1.

Review of: Highly resolved mapping of NO_2 vertical column densities from GeoTASO measurements over a megacity and industrial area during the KORUS-AQ campaign

We thank the reviewer's advice and agree with you for pointing out. In this reviewing process, according your advice, we recalculated NO_2 AMF using high resolution surface reflectance data. Furthermore, we selected optimal spectral range for DOAS fitting via sensitivity test. After that we also recalculated NO_2 SCD under from 435 nm to 475 nm. As a result, almost all figures were changed, and the output results were also changed (Fig. 5-8 in manuscript).

Major comments

-Please extend section 2.1. with campaign information: This comment is not addressed. A link is made to a pdf document but that doesn't contain all relevant info requested. Please add a table to the manuscript with at least the requested info that will improve interpretation of your data and analysis.

Response: We added table 1 to section 2.1. Table 1 shows average values from GeoTASO campaign flight observation data such as flight time, flight altitude, SZA, ect. And we added the contents of table 1 to P. 4, line 123 to 126.

Table 1. Summary of information on the dates when NO_2 VCD was retrieved during the KORUS-AQ period (LT=UTC+9 hr).

Date	5 Jun	9 Jun AM	9 Jun PM	10 Jun AM	10 Jun PM
ROI	Anmyeon	Seoul metropolitan		Busan metropolitan	
Flight time (LT)	13:11–17:20	7:48–12:00	13:46–17:52	8:02–11:38	13:05–15:19
Flight altitude (km)	8.6	8.4	8.5	8.6	8.5
Flight speed (ms^{-1})	117.0	116.2	117.6	117.2	117.1
SZA (°)	39.2	36.1	45.3	35.9	33.0
VZA (°)	168.1	167.4	117.6	117.2	117.1
AOD	0.27	0.40	0.21	0.13	0.09

SSA	0.966	0.980	0.949	0.981	0.968
Surface reflectance	0.07	0.09	0.09	0.06	0.06
Cloud fraction	0.08	0.31	0.55	0.16	0.20

-Ref. spectrum: you don't specify the value you are using for the residual amount while this was requested (please check also in TROPOMI or GEMS data what the background value is over your reference area; obviously not possible for the campaign dates but if it is a real background area, it should stay relatively stable). You don't demonstrate that the instrument is stable enough to use a sing ref. spectrum instead of a daily one, like requested. I provided details how you can easily do such a test based on the RMS on the fit.

Response: We investigated the time series of total NO_2 VCD using TROPOMI offline data on the south of Jeju Island in 2019 and 2020. Fig. R1 shows total NO_2 VCD on the south Jeju Island (32.983°N, 126.39°E) in 2019 (black) and 2020 (gray), respectively. In this period, the average of total NO_2 VCD is 4.81×10^{15} molecules cm^{-2} and standard deviation is 0.43×10^{15} molecules cm^{-2}, respectively. During this period, total NO_2 VCD is stable.

