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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Performance of MAX-DOAS measurements of aerosols at Tsukuba, Japan: a comparison with lidar and sky radiometer measurements" *by* H. Irie et al.

H. Irie et al.

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Reply to anonymous referee 3

We thank the reviewer very much for reading our paper carefully and giving us valuable comments. Detailed responses to the reviewer's comments are given below.

Comment 1: I suggest to change the title, for example to 'First retrieval of atmospheric aerosol profiles using Multi-Axis DOAS and comparison with lidar and sky radiometer measurements'. It needs to be mentioned in the abstract, introduction and conclusions that the very first aerosol profile retrievals from MAX-DOAS measurements are presented and that it has been proven that the method actually works.

Reply: Following the reviewer's comment, the title, abstract, introduction, and conclusions have been revised.

Comment 2: The advantages (and also shortcomings) of MAX-DOAS compared to established aerosol measurement techniques should be described in more detail in the introduction (e.g., simple instrumentation, the ability to perform long-term measurements also in remote areas, the ability to retrieve information on the vertical distribution of aerosol in contrast to sun photometers which only yield AOD).

Reply: The revised manuscript now states in more detail the advantages of MAX-DOAS in introduction.

Comment 3: The novel retrieval method should be described in more detail in Section 2.1. The optimal estimation method, as well as the diagnostic quantities derived (averaging kernels, error covariance, degrees of freedom for signal), should be briefly described, and I suggest to include the respective equations.

Reply: We have added some more descriptions and equations.

Comment 4: It should be mentioned that the inverse problem is non-linear and thus an iterative retrieval is necessary.

Reply: Done.

Comment 5: Although already published elsewhere, the general approach and the main features of the radiative transfer model need to be described. What is the general approach of the Monte Carlo model? Are the photon trajectories modelled backward or forward? How many photons are simulated? What is the typical random noise of the resulting airmass factors? How is Mie scattering on aerosols implemented? A definition of the box airmass factor is missing.

Reply: As the reviewer suggests, the revised manuscript now gives more information about our radiative transfer model in section 2.1.

Comment 6: The parametrisation of the aerosol profile using the parameters F1 ... F3 is obscure. Why are the partial optical depths (or alternatively the extinction coefficients) in the four retrieval layers not retrieved directly? The total optical depth could then be determined a posteriori by summing up the partial optical depths, and the respective error and averaging kernel of could be calculated as described in the book of Rodgers (Section 4.3).

Reply: The parameterization used in the present study has an advantage that the retrieval can be made without a priori knowledge of the absolute value of the aerosol extinction, while Frieβ et al. (2006) have argued that using inappropriate a priori constraints on the absolute values of the aerosol extinction can easily cause unrealistic or strongly biased results. Instead of the absolute values, we used a priori information of the profile shape, which is parameterized by the F values, because the (relative) variability of the profile shape, in terms of 1-km averages, was less than that of the absolute value for the period and time of the measurements presented here, as seen from the lidar data (Fig. 5 (Fig. 6 of the revised manuscript)). This is now stated in section 2.1 of the revised manuscript. However, the figure (Fig. 3 of the revised manuscript), showing how the profile is represented by the parameters and how it is dependent on each parameter, has been added to assist the readers' understanding of the parameterization used here.

Comment 7: The parametrisation used here makes the interpretation of the results very difficult, firstly regarding the errors (how do the errors in F1 ... F3 mentioned in Section 2.1 propagate into errors in partial optical depth or extinction coefficient?), and secondly regarding the averaging kernels shown in Fig. 3. What do the averaging kernels of F1 ... F3 and tell us about the vertical resolution and the sensitivity of the retrieved to the true aerosol extinction profile? I strongly recommend to convert the errors and averaging kernels from the F1 ... F3 to the σ1 ... σ3 vector space, which should not be too difficult to do.

Reply: While the parameterization has the above-mentioned advantage, we realize that

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there are disadvantages in that it is not easy to derive the vertical resolution and the sensitivity from the averaging kernels, as pointed out by the reviewer. This is now stated in section 2.1. To overcome these disadvantages, the comparisons with lidar and sky radiometer measurements were helpful, as discussed in section 3. Please note that the results are consistent with those of a simulation by Frieβ et al.(2006) for a MAX-DOAS measurement geometry, presumably because the measurement geometry used for the present study is basically the same as that used by Frieβ et al.(2006). However, we have converted the errors of τ and F values to the aerosol extinction space, as stated in section 2.1 of the revised manuscript.

