

Reply to Referee #3

First of all we want to thank this reviewer for the positive assessment and constructive comments.

We addressed these comments as explained in detail below.

The present manuscript addresses the issue of the difference between measured and simulated O₄ dSCDS. Many studies over the last year used correction factors on the measured O₄ dSCDs to achieve a better agreement without finding the physical explanation of these factors. Other studies support that the use of correction factor is not necessary. In previous studies, one possible explanation of this inconsistency was the uncertainties of aerosol information. It is very interesting that in this manuscript, this uncertainty is neglected because of the use of one day of measurements with very low AOD values.

I recommend the publication of the present manuscript. The content is clear, well explained and the manuscript falls into the scope of AMT.

Many thanks for the positive assessment.

Please consider some minor comments:

1. In Figure 1, I see that other days (or at least time windows during some days) have very small AOD values. Why these days are not included in your results? Would it be possible to include them and see if the results agree with the main findings of your study?

The extremely low AODs only occurred on the selected day. Only at the beginning of the following day, still low AODs were measured (< 0.05 at 360 nm). However, during this period, the measurements at low elevation angles were strongly affected by clouds. Nevertheless, we compared the MAX-DOAS O₄ measurements retrieved during that period with radiative transfer simulations. Here, we only made simulations for an aerosol-free atmosphere to limit the effort (and also because of the rapid temporal variation of the AOD). The comparison results are shown below:

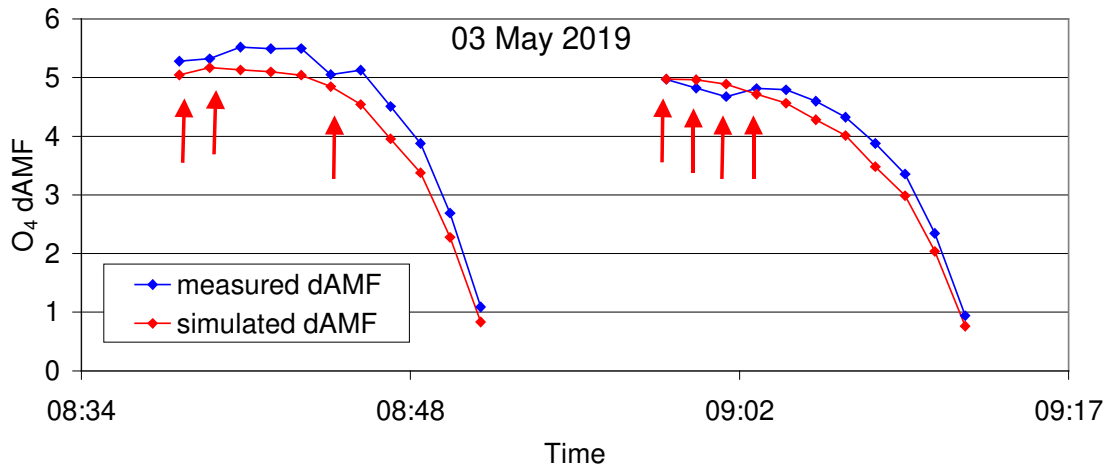


Fig. A13 Comparison of the measured and simulated O_4 dAMFs for two elevation sequences on 05 March 2019, when the AOD was rather small (<0.05 at 360 nm). The radiative transfer simulations were made for an aerosol-free atmosphere.

Like on 02 May 2019, the simulated O_4 dAMFs (for aerosol-free atmosphere) are smaller than the measurements (for cloud-free observations).

We added the following information to section 7.1.:

,It should be noted that that during the entire ship cruise, only during the beginning of 3 May 2019, similarly low (but still larger) AOD were measured as on 2 May 2019. We also compared the measured O_4 dAMFs for the first two elevation sequences on 3 May to radiative transfer simulations. For that comparison we only made simulations for an aerosol-free atmosphere in order to limit the effort (and also because of the rapid temporal variation of the AOD during that time period). The results (see Fig. A13) are similar to those on 2 May 2019: except for the cloud contaminated measurements, the simulations are smaller than the measurements.'

2. The uncertainties that are described in the text are very important for your findings. That would be very useful if you could create a table of uncertainties.

