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 at 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.

Response: First of all, we sincerely apologize for the late submission.

We thank the reviewer's kind comment and advice. The NO_2 AMF errors were recalculated based on the uncertainties of parameters (AOD, SSA, ALH, and surface reflectance) obtained from previous studies. Relevant references for the uncertainty were also added in the revised manuscript (P. 17-18, Lines 377–379):

"The σ of AOD, SSA, surface reflectance, and ALH are assumed as 30% (Ahn et al., 2014), 0.04 (Jethva et al., 2014), 0.005+0.05×surface reflectance (EOS Land Validation; https://landval.gsfc.nasa.gov), and 1 km (Fishman et al., 2012), respectively, in this study."

We showed the AMF which is native resolution (250 m) of GeoTASO by modifying Fig. 5(c), and we revised to the contents in the manuscript (P. 13, Line 300). Also, we have revised all the comments below.

- Ahn, C., Torres, O., & Jethva, H. (2014). Assessment of OMI near-UV aerosol optical depth over land. Journal of Geophysical Research: Atmospheres, 119(5), 2457-2473.
- Jethva, H., Torres, O., & Ahn, C. (2014). Global assessment of OMI aerosol single-scattering albedo using ground-based AERONET inversion. Journal of Geophysical Research: Atmospheres, 119(14), 9020-9040.
- EOS Land Validation (https://modis-land.gsfc.nasa.gov/ValStatus.php?ProductID=MOD09)
- Fishman, J., Iraci, L. T., Al-Saadi, J., Chance, K., Chavez, F., Chin, M., ... & Wang, M. (2012). The United States' next generation of atmospheric composition and coastal ecosystem measurements: NASA's Geostationary Coastal and Air Pollution Events (GEO-CAPE) mission. Bulletin of the American Meteorological Society, 93(10), 1547-1566.

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?).

Response: First, we appreciate the reviewer's comment and apologize for the confusion. We have therefore revised the OMI spatial resolution of the caption in Fig. 1 as you suggested.

Our paper more focused on the NO_2 retrieval from GeoTASO and the investigation of its spatial distribution. We did spatial binning after calculating NO_2 VCD. Chong et al. (2020) calculated SO_2 VCD using data observed in GeoTASO during the KORUS-AQ period, and binning was performed at a resolution of GEMS (7 x 8 km^2). As a result of previous studies, it was found that relative random uncertainty decreased as SO_2 VCD increased. This random uncertainty can be reduced even when additional spatial binning is performed, which provides a balance between random error and spatial resolution. In conclusion, they found that larger VCDs at 250 m resolutions do not necessarily lead to larger VCDs at 7 km x 8 km resolutions. A similar part of your comments exists in the comments below. Additionally, we showed the AMF by modifying Fig. 5(c) to native resolution to respond to the comments. Please, see P. 10, Lines 249-252.

After modification (P. 10, Lines 249-252):

"We showed the finally NO_2 VCDs by binning them with 0.01° × 0.01° from 250 m spatial resolution. Although the spatial binning NO_2 VCDs were compared to those at native resolution, we noted that the spatiotemporal variability was still able to be clearly distinguished from the background at 0.01° binning resolution. Chong et al.

(2020) showed that larger VCDs at 250 m resolutions do not necessarily lead to larger VCDs at wider resolutions."

- Chong, H., Lee, S., Kim, J., Jeong, U., Li, C., Krotkov, N. A., Nowlan, C. R., ... & Koo, J.-H. (2020). High-resolution mapping of SO_2 using airborne observations from the GeoTASO instrument during the KORUS-AQ field study: PCA-based vertical column retrievals. Remote Sensing of Environment, 241, 111725.

<u>-Treatment of the spectral reflectance-</u>

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 Roozendael, 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.)

<u>Response</u>: We agree with reviewer's comments. First of all, as you mentioned, the algorithms for calculating observed surface reflectance using GeoTASO data and surface reflection results such as Meier et al. (2017) were not provided. We tried to use surface reflectance (500 m resolution) of MCD43A3, but there were many pixels that were not produced within the observation area. We wanted to know the spatial distribution of NO_2 VCD. Therefore, we used MOD09CMG and MYD09CMG. Because the data exist in all pixels, the surface reflectance.

