Top-down ammonia emissions 22

IASI data and GEOS-Chem simulation

2021.10

- Method:
  - 1. IASI observations
  - 2. GEOS-Chem simulations
  - 3. NH3 emission flux estimations
  - 4. Uncertainty and sensitivity analysis
- Results and discussion:
  - 1. Observed and simulated NH3 concentrations
  - 2. Comparison of top-down NH3 emission estimations
    - compare lifetime of NH3 and NHx and their emissions
    - compare concentration based on updated emission over 2008 and 2018
  - 3. Global NH3 emissions distributions
  - 4. Regional NH3 emissions trends
    - Find evidence of biomass burning in northern high latitudes
  - 5. Validation against observations
  - 6. Relation to methane emissions

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

[a]The number of retrievals larger than 800, and the transport dominates over emissions or depositions when exclude grids.
[b-c]The lifetime is 50% and 200% in Eq. (1), respectively.
[d-e]The number of retrievals larger than 400 and 800 when exclude grids, respectively.
[f-g]The threshold used in the budget analysis is 0.2 and 5, respectively.

$$\hat{E}_{NH_3} = E_{NH_3,mod} + \frac{C_{NH_3,obs} - C_{NH_3,mod}}{\tau_{NH_x,mod}},$$

$$\tau_{NH_3,mod} = \frac{C_{NH_3,mod}}{D_{NH_3,mod} + Chem_{NH_3,mod}}.$$

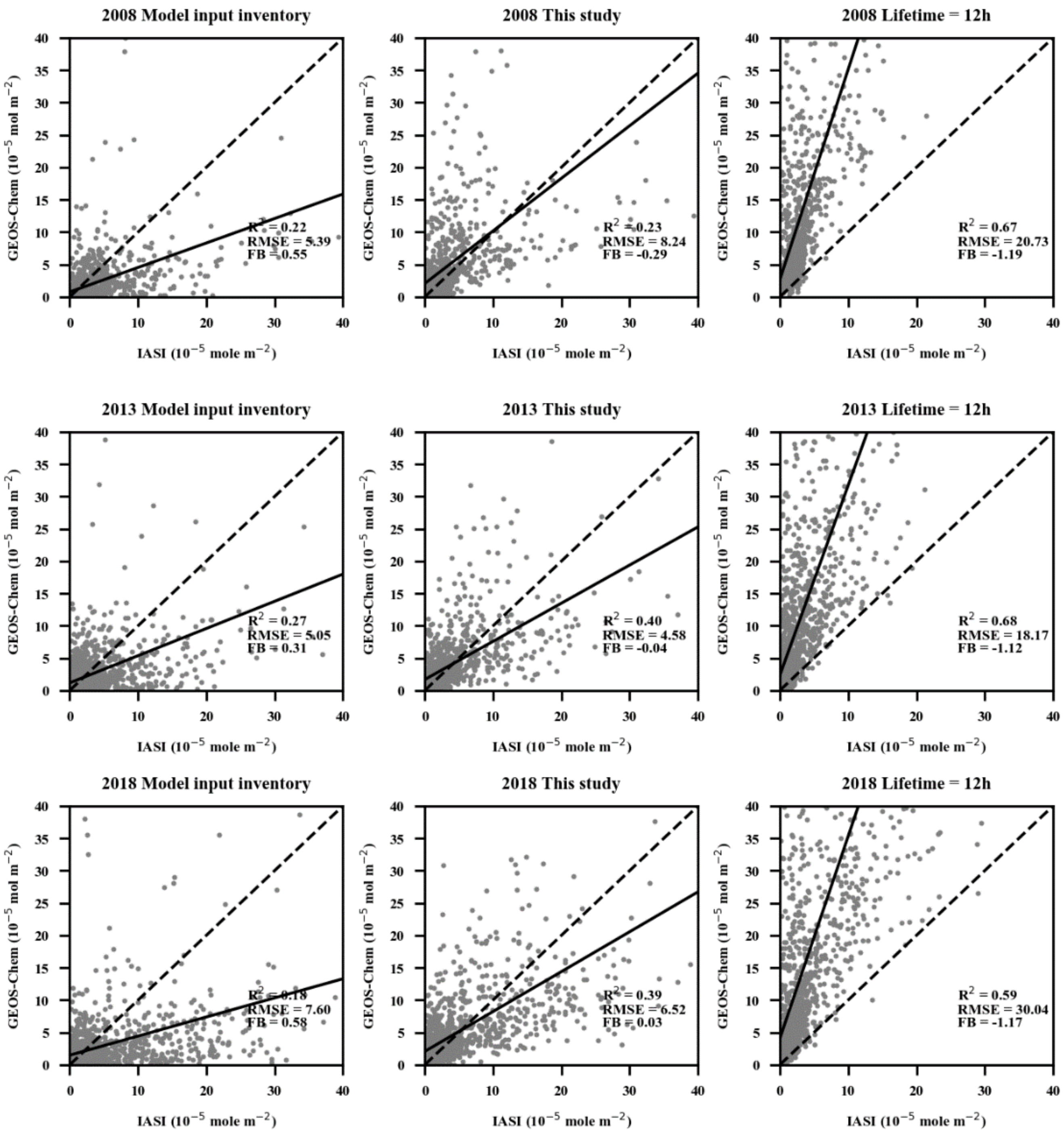
Update

- Validation against IASI observations
  - GEOS-Chem inventory
  - This study
  - Lifetime = 12h

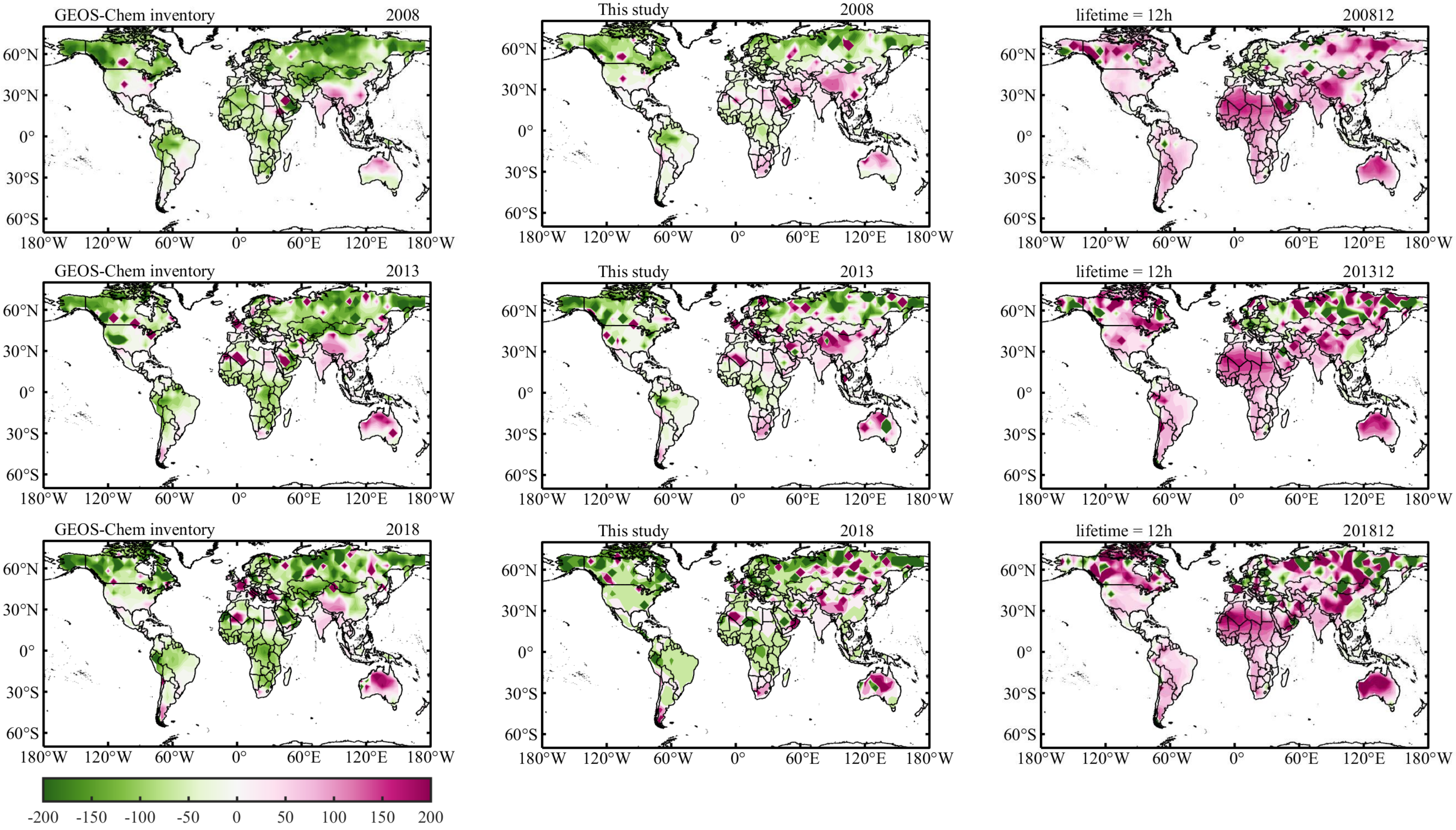
Scatterplots

$$RMSE = \sqrt{\frac{\sum_{\mathrm{i}=1}^{n}(C_{mod,i} - C_{obs,i})^2}{n}}$$

$$FB = 2 * \frac{\sum_{\mathrm{i}=1}^{n}(C_{\mathrm{mod},i} - C_{obs,i})}{\sum_{\mathrm{i}=1}^{n}(C_{\mathrm{mod},i} + C_{obs,i})}$$