A new table (table 3) was added to the paper (at the end of section 7.2):

Table 3 Uncertainties related to the different analysis steps

Spectral analysis		
Effect	Magnitude	Reference
Spectral fit	1 - 4%	Result of spectral fit
Temperature dependence	1.5%	Wagner et al., 2019
Fit paramaters	3.5%	Appendix A1, and Wagner et al., 2019
Total	4 – 5.5%	
RTM without aerosols		
O ₄ profile	1%	Wagner et al., 2019
albedo	1%	Section 6
RTM general	1%	Wagner et al., 2019
total	2%	
RTM with aerosols		
O ₄ profile	1%	Wagner et al., 2019
AP & SSA	3%	Section 6
Strat aerosols	1%	Section 6.1
albedo	1%	Section 6
Profile shape	2% for elevation angles < 4°, negligible for higher elevation angles	Section 6.1
RTM general	1%	Wagner et al., 2019
total	4%	
O₄ VCD		
	2%	This study, section 5, see also Wagner et al., 2019

Specific comments:

1. P.1, Line 21 : “aside from” instead of “aside”

Corrected

2. P.1, Line 22 : “e.g.,” instead of “e.g.”

Corrected

3. P. 1, Line 32 : “In this study,” instead of “In this study”

Corrected

4. P. 1, Line 25 : Please add some studies that used a scaling factor

We changed the text to:

‘Several studies found that a scaling factor ($SF < 1$) had to be applied to the observed atmospheric O₄ absorptions in order to bring them into agreement with radiative transfer simulations (e.g. Wagner et al., 2009; Clémer et al. 2010). Other studies, however, did not find the need to apply such a scaling factor (e.g. Spinei et al., 2015; Ortega et al., 2016). A more detailed discussion and overview on existing studies of both groups is provided in Wagner et al., 2019.’

We added the following references:

Spinei, E., Cede, A., Herman, J., Mount, G. H., Eloranta, E., Morley, B., Baidar, S., Dix, B., Ortega, I., Koenig, T., and Volkamer, R.: Ground-based direct-sun DOAS and airborne MAX-DOAS measurements of the collision-induced oxygen complex, O₂O₂, absorption with significant pressure and temperature differences, *Atmos. Meas. Tech.*, 8, 793-809, <https://doi.org/10.5194/amt-8-793-2015>, 2015.

Wagner, T., Apituley, A., Beirle, S., Dörner, S., Friess, U., Remmers, J., and Shaiganfar, R.: Cloud detection and classification based on MAX-DOAS observations, *Atmos. Meas. Tech.*, 7, 1289-1320, [doi:10.5194/amt-7-1289-2014](https://doi.org/10.5194/amt-7-1289-2014), 2014.

5. P. 4, Line 156 : Can you provide a possible explanation for this difference between ECMWF and in-situ measurements?

The most probable reasons for the discrepancies are originating from the rather coarse horizontal (~80 km) and temporal (6 h) resolution of the ECMWF interim data set: First, the given model data is the average for the modelled box. Moreover, the simulation uncertainties are increased for parameterized subscale processes (e.g. wave motion) which do affect the in-situ measurements. This information is added to section 5.

6. P. 5, Line 175 : “simulations,” instead of “simulations”
Corrected

7. P. 5, Line 208, 210, Figure 4 : Is there any explanation why the raw data vary more with altitude? Is it valid to use the data above 3 km?

We added the following information to the figure caption:
,The scatter of the range corrected backscatter profiles increases, because the received raw signal scales with the inverse of the square of the distance.’

We used the profile data for altitudes, for which the signal stays positive (after smoothing). The exact altitude, at which the signal is set to zero has negligible influence on the simulated O₄ dAMFs.
We added this information to section 6.1.

8. Figure A8 : y axis varies from 0 km to 10 km and not from 0 km to 0 km. Please correct
Corrected

9. P. 6, Line 221 : why the Angstrom coefficient is assumed equal to 2?

We took the value of 2 from existing publications (e.g. Malinina et al., 2019). However, most provided values are representative for larger wavelengths (typically 525 nm or larger). To estimate the uncertainties of the simulated O₄ dSCDs related to

the uncertainty of the Angström exponent, we performed additional radiative transfer simulations assuming a stratospheric AOD of 0.008 (corresponding to an Angström exponent of 1). We found that the O₄ dSCDs differ from those for a stratospheric AOD of 0.012 by less than 1%. We added this information to the paper.

10. P. 7, Line 276 : “be between” instead of “bebetween”

Corrected