

Figure S1: Synthetic cloud-sliced NO<sub>2</sub> in the upper troposphere (UT) for June-August 2016-2017. Data are obtained by applying cloud-slicing to partial NO<sub>2</sub> columns from GEOS-Chem nested domains simulated at 0.25° × 0.3125° to obtain UT NO<sub>2</sub> at 2° × 2.5° (top panel) and 8° × 10° (bottom panel). Grey grids have <5 data points.

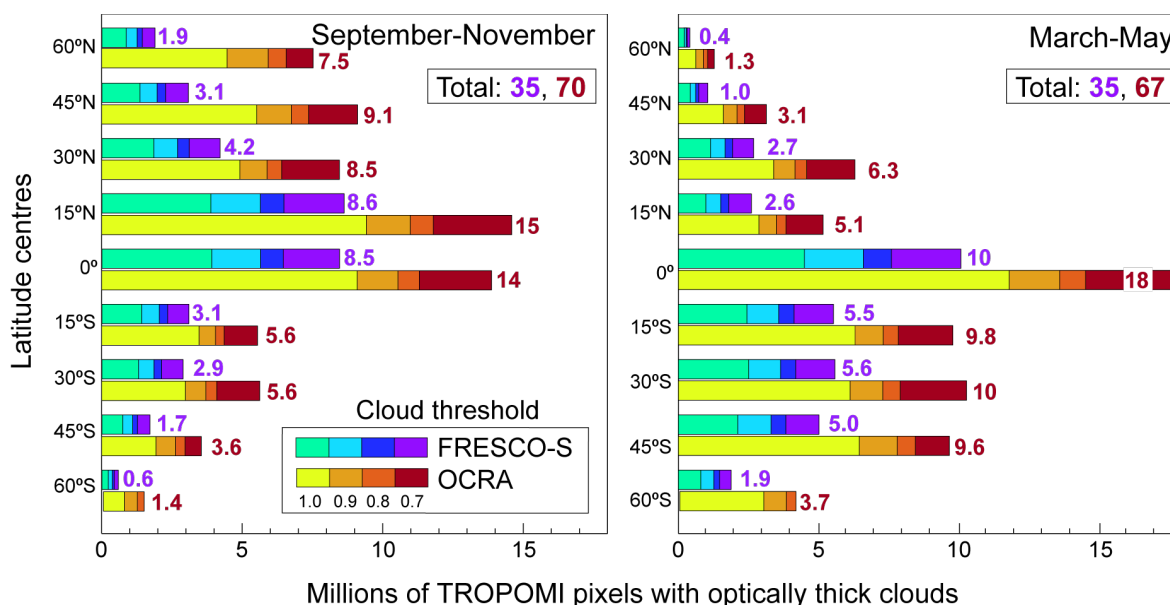


Figure S2: Meridional distribution of FRESCO-S and OCRA optically thick clouds in the upper troposphere. Bars count the occurrence of native TROPOMI pixels with cloud fractions  $\geq 1.0$ , 0.9, 0.8, and 0.7 binned into 15° latitude bands in September-November (left) and March-May (right) for FRESCO-S (cool colours) where FRESCO-S cloud top pressures are at 450-180 hPa, and OCRA (warm colours) where ROCINN-CAL cloud top pressures are at 450-180 hPa. Values inset are latitude band and global total number of TROPOMI pixels with cloud fraction  $\geq 0.7$ .

