

Supplementary Information

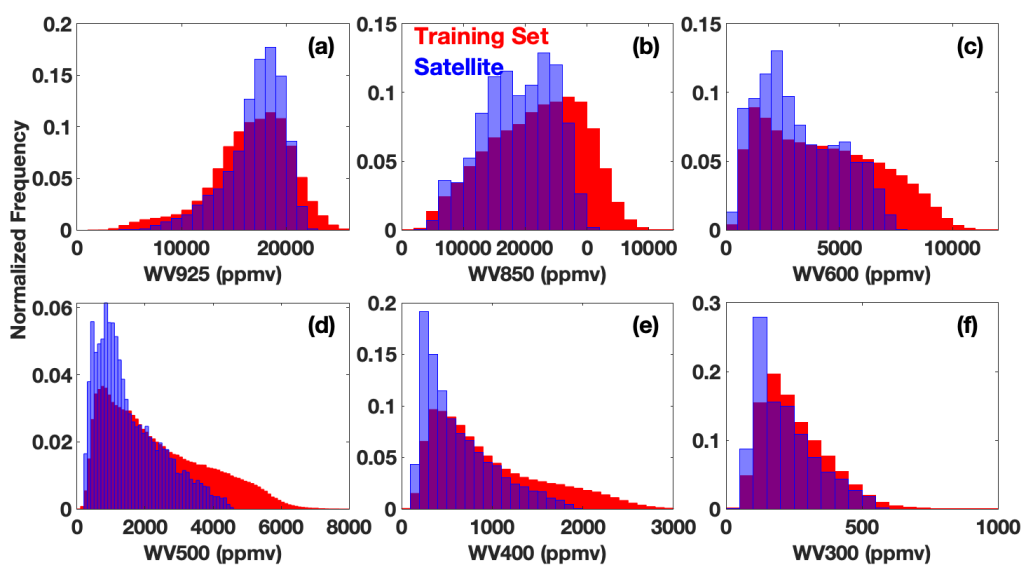


Figure S1: Comparison of the normalized distributions of the training dataset (red) for the February model and satellite observations of the indicated species for February 2017 (blue). Purple indicates regions of overlap.

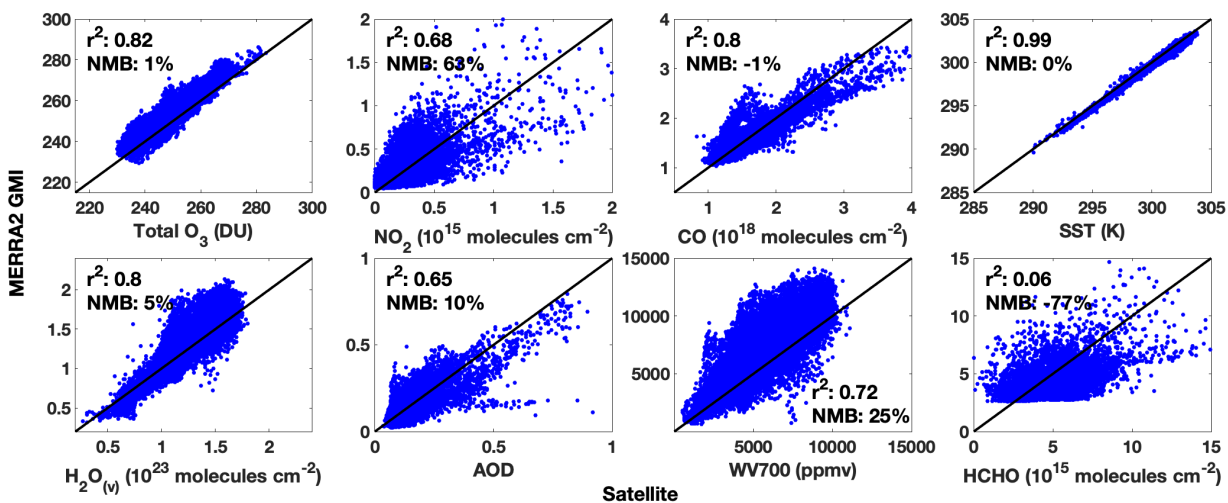


Figure S2: Regression of the indicated species from the MERRA2 GMI simulation against observations from the satellites listed in Table 1 for Feb. 2017. The r^2 of a linear least squares regression as well as the normalized mean bias is also indicated.

