

1.5 years of TROPOMI CO measurements: Comparisons to MOPITT and ATom - SUPPLEMENT MATERIALS

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S1 TROPOMI versus MOPITT over land: non-colocated retrievals

Here we describe results from the analysis of daily (from 7 November 2017 to 10 March 2019) non-colocated TROPOMI and MOPITT retrievals over 8 ROIs: 2 hemispheric, 4 representative of polluted regions (USA, Europe, India, and China), and 2 of clean regions (Sahara and Australia). TROPOMI and MOPITT retrievals were filtered to keep only clear daytime data over land. Daily mean retrievals for each dataset as well as relative bias between TROPOMI and each of the three MOPITT products (TIR, NIR, and TIR+NIR) were calculated; relative bias = $100 \times (\text{TROPOMI} - \text{MOPITT}) / \text{MOPITT}$. By utilizing non-colocated retrievals we maximized the size and diversity of the populations analyzed. Results from this analysis are summarized in Fig. S1.

S1.1 TROPOMI versus MOPITT TIR

Results summarized in Fig. S2 show that during the ~1.5 year analyzed, TROPOMI and MOPITT TIR total CO column retrievals were close to each other both in magnitude and temporal variation. The two datasets show strong differences between clean ROIs (Sahara and Australia; $10\text{-}20 \times 10^{17}$ molec. cm^{-2}) and highly polluted ROIs (India and China; up-to- 40×10^{17} molec. cm^{-2}). They also show the expected differences between the two hemispheres: retrievals are, overall, lower in the S Hemisphere ($10\text{-}20 \times 10^{17}$ molec. cm^{-2} versus $16\text{-}22 \times 10^{17}$ molec. cm^{-2}) due to less land area, population, and industrial activity. Both TROPOMI and MOPITT TIR show equivalent seasonal variability. ROIs located in the Northern hemisphere present an absolute maximum during boreal winter and a relative maximum in late boreal summer. The absolute maximum

is consistent with winter CO accumulation due to shorter days and larger zenithal angles, resulting in less photolysis, and to increased emissions due to biomass burning north of the Equator in Africa. The relative maximum is most likely due to fire emissions. Conversely, seasonal trends in Southern hemisphere ROIs show a maximum in September-October, consistent with CO accumulation during austral winter and emissions from biomass burning S of the equator. Daily relative bias values are generally within a $\pm 10\%$ range for all the ROIs except the two most polluted (India and China), where most values are between -20 to +40 %. When averaged over time (Table S1), relative biases are between -10.07 % (S Hemisphere) and 11.73 % (China), with a mean for all the ROIs of -3.81 %. We note that biases for most ROIs are predominantly negative, except for China.

S1.2 TROPOMI versus MOPITT NIR

Figure S3 shows daily results from the comparison of non-colocated TROPOMI and MOPITT NIR land retrievals; time-averaged results are summarized in Table S1. The ranges of daily mean retrievals and seasonal trends observed in each ROI are in general analogous to those described in Sect. S1.1. Relative bias values averaged for the period analyzed range between -10.60 % (S Hemisphere) and 6.88 % (China), while the mean for all the ROIs is -2.99 %. Bias values for the Sahara ROI (mostly in the -5 to 10 % range) contrast strongly with those shown in Fig. S2.g (-10 to -5 %). For all the other ROIs, relative biases with respect to MOPITT NIR are broadly similar in magnitude to those respect MOPITT TIR, albeit the former present larger oscillations along time. This is consistent with the MOPITT NIR retrievals being more sensitive to geophysical noise due to changes in albedo during MOPITT observation associated with spacecraft motion (Deeter et al., 2011).

S1.3 TROPOMI versus MOPITT TIR+NIR

Daily results from non-colocated TROPOMI and MOPITT TIR+NIR retrievals are shown in Fig. S4; temporally averaged results are summarized in Table S1. Results are similar to those described in Sect. S1.1 in terms of daily means, seasonal trends, and relative biases. The latter range between -9.96 % (S Hemisphere) and 12.73 % (China); the mean for all ROIs is -3.50 %.

