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1.5 years of TROPOMI CO measurements: comparisons to MOPITT and ATom

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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 (regions of interest). Polluted ROIs include: south-eastern USA (USA; 35°N, 95°W to 40°N, 75°W), central Europe (Europe; 45°N, 0°E to 55°N, 15°E), northern half of the Indian Subcontinent (India; 20°N, 70°E to 30°N, 95°E), and north-eastern China (China; 30°N, 110°E to 40°N, 123°E). Clean ROIs are: northern Africa and Arabia (Sahara; 15°N, 20°W to 30°N, 50°E) and western Australia (Australia; 32°S, 112°E to 17°S, 138°E). Two additional ROIs represent most of the northern and southern (N and S) hemispheres (0°N to 60°N and 60°S to 0°N, respectively). 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*(TROPOMI-MOPITT)/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; 10-20 x 10^{17} molec. cm^{-2}) and highly polluted ROIs (India and China; up-to-40 x 10^{17} molec. cm^{-2}). They also show the expected differences between the two hemispheres: retrievals are, overall, lower in the S Hemisphere (10-20 x 10^{17} molec. cm^{-2} versus 16-22 x 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 MOPITT TIR over water: above/below cloud analysis

The goal of this analysis was to calculate the maximum error caused by the use of reference CO profiles in TROPOMI retrievals over water. To this effect, 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 reference profiles. This scenario would be most accurate in case of optically thick clouds. To quantify this error, we compared TROPOMI retrievals over bodies of water (total columns and their above cloud partial column components) to their colocated MOPITT TIR counterparts. 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, available in a 25-level vertical grid. Scaling factors produced in the TROPOMI retrieval process are not included in the TROPOMI product; we obtained those by dividing each reported TROPOMI total CO column retrieval by the total CO column of its colocated reference profile. Total and partial above cloud column values were also calculated for the colocated MOPITT TIR profiles interpolated to match the 25-level vertical grid of the reference profiles.

Figure S5 and Table S2 summarize results from our comparison of colocated TROPOMI and MOPITT TIR retrievals over bodies of water in the N Hemisphere ROI. The top panels in Fig. S5 illustrate a comparison between total column (above and below cloud top) and partial column (above cloud top) retrievals for a single day, 1 January 2018. Partial column values from TROPOMI and MOPITT are more strongly correlated in this particular date, as shown by a larger R (0.87 versus 0.73) and a smaller relative bias (2.77 versus 2.92 %). The bottom panels in Fig. S5 summarize similar daily results for the entire ~1.5-year period analyzed. Relative biases between TROPOMI and MOPITT TIR for total or partial columns are small (in the -2 to 11 % range, ~4 % on average) and follow the same temporal patterns; their differences (total column bias - partial column bias) range from -1.79 to 1.56 p.p. (percentage points), with a -0.53 p.p. mean. Standard deviation values are on average around 13-15 %.

Similar results for the S Hemisphere ROI are summarized in Fig. S6 and Table S2. Partial column values for 1 January 2018 (Fig. S6.b) have a larger R (0.84 versus 0.79) and appear more strongly correlated than their total column counterparts (Fig. S6.a). They, however, show a larger relative bias (2.16 versus 0.36 %). Similar results for the entire period analyzed (Fig. S6.c and .d) indicate that relative biases for either total or partial columns are similarly small, ranging from -5 to 7 (~3 % mean). Their differences are in the -3.62 to 0.97 p.p. range, with a -1.02 p.p. mean. Standard deviations are in the 18-21 % range.

Based on the difference in relative bias between the total (above and below cloud) and partial (above cloud) column analyses, we estimate that approximating TROPOMI CO below cloud top by scaled reference profiles results, on average, in a ~-0.78 p.p. error. This approach would be most accurate in the presence of optically thick clouds which would preclude TROPOMI sensitivity below cloud top.

S3 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. S7 and Table S3. Colocation criteria were same day acquisition and horizontal distance $\leq$ 50 km; each ATom-4 profile was paired with the closest valid TROPOMI retrieval that met the colocation criteria.

The goal of this analysis was to calculate the maximum error caused by the use of reference CO profiles in TROPOMI retrievals over water. To this effect, 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 this error, 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. 3 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. S7.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 S7.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.

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.

