Reply to Reviewer 3:

1. "My major concern comes from the measurement of babs. The author used the difference between the bext and bsca to derive babs; however, they do not explain, in a convincing way, how they corrected for truncation effects to calculate bscat. They mentioned they used the correction scheme developed by Qian et al. (2012), but Qian et al. (2012) states "truncation error largely depends on particle size and refractive index," and the authors do not have a size distribution measurement and do not know the refractive index of the measured aerosols. How then they apply the correction? The authors specify they measured without size selection (page 29416 line 23). If the authors assumed a log-normal size distribution and an effective refractive index of the sampled particles, it should me mentioned, supported, and a particle loss calculation or measurement done with it."

REPLY -The experimentally measured correction factor of Qian et al. was employed for this work along with the information presented in figure 2. Figure 2b lists the actual bscat corrections used as a f(RH). At high RH values – the correction model essentially assumes behavior consistent with ammonium sulfate aerosol (quite hygroscopic). In addition, the measurements from Qian et al. considered ammonium sulfate aerosol which has a higher refractive index than aqueous phases. Both of these factors may suggest an overcorrection of bscat at high RH (this would underestimate absorption).

2. Furthermore, how often did the authors do zero measurements for their albedometer? Did they interpolate between each zero measurement to have a 'more realistic' zero for the corresponding particle measurement?

REPLY -In this case the user periodically zeroed the instrument. In general, there was not a very large drift observed between zero points.

3. From high to low relative humidity the CRD mirrors could get 'dirty' and 'clean' affecting their zero particle decay time and consequently affecting their bext measurement, which will in turn affect the babs calculation. The authors even mentioned that incorrect zero measurements could lead to falsely high MAC values, how sure are the authors the high MAC values of Fig. 6 don't come from erroneous zero measurement?

REPLY - The mirrors are unlikely to clean themselves (in my experience this has never, ever occurred). It sounds like you may be referring to condensation/evaporation of water? The purge flow helps prevent condensation on the mirrors. If the mirrors get 'dirty' or acquire condensate,
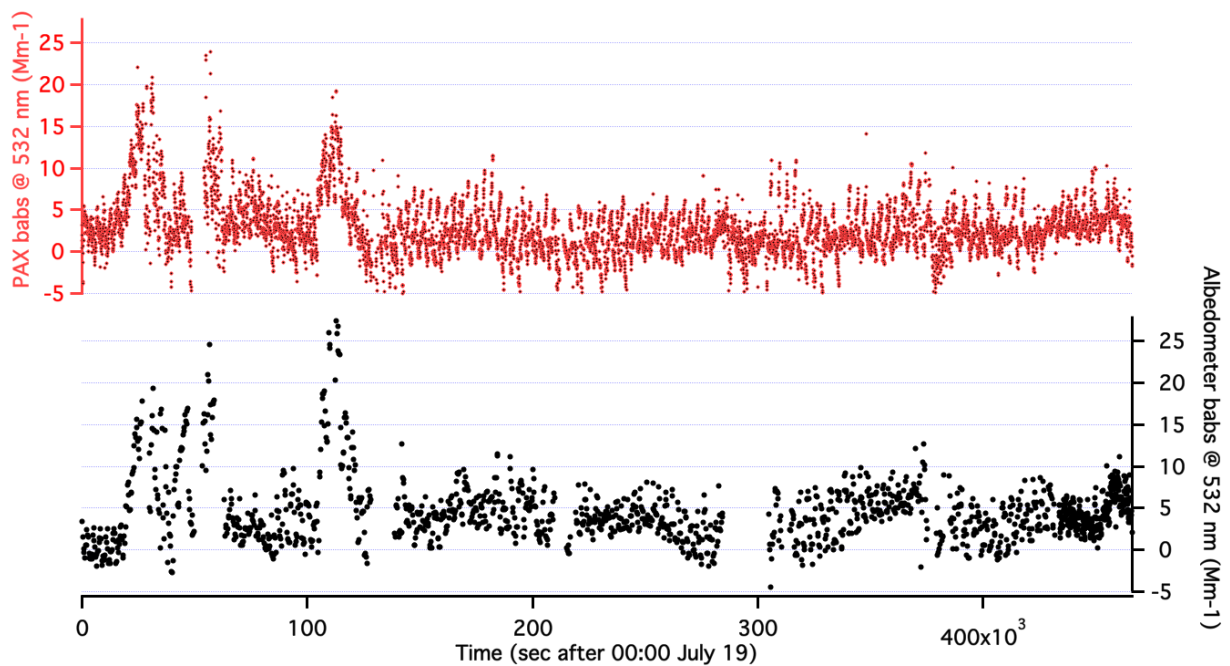
the operator observes an immediate and obvious change in the signal (RD curve completely disappears) and this would persist (as expected) during a filtered air blank. We don't think the results are caused from faulty zeroes because that effect would need to be both correlated with high RH <u>AND</u> high BC coat state for us to see the result of Fig 7. I cannot think of any physical reason why the CRD mirrors would tend to get especially dirty at high RH and high BC coat state, and then clean themselves for the next RH cycle.

