

First Estimate of NO₂ in the upper troposphere from TROPOMI



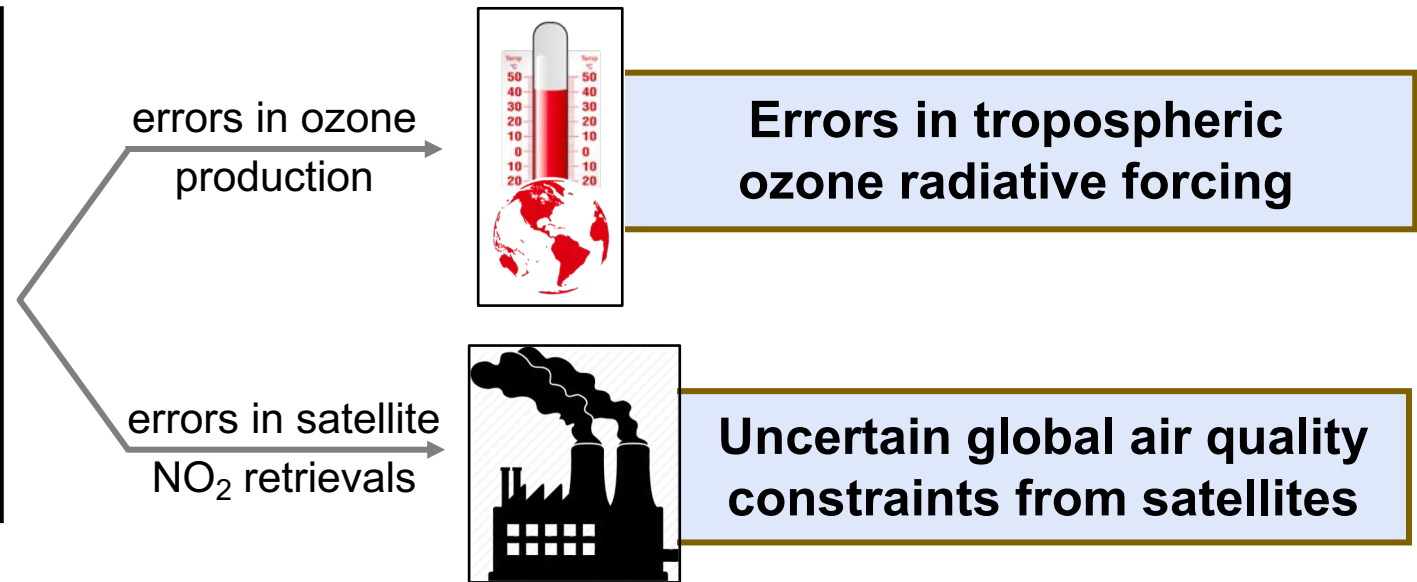
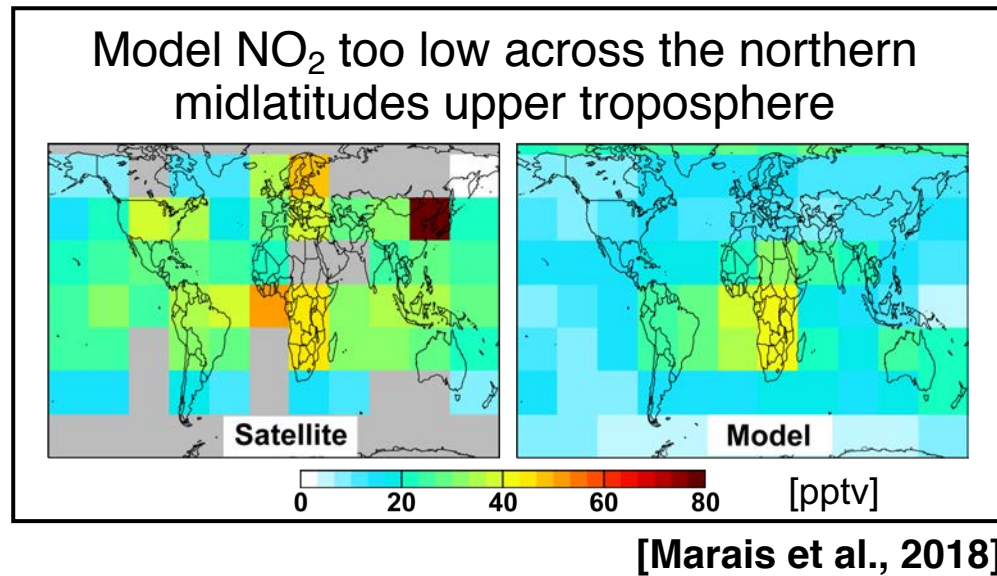
Eloise A. Marais, Henk Eskes, Sungyeon Choi, Joanna Joiner, Alexander Cede

