Observed and simulated Ammonia concentration 11

IASI data and GEOS-Chem simulation

2021.1

- Accomplished:
  - 1. calculate the model lifetime (effective) from the GEOS-Chem
  - 2. mean distribution of K (NH3/NH4)
  - 3. mean distribution and timeseries of IASI emissions (two estimated ways) based on GEOS-Chem's lifetime
- 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/chemistry/deposition rate of change
- 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 of trend

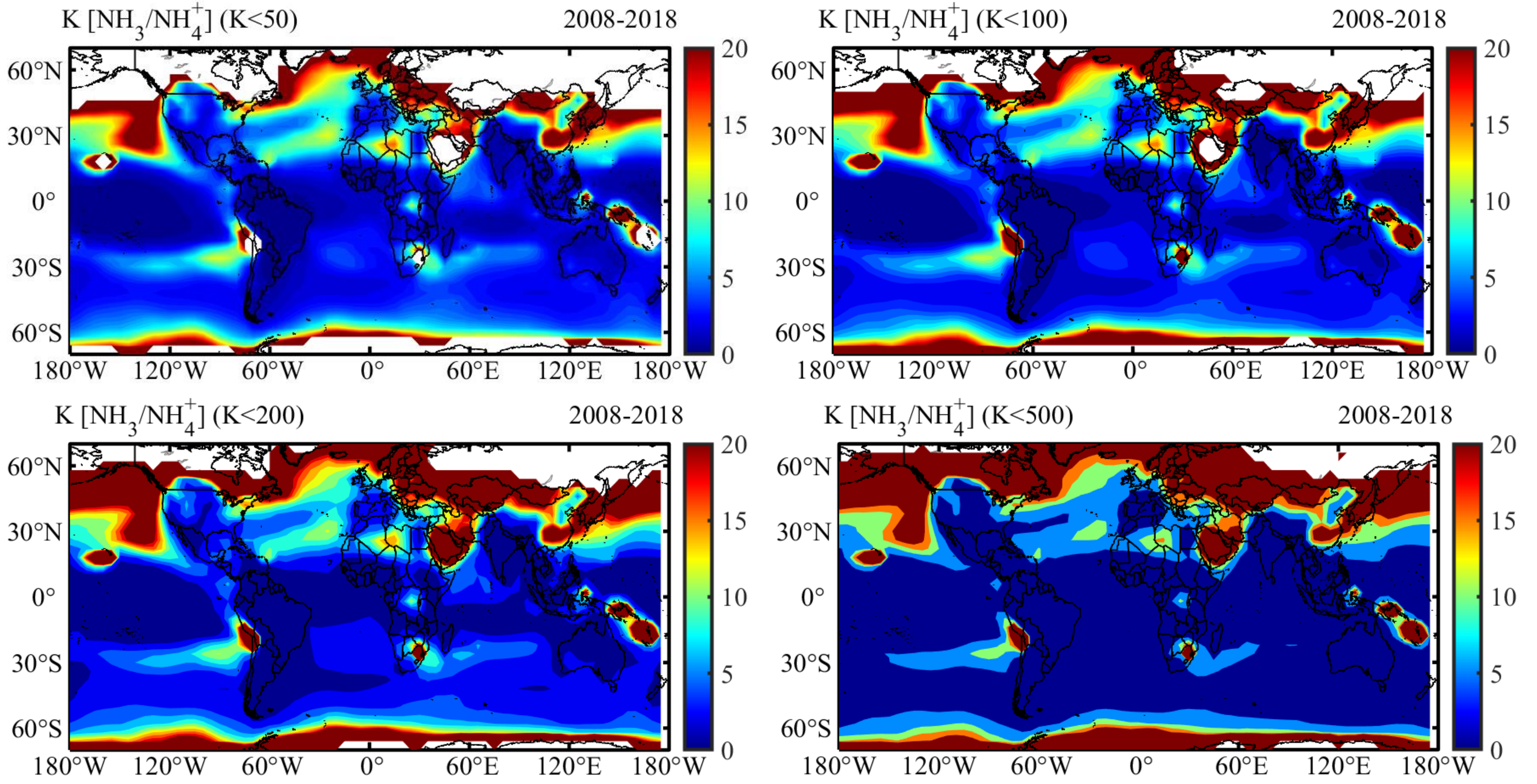
Lifetime (Ammonia-water equilibrium)

- $\frac{1}{\tau} = \frac{1}{\tau_{trans}} + \frac{1}{\tau_{chem}} + \frac{1}{\tau_{depo}}$
  - t_{trans}: NH3 transport in and out
  - t_{chem}: NH3 chemical loss
  - t_{depo}: NH3 deposition (dry deposition and wet deposition)
- $\frac{dC(t)}{dt} = S(t) - \frac{C(t)}{\tau(t)}$ species mass balance equation (Croft et al., 2014)
  - $C(t)$: the NH3 concentration at time t
  - $S(t)$: the time-dependent source emission fluxes
  - $\tau(t)$: the removal timescale

- $NH_3 + H_2O \rightleftharpoons NH_3 \cdot H_2O$
- $H_{NH_3} = \frac{[NH_3 \cdot H_2O]}{\mathrm{p}_{NH_3}}$, Henry's Law
- $NH_3 \cdot H_2O \rightleftharpoons NH_4^+ + OH^-$
- $K_1 = \frac{[NH_4^+][OH^-]}{[NH_3 \cdot H_2O]}$
- $[NH_4^+] = \frac{K_1 * H_{NH_3}}{[OH^-]} \mathrm{p}_{NH_3} = K_{NH_4^+/NH_3} * C_{NH_3}$
- $K_{NH_4^+/NH_3} = \frac{[NH_4^+]}{C_{NH_3}}$
- $\tau_{mod} = \frac{M_{NH_3} + M_{NH_4^+}}{-(\nabla M_{NH_3,}^{drydep,wetdep} + \Delta M_{NH_3}^{drydep,wetdep})} \rightarrow \frac{(K_{NH_4^+/NH_3}^{\mathrm{mod}} + 1) M_{NH_3}}{-\Delta M_{NH_3,NH_4^+}^{drydep,wetdep}}$

