

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

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The authors thank Referee #1 for the insightful suggestions and thoughtful comments. The points brought up by the referee are discussed 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.

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To clarify this oversight, the authors have added the following sentence below the introduction of the Angstrom exponent as alpha: The Ångstrom exponent formulation has also been used to refer to the wavelength dependent absorption, extinction or scattering of particulate mater and these alternate representations will be noted in our manuscript with the addition of subscripts.

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.

We appreciate this suggestion and have added the clarification underlined below for each of the presented cases. We consider several values for the RI reported near 589 nm: French et al. (2007) report $1.591 + 0.047i$ for PS-677 at 590 nm, Ma et al. (2003) report 1.584 for PSLs at 590 nm, Nikolov and Ivanov (2000) report 1.592 for thin film polystyrene at 588 nm, while, the manufacturer, Thermo Scientific reports 1.59 for the PSLs at 589 nm (French et al., 2007; Ma et al., 2003; Nikolov and Ivanov, 2000).

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).

This is an excellent point and one that we had considered when writing the paper, however we have added additional clarification. Specifically, our error bars are reporting the precision of the measurements. We did not propagate the uncertainty associated with the CPC as it would be very large by comparison and since we are focusing on evaluating the instrument in this paper (and not reporting much in the way of new RI values for PSLs), this is the uncertainty we wished to report. However, comparisons

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with theory especially at $\lambda=590$ nm where the RI of PSL is well known is used as a confirmation of accuracy of our results. We have added two sentences to the results sections to indicate this: The accuracy of the extinction cross section measurements is governed largely by the CPC counting efficiency (Miles et al., 2010a). The accuracy of our extinction cross section results is confirmed by agreement with a Mie theory model near 590 nm where the RI of PSLs has been extensively studied and minimal absorption is reported.

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?

This is another excellent point. We have clarified this by adding the following to the experimental section: In a typical 45 minute experiment, the particle concentration rises quickly to a maximum in the first 10 minutes, decreases slightly to a steady state and remains within the CPC error for 25 minutes and decreases within 10 minutes after the atomizer pump is turned off. The steady state particle concentration window is used for the relatively short optical measurements. During specific optical measurement of 120 or 180 seconds integration, an equivalent average was used for particle counting results; in a representative experiment, the standard deviation (1σ) on the particle concentration during the steady state window was 4%. In addition, some of the points below plus select corrections made to address points from by Referee #2 will address this issue.

5. The integration time for the detection limit measurements shown in Figure 2 is given

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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?

The second sentence added to address point 4 also addresses this point.

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.

Another great point needing our clarification. We have added two sentences, the first deals with the turbulence: While these spacers may increase the likelihood of turbulence, flow rates are kept low to minimize the impact. And the second deals with possible spurious results: In addition, during the particle free background for aerosol sampling, particle counts reached zero as is necessary to avoid spurious results and thus give no indication of re-entrainment of surface bound particles. In addition, select corrections made to address points from by Referee #2 will address this issue, especially those related to purge flow on mirrors.

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,

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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.

This is a great suggestion. We have moved the text as suggested. Further, we have modified the text to include information about who performed the fits as: The second set of RI data is wavelength dependent and was derived from experimental data fit to determine the reported dispersion coefficients of Cauchy's equation (Ma et al., 2003; Nikolov and Ivanov, 2000). and The third set was determined experimentally and is reported as RI values from 207-1033 nm (French et al., 2007). As noted in the second correction, the French et al. (2007) data was reported directly as RI values.

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.

We have clarified the difference between closure loop (used for RI retrieval and including a larger range of RI values) versus the comparison tool used for the data presented in Fig. 6. The specific sentence mentioned above has been corrected to: The calculated Mie extinction cross sections based on different RI values from the literature were compared with the experimental results.

9. Pg. 6327, line 26 to pg. 6328, line 13: My understanding of this section is that in

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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.

We have added a clarification of the value imaginary part of the RI for the Ma et al. and manufacturer's models: For both the first and second calculations, the imaginary portion of the CRI (k) is set at the negligible value of zero, though other low values could have been selected.

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.

Thank you for catching our mistake in ordering here. We have corrected this as: This type of depiction of extinction efficiency displays broad maxima and minima called interference structure which is due to destructive and constructive interference, respectively, between rays of scattered light. In addition, there are often smaller sharp peaks which are referred to as ripple structure.

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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.

We have prepared and submitted such a figure with our revisions. The new figure presents both the literature refractive index data as well as that retrieved at the two wavelengths used as a case study in this work. The figure can be seen as a supplement to this comment.

We have also corrected the typographical errors pointed out by this reviewer.

Please also note the supplement to this comment:

<http://www.atmos-meas-tech-discuss.net/4/C2570/2012/amtd-4-C2570-2012-supplement.pdf>

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