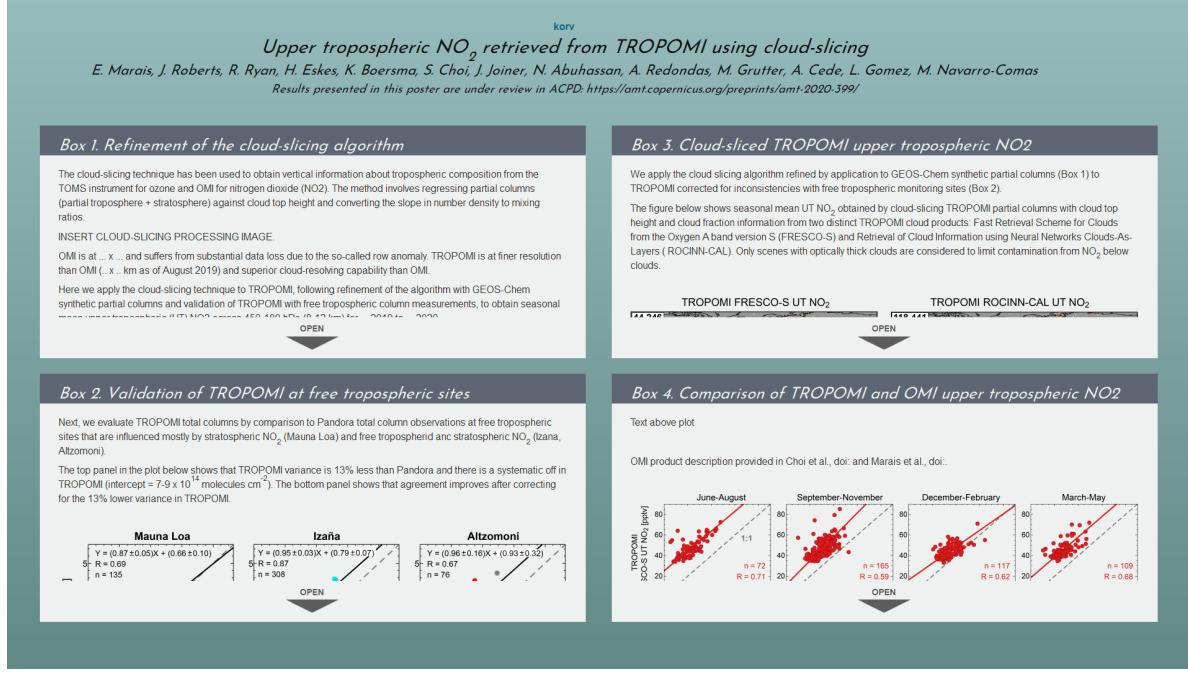


Upper tropospheric NO₂ retrieved from TROPOMI using cloud-slicing



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Results presented in this poster are under review in ACPD: <https://amt.copernicus.org/preprints/amt-2020-399/>

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BOX 1. REFINEMENT OF THE CLOUD-SLICING ALGORITHM

The cloud-slicing technique (Figure 1) has been used to obtain vertical information about ozone from the Total Ozone Mapping spectrometer (TOMS) (Ziemke et al., 2001) and NO₂ from the Ozone Monitoring instrument (OMI) (Choi et al., 2014; Belmonte-Rivas et al., 2015; Marais et al., 2018). Here we apply it to TROPOMI to obtain NO₂ concentrations in the upper troposphere.