Comment 8: The vertical resolution is very coarse, and the averaging kernels shown in Fig. 3 indicate that the profile is actually undersampled. What is the reason for choosing such a coarse resolution? Is it because of the high computational effort for calculating the lookup tables? What happens if only a thin aerosol layer is present which cannot be represented realistically by the state vector?

Reply: The main reasons why we have chosen a coarser vertical resolution are (1) to reduce the computational effort for creating the lookup tables and (2) to make the error (variation) of the a priori F values smaller. The presence of an aerosol layer, which cannot be represented by the state vector, would be a source of the systematic error, but the error is likely less than 30% (60%) for aerosol extinctions at 0-1 (1-2) km, based on our results of the MAX-DOAS/lidar comparisons made under various conditions.

Comment 9: The upper model layer extends from 3 - 100 km and is multiplied by a scaling factor. However, the stratospheric aerosol profile is well known from satellite measurements (e.g., SAGE II), and perhaps it would have been a better choice to keep the stratospheric part of the profile fixed and to vary only the upper troposphere, to which the measurements might have some sensitivity.

Reply: We agree that it would be a better choice to keep the stratospheric part of the profile fixed. While it is very unlikely that the choice of the stratospheric profile is a

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major problem with our retrieval algorithm, we will implement this in the future versions of our algorithm.

Comment 10: The a priori has been constructed from lidar measurements, which is a reasonable approach. Why are the non-diagonal elements of the a priori covariance matrix set to zero, although these covariance elements can be calculated from the lidar profiles?

Reply: This might be a good idea to optimize the comparisons. However, we still use the a priori covariance matrix, in which the non-diagonal elements are set to zero, to make the retrieval more independent of lidar measurements.

Comment 11: Aerosol profiles from lidar and DOAS are not independent from each other since the lidar measurements have been used as a priori for the DOAS retrieval. Therefore the comparison between lidar and DOAS needs to be treated with caution. A sensitivity study on the influence of the choice of the a priori on the retrieved quantities is necessary. What happens, for example, if the a priori profile is doubled?

Reply: We have done such sensitivity studies and summarized the results in Table 1.

Comment 12: It is only mentioned that the 'ΔSCD values were retrieved precisely' (P. 9774, L.1), but no errors are quantified. How has the measurement covariance matrix been constructed? Are any systematic retrieval errors considered?

Reply: For each retrieval, we derived two different ΔSCD errors; one from the measurement covariance matrix constructed based on the statistics of the dark counts in nighttime and one from the measurement covariance matrix constructed based on the residuals. Typical values of these errors (1e41 and 1e41 molecules^2 cm^{-5}, respectively) as well as the methods are now stated in section 2.1 of the revised manuscript.

Comment 13: Forward model parameter errors (regarding albedo, single scattering albedo, Henyey-Greenstein parameter, etc.) are discussed in Section 2.1. However, the overall error has been only estimated from comparison with lidar and sun photome-

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ter measurements. A comprehensive intrinsic error budget, as described in the book of Rodgers, is missing. I recommend to summarise the error components in a table and to provide an overall error estimate, which should also include smoothing error and retrieval noise.

Reply: We have summarized these errors in Table 1 of the revised manuscript.

Comment 14: It is mentioned that the degrees of freedom for signal range from 1.0 to 2.7 (P. 9776, L. 2). Is this variation caused by differences in atmospheric conditions, and if so, which conditions yield a higher information content?

Reply: We checked the correlations of the degree of freedom for signal with AOD and SZA, but could not find clear correlations. This may indicate that the variation of the degree of freedom is dependent not only on the atmospheric conditions but also on other factors, such as the quality of the DOAS fit.

Comment 15: How are the lidar profiles degraded to the coarse resolution of the state vector? The correct procedure would be the convolution of the lidar profiles with the averaging kernels of the DOAS retrieval.

Reply: Ideally, the true profile should be used to validate the MAX-DOAS retrieval. While such a profile was unavailable, the present study regarded the high-resolution lidar profile as the true profile. Since a lidar profile degraded with the averaging kernels of the MAX-DOAS retrieval can depart from the true profile, we simply take an average to degrade a lidar profile for each 1-km layer.

