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Atmospheric Measurement Techniques Discussions

Interactive comment on "Multiple wavelength retrieval of tropospheric aerosol optical properties from MAXDOAS measurements in Beijing" *by* K. Clémer et al.

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Answer to anonymous Referee #2:

The revised version of the manuscript is added as a supplement.

The discussion concerning the correction factor for the cross section has been rephrased in the revised version of the manuscript. The difference observed between the measured and simulated O4 DSCDs could possibly originate from a different source than errors in the O4 cross section. Therefore we do not want to put the emphasis on a correction for the cross section, but on a correction for the O4 DSCDs thereby accounting for systematic errors. One possible error source is the cross section, but

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since this is only a suggestion we believe that it is more appropriate to use a more general description.

General comments

-The retrieval of aerosol properties from MAX-DOAS is based on the measurement of the O4 dSCD, which serves as an indicator for the length of the light path and thus for aerosol extinction in the atmosphere. It is therefore crucial for the quality of the results that the retrieved O4 dSCDs are not subject of systematic errors, which would introduce artifacts in the resulting aerosol extinction profiles. As pointed out in the manuscript, the uncertainty in the absolute calibration of the O4 cross section is likely to represent a significant error source. In section 2.2, it is suggested that the true O4 absorption cross-section is 25% larger than measured by Hermans. This correction factor is determined from comparisons of modelled and measured O4 dSCDs at 30_ elevation angle for days with low aerosol load. The description of this method for deriving this crucial correction factor is, however, not convincing since the actual comparison of modelled and measured data is not shown. Please provide a plot showing the comparison between measured and modelled O4 dSCDs at all four wavelengths.

Plots comparing the measured (with and without correction) and simulated O4 DSCDs at the four wavelengths have been included in the revised version of the manuscript.

-Other possible reasons for the observed disagreement are that the radiative transfer model is wrong,

We performed tests using different radiative transfer codes LIDORT, DISORT, and the DAK code and obtained identical results for the radiance simulations. Explicit validation tables of LIDORT against other radiative transfer codes can be found in following references (Spurr et al., 2001; Spurr, 2006).

Spurr, R.J.D., Kurosu, T.P., and Chance, K.V.: A linearized discrete ordinate radiative transfer model for atmospheric remote sensing retrieval, J. Quant. Spectrosc. Radiat.

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Transfer, 68, 689-735, 2001. Spurr, R.J.D.: VLIDORT: A linearized pseudo-spherical vector discrete ordinate radiative transfer code for forward model and retrieval studies in multilayer multiple scattering media, Journal of Quantitative Spectroscopy & Radiative Transfer, 102, 316–342, 2006.

-or that any of the RTM input parameters are inappropriate (e.g., phase function, single scattering albedo, pressure profile, surface albedo). Can you be sure that this is not the case?

Differences caused by errors in the input parameters such as pressure profile, surface albedo, and single scattering albedo are much smaller than the observed differences between measured and simulated O4 DSCDs. They are of the order of a few percent. In general the phase function of the aerosol has a substantial influence on the retrievals. However in cases with a very low aerosol load (i.e., the situations used to determine the correction factor) the phase function and even the shape of the extinction profiles have little influence on the simulations.

-The data set presented here, covering about ten months of measurements and a large range of aerosol loads, offers the opportunity for a thorough investigation of the influence of systematic errors on the retrieval, as described by Rodgers et al. (2000).

A discussion concerning the forward model errors has been included in the revised version of the manuscript.

-It is mentioned that systematic errors might exist (P121, LL2), but such a discussion of systematic errors is completely missing. It is only stated that their impact is small (P123, LL5), but no evidence is provided.

A discussion concerning the forward model errors has been included in the revised version of the manuscript.

-How can the assumption that 'the correction factor applied on the measured O4 DSCDs eliminates all systematic errors on the measurements' (P122, L2) be justified,

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if the uncertainty in the O4 cross section from the calibration procedure described in section 2.2 is still 10% (P118, L26)?

The word “all” has been replaced by “most” in the revised version of the manuscript.

-It is stated that ’small errors in the O4 cross sections lead to large changes in the bias’ (P128,L6). How does this uncertainty in the O4 cross section propagates into the retrieved aerosol profiles quantitatively? I suggest to add examples for comparisons of CIMEL and MAX-DOAS AODs retrieved with/without O4 correction factor for all wavelengths, in order to quantify the uncertainty caused by a wrong O4 cross section.

This has been rephrased in the revised version of the manuscript. Figures have been added illustrating the effect of the correction factor: a correlation plot has been added comparing the retrieved AOD from the uncorrected and corrected O4 DSCDs and a correlation plot has been added comparing the AODs retrieved from the uncorrected O4 DSCDs and the CIMEL values.