In this study, the uncertainty according to each input variable was calculated and provided in Section 3.2. In our result, the variable with the largest AMF error was aerosol loading height (26.4%), which is SSA (4.2%), AOD (3.0%), and surface reflectance (2.8%). In addition, the average of the surface reflectance during KORUS-AQ period is 0.055, and there is a study using a fixed ground reflectance of 0.05 to retrieve SO_2 VCD (Chong et al., 2020). As mentioned below, AMF uncertainties were recalculated based on the uncertainties of input parameters from previous studies. Higher AMF uncertainty (27.8%) has been gained. The numbers in Table 6 are the average of the flight on 9 June 2016. To clarify it, a sentence and the caption for Table 6 have been revised. Please, see P. 18, Line 287.

-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.

Response: The NO_2 AMF errors were recalculated based on the uncertainties of parameters (AOD, SSA, ALH, and surface reflectance) obtained from previous studies. Relevant references for the uncertainty were also inserted in the revised manuscript (P. 17, Lines 377–379):

"The σ of AOD, SSA, surface reflectance, and ALH are assumed as 30% (Ahn et al., 2014), 0.04 (Jethva et al., 2014), 0.005+0.05×surface reflectance (EOS Land Validation; https://landval.gsfc.nasa.gov), and 1 km (Fishman et al., 2012), respectively, in this study."

- Ahn, C., Torres, O., & Jethva, H. (2014). Assessment of OMI near-UV aerosol optical depth over land. Journal of Geophysical Research: Atmospheres, 119(5), 2457-2473.
- Jethva, H., Torres, O., & Ahn, C. (2014). Global assessment of OMI aerosol single-scattering albedo using ground-based AERONET inversion. Journal of Geophysical Research: Atmospheres, 119(14), 9020-9040.
- EOS Land Validation (https://modis-land.gsfc.nasa.gov/ValStatus.php?ProductID=MOD09)
- Fishman, J., Iraci, L. T., Al-Saadi, J., Chance, K., Chavez, F., Chin, M., ... & Wang, M. (2012). The United States' next generation of atmospheric composition and coastal ecosystem measurements: NASA's Geostationary Coastal and Air Pollution Events (GEO-CAPE) mission. Bulletin of the American Meteorological Society, 93(10), 1547-1566.

(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?)

Response: The resulting uncertainty of the surface reflectance of a GeoTASO ground pixel was not considered in this study. However, this analysis should be carried out in the future. Therefore, the authors added the following sentence in the revised manuscript:

Page 19, Lines 406-408: "Moreover, the resulting uncertainties of input parameters of a GeoTASO ground pixel need to be considered by combining the initial uncertainties of CTM and satellite-based products, and by the variability of the parameters within a grid box. This kind of analysis should be taken into account in further study."

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.

Response: The recalculated NO_2 AMF errors due to uncertainties in AOD, ALH, and SSA are 3.0%, 26.4%, and 4.2%, respectively. The influence of aerosol properties seems smaller than those in Leitão et al. (2010). It can be explained by aerosol profile (AOD and ALH) and aerosol type (SSA) values on the flight day when error analysis is carried out. The average values of AOD, ALH, and SSA were 0.39, 0.27 km, and 0.98, respectively. Especially, the AOD ranged from 0.15 to 0.68

including low and moderate AOD conditions. As stated in Leitão et al. (2010), the effect of aerosol properties become large in high AOD condition (AOD = 1.05). Therefore, the NO2 AMF errors calculated in this study is smaller than those in the previous study since these were calculated under observation conditions with moderate aerosol loading on 9 June 2016. For better understanding of the readers and according to other comment by the reviewer, we have added the observation conditions on the flight day.

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.

Response: The values in lines 281-286 are averaged AMF errors derived from error propagation method on the flight day. However, values in Figure 8 indicates the percent difference of NO_2 AMF on each spatial pixel. Moreover, there are difference in calculation methods. To clarify the difference in the calculation methods, sentences have been added to explain in the revised manuscript.

P. 19, Line 409:

“$$AMF_{percent\_diff} = \frac{\partial AMF}{(AMF_{true} + AMF_{new}) \div 2} \times 100 \quad (14)$$”

P. 20, Lines 417-419: In this present study, we additionally investigated the spatial distribution of AMF calculation errors associated with uncertainties in aerosol properties (AOD, SSA, ALH, and SFR). Percent difference of NO_2 AMF ($AMF_{percent\_diff}$) was calculated on each spatial pixel using Eq. (14).”

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.

