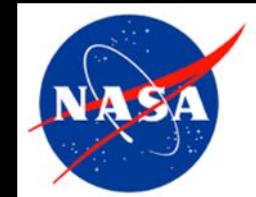


First Estimate of Upper Tropospheric NO₂ from TROPOMI



Eloise A. Marais, Nana Wei, Henk Eskes, Sungyeon Choi, Joanna Joiner, Alexander Cede

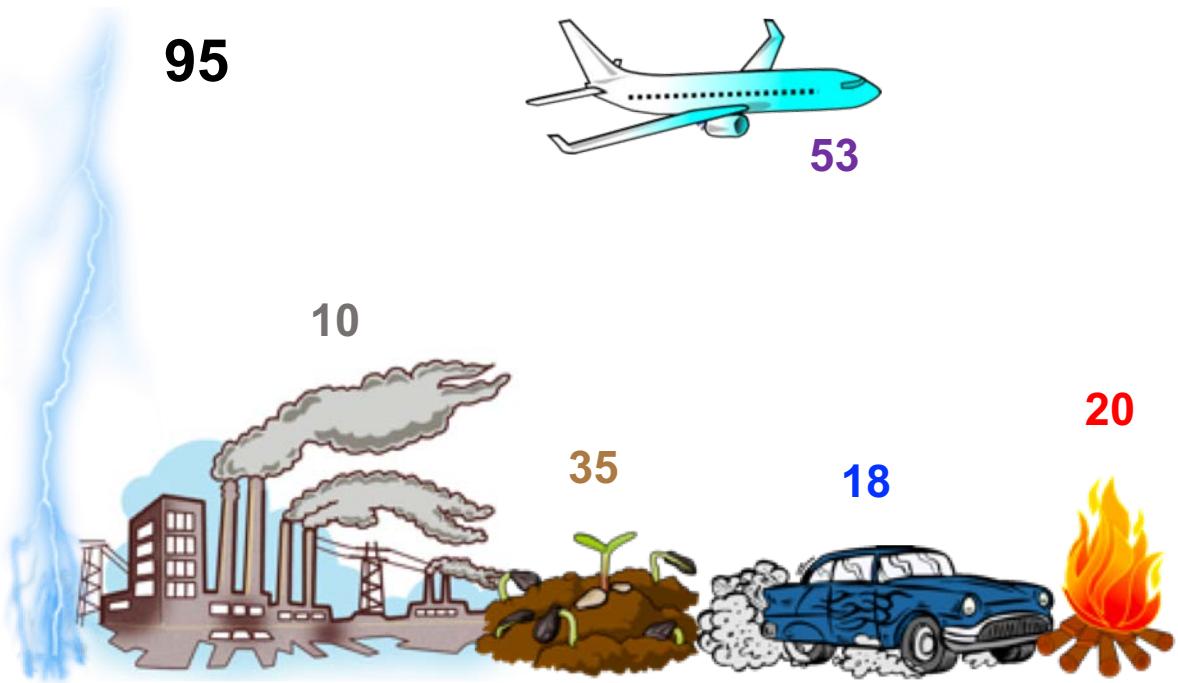
EOS Group Meeting
3 April 2020

eloise.marais@le.ac.uk
<http://maraisresearchgroup.co.uk/>

Global Relevance of the Upper Troposphere (~8-12 km)

NO_x ($\text{NO} + \text{NO}_2$) is very efficient at forming ozone in the upper troposphere where ozone is a potent greenhouse gas

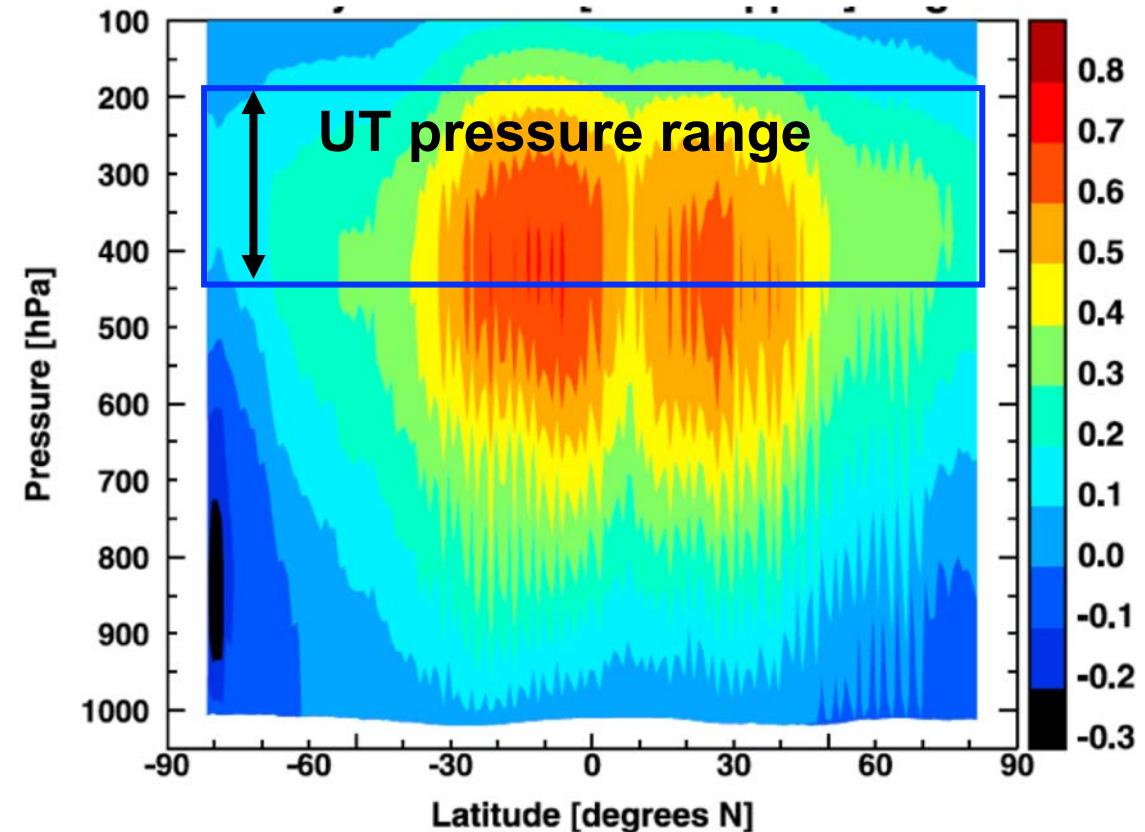
OPE (molecules O_3 /molecule NO_x) for individual NO_x sources



[adapted from Dahlmann et al., 2011]

Longer NO_x lifetime at higher altitude → greater OPE

Sensitivity of outgoing longwave radiation to variations in ozone concentrations

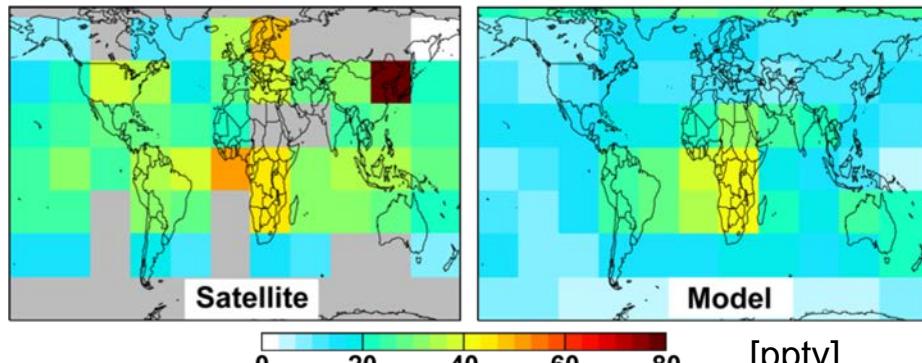


[Aghedo et al., 2011]

Errors in the UT Impact the Whole Atmosphere

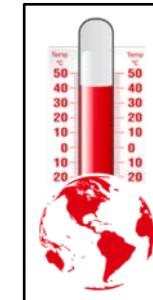
Small uncertainties in NO_x in the UT induce large climate and air quality errors

Model NO₂ too low across the UT in the northern midlatitudes



[Marais et al., 2018]