EGU General Assembly
7 May 2020

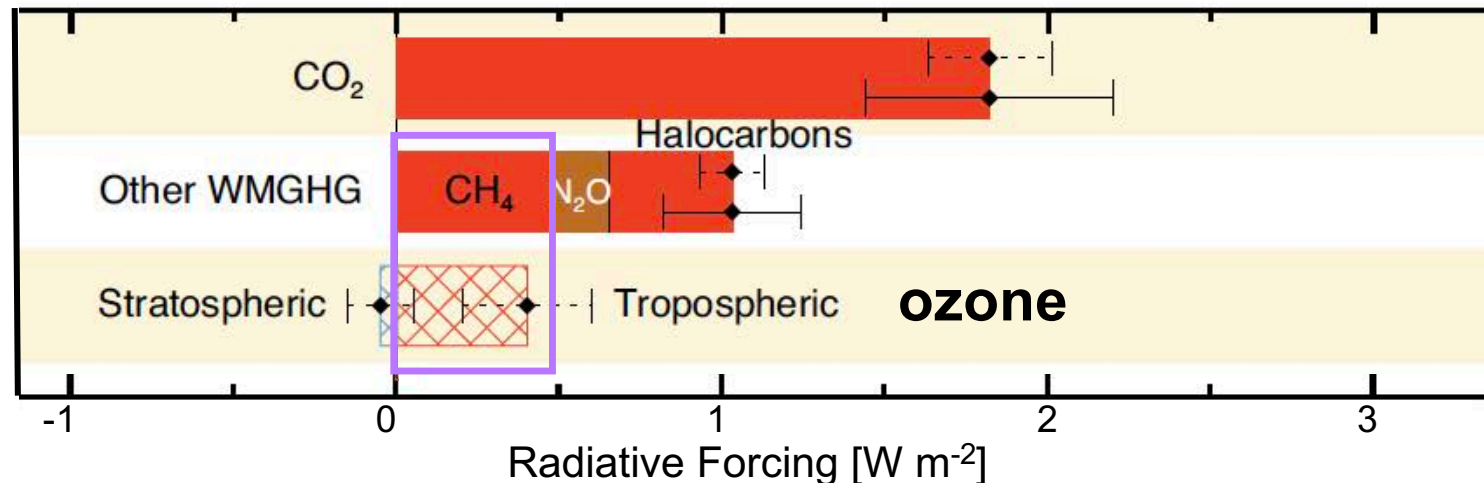
eloise.marais@le.ac.uk
<http://maraisresearchgroup.co.uk/>

The Impact of Errors in the Upper Troposphere are Widespread

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



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



Near-equivalent warming due to ozone and methane

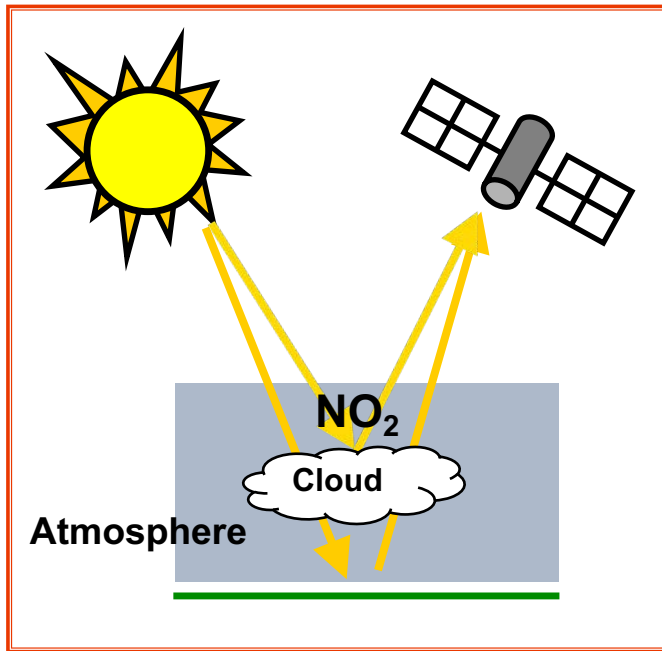
[IPCC AR5, 2013]

