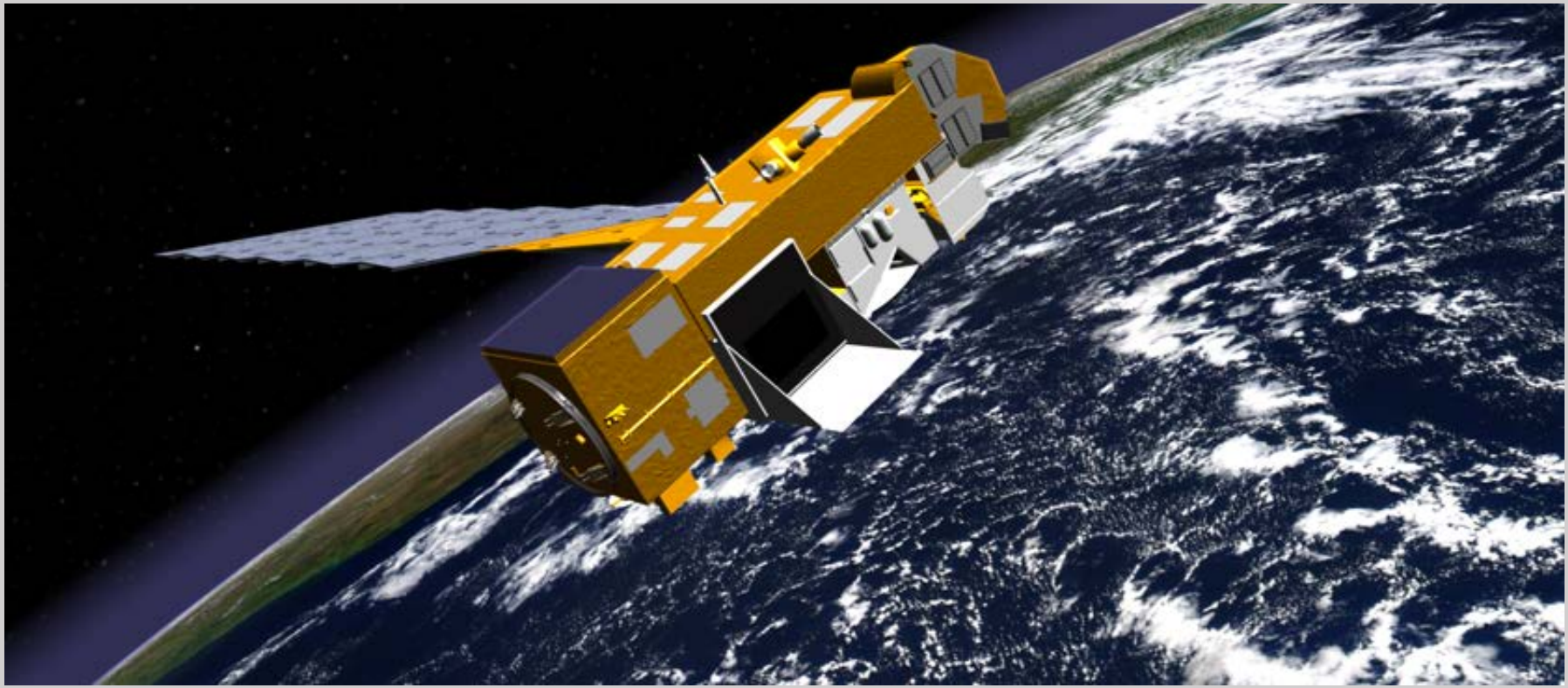
Using OMI cloud-sliced NO_2 and GEOS-Chem to better understand dynamics of NO_x in the upper troposphere



E. A. Marais, D. J. Jacob, S. Choi, J. Joiner, M. Belmonte-Rivas, R. C. Cohen, T. B. Ryerson, A. J. Weinheimer, A. Volz-Thomas, L. T. Murray, V. Shah, L. Jaeglé, S. Beirlie

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Cambridge, MA
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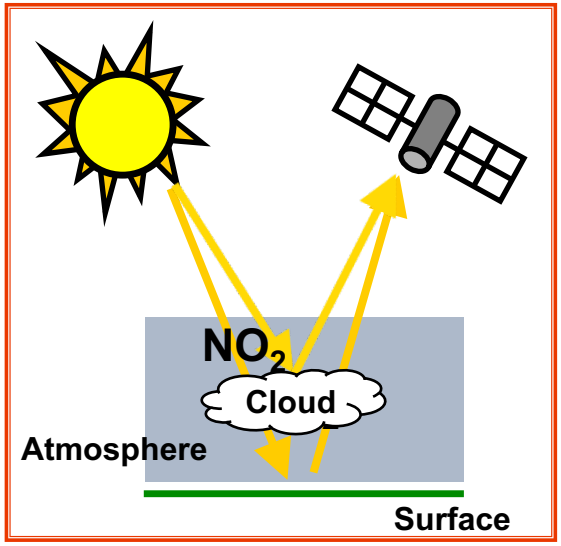
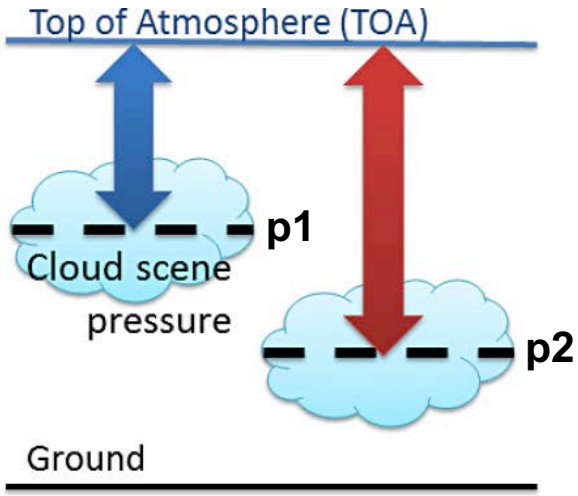
Global UT OMI NO_2: new dataset to better understand UT NO_x

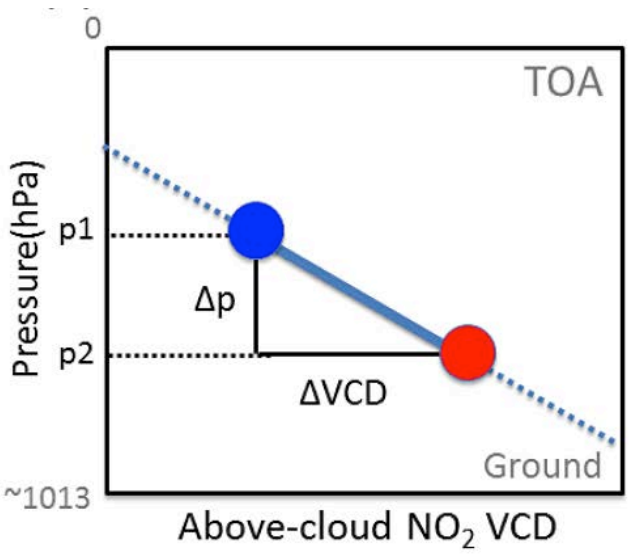
Exploit differences in cloud height to retrieve partial NO_2 columns (VCD) and calculate vertically resolved NO_2 concentrations (VMR).