- A quasi-equilibrium between sources and removals of ammonia (Dentener and Crutzen, 1994), $S(t) = 0 \rightarrow \frac{dC(t)}{dt} = -\frac{C(t)}{\tau(t)}$
- $\tau(t) = -\frac{C(t)}{\nabla C(t)}$
  - $\Delta C(t)$: NH3 total concentration rate of change
- $\frac{-1}{\tau_{mod}} = \frac{\nabla C_{trans}}{C} + \frac{\nabla C_{chem}}{C} + \frac{\nabla C_{drydep}}{C} + \frac{\nabla C_{wetdep}}{C} \rightarrow \tau_{mod} = \frac{C}{-(\Delta C_{trans} + \Delta C_{chem} + \Delta C_{drydep} + \Delta C_{wetdep})} = \frac{M}{-\Delta M_{trans,chem,drydep,wetdep}}$
  - $\Delta M_{trans,chem,drydep,wetdep}$: the total mass rate of change due to transport, chemistry, deposition
  - M: the NH3 mass in each grid (column)
- $\tau_{mod} = \frac{M}{-\Delta M_{trans,chem,drydep,wetdep}}$

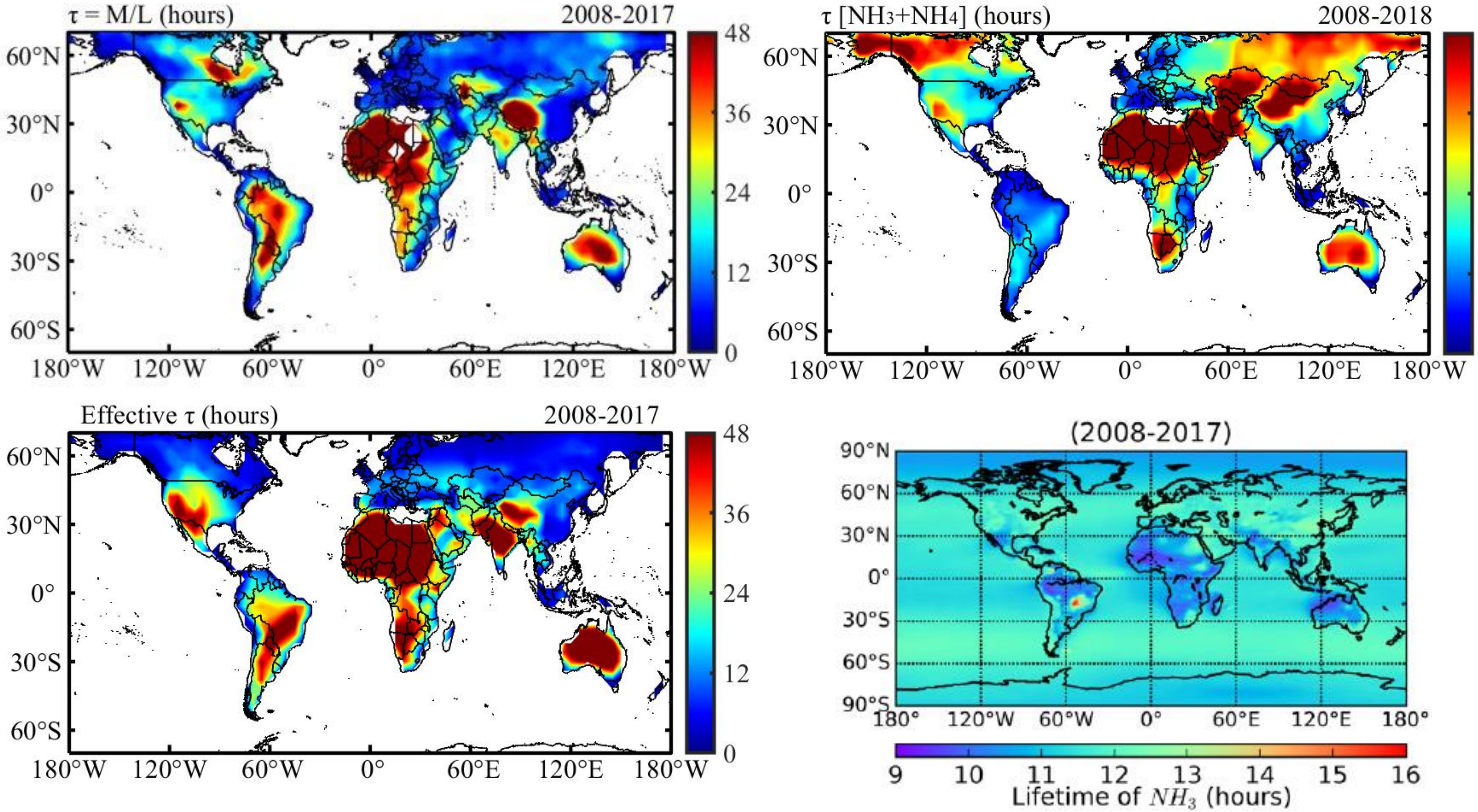
Average K[NH4/NH3]



Effective lifetime and correspond emissions

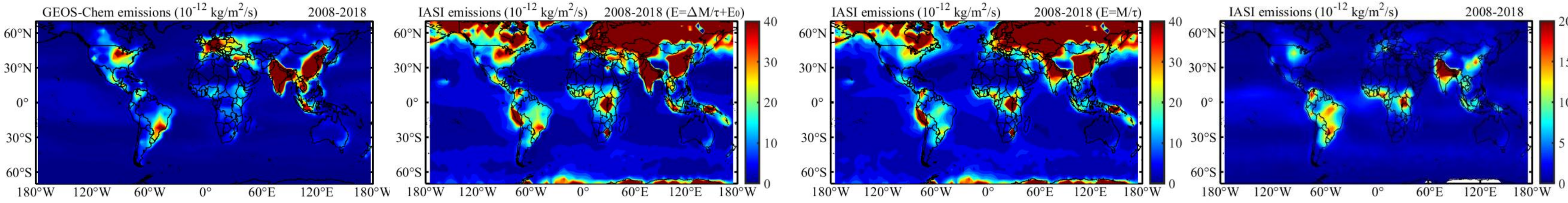
- $\tau_{mod} = \frac{(K^{\text{mod}}_{NH_4^+/NH_3}+1)M_{mod}}{-\Delta M^{drydep,wetdep}_{NH_3,NH_4^+}}$
- $\tau'_{mod} = \frac{\tau_{mod}}{K^{\text{mod}}_{NH_4^+/NH_3}+1} = \frac{M_{mod}}{-\Delta M^{drydep,wetdep}_{NH_3,NH_4^+}}$
- $E_{obs} = \frac{M_{\text{obs}}}{\tau'_{mod}} = \frac{M_{\text{obs}}}{M_{mod}}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