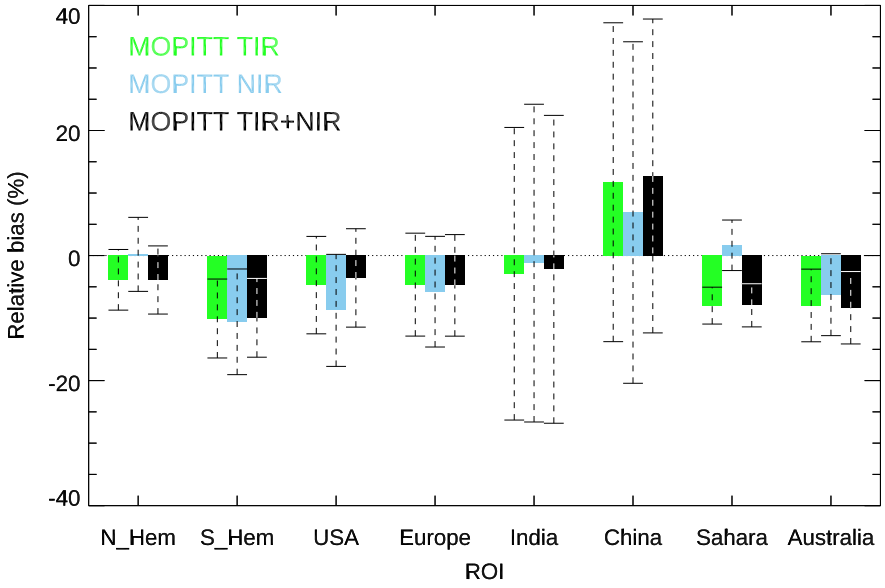

Figure S1. Summary of non-colocated land comparison results. Colored bars represent relative bias between TROPOMI and each of the three MOPITT products (TIR, NIR, and TIR+NIR). Dashed lines show ± 1 standard deviation of mean daily relative biases (i.e., inter-daily bias variability). Same as Fig. 13 in the main article.

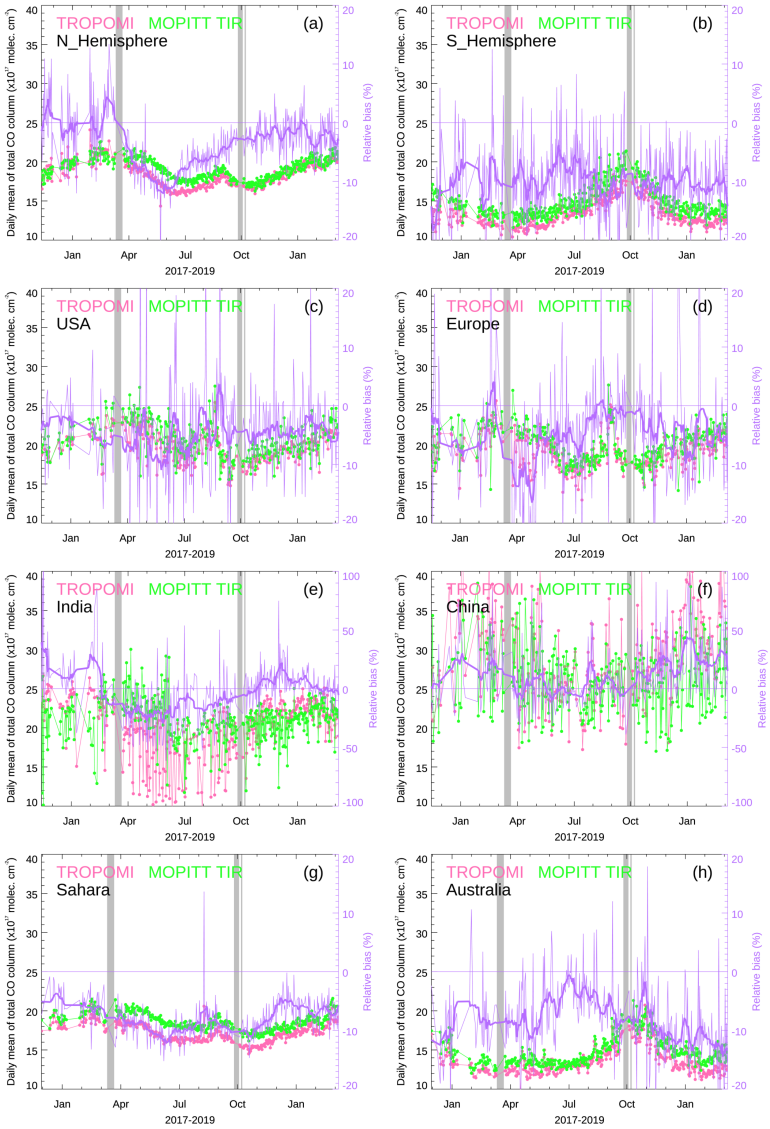

Figure S2. Comparison of non-colocated land retrievals from TROPOMI (pink) and MOPITT TIR (green) for each ROI analyzed. Filled circles show daily mean. Thin purple lines indicate daily relative bias between the two datasets, thick purple lines are a 11-day smoothed version with high-frequency variability 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.

