Response to Anonymous Referee #2

We want to thank the referee for the constructive examination of our paper entitled "Horizontal distribution of tropospheric NO_2 and aerosols derived by dual-scan multi-wavelength MAX-DOAS measurements in Uccle, Belgium".

Please find below our responses to each comment individually.

Please consider that:

A) Green bold: Comments of the Referee

B) Black: The response to each comment posed by the referee.

C) Black bold: Already existing text in the manuscript.

D) Red bold: Added text in the manuscript according to referee's comments.

General comments

A. Dimitropoulou et al. 2021 present an interesting and new way to retrieve horizontal trace gas and aerosol profiles from MAX-DOAS data measured at various geometries and wavelengths. The approach utilizes data from profiling algorithms, further RTM calculations, and makes several assumptions on the atmosphere and the spatial distribution of its absorbers.

I recommend the publication of this manuscript after addressing some critical comments, because of the interesting idea rather than the convincing approach/results.

Before I explain further on the issues below, please let me give you my personal opinion on the approach and its assumptions. You start with the calculation of vertical profiles of MAX-DOAS measurements and calculate MLH. Even though validated, a MAX-DOAS profiling algorithm is not the truth but a smoothed representation of the true atmosphere constrained by the limited vertical resolution. The calculation of MLH is again an assumption of a box like distribution even though it was shown with MMF before that the profile was most likely not box-like at all. Then, further assumptions about a homogeneity of the MLH, calculated effective light path lengths and AODs are made (e.g. aerosol parametrization, profile shapes with percentages of bulk load based on statistics, fix values of all input parameters). The estimation of VMR based on Sinreich et al. 2013 is again just an approximation which is only valid under certain conditions. And then, everything is used in another inversion step. Knowing all this, it appears to be a miracle that your correlations are still good! To remove some of my doubts, please assess the error budget and the propagation of errors between the individual steps in more detail.

Response: First, it is important to address this general comment concerning the assumptions made in the different steps of the proposed retrieval methodology.

The retrieval of profile information from MAX-DOAS measurements is typically made of several assumptions (i.e., the a priori profiles, the layering of the atmosphere, aerosol properties and other retrieval parameters). Given the fact that the retrieval problem is under constrained, these assumptions cannot be avoided.

To validate these retrievals, which is a crucial step, we perform validation studies based on available independent correlative datasets at the measurement site.

For the present study, we would like to underline that the application of the parameterization technique of Sinreich et al. 2013 has been widely used in the literature for pollution conditions similar to those encountered in the Brussels-Capital Region. In Dimitropoulou et al. (2020), we have validated the retrieved MAX-DOAS near-surface concentrations of NO_2 by performing a comparison with the in-situ stations located in the Brussels-Capital Region. For this reason, we have not performed again a similar comparison.

Additionally, the error budget is now estimated in more detail compared to the initial version of the manuscript. For instance, concerning the estimated error of the new OE-based inversion approach, we present the smoothing error in addition to the measurement error (see Figure 16).

The new Horizontal Distribution inversion approach has been validated by using independent remote sensing measurements (i.e., airborne and car-mobile DOAS) during one day of observations. Additionally, to this one-day comparison, we have added a comparison concerning the year 2018 between RIO and MAX-DOAS near-surface NO_2 concentrations in the Brussels-Capital Region (see Section S1). The main reason of not including this comparison in the principal manuscript is that the a priori NO2 horizontal profiles are estimated with the aid of the RIO model. Consequently, the RIO model is not a completely independent dataset to perform a proper validation.

In a future study, it is important to apply this novel approach in different measurement sites, in which more independent correlative datasets are available than those used in the present work.

B. Three further issues of this manuscript need to be solved before final publication and will be addressed in detailed below: 1. This manuscript is much too long. 2. The presented approach is not validated sufficiently. 3. The explanation of the approach in Section 4.2.1 needs a revision.

Response: Please consider the provided answers and modifications to our manuscript according to each of your specific comments below.

