First Estimate of Upper Tropospheric NO_2 from TROPOMI

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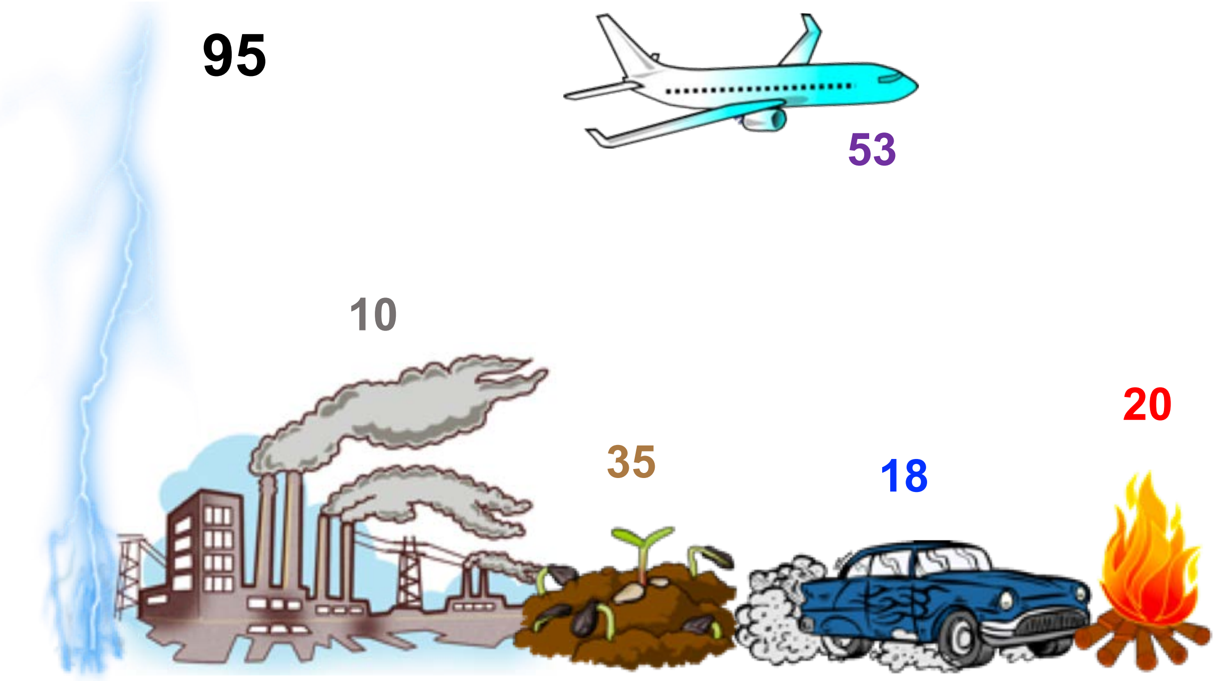
Global Relevance of the Upper Troposphere (~8-12 km)

NO_x (NO + NO_2) is very efficient at forming ozone in the upper troposphere where ozone is a potent greenhouse gas

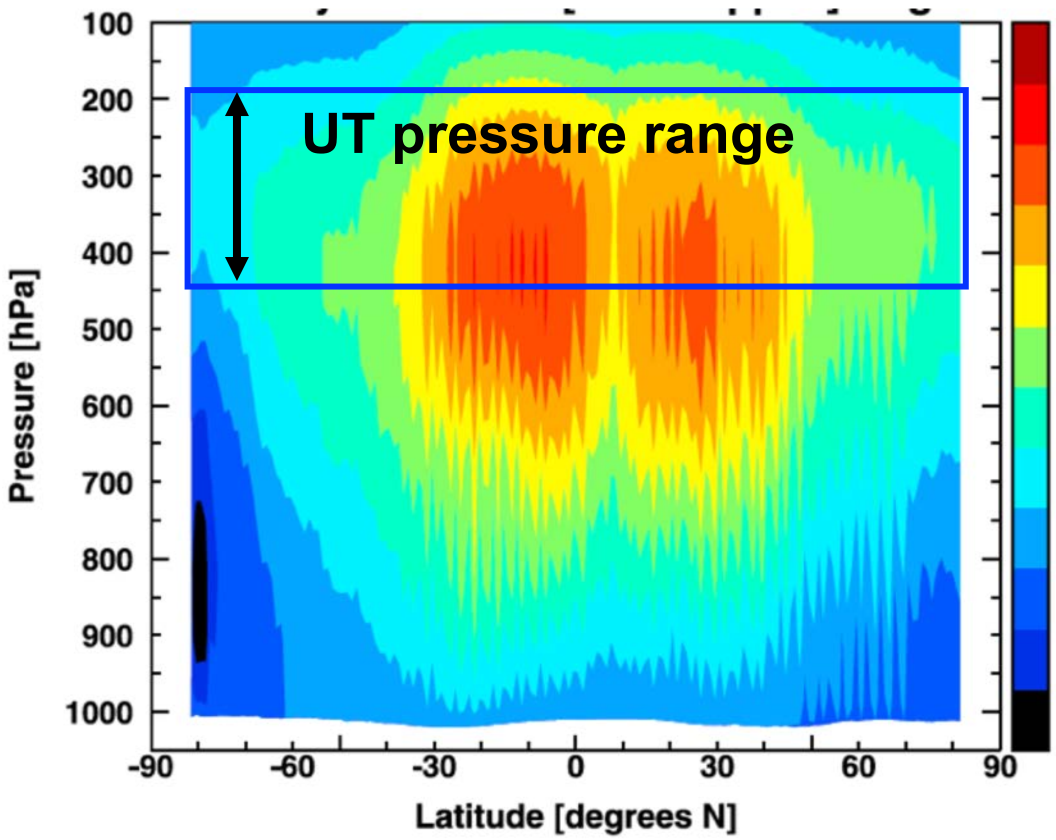
OPE (molecules O_3/molecule NO_x) for indivdual NO_x sources



[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

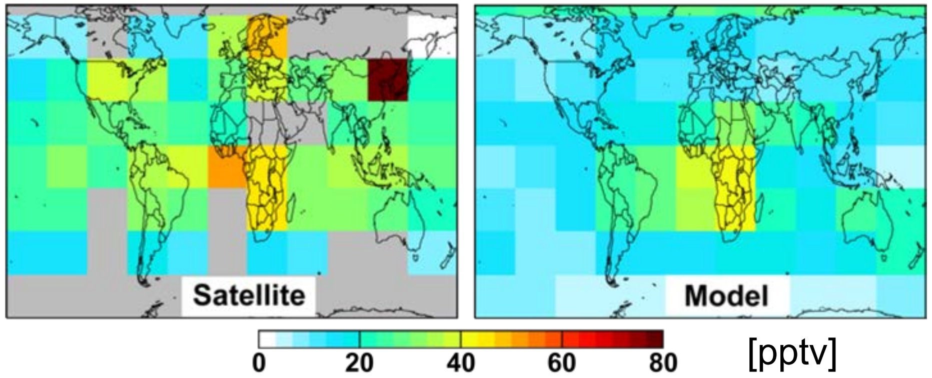


[Aghedo et al., 2011]

Errors in the UT Impact the Whole Atmosphere

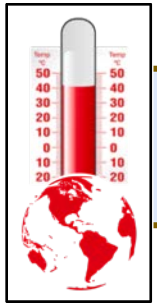
Small uncertainties in NO_x in the UT induce large climate and air quality errors

Model NO_2 too low across the UT in the northern midlatitudes



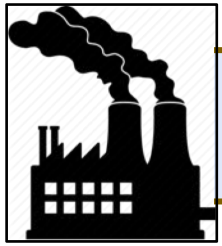
[Marais et al., 2018]

efficient ozone production



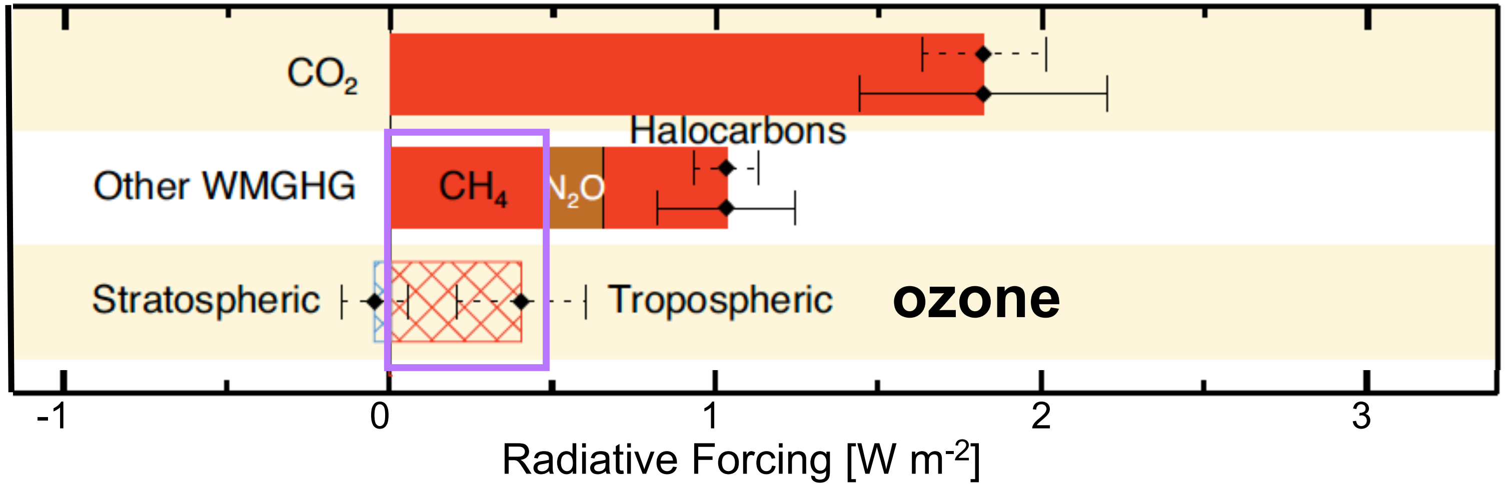
Errors in tropospheric ozone radiative forcing

