

We would like to thank the reviewers for their valuable comments and suggestions. It took more time than expected to go through all of the individual points, in particular as some of them required a significant extension of our original investigations. E.g., we included additional evaluations of ceilometer data to provide consistency checks for the MAX-DOAS retrievals, as well as additional examples of synthetic data retrieval. As a consequence we want to apologize for the delay of our point to point responses.

Reply to RC2

We thank reviewer #2 for the detailed comments. These comments are useful for use to improve the quality of the manuscript. We have supplemented a more examples of synthetic data retrieval to support our statements. In addition, we have further evaluated the retrieved MAX-DOAS aerosol profile by comparing to ceilometer measurements. We have addressed the reviewers' comments on a point to point basis as below for consideration. Our answers are presented in blue texts. Please note that all the page and line numbers mentioned below refer to the pages and lines in the manuscript with revision marks.

Anonymous Referee #2

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General comments

Wang et al. introduce a new MAX-DOAS aerosol profiling algorithm for high altitude sites. The algorithm itself is based on a parameterized approach using a pre-calculated look-up table and is optimized to retrieve profiles from data measured at high altitudes. The authors include an extensive sensitivity study and discuss the most important errors thoroughly. Furthermore, an attempt to validate the performance of the algorithm with ancillary measurements is shown. The AMTD version of this manuscript was also added with one retrieval example of synthetic data and the comparison with an OEM algorithm.

First of all, I would like to comment that the manuscript has improved considerably since the first submission. Unfortunately, my main concern from the first manuscript assessment is still valid. The validation part and the retrieval of synthetic data is not enough to show that this algorithm is not only capable of retrieving accurate profiles but performs better than state-of-the-art OEM algorithms at high altitudes (as the authors claim). In order to solve this issue, I suggest to extend the corresponding sections with the following tests:

1. The retrieval test of synthetic data (Section 4.3) should be complemented with further examples (different exponential profiles and elevated profiles).

Response: We have supplemented two more examples with different profile shapes, one exponential profile, and the other profile with a weak elevated layer. The result shows that our retrieval method can reproduce the true profiles within the measurement error. In addition, the synthetic data were also retrieved by an OEM based algorithm. The results are shown for reference. This result is supplemented in Sect. 4.3 (see Pages 32-34).

2. Comparisons of retrieved profiles with Ceilometer profiles should be added as well. This could be done in an additional section or with similarly averaged Ceilometer profiles in Fig. 10.

Response: We have supplemented the ceilometer results in Sect. 2.3. The ceilometer reports attenuated backscatter, while the MAX-DOAS measures aerosol extinction coefficients. As these parameters are not directly comparable, we have converted ceilometer measurement of attenuated backscatter profiles to aerosol extinction profiles using auxiliary AOD information from the co-located sun photometer. The ceilometer data and the comparison result are shown in Sect. 2.3 (see Pages 6-7).

3. Fig. 3, 6 and 7 are shown for one example only. It would be interesting to see how the depicted parameters look like for a not so ideal profile retrieval (e.g. smaller (larger) RAA (SZA) or different profile shapes).

Response: We have supplemented a few more examples with a smaller SZA and a larger RAA. The new results are shown in Fig. 5 (Page 20), Fig. 9 (Page 29) and Fig. 11 (Page 30).

Specific comments

P1, L4-5 and P3, L20-23 and P28, L22-23: The authors claim that commonly used MAX-DOAS algorithms are not suitable for profile retrievals at high altitudes. Since this is neither shown properly in this manuscript nor do the authors cite a publication which addresses this issue, I think that these sentences should be reworded or removed (see also comment to **Section 4.3**).

Response: We have revised the sentences (see Page 1, Lines 5-7, Page 3, Lines 31-34 and Page

38, Lines 14-16). In addition, we have added more examples of synthetic data retrieval. The retrieval of the synthetic data presented in Sect. 4.3 suggested that OEM-based retrievals cannot fully reproduce the true profile. The new results are shown in Sect. 4.3 (see Pages 32-34).

P3, L29: area → areas?

Response: This typo has been corrected (see Page 4, Line 7).

P4, L15-16: "The exposure time and number of scans of each measurement are adjusted automatically (...)." Could you please explain what the automatic adjustment of the number of scans of each measurement means?