Details on the three main issues:

1. **Nowadays, many paper show more and more results which are sometimes extremely insignificant or do not match the purpose of the publication. The main purpose of this manuscript is the introduction of a novel approach about the retrieval of horizontal absorber concentrations. A manuscript with this topic should introduce and explain the novel approach and show a thoroughly performed validation study. In this manuscript, you also address the question of how to optimize and validate Tropomi measurements. This has nothing to do with the content of your paper and is addressed in more detail elsewhere. So please remove Section 5.3.2 fully (or move it to the supplement) and remove parts of section 5.3.1! E.g. the three concluding points on page 40 can be removed as they are already content of the conclusions. When you add a proper validation (see point below), please move Section 4.2.3 to the supplement.**

Response: Following the above-mentioned recommendations, significant modifications are made to the present manuscript. First, we have added a comparison between the RIO air-quality model and the MAX-DOAS horizontal profiles (Section S1). Secondly, in Section 5.3, we present the comparison between TROPOMI and MAX-DOAS retrievals and the previously numbered Section 5.3.2 has been moved to the Supplement as Section S2. Finally, Section 4.2.3 has not been removed from the manuscript because it concerns a verification of the dual-scan MAX-DOAS retrieval method and not a validation.

2. **Validation of a novel approach is a necessary step but needs independent measurements! Validation can never be done with the same instrument. Since you already mentioned the in-situ air quality network in Brussels, I would recommend the use of this data or another independent data set. However, no validation of other instruments/data sets by your novel approach can be shown without first validating itself! This means of course that validation of Tropomi data with your approach is not appropriate. It would make more sense to validate/verify your approach with an already validated Tropomi data set.**

Response: We agree that a validation of a novel approach is necessary and crucial. For this reason, we have performed a comparison between the air-quality model dataset and the horizontal profiles derived by the new OE-based inversion approach. This comparison can be found in Section S1 of the manuscript, in which hourly NO_2 concentration maps from the RIO model are compared with hourly averaged NO_2 horizontal profiles in the Brussels-Capital Region. However, the comparison between profiles retrieved using the standard OEM approach and the parameterization results (see Section 4.2.3) is an important sanity check for the consolidation and verification of the dual-scan MAX-DOAS retrieval method presented in the present manuscript. It should be noted again that this comparison is not a real validation since the two datasets are not independent since we use the information about the MLHNO2 in the dual-scan MAX-DOAS retrieval method.

3. **It was difficult to understand your explanation in section 4.2.1 because it is not directly clear for the reader which quantities are fitted how and when. Please revise this section and explain specifically which measured or inverted quantity goes in which polynomial-fitting or RTM-calculation step. Maybe a small flowchart would help or adapt and refer to the existing flowchart in Fig. 3. Furthermore, Fig. 4 and Fig. 5 show simulated columns. Please add also an example with real measurements and the polynomial fit in the same figures.**

Response: In Section 4.2.1, we have added additional information, which can help the reader understand the developed dual-scan MAX-DOAS retrieval method. The additional changes are the following:

1. **Here, NO_2 dSCDs and consequently L_{NO2} are simulated using the radiative transfer model VLIDORT version 2.7 (Spurr, 2006).**
2. **The MLH_{NO2} is a known parameter and it is estimated per measurement scan, as the ratio of VCD_{NO2} to the NO_2 near-surface concentration as retrieved in the main azimuthal direction by the MMF inversion algorithm.**
3. **For the six different wavelengths (343 nm, 360 nm, 380 nm, 447 nm, 477 nm, and 530 nm), we separately perform RTM simulations and L_{NO2} (see Eq. 5) are simulated for the assumed SZA, RAA, MLH_{NO2}, c_{NO2}, and AOD input scenarios presented in Table 2.**

4. **The simulated O_4 dSCDs are a function of the input parameter AOD. The relation between the simulated O_4 dSCDs and the input AOD values is shown in Fig. 4a.**
5. **Additionally, in Fig.4b, we can see that the relation between the O_4 dSCDs and the AOD values is valid for MAX-DOAS measurements.**
6. **We observe, also, in Fig. 5b, in which an example day of MAX-DOAS measurements is presented, that the L_{NO2} as a function of the measured O_4 dSCDs have the same relation as the simulated quantities.**
7. **Since NO_2 is an optically thin absorber, L_{NO2} is not a function of c_{NO2} and consequently, a L_{NO2} value can be estimated by using the measured O_4 dSCD for each measurement.**
8. **Based on this approach, the near-surface NO_2 concentration can be calculated at the six different wavelengths by using the measured $dSCD_{NO2}$ together with the simulated L_{NO2} value (Eq. 2).**

Additionally, in Figures 4 and 5, an example with real measurements and their polynomial fit is included as well.