Obtain NO₂ in the Upper Troposphere (UT) with Cloud-Slicing

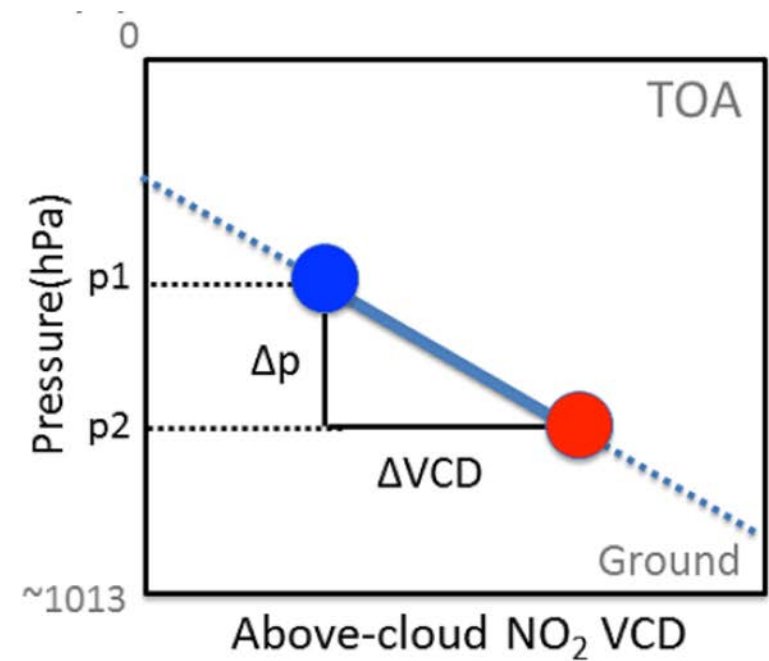
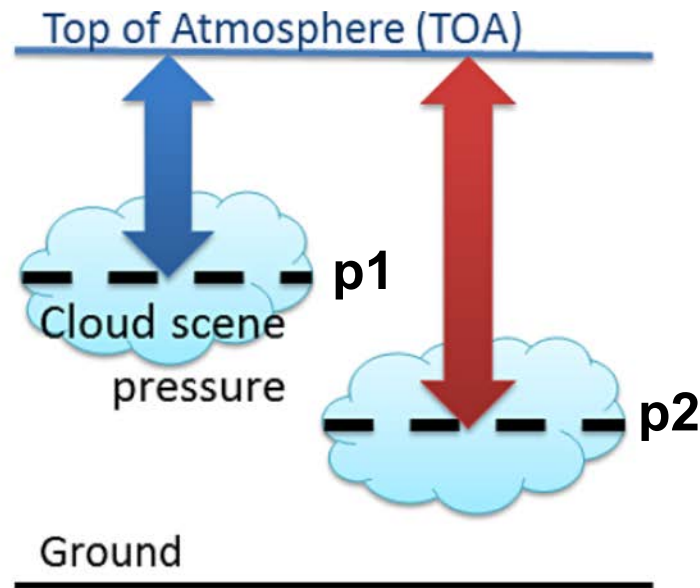
First applied by Ziemke et al. [2001] to TOMS ozone

Apply to partial NO₂ 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]