Response: We have supplemented the information in Sect. 2.1 (see Page 4, Line 32 to Page 5, Line 4).

P5, L5-6: "(...) the derivation of Angström exponents is critical and thus omitted." If this is critical, why is it omitted? In Section 4.5, you derive Angström exponents from MAX-DOAS results. It appears inconsistent to me that Angström exponents are only discussed from MAX-DOAS alone without validating with sun photometer results. This is even more problematic as you found that the MAX-DOAS AODs are much smaller than the sun photometer results (when the AOD lower 0.02 is the main reason for the omission). You could also compare with AERONET data in case the derivation from the available sun photometer is problematic.

Response: We are sorry for the confusion. As the uncertainty of the AOD measured by the sun photometer is relatively large, the uncertainty of the derived Angström exponent would be further amplified. Consequently, the Angström exponent is not very reliable, and this is the reason, that they are not discussed in the results. We have supplemented the explanation to Sect. 2.2 (see Page 5, Lines 26-28).

P5, L13: preformed → performed

Response: This typo has been corrected (see Page 8, Line 1).

P7, L19-20: Which phase function and SSA values were used for the simulations? Which climatology was used?

Response: We have supplemented the definitions (see Page 10, Line 20 to Page 11, Line 1).

Section 3.5: Which climatology was used for the LUT creation? Are different pressure and temperature conditions/profiles are taken into account (in addition to the cross section temperature discussion)?

Response: We used the US Standard climatology (Anderson et al., 1986). We have supplemented this information in the text (see Page 17, Table 5). The variation of atmospheric profile (i.e. temperature and pressure) is not considered in the look-up table, while we consider this effect as 'other possible error sources' in the error estimation of the retrieval. We have done a sensitivity analysis with summer and winter atmospheric profiles to estimate the uncertainty related to temperature and pressure variation. We estimated the corresponding uncertainty is less than 2% which is well covered by the 'other possible error sources'. We have supplemented the discussion about this issue in Sect. 3.7.4 (see Page 21, Line 20 to Page 22, Line 3).

P10, L6: A fixed median phase function was used for the LUT but Fig. 3 and Section B3 tell me that it is quite important to use a proper phase function (especially for small RAA). Do you plan to add more dimensions to the LUT for different phase functions or how do you deal with this problem?

Response: Since accurate estimation of phase function is in general not available, it is not feasible to add phase function as an additional dimension. We have further clarified in Sect. 3.6 that only well-known input parameters are defined as dimensions of the look-up table (see Page 15, Line 30).

P11, L22: ceiometer → ceilometer

Response: This typo has been corrected (see Page 15, Line 11).

P13, L11-12: "(...) we found that O₄ DSCD at 5° is almost negatively correlated with AOD." This is new for me. Could you please show this in a plot (maybe in the appendix)? Something only valid for high altitude sites?

Response: Theoretically, aerosol reduces the optical path length for off-zenith measurements in the atmosphere due to enhanced Mie scattering, and the optical path is expected to be the longest under pure Rayleigh atmosphere. Therefore, O₄ DSCD is expected to reduce with enhanced aerosol load. Fig. 1 show the correlation between O₄ DSCD at 5° and AOD (0-2 km) for all the

profiles in the look-up table (SZA = SAA = 60°). In each chart, the trend line is derived by moving average, and the r value is the Pearson correlation coefficient between the original data and the expected values obtained from the trend line. We have revised the description in Sect. 3.7.2 (see Page 18, Line 31 to Page 19, Line 4).

