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Atmospheric Measurement Techniques Discussions

Interactive comment on "Initial investigation of the wavelength dependence of optical properties measured with a new multi-pass aerosol extinction differential optical absorption spectrometer (AE-DOAS)" *by* R. T. Chartier and M. E. Greenslade

Anonymous Referee #1

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This manuscript describes the results of calibration experiments performed on a newly developed multi-pass aerosol extinction differential optical absorption spectrometer (AE-DOAS). Uncertainty in the aerosol direct effect has been identified in successive IPCC reports as a barrier to accurately predicting future climate change and thus instruments with the capability of taking in situ measurements of aerosol optical properties are highly desirable. The particular advantage of the AE-DOAS lies in its ability to record particle extinction simultaneously at all wavelengths spanning a nominal range

of 220 - 1050 nm, giving it the potential to determine the Angstrom exponent from high resolution extinction data. Such wavelength dependent optical property measurements are crucial for accurately calculating the aerosol direct effect. The manuscript therefore represents an advance in the field of optical property research and I would recommend its publication in AMT subject to the revisions detailed below:

1. On pg. 6317, line 17 the Angstrom exponent is defined as alpha, whereas the discussion starting at line 21 on pg. 6318 quotes values of (alpha)abs. Please could the authors make clear any distinction between these two quantities.

2. pg. 6319, line 20. Please state explicitly that the quantity referred to as being manufactured by Thermo Scientific are the PSLs. This is not made clear.

3. The CPC counting efficiency is stated in the manuscript as $\pm$ 10%. Is this uncertainty in the particle number density carried forward into the determination of the extinction cross-section of the particles (such as shown in Figure 6)? The text on pg. 6327 states only that the error bars on the data arise from the variation between 11 repeat measurements. A recently published paper highlights the importance of accurately knowing the counting efficiency of CPCs when using them for making measurements of aerosol optical properties (Aerosol Science and Technology 45, 11, (2011) 1360-1375).

4. To extract the aerosol extinction cross-section shown in Figure 6 from the raw data the aerosol number density must be known. My understanding of this analysis is that it requires the particle concentration to be assumed to be uniform along the entire instrumental path length. Given that the transit time of the aerosol through the multi-pass cell was found to be at least 8.5 – 10.5 minutes and that the particle concentration measured by the CPC was observed to vary as a function of time, can the authors please comment as to how representative the assumption of uniform particle concentration will be for their instrument ie. on what time scale did the CPC count vary with respect to the transit time?

5. The integration time for the detection limit measurements shown in Figure 2 is given as 180 s. What was the integration time for the PSL measurements and how does this relate to fluctuations in the number density measured by the CPC?

6. Given that spacers are used inside the multi-pass cell to reduce the internal volume, the aerosol flow will be turbulent and this may give rise to deposition of particles in corners/along edges etc. When performing our own optical property measurements we found that such deposited particles could be re-entrained in the flow during a background, supposedly particle free measurement , leading to spurious results. Please could the authors comment on whether they observed such effects in their measurements or if there was any evidence of particle deposition within the cell eg. if they performed transmission efficiency measurements for the PSLs through the apparatus for example.

7. Please consider moving the section starting on line 18, pg 6325 and ending on line 7, pg 6326 into the results section rather than having it in the experimental section. It provides specific details of the three RI formalisms that the data is compared against rather than just the mechanics of the comparison process that is used and in my view would tie in better with the discussion starting on line 26, pg 6327. Please also make it clear by writing explicitly in the text that it was Ma et al. and French et al. who performed the fits to experimental data to determine the RI values that are discussed, rather than just putting in the references. On first read through it was not apparent whether these were experimental fits performed by the authors themselves.

8. Pg. 6326, line 7-8: 'The calculated Mie extinction values were then compared with the experimental results to complete a closure loop'. Its current form and positioning in the text make this sentence confusing. My understanding of what you mean by a closure loop is that you are comparing the experimental extinction values with predictions from Mie theory for a range of refractive indices to determine which RI value best fits the data, such as in Figures 7 and 8. The fact that this sentence appears directly after the discussion of the three different RI formalisms that you identify implies that it is only

RI values which are generated by these specific formalisms which you are considering. Is this the case and if so, why were a wider range of RI values not considered? Also, the data shown in Figure 6 does not seem to be a closure study, rather a comparison of the experimental data with predictions for the extinction cross-section calculated using the three different RI formalisms. Please make this distinction clearer.

9. Pg. 6327, line 26 to pg. 6328, line 13: My understanding of this section is that in the simulations shown in Figure 6 performed using the manufacturers RI and the Ma et al. RI treatments only the real part of the refractive index has been considered. This is in contrast with the French et al. RI treatment where both the real and imaginary parts of the refractive index have been varied with wavelength. Please explicitly state the constant values of the imaginary part of the RI that were assumed for the manufacturers and Ma et al. RI calculations. It should be more transparent to the reader that the French et al. treatment is the only one which considers a variable imaginary part of the refractive index and thus this may be the reason why it agrees best with the experimental data.

10. Pg. 6331, lines 9 – 12: 'This type of depiction of extinction efficiency displays broad maxima and minima called interference structure which is due to constructive and destructive interference, respectively, between rays of scattered light'. This statement is incorrect. Maxima in the extinction efficiency are caused by destructive interference with minima being caused by constructive interference. The extinction efficiency is the ratio of the particle extinction cross-section to its geometrical cross-section and tells you how much more light a particle interacts with than its geometric area suggests. Destructive interference between light rays passing through and around a particle would increase its extinction cross-section, thus increasing the value of its extinction efficiency and giving rise to a maxima, not a minima.

11. I feel that it would be of great benefit to the reader to include a figure which shows how the real part of the RI for the Ma et al. formalism, and the real and imaginary parts of the RI for the French et al. formalism vary over the wavelength range under

consideration. Figure 6 shows the difference in the extinction cross-sections calculated with the three RI formalisms but a figure showing the refractive index explicitly would provide more detailed information about how much the RI has to change by to give the differences observed in Figure 6. This would immediately provide the reader with a feel for how accurately the refractive index must be known to reliably predict aerosol optical properties. Such a figure would be especially pertinent for the imaginary part of the refractive index as small changes in this value are observed to have a large effect on the particle extinction efficiency, as shown in Figure 8. I would therefore strongly recommend the inclusion of such a figure in the manuscript.

Typographical errors:

Pg 6317, line 27: change later to latter pg 6319, line 9: change later to latter pg 6330, line 17: change provide to provided pg 6333, line 9: change retrieve to retrieved Figure 6 caption: Ma et al. data is shown by solid triangles not solid diamonds.

Interactive comment on Atmos. Meas. Tech. Discuss., 4, 6315, 2011.