efficient ozone production



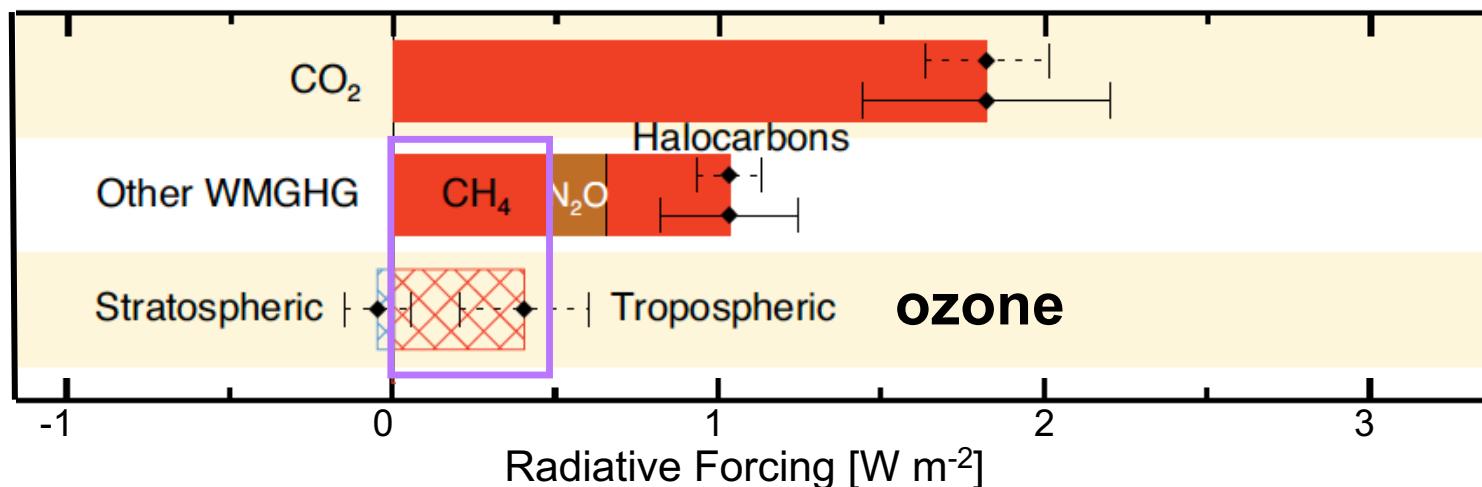
satellite NO₂ retrievals



Errors in tropospheric ozone radiative forcing

Uncertain global air quality constraints from satellites

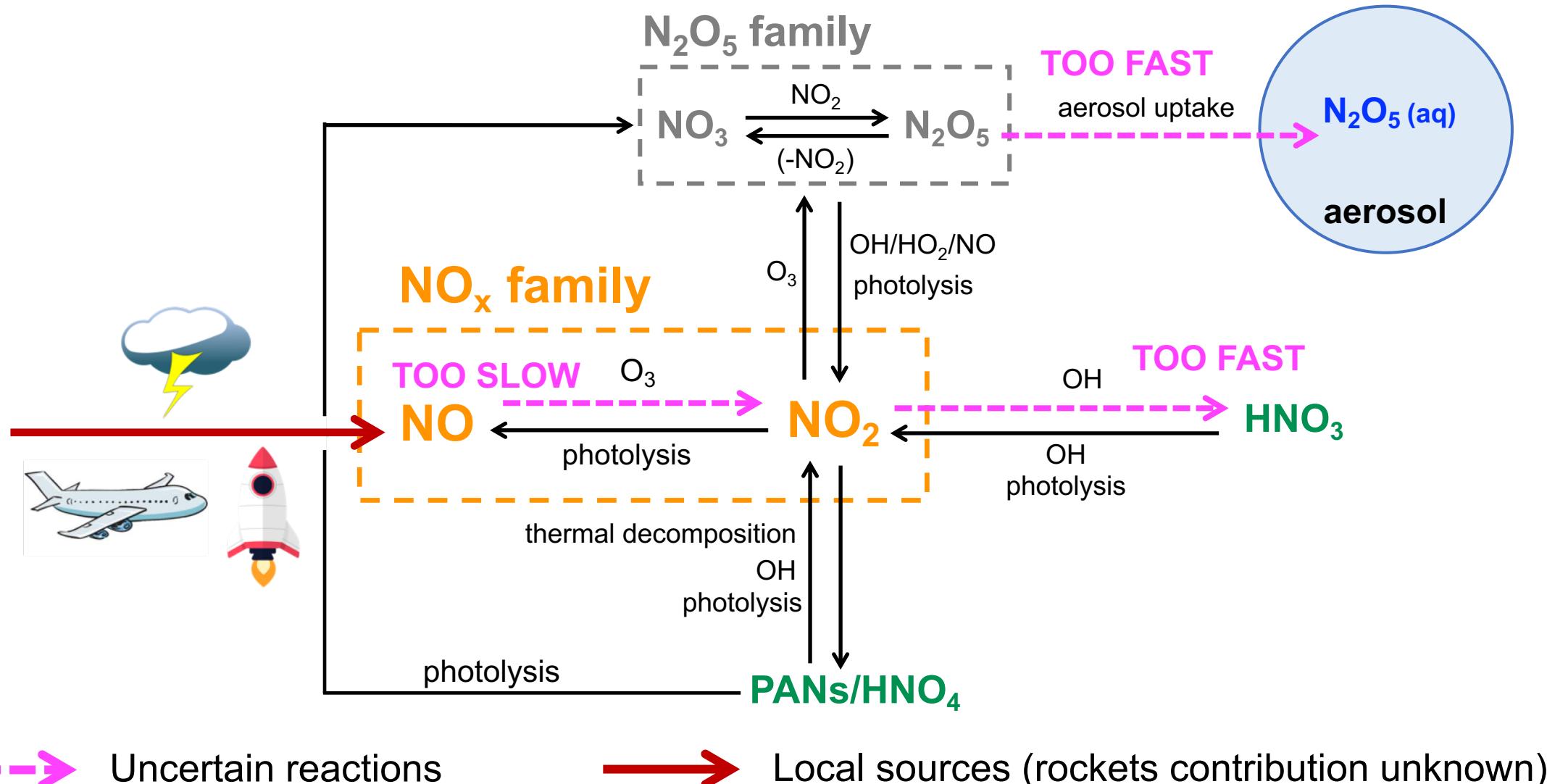
Tropospheric ozone and methane have near-equal climate impacts



[IPCC AR5, 2013]

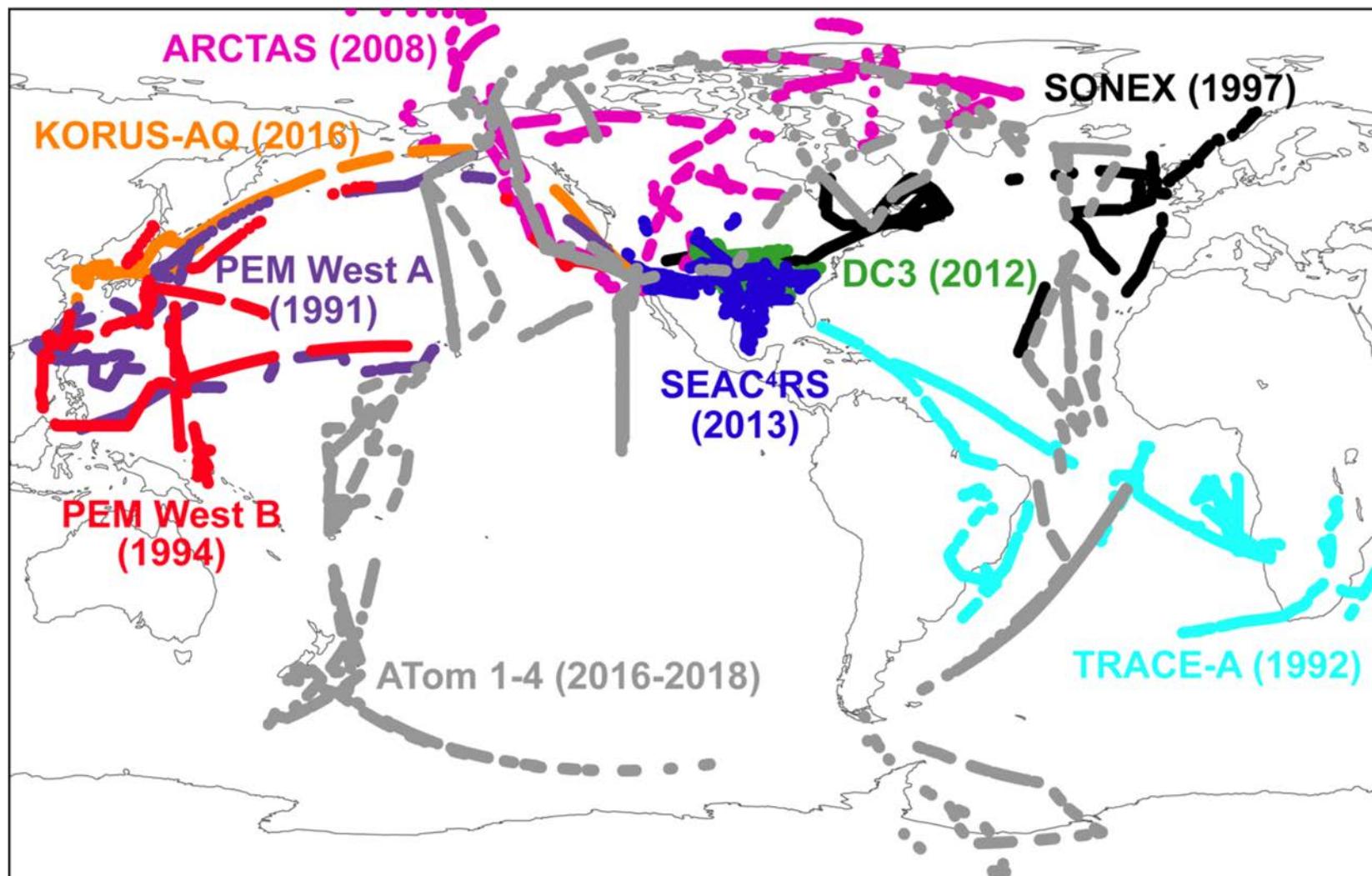
Large Uncertainties in Chemistry of Reactive Nitrogen

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



NO_x Measurements are Limited in Time and Space

Spatial distribution of data from NASA DC8 aircraft campaigns in the upper troposphere

