Decadal trends in US OMI NO_2 observations and the role of the upper troposphere

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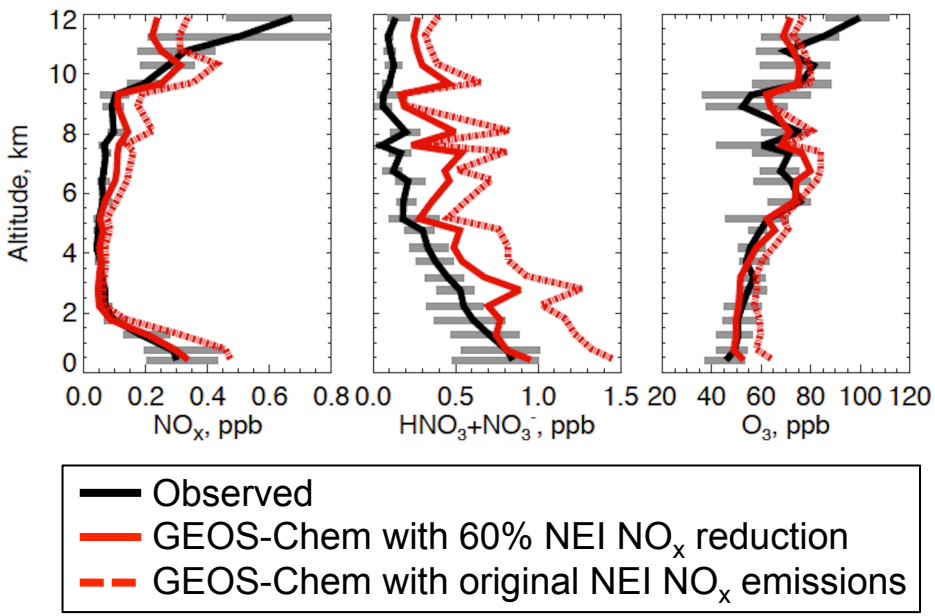
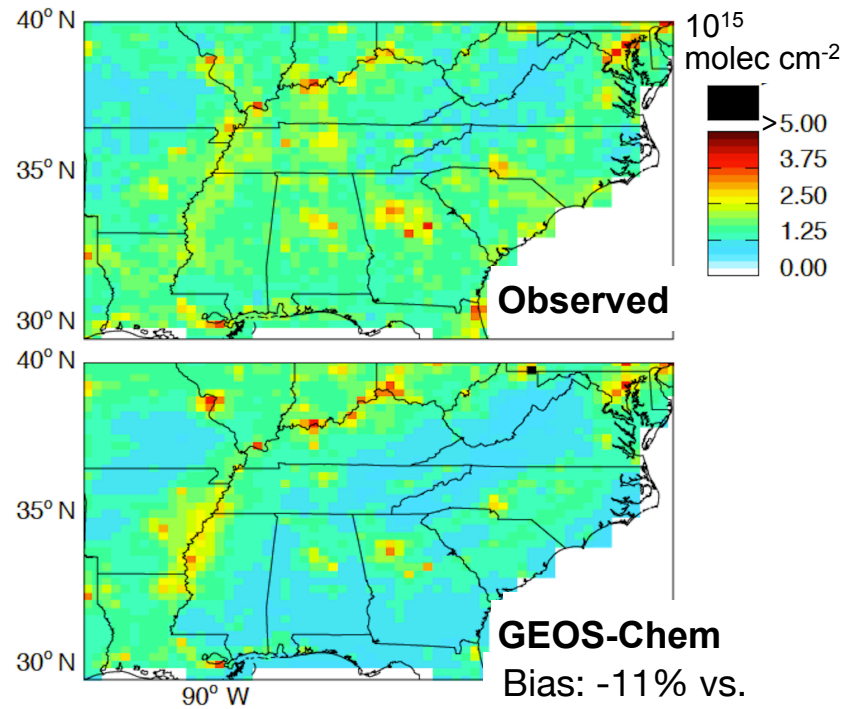
Previous work showed 60% reduction of EPA National Emission Inventory (NEI) NO_x required to reconcile GEOS-Chem with observations

Median vertical profiles from SEAC^{4}RS aircraft campaign show impact of reducing NEI NO_x emissions by 60%



Travis et al. (2016)

