Optimized ammonia emissions 17

IASI data and GEOS-Chem simulation

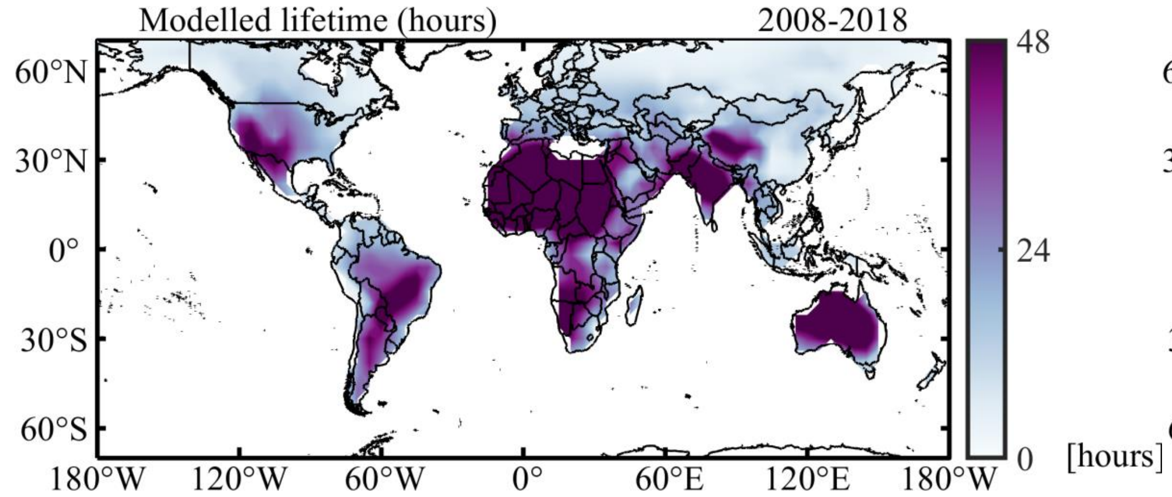
2021.4

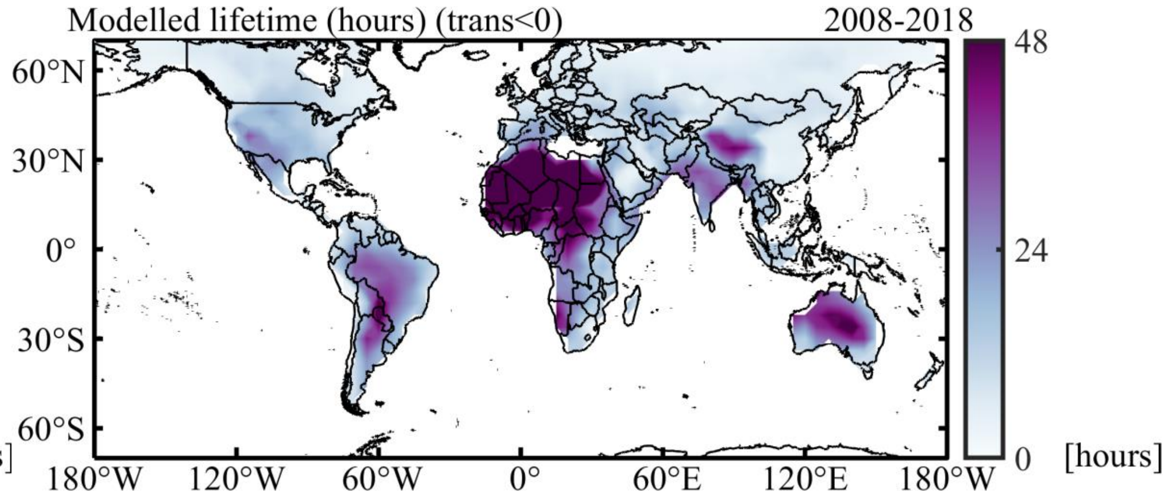
- Accomplished:
  - 1. recalculte the number of retrievals
  - 2. check the lifetime
  - 3. uncertainty of depositions and IASI total columns
- Ammonia Data:
  - IASI total columns: Reanalyzed IASI/Metop-A
    - Daily, L2, 1°×1° (2008-2018)
  - GEOS-Chem simulation, 4°×5°, daily, 2008-2018
    - Total column concentration
    - Total column transport/deposition rate of change
    - Emissions
- meteorological input data
  - ECMWF ERA5 skin temperature, 0.25°x0.25°
    - hourly data on single levels (2008-2018), 9:00/10:00
- Ongoing:
  - 1. regional details

FAOSTAT

- Input:
  - Fertilizers: totals agricultural use N for fertilizers——1.37E9 kg
- Agricultural emissions:
  - Manure management: totals N (N2O) from aerobic and anaerobic manure decomposition processes——3.96E10 kg
  - Manure applied to soils: N2O emissions from N of manure added to agricultural soils——4.81E10 kg
  - Manure left on pasture: N2O emissions from manure N left on pastures by grazing livestock——1.43E11 kg
  - Crop residues: N2O emissions from N in crop residues and forage/pasture renewal left on agricultural fields——4.36E10 kg

lifetime





Emission fluxes of ammonia are continuous

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

- M: NH_3 mass
- F_{out}: NH_3 mass rate of export
- L: NH_3 mass rate of chemical reaction
- D: NH_3 mass rate of deposition, including the wet deposition and the dry deposition

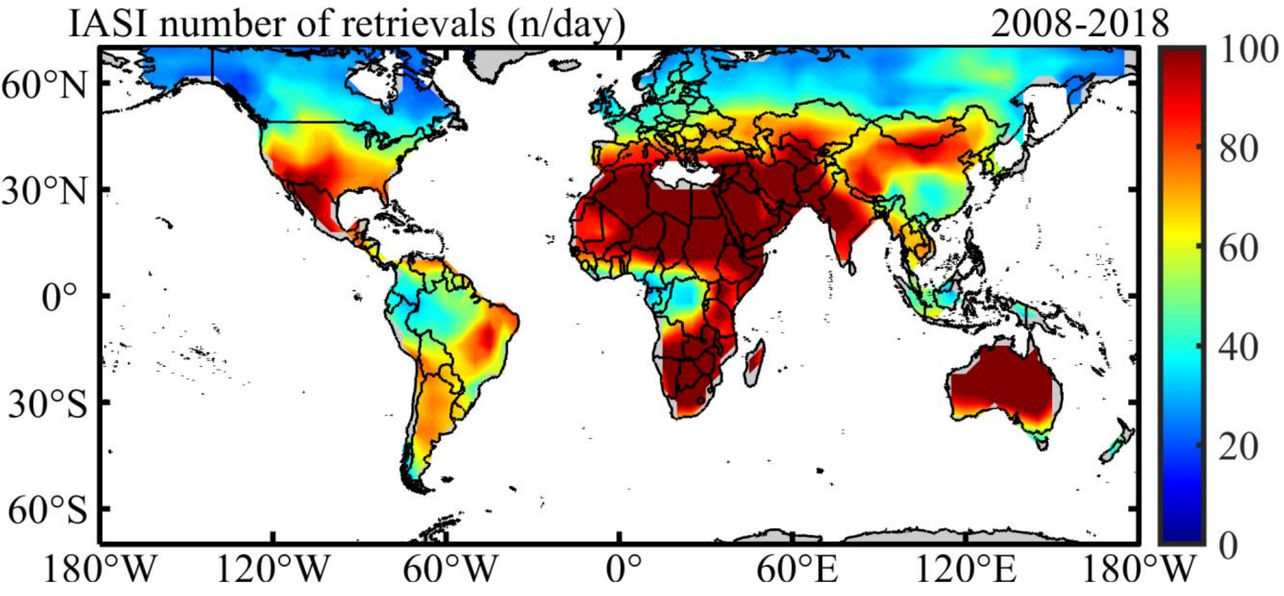
The NH_4^+ concentration ($[NH_4^+]$) is proportional to NH_3 concentration ($[NH_3]$)

