

# Nitrogen oxides in the global upper troposphere interpreted with cloud-sliced $\text{NO}_2$ from the Ozone Monitoring Instrument



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**EGU, Session AS3.12**  
Vienna, Austria  
26 April 2017

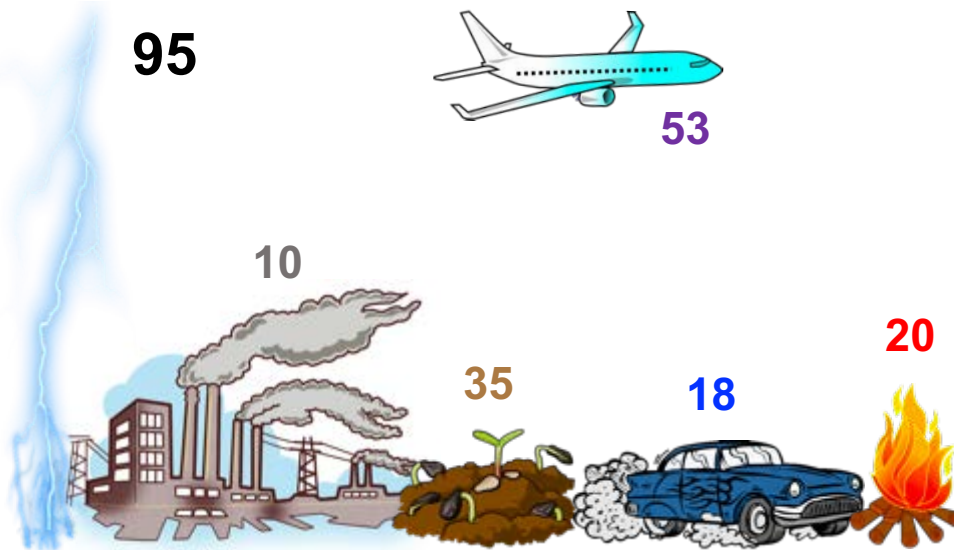


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# Nitrogen oxides impact ozone chemistry in the upper troposphere

$\text{NO}_x$  ( $\text{NO} + \text{NO}_2$ ) has a high ozone production efficiency (OPE) in the upper troposphere where ozone is a powerful greenhouse gas

## OPE (molecules $\text{O}_3$ /molecule $\text{NO}_x$ ) for individual $\text{NO}_x$ sources

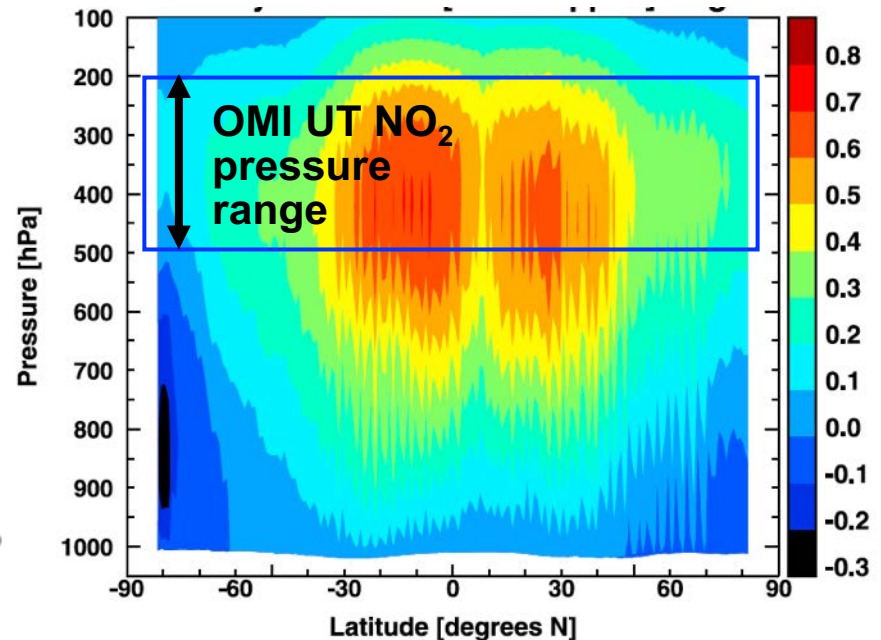


[adapted from Dahlmann et al., 2011]

Longer  $\text{NO}_x$  lifetime at higher altitude, so sources there have greatest OPE

Aircraft observations to constrain upper troposphere  $\text{NO}_x$  are limited to summertime aircraft campaigns in North America and commercial aircraft at cruise altitude

## Sensitivity of outgoing longwave radiation to variations in ozone concentrations

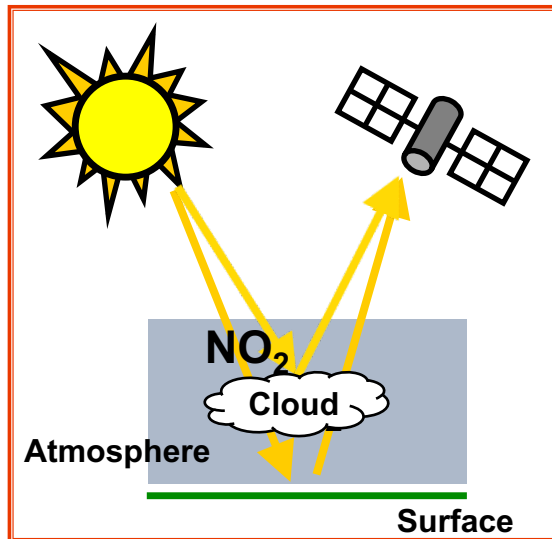


[Aghedo et al., 2011]

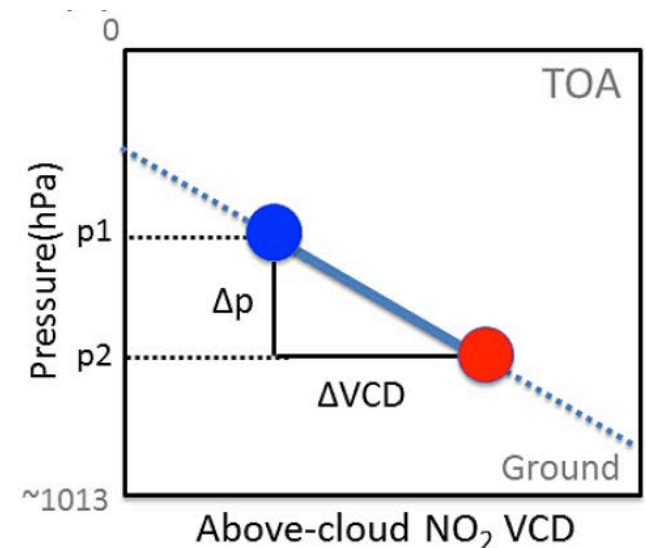
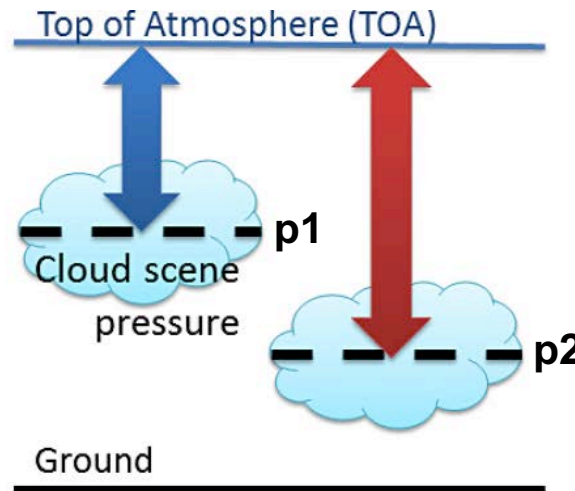
# Global UT OMI NO<sub>2</sub>: new dataset to better understand UT NO<sub>x</sub>

Exploit differences in cloud height to retrieve partial NO<sub>2</sub> columns (VCD) and calculate vertically resolved NO<sub>2</sub> concentrations (VMR).

