Anonymous Referee #1
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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.

This paper uses the multi-sphere T-matrix method (MSTM) to analyze how BC size, aggregate fractal dimension, and mixing state affects the absorption Angstrom exponent (AAE). The article is well organized and well written, although it could benefit from some minor copy editing in some places. I find it suitable for publication after the corrections listed below.
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.

The introduction should be expanded somewhat, as there is significant work on this topic that the authors do not mention. For instance, see Liu, JQSRT 2019, Liu and Mishchenko, Rem. Sens. 2018 for aggregated BC computations. I would also search for more. As can be gathered from my comments below, much of the work cited in the intro is not consistent with what I have read in those articles.
Response: For the comments regarding inconsistence of cited articles, we will present in following point-to-point responses. We have added the aggregate BC computations in the Introduction, and cited both papers (Page 3, lines 7-8).

Page 2, line 1, authors state: "... the absorbing organics, named brown carbon (BrC), is one type of organic carbon absorbing radiation in the ultraviolet and visible spectra [Clarke et al., 2007]."
This is a little misleading. BrC is not one type of organic carbon; rather, BrC is composed of many different absorbing organic species.
Response: We have modified it accordingly (Page 2, lines 6-7).

Page 2, line 5, authors state: "The lack of accurate understanding and parameterization of the AAE of aged BC has been a pivotal limitation on the assessment of BC radiative effects [e.g., Ramanathan and Carmichael, 2008; Bond et al., 2013]."
This is very misleading, as these articles do not attribute such large importance to AAE. In fact, I did a search for "Angstrom" in RC08 and did not get a single hit.
Response: We have revised it accordingly, and abandoned citing both articles (Page 2, lines 11-12).

P2, L16, authors state: "Hence, the AAE can be utilized to quantify the separation of BrC absorption from BC absorption based on their distinctive functions of incident wavelength [e.g., Lu et al., 2015]."
This is an oversimplification of current AAE discussions, as there are plenty of articles in the literature stating that AAE can not unambigously separate BrC from BC (e.g., see Lack and Cappa (2010) and

Lack and Langridge (2013), Schuster ACP 2016, part 2, etc.). If you want readers to take this article seriously, you should highlight the current AAE issues that are being discussed in the literature and then tell readers how your contribution fits into the overall discussion.

Response: Thanks for the constructive comments. We have modified it accordingly (Page 2, lines 20-21).

P2, L22, authors state: "The AAE values of BC-dominated aerosols produced with burning oil, are observed in the range of 0.8–1.1 [e.g., Chakrabarty et al., 2013]."
But C13 concluded that only mustard oil was dominated by BC, and they measured an average AAE = 1.32 for mustard oil. How did the authors arrive at 0.8-1.1 from the C13 article?

Response: Thanks for the careful check from the reviewer. We have revised it accordingly (Page 2, lines 27-28).

P2, L24: I don't see AAE > 4 anywhere in Kirchstetter 2004.
P2, L26: I don't see BrC AAE ~8 anywhere in Clarke 2007.

Response: These AAE values not directly shown in the articles are based on our estimations, and we have abandoned citing both references (Page 2, line 29).

METHOD:
The aggregates used to represent BC in this study seem to have been drawn out of thin air. The authors do not discuss why they chose N=200 or Df = 1.8, 2.8 in detail. Later, the authors draw some fairly broad conclusions based upon this numerical work, but the reader is left wondering how the results might differ if the authors had chosen different aggregates. This is especially important, since the spherical coatings in Figure 1 do not look terribly realistic. How might the results change if the authors used less particles per aggregate (e.g., N = 40, as in Adachi, JGR 2010) and non-spherical coatings? How big are the primary spherules in this work? How would the results change if one alters the spherule sizes? What if one alters N? What role does shielding play? Large N –> more shielding –> less efficient absorption. It would be nice to see one of these aggregate papers address the shielding issue. I realize that shielding is probably too much to add to this paper, but acknowledging that shielding is an important topic that is still unaddressed would be nice.