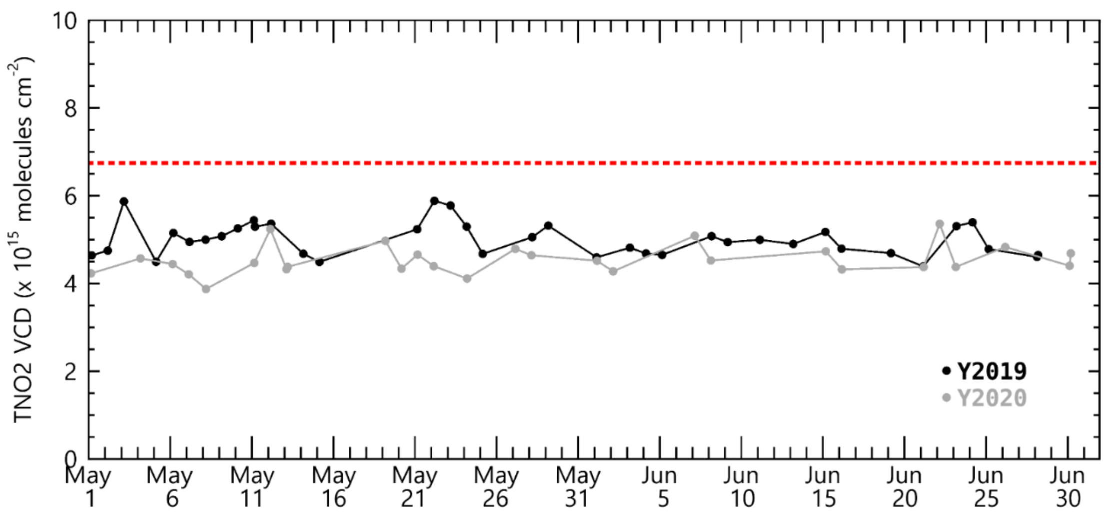

Figure R1. Total NO_2 VCD obtained from TROPOMI in 2019 (black) and 2020 (gray) on South Jeju Island. The red dot represent NO_2 VCD obtained from CAMQ.

-Spectral window: You agree that 425-490 nm would be more stable but you don't explain why you took a 425-450 nm window for your analysis. I'm not asking to change to 425-490 nm but I ask to give a proper argumentation for this choice. Moreover, in your answer and subsequent scatter plot you do the analysis for 425-460 nm, while you use the 425-450 nm range in your

manuscript??? This is not the same…

Response: We recalculated all NO_2 SCD in this present study using the radiance from 435 nm to 475 nm and recalculated NO_2 AMF at 455 nm. To select optimal wavelength range for DOAS fitting, we carried out sensitivity test with 17 fitting window candidates from 420 nm to 480 nm with the length of the fitting window from 25 nm to 60 nm. We presents this results to Fig. 3 and P. 7, line 197 – 207 in manuscript.

After modification (P. 7, Line 199 - P. 8, Line 211):

"The spectral fitting window was selected based on the sensitivity test with 17 fitting window candidates from 420 nm to 480 nm with the length of the fitting window from 25 nm to 60 nm. Spectral fitting residuals and NO_2 SCD errors have been investigated for 17 spectral fitting window candidates (Fig. 3). In terms of the residual, when the NO_2 fitting window includes a wavelength region less than 430 nm, it tends to have a larger residual compared to the case where it does not. The higher residual can include the more noise signals that cannot be calculated mathematically, which can become an uncertainty for the NO_2 SCD retrievals. Therefore, we excluded the fitting window which includes wavelength less than 430 nm for the GeoTASO NO_2 retrievals during KORUS-AQ campaign. In case of NO_2 SCD error, it was confirmed that the longer the fitting window length, the lower the NO_2 SCD error appeared regardless of including the wavelength region less than 430 nm. Therefore, for the stable NO_2 SCD retrieval, an appropriate spectral fitting window needs to be selected which can minimize the residual with a moderate length of the fitting window. To find the optimal fitting window, we set the threshold value based on the results above: residual < 0.001, NO_2 SCD error < 1.4×10^{15} molecules cm^{-2}, the length of fitting window > 30 nm. Then, the fitting window of 435–475 nm was finally selected for the GeoTASO NO_2 retrievals during KORUS-AQ campaign."

