First Estimate of NO_2 in the upper troposphere from TROPOMI

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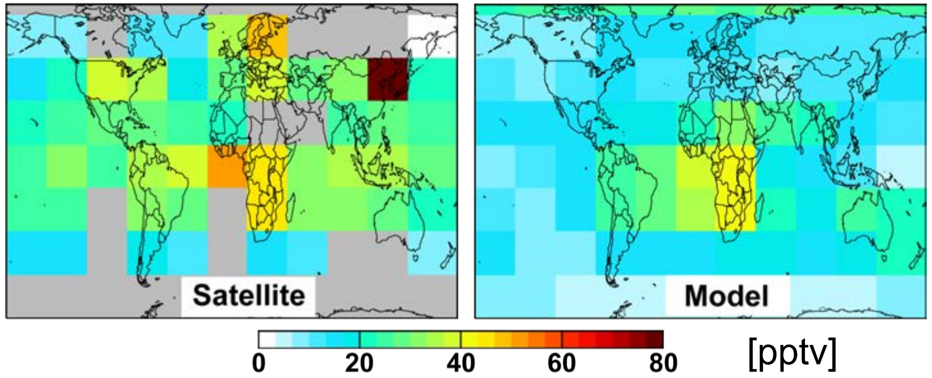
The Impact of Errors in the Upper Troposphere are Widespread

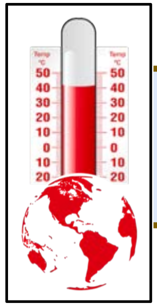
Small uncertainties in upper tropospheric NO_x induce large climate and air quality errors

Model NO_2 too low across the northern midlatitudes upper troposphere



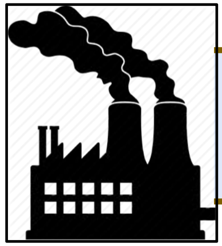
[Marais et al., 2018]

errors in ozone production



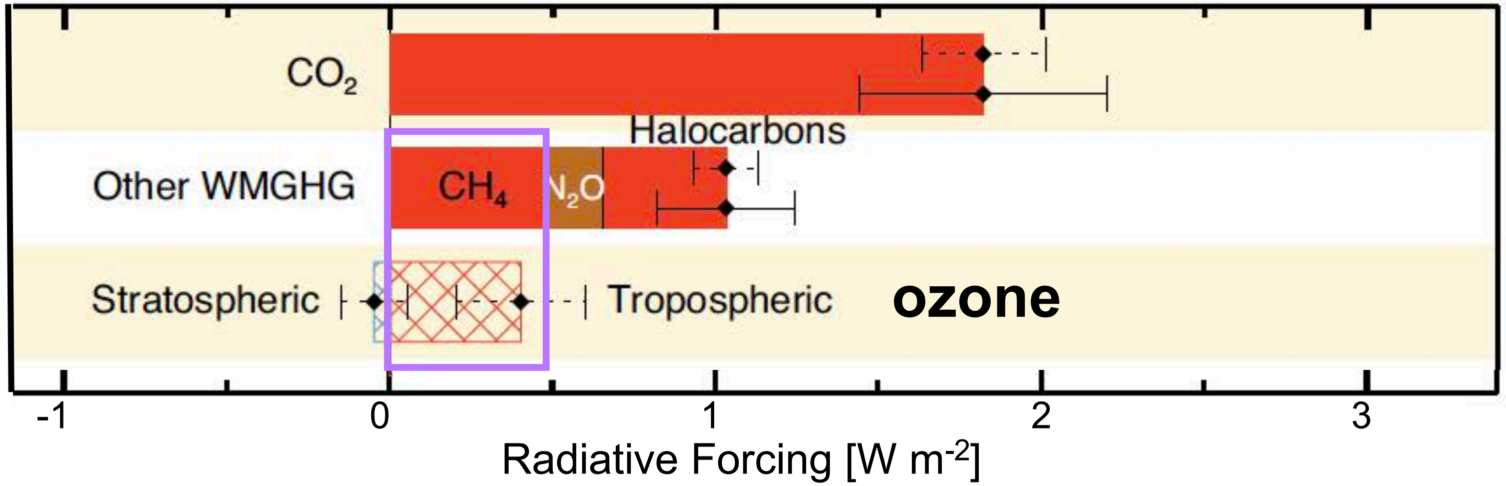
Errors in tropospheric ozone radiative forcing