Response: Thanks for the concerns from the reviewer. The aggregates used follow our previous papers, and detailed microphysical parameters and construction of coated BC aggregates have been illustrated therein (such as Zhang et al., 2018, 2019). Meanwhile, used parameters of coated BC aggregates (N=200 or Df = 1.8, 2.8) are commonly seen in various papers, such as Liu et al., 2017; Doner et al., 2017; Teng et al., 2019; Zeng et al., 2019. N=200 is often applied to model BC aggregate at accumulation mode, while BC Df of 2.8 and 1.8 represent compact and lacy BC aggregates, respectively.

For the monomer size, we follow Zhang et al., 2018. Only the accumulation mode is considered, as BC is observed to be mostly in accumulation mode. For the accumulation mode considered, the radius range is set as 0.05–0.5 μm in steps of 0.005 μm for the averaging. The exact sizes of BC aggregates can be known based on these coated BC sizes and shell/core ratios.

For the effects of BC monomer size or monomer number on the absorption of coated BC aggregates, both are the same question actually as we consider polydisperse coated BC aggregates with lognormal

size distributions. For coated BC aggregates with a fixed lognormal size distribution, more BC monomer number corresponds to smaller BC monomer size. We take N=1 as an extreme example (this is core-shell model with a spherical BC core), the absorption of fully coated BC aggregates is almost the same (see Fig. 2a in Zhang et al., 2017). So it is expected that the effects of BC monomer size or monomer number on our AAE results of polydisperse coated BC aggregates are trivial.

We assume spherical coatings, but it doesn't mean that the organics is a homogeneous sphere within the overall partially coated BC particle (the organics is a homogeneous sphere only in the case with F=0.0). To build the particle model of partially coated BC, we first generate a BC fractal aggregate and a homogeneous organics sphere, and after BC coated by organics, some BC monomers (volume fraction of F within all BC monomers) will take the place of some organics within the original homogeneous organics sphere. The assumption that the organics are spherical, are based on three aspects in this study. Firstly, the exact numerical method, MSTM, employed in this study is robust and fast in the calculation of optical properties of fractal BC particles, which is in the framework of the T-matrix method. Another powerful DDA method is almost two orders of magnitude slower than the MSTM for coated BC, as shown in Liu et al. [2017]. But the MSTM has the only limitation that the spherical surfaces are nonoverlapping (i.e., for spheres or a cluster of spheres). Secondly, no representative morphology of coating of organics is observed for ambient aged BC aerosols. Some observations of individual aged BC particles actually do show the spherical coating geometry [e.g., Alexander et al., 2008; Zhang et al., 2008; Wu et al., 2016], although some coatings may depict other geometries. While the fractal aggregates have been successfully employed to model BC geometries, simulating the geometry of organics for coated BC is difficult. Thirdly, however, for coated BC, the simple spherical coating is found to have similar effects on the optical properties to those based on more complicated coating structure [e.g., Dong et al., 2015; Liu et al., 2015; Liu et al., 2017]. Therefore, it is expected that similar absorption results and further AAE will be presented if the BC aggregates are modeled with a non-spherical coating.

For the shielding effect, we have mentioned this important topic (Page 4, lines 13-15).

References:

Alexander, D. T. L., Crozier, P. A., and Anderson, J. R.: Brown Carbon Spheres in East Asian Outflow and their Optical Properties, Science, 321, 833–836, 2008.

Doner, N., Liu, F., and You, J.: Impact of necking and overlapping on radiative properties of coated soot aggregates, Aerosol Sci. Tech., 51, 532–542, 2017.

Dong, J., Zhao, J. M., and Liu, L. H.: Morphological effects on the radiative properties of soot aerosols in different internally mixing states with sulfate, J. Quant. Spectrosc. Radiat. Transfer, 165, 43–55, 2015.

Liu, C., Li, J., Yin, Y., Zhu, B., and Feng, Q.: Optical properties of black carbon aggregates with non-absorptive coating, J. Quant. Spectrosc. Radiat. Transfer, 187, 443–452, 2017.

Liu, F., Yon, J., and Bescond, A.: On the radiative properties of soot aggregates - Part2: effects of coating, J. Quant. Spectrosc. Radiat. Transfer, 172, 134–145, 2015.