-Also, a quantification of forward modelling parameter errors, caused by uncertainties in parameters such as pointing accuracy, aerosol optical properties (phase function, single scattering albedo), pressure profile, surface albedo, etc., is missing. In particular, I could imagine that the optical properties of aerosol particles vary strongly due to the occurrence of different aerosol types in this heavily polluted environment, and that the SSA and phase function adapted from the sun photometer measurements are subject to significant uncertainties.

We studied the effect of the forward modelling parameters on the retrievals and found that only the aerosol phase function has a substantial influence on the results. Figures showing the influence of changing the phase function asymmetry factor by 10% have been included in the revised version of the manuscript.

-Furthermore, it is speculated that there might be an ’additive error’ on the O4 DSCDs from ’poorly fitted H2O and O2’ (P127, LL19), but no attempt made to quantify these

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errors. To what extent does the RMS of the retrieval, as well as the deviation between MAX-DOAS and Cimel AOD increase with increasing H2O absorption?

Quantification of the effect of the H2O and O2 absorptions on the O4 DSCDs is not very straightforward. As suggested we looked at possible correlations between the DOAS fit error, RMS of the retrieval, difference between AODs retrieved from MAXDOAS and the CIMEL values with the water vapour vertical column, but no correlations were found.

-The statement that modelling the radiative transfer without including polarisation yields wrong intensities (P121) remains without further explanation. What is the physical reason, i.e. how can these differences be explained from radiative transfer theory? Radiances simulated by radiative transfer models without explicitly treating polarisation are used in numerous remote sensing applications (also for the Aeronet inversions of aerosol optical and microphysical properties used within this study), and saying that these simulations are in general incorrect is a very strong statement. Since this discussion appears to be beyond the scope of the paper, I suggest to remove it.

In this paper we do not intend to give a full discussion of the effect of polarisation on radiative transfer. However, we do want to illustrate that for the specific MAXDOAS set-up we used, polarisation has a small effect on the observed O4 DSCDs and a somewhat larger effect on the intensities. In our opinion, it is not inconceivable that polarisation has a larger effect on the light intensity than on the length of the light path, and hence on the O4 DSCDs. We believe that this information is of importance for the scientific community developing retrieval algorithms for aerosol optical properties from MAXDOAS measurements. When not accounting for polarisation, a systematic error of up to 15% is added to the simulated intensities (depending on the solar angles and viewing direction), while for the O4 DSCDs a maximum error of 5% is added. The latter illustrates that not accounting for polarisation can not account for the difference we observe between measured and simulated O4 DSCDs. Some changes have been made in the revised version of the manuscript to point this out. It is true that AERONET does not include polarisation in their retrieval algorithm yet. For the direct sun measurements

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this is not a problem, evidently. For the other measurement modes, polarisation could have an effect. However the measurement geometries are different from the MAX-DOAS geometries, and it possible that polarisation has less effect in these situations. In addition it is possible that the errors induced in the aerosol optical and microphysical properties due to neglecting the effect of polarisation are small in comparison to other uncertainties.

-Why is the intensity, compared to the O4 absorption, more sensitive to the pressure profile (P121, L16)?

This statement was indeed an error. It has been removed in the revised version of the manuscript.

-It could be mentioned that, in principle, the stronger sensitivity of the intensity to certain atmospheric parameters offers the opportunity to retrieve these quantities, as outlined by Friess et al. (2006).

This has been done in the revised version of the manuscript.

-Is the smaller information content at 630 nm really due to a higher error in O4 DSCD, as mentioned in Section 4.2 and in the conclusions? The relative error of the O4 DSCD is still only 1.5% at 630 nm (see Fig. 2), and the noise error, quantifying the impact of the measurement error on the retrieval, is even smaller than at 360 and 477 nm (see Fig. 10). Can you exclude the possibility that the smaller information content comes from differences in radiative transfer (e.g., higher visibility at longer wavelengths)?

The theoretical sensitivity study by Friess et al. (2006) concluded that the highest DFS was obtained at 577 and 630 nm due to (1) the higher transparency of the atmosphere and (2) the larger O4 cross section. This is opposite to what we observe here. Therefore we believe that the difference in DFS is not caused by differences in the radiative transfer.

-In addition to the general statements on the impact of weather conditions on the qual-

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ity of the retrieval on P128, it would be useful to have some more quantitative information. For example, how does the RMS difference between modelled and measured O4 dSCD changes when clouds are present?