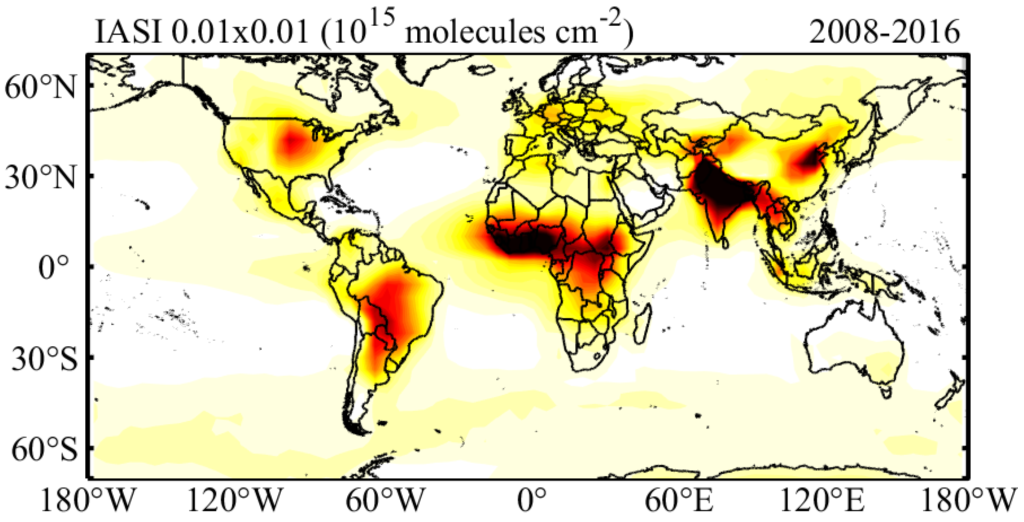


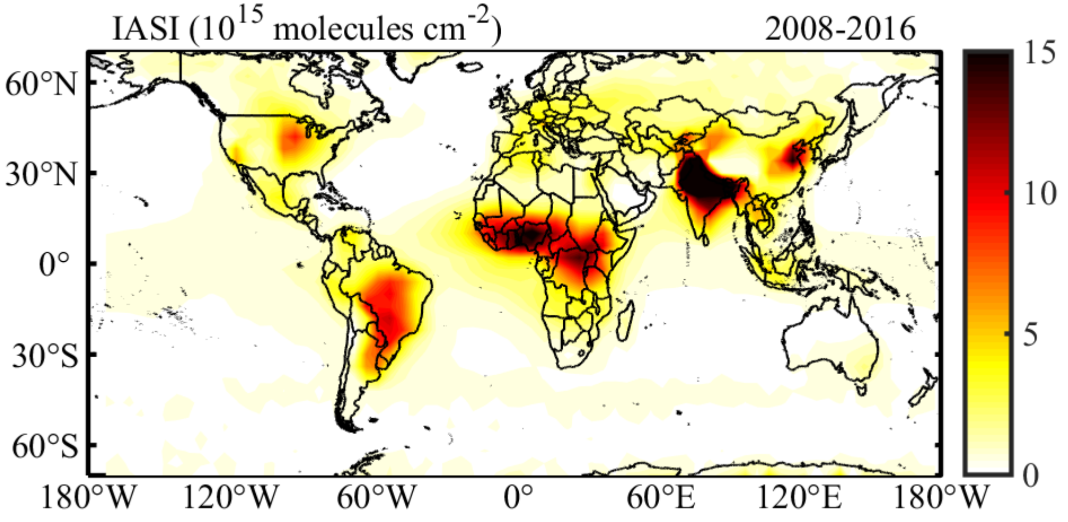
Spatial FB



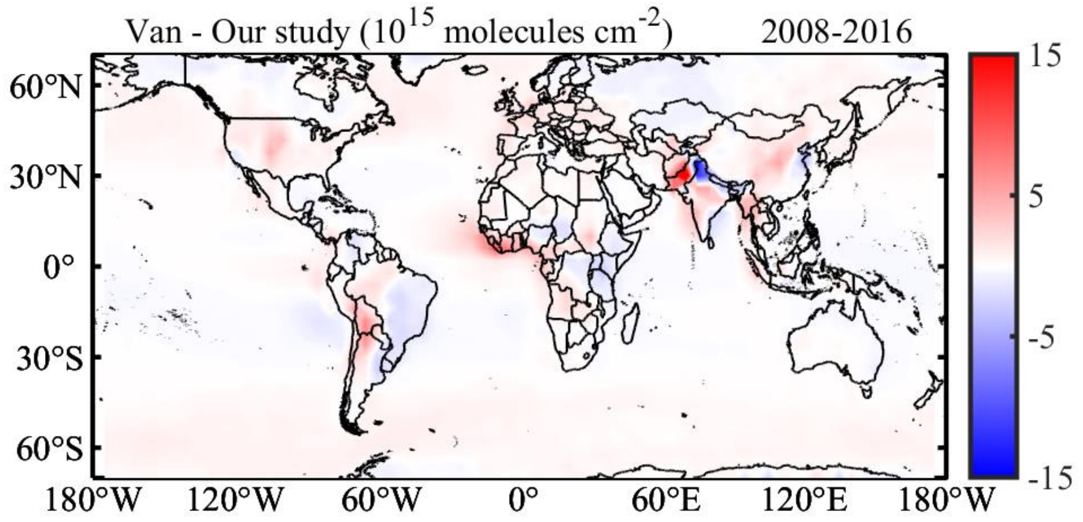
GEOS-Chem inventory
2008
This study
2008
lifetime = 12h
200812
GEOS-Chem inventory
2013
This study
2013
lifetime = 12h
201312
GEOS-Chem inventory
2018
This study
2018
lifetime = 12h
201812
60°N
30°N
0°
30°S
60°S
180°W
120°W
60°W
0°
60°E
120°E
180°W
-200
-150
-100
-50
0
50
100
150
200

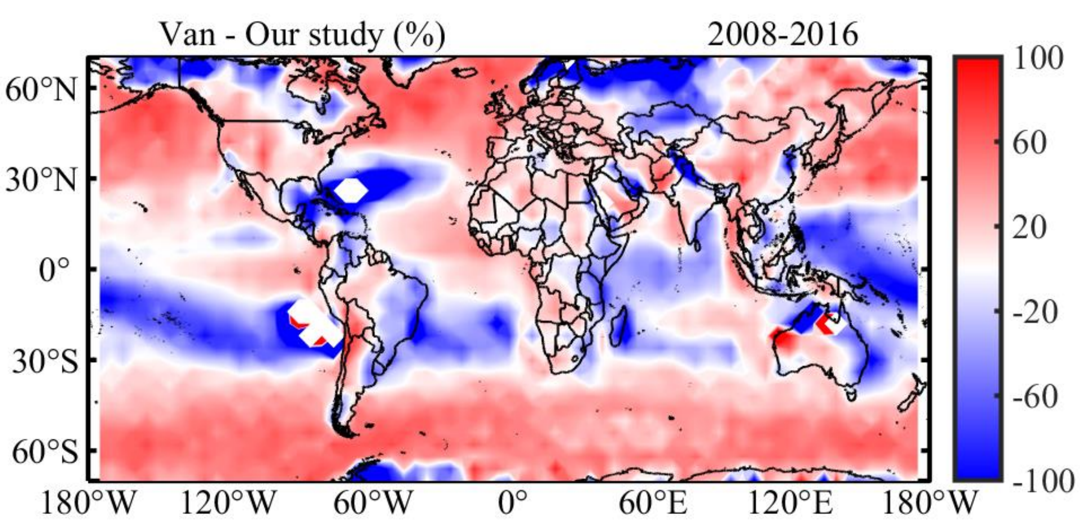
IASI – compare with Van Damme 0.01 degree



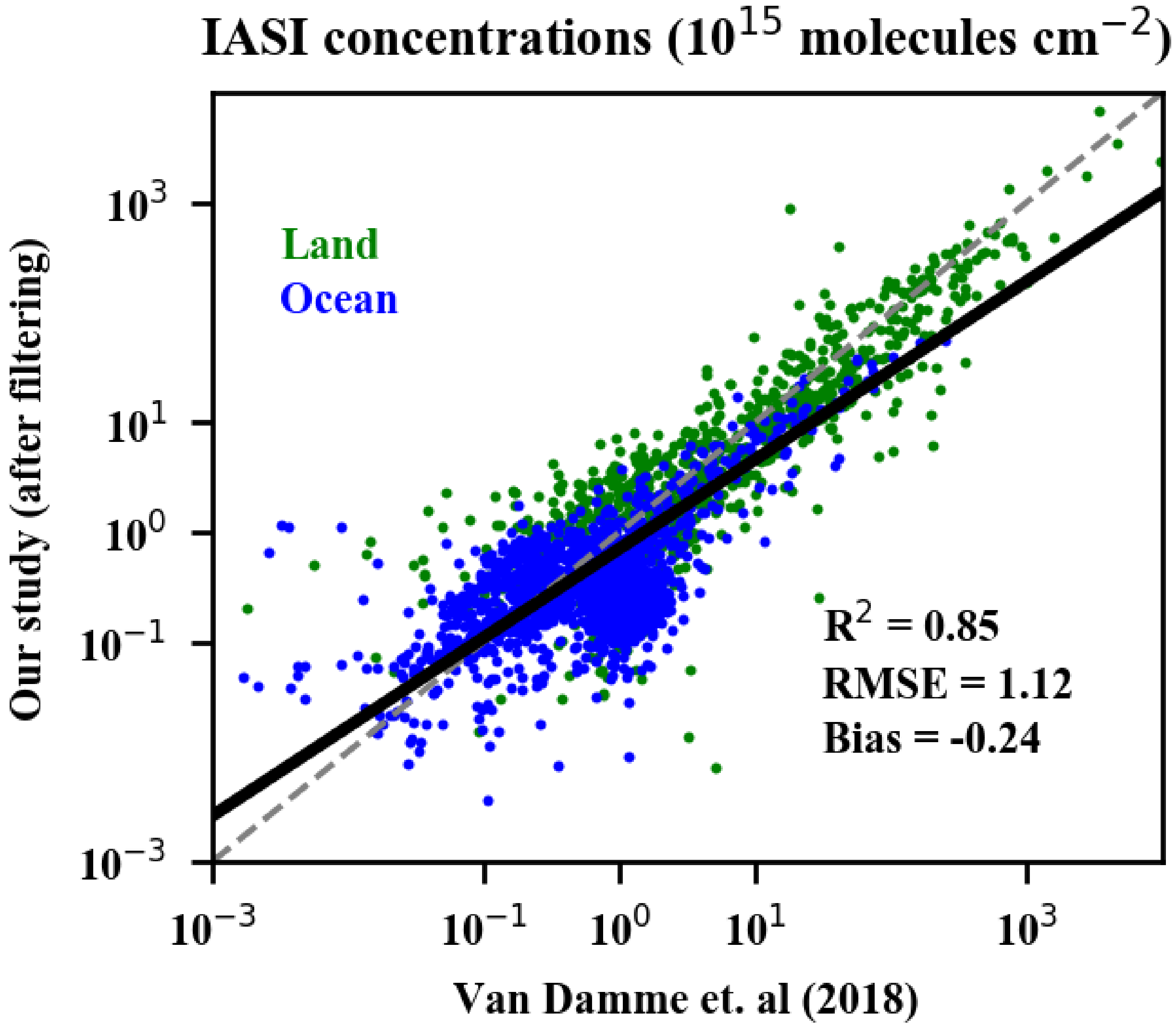


Difference: Ocean and background area, even some hotspot?





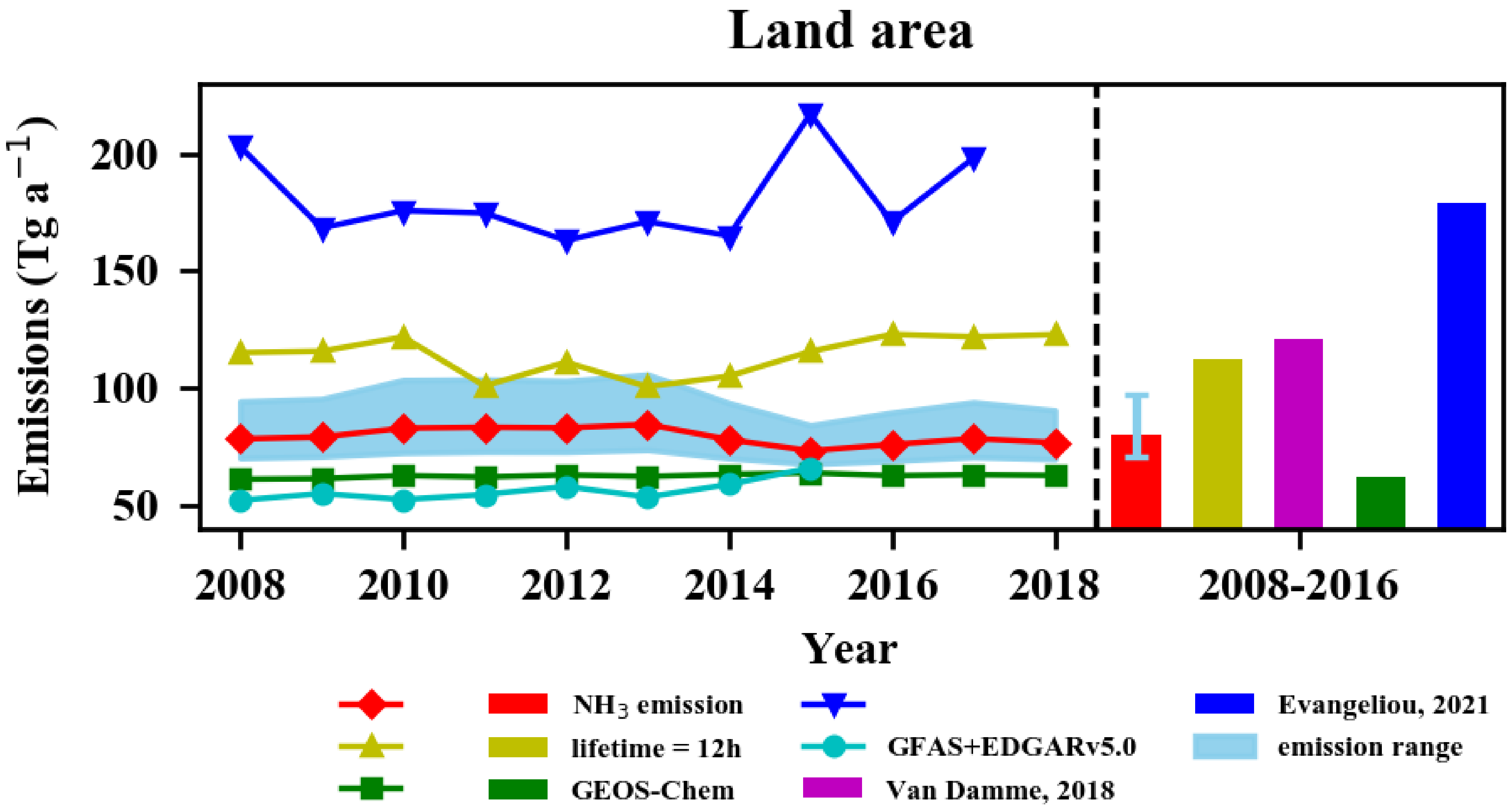
IASI – compare with Van Damme 0.01 degree