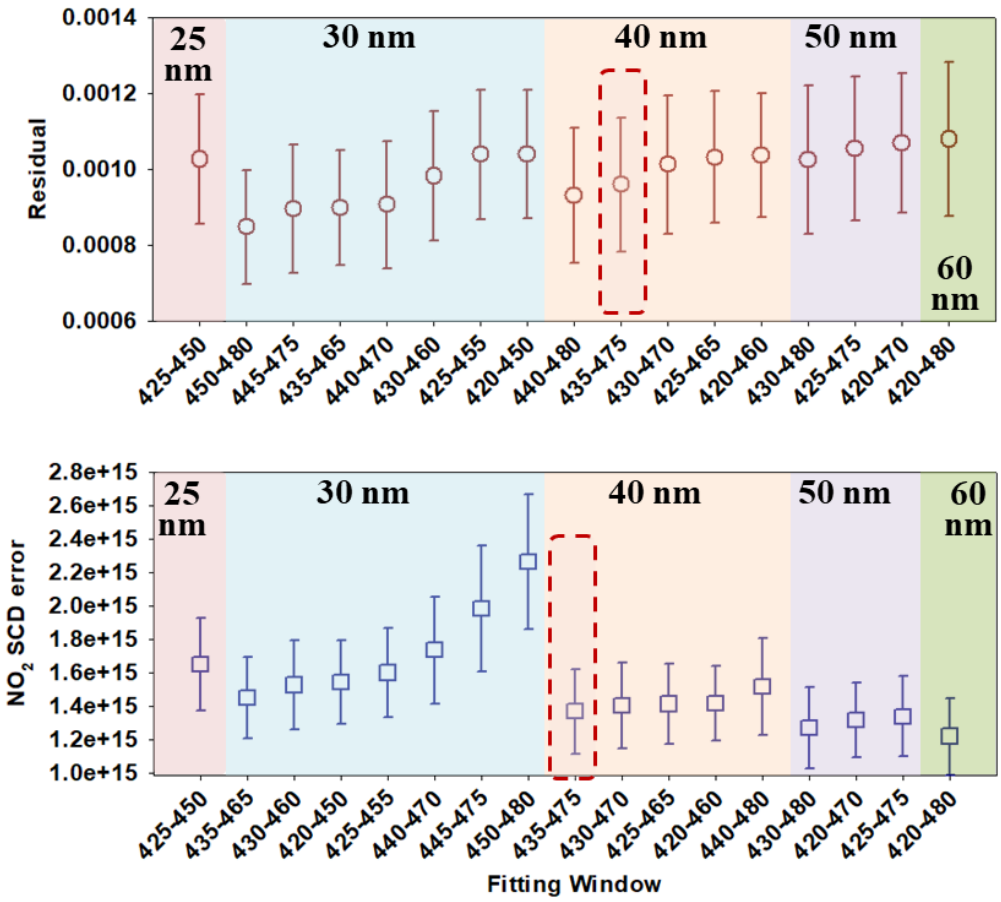

Figure R2. Residuals and NO_2 SCD errors of 17 spectral fitting window candidates (May 17, 2016, across track number: 15).

-Surface reflectance and impact on AMF: This answer cannot be accepted and I still have a strong concern that this course albedo product has a strong impact on your VCD retrievals. First of all it is not clear how the error on the surface reflectance product is determined. Secondly it is not only the error on the surface reflectance product that counts but also the spatial representativity error. As mentioned, several studies demonstrate strong variability of the albedo in an urban context and sensitivity tests show a very strong impact on the AMF, thus on the VCD. I would like to see at least one data set where you apply a better albedo product, e.g. MODIS product MCD43A1 or MCD43A, and point out differences between the VCD retrievals.

Response: Thanks to your advice, we recalculated AMF using surface reflectance obtained from MCD43A3 data of MODIS not MOD09CMG. The spatial resolution of MCD43A3 is 500 m × 500m. The average difference of surface reflectance and AMF between using MCD43A3 and using MOD09CMG is -0.047 and 0.330, respectively. Mean absolute error is 0.049 and 0.296, respectively. Fig. R3 shows the difference in AMF between new surface reflectance (MCD43A3) and MOD09CMG.