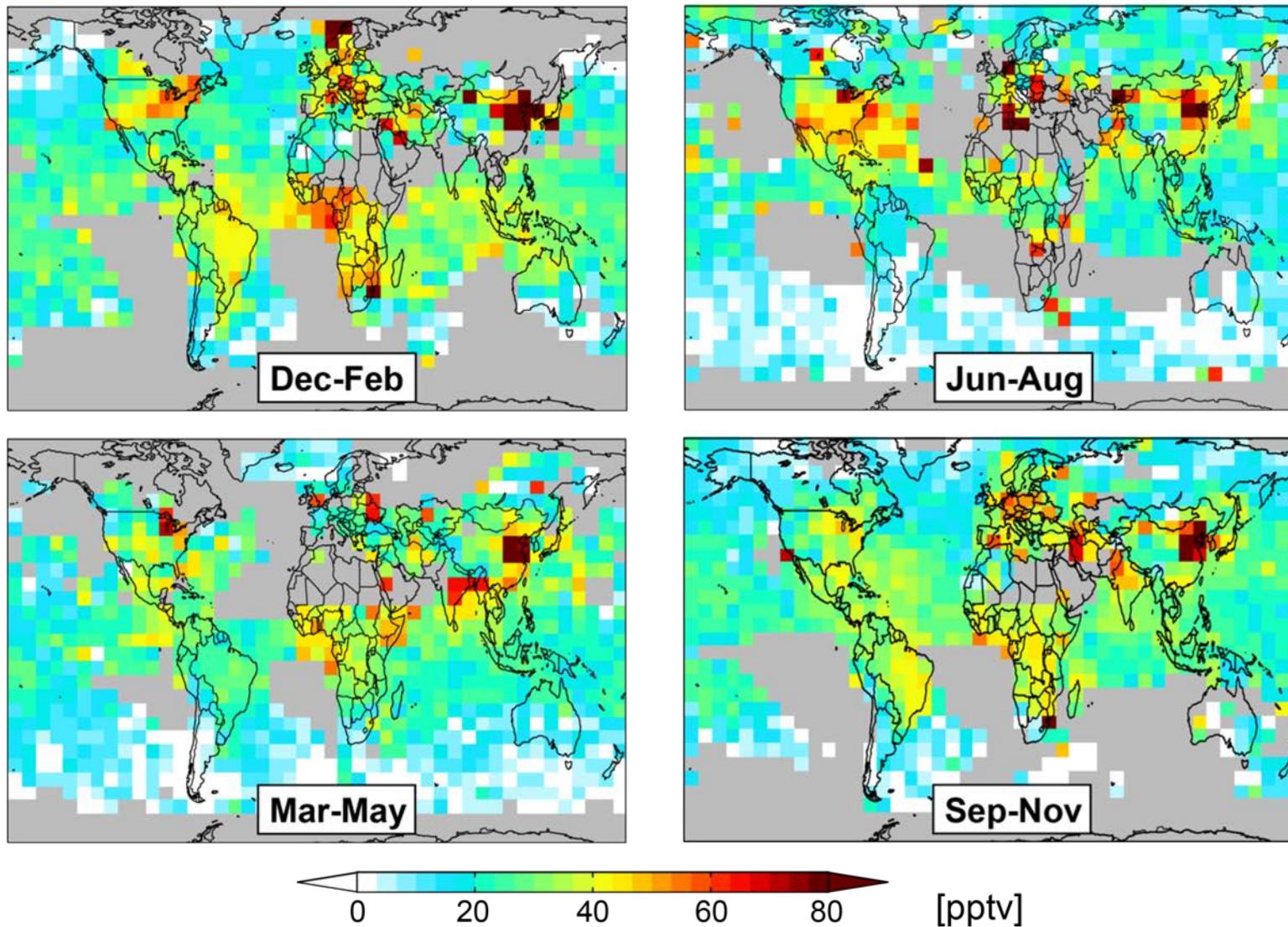


NO_2 in situ instruments are also susceptible to positive biases in the upper troposphere

New UT NO₂ products from the Ozone Monitoring Instrument (OMI)

Near global spatial coverage of seasonal mean UT NO₂ at $5^\circ \times 8^\circ$ (latitude \times longitude)

NASA OMI upper troposphere NO₂ (2005-2007)



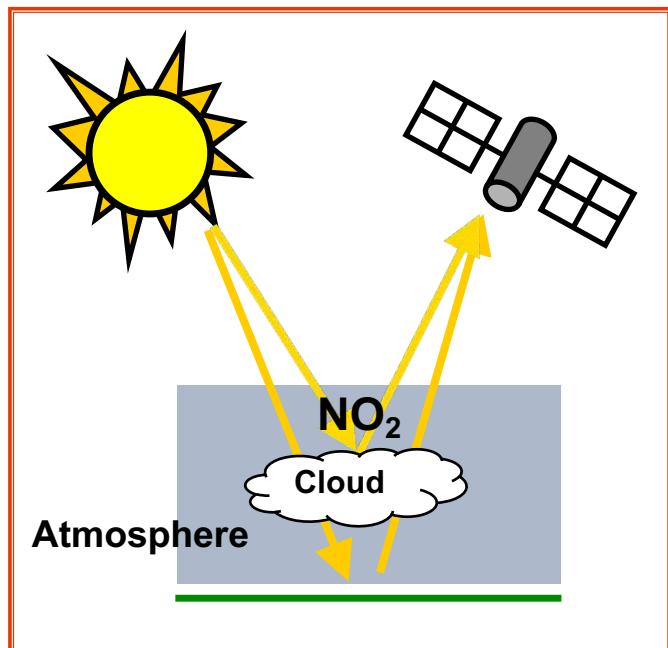
[Marais et al., 2018;
Choi et al., 2014]

UT NO₂ Obtained with the Innovative Cloud-Slicing Technique

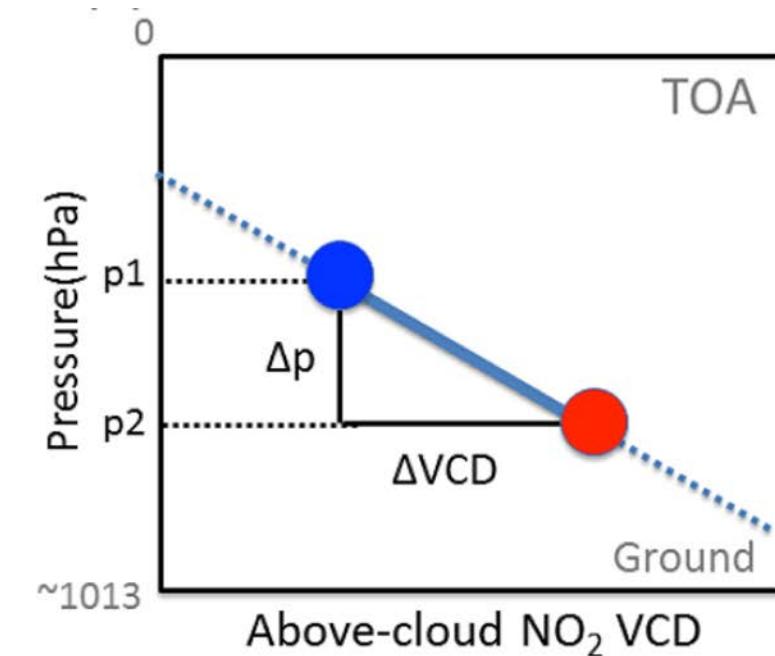
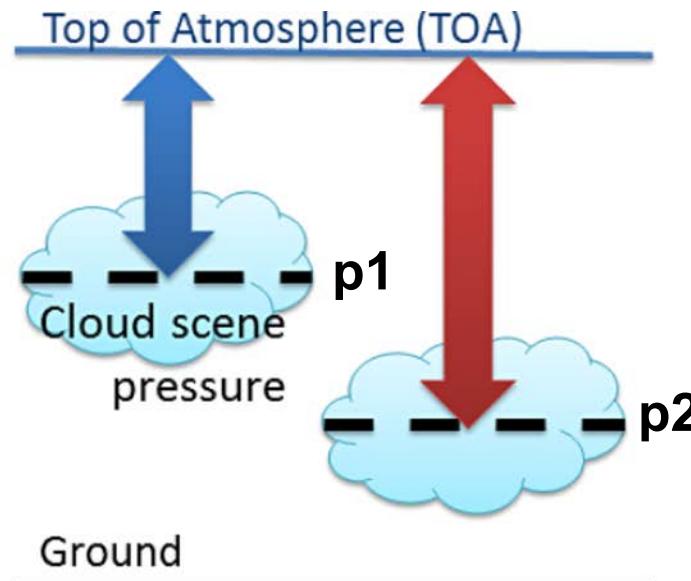
First applied by Ziemke et al. [2001] to TOMS ozone

Retrieve partial NO₂ columns over cloudy scenes at different heights

APPROACH



Use cloud height variability to derive partial columns



[adapted from Choi et al., 2014]

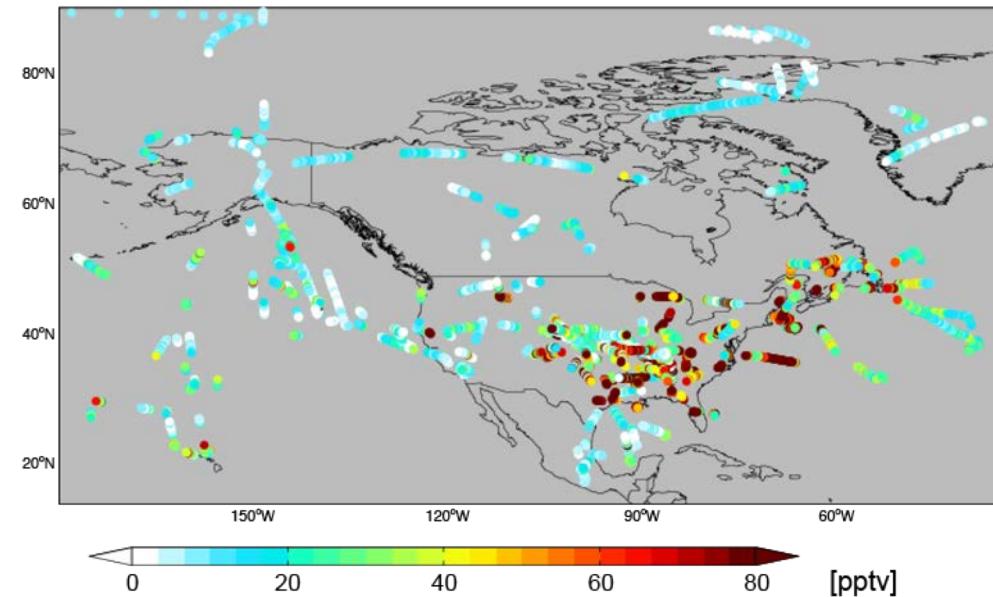
NO₂ volume mixing ratio (VMR) between clouds at p1 and p2

