Atmospheric
Chemistry
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Discussions

Interactive comment on "Atmospheric black carbon can exhibit enhanced light absorption at high relative humidity" *by* Y. Wei et al.

Anonymous Referee #3

Received and published: 29 December 2013

The manuscript, Atmosphere black carbon can exhibit enhanced light absorption at high relative humidity, by Wei et al. investigates the change in the mass absorption cross section (MAC) at different relative humidities in an urban city. The MAC was derived from the measured extinction coefficient (bext) and scattering coefficient (bsca) (by the difference of bext and bsca they present the change in the absorption coefficient (babs)), using an aerosol albedometer together with measured mass concentration of black carbon (BC) by an SP2.

The results from the measurements they performed are important for the community and will be helpful in filling gaps in the knowledge in how the optical properties of BC change as it is transported in the atmosphere. However, the authors seem to have neglected crucial data analysis to support their concluding claim. I believe, as

I'll explain below, that their conclusion can be changed once the missing data analysis is incorporated to the results. I cannot recommend publication of the manuscript in the present form. I believe the manuscript needs modifications and improved analysis before I recommend it for publication.

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 bsca. 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. 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? 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? 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.

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?

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.

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 asses if they are presenting an upper or lower limit at best.

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.

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 ïňĄrst period, cycling of MAC in phase with RH was observed. As local traïňČc/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.

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.

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?

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 $\pm$

Page 29418 lines 9-10: "During ïňＡeld measurements, the SP2 was conïňＡgured 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. . ."

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 29413, 2013.