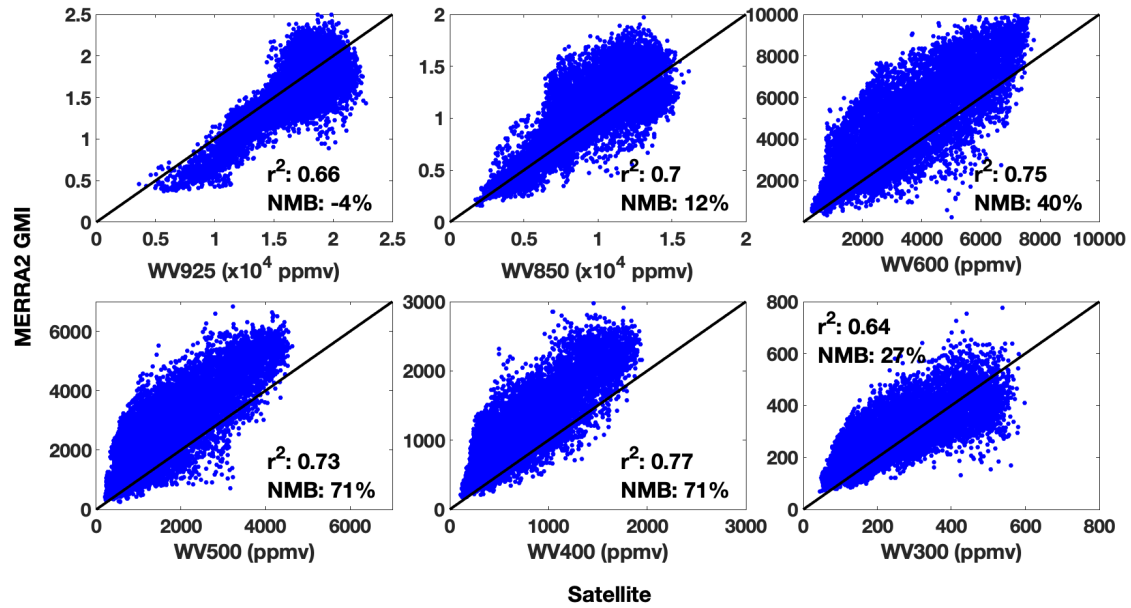


Figure S3: Same as Figure S2 except for the remaining water vapor layers.

