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Comment on amt-2022-51

Anonymous Referee #2

Referee comment on "Highly resolved mapping of NO₂ vertical column densities from GeoTASO measurements over a megacity and industrial area during the KORUS-AQ campaign" by Gyo-Hwang Choo et al., Atmos. Meas. Tech. Discuss.,
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Referee Comment on Atmos. Meas. Tech. Discuss. [preprint],
<https://doi.org/10.5194/amt-2022-51>, in review, 2022.

Highly resolved mapping of NO₂ vertical column densities from GeoTASO measurements over a megacity and industrial area during the KORUS-AQ campaign by Gyo-Hwang Choo et al.

The article presents airborne and ground-based measurements during the KORUS-AQ field study in South Korea, focussing on NO₂ column density measurements above two metropolitan regions and one industrial region in May/June 2016. Observations are performed on several days, in the morning and/or in the afternoon. For the airborne measurements, the GeoTASO instrument is applied. Its specifications, NO₂ retrieval details as well as information on the AMF determination are given. Results above the different probed areas are presented and discussed. The probed areas have sparse coverage with trace gas measurements otherwise. Error analysis is included as well as a short comparison with ground-based and OMI observations. Similar to other airborne DOAS sensors, the GeoTASO instrument constitutes a valuable tool for tropospheric trace gas monitoring and mapping, also above areas that are less well accessible. The presented measurements are relevant for a better understanding of the spatial variation of NO₂ above South Korean polluted sites that are less well monitored otherwise.

General Comments

The overall structure of the article is well understandable. Although there are some typing

and grammar mistakes, the text is well readable.

The GeoTASO instrument is introduced and further publications are cited where details of interest can be found. The different assumptions necessary for the conversion from detected slant column densities to meaningful tropospheric vertical column densities are explained.

However, some relevant aspects could be treated with more caution. The respective error analysis could point out more clearly the limitations. While the GeoTASO good spatial resolution is emphasized, this is not really shown, but all data is binned to a 0.01° grid. The authors rightly consider the comparison between GeoTASO and OMI relevant and mention this in the abstract and conclusions. Therefore, a dedicated figure would certainly support this analysis.

After consideration of the comments and suggestions below, and after submitting a revised version, I recommend publication of this article in AMT.

Major Comments

-Spatial resolution-

The article presents observations with different spatial resolution (airborne as compared to satellite), and the usefulness of good spatial resolution is emphasized. Therefore, two aspects should be treated with more care. Firstly, the correct resolution information should be stated. Please update either the figure or correct the caption text of Fig. 1 (the grid for the OMI data is 0.25° here). More importantly, the best possible presentation of the GeoTASO spatial resolution should be aimed for. In all figures of the publication, GeoTASO measurements are gridded to a 0.01° grid, corresponding to a side length on the order of 1km, while spatial resolution of the instrument is 250m. It is confusing, when 250m resolution is announced and emphasized but never shown. If it is not possible or not aimed at to use the mentioned best resolution, it should be explained in the text why (is the signal-to-noise ratio otherwise too bad? is there another reason?).

-Treatment of the spectral reflectance-

Spatial variation of the ground albedo is large, e.g., darker vegetation in parks as compared to paved areas of parking areas, flat roof tops, or similar. The immediate influence on the retrieved slant column of NO₂ is substantial, with clear enhancements above the brighter surfaces. The spatial resolution of about 5.6km used for surface characterisation is rather coarse in comparison to the GeoTASO resolution.

Could the intensity of GeoTASO measurements be used to retrieve a pixel-by-pixel ground albedo similar to what is done in Meier et al. 2017 or could another product with better spatial resolution be used? If this is not possible and the coarser spatial resolution shall be retained for the data analysis, the authors should consider a more careful investigation and critical discussion of the albedo treatment and the resulting influence on the error budget. This is also part of the next comment. The uncertainty of the albedo has at least two influencing aspects, (a) the uncertainty in the determination itself in addition to (b) the variability of the albedo within one MODIS ground pixel. Are both aspects considered in the error budget? Is the uncertainty of the combined effect only 20% as stated in the error analysis section?