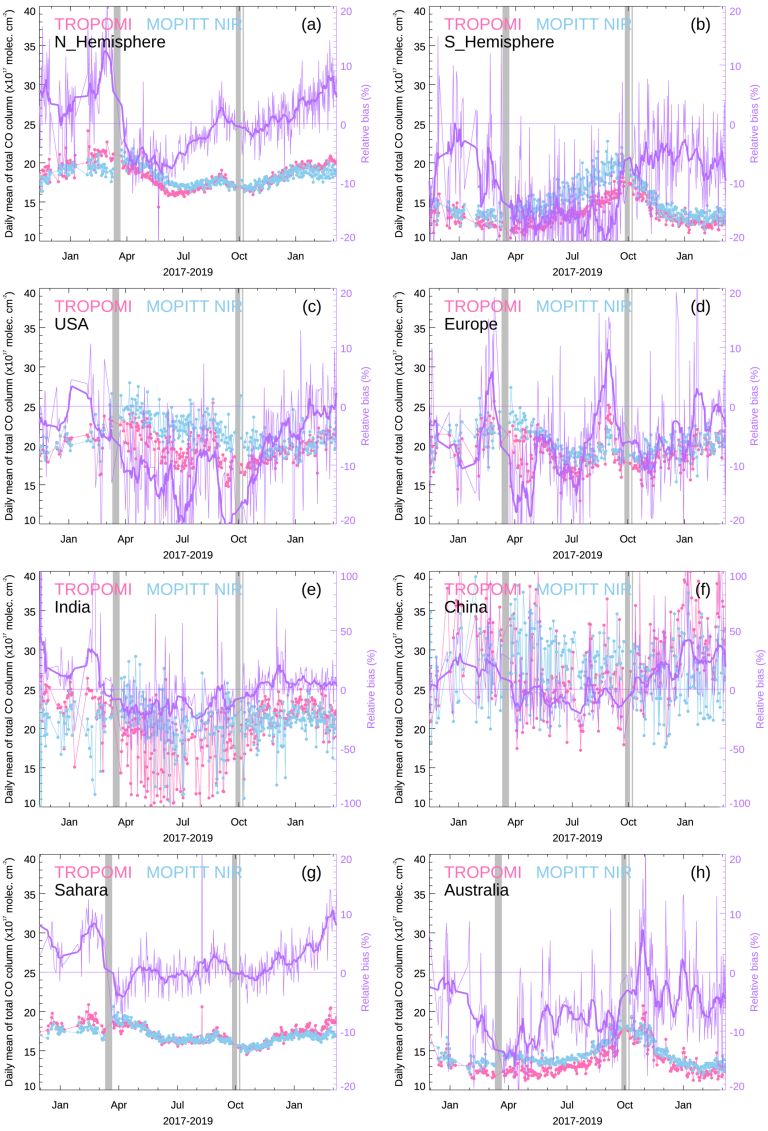

Figure S3. Comparison of non-colocated land retrievals from TROPOMI (pink) and MOPITT NIR (blue) for each ROI analyzed. See caption to Fig. S2 for details.

