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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The Absorption Ångström Exponent of black carbon: from numerical aspects" *by* Chao Liu et al.

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Overall: First of all, we would like to thank the two anonymous reviewers and Dr. Corbin for their thoughtful review and valuable comments to the manuscript. In the revision, we have accommodated all the suggested changes into consideration and revised the manuscript accordingly. All changes are highlighted in RED in the revised manuscript.

This paper systematically tested the sensitivity of the AAE of BC in three representative morphology, and point out which factors should be considered when deriving AAE from possible available measurements. Though the calculation itself is not new, but the concept and focus is scientifically important. This paper is well organized and generally well written, but in this version it reads a bit too technical, so I would recommend final

publication after incorporating a bit more work, to allow this work within the scope of ACP.

Response: Thanks for the comments. We agree with the review that the technique may be not new, while the topic and focus of this study is really important. First, we would like to stress that all results were bulk properties averaged over a given particle size distribution. Because the simulations for the bulk properties are not well emphasized in the original version, it may be missed by the readers. Considering that some comments are related to the ensemble average and bulk properties, we improved the discussion to avoid misunderstanding. Meanwhile, all comments are constructive and important to improve the manuscript, and we followed the suggestions to incorporate our work.

Major points:

1) The most lack of this study is the authors have not calculated the AAE of BC in bulk but only for single BC particle. If I understand correctly, the authors have only given the BC lognormal size distribution, coating distribution as a guidance of size range selection for sensitivity test, however the single BC particle calculation has not been applied in the particle distribution to work out how these calculation will influence the whole. The information in bulk may be more valuable for the ambient measurement as most of the instruments measure in bulk.

Response: We totally agree with the reviewer that the bulk properties are more valuable for ambient measurements and downstream applications than that of a single particle, and, actually, we only discuss bulk properties in the work. However, the simulations for the bulk absorption is not well introduced in the original manuscript, and readers may easily miss the point. All results shown in this study are bulk properties averaged over a lognormal size distribution. We considered size distributions with geometric mean dimeters ranging from 50 nm to 200 nm, and a fixed geometric standard deviation of 1.5. To avoid similar misunderstanding, we emphasize the process of the corresponding simulations. (Line 15 of Page 1, Line 3 of Page 9, and Line 25 of Page

10)

2) The particle size as called GMD in this study is a bit confusing. For the coated size, I presume this is the size as entire BC particle, i.e. the coated particle, but if we compare everything all in GMD, would the coated BC has a less content of BC core? I'm not sure how comparable are they if in a same figure. Also, given the BC has complex morphology, what is GMD, is it supposed to be volume-equivalent diameter? This is important to be clarified.

Response: The GMD is one of the two parameters in the lognormal size distribution to determine BC size distribution, and the other parameter, GSD, is fixed to be 1.5. At this point, we want to clarify two points: (1). All GMDs are specified for the BC part to keep the BC amount consistent. Thus, the coated BC will have larger overall sizes than those of bare BC with the same GMD, but the amount and size distribution of BC component are consistent for a fare comparison. (2). For different BC particles with complex morphology, the size is defined by the diameter of equivalent volume sphere. Both statements are not well mentioned in the manuscript, and we have clarified this in the revision. (Line 17 of Page 8 and Line 4 of Page 9)

3) How the coating has been associated with BC core is not clearly presented, are they partly coated or embedded? How did you treat the coating interaction with BC? One recent study (Liu et al., 2017, DOI: 10.1038/ngeo2901) could be referenced in page 6 line 10 or page 10 line 28 etc. to support your discussion.

Response: As shown in Figure 1(c), the BC core is totally embedded by the spherical coating. We simply introduce a coating sphere at the mass center of the BC aggregates, and the details of this model can be found in Liu et al (JQSRT, 2017). The interaction between the BC and coating is rigorously considered by the MSTM method, which is also one of the advantages of the model. To be more specific, with the inhomogeneous particle shape of Coated BC determined, the MSTM can consider the absorption and scattering properties of the particular particle accurately. (Line 23 of

Page 7 and Line 32 of Page 10) Liu et al. (DOI: 10.1038/ngeo2901) present an excellent work on the influence of coating and aging on BC optical and radiative properties, which is highly related to our work. The results of the paper may greatly benefit our studies, and we will study their results to get a better representation on Coated BC particles in further studies. Liu et al.'s work has been cited in the revised manuscript to support our discussion in the revision. (Line 16 of Page 6 and Line 16 of Page 7)