Emission reduction consistent with OMI NO_2 observations

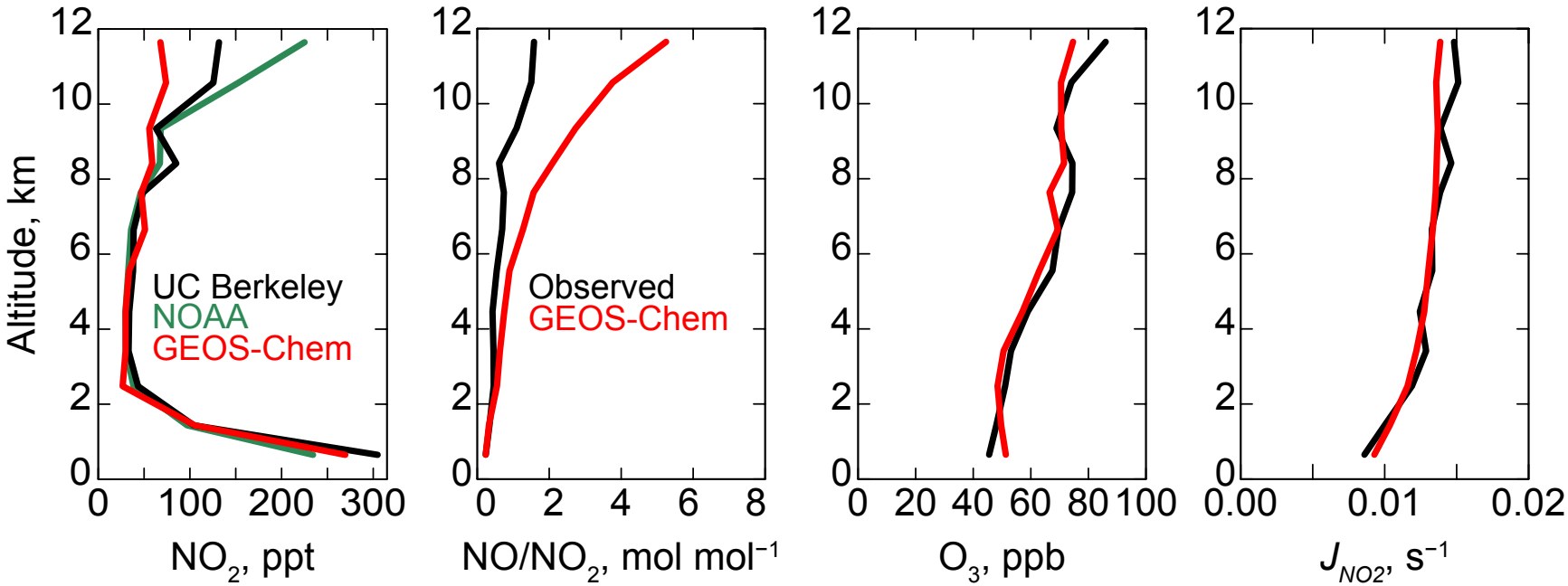


Bias: -11% vs. +26-31% for original emissions

Remaining model bias compared to satellite observations due to underestimate of NO_2 in upper troposphere

Observations show elevated NO_2 concentrations in the upper troposphere not captured by models

SEAC^{4}RS daytime median vertical profiles

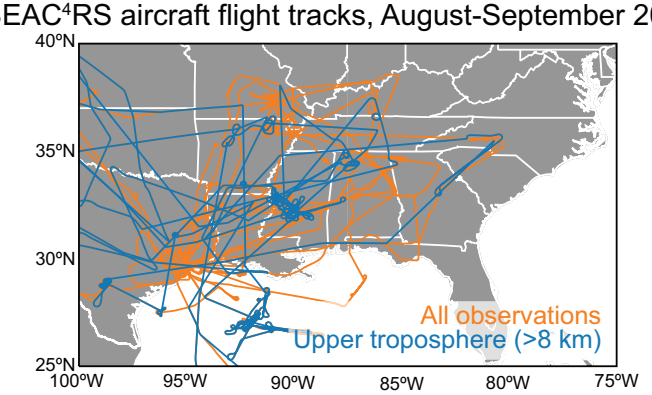


NOAA measurements: T. Ryerson
Berkeley measurements: R. Cohen

O_3 measurements: T. Ryerson

J_{NO2} measurements: S. Hall

SEAC^{4}RS aircraft flight tracks, August-September 2013

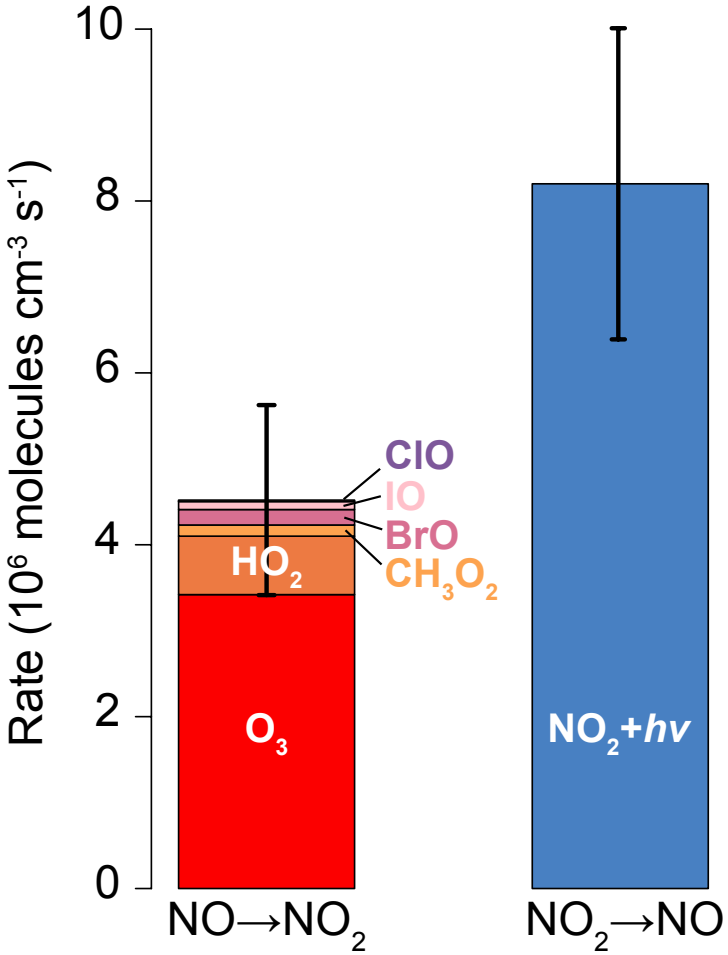


- GEOS-Chem overestimates the NO/NO_2 ratio by over a factor of 2 in the upper troposphere
- Boundary layer (<2 km) accounts for only 20-35% of the total NO_2 column, the upper troposphere (>8 km) accounts for 35-50%