## APPROACH



## Use cloud height variability to derive pseudoprofiles



[Choi et al., 2014]

NO<sub>2</sub> volume mixing ratio (VMR) between clouds at p1 and p2

$$\text{NO}_2 \text{ VMR} = \frac{\Delta \text{VCD}}{\Delta p} \times \frac{k_B g}{R_{\text{air}}}$$



# Two global UT OMI NO<sub>2</sub> products: KNMI and NASA

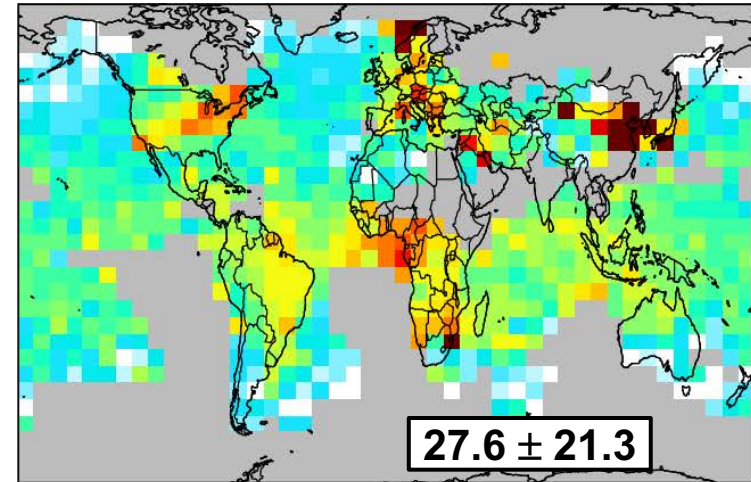
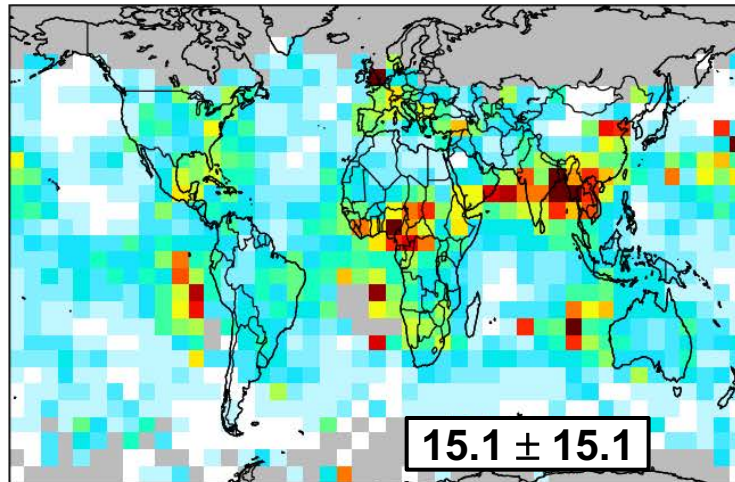
Seasonal mean OMI NO<sub>2</sub> from KNMI (~380 hPa) and NASA (~350 hPa)

*Data are at 8° × 5° (longitude × latitude)*

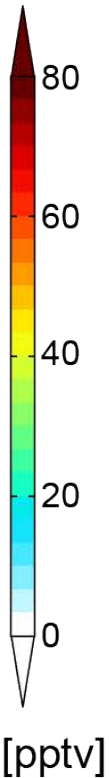
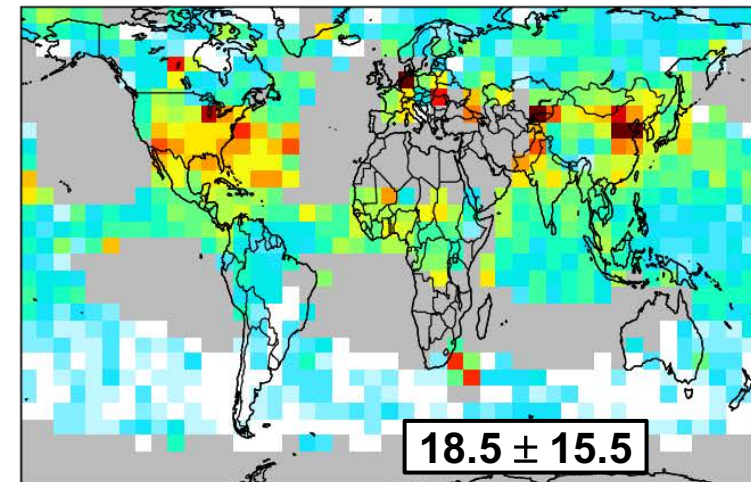
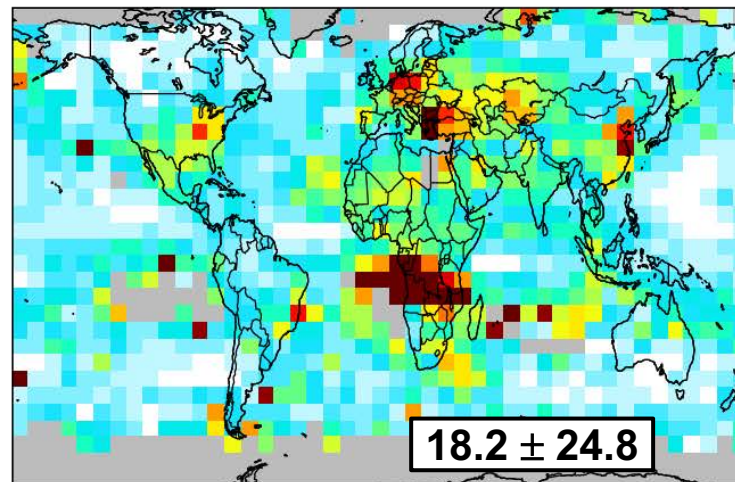
## KNMI (2006)