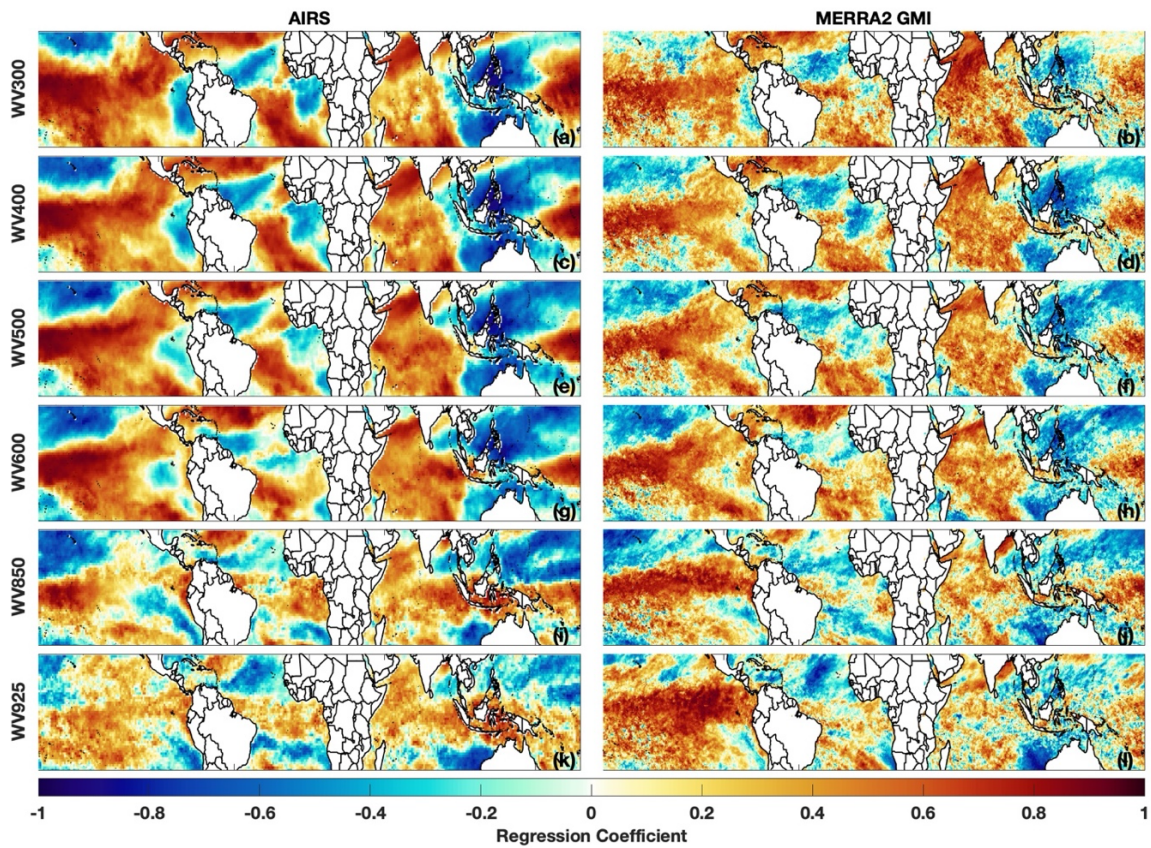


Figure S4: Same as Figure 2 except for the remaining water vapor layers. Regressions are for 2005 – 2019.

