Response to Anonymous Referee #1

We want to thank the referee for the thoughtful 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:

1. **In my opinion a distinction between the VCD_NO2 and c_NO2 measured in main viewing direction (retrieved by traditional profile inversion) and the values retrieved in other azimuthal directions would help a lot for clarification (e.g. page 9 line210, page 10 equation 1, page 11 equation (4))**

   Response: The VCD_{NO2} and c_{NO2} derived by applying the MMF inversion algorithm (i.e., traditional profile inversion) are named as $VCD_{NO2,main}$ and $c_{NO2,main}$ (please see Section 4.1), while the VCD_{NO2} and c_{NO2} derived by applying the parameterization technique are kept the same.

2. **Inventing a new name for this new technique would help especially when comparing to other data-sets (e.g. in chapter 5.2). Usually, elevation scans are called "MAX-DOAS". Something like "Mapping MAX-DOAS" would help to distinguish between the different kind of retrievals.**

   Response: The ensemble of the developed methodology, as described in Section 4, is now called "Mapping MAX-DOAS technique" and the title of this Section is "**Description of the mapping MAX-DOAS technique**". The authors have made the necessary changes to help the reader to distinguish the different retrievals used in the present study. The changes are in the following sections of the revised manuscript: Abstract, Introduction, Section 4, Figure 3, Section 5.2, and Section 5.3.

3. **Is a seasonal analysis really useful for NO2? An analysis sorted for different temperature regimes could be more useful. Also, an analysis sorted for several wind regimes could be interesting, especially as 3 huge NO2 emitters are in the region.**

   Response: Our primary motivation for presenting a seasonal analysis for NO_2 is to allocate the three huge NO_2 emitters in the Brussels Capital Region on a seasonal basis with the aid of seasonal NO_2 maps. Based on this motivation, we present the comparisons between MAX-DOAS and TROPOMI on a seasonal basis, too.
   During the preparation of the present manuscript, we have investigated the effect of temperature on NO_2 VCD by using co-located temperature measurements, and no

temperature dependence is found for NO_2 (in contrast with HCHO; please see the article: https://ieeexplore.ieee.org/abstract/document/9553326).
The effect of wind direction on NO_2 VCD has been investigated in our previous publication. Generally, when the wind is blowing from the NE and SE direction, higher NO_2 are retrieved. The corresponding results can be found at: https://amt.copernicus.org/articles/13/5165/2020/.

4. **The RIO data is available in an hourly model. It would be interesting to see a comparison to this data set.**

   Response: In the revised version of the present manuscript, a comparison between the NO_2 horizontal profiles and the RIO data on an hourly basis is performed and presented in Section S1.

5. **In a future study the comparison could also be performed for AOD with the 1km resolution data from MODIS.**

   Response: In a future study, a comparison between the retrieved near-surface aerosol extinction coefficient and the AOD from MODIS could indeed be done for validating the retrieved dual-scan aerosol horizontal profiles.

6. **Harmonize axes and color bars of plots in one figure.**

   Response: Following the specific comments, we have harmonized axes and color bars of plots in the following figures: Fig. S1, S2, S3, S4, 12, and 19.

Specific comments:

1. **Page 2 line 29: If possible, restructure sentences for a better reading flow.**

   Response: The sentences have been rephrased as follows:
   Aerosols with a small diameter can penetrate deeply into the lungs, causing millions of premature deaths around the world per year (Khomenko et al., 2021). Additionally, aerosols influence the Earth's climate system by changing its radiation budget by scattering and absorbing sunlight (Quaas et al., 2008).

2. **Page 6 figure 2: Why are the reference lines not exactly at the maximum of O4 for all windows?**

   Response: In each chosen fitting window, the reference wavelength corresponds to the maximum of O4 absorption peak or close to this maximum. In some windows, the reference wavelength is close to the maximum. This choice is because a better DOAS fit in terms of RMS is achieved.

3. **Page 7: line 55: DDS is defined, what is the difference between DDS and DDS2?**

   Response: In the revised version of the present manuscript, we refer to this dataset as DDSv2 because the DDS data are made with version v2.1 and v2.2 (see Section S2).

4. **Page 7 table 1: Data products 01.03.00 and 01.03.01 have an overlap in time. What is used in this period?**

   Response: In reality, there is no overlap in time but a typo mistake in Table 1. We have corrected the end date of v01.03.00, which is 23/04/2019.