## NASA (2005-2007)

Dec-Feb



Jun-Aug



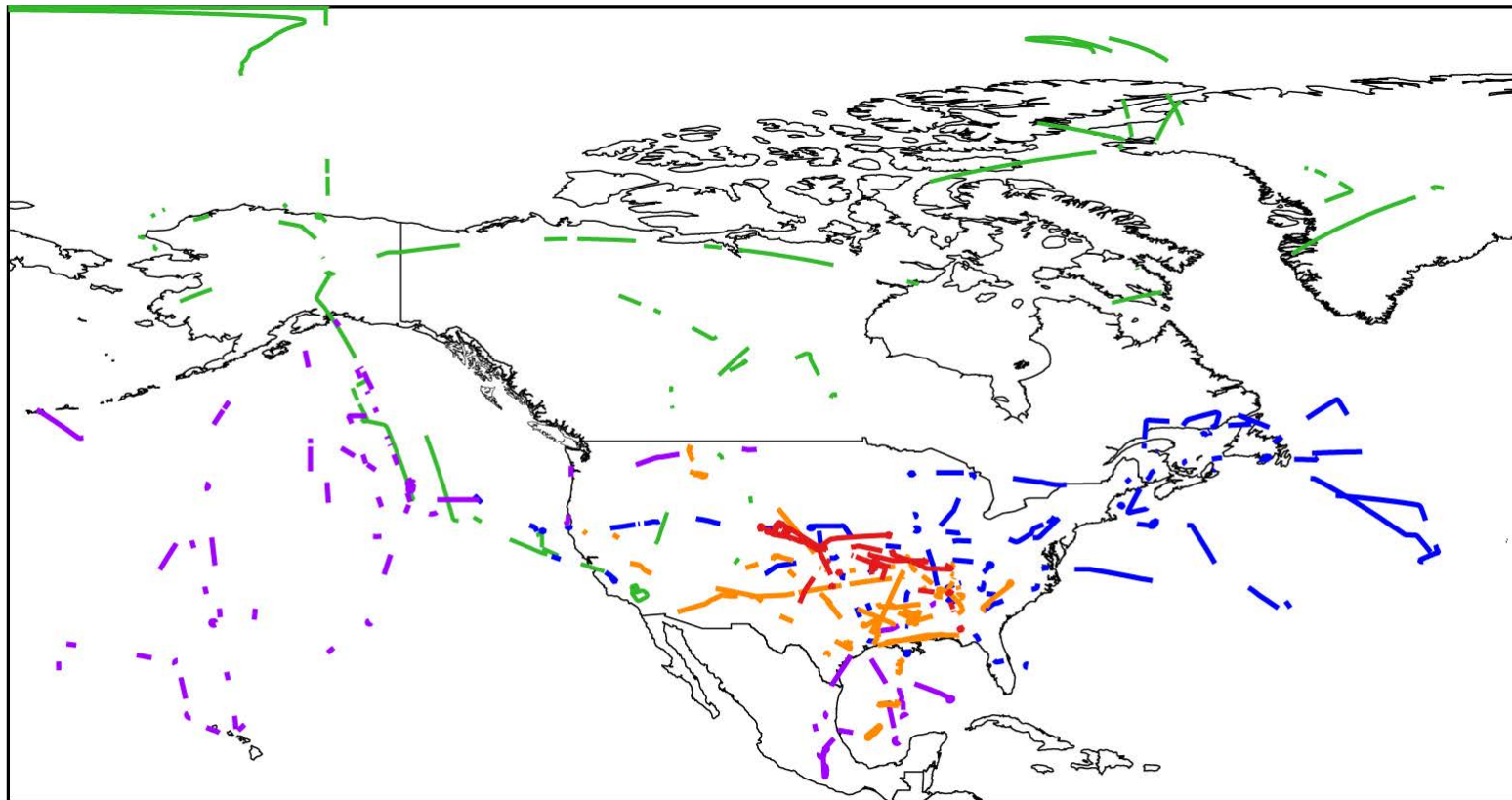
[Belmonte-Rivas et al., 2015]

[Choi et al., 2014]

# Evaluate OMI UT NO<sub>2</sub> with aircraft observations

NO<sub>2</sub> is challenging to measure in UT: NO<sub>2</sub> concentrations relatively low and susceptible to contamination from thermal decomposition of NO<sub>x</sub> reservoir species (PANs, HNO<sub>4</sub>)

**Flight tracks at midday (11h00-16h00) in the UT (280-450 hPa) and without stratospheric influence (CO:O<sub>3</sub> < 1.25)**



— INTEX-A (Jun-Aug 2004) — ARCTAS (Mar-Apr & Jun-Jul 2008)  
— INTEX-B (Mar-May 2006) — DC3 (May-Jun 2013) — SEAC<sup>4</sup>RS (Aug-Sep 2013)

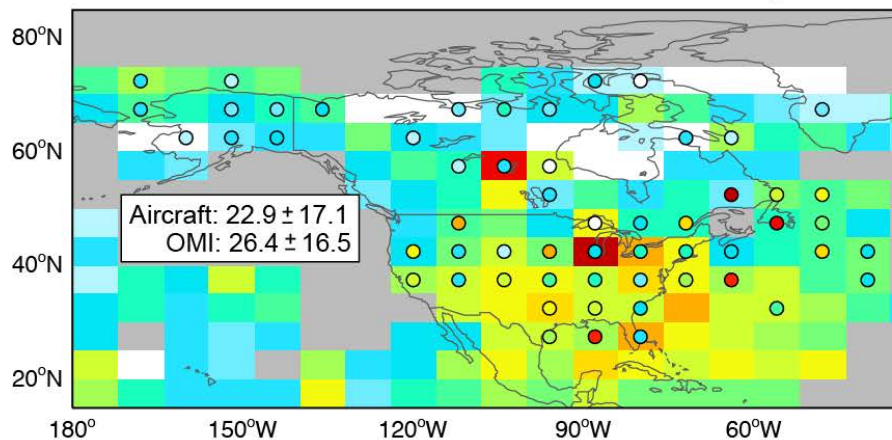
Majority of flights to evaluate OMI UT NO<sub>2</sub> are in summer over North America



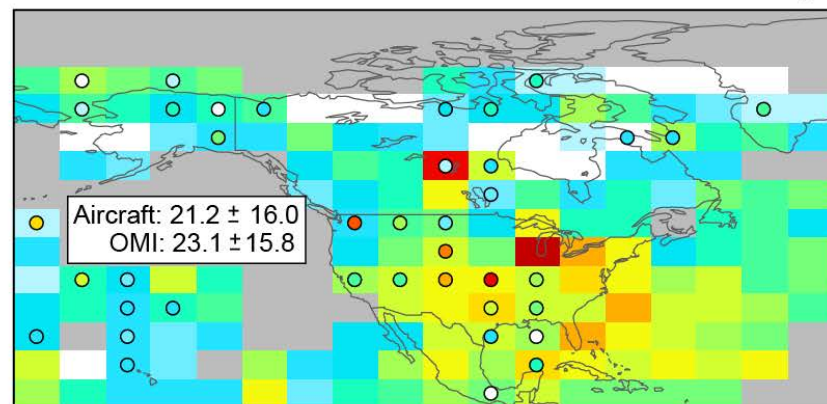
# Evaluate OMI UT NO<sub>2</sub> with aircraft observations

