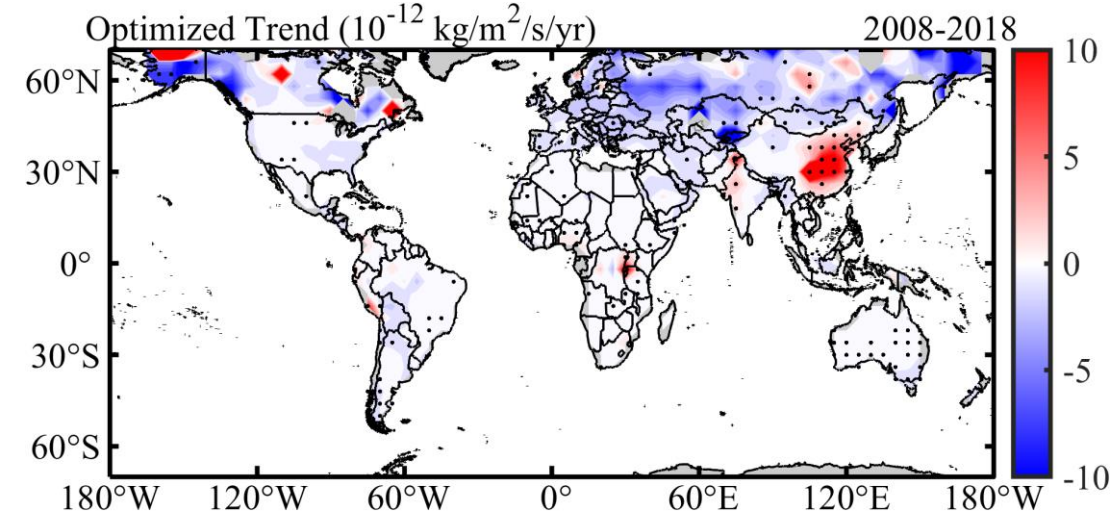
2021.5

- Accomplished:
  - 1. Diagram and Flowchart
  - 2. IASI uncertainty for emission
  - 3. FAO data
- Ammonia Data:
  - IASI total columns: Reanalyzed IASI/Metop-A, V3.0.0R
    - Daily, L2,  $1^{\circ} \times 1^{\circ}$  (2008-2018)
  - GEOS-Chem simulation,  $4^{\circ} \times 5^{\circ}$ , daily, 2008-2018, V12.9.3
    - Total column concentration
    - Total column transport/deposition rate of change
    - Emissions
- meteorological input data
  - ECMWF ERA5 skin temperature,  $0.25^{\circ} \times 0.25^{\circ}$ 
    - hourly data on single levels (2008-2018), 9:00/10:00
- Ongoing:
  - 1. regional details

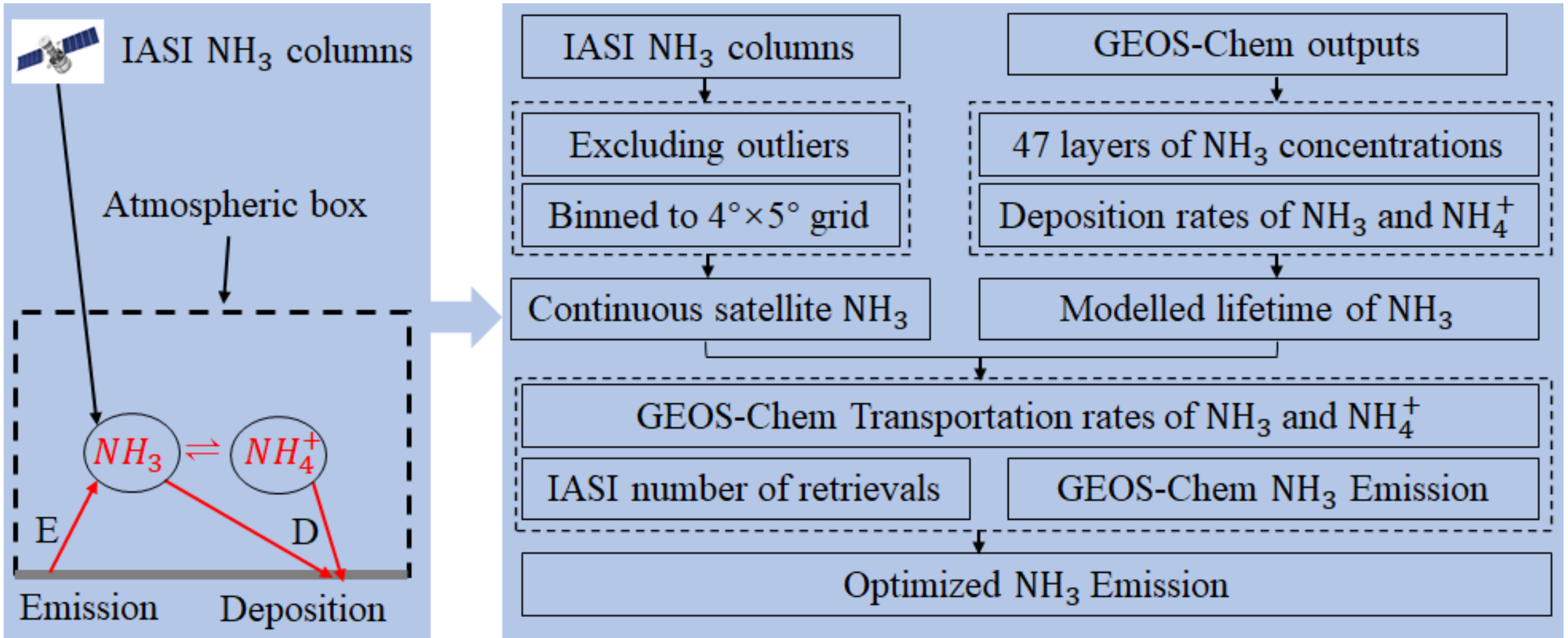
# FAOSTAT Agricultural N Use



# Optimized emissions trend ( $p < 0.05$ )

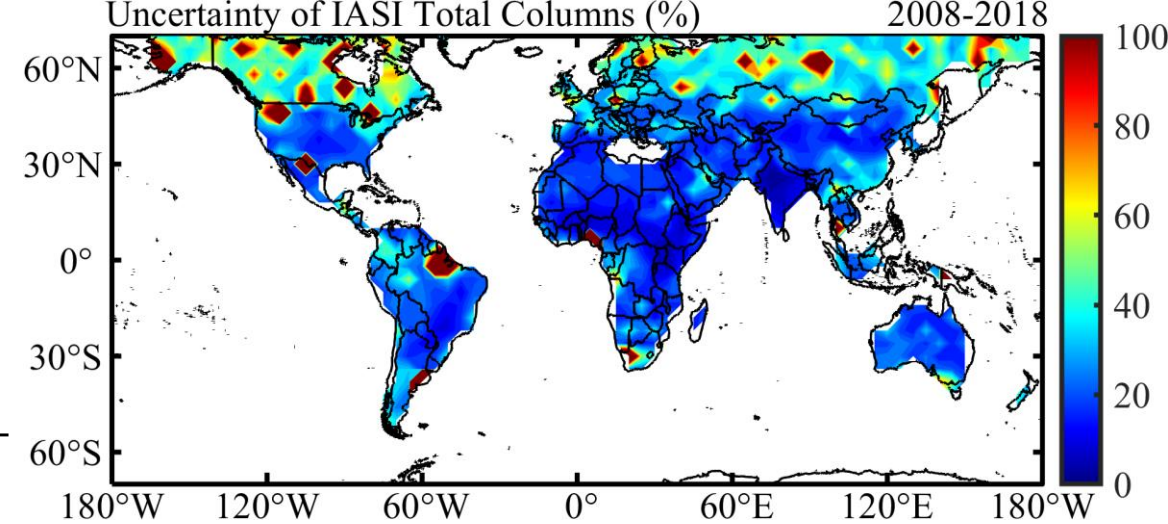


# Diagram and Flowchart of using IASI data and GEOS-Chem simulation to optimized $\text{NH}_3$ emission.



# Uncertainty

- IASI total columns: •  $Error = \frac{relative\_error}{\sqrt{n}}$
- Lifetime: **~20%**
  - 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