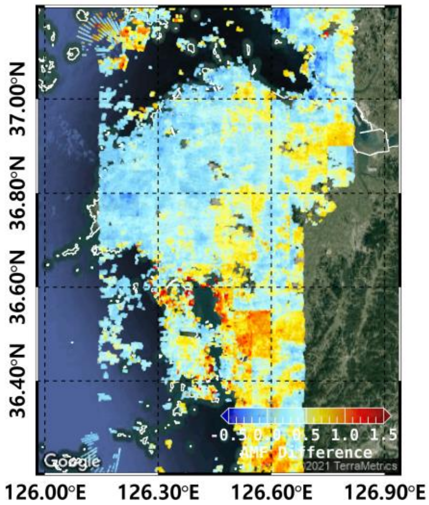

Figure R3. Difference of NO_2 AMF calculated using VLIDORT over Anmyeon in South Korea on 5 June 2016.

-spatial binning: I still don't fully get it and the explanation in the manuscript is certainly not clear at all (please revise). Yes indeed, random noise will decrease (but then still it would be better to bin before spectral fitting) and it is indeed a trade-off, but the objective/title of the paper is highly resolved mapping of NO2... I don't really see the point of binning to a spatial resolution courser than TROPOMI. The reference to the SO2 paper is not really valid as the SO2 signal is lower + less spatiotemporal variability. I'm also confused by your plots R1 and R2 in next answer. It's a pushbroom imaging system so where are the data gaps coming from?

Response: Gladly taking your advices, we recalculated NO_2 VCD using high spatial resolution (500 m × 500m) surface reflectance data (MODIS MCD43A3), the results were also re-presented as a figure without spatial binning (Fig. 5-8 in manuscript).

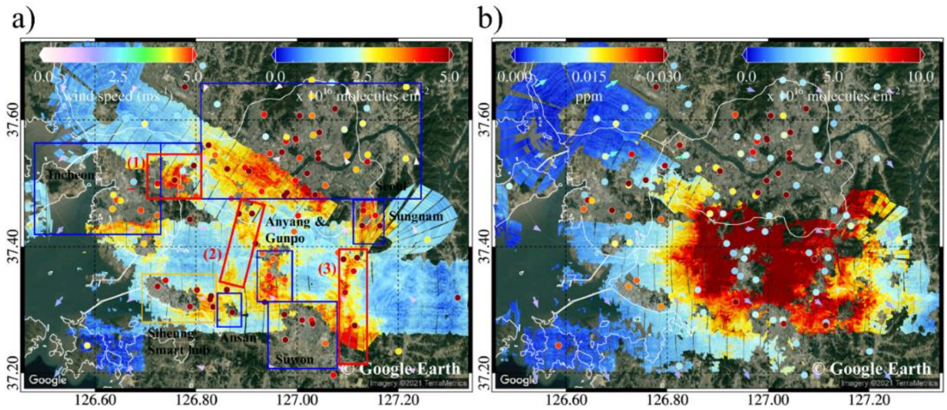

Figure R4. Tropospheric NO_2 VCD, in the Seoul metropolitan region on 9, June 2016 retrieved from GeoTASO: a) at 9 AM and b) at 3 PM. The red boxes represent expressways (counterclockwise from left to right, (1) Gyeongin expressway, (2) Seohaean expressway, and (3) Gyeongbu expressway), the orange box indicates the industrial complex, and the blue boxes indicate the major cities (Seoul, Incheon, Suwon, Bucheon, Anyang, Gunpo, Sungnam, and Ansan) of the Seoul metropolitan region. Colours of the circles depict the NO_2 surface mixing ratio obtained from Air-Korea. The colour arrows show the wind direction and speed at 1000 hPa over Seoul metropolitan region, obtained via the Unified Model (UM) simulations (background RGB image is from Google Earth; https://www.google.com/maps/).

