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

**Chao Liu et al.**

chao_liu@nuist.edu.cn

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Overall: First of all, we would like to thank Dr. Corbin and the two anonymous reviewers 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 the revised manuscript in RED in the revision.

The manuscript "The absorption Angstroem exponent of black carbon from numerical aspects" by Liu, Chung and Yin (ACPD 2017, doi:10.5194/acp-2017-836) is on a very interesting topic. The authors present numerical simulations of BC properties for 3 test cases of morphology using computer-modelled soot aggregates.



Response: Thanks Dr. Corbin for the constructive comments on the manuscript, and we revised the manuscript following all the suggestions

The value of these results is clear; the effects of variations in BC microphysical properties on the AAE are clearly seen. However, the limitations of the results could be discussed in much more depth. I have several comments related to this:

1. The words "diameter" and "size" are used in multiple ways throughout the study. Especially when discussing the results of Schnaiter et al., with respect to coating thicknesses of BC, the word diameter is poorly defined as it would refer only to the apparent mobility of the particles (increased by coatings but decreased by restructuring).

Response: During the revision, we improved the clarity of the terms "diameter" and "size" by adding exact definition for diameter, and detailing and improving the discussion about the coating thickness. The definition for BC size can be quite different considering the different principles for size measurements. In this study, we try to keep the discussion simple, and both diameter and size refer to the diameter of equivalent volume sphere for consistency. We clarified this definition in the revision, and, in the section for the coating thickness, we illustrate the definition of three quantities in the figure and specify each of them clearly in the discussion, which will help readers to better understand the discussion. In this revised version, there will be little chance for misunderstanding the definition of diameter we used. (Line 16 of Page 8, Line 30 of Page 6, and Figure 2)

2. The use of a change in mobility diameter (Schaiter et al.) to infer size-dependent coating thicknesses is invalid, as noted above. As a reviewer noted, Liu et al. (Nat. Geosci 2017) should be referenced here. In any case, I am not sure of the value of this size-dependent coating thickness. Since all of the results were plotted against diameter, there is no possibility that the reader misunderstands that coating thickness depends on particle size and history. A constant volume fraction of coatings would allow a fair evaluation of the size dependence to be made. Currently the x axis of

the figures (labelled GMD) reflects changes in both size and coating thickness, and is therefore difficult to interpret.

Response: First, our paper discusses the relationship between the coating thickness and the BC core size. To be clear, the paper does not discuss how this relationship changes over time as the coating progresses. What is discussed in the paper is a snapshot relationship after 24 hours of coating. Such a snapshot relationship (whether after 24 hours or after 48 hours) is what we need for our study.

Many papers do provide the size distribution of coated particles, but Schnaiter et al. (2005) provide the size distribution of BC cores as well. In reality, Schnaiter et al. (2005) provide the size distribution of coated particles and that of fresh BC. Since a fresh BC particle becomes a BC core after coating, both the fresh BC and the BC core have the same diameter of the corresponding equivalent volume. Schnaiter et al. (2005) actually employed an SMPS (Scanning Mobility Particle Sizer) to measure the particle sizes, and SMPS gives mobility diameter. Mobility diameter is very close to the diameter of the corresponding equivalent volume for spherical particles (and thickly coated BC are nearly spherical) but mobility diameter may deviate from the diameter of the corresponding equivalent volume for fresh BC. However, without information for particle shape and density, it is difficultly get the relationship between the two diameters, and we made this assumption in the study. (Line 30 of Page 6)

A constant volume fraction of coating assumes that small BC cores have thin coating and large BC cores have thick coating, but there is little backing evidence for this assumption. Originally, we thought of a constant volume fraction of coatings too, but later realized that this assumption may be wrong. The relationship between coating thickness and core size is developed in our paper with two ideas: (1) We try to derive and use a more realistic relationship between coating thickness and core size that has an observational basis. Although the derived relationship in our paper is based only one experimental study (i.e., Schnaiter et al. 2005) and used mobility diameter data, it is the first derived relationship in the community that has any observational support. (2).



As we have emphasized in the manuscript, we understand the results for Coated BC as a special case, and much more work should be done to give a more general conclusion considering the complex properties of Coated BC. Thus, we prefer to keep using the coating thickness we obtained, but better clarify the discussion in the revision.

Liu et al. (2017) presented a state-of-art study on the BC absorption enhancement due to particle mixing. Their work can definitely improve our understanding on the relationship between BC core size and coating, and we would try to obtained such relationship for sensitive studies in the future. (Line 16 of Page 6 and Line 16 of Page 7)

3. The number of primary particles in an aggregate was mentioned as hundreds or even thousands, but what is the most likely number?