satellite NO_2 retrievals



Uncertain global air quality constraints from satellites

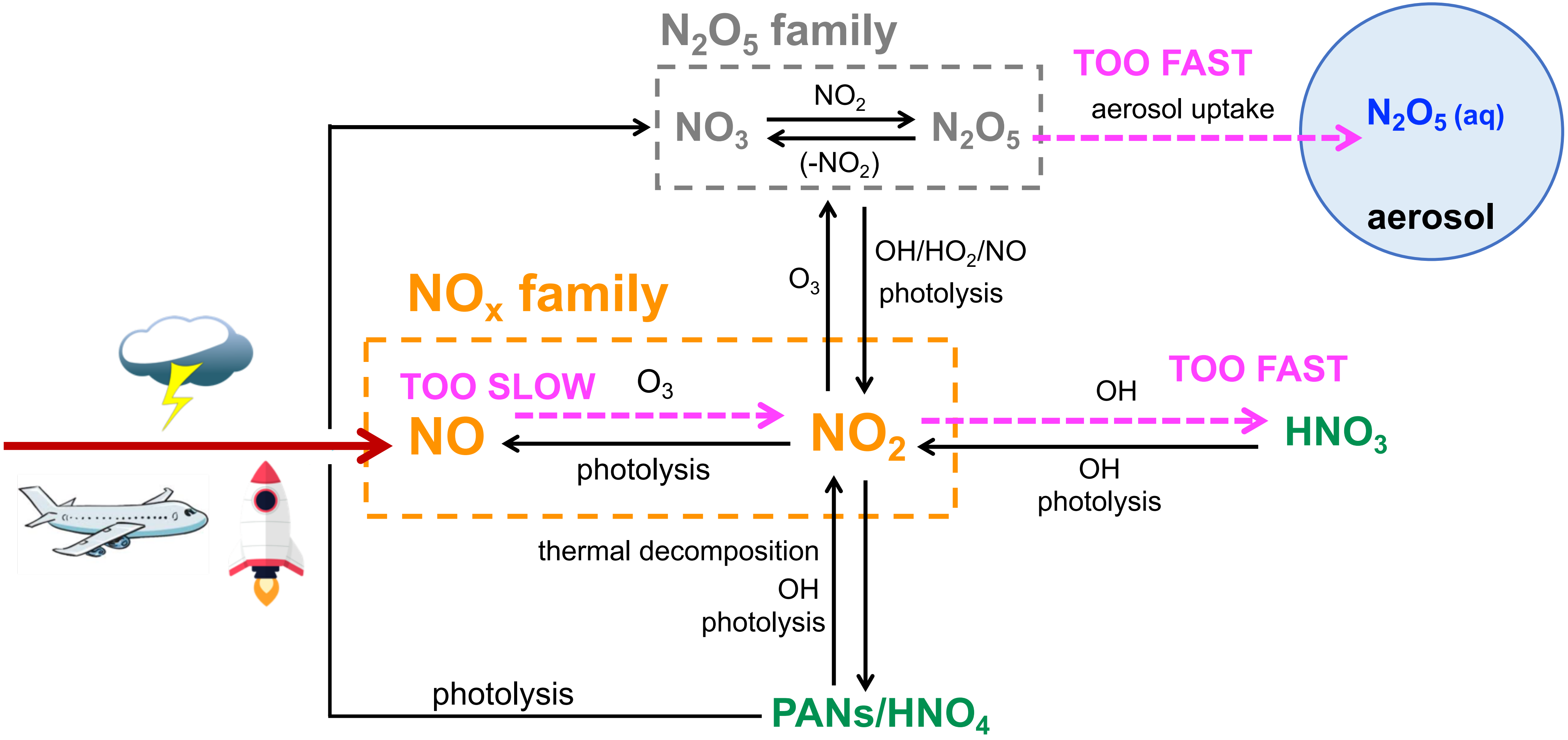
Tropospheric ozone and methane have near-equal climate impacts



[IPCC AR5, 2013]

Large Uncertainties in Chemistry of Reactive Nitrogen

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

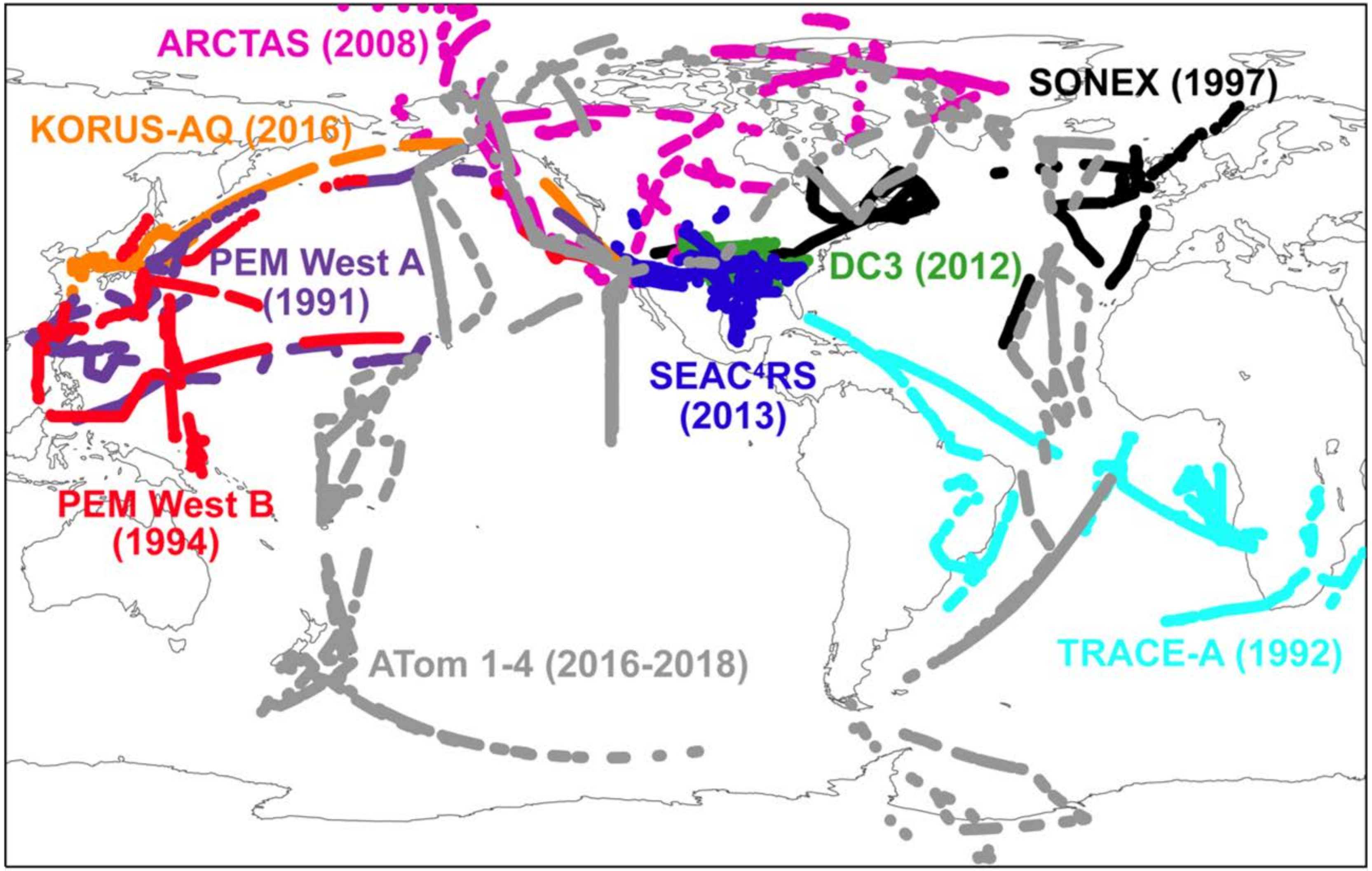


Uncertain reactions

Local sources (rockets contribution unknown)