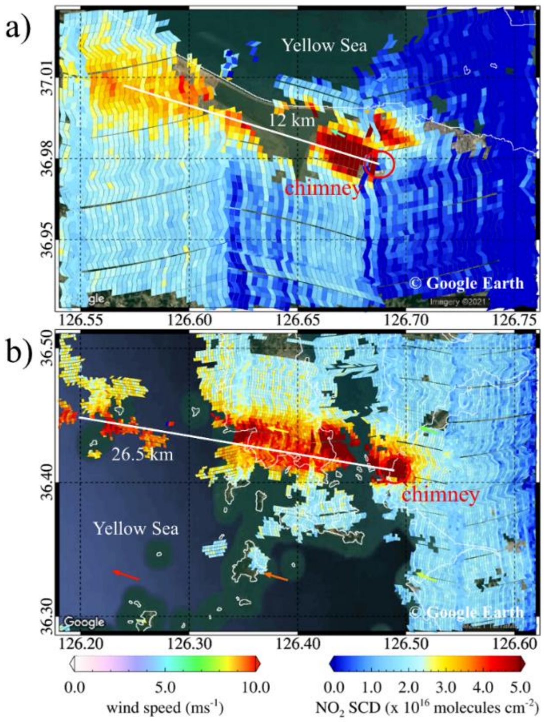

Figure R5. Enlarged view of GeoTASO tropospheric NO_2 VCD observation over a) Hyundai steel works, indicated by the red box in Figure 6, and b) the Boryeong power plant, indicated by the white box in Figure 6. The arrows represent the wind direction and speed at 850 hPa from the Unified Model (UM) simulations, respectively (background RGB image is from Google Earth; https://www.google.com/maps/).

- The error on the SCDref is an important source of uncertainty: Yes but you still don't take it into account in your error budget (similar for the error on the NO2 profile)…The answer to the next uncertainty question is a little weird. Either you don't include an uncertainty budget, either you do it properly.

Response: we added this sentence "We confirmed the stability of NO_2 distribution in this region using TROPOMI offline data from 2019 to 2020. In this period, the average of NO_2 VCD is 4.81×10^{15} molecules cm^{-2} and standard deviation is 0.43×10^{15} molecules cm^{-2}, respectively."

- Obtained AMF: Still confusing what you obtain. Can you provide a plot/timeseries of your obtained AMF against the input surface reflectance for one data set or just one flightline? As indicated your AMF should be strongly correlated with the surface reflectance.

Response: Fig. R6 shows the correlation between AMF and surface reflectance. We discovered that there is strong correlation between AMF and surface reflectance. As a result, the difference in NO_2 distribution was clearly revealed when high resolution surface reflectance data were used, especially over road and industrial area with high NO_2 VCD.

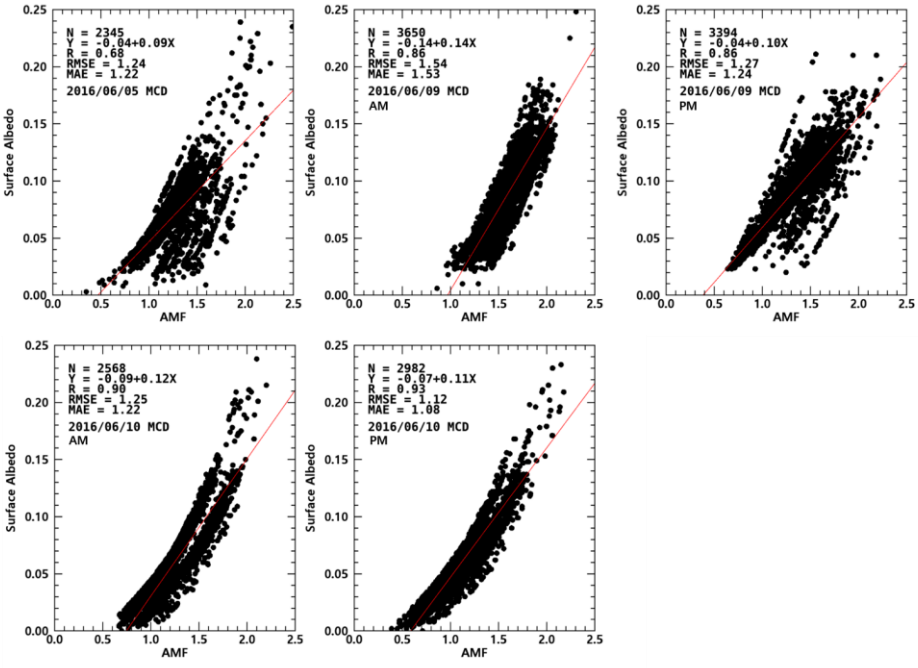

Figure R6. Scatter plot between (x-axis) AMF and (y-axis) surface reflectance.