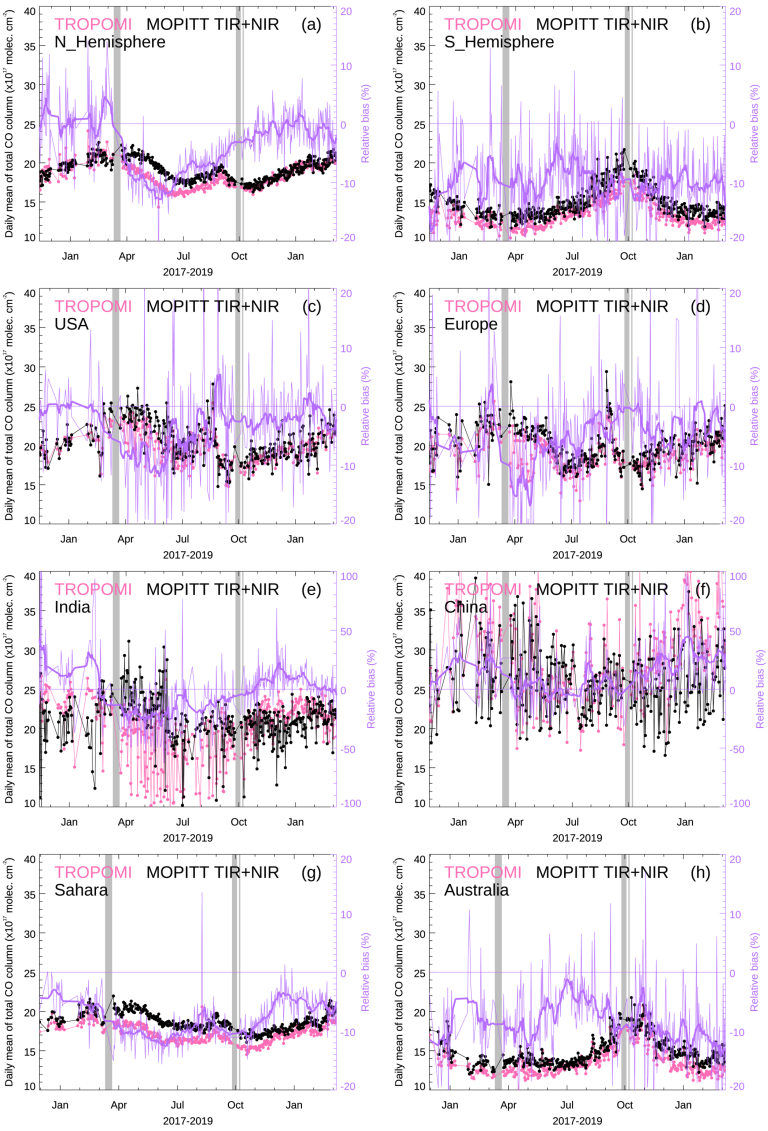

Figure S4. Comparison of non-colocated land retrievals from TROPOMI (pink) and MOPITT TIR+NIR (black) for each ROI analyzed. See caption to Fig. S2 for details.

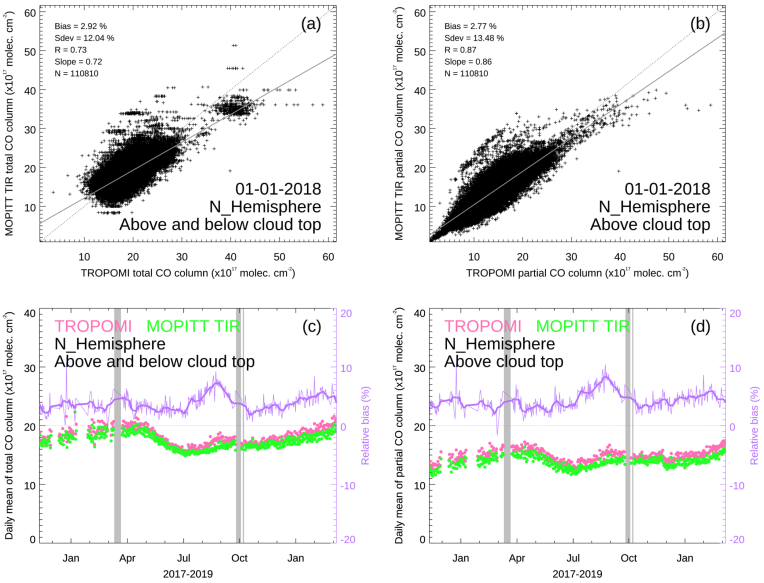

Figure S5. Comparison of colocated retrievals over bodies of water from TROPOMI and MOPITT TIR for the N Hemisphere ROI. a) Total CO column values (above and below cloud top) for a single day, 1 January 2018. b) Partial CO column values (above-cloud only) for the same day. c) Compilation of means and relative biases of total CO column values (above and below cloud top) from 7 November 2017 to 10 March 2019. d) Same for partial CO column values (above-cloud only).

