Review of "New Observations of Upper Tropospheric NO_2 from TROPOMI"

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Marais et al. "New Observations of Upper Tropospheric NO_2 from TROPOMI" presents an application of their cloud slicing technique previously used with OMI NO_2 data to TROPOMI NO_2 data. The high resolution of TROPOMI pixels allows the cloud slicing approach to be applied to much finer resolutions, comparable to typical global chemical transport model resolutions. This will be valuable to study the representation of upper tropospheric (UT) processes in these models since, as the authors say, in situ UT observations are sparse.

This manuscript should be published in AMT with minor revisions, mostly for clarity. I will address my comments by section below. I do deeply appreciate the authors providing their code in a proper repo!

Sect 2: Cloud slicing GEOS-Chem

- Line 132: "These are then screened to remove clusters with non-uniform GEOS-Chem stratospheric NO2.... Additional filtering is applied to clusters to remove extreme NO2 partial columns...." I don't see any comparable filters applied to TROPOMI data. Have you applied such filters, and if not, how does the GEOS-Chem test look without them? My concern is that having additional filters on the GEOS-Chem data not possible on the TROPOMI data limits its utility as a synthetic test.

- Line 148: Recommend citing Ziemke et al. 2001 here for the conversion from slope to mixing ratio.

- Line 151: Could you give more detail on what you mean by "Gaussian weighting individual estimates of cloud-sliced UT NO2 to the pressure centre"?

- Line 170-175: Do you have any hypothesis for why the size of the binning grid squares affects the magnitude of the cloud-sliced NO2, both here and with the TROPOMI data in Sect 4? This kind of systematic difference from resolution raises the question of which resolution is the most accurate, so an understanding of what drives that behavior would help.

- Line 202: "The cloud fraction retrieved from TROPOMI is an effective or radiometric cloud fraction that is systematically **less** than the physical cloud fraction from the model." Is "less" correct? Usually radiative cloud fractions are greater than geometric cloud fractions because they are weighted by the amount of light coming from the clear vs. cloudy parts of the scene, and since cloud tops are highly reflective, this boosts the radiance cloud fraction (Bucsela et al. 2013, p. 2611 below Eq. 5; Laughner et al. 2018 Fig. S3c).

- Line 210: "This is because the decrease in cloud top altitude leads to a larger increase in the vertical extent of partial columns above high-altitude clouds than those above low-altitude clouds leading to steeper regression slopes and larger UT NO2." This is a bit confusing—did you mean that a 1 km difference is a larger *relative* increase in column for high altitude clouds than low altitude, since there is less column above high altitude clouds?

Sect 3: TROPOMI evaluation

- Around line 300: are you comparing clear-sky and cloudy-sky TROPOMI VCDs calculated with the geometric-only AMF to Pandora, or just cloudy TROPOMI pixels where the cloud top height is similar to the elevation of these Pandoras? If the former, I'm not sure I understand the reason to include this comparison. The AMF used here relies on assumptions that may hold above clouds, but become more uncertain when the total column is considered.

- Line 328: "The underestimate in stratospheric NO2 variance would lead to an overestimate in the relative contribution of the stratosphere to the total column for small column densities and vice versa." A few points:

  - I'm not convinced by this. An underestimation in the variance doesn't indicate when it is under- or over- estimated. I think I understand what you are implying - that e.g. when the total column decreased by 50% the stratosphere *typically* changes by less than that, and if we assume that the stratosphere is proportional to the total column, then your argument holds. But this would be easier to demonstrate by comparing the stratospheric columns directly (e.g. scatter plot of TROPOMI strat. vs. Pandora strat.) rather than invoking variability.
  - Given that the stratosphere is assumed to be fairly uniform over moderate spatial scales, I'm not automatically convinced that even a 1x1 deg model significantly smoothes the stratosphere. Dirksen et al. (2011) for example showed that the OMI DOMINO stratospheric product captured a polar vortex event reasonably well by comparison with ground based data.
  - One final point I am concerned about is the difference in stratospheric dynamics over Mauna Loa and the rest of the tropics compared with the extratropics due to the Brewer-Dobson circulation. Have you considered the Pandora vs. TROPOMI stratospheric columns at, for example, Fort McKay, Fairbanks, Eureka, or Ny-Alesund to see if this same error in the stratosphere occurs in the mid- and high-latitudes?