errors in satellite NO_2 retrievals



Uncertain global air quality constraints from satellites

Ozone is a powerful greenhouse gas (particularly in the upper troposphere)



Near-equivalent warming due to ozone and methane

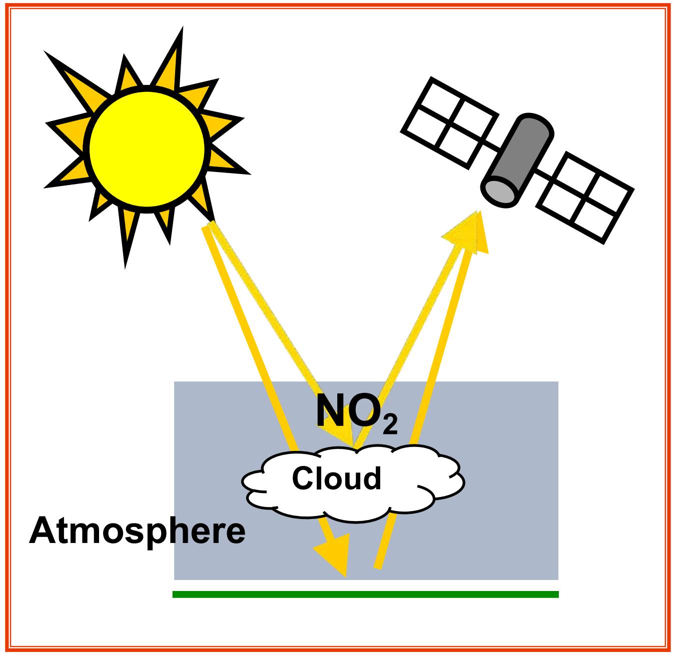
[IPCC AR5, 2013]

Obtain NO_2 in the Upper Troposphere (UT) with Cloud-Slicing

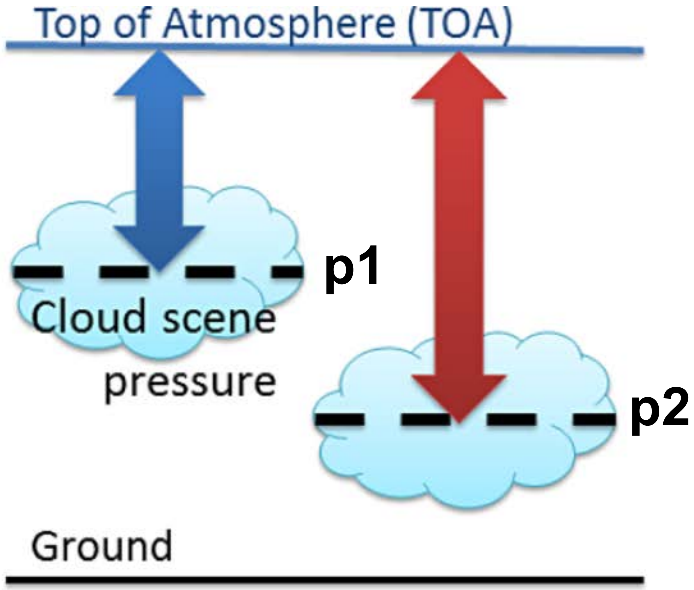
First applied by Ziemke et al. [2001] to TOMS ozone