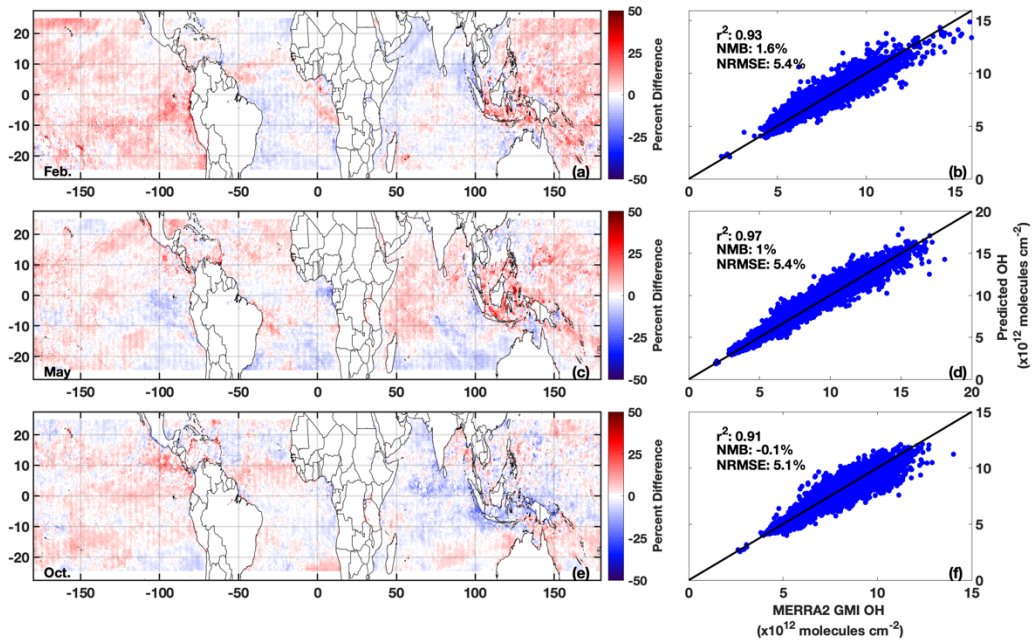


Figure S5: The percent difference between TCOH predicted from the GBRT model and from the MERRA2 GMI simulation for Feb. (a), May (c), and Oct. (e) 2017 is shown on the left. The regression of TCOH from MERRA2 GMI against that predicted from the GBRT model for those months is shown on the right. In addition, the r^2 of a linear least squares fit, the normalized mean bias, and the normalized root mean square error for each month is also indicated.

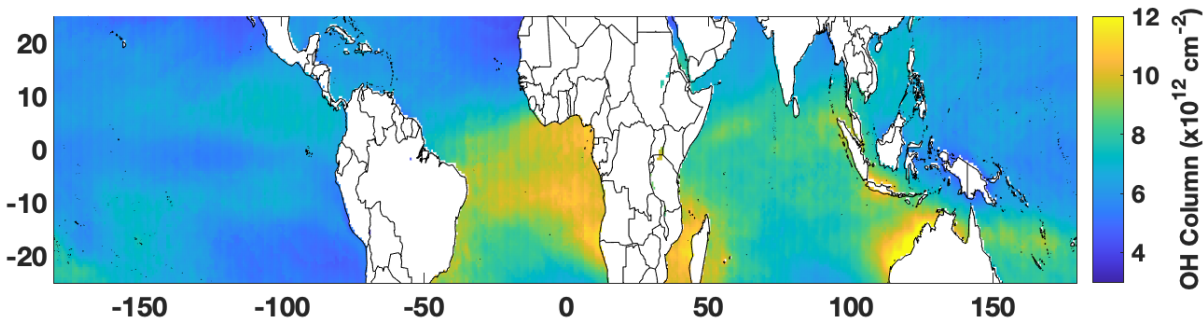


Figure S6: TCOH from MERRA2 GMI for Feb. 2017.

