**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.

Anonymous Referee #2

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The manuscript entitled 'Multiple wavelength retrieval of tropospheric aerosol optical properties from MAXDOAS measurements in Beijing' by Clémer et al. describes measurements of aerosols by Multi-Axis DOAS over a period of 10 months in a heavily polluted environment. The results are obtained using a newly developed retrieval algorithm based on optimal estimation. To my knowledge, this is the first time that data from all four dominant O_4 absorption lines in the UV/Vis are presented. The retrieval of aerosol properties from MAX-DOAS represents a new technique, and the comparison of aerosol optical depths determined at different wavelengths with well established sun photometer data is very valuable for the validation of this method, in particular in a polluted environment like Beijing where a wide range of aerosol loads occurs.

The paper is well written and presents the concepts and results in a concise manner. Since the potential of MAX-DOAS to retrieve aerosol vertical profiles is a novel concept, the paper addresses relevant scientific questions within the scope of AMT, and I recommend the publication after some revisions as outlined in the following. These are mainly related to the correction of the O_4 cross section and the missing discussion of systematic errors.

General comments

The retrieval of aerosol properties from MAX-DOAS is based on the measurement of the O_4 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 O_4 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 O_4 cross section is likely to represent a significant error source. In section 2.2, it is suggested that the true O_4 absorption cross section is 25% larger than measured by Hermans. This correction factor is determined from comparisons of modelled and measured O_4 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 O_4 dSCDs at all four wavelengths. Other possible reasons for the observed disagreement are that the radiative transfer model is wrong, 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?

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 in-

fluence of systematic errors on the retrieval, as described by Rodgers et al. (2000). 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. How can the assumption that 'the correction factor applied on the measured O_4 DSCDs eliminates all systematic errors on the measurements' (P122, L2) be justified, if the uncertainty in the O_4 cross section from the calibration procedure described in section 2.2 is still 10% (P118, L26)? 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 O_4 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 O_4 correction factor for all wavelengths, in order to quantify the uncertainty caused by a wrong O_4 cross section. 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. Furthermore, it is speculated that there might be an 'additive error' on the O_4 DSCDs from 'poorly fitted H_2O and O_2' (P127, LL19), but no attempt made to quantify these errors. To what extent does the RMS of the retrieval, as well as the deviation between MAX-DOAS and Cimel AOD increase with increasing H_2O absorption?

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.

Why is the intensity, compared to the O_4 absorption, more sensitive to the pressure profile (P121, L16)? 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).

Is the smaller information content at 630 nm really due to a higher error in O_4 DSCD, as mentioned in Section 4.2 and in the conclusions? The relative error of the O_4 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)?

In addition to the general statements on the impact of weather conditions on the quality 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 O_4 dSCD changes when clouds are present? How does the agreement between measured and modelled O_4 dSCDs, but also the DFS and vertical resolution, depend on aerosol load?

Specific comments

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

P112, L15: 'The results indicate that good quality O_4 slant column measurements are essential...': I suggest to change 'indicate' to 'confirm' since this has already been pointed out elsewhere (e.g, Friess, 2006).

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). 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.

P114, L6: It would be useful to describe the O_4 vertical profile more precisely, by stating that the O_4 is the collision complex of O_2 and thus its concentration is proportional to the square of the O_2 concentration.

P114, L24: Compared to other MAX-DOAS instruments, why can this instrument be denoted 'new generation', and in which way has it been 'optimized for the retrieval of tropospheric aerosol and trace gas properties'?

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

P115, L6: '... full dual-channel system': what does 'full' mean in this context? Please explain the term 'two-way splitter'.

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'.

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?

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 O_4, the reason for this approach is certainly not to 'eliminate the stratospheric contribution' (P116, L24) of O_4, which is negligible.

Top of P117: Provide references for the absorption cross sections used for the spectral analysis, and specify the polynomial degree. 'Ring interference spectrum': why 'interference'?

P118, L4: Replace 'the O_4 absorption band' with 'the peak cross section of the O_4 absorption band' if that's what you mean. Specify the peak value of the Hermans O_4 cross section at 360 nm for comparison with the range of values provided here.

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

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

P119, L11: Why are measured O_4 DSCDs 'pseudo-measurements'?

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

Equation 2 describes the Gauss-Newton method which is known to have a slow convergence rate. Did you consider to use more efficient algorithms, such as Levenberg-Marquardt, with a faster convergence rate?

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

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

P123, L7: Do NO_2 and O_3 profiles really influence the O_4 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 O_4 DSCDs.

P124, L20: Provide information on the weather conditions for the example shown in

Figure 6.

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'.

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

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).

I would appreciate some remarks on the impact of the uncertainty of the O_4 cross sections on the retrieval, and on the necessity for better O_4 cross sections, in the conclusions.

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

Figure 6: Please show the real O_4 fit errors, and not 10% of the O_4 DSCDs as error bars.

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.

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.

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