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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The absorption Angstrom exponent of black carbon with brown coatings: effects of aerosol microphysics and parameterization" *by* Xiaolin Zhang et al.

Anonymous Referee #2

Received and published: 1 May 2020

The paper describes a numerical study of the Aerosol Absorption Angstrom Exponent (AAE) for aged BC particles. The authors use the multi-sphere T-Matrix method to calculate the optical properties of coated BC particles. One of the "surprising" findings of the study is that, in some circumstances, BC coated by brown carbon exhibits AAE lower than even "pure" BC (I've put quotations because probably there is no such thing as pure BC, apart from a modeling perspective). I think the work is interesting and adds important results useful to the community. Therefore I think the work is worth publishing after the following comments are carefully addressed.

General comments

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- The English language should be improved significantly before the manuscript can be published. I would encourage the authors to have a native speaker read over and edit the paper to improve readability. As it is now, grammar and sentence construction issues seriously hamper the readability and therefore the understandability of the paper.

- I found it difficult to clearly understand the different parameters defined in the paper, especially F until much later in the paper. I think it would help a lot to provide the value of F, f, Dp/Dc, Df, etc. and not just the coated volume fractions in Figure 1 and to clearly define these parameters at the very beginning.

- Refractive index: please provide the values used for each wavelength not just references to the literature, maybe provide a table (or a graph) with all the values used (most importantly obviously for BrC.

- It would be interesting to have some sort of physical explanation (or tentative interpretation) for why the Mie calculations result in generally lower AAE.

- The strong dependence of AAE on the shell/core ratio seems quite reasonable because the AAE increases with the increased amount of absorption ascribable to coating, which has a high AAE in the first place, vs. “pure” BC. Less intuitive, but also quite interesting, is maybe the dependence on F.

- For some of the plots, it would be interesting to provide bands instead of point to account of slight variations of different parameters as in a sensitivity study, but I understand that might require a substantial amount of additional work which might not be doable at the time.

- Is there a rationale behind choosing a power laws model vs. a polynomial or any other type of fits for equation 9? I mean, did the authors consider other potential models, or did they pick this one for a specific reason? Also, please provide the fitting parameters’ confidence (e.g., 95%) ranges. More on this later (in the specific comments)

- Related to the previous comment, the proposed parametrization does a decent job in the middle of the ranges of f and Dp/Dc, but not so well at all at the extreme values. Although the authors mention that in passing, I think this is an important caveat to point out very clearly in the paper, including in the abstract so that future research will use caution in applying the model for cases it might not be applicable to (for example for F=0, Dp/Dc higher than 2.5 and f near zero, the parametrization-numerical simulation difference in AAE is about 1, which is a very large discrepancy, and 0f 0.5 at the other extreme of Dp/Dc)

Specific comments

Lines 14-16, page 1. The sentence describes an important finding, but I think it is a bit confusing. The reader might ask if the AAE<1 is for BC thinly coated by BC, or BC thickly coated by some other material, or BC coated by a large amount of BrC, or BC coated by a thin layer of BrC and then further coated by a large amount of other material. I would suggest clarifying the sentence.

Line 18, page 1: By “trivial” do the authors mean negligible?

Line 19, page 1: “more small coated BC. . .” and “more brown carbon. . .” the comparative “more” should always be accompanied by a clear indication of what we should compare with. In other words, “more” than what or with respect to what? Also "more small" should be "smaller"

Line 20, page 1: “. . .shows weakly sensitive. . .” consider rephrasing. Maybe “shows weekly sensitivity. . .” or “appears to be weakly sensitive. . .” or similar.

Lines 12-13, page 2: “. . .AAE is considered to be aerosols originating. . .” consider revising the wording, this makes it appear as if AAE is an aerosol, while it is the property of the aerosol.

Line 9, page 3: “This limits its applications. . .” what does “its” refer to?

Lines 6 and 7, page 4: the definition of F is not very clear to me. What does “BC

monomers within coating" mean?

Line 11, page 5: I would not say that "absorption universally decreases exponentially". The power law is a useful practical tool, an approximation, but I would definitely not say that it is a universal law for the wavelength dependence of absorption.

Line 20, page 5: The sentence is not clear.

Line 28, page 5: "the bias induced by chosen absorptions at two wavelengths may be averted". This sentence is not clear. What bias? How is "averted"?