S2 TROPOMI versus ATom-4 over water: above/below cloud analysis

Results from an analysis of colocated TROPOMI and true (unsmoothed) ATom-4 profiles over bodies of water performed for the period between 24 April and 21 May 2018 are summarized in Fig. S5 and Table S2. Colocation criteria were same day acquisition and horizontal distance ≤ 50 km; each ATom-4 profile was paired with the closest valid TROPOMI retrieval that met the colocation criteria.

For this comparison we assumed that TROPOMI retrievals are only sensitive to CO above cloud top, while CO below cloud top is fully approximated by TROPOMI's scaled model-based reference profiles. This scenario would be most accurate in case of optically thick clouds. To quantify the error introduced by approximating below-cloud-top CO with TROPOMI reference profiles, we compared TROPOMI retrievals over bodies of water (total columns and their above cloud partial column components) to their colocated ATom-4 counterparts. Complete (e.g., from the surface to the top of the atmosphere) ATom-4

CO profiles were generated following the standard method for MOPITT validation with airborne data, as described in the main article. The complete profiles were then interpolated to match the TROPOMI reference profile 25-level vertical grid. ATom total CO column values were calculated applying Eq. 1 in main article. The corresponding ATom partial column values were also calculated, including only the layers above cloud top. For each TROPOMI observation, a partial above cloud column was calculated by subtracting from the reported total TROPOMI column the below cloud partial column of its colocated, scaled TROPOMI reference profile.

Fig. S5.a shows total CO column retrievals which, for TROPOMI, according to our assumption, would include a measured component (partial column above cloud top) and a reference component (partial column below cloud top). TROPOMI and ATom-4 total CO column values show very strong correlation ($R = 0.93$, slope of linear fit = 0.96) and a small negative relative bias (-4.76 %) indicative of slightly low TROPOMI values with respect to ATom-4. Figure S5.b shows results for partial (above cloud) CO column values. The relative bias in this case is closer to zero (-1.11 %) and the linear fit has a slightly larger R (0.95), indicative of an even stronger correlation between the above-cloud-only component of the two datasets; the slope of the linear fit is slightly lower (0.92). We interpret the difference between these two relative bias values (-3.65 p.p.) as an estimate of the error introduced by assuming that below-cloud partial CO columns can be approximated by TROPOMI scaled CO reference profiles. Results from this analysis characterize a worst-case scenario (where TROPOMI has no sensitivity to CO below cloud top) and they complement results from the TROPOMI versus ATom-4 analysis presented in the main article, where it is assumed that TROPOMI has some sensitivity to CO below cloud top.

65 **References**

Deeter, M. N., Worden, H. M., Gille, J. C., Edwards, D. P., Mao, D., and Drummond, J. R.: MOPITT multispectral CO retrievals: Origins and effects of geophysical radiance errors, *JOURNAL OF GEOPHYSICAL RESEARCH-ATMOSPHERES*, 116, <https://doi.org/10.1029/2011JD015703>, 2011.