Teng, S., Liu, C., Schnaiter, M., Chakrabarty, R. K., and Liu, F.: Accounting for the effects of nonideal minor structures on the optical properties of black carbon aerosols, Atmos. Chem. Phys., 19, 2917-2931, 2019.

Wu, Y., Cheng, T. H., Zheng, L. J., and Chen, H. Optical properties of the semi-external mixture composed of sulfate particle and different quantities of soot aggregates, J. Quant. Spectrosc. Radiat. Transfer, 179, 139–148, 2016.

Zhang, R., Khalizov, A. F., Pagels, J., Zhang, D., Xue, H., and McMurry, P. H.: Variability in morphology, hygroscopicity, and optical properties of soot aerosols during atmospheric processing, P. Natl. Acad. Sci. U.S.A., 105, 10291–10296, 2008.

Zhang, X., Mao, M., Yin, Y., and Wang, B.: Absorption enhancement of aged black carbon aerosols affected by their microphysics: A numerical investigation, J. Quant. Spectrosc. Radiat. Transfer, 202, 90–97, 2017.

Zhang, X., Mao, M., Yin, Y., and Wang, B.: Numerical investigation on absorption enhancement of black carbon aerosols partially coated with nonabsorbing organics, J. Geophys. Res., 123, 1297–1308, 2018.

Zhang, X., Mao, M., and Yin, Y.: Optically effective complex refractive index of coated black carbon aerosols: from numerical aspects, Atmos. Chem. Phys., 19, 7507–7518, 2019

Zeng, C., Liu, C., Li, J., Zhu, B., Yin, Y., and Wang, Y.: Optical properties and radiative forcing of aged BC due to hygroscopic growth: Effects of aggregate structure, J. Geophys. Res., 124, 4620-4633, 2019.

How do the authors' results compare to other work, such as Liu and Mishchenko (Remote Sensing, 2018)? LM18 computed AAE for particles with many different aggregate configurations and mixing scenarios. Placing the author's results in the context of this wider study could help the reader understand the range of applicability of the results presented here.

Response: We have compared our results with this important work (Page 9, lines 14-15).

The authors frequently state that their calculations are "more realistic," but I have never seen TEM pictures that look like Figure 1b. There are also many articles with non- spherical aggregate coatings and therefore more realistic than Fig 1c (e.g., Adachi 2010). Many of these articles only address single particles, though.

Also, how do the fractal dimensions Df = 1.8, 2.8 shown in Fig 3 relate to the morphologies shown in Fig 1? That is, what are the Df for the morphologies of Fig 1? More importantly, what do the BC aggregates look like when Df = 1.8, 2.8 and N = 200?

Response: Thanks for the constructive comments. Some observations of individual aged BC particles actually do show the spherical coating geometry [e.g., Alexander et al., 2008; Zhang et al., 2008; Wu et al., 2016], which generally look like Figure 1b. Moreover, for coated BC, the simple spherical coating is found to have similar effects on the optical properties to those based on more complicated coating structure [e.g., Dong et al., 2015; Liu et al., 2015; Liu et al., 2017]. Therefore, it is expected that similar results of absorption and further AAE will be presented if the BC aggregates are modeled with a non-spherical coating.

The BC aggregate shown in Fig. 1a has a Df of 1.8, while its Df is 2.8 in Fig. 1c. The BC aggregate in Fig.1 has N=200, and BC Df of 2.8 and 1.8 represent compact and lacy BC aggregates, respectively.

For References, see previous Response.

P4, L22: Authors should make clear that these numbers pertain to aggregate sizes, not the monomers. Presumably these radii correspond to equivalent volume spheres, which should also be mentioned. Also, how is r_g related to the gyration radius of Eq 1, R_g?

Response: We have revised accordingly and mentioned these (Page 5, lines 3-4, 8-9).

The r_g in the size distribution is spherical volume-equivalent radius, which is different from the gyration radius Rg in Equation (2).

P5, L28, authors state: "...and the bias induced by chosen absorptions at two wavelengths may be averted."

The authors seem to be stating that the AAE errors are not subject to absolute measurement errors of absorption. However, the AAE is an exponent; as such, it is highly sensitive to absorption measurement errors when AAE is derived from two wavelengths. A simple perturbation analysis using "typical" measurement errors will illustrate this.