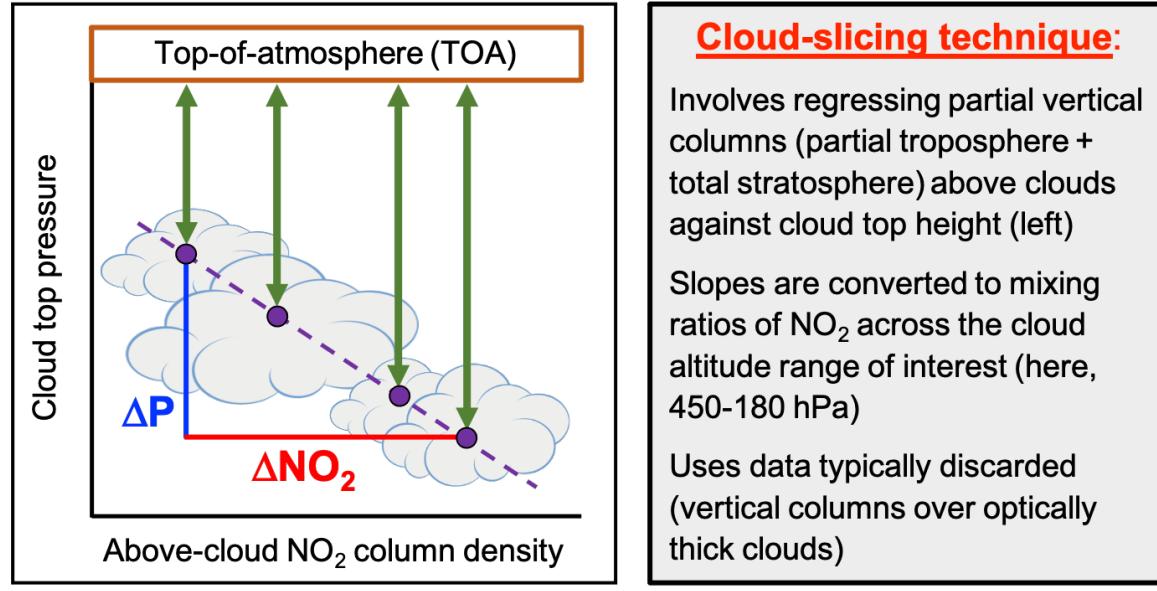


Figure 1. Schematic of the cloud-slicing technique used to calculate mixing ratios of NO₂.

We start by refining the cloud-slicing algorithm with GEOS-Chem synthetic partial columns simulated at 25-32 km in June-August for domains nested over North America, China, and Europe.

We assess the skill of the cloud-slicing technique by comparing NO₂ mixing ratios calculated by the model ("true" UT NO₂) and obtained by cloud-slicing partial synthetic columns above clouds at an altitude range of 450-180 hPa. The regression plot (Figure 2) confirms that cloud-sliced UT NO₂ is consistent with the "truth".

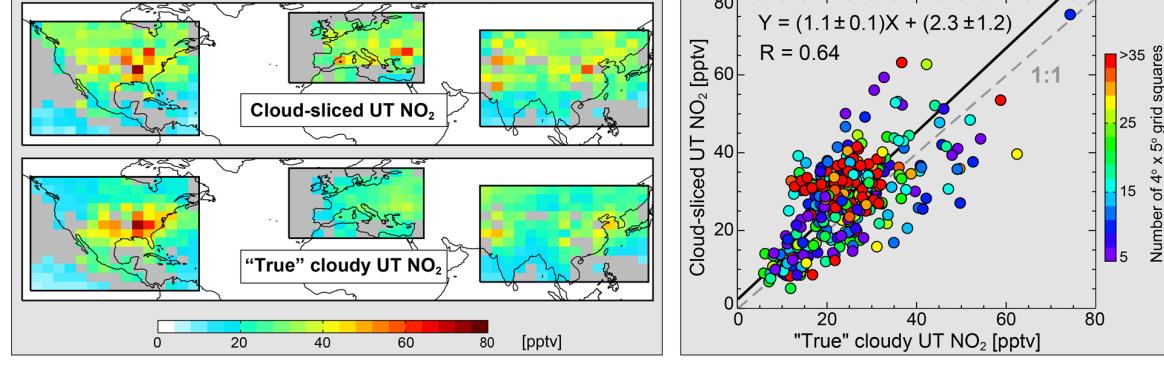


Figure 2. Comparison of cloud-sliced and "true" UT NO₂ obtained using synthetic data from GEOS-Chem.