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

Consistency with Aircraft Observations of NO₂

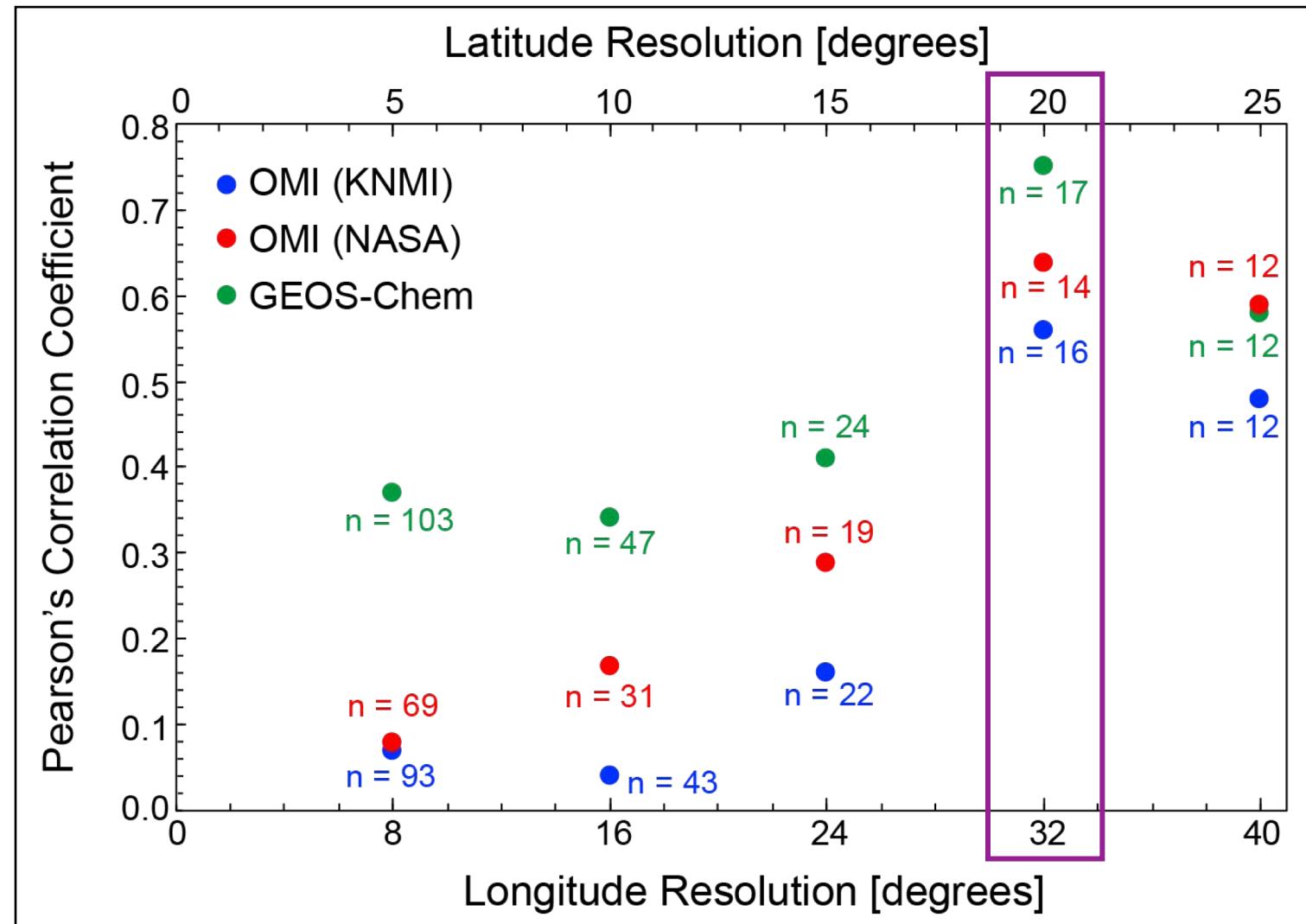
Agreement in spatial distribution between (reliable) aircraft and OMI UT NO₂ observations, but at coarse scales (seasonal, 20° latitude (2000 km) × 32° longitude (3200 km))

NASA DC8 NO₂ in Spring-Summer



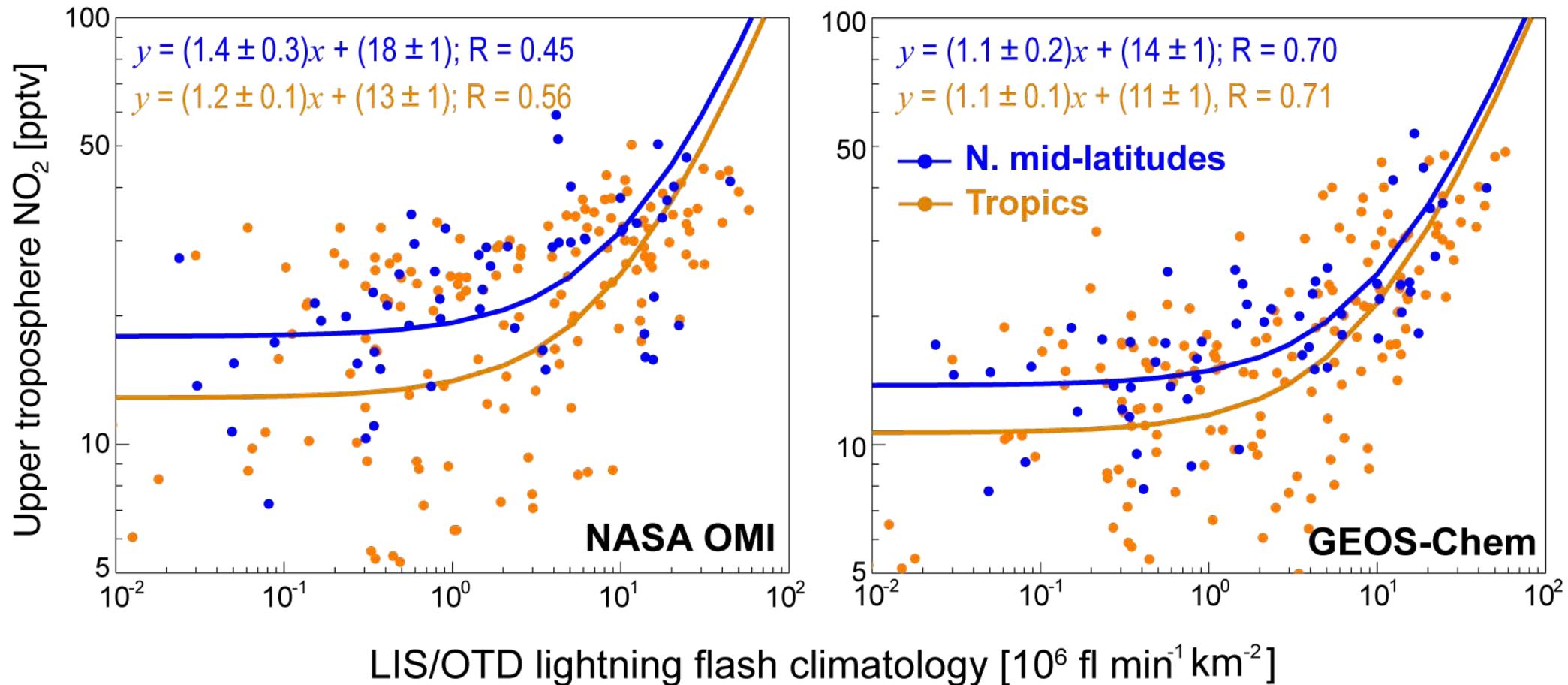
NO₂ obtained for multiple campaigns
from Ron Cohen's TD-LIF instrument

[Marais et al., 2018]



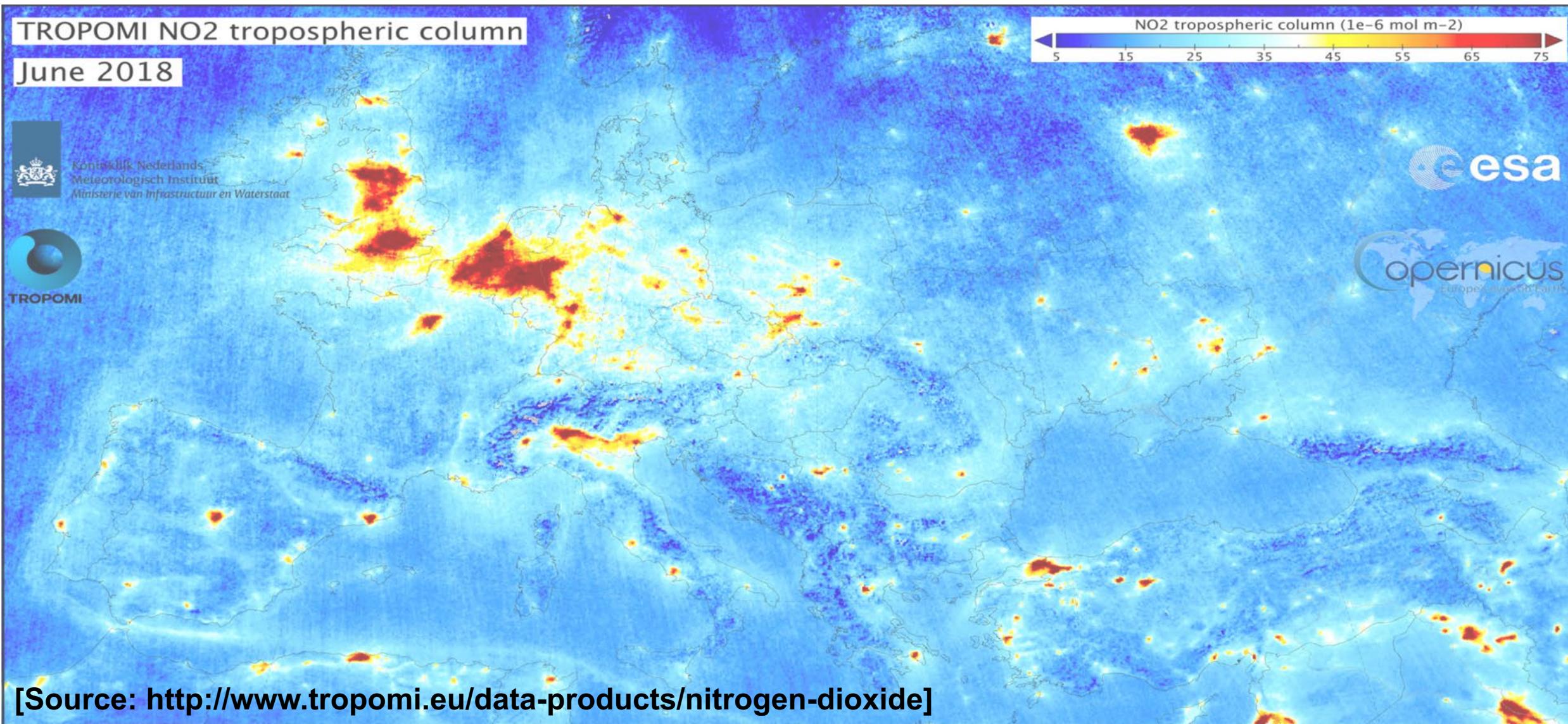
Used OMI UT NO₂ to Better Constraint Global Lightning NO_x

Log-log relationship between UT NO₂ from OMI and GEOS-Chem and satellite observations of lightning flashes in the **northern midlatitudes** and **tropics**



Similar slope in northern midlatitudes and tropics supports similar lightning NO_x production rates

Can We Do Better with the High-Resolution TROPOMI?