APPROACH



Use cloud height variability to derive pseudoprofiles





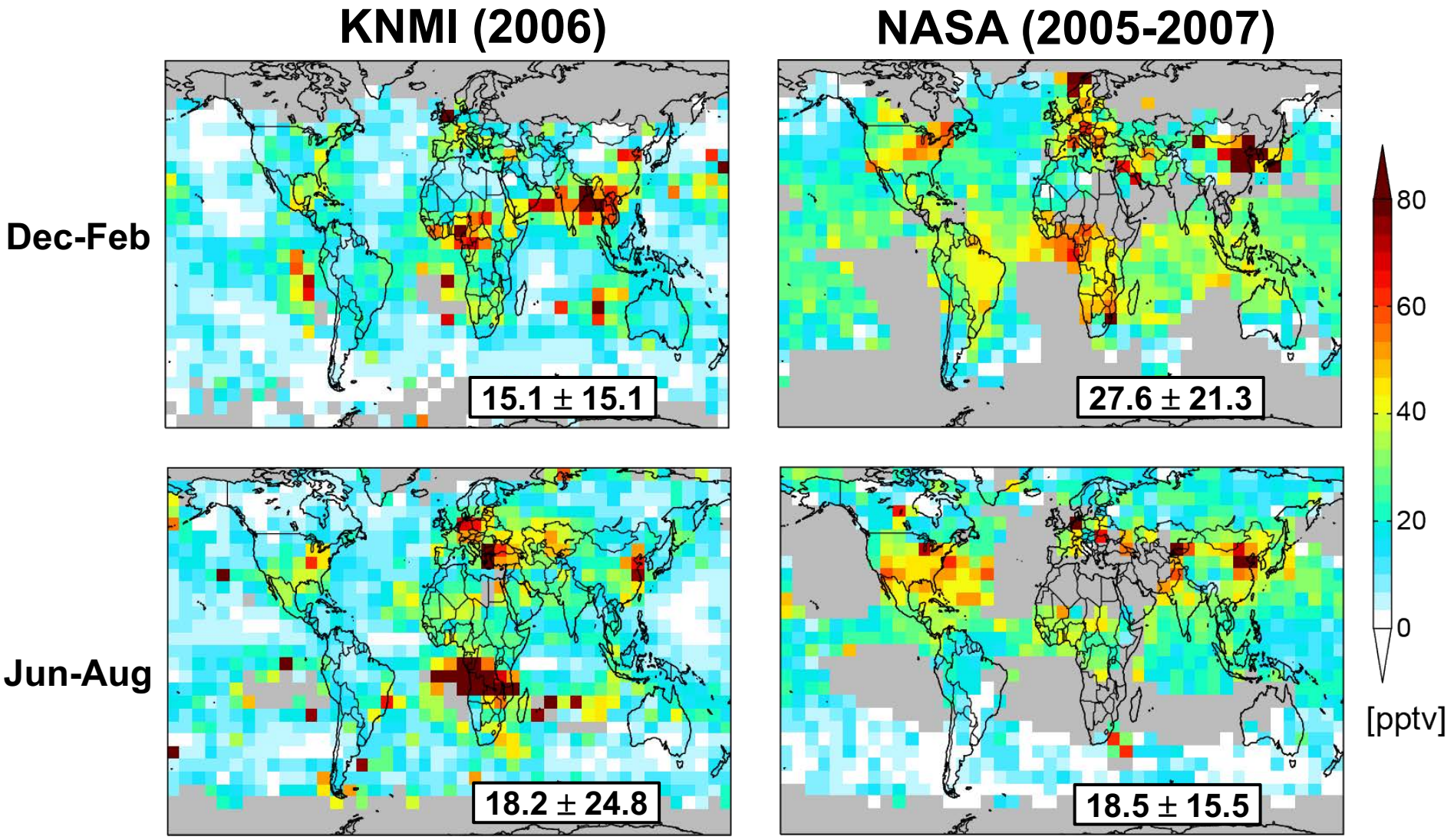
[Choi et al., 2014]

NO_2 volume mixing ratio (VMR) between clouds at p1 and p2

$$\mathrm{NO_2\ VMR} = \frac{\Delta \mathrm{VCD}}{\Delta \mathrm{p}} \times \frac{k_B g}{R_{\mathrm{air}}}$$

Two global UT OMI NO_2 products: KNMI and NASA

Seasonal mean OMI NO_2 from KNMI (~380 hPa) and NASA (~350 hPa)
Data are at 8º × 5º (longitude × latitude)

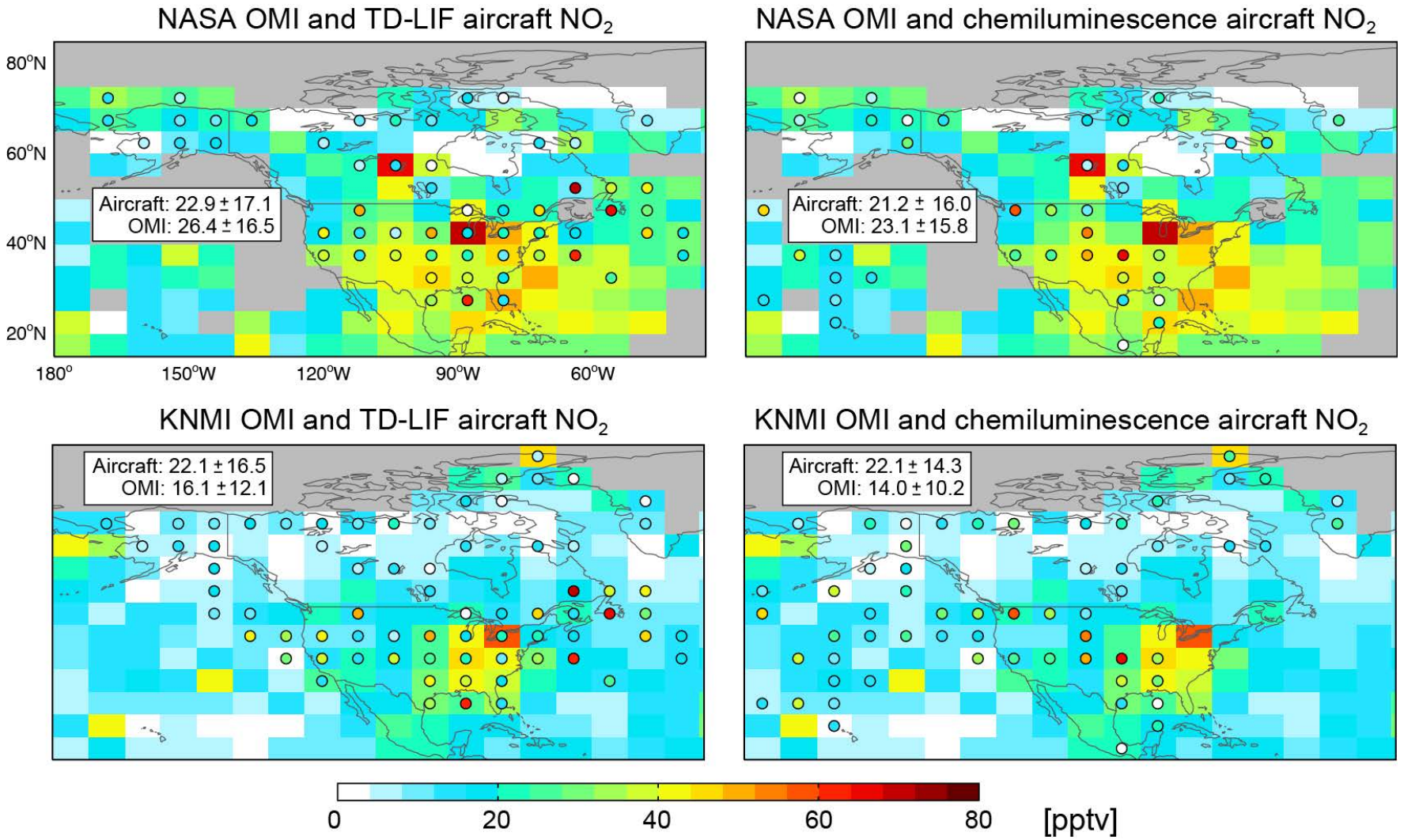


[Belmonte-Rivas et al., 2015]