Comparison of May-August OMI and aircraft UT NO<sub>2</sub> during DC8 campaigns

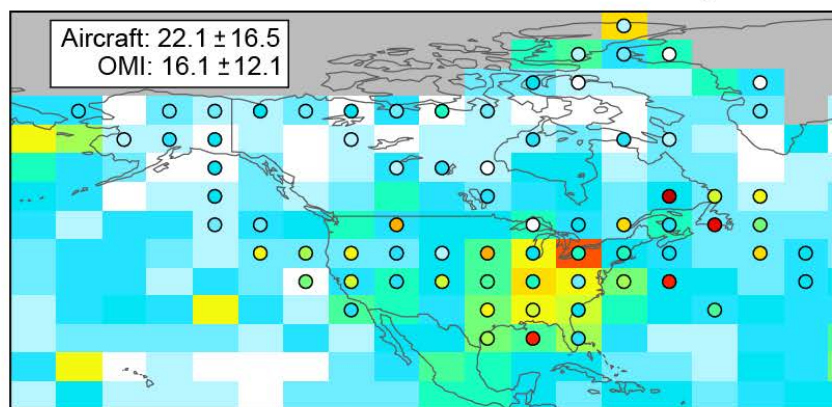
NASA OMI and TD-LIF aircraft NO<sub>2</sub>



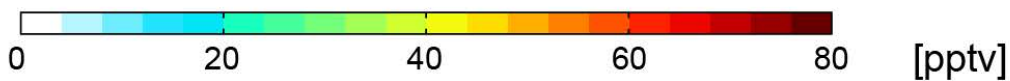
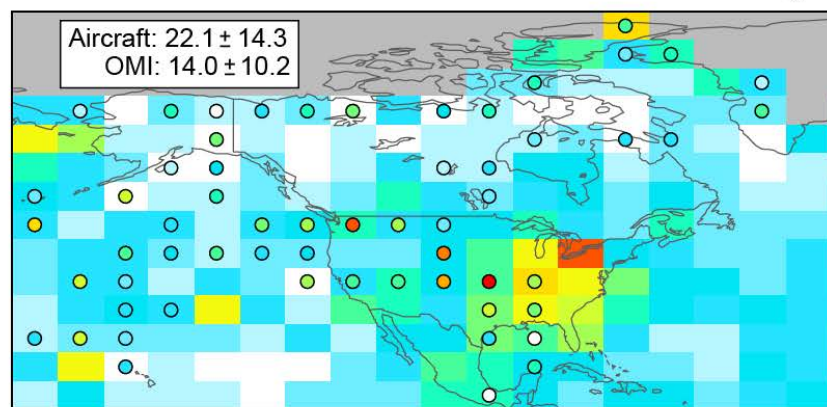
NASA OMI and chemiluminescence aircraft NO<sub>2</sub>



KNMI OMI and TD-LIF aircraft NO<sub>2</sub>



KNMI OMI and chemiluminescence aircraft NO<sub>2</sub>

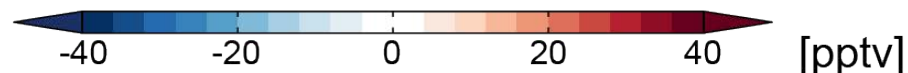
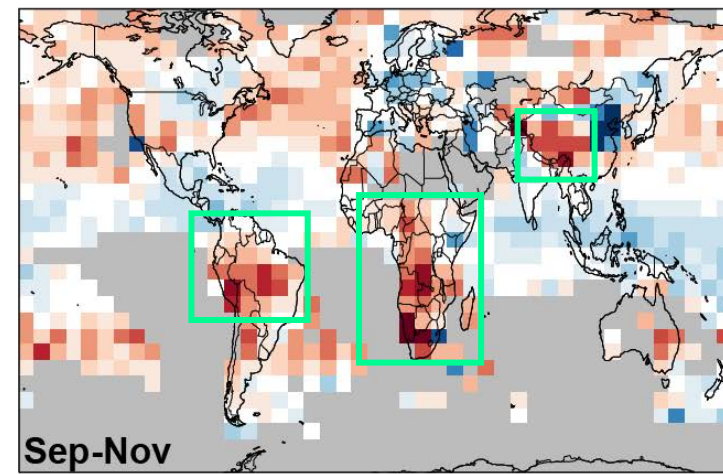
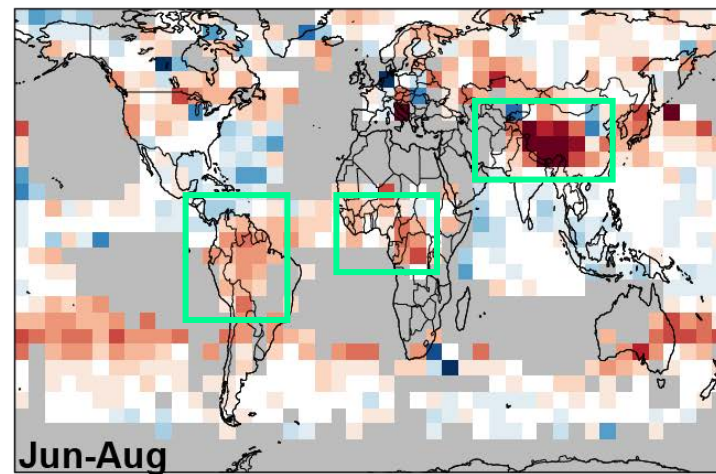
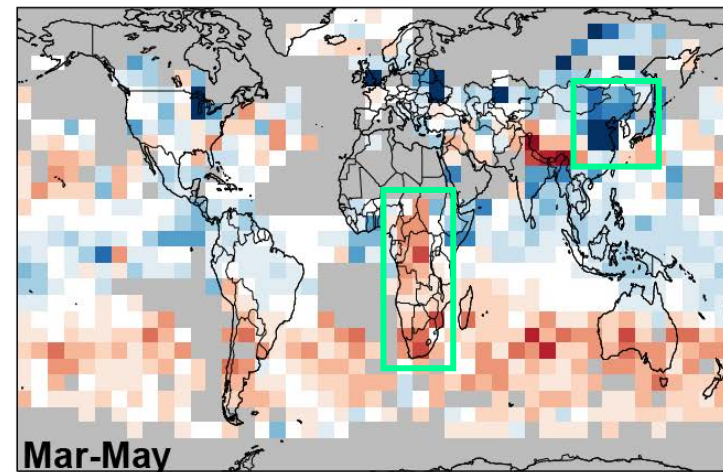
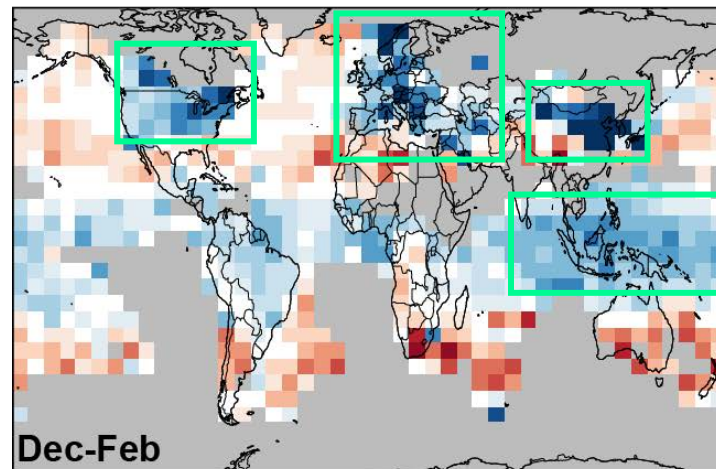