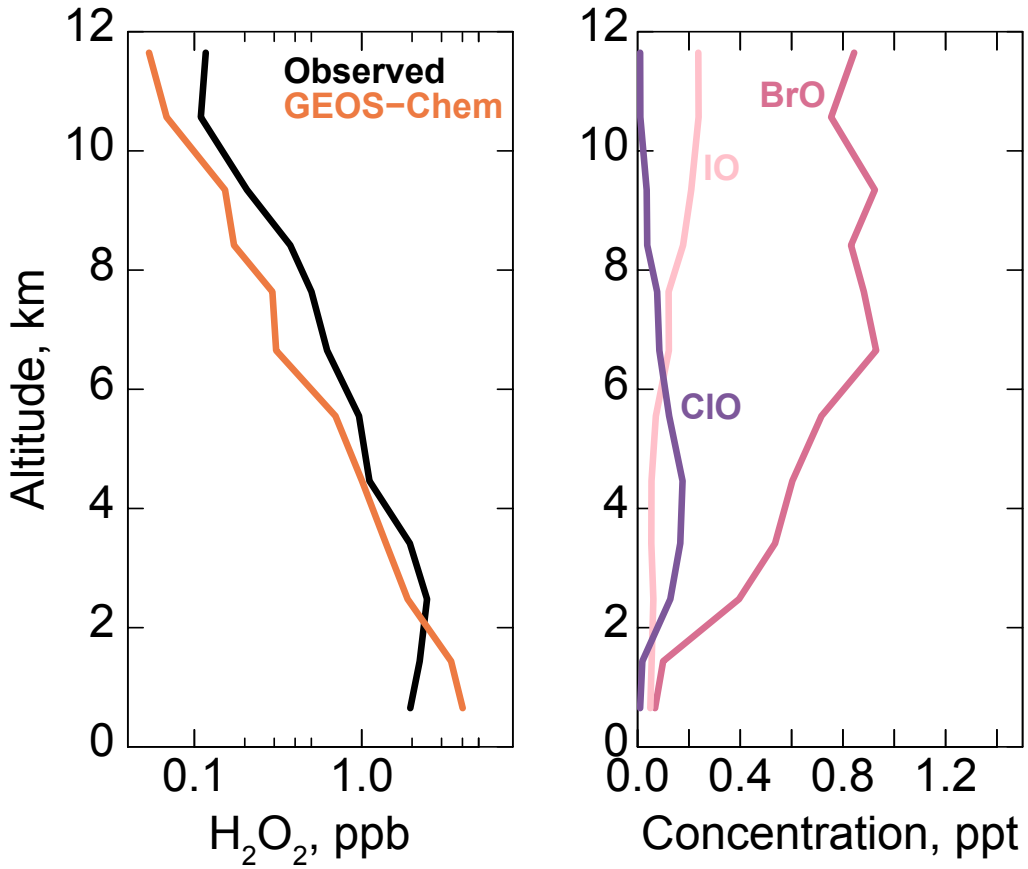
Silvern et al. (2018)

Conversion of NO to NO_2 balances only half of NO_2 photolysis and cannot be explained by errors in GEOS-Chem radical concentrations

NO-NO_2 cycling in the upper troposphere during SEAC^{4}RS

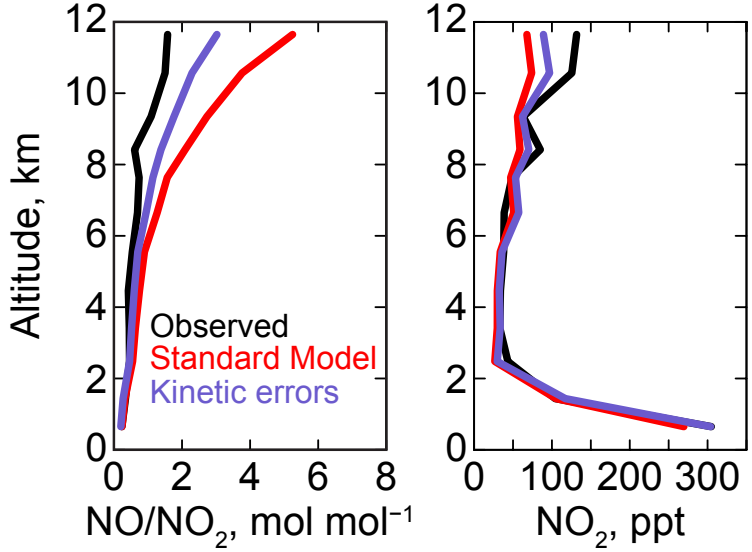


Underestimate in peroxy and BrO radicals would have to be factor of 5 and 21 to close NO-NO_2 budget