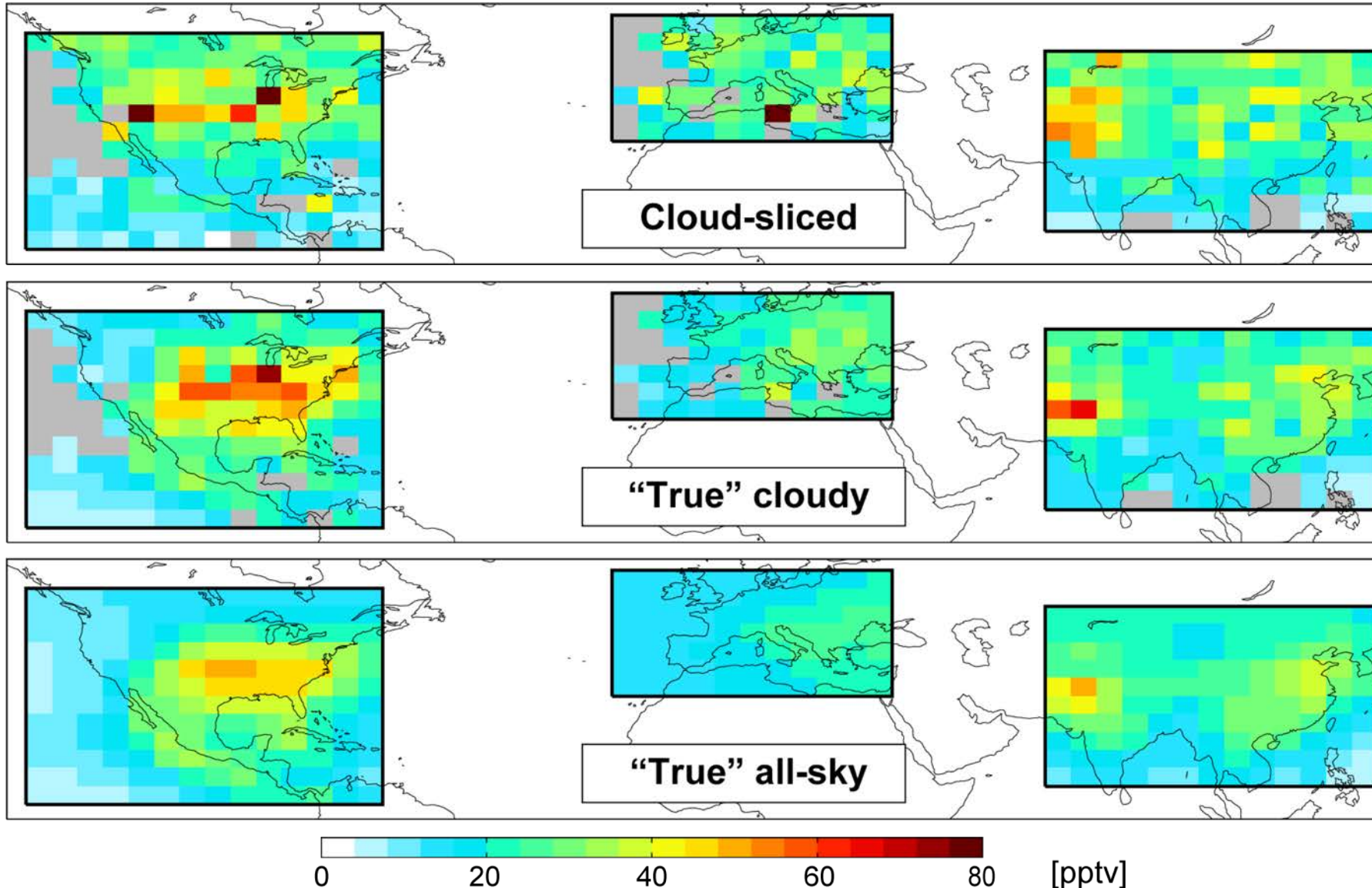
NO₂ volume mixing ratio (VMR) between clouds at p1 and p2

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

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 $0.5^\circ \times 0.3125^\circ$ to obtain UT NO_2 at $4^\circ \times 5^\circ$.

