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

Anonymous Referee #2

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Major comments

The paper entitled “Atmospheric black carbon can exhibit enhanced light absorption at high relative humidity” addresses a topic of major importance within the aerosol sciences, i.e., the light absorption enhancement of black carbon containing particles under ambient conditions (e.g., high relative humidity). The paper presents the results of ambient measurements conducted with the aerosol albedometer (where the absorption is measured by difference of extinction and scattering) and reports significant enhancements of light absorption for coated particles under ambient humidity conditions. The subject is relevant for the scientific community and it would be proper for publication on Aerosol Chemistry and Physics. Recently, several studies have looked at light absorption enhancement, mainly by using photo acoustic spectrometers (which make direct observation of light absorption) sometimes in conjunction with thermal denuders, which allow to remove the effect of coatings and have a direct measurement of the true absorption enhancement by referring to the absorption measured by black carbon cores. Here, the enhancement is measured as a ratio of light absorption at high and low RHs without the use of a thermal denuder (in other words, the coatings are never removed) the results are not really a measure of absorption enhancement by black carbon per se, rather this is a measurement of the response of ambient black carbon-containing particles to high humidity. I realize that this is almost a philosophical issue, but a rather important one, in my opinion. Beside, I see a few major issues with the study. First, the absorption is obtained by difference, and this poses the question of what kind of errors are propagated into the absorption values, especially at elevated relative humidity (RH). We know from previous studies that the absorption enhancement can be very small, and perhaps even smaller that the propagated uncertainties due to a) absorption by difference and b) measurements made at high RH. Because a full uncertainty analysis is missing from this paper, there is no way of establishing in the observations are significant. Second, the truncation correction of the scattering portion could introduce further error and increase the uncertainty. It is not clear to me what may be the effects of truncation (and truncation correction) on the results, especially because the correction should depend on RH, size and chemistry as well (i.e., refractive index). I am also very confused by what size cut was applied to the data. it seems to me that there was not impactor at all for the optical instruments, whereas we know that the SP2 - to which the authors refer for the measurement of mixing state and size - measures in the submicron range. So there is a clear disconnect here between the mass-based and optical-based methods. Furthermore, the lack of PM1 cut to the optical data makes it difficult to isolate the effect of dust and seasalt and in general of supermicron particles on the data. it is likely that supermicron particles do not contribute to the absorption, however they have large truncation, are highly hygroscopic (if seasalt is present as likely given the location) and this might propagate into even larger errors which at this point are not accountable for. For these reasons, I find that the results reported in this...
study to be inconclusive and I am not convinced that there is enough evidence to support the substantial enhancement observed in the data. Therefore, I have to suggest that the paper is rejected because the results cannot be fully verified.

Minor comments

Introduction Page 29415, line 6: the MAC at 550 nm reported in Bond and Bergstrom (2006) is 7.5 m2g-1, and not 4-8 m2g-1

Experimental method Pages 29416-29417: The sampling is not very well described. Were the PAX data used at all? What wavelengths were the optical measurements performed at? Also, it is not clear to me if there is a RH history (cycle) for the sample that the authors have omitted. It is not clear what the actual high and low RH values were in the instruments for the measurements. If the lowest RH was 45%, then I would not define it "dry". There is quite a bit of water at 45% RH.

Optical Measurements Results Page 29420, line 10 (and Figure 5): The MSC seems very low to me. What is the wavelength measurements were performed at, and what were the particle sizes? Page 29421, line 25 (and Figure 7): the MAC seems very low here for 532 nm and dry RH. it should be around 8 m2g-1 and not half of the value. Other studies (Cross et al., 2010) have shown that MAC for pure BC is around 8 m2g-1. Have the authors tried to use the PAX measurement to compare to the albedometer-based absorption to see if MAC changes? Or perhaps the SP2 BC mass are overestimated due to e.g., SP2 data corrections? Also, from the discussion related to the figure it seems to me that enhancement is only observed at RH > 60% and only if the coating fraction is high. Are the increase in MSC and MAC with RH and coating fraction correlated? It seems to me that there are several factor all tangled together here Page 29422, line 15-16: it seems to me that an uncertainty of 2 Mm-1 in absorption by difference at high RH is a really low number

Mechanism of enhanced absorption How is it possible that the authors calculate a MAC of 7 m2g-1 whereas the observations indicate a MAC of 4 m2g-1 for "dry" particles?

Also, they indicate an increase in MAC from 7 to 15 m2g-1 for "wet" and largely coated particles. This is a very large enhancement factor. At the same time the calculated and measured MAC for wet particles seem to agree, and I find this strange given that the "dry" case does not agree at all.

Significance and conclusions The authors state that the measurements may have large uncertainties and that no conclusive statements can be made without knowing the chemistry of the coating fractions. I would add that even the way MAC is derived might have serious issues here.

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