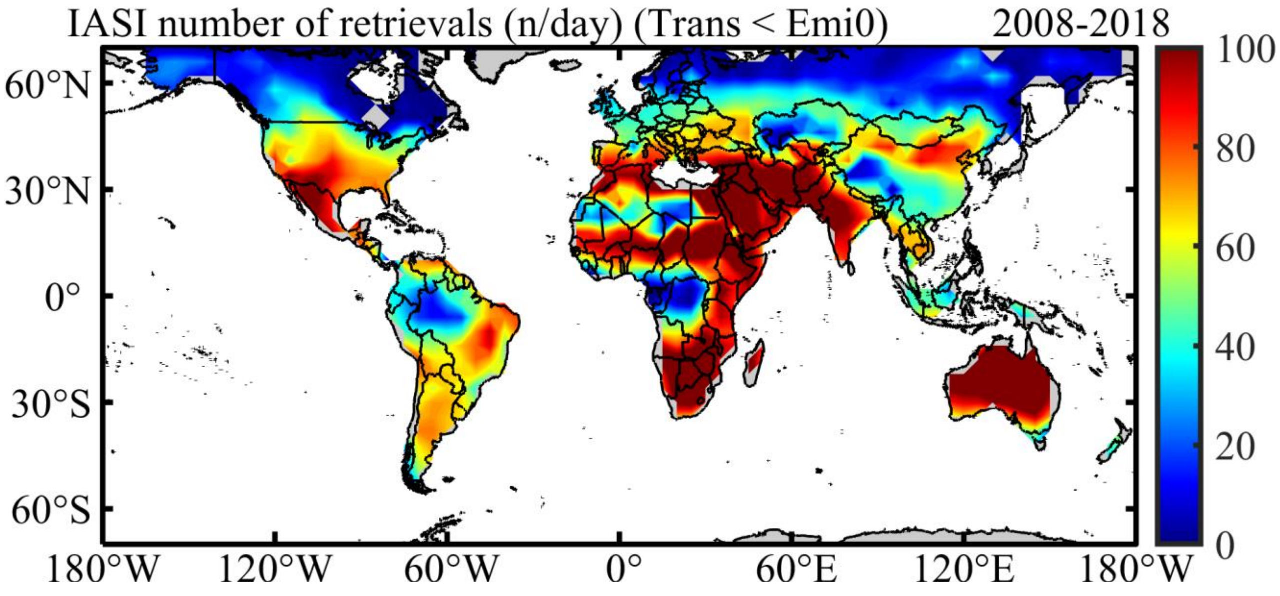
$$[NH_4^+] = K \times [NH_3]$$
$$D = D_{NH_3} + D_{NH_4^+}$$

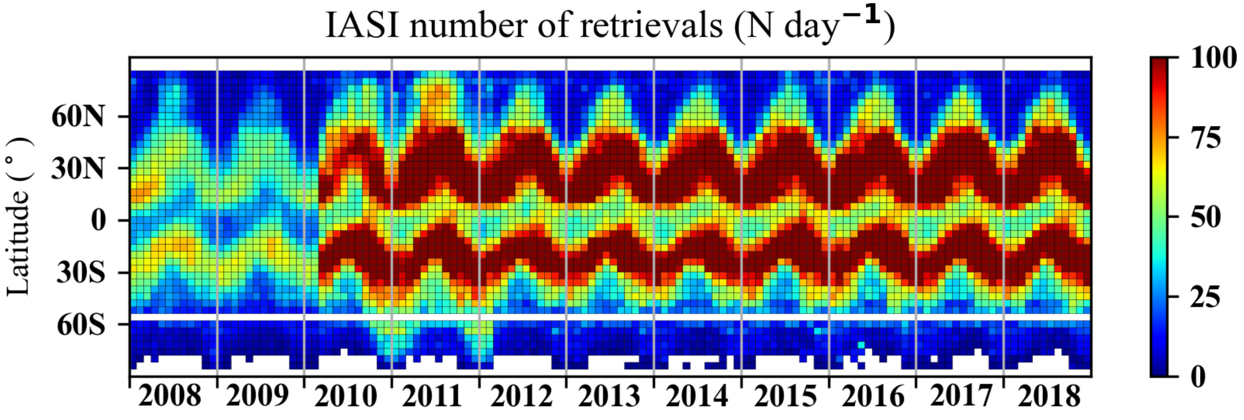
Neglect the NH_3 export (F_{out})

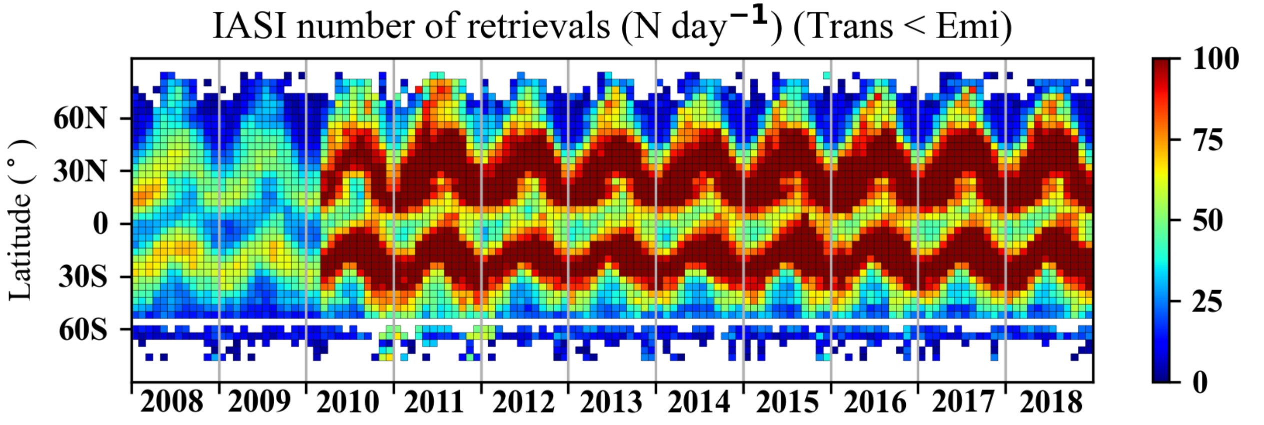
$$\tau_{mod} = \frac{M_{NH_3}}{D_{NH_3} + D_{NH_4^+}}$$