BOX 2. VALIDATION OF TROPOMI AT FREE TROPOSPHERIC SITES

Next, we evaluate TROPOMI total columns with Pandora observations at free tropospheric sites that range in tropospheric influence from near negligible (Mauna Loa) to 5-15% (Izaña) to >20% (Altzomoni).

The top panel in Figure 3 shows that TROPOMI stratospheric column variance is 13% less than Pandora that would cause an overestimate in the contribution of the tropospheric column to the total and an overestimate in cloud-sliced UT NO₂.

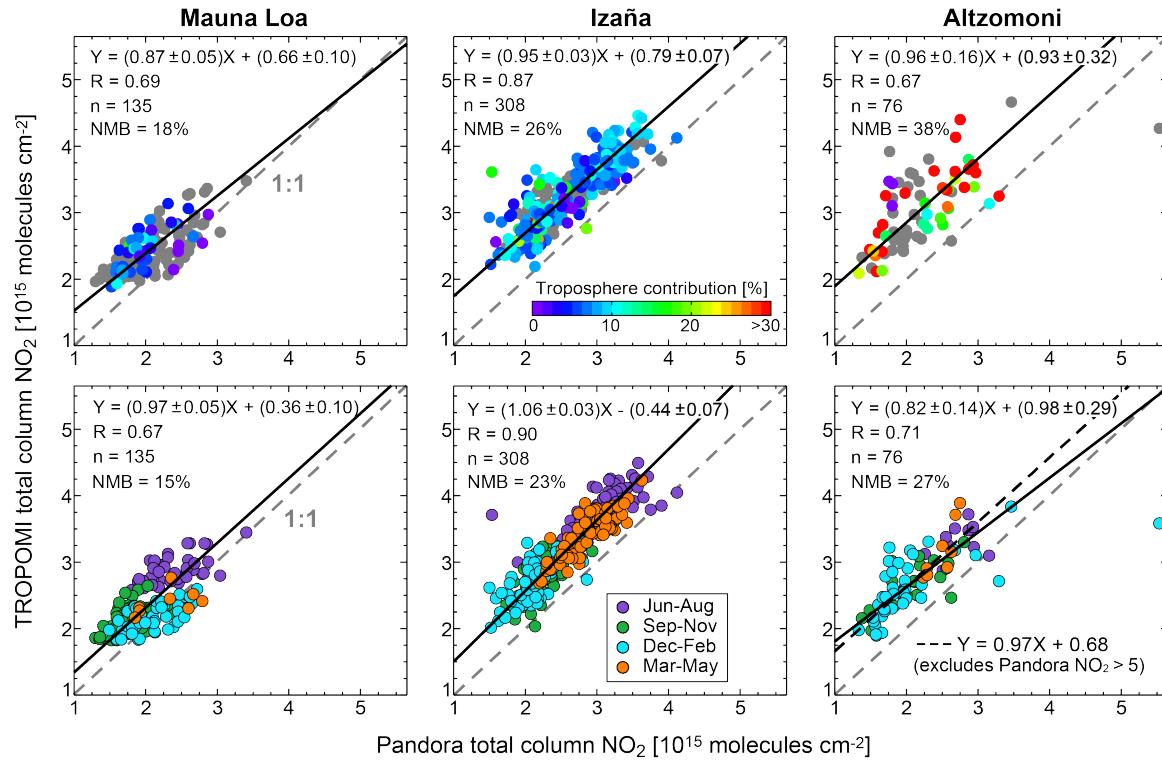


Figure 3. Comparison of midday mean TROPOMI and Pandora total column NO₂ at free tropospheric sites.

We go on to compare TROPOMI, MAX-DOAS and Pandora tropospheric column NO₂ at Izaña to identify that TROPOMI is 90% more than the ground-based observations, decreasing to 30-40% more after applying a variance correction to TROPOMI stratospheric columns.