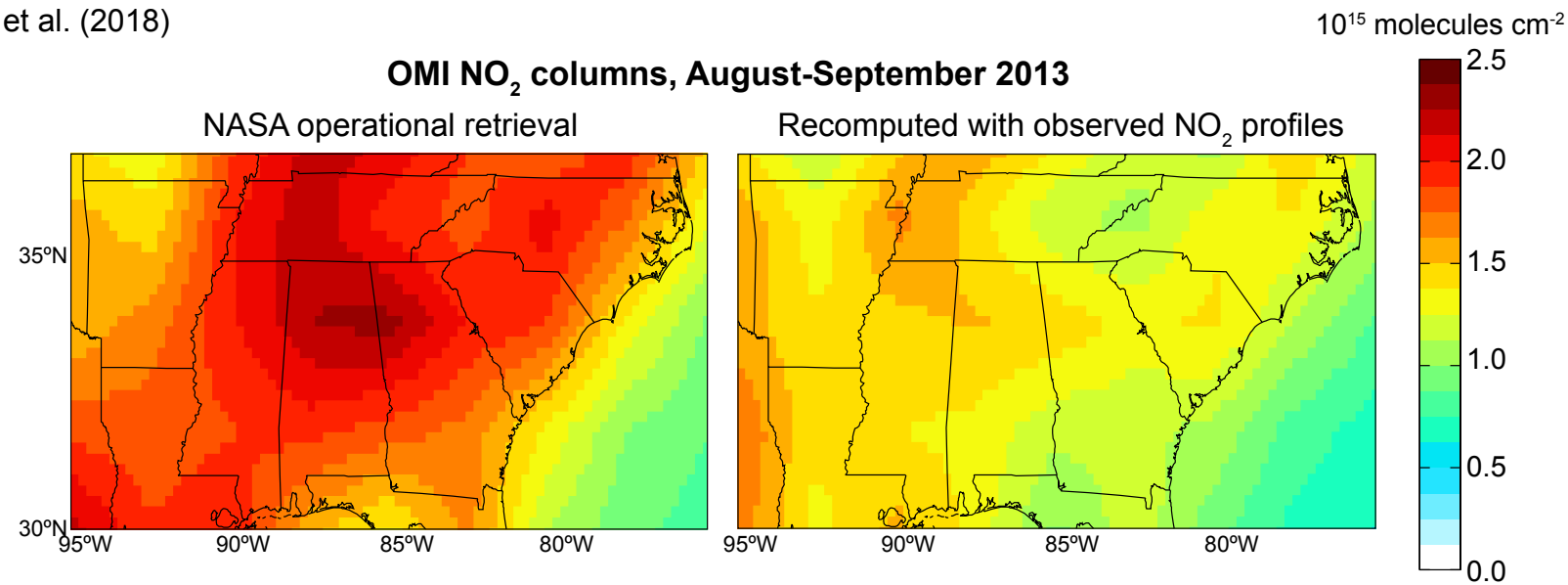
Silvern et al. (2018)

Errors in model NO_2 concentrations in the upper troposphere results in biases in the interpretation of satellite observations

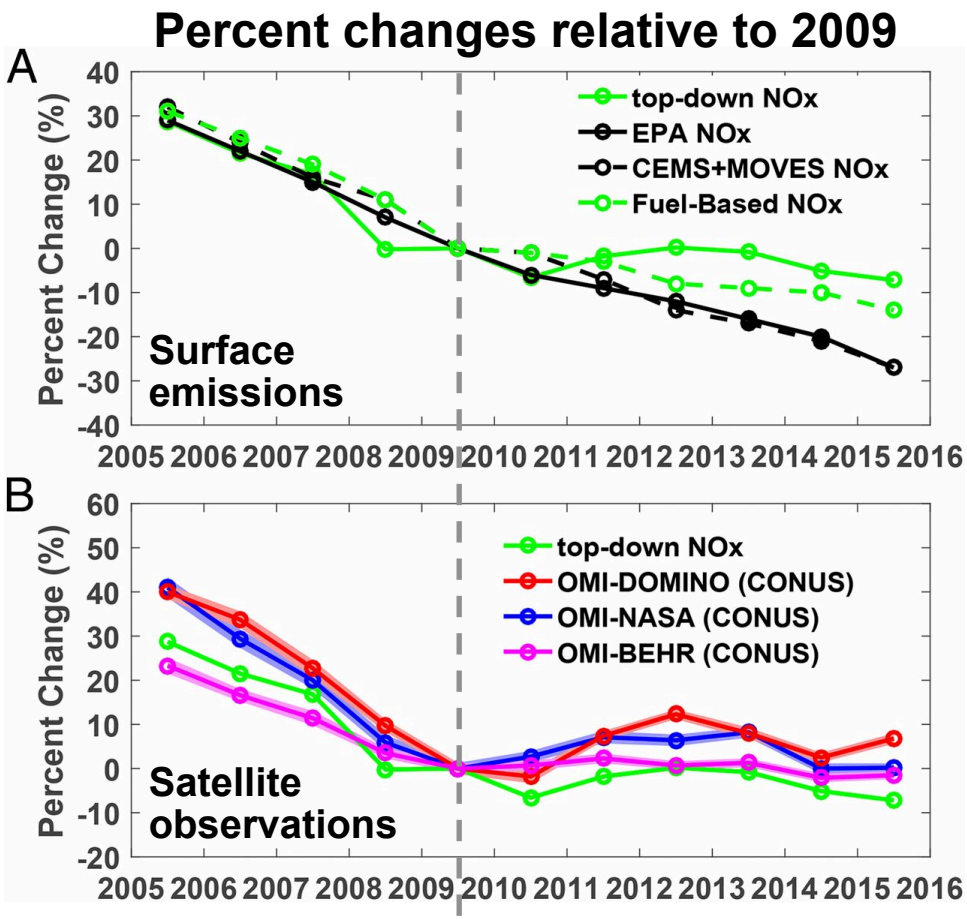


- GEOS-Chem may underestimate NO_2 due to kinetic errors or would not if a missing organic NO_x reservoir were present
- GEOS-Chem simulates upper troposphere lightning NO_x consistent with observations
- If observed NO_2 is correct, the NASA retrieval may be biased high by 30%

Silvern et al. (2018)