NO_x Measurements are Limited in Time and Space

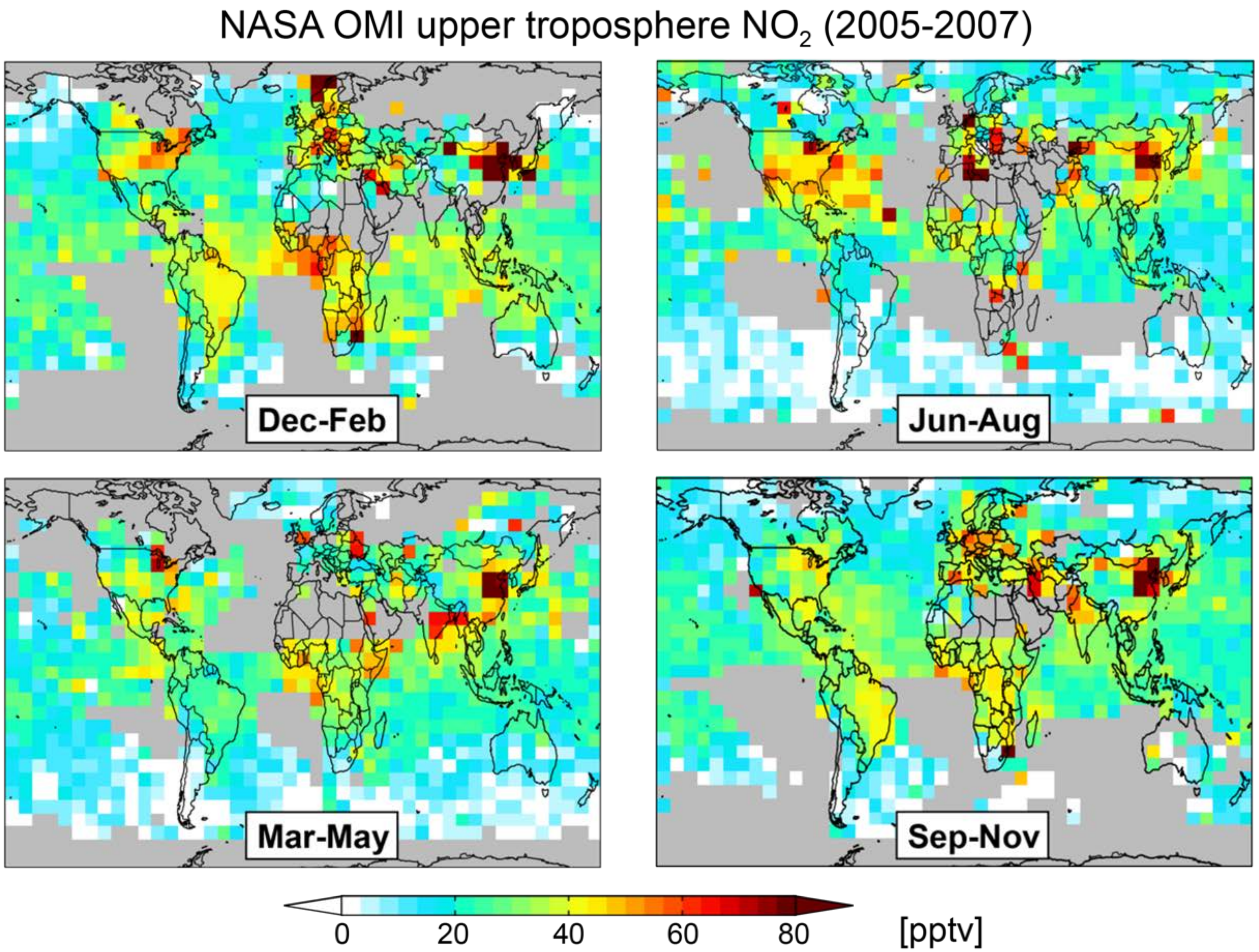
Spatial distribution of data from NASA DC8 aircraft campaigns in the upper troposphere



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

New UT NO_2 products from the Ozone Monitoring Instrument (OMI)

Near global spatial coverage of seasonal mean UT NO_2 at 5° × 8° (latitude × longitude)



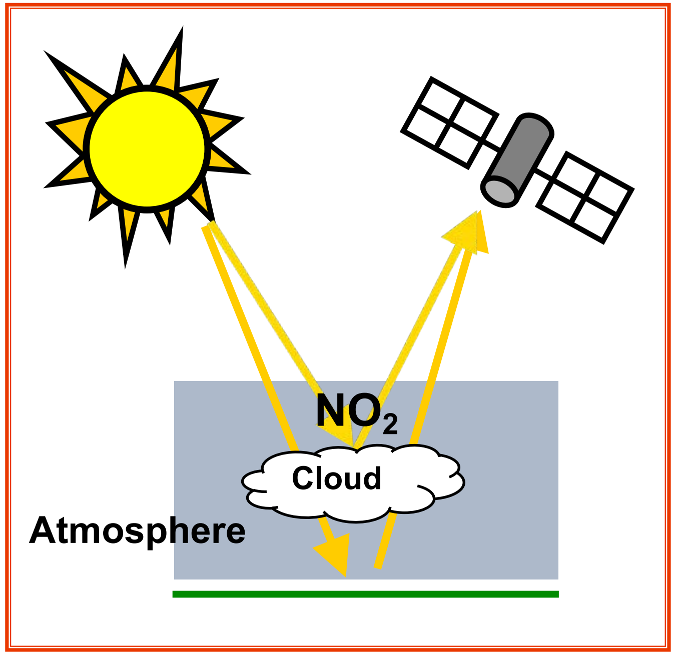
[Marais et al., 2018; Choi et al., 2014]

UT NO_2 Obtained with the Innovative Cloud-Slicing Technique

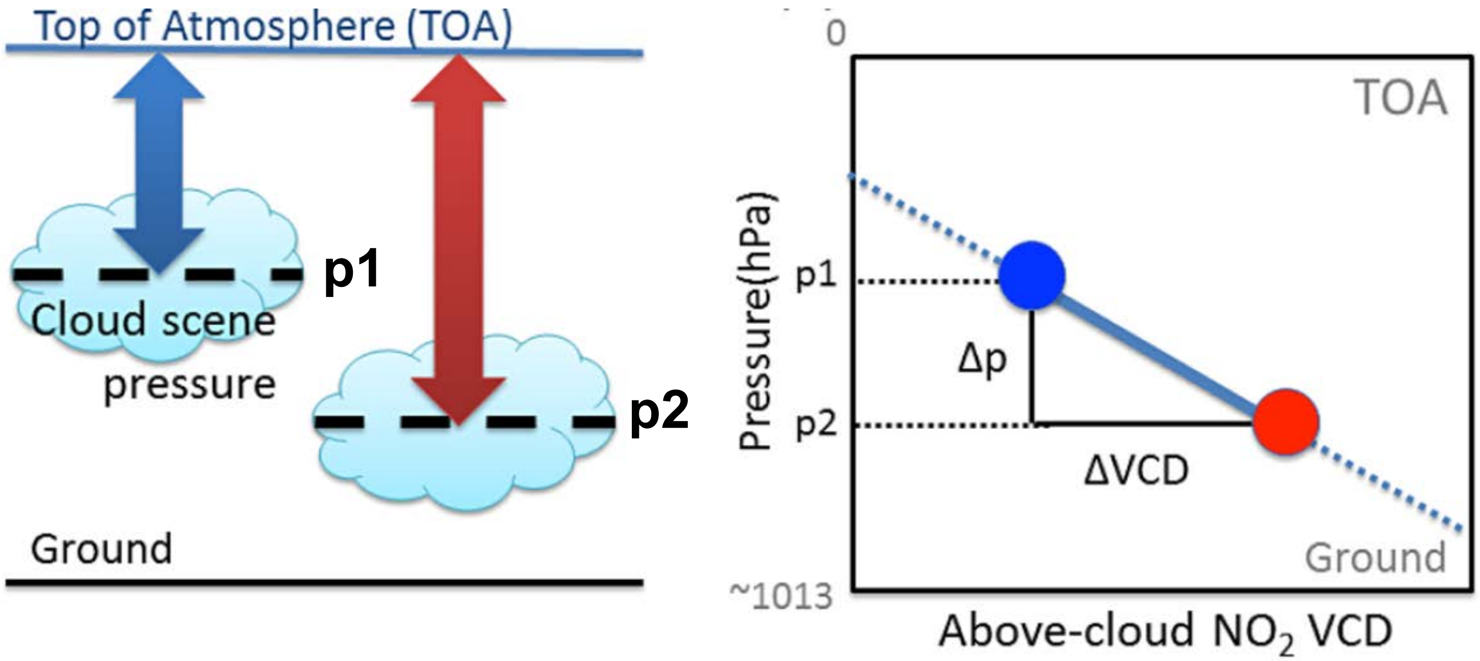
First applied by Ziemke et al. [2001] to TOMS ozone

Retrieve partial NO_2 columns over cloudy scenes at different heights

APPROACH



Use cloud height variability to derive partial columns



[adapted from Choi et al., 2014]