Number of retrievals

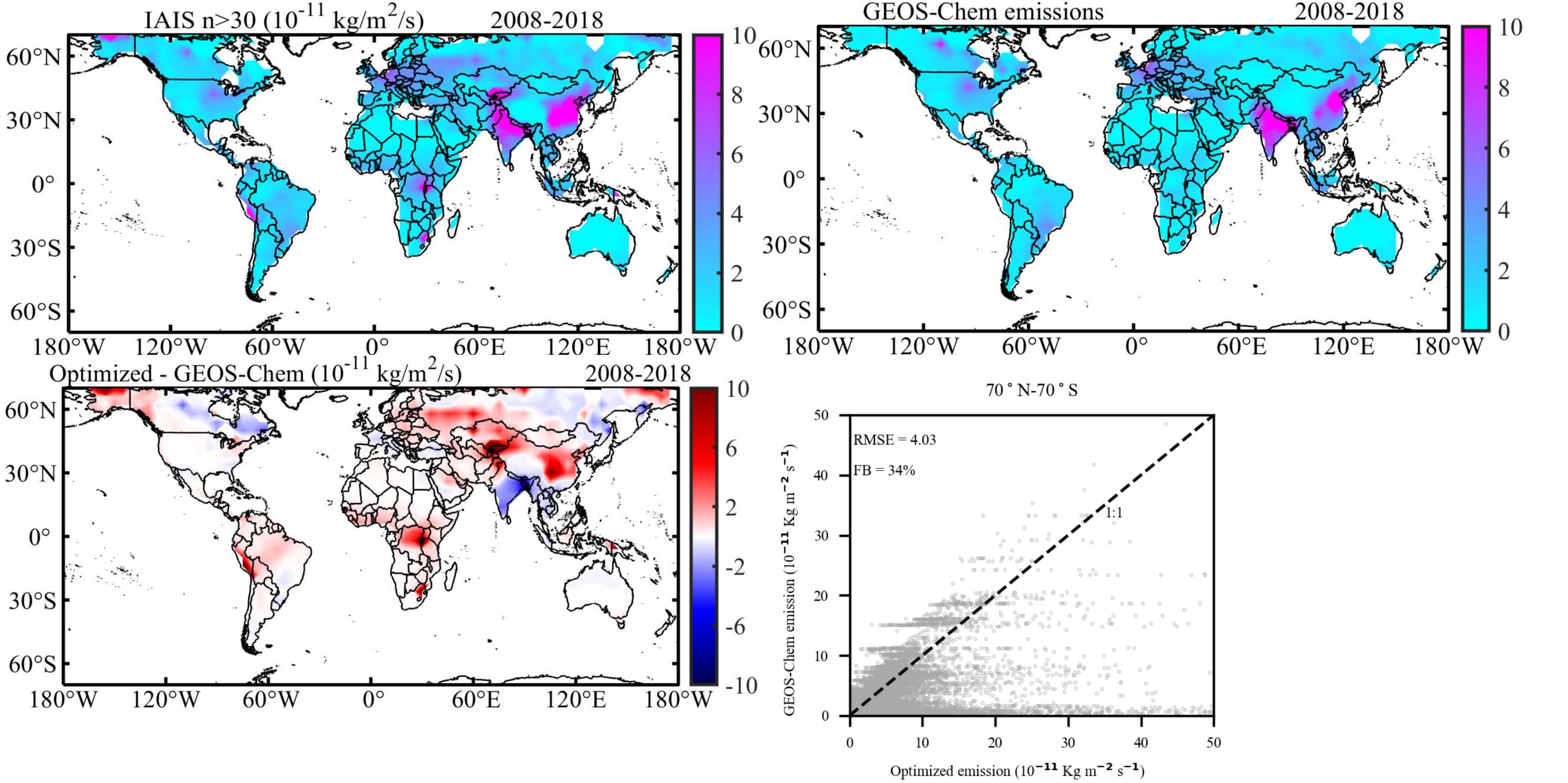




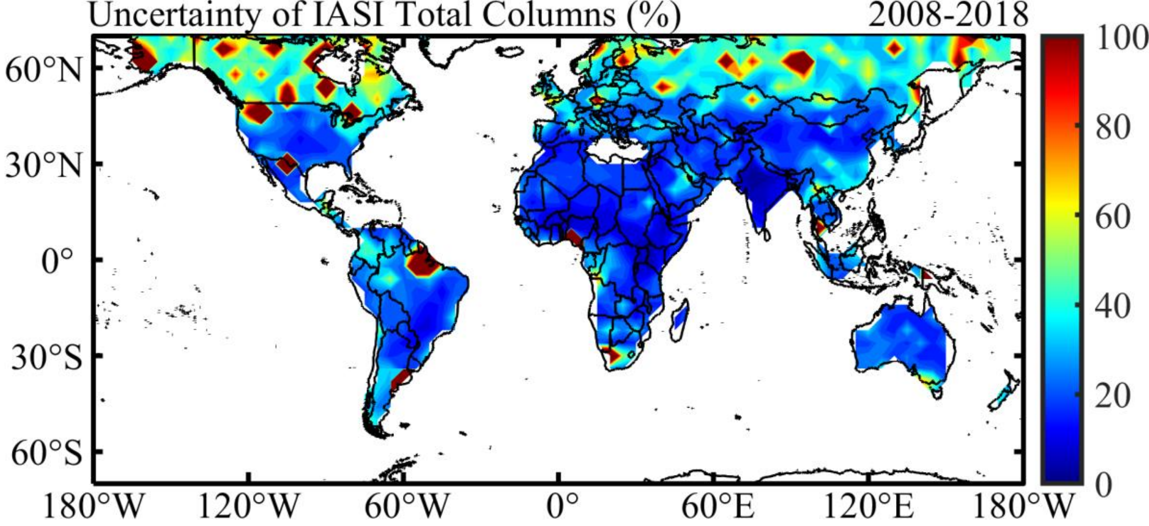




Optimized versus GEOS-Chem



Uncertainty



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

- Lifetime:
  - Transportation (-)
- Transportation (+)/emission ratio: 0.3/0.5/0.8/1/1.2/1.5/2
- Number of retrievals: 0/15/25/30/35/40/50

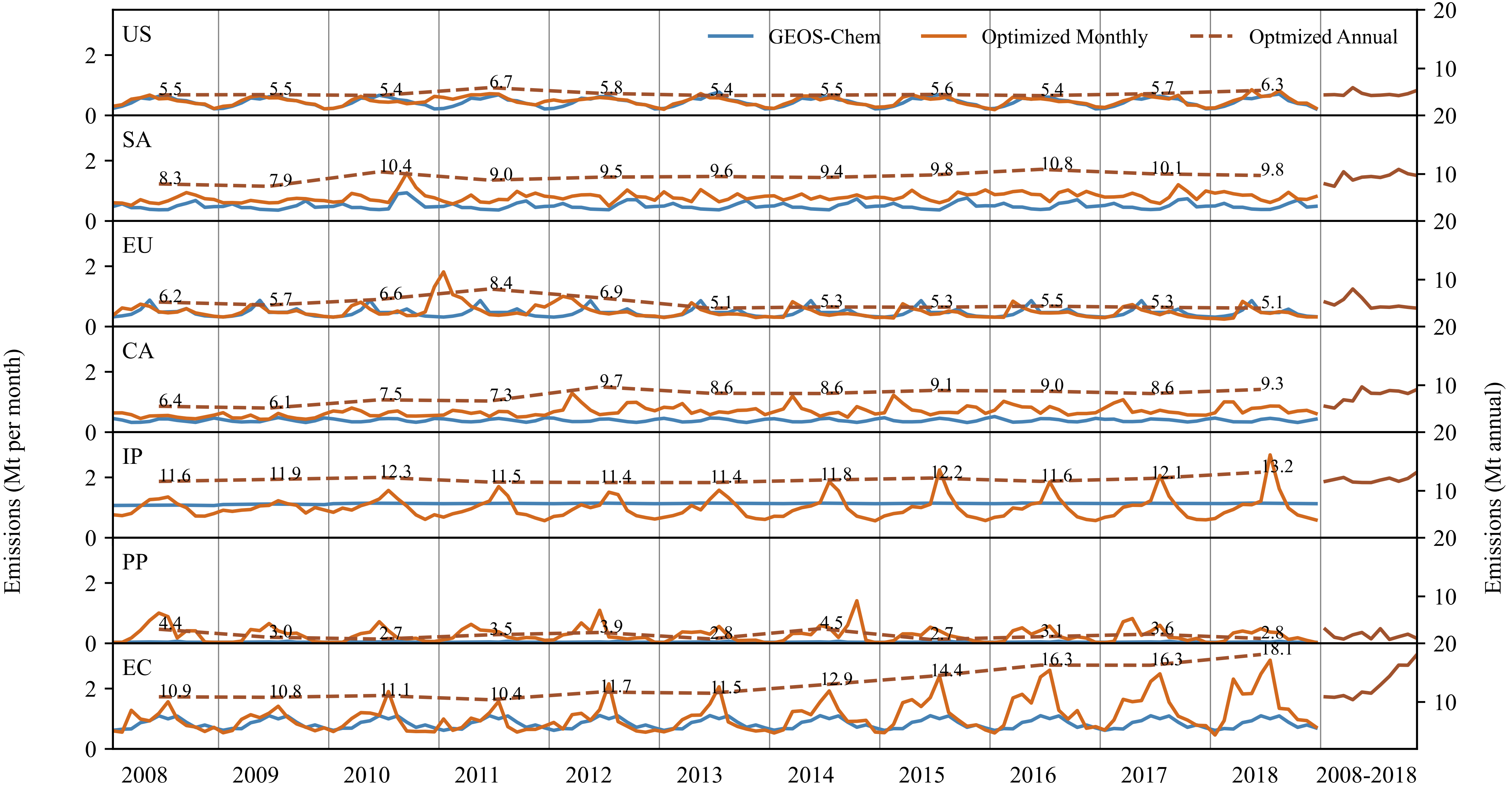
Parameter perturbed	Averaged emission	Standard deviation
Initial: ratio < 1, n > 30	94	7.4
Transportation (-)	101	9.1
Transportation(+)/emission ratio < 0.3	88	5.4
Transportation(+)/emission ratio < 0.5	90	6.2
Transportation(+)/emission ratio < 0.8	92	7.7
Transportation(+)/emission ratio < 1.2	95	7.9
Transportation(+)/emission < 1.5	96	8.8
Transportation(+)/emission < 2	97	10.7
Number of retrievals > 0	119	13
Number of retrievals > 15	99	11
Number of retrievals > 25	95	9.6
Number of retrievals > 35	92	7.2
Number of retrievals > 40	90	7.7
Number of retrievals > 50	85	13.9

Table 1. Model ensemble simulations using different emissions for ammonia that were used in the calculations of uncertainty. Uncertainties were calculated as the standard deviation of the surface concentrations of ammonia from the 10 ensemble members for the 10-year period (2008–2017).