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

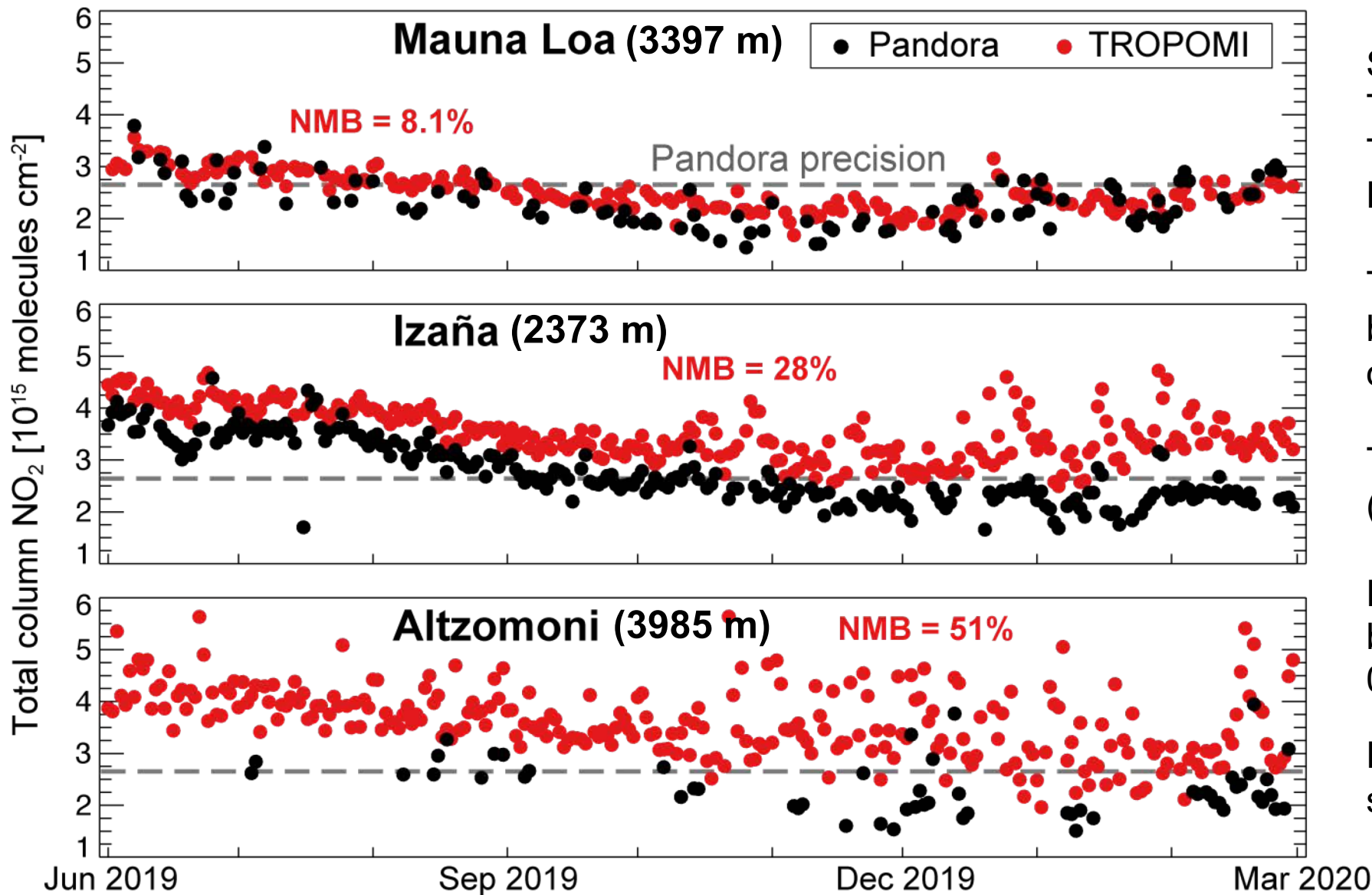
Regression slope for cloud-sliced versus “true” cloudy UT NO_2 is 0.92 ± 0.05 and the intercept is 2.3 ± 1.1 pptv.

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 ($R > 0.7$)