5. **Page 8 figure 3: In yellow box last line: "1 EA, many TAA"**

Response: The remark is correct and Figure 3 has been updated. Additionally, figure's caption has been modified as follows:
Figure 3. Mapping MAX-DOAS technique flow chart.

6. **Page 8 line 205: Is all data flagged for clouds or only the main viewing direction?**
Response: The information about the cloud coverage comes from the main viewing direction. In every full scan (i.e., vertical and azimuthal scan), we use the main viewing direction to estimate the MLH_{NO2}. When a vertical scan is characterized as cloudy, the full scan is excluded from the analysis (i.e., vertical and azimuthal scan).

7. **Page 9 line 211: Add an equation, as this value is used in all future calculations.**
Response: An equation for estimating the MLH of NO_2 has been added (Equation 1).

8. **Page 10 line 235: Replace ":" by ",".**
Response: We have replaced ":" by ',".

9. **Page 10 line 252: State clearer, that this sentence refers to the old paper.**
Response: The paragraph has been slightly modified as follows:

Assuming a homogeneous NO_2 distribution inside the MLH, the MLH is derived from the NO_2 vertical profiles in the main azimuthal direction and is defined as the ratio of the NO_2 VCD to the near-surface concentration of NO_2. In Dimitropoulou et al. (2020), the RTM simulations were performed for eight different MLH values of aerosols and NO_2 in the range of 500-2000 m (i.e. eight different combinations) and for different measurement viewing geometries (Solar Zenith Angle (SZA), Relative Azimuth Angle (RAA) and the corresponding elevation angle of 2°). For every MAX-DOAS measurement, one value of the correction factor is given according to its viewing geometry and MLH value during the measurement. For further information, we refer the reader to Dimitropoulou et al. (2020).

10. **Page 11 line 276: Equation (4) and the following sentence are confusing next to each other (I understood it after reading the next two pages). In the equation is written dSCD_NO2_simulated, in the text are mentioned simulated O4 DSCDs. Explain in more detail.**
Response: Section 4.2.1 has been revised. Additionally, this 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).

11. **Page 11 line 281: Was the impact of this fixed NO2 concentration investigated? If typical high values for Brussels are used, does the retrieval significantly change?**
Response: In the present study, the impact of this fixed NO_2 concentration is used to estimate the uncertainty related to the estimation of L_{NO2} from the RTM simulations. In the case of a box profile, the values of $1.5*10^{11}$ molec.cm^{-3} and $1*10^{11}$ molec.cm^{-3} are used. When other profile shapes are used, such as linearly decreasing vertical profiles, the near-surface NO_2 concentration was fixed to $1.5*10^{11}$ molec.cm^{-3}, with decreasing values in the

upper layers. This impact is estimated to be about 9.6% in the visible range (see Section 4.2.2).

12. **Page 12 line 298: "are" instead of "is" (or restructure sentence)**
Response: Corrected (see page 12).

13. **Page 13 line 307: Which order is the polynomial fit?**
Response: The polynomial fit is of second order. This information is already given in the caption of Figure 5.

14. **Page 13 line 310: Is this statement also true for the high pollution cases in Brussels?**
Response: In the Brussels-Capital Region, the pollution NO_2 levels will never be so high that the assumption of NO_2 as an optically thin absorber is not fulfilled.

15. **Page 13 line 314: (see Fig. 6 (a))**
Response: The sentence has been modified as follows:
For example, a MAX-DOAS measurement with SZA=30°, RAA=60°, MLH_{NO2}=1km, and measured O_4 dSCD=6.10^{43} $molec^2.cm^{-5}$ will have a L_{NO2} equal to 15 km at 477 nm (see Fig. 6a).

16. **Page 16 equation (6) and (7): For readability it would be good to include the missing step between the two equations**
Response: The missing step between equations (6) and (7) is now added.

17. **Page 17 equation (7): possible space character in subscript**
Response: Equation (7) has been checked and no space character was found in subscript.

18. **Page 22 equation (11), middle line: Confusing definition. If written like this, it should be x(first) >L_NO2 (the first value which extends over L_NO2).**
Response: In Equation 11 (now equation 14), as x(last), we are referring to the last point of the horizontal grid. To be more clear, we named x(last) as x(max).

19. **Page 23 line 536: If this is true, why do the lines differ in figure 13 and 14 for the first km from the a priori? And why is the information of this region used in the later comparisons if this information cannot be trusted?**
Response: In this region (extending from the MAX-DOAS instrument until the location of the L_{NO2} estimated from the 344nm measurements), the information that we obtain from the MAX-DOAS measurements is a constant NO2 near-surface concentration. The role of the a priori profile is to give this region the shape of the NO2 horizontal profile, as shown in Figure 13. In our under-constrained problem, the a priori information is derived by an air-quality model.