2.

Review of: Highly resolved mapping of NO_2 vertical column densities from GeoTASO measurements over a megacity and industrial area during the KORUS-AQ campaign

We thank the reviewer's advice and agree with you for pointing out. In this reviewing process, according your thankful advice, we recalculated all NO_2 AMF using high resolution surface reflectance data. Furthermore, we selected optimal spectral range for DOAS fitting via sensitivity test. After that we also recalculated all NO_2 SCD under from 435 nm to 475 nm. As a result, almost figures were changed, and the output results were also changed (Fig. 5-8 in manuscript).

Major comments

Comment 1 (VCD stripe in Fig. 5a):

The surprising VCD values close to the Boryeong power plant (latitudinal stripe at 126.4°E) have not been improved and not commented, see Fig. 5a. It is not clear, how the stripe of large VCDs evolve from the given SCD and AMF. The SCD shows well the strong plume from the Boryeong power plant, while the VCD is dominated by artefacts. Even if the AMF calculation is never perfect, it should be clarified where this problem evolves from. Even if the data cannot be improved, it is recommended to state the cause of the artefact and a reason for not correcting it.

(The explanation and Figure R1 in the AC2 supplement would be more helpful if they were in better quality (the maps are somewhat hazy and blurred), and ideally with the same aspect ratio and grid lines as Fig. 5.)

Response: Following your advice, we examined the results of AMF calculation and we found some issues which is the fact that the AMF over some region is calculated using negative value of surface reflectance.

In this reviewing period, we recalculated all AMF using high resolution surface reflectance data (MODIS MCD43A3) instead of the previous MxD09CMG. But in MCD43A3, there are many pixels that do not provide surface reflectance, so the area where VCD cannot be calculated has increased compared to the previous results (Fig. R1 in review, Fig. 7 in manuscript).

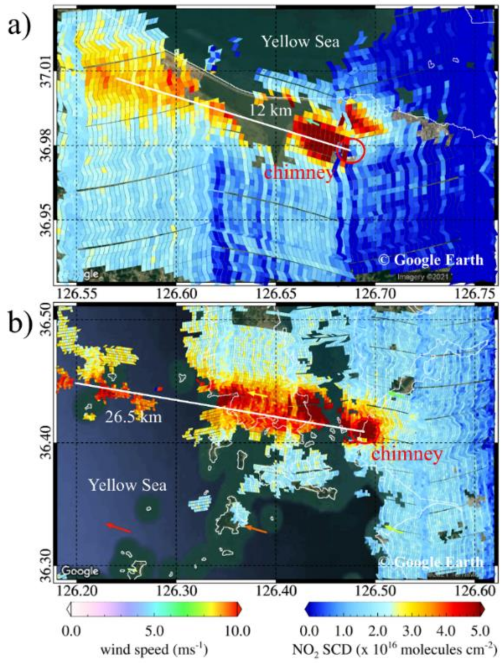

Figure R7. Enlarged view of GeoTASO tropospheric NO_2 VCD observation over a) Hyundai steel works, indicated by the red box in Figure 6, and b) the Boryeong power plant, indicated by the white box in Figure 6. The arrows represent the wind direction and speed at 850 hPa from the Unified Model (UM) simulations, respectively (background RGB image is from Google Earth; https://www.google.com/maps/).

