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Atmospheric Chemistry and Physics Discussions

# *Interactive comment on* "Amplification of light absorption of black carbon associated with air pollution" *by* Yuxuan Zhang et al.

**Anonymous Referee #2**

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The authors present measurements from Beijing, focusing on analysis and interpretation of data from a single particle soot photometer. The use the SP2 measurements to infer light absorption and light absorption amplification factors. The technical analysis is of reasonably high quality. The interpretation and discussion could benefit from some stronger quantitative analysis to discern process-based information. The authors also need to clarify how they have calculated averages, and whether they are mass-weighted or not. My specific comments follow below.

General comment on terminology: Throughout the manuscript, the authors need to clarify when they are talking about actual absorption measurements or absorption enhancement measurement, and when they are talking about calculated/theoretical values. There are many, many points where this distinction needs to be made clearly,



starting from the abstract and continuing through the conclusions. I will note only a few points where this is necessary as examples, but there are many more beyond what I have noted.

P1 L20 & 27, and P2 L1 & L3: This should be changed to "theoretical light absorption capability"

P2, L14: The Moffett study does not directly measure light absorption. Their conclusions are based on theoretical calculations. Thus, it is not appropriate here. Same for the Zhang et al. 2016 reference.

P2, L15: It is unclear as written when the authors are referring to theoretical studies versus observations studies. This must be clarified.

P2, L23: This should be revised to clarify that the concept of "more coating materials results in stronger light absorption capability" depends on whether one considers coatings on individual particles versus coatings averaged over the ensemble of particles.

P2 ,L29: It should be clarified that the idea that more materials coat BC under polluted conditions is only true so far as the total amount of BC does not scale equally with the overall amount of pollution. If there were more secondary aerosol but also more BC particles, then it is possible that the average coating per particle is unchanged in more versus less polluted conditions. Also, this statement oversimplifies issues related to mixing state, and whether that secondary material condenses on BC versus on non-BC containing particles. The authors oversimplify here, in my opinion.

P2, L31: it is unclear what "quasi-atmospheric" means. Also, this study ultimately indicates very simply that when you have greater amounts of coating on monodisperse particles the absorption enhancement is larger.

P4, L3: Fig. S2 indicates that the uncertainty in the aethelometer measurements is 10% based on the uncertainty in the "compensation factors." However, this inherently assumes that the measurements by the reference instrument, a MAAP, are perfect and

without uncertainty, which is not true. The actual uncertainty is larger and this should be noted. Also, this assumes that the MAAP perfectly accounts for filter-loading and multiple-scattering effects.

Eqn. 1: This is not so much an equation as a relationship. It does not seem to me that it needs to be called out as an equation.

SP2 limits: The authors indicate a lower size limit of 75 nm in the main text. But, Fig. S4 makes clear that the lowest two size bins are strongly biased low in terms of their concentrations since there is no physical reason for such a sharp fall off in concentration below $\sim$95 nm. It is unclear whether this is taken into account, which would be particularly important when the [PM1] is < 50 ug/m3. Fig. S4 makes clear that there is a counting artifact in the SP below 95 nm, where the detection efficiency falls off rapidly giving rise to an apparent sharp decrease in concentration.

Fig. S7 and P6/L5: it is unclear how this figure addresses uncertainties in the absorption calculations. However, the authors do argue in the supplemental that the "absorption. . .was underestimated by no more than 50%." A 50% uncertainty is very large and this information should not be buried in the supplemental. Further, additional details are required as to how this was determined. The MAC from Mie theory varies as a function of particle size while that from RDG is constant. The RDG MAC for bare BC, using the RI given here, is $\sim$3.2 m2/g at 880 nm. This is relatively small to begin with, so how is the 50% number determined. Also, the argument that "the uncertainty of BC light absorption from the calculation of bare BC properties using Mie theory is no more than 2%" is demonstrably not correct. If the absolute absorption can be underestimated by 50%, then the uncertainty cannot be only 2%. Just because there were few times that bare BC particles were observed does not change this fundamental issue. The uncertainty in the absolute absorption from the calculations is much larger than 2%. And then on P8, L5 it is stated that the uncertainty is 10%. It is ultimately unclear what the actual uncertainty on the calculations is.



P7/L19: This should be Fig. S4a.