20. **Page 25 figure 11: Add wavelengths and change colors (see https://www.color-blindness.com/coblis-color-blindness-simulator/). Add also time of the measurement.**
Response: Figure 11 has been modified according to this comment and the measurement time has been added to figure's caption as follows:

Examples of weighting functions used in the new horizontal distribution inversion approach (11 September 2018, 11:26 UTC).

21. **Page 26 figure 12: Same y-axis for all**
Response: The y-axis is now the same for all the subplots in Figure 12.

22. **Page 27 figure 12: Mention RIO in caption**
Response: The legend of Figure 12 has been modified as follows:

"Figure 12. Example of seasonal RIO *a priori* NO_2 horizontal profiles for the new horizontal distribution inversion approach as a function of the horizontal distance from the MAX-DOAS instrument in six different azimuthal viewing directions, before the application of the scaling factor."

23. **Page 27 figure 13: Adjust both axes to the same values, add (a) and (b)**
Response: Figure 13 has been modified according to a comment of Reviewer #2. Three different example days are displayed for low, medium and high NO_2 concentration values. We have added (a), (b), and (c) for the three different examples.

24. **Page 28 figure 14: Add a priori of aerosol distribution. Are errors available?**
Response: Similarly as in Figure 13, three examples of near-surface aerosol extinction coefficient profiles have been added. Additionally, the a priori near-surface aerosol extinction coefficient profiles and the retrieval error have been added in Figure 13.

25. **Page 32 figure 16: For consistency it would be good to see the values corresponding to figure 13**
Response: Figure 16 has been modified and the errors refer to the example used in Figure 13b.

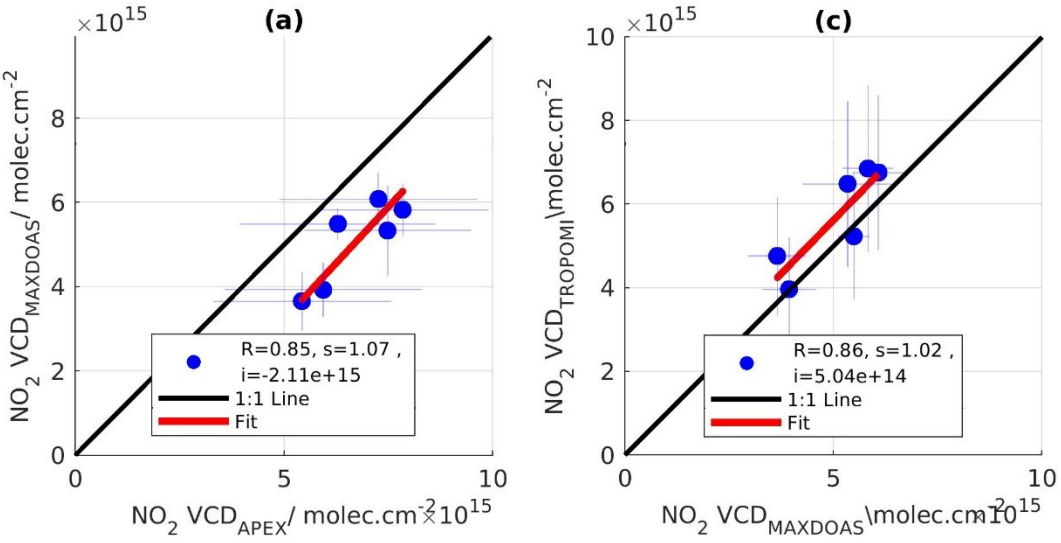
26. **Page 33 figure 17: It is claimed before, that the measurement is insensitive for the first kilometers. It would be good to mark this range (e.g. draw a line). Why is the y-axis limited to 11km?**
Response: It is true that maximum and minimum NO_2 values cannot come from the MAX-DOAS measurements in this region. This horizontal region of insensitivity depends on the L_{NO2} of the first MAX-DOAS measurement at the smaller wavelength, and consequently, is different for each retrieved horizontal profile. We have decided to not add this information in Figure 17 as it will become difficult to interpret. Concerning the y-axis, we have extended the range of this axis until the maximum L_{NO2} estimated during that day.