# TROPOMI ROCINN-CAL upper tropospheric NO<sub>2</sub> for cloud fraction $\geq 1.0$

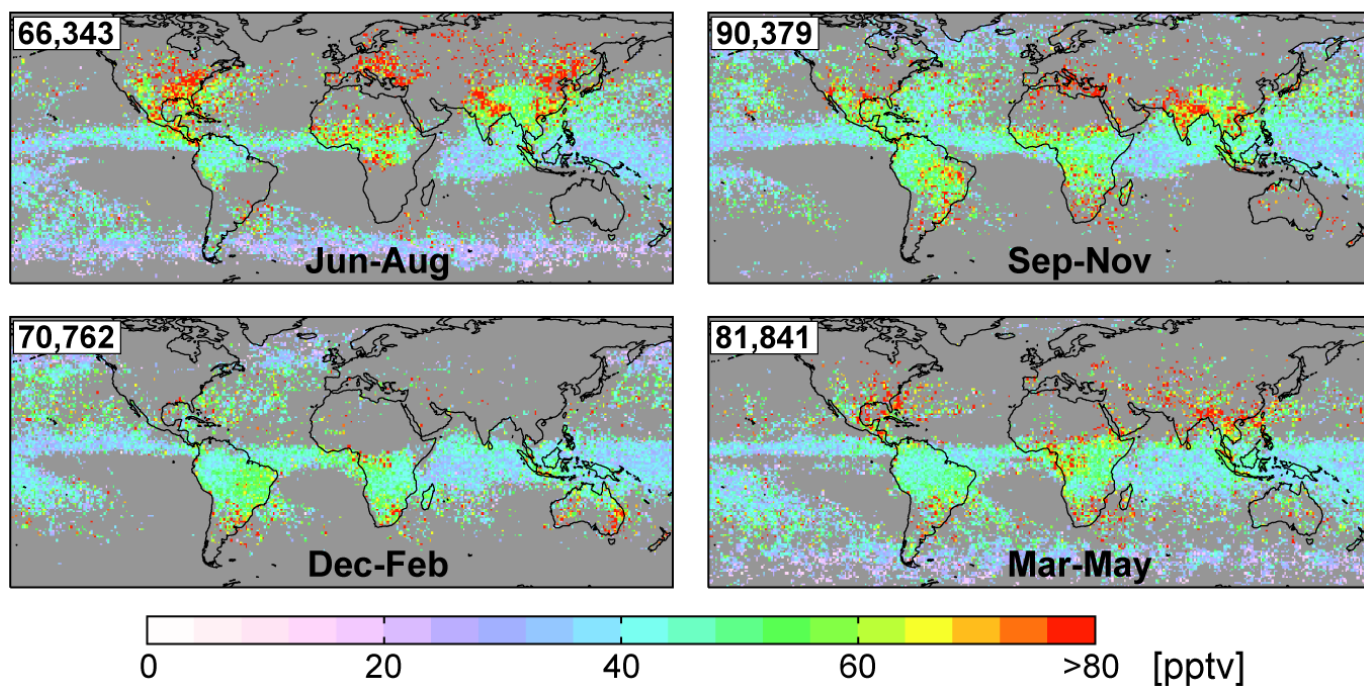


Figure S3: Seasonal mean upper tropospheric (UT) NO<sub>2</sub> from TROPOMI at  $1^\circ \times 1^\circ$  using a ROCINN-CAL cloud fraction threshold of 1.0 instead of 0.7 (Figure 5). Results are obtained after applying correction factors to TROPOMI stratospheric and tropospheric columns (see manuscript for details).