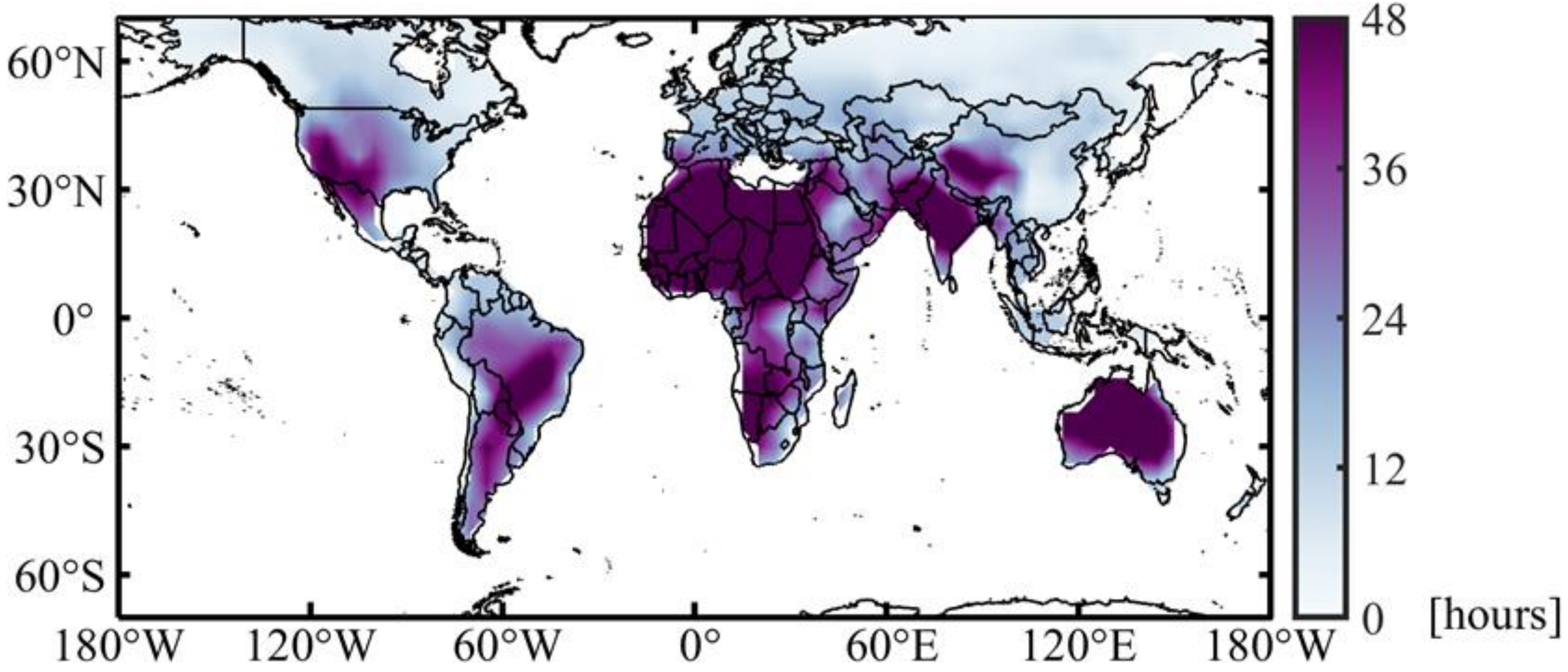
$$RMSE = \sqrt{\frac{\sum_{i=1}^{n}(C_{Van,i} - C_i)^2}{n}}$$

$$Bias = \frac{\sum_{i=1}^{n}(C_{Van,i} - C_i)}{n}$$

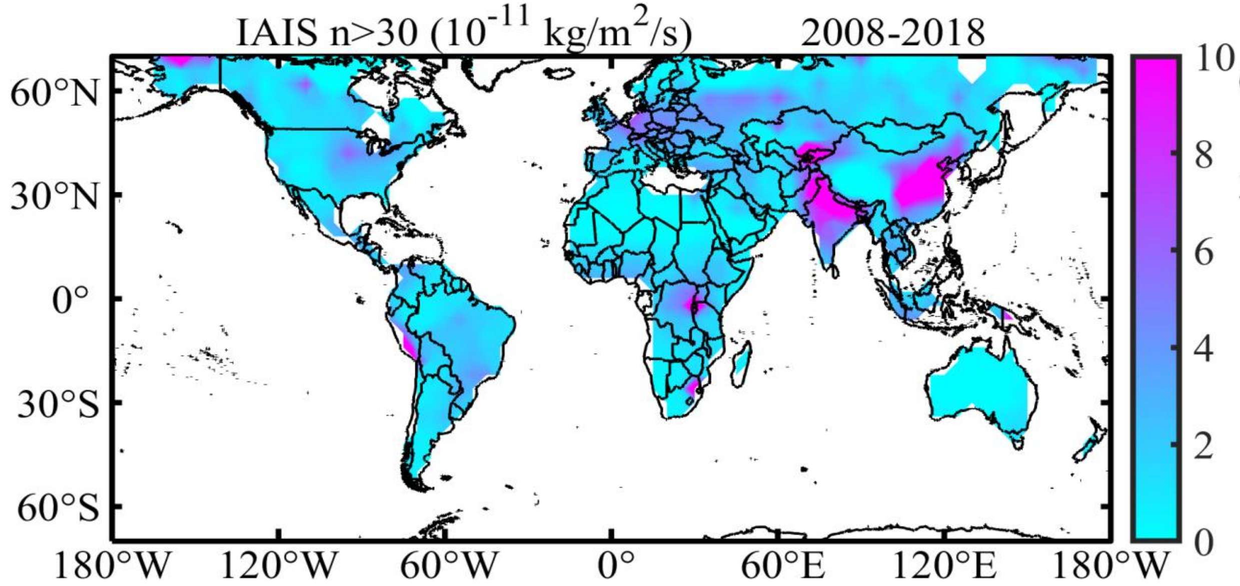
Emission

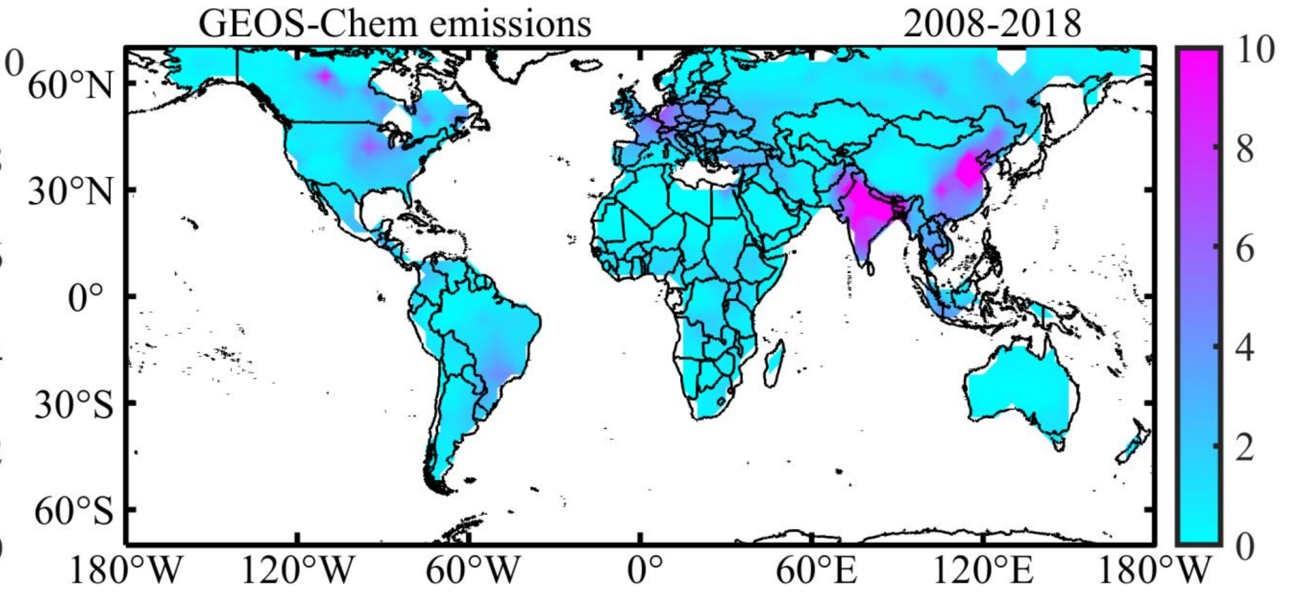

Land area
Emissions (Tg a^{-1})
200
150
100
50
2008
2010
2012
2014
2016
2018
2008-2016
Year
NH$_3$ emission
lifetime = 12h
GEOS-Chem
GFAS+EDGARv5.0
Van Damme, 2018
Evangeliou, 2021
emission range

lifetime

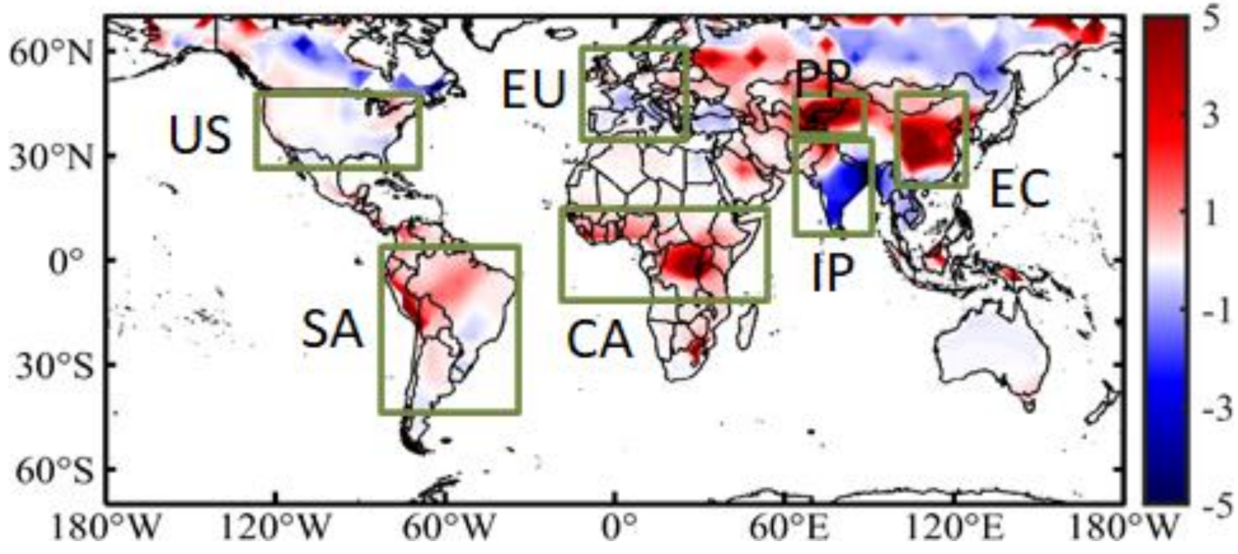

60°N
30°N
0°
30°S
60°S
180°W
120°W
60°W
0°
60°E
120°E
180°W
48
36
24
12
0
[hours]