(Meier, A. C., Schönhardt, A., Bösch, T., Richter, A., Seyler, A., Ruhtz, T., Constantin, D.-E., Shaiganfar, R., Wagner, T., Merlaud, A., Van Roozendaal, M., Belegante, L., Nicolae, D., Georgescu, L., and Burrows, J. P.: High-resolution airborne imaging DOAS measurements of NO₂ above Bucharest during AROMAT, *Atmos. Meas. Tech.*, 10, 1831–1857, <https://doi.org/10.5194/amt-10-1831-2017>, 2017.)

-Uncertainties-

The given uncertainties that directly enter the error analysis are not well motivated. Uncertainty values of AOD, SSA, ALH and surface reflectance are assumed and applied (cf. page 9, l. 273), however, no reference or additional information is given. What is the origin of these numbers? The uncertainties seem to be rather small.

(For example, as stated above, the surface reflectance within one MODIS 0.05° grid box can vary quite substantially. The resulting uncertainty of the surface reflectance of a GeoTASO ground pixel is given by a combination of the initial uncertainty of the MODIS value, and in addition by this variability of the albedo within one grid box. Is this variability taken into account here?) Also when comparing with typical urban scenarios (Leitao et al., 2010), the influence of aerosol properties (different aerosol types and optical properties) on the AMF would be assumed to be larger than a percent given in ll. 284-286 and Table 6.

Furthermore, the results from the spatial variations of the error (ll. 290-305 and Fig.8) yield larger values than stated for the calculated impact (ll. 281-286). This should be reconsidered.

In order to understand the situation treated in the study, the field of values (spatial distribution) shown as maps would be helpful, i.e. similar to Fig. 8, four maps giving the applied (unperturbed) values of AOD, SSA, ALH and surface reflectance for an example flight. At least some information of the applied values is needed, such as the average and spread of values used (mean and standard deviations within the measurement area for the above parameters). This would be necessary for the reader to understand the situation.

As correctly stated in the introduction and conclusions, the NO₂ vertical profile influences the NO₂ AMF. However, this is not explicitly mentioned in the error analysis. For typical urban scenarios, the uncertainty in NO₂ profile can add another 10% uncertainty to the AMF (Leitao et al., 2010, Meier et al., 2016). It would be good to take this additional uncertainty into account.

-Resulting error calculation-

The explanations following eq. (10) especially lines 274-277, are not very well described. Please revise this short part. Was the influence of varying the parameters only determined for positive perturbation as stated (i.e. only for $c_i + sc_i$ and not for $c_i - sc_i$)? The AMF dependence on the four parameters is non-linear, so that the relative contribution/error source of a negative perturbation could be larger than in positive direction. Both variations ($\pm sc_i$) should be investigated. In the analysis of the spatial variations of the error (ll. 290-305), on the other hand, the both-sided influence is rightly investigated.

l. 277 states that $c_i + sc_i$ is the uncertainty of each parameter, which is not correct. This would be only sc_i . Maybe what is meant is "... the new NO₂ AMF simulated using the perturbed input parameters $c_i + sc_i$ (i.e. the original input parameters modified by the uncertainty)".

-AMF results-

Especially when regarding Figure 5 for the Anmyeon region some questions about the AMF calculation arise. While the SCD result from GeoTASO looks reasonable, especially above the two emission plumes, the VCD map shows a stripe of large NO₂ values (in latitude direction at about 126.4°E) downwind of the power plant. Looking at the AMF map, the AMF exhibits a sudden low value within this stripe. So the enhanced VCDs seem to be provoked by the low AMF values in this location. In addition, the overall impression of the AMF figure is quite stripy. Hence, the applied AMF values should be checked and potentially corrected if some mistake can be found. It becomes obvious that the AMF influence on the VCD is large, and that possibly the error budget is underestimating the AMF uncertainty. To find the reason, it could be helpful to investigate the spatial distributions of the influencing parameters (AOD, ALH, SSA and SFR).

-OMI NO₂ product-

As stated above, the comparison to OMI data is a relevant aspect of the study and is also mentioned in the abstract and conclusions. However, hardly any information is given on the OMI data (NO₂ product). Please add the basic necessary information, especially which data version has been used, and include a reference, where all further details of the product can be found.