27. **Page 33 line 664: Is the wind direction and speed stable over the measurement period? What is the wind speed?**
Response: As you can see in Figure 18, the wind direction is almost stable over the measurement period and its main directions are NE, E, and SE during that day. The wind

speed is higher during the morning hours (3.7 m/s for 5 UTC) and lower around 10 UTC (i.e., 2.2 m/s).

28. **Page 34 figure 18: The wind direction could be added to the plots. If possible, the latitude and longitude range could be shortened to make the plot bigger.**
Response: The wind direction as measured by the meteorological station in the rooftop of BIRA-IASB has been added to each subplot. Additionally, the subplots are more zoomed.

29. **Page 35 line 695: Mention typical overpass time of TROPOMI. Is every measurement from TROPOMI used? Even when several are available for one day? And is the exact time of the overpass used or the typical overpass time? The NO2 distribution can change quickly. Were also other time ranges used for comparison? (+/-30Min)?**
Response: We have added the following sentence to mention the overpass time of TROPOMI during this day:
During this day, the TROPOMI overpass time was at 12:19 UTC.
For other days, TROPOMI can have two overpasses above Brussels-Capital Region. When this is the case, the exact time of each overpass is used and accordingly, the MAX-DOAS horizontal profiles that are retrieved +/- 1 hour of each overpass are used.
During the 28th of June 2019, we performed the same comparison as in Section 5.2, by selecting a smaller time range (+/-30min) for the MAX-DOAS horizontal profiles and by applying a weighting (following comment). The results are the following:



As the comparison points diminished, we have decided to perform the comparisons between MAX-DOAS and TROPOMI observations around 1 hour of each TROPOMI overpass time.

30. **Page 35 line 698: How does this simplification (no weighting) influence the results?**
Response: In this revised version of the manuscript, we have weighted the MAX-DOAS segments by their relative length inside each TROPOMI pixel. As you can see in Figure 20 of Section 5.2, the resulting correlation coefficient, slope and intercept change slightly.

31. **Page 35 line 706: Can this be explained/expected? (Different sensitivity…)**
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 that day, the APEX (airborne) instrument detected an additional NO_2 layer at an altitude higher than the estimated MLH_{NO2} (which is around 700m during the APEX flights).
Another possible explanation could be the different measurement techniques of both instruments (i.e., ground-based and airborne) and consequently, different instrument sensitivities as well as the different reference spectrum used in the DOAS analysis of measured spectra.

32. **Page 36 line 723: Are the retrieved values high/low, compared to typical values for this region?**
Response: For the Brussels-Capital Region, the retrieved values during that day are low/medium. As we can see in Figure 23, the retrieved NO_2 tropospheric columns can be up to $2.5*10^{16}$ molec/cm^2.

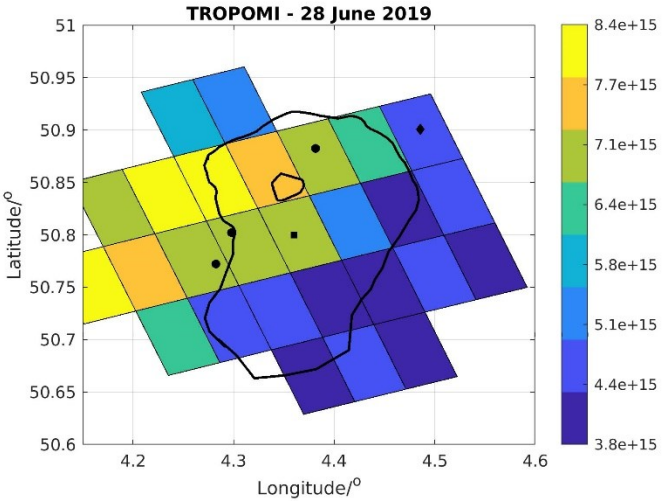
33. **Page 37 figure 19: Add MAX-DOAS maps in original resolution for comparison with original APEX. Also, the RIO analysis could be added. Use the same color scale for all plots!**
Response: In Figure 19, the MAX-DOAS map in original resolution has been added. Following a comment from Reviewer #2, the color-scale is different for the original APEX and AEROMOBIL measurements and the same for all the other subplots. A comparison with RIO dataset is presented in Section S1.

34. **Page 38 line 759: Point number 3 is unclear to me. A sketch could help to clarify.**
Response: Point number 3 is indeed hard to be interpreted by the reader. In addition to that, an application of such a weight in the mean MAX-DOAS NO_2 columns does not change the comparison outcome. For these reasons, point number 3 has been modified as follows:
The MAX-DOAS segments are weighted by their relative length inside each TROPOMI pixel.