Parameter perturbed	Averaged emission	Standard deviation
Initial: ratio < 1, n > 30	94	7.4
Transportation (-)	101	9.1
Transportation(+)/emission ratio < 0.2	87	5.2
Transportation(+)/emission < 5	102	17.4
Number of retrievals > 10	104	11.2
Number of retrievals > 50	85	13.9
Lifetime -30%	105	11.0
Lifetime -20%	100	9.5
Lifetime -10%	96	8.4
Lifetime +10%	91	6.7
Lifetime +20%	89	6.1
Lifetime +30%	88	5.6
IASI column lower	77	19
IASI column upper	111	26

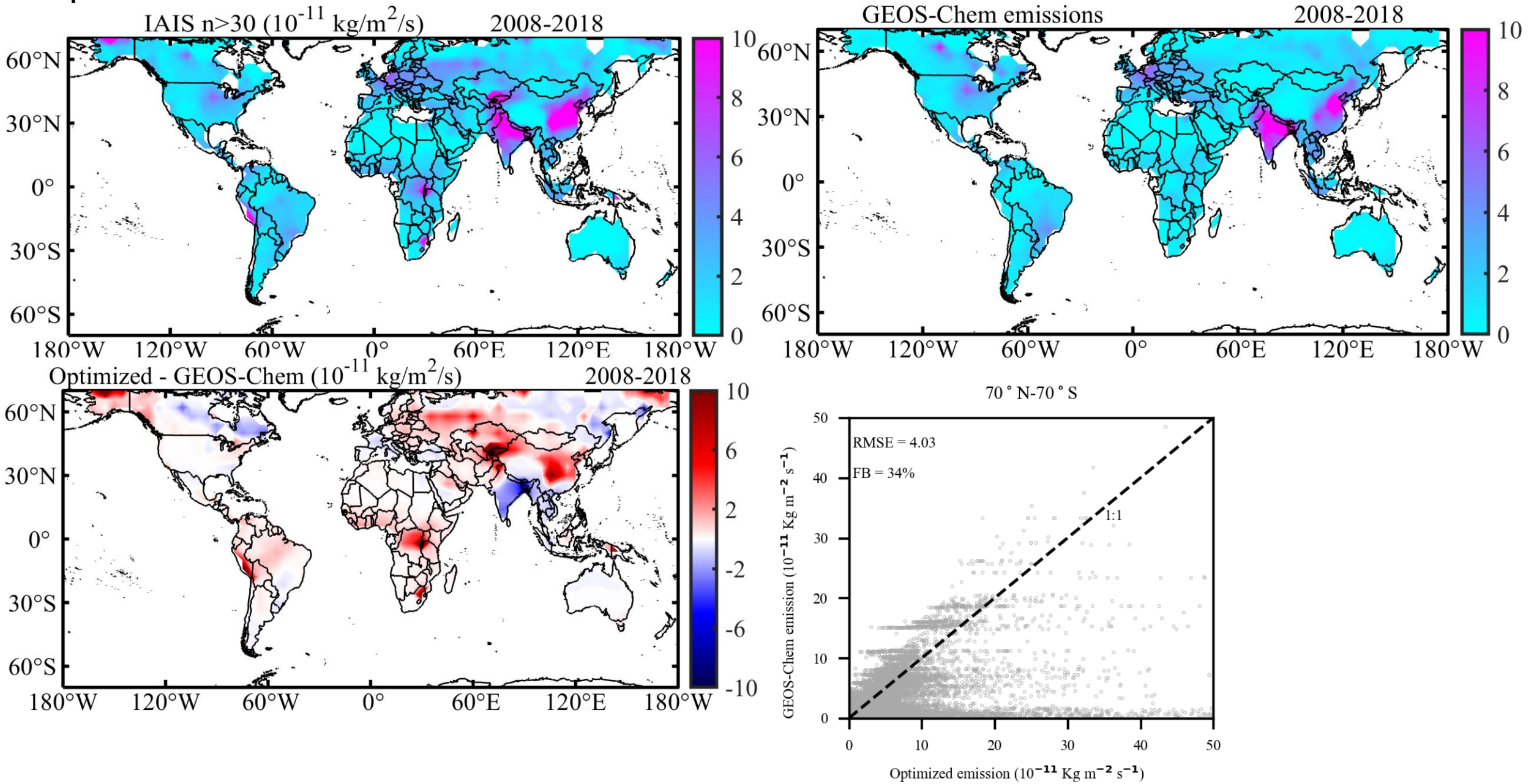