Response: To improve reader’s understand, some information (mean and standard deviation values) of the parameters (AOD, SSA, ALH, and surface reflectance) have been provided in the revised text.

P. 18, Lines 384-385: “On the flight day, average (standard deviation) values of AOD, SSA, ALH, and surface reflectance were 0.39 (0.10), 0.98 (0.001), 0.27 km (0.10 km), and 0.09 (0.04), respectively.”

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.

Response: Our paper more focused on the NO_2 retrieval from GeoTASO and the investigation of its spatial distribution. Therefore, for now, we considered AOD, SSA, ALH, and surface reflectance in the error analysis section. The authors fully agree with the necessity of AMF error analysis due to the uncertainty related to the a priori NO_2 profile shape and preparing it now for the next paper. Some sentences were added to point out the necessity related to NO_2 profile shape (Page 19, Lines 404-408):

"A priori NO_2 profile shape also can be one of factors to cause calculation error for NO_2 AMF as reported in the previous studies (Leitao et al., 2010, Meier et al., 2016). It is necessary to calculate the effect of a priori NO_2 profile shape on airborne NO_2 AMF error in the future."

-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 ci + sci and not for ci - sci)? 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 (+/- sci) 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.

Response: In the error analysis, a negative perturbation was also considered, however, it was not specified in Eq. (13). It has been corrected in the revised manuscript.

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

Response: The authors agree that. The sentence has been modified in the revised manuscript.

-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).

Response: We agree with the reviewer's advice. As you advised, we showed the AOD, ALH, SSA, and surface reflectance used in the AMF calculation in Figure R1. AOD and SSA represent CMAQ model resolution and SRF represent MODIS resolution. The reason why the binned AMF (Fig. 5(c)) caused confusion was that there was a technical problem in the plotting process for duplicate pixels when expressing them as a mapping. Therefore, we showed the AMF by modifying Fig. 5(c) to native resolution (250 m) of GeoTASO, and revised to the contents in the manuscript (P. 13, Lines 300-305).

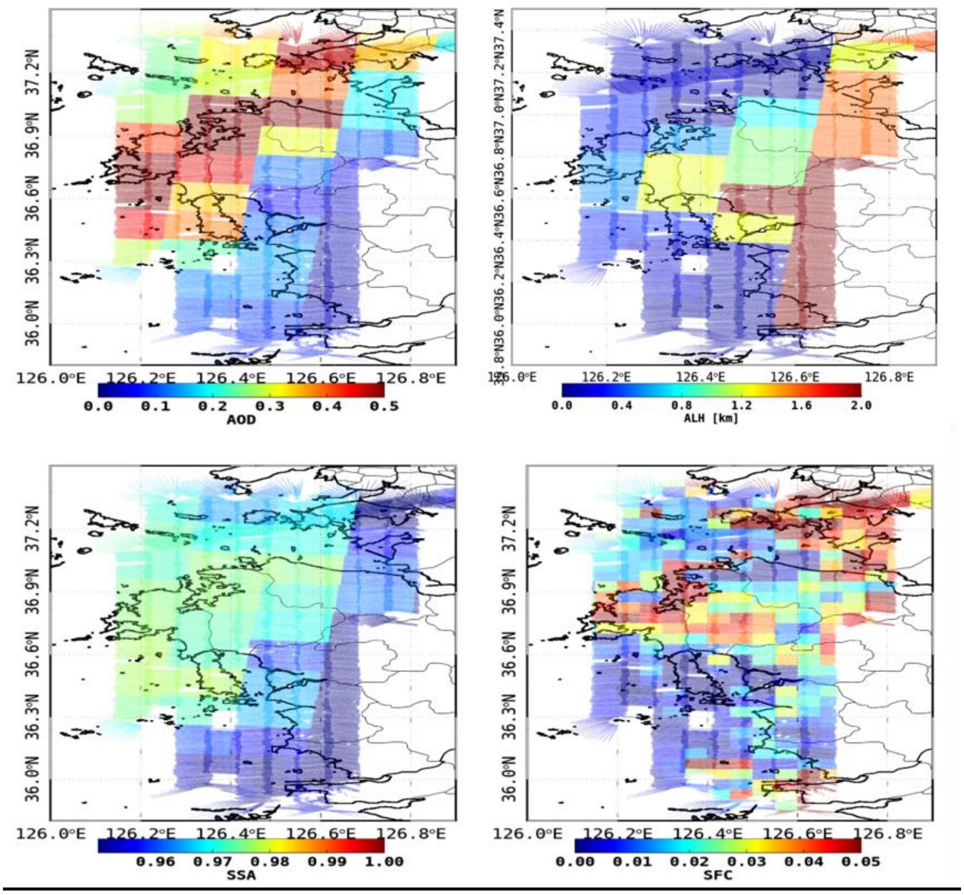


Figure R1. AOD, ALH, SSA for AMF calculated using the CMAQ model and surface reflectance (SFC) for AMF calculated MODIS data over Anmyeon on 5 June, 2016.