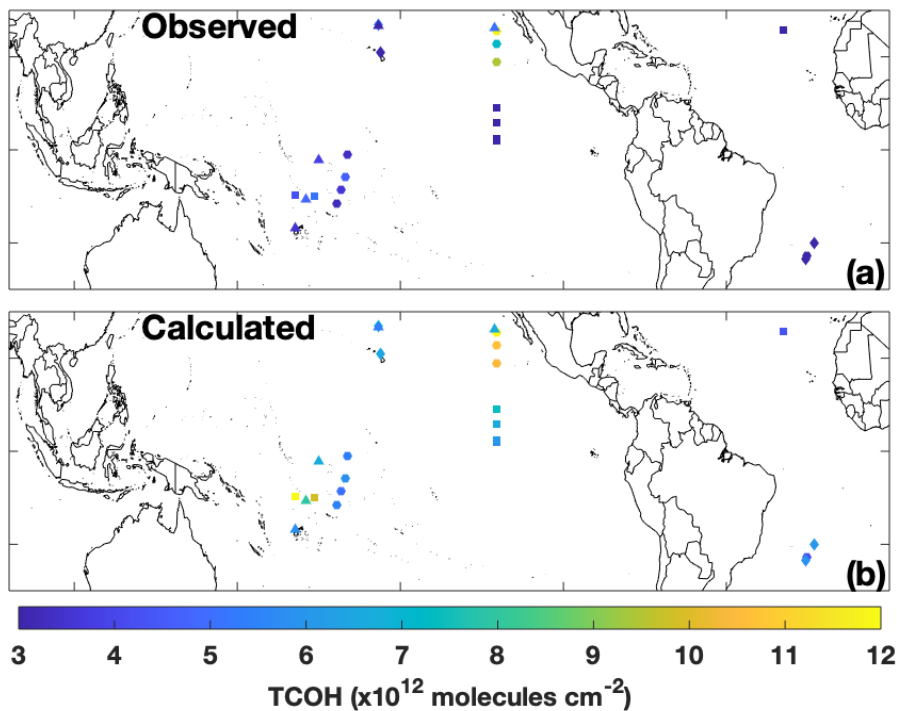


Figure S7: Distribution of observed (a) and GBRT calculated (b) TCOH columns for the four ATom deployments. Only columns with the required GBRT model inputs are shown. Individual ATom deployments are shown with circles (ATom 1), squares (ATom 2), diamonds (ATom 3), and triangles (ATom 4).

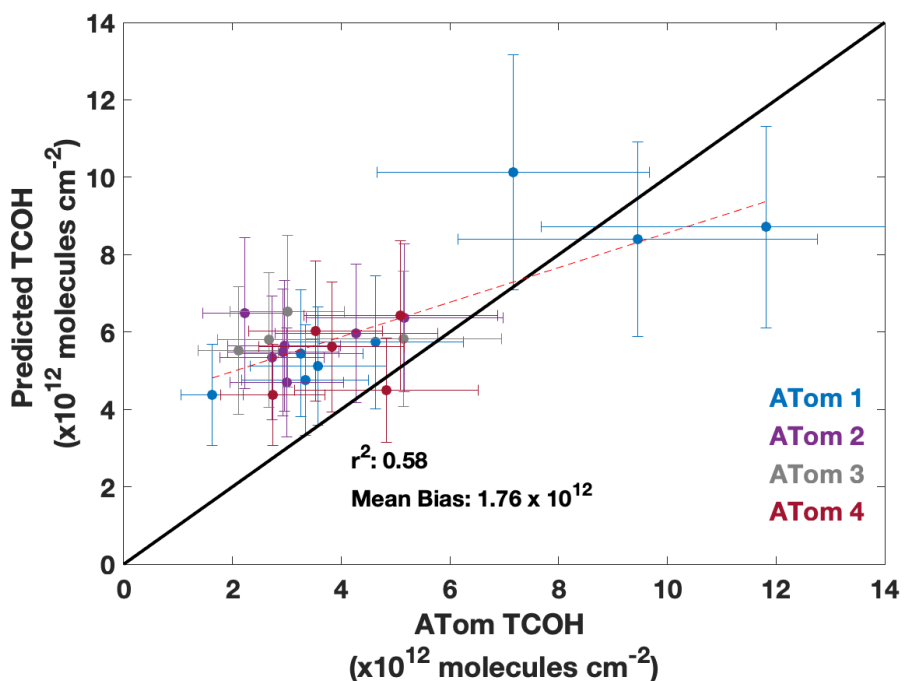


Figure S8: Same as Figure 5 except using NO_2 calculated with a box model. While there is significant overlap in the columns represented, because of data availability for box modeling, the columns evaluated here are a slightly different subsample of the ATom columns than those shown in Figure 5.