# Structure

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

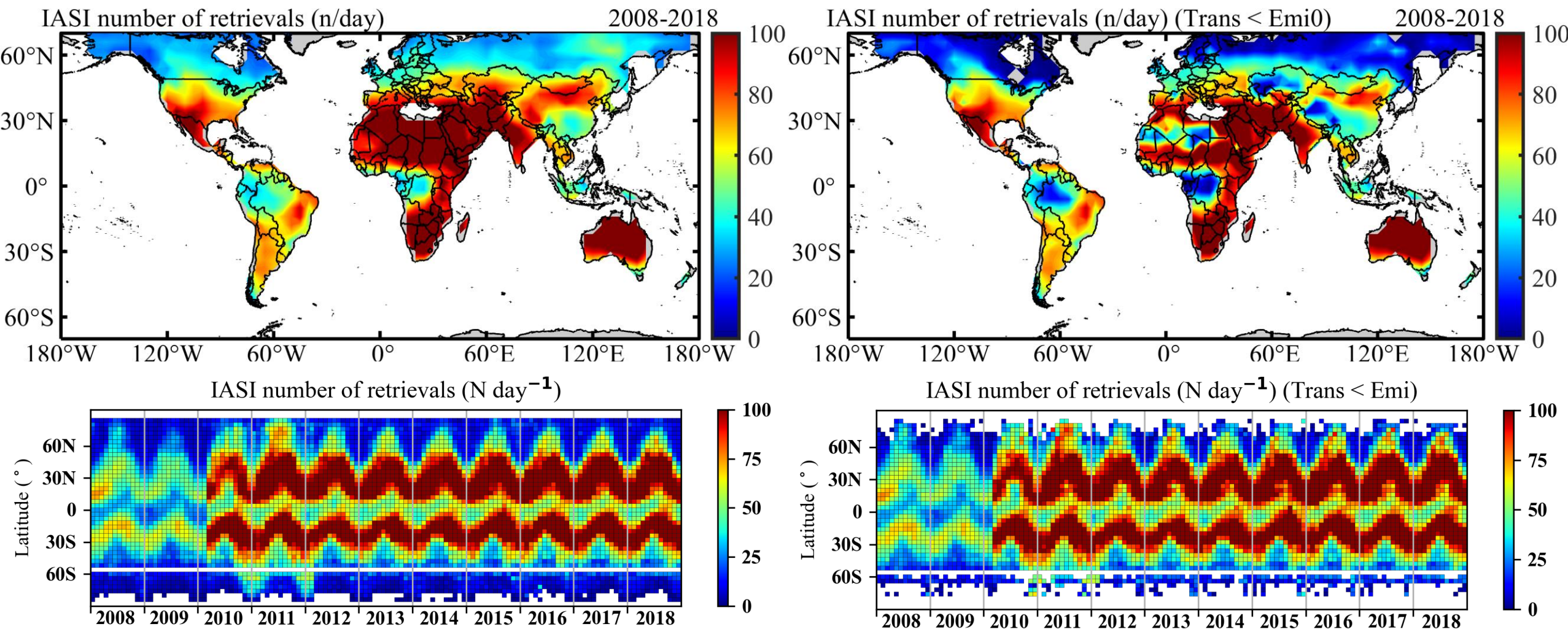




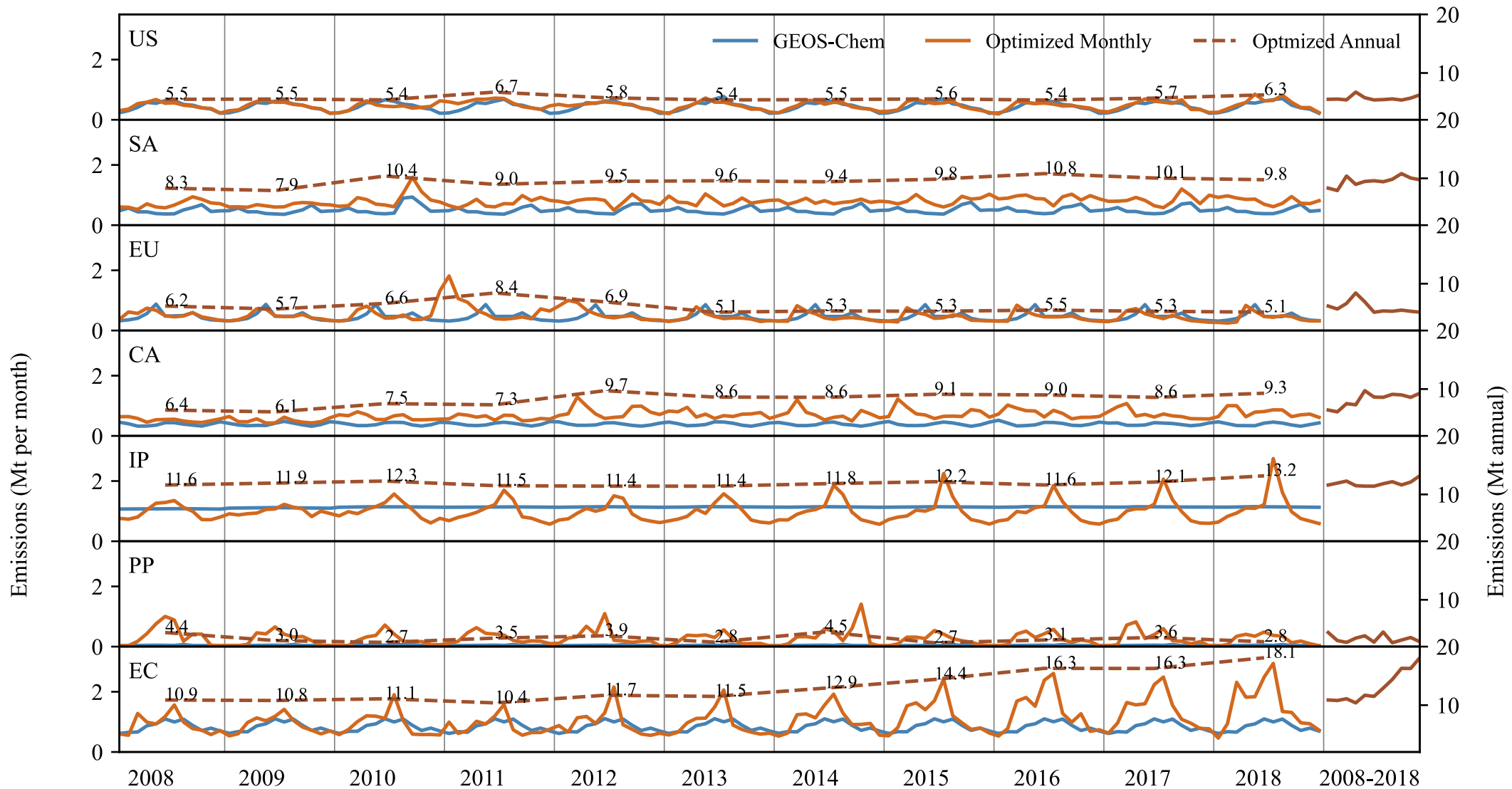
# Optimized versus GEOS-Chem



# Number of retrievals

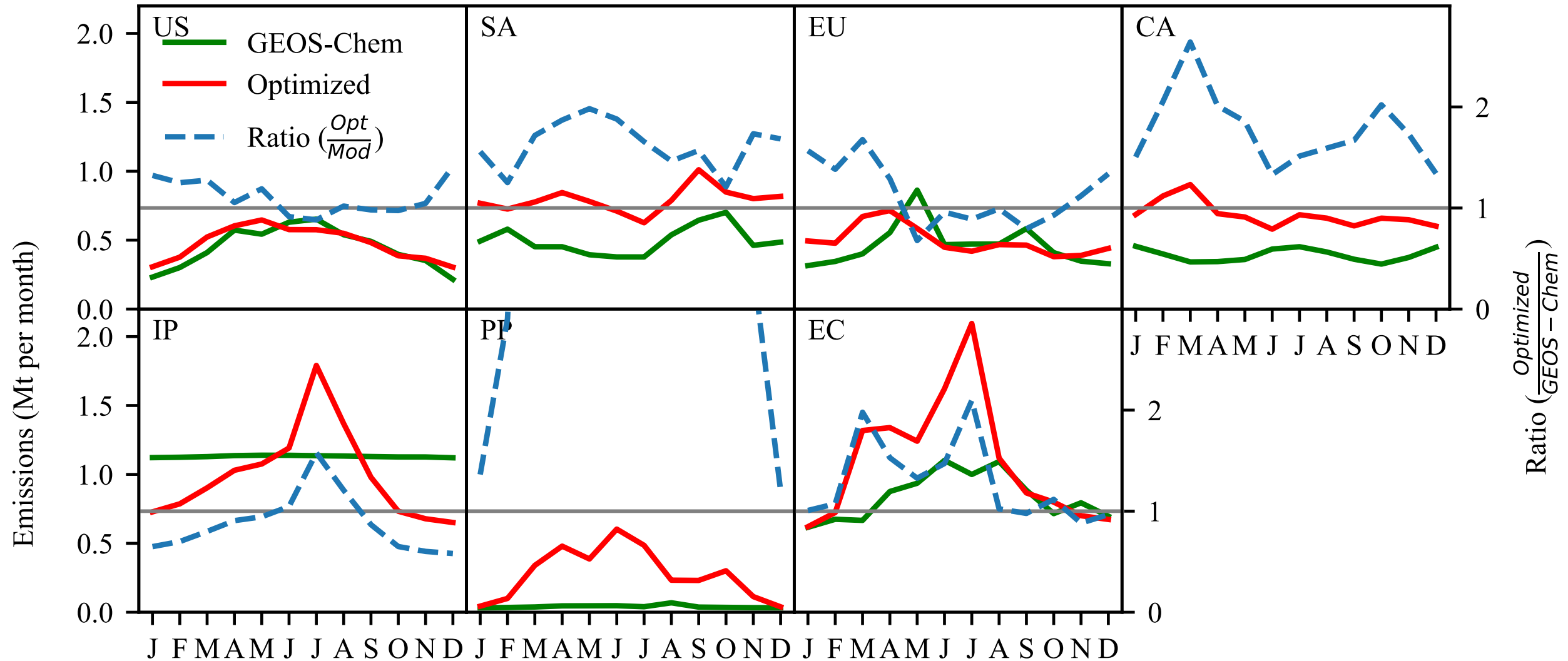