4) The empirical equation (equation 6) is almost all about refractive index uncertainty, and they are separately discussed for three different morphology cases. Though the refractive index has large variation from different literatures, but mostly we are using a fixed refractive index or fixed spectral dependence of refractive index, otherwise there will be no real value for anything. However, the authors have not really given how the BC morphology has actually influenced AAE, such as Df value, the amount of coatings associated. These are most interested to communities who care about how the BC ageing will influence its mixing state/morphology and how the AAE will be modified by these factors.

Response: BC morphology shows the most complicated influence on BC AAE, which can be quite different for particles with different sizes or refractive indices. As we can see from Figure 6, the AAE for BC at different sizes decreases to a totally different degree as BC becomes compact (from Fresh to Compact BC). Meanwhile, the influence of coating on the AAE would be much more complicated considering the realist particle geometries in the ambient atmosphere. Thus, we take the geometry as an independent factor for Equation 6, and give the empirical equation for each particle geometry. To qualitatively understand the effects of morphology, the AAEs of BC in the three different formats can be easily estimated by our empirical equations if its size and refractive indices are known, and, then, the influence of morphology can be derived. This means that we only consider the influence of BC morphology at certain particle size and refractive index, and the effects of geometry can be qualitatively given by the differences between two empirical equations representing particles with different geometries. We add some discussions about the influence of BC morphology in the revision. (Line 24 of Page 1 and Line 26 of Page 13)

Others:

In page 6, the representation of coating thickness according to Schnaiter et al. (2005), could you point out which source of BC are they, and are they fresh BC, how long have they been aged?

Response: Thanks for the suggestion. The experiment given by Schnaiter et al. (2005) was carried out at a large aerosol chamber facility, and diesel soot particles were coated with secondary organic compounds produced by the in situ ozonolysis of -pinene. The particles are aged for 24 hours. We include additional informations in the revisions. (Line 10 of Page 7)

Page 6 line 30 to page 7 line 10, there are many parameter assumptions which have not been clearly explained: the 100 monomers are used, so are we actually only testing one BC core size? Have we tested the sensitivity to different monomer sizes (only 30nm is used here)? Liu et al., 2015 (DOI: 10.1002/2014GL062443) point out the AAE could be sensitive to the monomer size, also give the reference you choose 30nm.

Response: We considered the lognormal size distribution for BC particles, and clarified this point in the revision. Aggregates with 100 monomers are only an example to illustrate the particle geometries. As suggested by the reviewer, we have added the results with different monomer sizes in Figure 6. For Fresh BC with lacy structure, the monomer size doesn't change BC AAE significantly, whereas may decreases the AAE of Compact BC by approximately 0.1 as the monomer diameter increasing from 20 to 40 nm. Those discussions are also included in the revision. (Line 24 of Page 11)

For compact BC, the Df is used as 2.8 which is nearly sphere, any reference for this value? As above, it would be useful to test the sensitivity to Df.

Response: For compact BC, we considered the particles as compact as possible, and

thus a value of 2.8 is used in this study. To reveal the sensitivity of AAE to Df, and, thus, both a small (1.8) and a large (2.8) Df value are used in this study. Actually, there are not too many observations that give such a large Df, whereas some electronic microscopic images of BC show really compact structure. All other parameters are chosen based on observational data, and we had added the corresponding references. To better illustrates the sensitivity, we include one more curve in Figure 6 for results of aggregates with a Df of 2.3. For better understanding on the influence, the references for the Df values are included, and the sensitivity to Df is also discussed. (Line 3 of Page 12)

Page 7 line 8-16: the whole discussion here is rather confusing, you should point out what size previous instruments actually measured, the coated particle size or only BC core size, currently they are mixed up. You should point out SMPS measured mobility diameter is very sensitive to the particle shape (which is different from the volume equivalent diameter you present here), but references you referred Reddington et al., 2013; Wang et al., 2015 used the BC core size, measured by the single particle soot photometer.

Response: The discussion related to BC size is completely reorganized, and, with additional discussions, it should be easier to understand right now. (Line 9 of Page 8)

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