Consequently, the MAX-DOAS and TROPOMI comparisons presented in Section 5.3 have been modified.

35. **Page 39 line 768: Regridding routine is not completely clear to me. Are the averaged pixels weighted by their ratio by which they cover the reference pixel?**
Response: To create the seasonal maps for MAX-DOAS and TROPOMI datasets, we have chosen the TROPOMI pixels (i.e., reference pixels) recorded during the APEX flights (28 June 2019; see figure below). For every measurement day, the TROPOMI pixels and MAX-DOAS grids that fell inside each reference pixel have been averaged without applying a weight equal by the ratio by which they cover each reference pixel.

The reference pixels are the following:



36. **Page 39 line 776: Agreement for summer seems to be worse than for other seasons. The maximum in TROPOMI is seen in the NW, in MAXDOAS it is south.**

Response: Indeed, the agreement for summer is worse than for other seasons. We have modified the text as follows:

It is found that the locations of the NO_2 peaks and dips show a reasonably high degree of similarity between TROPOMI and MAX-DOAS during all seasons, except summer.

37. **Page 39 line 782: Can this be explained?**

Response: As we can see in Figure 22c, large positive biases does not occur for our study. Although, the most common case (in each pixel) is to have a negative bias, which means that TROPOMI underestimates the tropospheric MAX-DOAS NO_2 columns. This finding is in agreement with several studies (Verhoelst et al., 2021; Tack et al., 2021; Judd et al., 2020; Dimitropoulou et al., 2020; Ialongo et al., 2019).

38. **Page 39 line 792: Explain SEAS, I guess season?**

Response: The sentence has been slightly modified as follows:

When seasonally-averaged TROPOMI and MAX-DOAS pixels (the pixels shown in Fig. 21) are compared one-by-one (see seasonal (SEAS) in Fig. 23), both correlation coefficient (R in the range of 0.57-0.93) and slope values (s in the range of 0.65-0.94) improve considerably.

39. **Page 40 line 806: "… presented in Dimitropoulou et al. (2020), see table 4."**

Response: The sentence has been modified.

40. **Page 41 figure 21: If possible, remove one color bar and use the free space to widen the plots in x-axis.**

Response: Concerning Figure 21, one color bar per season has been removed and we have zoomed more in the subplots.

41. **Page 44 figure 23: Red line is hardly visible**

Response: In Figure 23 and Figure S11, we have extended the red line to cover the whole range of tropospheric NO_2 columns.

42. **Page 45 table 4: Clarify R (seasonal) and s (seasonal). It probably corresponds to SEAS? Are the number of points the number of days or the number of pixels?**

Response: In Table 4, the legend has been modified as follows:

Table 4. Summary of the regression analysis parameters (e.g., correlation coefficient (R) and slope (s)) and the number of data points (N) derived in the present study during only one year of observations (i.e., number of pixels) and in Dimitropoulou et al. (2020). Please note that R (seasonal) and s (seasonal) corresponds to SEAS in Figure 23.

43. **Page 45 line 885: Add name of the improved FRESCO-S cloud retrieval.**

Response: As mentioned above, the TROPOMI tropospheric NO_2 column product with an improved FRESCO-S cloud retrieval is called DDSv2. We have added:

First, a TROPOMI tropospheric NO_2 column product (DDSv2 product) with an improved FRESCO-S cloud retrieval was tested.

44. **Page 45 line 887: "Fig. 24 (a) and (b) …"**

Response: (a) and (b) have been added to the text.

45. **Page 46 line 910: Using MAX-DOAS profiles as a priori profiles for TROPOMI is suitable for a consistency check of the method. It should not be mistaken to be the "truth".**

Response: The sentence has been modified as follows:

The present study suggests that in urban conditions, the NO_2 profile shapes from the CAMS regional CTM ensemble are not the most suitable a priori information that can be applied in the TROPOMI retrieval.

46. **Supplement figure S1: Same y-axis for all plots, MLH could be in the title of the individual sub-plots.**

Response: Figure S1 and S2 have been corrected according to this comment.

47. **Supplement figure S3: Same color bar for all plots**

Response: Figure S3 has been modified accordingly.

48. **Supplement figure S4: Same y-axis, at least for AOD>0, this highlights the differences much stronger**

Response: In Figure S4, same y-axis has been used in all the subplots, except for AOD equal to zero.