Anonymous Referee #2

First of all, we would like to thank the anonymous reviewer for his/her 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 revision. In this point-to-point response, the reviewer's comments are copied as texts in BLACK, and our responses are followed in BLUE.

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.

Response: Thanks for the constructive comments. The comments are significantly helpful to improve the manuscript, and make the paper more solid. The following presents our point-to-point responses as well as the revision for the manuscript.

General comments

- 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.

Response: Thanks for the constructive comment, and the manuscript has been polished by an English editor.

- 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.

Response: We have modified accordingly and defined these parameters at the beginning of the Methodology as:

"It is observed that BC particles can externally attached to, partially coated in, or fully encapsulated in coatings [China et al., 2013, 2015]. This study considers BC aggregate core with a spherical coating, following the coated BC models built by Zhang et al. [2018], and the sketch maps of three typical coated BC structures considered (i.e., externally attached, partially coated and fully coated) are portrayed in Figure 1. For coated BC, the coated volume fraction of BC (F) is a crucial microphysical parameter characterizing its mixing state, and it follows

$$F = \frac{V_{BC\ inside}}{V_{BC}}, \quad (3)$$

where $V_{BC\ inside}$ and V_{BC} are the volume of BC monomers encapsulated in coating and overall BC volume, respectively (see Fig. 1). With the definition, the externally attached, partially coated, and fully coated BC aggregates show F=0, 0<F<1, and F=1, respectively. For coated BC, shell/core ratio Dp/Dc is an important microphysical parameter and is defined as spherical equivalent particle diameter divided by BC core diameter (Dc).

The fractal dimension D_f is a parameter describing the compactness of BC aggregates, and due to aging process in ambient air, BC can be coated by other species, such as organics, becoming compact (i.e., large D_f) [e.g., Coz and Leck, 2011; Tritscher et al., 2011].”

- 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.

Response: We have provided a graph for BrC refractive index accordingly in Fig. S1 as:

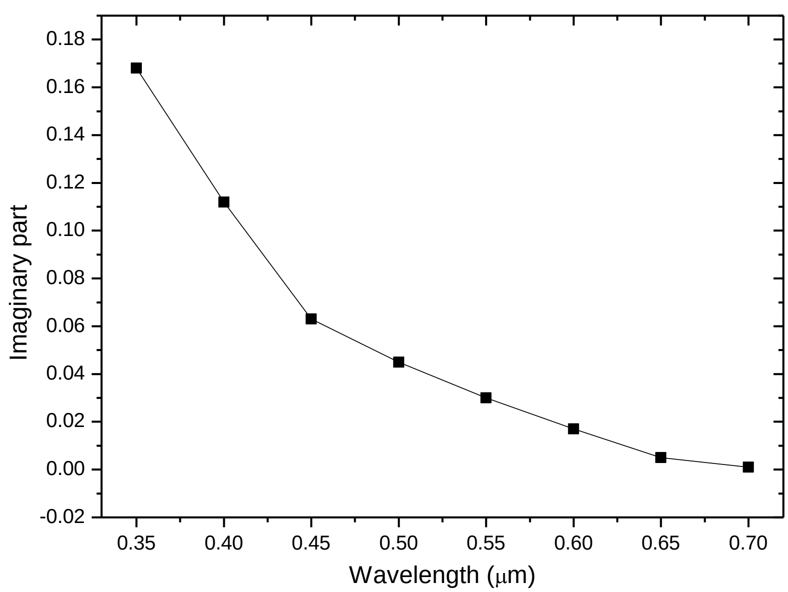

Fig. S1. Imaginary part of refractive index of brown carbon as a function of wavelength based on Kirchstetter et al. [2004].

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

Response: Thanks for the constructive suggestion, and we have tried to give some tentative interpretations in the revision as:

“This is probably due to that the absorption of BC coated by BrC with core-shell Mie model show slower increase with decreased wavelength than that of coated BC with realistic particle geometry.”

- 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.
Response: Thanks for the reviewer's constructive comments.

- 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.
Response: Thanks for the constructive comments, and we will do it in our further work in the future as it requires 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)
Response: Thanks for the constructive comments from the reviewer. The parameterization of the AAE of coated BC is challenging and difficult, as there are three microphysical parameters (i.e., *F*, Dp/Dc and *f*) for fitting (Common fitting is only for one parameter). The rationale behind choosing a fit type may be that we first have to select a model to well fit one parameter and then guess the possible model for fitting three parameters. We have tried the polynomial fit and multiple variable linear regression fit, and both are failed (The polynomial fit even cannot converge, while linear regression fit show very low correlation coefficient R^2). Therefore, the power law model shown already is the best one at present.
For the fitting parameters' confidence ranges, we have added accordingly in the revision as:
"the coefficients (with 95% confidence range) can be fitted and the AAE of coated BC is given by".