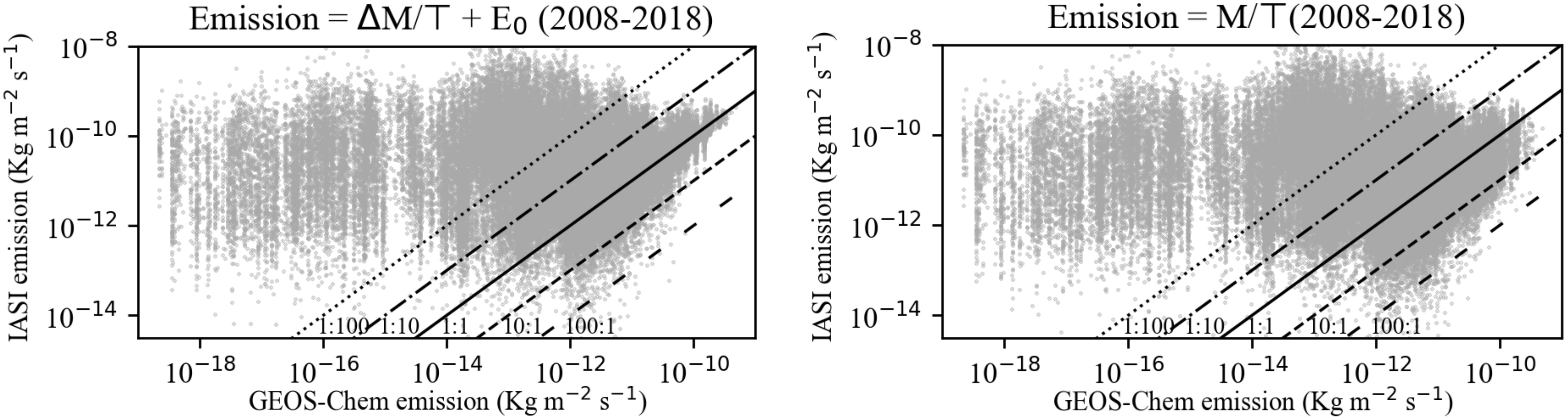
Average lifetime of NH3

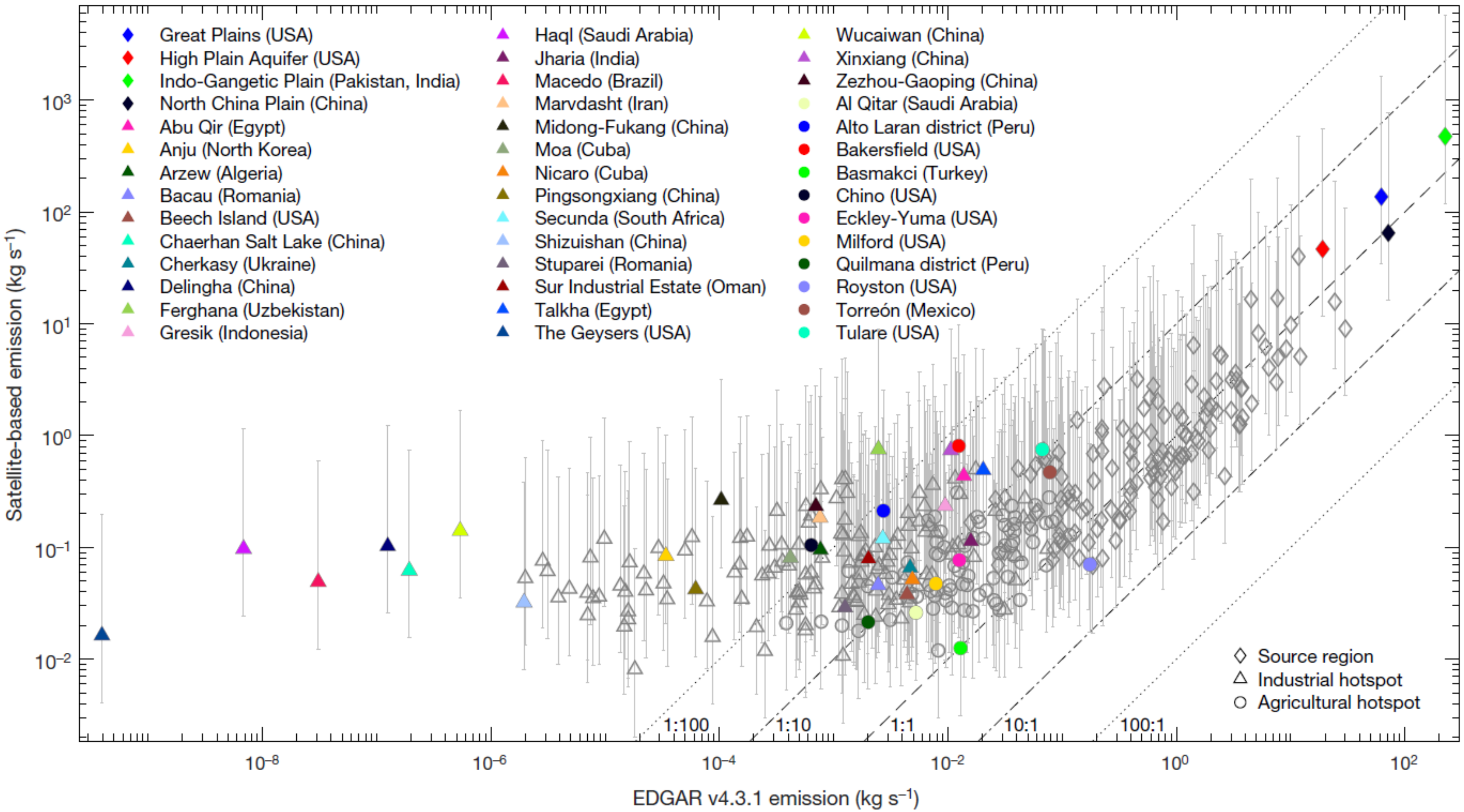


(Evangeliou et al., 2020)