<u>-OMI NO2 product-</u>

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.

<u>Response</u>: Following reviewer's suggestion, we have described the information about the OMI NO_2 data in P. 5, Lines 134-137.

After modification (P. 5, Lines 134-137):

"The OMI data obtained by the Level 2.0 OMNO2 version 3.0 and downloaded from the NASA's Earthdata search (http://search.earthdata.nasa.gov/search/). We calculated the arithmetic means of the tropospheric NO_2 VCDs, similar to Choo et al. (2020), to obtain the grid data (0.25° × 0.25°) during KORUS-AQ period."

-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).

Response: we revised section 3.3 as follow:

After modification (P. 21-24, Line 432-494):

"Tropospheric NO_2 VCDs retrieved from GeoTASO L1B data ($NO_{2,G}$) were compared with those obtained from OMI NO_2 VCDs ($NO_{2,O}$) and Pandora ($NO_{2,P}$). The $NO_{2,O}$ were only available for 10 June during the campaign period. Therefore, we only compared 37 $NO_{2,G}$ and $NO_{2,O}$ data points within a radius of 20 km and 30 min, which yielded a correlation coefficient of 0.70 with a slope of 0.41 (Fig. 9 a)). In order to validate, All $NO_{2,G}$ within a radius 20 km of the OMI center coordinate were averaged.

The NO_2 values are relatively low since GeoTAOS observation is carried out in a region with low NO_2 compared to Seoul metropolitan and the overpass time of OMI is about 13:30 LT when NO_2 decreased. It is thought that the reson the low slope value is because the OMI with low spatial resolution does not reflect the spatial NO_2 inhomogeneity in the pixel.

To validate the accuracy of $NO_{2,G}$ data, we made a comparison with NO_2 VCD obtained from the Pandora system ($NO_{2,P}$) during the KORUS-AQ campaign period. $NO_{2,P}$ obtained from Busan University, Olympic Park, Songchon, Yeoju, and Yonsei University Pandora sites on June 5, 9, and 10 were used for the GeoTASO validation (Fig. 1). $NO_{2,G}$ and $NO_{2,P}$ columns at these sites are compared in Fig. 10. In order to compare $NO_{2,G}$ and $NO_{2,P}$, we used averaged $NO_{2,G}$ retrieved from 16 across track with smallest viewing zenith angle and averaged 30 min NO_2 obtained from pandora measurement within a radius of 0.05 degree. $NO_{2,G}$ and $NO_{2,P}$ were correlated (R = 0.79, with a slope of 1.15), however, when $NO_{2,P}$ was lower than 1×10^{16} molecules cm^{-2}, the correlation coefficient between $NO_{2,G}$ and $NO_{2,P}$ was < 0.1. The weak correlation at low NO_2 levels are most likely to reflect the differences in viewing geometries and the horizontal inhomogeneity of the measured NO_2 between Pandora and GeoTASO. Also, from this result, it is thought that it can be used for NO_2 validation of geostationary satellite such as GEMS using Pandora and GeoTASO. However, since the number of pandora is limited in this campaign, we had difficulties to validate NO_2 retrieved from GeoTASO under various conditions. I believe that many ground-based remote sensing measurements are needed to validate GEMS under various conditions.