P7/L26: This should say "calculated absorption coefficient."

P8/L2: As noted above, the uncertainty on the AE33 measurements is >10%.

P8/L5: is the agreement similarly good at the other wavelengths?

P8/L9: It should be clarified that these are all theoretical studies of the enhancement. None of the citations is to a direct measurement of the enhancement.

P8/L14: It should be clarified to "the calculated Eab".

P8/L16: It would be helpful to show a graph that explicitly has the Dp/Dc and calculated Eabs as a function of PM1 concentration, to help illustrate this point. This could be shown as a mean across all sizes, a weighted mean across all sizes, or for a few select sizes. This would help to illustrate the magnitude of the changes.

P8/L24: I find the term "aging degree" to be ambiguous because it could apply to almost anything. I suggest that here, and throughout, the authors change to a more specifically descriptive language. Perhaps "coating-to-core ratio"?

P8/L25: It would be helpful to frame this in the context of the overall size distribution, i.e. to report the weighted-average values based on the observed PM1-dependent BC core size distributions.

P8/L23: Better as "exponential function with a y-offset". However, this by itself gives little physical insight. In Metcalf et al. (2013), for example, there was a comparison to the expected decay based on the SA/V ratio of the particles and diffusion controlled growth. Here, the authors mention this study but do not connect to it quantitatively. The authors should strongly consider introducing a physical explanation using a semi-quantitative analysis, rather than just an empirical fit. I believe this would strengthen the paper.

P8/L30: it is unclear how the "change rates" were calculated. Are these point-by-point

differences? And, how is the Eab calculated? Is this a weighted average? Also, it's not entirely demonstrated how this is an especially meaningful metric. Isn't the same general information obtained by plotting Eab vs. rBC (for example)? Assuming this is from point-by-point differences, then one would expect that:

k_Eab/k_pm1 =((E_(ab,t2)-E_(ab,t1))/E_(ab,t1) )/((PM1_t2-PM1_t1)/(PM_t1 ))=(PM1_t1)/E_(ab,t1) ·(E_(ab,t2)-E_(ab,t1))/(PM1_t2-PM1_t1 )

How is this generally useful? This could be elaborated upon. Also, given that negative values are allowed, it could be clarified that these are not just "growth" rates. This is really just a susceptibility curve.

P8/L31: The units on the equation are incorrect. It is keab = 4.8kPM1, not 4.8%. The percents cancel.

P9/L1: The statement here relates to one point on a graph of hundreds of points. What is the uncertainty on a single point? Is this meaningful to state? I question whether it is especially meaningful to state the results for this one point. I could randomly pick another point, based on the maximum kPM1 (for example) and conclude that kEab varies slowly with kPM1. This feels to me too selective to be meaningful to include and I suggest it is removed or put in a fuller context.

P9/L2: The authors relate their observations to other studies. However, I do not understand why they only consider values with kEab > 0. Why exclude the negative numbers? Also, to reiterate my above point, are the individual points truly meaningful once one accounts for the uncertainty in the individual points? Using confidence intervals for the slopes would, in my opinion, be more meaningful. Or looking at the distribution of kEab values. It is evident from Fig. 3 that if a histogram of kEab values was made the peak would be around zero, i.e. that the particles are shrinking as often as they are growing, on average. While I do see some value in providing the range of values here, a much more statistical picture would provide much greater value.



Fig. S9 and P9/L5: If the authors were to introduce a more quantitative picture that included an interpretation of why this type of behavior might be expected that would be most welcome. Most likely, this is simply because the net change in diameter for a given amount of material deposited decreases with the size of the particle due to surface-to-volume scaling. Since the particles are larger when PM1 is larger, one would expect the deltaDp/time to decrease with PM1 and thus the Eab/time would also decrease. Of course, this oversimplifies because Eab is not a linear function of deltaDp. But, it would be great if the authors could introduce some physical discussion of why this observed behavior is/is not expected this would increase the value of this observation. P9/L9: I find this discussion about previous studies "ignoring" an aspect to be unclear and suggest it be expanded/clarified. How does the current observation help, specifically, explain these previous studies? It is not abundantly clear. Consider that the variability in the calculated Eabs is actually only $\sim$15% between the low and high PM1 periods in the size range that matters (the BC mass weighted size), based on Fig. 2. These previous measurements would not have been able to discern a 15% difference easily, most likely, in their data anyway. While the current study finds a large theoretical enhancement, what is not found is substantial variability in the enhancement (Fig. 2). The variability in the "growth rate" is inconsequential in the context of the actual enhancement dependence on PM1 (Fig. 2). Related, it is not clear where the 28% on L19 comes from. What matters is the mass-weighted enhancement. Assuming this is a straight average over the points in Fig. 2, this is not relevant to the actual measurement of the enhancement, which is weighted. I suggest that the authors rethink this discussion entirely.