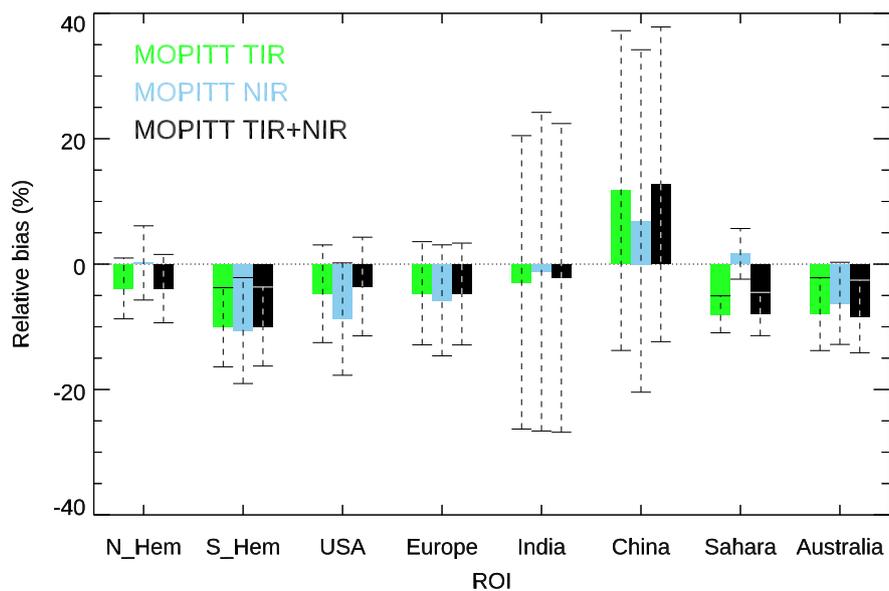


Figure S1. Summary of non-located land comparison results. Colored bars represent relative bias between TROPOMI and each of the three MOPITT products (TIR, NIR, and TIR+NIR). Solid lines show ± 1 standard deviation of mean daily relative biases (i.e., inter-daily bias variability).