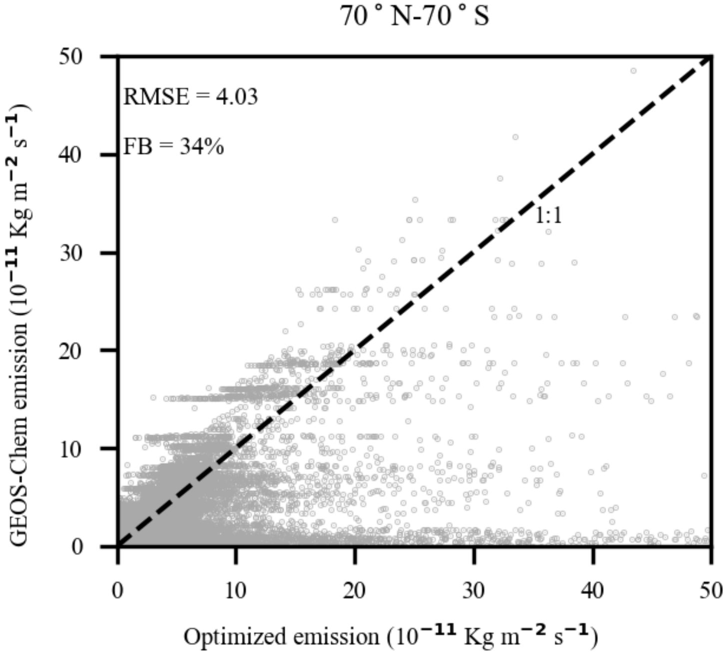
Optimized versus GEOS-Chem



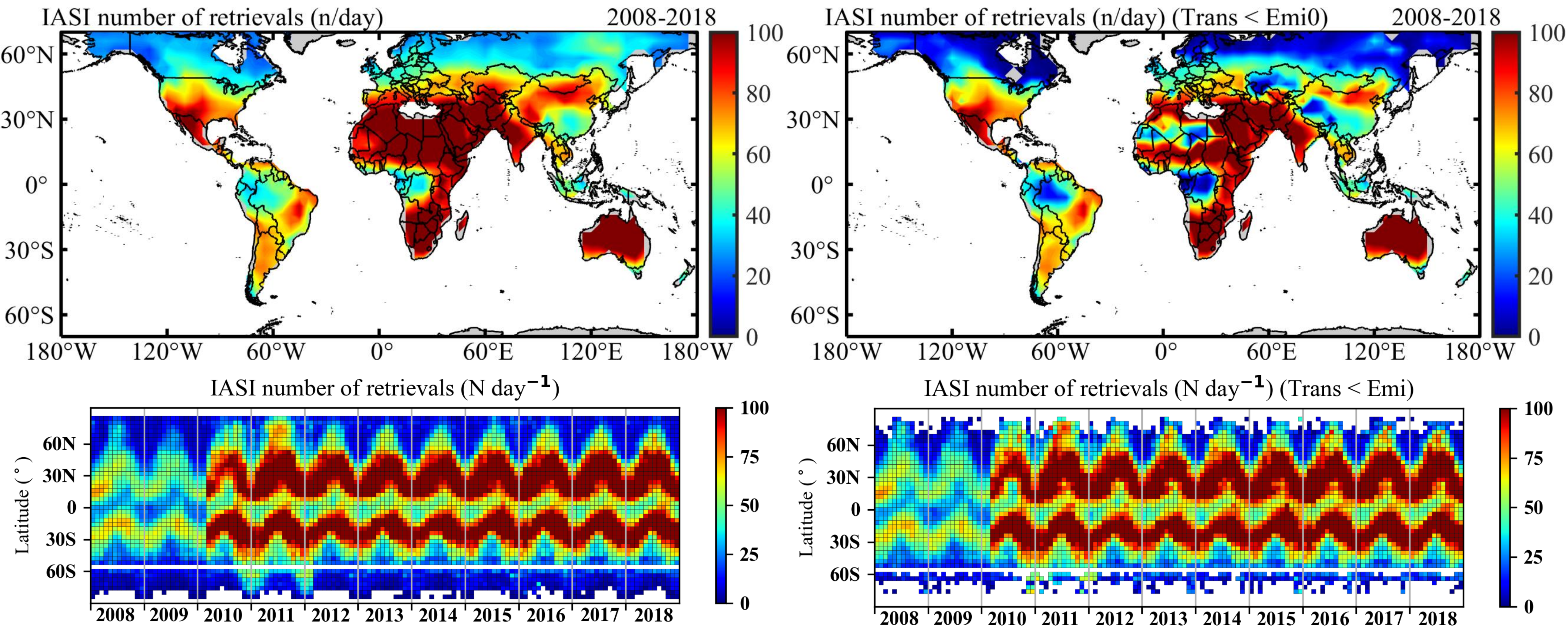


Optimized – GEOS-Chem

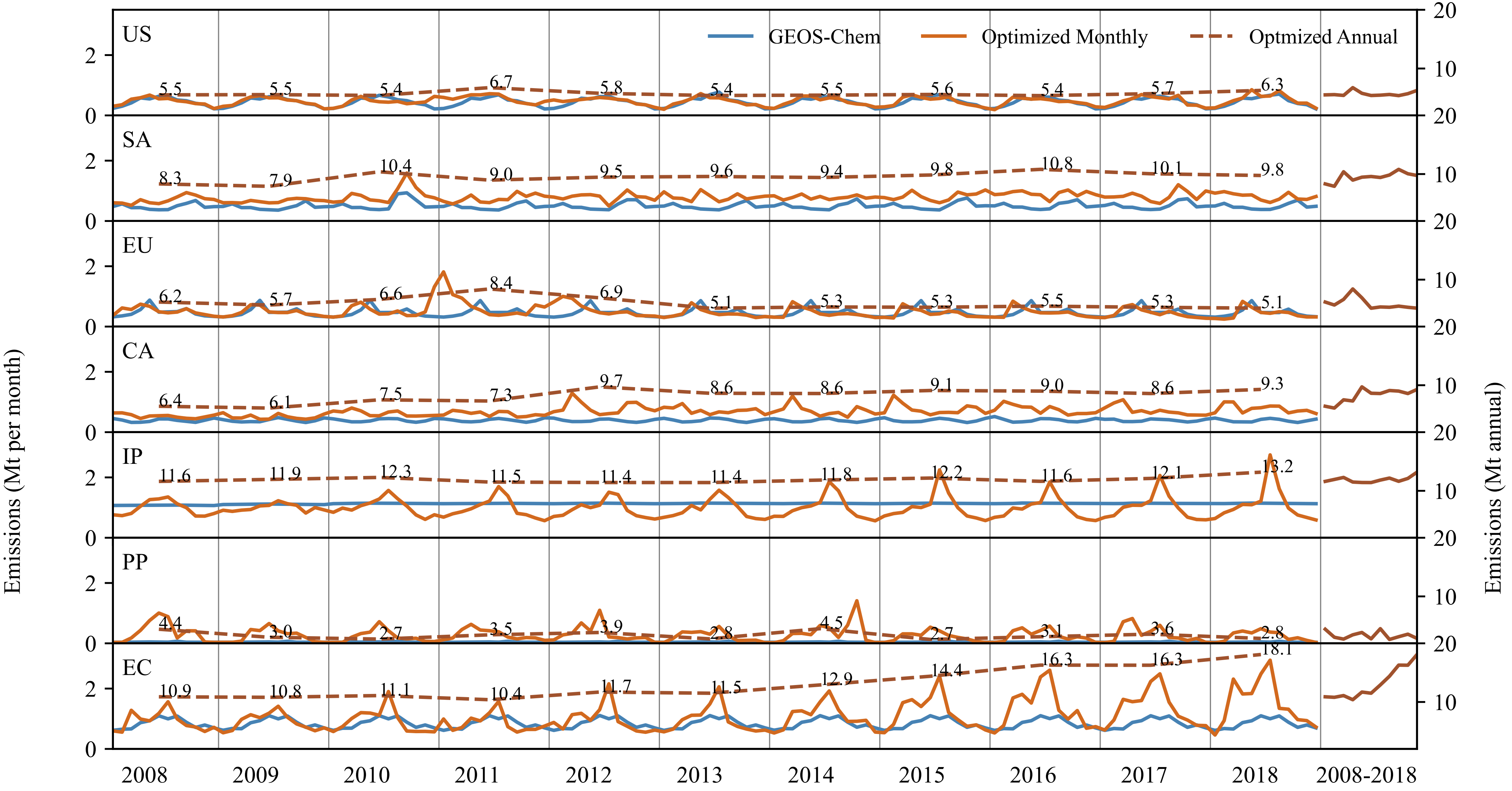




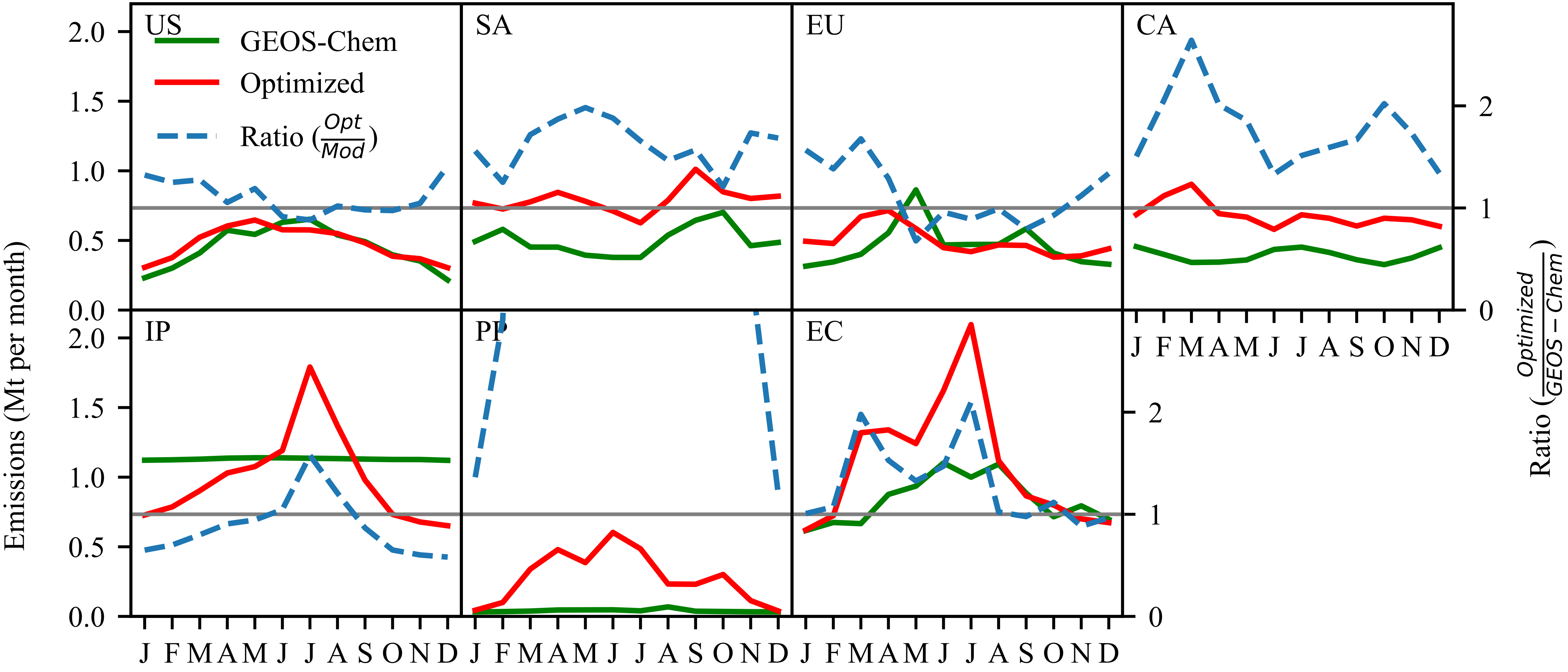
Number of retrievals



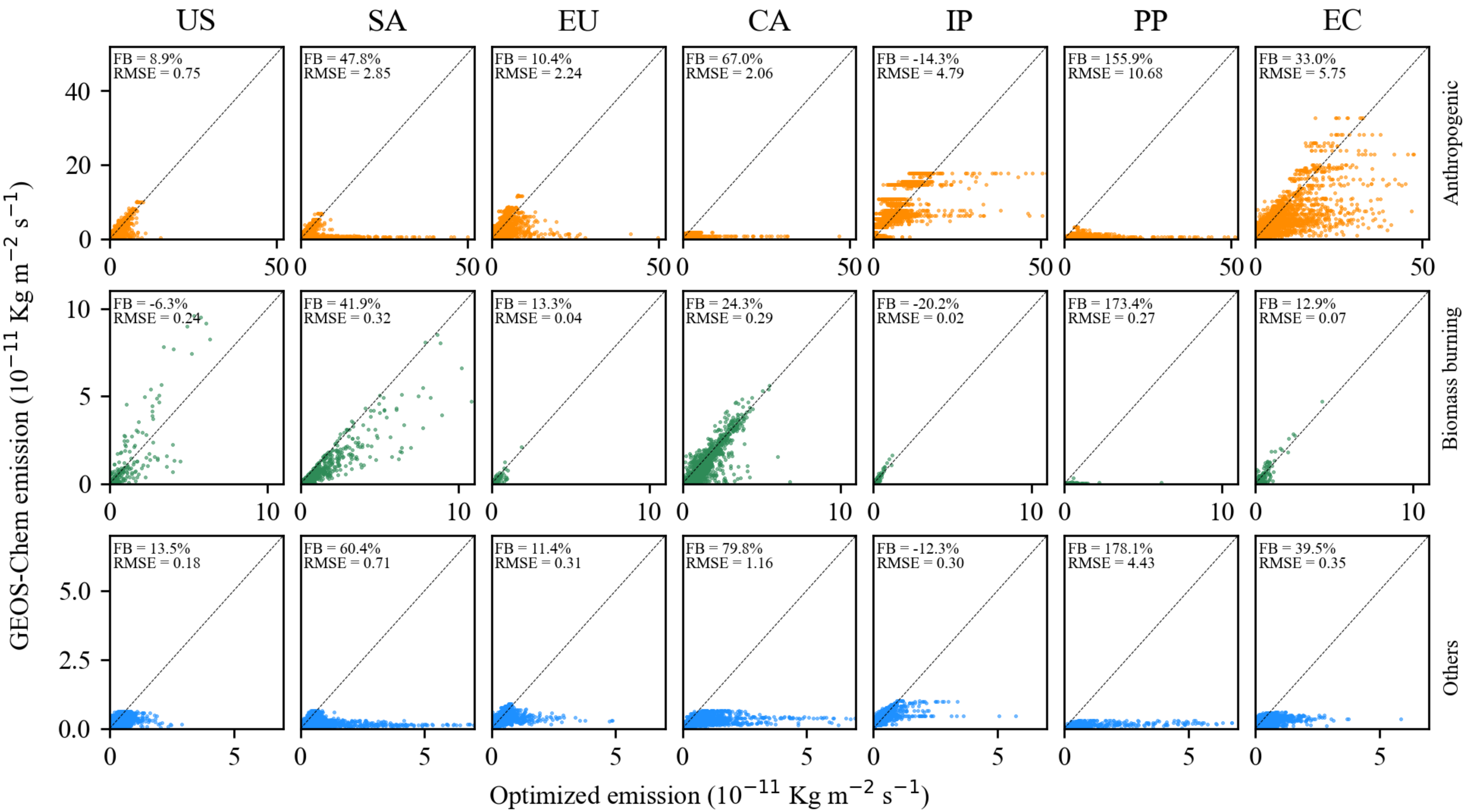
Optimized emissions timeseries



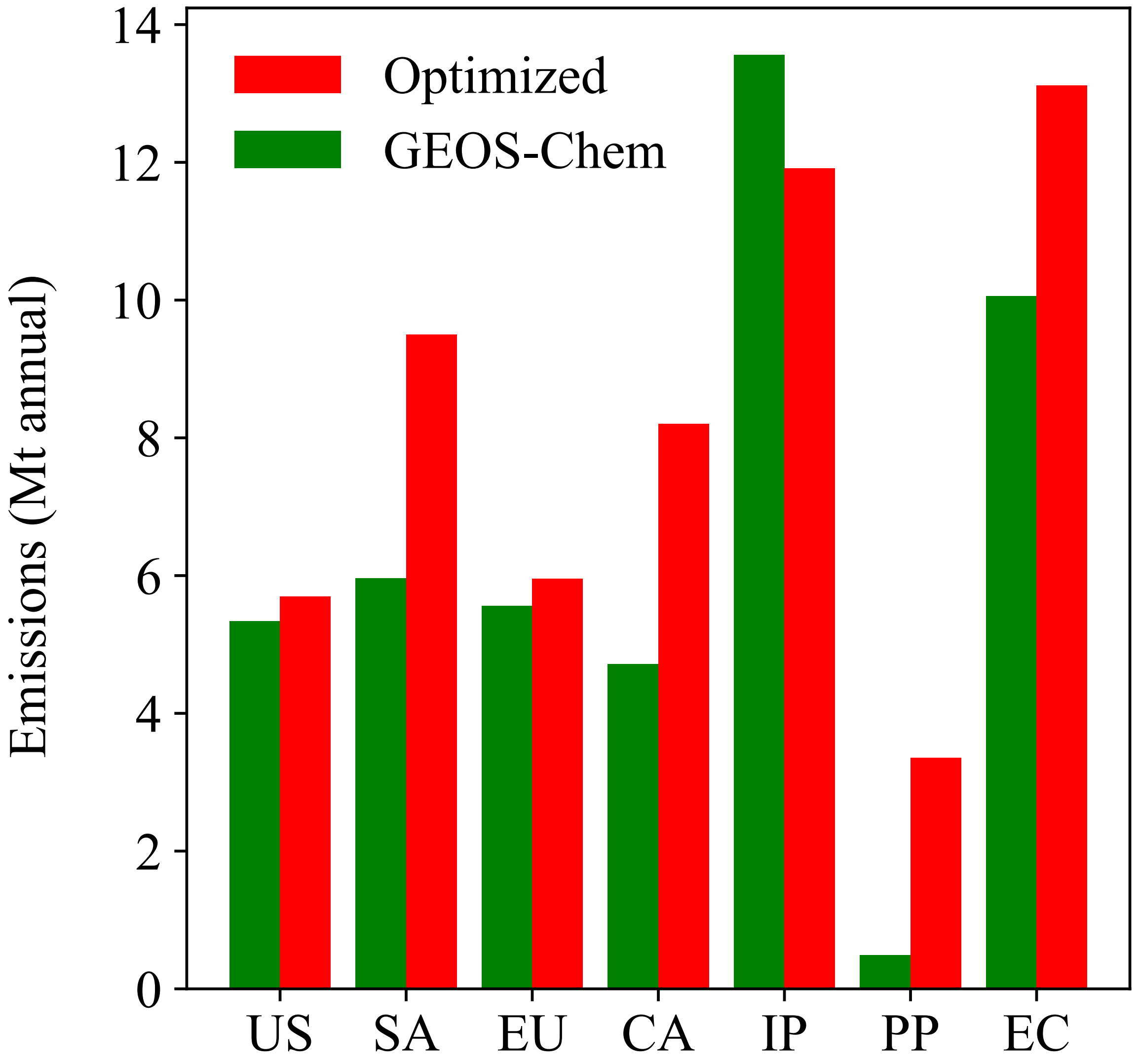
Optimized emissions monthly variations