- 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)
Response: Thanks for the constructive comments. We have pointed it out in the Abstract as:
"The proposed parameterization of coated BC AAE does a decent prediction for moderate BC microphysics, whereas caution should be taken in applying it for extreme cases, such as externally attached coated BC morphology."

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.

Response: We have modified it accordingly as:
"more large BC particles coated by thin brown carbon can have an AAE smaller than 1.0".

Line 18, page 1: By "trivial" do the authors mean negligible?
Response: We have modified "trivial" to "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"
Response: We have revised it accordingly as:
"if there are plenty of small coated BC particles, heavy coating, or a large amount of brown carbon".

Line 20, page 1: ": : :shows weakly sensitive: : :" consider rephrasing. Maybe "shows weekly sensitivity: : :" or "appears to be weakly sensitive: : :" or similar.
Response: We have revised "shows weakly sensitive" to "appears to be weakly sensitive".

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.
Response: We have revised it in the revision as:
"Therefore, in ambient measurements, large AAE is considered to be that aerosols originate from dust or biofuel/biomass burning, while small AAE near 1.0 is understood to be that aerosols are BC-rich particles due to the burning of fossil fuel [Russell et al., 2010]."

Line 9, page 3: "This limits its applications: : :" what does "its" refer to?
Response: We have revised "its" to "the AAE".

Lines 6 and 7, page 4: the definition of F is not very clear to me. What does "BC monomers within coating" mean?
Response: We have changed "BC monomers within coating" to "BC monomers encapsulated in coating" and added "see Fig. 1" to make it clear.

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.
Response: We have deleted "universally".

Line 20, page 5: The sentence is not clear.
Response: We have modified it in the revision as:
"Nonetheless, the AAE obtained from Eq. (7) is rather sensitive to observational wavelengths selected, and notable distinct AAE values can be obtained for different wavelength ranges [Moosmuller and Chakrabarty, 2011]."

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"?

Response: We have modified it accordingly following:

"the AAE bias induced by wavelength selection may be averted by this fitting method".

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".

Response: We have deleted this sentence as it is only for a smooth transition.

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

Response: We have deleted "averagely".

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: : :"

Response: We have revised it following:

"When BC aggregates are fully coated by BrC, with the increase of Dp/Dc from 1.9 to 2.7, the AAE varies in a range of 1.5–2.6."

Lines 9 and 10, page 6: ": : :an outmost off-center core-shell and concentric coreshell : : :" 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.

Response: We have added a direct reference (i.e., *[Zhang et al., 2019])* to 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).

Response: We have highlighted it in the Abstract

"The currently popular core-shell Mie model reasonably approximates the AAE of fully coated BC by brown carbon, whereas it underestimates the AAE of partially coated or externally attached BC, and underestimates more for smaller coated volume fraction of BC."

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 (sigma g)?

Response: Thanks for the reviewer's constructive comments.

(a) The size distribution refers to the entire mixed particle, and we have modified it to make it clear as: *"The lognormal size distributions for coated BC with r_g (x axis) in the range of 0.05–0.15 μm and σ_g assumed as the aforementioned 1.59 are considered."*

(b) We have added the AAE dependence on size distribution for low BC fractal dimension, and the comparisons of the AAE between low and high BC fractal dimension are shown in Fig. S2. The differences of the AAE of BC with brown coating induce by BC fractal dimension are generally trivial.