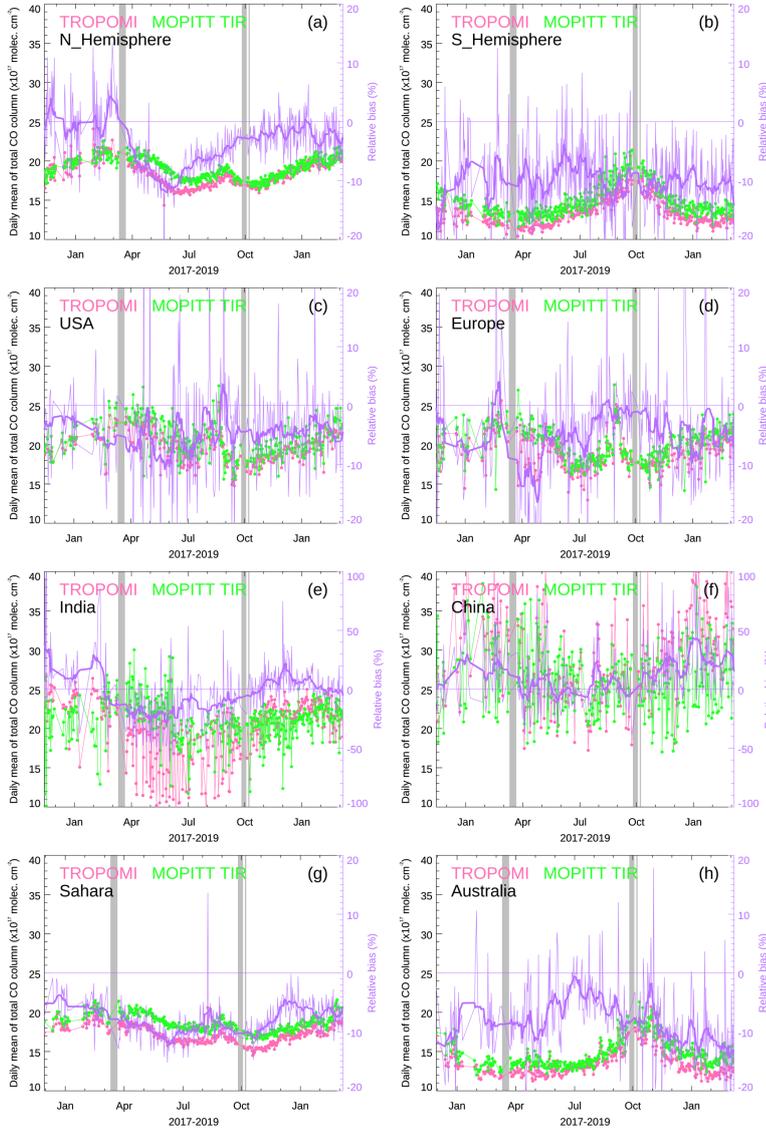


Figure S2. Comparison of non-located land retrievals from TROPOMI (pink) and MOPITT TIR (green) for each ROI analyzed. Filled circles show daily mean. Thin purple lines indicate daily relative bias between the two datasets, thick purple lines are a 11-day smoothed version with high-frequency variability removed. Gray bars show periods without MOPITT measurements because of hot calibrations (March and October 2018) or a safe mode maneuver (October-November 2018). Note that for the India and China ROIs the relative bias scale is different than for the other ROIs.

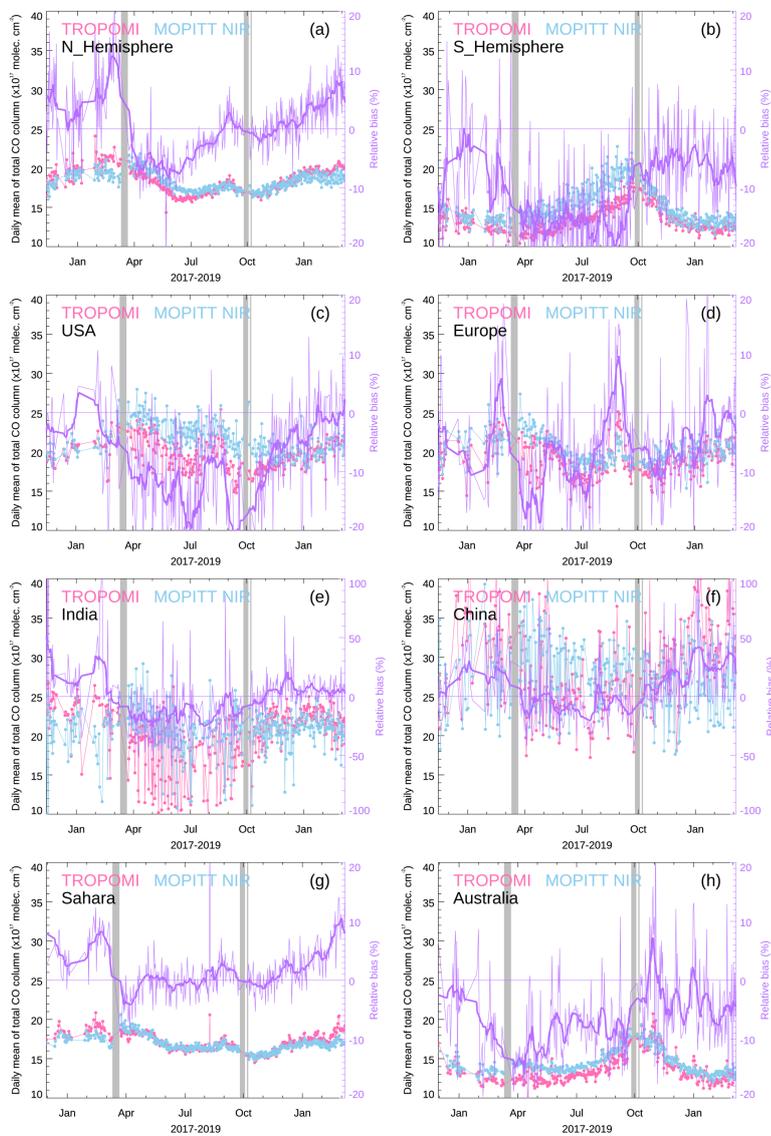


Figure S3. Comparison of non-located land retrievals from TROPOMI (pink) and MOPITT NIR (blue) for each ROI analyzed. See caption to Fig. S2 for details.