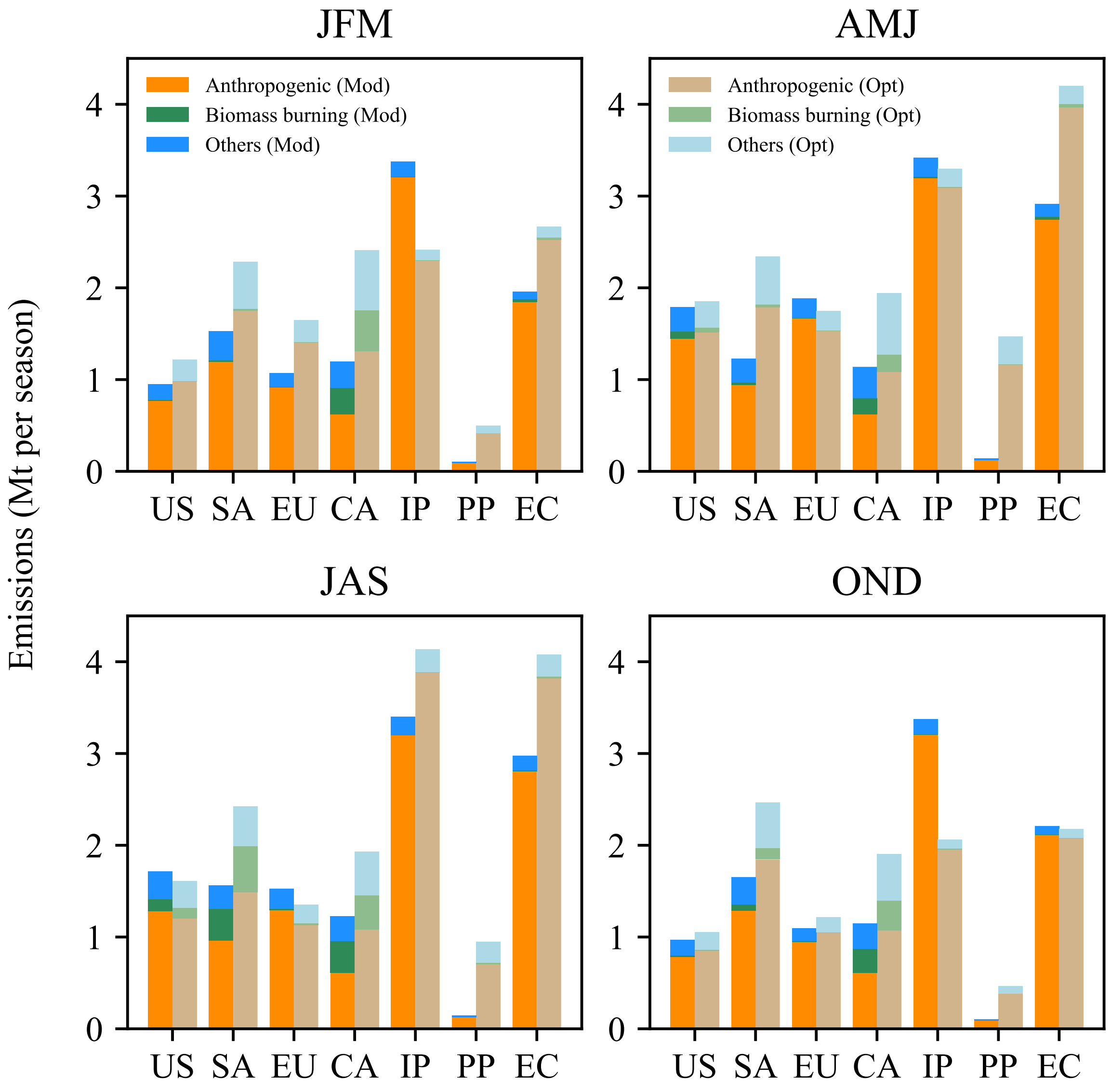


Optimized versus GEOS-Chem by sectors

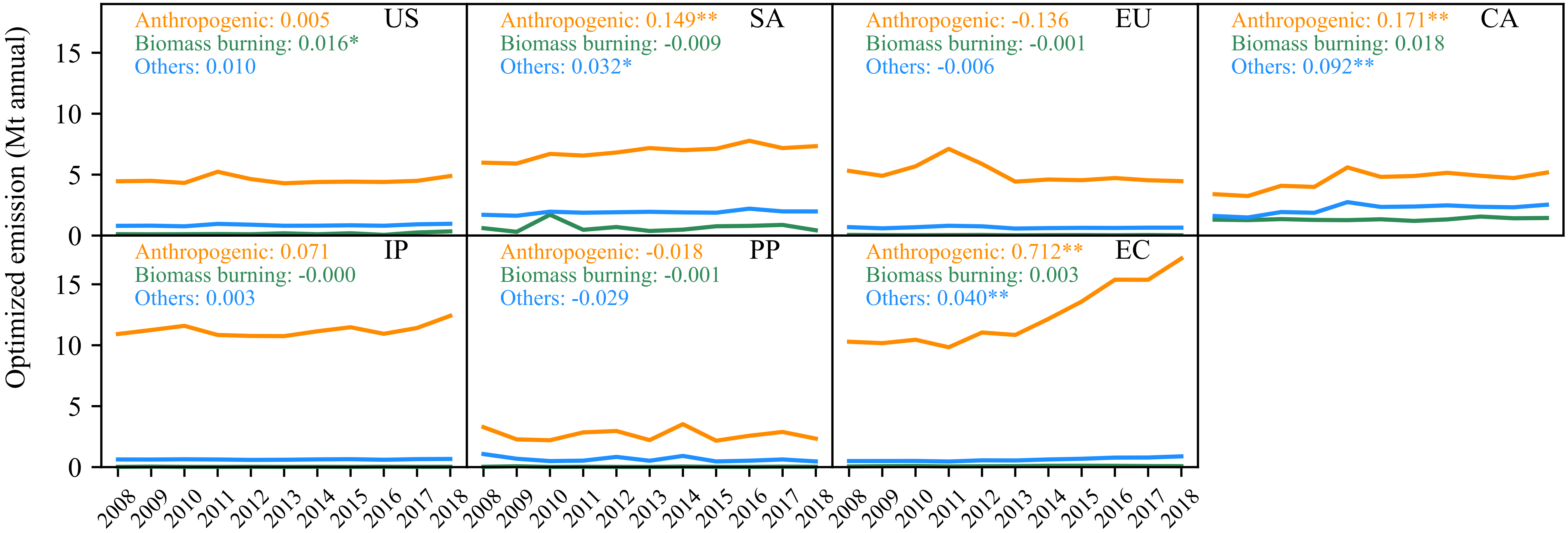


Optimized emissions by sectors

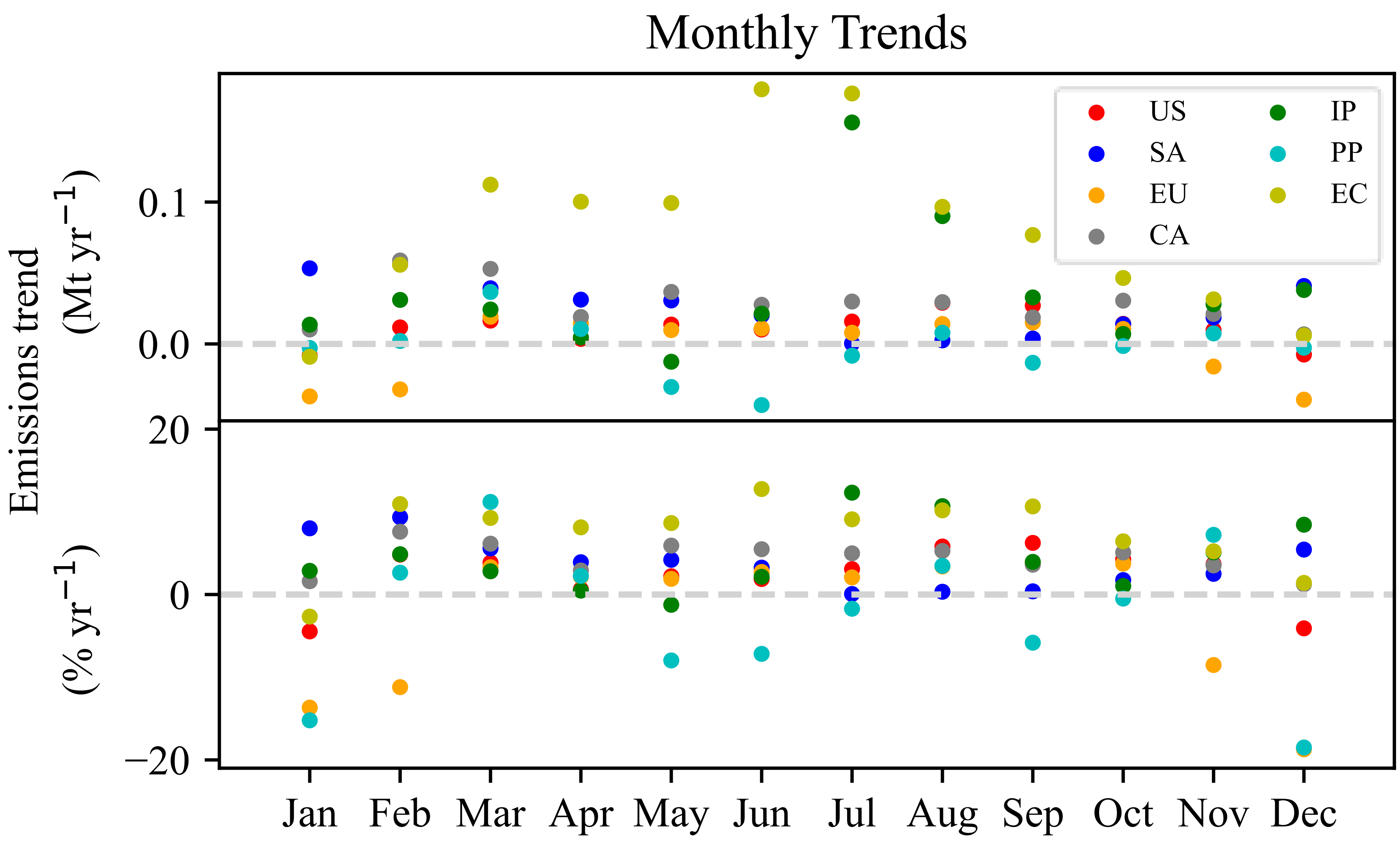




Optimized emissions trend by sectors

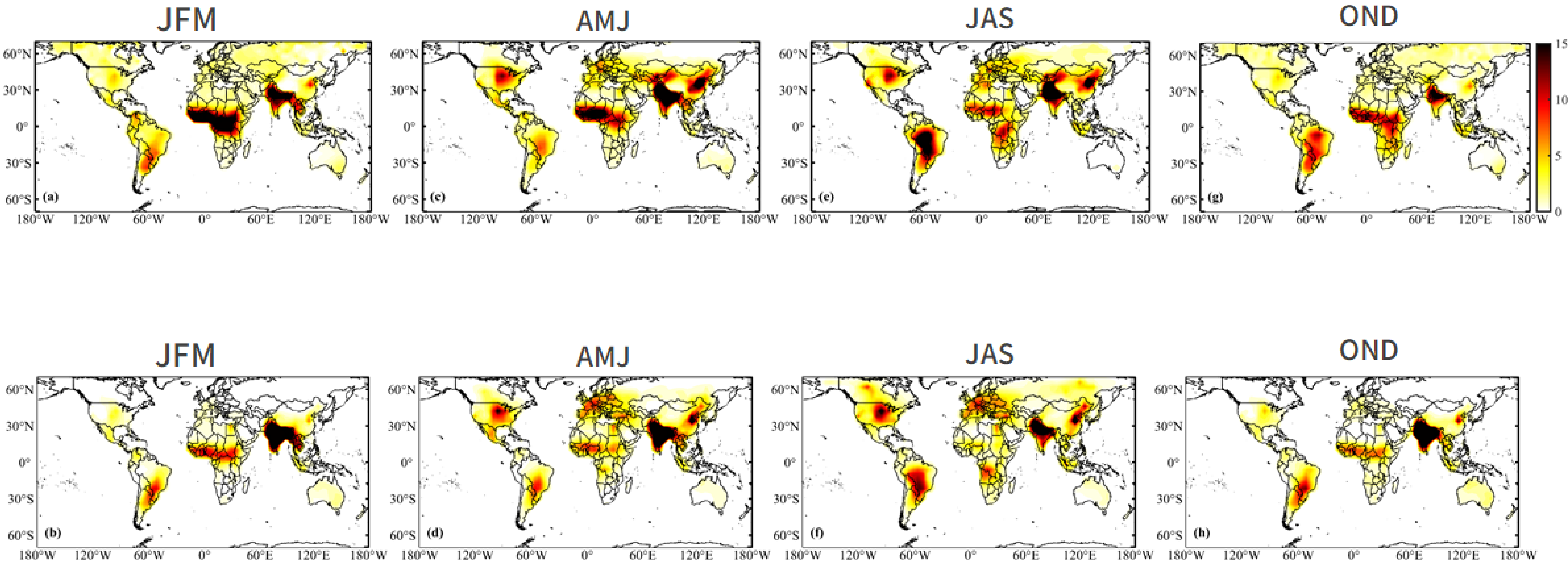


Optimized emissions monthly trend



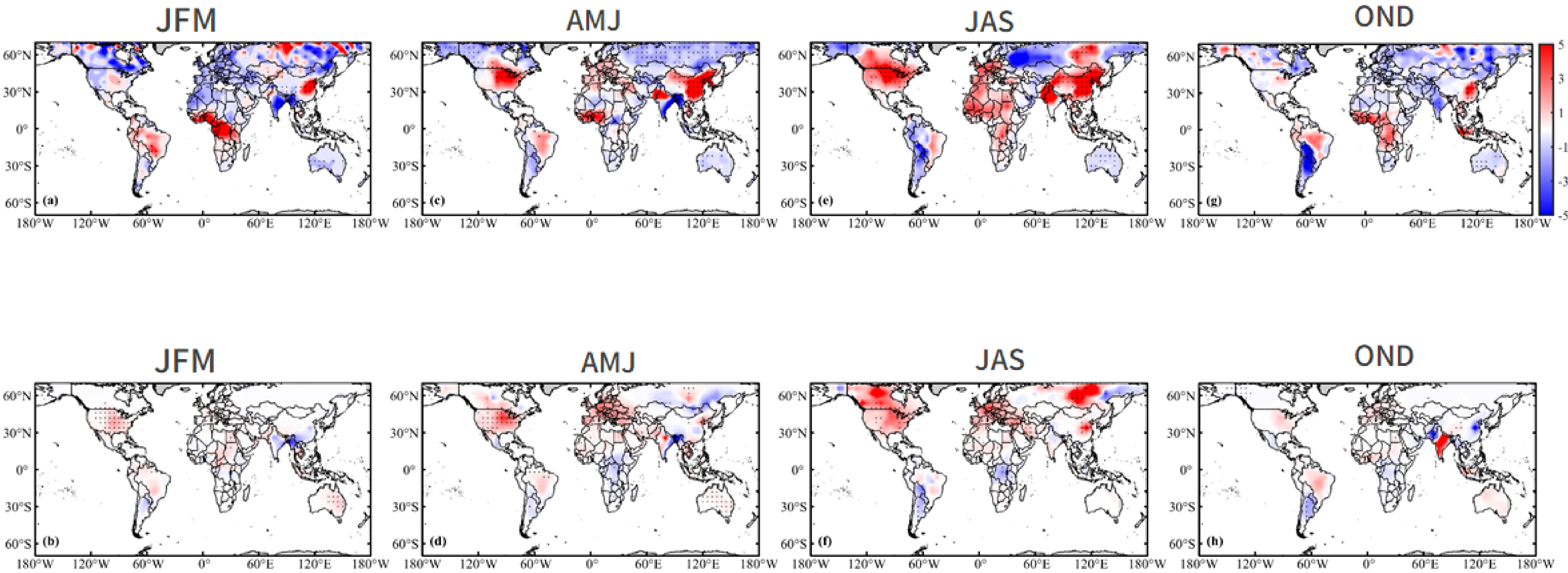
NH_3 Seasonal Concentrations

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