US NO_x emissions show linear decrease while OMI NO_2 observations show slowdown



EPA NO_x emissions = -6.4% a^{-1}
OMI NO_2 columns = -8.8±1.0% a^{-1}

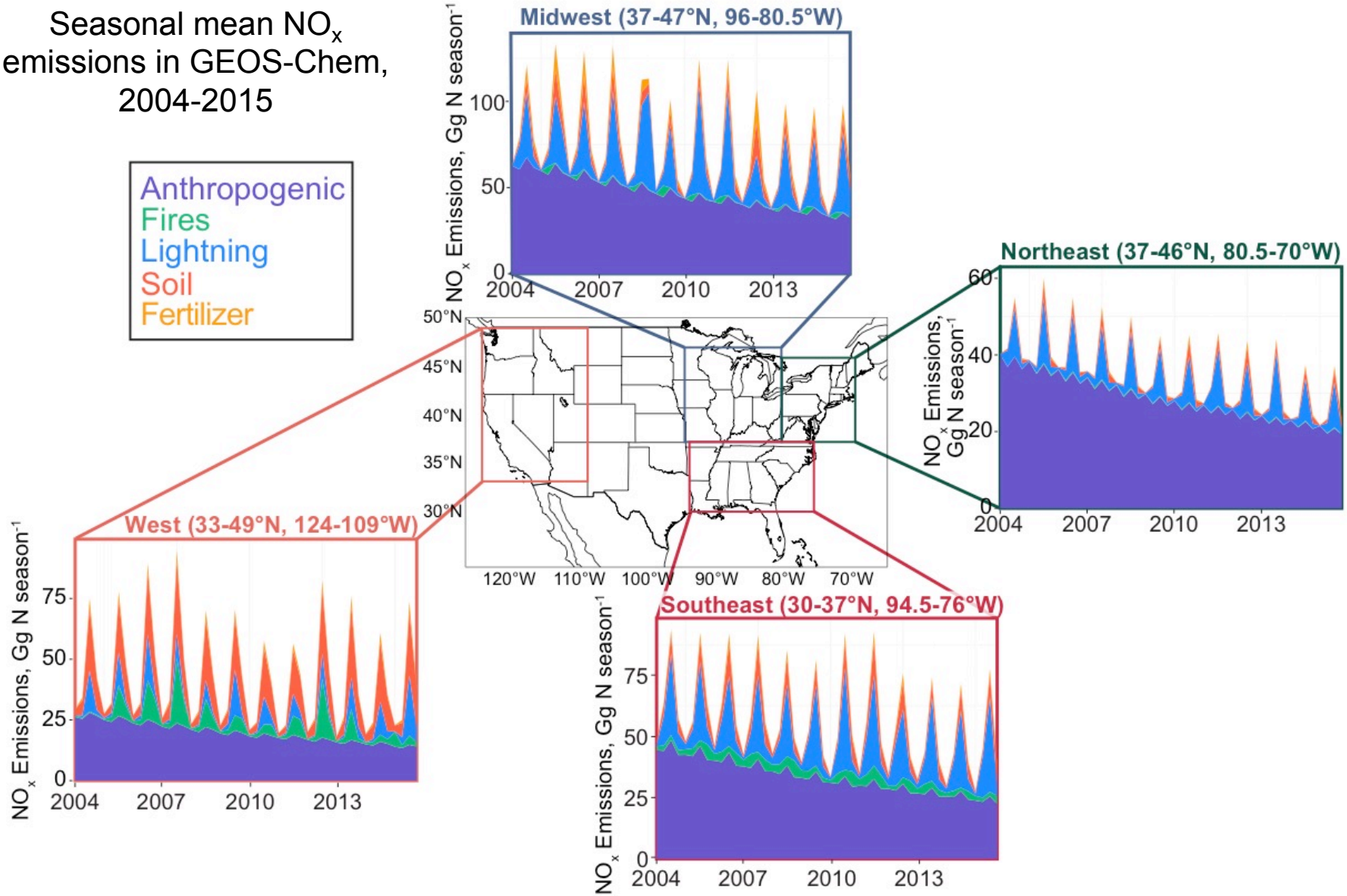
EPA NO_x emissions = -5.3% a^{-1}
OMI NO_2 columns = -1.7±1.4% a^{-1}

- Jiang et al. (2018) show discrepancy in trends must be due to emission errors
- Assume response of NO_2 columns to changes in emissions to be constant over time