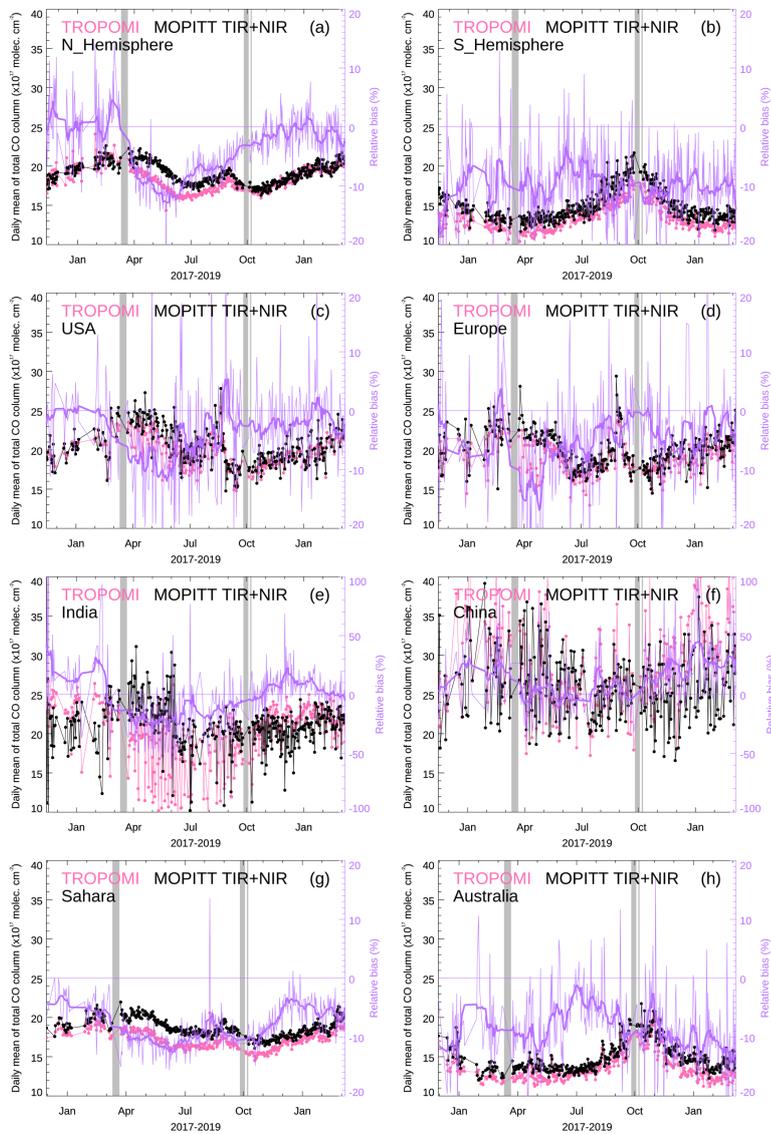


Figure S4. Comparison of non-located land retrievals from TROPOMI (pink) and MOPITT TIR+NIR (black) for each ROI analyzed. See caption to Fig. S2 for details.

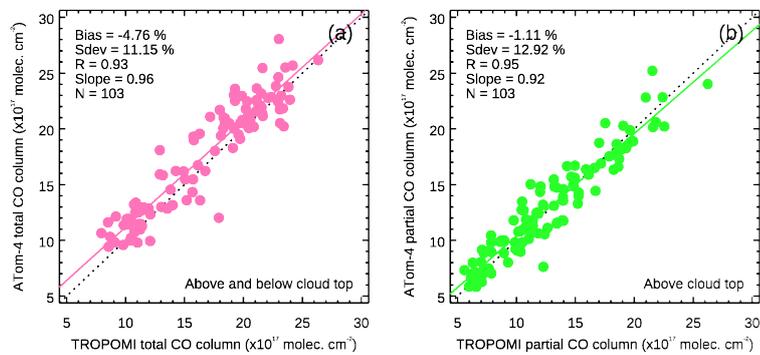


Figure S5. Comparison of collocated retrievals over bodies of water from TROPOMI and true ATom-4 (unsmoothed), performed for the period between 24 April and 21 May 2018. a) Total column retrievals (above and below cloud top), b) Partial column retrievals (above cloud top only).