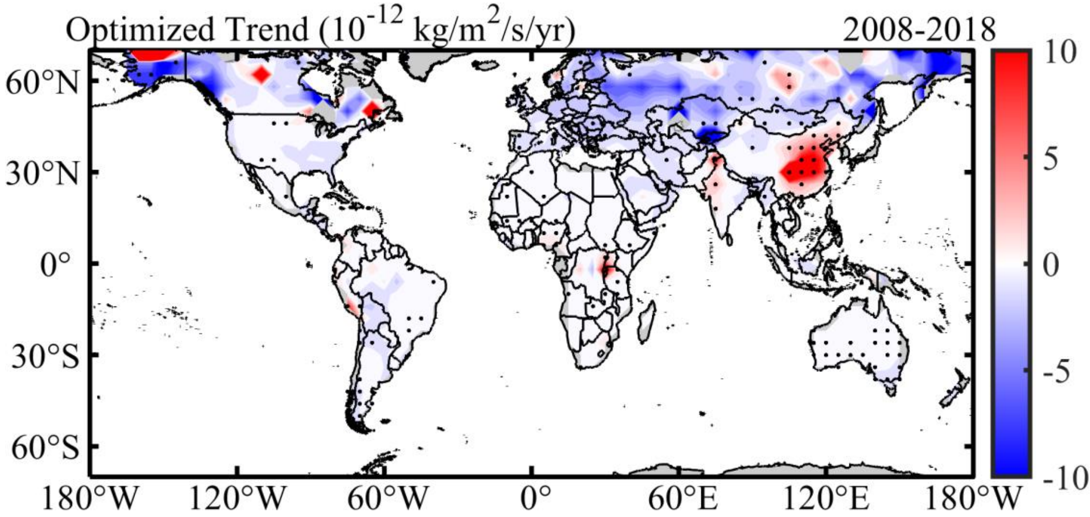
	Parameter perturbed	10-year average emissions ($Tg\,yr^{-1}$)
Ensemble 1	$d_k = 0$ in Eq. (2)	121 ± 50.6
Ensemble 2	$d_k = 10$ in Eq. (2)	175 ± 33.3
Ensemble 3	$d_k = 20$ in Eq. (2)	189 ± 28.7
Ensemble 4	$d_k = 60$ in Eq. (2)	218 ± 15.5
Ensemble 5	$d_k = 100$ in Eq. (2)	208 ± 51.8
Ensemble 6	$d_k = 500$ in Eq. (2)	223 ± 26.5
Ensemble 7	EGG	65 ± 2.8
Ensemble 8	VD0.5	189
Ensemble 9	NE	213 ± 18.1
Ensemble 10	VDgrlf	201 ± 10.4

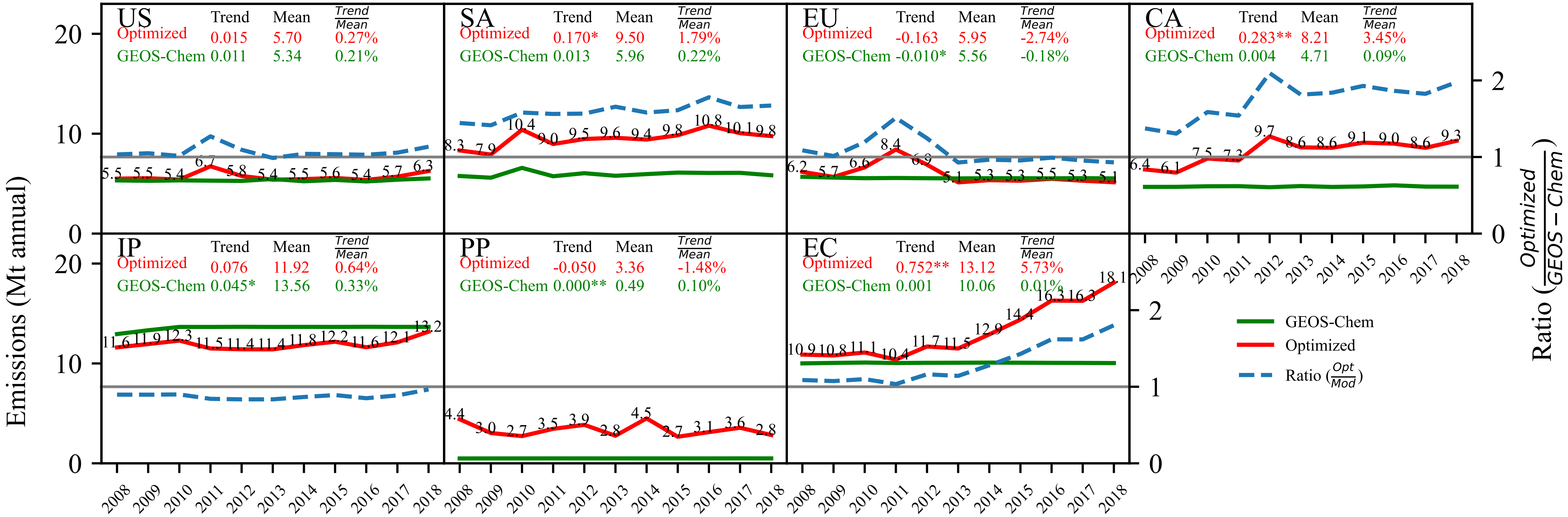
(Evangeliou et al, 2021)