[Choi et al., 2014]

Evaluate OMI UT NO_2 with aircraft observations

Comparison of May-August OMI and aircraft UT NO_2 during DC8 campaigns

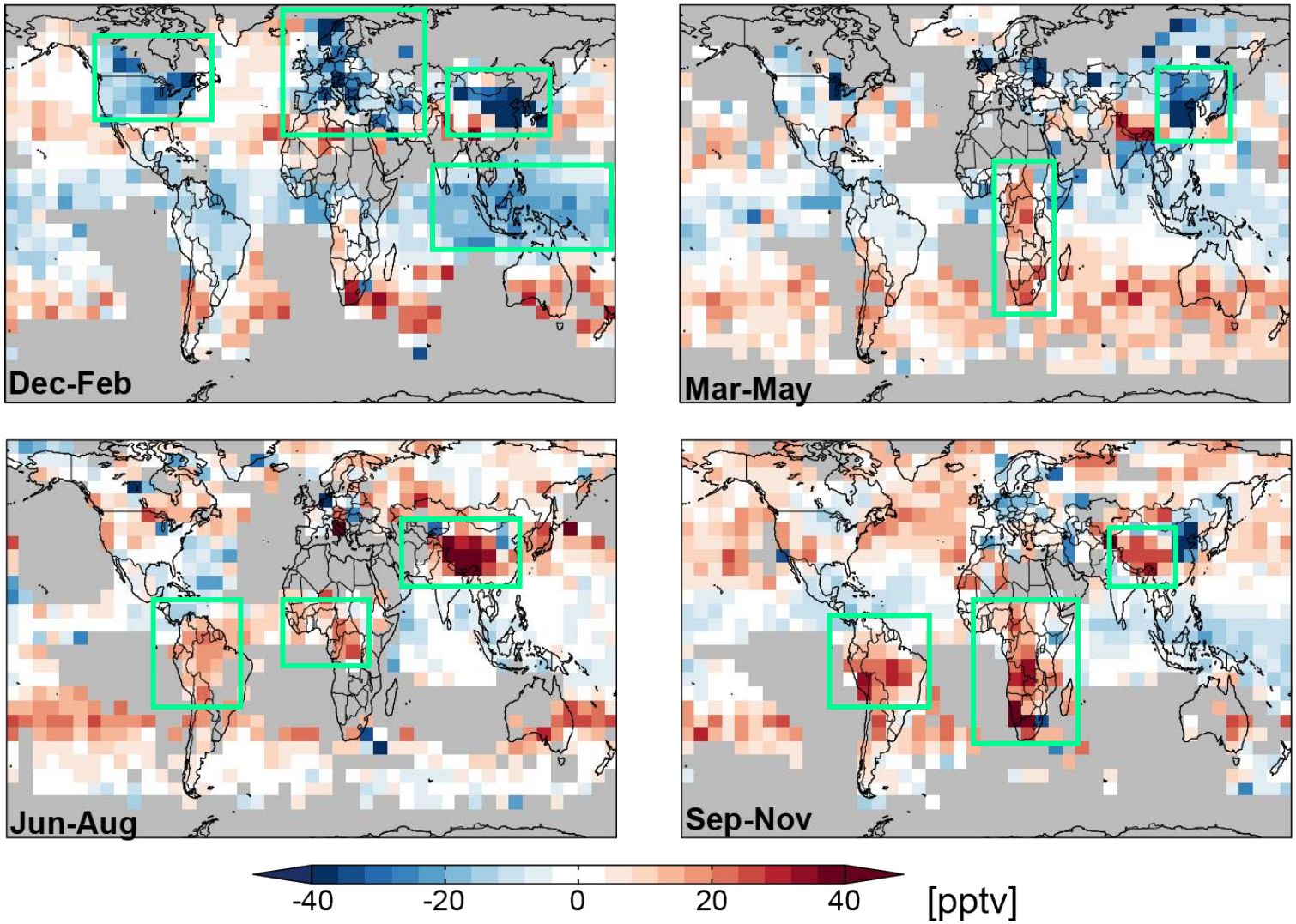


[Aircraft NO_2 from T. Ryerson, R. Cohen, A. Weinheimer]

NASA product is more consistent with aircraft measurements than KNMI (KNMI is consistently lower).

Compare GEOS-Chem and NASA OMI UT NO_2

Difference between NASA OMI and GEOS-Chem UT NO_2 (GEOS-Chem minus NASA)