# Optimized emissions timeseries

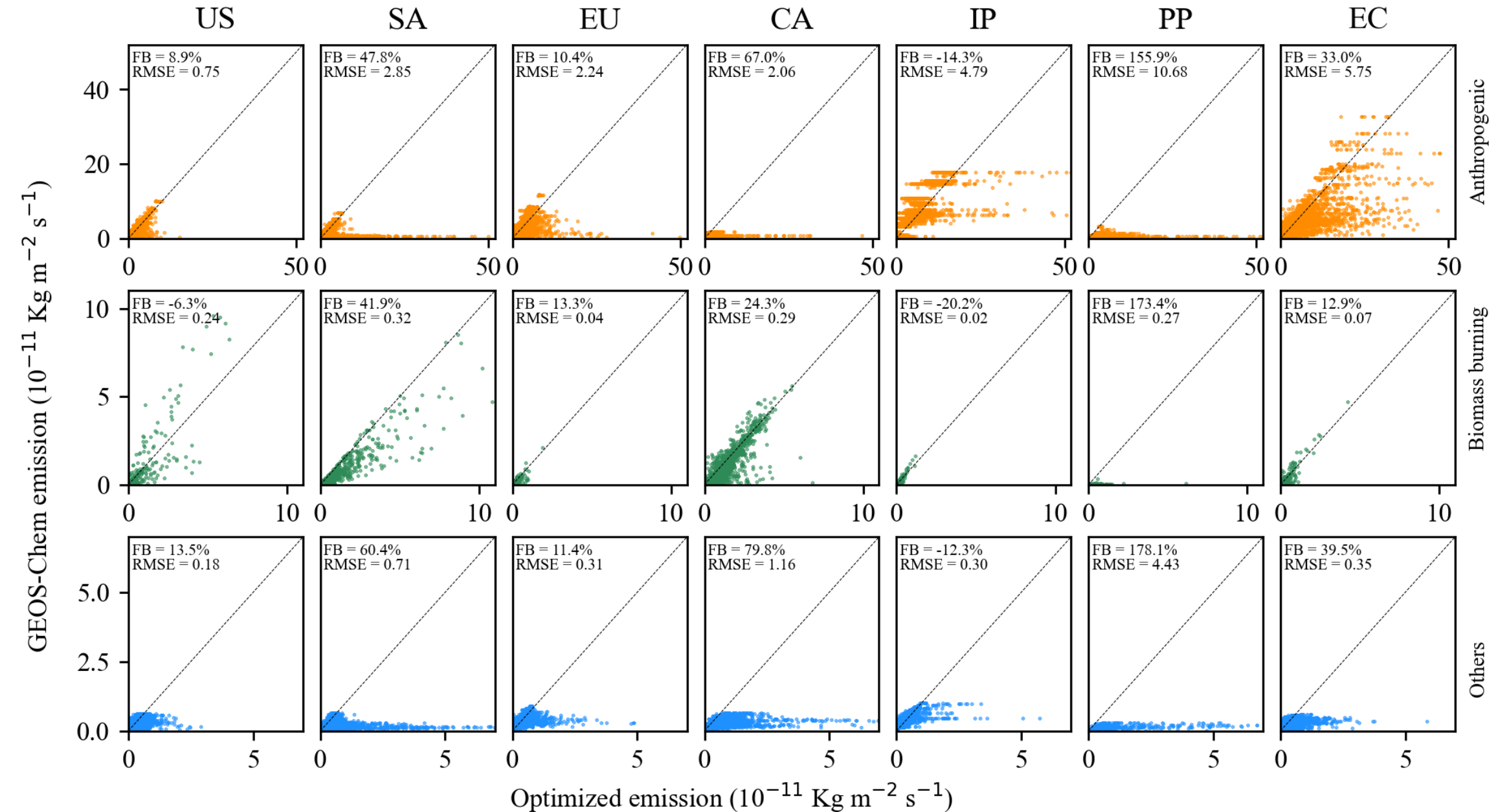




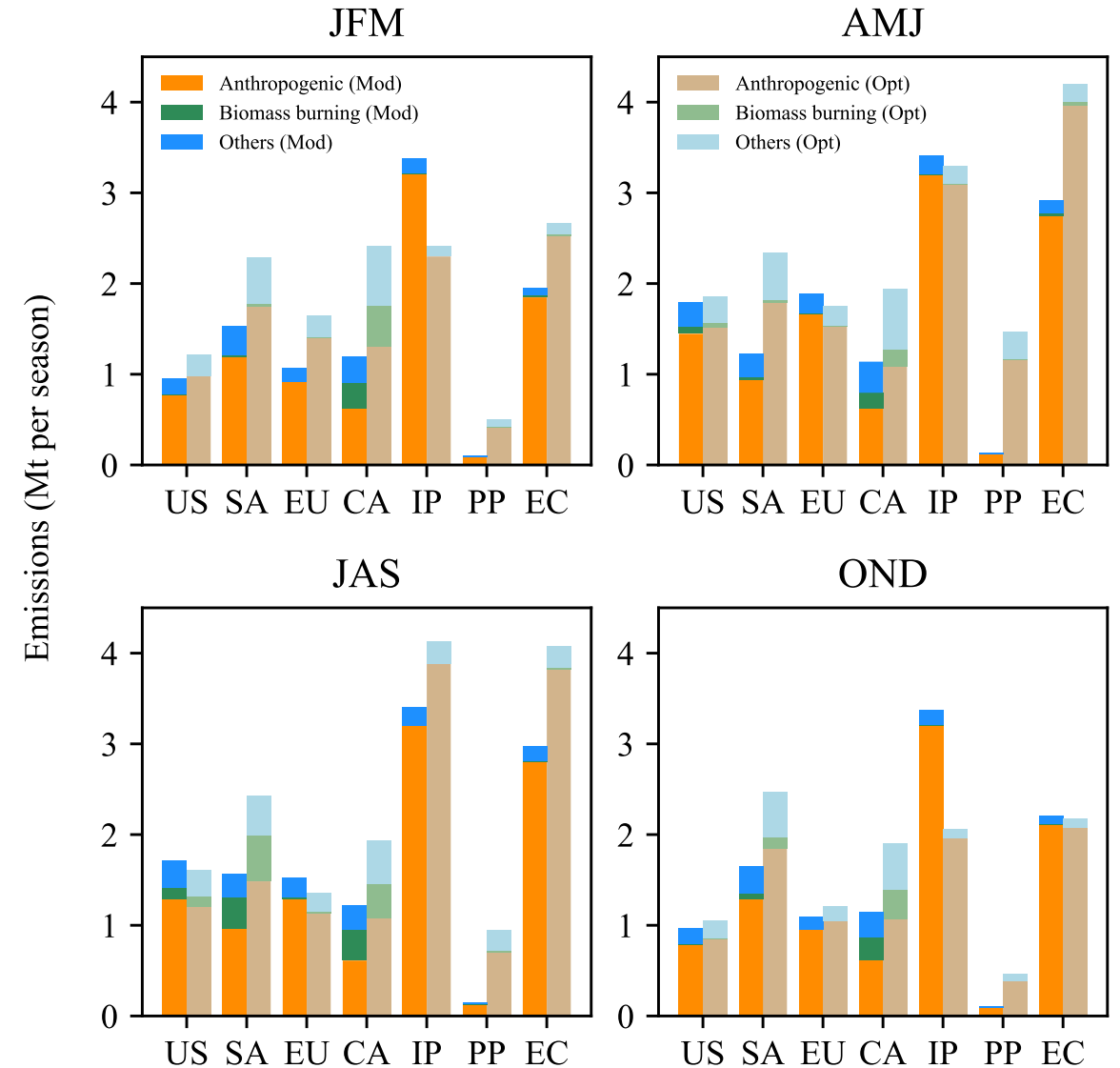
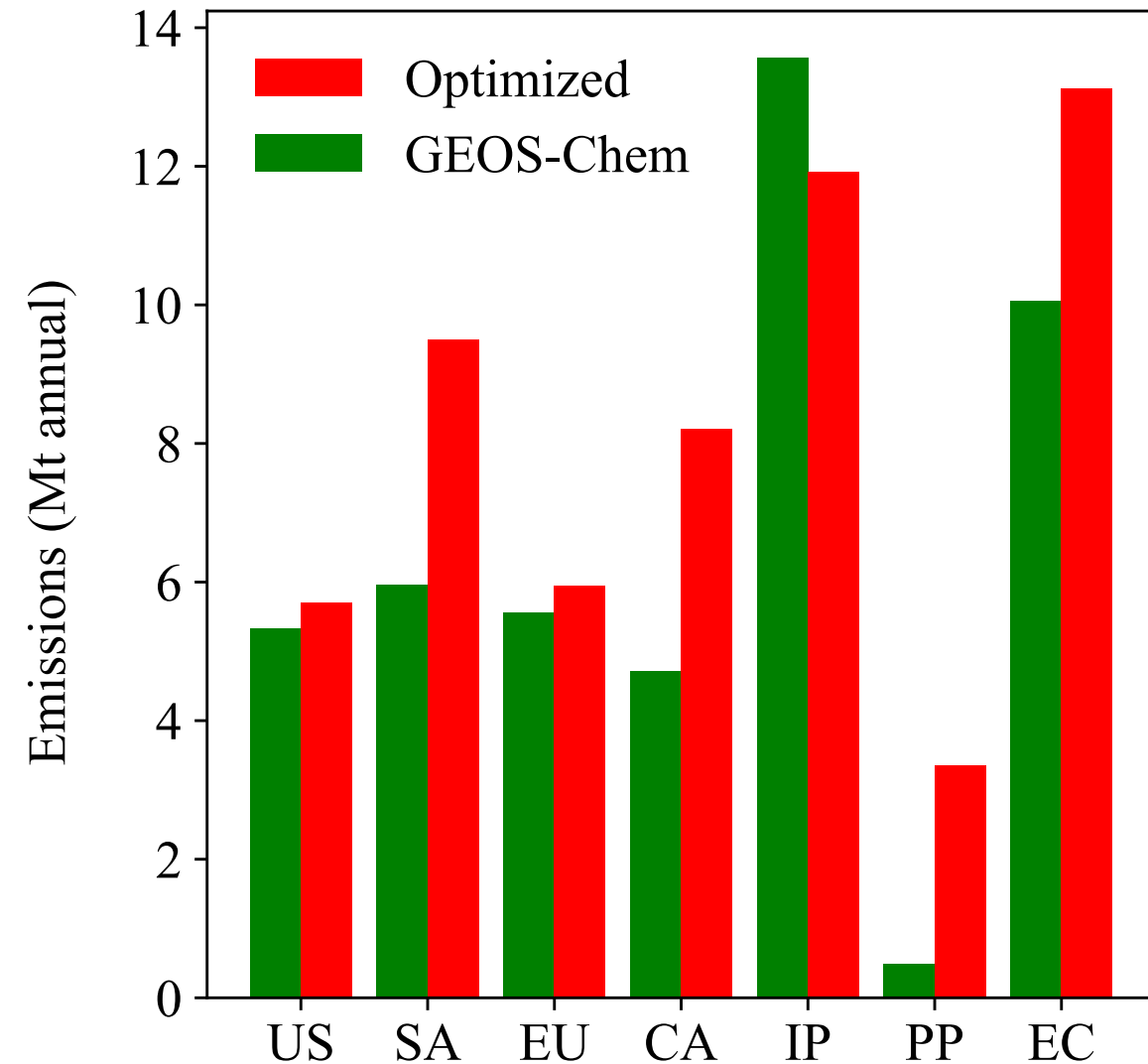
# Optimized emissions monthly variations