Lines 1 and 2, page 6: I don't understand the sentence "Since the AAE of coated BC is acquired, systematic studies of the impacts of brown coating on the AAE of BC particles follow".

Line 7, page 6: what does "averagely" mean in this context?

Line 18, page 6: "...with the augment of Dp/Dc from 1.9 to 2.7, the AAE alters in the range of 1.5–2" awkward wording, consider revising. What is the "argument of Dp/Dc", what does it mean "AAE alters..."

Lines 9 and 10, page 6: "...an outmost off-center core-shell and concentric core-shell..." is not completely clear to me what the authors refer to. Maybe a drawing similar to Figure 1 or a direct reference to the existing figure 1 (if relevant) would help to understand what exactly is the configuration considered.

Lines 4 to 6, page 7: I think this is an important finding that is worth highlighting (e.g., in the abstract).

Section 3.2, page 7: (a) Does the size distribution refer to the BC component or to the entire mixed particle (BC plus BrC size)? (b) Is the dependence on size distribution evaluated only for the high fractal dimension case? Did the authors also look at the dependence for low fractal dimension? It would be interesting to see the results. (c) Also, did the authors explore potential dependencies on the width of the distribution

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(sigma g)?

Lines 9, 10, page 7: The definition of F is provided more clearly here than initially. This definition should be provided much earlier on in the paper.

Line 23, page 7: I would not consider this to be a “contamination”

Lines 25 to the end of page 7: f is finally defined here. I think a reference to its meaning earlier on would help the readability of the paper.

Line 4, page 9: “shows weakly sensitive. . .” maybe should be “show weak sensitivity” or “is weakly sensitive”

Line 10, page 23” “remove “in” from “This is generally in consistent with the findings. . .”

Line 21, page 9: I suggest put the defined parameters in parenthesis to assure a clear understanding of what is what even if previously defined already. Such as in: “the absorbing volume fraction of coating (f), coated volume fraction of BC (F), and shell/core ratio (Dp/Dc)”

Line 22, page 9: “. . .whereas the size distribution is considered independently (i.e., to be fixed).” This is not clear to me.

Line 25, page 9: Maybe “power laws” is more appropriate than “exponential”.

Lines 2-4, page 10: This finding and explanation are confusing to me.

Lines 4 to 5, page 10: “The influences of particle microphysics on the AAE of coated BC are obviously confirmed by corresponding coefficients in Equation 5 (10).” I am not sure I understand this sentence. Do the authors mean that the coefficients are large and therefore the dependence is strong, or something else? I guess that becomes clearer in the following sentences.

Line 8, page 10: “. . .the capability of the expresses. . .” what does that mean?

Line 12, page 10: “dominated” maybe should be “dominant”? Also, the fully coated

morphologies might be dominant in many circumstances such as biomass burning plumes, but not always, for example not always in urban environments.

Lines 24-25, page 10: “Although the volume of BrC seems to be responsible for the large AAE of coated BC, more BC encapsulated in brown coating or more large coated BC particles reduce this effect.” This seems reasonable, what matters more is the volume ratio because that is the determinant variable that splits between the absorption being dominated by BC with low wavelength dependence (low AAE) and the absorption due to the coating (with high AAE for BrC coating). More counter-intuitive, but also interesting seems to be the following sentence; is there any hypothesis on why that might be (meaning why the AAE might be significantly lower than 1 for thin BrC coatings)?

Line 30, page 10: “might be made. . .” or “might not be made. . .”. Same in the conclusion section.

Line 31, page 10: “which is a replenishment of related findings” consider rewording, the use of “replenishment” here does not seems to be the most appropriate.

Figure 5-7: How does f differ (or how is related to) Dp/Dc?

Figure 7: That is an interesting comparison. It seems like the model does well for intermediate values of f and Dp/Dc values. The model does less well at the extremely lower or higher values of f or Dp/Dc. This might suggest a bias in the model that tends to fit better the center but less well the tails. That might also be due to the power-law fit choice, so, as mentioned in the general comments, it could be good to also explore other parametrizations (such as a polynomial or even just a simple multiple variable linear regression or so) to understand if the power fit is truly justified and appropriate, or if a different model would perform better.

Table 1: Re-define what the different parameters are in the caption so the reader does not have to search for the definitions in the text. F, Dp, Dc, f, etc.

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