To evaluate the spatiotemporal distribution of NO_2 VCDs retrieved from GeoTASO, $NO_{2,G}$ in comparisons to surface spatial patterns, $NO_{2,G}$ was compared with $NO_{2,A}$ for GeoTASO data within a radius of approximately 0.05 km and 30 min (Fig. 9). In order to compare $NO_{2,G}$ and $NO_{2,A}$, we used averaged $NO_{2,G}$ retrieved from 16 across track and averaged 30 min within a radius of 0.05 degree. Since in-situ measurements provides NO_2 VMR ($NO_{2,A}$)(ppmv) once per hour, $NO_{2,A}$ of the nearest time is used to compare with $NO_{2,G}$.The correlation coefficient (R) between $NO_{2,G}$ (molecules cm^{-2}) and $NO_{2,A}$ at 9 AM and 3 PM LT in the Seoul metropolitan region was 0.45

and 0.81, respectively. When using only roadside station data from Air-Korea, the R-value for the morning increased to 0.83, which implies GeoTASO is more sensitive to emissions from NO_2 source areas, such as roadsides. As a result of the comparison, there were large differences in the morning and afternoon. These results were identified because synoptic meteorology played an important role from June 1 to June 10, 2016 (Choi et al., 2019). As described by Judd et al. (2018), the spatial distribution for NO_2 VCDs appears that reflects the emission source in local industrialized regions and transportations in the morning with relatively weak winds. In general, NO_2 concentration increases to late morning, indicating that the emissions process proceeds faster than the NO_2 removal process. As the planetary boundary layer heights (PBLH) in early afternoon increase and surface NO_2 is mixed through a deeper PBLH, the NO_2 VCDs distribution showed a wider increase in most of the Seoul metropolitan area and the overall column amounts continue to increase (Judd et al., 2018).

In addition, when comparing NO_2 VCDs with surface NO_2 concentrations, it should be interpreted carefully that it is a non-linear relationship between $NO_{2,G}$ and $NO_{2,A}$. Although it may vary depending on weather conditions, high NO_2 VCDs from airborne observations may sometimes be detected with low surface NO_2 concentrations. In particular, when exhaust gases emitted from industrial facilities are happen at a certain altitude (stacks/chimneys), $NO_{2,G}$ show high NO_2 VCDs, but $NO_{2,A}$ may be observed to have a low concentration. Unfortunately, in Anmyeon industrial region, $NO_{2,G}$ and $NO_{2,A}$ could not be compared due to spatial restrictions because the distribution of ground observation stations is concentrated in metropolitan areas.

In the Busan metropolitan area, the R-value of the $NO_{2,G}$ and $NO_{2,A}$ data had a correlation coefficient greater than 0.78. This reflects the more even horizontal distribution of NO_2 in the afternoon, when diffusion from the source areas had taken place. However, for a more accurate comparison, NO_2 VCD data should be converted to NO_2 MR based on mixing layer height, temperature, and pressure profile data (Kim et al., 2017; Qin et al., 2017; Jeong and Hong, 2021a). However, since the number of pandora and satellite data is limited in this campaign, we had difficulties to validate NO_2 retrieved from GeoTASO under various conditions. Since ground-based, airborne and space borne remote sensing measurements has their own advantage and disadvantage, I believe that a comprehensive observation campaign involving all of groud-based, airborne and space borne measurements should be carried out continuously for upcoming new era of geostationary environmental satellite."

Further Comments

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

Response: "regional radiative transfer models" has been replaced with "regional air quality models" at P. 3, Line 82.

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

Response: The time is 09 UTC. I know it is good to use the reference spectrum when light path is short, however in south Korea where NO2 is very high, I think it is better to use the reference spectrum in a clear area rather than a short light path.

l. 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 x 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.

Response: In this present study, we used radiance value averaged by 250 m × 250 m in each 33 across tracks as a reference over south ocean of Jeju Island which is one of the most clean region in south Korea. The NO_2 VCD and standard deviation in OMI in this region (SZA<85, cloud fraction < 0.5) is 4.77×10^{15} molec. cm^{-2} and 1.33×10^{15} molec. cm^{-2}, respectively.