4. Especially because the authors say in page 29422 lines 9-11, that the highest measurements of MAC correspond to the periods of low babs. Assuming the truncation correction is robust and particle loss correction was done, from their Fig. A4 it is seems their errors are as large (even larger at some points) than their measured data presented in Fig. 7. It does not seem possible to draw any conclusion from the two figures.

REPLY: Yes, the uncertainty is significant. I suppose that is part of making such a measurement. We would like to make more measurements in the future at a location where BC is higher – this would help reduce relative uncertainty. Maybe one day…. We produced a supplementary file in response to reviewer #2. It shows a plot of babs vs. BC mass conc for RH< 60% and high RH+high coat fraction. The slope of the line for high coat fraction+high RH is larger than the low RH case. In addition it can be seen that a lot of data points for the high coat fraction + high humidity (but low BC conc) cases are a bit above the line near the origin. Those points would have the highest MAC – but are most difficult to pin down quantitatively because they produce the smallest signals on all detectors (e.g. highest propagated relative uncertainty).

5. I do not understand why the authors show they have a PAX (photoacoustic extinctiometer), which according to the DMT website measures babs and bsca at either 870, 532, or 405 nm wavelength (the authors do not mention at what wavelength it was working), from which it derives single scattering albedo (SSA) and BC mass concentration, and not use the data. I think they could have compared their derived babs and SSA with the one from the PAX for their dry measurements. This could have help improve the confidence in their babs measurement after the truncation correction. Why not also use the BC mass concentration from the dry measurements?

REPLY - This is a very reasonable idea. We suspect the BC mass concentration provided by the SP2 LII sensor is more accurate though. The plot below shows the time series babs data for both sensors (red PAS; black difference method) both at 532nm.

6. In section 3.1 the authors mentioned "individual particles measured in Houston were assigned to either being "coated" or "not coated" based on observed lag time – no distinction was made as to the thickness of coating present." But then show in Fig 4b the rBC coated fraction. If I'm not mistaken, the coated fraction comes from the measured optical size from the 1064nm laser and the mass-equivalent diameter distribution of BC cores. The optical size distribution from the SP2 is done through a scattering measurement calibrated with PSL or size selected ammonium sulfate particles. Which calibration did the authors use? If the PSL was used did they correct for refractive index? What are the errors in the coated fraction? Also, regardless of the calibration used, some correction to the measured optical diameters should be applied; the refractive index of water is significantly lower than that of either PSL or ammonium sulfate.

