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Atmospheric Measurement Techniques Discussions

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Interactive Comment

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 #2 for the insightful suggestions and thoughtful comments. The points brought up by the referee are discussed below:

Major Points 1. As noted by Referee 1, the issue of particle flow through the system is a serious issue and not well addressed. It is difficult to ascertain how the particles flow through this system. The fact that there is a 8.5-10.5 minute delay between the peak of the extinction measurement and the peak of the particle concentration measured by a CPC at the output of the monitor indicates to me that the particle concentration is not uniform throughout the cell, at least for a fairly long period of time. Thus, how exactly were the measured particle extinction cross sections calculated from the raw data? My guess is that after some period of time, a steady state is reached where the measured extinction matches the measured number density at the output of the cell. If so, what implications would this have for sensitivity and time response (see next section)? A plot of some data accompanied by a clearer explanation of how the extinction cross sections are calculated would be useful.

We appreciate this information as it informs us that our explanations were not clear to the reviewers. We have chosen to address this with clarifications within the text which we believe improve the explanation of the experimental conditions with regard to the reviewer comments. Specifically this section has been expanded to the following paragraph: Particle concentration varies as a function of time during an experiment. The highest AE-DOAS extinction signal for an experiment is assumed to correspond to the maximum aerosol concentration for the experimental trial. A time offset is defined as the difference between the start time of the AE-DOAS measurement with the highest extinction and the time the maximum concentration was measured by the CPC and is due to particle build up in the volume of the gas cell and the travel time between the gas cell and the CPC. For all PSL measurements discussed in this work, the time offset from the AE-DOAS measurement to the aerosol concentration measurement was between 8.5-10.5 minutes. 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%.

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We have also added clarification of the error bars reported on our optical data, which are the precision of the measurements and give an indication of stability with the following: The detection limit of the AE-DOAS is variable, but can be calculated for any experiment by finding 3σ of the baseline for the last two zero measurements before the sample is introduced. In addition, zeros can be compared before and after sample measurement to indicate a change in baseline, though experimental times are kept as short as possible to minimize drift. Based on the lamp spectrum and detection limit, we will primarily use the instrument in the 235-700 nm wavelength range. The experimental results presented in the subsequent sections focus on the precision of our measurements over a number of trials and also give an indication of the noise levels of the instrument.

2. The description of the noise characteristics of the instrument is somewhat lacking. It is unclear to me how the reported levels of detection were established. It appears that the authors simply accumulate 6 minutes of data before introducing sample and compare two 3 minute averages to one another. If so, that is not particularly reassuring. The authors also ignore the issue of baseline drift which could be significant, especially when using an arc lamp source. Must particle-free baselines be taken every other measurement period (with minimal duty cycle once time has been allotted for cell purging which can take 6-8 minutes at the flow rates quoted) or can this restriction be relaxed in the absence of gas phase interferences? Recording data using particle-free zero air for several hours and then using an Allan analysis would provide a much more complete picture.

The text added in response to Major Point 1 above also addresses these concerns. Further, we have added some notes, which largely resulted from reviewer comments, about future improvements we plan to make to our instrument in the discussion section which address these issues, specifically: We will also be studying a variety of samples in the laboratory and preparing the AE-DOAS for ambient measurements, where the latter will include investigating increased flow rates through the system to reduce the

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duty cycle for obtaining data and adding pressure and temperature monitors to allow Rayleigh scattering corrections. In addition, we will consider substituting the current Xe arc lamp for high power light emitting diodes (LEDs) that are beginning to be used in spectroscopic equipment for increased stability and sensitivity or moving to a broad band cavity enhanced system.

Minor Points 1. Figure 2, as plotted, is not particularly useful except to let the reader know that the instrument does not work well past 750 nm. I would suggest that the wavelengths range be restricted to 250-750 nm which would allow the reader to see wavelength sensitivity variation over the monitor's useful range. I also suggest that the sensitivity be plotted in Mm-1 since that nomenclature is used in the text and is considered standard anyway.

We appreciate this suggestion, however, we have chosen to keep this figure as it was initially presented because we are trying to make the point, as the reviewer notes the figure successfully achieves that the most useful wavelength range for the instrument is from 250-750 nm despite the wider range of the lamp and mirrors. This indicates to us that with a better light source, the instrument could be used over a wider range and we note this at the end of the discussion section. The precision on our measurements reported as error bars in Figures 7, 8, and 9 also give an indication of the sensitivity and use of the instrument which is important and meaningful.

2. Figure 6 is also quite difficult to decipher. Most of the error bars could be eliminated to allow for easier inspection of the actual experimental data and comparison with the French, et al. model. Also, why not plot the abscissa as extinction efficiency as are the other plots that follow? If you wish to use absolute units, why not stick with square micrometers.

Thanks to the referee for this comment. We selected these units to improve the ease of comparison with other values in the literature and have decided to keep these value as extinction cross section. Changing to the extinction efficiency complicates the plot and

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makes it difficult to keep parallel with most other literature figures which plot extinction efficiency versus size parameter yet the typical size which results in a variable size parameter is not changed in the data presented in this figure. We feel that such a change would be confusing to readers. It would also be easy enough for a reader to mentally convert to square micrometers and so we wish to keep the units as initially presented.

3. Another issue is that of mirror contamination. Is there a purge flow to protect the mirrors from contamination? My assumption is that there is not. If so, this should be stated. It is possible that under high particle loadings, and long data accumulation periods, mirror reflectivity could degrade to the point where sensitivity was affected.

To clarify this oversight, the authors have added the following sentences to the experimental section of the manuscript: The mirrors are not protected from contamination by a purge flow; however, sensitivity experiments following heavy use conducted before and after mirror cleaning and realignment show results within experimental uncertainties. 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.

4. As part of the Introduction or Discussion sections, the authors might want to make note of the introduction of "cavity-enhanced" broad band absorption monitors which utilize ICOS techniques over 50-100 nm wavelength ranges. Another possible point is that there are now high power LEDs (up to 1 W) that cover the 360-680 nm range that might offer an upgrade with respect to the use of an arc lamp. LEDs are extremely stable and might increase the

We appreciate this additional information and have included the following sentence in the discussion to address these new technologies: In addition, we will consider substituting the current Xe arc lamp for high power light emitting diodes (LEDs) that are beginning to be used in spectroscopic equipment for increased stability and sensitivity or moving to a broad band cavity enhanced system.

5. At the current sensitivity levels, Rayleigh scattering of air is not much of an issue. However, at 250 nm, it is almost 300 Mm-1 per standard atmosphere. If one uses a particle filter which introduces a pressure drop of 2-3%, this could be a problem if instrument sensitivity increases (e.g., by averaging wavelength bins). Of course, monitoring for pressure and temperature in the sampling cell allows one to correct for this.

Yes, this is indeed a problem for possible future work (especially ambient sampling) with the AE-DOAS. We have added a sentence in the discussion to address these improvements needed for future use as: We will also be studying a variety of samples in the laboratory and preparing the AE-DOAS for ambient measurements, where the latter will include investigating increased flow rates through the system to reduce the duty cycle for obtaining data and adding pressure and temperature monitors to allow Rayleigh scattering corrections. We have also included some clarification in the experimental section that this is not of concern for the work presented in this manuscript: We do not use filters in the experiments presented here, but future experiments will include temperature and pressure monitoring in the gas cell to allow corrections for Rayleigh scattering.

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

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