Optimized emissions timeseries

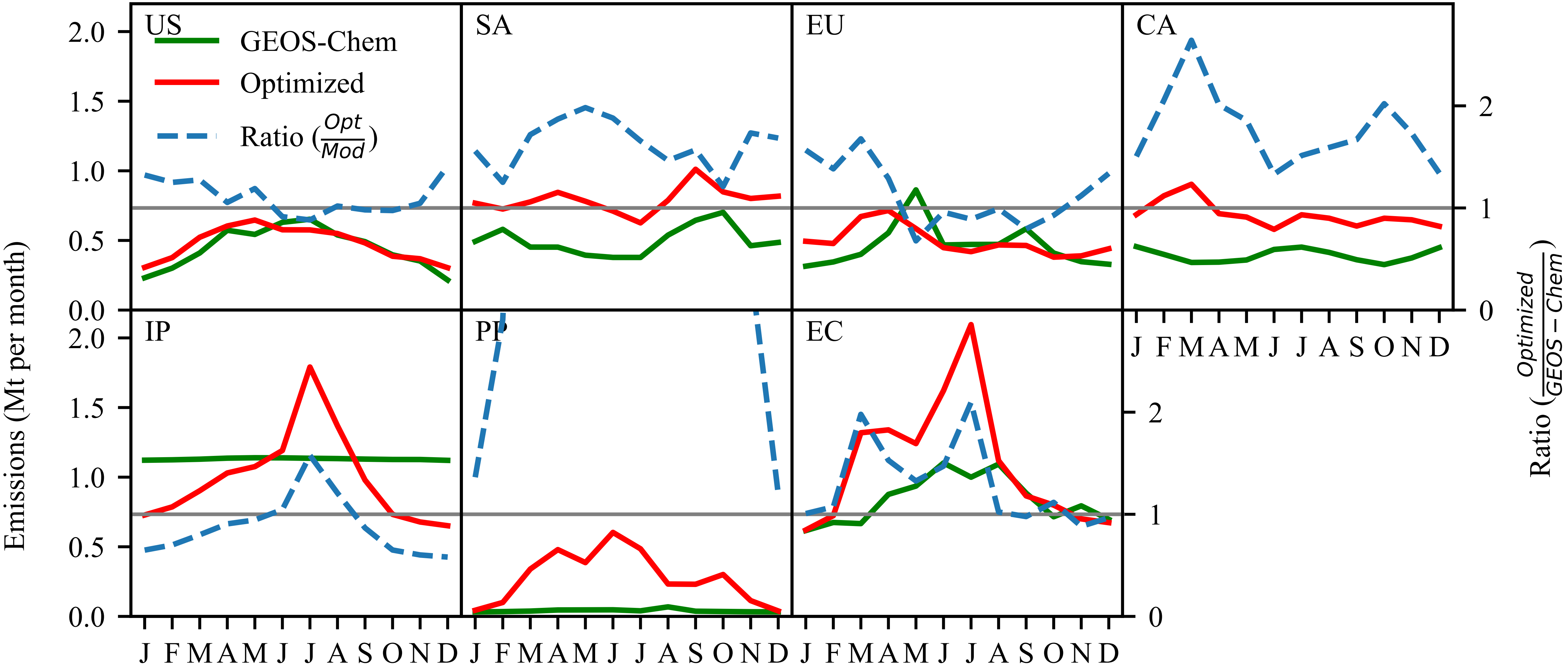


Optimized emissions trend ($p < 0.05$)

