Dear Thomas von Clarmann,

We thank you for your valuable contribution to ameliorating the present paper.

Following the recommendation of the third reviewer concerning the weighting functions, we have decided to use the proposed formula to estimate the weighting functions, as it is more physically more adequate. Please note that all the analysis and, consequently, Figures 11 to 23 and S5 to S11 have been modified.

In the present document, we list a point-to-point response to the reviews.

Please find below our responses to each comment individually.
Please consider that:
A) Green bold: Comments of the Referee
B) Black: Response to each comment addressed by the referee.
C) Black bold: Already existing text in the manuscript.
D) Red bold: Added/corrected text in the manuscript according to referee's comments.

Response to Anonymous Referee #1

1. **Thanks for adding more examples to Fig 13. and 14. Please discuss the new examples in 2-3 sentences. Especially the larger deviation of a priori and retrieval result in Fig. 14 is interesting. Would you consider the a priori information as good enough?**

   Response: Please consider the following additional discussion concerning Fig. 13 and 14:

   Three examples of the retrieved NO_2 horizontal profile are presented in Fig. 13, together with corresponding measured and simulated $\bar{c}_{NO2}$ at the six different wavelengths for July 2, 2018 (fig. 13a; low NO2 abundance condition), September 11, 2018 (fig. 13b; medium NO2 abundance condition), and September 30, 2018 (fig. 13c; medium NO2 abundance condition). RMS is calculated between measured and simulated NO_2 near-surface concentrations of the horizontal retrieval normalized by the mean of the measured NO_2 near-surface concentrations (upper panels in Fig. 13).. As the NO_2 values become larger, the agreement between measured and simulated $\bar{c}_{NO2}$, expressed via the RMS value, is improved.

   Similarly, examples of measured and retrieved near-surface aerosol extinction coefficient and retrieved aerosol horizontal profile are shown in Fig. 14, for different aerosol load conditions (low (Fig. 14a), medium (Fig. 14b) and high (Fig. 14c)) over the Brussels-Capital Region. We observe that the agreement between simulated and measured near-surface aerosol extinction coefficient at the six different wavelength tends to be worse than for NO_2. This could be due to the use of a constructed (constant)

a priori aerosol horizontal profile due to the lack of information on the aerosol extinction horizontal distribution in the Brussels-Capital region.

2. **Please add a small discussion of the smoothing error in Fig 16.**

   Response: A small discussion of the smoothing error in Fig. 16 has been added as follows:

   The horizontal profiles of the measurement and smoothing error in percentage are shown in Fig. 16. As can be seen, the smoothing error is significantly larger than the measurement error (range of 3%-10% and 14%-40%, respectively). The smoothing error becomes also larger as the horizontal distances from the instrument become larger. This is mainly because of the exponential decrease of the sensitivity as a function of the horizontal distance (see weighting functions in Fig. 11), and consequently, the larger impact of the difference between the a priori profile and the true state of the atmosphere.

3. **Thanks for adding the validation Section S1 in the supplement. Please refer to this section in the main manuscript.**

   Response: We are referring the reader to Section S1 in Section 5.3 in the following point:

   A mean scaling factor equal to the mean ratio between the measured and RIO NO_2 near-surface concentrations is applied because of the systematic underestimation of NO_2 near-surface concentrations by MAX-DOAS when compared to in-situ measurements (see Dimitropoulou et al., 2020 and Section S1).

Response to Anonymous Referee #3

General comments

1. **A main problem of the manuscript is that the description of the methods is lacking conciseness, and it is not always clear whether certain approaches are based on physical principles or rather represent simplifying assumptions. In particular, the description of the horizontal distribution inversion approach (Section 4.3.) conveys the impression that a constant weighting function as a function of distance with a sharp drop at the estimated light path length is a fact, while this is in actually a very simplifying and physically incorrect assumption. In reality, I would rather expect that the sensitivity decreases exponentially with distance from the observer in accordance with the Beer-Lambert law, as already discussed by Kern et al. [2010] and Vogel et al. [2011]. In the light of these (over-) simplifications, it is surprising to see that the retrieved horizontal distribution compares very well with airborne and satellite measurements.**

   Response: We would like to thank the reviewer for rising this issue concerning the weighting function of the inversion approach. Please consider our response to specific comment #1.