It would indeed be very interesting to have more quantitative information on the effect of clouds, and different types of cloud cover in the retrievals. This would however require more detailed information on the cloud coverage from ancillary measurements or from a cloud-identification algorithm based on intensities and O4 DSCDs obtained from the MAXDOAS measurements. At the moment we do not have such measurements or method to provide us with the necessary information. But it will be something we will study in more detail in the future.

-How does the agreement between measured and modelled O4 dSCDs, but also the DFS and vertical resolution, depend on aerosol load?

We had a look at the relation between the AOD and the RMS, DFS, and vertical resolution, but no correlations were found.

Specific comments

-P112, L4: Move the coordinates of the measurement site from the abstract to the beginning of Section 2.

This has been done in the revised version of the manuscript.

-P112, L15: 'The results indicate that good quality O4 slant column measurements are essential...': I suggest to change 'indicate' to 'confirm' since this has already been pointed out elsewhere (e.g, Friess, 2006).

This has been done in the revised version of the manuscript.

-P113, L8: Aerosols can not always be considered as 'pollutants', in particular if they occur naturally (although perhaps not that much in Beijing). For the same reason, I suggest to replace 'aerosol pollution' with 'aerosol load' elsewhere (e.g., P118, L7).

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Situations with low aerosol load are characterised as 'low-pollution' (e.g., P118, L21), although the concentration of gaseous pollutants is unknown and might be high.

This has been done in the revised version of the manuscript.

-P114, L6: It would be useful to describe the O4 vertical profile more precisely, by stating that the O4 is the collision complex of O2 and thus its concentration is proportional to the square of the O2 concentration.

This has been done in the revised version of the manuscript.

-P114, L24: Compared to other MAX-DOAS instruments, why can this instrument be denoted 'new generation',

In the revised version of the manuscript the term "new generation" has been changed into the more appropriate term "state-of-the-art".

-and in which way has it been 'optimized for the retrieval of tropospheric aerosol and trace gas properties'?

By mounting the optical head on the suntracker we are now able to perform in addition to the MAXDOAS scans almucantar scans. These measurements can provide additional information on the aerosol (phase function) and on the trace gases (e.g., horizontal distribution). In addition direct sun measurements can be performed. Furthermore the optical design and spectrometers are chosen as such that all main absorptions of interest are measured (e.g., the four main absorption bands of O4) with a suiting resolution and signal to noise ratio.

-P115, L5: Delete the sentence 'Here we present a short description of the instrument.' This is evident because the Section is entitled 'The instrument'.

This has been done in the revised version of the manuscript.

-P115, L6: '... full dual-channel system': what does 'full' mean in this context?

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The word “full” has been removed in the revised version of the manuscript.

-Please explain the term ’two-way splitter’.

The phrase “A two-way splitter fiber optic bundle with rectangular terminations links the output of the optical head with the two spectrometers.” has been replaced by “Three optical fibers with rectangular terminations connected through a fiber optic splitter link the output of the optical head with the two spectrometers.” in the revised version of the manuscript.

-P115, L9: To my knowledge, most MAX-DOAS instruments are able to measure between zenith and horizon. Therefore this is not really a ’wide range of elevations’.

The words “wide range” also refer to the azimuth directions; The latter is not standard for all MAXDOAS instruments.

-P115, L10: It is not clear why this sentence starts with ’In addition’. Is the elevation angle controlled by something else but the movement of the sun tracker?

The words “In addition” have been removed in the revised version of the manuscript.

-P116, LL23: It is mentioned that a subtraction of the DSCD from the current scan can be done, but it is not clear whether this really has been done for the aerosol retrieval. In case of O4, the reason for this approach is certainly not to ’eliminate the stratospheric contribution’ (P116, L24) of O4, which is negligible.

In the revised version of the manuscript we added: “This approach is widely used for the retrieval of trace gas profiles from MAX-DOAS measurements and was adopted here for the aerosol retrievals even though the O4 is mainly located in the troposphere. The advantage is that by using a reference measured shortly after the off-axis DSCDs, we can assume that the aerosol extinction profile is mainly identical for both observations, facilitating the interpretation (Friess et al., 2006).”

-Top of P117: Provide references for the absorption cross sections used for the spectral

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analysis, and specify the polynomial degree.

A table with the DOAS fit settings and corresponding references has been added to the revised version of the manuscript.

-'Ring interference spectrum': why 'interference'?

The word "interference" has been removed in the revised version of the manuscript.

-P118, L4: Replace 'the O4 absorption band' with 'the peak cross section of the O4 absorption band' if that's what you mean. Specify the peak value of the Hermans O4 cross section at 360 nm for comparison with the range of values provided here.

This has been done in the revised version of the manuscript.

