

Interactive comment on “Improved aerosol correction for OMI tropospheric NO₂ retrieval over East Asia: constraint from CALIOP aerosol vertical profile” by Mengyao Liu et al.

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General comments

1. With this manuscript, the authors are delivering a new version of the POMINO product. This product is based on slant columns retrievals from DOMINO, uses nested GEOS-Chem simulation, and applies MODIS and CALIOP/CALIPSO data for AMF calculation, with each item presenting concerns for reporting it as a new version of the POMINO product. NO₂ slant column retrievals used here is version 2 product, which is reported to be erroneous. NO₂ profiles and other model dependent parameters are taken from two versions of nested model that switch from one resolution to another

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in 2013. They apply CALIOP/CALIPSO data over 2007-2015 to OMI data taken over 2004-present. MODIS AOD data are taken from two versions (collection 5.1 and collection 6 with a switch in 2013). There is enough ground to suggest that the product is erroneous, is not consistent over time, and should not be distributed to users as an updated version.

The latest study by Zara et al. (2018) shows that the uncertainty of SCD is $\sim 1.32 \times 10^{15}$ molec/cm² in DOMINO v2, which is reduced to $\sim 0.84 \times 10^{15}$ molec/cm² in the newly released QA4ECV product (used for comparison here). The SCD errors are thus much smaller than the errors introduced from AMF calculation over polluted regions like China (Boersma et al., 2011; Lin et al., 2015; Lorente et al., 2017). Also, any bias in the total SCD is mostly absorbed by the stratospheric separation step, and may not propagate into the tropospheric SCD (van Geffen et al., 2015). In other words, the tropospheric SCDs used in POMINO (from DOMINO v2) are well-established. We are considering moving to using tropospheric SCDs from QA4ECV, once these have been evaluated more thoroughly (something that this paper contributes to).

We had to choose two model versions according to the availability of meteorological data – GEOS-5 data on the 0.5×0.667 grid were replaced by GEOS-FP on the 0.25×0.3125 grid after May 2013). Despite the differences in horizontal resolution and driving meteorology, the two model versions have the same vertical coordinate and use the same schemes for advection, PBL mixing and convection, which is important for vertical distribution of NO₂ and aerosols. We consider the discontinuity in GEOS-Chem model resolution as a minor limitation for our NO₂ product for long-term trend studies.

We used the CALIOP/CALIPSO data over 2007-2015 according to data availability. Our correction of vertical profiles is on the basis of monthly climatology, thus applying the correction to other years is appropriate. Analogous approaches have been used in previous studies, for example, applying the 3-year average or 5-year average OMI surface albedo data to all years in DOMINO (Boersma et al., 2007; 2011), and applying

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4-year average (2004-2007) monthly mean NO₂ profile shapes derived from GMI CTM simulation to retrieve tropospheric NO₂ in NASA's SPv2 (Bucsela et al., 2013).

MODIS AOD data were taken from two versions in our POMINO product. We agree that using the same version of MODIS AOD data would be better. However, the difference in C5.1 and C6 is relatively small (C6 is smaller by 13.7% averaged over East China in 2012), compared to the difference between GEOS-Chem and C5.1 or between CEOS-Chem and C6. Our one-year test by using C5.1 versus C6 AOD (to correct model AOD) leads to 3.8% decrease in the retrieved NO₂ averaged over East China in 2012.

As suggested by the second reviewer, we have included the newest QA4ECV NO₂ product in the revised manuscript. Figure 9, Table 2 and Table 3 have been updated accordingly. QA4ECV is biased low in cases with high aerosol loading, but its R₂ with respect to MAX-DOAS is better than DOMINO v2. This additional comparison further strengthens the importance of aerosol correction in NO₂ retrieval over East Asia. Despite its various limitations discussed here, POMINO v1.1 is closer to MAX-DOAS than QA4ECV is, especially in hazy days, highlighting the capability of POMINO v1.1.

Given these above discussions, we have decided to not release POMINO v1.1 to users. Rather, we will eventually release POMINO v2, which will include MODIS C6 merged AOD and MCD43C2 C6 daily BRDF. The POMINO v1.1 will be used as an intermediate (and the most important) step between POMINO and POMINO v2. And this paper documents how improvement in aerosol vertical distribution affects the POMINO NO₂ product, such that all other factors are consistent between POMINO and POMINO v1.1. We have clarified this point in the revised abstract and conclusion.

2. To justify the improvement in the retrieved product, authors have used a small set of MAX-DOAS measurements. Improvements are justified based on improved correlation coefficient with the POMINO product. It appears from Figure 10 that the enhanced correlation might, in fact, be driven by changes in \hat{L}_{ij6} data points only with very large

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(>100 x 10¹⁵ molec cm⁻²) values. In many instances (for columns < 100 x 10¹⁵ molec cm²), the agreement between OMI and MAX-DOAS appears to be better for DOMINO. Author should use different means of validation, larger set of validation datasets, and various statistical methods to assess the products.

