Atmospheric Measurement Techniques Discussions

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Comment on amt-2021-308

Anonymous Referee #2

Referee comment on "Horizontal distribution of tropospheric NO_2 and aerosols derived by dual-scan multi-wavelength multi-axis differential optical absorption spectroscopy (MAX-DOAS) measurements in Uccle, Belgium" by Ermioni Dimitropoulou et al., Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2021-308-RC2, 2022

General comments

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.

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.

Details on the three main issues:

- 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.
- 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.
- 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.

Specific comments

P3, L88: Please write "telescope azimuth angle" so that the abbreviation (TAA) makes more sense.
P3, L89: Why did you use 2° instead of 1°? (see also **P9, L219**)
P3, L92: Remove "directions". It is not needed.
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!
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.
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).
P5, L116: Why are not all reference wavelengths in the peak center of their corresponding O_4 absorption bands?

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?
P7, L172: "measured radiance spectra ... is analyzed" to "are analyzed"
P9, L195: Please cite some of the "several studies" you are referring to.
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.
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!
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**)
P10, L251: Please move the full stop from the index of NO_2 to the normal level.
P11, L276: "Here, O4...". If "here" refers to the equation above it should be "Here, NO2..."
Section 4.2.1 Please revise according to the general comment.
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.
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.
P13, L335: "less pronounced Rayleigh scattering" to "less pronounced Rayleigh and Mie scattering"
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.
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?
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.
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.
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.
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.

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!
P29, L601: "close their" to "close to their"
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.
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!
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?
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.
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.
P35, L706: How do you explain this intercept?
P35, L709: "channels" to "elevation angles" or "geometries"
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.
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.
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.
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**)!
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.
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.

P40 L818-828: This belongs to the conclusion and should be removed from this section.
Section 5.3.2: Remove this section from the manuscript or move it to the supplement (see general comments).
P49, L953-954: and AODs
Conclusions: Please change the conclusions 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.