

Responses to Reviewers (ACP Manuscript # ACP-2017-836)

First of all, we would like to thank Dr. Corbin for his re-review on the manuscript. In the revision, we followed both major suggestions and significantly improved the manuscript, especially for the derivative related to coating amount. All changes are highlighted in RED. Meanwhile, in this point-to-point response, the reviewers' comments are copied as texts in BLACK, and our responses are followed in BLUE.

The Editor has requested my feedback on whether the revised manuscript sufficiently addressed my comments. Unfortunately, I cannot answer this question in the affirmative. I would define "sufficiently addressed" as either (i) disagreeing with a comment and providing an evidence-based argument for such disagreement, or (ii) agreeing with a comment and providing a modified manuscript which remains self-consistent. In the two most substantial cases, it appears that the authors have agreed with my comments, but not modified the manuscript in response. Therefore, I believe that major revisions to the revised manuscript would be required for it to send a self-consistent and unambiguous message.

Of my six comments, three addressed a lack of literature context (#3, 4, 5), one (#1) addressed the inaccuracy with which the manuscript uses technical language, and two (#2, #6) addressed the degree to which the manuscript contributes to scientific progress. The latter two (#2 and #6) are the ones I consider most substantial, although I do not see that #1 was addressed sufficiently either.

Regarding #2, my comment related to the fact that the authors have based their calculations on an invalid reinterpretation of literature results (the assumption that a change in mobility diameter can be used to infer coating thicknesses). In my original comment I provided a solution for the authors to avoid this problem without much more work. The modified manuscript has not changed this assumption, although the authors appear to agree with my comment (there is a new statement that "the conclusions must be viewed with some caution" on page 7). This makes the manuscript appear non-self-consistent.

Response: We agree with the reviewer that the assumption of using mobility diameter for fresh BC is problematic. To avoid such error, we made a more rigorous calculation to translate the mobility diameter into the equivalent volume diameter for both BC core and

Coated BC, and, then, the observation and our numerical model can be connected more accurately. To be more specific, this is done in the following ways:

- (1). For Coated BC, we assume the mobility diameter to be a reasonable representation of equivalent volume diameter, because they are compact and nearly spherical.
- (2). For Fresh BC, instead of directly using the mobility diameter from the SMPS, we first transfer the mobility diameter into equivalent volume diameter (Fig. 2b) following the relationship between mobility diameter and aggregate parameters given by Naumann (2003) and the diesel soot geometric parameters given by Schnaiter et al. (2003; 2005). In this way, we significantly improve the accuracy for the description of the Fresh BC (i.e. BC core sizes) size distribution.

With the simplifying assumptions that are much more reasonable, we can get the volume fraction of coating as a function of equivalent volume diameter of BC core, which is what we need for our study and not conflict with our aggregate models. As shown in Figure 2d, the volume fraction of coating decreases as BC core size increases. This coating amount variation we obtained agrees with the trend obtained by Fierce et al. (2016). This also indicates that our coating may be not quantitatively perfect, but is qualitatively right, at least in the right direction.

To further address this concern on the coating amount distribution, we perform additional simulations for Coated BC with fixed coating thickness and fixed coating volume fraction for comparison. The following figure illustrates absorption enhancement due to coating with different coating assumptions (i.e., the one we derived, fixed coating fraction, and fixed coating thickness (based on equivalent volume sphere)). For all three cases, the total coating amount for the ensemble of different-sized BC particles is the same, whereas the coating distribution is different for BC cores with different core sizes. We can see that three cases lead to similar absorption enhancement, so we conclude that our results based on the derived coating amount should be similar with those based on fixed coating fraction. However, our results are supported by the observations.

To conclude, by mainly changing the mobility diameter into equivalent volume diameter for Fresh BC, we have significantly improved the discussion about the coating derivation (see entire Page 7 of the revision), and reconstructed the Figure 2 with more details. The coating amount given should be much improved and reasonable.

Furthermore, we would really like to continue work on this task based on Liu et al. (Natrue Geoscineces, 2017) to derive more reasonable and generally coating thickness dependence. Meanwhile, we have demonstrated in the text that the results should be carefully understood as those from a special case.

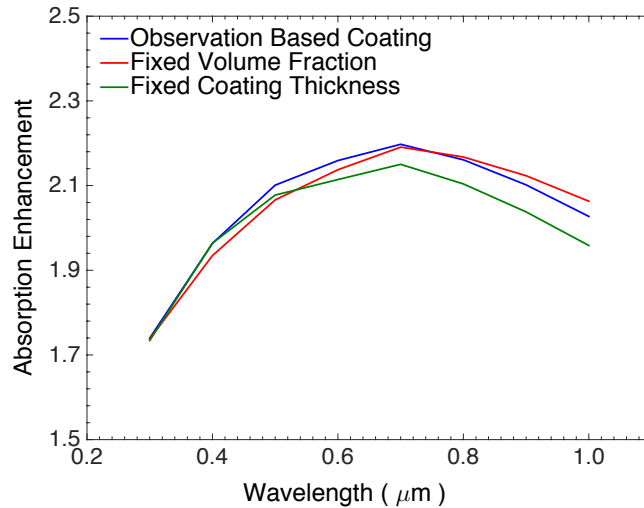


Figure 1. Absorption enhancement due to non-absorption coating with different coating assumptions, and the total The lognormal size distribution with a GMD of 0.12 micron and a GSD of 1.5 is assumed for the BC core.

Regarding #6, my comment related to the treatment of the real and imaginary parts of the complex RI as independent being inconsistent with literature. The authors appear to agree (according to their adding a similar statement on page 10), but have not altered the analysis as a result. Again, this makes the manuscript appear not to be self-consistent. Reading between the lines, I believe that the authors intended for their calculations to demonstrate the impact of uncertainty in the RI values. This I would understand, but according to the current revision I cannot be sure that this is what the authors intended. If it is, then it should be clearly stated and the entire manuscript adjusted accordingly.

Response: Yes, we agree with the reviewer that the wavelength variation on the real and imaginary parts of refractive indices (i.e. parameters A and B in the manuscript) should be dependent, and we add extra discussions to stress this point to keep the manuscript consistent. In the revision, we kept Figure 8 to illustrate the uncertainties related RI wavelength dependences, whereas the quantitative discussion on A and B based on Equation (6) is removed. Because independent A and B may overestimate the variability of BC AAE, Equation (6) is simplified to consider only wavelength-independent refractive index. The modifications are made at Lines 5-9 of Page 11, Lines 13-14 of Page 14, and Line 33 of Page 14 – Line 2 of Page 15, and we also remove some statements that may confuse the readers as suggested by Dr. Corbin.

Because the reviewer thought that some other comments from the original review are not addressed sufficiently, the following lists the original comments #1 and #4 as well as the response.

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: The section related to the coating amount is corrected and totally rewritten, and we use the “diameter” and “size” in the revision really carefully. For coating, we consider only coating amount in this revision, and there will be much less confusing statements.

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: The reference is added, and we further specify the uniqueness of our study in the revision.

Last but not the least, we thank Dr. Corbin again for the constructive comments, and both suggestions have significantly improved our paper.