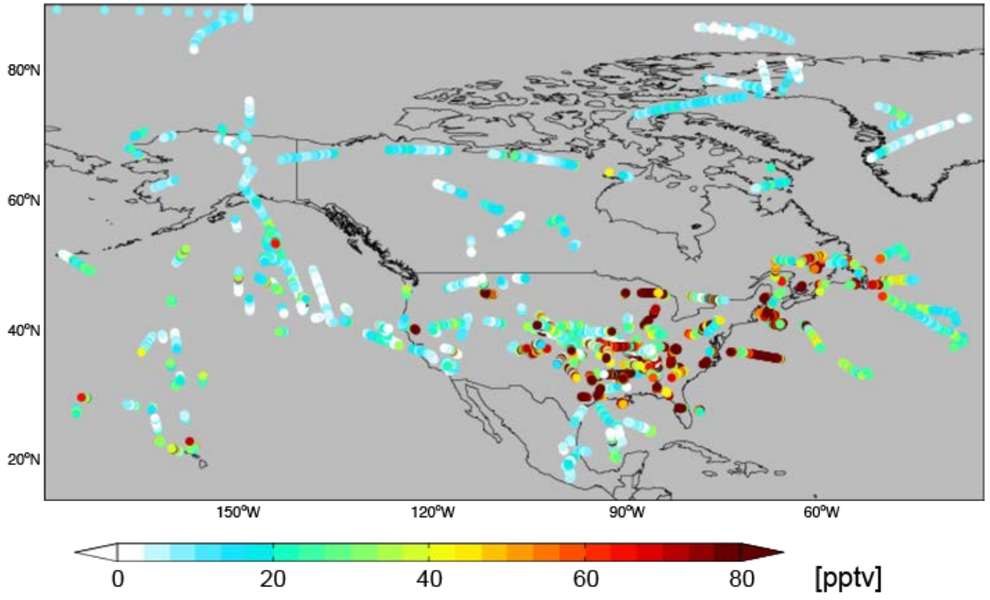
NO_2 volume mixing ratio (VMR) between clouds at p1 and p2

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

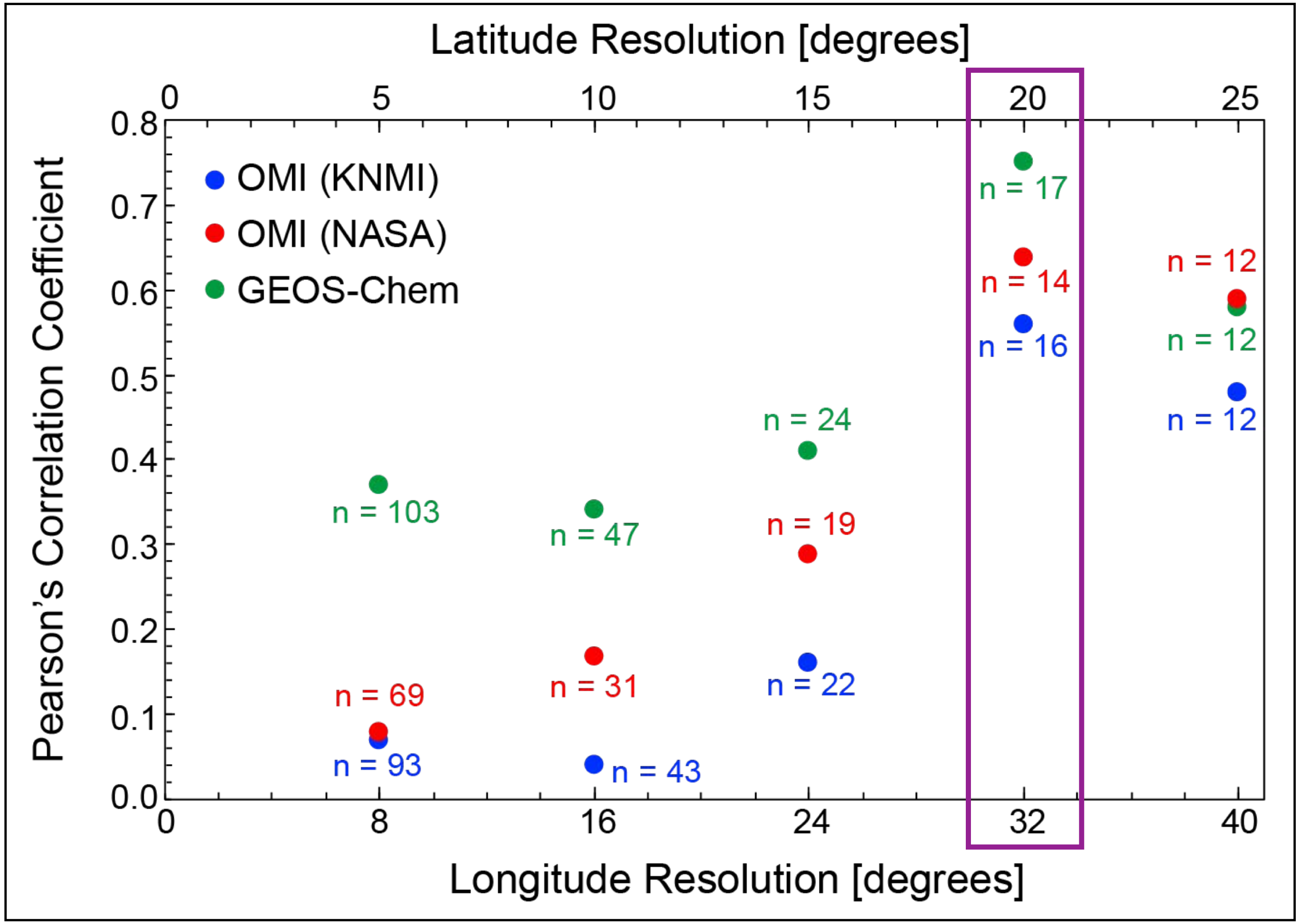
Consistency with Aircraft Observations of NO_2

Agreement in spatial distribution between (reliable) aircraft and OMI UT NO_2 observations, but at coarse scales (seasonal, 20° latitude (2000 km) × 32° longitude (3200 km))

NASA DC8 NO_2 in Spring-Summer



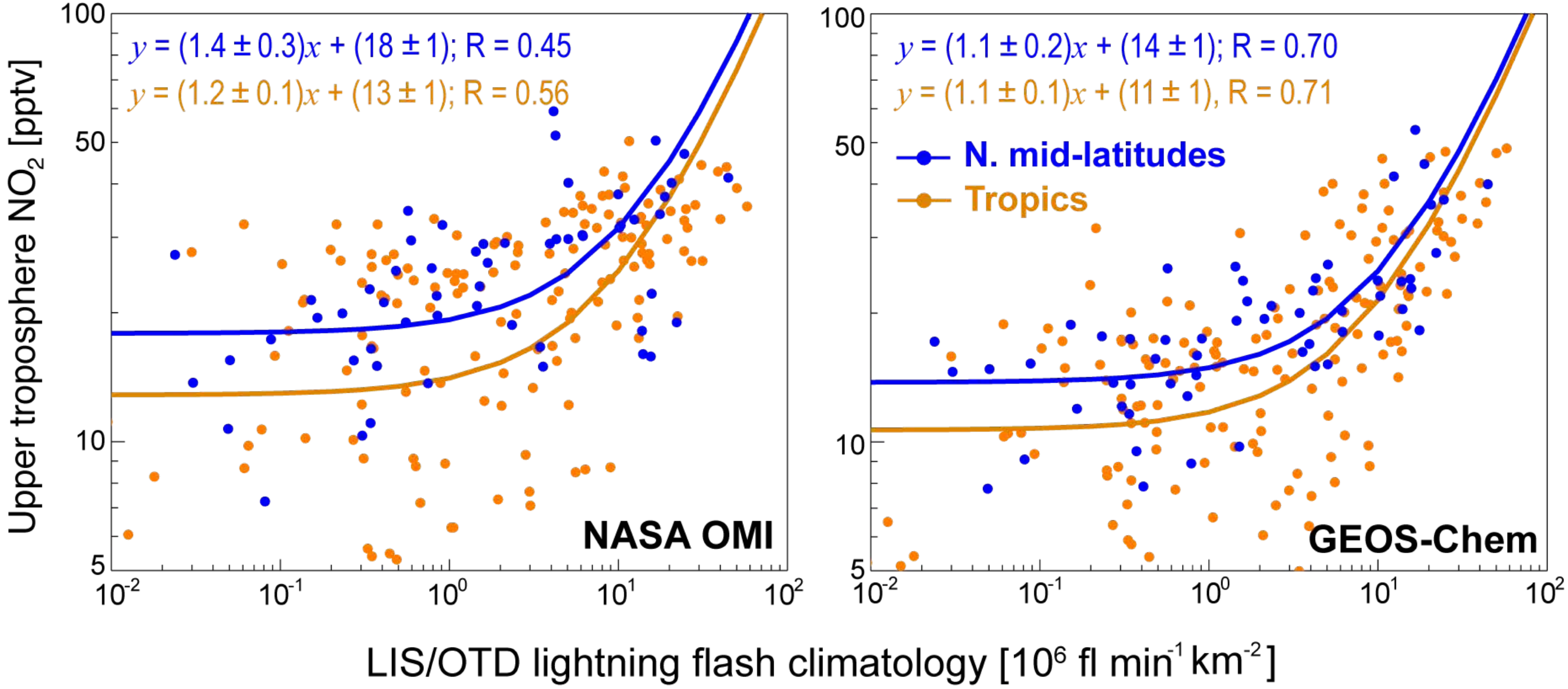
NO_2 obtained for multiple campaigns from Ron Cohen's TD-LIF instrument



[Marais et al., 2018]