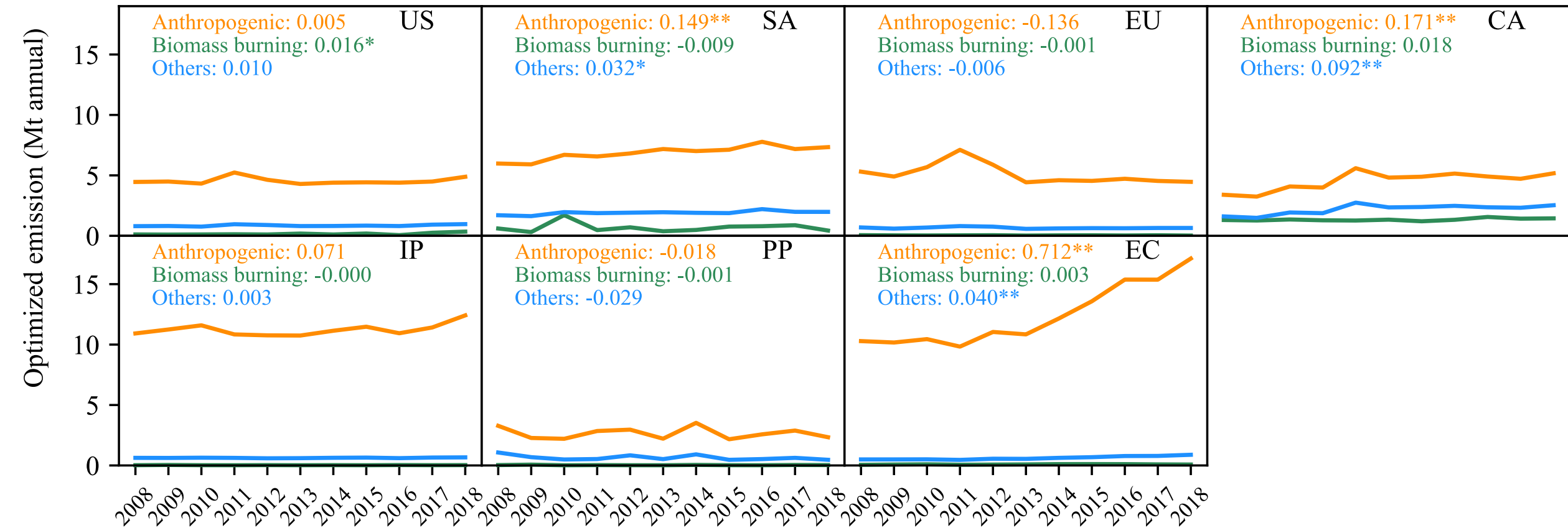
# Optimized versus GEOS-Chem by sectors



# Optimized emissions by sectors

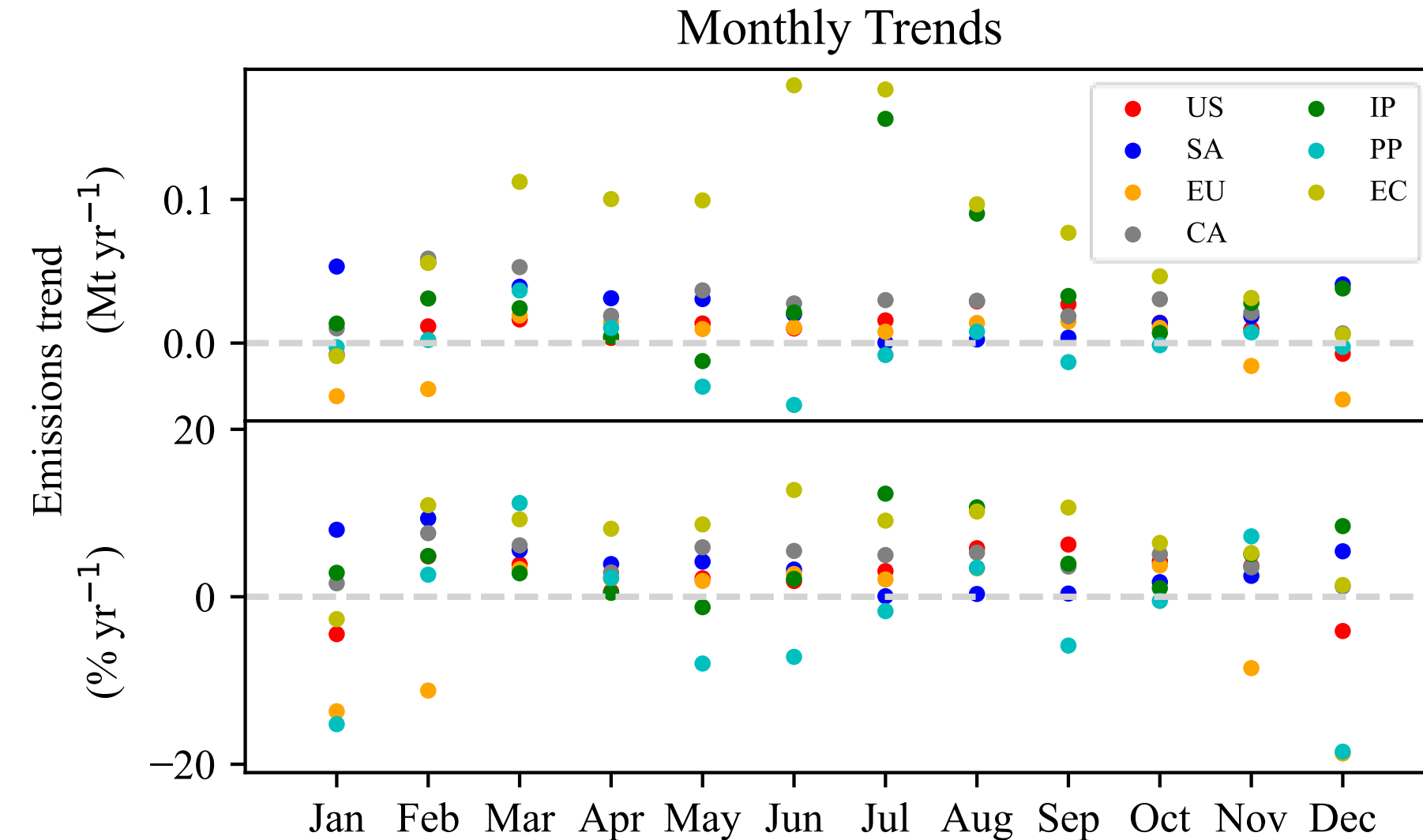


# Optimized emissions trend by sectors





# Optimized emissions monthly trend



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

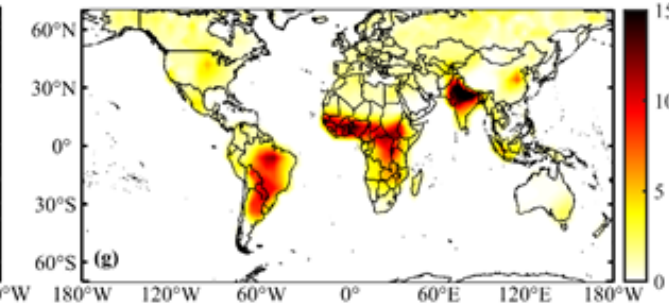
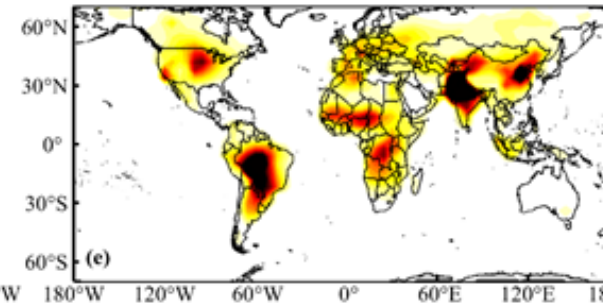
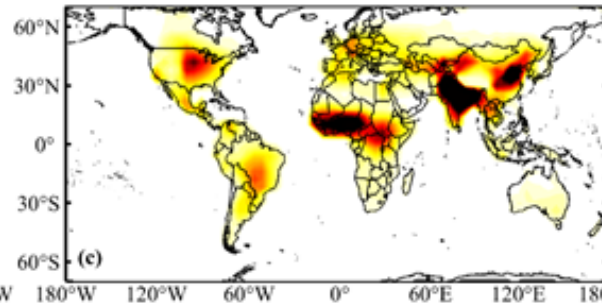
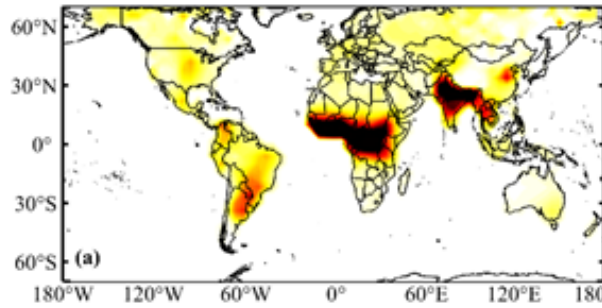
Mean ( $10^{15}$  molecules cm<sup>-2</sup>)

