Response to Comment by G. Smith
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Manuscript Title: Broadband measurements of aerosol extinction in the ultraviolet spectral region

Response to G. Smith Comments:

As the anonymous reviewers have already pointed out, this manuscript is very well written and describes a significant and important advance in the measurement of aerosol extinction. By combining two broadband cavity-enhanced extinction spectrometers, the authors have been able to measure aerosol extinction in the UV/near-UV region of the spectrum that is otherwise difficult to measure.

Extinction using cavity ringdown spectroscopy at 355 nm (Sappey et al., 1998; Baynard et al., 2007; Nakayama et al., 2010; Adler et al., 2010) and 390 nm (Dinar et al., 2008) has been measured previously, but the present instrument provides a complete extinction spectrum from 360 nm to 420 nm.

One of the advantages of this approach is the ability to measure the absorption spectrum, and not just absorption at a few discrete wavelengths, of aerosols in this region which may be of utility in identifying and quantifying the so-called "brown carbon" component of ambient aerosols.

I would like to offer the following points for the authors to consider so as to increase the utility of this excellent paper:

We thank Geoffrey Smith for the positive summary, and we are pleased that our paper has attracted interest within the aerosol community.

1. The anonymous reviewers have mentioned some of the limitations that size selection with a DMA might pose for using the instrument to measure index of refraction of ambient aerosols (e.g. water content, assumption of spherical shape, correction for multiply-charged particles).

In addition to these issues, I wonder if there will be enough particles after size selection to make extinction measurements with the precision required to extract an index of refraction using the Mie fitting algorithm.

For example, in Petterson et al., 2004, the aerosol extinction in a polluted urban boundary layer (Los Angeles, CA) was calculated to be about 5 x 10-7 cm-1 for sub-micron particles. Extinction of aerosol size selected by a DMA would then be something on the order of 100x smaller because of the single-charging efficiency (~ 10%) and selection of a narrow range of mobility diameters from the ambient size distribution (also assumed conservatively to be ~10%). Thus, the size-selected extinction might be 5 x 10^{-9} cm^{-1} or smaller. Does the broadband instrument have the sensitivity to make such measurements with small enough uncertainty so as to make index of refraction retrieval meaningful for ambient aerosols?

In Sect. 3.4, we report the precision (1σ) for our 1 min extinction measurements to be 2.1×10^{-9} and 1.7×10^{-9} cm^{-1} at the center wavelengths of 365 nm and 405 nm. For ground-based measurements with a slowly varying aerosol population, averaging for longer time periods would further improve the precision. Consistent with Smith's calculations, we expect that this technique has the potential to retrieve aerosol refractive index as a function of wavelength in polluted, urban environments. In addition, it is appropriate for biomass burning and smog chamber studies.

That said, we agree with Smith and Reviewer #3 that it will be challenging to measure refractive indices as a function of wavelength in ambient air. For this reason, we present three potential applications of our technique in the Summary and Conclusions. The first of these is straightforward and represents the broadband, ultraviolet equivalent of measurements from existing CRDS field instruments: measurement of total optical extinction as a function of wavelength.

2. Do the authors have an idea of the uncertainty on the correction for doubly- and triply-charged particle contribution to the measured extinction? For example, what are the uncertainties on the DMA transfer theory

and steady-state charge distribution approximation calculations? And, how do these propagate to calculation of the extinction cross section of the singly-charged particle and to the retrieval of the index of refraction? Such an analysis might be beyond the scope of this paper, but it might be worth discussing the estimated uncertainties in the correction.

The uncertainty of the measured size distribution was the one aspect of the error budget that we were not able to quantify experimentally for this paper. We attempted to measure this using two DMAs in series, with one used to select a diameter and the second used to scan the full size distribution. However, two DMAs in series have a total particle transmission of less than 1% (10% × 10%) and we were not able to generate sufficient particle concentrations to achieve useful signal-to-noise. In the future, we would like to use an optical particle counter to measure the size distribution from the DMA. Wiedensohler (1988) indicates that the theoretical uncertainty is less than 3% for the charge distribution.

3. It seems to me that one of the great advantages of this instrument is its ability to measure an extinction spectrum in the 360-420 nm range instead of at a single wavelength. This capability could be very useful in subtracting contributions to extinction by other species, for example NO2. The authors might want to mention this benefit of their approach specifically when discussing ambient measurements (page 138, lines 4-7). In theory, there would be no need to make measurements of NO2 with a separate instrument or even to remove it using an activated charcoal filter.

Simultaneously retrieving gas concentrations and aerosol extinction is a possibility, and we have added this:

Pg. 138, lines 4-8: "The strong absorption by gases in the ultraviolet and visible spectral region, particularly by NO_2, requires either a correction based on independent measurements of the absorbing species (e.g. Langridge et al., 2011), ~~or~~ selective removal of gas-phase absorbers (e.g. using an activated charcoal filter)*, or fitting the absorptions in the measured spectra*."