Response: We acknowledge that the absolute measurement errors of absorption can induce AAE errors, whereas what we try to talk about here is the issue of wavelength selection. We have modified it to make it clear (Page 6, line 3).

RESULTS:

P7, L1, authors state: "On the whole, the impacts of ... BC position within brown coating on the AAE of coated BC are generally negligible."

That's because the shells are not that much larger than the cores (Dp/Dc > 1.6). There are many early papers that investigated the effect of "randomly placed inclusions" vs. a "concentric inclusion." See Fuller JGR 1999, for example. It is worth noting the similarities and differences between your results and the early core/shell work, here.

Response: Thanks for the suggestion from the reviewer, whereas we are sorry that we cannot find this old paper (Fuller JGR 1999?) for a comparison.

P7, L23, authors state: "The above simulations assume BC coated by BrC, whereas it may be contaminated by non-absorptive organic carbon in ambient air."

Well, BrC is always "contaminated" with OC. That's because no-one has ever definitively separated BrC from OC. For instance, Kirchstetter separated OC from BC, so Kirchstetter's refractive indices represent a mixture of absorbing OC (now widely called BrC) AND non-absorbing OC. These are not two separate compounds, as both BrC and OC represent hundreds (thousands?) of compounds. I believe that this is why there is such a huge range of refractive indices for BrC in the literature. I believe that if anyone ever isolated the absorbing compounds of BrC from other OC, that the resulting BrC refractive index would be higher than the values that the community is using right now.

I really like the concept of this section, but the phrasing is misleading. What you are basically doing is assuming that the Kirchstetter BrC IRI is the upper extreme for BrC absorption, and then considering cases of BrC that are less absorbing than the Kirchstetter values. You could also look at the range of values provided by other groups as another (perhaps better) way of discussing variable BrC absorption. See Schuster ACP 2016 figures, for instance. Whatever you do, though, the wording should not convey the idea that Kirchstetter measured "pure" BrC. I don't believe that K04 meant to convey this.

Response: Thanks for the constructive comments from the reviewer, and we have revised accordingly

(Page 8, lines 10-13).

P9, L8, authors state: "In addition, our results with more realistic geometries indicate that occurrence of BrC can only be made with confidence if the AAE of coated BC is larger than 1.4, as the AAE smaller than 1.4 can not necessarily exclude BrC as an important contributor to particle absorption."
This sentence does not make sense to me.
Response: We have revised it to make it clear (Page 9, lines 23-24).

P10, L25, authors state: "Interestingly, BC coated by thin BrC with a large size distribution (i.e., large r_g) can have the AAE smaller than 1.0, and this implies that BC aerosols containing BrC can even show lower AAE than pure BC particles, which challenge conventional beliefs."
Pure and uncoated BC can also have AAE < 1 if the particles are large, according to Fig 4 when F=0. This corresponds to the geometry of Fig 1a, right? It would be nice if the authors are also able to present the AAE for a particle that are not touching another sphere, but I believe that they would still obtain AAE < 1 for large aggregates of BC. This should be mentioned here, because AAE is sensitive to particle size. See Fig 6, models 2 & 3 in Liu and Mishchenko (Rem. Sens., 2018); see also Gyawali (ACP, 2009) and Schuster (ACP, 2016).
I don't know what is considered to be "conventional belief," but the AAE = 1 assumption for BC is a by product of the Rayleigh small particle limit for absorption. Aggregates of BC do not necessarily satisfy the "small" criteria, so AAE = 1 does not necessarily hold (especially for collapsed aggregates with significant shielding). Open aggregates can be reasonably modeled as a loose collection of spheres, though, so the AAE = 1 approximations may hold for those cases. Thus, we expect a range of AAE for BC.
Response: Thanks for the reviewer's constructive comments. The results here not only correspond to the geometry of Fig. 1a (i.e., F = 0.0), but also relate to other geometries (see Fig. 4 and Fig. 6). The conventional belief here is that BC aerosols containing BrC should show larger AAE than pure BC particles (see page 11, lines 11-12).