- Line 341 (whole paragraph)

1. Is this really relevant to this paper? It seems focused on general TROPOMI validation, not cloud slicing.
2. If it is relevant, I'm not convinced that the stratospheric variance is the largest reason for these urban/rural differences. Underestimation of satellite NO_2 in urban areas and overestimation outside them is a common result of coarse a priori profiles that are too coarse (Russell et al., 2011) because the prior profile is a mix of urban & rural characteristics. Since Pandora is substantially less sensitive to the vertical profile of NO_2, it follows that comparisons with Pandora would illuminate this difference.

- Line 349: "After applying the stratospheric NO2 variance correction"— please explain how this correction is applied.
- Line 364: "We impose a modest threshold" - does this mean only TROPOMI columns >4e13 are used? TROPOMI columns <4e13 are set to 4e13? Something else?
- Line 375: "TROPOMI free tropospheric columns (red crosses in Figure 4)" - I am a little confused here. Has this section been discussing TROPOMI observations with just the free tropospheric (excluding boundary layer) or the total tropospheric columns? From Eqs. 1 & 2, there is no indication that the boundary layer has been removed, just the tropospheric VCD was recalculated with a geometry only AMF.

Sect 4: UT NO2 retrieval

- Line 416, paragraph: it is confusing to use "The standalone product version number changes from 01-01-07 to 01-01-08..." as the only transition from describing the FRESCO cloud product to describing the offline cloud product. This is aggravated because, earlier in the paper, FRESCO and ROCINN were used as the names of the products rather than "in the NO2 files" and "the standalone product". Also "These are FRESCO-S from the same data file as TROPOMI NO2 and the standalone offline (OFFL) cloud product," is ambiguous as to whether it means one product is FRESCO-S from the NO2 files and the other is the standalone product *or* FRESCO-S is the product in both the NO2 and standalone files. This paragraph could be made clearer.
- Moving the descriptions of the retrievals to the supplement (or removing altogether) could streamline the paper and focus on the cloud slicing aspects.
  - It seems that the most important difference between these products is whether they treat clouds as reflective surfaces or 3D objects. Focusing this paragraph on that, rather than the details of how these algorithms work, would likely help readability.
- Why is there so much more data in the northern high latitudes in Sept-Nov than Jun-Aug (Fig 5)? It seems like the greater summer illumination would make it easier to get data in Jun-Aug. According to Figs. 6 and S2, while there are more clouds in Sep-Nov than Jun-Aug, there are still 1.2 million cloudy pixels in Jun-Aug.

- Line 459: “Coincident gridsquares of the two data sets...” Since the previous sentence was talking about anthropogenic contamination, it isn’t immediately clear that the “two data sets” are the two cloud sliced ones. Perhaps a paragraph break would help?
- Line 518: “...as clusters of TROPOMI pixels in the midlatitudes and polar regions overcome the 140 hPa cloud top pressure range...” a bit awkwardly phrased, as ”overcome” somewhat implies more clouds exceeding the 140 hPa cutoff, which would imply fewer, not more clusters passing the filter.

Sect 5: Comparison between TROPOMI and OMI

- Line 531: Was 2005-2007 (rather than a multiyear period overlapping TROPOMI) chosen because that was before the row anomaly began degrading OMI retrievals? If so, would be good to say so to explain why this time period was chosen even though the impact of the 10+ year gap is difficult to quantify.
- Line 540: Conversely, Romps et al. (2014) suggest lightning will increase 12%/°C. Given that Schumann and Huntrieser published at the beginning of this 10 year gap, a more recent reference would strengthen the claim that lightning has not increased.
- Line 559: “The underestimate in variance in December-February in both products could reflect the need to account for seasonality in the stratospheric variance correction.” Cannot tell that the variance is underestimated in both from Fig. 8 since it is just comparing the two satellites’ values, without a truth metric. Please indicate how you know both are underestimated.
- Line 560: “UT NO2 obtained without applying correction factors to TROPOMI stratospheric and tropospheric columns (Figure S5) results in greater data density due to less variance in TROPOMI stratospheric columns, but the discrepancy with OMI is much greater.” This raises the question of why TROPOMI needed the 50% reduction in tropospheric columns. Since the TROPOMI:OMI slopes increase, that suggests (assuming that 2005-2007 and 2019-2020 are comparable) that TROPOMI has a multiplicative bias vs. OMI. Which OMI product (DOMINO v1/v2, NASA SP v1/v2/v3) are you using in this comparison? The QA4ECV product is probably the most similar to the TROPOMI retrieval; in the past, DOMINO has been clearly higher than NASA SP (e.g. Hains et al., 2010), so if the OMI 2005-2007 product uses NASA, then it’s possible this differences are retrieval driven (though that comparison is muddled since you’re not using the standard tropospheric AMF).