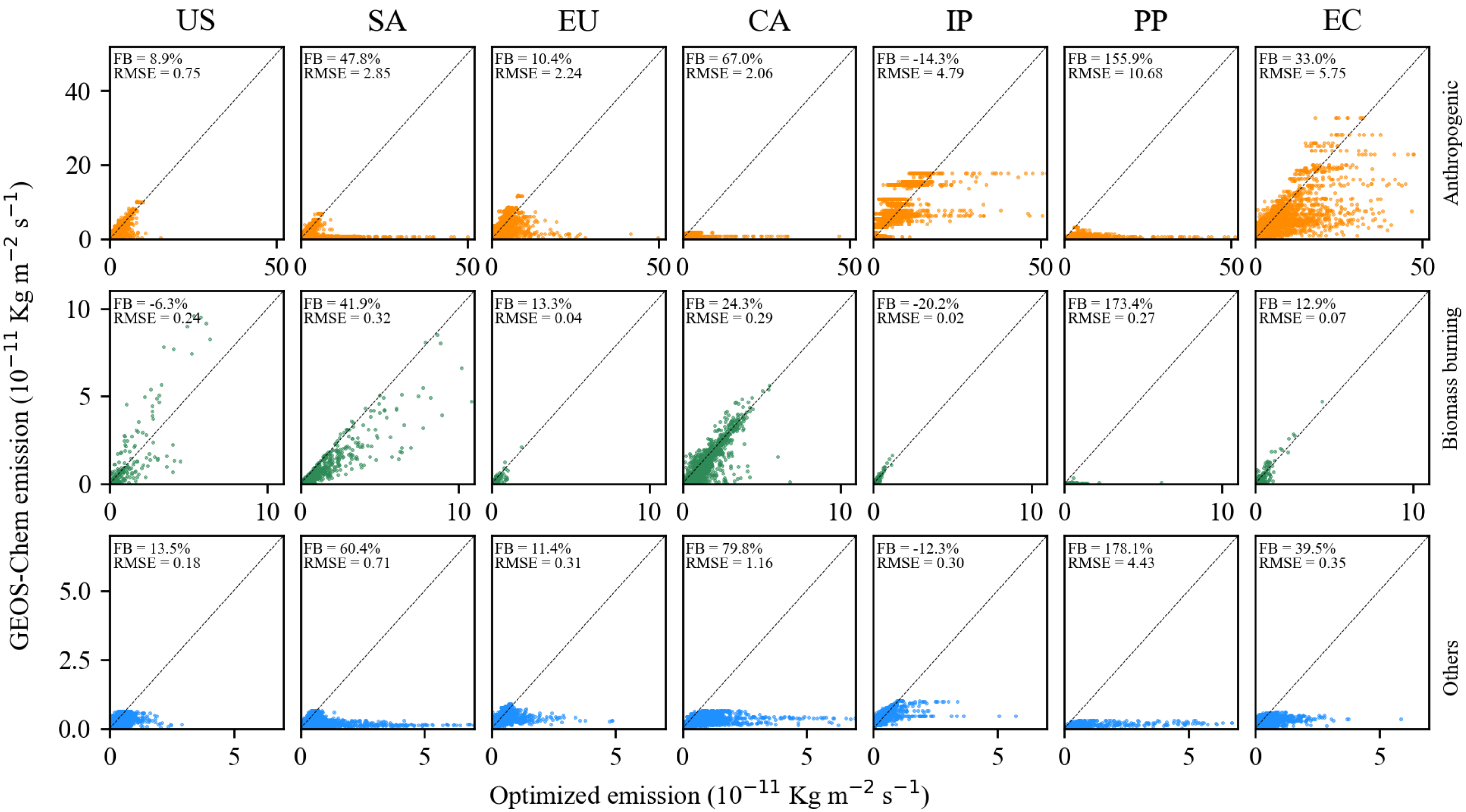




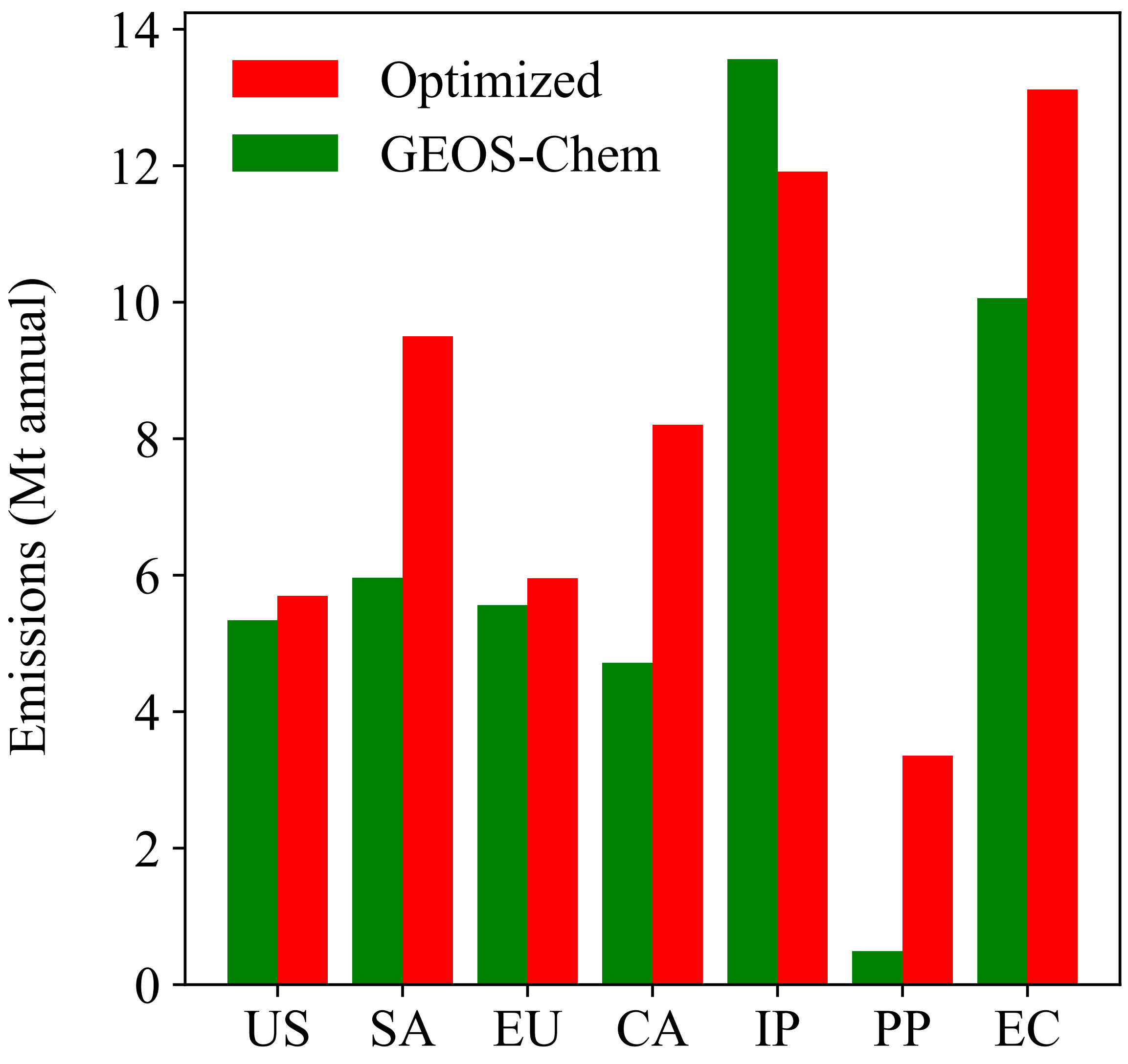
Optimized emissions monthly variations

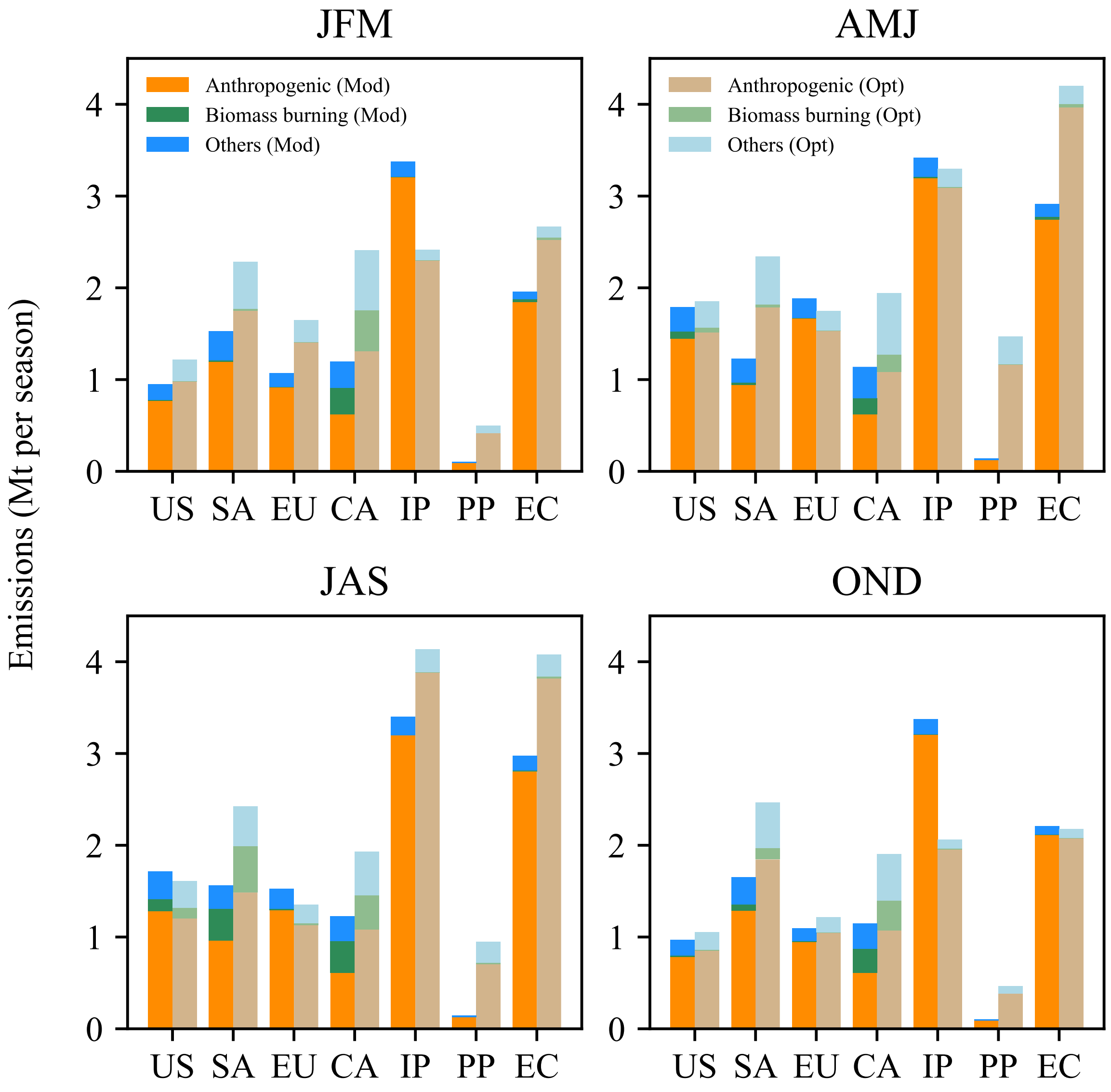


Optimized versus GEOS-Chem by sectors

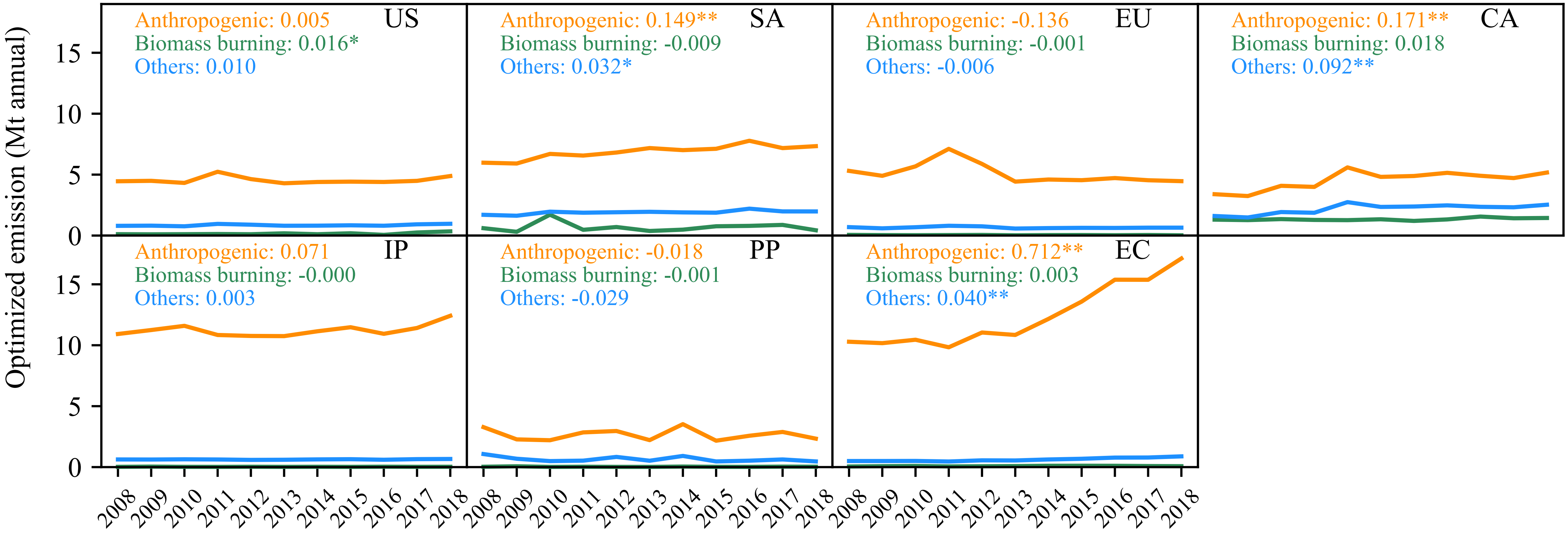


Optimized emissions by sectors

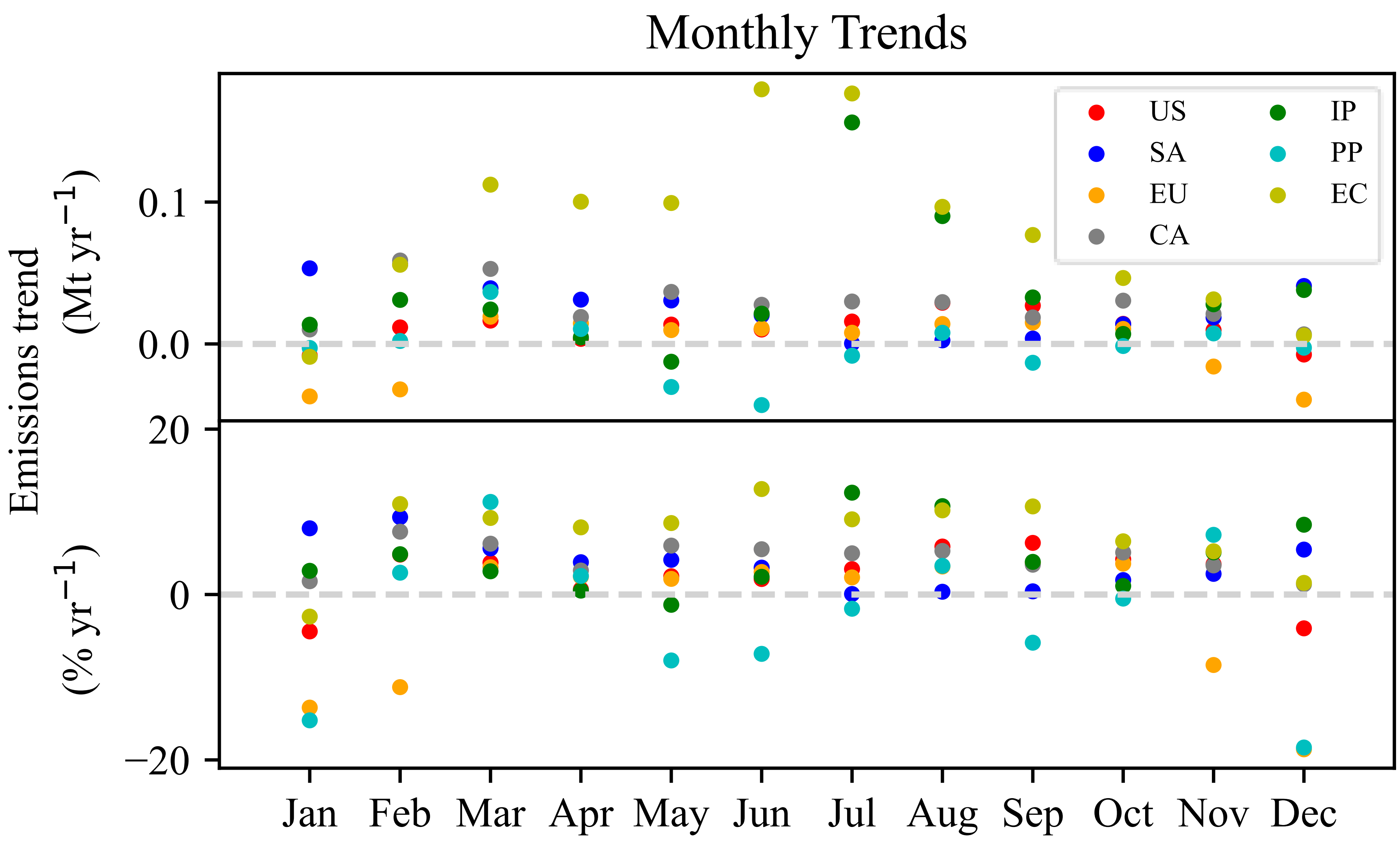




Optimized emissions trend by sectors

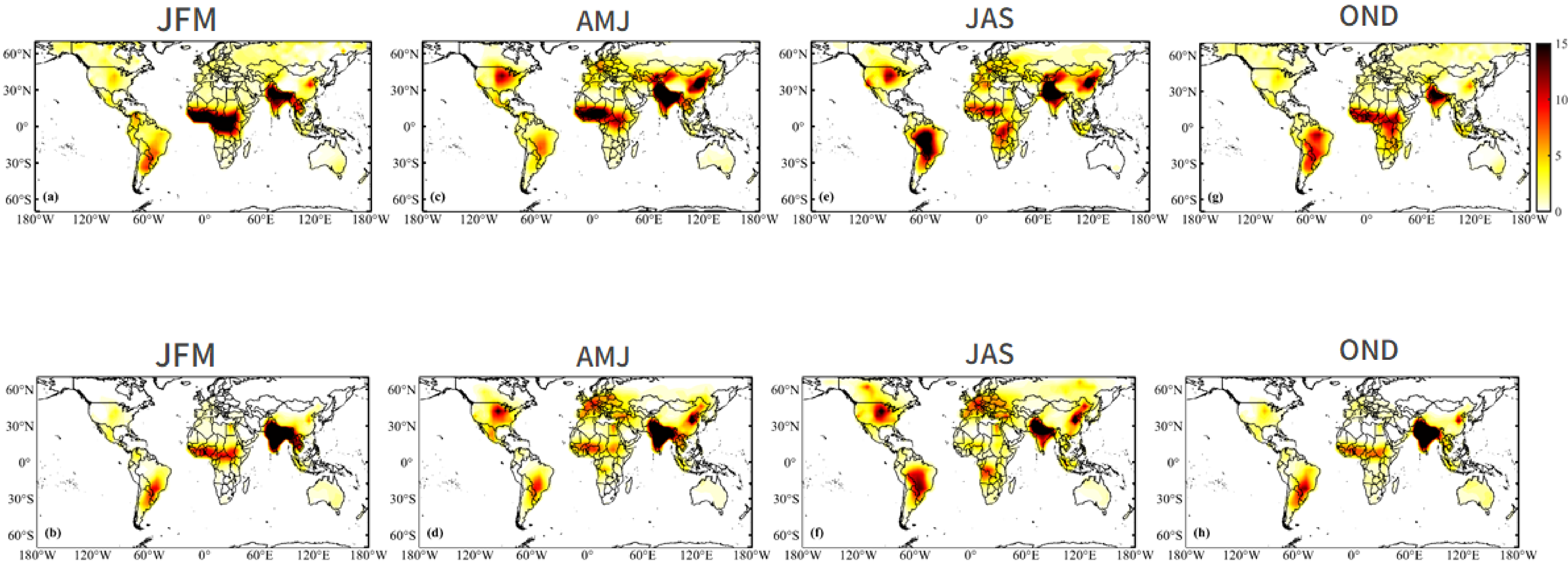


Optimized emissions monthly trend



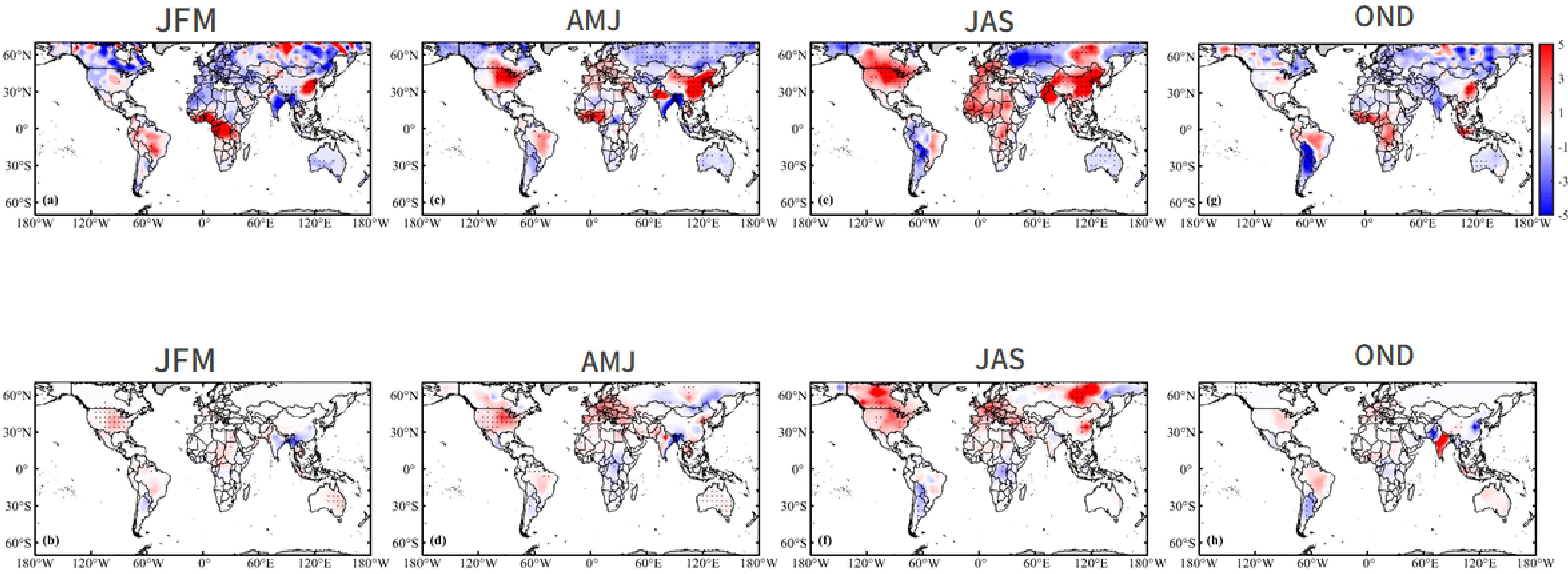