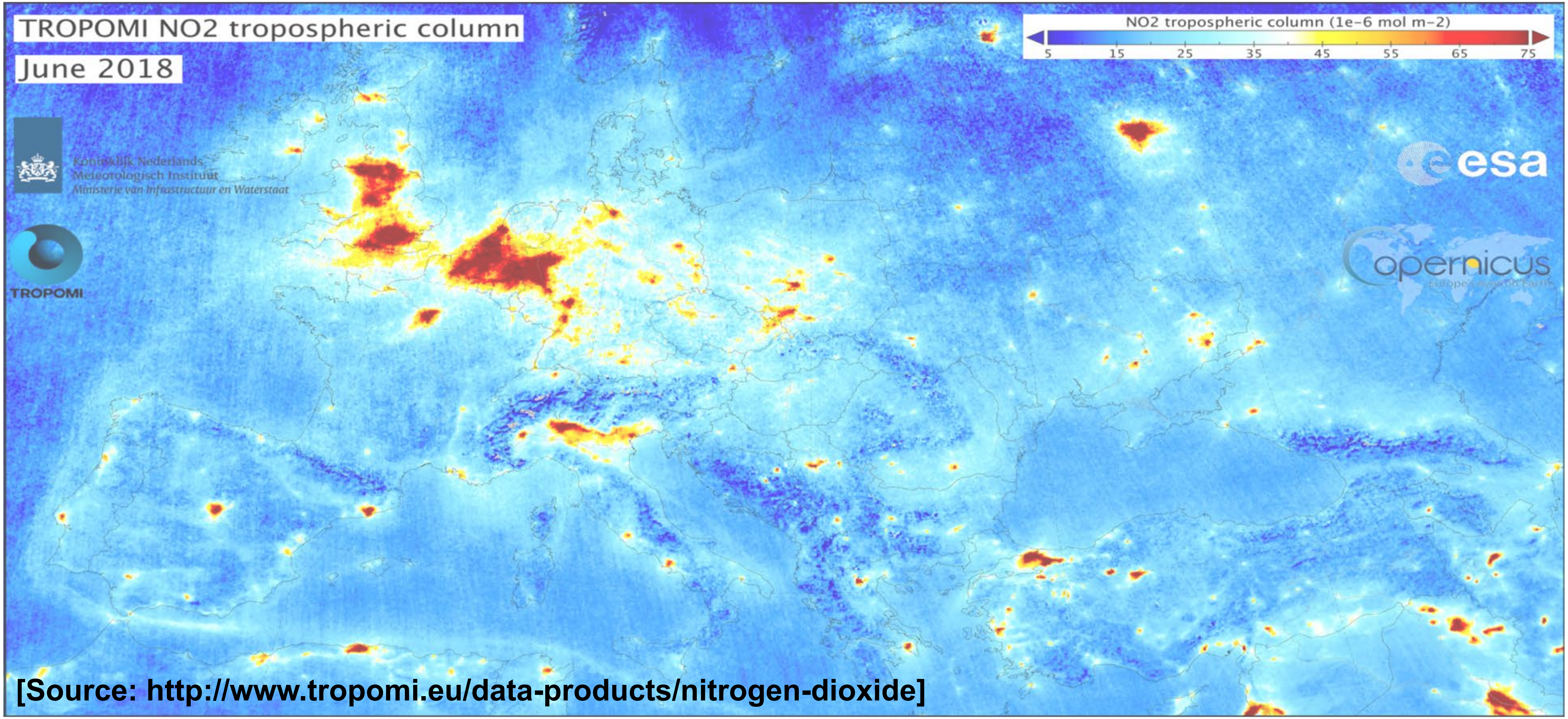
Used OMI UT NO_2 to Better Constraint Global Lightning NO_x

Log-log relationship between UT NO_2 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?

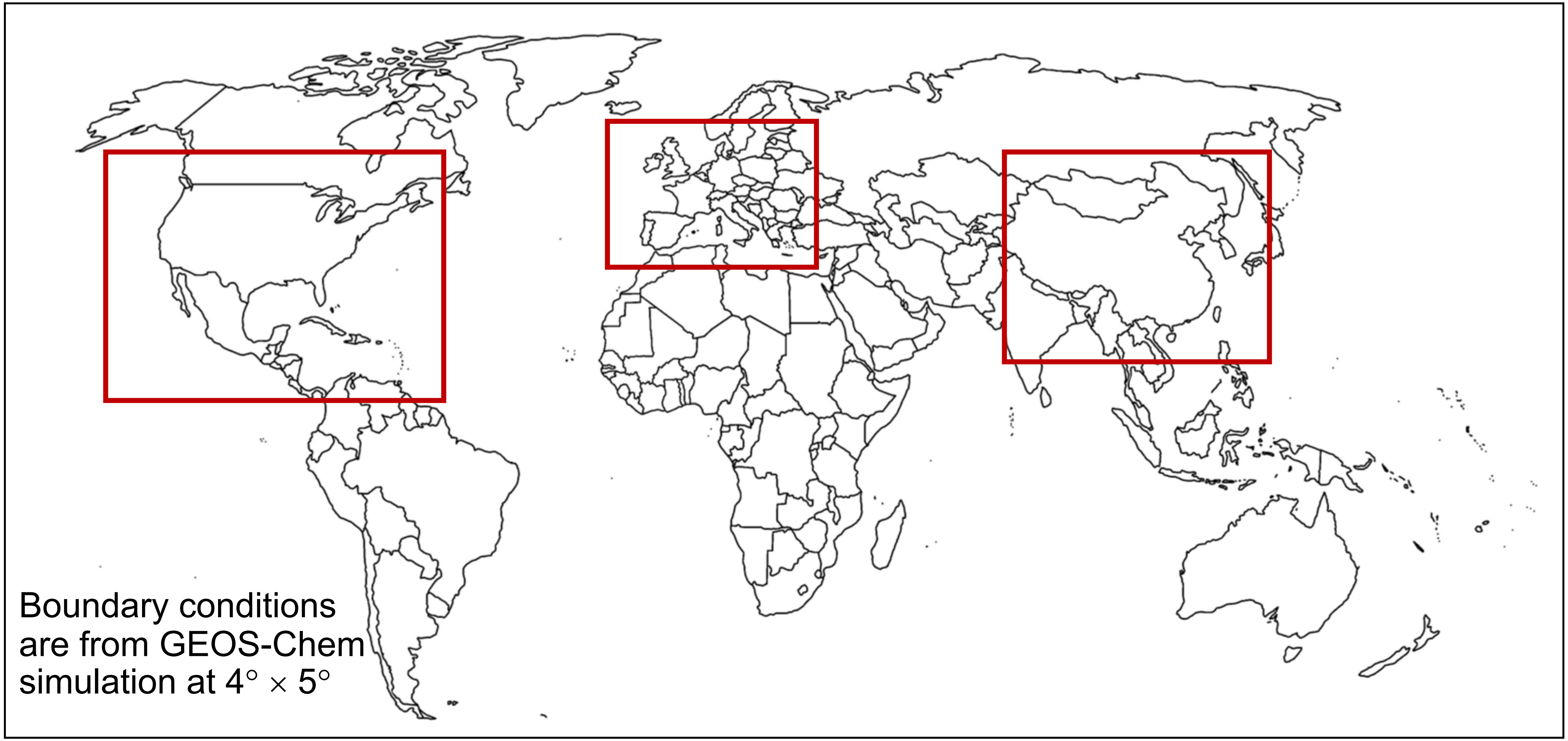


Nadir spatial resolutions in km (along × across): **13 × 24** (**OMI**); **5.6 × 3.5** (**TROPOMI**)

Synthetic Cloud-Slicing Experiment with GEOS-Chem

Determine whether UT NO_2 obtained with cloud-slicing reproduces UT NO_2 obtained by averaging across the UT (450-180 hPa)

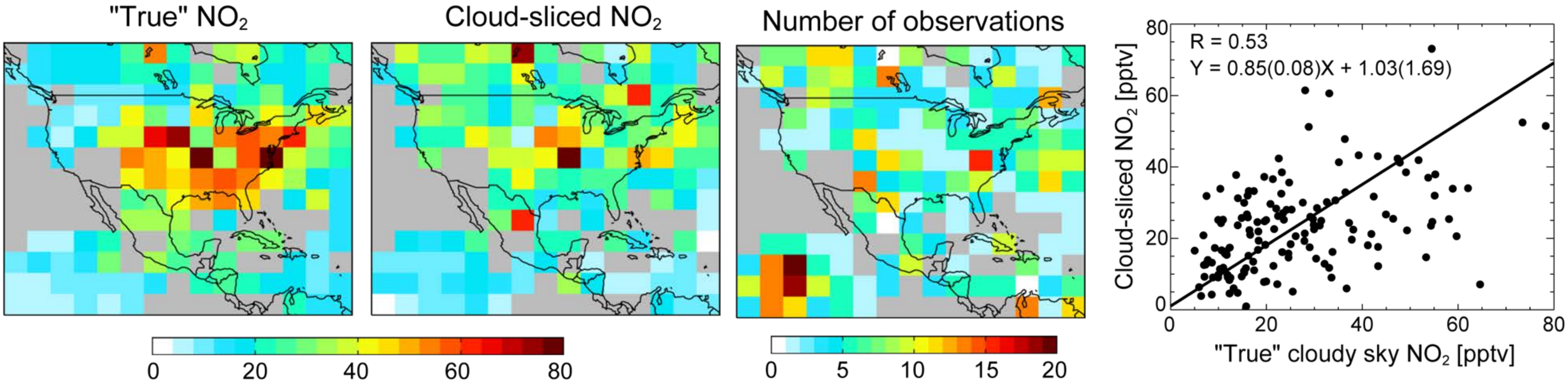
GEOS-Chem nested domains at $0.25^\circ \times 0.3125^\circ$ driven with NASA GEOS-FP meteorology