-GeoTASO/OMI comparison-

The aircraft/satellite comparison is not sufficiently supported by data. In addition to merely stating the correlation and slope (p.10), it would be necessary to actually show a direct comparison, ideally in a scatter plot. The section on p. 10 is rather short and unclear. Does the number 53 refer to the number of OMI pixels considered for the comparison? From GeoTASO, there are then presumably much more observations used. Are all aircraft data from within one satellite pixel averaged prior to the comparison, or are the individual GeoTASO observations compared to the OMI data?

Considering the reference Judd et al (2019), the authors could put their results, especially the resulting correlation, into context with conclusions therein.

The slope of 0.43 is not commented. This result should be critically discussed giving some idea of the reasons. Finally, the comparison of GeoTASO and OMI NO₂ could receive a section of its own (like it is done with 3.3.1 and 3.3.2 for GeoTASO/insitu and

GeoTASO/Pandora, respectively).

Further Comments

I. 81: What is meant here? Maybe something else is addressed and not radiative transfer models, maybe regional air quality models?

I. 151: From which time of the day is the reference spectrum taken?

I. 152: Probably there is a typo in the exponent, as this value seems to be too large. The background OMI NO₂ in the reference region (red circle in Fig. 1) is below 1×10^{15} molec/cm², the stratospheric amount is also much smaller than the given value. Therefore, the CMAQ probably has a different output than stated here. Also it is not suitable to give three digits precision here. Please check this number and correct.

I. 154: Please explain the spread in FWHM values for the GDF. These are probably differences for the different viewing directions of GeoTASO. Please specify.

II. 154-164: The settings in the NO₂ retrieval are different from what was documented in previous studies (e.g. Judd et al., 2019). Especially, the polynomial order of 8 is exceptionally large. Is this correct? Is it clear, why this is necessary? In addition, please give a reference for the applied H₂O absorption.

II. 182-185: Part of the upward looking NO₂ column (roughly the stratospheric column) is effectively subtracted by the use of the reference in the DOAS fit. This could be mentioned here. However, it is the change in the upward looking NO₂ column between reference and actual measurement that is then neglected in the further analysis. Morning and afternoon measurements are used, and the stratospheric NO₂ column changes during the day. Therefore, the time of the reference measurement on 1 May 2016 is relevant (I. 151). Although the change is not large, it enters the error budget and could/should be quantified.

II. 312-317: The correlation values for AM and PM comparisons between GeoTASO and insitu NO₂ are considerably different. A few words of discussion would be appreciated here (e.g. influence of boundary layer height, meteorology, or other influencing factors?). Maybe also put these results into perspective with expectations (no perfect correlation

expected for this type of comparison). The explanation in I.316 is confusing. It is rather the in situ measurement that is specifically sensitive to local NO₂ sources such as roadsides. In case of more distant NO₂ sources, or vertically elevated sources such as power plant exhaust plumes, the NO₂ is potentially not detected by the in situ instruments (depending on conditions) but well visible for GeoTASO, thereby reducing correlation between the two observations.

II. 328-330: The slope of 1.48 between GeoTASO and Pandora data is not explained. What could be the reason for such a value? Please discuss this shortly. The differences in viewing geometry can cause some scatter in the results, but cannot readily explain a slope much larger than 1.

II. 348-349: The description of changing NO₂ VCDs in Seoul and Busan from morning to afternoon flights sounds as if this was due to the different locations/cities. Isn't this rather due to different conditions, either different meteorological conditions or other?

II. 350-351 (and I. 125): The comparison of GeoTASO results with insitu measurements is addressed here as "validation". It is certainly interesting to compare ground level mixing ratios of NO₂ with tropospheric column densities. However, it is not possible without detailed information on the vertical profile of NO₂, to use point measurements of mixing ratios to actually validate column density measurements observed from aircraft. This is correctly stated in II. 320-321. A comparison of the two measurement strategies is interesting and has a certain information content of its own, but it is preferred to describe this as a comparison and not as a "validation".

In addition, the local mixing ratio at the surface is a different physical quantity than the tropospheric or total column densities that integrate over a considerable vertical range, i.e. there is not only a difference in the "physical units" (as stated in I. 125) but in the basic physical quantity. Please rephrase.

I. 352: The distance was restricted to these maximum values. So this should not say "approximately", but: "When the distances between two observations were below 25, 0.5 or 1 km..." or similar.