NH_3 Seasonal Concentrations

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

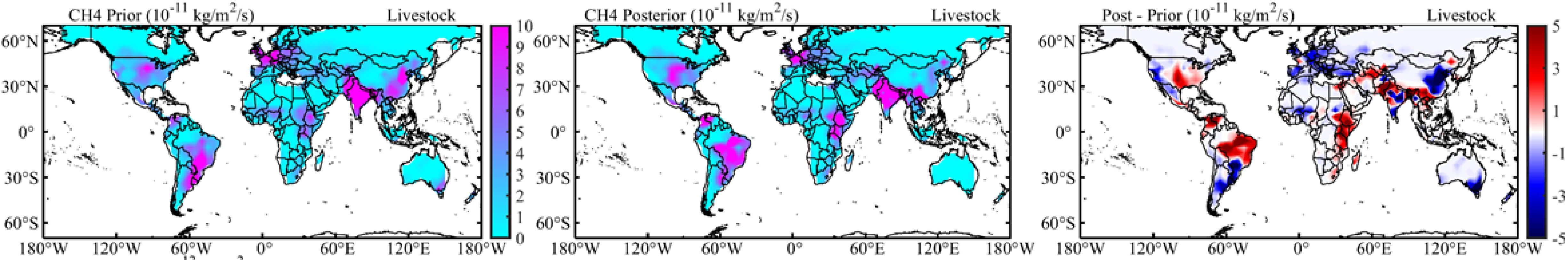


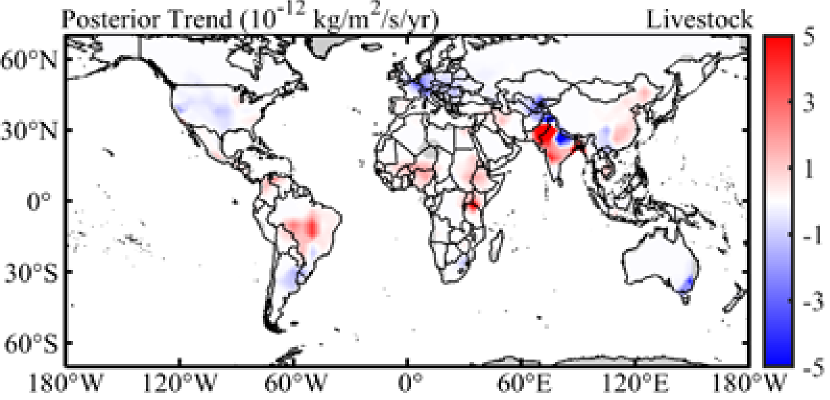
NH_3 Seasonal Concentrations

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

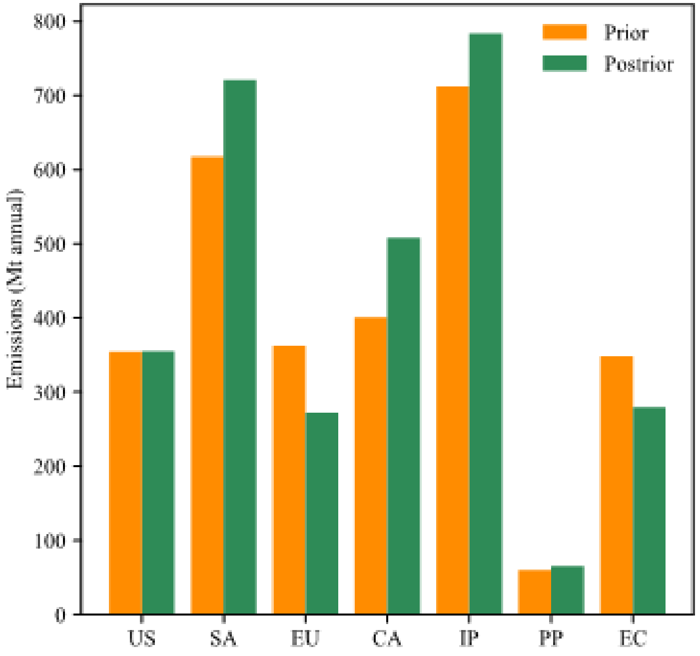


CH4 livestock emissions





	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



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

Table SI1: NH_3 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