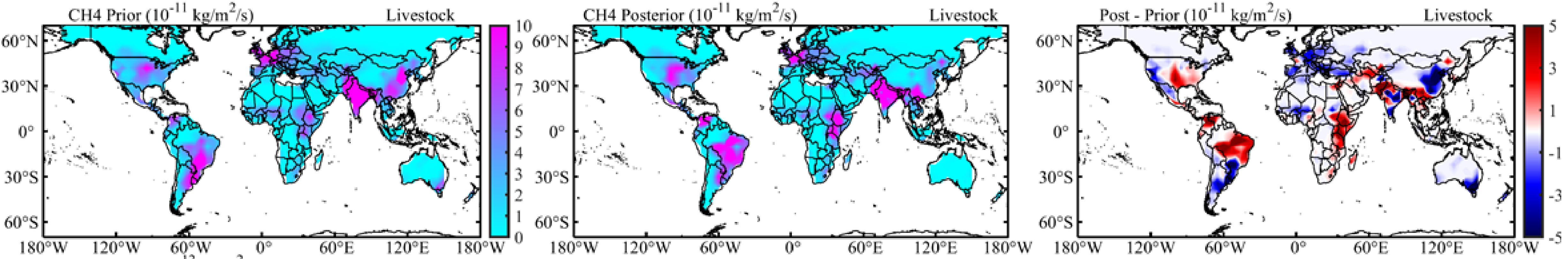


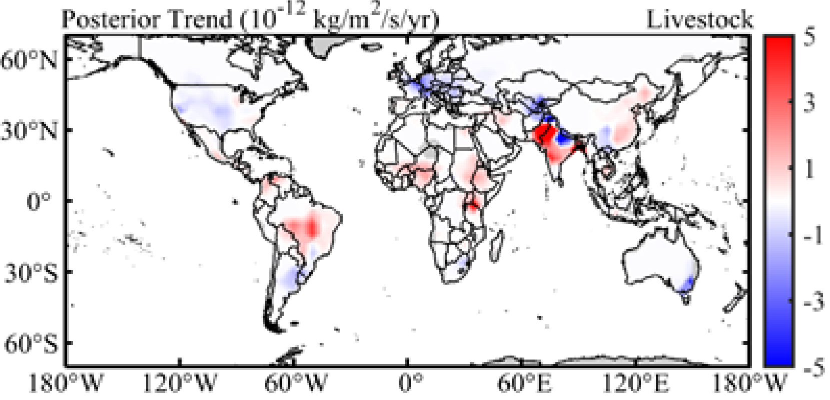
NH_3 Seasonal Concentrations

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

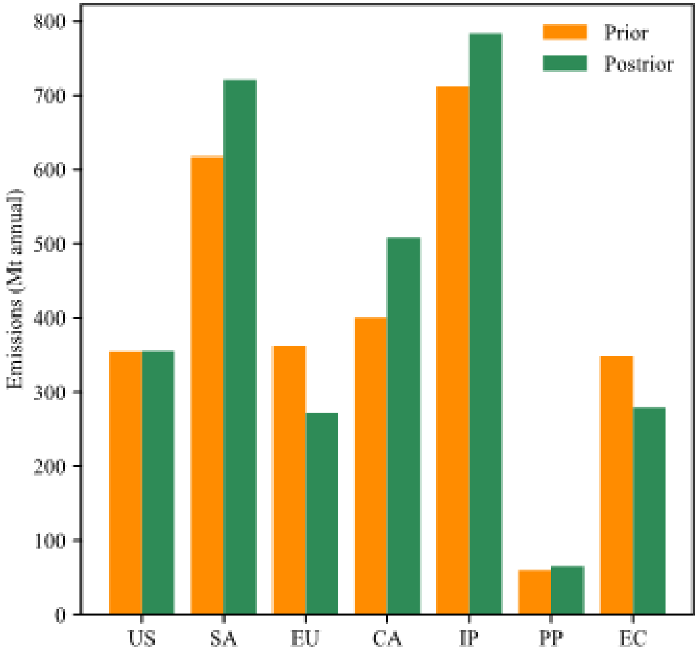


CH4 livestock emissions

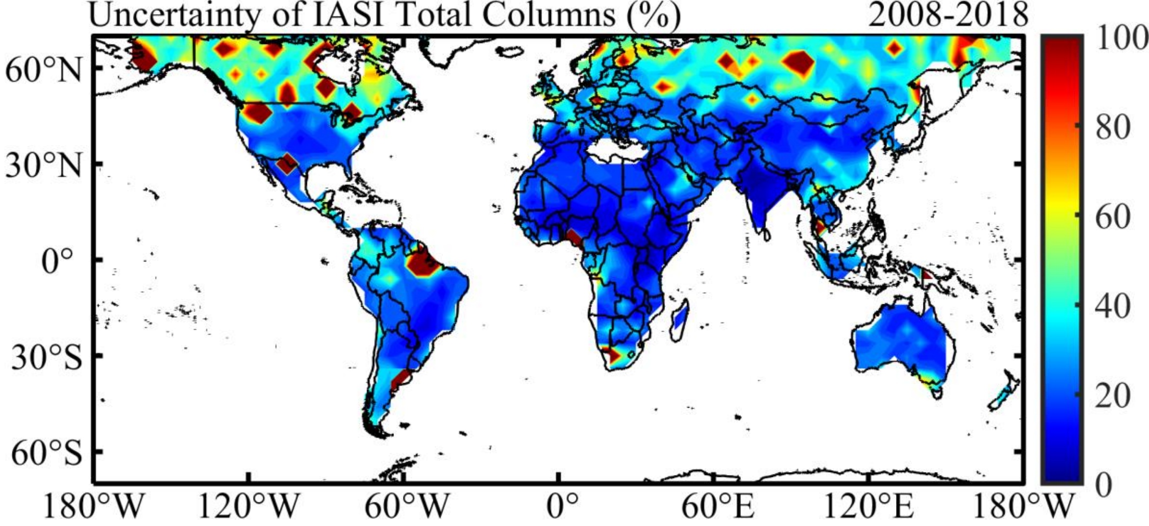




	Mean (Mt)	Trend (Mt/yr)	Relative trend (%)
US	355	-1.11	-0.14
SA	721	5.05	0.46
EU	272	-2.67	-0.24
CA	508	8.02	0.86
IP	783	10.68	0.71