REPLY - We assigned fraction coated from the lag time analysis only (difference in time between incandescence and scattering peaks from sp2 raw data). Our coated fraction refers to the fraction of particles exhibiting lag time > 1 microsecond. It had nothing to do with scatter channel signal for the SP2. In fact we did not use the SP2 scatter intensity for any important measured quantity in this work. Dry air was also used as purge flow for the SP2.
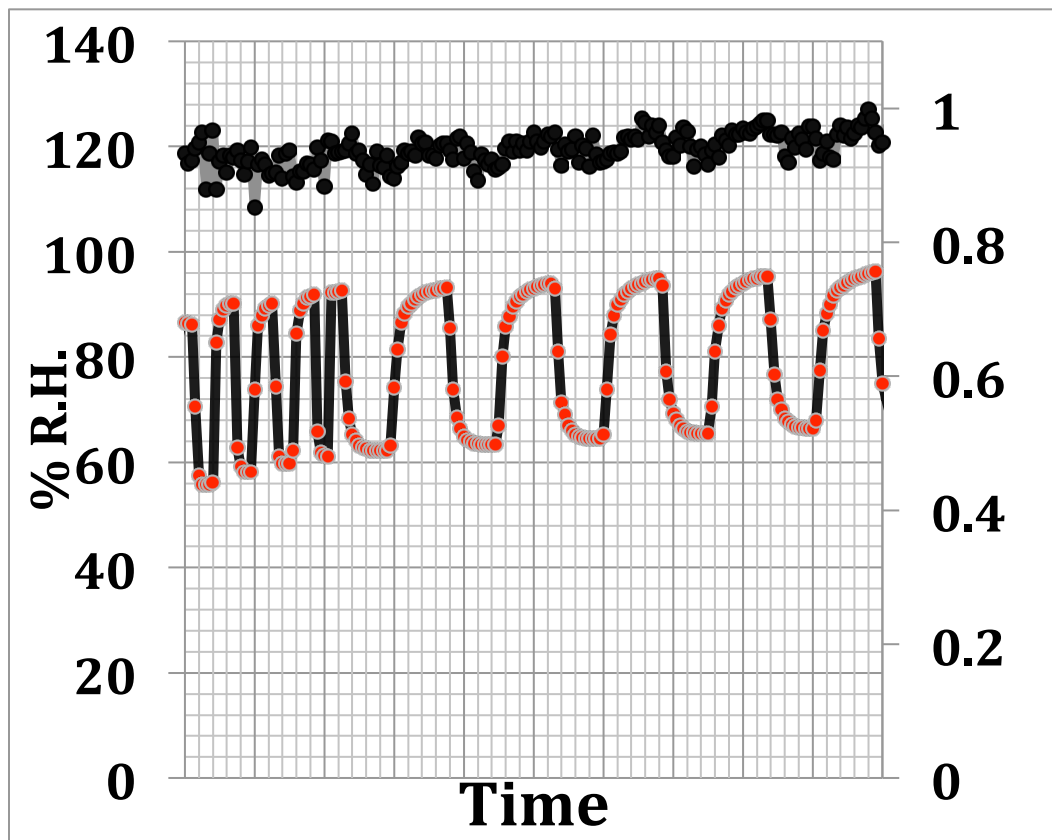
7. The authors show in Fig. 5 the mass scatter coefficient vs RH. They calculated this by taking PM 2.5 measurements which were 100 m away from their sampling line. There is no mention of particle loss calculations through their system, and having a couple of 3-way valves together with their sample rates (up to 4 LMP), they'll most likely have significant particle loss, especially of particles greater than 1 μm. This

together with the truncation calculations makes it difficult, if not impossible, to assess if they are presenting an upper or lower limit at best.

REPLY - Sorry for the apparent confusion. We were in a different building 100 m away – we did not actually sample the aerosol through 100 m long length of tubing. The sampling line for optical measurements was only a few meters in length.

7. Why did the authors decide not to show single scattering albedo? It is a useful variable for modellers. The authors could have also used it to assess differences between the dry and wet aerosol, and to help explain the changes in the MAC. Maybe plot a 'wet' SSA and 'dry' SSA.