Comment 2 (treatment of GeoTASO resolution):

One open question is the resolution of GeoTASO and how this is advertised and used in the manuscript. The authors' explanations seem to motivate that the fine resolution is not necessary (e.g., emphasizing the reduction of random uncertainty at coarser resolution), in contrast to the advertisement of the GeoTASO 250 m resolution. The fine resolution is suitable for detection of the largest NO2 values. Larger spatial pixels always dilute the signal. Clearly, the 0.01° resolution also shows the industrial enhancements and variability of NO2, although the 250 m resolution is even more suited for smaller scale variations. In any case, it is acceptable to use 0.01° resolution throughout the manuscript, however, in that case the 250 m should not be advertised in the abstract without any restriction. At least some modification of the text is recommended, if the authors decide not to show any native 250m resolution NO2 data. From the abstract:

"For the first time, we examine highly resolved (250 m × 250 m resolution) tropospheric NO2 over the Seoul and Busan metropolitan regions, and the industrial regions of Anmyeon."

Could be changed to something similar to this:

"For the first time, we examine highly resolved tropospheric NO2 over the Seoul and Busan metropolitan regions, and the industrial regions of Anmyeon. The native 250 m × 250 m resolution data of the instrument is binned to 0.01° resolution for the analysis of NO2 presented here."

p. 10, ll. 249-252: Here, the authors include some modified text, and this states one of the disadvantages of the binning: "… larger VCDs at 250m resolutions do not necessarily lead to larger VCDs at wider resolutions." So some larger signals are diluted, but this is a trade-off with

better signal quality.

Overall, if the native data is not the data shown and analysed, the reasoning for which data resolution is used should be stated very shortly once at the beginning with the main aspects for the choice, advantages and disadvantages, making clear that the final choice is finding a balance between the well-known influences (more details and stronger variability in the finer resolution, better data quality while still retaining a large portion of the variability at slightly coarser resolution).

Response: Thank you for your kind advice. We represented the pictures of all the results at 250 m × 250 m resolution. For this reason, it is considered that there is no resolution related problem to spatial binning.

Comment 3 (variability of AMF parameters on GeoTASO scale):

In the error analysis, the authors do not take into account the error that results from the fact that the values of AOD, ALH, SSA and SR used for AMF determination might not fit to the small GeoTASO ground pixel. Although the uncertainties might be suitable for the data products used, the question on how representative these values on a larger spatial grid are for the finer GeoTASO pixels, is not answered. The authors now mention that there is an additional error resulting from this aspect, but some further clarification would help.

p.19, ll.406-408: Here, it should be clarified which grid box is referred to. For example: "…variability of the parameters within the respective CTM or satellite grid box."

Please add a sentence directly after the above in the sense of:

"If values such as surface reflectance are assumed constant over larger areas, the fundamental spatial variability in this input data increases the uncertainty of the AMF and hence of the determined NO2 VCD on the respective finer spatial scale."

An idea of the variability of the surface reflectance of GeoTASO measurements within a MODIS grid box can be gained by looking at the relative intensity of the GeoTASO spectral data within an area, for which constant SR is assumed. Even if no specific calculations or refinements of the input data are made for the current manuscript, this influence can be roughly estimated.

Response: As you suggested, we have modified the manuscript on P. 17, Line 425-426 and added to P. 17, Line 426-428.

Minor comments

p.7, l. 186: Considering the response to the earlier question regarding the GDF spread, the numbers are still surprising. A spread in GDF is expected, but in the author's response the FWHM at 438.4 nm varies only between 0.8788 nm and 0.8985 nm. Does the GDF vary that strongly (0.7 to 1.0 nm as stated) at different wavelengths? This is a bit surprising, if variation is that small at 438.4nm. The table given in the supplement to AC2 is not needed in the manuscript.

Response: In fitting range (435 nm – 475 nm), the GDF vary from about 0.88 nm to 0.92 nm. But it isn't changed strongly as you mentioned.

p. 8-9, ll. 197-202 (Figure 3 and caption): Description and label of the x-axis (wavelength / nm) is missing. This information needs to be added.