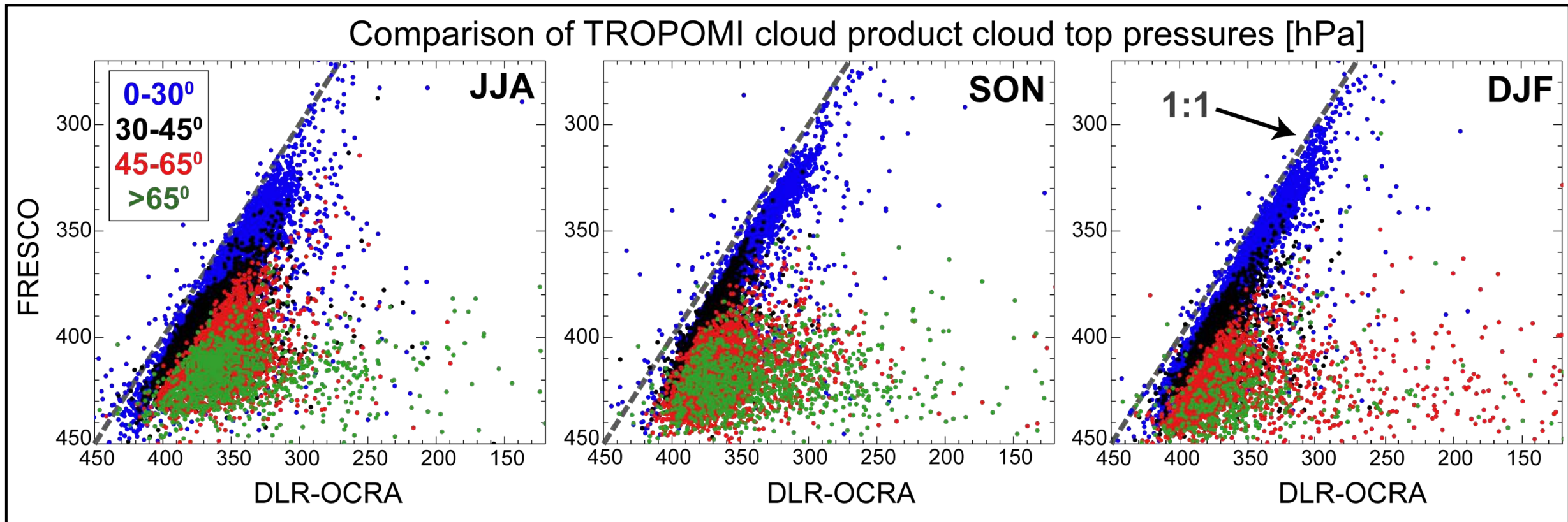
High bias in TROPOMI background NO_2 of $0.8\text{-}1.3 \times 10^{15}$ molecules cm^{-2}

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 NO₂ (FRESKO) and operational (DLR-OCRA) cloud product seasonal means at 2° × 2.5° for FRESKO clouds at 180 to 450 hPa