2. **I wonder if it is not possible to infer the correction factor discussed in Section 4.2 directly from the aerosol and NO2 profiles retrieved by MMF, which directly provide dSCD(NO2) and c(NO2) and thus L(NO2) as the ratio of both.**

   Response: The experimental set-up in Uccle contains 10 different azimuthal directions. In order to ensure that L(NO2) is appropriate for each of them under different viewing geometries (i.e., SZA, RAA, and elevation angle), we preferred to perform simulations separately from the OEM-based MMF algorithm, which can only retrieve information in the main azimuthal direction in which elevation scans are performed.

3. **Some of the measurements used for intercomparison are not explained, or only later in the manuscript. For example, it is not clear what CIMEL measurements are, and airborne measurements are mentioned already in Section 4.3, but the context is only given much later in the manuscript in Section 5.2. I suggest to add a short Section (4) with a short description of the ancillary data used for intercomparison (CIMEL, APEX, car DOAS, etc.).**

   Response: In the revised version of the manuscript, we have added a new Section (**4.Ancillary measurements**), in which short descriptions of CIMEL, APEX, RIO, in-situ and car-mobile DOAS measurements are presented.

4. **The level of agreement between the horizontal distributions of NO2 from MAX-DOAS on the one hand and from satellite, car- and airborne measurements on the other hand presented in Section 5.2. is quite impressive. However, I recommend a major revision of the manuscript due to significant deficiencies in the methodology and in the description of the inversion approach. The inversions of the horizontal distribution need to be re-done using physically correct weighting functions.**

   Response: Please consider our response about the issue that you have raised concerning the weighting functions in the Specific Comment #1.

Specific Comments

1. **Section 4.3: I find the description of the forward model quite confusing, and I feel that this Section requires substantial revision. Moreover, I think the inversion needs to be re-done with appropriate weighting functions. Equation 12 suggests that the forward model for the calculation of a mean concentration is given as the integral over the concentration divided by the light path length. First of all, I guess that the model is not based on numerical integration, but that it is rather based on a discrete sum over the horizontal grid. Second, what is missing in this equation is an appropriate weight as provided by a weighting function that represent correct physics. The assumption of a constant sensitivity between the instrument and the effective light path length L is not**

realistic. Instead, an exponential decrease should be chosen as weighting function in accordance with the Beer-Lambert law. Also, the light path through the boxes is not horizontal but slanted, so the weighting function needs to contain the cosine of the elevation angle (this is however only a small effect at 2° elevation angle). I furthermore think it would be more appropriate to use the observed dSCDs directly as measurement vector y instead of (weighted) mean concentrations, which are not a very useful quantity. The weighting function as a function of wavelength λ and distance x to the box would then be

$K(\lambda, x) = (\exp(-x/L(\lambda))) * \Delta x/\cos(\alpha)$

with Δx being the width of the boxes, α the elevation angle, and $L(\lambda)$ the effective light path estimated from the measurements. The forward model would then simply be

$F(\lambda) = \Sigma(\lambda, x_i) * c_i$

with x_i being the distance between observer and box i and c_i the NO2 concentration in this box. This equation can be readily inverted using OEM.

Response: After the issue raised with the weighting functions, we have decided to re-do the inversion with appropriate weighting functions.