IASI emission fluxes versus GEOS-Chem emission inventory

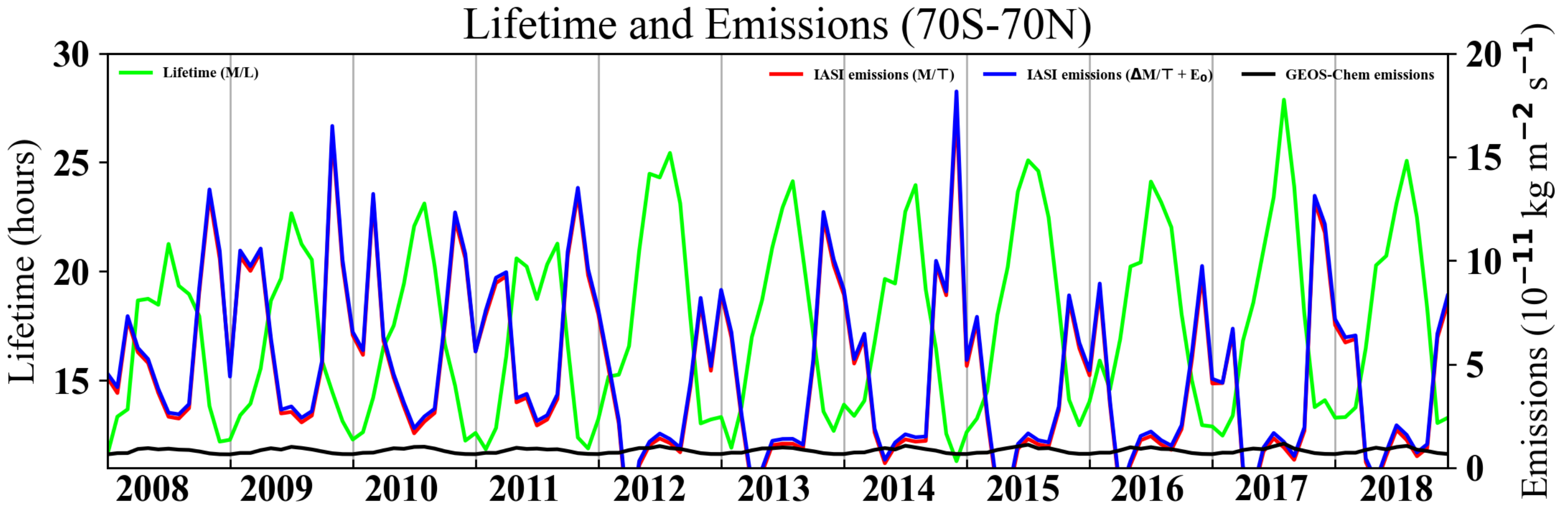


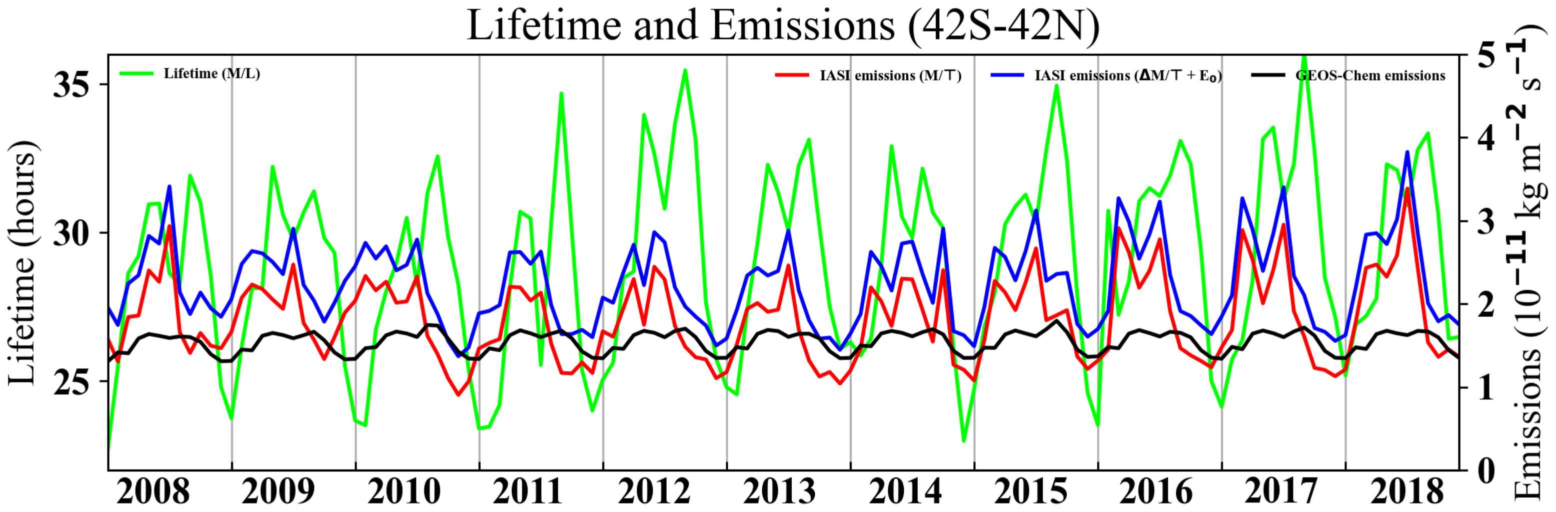


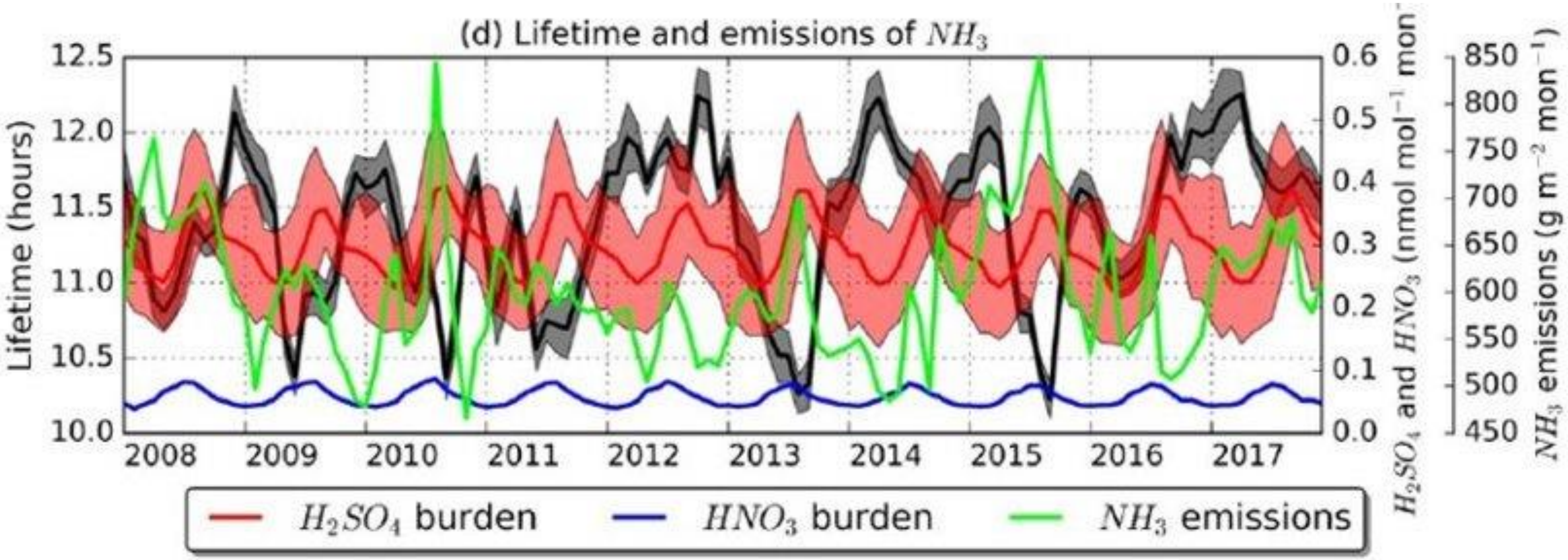


(Damme et al., 2018)