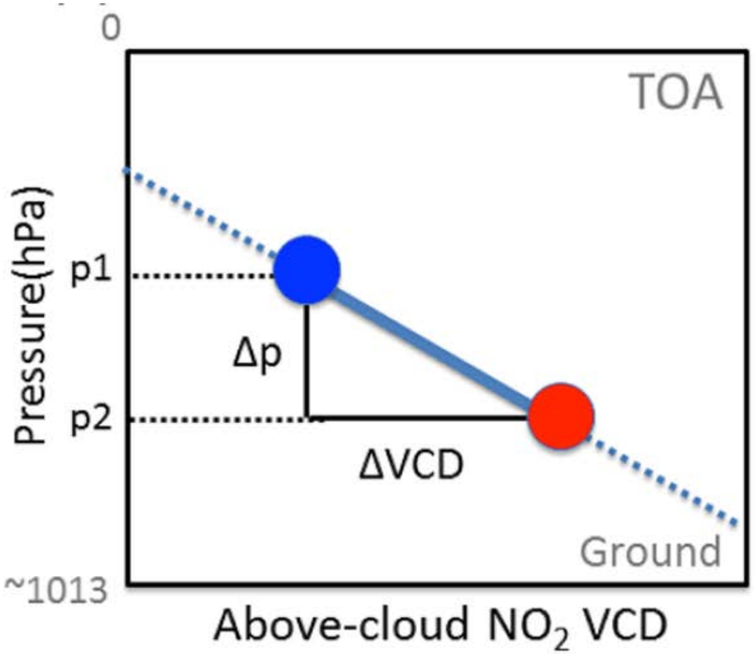
Apply to partial NO_2 columns over optically thick clouds at different altitudes from 450 to 180 hPa for June 2019 to February 2020

Partial columns above thick clouds