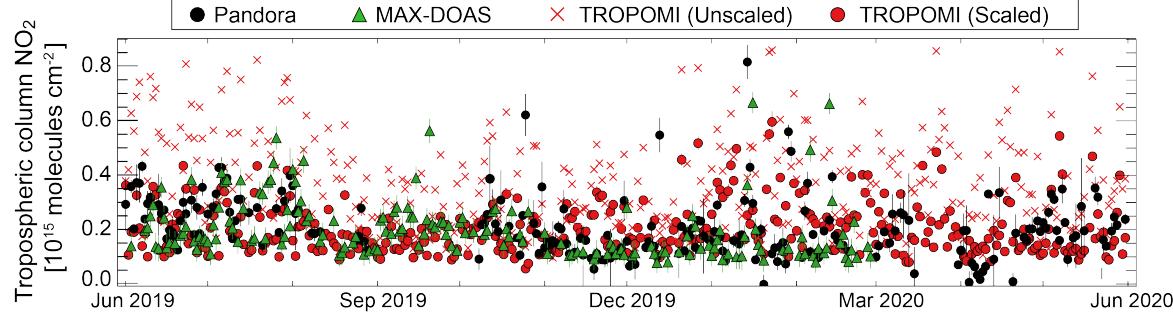


Figure 4. Comparison of midday TROPOMI, MAX-DOAS and Pandora daily mean tropospheric column NO₂

at Izaña.

The bottom panel of Figure 3 compares TROPOMI to Pandora after applying correction factors to TROPOMI. The offset decreases to $4-7 \times 10^{14}$ molecules cm^{-2} . There is only a marginal improvement in the TROPOMI normalized mean bias (NMB). This will be removed with cloud-slicing, assuming that the stratosphere is the source of the positive NMB.

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BOX 3. CLOUD-SLICED TROPOMI UPPER TROPOSPHERIC NO₂

We obtain seasonal mean mixing ratios of UT NO₂ at $1^\circ \times 1^\circ$ for June 2019 to May 2020 (Figure 5) by applying the refined cloud-slicing algorithm (Box 1) to corrected TROPOMI total columns (Box 2) above optically thick clouds. Cloud top height and fraction are from 2 distinct cloud products: FRESCO-S cloud top height and fraction and combined OCRA cloud fraction and ROCINN-CAL cloud top heights.