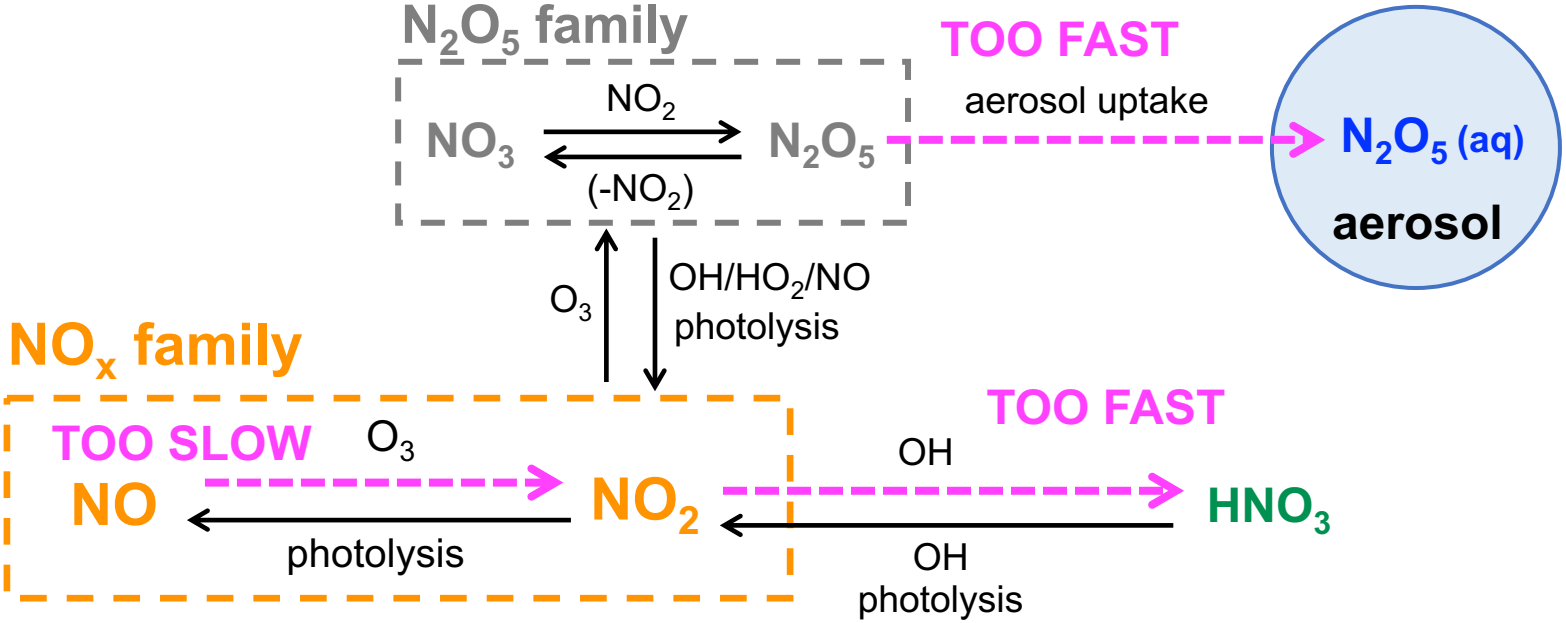
BLUE: model too low

RED: model too high

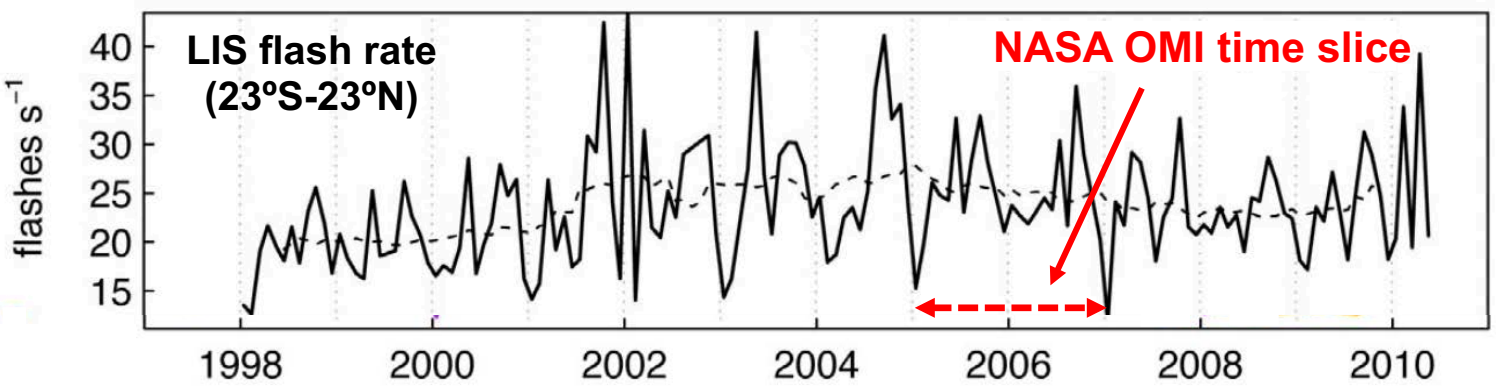
Notable regional discrepancies

Chemistry and lightning interannual variability

Uncertain reaction rates impact NO:NO_2 and NO_x abundances, but updating these does not resolve the discrepancies between OMI and GEOS-Chem.



Lightning interannual variability from the LIS sensor also does not improve agreement between the model and observations

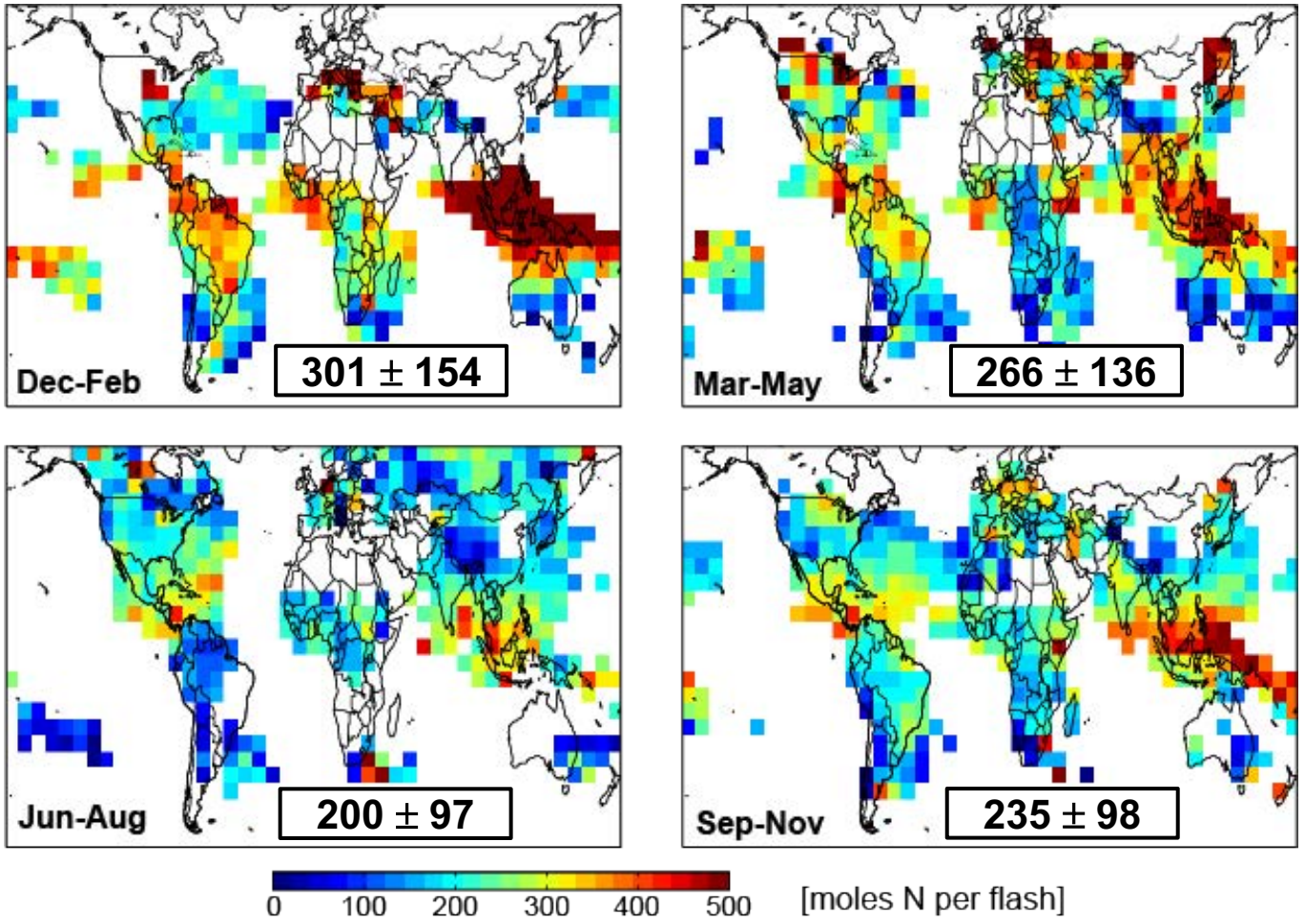


[Adapted from Murray et al., 2012]

NO_x production per lightning flash

Single value (260 moles NO per flash) used in GEOS-Chem based on limited data

Spatially resolved resolved NO_x production per flash (*Data are at 8º × 5º*)



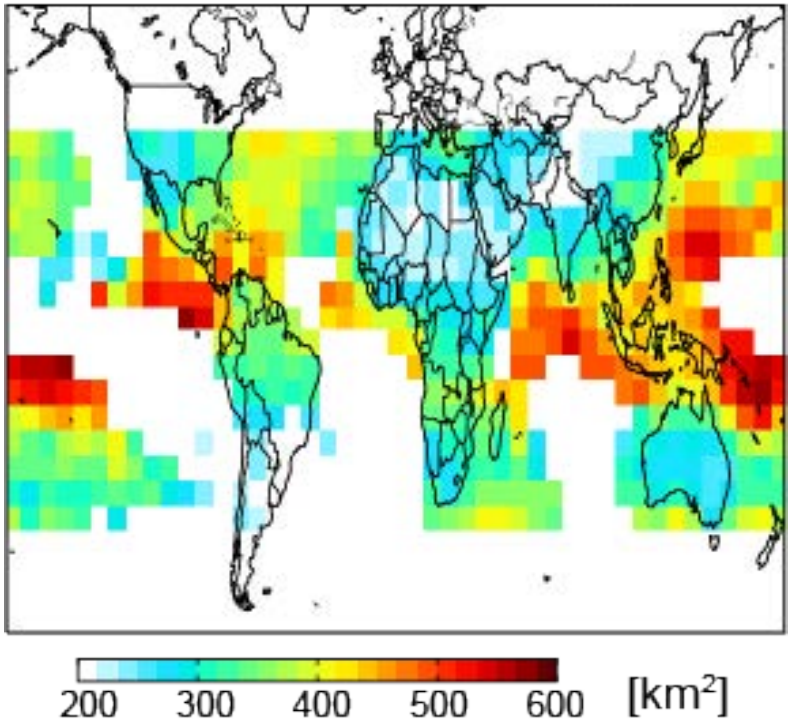
Obtained by scaling 260 mol NO/fl by discrepancies between GEOS-Chem and OMI

Spatial features: higher values over oceans than land.