The high values represent very polluted cases that our algorithm intends to capture. Excluding these polluted cases would lead to a substantial sampling bias over polluted regions. We have made the distinction between hazy cases and less hazy situations. The latter are more representative for retrievals over the US and Europe. In those cases, QA4ECV may perform better and POMINO is more likely to be biased high (Table 3). We would definitely prefer to have a larger set of MAX-DOAS NO₂ data. Unfortunately, very few high-quality MAX-DOAS measurements are available over China. We have made efforts to get data from multiple sites to enhance the spatial representativeness. Our criteria to select MAX-DOAS data and OMI data mainly follow Wang et al., (2017b) and Lin et al., (2014), who have already discussed the influence from various statistical methods. We have included a statement in the end of Sect. 6 that "Further research may use additional MAX-DOAS datasets to evaluate the satellite products more systematically."

3. The whole discussion about processing (filtering, regridding) and comparison of CALIPSO data is distracting and unnecessary. These could be completely removed, shortened, or moved to the Appendix/Supplementary section. Also, data processing is largely subjective. Why not use more mature data assimilation technique instead?

We have revised the manuscript accordingly. The discussion on the treatment of CALIOP data has been moved to Appendix B. Data assimilation is subject to the very limited availability of CALIOP data. It is also computationally prohibitive for our application here (multiple years over a large domain on a high-resolution grid).

Specific comments

1. Page 9, line 225: This statement may not be true. Please, replace "will not" to "may

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not”.

Changed.

2. Page 9, line 227-231: Please be more specific on AMF calculation. What wavelength range is used for AMF for POMINO/DOMINO? I assume this is more important than the difference between online and look-up table approach.

Changed. The wavelength is 438 nm in both DOMINO and POMINO. The dependence of AMF on the wavelength is weak (actually 0.25%/nm, Boersma et al. (2018)). Other details of AMF calculations can be found in Lin et al. (2014b, 2015).

3. Page 9, line 228: This paper is all about POMINO and DOMINO. Please, say “DOMINO” instead of “in most retrieval algorithms”.

As far as we know, most algorithms use look-up tables, including but not limited to NASA’s SP product, DOMINO, and others participating the QA4ECV project.

4. Page 10, line 237: What are those “Other aspects”? Please, list them.

Changed.

5. Page 10, lines 237-239: This statement is likely misleading as look-up table may have been used in certain aspect of your calculation. Please, remove “without use of look-up tables”.

Changed.

6. Page 10, lines 239-244, 257-259: See my general comment. The same product cannot use simulated fields from two different models. The retrievals should be based on single model.

See response to general comment.

7. Page 13, lines 314-316: How does the se of CALIPSO constraints affect cloud pressure, cloud fraction, and radiative cloud fraction? Please include relevant results

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and discussions. The detailed results can be found in Sect. 4.

8. Page 13, lines 321-325: Please, clarify this statement.

Clarified.

9. Page 14, line 360: What is the justification of 2-hour averaging of MAX-DOAS? Why do you expect instantaneous OMI measurements compare well with MAX-DOAS averaged over 2-hours? Is this exercise described in the following sentences motivated to show only good results?

As already clarified in manuscript, we used the criteria based on several previous studies (Lin et al., 2014; Wang et al., 2015, 2017b). These previous papers have already discussed the most appropriate criteria to balance data coverage, passing time, spatial domain around the pixel center, etc.

10. Page 15, lines 366-367: “to some degree” is redundant.

Changed.

11. Page 15, lines 374-375: Why is this necessary? How do cloud and haze differ for their impact on measurement sensitivity of OMI?

As emphasized in the manuscript, we wanted to separate the hazy days from cloudy days. Some days are cloud-free but hazy (with heavy NO₂ pollution as well). These days were filtered out in DOMINO and QA4ECV through the criteria on cloud radiance fraction. By comparison, our algorithm was able to retain these days and avoid sampling bias (by missing polluted days) while preserving the overall accuracy of NO₂ product. As explained in the manuscript, neither the OMI cloud product nor the MODIS cloud product is able to provide the true cloud fraction, so we used the meteorological monitoring stations and the MODIS RGB product to manually check whether a day is cloudy or hazy.

12. Page 16, line 395: Please, add citations for this statement.

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Changed.

13. Page 17, line 418: How does the emission strength affect the height of peak extinction?

The effect of emission strength on aerosol vertical profiles is season and location dependent. For the case here (Figure 4), emissions over Eastern China are higher in winter, in which season the atmosphere is more stagnant vertically. This means that more aerosols are concentrated near the surface, thus decreasing the height of peak extinction.

14. Page 19, lines 470-472: The spatial correlations suggest that GEOS-Chem performs very poorly in simulating aerosol fields. Why do you still use GEOS-Chem? Could not you just use CALIPSO-based aerosol information?

GEOS-Chem provides daily and spatially resolved information, which is what is needed by the satellite retrieval. CALIOP, in contrast, has poor temporal and spatial coverage, preventing fully CALIOP-based aerosol profile information to be used to retrieve the NO₂ product. The spatial correlation between GEOS-Chem and CALIOP is not as good as their temporal correlation. We thus used CALIOP for monthly climatological corrections, while retaining the GEOS-Chem simulated day-to-day variability.

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