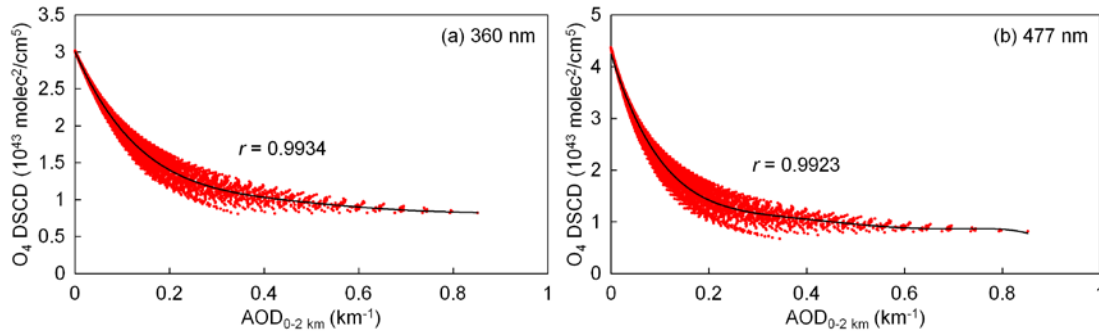


Fig. 1: Correlation between O4 DSCD at 5° and AOD (0-2 km) for all the profiles in the look-up table (SZA = SAA = 60°).

Fig. 3 Please add SZA and RAA values to the description of your example cycle as it is used throughout the manuscript.

Response: We have added SZA and RAA values to the captions of the figures (see Page 20, Fig. 5, Page 21, Fig. 6, Page 25, Fig. 7, Page 26, Fig. 8, Page 29, Figs. 9 and 10, Page 30, Fig. 11 and Page 31, Fig. 12).

P16, L14-15: "This is because the a priori profile is not needed in our retrieval algorithm". To be more accurate, you include a priori assumptions of aerosols above retrieval height in your total uncertainty. Furthermore, since your layer σ_3 and σ_2 depend on σ_1 you have another constrain for your solution which could be understood as a priori information. The question is how do you account for this kind of uncertainty?

Response: Maybe our description was confusing. In the profile set, we excluded profiles with strong elevated layers, but the aerosol extinction in different layers is independent in the retrieval. We have refined the description of the look-up table in Sect. 3.5 to make it easier to be understood (see Page 14, Line 17 to Page 15, Line 21).

P16, L17: In $\chi^2 < 1.5M$, is M the number of LOS? From an OEM point of view, more LOS mean usually a higher information content. But for your approach, more LOS mean a larger χ^2 criterium and

therefore more possible profiles in your weighted mean calculation. Could you please explain this issue?

Response: Yes, M is the number of elevation angles, which is defined in the text (see Page 22, Line 19). According to the definition of χ^2 , when M increases, χ^2 would also increase. Therefore, the criterion is not really changed. This is also the case for the stopping criterion of the OEM retrievals. For more details, please refer to Rodger, 2000.

Section 4.3: Please add information on the used OEM algorithm and the RTM including parametrization of the OEM retrieval (e.g the definition of a priori and measurement covariance matrices, climatology, vertical grid...). Maybe in a table? The OEM solutions do not seem to be constrained enough (too many oscillations).

Furthermore, box-like true profiles would also be problematic for lower altitude sites and higher AODs due to the a priori smoothing. Please add also a retrieval for an exponential true profile (an elevated true profile would also be interesting). One problem arises by saying that the shown true profile (nearly box-like) is representative for UFS but you use an exponential a priori profile for the OEM. Since the a priori profile is the best (first) guess of the true atmosphere, an exponential profile is insufficient here (in contrast to typical retrievals in the PBL). A better a priori would be a Boltzman distribution or maybe an exponential profile with an even larger scaling height.

Response: We have supplemented two more examples of synthetic data retrieval, one with an exponential profile and one with an elevated layer. The retrieval settings are also supplemented (see Page 34, Lines 5-14). Compared to the previous version, we set a stronger constraint to the a priori profile for the OEM retrieval. However, further optimization of the OEM retrieval is beyond the scope of this paper.

Additionally, please add a graph showing the simulated and retrieved DSCD including an RMS value of the difference between both DSCD as I don't think that the noise-free OEM solutions are that bad but might describe the measurement well.

Response: The simulated and retrieved DSCDs as well as the RMS of the difference are shown in Fig. 2. Since the manuscript is already very long and the OEM retrieval is not the main focus of the paper, we only briefly summarized the results in the text (see Page 34, Lines 17-18) without showing the plots in the manuscript.