REPLY- the table reports mean 'dry' SSA. After reviewing the data, on average the SSA tends to increase <u>very</u> slightly at higher RH values. However, the effect is not as large as one might expect at first guess. It seems the apparent increase in scatter and absorption tend to cancel each other to a large degree. A sample figure is below taken during RH cycling. SSA is plotted in black on second y-axis.

Other time periods illustrated a bit more of an increase at higher RH, but again nothing too dramatic.

8. For Fig. 6, why only plot two specific times and not the whole sampling period? The authors mention: "Prior to 450 ks BC aerosol was fairly dilute and was highly coated. During this period, cycling of MAC in phase with RH was observed. As local activities increased during the morning commute (beginning at approx. 450 ks), additional fresh BC was added to the local atmosphere and the fraction of BC coated was observed to drop" (page 29421, lines 3-7). This phenomenon should then be seen during every morning the sampling lasted (unless there was a weekend in the middle). Furthermore, plotting the whole campaign can help see differences with aerosols coming from different origins (as their Fig. A3 shows). A paragraph in the discussion of the different origin and properties of the aerosol measured should be added.

REPLY - We only highlighted certain times for clarity of presentation. In general we did observe the rush hour peak in BC on weekdays- it was fairly predictable – particularly if winds were low. Weekend behavior was different. In addition, wind direction changed from day to day and this seemed to be an important parameter for the site. Unfortunately, the sampling period was not long enough to draw definitive conclusions about the directional and weekday vs. weekday effects.

9. In page 29421 lines 23-26, the authors mentioned the MAC increases from 3.8 to 4.4 m2g-1 as the coated fraction increased. However, from Fig.7 Inset A, it seems more as a fluctuation. The graph does not seem to show a trend of lower to higher values. Also, the authors should make the markers more easily visible.

REPLY - For 7A this is true. The 'increase' is not significant. Keep in mind this experiment was for dryer aerosol with RH< 60%. We did not see a significant increase in MAC at all when coat fraction increased when RH was low. This is consistent with previous literature on the subject.

10. Minor comments.

The authors should expand on the description of the experimental methods What were the purge flows they used? Did the purge flows affect the RH measurement, by how much?

REPLY -The purge flow is generally small – about 0.25 lpm combined. We measure RH right after the cell (after mixing flows).

-The authors should change the x-axis labels to local time.
-Fig. 4a, the labels are difficult to read, you can increase the font.
-Fig. 6 caption, what does abrupt changes in behaviour mean? And "similar changes"? "In 4b the period. . ." do they mean Fig. 4b or Fig. 6b? Then you mention: "However, such a large drop in coat fraction was not observed in (A)". But in (A) the coat fraction goes down to around 0.5 which is at least similar to the observed in Fig. 6b if not smaller. Wording should be changed in the caption.
-MAC is not defined in the abstract.
-There is no description of the PAX
-Page 29417 line 19: + should be ±
-Page 29418 lines 9-10: "During measurements, the SP2 was configured to save data for between 1 of 5 and 1 of 20 particles observed" Should be rephrased.
Page 29420. I think the explanation of the TEOM instrument should be done in section 2.2.
Page 29420 lines 24-25: "Water vapor condensed into hygroscopic materials at high RH causing deliquescence of particles. . ." How do the authors know the particles deliquesced? Are the authors saying that first some particles deliquesced and then these "new particles" coated the BC cores? Please rephrase or clarify.
Page 29422 line 18: "...because high coat fraction tended to be observed for very dilute, aged BC plumes" How do they define aged BC plumes? From Fig A3. which measurements do they refer to?
Page 29423 line 17: missing "(" before "here called. . ."

REPLY: Thanks for the editing suggestions. We are not opposed to making many of the suggested changes, if the manuscript is accepted. Perhaps we can address these concerns in a revised version after acceptance of data presented.