[Aircraft NO<sub>2</sub> from T. Ryerson, R. Cohen, A. Weinheimer]

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

# Compare GEOS-Chem and NASA OMI UT NO<sub>2</sub>

Difference between NASA OMI and GEOS-Chem UT NO<sub>2</sub> (GEOS-Chem minus NASA)



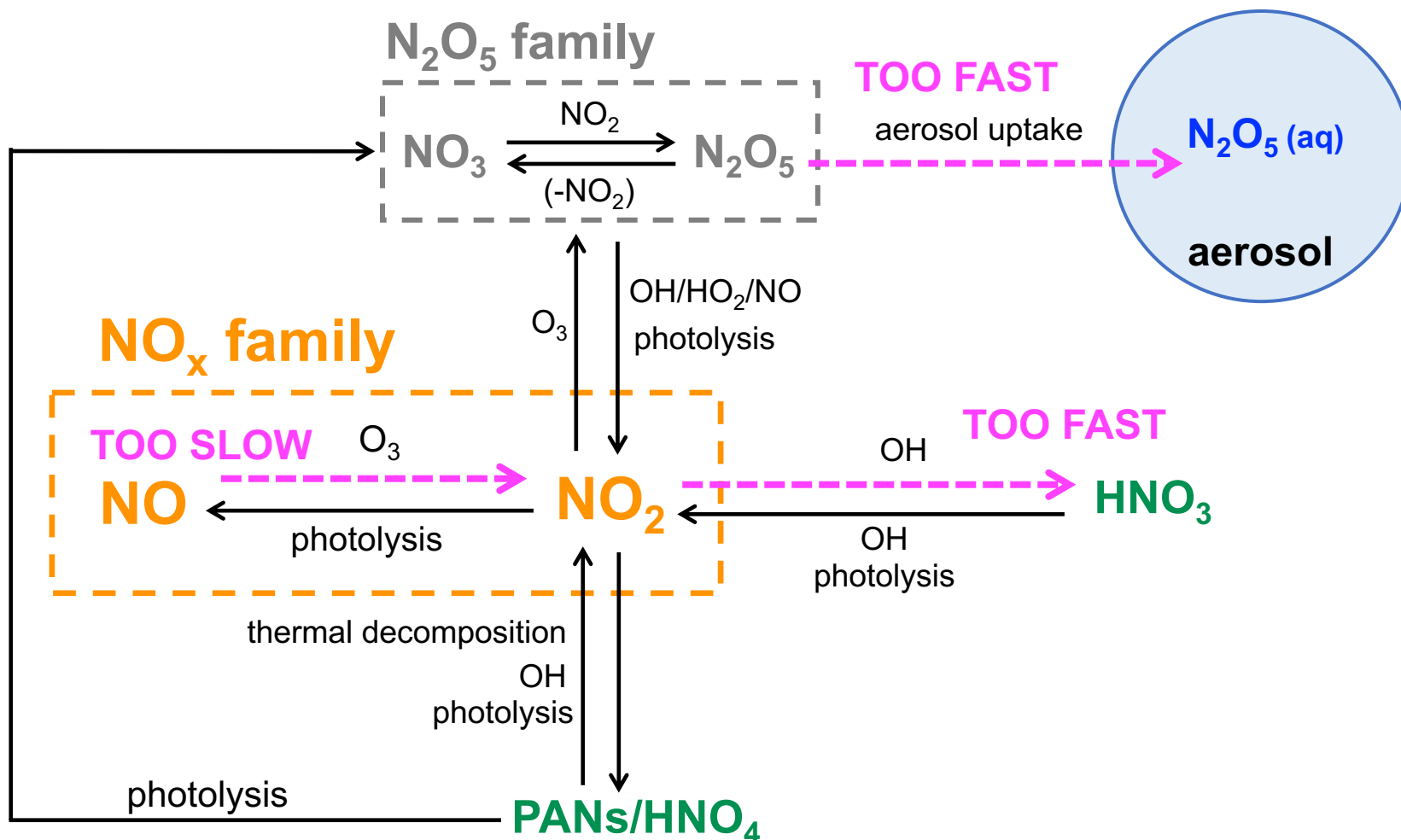
**BLUE:** model too low


**RED:** model too high

 Notable regional discrepancies

# Reactive Nitrogen Chemistry Uncertainties

Uncertainties in  $\text{NO}_x$  chemistry have implications for relative ( $\text{NO}:\text{NO}_2$ ) and total  $\text{NO}_x$  abundances and  $\text{NO}_x$  lifetimes.