Use cloud height variability to derive partial columns





[adapted from Choi et al., 2014]

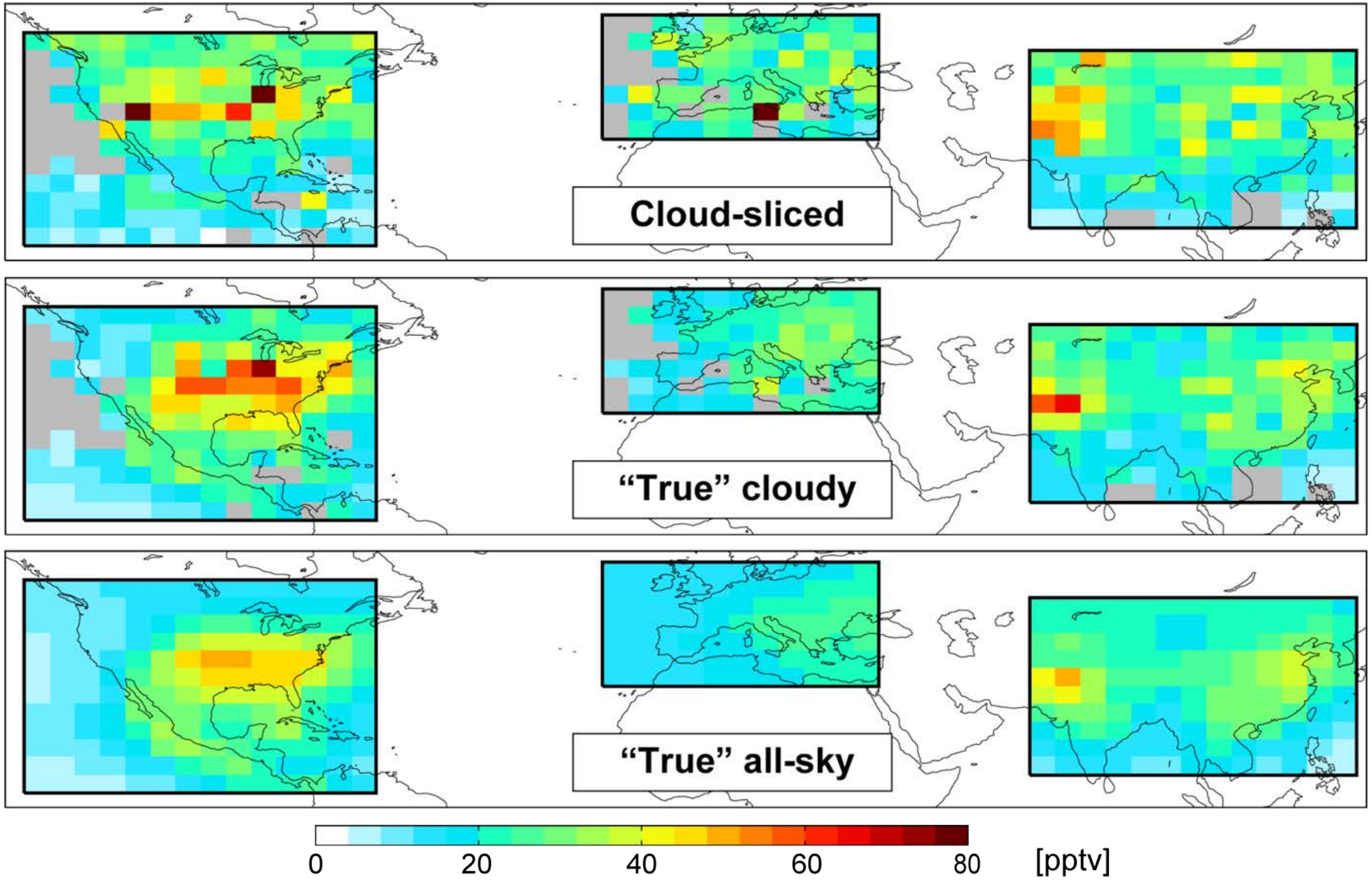
NO_2 volume mixing ratio (VMR) between clouds at p1 and p2