monthly timeseries of lifetime and emissions

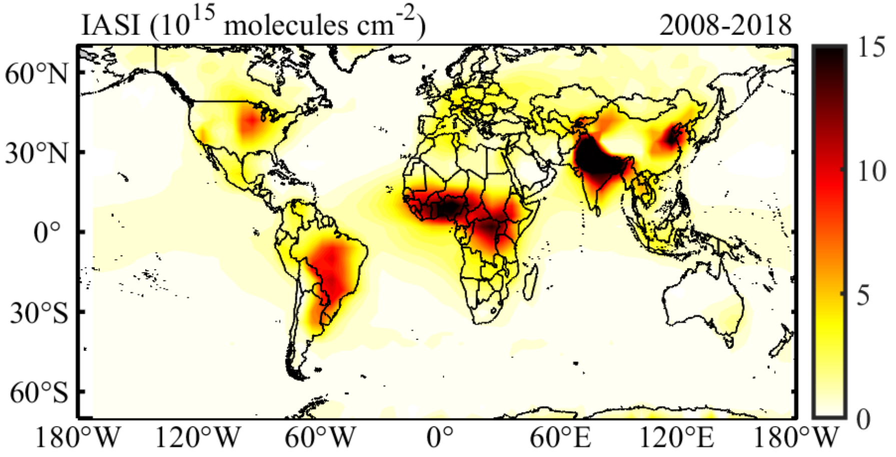


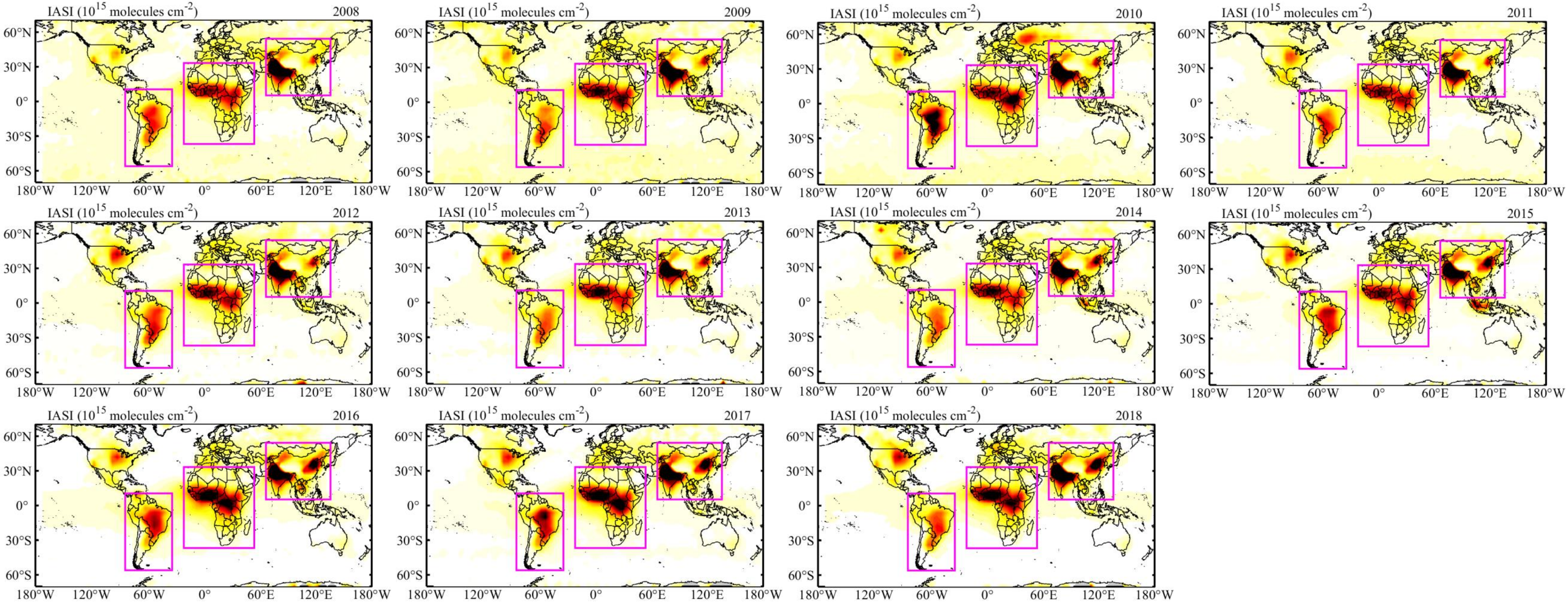




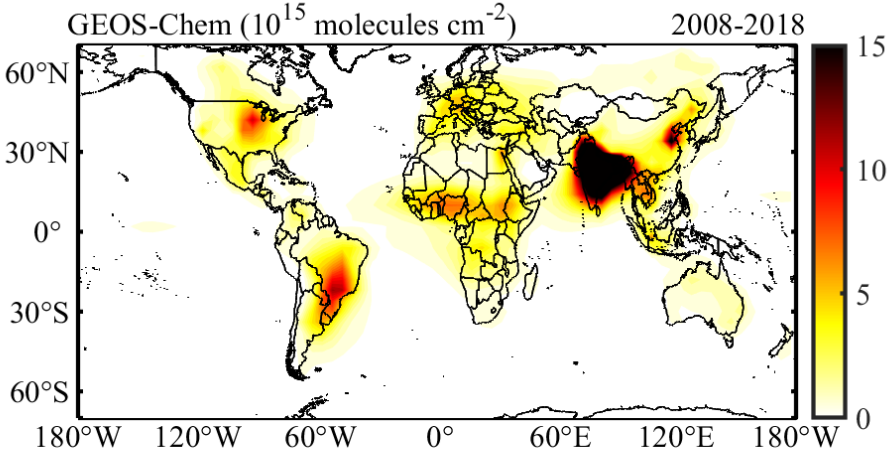
(Evangeliou et al., 2020)