Page 10, L30, authors state: "Our results with more realistic geometries also indicate that occurrence of BrC may be made confidently unless AAE>1.4, which is a replenishment of related findings of Lack and Cappa [2010] produced by the core-shell Mie model."
This is exactly opposite of LC2010, per their abstract: It has often been assumed that observation of an absorption Angstrom exponent (AAE)>1 indicates absorption by a non-BC aerosol. Here, it is shown that BC cores coated in C_Clear can reasonably have an AAE of up to 1.6, a result that complicates the attribution of observed light absorption to C_Brown within ambient particles. However, an AAE<1.6 does not exclude the possibility of C_Brown; rather C_Brown cannot be confidently assigned unless AAE>1.6. – LC2010
Response: We have revised it accordingly and abandoned the comparison in this way (Page 11, lines 15-16).

CONCLUSIONS:
P11, L16, authors state: "Meanwhile, BC coated by thin brown carbon with a large size distribution can show an AAE smaller than 1.0, implying that BC aerosols containing brown carbon can even show

lower AAE than pure BC particles, and this challenges conventional beliefs."
Here again, a BrC coating is not necessary to achieve AAE < 1.
Also, AAE = 1 for all BC is not a "conventional belief," as many of us know that particle size is important. Lack and Cappa (2010) discuss this, for instance. See also Gyawali (ACP, 2009) and Schuster (ACP, 2016 part 2).
Response: Thanks for the constructive comments. The results here not only correspond to the geometry of Fig. 1a (i.e., F = 0.0), but also relate to other geometries (see Fig. 4 and Fig. 6). The conventional belief here is that BC aerosols containing BrC should show larger AAE than pure BC particles.

MINOR ISSUES:
P4, L7, authors state: "...the volume of BC monomers within coating and overall BC volume..." It took me awhile to discern the meaning of this phrase. It would be helpful if the authors point the readers to Fig 1b, here.
Response: We have modified it accordingly (Page 3, lines 29-30).

P4, L8: k_f has not been defined thus far. Is this the same as the k_0 of Eq 1?
Response: We have changed k_f to k_0 (Page 4, line 12).

P5, Lines 1-7: This paragraph would be much stronger with an active voice. The authors are discussing things that are "normally" done and providing citations, which sounds like a literature review. The paragraph would be much clearer if the authors tell the reader what they are doing with an active voice; then the citations become the justification.
Response: We have revised it accordingly (Page 5, lines 10-16).

P5, L10 and throughout: I would avoid using the word "bulk" in this context, as bulk optical properties refer to bulk matter that is much much larger than the wavelength, which is not the topic of this paper.
Response: We have revised it accordingly (Page 5, line 24; Page 6, line 1).

P5, L9, authors state: "... can be calculated." Here again and throughout – get rid of passive voice. Tell the reader what you did, not what can be done.
Response: We have modified it accordingly (Page 5, lines 18-19).

P5, line 27: authors state that the slope of the line in Fig 2 is 2.1, but the figure indicates a negative slope. More precise wording is needed.
Response: We have modified it accordingly (Page 6, line 10).

P6, L12: The authors state that "the AAEs of BC coated by BrC are sensitive to fractal dimension,..." but their Figure 3 indicates that this sensitivity is small when Dp/Dc >1.5 or so for F =0, and that there is no sensitivity at all when F > 0. This should be mentioned in this paragraph.
Response: We have revised it (Page 7, lines 2-5).

P6, L22 and elsewhere: The authors frequently discuss the difference between compact and lacy BC aggregates, but they never tell the reader which Df is more compact (i.e., Df=1.8 or Df=2.8).
Response: We have revised it accordingly (Page 7, line 4).

Figures 4-7: It is annoying that the colorbar in Figs 4-7 unconventionally decreases upward.
Response: We consider coated BC microphysical parameters with many discrete points in the numerical study as shown in Table 1, and this may be the reason why the color bars do not look perfectly smooth.

P9, L3 and throughout: "In general, among all sensitive microphysical parameters of coated BC, the absorbing volume fraction of coating plays a more substantial role in the AAE determination."
More substantial than what? Comparative words like 'more' have to be 'more than' something. This seems to happen fairly frequently in this paper (e.g., "more realistic geometries" – more realistic than what?).
Response: Thanks for the comments, and we have modified it accordingly (Page 9, lines 18-19).