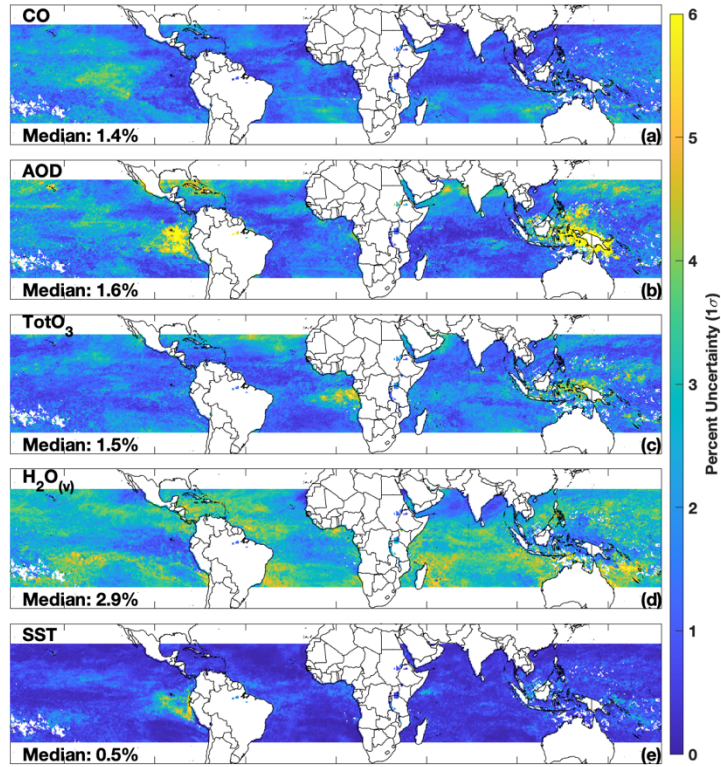


Figure S9: Normalized 1σ uncertainty in the satellite TCOH product due to uncertainties in the indicated retrievals for February 2017.

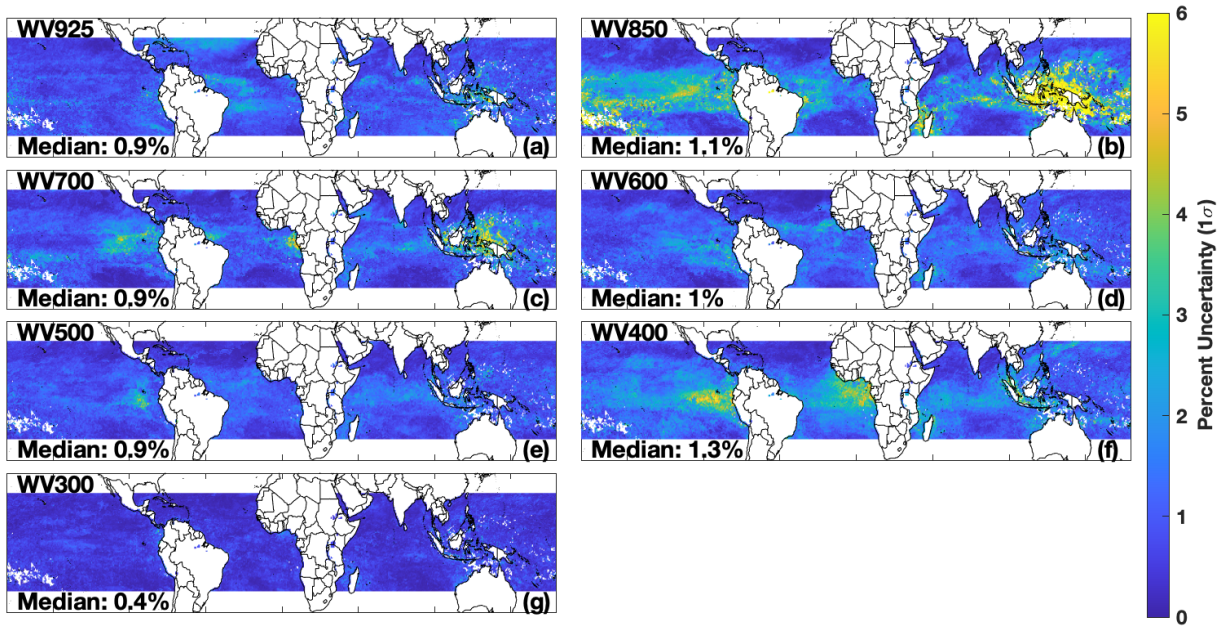


Figure S10: Same as Figure S9 except for the water vapor layers.