-P118, L28, and Eq. 1: Use the common symbol _ instead of 'xs' for the cross section.

Not applicable due to the changes made in the discussion. The symbol is not used anymore.

-P119, L5: Replace beginning of the sentence with, e.g., 'An inversion algorithm was developed, dedicated for ...'

This has been done in the revised version of the manuscript.

-P119, L11: Why are measured O4 DSCDs 'pseudo-measurements'?

The term "pseudo-measurements" has been replaced by "measurements" in the revised version of the manuscript.

-P120, L3: 'The non-linear aerosol inversion problem can then be determined...': The aim is not to determine the problem, but the solution.

The word "determine" has been replaced by "solved" in the revised version of the manuscript.

-Equation 2 describes the Gauss-Newton method which is known to have a slow con-

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vergence rate. Did you consider to use more efficient algorithms, such as Levenberg-Marquardt, with a faster convergence rate?

At the moment we did not yet explore other algorithms. Using our approach of changing the Sa matrix each iteration allows to converge quite fast ($\sim$5 iterations) even when the solution is far from the apriori. But, it could indeed be useful to explore other algorithms, such as the Levenberg-Marquardt algorithm, in the future.

-P122: Since the usage of a fixed a priori poses problems under strongly varying conditions, an iterative scaling approach has been applied. It is understandable that this approach is necessary for practical reasons. However, it should be pointed out that in this case the retrieval algorithm is not optimal estimation anymore and the underlying statistics is not Bayesian.

This has been done in the revised version of the manuscript.

-Did you consider using regularisation methods to overcome these problems?

We did some test with Tikhonov regularisations, but this approach was less successful than using our approach with the changing Sa matrix.

-P123, L4: Please describe how the DFS can be optimised by varying the correlation length.

This optimization was done using the empirical method reported by Hendrick et al. (2004). The reference has been added to the revised version of the manuscript.

-P123, L7: Do NO2 and O3 profiles really influence the O4 airmass factors? This would only be the case if they would have a significant impact on the tropospheric light path. On the other hand, aerosol optical properties are not mentioned as parameters influencing O4 DSCDs.

The NO2 and O3 profile do not really influence the O4 DSCDs. The influence of the aerosol optical properties is discussed in the next paragraph. In the revised version of

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the manuscript the formulation has been changed to make this clearer.

-P124, L20: Provide information on the weather conditions for the example shown in Figure 6.

We do not have ancillary information on the weather conditions. However, from the direct sun measurements and the smoothness of the diurnal variation of the O4 DSCDs, it was most likely a clear-sly day.

-P126, L6: An increase in DFS from 1.79 at 360 nm to 2.14 at 577 nm, i.e. of more than 20%, is more than 'slight'.

The word "slight" has been removed in the revised version of the manuscript.

-P126, L21: Again, the forward model errors can not simply be neglected. A thorough discussion forward model errors is missing.

A discussion concerning the forward model error has been added to the revised version of the manuscript

-P129, L9: I suggest to replace 'We showed' with 'We confirmed' since the strong sensitivity to near-surface aerosols and the quantification of the vertical resolution has been discussed previously (e.g., Friess et al., 2006; Irie et al., 2008).

This has been done in the revised version of the manuscript.

-I would appreciate some remarks on the impact of the uncertainty of the O4 cross sections on the retrieval, and on the necessity for better O4 cross sections, in the conclusions.

This has been done in the revised version of the manuscript.

-I am not sure if Li et al. (ACPD, 2008) should be cited since it has not been published in ACP.

Recently the paper has been published in ACP; the reference has been changed in the

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revised version of the manuscript.

-Figure 6: Please show the real O4 fit errors, and not 10% of the O4 DSCDs as error bars.

This has been done in the revised version of the manuscript.

-Figure 10: The x-axis units (1/km) are missing in the first two columns. For better comparison, it would be useful to show all profile graphs (1st column), but more importantly all error graphs (2nd column), with the same axis scale.

This has been done in the revised version of the manuscript.

Technical corrections

P122, L22: Replace 'decrease ... up to' with 'decrease ... down to' P125, L22: (Xia et al., 2006) is not listed in the references. P126, L6: Replace '2' with 'two'. P128, L3: 'At the longer wavelengths' - remove 'the'. P128, L5: Replace 'are high quality' with 'are of high quality'. Reference Chance and Spurr, 1997: 'Rayleigh': first letter in capital. Reference de Rooij et al., 1984: 'Mie': first letter in capital.

All technical corrections have been made in the revised version of the manuscript.

Please also note the supplement to this comment:
http://www.atmos-meas-tech-discuss.net/3/C517/2010/amtd-3-C517-2010-supplement.pdf

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