PP	66	-1.65	-1.15
EC	280	1.37	0.49



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	Zhang et al., 2012
NH4 wet deposition	-23-25%	2006-2009	US	NADP/NTN	Zhu et al., 2013
NH4 wet deposition	-1% (-25-12%)	2008-2012	China	EANET	Zhao et al., 2017
dissolved inorganic N deposition	9% (-4-52%)	2000-2014	Southern China	Literature review	Xu et al., 2018

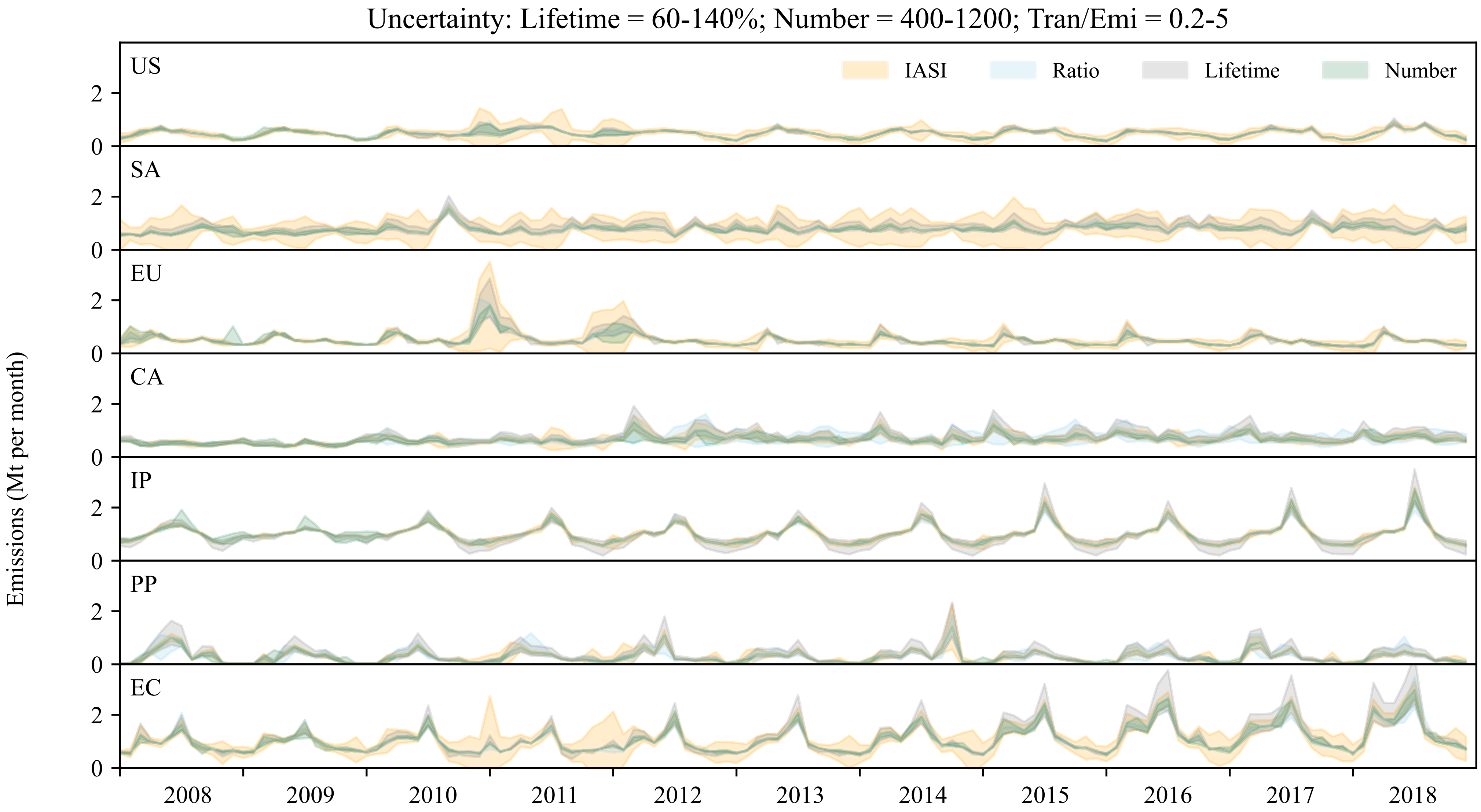
Uncertainty (Mt)