Specific comments

1. P3, L88: Please write "telescope azimuth angle" so that the abbreviation (TAA) makes more sense.

Response: The text has been modified according to this comment:

… **(1) a vertical scan in nine different elevation angles (EAs) in one fixed telescope azimuth angle (TAA; Northeast direction i.e., towards the city center and the national airport)** …

2. P3, L89: Why did you use 2° instead of 1°? (see also P9, L219)

Response: The primary motivation of using the elevation angle of 2° instead of 1° is to avoid obstacles across the line of sight of the different azimuthal directions used in the present study. According to Sinreich et al. (2013), the parameterization technique is valid for low elevation angles. In their study, they have tested the elevation angles of 1° and 3°. In Ortega et al.'s (2015) study, the parameterization technique is also applied for an elevation angle of 2°.

3. P3, L92: Remove "directions". It is not needed.

Response: The word “directions” has been removed from this sentence:

The selection of more azimuthal directions towards the North, Northeast, and Northwest ~~directions~~ was made considering the location of the main NO_2 emission sources and, consequently, the highly variable NO_2 horizontal distribution towards these directions.

4. P3, L94: Integration time of 60s? Why was it set to such a long time? Depending on the wind speed, many things can happen within one minute in an area with that strong spatial inhomogeneities!

Response: By changing the experimental set-up of the MAX-DOAS instrument in Uccle from one to multiple azimuthal directions, our aim was to study the horizontal distribution of aerosols, NO_2, and HCHO. The selected experimental set-up is a trade-off between acceptable S/N ratio and having a maximum number of azimuthal and elevation directions.

HCHO is a weak absorber and for this reason, we had to adopt an acquisition time of the spectrum large enough to ensure that the measurements are not too noisy. HCHO results are not presented in this study but you can find the study in the following link:
https://ieeexplore.ieee.org/abstract/document/9553326

5. Figure 1: When I remember correctly, you will never again talk about the individual tests shown in this plot. So please remove these tests and show only the applied azimuthal directions. Please zoom slightly in and show the power plants, ring freeway, and MAX-DOAS site, similar to Fig. 18.

Response: The period we have used for the present study extends from March 2018 to February 2020. Consequently, different experimental set-ups are exploited during this study and we think it is important to make it clear to the reader. For this reason, we have decided to show the individual experimental set-ups in Figure 1. Additionally, we have modified this figure to show the main NO_2 emitters in the Brussels-Capital Region and the figure's caption accordingly.

Figure 1. The experimental set-up of the BIRA-IASB dual-scan MAX-DOAS instrument. Each line is color-coded according to the different set-ups that were used from March 2018 to February 2020. The length of each line is equal to 20 km, which corresponds to the typical horizontal sensitivity for the MAX-DOAS measurements in the present study (see Fig. 18). The black square shows the MAX-DOAS instrument location, the black polygon the National Airport, the black dots the NO_2 hotspots emitting more than 10 kg of NO_x per hour (Emission Inventory of the Belgian Interregional Environment Agency, 2017) , and the black line represents the Brussels Ring motorway.

6. Section 2.2: Please explain how you decided on these wavelength intervals. Did you do some optimization for the shown fit settings? For example, I was wondering why the window 510-540nm was chosen like that even though large H_2O absorption is present at the start and end wavelengths. These absorption features might also explain slightly larger residual structures (compare Fig. 6).

Response: The main criterion of deciding these wavelength intervals was to include, in each of them, one of the main maximum O_4 absorption bands that are available in the UV-Visible wavelength range. The choice of the optimal wavelength interval to retrieve the trace gas of interest is a compromise between maximizing the sensitivity of the trace gas and minimizing interference with other absorbers and the presence of residual structures in the DOAS fit results.
Concerning the last fitting window (510-540 nm), H_2O is included in our fit.

7. P5, L116: Why are not all reference wavelengths in the peak center of their corresponding O_4 absorption bands?

Response: The reference wavelength, in each chosen fitting window, corresponds to the maximum of O_4 absorption peak or close to this maximum. In some windows, the reference wavelength is not exactly at the maximum because a better fit in terms of RMS is achieved.

8. P5, L123-L125: You said that the O_4 cross section of Finkenzeller improves results in the UV. Does it show any change in the visible range? Why did you decide against using it here as well?

Response: According to the presentation of H. Finkenzeller at the DOAS workshop in 13 July 2020, the main difference between the O_4 cross section of his study and the previously published O_4 cross section (Greenblatt et al., 1990; Thalman & Volkamer 2013; Hermans et al 2011) in the wavelength range of 308-500 nm is the transition at 344nm. More precisely, in the study of H. Finkenzeller, the O4 cross section values before and after the peak at 344 nm are between the O4 cross section values of Greenbalt et al., (1990) and Thalman & Volkamer (2013). Following this finding, we have decided to use the O_4 cross section of Finkenzeller only in the UV wavelength region.