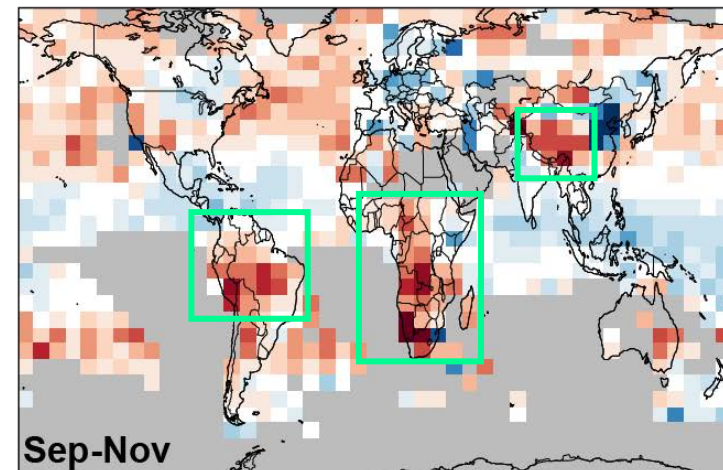
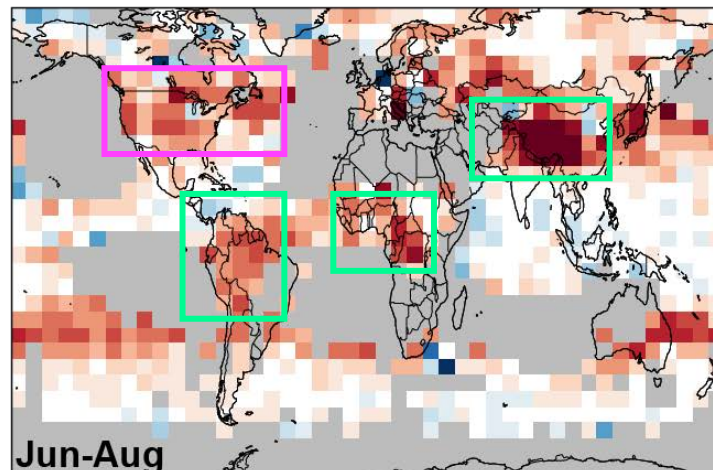
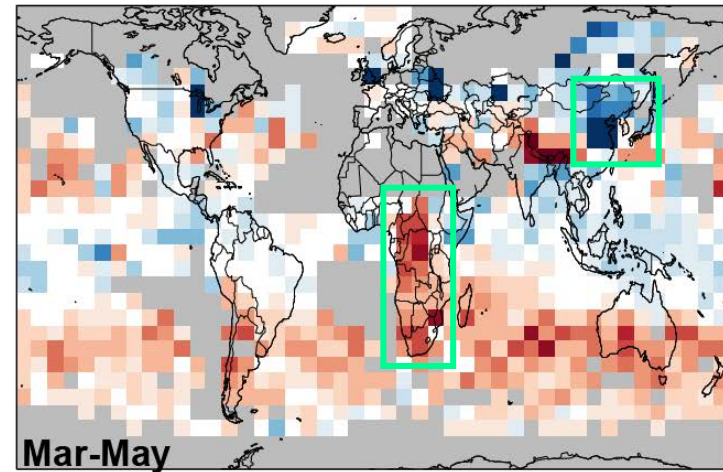
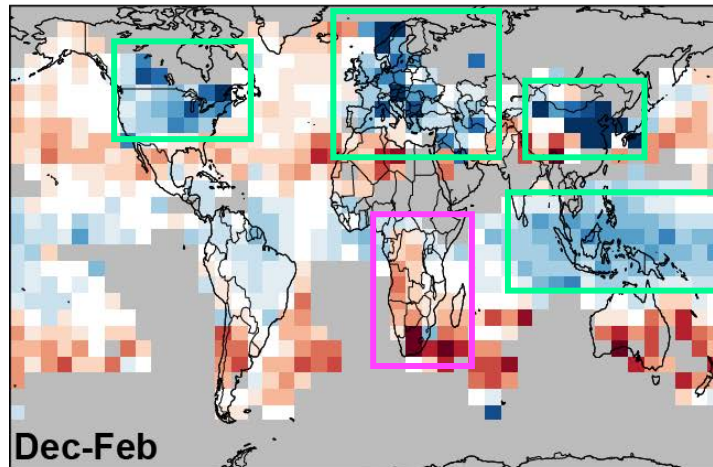


 Uncertain reaction rates that impact  $\text{NO}:\text{NO}_2$  and  $\text{NO}_x$  abundances. Updating these increases  $\text{NO}_2$  abundances.



# Compare GEOS-Chem and NASA OMI UT NO<sub>2</sub>

Updating chemistry increases UT NO<sub>2</sub>, predominantly in the tropics and the Southeast US (Jun-Aug), exacerbating model overestimates in these locations.



**BLUE:** model too low

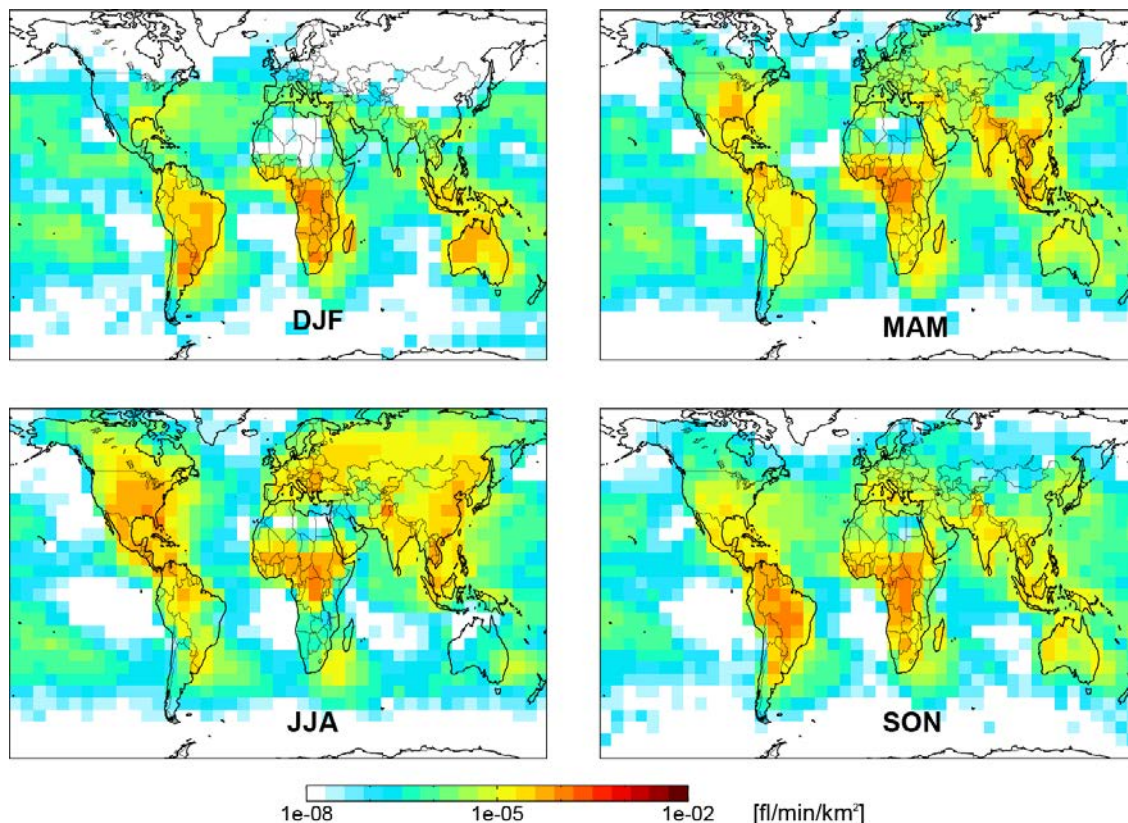
**RED:** model too high

 Additional discrepancies

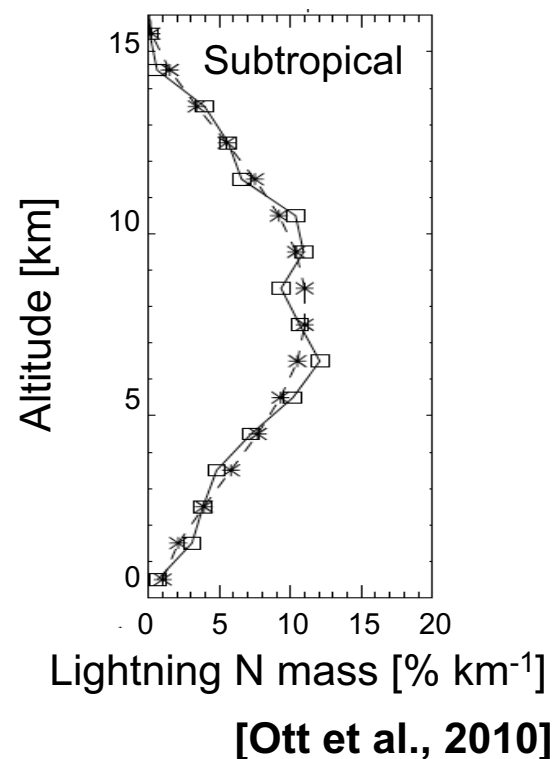
# Lightning $\text{NO}_x$ as a source of discrepancy