JFM

AMJ

JAS

OND

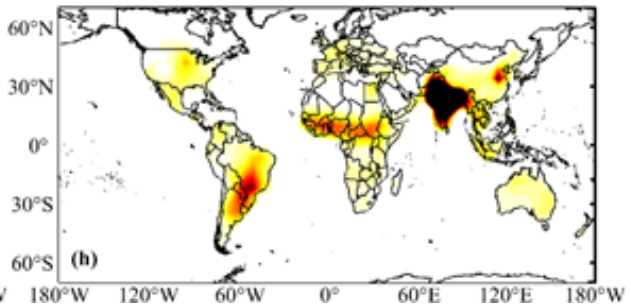
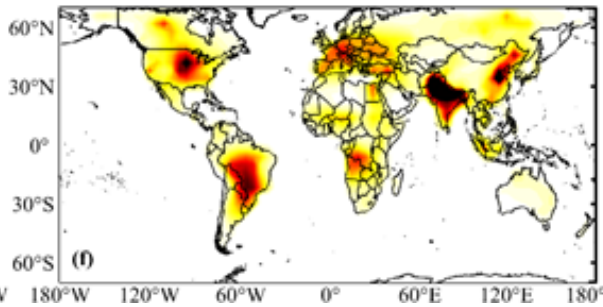
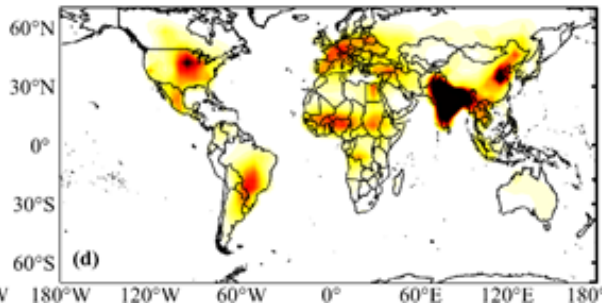
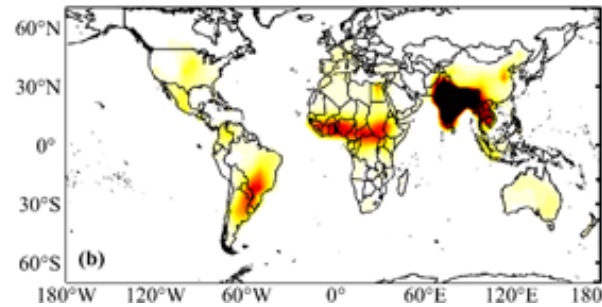


JFM

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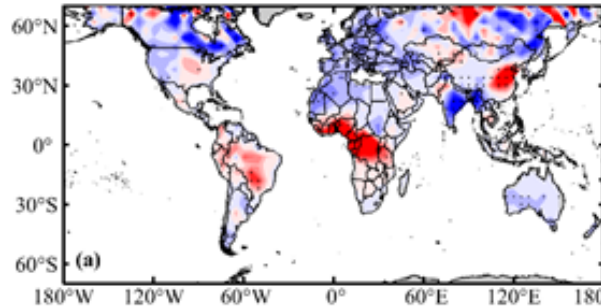
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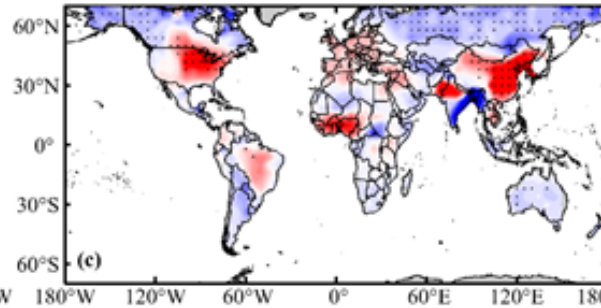
# NH<sub>3</sub> Seasonal Concentrations