I. 353 and also I. 25: In both locations (conclusions and abstract), a correlation coefficient of 0.84 is given for the GeoTASO/Pandora validation. However, for data within 1km, the correlation was 0.94 as stated in I.328. The 0.84 value resulted for 5km radius.

It is not clear, why different values are chosen for the conclusions than for the abstract. A decision should be taken, which are the most important results. One section can state more of the results than another section, but it is confusing, if these two main summarizing sections choose to emphasize different results. For the comparison with the

insitu data, the abstract states $r=0.78$ for the Seoul afternoon values, while the conclusions state $r=0.67$ (which is for Busan, actually $r>0.67$) as well as $r=0.38$ for the Seoul morning values.

II. 356-357: As the sentence addresses the current study ("This demonstrates..."), please delete GCAS and APEX, or rewrite "This demonstrates that airborne remote sensing measurements from GeoTASO, similar to GCAS, APEX and others, can be a very effective tool for..." or similar.

II. 360-361: Please rewrite. The determination of the aerosol properties is not based on the error estimation, rather "Based on the error estimation, it can be concluded that aerosol properties are relevant and should be determined.." or similar.

Figures 5 and 7: Wind arrows are missing and could be added.

Figure 8: Overall, the maps in this figure are too small. A better structure would probably be two columns and four lines (instead of two lines and four columns) of maps. Then the single maps could be larger.

Tables 2-5: It remains somewhat open why these tables with many numbers are given in great detail, while they are not specifically used in the analysis. It is mentioned in the text, e.g. that the cities are highly populated and the expressways are heavily used. But no quantification of emissions with explicit comparison of different roads or regions is performed. Are these numbers needed at all / in such detail? Can they further support the analysis? For the cars, e.g., lines 212-213 give a good summary of the large numbers and might be sufficient for the level of discussion and analysis here.

Similarly, the numbers for the power plants or steelwork NO_x emissions (l. 235) are too detailed. These could rather be rounded to 10.3, 11.9 and 16.8 kt/year, respectively, in order to improve readability.

References

II.64-70: For the history of airborne DOAS instruments, the HAIDI (General et al., 2014) is missing. Further airborne DOAS studies with partly different objectives exist. To motivate the selection of mentioned instruments here, it would be helpful to state that these are all

mapping instruments (with according spatial coverage).

General, S., Pöhler, D., Sihler, H., Bobrowski, N., Frieß, U., Zielcke, J., Horbanski, M., Shepson, P. B., Stirm, B. H., Simpson, W. R., Weber, K., Fischer, C., and Platt, U.: The Heidelberg Airborne Imaging DOAS Instrument (HAIDI) – a novel imaging DOAS device for 2-D and 3-D imaging of trace gases and aerosols, *Atmos. Meas. Tech.*, 7, 3459–3485, <https://doi.org/10.5194/amt-7-3459-2014>, 2014.

Technical Corrections

l. 38: replace "size" by "sized"

l. 69: Please replace "emissions sources" by "emission sources".

l. 77: Please put a comma behind APEX.

l. 87: Please replace "campaign" by "campaigns".

l. 117: Please add "nm" behind 0.6.

l. 118: Please replace "O3" with subscript "O₃", and "depicted as" with "depicted by".

l. 127: Please replace "compared" with "compare".

l. 133: Please replace "measurement" by "measurements".

l. 136: Please replace "collection" with "correction".

I. 153: Please delete "convoluted", replace by "convolved". And then rephrase to read: „... represents the differential gas phase absorption cross-section convolved with the Gaussian...“

I. 159: Please delete "ring" and replace by "Ring".

I. 174: Please replace "product" by "products".

II. 176-178: Please correct this sentence. There seems to be a mix up of two parts, and the grammar is not correct.

I. 184: Please replace twice the term "concentrations" by "column densities".

I. 190: Please use capital letters (Weather Research and Forecasting Model, WRF. Advanced Research WRF, ARW).

I. 236: At the beginning of the sentence, please replace "Fig.5" with "Figure 5".

I. 246: 6m/s (or -1 as superscript)

I. 256: Replace "which" by "with".

I. 257: Add and "s": "Figs. 4 and 7"

I. 262: Include "the": "fitting error of the SCD"

I. 346: Replace "of" by "off".

I. 362: Replace "affect" by "affects".

Full stops at the end of sentences are sometimes missing (e.g. l. 122, 132, 312...)