P9, Eqs 9 & 10: I don't understand the utility of these empirical equations. The authors are using 3 parameters that are difficult or impossible to measure in order to approximate something that is relatively easy to measure (the AAE). I don't understand the point.
Response: Thanks for the concerns from the reviewer. There are considerable inconsistences associated with AAE observations, and the uncertainties in absorption measurements at multi-wavelengths (such as using aethalometer) may be one significant reason. The Equations 9 and 10 can be act as the AAE response to the key sensitive microphysical parameters (i.e., absorbing volume fraction of coating, coated volume fraction of BC, and shell/core ratio) for a quantitative understanding. Moreover, the absorbing volume fraction of coating may be acquired with the chemical measurements by a single particle aerosol mass spectrometry (SPAMS) (e.g., Wang et al., 2019). The coated volume fraction of BC can be observed with a scanning electron microscopy (SEM) (e.g., China et al., 2013, 2015), while the shell/core ratio can be obtained using a single-particle soot photometer (SP2) (e.g., Liu et al., 2015; Zhang et al., 2016).

Anonymous Referee #2

Received and published: 1 May 2020

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 (Page 3, lines 23-30; Page 4, lines 1-2, 8-10).

- 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.

- 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 (Page 7, lines 8-10).

- 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 (Page 10, line 13).

- 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 (Page 1, lines 29-31).

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 (Page 1, lines 16-17).

Line 18, page 1: By “trivial” do the authors mean negligible?
Response: We have modified it accordingly (Page 1, line 20).

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 (Page 1, line 21).

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 it accordingly (Page 1, line 22).

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 (Page 2, lines 16-18).

Line 9, page 3: “This limits its applications: : :” what does “its” refer to?
Response: We have revised it accordingly (Page 3, line 11).

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 (Page 3, lines 29-30).

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” (Page 5, line 20).

Line 20, page 5: The sentence is not clear.
Response: We have modified it (Page 6, line 3).

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 (Page 6, lines 11-12).

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 (Page 6, line 13).

Line 7, page 6: what does “averagely” mean in this context?
Response: We have deleted “averagely” (Page 6, line 18).

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 (Page 6, line 29).

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 to help understand what exactly is the configuration considered (Page 6, line 21).

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 (Page 1, lines 25-27).

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 (Page 7, line 23).

(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.

