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 #3 Comments:

Washenfelder et al. present a new method for the measurement of wavelength dependent aerosol optical properties in the near-UV wavelength region. They demonstrate the utility of this instrument through the measurement of wavelength dependent refractive indices for a variety of absorbing and non-absorbing substances. The manuscript is exceptionally well written and the results presented clearly. I recommend publication after the authors address the below questions.

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.

Are the LED's mounted on adjustable mirror mounts or are they fixed position?

We have modified the text:

Pg. 118, lines 19-20: "~~The output from each LED is collimated using a single F/1.2 fused silica lens and passively coupled into a cavity consisting of two 2.5 cm, 1 m radius of curvature mirrors (Advanced Thin Films, Boulder, CO, USA).~~ *Each LED is mounted in a fixed position, and collimated with a single F/1.2 fused silica lens mounted on a three-axis translation stage. The collimated light is passively coupled into a cavity consisting of two 2.5 cm, 1 m radius of curvature mirrors (Advanced Thin Films, Boulder, CO, USA).*"

P126, Line 19: Although it is certainly challenging to account for the effects of multiplycharged particles on the extinction for "monodisperse" size-selected particles, it should be noted that the actual assumption that needs to be made is that the multiply charged particles have the same refractive index as the singly charged particles. A minimization can then be done that accounts for the contributions of the multiply charged particles in a self-consistent manner.

This is an important point, and we were not explicit about it. We have modified the text:

Pg. 126, line 27: "*This approach assumes that the multiply-charged particles have a refractive index identical to that of the singly-charged particles, which was true for these laboratory experiments.*"

P126, Line 24: Do the authors know if the method they apply here for multiple charge correction is essentially the same as clicking the button "multiple charge correction" on the TSI SMPS AIM software? An answer to this question would benefit the community as there are many such systems in use. My interpretation is that it is the same (or at least similar).

We expect that our calculation of the full size distribution (described in Sect. 3.3.2) is equivalent to the method used in the TSI SMPS AIM software. However, we do not use the TSI SMPS and have not examined the details of the TSI software. We use the Wiedensohler approximation to the steady-state charge distribution, applied to the experimentally determined size distribution, to determine the number of multiply charged particles.

Section 4.2.1: The authors discuss here multiple-charge corrections that are necessary. It would be helpful if they were to indicate (i.e. remind the reader) that this is because the wavelength-independent approach necessitates the use of size-selected particles, and thus they must use the method that they do not favor (as discussed in 3.3.2) to derive the refractive index values.

We added two sentences to clarify this:

Pg. 132, lines 13-15: "Using the wavelength-independent approach (Sect. 3.3.1), we retrieved the complex refractive index from extinction measurements shown in Fig. 5~~, after correcting for multiply-charged particles~~. ***This method is only appropriate for a monodisperse size distribution, and required that we correct the measured optical extinction for multiply-charged particles.*"**

Pg. 132, lines 23-25: "Results from the full retrieval of complex refractive indices as a function of wavelength, derived using the full size distribution, are shown in Fig. 6 for the two ~~BAES~~BBCES channels. ***This method requires no assumption for the refractive index of the multiply-charged particles.*"**

Section 4.2.3: Can the authors offer an explanation for the difference in the Trainic et al. ammonium sulfate refractive index?

We do not have an explanation for the difference between our measurements and Trainic et al. (2011) at 355 nm.

Section 4.3 and 4.4: The authors do not correct their Nigrosin dye or Suwanee River fulvic acid measurements for multiply charged particles because literature refractive index values do not exist. I would encourage them to think about including the multiple charge correction as part of their fitting algorithm explicitly, where the RI being tested during the fitting is used to determine the multiple-charge correction. This allows for correction of multiple-charged particles even if the refractive index is not known a priori with the only assumption being that the single and multiply charged particles have the same refractive index (which is a very reasonable assumption). Alternatively, they could use the results from their multi-wavelength retrieval.

The method that the reviewer proposes is an iterative retrieval, where the correction of the extinction cross section for multiply-charged particles uses the derived refractive index, eventually approaching a convergence. We analyzed our data using this method, and originally described it in the text. However, it is mathematically equivalent to the retrieval method described in Sect. 3.3.2, which uses the full size distribution, and the results from the two methods agreed. To simplify the manuscript, we decided to remove the description of the iterative retrieval approach. We also decided to present the measured optical extinction cross sections for nigrosin and Suwannee River fulvic acid, without a correction for multiply-charged particles.

Section 4.4.2: Can the authors put forward a reason for why the Dinar et al. refractive index for the same Suwanee River fulvic acid is so different than in this study?

In the text, we indicate that Dinar et al. (2008) used SRFA 1R101F and we used SRFA 1S101F. Sample 1R101F is no longer available for sale from the International Humic Substances Society. This is our best explanation for the difference.

Section 5: This is a very useful discussion.

Thank you.

Section 6, P138, L20: Although it is useful to note that uncertainties with respect to particle mixing state, composition of single vs. multiple charges, etc. can be overcome by using the method of Beranek et al. (2012), which uses a DMA in tandem with an aerosol particle mass analyzer, I question whether the BAES method would have the requisite sensitivity to deal with the particle losses that would occur going through both a DMA and APM for ambient measurements (certainly such an approach would work in the lab). That said, the authors are encouraged to try.

We agree that ambient measurements will be challenging, and that low ambient particle concentrations and losses through the DMA and APM will be concerns.