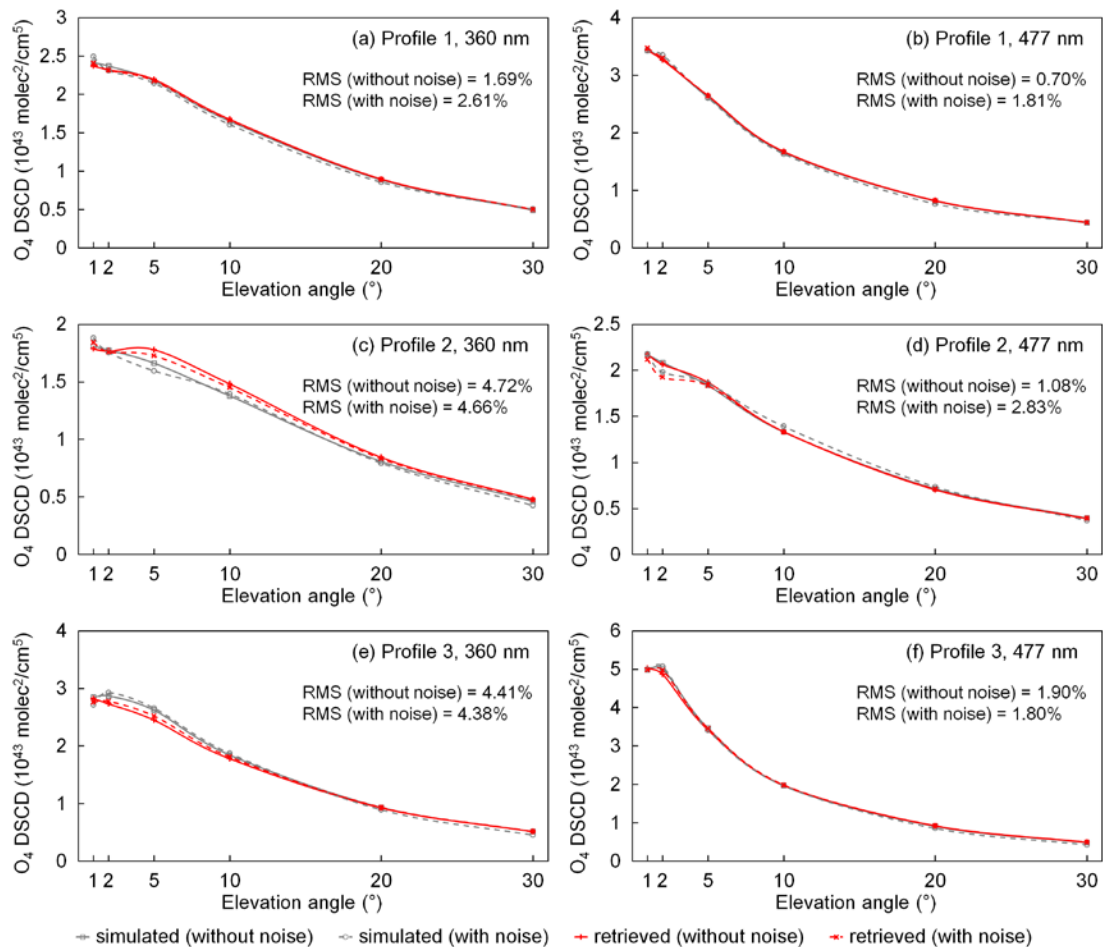


Fig 2. Simulated and retrieved DSCDs and the RMS for the synthetic study.

Section 4.4 and Fig. 9: The reason for such a large difference between sun photometer and MAX-DOAS AOD is still unclear to me. I agree that aerosols in higher altitudes might be responsible for a difference but this is true as well for measurements in the PBL. But here, the introduction of a scaling factor leads to a much better agreement. The relative amount of aerosols in altitudes higher than 2km might be responsible but this is just a guess without prove. Could the authors please take a look at the Ceilometer profiles to solve this issue (see also the following comment for Fig.10)?

Response: From the averaged ceilometer profiles we observed that there are significant amounts of aerosols above the MAX-DOAS retrieval height, see Fig. 1. The results indicate that the aerosols above the retrieval height contribute 30-50% to the total AOD. We have supplemented this result in Sect 2.3 (see Page 7, Lines 15-16).

Furthermore, which kind of scaling factor (SF) is needed to bring the MAX-DOAS AOD to the sun photometer level and how large is the difference to the actually applied SF? Is there also a clear seasonal pattern in your SF? If yes, maybe your way of how to retrieve LOS depending SF is not

optimal?