- $\Omega = \sum_{i=1}^{47} c_{\mathrm{i}} \times rho_i \times h_i \times k$
  - Ω: **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

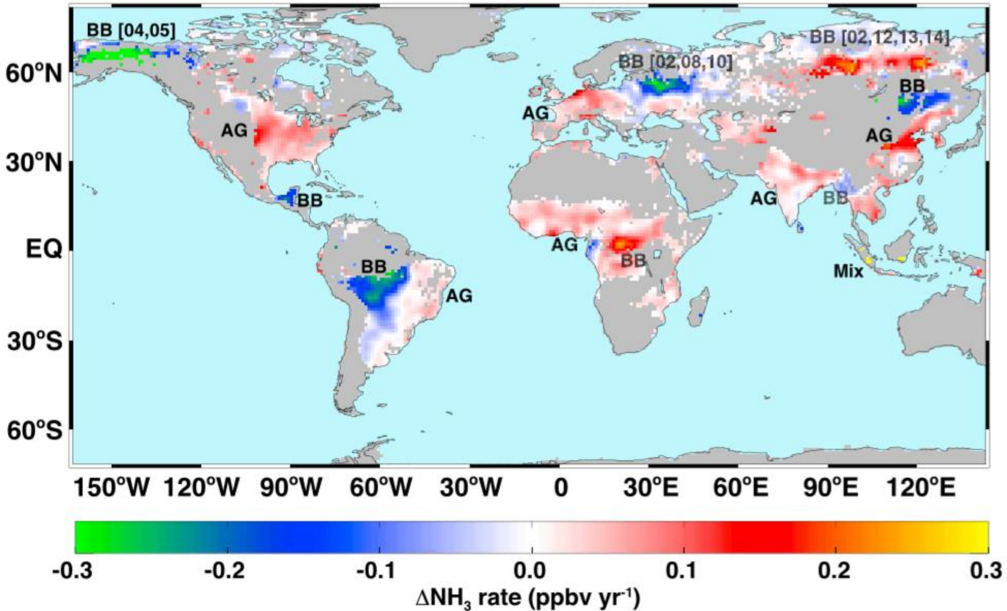
- Latitude: 46 degrees
  - 88°-90°: 2x5 to 1x1, 2 degrees
  - 0-88°: 4x5 to 1x1, 44 degrees
- Method:
  - Step1: mask ocean, set as NaN
  - Step2: calculate mean value in each upscaling grid

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 worlds 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