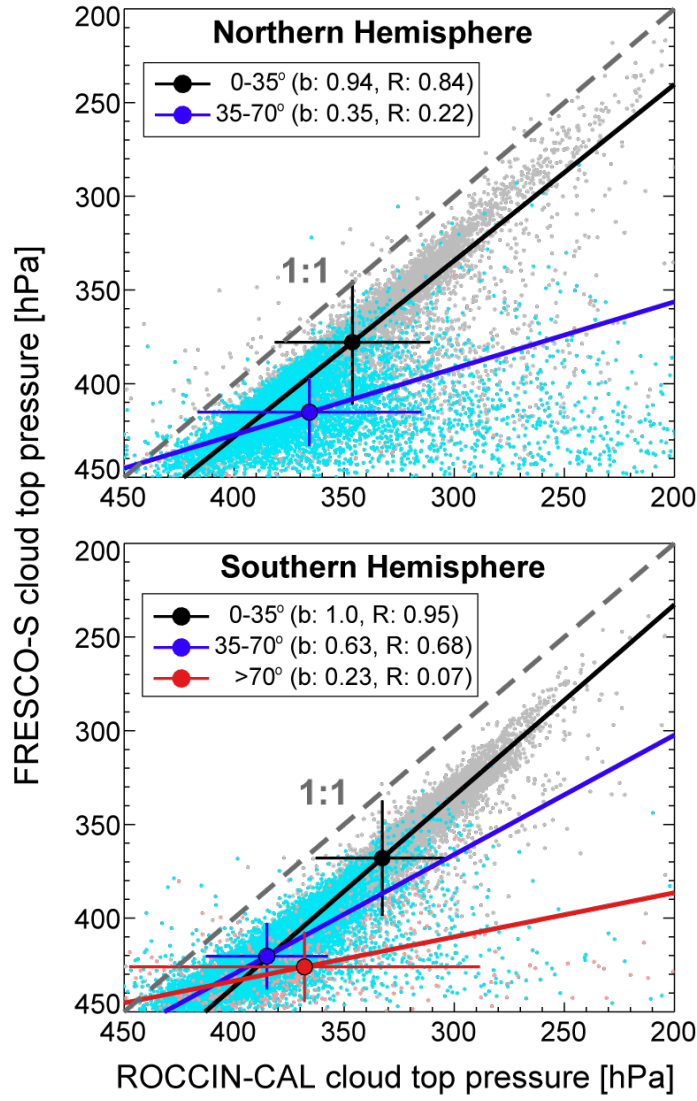
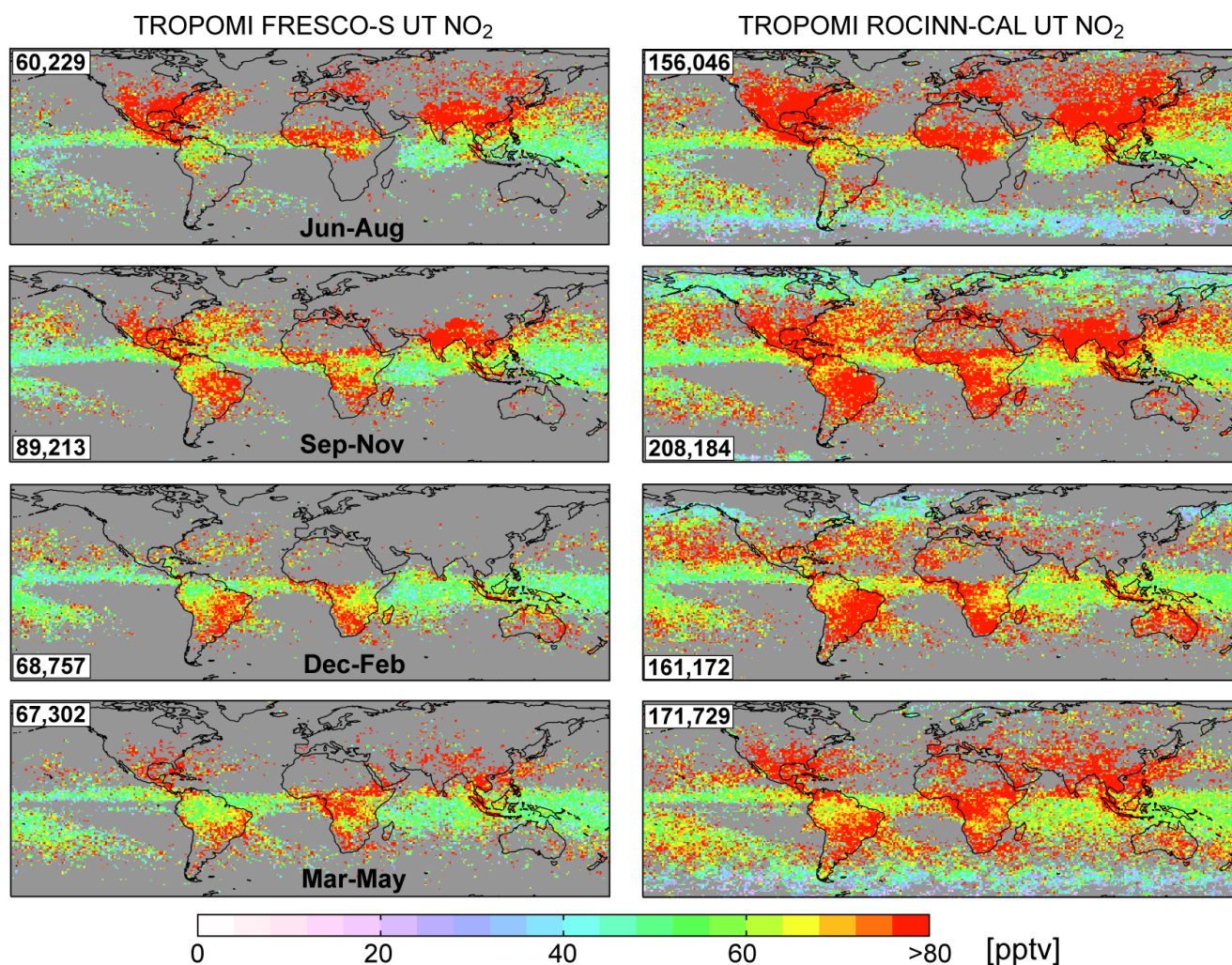


Figure S4: Comparison of FRESKO-S and ROCINN-CAL cloud top pressures from optically thick clouds in the upper troposphere for December 2019 to February 2020. Data are gridded to  $1^\circ \times 1^\circ$  for TROPOMI pixels with FRESKO-S cloud fractions  $\geq 0.7$  and cloud top pressures at 450-180 hPa. Small points are gridded seasonal means and lines are RMA regressions for the tropics (grey points, black regression line), subtropics and midlatitudes (cyan, blue), and poles (pink, red). Large points are latitude band means and error bars are corresponding standard deviations. Grey dashed lines show the 1:1 relationship. Values in the legend are Pearson's correlation coefficients (R) and RMA regression slopes (b).



35

40

**Figure S5: Seasonal mean upper tropospheric (UT) NO<sub>2</sub> from TROPOMI at 1° × 1° with no correction factors applied to TROPOMI (see manuscript for details). Data are obtained with cloud fraction and cloud top pressure information from the FRESCO-S (left) or ROCINN-CAL (right) TROPOMI cloud product for June-August 2019 (first row), September-November 2019 (second), December 2019 to February 2020 (third), and March-May 2020 (fourth). The colour scale saturates at 80 pptv.**