9. P7, L172: "measured radiance spectra ... is analyzed" to "are analyzed"

Response: Corrected.

10. P9, L195: Please cite some of the "several studies" you are referring to.

Response: We have added three related studies. The interested reader is now referred to Table 1 of Wagner et al. (2019) for more studies.

11. P9, L196: I would write of a scaling factor ≠ 1 because some studies suggest also larger scaling factors depending on spectral range, location and season.

Response: The sentence has been corrected.

12. P9, L208-L209: Why do you accept retrievals with homogeneous cloud coverage? The corresponding aerosol profile is wrong for sure! This means that your MLH is inaccurate because is is negatively affected by the wrong radiative transfer. All your RTM calculations of L_{NO_2} are wrong as well and, therefore, your horizontal profiles!

Response: The effects of the clouds on trace gas retrieval are very important when broken clouds are present because during an elevation scan, some measurements are influenced by clouds while some others are not depending on the elevation angle. Under homogeneous cloud cover, the quality of MAX-DOAS are much less affected by clouds (see e.g. Gielen et al., 2004 and Wagner et al., 2015). For this reason, we have chosen to only filter out elevation scans that are measured under broken cloud conditions.

13. P9, L219: Why did you select an elevation angle of 2°? I would assume that 1°? is better suited for your purpose and assumptions about homogeneity are closer to the truth. (see also P3, L89)

Response: This comment has been addressed in comment no. 2.

14. P10, L251: Please move the full stop from the index of NO_2 to the normal level.

Response: The text has been modified accordingly.

15. P11, L276: "Here, O4...". If "here" refers to the equation above it should be "Here, NO2..."

Response: The sentence has been modified as follows:

Here, NO_2 dSCDs and consequently, L_{NO2} are simulated using the radiative transfer model VLIDORT version 2.7 (Spurr, 2006).

16. Section 4.2.1 Please revise according to the general comment.

Response: Following the general comment, we have revised Section 4.2.1, but it should be noted that it is still included in the main manuscript.

17. P13, L324: "As discussed above, around 30%...". I would not call this a real discussion. You just mentioned it without showing any results of the analysis.

Response: The sentence has been modified as follows:

As mentioned above, around 30% of the total aerosols is expected to be found inside this layer.

18. Fig. S4, S5: Please discuss the dependence on SZA and RAA for Fig. S1 and S2, respectively, as well. I would assume that a significant contribution of the dependence is due to aerosols and the applied phase-function.

Response: The dependence on SZA and RAA for Figures S1 and S2 has already been discussed in Dimitropoulou et al. (2020). The difference is that we present the L_{NO2} directly in the present manuscript. Only the correction factors were presented in Dimitropoulou et al. (2020) (Sinreich et al., 2013).

More precisely, for small RAA, a large correction factor value is found, which means small values of L_{NO2}. L_{NO2} increases with the RAA. The dependency of L_{NO2} with SZA is linear until SZA reaches the value of 45°, where it increases rapidly. This result, together with the correction factor values for SZA values in the range of 45°-80° (Figure 2 in Dimitropoulou et al., 2020), are indicators concerning the limitations of the parameterization technique for low sun conditions.

19. P13, L335: "less pronounced Rayleigh scattering" to "less pronounced Rayleigh and Mie scattering"

Response: Corrected.

20. Fig. 6: Please explain why the smaller deviations of L from the polynomial fit propagate into much larger deviations for the near surface concentration and vertical column density.

Response: In Figure 6, we can see that the near-surface NO_2 concentrations at 360nm and 380 nm deviate more from the polynomial fit than the other points at larger wavelengths. Consequently, the L_{NO2} at 360nm and 380nm (Figure 6a) are the points that deviate the most from the polynomial fit.

The error related to these near-surface NO_2 concentrations reflects this deviation.

21. Eq 6 and 7: The step from Eq. 6 to 7 can not happen without further assumptions. I would rather prefer the calculation of the partial derivative than this assumption. Please explain this step in more detail. How is the uncertainty of the O_4 dSCD used?

Response: Between Equations 6 and 7, we have included the intermediate step for the calculations of the error.

The uncertainty of the O_4 dSCD is not included in the error calculation of this study. According to Dimitropoulou et al. (2020), the O_4 DOAS fit error is up to 5 % and 6 % in the visible and UV range, respectively.

As first error source, we take into account the uncertainty related to the NO_2 dSCD. As second error source, we include the uncertainty related to the estimation of the L_{NO2} from the RTM simulations. As third error source, we include the error related to the MLH_{NO2} estimation (see Dimitropoulou et al., 2020).