First, we confirm that the model is based on a discrete sum over the horizontal grid. Second, we agree that the assumption of a constant sensitivity (i.e., constant weighting function) between the instrument and L and no sensitivity beyond L is likely not the most realistic one . For this reason, we have modified the weighting function as suggested:

$K(\lambda, x) = [\exp(-x/L(\lambda))] * \Delta x/\cos(\alpha)$

By using these weighting function, L(NO2) corresponds to an effective distance, which can be interpreted as the distance at which the sensitivity drops to 1/e. Beyond L(NO2), the sensitivity continues to reduce. In the revised version of this paper, we have decided to consider also distances beyond L(NO2) in the inversion with the maximum distance being defined as the distance where the sensitivity is 10% or less of the sensitivity close to the measurement site.

Consequently, we have performed again the analysis with the new weighting functions and Figures (11 to 23 and S5-S11) and Tables (3 to 4) have been modified accordingly. The main changes and improvements/degradations in the results are the following:

- An increase in the retrieved NO_2 near-surface horizontal profile
- An increase in the RMS between the measured and retrieved NO_2 near-surface concentrations

- A decrease in the DOFs values of the retrieved NO_2 near-surface horizontal profile
- A change in the horizontal shape of the averaging kernels, measurement error and smoothing error
- A better agreement between tropospheric NO2 columns derived by the MAX-DOAS observations and the airborne, car mobile-DOAS and satellite observations on 28th of June 2019
- An improvement of the correlation coefficient value and a degradation of the slope values in the seasonal comparisons between TROPOMI and MAX-DOAS tropospheric NO2 columns

2. **P29, L527ff: It cannot be 'seen' from Figure 11 that the weighting functions are constant up to a distance L, but instead this is an assumption, which is physically incorrect – see my comments above.**

   Response: Please consider our response to your comment above. The sentence has been modified as follows:

   The sensitivity decreases exponentially up to a distance corresponding to the differential effective light path length of each measurement. More precisely, each measurement is highly sensitive to the MAX-DOAS instrument location. This sensitivity decreases exponentially as a function of the horizontal distance. Then, it reaches a value equal to 1/e to the horizontal distance equal to the differential effective light path length of each measurement.

3. **P29, L552: Here you mention CIMEL observations without explaining what the nature of these measurements are and where they have been perfromed. A short description of these measurements should be part of an extra Section on ancillary measurements further up in the manuscript – see general comments.**

   Response: After considering General comment #3, a Section (now, Section 4 – Ancillary measurements) has been added.

4. **P29, L550ff: Do I understand it right that the linear decrease in a priori AOD is a decrease in the horizontal dimension? If so, what are the motivations for this assumption? A higher AOD at your measurement site than anywhere else in the surroundings is hard to justify (except if there were strong aerosol sources next to your instrument).**

   Response: Indeed, as an a priori AOD horizontal profile, we have chosen a horizontally decreasing profile. Given the fact that there are no strong aerosol sources close to the instrument, in the revised version of the manuscript, we now use a horizontally constant a priori AOD profile.

5. **P30, L567: What kind of airborne observations are these? This is only explained later in the manuscript– I suggest to move the introduction of APEX from Section 5.2. to a Section further up in the manuscript describing all ancillary data used in this study (see general comments).**

   Response: Please consider our response to General comment #3.

6. **P30, L580ff, and Section 4.4: It is stated several times that there is no information on the horizontal distribution at distances closer than the shortest scattering distance. The averaging kernels are, however, not zero at these regions (see revised Fig. 15). Instead, it seems that parts of the information coming from short distances are falsely attributed to distances further away. For example, the 8.75 km averaging kernel has a constant value of 0.04 up to a distance of approx. 8 km. Unfortunately, no averaging kernels for distances closer to the instrument are shown.**

   Response: As expected, the use of the new weighting functions, (see Fig. 11) have an impact to the retrieved NO2 horizontal profile and the form of the averaging kernels. In Figure 15 of the revised manuscript, we observe that the AKs for distances closer to the instrument (d=2.25 km and d=4.75 km) have the maximum values and decrease exponentially as a function of the horizontal distance from the instrument. Consequently, the statement that there is no information on the horizontal distribution at distances closer than the shortest scattering distance is not correct and has been removed from the revised manuscript.