Response: As we assume that the scaling factors are constant (for each elevation angle), it is impossible to derive a scaling factor which can bring the MAX-DOAS measurements to the sun photometer observations for all conditions. In addition, since the scaling factors were derived based on a huge amount of data and the AOD varies within a relatively narrow range in a single season, data within a single season are insufficient to derive a representative scaling factor. For example, our determination method for the scaling factors at high elevation angles requires measurements under low aerosol load ($AOD < 0.03$), but such measurements are not available in summer. Therefore, it is not feasible to derive a seasonal pattern of the scaling factors. We have further clarified in Sect. 3.9 (see Page 26, Lines 9-10).

Fig. 10: Please add also similarly averaged Ceilometer profiles and an error range for your profiles. Since a validation instrument is available, a comparison should be shown. The ceilometer backscattering signal could be scaled with the sun photometer AOD (see e.g. Wagner et al. 2019 for details). With this kind of comparison you could also assess how much aerosol is located at even higher altitudes than 2km.

Response: Please note, that the retrieval of the AOD from ceilometer data is *per se* not possible, at least with a sufficient accuracy to allow a strict validation. To cover the reviewer's point, we can however use ceilometer measurements for a consistency check by considering the paper suggested by the reviewer: we have supplemented the ceilometer profiles scaled using the method described in Wagner et al. 2019 in Fig. 1, and also supplemented error ranges for Fig. 15 (Fig. 10 in the discussion paper). The ceilometer profiles show that the aerosols above retrieval height contribute in the order of 30-50% to the total AOD. This explains the differences between MAX-DOAS and sun photometer AODs. This result was supplemented in Sect 4.4 (see Page 34, Line 32 to Page 35, Line 1).

Section 4.5: See comment to P5, L5-6.

Response: The uncertainty of AOD measured by sun photometer is relatively large, and the deviation of Angström exponent would further amplify the uncertainty. We have clarified in Sect. 2.2 (see Page 5, Lines 26-28).

P29, L6: profile → profiles

Response: This typo has been corrected (see Page 38, Line 22).

Appendix B: Here, important information are missing. For example, when you use aerosols in B1, which SSA and phase function is used? In B3 which SSA? In general, which climatology.

Response: We have supplemented the settings in the text (see Page 41, Lines 11-14 and Page 42, Table B1).

P34, L8-9: "using phase functions from Hohenpreißenberg should not have a significant impact on the aerosol retrieval". But the results are only shown for RAA = SZA = 60°. For other geometries it might be important to have an accurate phase function. Especially since you show in Figure 3 that the phase function is one of the largest error sources.

Response: Fig. B3 only shows an example. In our retrieval, the error caused by phase function is estimated using a look-up table which considered all possible solar and viewing geometries. We have revised the description in Sect. B3 (see Page 45, Lines 12-13).

P35, L5-6: the averaging kernels (...) are all close to zero at the altitudes above 2km. That is correct but OEM based aerosol retrievals are iterative approaches which might still get an elevated layer more or less correct even though the kernels look like that. The sensitivity in these altitudes is lower for sure but if there is a dominant elevated aerosol layer, your retrieval is not capable of retrieving it accurately due to the dependencies of the individual layers while OEM algorithms might find an accurate solution (see also comment to **Section 4.3** for a test of an elevated layer).

Response: Our look-up table does not consider extreme cases, i.e. strong elevated layers, as the measurement site is located at a high altitude (2650 m a.s.l.), strong elevated layer is typically either close to our instrument or above the retrieval height. Strong elevated layers can also be included in the look-up table if it is used for the retrieval of aerosol profiles at low altitude sites. In order to reduce computational efforts, we have limited the formulations of the look-up table with only weak elevated layers. On the other hand, the retrieval of the synthetic data showed that OEM based retrieval cannot fully reproduce the elevated layer, either (see Page 32, Fig. 13). We have revised the descriptions in Sect. 3.5 (see Pages 14-15).

References:

Anderson, G. P., Clough, S. A., Kneizys, F., Chetwynd, J. H., and Shettle, E. P.: AFGL atmospheric constituent

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