The error source related to the MLH_{NO2} estimation is included only in the error estimation of the near-surface NO_2 concentration and not in the VCD_{NO2} because it would be accounted twice and conduct to a falsely larger error budget for the VCD_{NO2}.

Please find the modifications in Section 4.2.2.

22. Section 4.2.3: This section needs to move to the supplement and has to be replaced by a real validation. The sanity check is interesting but should not be content of the main manuscript. The validation part is unfortunately no validation but rather a verification. Validation only works with independent measurements. Using just a different azimuthal direction from the same instrument is not at all independent. For a study like this, I would assume comparison with e.g. in-situ instruments. Please add a validation study from an independent instrument as e.g. in-situ data from the air quality network in Brussels.

Response: Section 4.2.3 has been named as: **Verification of the dual-scan MAX-DOAS retrieval method**

The present parameterization technique is an updated form of the parameterization technique used in Dimitropoulou et al. (2020), in which a comparison between MAX-DOAS and in-situ observations has been already performed. For this reason, in the present manuscript, we did not perform a comparison with the in-situ air quality network in the Brussels-Capital Region.

Additionally, in the revised manuscript, we now perform a comparison between the RIO air quality model dataset and the MAX-DOAS NO_2 horizontal profiles over one year of observations (now, Section S1).

Please consider our response about this section in the general comments.

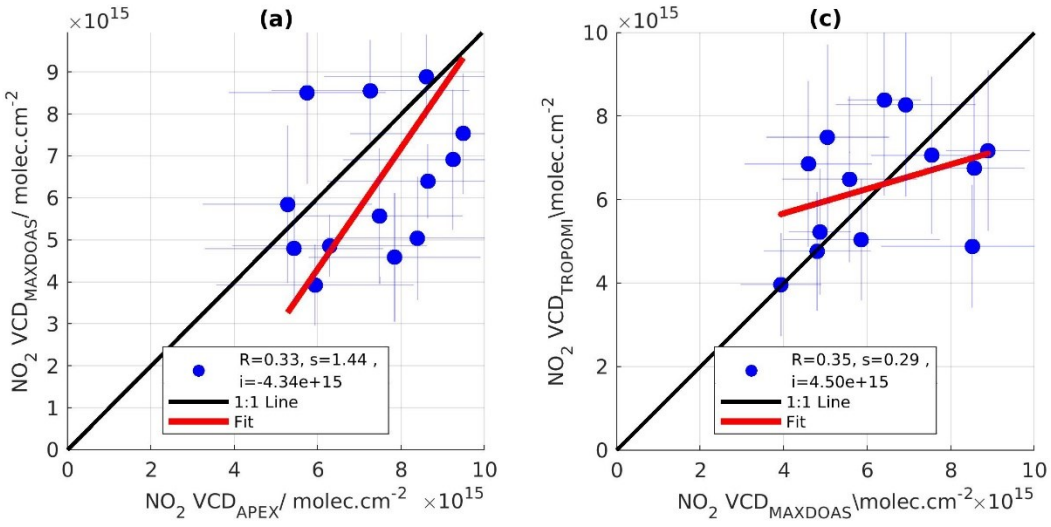
23. Eq 11: Please give more information on this approximation/definition. Is the weighting function for aerosol extinction coefficients defined in a similar way? With this definition, you suppress variability in the horizontal direction which means that you consider your effective light path lengths as perfect. I also have a problem with the fact that the weighting itself was arbitrarily defined as horizontal step width divided by whatever is found from your simulation of L. Is the information content not large enough to allow a more flexible implementation? Please discuss this further.

Response: First, the weighting functions for the aerosol extinction coefficients are defined in a similar way as for NO_2. Concerning the approximation/definition of the weighting functions, we have used a simple horizontal box model in which we assume a geometric approximation where the sensitivity along the horizontal distance per measurement is constant. The comparisons performed during the 28th of June 2019 show that the NO_2 horizontal profiles are in good agreement with ancillary data. Additionally, the comparison between the MAX-DOAS NO_2 horizontal profiles and the RIO model data show a good agreement, too. These findings indicate that the approximation concerning the weighting functions is reasonable and lead to realistic results.
In a future study, the calculation of more appropriate weighting functions, considering the horizontal variability of the light intensity along the horizontal dimension could be investigated.

24. P22, L502: How large is this mean scaling factor? You already add a bias here by applying a mean factor. It would be interesting to know if the unscaled a priori profiles would lead to a better agreement with ancillary instruments or if it just destabilizes your retrieval.