Comment 16: Apparently, no cloud screening has been performed for the MAX-DOAS measurements. A sensitivity test has been performed to investigate the impact of clouds on the retrieval, but apparently only under the assumption of a homogeneous cloud cover. What happens if the cloud cover is inhomogeneous, for example if there is clear sky above the instrument, but clouds are present near the horizon (or vice versa), a situation that is not unlikely to occur?

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Reply: We are also interested in such sensitivity studies on clouds. We think, however, that these are beyond the scope of the present work, because a comprehensive study for various cloud conditions (different cloud altitudes, different cloud thicknesses, different spatial distributions, etc.) is needed.

Comment 17: To allow for a better comparison of measured and modelled O4 ΔSCDs, Figures 2 and 3 should be merged to one graph. The scale of the y-axis should only extend to 10 units. Error bars for the measured O4 ΔSCDs should be shown.

Reply: We attempted to merge the two plots, but the merged plot looked very busy, as ten different colors or lines were needed. Instead, the residuals between the measured and modeled ΔSCD values have been added in Fig. 4 (Fig. 5 of the revised manuscript) to show the differences, which could be seen in the merged plot. The scale of the y-axis has been fixed, as the reviewer suggests. Error bars of the measured O4 ΔSCDs are now mentioned in the revised manuscript, but omitted in the figure, because the errors are too small.

Comment 18: Fig. 5 is difficult to read, consider using a different colour scale.

Reply: We attempted to use different color scales, but decided to use the same color scale as in the original manuscript, as it is the best one.

Comment 19: I suggest to prepare a further figure showing the direct comparison between some profiles from lidar and DOAS (e.g., for the period of four days shown in Fig. 2), including the errors from both instruments.

Reply: We attempted to prepare a figure showing the comparison between vertical profiles from lidar and MAX-DOAS. However, it was difficult to decide which profiles should be shown, because the agreement of the profiles above 1 km is dependent strongly on the choice of the profiles, as the agreement is not very good there. We were able to choose the best case (or some specific cases), but we are afraid that it is unfair and may confuse the readers. However, we have added one more plot (Fig. 8 of

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the revised manuscript) of the lidar/MAX-DOAS comparisons in the revised manuscript, although more direct comparisons have been made using a correlation plot (Fig. 7 (Fig. 9 of the revised manuscript)).

Comment 20: Again, the novelty of the work presented here should be stronger emphasised in the conclusions. Are there any plans to improve the retrieval algorithm, e.g., by including the O4 absorption at other wavelengths and/or the measured intensity?

Reply: We have made the argument stronger, as suggested by the reviewer. While we are interested in including the O4 absorption at other wavelengths and/or the measured intensity, we are currently planning to accumulate data for various atmospheric conditions to evaluate the current version of our retrieval method more systematically and also planning to develop a proper cloud screening method, as stated in the manuscript.

Comment 21: The symbol σ, which usually refers to the absorption cross section, is used for the aerosol extinction coefficient. I recommend to use another symbol (e.g., k) to avoid confusion.

Reply: Done

Comment 22: P. 9770, L.26: replace 'dimer' with the more accurate term 'collision complex'.

Reply: Done

Comment 23: P. 9773, L.8: are two polynomials (2nd and 3rd) order fitted simultaneously (which makes no sense)?

Reply: The revised manuscript now states how the two polynomials are implemented in our DOAS fitting forward model.

Comment 24: P. 9773, L. 20: merge the two sentences ('... shown in Fig. 1, where two spectra ... are plotted').

Reply: Done

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Comment 25: P. 9773, L. 21: specify the relationship between differential slant column and optical depth: $\tau = \sigma\Delta$ SCD (with σ being the absorption cross section).

Reply: Done

Comment 26: P. 9774, L. 11: I suppose the 30 min interval corresponds to a complete sequence of elevation angles - this should be mentioned.

Reply: Done

Comment 27: P. 9775, L. 7: replace '300 thousand' with '300,000'.

Reply: Done

Comment 28: Last sentence on P. 9775: it should be mentioned that a value close to one of the diagonal element of the averaging kernel corresponding to indicates that the total aerosol optical depth can be retrieved precisely.

Reply: This is now discussed using the area in section 2.1.

Comment 29: P. 9779, L. 20: replace 'the underestimate in' with 'the underestimation of the'

Reply: Done

Comment 30: P. 9780, L. 4: replace 'nonnegligible' with 'non-negligible'.

Reply: Done

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 9769, 2007.

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