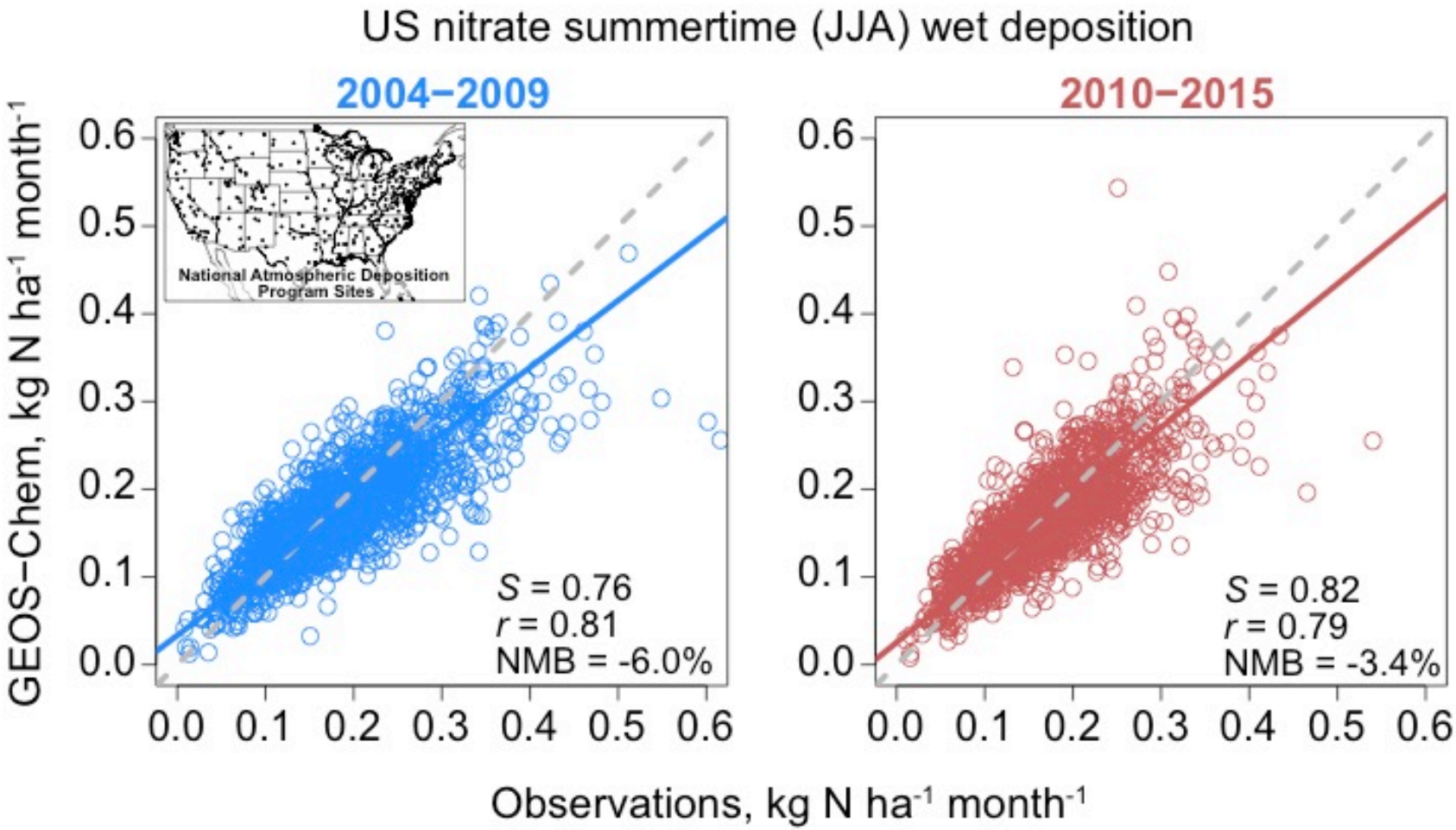
Jiang et al. (2018)

Contribution of natural sources to total NO_x budget varies by region and season

Seasonal mean NO_x emissions in GEOS-Chem, 2004-2015



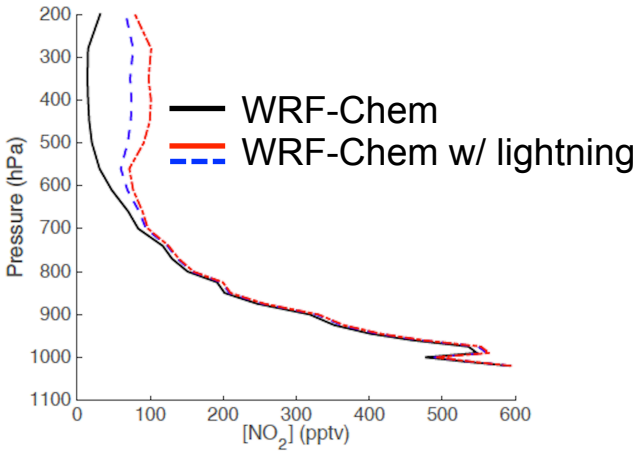
Nitrate wet deposition shows 60% anthropogenic NO_x emission scaling applies to 2004-2015 summertime and consistency with linear EPA trend



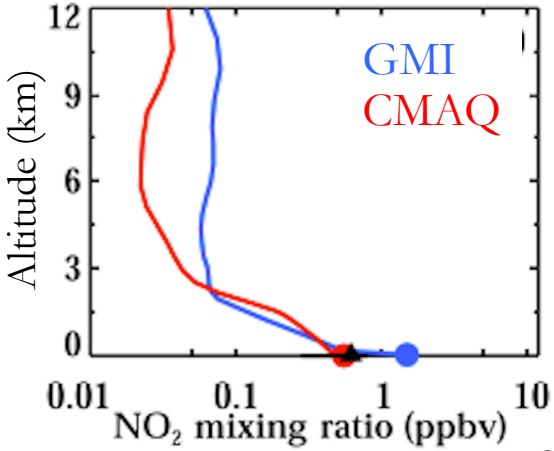
- Agreement consistent across US for summertime
- GEOS-Chem underestimates nitrate wet deposition by 20-30% in seasons other than summer indicating emissions or NO_x lifetime errors

We simulate daily NO_2 profiles to improve representation of the upper troposphere in interpretation of OMI observations