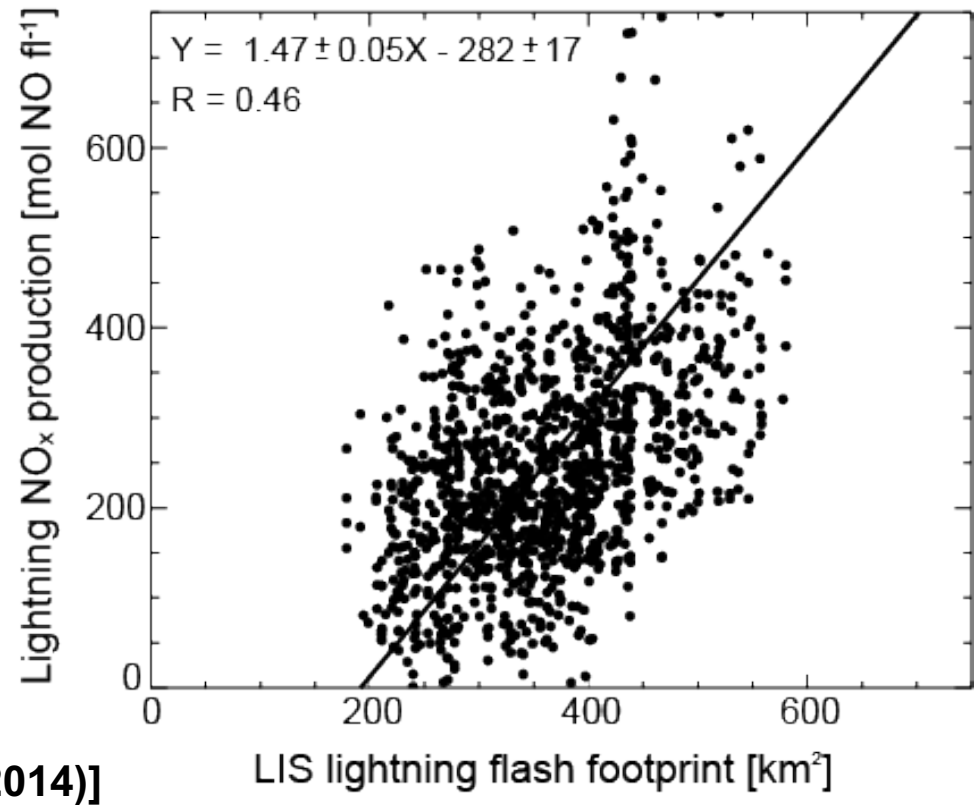
NO_x per flash as a function of lightning intensity

Combine OMI-derived NO per flash and OTD- and LIS-derived lightning footprints

LIS Lightning flash footprint



NO_x production rate versus flash footprint



[Lighting flash footprint from Beirle et al. (2014)]

Large lightning footprint is associated with high flash intensity and long flash duration [Beirle et al., 2014]. The reason is not clear.

Linear relationship between lightning NO_x production rate and flash footprint.

Potential to move towards a more mechanistic representation of NO_x production rates.

Summary and Concluding Remarks

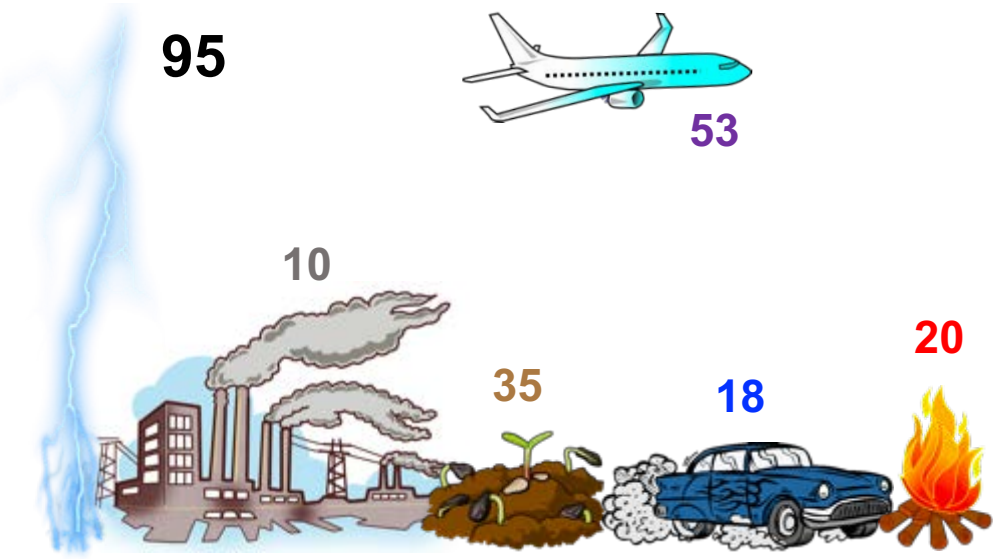
Used new OMI products from NASA and KNMI to provide constraints on upper troposphere (UT) NO_x where production efficiency and global warming potential of ozone are high.

- NASA OMI UT NO_2 observations are more consistent with aircraft DC8 observations than is KNMI
- GEOS-Chem chemical transport model UT NO_2 has large regional discrepancies when compared to NASA UT NO_2
- Uncertainties in chemistry and accounting for lightning flash interannual variability (IAV) does not address model biases
- Likely culprit in the model is the assumption that the NO production rate per flash is the same throughout the world (260 mol NO per flash)
- We find a robust linear relationship between OMI-derived lightning NO_x production per flash and LIS flash footprints
- Next steps: independently evaluate with NO_2 observations from the ATom aircraft campaign and determine the implications for UT ozone
- Caveats: the northern hemisphere wintertime (Dec-Feb) discrepancy remains

Nitrogen oxides impact ozone chemistry in the upper troposphere

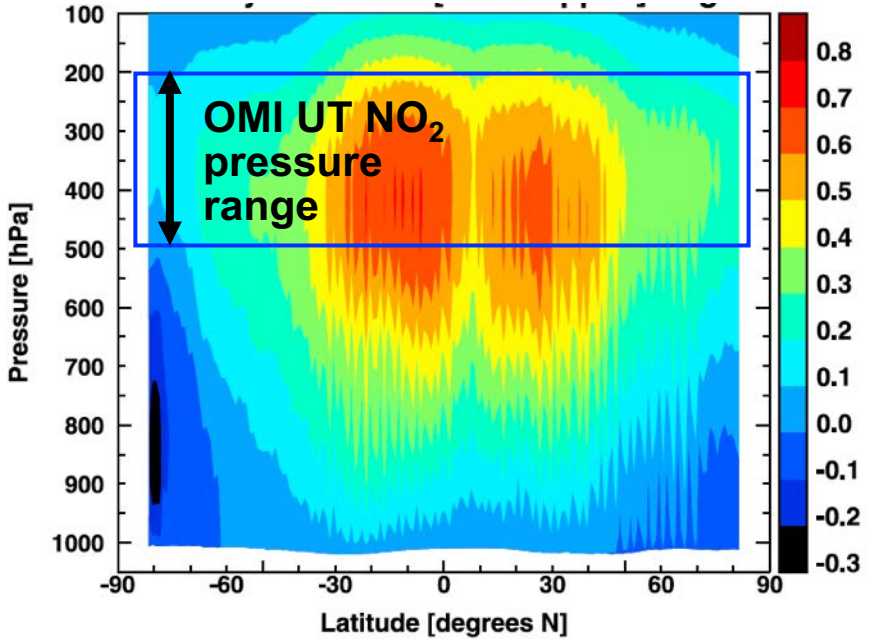
NO_x ($NO + NO_2$) has a high ozone production efficiency (OPE) in the upper troposphere where ozone is a powerful greenhouse gas