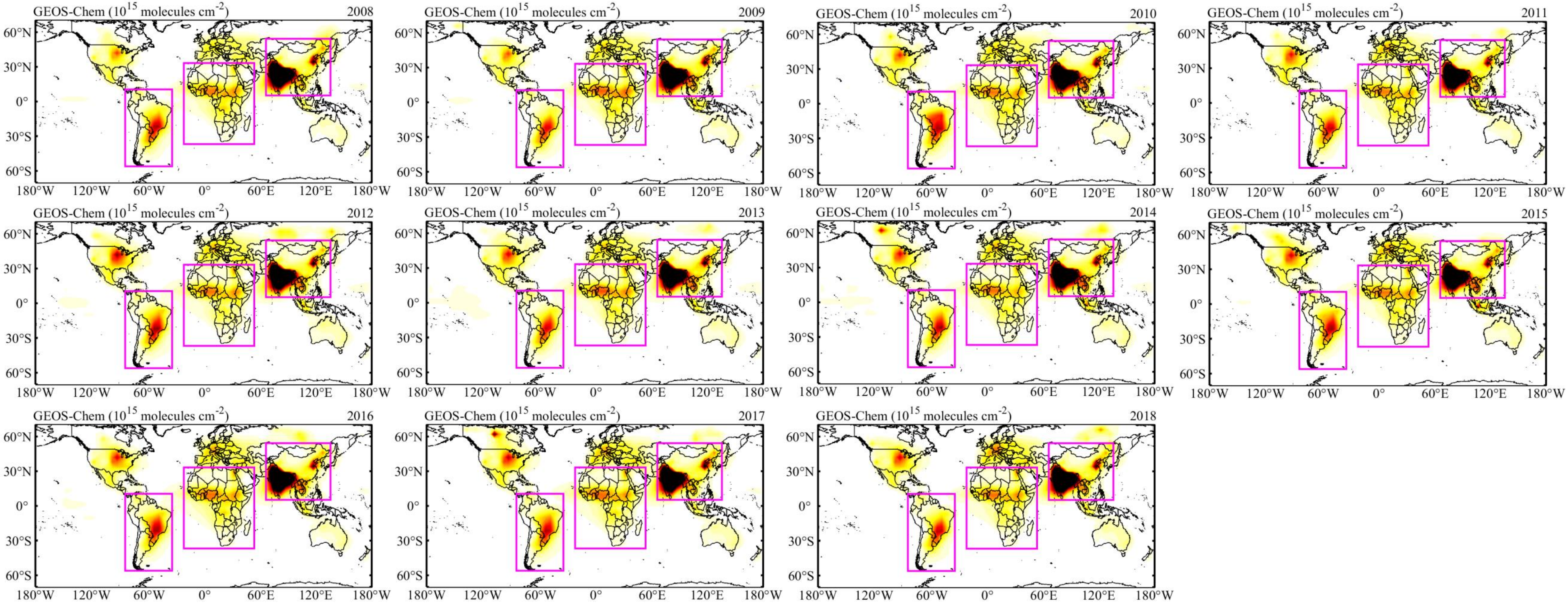
IASI mean in annual concentration distribution



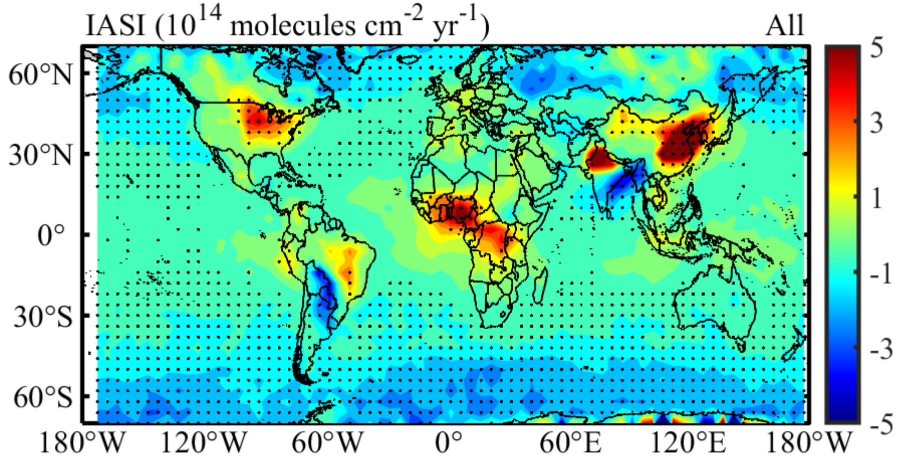


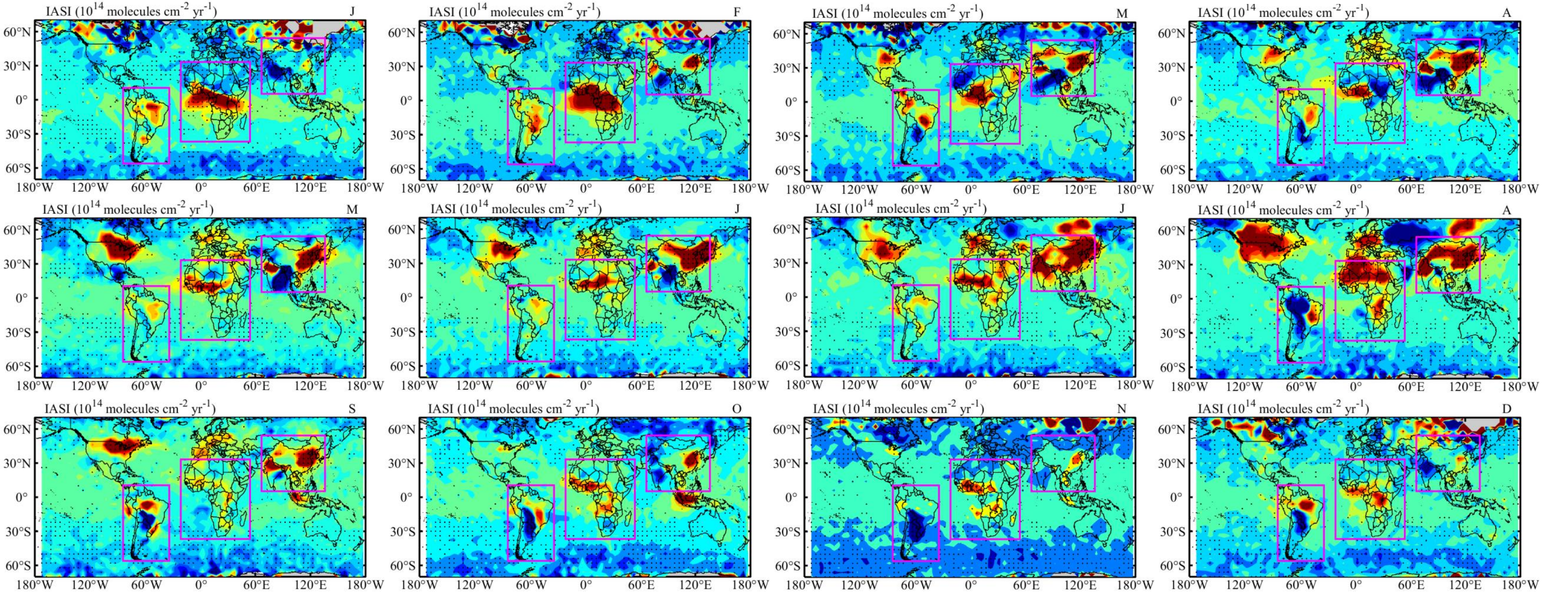
GEOS-Chem mean in annual concentration distribution