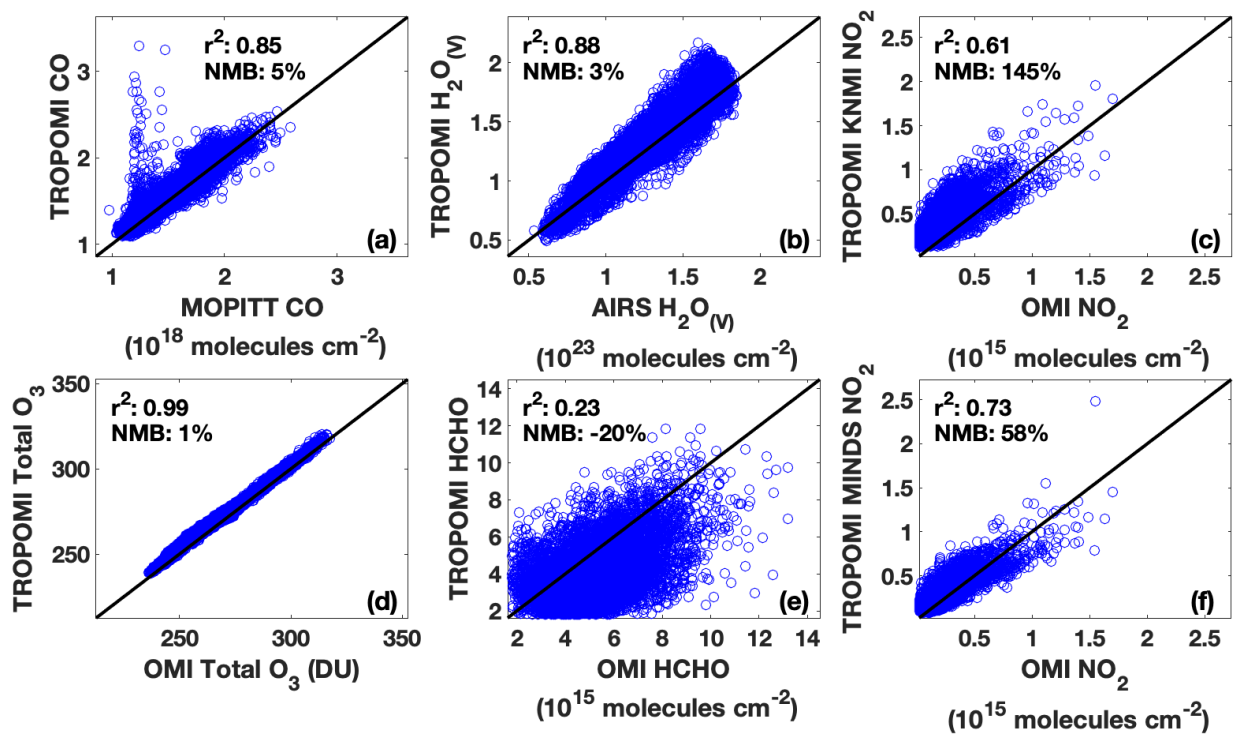


Figure S11: Regression of the indicated species from TROPOMI against retrievals from the satellites listed in Table 1 for May 2018. Both the KNMI (c) and MINDS (f) TROPOMI NO_2 retrievals are shown. We also indicate the r^2 of a linear least squares fit to the data as well as the normalized mean bias of TROPOMI with respect to the other satellite product.