Response: We consider BC aggregates with different monomer number, and only the bulk properties averaged over given size distributions are discussed. As we have demonstrated in the manuscript, we consider aggregates with monomer number from 1 to 2000. To know the most likely number, we can give an example here. With monomer diameter defined as 30 nm, an aggregate with approximately 300 monomers corresponds to an equivalent volume diameter of 0.2 m. The most likely number could vary from case to case, and, thus, we consider a relatively wide range of size distribution for sensitivity study. We have also give more detailed relationship between monomer number and equivalent-volume diameter in the revision. (Line 32 of Page 8)

4. (page 6) The authors might cite the previous studies which have modelled coatings on complex BC morphologies, e.g. Liu et al. 2016 (doi:10.1016/j.jqsrt.2015.08.005) and references therein. As the authors noted, however, no previous study discussed the AAE.

Response: Yes, we added some discussion about this point in the revision at Line 29-34, Page 6, and Liu et al. (2016) as well as Dong et al. (2015) and Liu et al. (2012) is cited now. (Line 8 of Page 6)

5. To what degree might the results change with different choices of monomer diameter and monomer number?

Response: In the revision, we include the results for particles with different monomer diameter. As shown in the updated Figure 6, the AAE increases as the monomer diameter deceases. The monomer diameter is not a very big problem for Fresh BC, but becomes important for Compact BC. Meanwhile, the variation over monomer number, i.e. particle overall size, can be understood by that over the GMD in this study. With the increases of monomer number, the GMD increases, and the AAE decreases. (Figure 6 and Line 24 of Page 11)

6. On page 11, lines 10-14 do not follow from the previous discussion. The authors are saying that the BC RI may change between sources, so that the AAE may not be fixed. But it has been assumed that the imaginary and real RIs are free to vary independently, which is not a justified assumption (Bond et al. 2006; Moteki et al. JAS 2010, doi:10.1016/j.jaerosci.2010.02.013). This assumption would have led to overestimated variability in AAE. Also, this assumption was necessary to define A and B as independent in equations 4 and 5.

Response: The discussion is unappropriated here in the discussion, and we omitted them in this paragraph. Although the real and imaginary parts cannot vary independently, and this study defines A and B independently to better understand the effects of each parameter on BC AAE. For realistic applications, A and B can be obtained based on the given wavelength-dependent refractive indices. (Line 20 of Page 10)

Also I have a couple of minor comments which were not noted by the reviewers:

âĂć Palas soot (page 3) should be referred to as spark-generated carbon nanoparticles.

Response: We modified the text following the suggestion. (Line 12 of Page 3)

âĂć At page 4 line 20, Moosmüller et al. (2011, doi:10.5194/acp-11-1217-2011) should



be cited.

Response: This is a really great paper to support our work, and we cited it in the revision. (Line 4 of Page 4)

âĂć In Section 2.2, please rewrite the discussion. It currently sounds like you are saying that an SMPS measures the diameter of volume equivalent spheres.

Response: SMPS measures the mobility diameter of the particle, and we clarified this point in the revision. (Line 9 of Page 8)

âĂć On page 10, I don't understand why a linear regression (I assume of log- transformed data?) should give the best representation of the AAE if the relationship is not exponential. A linear regression of log-transformed data is not reliable even when the data follow a power-law relationship (Clauset et al., 2009, doi:10.1137/070710111)

Response: The linear regression is applied for the log-transformed data, i.e., for the linear relationship shown in Figure 5. Considering the highly agreement between the simulated data and the fitting, the method to get the fitted data doesn't influence the results too much. We have clarified that the data is obtained by linear regression of the log-transformed data. (Line 7 of Page 11)

Finally, the authors have proposed that their numerical results are a suitable absolute reference for BC properties. This is clearly not the case, as a closure between experiment and theory is still lacking (Bond et al., 2006; Radney et al., 2014). Radney et al. (2014, doi:10.1021/es4041804) is particularly relevant to this study because they have used the same T-matrix approach in combination with direct measurements of uncoated BC, yet were unable to satisfactorily model particle extinction. This shows that T-matrix models of BC aggregates should not be used to make strong conclusions on BC properties. Rather than interpret these numerical results as a comprehensive guide and absolute reference, the authors could interpret these results as a clear demonstration of the importance of various parameters on the optical properties of BC. As noted above, I found these results to be quite interesting, because they demonstrate quantitative the theoretical impacts of changes in various properties on BC in a way that has not been done before.

Response: Thanks for the suggestion, and we agree with Dr. Corbin on the limitation of the numerical models and the suggested interpretation. Although the results can hardly be understood as an absolute reference for BC AAE, we think it is fine enough to illustrate the response of BC AAE to BC size, refractive index, and geometry, at least qualitatively. Thus, we have incorporated these into the revised version, and discussed the corresponding studies. (Line 21 of Page 15)

References

All references were either made in the submitted manuscript or were cited using doi's. These doi's provide direct links to the articles via http://dx.doi.org

Response: Thanks again for showing us those important references, and most of them are helpful for our work and discussed in the revision.

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