Standard retrievals underestimate NO_2 in upper troposphere

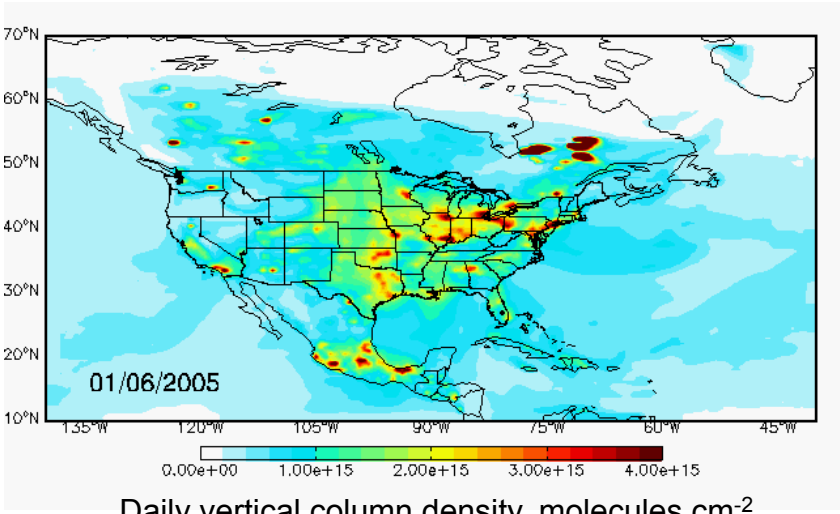


Laughner and Cohen (2017)



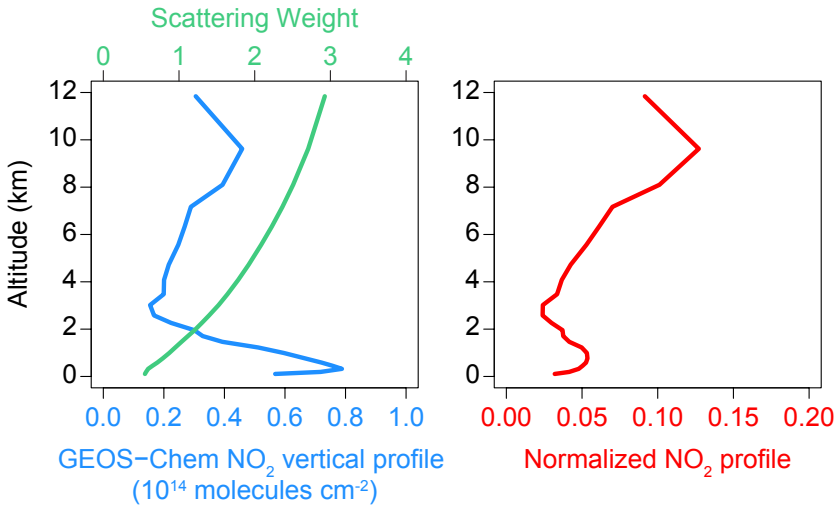
Goldberg et al. (2017)

Sample GEOS-Chem at local OMI overpass time

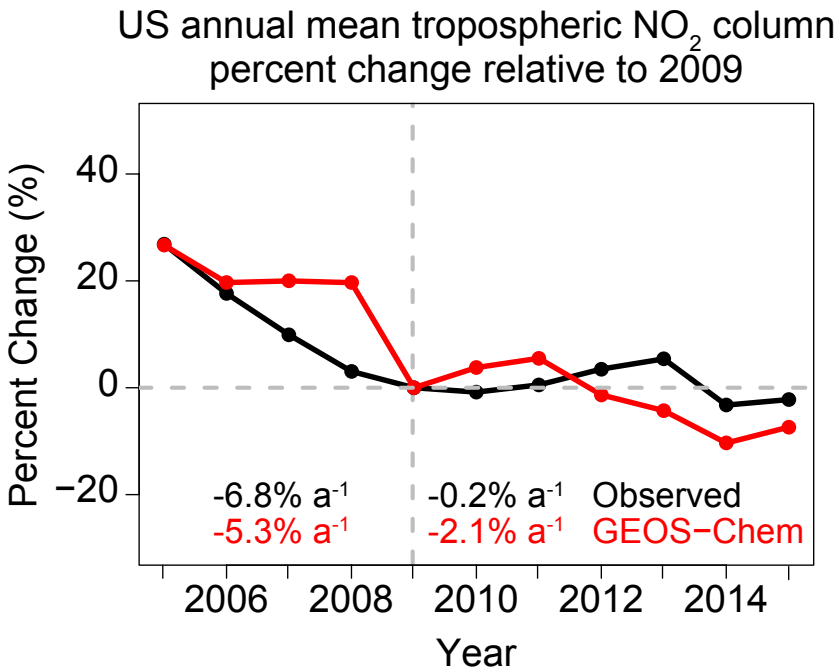


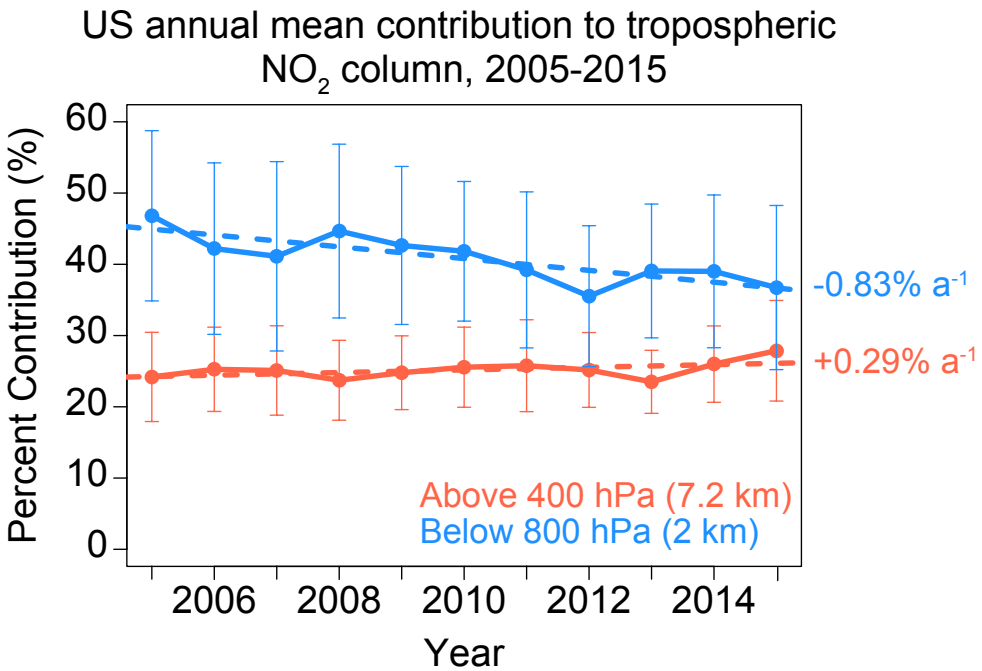
Daily vertical column density, molecules cm^{-2}