OPE (molecules O_3/molecule NO_x) for indiviual NO_x sources



[adapted from Dahlmann et al., 2011]

Sensitivity of outgoing longwave radiation to variations in ozone concentrations



[Aghedo et al., 2011]

Longer NO_x lifetime at higher altitude, so sources there have greatest OPE

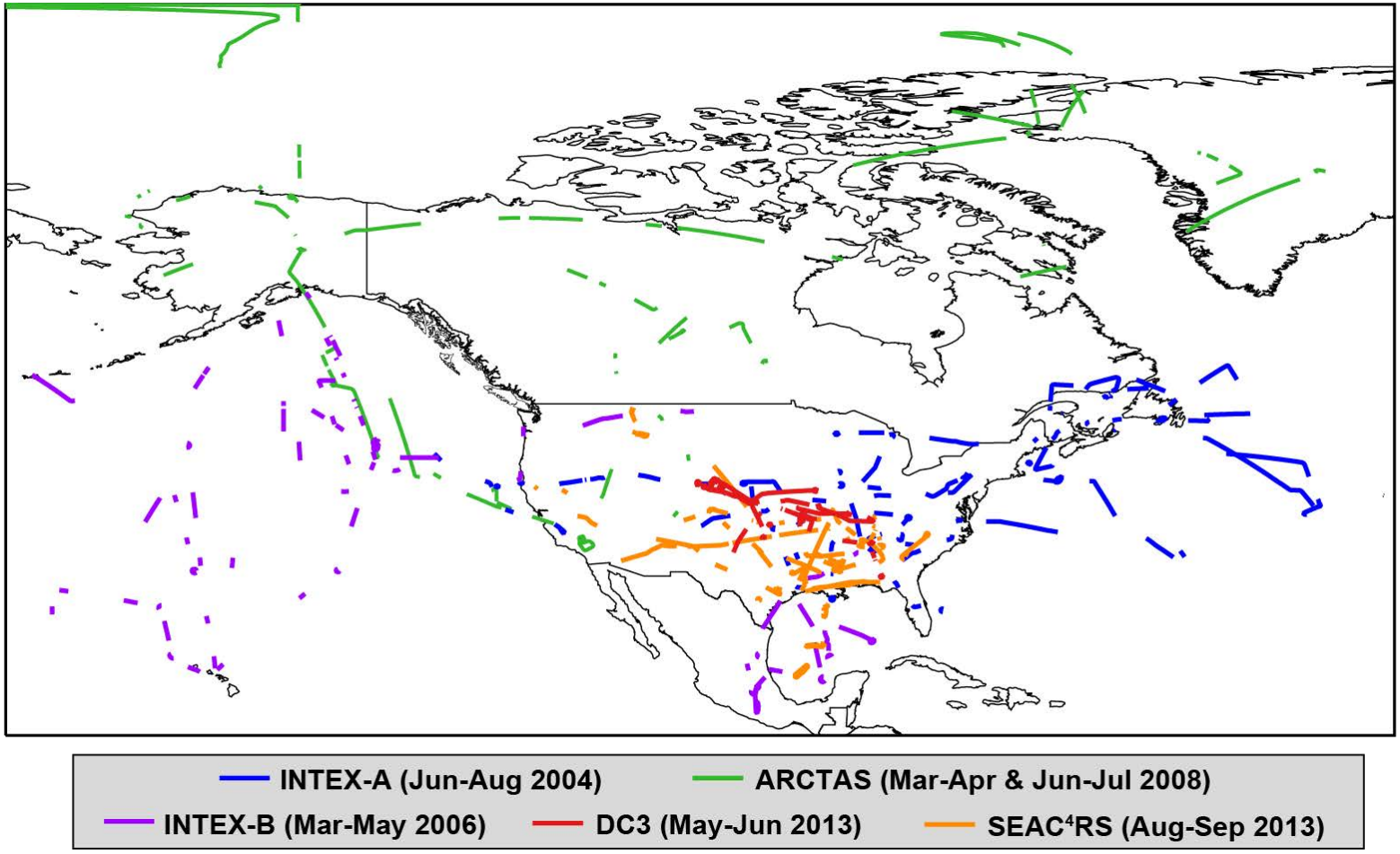
Aircraft observations to constrain upper troposphere NO_x are limited to summertime aircraft campaigns in North America and commercial aircraft at cruise altitude

Supplementary Slides

Evaluate OMI UT NO_2 with aircraft observations

NO_2 is challenging to measure in UT: NO_2 concentrations relatively low and susceptible to contamination from thermal decomposition of NO_x reservoir species (PANs, HNO_4)

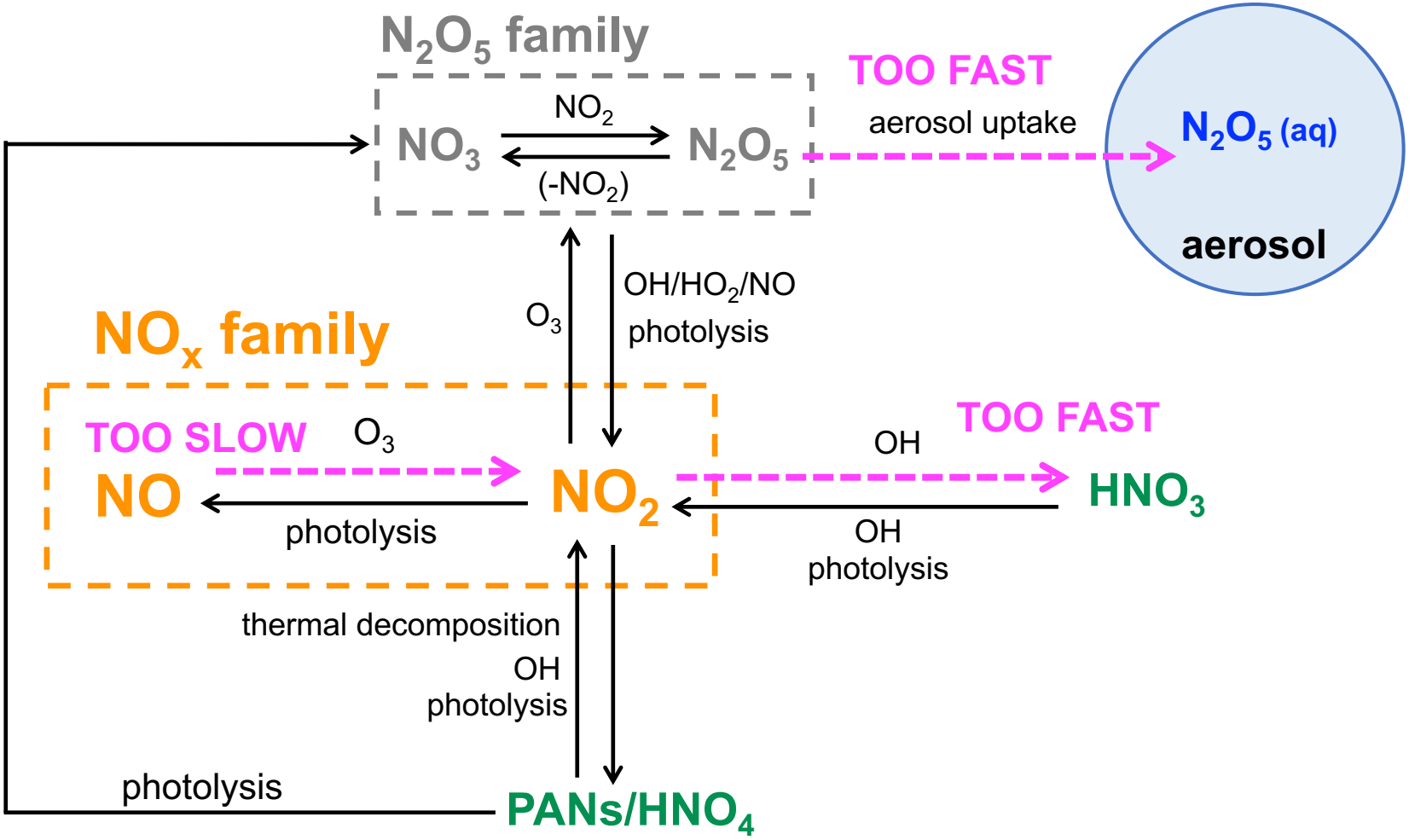
Flight tracks at midday (11h00-16h00) in the UT (280-450 hPa) and without stratospheric influence ($CO:O_3 < 1.25$)