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

Synthetic Cloud-Slicing Experiment with GEOS-Chem

Test that cloud-sliced NO_2 reproduces the "truth" with synthetic data from GEOS-Chem

Upper tropospheric (180-450 hPa) NO_2 obtained with GEOS-Chem



Use GEOS-Chem output at $0.5° \times 0.3125°$ to obtain UT NO_2 at $4° \times 5°$.

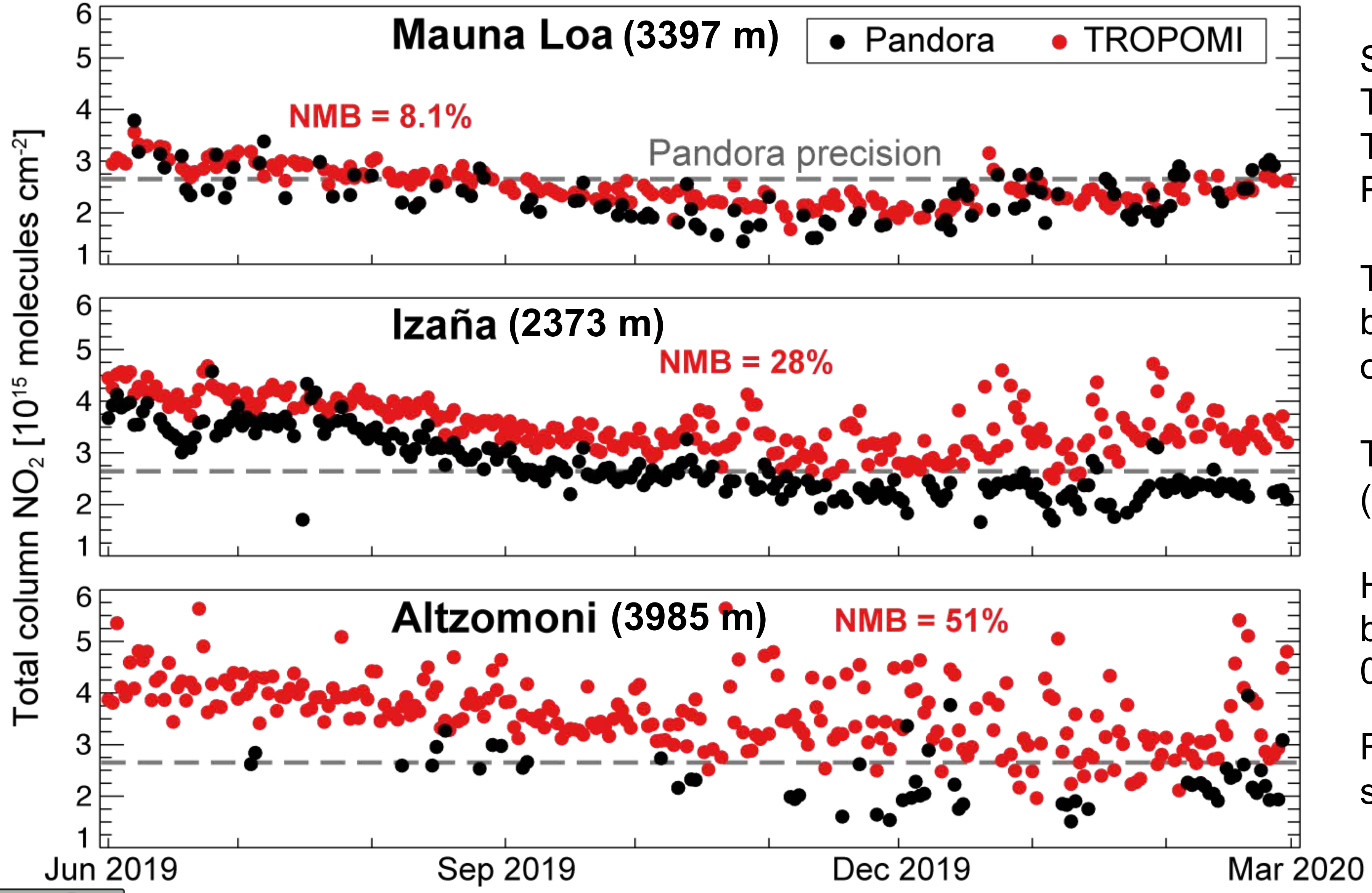
Cloud-sliced and "true" cloudy scene UT NO_2 are spatially consistent (R = 0.66).

Regression slope for cloud-sliced versus "true" cloudy UT NO_2 is 0.92 ± 0.05 and the intercept is 2.3 ± 1.1 pptv.

Targeting cloudy conditions leads to 10% higher UT NO_2 than all-sky conditions.

Evaluate TROPOMI against Pandora at High-Altitude Sites

Total column NO_2 time series at 3 Pandora long-term measurement stations



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

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

Temporal correlation at all sites ($R > 0.7$)