Response: As mean scaling factor, we are referring to the mean ratio between the six MAX-DOAS measurements (i.e., NO_2 near-surface concentrations as retrieved in the six different wavelengths) and the unscaled a priori profile in every measurement direction. Taking the example of 28th of June 2019, the scaling factor are within a range of 0.27 – 0.56. This finding is in agreement with several studies in which comparisons between remote sensing and in-situ instruments are performed (see e.g. Section 4.2 of Dimitropoulou et al., 2020).

If we do not apply these scaling factors, the comparisons between MAX-DOAS and airborne, and MAX-DOAS and TROPOMI for June 28, 2019 are the following:



As you can see, if we don't apply any scaling factor, a worse agreement is found between the MAX-DOAS horizontal profiles and the ancillary data.

25. Fig. 13 and 14: Please add two further examples for profiles with larger concentrations and examples with a small aerosol/NO_2 load. The readability should not suffer with two more curves in these plots. Please add the a priori profiles and errorbars for the aerosol horizontal profiles as well. In Fig. 14, you see a larger deviation of measured and simulated extinctions for the middle L_{O_4} values as well as for the first data point. Since the reader cannot see the a priori, it is hard to assess how this propagates into your retrieved profile. Please discuss this deviation together with the a priori profile and the retrieval errors.

Response: In Figures 13 and 14, we have added two additional examples with profiles that have larger and smaller NO_2 values and aerosol load, respectively. Additionally, a discussion concerning these two figures is included in Section 4.3.

26. Fig. 15: Since AK are the multiplication of the gain matrix G with your weighting function matrix K, your averaging kernels show the sensitivity based on your definition of the weighting function. Please show for this scenario the corresponding a priori profile and retrieval result. I would assume that the features of your AK matrix are strongly dominated by L_i and your a priori profile. It is difficult to understand why the sensitivity for blue and red are the highest at exactly the same distance (similar with purple and green). If this figure would be a good representation of the derivative of concentration with respect to the true concentration, individual peaks should be found at different distances. Please discuss!

Response: As it is stated in the present comment, the AKs are calculated by multiplying the gain matrix G with the Weighting function matrix. Figure 15 has been modified and is referring now to the a priori profile and retrieval result of Figure 13a.

Indeed, the features of the AK matrix depend strongly on the location of L_i and the a priori profile shape, as we can see in Figure 13a and Figure 15. For the first three sampling grids of Figure 15 (blue, red, yellow), the information is coming mainly by the two first measurements (i.e., two first L_i), which can explain the co-located peaks of these sampling grids.

27. P29, L601: "close their" to "close to their"

Response: corrected.

28. P29, L610-L611: Why should this be the case? What about the smoothing error? Since you have many constraints due to your a priori assumptions, I would assume that the smoothing error has a significant contribution to your total error.

Response: Indeed, the smoothing error has a significant contribution to the total error of the retrieval. In order to estimate the smoothing error, we have used RIO horizontal NO_2 profiles (after applying a mean scaling factor) to construct a covariance matrix.

The measurement error and the smoothing error are now shown in Figure 16.

29. Table 3: "Medium RMS" to "Median RMS". What do the DOFS and RMS values in brackets in the last row mean? If this refers to the total accepted retrievals, why did you write different thresholds in the text? Furthermore, it is hard to assess the range of RMS and DOFS values based on these numbers only. Please add a figure of the frequency distribution of RMS and DOFS values and discuss it together with this table!

Response: In Table 3, we have corrected *medium RMS* to *median RMS*.

Please consider a correction in the last row, which was written by mistake in the first version of the manuscript.

We have added Figure S7 and S8 in the Supplement showing the probability density function (PDF) of RMS and DOFS values for our study.

Additionally, in Section 4.4, the text has been modified as follows:

To eliminate the unsuccessful retrievals, the percentage of accepted retrievals with respect to the total number of retrievals during the four seasons is investigated when a specific filtering on RMS and DOFS is applied (see Table 3 and Figures S7 and S8). As we can see in Fig. S8, DOFS are in the range of 1.2-2.5. From these tests, it is found that most of the retrievals have DOFS larger than 1.5 (see Fig. S8). RMS is defined as the root-mean-square deviation between measured and simulated c_{NO2} normalized by the mean of the measured c_{NO2} (e.g., same RMS as in Fig.10). Table 3 and Fig. S7 indicate that RMS values are in the range of 0-30% and most of the retrievals have an RMS smaller than 6% with a

median RMS value of around 4.5% during all seasons. Based on these investigations, DOFS>1.5 and RMS<6% are used as retrieval quality control criteria.