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

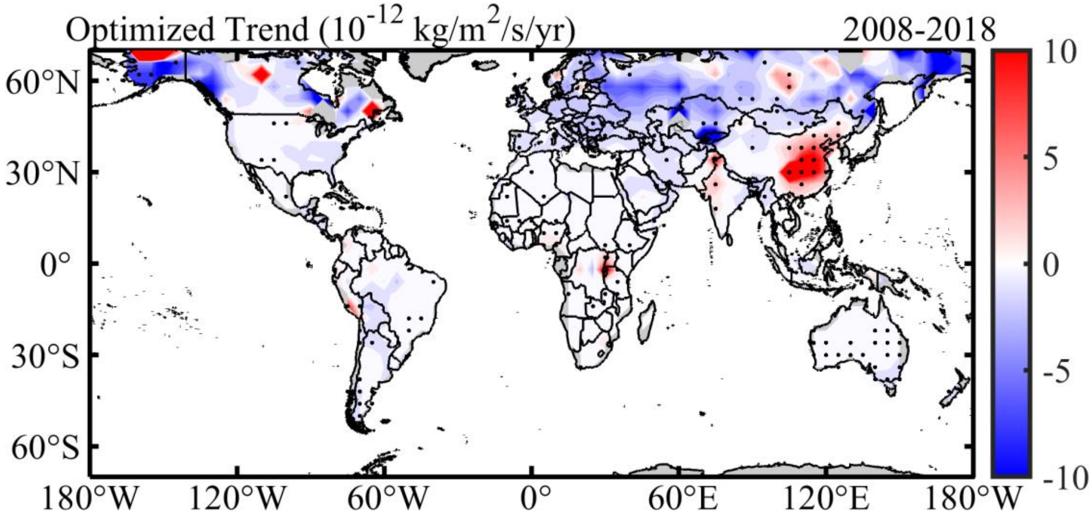
Parameter perturbed	Averaged emission	Standard deviation
Initial: ratio < 1, n > 800	92	7.4
Transportation (-)	101	9.1
Transportation(+)/emission ratio < 0.2	87	5.2
Transportation(+)/emission < 5	102	17.4
Number of retrievals > 400	95	11.2
Number of retrievals > 1200	87	13.9
Lifetime -40%	107	11.0
Lifetime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

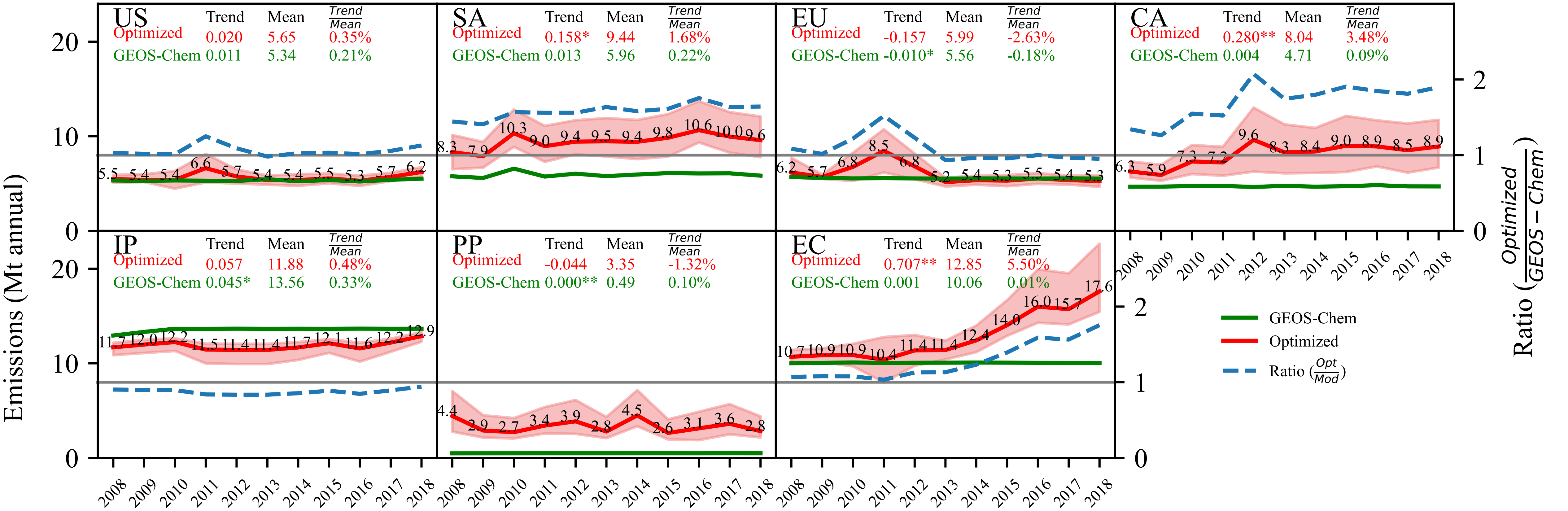
- $\sigma_C = \sqrt{\frac{\sum(\sigma_i \times \Omega_i)^2}{n-1}}$
  - σ_C: the total column error in each grid, [mol m-2]
  - σ_i: the ith retrieval relative error, [mol m-2]
  - Ω_i: the ith retrieval total column, [%]
- $\sigma_{IASI} = \frac{\sigma_C \times M}{\tau_{mod}}$
  - σ_{IASI}: the emission error in each grid associated with IASI total column error, [kg m-2 s-1]
  - M: relative molecular mass, 17 [kg mol-1]
- $\overline{\sigma_{IASI}} = \sqrt{\sum(\sigma_{IASI_j} \times A_j \times t)^2}$
  - $\overline{\sigma_{IASI}}$: the domain mean error, associated with IASI total column error, [kg]
  - σ_{IASI_j}: the emission error in jth grid, [kg m-2 s-1]
  - A_j: the area of jth grid, [m2]
  - t: the defined time period, [s]

Uncertainty



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 τ

- $E = M/\tau$
  - E: emission fluxes, assumes stationarity and constant first-order loss terms
  - M: the total mass contained within the assumed box
  - τ: The effective lifetime or residence time of NH3 within a given box

- $\tau_{mod} = \frac{(K^{\mathrm{mod}}_{NH_4^+/NH_3}+1)M_{mod}}{-\Delta M^{drydep,wetdep}_{NH_3,NH_4^+}}$
- $\tau'_{mod} = \frac{\tau_{mod}}{K^{\mathrm{mod}}_{NH_4^+/NH_3}+1} = \frac{M_{NH_3}}{-\Delta M^{drydep,wetdep}_{NH_3,NH_4^+}}$
- $\hat{E}_{obs} = \frac{(M_{\mathrm{obs}}-M_{mod})}{\tau'_{mod}} + E_{mod}$

Table SI1: NH₃ lifetime estimates reported in the literature.

REFERENCE	LIFETIME	COMMENT
Norman and Leck, 2005	Few hours	Clean remote ocean
	Several days	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

- $\Omega = \sum_{i=1}^{47} c_{\mathrm{i}} \times rho_i \times h_i \times k$
  - $\mathbf{\Omega}$**: total column concentration,** [mol/m2]
  - c_{i}: 'IJ-AVG-$_NH3', mixing ratio for each level, [ppbv] to [v/v] (*1E-9)
  - rho_i: 'TIME-SER_AIRDEN', air density for each level, [molecules/cm3]
  - h_i: 'BXHGHT-$_BXHEIGHT', grid box height for each level, [m] to [cm] (*100)
  - k: 1/6.02214179E19, multiplication factor to convert [molecules/cm2] to [mol/m2]

Regrid 180x360 to 46x72

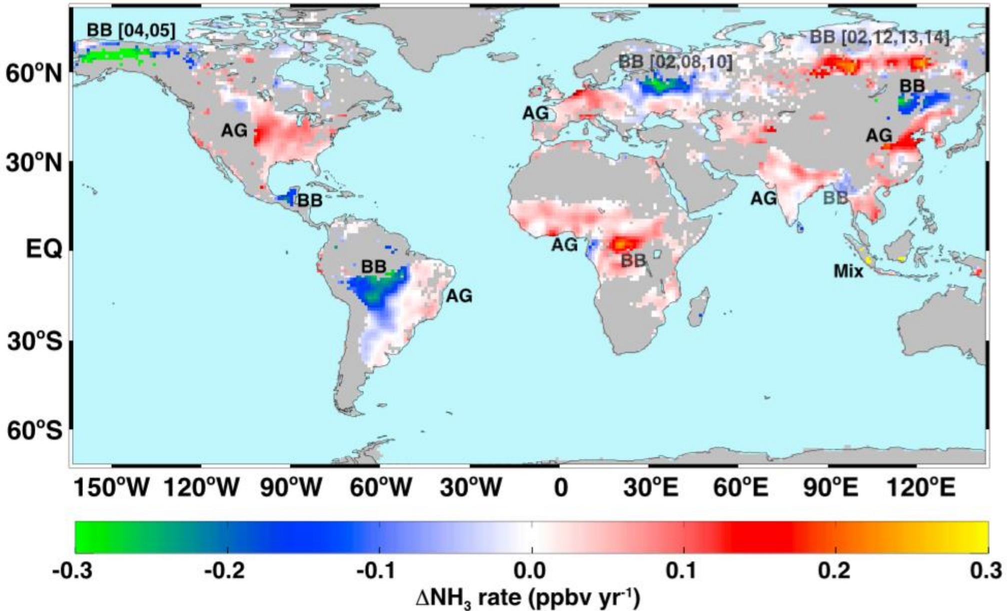
- Latitude: 46 degrees
  - 88°-90°: 2x5 to 1x1, 2 degrees
  - 0-88°: 4x5 to 1x1, 44 degrees
- Method:
  - Step1: mask ocean, set as NaN
  - Step2: calculate mean value in each upscaling grid

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 C2H6 for 2010
  - XIAO_C3H8: C2H6 and C3H8
  - AFCID: PM2.5 dust emission
- Natural
  - GEIA_NH3: 1990 (obsolete now)
  - SEABIRD_DECAYING_PLANTS: the oceanic emissions of acetaldehyde
  - NH3: 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 (NH3) concentrations (14year) over several of the world's major agricultural regions
- The rate of change of NH3 volume mixing ratio (VMR) in parts-per-billion by volume (ppbv) per year computed
  - BB: biomass burning
  - AG: agricultural



(Warner et al, 2017)

End