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

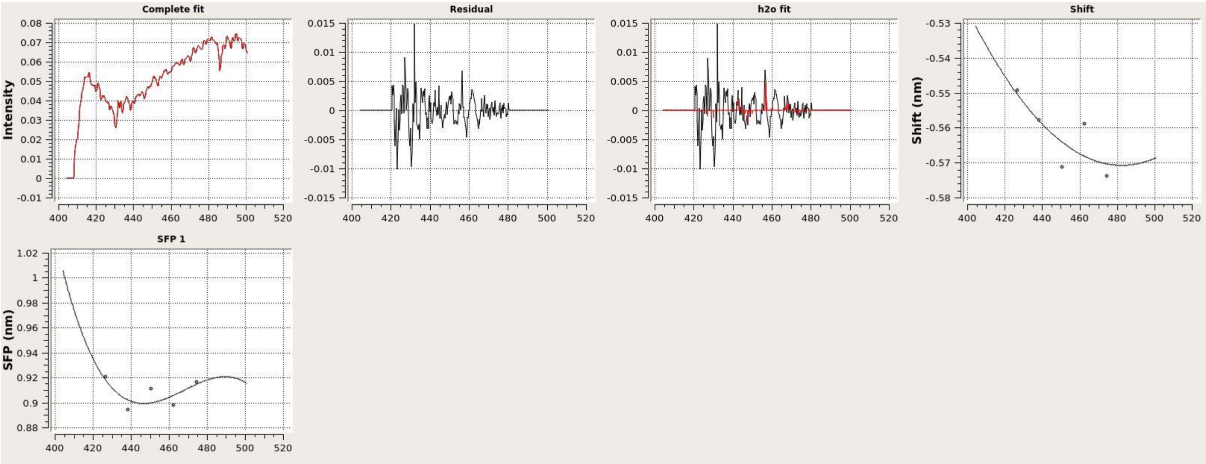
Response: As you mentioned, SRF (Spectral Response Function) varies across track and wavelength. The FWHM along the across track at about 438.4 nm is as follows:

Across track	FWHM	Across track	FWHM	Across track	FWHM	Across track	FWHM
0	0.8915	9	0.8938	18	0.8849	27	0.8884
1	0.8904	10	0.8862	19	0.8794	28	0.8835
2	0.8902	11	0.8948	20	0.8857	29	0.8806
3	0.8907	12	0.8915	21	0.8985	30	0.8788
4	0.8898	13	0.8876	22	0.8880	31	0.8923
5	0.8941	14	0.8880	23	0.8920	32	0.8880
6	0.8979	15	0.8890	24	0.8911		
7	0.8895	16	0.8903	25	0.8888		
8	0.8922	17	0.8878	26	0.8894		

If you think it is necessary to add this results to manuscript, we will follow it.

ll. 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.

Response: We re-retrieved NO_2 VCD under polynomial order of 3. And we revised figure.3. The reference for the applied H_2O absorption is as follows:

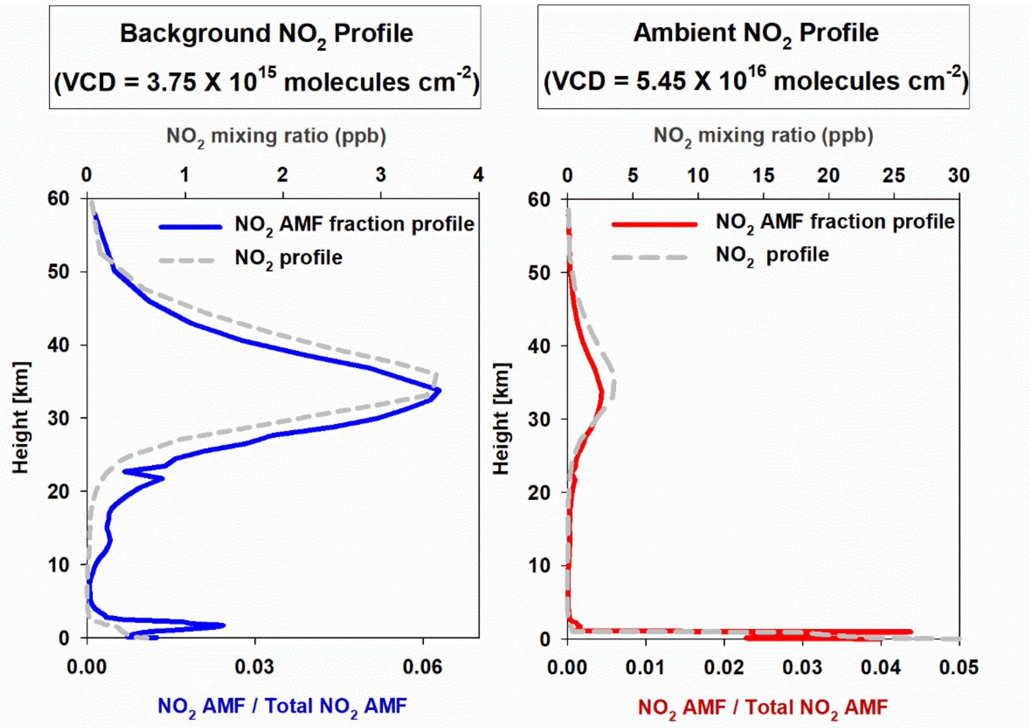


ll. 182-185: Part of the upward looking NO2 column (roughly the stratospheric column) is effectively subtracted by the use of the reference in the DOAS fit. This could be mentioned here. However, it is the change in the upward looking NO2 column between reference and actual measurement that is then neglected 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 (l. 151). Although the change is not large, it enters the error budget and could/should be quantified.

