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Interactive Comment

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.

Anonymous Referee #3

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The manuscript entitled 'Performance of MAX-DOAS measurements of aerosols at Tsukuba, Japan: a comparison with lidar and sky radiometer measurements' by Irie et al. presents the first retrieval of aerosol profiles by Multi-Axis DOAS measurements.

Aerosols represent the largest uncertainty in the assessment of the prediction of changes in the Earth's climate. Therefore new approaches for the measurement of atmospheric aerosol properties, in particular regarding their vertical distribution, are highly desirable. The ability MAX-DOAS measurements of O_4 to retrieve information on atmospheric aerosol measurements has only been shown qualitatively (Wagner et al, 2004) and investigated in a theoretical study (Frieß et al., 2006) so far. The work of

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Irie et al. presents the first application of this approach to real data. The comparison to other aerosol measurement techniques (lidar and sun photometer) shows remarkably good agreement. Therefore this manuscript describes pioneering work, which should be emphasised much stronger. From the way the paper is written, the reader could have the impression that the method is already well established and that this is only one of many intercomparison papers. 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.

I recommend the publication in ACP after some minor modifications as described below.

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).

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

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

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airmass factors? How is Mie scattering on aerosols implemented? A definition of the box airmass factor is missing.

The parametrisation of the aerosol profile using the parameters $F_1 \dots F_3$ 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 τ could be calculated as described in the book of Rodgers (Section 4.3). The parametrisation used here makes the interpretation of the results very difficult, firstly regarding the errors (how do the errors in $F_1 \dots F_3$ 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 $F_1 \dots F_3$ and τ 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 $F_1 \dots F_3$ to the $\sigma_1 \dots \sigma_3$ vector space, which should not be too difficult to do.

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?

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.

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

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zero, although these covariance elements can be calculated from the lidar profiles?

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?

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?

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 photometer 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.

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?

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.

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 situa-

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tion that is not unlikely to occur?

To allow for a better comparison of measured and modelled O_4 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 O_4 ΔSCDs should be shown.

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

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.

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 O_4 absorption at other wavelengths and/or the measured intensity?

Technical corrections:

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.

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

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

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

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

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

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P. 9775, L. 7: replace ’300 thousand’ with ’300,000’.

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 τ indicates that the total aerosol optical depth can be retrieved precisely.

P. 9779, L. 20: replace ’the underestimate in’ with ’the underestimation of the’

P. 9780, L. 4: replace ’nonnegligible’ with ’non-negligible’.

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