Majority of flights to evaluate OMI UT NO_2 are in summer over North America

Reactive Nitrogen Chemistry Uncertainties

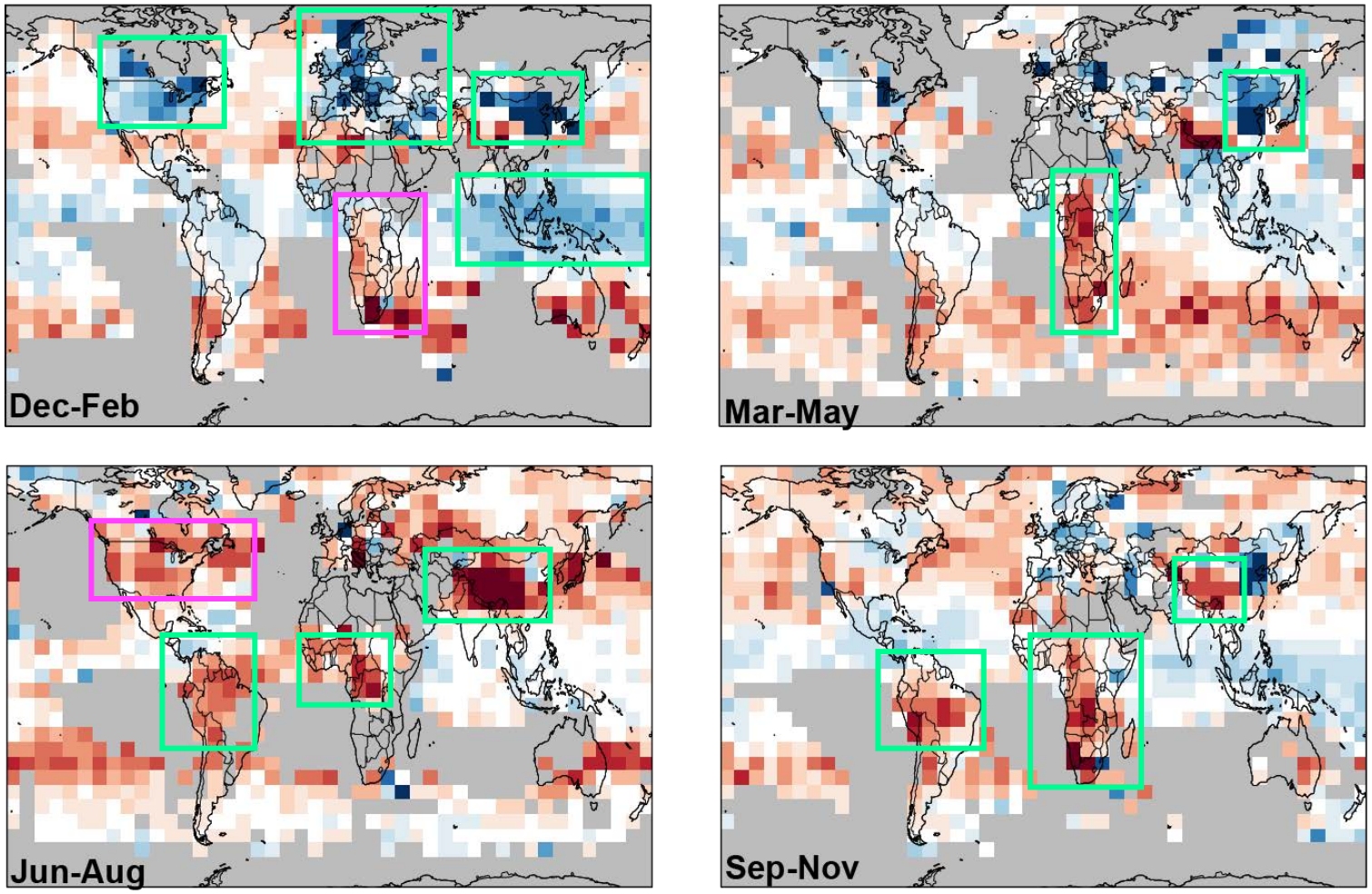
Uncertainties in NO_x chemistry have implications for relative ($NO:NO_2$) and total NO_x abundances and NO_x lifetimes.



Uncertain reaction rates that impact $NO:NO_2$ and NO_x abundances. Updating these increases NO_2 abundances.

Compare GEOS-Chem and NASA OMI UT NO_2

Updating chemistry increases UT NO_2, predominantly in the tropics and the Southeast US (Jun-Aug), exacerbating model overestimates in these locations.