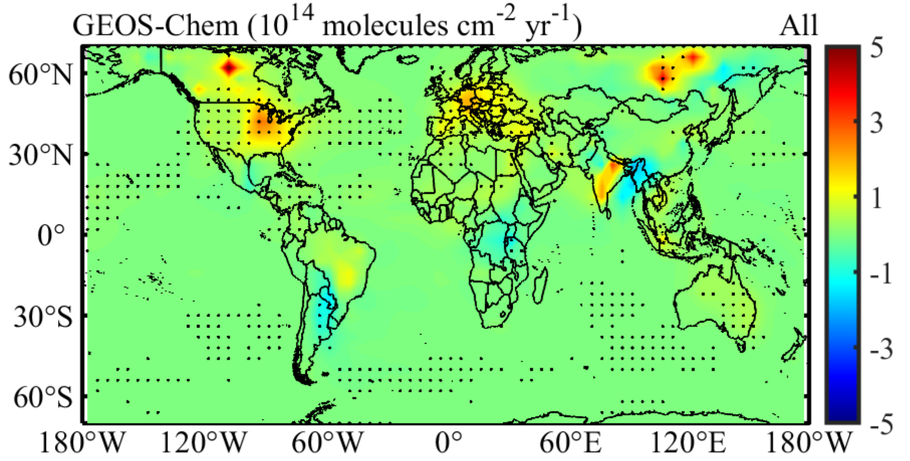


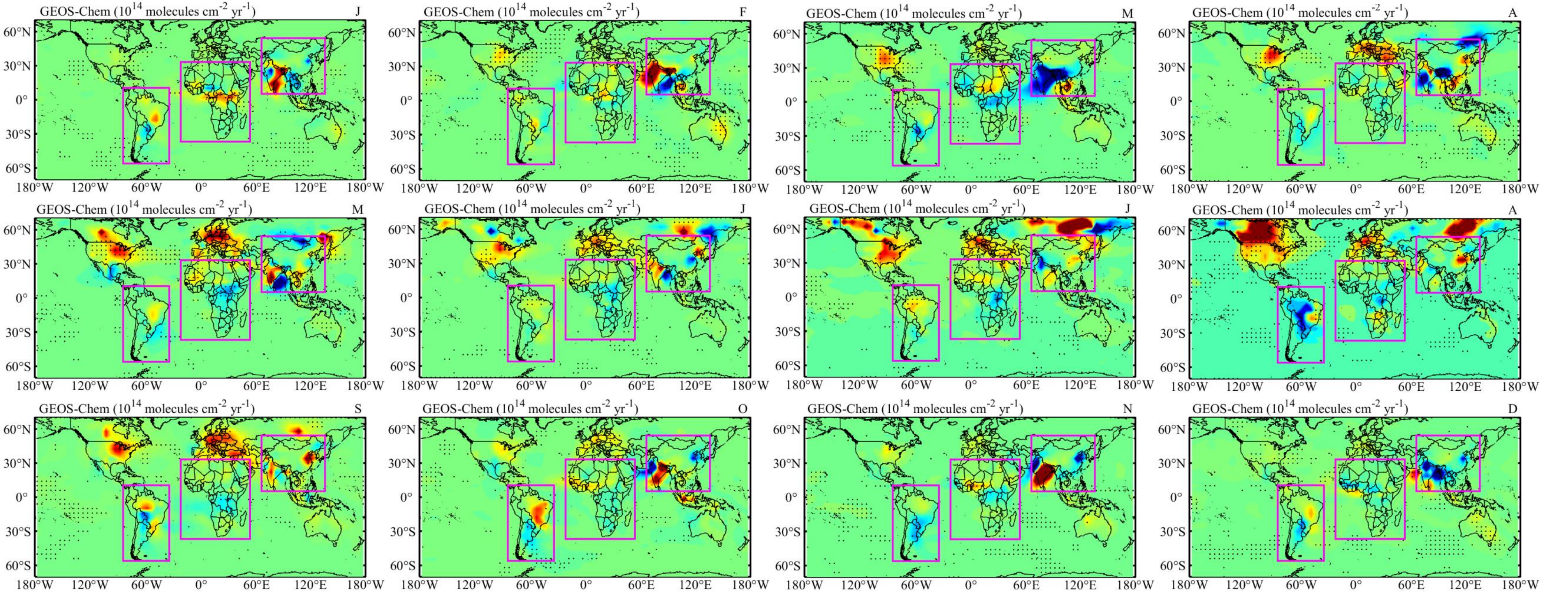
IASI trend in monthly concentration distribution