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



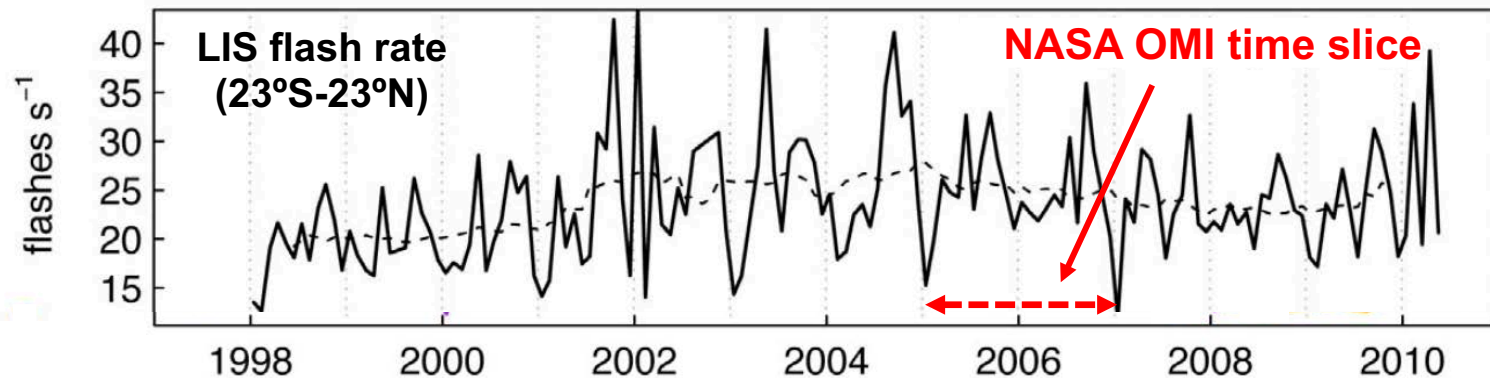
Total GEOS-Chem lightning  $\text{NO}_x$  emissions for 2006: **5.4 Tg N**

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



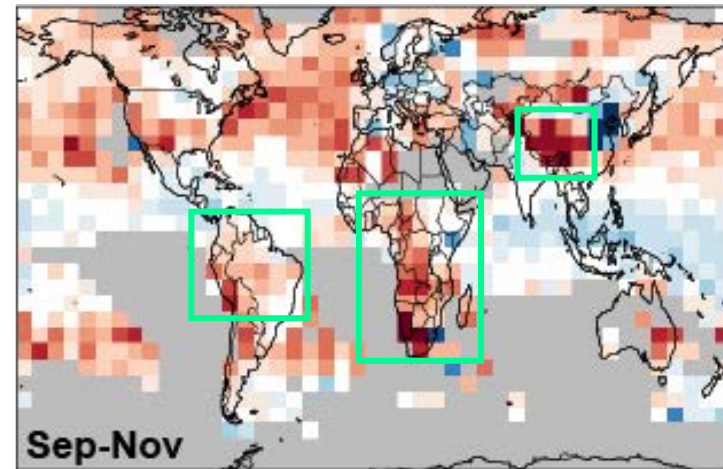
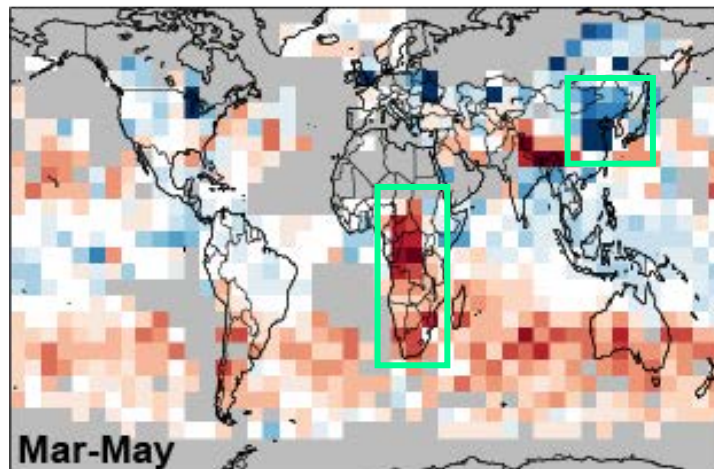
# Lightning activity interannual variability (IAV)