Response: Because the fitting range has changed, we redraw Fig. 3 (now Fig. 4) and added a label of x-axis.

p.9, ll.224-227: The discussion of the NO2 VCD retrieval still misses some explanation. If the NO2 SCD used in the equation is the original SCD from the DOAS retrieval, part of the stratospheric SCD is already taken into account, because it is part of the reference measurement South of Jeju as well as of the current spectral measurement. The earlier comment here (originally ll. 182-185 in the original manuscript version), did not ask for new data or new model calculation, but simply for mentioning the correct relevant factors in the text. Since the retrieved SCD is the difference between the actual measurement and the reference measurement, the stratospheric, free tropospheric and background NO2 from the reference region are effectively subtracted from each measurement. Only the changes in time and space of these values are still included in the final NO2 product. These aspects of the retrieval and the assumptions behind should be stated in the text. The original question addressed the changes of these values, especially the changes in the stratospheric part over time. The stratospheric NO2 during the morning reference is different from the stratospheric NO2 during the GeoTASO measurement time. This change in stratospheric NO2 could be mentioned (and potentially quantified) in the text, as well as the potential influence of changes in free tropospheric and background NO2.

Response: Thank you for your kind advice. We have added the sentence “However, in this case, it was hard for us to consider the temporal variation of NO_2 in stratosphere in this present study since NO_2 present in the stratosphere could not be removed.” (P. 9, Line 251-253)

p.19, l.409: The values AMF_true and AMF_new are not explained in the text. Maybe “centre” or “mean” would be rather suitable than “true” and “perturbed” would be rather suitable than “new”?

Response: As you suggested, we have modified the manuscript on P. 16, Line 399-401.

Why is the change in AMF divided by the arithmetic mean of AMF_true and AMF_new, while the AMF_true is the point of reference? This way, changes to larger AMF (AMF_new > AMF_true) always cause smaller relative differences than changes to lower AMF. This is not clear.

Response: We misrepresented the existing Eq. (14). The revised formula is written in P. 17, Line 429.

After modification (P. 17, Line 429):

“$$AMF_{percent\_change} = \frac{AMF_{perturbed} - AMF_{centre}}{AMF_{centre}} \times 100 \quad (14)$$ ”

References:

p. 8, l.190: Probably this point was overlooked in the revision. It would be recommended to add a reference (a citation) for the applied H2O cross section.

Response: We have added “H_2O (Rothman et al., 2010)” to P. 8, Line 214.

Rothman, L. S., Gordon, I. E., Barber, R. J., Dothe, H., Gamache, R. R., Goldman, A., Perevalov, V. I., Tashkun, S. A., Tennyson, J. HITEMP, the high-temperature molecular spectroscopic database. Journal of Quantitative Spectroscopy and Radiative Transfer, 111(15), 2139-2150, 2010.

p.19, ll. 404-408: There are also earlier papers that analyse the NO2 profile influence on the AMF error.

Response: We have added the references to P. 17, Line 421-422.

Technical Corrections:

p.18, ll. 384-385: Typo in the value for SSA. Is the uncertainty really only 0.001? In that case the value should read 0.980 (0.001). The same number of digits is required for value and uncertainty. Possibly, there is a typo in the value of the uncertainty, which is probably much larger?

Response: The sentence was deleted because it was already in table 1 added to the manuscript.

p.21, l. 439: some typos, sentence should read “… It is thought that the reason for the low slope …”

Response: P. 19, Line 429 have been modified.

p.21, l.438: Typo in GeoTASO, here it says “GeoTAOS”.

p.22, l. 443: Same typo in GeoTASO.

Response: P. 19, Line 458 and 463 has been modified.

Please change sentences with “I” as subject to “We” or even better to passive speech. (e.g. p.22, l. 455 and others).

Response: “I” has been replaced with “We” on P. 19, Line 475.