30. Fig. 18: Please zoom in and increase the quality of this figure so that details can better be seen. Please add a similar plot showing the near-surface concentration/extinction in the supplement. Do near-surface values support your finding that air pollution in Brussels is mainly driven by the power plant and traffic emissions? Can larger values be found in the distance of the Ring-motorway?

Response: Figure 18 has been modified according to this comment. Additionally, the wind direction as measured in the meteorological station at the rooftop of BIRA-IASB has been added to each subplot.

In the supplement, you can find two new figures: one for the near-surface NO_2 concentration (Figure S5) and one for the near-surface aerosol extinction coefficient (Figure S6).

In Section 5.1, we have added the following sentences:

Same NO_2 horizontal distribution is found when investigating the NO_2 near-surface concentrations for this day (see Fig. S5).

and,

Maximum near-surface aerosol extinction coefficient values are observed during all day long and detected in the N, NW and NE direction (see Fig. S6). NO_2 and aerosol peaks are co-located towards the N and NW direction.

31. P35, L698: Why are the segments not weighted? If there is just a tiny fraction of light path within a pixel, it should not be weighted with a similar factor as contributions with much longer light paths. Please change this or discuss why it is not possible/reasonable.

Response: Following your comment, we have weighted the MAX-DOAS segments by their relative length inside each TROPOMI pixel for the results presented in Section 5.2 and Section 5.4.

The text has been modified as follows:

It should be noted that the MAX-DOAS segments are weighted by their relative length inside each pixel.

32. Fig. 19: Please change the range of colors for subfigure a) so that the plume is better visible. For the original data of APEX and the AEROMOBIL I would not assume the same color-scale. However, subplot b), c) and d), should have the same color-scale! Please change this.

Response: Figure 19 has been modified according to this comment.

33. P35, L706: How do you explain this intercept?

Response: Given the fact that with the dual-scan MAX-DOAS parameterization technique, we are sensitive to the near-surface NO_2 layer, it is possible that during this day, the APEX (airborne) instrument detected an additional NO_2 layer in an altitude higher than the estimated MLH_{NO2} (which is around 700m).

Another possible explanation could be the different measurement techniques of both instruments (ground-based and airborne) and their different sensitivities as well as the different reference spectrum used in the DOAS analysis of both measured spectra.

34. P35, L709: "channels" to "elevation angles" or "geometries"

Response: The sentence has been corrected.

35. Fig. 20b: Please correlate the not averaged data as well by comparing MAX-DOAS and AEROMOBIL data points which are close to each other. 6 data points are not statistically significant enough for such a comparison. Especially not when you can compare data in higher resolution.

Response: We have performed a comparison between the not averaged MAX-DOAS and AEROMOBIL data for 28 June 2019, which is shown in Fig.17b. Moderate correlation coefficient is found between both datasets (R=0.74) and a slope equal to 0.55.

36. Section 5.2: Please add the exact overpass times you used for the comparison of TROPOMI and add a reference to Fig. 18 in the text so that the reader can also compare the MAX-DOAS data in higher resolution at the overpass times with Fig. 19. Furthermore, add the start and end time for the AEROMOBIL measurement in the text and the caption of Fig. 19.

Response: The exact overpass time used for the comparison of TROPOMI has been added to the text as follows:

During this day, the TROPOMI overpass time was at 12:19 UTC.

A reference to Figure 18 has been added so that the reader can compare the MAX-DOAS data at the overpass time with Figure 19 as follows:

During the TROPOMI overpass (i.e., 12:19 UTC) above the Brussels-Capital Region, dual-scan MAX-DOAS tropospheric NO_2 columns are retrieved, as it can be seen in Fig. 18. The correlation between TROPOMI and MAX-DOAS tropospheric NO_2 columns during the day of the airborne measurements above Brussels is presented in Fig. 20c.

The start and end time for the AEROMOBIL measurements have been added to the text as follows:

The AEROMOBIL was used to measure the spatial distribution of tropospheric NO_2 columns mainly over the Ring road of Brussels with start measurement time at 8:30 UTC and end time at 15:42 UTC.