Table S1. Statistics from non-colocated TROPOMI (T) versus MOPITT (M) retrievals over land for the period between 7 November 2017 and 10 March 2019. Relative bias in %. Column bias in units of 10^{17} molec. cm^{-2} .

| | | TROPOMI vs MOPITT _{TIR} | TROPOMI vs MOPITT _{NIR} | TROPOMI vs MOPITT _{TIR+NIR} |
|---------------|---------------------------|----------------------------------|----------------------------------|--------------------------------------|
| N Hemisphere | Relative Bias | -3.88 | 0.19 | -3.91 |
| | Column Bias | -0.74 | 0.04 | -0.75 |
| | Mean Daily Samples (T, M) | 151685, 15716 | 151685, 15855 | 151685, 14764 |
| S Hemisphere | Relative Bias | -10.07 | -10.60 | -9.96 |
| | Column Bias | -1.55 | -1.69 | -1.53 |
| | Mean Daily Samples (T, M) | 26551, 6287 | 26551, 6323 | 26551, 5992 |
| USA | Relative Bias | -4.73 | -8.77 | -3.58 |
| | Column Bias | -1.07 | -1.99 | -0.84 |
| | Mean Daily Samples (T, M) | 1559, 144 | 1559, 143 | 1564, 142 |
| Europe | Relative Bias | -4.65 | -5.78 | -4.77 |
| | Column Bias | -1.00 | -1.20 | -1.04 |
| | Mean Daily Samples (T, M) | 1680, 146 | 1680, 146 | 1680, 142 |
| India | Relative Bias | -2.91 | -1.21 | -2.20 |
| | Column Bias | -0.98 | -0.68 | -0.92 |
| | Mean Daily Samples (T, M) | 3831, 654 | 3822, 657 | 3852, 624 |
| China | Relative Bias | 11.73 | 6.88 | 12.73 |
| | Column Bias | 2.55 | 1.20 | 2.80 |
| | Mean Daily Samples (T, M) | 1395, 197 | 1392, 198 | 1395, 191 |
| Sahara | Relative Bias | -8.01 | 1.64 | -7.96 |
| | Column Bias | -1.50 | 0.27 | -1.50 |
| | Mean Daily Samples (T, M) | 50605, 4117 | 50605, 4143 | 50605, 3872 |
| Australia | Relative Bias | -7.98 | -6.26 | -8.35 |
| | Column Bias | -1.20 | -0.90 | -1.26 |
| | Mean Daily Samples (T, M) | 5918, 1311 | 5918, 1316 | 5918, 1263 |
| Mean all ROIs | Relative Bias | -3.81 | -2.99 | -3.50 |
| | Column Bias | -0.69 | -0.62 | -0.63 |
| | Mean Daily Samples (T, M) | 30403, 3572 | 30402, 3598 | 30406, 3374 |

Table S2. Comparison of colocated retrievals over bodies of water from TROPOMI and true ATom-4 (unsmoothed): Statistics from above/below cloud analysis performed for the period between 24 April and 21 May 2018. Relative bias in %. Column bias in units of 10^{17} molec. cm^{-2} .

| | | TROPOMI vs true ATom-4 Above & Below Cloud Top | TROPOMI vs true ATom-4 Above Cloud Top |
|------------------|--------------------------------|---|---|
| Atlantic/Pacific | Relative Bias \pm St. Dev. | -4.76 \pm 11.15 | -1.11 \pm 12.92 |
| | Column Bias \pm St. Dev. | -0.89 \pm 1.80 | -0.17 \pm 1.51 |
| | Number of Colocated Pairs | 103 | 103 |
| | Change in Relative Bias (p.p.) | | -3.65 |