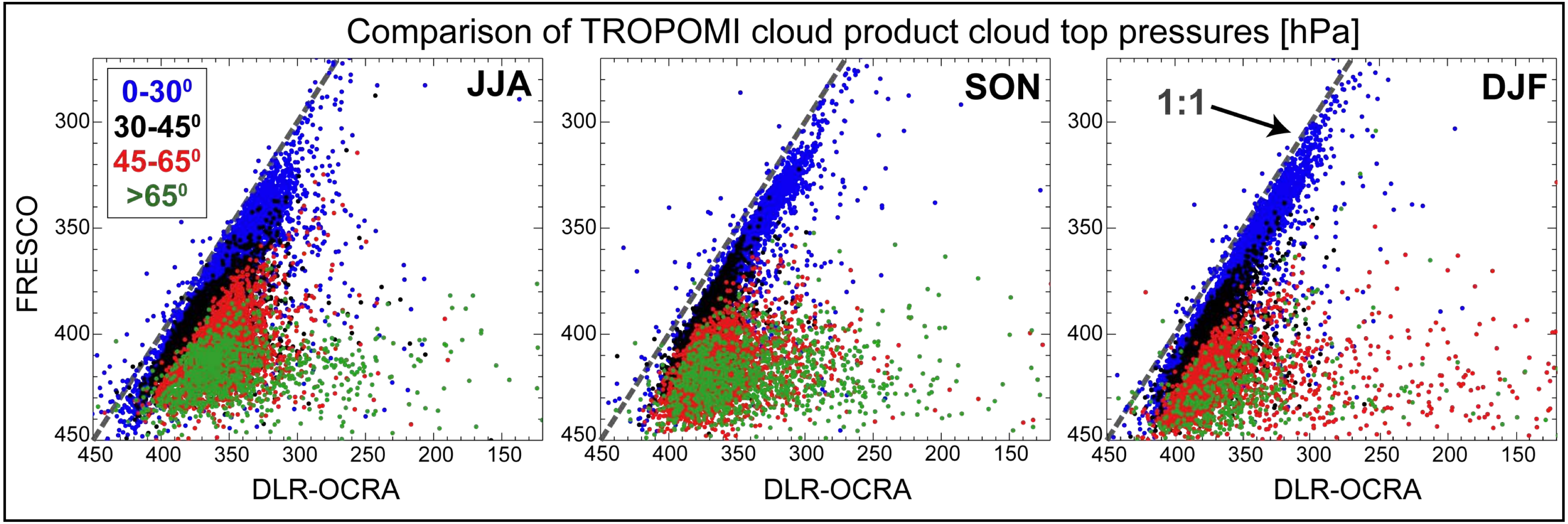
High bias in TROPOMI background NO_2 of 0.8-1.3 $\times$ 10^{15} molecules cm^{-2}

Reported TROPOMI NO_2 stratospheric column is ~10%

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

Evaluate TROPOMI Cloud Top Heights

Comparison of TROPOMI cloud top pressure products at different latitude bands



Compares TROPOMI NO_2 (FRESCO) and operational (DLR-OCRA) cloud product seasonal means at $2° \times 2.5°$ for FRESCO clouds at 180 to 450 hPa