Calculate new air mass factor with GEOS-Chem NO_2 profile



GEOS-Chem can generally reproduce observed flattening trend due to decreasing contribution of the boundary layer as emissions decline





Flattening trend in satellite NO_2 columns in part due to increasing importance of the upper troposphere, not recognized previously

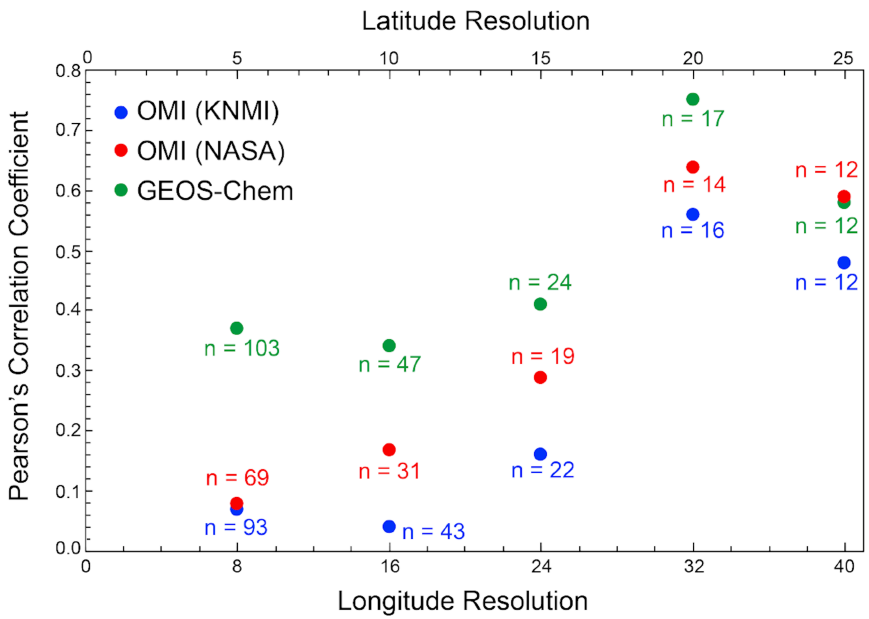
Implications for TEMPO

- NO_2 in the free and upper troposphere makes a large contribution to the total NO_2 vertical column observed from satellites
  - TEMPO retrieval will need to properly account for upper tropospheric NO_2
  - Large discrepancy between observed and modeled NO_2 in the upper troposphere, which may bias satellite retrievals

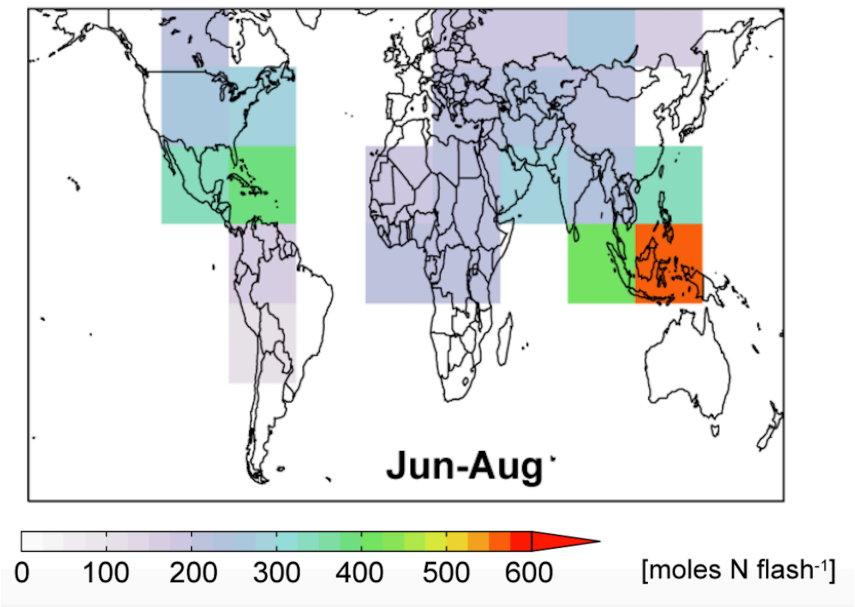
- Cloud-slicing technique developed for OMI NO_2 (Choi et al., 2014; Belmonte Rivas et al., 2015) could be leveraged by TEMPO to observe upper tropospheric NO_2 with higher temporal and spatial resolution

Current cloud-sliced NO_2 products provides constraint on upper tropospheric NO_x but only at coarse spatial resolutions

Correlations between upper troposphere NO_2 and aircraft observations



OMI-derived seasonal mean lightning NO_x production rate



TEMPO could offer improved constraints on upper tropospheric NO_2 at higher spatial and temporal resolution than current cloud-slicing products

Marais et al., submitted