Authors' response to comments from Anonymous Referee #1

General comments:

The paper "Global retrieval of TROPOMI tropospheric HCHO and NO₂ columns with improved consistency based on updated Peking University OMI NO₂ algorithm", by Zhang and co-authors, presents an updated version of the POMINO algorithm for HCHO and NO₂ and its structural uncertainty in the AMF calculation. The paper is well structured. The topic fits in the scope of AMT.

We sincerely thank the Referee #1 for reviewing our paper and providing constructive comments for improvement. Responses to the comments are provided below.

Specific comments:

One concern is the application of aerosol correction on FRESCO parameters. The FRESCO CF has been recalculated and corrected using aerosol information in UV and VIS, but the original O₂-A band may be less affected by aerosol than UV/VIS, possibly introducing an overcorrecting issue. Such overcorrecting can be amplified by using the un-corrected CP.

Thank you very much for this comment. A similar comment was raised by Referee #3.

We agree that the aerosol overcorrection issue occurs for partly cloudy pixels because we only recalculate the cloud fraction with explicit aerosol corrections, but use the cloud top pressure from FRESCO-S product which already implicitly includes aerosols. Liu et al. (2020) quantified such overcorrection issue for aerosols by conducting a sensitivity study using a "semi-explicit" aerosol correction approach. In this approach, aerosol optical effects are explicitly corrected for clear-sky AMFs, but are excluded for the cloudy-sky portion of partly cloudy pixels. Results show that NO₂ differences due to the aerosol correction choice for cloudy-sky AMFs vary from 3.1% to 11.2% over East Asia in July 2018, depending on the NO₂ pollution level (Section 3.3 in Liu et al., 2020). It should be noted that the FRESCO-S cloud top pressure data stored in the v1.2–v1.3 NO₂ data product, as used by Liu et al. (2020), are reported with a high bias over scenes with low cloud fractions and/or a considerable aerosol load. An improved version based on the FRESCO-wide approach is applied in v1.4 and subsequent NO₂ products, which was proven to be more realistic compared with the old version (Van Geffen et al., 2022a, b).

Using the updated FRESCO-S cloud top pressure data stored in the RPRO v2.4.0 NO₂ product, we attempt to estimate the impact of the aerosol overcorrection issue without conducting a sensitivity study. Given the fact that, in the retrieval algorithm, the cloud is assumed to be an optically thick Lambertian reflector with a high albedo of 0.8, it is reasonable to assume that the impact of this overcorrection becomes non-negligible when the FRESCO-S cloud top pressure is too high, meaning the cloud is very close to the surface and therefore vertically mixed with aerosols and trace gases; while for pixels where the cloud is higher than the trace gases, the aerosol correction should have little influence on the cloudy-sky AMFs. Based on this strategy, we selected all valid pixels where the difference between the surface pressure and the FRESCO-S cloud top pressure is equal to 100 hPa or less in July 2021 and January 2022. In such case, we can mitigate the potential aerosol overcorrection by using aerosol-corrected clear-sky AMFs instead of aerosol-corrected total AMFs.

The comparison result in Figure S6 (shown below) shows that the normalized mean bias (NMB) is around

14% on average for HCHO retrievals (~16% for clean pixels with HCHO column $\leq 10 \times 10^{15}$ molec.cm⁻², and ~8% for polluted pixels with HCHO column > 10 × 10¹⁵ molec.cm⁻²), and around 8% on average for NO₂ retrievals. The NO₂ results are also qualitatively in line with those in Liu et al. (2020). Therefore, we tentatively estimate the uncertainty due to the aerosol overcorrection to be in the range from 10% to 15% for HCHO and 10% for NO₂.

In line 397-423 in the revised manuscript, we added:

"One issue existing in the process of cloud correction in the POMINO retrieval is that only the cloud fraction is re-calculated with explicit aerosol corrections, while the cloud top pressure is taken from the external dataset, i.e., the FRESCO-S cloud product, in which the aerosols are implicitly accounted for. As a result, this step introduces presumably an aerosol overcorrection issue in the cloud top pressures of partly cloudy pixels, and therefore brings in additional uncertainties in the AMF calculations. Lin et al. (2015) reported that excluding aerosols leads to an increase of O₂-O₂-based cloud top pressures (from 700–900 hPa to 750–950 hPa) over eastern China, but it is difficult to clarify the mechanism due to its complexity (Lin et al., 2014). Currently there is no direct way to estimate the effect of aerosol correction on the FRESCO-S cloud height retrieval without doing O₂ A-band cloud retrieval tests, which is beyond the scope of this study. However, below we give an estimation of the uncertainty in POMINO HCHO and NO₂ vertical columns caused by this issue.