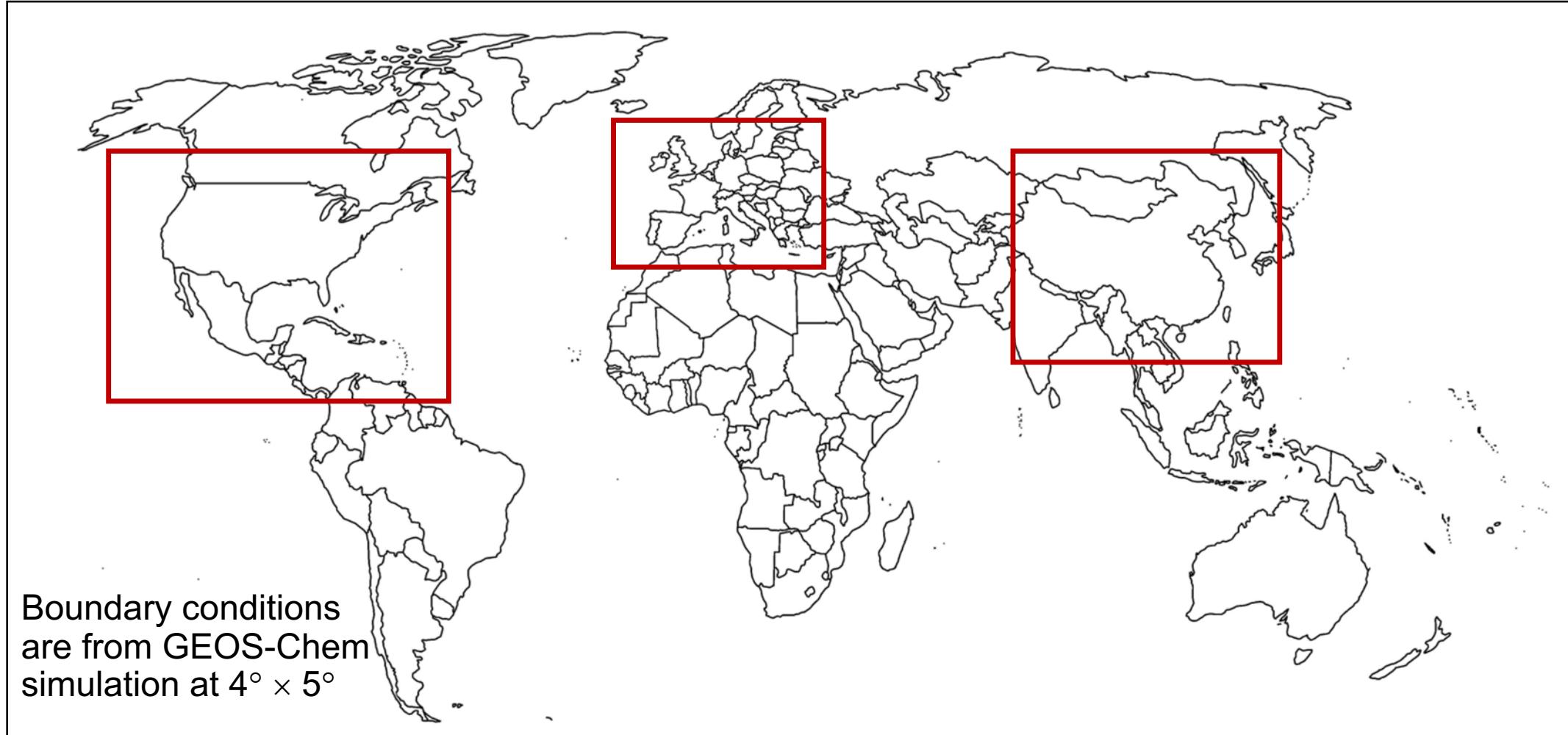


Nadir spatial resolutions in km (along × across): **13 × 24 (OMI)**; **5.6 × 3.5 (TROPOMI)**

Synthetic Cloud-Slicing Experiment with GEOS-Chem

Determine whether UT NO₂ obtained with cloud-slicing reproduces UT NO₂ obtained by averaging across the UT (450-180 hPa)

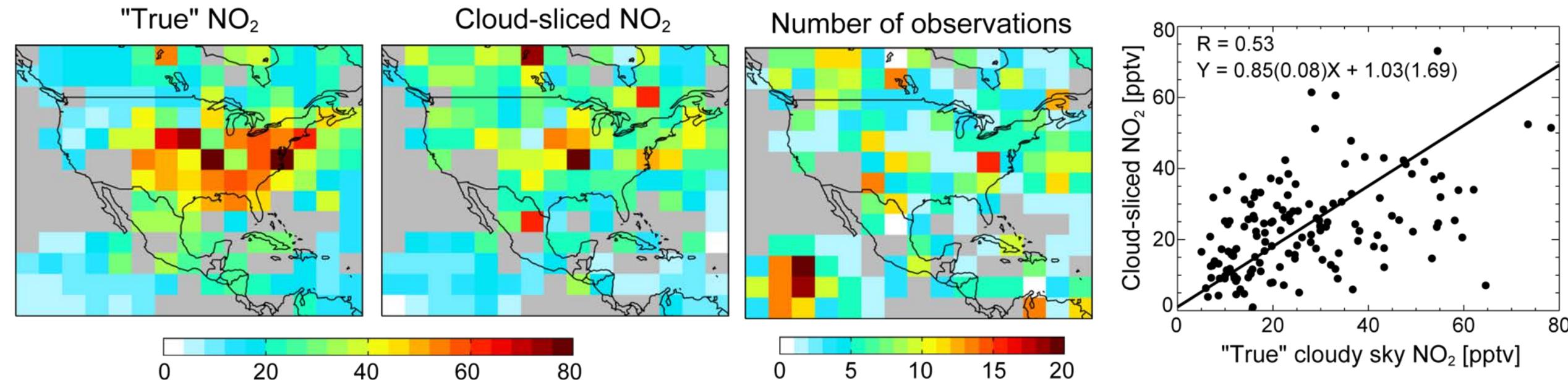
GEOS-Chem nested domains at $0.25^\circ \times 0.3125^\circ$ driven with NASA GEOS-FP meteorology



Synthetic Cloud-Slicing Experiment with GEOS-Chem

Compare “true” and cloud-sliced UT NO₂ from GEOS-Chem over North America in June-August

Daily cloud-sliced NO₂ obtained at $4^\circ \times 5^\circ$ from daily nested grid GEOS-Chem partial columns sampled around the satellite overpass



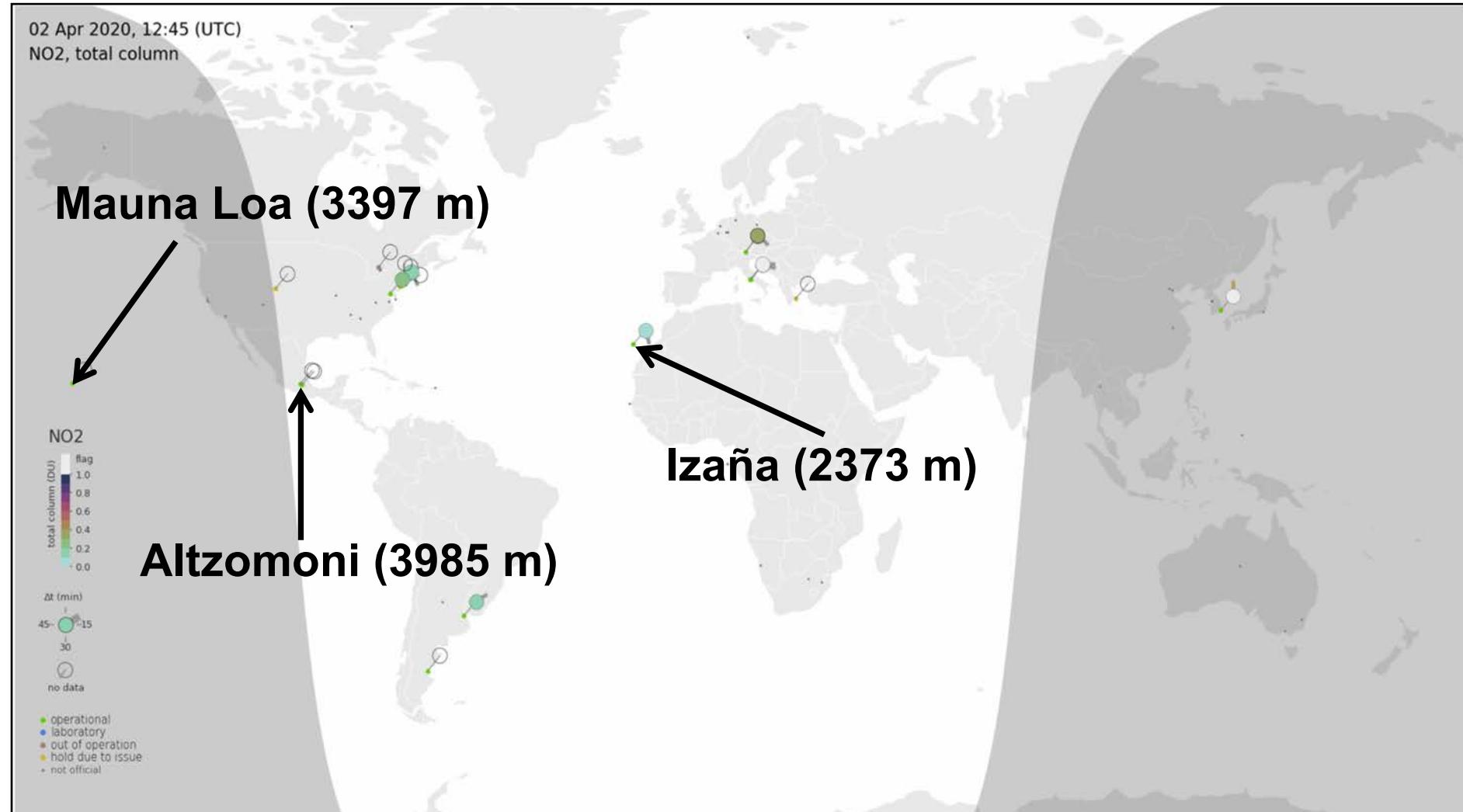
Sampling conditions informed by Choi et al. [2014] approach:

Satellite overpass (12h00-15h00), dynamic cloud top height range (standard deviation > 35 hPa, range > 160 hPa), and UT NO₂ with relative error < 100%, < 200 pptv, significantly greater than zero.

Initial results are encouraging ($R > 0.5$). Data density is an issue. Simulating more years.

Evaluate TROPOMI total column NO₂ against Pandora

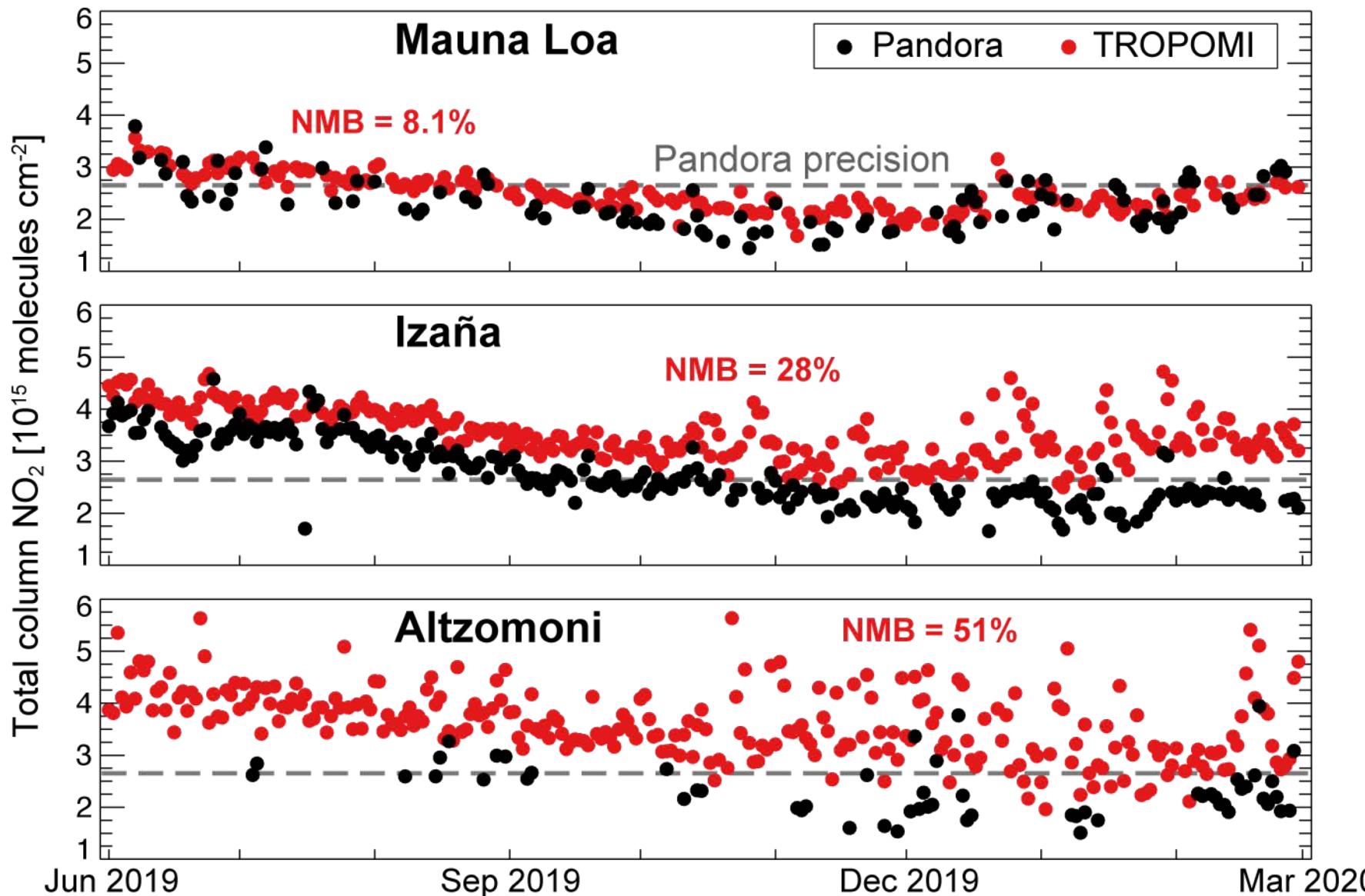
Global Pandora network, indicating locations of high-altitude sites (large relative contribution from the UT) used to evaluate TROPOMI



[Source: <https://www.pandonia-global-network.org/>]

Evaluate TROPOMI total column NO₂ against Pandora

Pandora and TROPOMI total column NO₂ time series at the 3 measurement stations



Sample Pandora \pm 30 min of TROPOMI overpass and TROPOMI within 20 km of Pandora site

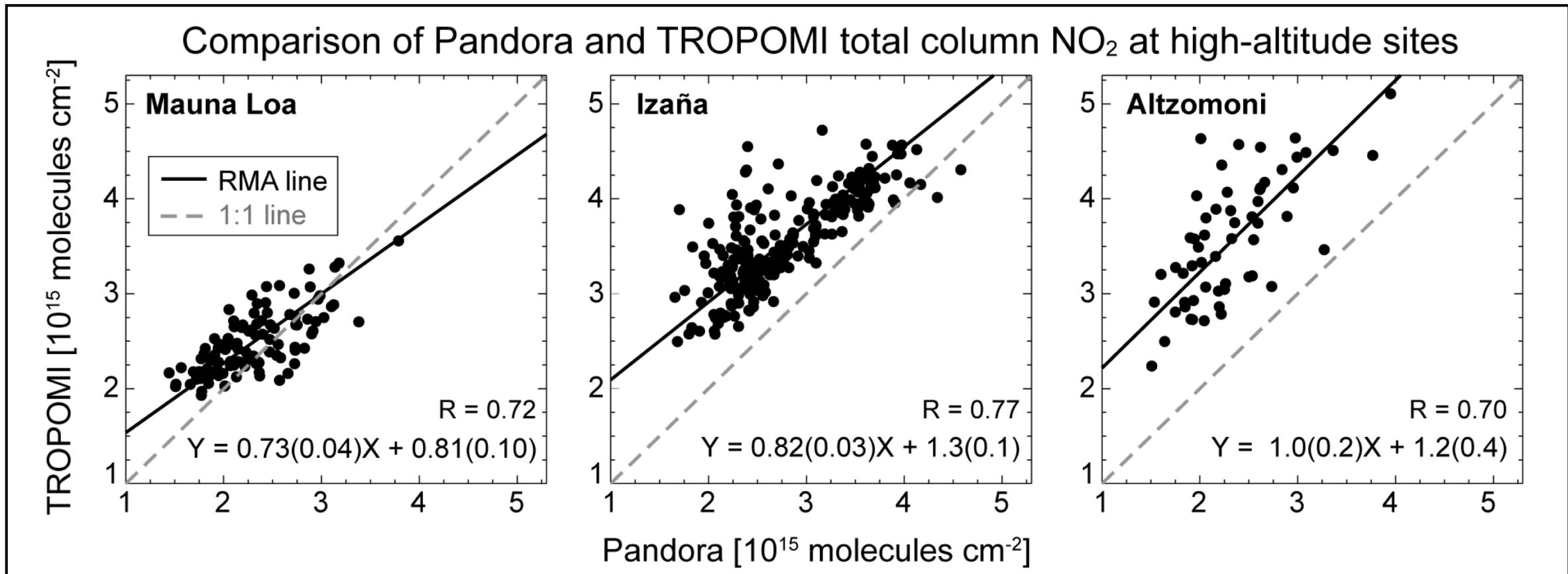
Focus is on cloudy scenes, so use a TROPOMI qa_value threshold of 0.45