Response:

We agree that stratospheric NO_2 AMF is unignorable particularly in regions with low NO_2 concentrations. But since this study is focused on a polluted area in Korea with high NO_2 pollution, We thought NO_2 AMF in stratospheric can be exceptional. The fraction profile of NO_2 AMF by altitude is illustrated in the figure below. When NO_2 VCD is 5.45×10^{16} molecules cm^{-2}, the AMF effect at 8 km or higher is less than 8%.

In addition, the model used in this study was CMAQ during KORUS-AQ, which did not include stratospheric composition. We acknowledge that there is still a room to be improved, but applying another model seems unavailable at this stage, because if we do so, we have to do the analysis and writing all over again.



ll. 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 l.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.

Response: We agree with the reviewer's advice and have therefore revised the manuscript as you suggested. We have further explained in P. 23-24, Line 473-486.

After modification (P. 23-24, Lines 473-486):

"As a result of the comparison, there were large differences in the morning and afternoon. These results were identified because synoptic meteorology played an important role from June 1 to June 10, 2016 (Choi et al., 2019). As described by Judd et al. (2018), the spatial distribution for NO_2 VCDs appears that reflects the emission source in local industrialized regions and transportations in the morning with relatively weak winds. In general, NO_2 concentration increases to late morning, indicating that the emissions process proceeds faster than the NO_2 removal process. As the planetary boundary layer heights (PBLH) in early afternoon increase and surface NO_2 is mixed through a deeper PBLH, the NO_2 VCDs distribution showed a wider increase in most of the Seoul metropolitan area and the overall column amounts continue to increase (Judd et al., 2018).

In addition, when comparing NO_2 VCDs with surface NO_2 concentrations, it should be interpreted carefully that it is a non-linear relationship between $NO_{2,G}$ and $NO_{2,A}$. Although it may vary depending on weather conditions, high NO_2 VCDs from airborne observations may sometimes be detected with low surface NO_2 concentrations. In particular, when exhaust gases emitted from industrial facilities are happen at a certain altitude (stacks/chimneys), $NO_{2,G}$ show high NO_2 VCDs, but $NO_{2,A}$ may be observed to have a low concentration. Unfortunately, in Anmyeon industrial region, $NO_{2,G}$ and $NO_{2,A}$ could not be compared due to spatial restrictions because the distribution of ground observation stations is concentrated in metropolitan areas."

ll. 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.

Response: We revised section 3.3 as follow:

ll. 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?

Response: As you suggested, we have modified the manuscript. In addition, we show the wind speed and wind direction obtained from the UM-RDAPS in Figs. 4, 5, and 7, and write it in P. 11, Line 267, P. 13, Line 303, and P. 15, Line 330.

After modification (P. 24, Lines 506-507):

"However, in contrast to Seoul, tropospheric NO_2 VCDs in Busan decreased in the afternoon due to different weather conditions locally."

ll. 350-351 (and l. 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 ll. 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".

Response: We thank the reviewer's comments. We have modified the sentence as follows:

After modification (P. 24, Line 508-509):

"To compare the data retrieved from the GeoTASO system, we compared $NO_{2,G}$ with $NO_{2,O}$ obtained from the OMI, $NO_{2,A}$ obtained from Air-Korea, and $NO_{2,P}$ obtained from the Pandora observation system."

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 l. 125) but in the basic physical quantity. Please rephrase.

Response: As you suggested, the sentence was replaced as written in P. 6, Line 152.

After modification (P. 6, Line 152):

"Although the basic physical quantity of VCD and surface mixing ratio from in-situ measurements are different, ~"

l. 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.

Response: "regional radiative transfer models" has been replaced with "regional air quality models" at P. 3, Line 82.

After modification (P. 24, Line 510):

"When the distance between two observations was below 20 km or 0.05 degree within 30 min, …"

l. 353 and also l. 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 l.328. The 0.84 value resulted for 5km radius.

Response: Corresponding Section 3.3 has been modified.

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.