Given the fact that, in the retrieval algorithm, the cloud is assumed to be an optically thick Lambertian reflector with a high albedo of 0.8, the cloudy-sky AMF (and hence tropospheric AMF) is very sensitive to the accuracy of the cloud height when the cloud is low and vertically mixed with the aerosols and trace gases. In these cases, we can assume that the retrieved cloud height is primarily influenced by aerosols (Van Geffen et al., 2022a), therefore the aerosol overcorrection issue becomes non-negligible. Focusing on valid pixels for which the difference between the surface pressure and the FRESCO-S cloud top pressure is equal to 100 hPa or less (~17.5% and ~19.9% of total pixels in July 2021 and January 2022, respectively), the aerosol overcorrection uncertainty can be roughly estimated from the difference of HCHO and NO₂ vertical columns retrieved using either aerosol-corrected clear-sky AMFs (aerosol correction applied; cloud correction not applied) or aerosol-corrected total AMFs (both aerosol and cloud corrections applied). Based on the results shown in Figure S6, we tentatively estimate the uncertainty to be in the range from 10% to 15% for HCHO, and within 10% for NO₂. The estimated NO₂ uncertainty level is also supported by the sensitivity test results in Liu et al. (2020). They implemented a "semiexplicit" aerosol correction approach, in which aerosol optical effects are explicitly corrected for clearsky AMFs, but are excluded for the cloudy-sky portion of partly cloudy pixels, and found the NO₂ differences due to the aerosol correction choice for cloudy-sky AMFs vary from 3.1% to 11.2% over eastern China in July 2018. The tentatively estimated uncertainty range above is comparable to or less than that from other ancillary parameters (Sect. 5), and only needs to be taken into account for partly cloudy pixels with low clouds."

In the revised Supplement, we added:

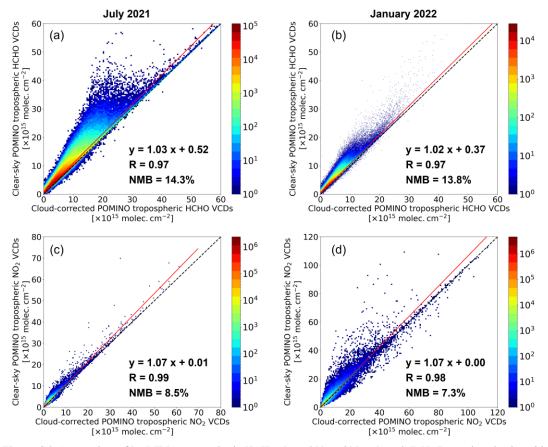


Figure S6. Scatterplots of POMINO tropospheric HCHO (**a** and **b**) and NO₂ (**c** and **d**) VCDs retrieved using either aerosol-corrected and cloud-corrected total AMF (x-axis) or aerosol-corrected clear-sky AMF (y-axis), from all pixels where the difference between surface pressure and FRESCO-S cloud top pressure is equal to 100 hPa or less. The left column shows the results for July 2021, and the right column for January 2022. The slope, offset and correlation from a linear regression using the robust Theil-Sen estimator and normalized mean bias (NMB) are given in each panel and plotted as the red line. The black dashed line is the 1:1 line.

Table 1 is a bit heavy, and I recommend to remove redundant information, such as "daily" in a priori profiles. One typo in POMINO HCHO CF is 340 instead of 440.

Thank you for the recommendation. We have simplified Table 1 in the revised manuscript.

Regarding the cloud fraction in POMINO HCHO retrievals, we use values re-calculated at 440 nm (in the NO₂ retrieval) instead of re-calculating them at 340 nm, because (1) the cloud fraction derived at 440 nm is expected to be more reliable than at 340 nm due to the larger noise in the UV band; (2) we want to perform fully consistent cloud corrections in both POMINO HCHO and NO₂ retrievals. We have changed the description to "CF and CP: same as POMINO NO₂" in Table 1 to make it more clear in the revised manuscript.

What is the meaning of "VCD from QA4ECV" for Mohali in Table 2?

It means a retrieval strategy where MAX-DOAS vertical columns are calculated using tropospheric AMFs based on climatological profiles of both trace gas and aerosol loads, as developed during the QA4ECV project (<u>http://uv-</u>

<u>vis.aeronomie.be/groundbased/QA4ECV_MAXDOAS/QA4ECV_MAXDOAS_readme_website.pdf</u>). These data are more accurate than the simple geometric approximation strategy as used in previous studies (De Smedt et al., 2021). We have updated the description and added the reference in the revised manuscript.

Please enlarge the font size of xaxis label in Figures 10 and 11.

Done.

References:

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- Liu, M., Lin, J., Kong, H., Boersma, K. F., Eskes, H., Kanaya, Y., He, Q., Tian, X., Qin, K., Xie, P., Spurr, R., Ni, R., Yan, Y., Weng, H., and Wang, J.: A new TROPOMI product for tropospheric NO₂ columns over East Asia with explicit aerosol corrections, Atmospheric Meas. Tech., 13, 4247–4259, https://doi.org/10.5194/amt-13-4247-2020, 2020.
- Van Geffen, J., Eskes, H., Compernolle, S., Pinardi, G., Verhoelst, T., Lambert, J.-C., Sneep, M., ter Linden, M., Ludewig, A., Boersma, K. F., and Veefkind, J. P.: Sentinel-5P TROPOMI NO₂ retrieval: impact of version v2.2 improvements and comparisons with OMI and ground-based data, Atmospheric Meas. Tech., 15, 2037–2060, https://doi.org/10.5194/amt-15-2037-2022, 2022a.
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