Response to Reviewers Manuscript Number: AMT-2015-100
Manuscript Title:
Spectral Aerosol Extinction (SpEx): A new instrument for in situ ambient aerosol extinction measurements across the UV/visible wavelength range

The comments from the referee are italicized, with our responses in plain text. All of the line numbers refer to the original manuscript.

Anonymous Referee #1

Received and published: 21 July 2015

This paper reports on a new instrument for measuring aerosol extinction. A number of instrument already exist that are capable of this type of measurement, the advantage of this approach, according to the authors, is that it uses a broad band (xenon) light source versus a few specific wavelengths (ie LEDs). This has a number of advantages; better determination of wavelength dependent properties (e.g., assess accuracies of extinction Angstrom exponents), and by extending to lower wavelengths than typical (300 nm) provides a better measure of brown carbon (more on this below). Whether it proves to be an effective approach for measuring brown carbon outside of large plumes remains to be seen; as an indirect measurement, this is challenging. But, overall, the paper is well written and describes a range of careful experiments validating the instruments performance – a very nice paper.

We thank Referee #1 for their compliment.

Abstract lines 12 to 14 relating to brown carbon; while strictly true this instrument will have major limitations measuring brown carbon since it is an indirect measurement (extinction). Furthermore, there are other instruments currently available that also measure absorption in the 300nm wavelength range (7 wavelength aethalometer, although smallest wavelength is over 350 nm).

The sentence in the abstract lines 12-14 has been modified:

"SpEx measurements are expected to help identify the presence of ambient brown carbon due to its 300 nm lower wavelength limit compared to measurements limited to longer UV and visible wavelengths."

An advantage of this instrument is that is does not disturb the aerosol,

measuring it in it's native state (ie not collected on filters etc). How is the instrument likely to be operated in the field, ie at what cell T, and RH, ambient? What about issues with heating of the cell, etc? Ie, how is the large effect of particle liquid water going to be accounted for? Also, enhanced UV light absorption may be due to Brown Carbon or clear shells over absorbing cores (ie, no brown carbon present). Will this method be able to distinguish this? If not, it should be noted, since the paper tends to emphasis the use of this instrument for investigating extinction in the UV range (also, pg 6492 line 2and 3 are not strictly correct when considering this).

In the initial field deployments of SpEx during DISCOVER-AQ, we directly sampled ambient aerosol at ambient RH without drying the particles. We did not control the temperature of the lamp or the temperature of the optical cell. During the first deployment in Houston aboard NASA Langley's MACH-2 ground-based mobile laboratory, we learned a great deal about the initial instrument design and operation under field conditions. Prior to the DISCOVER-AQ Colorado deployment several important modifications were made to the instrument, however additional upgrades remained including monitoring/controlling the T, P, and RH within the cell, controlling the T of the xenon lamp, and making changes to the fiber optics to reduce sensitivity to mechanical noise. These additional modifications are underway and we anticipate future work will be able to report significant reductions in the limit of detection. However, we felt it was important to publish the instrument characteristics as described in this manuscript to provide context for data obtained in Colorado (manuscript in preparation).

The question about RH raises an important point. Typically, ambient measurements are made behind dryers to facilitate comparisons of particle characteristics made at different times and locations throughout the troposphere. However, to compare in situ aerosol extinctions with those from remote sensors, extinctions will also need to be measured at ambient RH for an apples-to-apples comparison of ambient extinction (as opposed to apples-to-apples comparisons of dried particles). Hence, we anticipate making measurements both at ambient RH and behind a dryer depending on the research objectives at hand. In either case, it will be important to monitor and record the RH of the measurement, which future measurements will include.

While future data sets (i.e., SpEx plus additional data) may permit an assessment of BrC v. non-BrC clear shells over absorbing cores, along with

theoretical Mie calculations or other modeling approaches, it is premature to include any sort of speculation along those lines in this manuscript since we lack adequate data to address it. We have softened the language of our expectations for future BrC studies modifying the sentence on pg. 6492 lines 2-3 as follows:

"With its lower wavelength limit of 300 nm, SpEx will facilitate exploration of spectral UV optical characteristics."

Pg 6481, lines 10 and following discussing LODs; what about sampling in the FT (ie, the intro discusses aircraft based deployment of the instrument). Can the instrument be effectively used for FT measurements?

Current LODs suggest that SpEx would not provide adequate measurements in the FT. However, the modifications underway are expected to reduce the LOD and allow for possible airborne deployments. To clarify the restriction to boundary layer measurements for the version of SpEx described in this manuscript two modifications have been made to the text: in the introduction (page 6477 lines 5-8):

"The rack-mounted prototype configuration was deployed aboard the ground-based NASA Langley Mobile Aerosol Characterization (MACH-2) laboratory during DISCOVER-AQ (Deriving Information on Surface conditions from Column and Vertically Resolved Observations Relevant to Air Quality) and obtained ambient spectra at while parked at several ground-sites."

and on page 6481 lines 20-21:

"Thus, this data reduction scheme is able to sufficiently characterize both concentrated plumes and background conditions on reasonable time-scales at ground sites. Future modifications to SpEx are anticipated to reduce the limit of detection such that airborne deployments for measurements in the free troposphere will be feasible."

How will the instrument perform if used for size-selected extinction measurements? Is there sufficient sensitivity? (Noting that in the conclusion it is stated that this is not the major application envisioned).

In the lab, several of our tests were size selected however, as noted in the manuscript for some tests we were unable to generate large enough concentrations to obtain data above our detection limit when attempting to

select for certain sizes. Under typical ambient conditions where extinctions are smaller than those in the laboratory tests, we do not anticipate being able to size select ambient aerosols and obtain spectra above our LOD. This would be an exciting capability if our instrument improvements provide a sufficient reduction in our LOD. We have slightly modified the sentence in the conclusions on page 6492 lines 13-16 to note the difference in LOD between SpEx and BBCES:

"The smaller cavities (each with an approximately 1.5 l volume), slower flow rate, and lower detection limit makes BBCES more suitable than SpEx for retrievals of m(λ) due to the need to size select aerosols to perform the retrieval."

At 300 nm are there aerosol species that absorb other than brown carbon. Eg, were any tests made with nitrate?

This is a great question. There may well be UV absorbers that are not carbonaceous. Unfortunately, we did not test for any nitrate compounds in the tests presented here (see Table S2). However, we will make a point to do so in future laboratory tests and will report those spectra when we have them.

Conclusions, pg 6492, lines 10 – 12. Washenfelder used data from a PILS-LWCC measurement of BrC to do the source apportionment (or any measure of BrC, for that matter) since the BBCES could not detect it. This is a very important point about trying to measure BrC from an extinction measurement.

This is a very good point. We have added the following sentence after that on lines 10-12 on page 6492:

"Note, their source apportionment relied both on BBCES measurements and absorption measurements obtained with the PILS-LWCC (Particle Into Liquid Sampler coupled to a Liquid Waveguide Capillary Cell)."

In addition, on page 6492 lines 24-25 of the conclusions, we have modified the text as follows:

"Such measurements offer a complement to related extant in situ measurements of aerosol chemical, physical, and optical properties. For example, SpEx extinction spectra coupled with PILS-LWCC absorption

spectra will allow for the calculation of scattering spectra, single-scattering albedo spectra, and other spectral optical properties."