Synthetic Cloud-Slicing Experiment with GEOS-Chem

Compare “true” and cloud-sliced UT NO_2 from GEOS-Chem over North America in June-August

Daily cloud-sliced NO_2 obtained at 4° × 5° from daily nested grid GEOS-Chem partial columns sampled around the satellite overpass



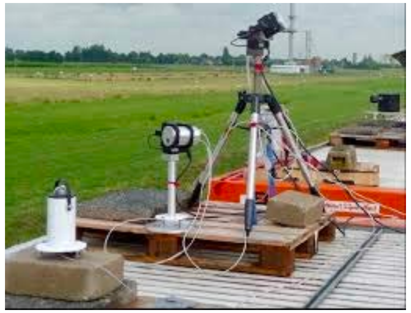
Sampling conditions informed by Choi et al. [2014] approach:

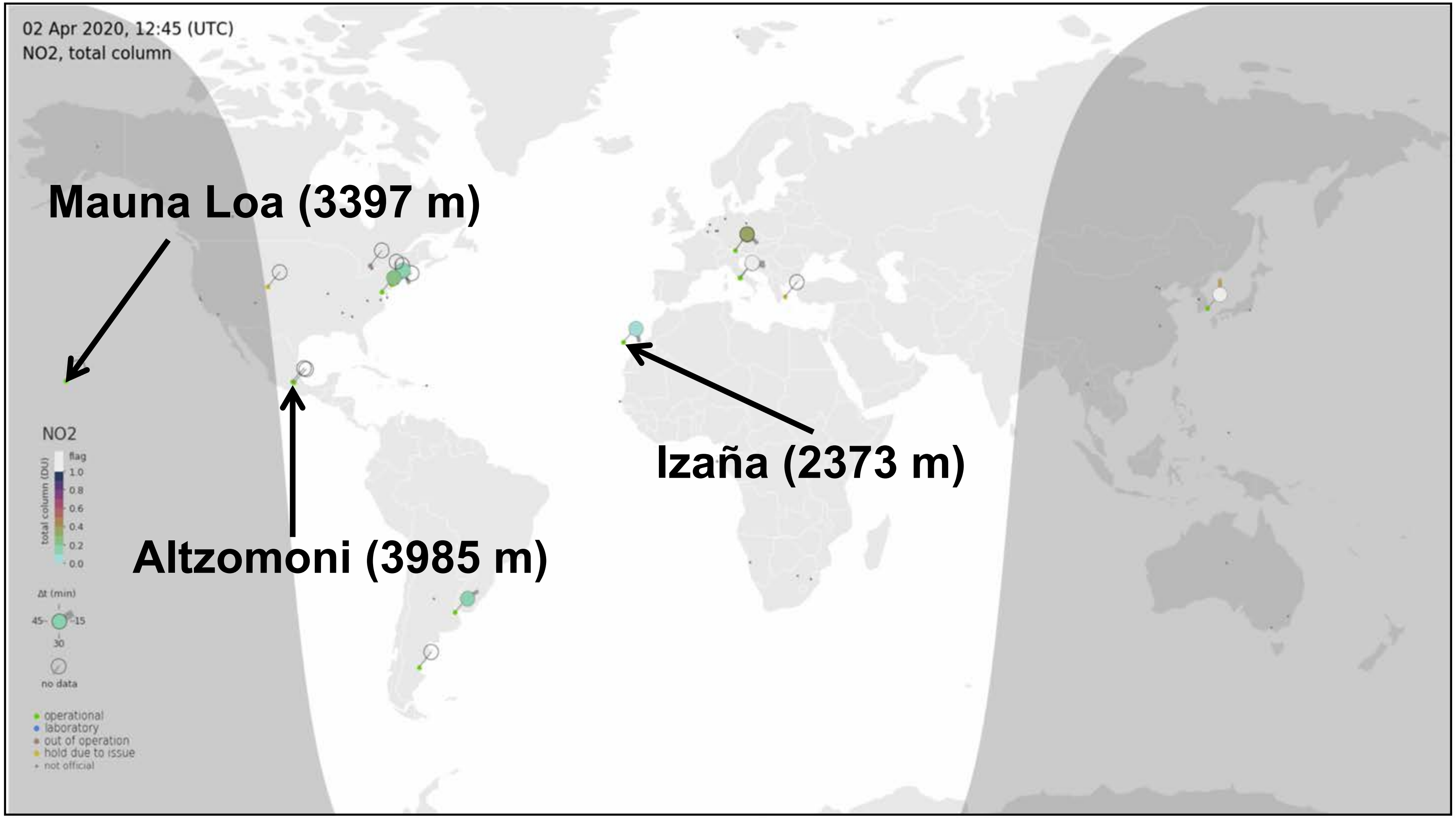
Satellite overpass (12h00-15h00), dynamic cloud top height range (standard deviation > 35 hPa, range > 160 hPa), and UT NO_2 with relative error < 100%, < 200 pptv, significantly greater than zero.

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

Evaluate TROPOMI total column NO_2 against Pandora

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

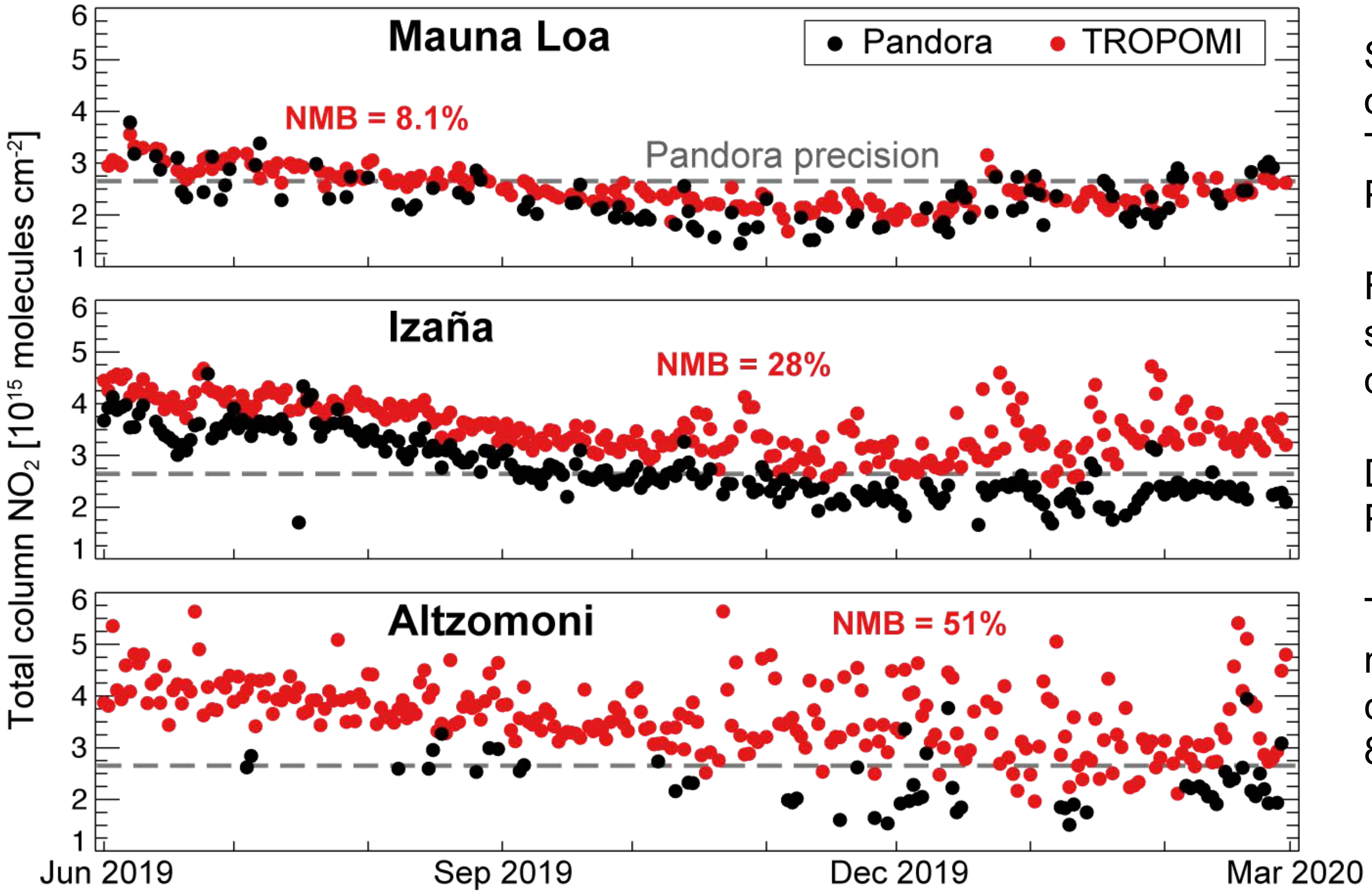




[Source: https://www.pandonia-global-network.org/]