References

Bucsela, E. J., Krotkov, N. A., Celarier, E. A., Lamsal, L. N., Swartz, W. H., Bhartia, P. K., Boersma, K. F., Veefkind, J. P., Gleason, J. F., and Pickering, K. E.: A new stratospheric and tropospheric NO_2 retrieval algorithm for nadir-viewing satellite instruments: applications to OMI, Atmospheric Measurement Techniques, 6, 2607–2626, doi:10.5194/amt-6-2607-2013, URL https://amt.copernicus.org/articles/6/2607/2013/, 2013.

Dirksen, R. J., Boersma, K. F., Eskes, H. J., Ionov, D. V., Bucsela, E. J., Levelt, P. F., and Kelder, H. M.: Evaluation of stratospheric NO2 retrieved from the Ozone Monitoring Instrument: Intercomparison, diurnal cycle, and trending, Journal of Geophysical Research: Atmospheres, 116, doi:https://doi.org/10.1029/2010JD014943, URL https://agupubs.onlinelibrary.wiley.com/doi/abs/10.1029/2010JD014943, 2011.

Hains, J. C., Boersma, K. F., Kroon, M., Dirksen, R. J., Cohen, R. C., Perring, A. E., Bucsela, E., Volten, H., Swart, D. P. J., Richter, A., Wittrock, F., Schoenhardt, A., Wagner, T., Ibrahim, O. W., van Roozendael, M., Pinardi, G., Gleason, J. F., Veefkind, J. P., and Levelt, P.: Testing and improving OMI DOMINO tropospheric NO2 using observations from the DANDELIONS and INTEX-B validation campaigns, Journal of Geophysical Research: Atmospheres, 115, doi:https://doi.org/10.1029/2009JD012399, URL https://agupubs.onlinelibrary.wiley.com/doi/abs/10.1029/2009JD012399, 2010.

Laughner, J. L., Zhu, Q., and Cohen, R. C.: The Berkeley High Resolution Tropospheric NO_2 product, Earth System Science Data, 10, 2069–2095, doi:10.5194/essd-10-2069-2018, URL https://essd.copernicus.org/articles/10/2069/2018/, 2018.

McKenna, D. S., Konopka, P., Grooß, J.-U., Günther, G., Müller, R., Spang, R., Offermann, D., and Orsolini, Y.: A new Chemical Lagrangian Model of the Stratosphere (CLaMS) 1. Formulation of advection and mixing, Journal of Geophysical Research: Atmospheres, 107, ACH 15–1–ACH 15–15, doi:https://doi.org/10.1029/2000JD000114, URL https://agupubs.onlinelibrary.wiley.com/doi/abs/10.1029/2000JD000114, 2002.

Romps, D. M., Seeley, J. T., Vollaro, D., and Molinari, J.: Projected increase in lightning strikes in the United States due to global warming, Science, 346, 851–854, doi:10.1126/science.1259100, URL https://science.sciencemag.org/content/346/6211/851, 2014.

Russell, A. R., Perring, A. E., Valin, L. C., Bucsela, E. J., Browne, E. C., Wooldridge, P. J., and Cohen, R. C.: A high spatial resolution retrieval of NO_2 column densities from OMI: method and evaluation, Atmospheric Chemistry and Physics, 11, 8543–8554, doi:10.5194/acp-11-8543-2011, URL https://acp.copernicus.org/articles/11/8543/2011/, 2011.