Data close to estimated Pandora precision

TROPOMI normalized mean bias (NMB) for coincident observations is 8.1-51%

Evaluate TROPOMI total column NO₂ against Pandora

Scatterplots of TROPOMI versus Pandora total column NO₂ at the 3 stations



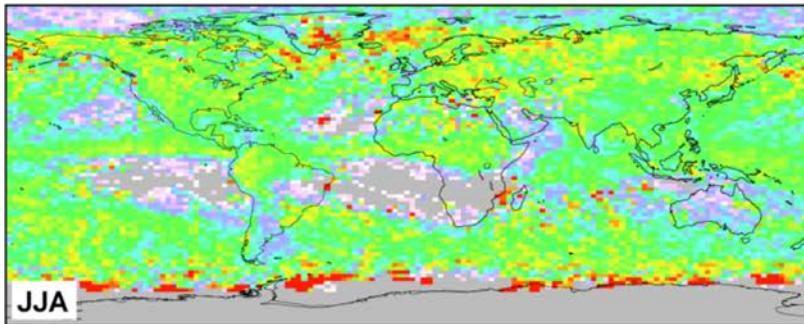
Positive intercept supports systematic bias in TROPOMI total column

TROPOMI validation documents show a slight underestimate (10%) in the stratospheric column

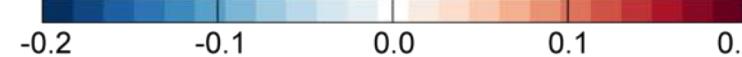
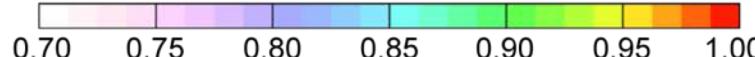
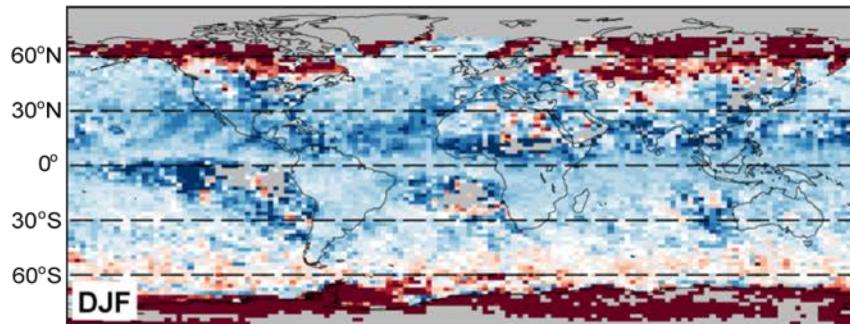
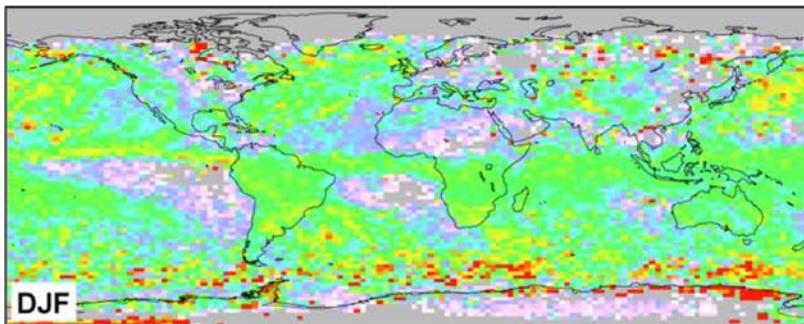
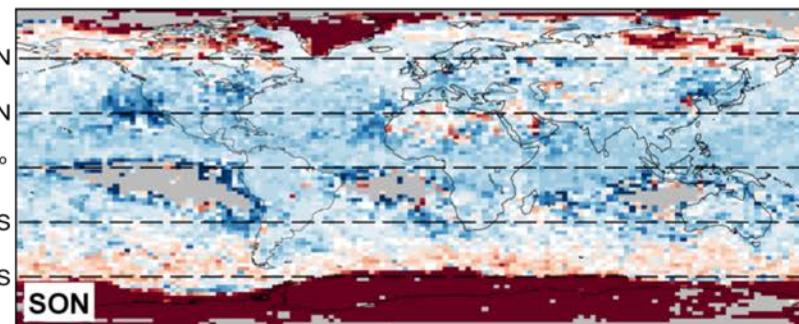
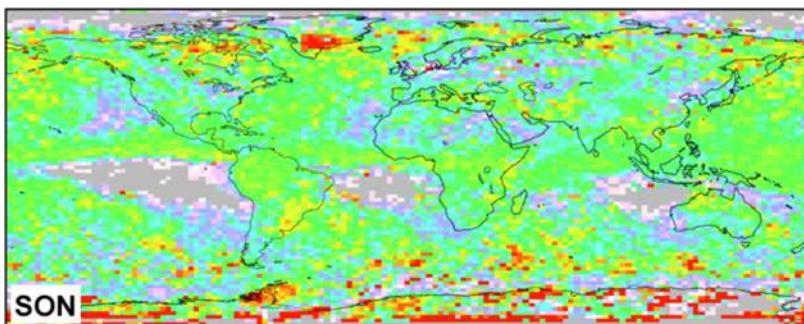
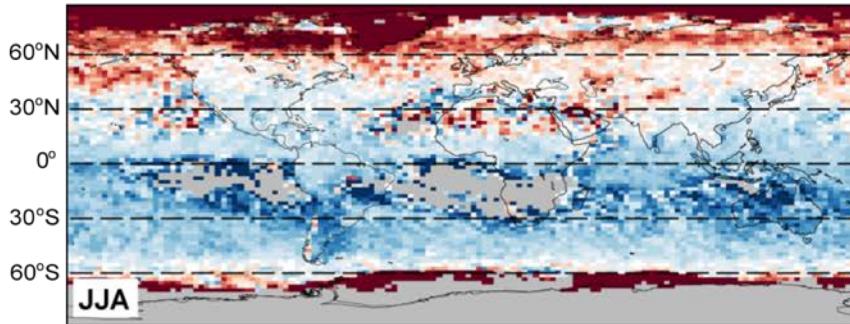
Evaluate TROPOMI Cloud Fractions

Seasonal mean FRESCO cloud fraction and the difference between FRESCRO and DLR-OCRA

FRESCO cloud fraction



FRESCO minus DLR-OCRA cloud fraction



Only consistent data versions are used from June 2019 to February 2020

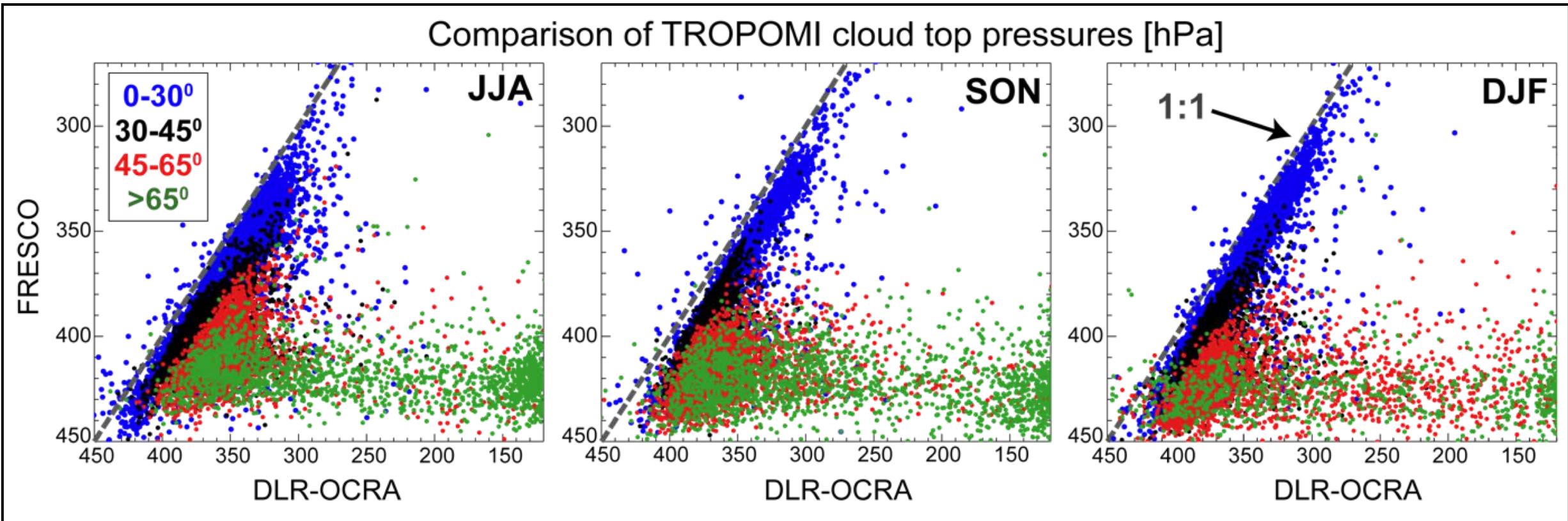
FRESCO: cloud product used in the TROPOMI NO₂ column retrieval

DLR-OCRA: Official TROPOMI cloud product (*OFFL_L2_CLOUD*)

Compare scenes with FRESCO cloud fraction > 0.7

Evaluate TROPOMI Cloud Top Heights

Scatterplots of seasonal mean of TROPOMI cloud products at $2^\circ \times 2.5^\circ$ colored by latitude bands



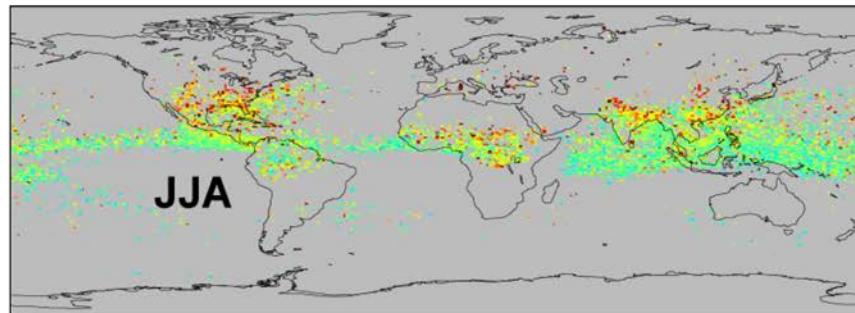
Compare scenes with FRESCO cloud top pressure range of 180 to 450 hPa

DLR-OCRA systematically lower than FRESCO and discrepancy increases with latitude