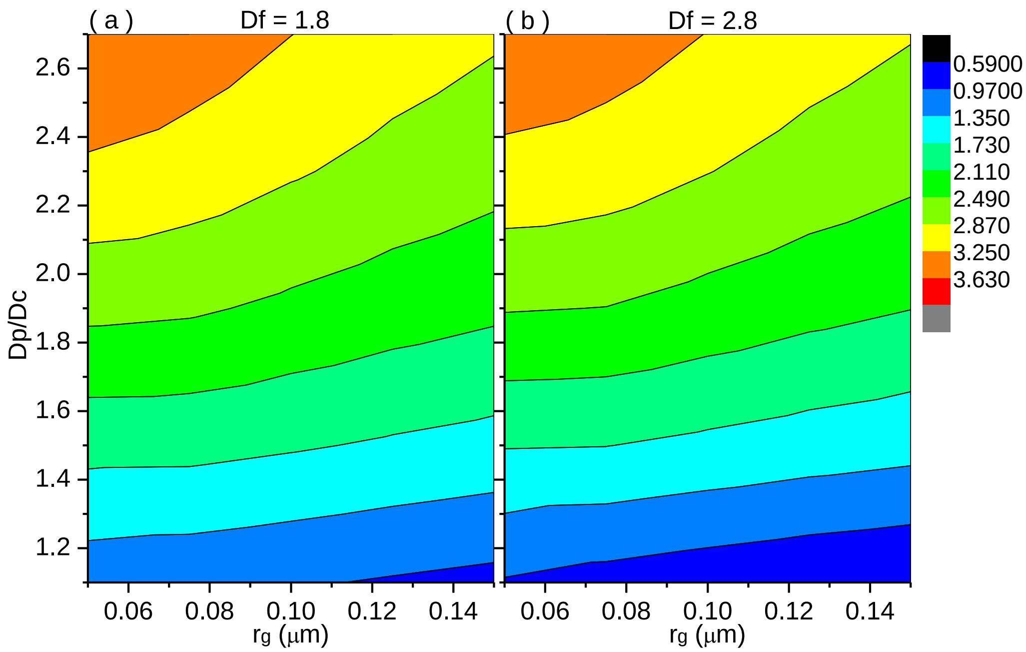

Fig. S2. The absorption Angstrom exponent (AAE) of BC aggregates coated by brown carbon with different shell/core ratio (Dp/Dc) and particle size distribution. BC fractal dimension of 1.8 and 2.8 are shown from left to right. Coated volume fraction of BC is 0.0, and geometric standard deviations (σg) for applied lognormal distribution are 1.59.

(c) We have explored the AAE dependence on the width of particle size distribution, which is shown in Fig. S3. As aerosol-climate models generally consider particle size distributions with fixed width (i.e., σ_g) but varying radius (i.e., r_g), we show the AAE dependence on σ_g in Fig. S3. The AAE of BC with brown coating generally decrease with increased width of size distribution, except for externally attached BC-BrC with small width of size distribution (i.e., $F = 0.0$ and $\sigma_g < 1.39$).

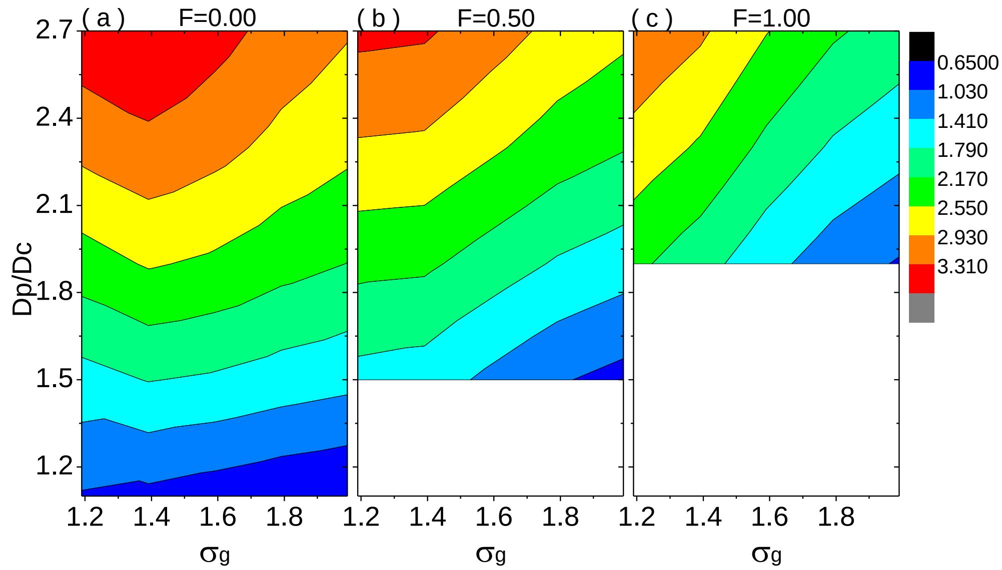

Fig. S3. The absorption Angstrom exponent (AAE) of BC aggregates (BC fractal dimension of ~2.8) coated by brown carbon with different shell/core ratio (Dp/Dc) and particle size distribution. Three coated volume fractions of BC, being 0.00, 0.50, and 1.00, are shown from left to right. For fully coated BC structure, BC is located at the particle geometric center. The geometric mean radii (rg) for applied lognormal distribution are 0.075 μm.