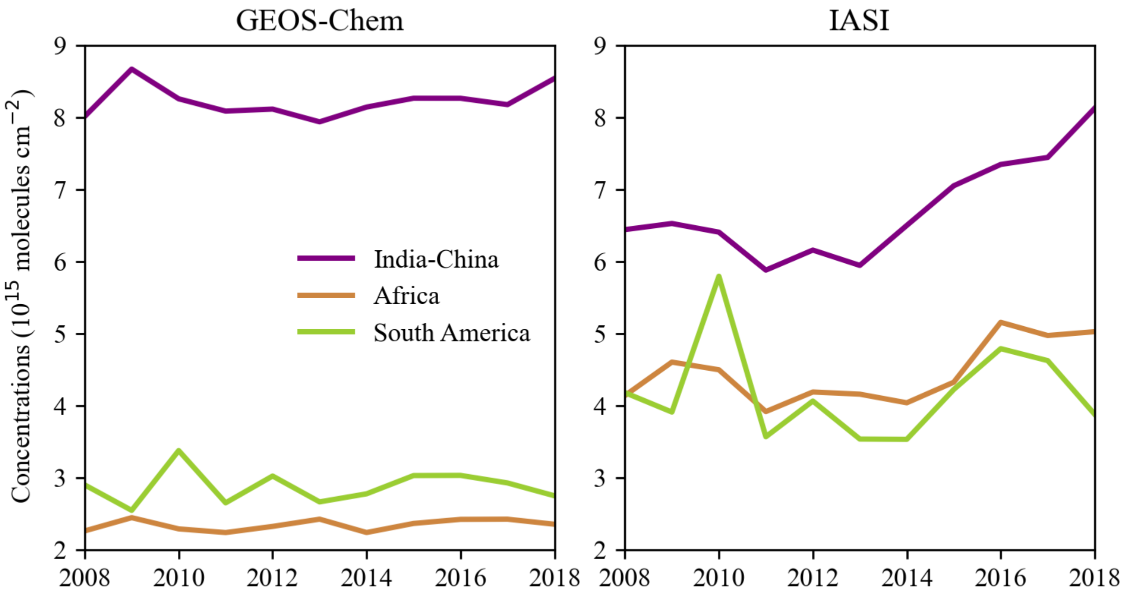


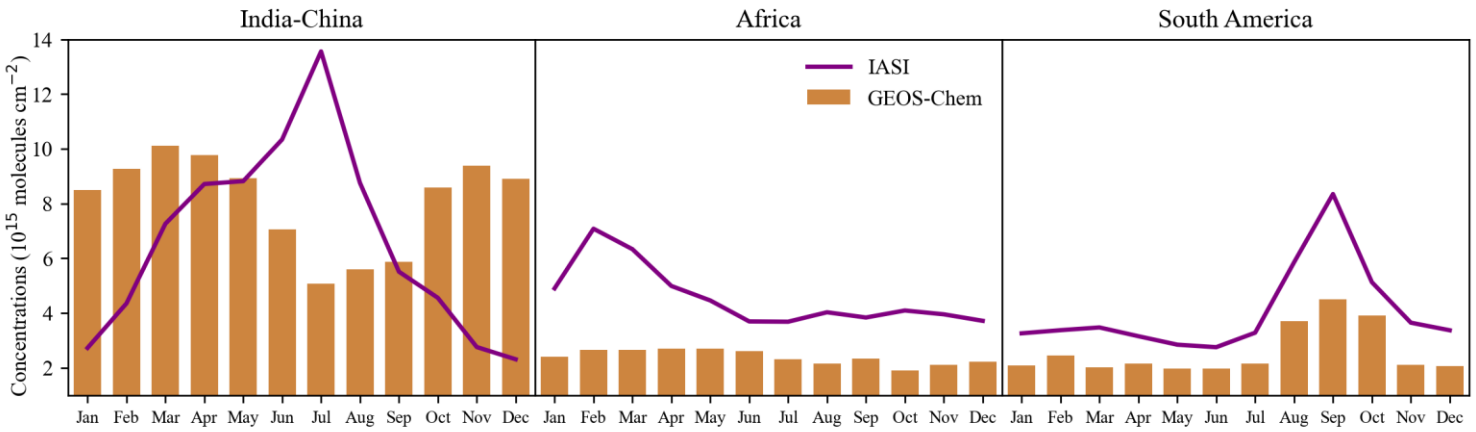
GEOS-Chem trend in monthly concentration distribution





Annual and mean monthly concentration mean of GEO-Chem and IASI over the India-China, Africa and South America





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
- $\tau_{\mathrm{mod}} = \frac{M_{mod}}{E_{mod}}$
- $E_{obs} = \frac{M_{\mathrm{obs}}}{\tau_{\mathrm{mod}}} = M_{\mathrm{obs}} * \frac{E_{mod}}{M_{mod}} = E_{mod} * \frac{c_{\mathrm{obs}}}{c_{mod}}$

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

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

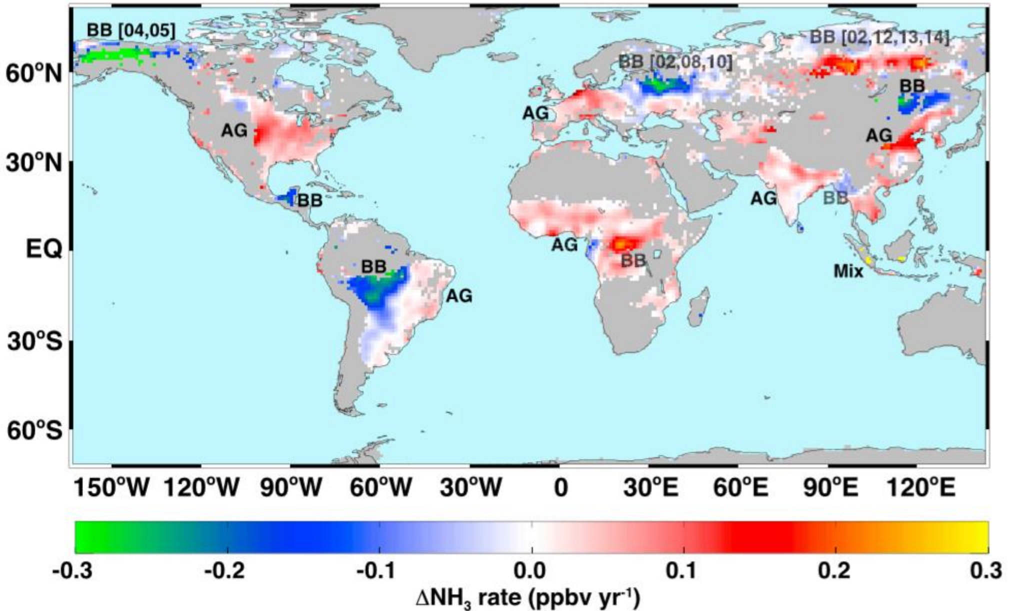
- Latitude: 46 degrees
  - 88°-90°: 2x5 to 1x1, 2 degrees
  - 0-88°: 4x5 to 1x1, 44 degrees
- Method:
  - Step1: mask ocean, set as NaN
  - Step2: calculate mean value in each upscaling grid

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 C_2H_6 for 2010
  - XIAO_C3H8: C_2H_6 and C_3H_8
  - AFCID: PM2.5 dust emission
- Natural
  - GEIA_NH3: 1990 (obsolete now)
  - SEABIRD_DECAYING_PLANTS: the oceanic emissions of acetaldehyde
  - NH_3: the Arctic seabird
- 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