Response to Reviewers
Manuscript Number: AMT-2012-280
Manuscript Title: Broadband measurements of aerosol extinction in the ultraviolet spectral region

The discussion below includes the complete text from the reviewer, along with our responses to the specific comments and the corresponding changes made to the revised manuscript. All of the line numbers refer to the original manuscript.

Response to Reviewer #1 Comments:

Washenfelder et al. describe a novel, broadband optical extinction spectrometer based on the sensitivity enhancement resulting from the use of optical cavities. The broadband spectrometer comprises dual optical cavities using near-UV LEDs to monitor aerosol extinction from 360-420 nm. Extinction measurements of the long-wavelength channel are found to be in excellent agreement with an additional cavity ringdown channel at 407 nm. This result is an important validation of the broadband measurements because CRDS is now an established technique for measuring the extinction of aerosols. Additionally, the measurements of the two broadband channels agree closely in the spectral overlap region. The authors give a considered analysis of the various contributions to the measurement uncertainty; the combined uncertainty for the extinction measurements is very good for this type of spectrometer. The broadband channels are then used to retrieve the complex refractive index of several model particle systems.

Two approaches are adopted to retrieve the complex refractive index of the sample depending on whether the index varies strongly or weakly with wavelength. The results in this work agree with the few near-UV measurements in the literature and provide high quality values for the refractive index of these aerosols.

The advance made by this work is the application of the broadband optical cavity technique to the study of aerosol optical properties across a continuous spectral region. The near-UV region is particularly significant as there are relatively few of measurements of aerosol extinction in this region, apart from 355 nm (the third harmonic of the Nd:YAG laser). Recent studies of aerosol properties have suggested that some aerosol types, most notably brown carbon, start to absorb strongly at short wavelengths. This effect may have a significant effect on the local actinic flux – as for example, around megacities.

This is a careful and thorough study and I recommend publication of this article.

We thank the reviewer for the positive summary. Listed below are our responses to the specific comments and the corresponding changes made to the revised manuscript.

(1) Although the effect of multiply-charged particles in the DMA is addressed, the reader is not informed whether this is potentially a big effect or not. What proportion of particles would be multiply charged?

We have added text to emphasize the importance of accounting for multiply-charged particles:

Pg. 126, line 15: *"Multiply-charged particles transmitted through the DMA make a significant contribution to the measured cross section, and must be correctly accounted for. For example, with an ammonium sulfate solute concentration of 2.0 g L^{-1} and a DMA size selection of 200 nm, the size distribution includes 71% singly-charged particles (D_p = 200 nm); 23% doubly-charged particles (D_p = 315 nm); and 6% triply-charged particles (D_p = 421 nm). In addition, the optical cross sections of the multiply-charged particles are typically greater than that of the singly-charged particles, with a fractional contribution to the optical cross section that is greater than their fractional abundance."*

(2) The renaming of the technique as BAES is not helpful – the system is a two-channel broadband optical cavity spectrometer and no different in any substantive way than their earlier three channel LED system (Axson et al. ACP, 2011) or their single channel Xe arc lamp system (Washenfelder et al. 2008). The proliferation of different names for the same approach (IBBCEAS/BBCEAS/CE-DOAS) is unhelpful to the community and implies a methodological novelty that does not exist. This is not the first time that aerosols have been measured with broadband systems. Axson et al. should be included in the references.

We agree with the reviewer. The proliferation of names for IBBCEAS/BBCEAS is not useful, and we have used "IBBCEAS" (Fiedler et al., 2003) as the acronym in our previous work.

We were not trying to imply a methodological novelty here. The problem is that "Incoherent Broadband Cavity Enhanced Absorption Spectroscopy" is misleading for this manuscript, because we are measuring the sum of absorption and scattering. The closest correct, general name would simply be "Broadband Cavity Enhanced Spectroscopy (BBCES)." We have decided to adopt this name, and have removed "Broadband Aerosol Extinction Spectroscopy (BAES)" throughout.

We have added this explanation:

Pg. 117, lines 18-19: "~~We refer to our new method and instrument as the Broadband Aerosol Extinction Spectrometer (BAES).~~ *Instead of using the existing acronym of IBBCEAS/BBCEAS, we refer to this method by the more general name of Broadband Cavity Enhanced Spectroscopy (BBCES), because it is a measurement of total optical extinction.*"

We have added the reference to Axson et al. (2011):

Pg. 118, lines 7-8: "Notable changes include the addition of a second channel, replacement of the Xenon arc lamp with LEDs, and changes to the collimating and filtering optics *(Axson et al., 2011)*."

(3) High power LEDs are a superb light source for broadband cavity systems but are limited to wavelengths above 360 nm. Further work is needed to study the optical properties of particles at very short actinic wavelengths (>300 nm), where particulate absorption affects J(O1D). This work goes some way to studying such short wavelengths.

We agree that although our work represents the best UV spectral range that is currently achievable with LEDs, measurements at shorter wavelengths are desirable. An IBBCEAS instrument with an arc lamp source has been used to measure gas-phase absorbers at shorter wavelengths (Chen and Venables, 2011), and this approach could be applied to aerosol measurements in the future.

Minor corrections:

p.115, l.5: "Composition" is a chemical, not physical property.

We have changed this to "…size, shape, and composition are intrinsic physical *and chemical* properties."

Sect. 3.2: The wavelength dependence of the complex refractive index and its components should be made explicit in Eq. (4).

We have changed this.

Fig. 11: Lines of the same colour on figure should be redrawn – it is not clear which line corresponds to which refractive index.

We have changed Fig. 11 to make the traces distinguishable.