Evaluate TROPOMI total column NO_2 against Pandora

Pandora and TROPOMI total column NO_2 time series at the 3 measurement stations



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

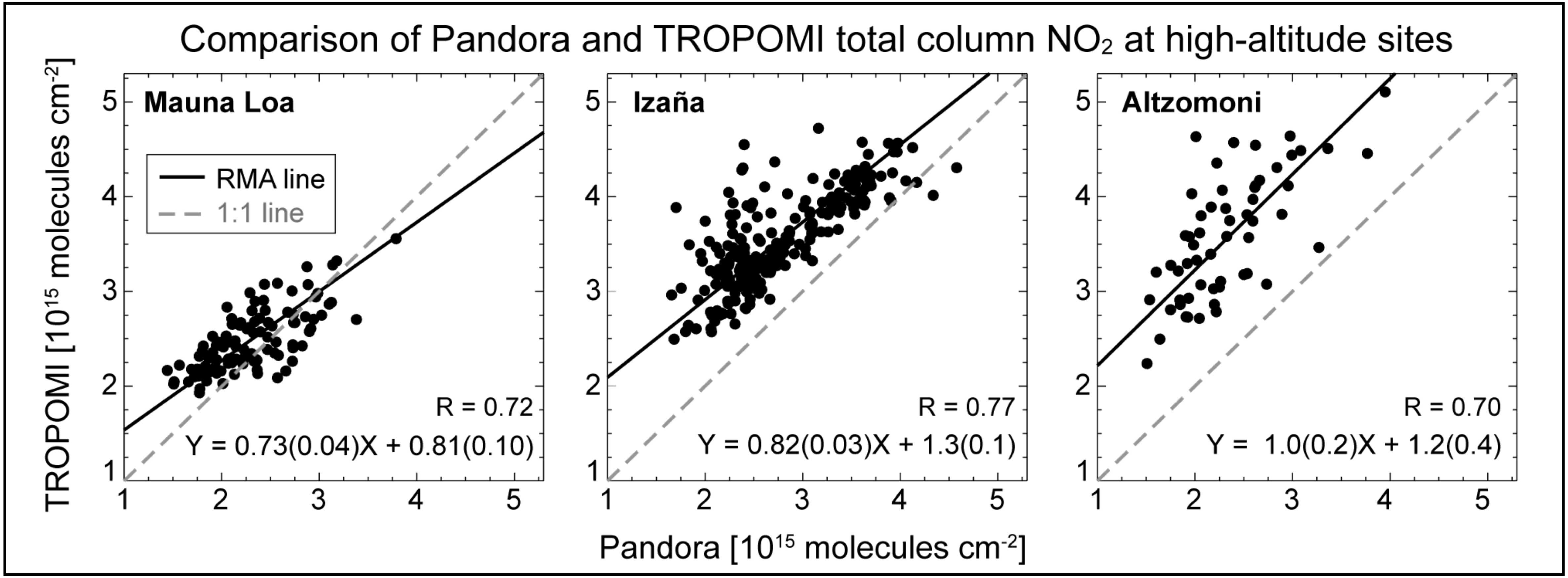
Focus is on cloudy scenes, so use a TROPOMI qa_value threshold of 0.45

Data close to estimated Pandora precision

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

Evaluate TROPOMI total column NO_2 against Pandora

Scatterplots of TROPOMI versus Pandora total column NO_2 at the 3 stations

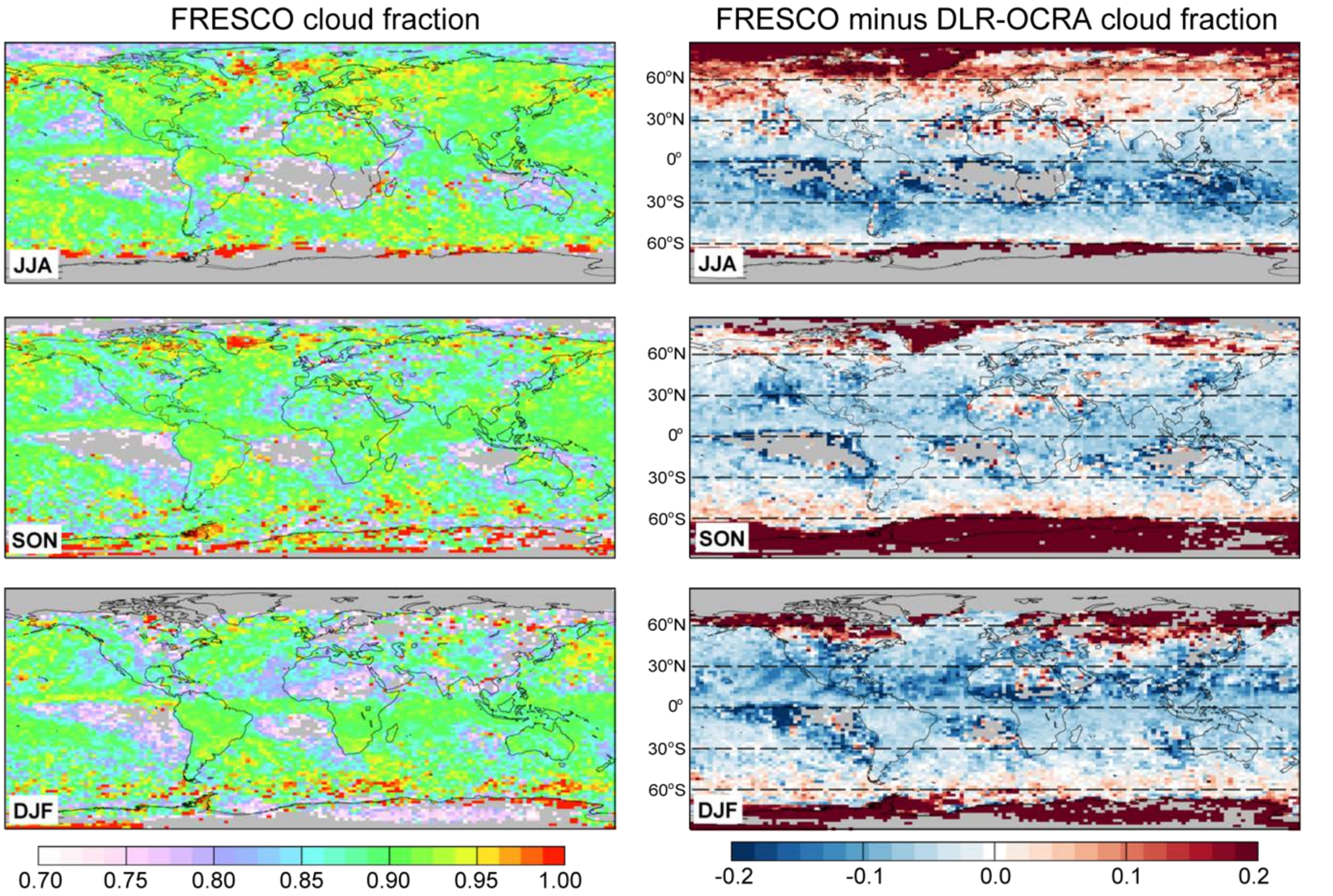


Positive intercept supports systematic bias in TROPOMI total column

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



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

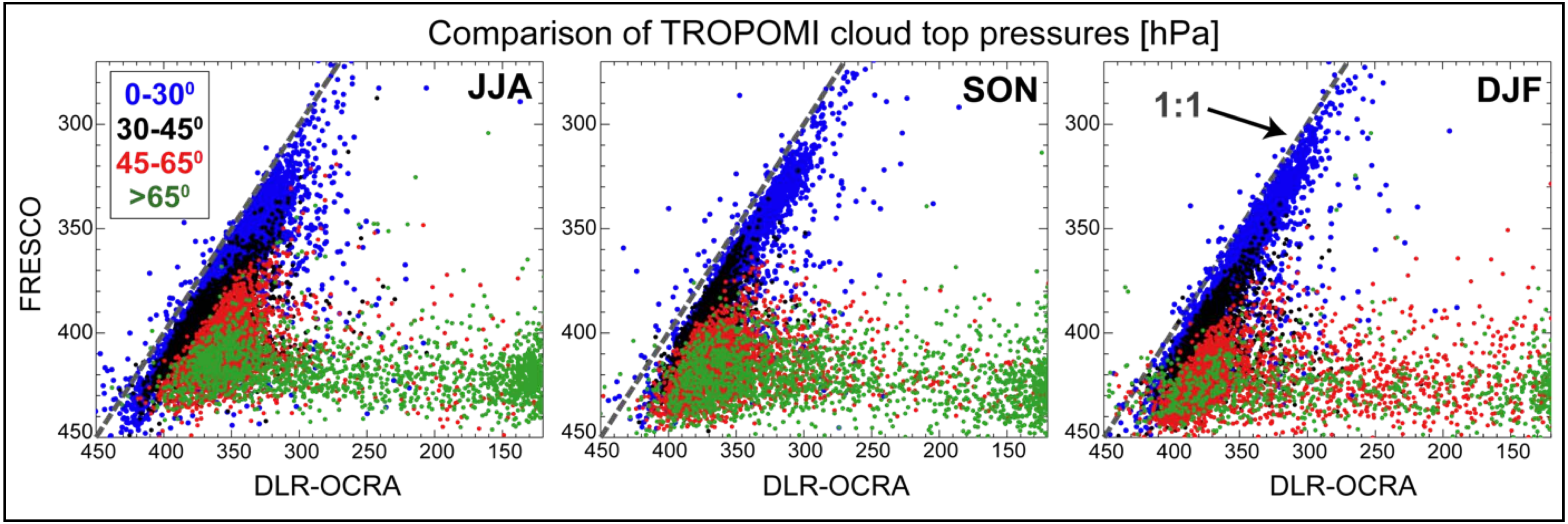
FRESCO: cloud product used in the TROPOMI NO_2 column retrieval