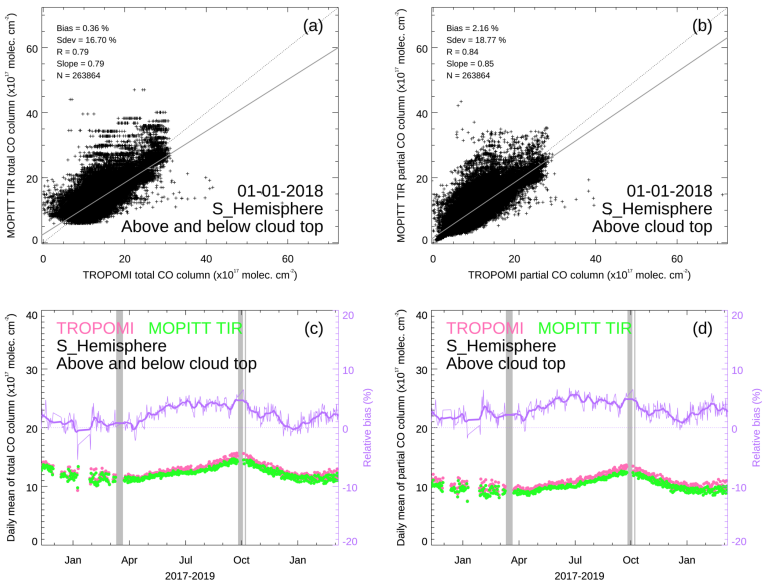

Figure S6. Comparison of colocated retrievals over bodies of water from TROPOMI and MOPITT TIR for the S Hemisphere ROI. a) Total CO column values (above and below cloud top) for a single day, 1 January 2018. b) Partial CO column values (above-cloud only) for the same day. c) Compilation of means and relative biases of total CO column values (above and below cloud top) from 7 November 2017 to 10 March 2019. d) Same for partial CO column values (above-cloud only).

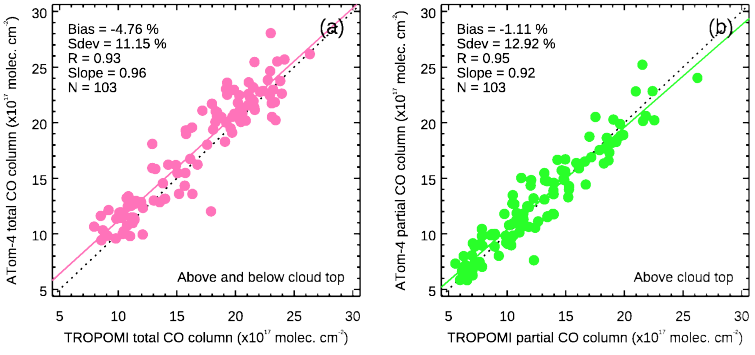

Figure S7. Comparison of colocated retrievals over bodies of water from TROPOMI and true ATom-4 (unsmoothed), performed for the period between 24 April and 21 May 2018. a) Total column retrievals (above and below cloud top), b) Partial column retrievals (above cloud top only).

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. Colocated TROPOMI versus MOPITT TIR CO retrievals over bodies of water: Statistics from above/below cloud analysis performed for the period between 7 November 2017 and 10 March 2019. Total column = above and below cloud top. Partial column = above cloud top. Relative bias and standard deviation in %. Column bias and standard deviation in units of 10^{17} molec. cm^{-2}.

		TROPOMI vs $MOPITT_{TIR}$ Total Column		TROPOMI vs $MOPITT_{TIR}$ Partial Column
N Hemisphere	Relative Bias±St. Dev.	3.82±13.27		4.35±14.72
	Column Bias±St. Dev.	0.53±2.35		0.48±2.04
	Mean Daily Colocated Pairs	127360		127360
	Change in Relative Bias (p.p.)		-0.53	
S Hemisphere	Relative Bias±St. Dev.	2.14±18.15		3.16±21.49
	Column Bias±St. Dev.	0.19±2.38		0.24±2.14
	Mean Daily Colocated Pairs	164935		164935
	Change in Relative Bias (p.p.)		-1.02	
Mean both Hemispheres	Relative Bias, St. Dev.	2.98±15.71		3.76±18.11
	Column Bias±St. Dev.	0.36±2.37		0.36±2.09
	Mean Daily Colocated Pairs	146148		146148
	Change in Relative Bias (p.p.)		-0.78	

Table S3. 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±St. Dev.	-4.76±11.15	-1.11±12.92
	Column Bias±St. Dev.	-0.89±1.80	-0.17±1.51
	Number of Colocated Pairs	103	103
	Change in Relative Bias (p.p.)	-3.65	