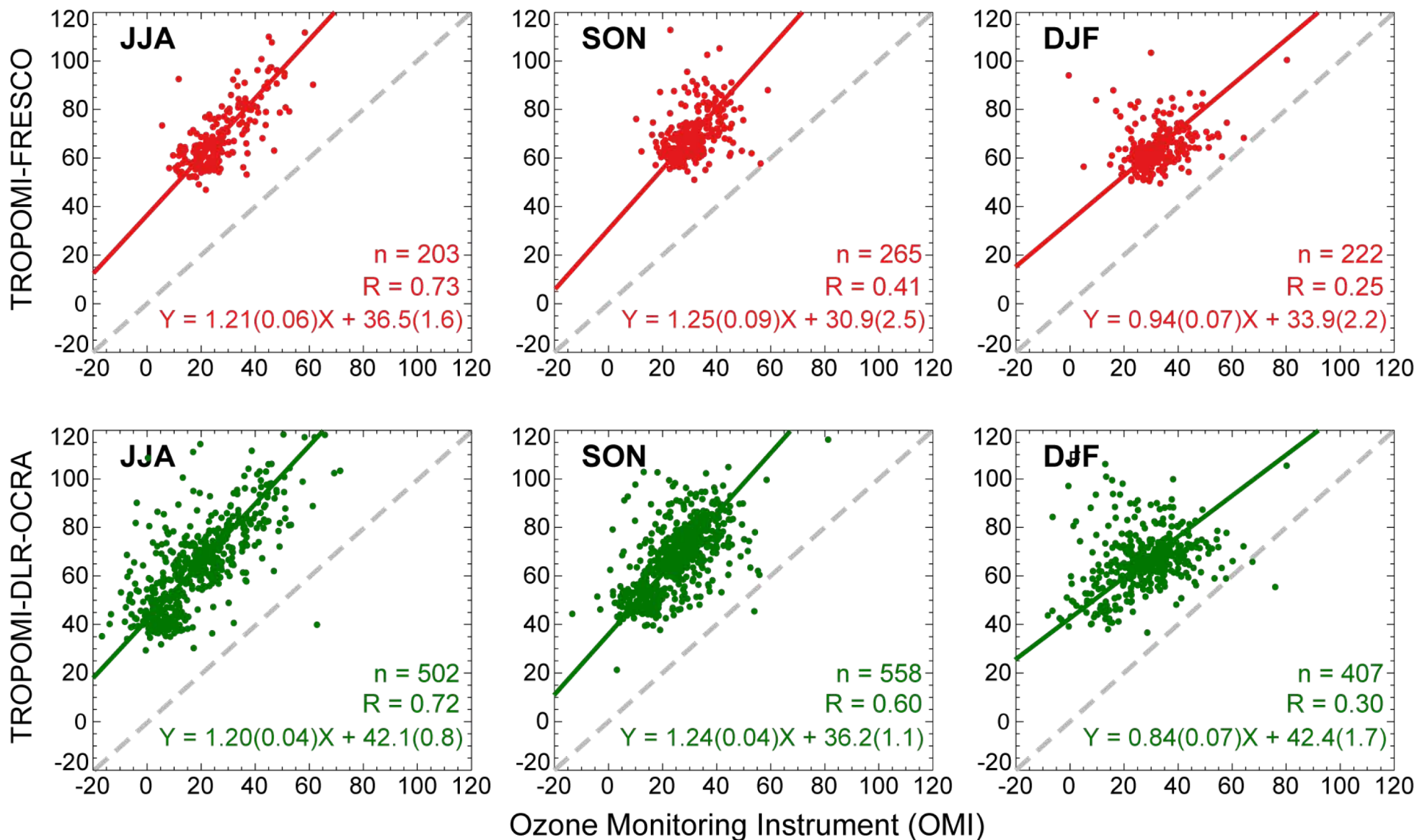
DLR-OCRA systematically lower clouds than FRESKO. Difference increases with latitude.

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

TROPOMI and OMI UT NO₂ Comparison

TROPOMI UT NO₂ [pptv] obtained with **FRESCO** clouds or **DLR-OCRA** clouds compared to OMI UT NO₂

OMI product obtained at $8^\circ \times 5^\circ$ 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 NO₂ of 8-51% compared to Pandora, likely due to underestimate in free tropospheric a priori NO₂ leading to underestimate in air mass factor.
- Synthetic cloud-sliced NO₂ in the upper troposphere (UT) from GEOS-Chem partial columns broadly reproduces the spatial distribution of “true” UT NO₂ obtained under cloudy conditions in summer.
- Preferentially selecting very cloudy scenes for cloud-slicing leads to a 10% higher NO₂ than is obtained under all-sky conditions from GEOS-Chem.
- The different TROPOMI cloud products yield similar TROPOMI UT NO₂, 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 NO₂ 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 NO₂ mixing ratios in the upper troposphere by a factor of 2 to obtain synthetic partial columns also yields cloud-sliced UT NO₂ that is 30-40 pptv more than the “truth”.

Questions? Contact [Eloise Marais](mailto:eloise.marais@le.ac.uk) (eloise.marais@le.ac.uk)

References

- Choi et al.**, First estimates of global free-tropospheric NO₂ abundances derived using a cloud-slicing technique applied to satellite observations from the Aura Ozone Monitoring Instrument (OMI), ACP, 14, 10565–10588, doi:10.5194/acp-14-10565-2014, 2014.
- Myhre et al. (IPCC, AR5)**, Anthropogenic and Natural Radiative Forcing. In: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to AR5 in the IPCC [Stocker et al. (eds.)]. Cambridge University Press, Cambridge, UK and NY, NY, US, 2013.
- Marais et al.**, Nitrogen oxides in the global upper troposphere: interpreting cloud-sliced NO₂ observations from the OMI satellite instrument, ACP, 18, 17017–17027, doi:10.5194/acp-18-17017-2018, 2018.
- Ziemke et al.**, “Cloud slicing”: A new technique to derive upper tropospheric ozone from satellite measurements, JGR, 106, 9853–9867, doi:10.1029/2000jd900768, 2001.