Related to the previous comment, it is unclear how the references on P9/L9 relate to the references on P9/L13. The authors appear to be linking these, I think, but it is not clear. Also, this seems selective, as there are studies (e.g. Liu et al. 2015) in which variability was observed. The authors should aim to provide a more comprehensive picture. Finally, it is not at all clear that the conditions in the cited studies are similar enough to those here to be relevant. This aspect needs to be discussed.

P9/L27: Should be Figure 4.

Figure 4: The site location should be clearly indicated. In addition, the boundaries of the in-region vs. out of region should be indicated clearly. Also, it is not clear whether these regions are defined based on some physical parameter (e.g. as air basins) or simply based on political boundaries. It would be useful if this were addressed.

P10/L3: It is unclear where the 62% number comes from.

P10/L14: It is unclear how Fig. 4c indicates that there were "higher aging rates." Can this be clarified?

P10/L15: "observed" should be "calculated." Also, is this a weighted average? A straight average across size? This needs to be clarified. A weighted average is most appropriate. This comment applies to everywhere in the manuscript that values for Eab or Dp/Dc are mentioned. What sort of averages are these? This needs to be clarified.

P10: I find the discussion with respect to O3 is somewhat lacking in detail and nuance. While O3 is lower during the polluted events, the concentration of precursors may be higher and this would contribute to aging. Additionally, photochemical processing is not the only possible pathway. Have the authors considered to what extent NO3 oxidation at night might be important?

P10/L24: The meaning of "taking more EEItotal more BC" is unclear.

P11/L17: Is the MAC range given here the increase over the baseline or the actual MAC range at 550 nm? I find this unclear. Also, is this mass weighted? The MAC varies with particle size.

P11/L19: Are the DRF values given related to the total BC? It is surprising that the increase is so small, given that the BC concentration itself increased by a factor of 7 or so. This could be clarified.

P12/L5 and P12/L8: It is not clear to me that the authors have demonstrated that there



is a "speeding up" or "acceleration" of the coating process in the more polluted air. In fact, they seem to be arguing that photochemical processing is slower, but that there is longer time. This would actually go against the idea that there is a speeding up. This should be revisited. Associated with this, it is not clear to me that Fig. 7 is necessarily correct. The EEI analysis indicates that the contribution from the regional sources is smaller during less polluted periods. This does not mean that those particles from regional sources are less coated just because the overall particle distribution has less coating during low pollution periods. In fact, it is possible that the regional particles are more coated due to higher photochemical activity (potentially). But, because their fractional contribution is smaller the net impact on the coating amount appears smaller in the average, which is now dominated by the local sources. I think that Fig. 7 and the discussion section need to be rethought a little bit to provide a more nuanced picture of what might be happening. It may be that the authors are correct, but I do not think that they have fully justified their conclusion here.

P12, conclusions: The authors should consider reporting mass-weighted averages of Dp/Dc and Eab in addition to the ranges to provide a fuller picture.

P13/L6: See previous comment regarding the reporting of single points without stated uncertainties. Is the 7.3%/h value believable? It is unclear, since it is a single outlier in the entire plot.

P13/L10: It is unclear how a 13-44% variation in the Eabs translates to a 28% underestimate in absorption. This appears to simply be an average of 13% and 44%, and not an appropriately mass-weighted average.

P13/L16: See previous comments about "speeding up". I do not think the authors have justified this conclusion.

P13/L23: It is unclear where the conclusion regarding heterogeneous chemistry comes from. This is pure speculation that is introduced at this point without justification. Why do the authors believe this to be the case? Also, this seems arbitrary. If the authors

had defined their periods differently then they could come to a different conclusion.

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