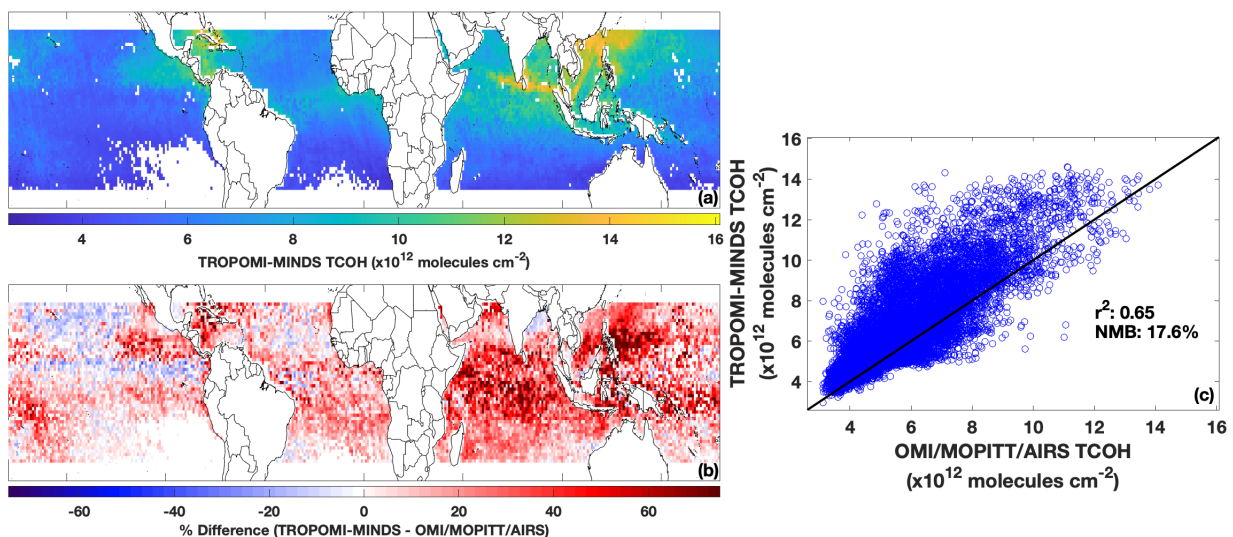


Figure S12: Same as Figure 7 except using the MINDS NO_2 retrieval for TROPOMI.

Table S1: Estimated uncertainty and source for each satellite retrieval.

Species	Estimated Uncertainty	Uncertainty Source and Notes
Total O ₃ Column	5%	Based on agreement with other observations (Labow et al., 2013)
Tropospheric NO ₂ Column	100%	Typical uncertainty of a single retrieval in the remote atmosphere (Choi et al., 2020)
CO Column	5.3%	Median reported uncertainty in L3 data file for study area
HCHO Column	100%	Median reported fit uncertainty of a single retrieval in data file for study area.
H ₂ O _(v) Column	12%	Median reported uncertainty in L3 data file for study area.
SST	0.35 K	Mean bias of dataset when compared to other SST records (Chin et al., 2017)
AOD at 550 nm	0.05 + 0.15τ	Reported value in (Sayer et al., 2014)
H ₂ O _(v) Layers	30 – 40%	Median reported uncertainty in L3 data file for study area.

References

Chin, T. M., Vazquez-Cuervo, J., and Armstrong, E. M.: A multi-scale high-resolution analysis of global sea surface temperature, *Remote Sensing of Environment*, 200, 154-169, <https://doi.org/10.1016/j.rse.2017.07.029>, 2017.

Choi, S., Lamsal, L. N., Follette-Cook, M., Joiner, J., Krotkov, N. A., Swartz, W. H., Pickering, K. E., Loughner, C. P., Appel, W., Pfister, G., Saide, P. E., Cohen, R. C., Weinheimer, A. J., and Herman, J. R.: Assessment of NO₂ observations during DISCOVER-AQ and KORUS-AQ field campaigns, *Atmos. Meas. Tech.*, 13, 2523-2546, 10.5194/amt-13-2523-2020, 2020.

Labow, G. J., McPeters, R. D., Bhartia, P. K., and Kramarova, N.: A comparison of 40 years of SBUV measurements of column ozone with data from the Dobson/Brewer network, *Journal of Geophysical Research: Atmospheres*, 118, 7370-7378, <https://doi.org/10.1002/jgrd.50503>, 2013.

Sayer, A. M., Munchak, L. A., Hsu, N. C., Levy, R. C., Bettenhausen, C., and Jeong, M. J.: MODIS Collection 6 aerosol products: Comparison between Aqua's e-Deep Blue, Dark Target, and “merged” data sets, and usage recommendations, *Journal of Geophysical Research: Atmospheres*, 119, 13,965-913,989, <https://doi.org/10.1002/2014JD022453>, 2014.