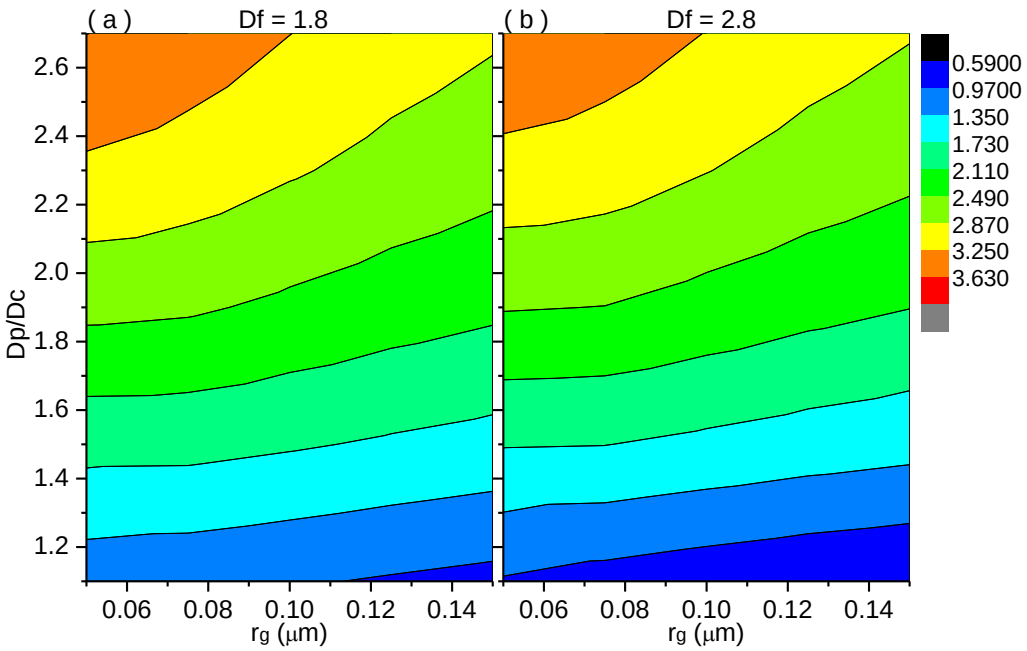

Fig. S2. The absorption Angstrom exponent (AAE) of BC aggregates coated by brown carbon with different shell/core ratio (Dp/Dc) and particle size distribution. BC fractal dimension of 1.8 and 2.8 are shown from left to right. Coated volume fraction of BC is 0.0, and geometric standard deviations (σg) 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 $\sigma_g < 1.39$).

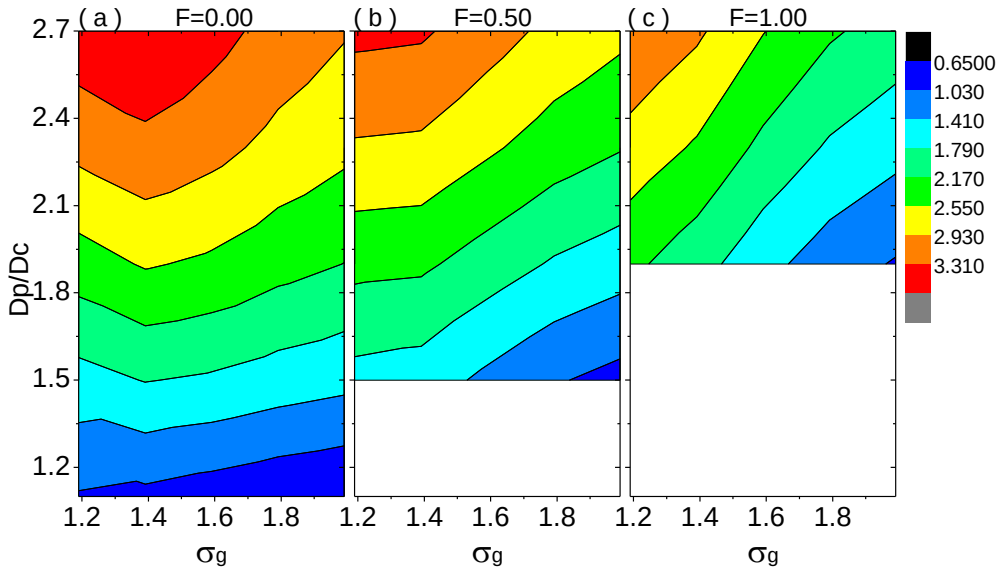

Fig. S3. The absorption Angstrom exponent (AAE) of BC aggregates (BC fractal dimension of ~2.8) coated by brown carbon with different shell/core ratio (Dp/Dc) and particle size distribution. Three coated volume fractions of BC, being 0.00, 0.50, and 1.00, are shown from left to right. For fully coated 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 revised it accordingly (Page 3, line 30; Page 4, line 1).

Line 23, page 7: I would not consider this to be a "contamination"
Response: We have revised it accordingly (Page 8, line 5).

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 it accordingly (Page 9, line 19).

Line 10, page 9" "remove "in" from "This is generally in consistent with the findings: : :"
Response: We have modified it accordingly (Page 9, line 24).

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 (Page 10, lines 4-5).

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 (Page 10, line 5).

Line 25, page 9: Maybe "power laws" is more appropriate than "exponential".
Response: We have revised it accordingly (Page 10, line 9).

Lines 2-4, page 10: This finding and explanation are confusing to me.
Response: We have revised it to make it clear (Page 10, lines 16-18).

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 (Page 10, lines 18-20).

Line 8, page 10: ": : :the capability of the expresses: : :" what does that mean?
Response: We have revised it to make it clear (Page 10, line 23).

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 (Page 10, line 27).

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 accordingly (Page 11, lines 15-16; Page 12, lines 2-3).

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 (Page 11, lines 15-16; Page 12, lines 2-3).

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.