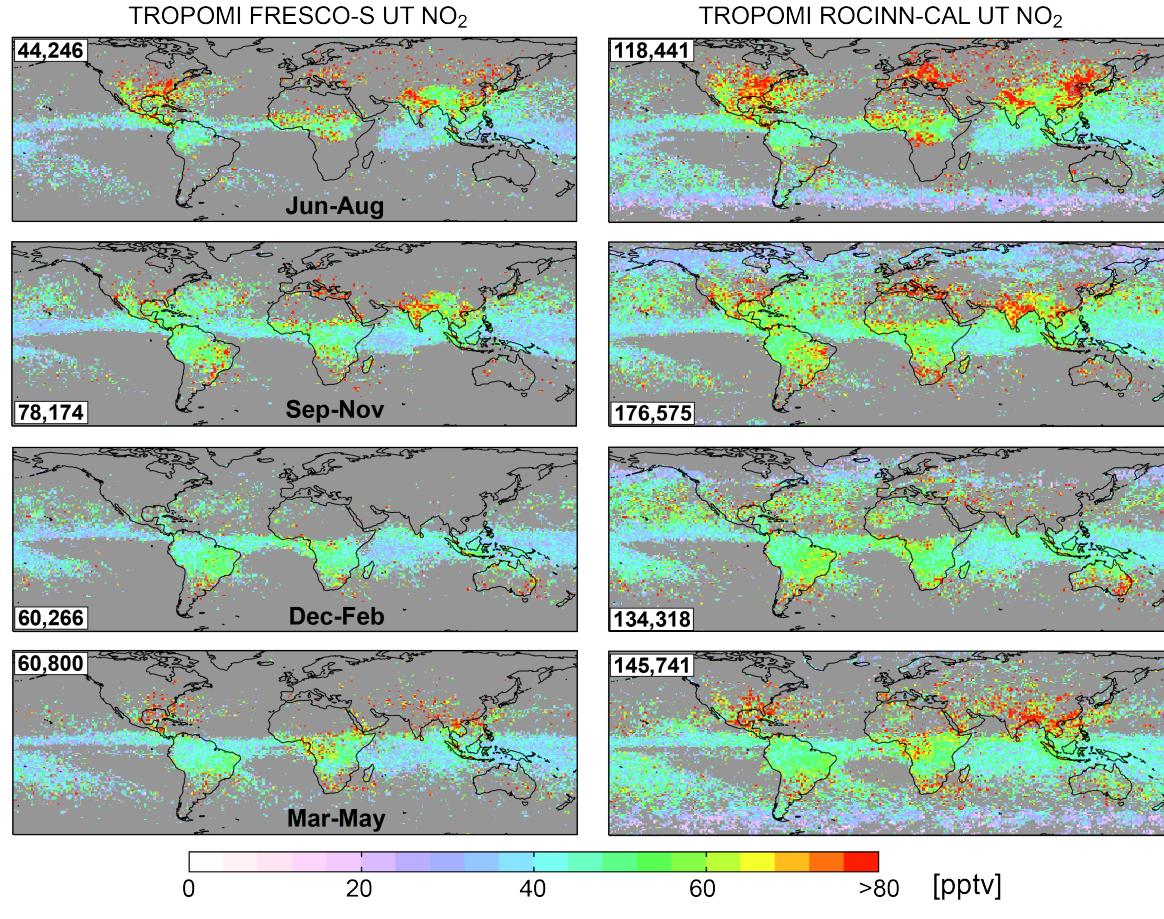


Figure 5. Seasonal mean TROPOMI cloud-sliced upper tropospheric (UT) NO₂ at $1^\circ \times 1^\circ$ using cloud information from Fast Retrieval Scheme for Clouds from the Oxygen A band version S (FRESCO-S) (left) or Retrieval of Cloud Information using Neural Networks Clouds-As-Layers (ROCINN-CAL) (right). Inset values are total successful cloud-sliced retrievals.

Enhancements in UT NO₂ in Figure 5 are as expected: over locations with intense year-round (tropics) and seasonal (subtropics and midlatitudes) lightning. Evidence of contamination is over anthropogenic hotspots in North China and the northeast US and unprecedented wildfires in Australia in December-January.

The latitudinal extent of TROPOMI UT NO₂ is greater using ROCINN-CAL than FRESCO-S, as ROCINN-CAL has twice as many optically thick clouds (cloud fraction ≥ 0.7) than FRESCO-S (Figure 6) and ROCINN-CAL has a wider cloud top height range than FRESCO-S (not shown).