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° × 2.5° colored by latitude bands



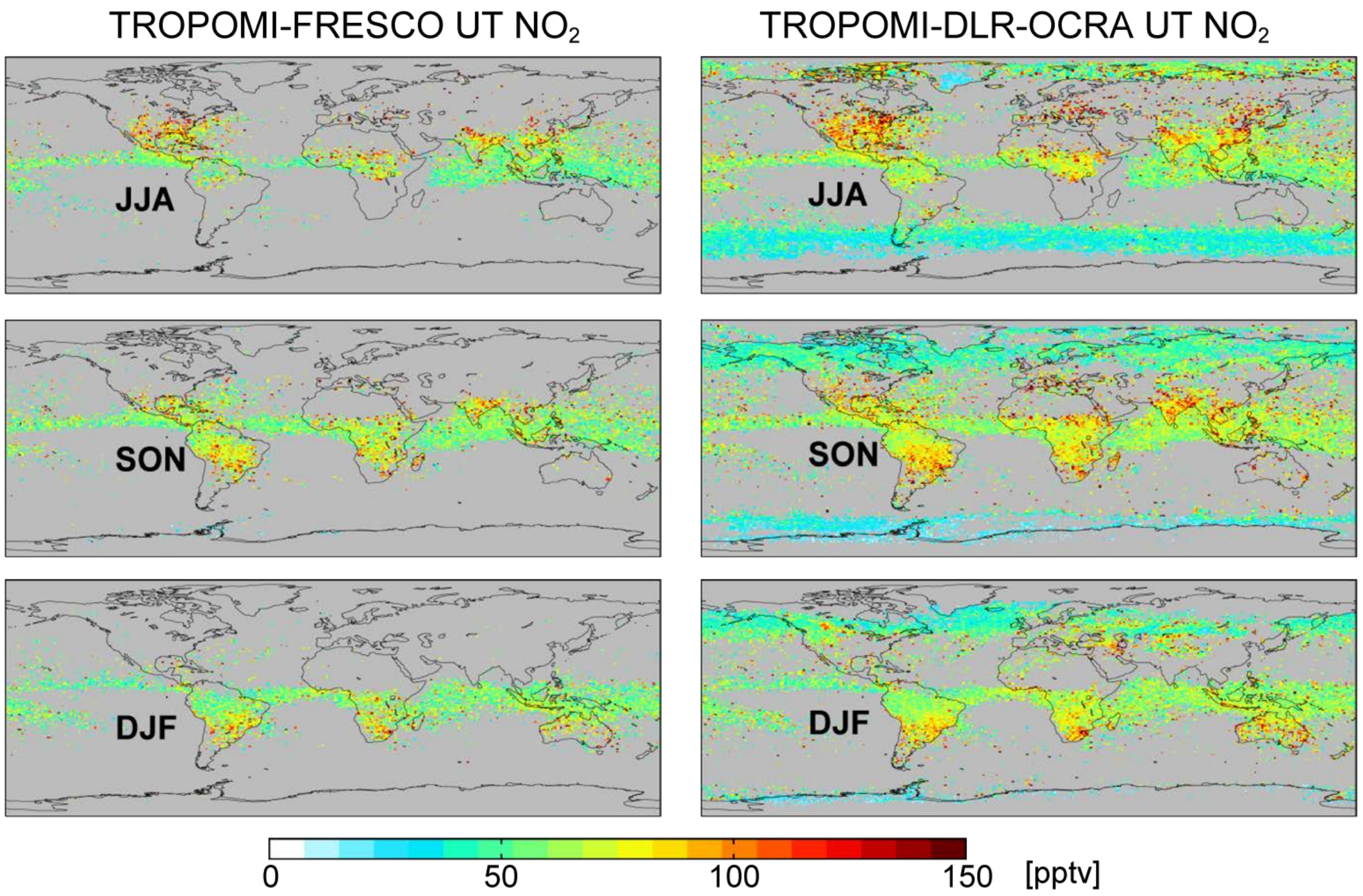
Compare scenes with FRESCO cloud top pressure range of 180 to 450 hPa

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

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

Seasonal Mean TROPOMI Cloud-sliced UT NO_2

Data are at 1° × 1°

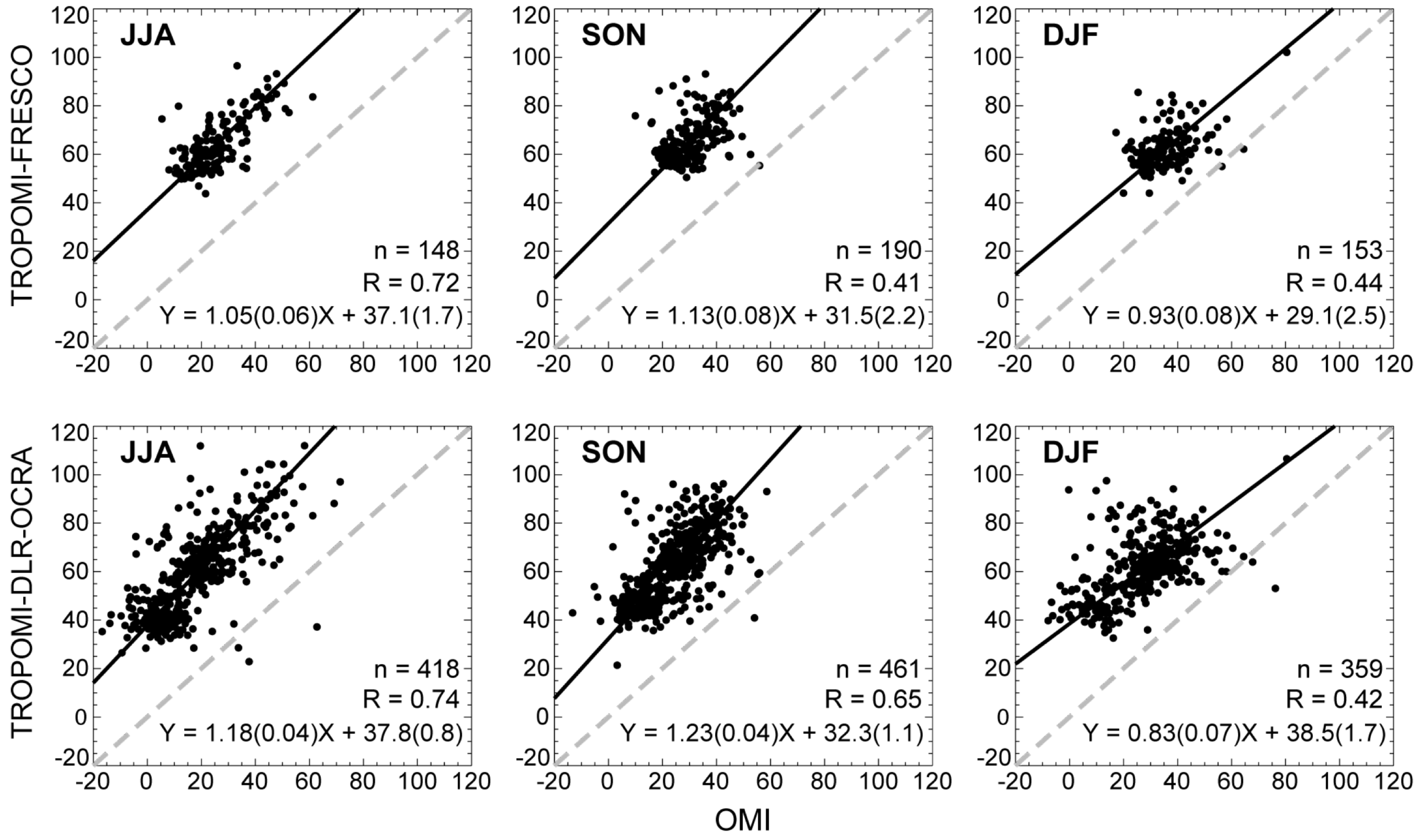


Results consistent ($R > 0.6$; slope ~1) for coincident scenes (mostly the tropics)

TROPOMI and OMI UT NO_2 Comparison

TROPOMI UT NO_2 obtained at 1° × 1° and gridded to the NASA product resolution (8° × 5°)

Comparison of TROPOMI and OMI seasonal mean UT NO_2 [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 NO_2 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 NO_2 products

What's Next

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

Independent assessment against reliable aircraft NO_2 observations from NASA DC8 campaigns