At poles ($>65^\circ$ N/S) DLR-OCRA ranges from <150-450 hPa, whereas FRESCO range is 400-450 hPa

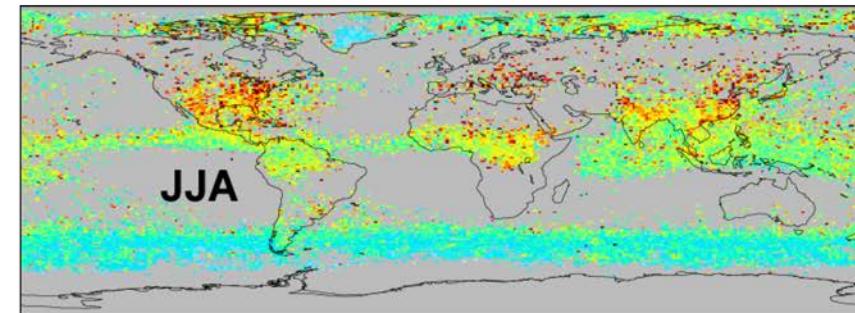
Seasonal Mean TROPOMI Cloud-sliced UT NO₂

TROPOMI-FRESCO UT NO₂

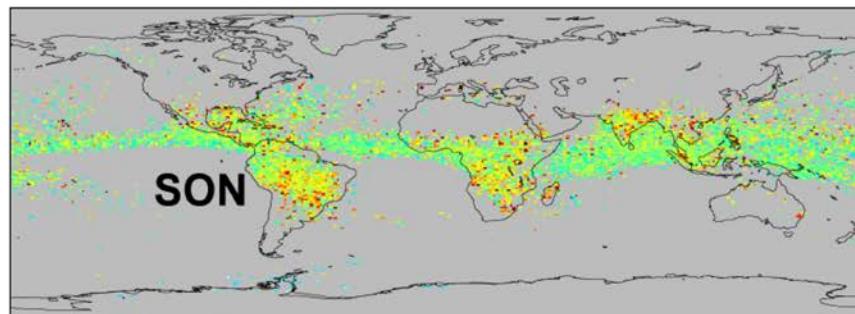


JJA

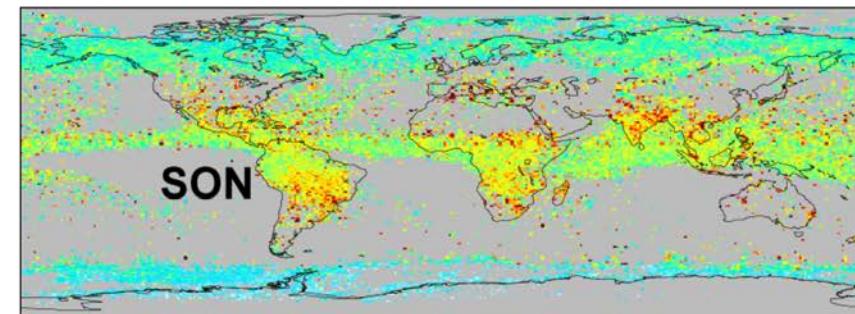
TROPOMI-DLR-OCRA UT NO₂



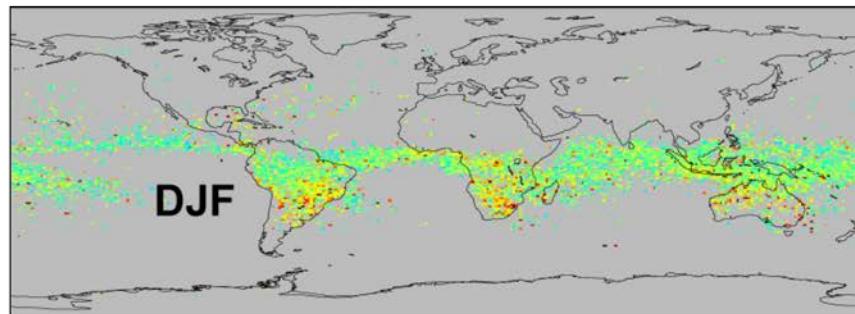
JJA



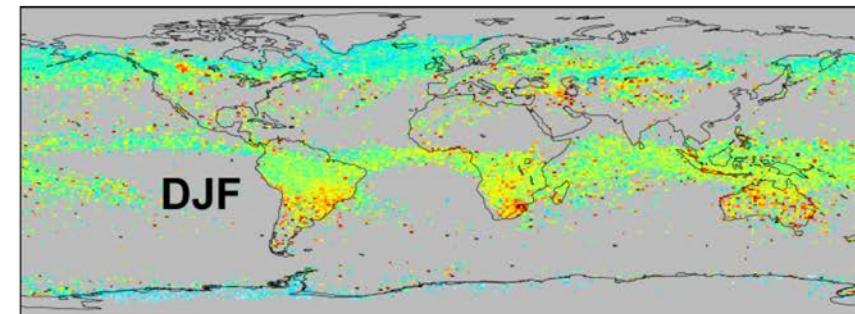
SON



SON



DJF



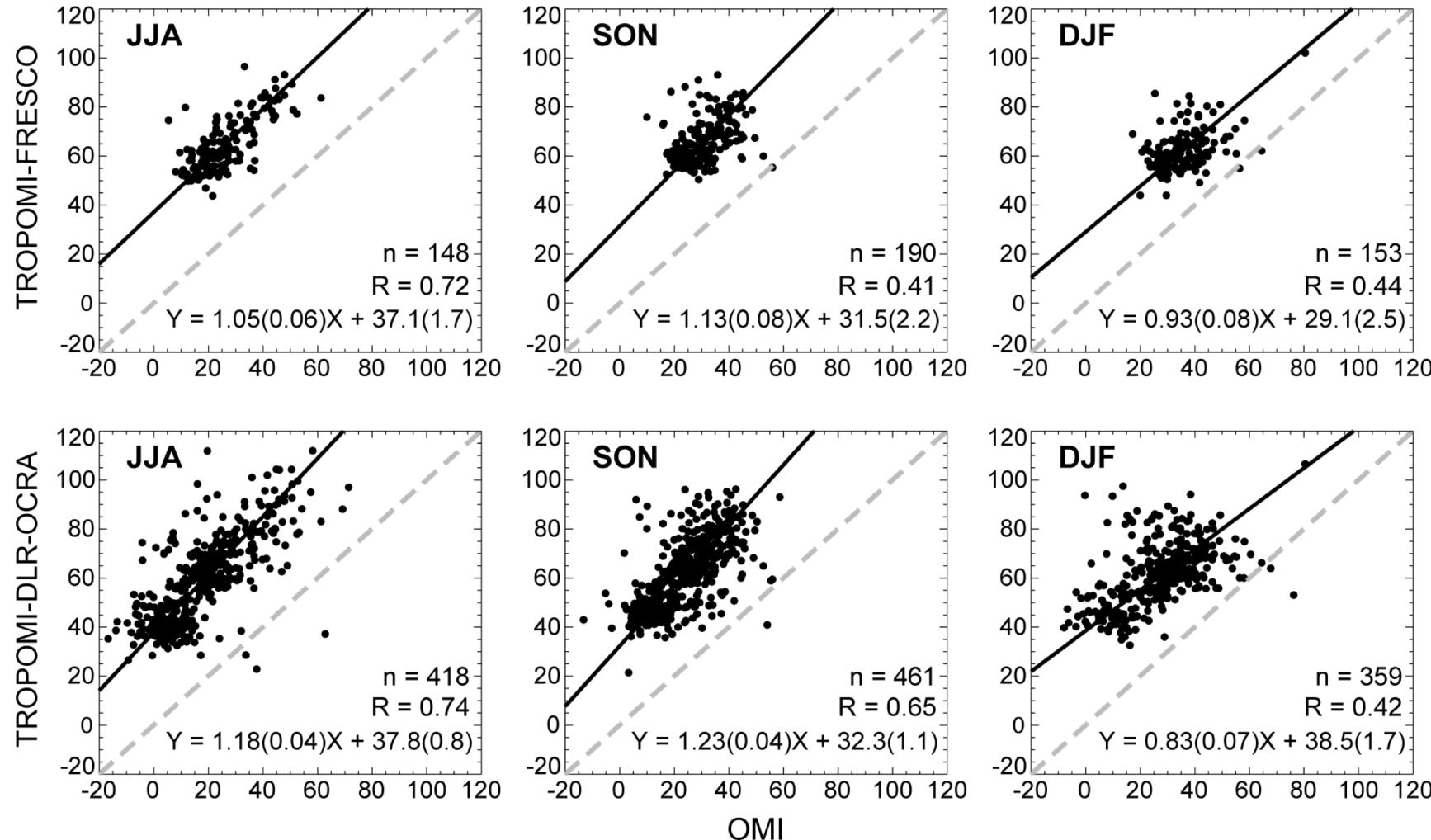
Results consistent ($R > 0.6$; slope ~ 1) for coincident scenes (mostly the tropics)

Data are
at $1^\circ \times 1^\circ$

TROPOMI and OMI UT NO₂ Comparison

TROPOMI UT NO₂ obtained at 1° × 1° and gridded to the NASA product resolution (8° × 5°)

Comparison of TROPOMI and OMI seasonal mean UT NO₂ [pptv]



Greater data density and better R with DLR-OCRA. TROPOMI bias consistent with Pandora comparison

Concluding Remarks

The cloud-sliced approach induces a small low bias in UT NO₂ variance, but this may be impacted by sampling density.

TROPOMI is biased high in the free troposphere compared to Pandora measurements. The cause is not yet apparent.

Large differences in cloud fractions and cloud top heights from different products beyond the tropics and subtropics.

Using DLR-OCRA product yields greater global coverage than using the FRESCO product.

Spatial consistency between OMI and TROPOMI UT NO₂ products

What's Next

Evaluate GEOS-Chem synthetic cloud-slicing over Europe and China

Independent assessment against reliable aircraft NO₂ observations from NASA DC8 campaigns