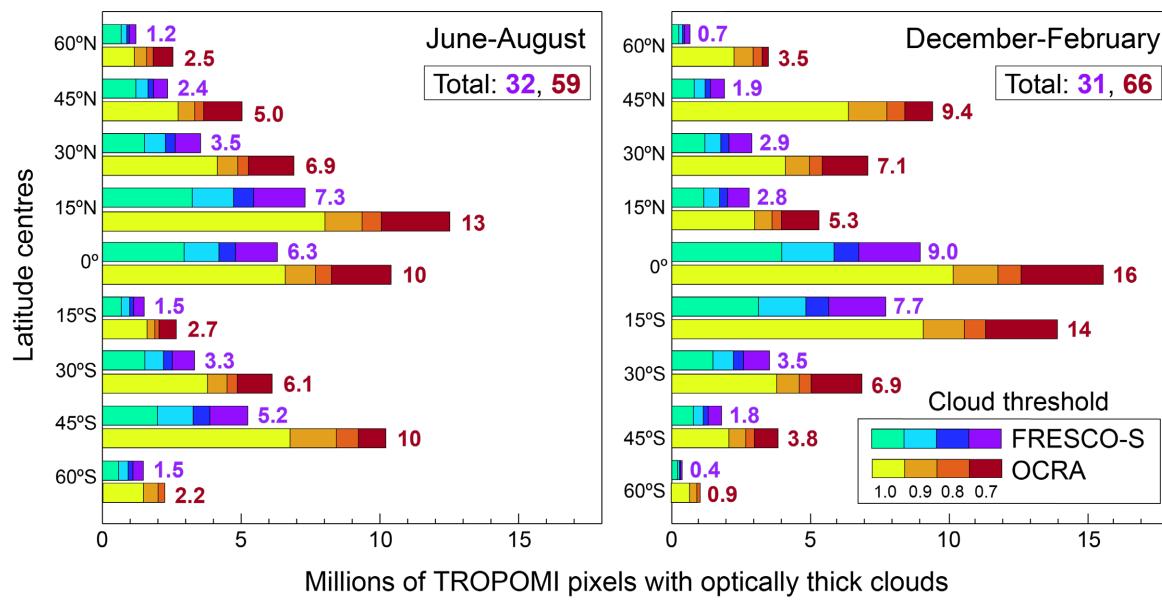


Figure 6. Meridional distribution of upper tropospheric optically thick clouds from FRESCO-S and OCRA (cloud fraction used in the ROCINN-CAL product).

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BOX 4. EVALUATION OF TROPOMI UPPER TROPOSPHERIC NO₂ WITH OMI

An OMI UT NO₂ product of multiyear (2005-2007) seasonal means is detailed in Marais et al. (2018) using a similar algorithm to that applied to TROPOMI, but at coarser spatial resolution ($5^{\circ} \times 8^{\circ}$) and across a narrower cloud top range (450 - 280 hPa) than TROPOMI. We use it to evaluate our TROPOMI product (Figure 7).