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.

Response: We have provided it earlier in the revision as:

"*With the definition, the externally attached, partially coated, and fully coated BC aggregates show F=0, 0<F<1, and F=1, respectively.*".

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

Response: We have revised it accordingly as:

"The above simulations assume BC coated by BrC, whereas non-absorptive organic carbon can also exist in BC coatings in ambient air."

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.

Response: We thank for the reviewer's constructive comment, whereas the absorbing volume fraction of coating (*f*) is defined by us. It may be the first time (to our knowledge) to define *f* here, and we are sorry that there is no reference for defining it earlier in the manuscript.

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

Response: We have modified "shows weakly sensitive" to "shows weak sensitivity" accordingly.

Line 10, page 9" "remove "in" from "This is generally in consistent with the findings: : :"

Response: We have deleted "in" accordingly.

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)"

Response: We have revised it accordingly following:

"Thus, to make the parameterization doable, the absorbing volume fraction of coating (f), coated volume fraction of BC (F), and shell/core ratio (Dp/Dc) are used for the AAE parameterization".

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

Response: We have revised it to make it clear as:

"whereas the size distribution is not considered (i.e., to be fixed)".

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

Response: We have revised "exponential" to "power law" accordingly.

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

Response: We have revised it to make it clear in the revision following:

"The correlation coefficient for parameterizing with three variables (i.e., f, Dp/Dc, and F) is mildly smaller than that with one variable (i.e., each of f, Dp/Dc, and F), and this is possibly associated with the lack of considering the combined interaction effects of f, Dp/Dc, and F on the AAE in the parameterization."

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.

Response: It means that the coefficients are large and therefore the dependence is strong. Meanwhile, The Equation can be treated as a quantitative understanding of the influences of particle microphysics on the AAE of coated BC. We have revised it to make it clear in the revision as:

"The influences of particle microphysics on the AAE of coated BC are obviously confirmed by corresponding coefficients in Equation (10) with a quantitative understanding."

Line 8, page 10: ": : :the capability of the expresses: : :" what does that mean?

Response: We have revised it to make it clear in the revision as:

"*To confirm the capability of this parameterization in approximating the AAE of coated BC*".

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.

Response: We have revised it accordingly in the revision as:
"considering that the partly and fully coated morphologies are dominant in aged BC based on observations".

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)?

Response: Thanks for the constructive comments from the reviewer. The hypothesis on why that might be is the one we do not know at present and still needs further studies.

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

Response: We have modified "might be made" to "might not be made" accordingly in the revision.

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.

Response: We have deleted it as we have no appropriate rewording.

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

Response: The *f* and Dp/Dc are two different microphysical parameters of coated BC, and they show no relations. The *f* is absorbing volume fraction of coating, characterizing the percentage of BrC in the whole coatings, while Dp/Dc is shell/core ratio of coated BC that is the spherical equivalent particle diameter divided by BC core diameter.

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.

Response: Thanks for the constructive comments. The parameterization of the AAE of coated BC is challenging and difficult, because there are three microphysical parameters (i.e., *F*, Dp/Dc and *f*) for fitting, and common fitting is only for one parameter. We have tried the polynomial fit and multiple variable linear regression fit, and both are failed (The polynomial fit even cannot converge, while linear regression fit show very low correlation coefficient R^2). The power law model shown already is the best one at present.

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.

Response: We have redefined these parameters in Table 1.

Table 1: Key microphysical properties of coated BC aggregates

Parameters		applied values
F[a]		0.0, 0.25, 0.5, 0.75, 1.0
D_p/D_c[b]		1.1, 1.5, 1.9, 2.3, 2.7
f[c]		0.0, 0.25, 0.5, 0.75, 1.0
BC D_f[d]		1.8, 2.8
Size distribution	r_g, µm	0.075 (0.05–0.15)
	σ_g	1.59

[a]Coated volume fraction of BC.

[b]Shell/core ratio of coated BC that is spherical equivalent particle diameter divided by BC core diameter.

[c]Absorbing volume fraction of coating.

[d]Fractal dimension of BC aggregate.