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

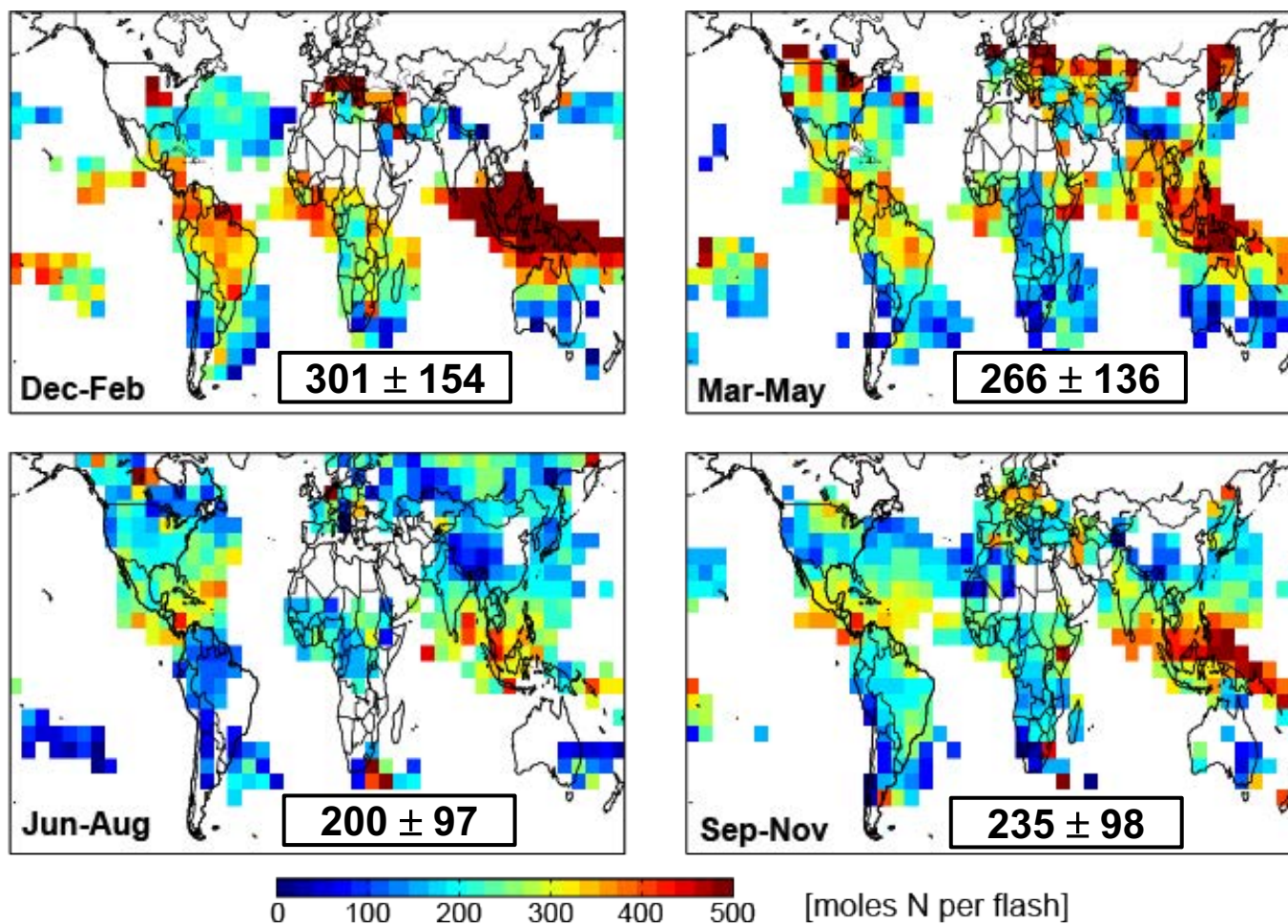
 Discrepancies



# NO<sub>x</sub> production per lightning flash

Single value (260 moles NO per flash) used in GEOS-Chem based on limited observations, so represents an under constrained parameter

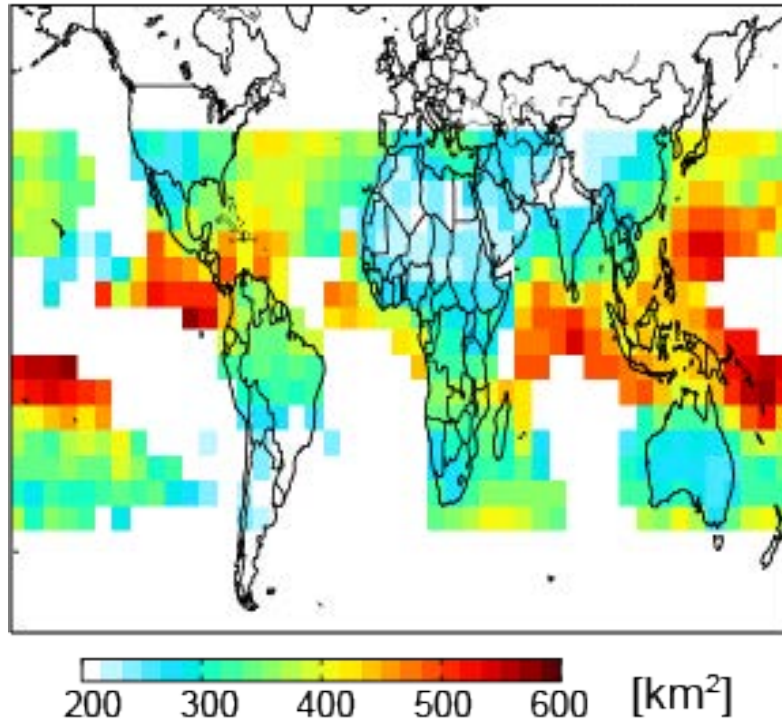
Scale 260 mol/fl by the discrepancy between the model and OMI to estimate spatially resolved NO<sub>x</sub> production per flash (*Data are at 8° × 5°*)



# NO<sub>x</sub> per flash as a function of lightning intensity

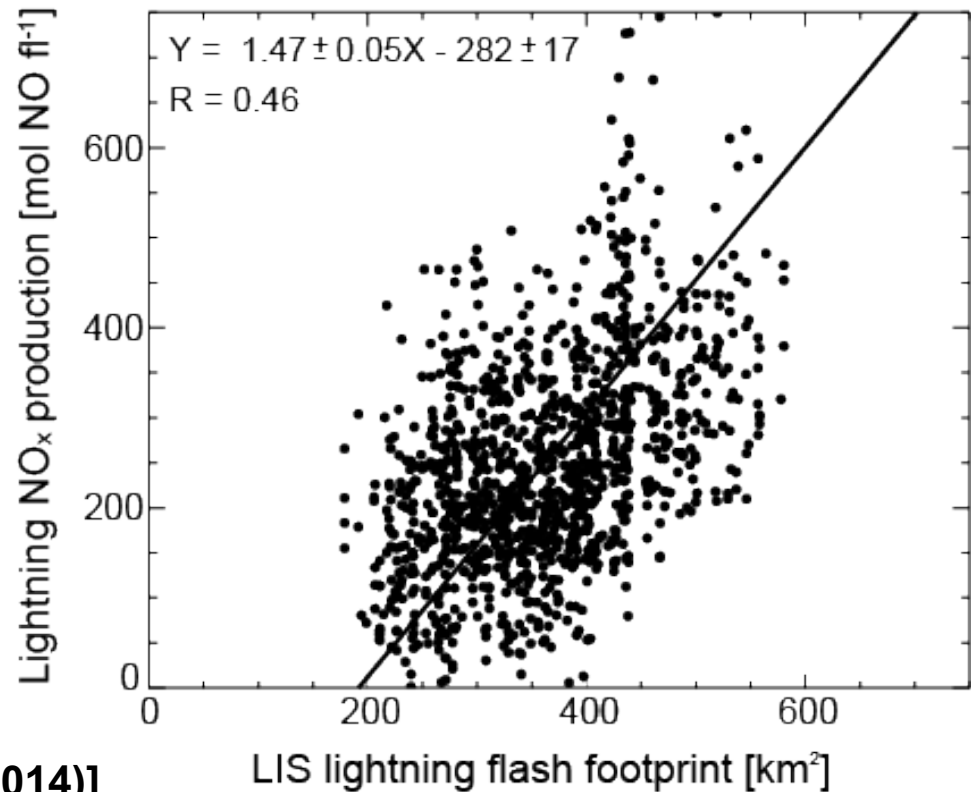
Combine OMI-derived NO per flash and OTD- and LIS-derived lightning footprints

LIS Lightning flash footprint



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

NO<sub>x</sub> production rate versus flash footprint



Extensive 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<sub>x</sub> production rate and flash footprint.

Potential to move towards a more mechanistic representation of NO<sub>x</sub> production rates.

# Summary and Concluding Remarks

Used new OMI products from NASA and KNMI to provide constraints on upper troposphere (UT)  $\text{NO}_x$  where production efficiency and global warming potential of ozone are high.

- NASA OMI UT  $\text{NO}_2$  observations are more consistent with aircraft DC8 observations than is KNMI
- GEOS-Chem chemical transport model UT  $\text{NO}_2$  has large regional discrepancies when compared to NASA UT  $\text{NO}_2$
- 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  $\text{NO}_x$  production per flash and LIS flash footprints
- Next steps: independently evaluate with  $\text{NO}_2$  observations from the ATom aircraft campaign and determine the implications for UT ozone
- Caveats: the northern hemisphere wintertime (Dec-Feb) discrepancy remains