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

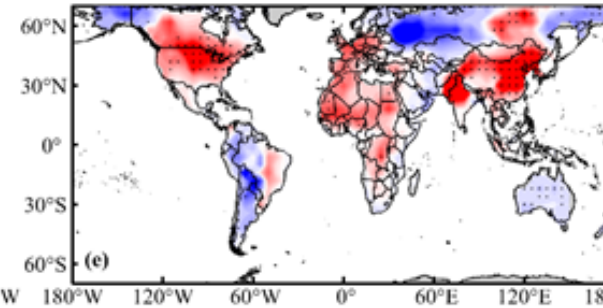
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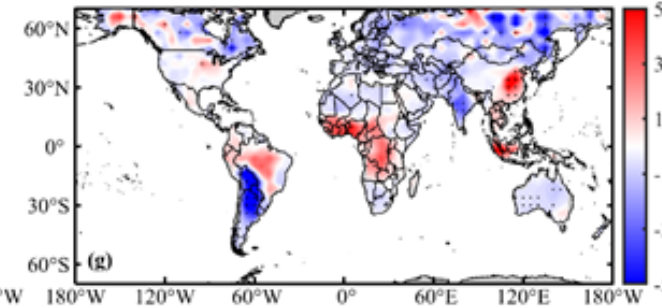
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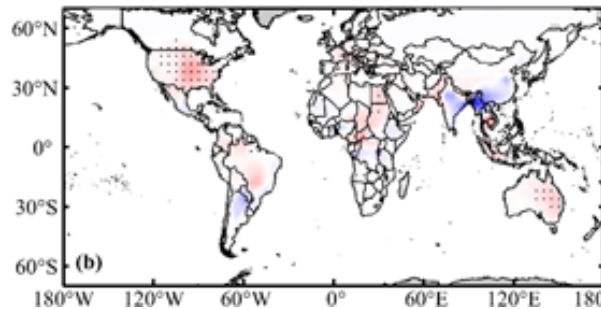
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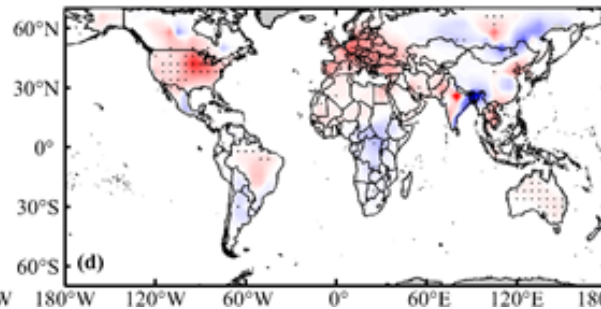
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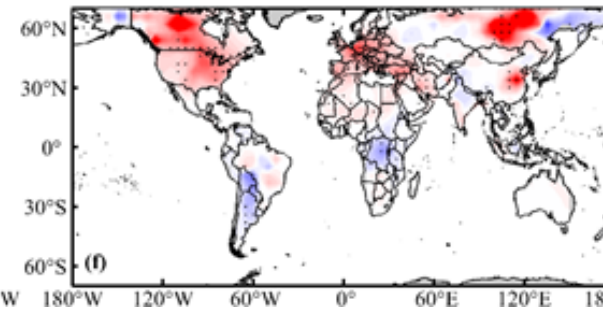
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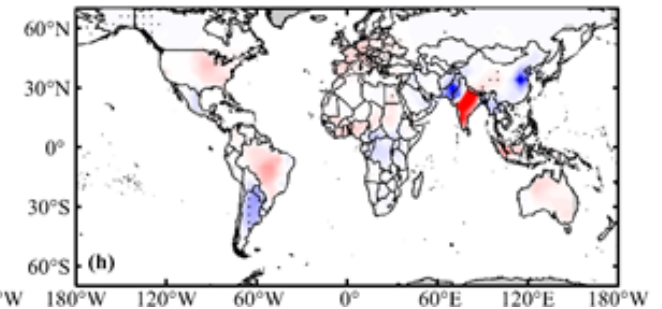
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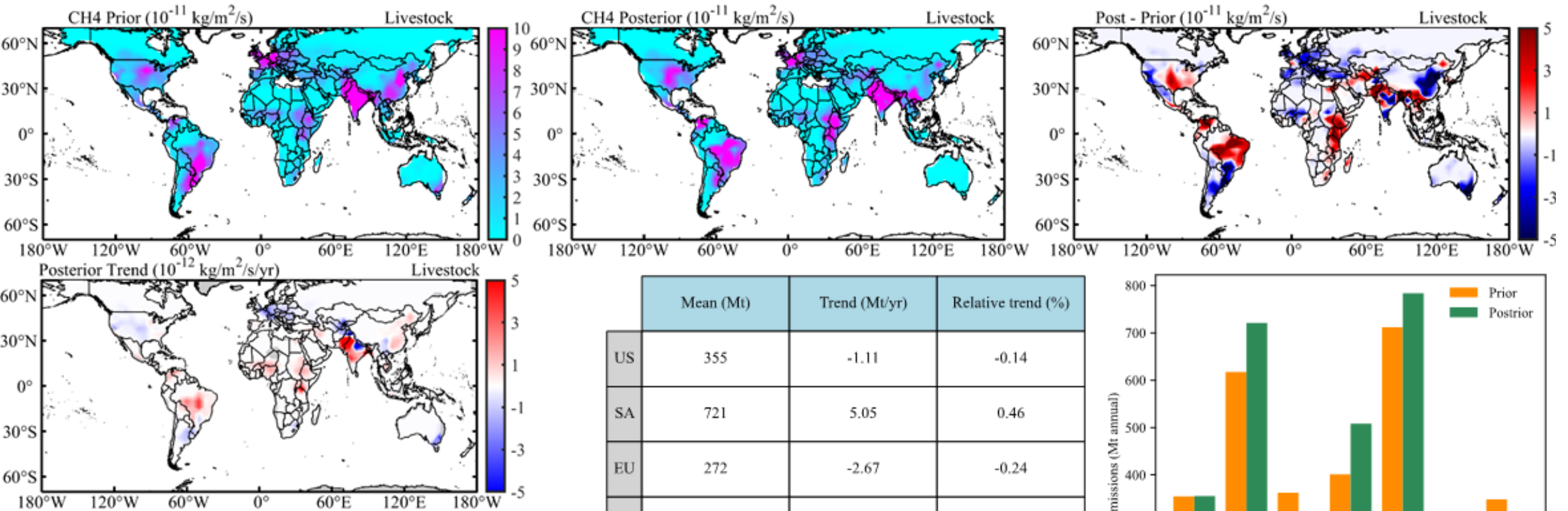
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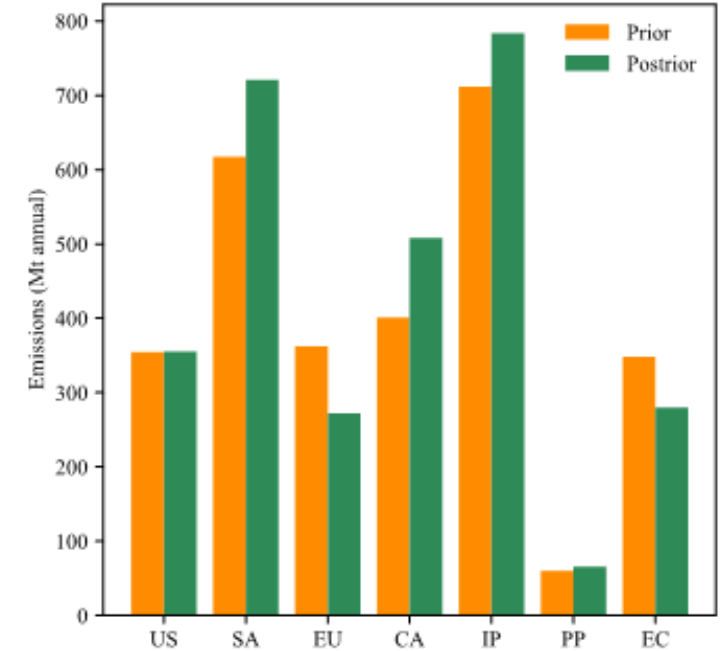
OND



# 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



# 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

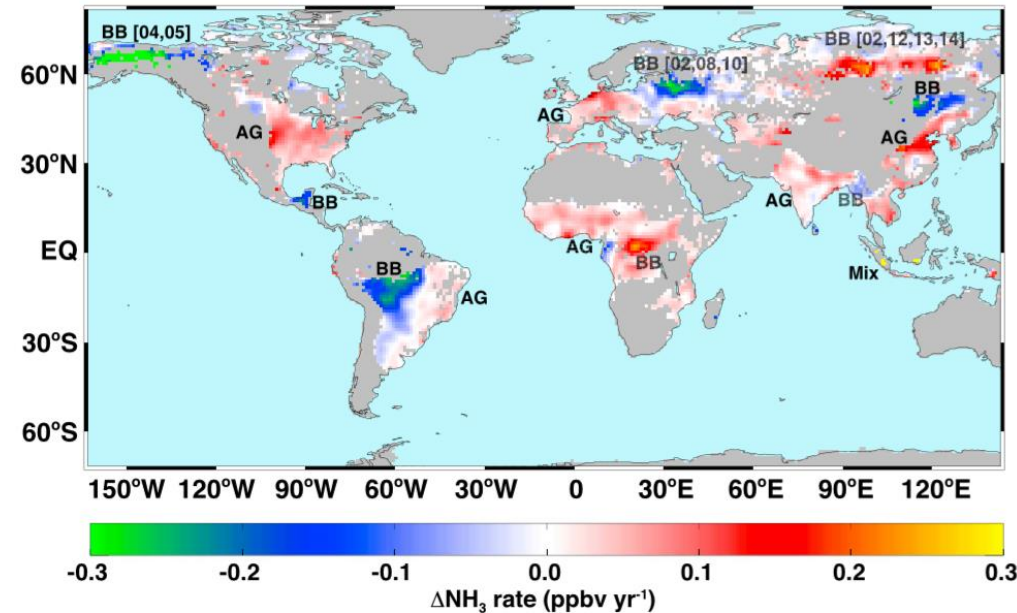
- Latitude: 46 degrees
  - 88°-90°: 2x5 to 1x1, 2 degrees
  - 0-88°: 4x5 to 1x1, 44 degrees
- Method:
  - Step1: mask ocean, set as NaN
  - Step2: calculate mean value in each upscaling grid

# 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