Response: We thank the reviewer's comments. We have modified the sentence as follows:

After modification (P. 1, Lines 24-26):

"The VCDs retrieved from the GeoTASO airborne instrument were well correlated with those obtained from the Ozone Monitoring Instrument (OMI) (r = 0.70), NASA's Pandora Spectrometer System (r = 0.79), and NO_2 mixing ratios obtained from in situ measurements (r = 0.45 in the morning, r = 0.81 in the afternoon over the Seoul, and r > 0.78 over Busan)."

ll. 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.

Response: We agree with the reviewer's advice and have therefore revised the manuscript as you suggested (P. 25, Lines 514-516).

After modification (P. 25, Lines 514-516):

"This demonstrates that airborne remote sensing measurements from GeoTASO, similar to GCAS, APEX and others, can be a very ..."

ll. 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.

Response: Again, We agree with the reviewer's advice and have therefore revised the manuscript as you suggested (P. 25, Lines 518-520).

After modification (P. 25, Lines 518-520):

"Based on error estimation, it can be concluded that aerosol properties are relevant and should be determined ..."

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

Response: We appreciate the reviewer's comment. As the reviewer mentioned, we have added that the wind arrows obtained from UM-RDAPS and shown them in Fig. 5 and 7. Additionally, an additional comment about the wind information has been added to the revised manuscript as follows. Please, see P. 13, Line 300, P. 16, Lines 340 and captions for Fig. 5 and 7.

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.

Response: We agree with reviewer's comment. We have modified the structure of Fig. 8.

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 NOx 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.

Response: We agree with the reviewer's advice and have therefore revised the manuscript as you suggested. Although we did not perform a very detailed analysis with the specific numbers presented in Table 2-5, we wanted to show the specific numbers them in detail in this research as a factor that affects the emitted NO_2.

After modification (P. 13, Line 315-316):

"In 2016, the annual NOx emissions by the Hyundai steelworks and the Dangjin and Boryeong power plants were about 10.3, 11.9, and 16.8 kt $year^{-1}$, respectively."

References

ll.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.

Response: The additional reference (General et al., 2014) has been added to P. 3, Line 65.

Technical Corrections

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

Response: "size" has been modified to "sized" on P. 1, Line 39.

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

Response: "emissions sources" has been replaced with "emission sources". Please, see P. 3, Line 69.

l. 77: Please put a comma behind APEX.

Response: We have added "," to P. 3, Line 77.

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

Response: "campaign" has been modified to "campaigns" on P. 3, Line 88.

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

Response: We have added “nm” to P. 5, Line 142.

l. 118: Please replace “O3“ with subscript “O_3“, and “depicted as“ with “depicted by“.

Response: P. 5, Line 143 have modified following the reviewer’s suggestion.

l. 127: Please replace “compared” with “compare”.

Response: “compared” has been changed to “compare” on P. 6, Line 154.

l. 133: Please replace “measurement” by “measurements”.

Response: “measurement” has been modified to “measurements” on P. 6, Line 151.

l. 136: Please replace “collection” with “correction”.

Response: “collection” has been modified to “correction” on P. 6, Line 163.

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

Response: As you suggested, we have modified the manuscript on P. 7, Line 185.

l. 159: Pleace delete “ring” and replace by “Ring”.

Response: P. 8, Line 190 have been modified.

l. 174: Please replace “product” by “products”.

Response: “product” has been replaced with “products”. Please, see P. 9, Line 215.

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

Response: Following reviewer’s suggestion, the corresponding sentence has been modified on P. 9, Line 217-219.

l. 184: Please replace twice the term “concentrations” by “column densities”.

Response: We have modified as the reviewer suggested in P. 9, Line 226.

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

Response: ″weather research and forecasting (ARW-WRF) model″ has been revised as "Weather Research and Forecasting (WRF)-Advanced Research WRF (ARW) Model". Please, see P. 10, Line 233.

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

Response: P. 15, Line 322 have modified following the reviewer's suggestion.

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

Response: P. 16, Line 337 have been modified.

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

Response: "which" has been replaced with "with" on P. 17, Line 356.

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

Response: "s" has been added to P. 17, Line 357.

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

Response: "the" has been added in front of "fitting error of SCD" in the revised manuscript.

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

Response: We couldn't find that part you mentioned in the manuscript.

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

Response: "affect" has been replaced with "affects". Please, see P. 25, Line 520.

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

Response: We added all the missing "." at the end of the sentences.