BLUE: model too low

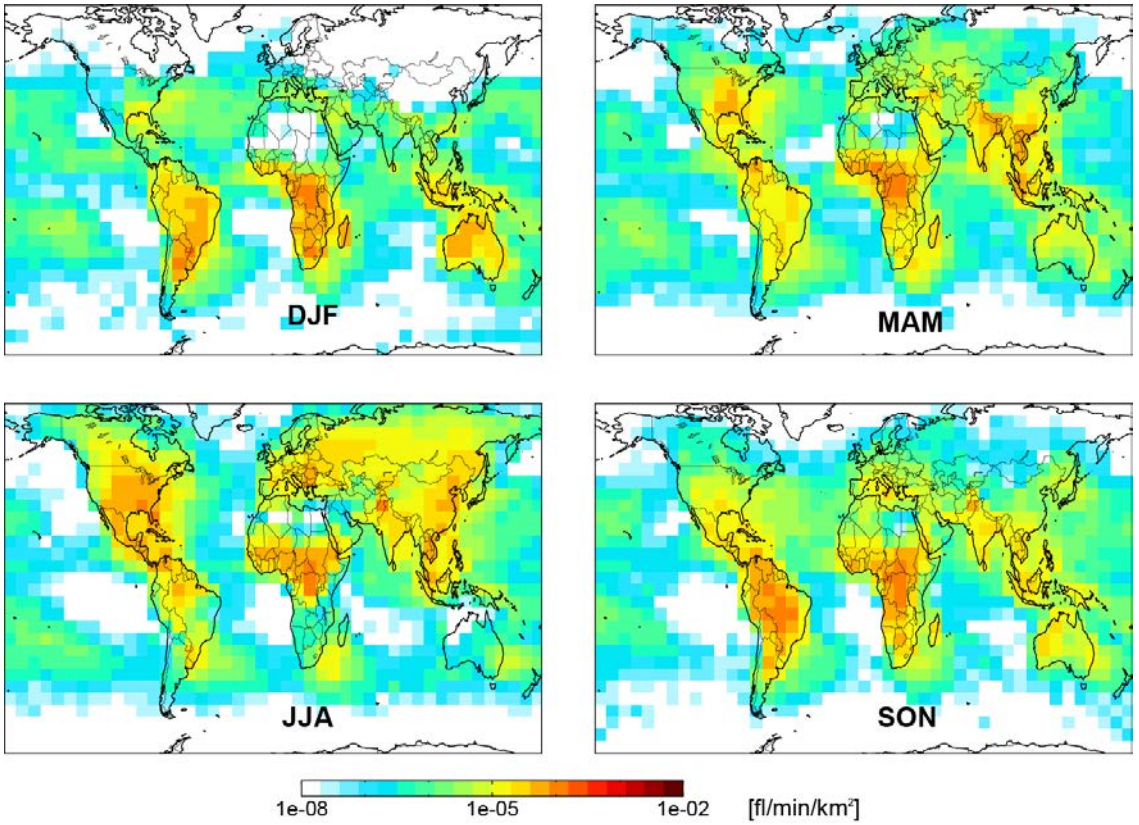
RED: model too high

Additional discrepancies

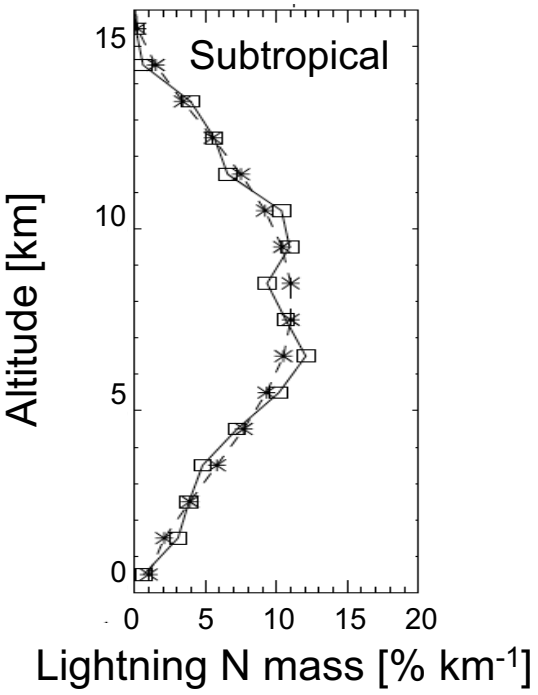
Lightning NO_x as a source of discrepancy

In GEOS-Chem lightning location is imposed with climatology of lightning flashes from OTD-LIS and vertical profiles from the literature [Murray et al., 2012]

Model lightning flashes



Model vertical lightning N distribution



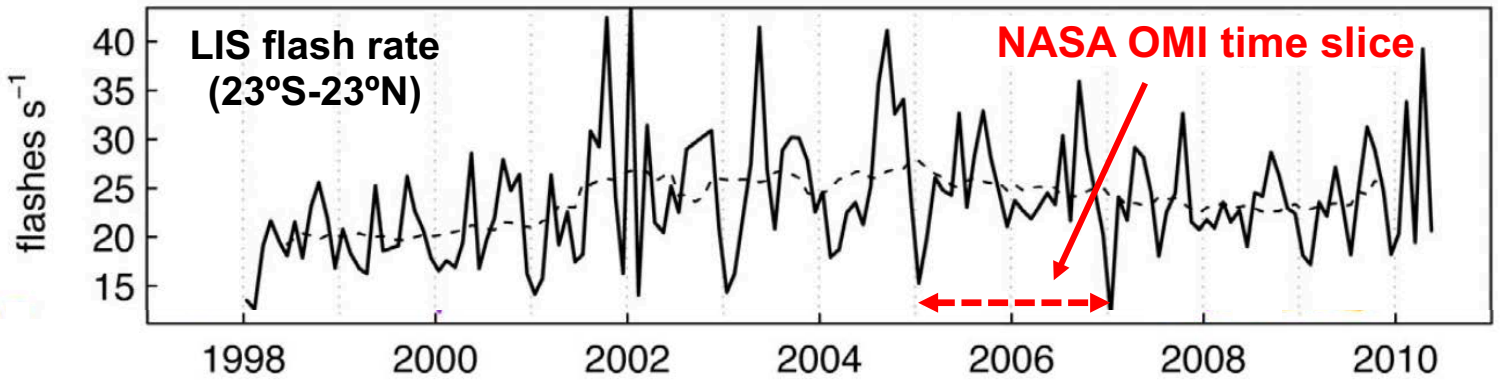
[Ott et al., 2010]

Total GEOS-Chem lightning NO_x emissions for 2006: **5.4 Tg N**

What is the effect of accounting for lightning flash interannual variability?

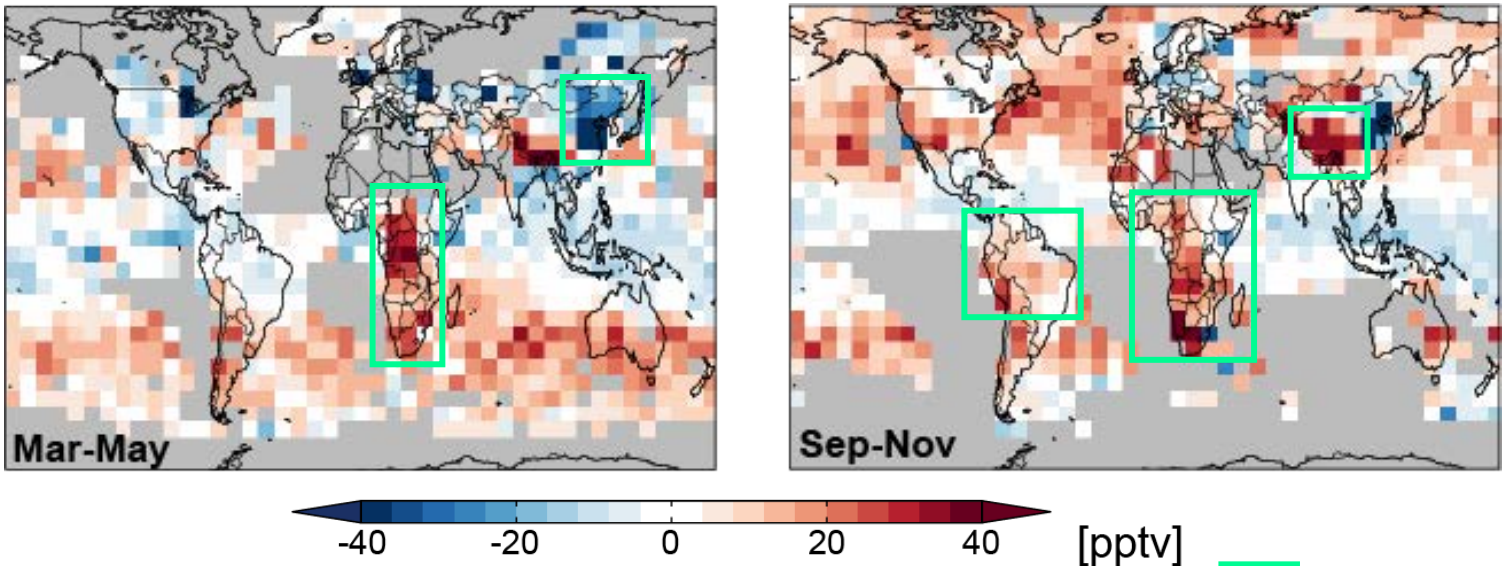
Lightning activity interannual variability (IAV)

Lightning flash rate interannual variability from LIS (40S-40N viewing domain).
(OTD only available until 2000).



[Adapted from Murray et al., 2012]

Again, does little to reduce the discrepancy in the model



BLUE: model too low **RED:** model too high Discrepancies