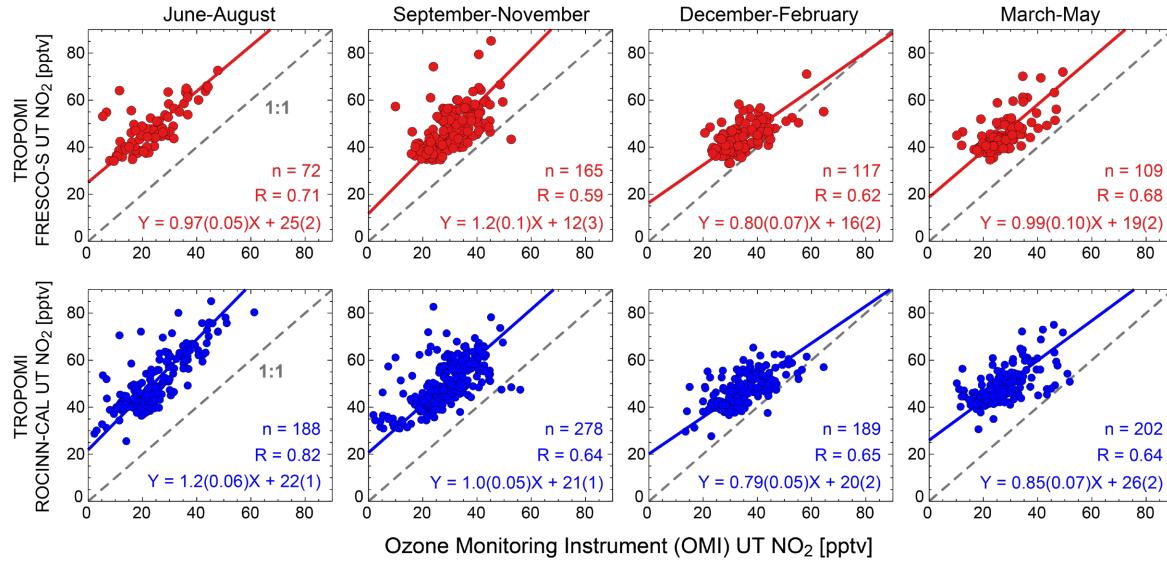


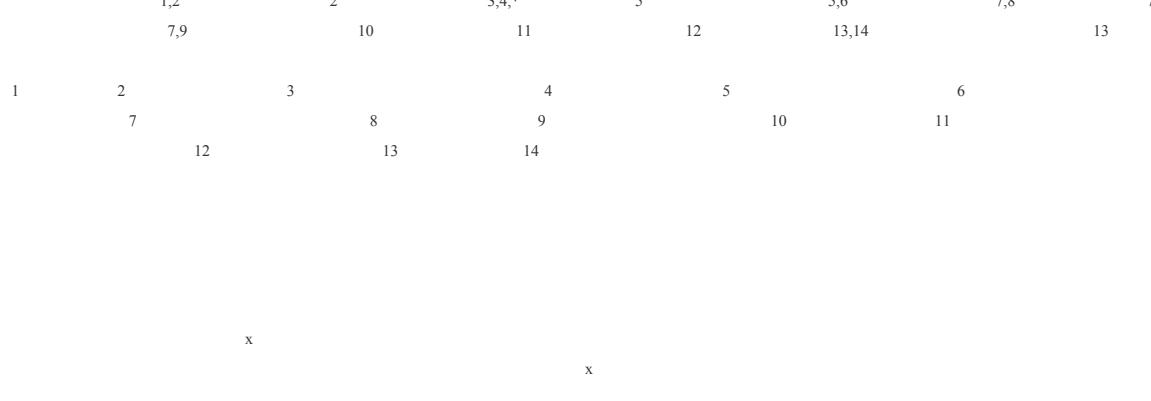
Figure 7. Evaluation of TROPOMI UT NO₂ with OMI UT NO₂ at the OMI grid resolution.

Both TROPOMI products are spatially consistent with OMI in all seasons for both products, but with a 12-26 pptv positive offset in TROPOMI likely due to increase in NO₂ with altitude across the sampling pressure ceilings of the 2 products. If no correction factors are applied to TROPOMI (Box 2), differences between the two products are much larger (intercepts = 16-36 pptv, slopes = 1.2-1.7).

Concluding Remarks

The TROPOMI UT NO₂ product described here offers the opportunity to address poor understanding of reactive nitrogen in the UT, constrain sources of reactive nitrogen to the UT, and assess spatial and temporal variability of reactive nitrogen in the UT, in particular interannual variability.

If you are interested in using the data, please contact Eloise Marais (e.marais@ucl.ac.uk).



ABSTRACT

Nitrogen oxides in the NO_x-limited upper troposphere (UT) have a large influence on the oxidizing capacity of the troposphere and formation of the greenhouse gas ozone. Models misrepresent NO_x in the UT and observations to address deficiencies in models are sparse. Here we obtain one year of near-global seasonal mean mixing ratios of NO₂ in the UT (450-180 hPa) by applying cloud-slicing to TROPOMI partial columns of NO₂. This follows refinement of the cloud-slicing algorithm by application to synthetic partial columns from the GEOS-Chem model and bias correction of TROPOMI compared to ground-based observations from Pandora and MAX-DOAS instruments at high-altitude sites. TROPOMI underestimates stratospheric NO₂ variance by 14%, overestimates the stratospheric column by 7×10^{14} molecules cm⁻² and overestimates the free tropospheric column by a factor of 2. Two cloud-sliced UT NO₂ products are retrieved using distinct cloud products, so-called FRESCO-S and ROCINN-CAL. These yield consistent UT NO₂, but with greater spatial coverage using ROCINN-CAL, due to its greater abundance of optically thick clouds and greater variability in cloud top heights at the midlatitudes and poles. The TROPOMI UT NO₂ products are spatially consistent with UT NO₂ from the existing Ozone Monitoring Instrument (OMI) product. A positive bias in the TROPOMI product of ~10 pptv is expected, due to finer resolution of TROPOMI pixels and the UT NO₂ product than OMI, and greater vertical extend of TROPOMI (up to 180 hPa) than OMI (280 hPa) over a pressure range in which NO₂ increases with altitude. The TROPOMI UT NO₂ product offers potential to evaluate and improve representation of UT NO_x in global chemical transport models and supplement in situ NO₂ observations from aircraft that are susceptible to large biases in the UT.

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