And the caption of Fig.19 had been modified as follows:

Figure 19. **(a) Tropospheric NO_2 VCD as detected by the APEX instrument in its initial spatial resolution. Tropospheric NO_2 VCD maps (TROPOMI pixels) as retrieved over Brussels on 28th of June 2019 by the (b) APEX, (d) MAX-DOAS and (f) TROPOMI (overpass time at 12:19 UTC) instruments. Tropospheric NO_2 VCD as retrieved by the (c) MAX-DOAS and (e) AEROMOBIL (between 8:30 UTC and 15:42 UTC) in its initial spatial resolution. The black square shows the MAX-DOAS instrument location, the black polygon the National Airport, the black dots the NO_2 hotspots emitting more than 10 kg of NO_x per hour (Emission Inventory of the Belgian Interregional Environment Agency, 2017) , and the black line represents the Brussels Ring road.**

37. Section 5.3.1: In addition to my general remark about the purpose of this document, I would like to ask you to change this section either to a "validation of the new algorithm" or to a "comparison only" section.

Response: Section 5.3.1 is now Section 5.3 entitled as:

Comparison of MAX-DOAS horizontal NO2 distribution versus TROPOMI observations

38. Fig. 21: By just looking at this plot, I would say the agreement is not that good. Why is the difference in the SW direction so large in Summer and Winter? Why are the values close to the MAX-DOAS site much higher for the MAX-DOAS data in Spring? This is especially interesting because your algorithm was described of having a poor sensitivity close to the instrument (compare P26 L599-L600)!

Response: If we observe Figure 22 and Figure 23 (i.e., seasonal results), one can see that indeed summer and winter show a less good agreement compared to spring and autumn. In Figure 22, in the SW direction, we obtain the highest positive bias (around 9%), and close to the instrument, the lowest bias (around – 36%).

Several studies (cited in the manuscript) have reported the same underestimation of tropospheric NO_2 columns as observed by TROPOMI when compared to ground-based remote sensing ones. Consequently, our findings is in agreement with previous studies.

With the phrase “poor sensitivity close to the instrument”, we aim to describe that in the first kilometers (i.e., starting from the instrument until the first measurement), the information of the tropospheric NO_2 columns in the horizontal profiles comes only from the first MAX-DOAS measurement and consequently, the horizontal distribution is highly influenced by the a priori NO_2 profile.

A possible explanation about the spatial distribution of the bias between TROPOMI and MAX-DOAS could be the change of the MLH_{NO2} along each line of sight, which is related to the elevation angle of observation and the horizontal sensitivity.

For our study, several factors, that can be improved in future studies, play an important role in the MAX-DOAS retrieval such as the estimation of the MLH_{NO2} from one azimuthal direction, the non-homogeneously distributed azimuthal directions, and the assumptions of the NO_2 vertical profile as a box profile to estimate the near-surface NO_2 concentration.

39. P39, L780-L782: Similar to the comment above, I am wondering why there is a large negative Bias close to the instrument and a positive one for the pixels in large distance to the MAX-DOAS site? I was wondering if the values at the contour-legend are correct? Is the positive bias just smaller than 1%? If yes, this bias might be negligible. Please check the values and the figure and give an explanation for theses biases if possible.

Response: Please find our response concerning the bias in the previous comment.

Additionally, Figure 22 has been modified and the contour-legend is now correct with values ranging from -36% to 9%.

40. P39, L790: I would not talk of an under-/overestimation by one of the instruments if it is not clear which instrument/algorithm shows the more accurate results.

Response: This sentence and the following one have been modified as follows:

During spring, the slope value is equal to 0.90, while during winter, summer, and autumn, the slope values are smaller (0.64 0.56, and 0.64, respectively). Similar findings have been reported in several studies (Verhoelst et al., 2021; Tack et al., 2021; Judd et al., 2020; Dimitropoulou et al., 2020; Ialongo et al., 2019).

41. P40 L818-828: This belongs to the conclusion and should be removed from this section.

Response: This part has been removed from this Section and has been placed to **Section 6: Conclusions**.

42. Section 5.3.2: Remove this section from the manuscript or move it to the supplement (see general comments).

Response: Section 5.3.2 has been removed from the main manuscript and placed to the Supplement as Section S2.

43. P49, L953-954: and AODs

Response: The sentence has been modified according to this comment.

44. Conclusions: Please change the conclusions according to the general and specific comments.

Response: Section 6, which gives the conclusions of the present study has been modified according to the general and specific comments.

References

Sinreich, R., Merten, A., Molina, L., and Volkamer, R.: Parameterizing radiative transfer to convert MAX-DOAS dSCDs into near-surface box-averaged mixing ratios, Atmos. Meas. Tech., 6, 1521–1532, doi:10.5194/amt-6-1521-2013, 2013.