7. **Figure 15: Given the small peak values of the averaging kernels (at most 0.05), I wonder if the fine horizontal grid of 500 m is really useful or if a coarser grid would have been more appropriate. Furthermore, I could imagine that the averaging kernels would look more smoothly if more realistic (exponentially decreasing) weighting functions rather than the arbitrary step-like functions would have been used.**

   Response: The information content of the measurements (DOFs) determine the choice of the retrieval grid. For our retrieval, the DOFs are generally larger than unity and smaller than two. This means that we can only retrieve the NO2 near-surface concentration in 1-2 horizontal boxes. In practice, in the atmospheric remote sensing, we use much thinner retrieval grid. During the development of the horizontal OEM-based inversion approach, we have tested the use of wider horizontal boxes and we found that this choice does not have a considerable effect on the NO2 horizontal retrieval.

   The use of more realistic weighting functions have a significant impact on the form of the averaging kernels, which are smoother and show maximum values for distances closer to the instrument.

8. **Section 5.3.1: Here you describe the methodology for comparison between the different datasets, but the NO2 gas maps based this method have already been shown in Section 5.2, if I understand it right. Is there something different in the data processing (filtering and spatio-temporal binning) for the production of Figure 21 compared to Figure 19 (except that Fig. 21 shows seasonal averages)? If not, then the description of this method should appear at the beginning of Section 5.2.**

   Response: In Section 5.3.1 and 5.2 (now, Section 6.3.1 and 6.2), the NO2 maps for the Brussels-Capital Region have been produced by using the same data processing. Please consider the following modification in the revised paper (Section 6.3.1):

   To compare the TROPOMI and MAX-DOAS tropospheric NO_2 columns, the similar approach is used as in Section 6.2. Additionally, TROPOMI and MAX-DOAS tropospheric NO_2 columns are compared in a seasonal basis, and the seasonally-averaged maps of those VCDs on the area covered by the TROPOMI pixels are created. To generate these maps, the ensemble of TROPOMI pixels recorded on 28 June 2019 is chosen as reference and TROPOMI pixels that coincide with this reference grid are averaged. The daily horizontal profiles of MAX-DOAS NO_2 columns are averaged on the daily TROPOMI grids and then, the reference grid is used to create the seasonally-averaged MAX-DOAS maps.

Technical Corrections

1. **P6, L133: The O4 cross sections by Finkenzeller have recently been published [Finkenzeller and Volkamer., 2022]. Please add the according reference.**

   Response: The reference has been added to the manuscript.

2. **P8, L185: 'in six different fitting windows' - > 'in the six different fitting windows listed in Section 2.2.'**

   Response: The sentence has been modified accordingly.

3. **P9, L187: Explain abbreviations/acronyms 'OEM' and 'MMF'**

   Response: The abbreviations/acronyms are explained in the revised version of this paper.

4. **P9, L189: Explain abbreviation 'MLH'**

   Response: The abbreviations is explained.

5. **P9, L195: Here you should state that the NO2 near-surface concentrations and VCDs and the near-surface aerosol extinction are retrieved as a function of distance from the instrument.**

   Response: The sentence is modified as follows:

Then, in the next step, a new dual-scan parameterization technique is applied to the O_4 and NO_2 dSCDs at the six different wavelengths and in all the azimuthal directions with MLH_{NO2}, measured O_4 dSCDs, and measurement geometry being the main input parameters to retrieve the horizontal sensitivity of NO_2 and, consequently, the NO_2 near-surface concentrations and VCDs, and near-surface aerosol extinction as a function of distance from the instrument (see Section 5.2).