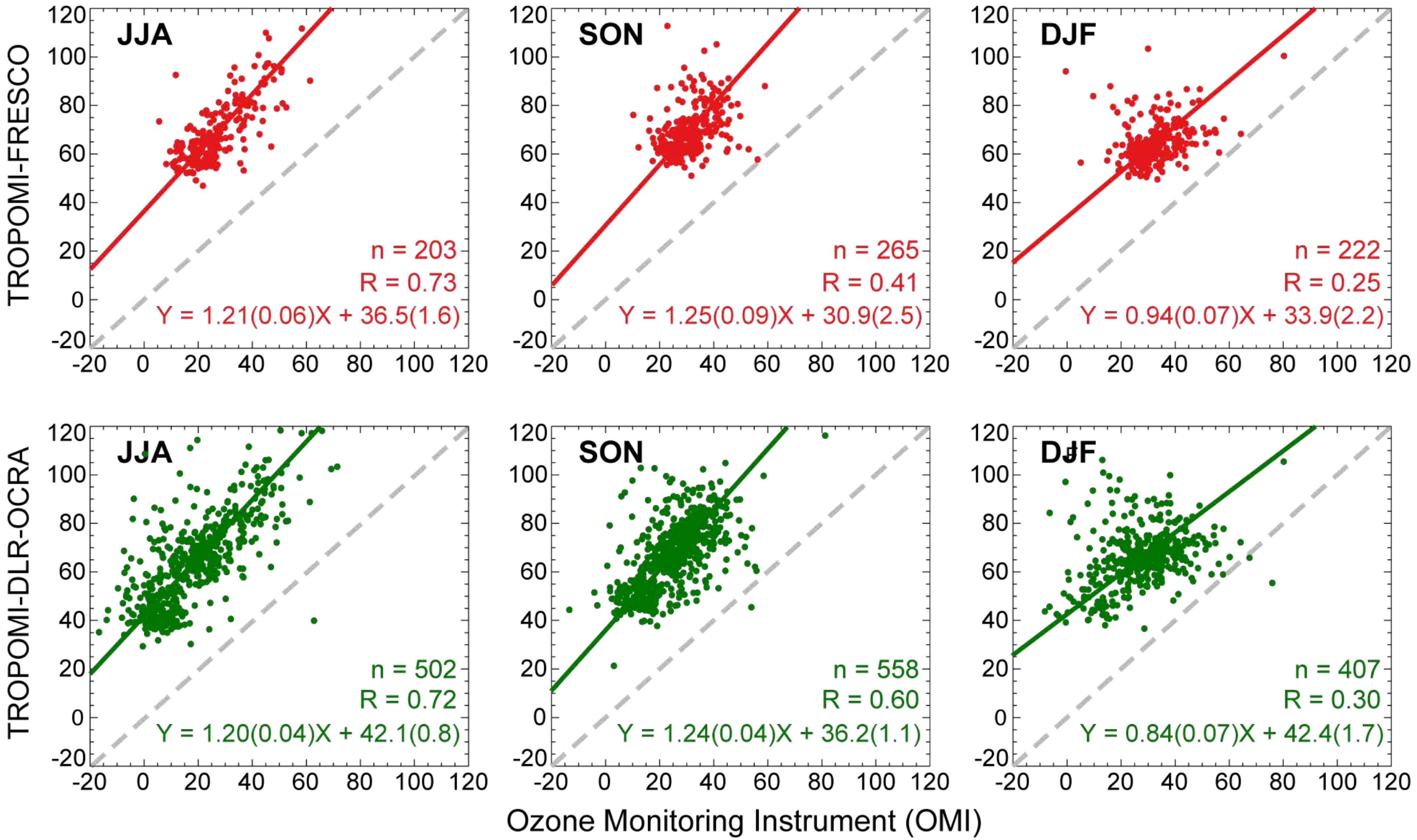
DLR-OCRA systematically lower clouds than FRESCO. Difference increases with latitude.

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

TROPOMI and OMI UT NO_2 Comparison

TROPOMI UT NO_2 [pptv] obtained with **FRESCO** clouds or **DLR-OCRA** clouds compared to OMI UT NO_2

OMI product obtained at 8° × 5° by **S. Choi** and **J. Joiner**. Details in Choi et al. [2014] & Marais et al. [2018]



Values are correlation and regression statistics.

DLR-OCRA cloud product yields greater data density and spatial consistency with OMI product.

TROPOMI positive intercept consistent with bias expect from Pandora comparison and supports upper troposphere as source of bias in column

Concluding Remarks of Preliminary Results

- Positive bias in TROPOMI NO_2 of 8-51% compared to Pandora, likely due to underestimate in free tropospheric a priori NO_2 leading to underestimate in air mass factor.

- Synthetic cloud-sliced NO_2 in the upper troposphere (UT) from GEOS-Chem partial columns broadly reproduces the spatial distribution of “true” UT NO_2 obtained under cloudy conditions in summer.

- Preferentially selecting very cloudy scenes for cloud-slicing leads to a 10% higher NO_2 than is obtained under all-sky conditions from GEOS-Chem.

- The different TROPOMI cloud products yield similar TROPOMI UT NO_2, but with greater spatial coverage with DLR-OCRA than with FRESCO-S due to more dynamic cloud height range in the UT from DLR-OCRA.

- TROPOMI and OMI UT NO_2 have similar spatial distribution and variance, but TROPOMI positive bias of 30-40 pptv supports upper (and perhaps also free) troposphere as source of bias in the column.

- Increasing GEOS-Chem NO_2 mixing ratios in the upper troposphere by a factor of 2 to obtain synthetic partial columns also yields cloud-sliced UT NO_2 that is 30-40 pptv more than the “truth”.

Questions? Contact Eloise Marais (eloise.marais@le.ac.uk)

References

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