6. **P9, L197: Three times 'horizontal' in one sentence. Please rephrase.**

Response: The phrase has been rephrased as follows:

In the final step, a new OEM-based horizontal distribution inversion approach is developed using the six near-surface NO_2 concentrations and aerosol extinction values per azimuthal direction to retrieve NO_2 and aerosol extinction horizontal profiles in an output grid of 500m thickness (see Section 5.3).

7. **Section 4.2: Equation (3) is just a trivial rearrangement of Equation (2) and therefore obsolete. Please remove one of these.**

Response: Equation (3) has been removed and the text has been adjusted as follows:

L_{O4} is calculated by using Eq. 2 for O_4.

8. **P16, L348: 'Regarding the aerosols' - > 'Regarding aerosols'**

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

9. **P23, L434: I suggest replacing the term 'sanity' with 'consistency'**

Response: The term sanity is replaced by the term consistency.

10. **Equation 14: I suggest to replace dx with Δx since dx can be confused with the differential in Equation 11.**

Response: dx is replace with Δx in Equation 14 (now, Equation 13).

11. **P30, L579: Insert a comma before the year number.**

Response: A comma has been inserted before the year number.

12. **P40, L670: Replace 'error' with 'covariance matrix', and mention that the error is given as the square root of its diagonal elements.**

Response: The word 'error' has been replaced with 'covariance matrix'. Additionally, the following sentence has been added:

Then, the retrieval noise error is given as the square root of the diagonal elements of the noise covariance matrix.

13. **Caption of Figure 16: 'measurement error' → 'measurement and smoothing error'**
    Response: The caption of Figure 16 has been modified accordingly.

14. **Figure 18: The bottom-right panel should have the same width as the other panels.**
    Response: Figure 18 has been modified accordingly.

15. **Title of Section 5.3: 'Comparison between MAX-DOAS horizontal NO2 distribution and TROPOMI observations'**
    Response: The title of Section 5.3 (now, Section 6.3) has been modified.

16. **P70, L1105: The approach is better than what?**
    Response: The word 'better' has been replaced by the word 'good'.

17. **P70, L1108: Do you mean a slope closer to unity?**
    Response: Indeed, we do mean a slope closer to unity. The text has modified accordingly.

18. **P70, L1114: Delete this paragraph on the role of the a priori as it refers to a Section that has been removed in the revised manuscript.**
    Response: This paragraph has been deleted from the manuscript.

A list of all relevant changes made in the manuscript is presented below. Please consider also the uploaded manuscript using track changes in Word.

List of changes in the manuscript

(Page 1, Line 22), (Page 3, Line 70-76), (Page 5, Line 131), (Page 8, Line 181-204), (Page 8-9, Line 205-212), (Page 9, Line 217-219), (Page 10, Line 227-228), (Page 10, Line 251), (Page 10, Line 253), (Page 11, Line 274-279), (Page 12, Line 297), (Page 12, Line 306-316), (Page 13, Line 323-326), (Page 13, Line 331), (Page 13, Line 337), (Page 14, Line 365), (Page 17-18, Line 413-415), (Page 18, Line 441-444), (Page 21, Line 473-480), (Page 21, Line 487), (Page 21-22, Line 494-503), (Page 22-23, Line 511-532), (Page 23, Line 537-554), (Page 23-24, Line 559-491), (Page 25, Line 600), (Figure 11), (Figure 13), (Figure 14), (Page 32-35, Line 655-710), (Figure 15), (Figure 16), (Page 38, Line 727-733), (Figure 17), (Figure 18), (Page 43, Line 771-776), (Page 43-44, Line 791-803), (Page 44, Line 810-814), (Figure 19), (Figure 20), (Page 48-49, Line 849-911), (Figure 21), (Figure 22), (Figure 23 and legend), (Table 4), (Page 57, Line 993), (Page 58, Line 1024-2043), (Page 60, Line 1095), and (Page 60, Line 1115-1119).

Best regards,

Ermioni Dimitropoulou