

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

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Anonymous Referee #1

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.

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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 inconsistency 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 as follows. “Nonetheless, the core-shell Mie structure is in debate [e.g., Cappa et al., 2012], as lacy or compact fractal aggregates are widely accepted for BC geometries [e.g., Liu and Mishchenko, 2018; Liu et al., 2019].”

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 in the revision as: “Among BC coatings, in addition to non-absorbing components, the absorbing organics, named brown carbon (BrC), absorbs radiation in the ultraviolet and visible spectra [Clarke et al., 2007].”

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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 as the following: "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."

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 unambiguously 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 following: "The AAE cannot unambiguously be utilized to quantify the separation of BrC absorption from BC absorption despite of their distinctive functions of incident wavelength [e.g., Schuster et al., 2016]."

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 as the following: "The AAE of BC-dominated aerosols produced with mustard oil,

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is observed to be  $\sim 1.3$  [e.g., Chakrabarty et al., 2013]"

P2, L24: I don't see AAE > 4 anywhere in Kirchstetter 2004. P2, L26: I don't see BrC AAE  $\sim 8$  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.

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  $D_f = 1.8, 2.8$  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  $N \rightarrow$  more shielding  $\rightarrow$  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  $D_f = 1.8, 2.8$ ) 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  $D_f$  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

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the accumulation mode considered, the radius range is set as 0.05–0.5  $\mu\text{m}$  in steps of 0.005  $\mu\text{m}$  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.

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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 in the revision as: "The shielding effect of  $N$  on the absorption of BC aggregates is an important topic, as larger  $N$  can induce more shielding and result in less efficient absorption [Liu and Mishchenko, 2007]." 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,

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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 as the following: "The AAE of BC coated by non-absorbing organics in our study is coincident with corresponding results presented in Liu C. et al. [2018] and Liu L. et al. [2018]."

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  $D_f = 1.8, 2.8$  shown in Fig 3 relate to the morphologies shown in Fig 1? That is, what are the  $D_f$  for the morphologies of Fig 1? More importantly, what do the BC aggregates look like when  $D_f = 1.8, 2.8$  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.,

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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  $D_f$  of 1.8, while its  $D_f$  is 2.8 in Fig. 1c. The BC aggregate in Fig.1 has  $N=200$ , and BC  $D_f$  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 in the revision as: "Coated BC follows this size distribution, while  $r$  is the radius of equivalent volume sphere that has the same volume as that of coated BC aggregate. The exact sizes of BC aggregates can be known on the basis of these coated BC sizes and shell/core ratios." The  $r_g$  in the size distribution is spherical volume-equivalent radius, which is different from the gyration radius  $R_g$  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 as the following: "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

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and Chakrabarty, 2011].”

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 ( $D_p/D_c > 1.6$ ). 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

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revised accordingly following: "It should be noticed that no one has ever definitively separated BrC from organic carbon, and to a certain extent, the concept of  $f$  here may be treated as that the cases of BrC with imaginary parts of refractive indices less than those of Kirchstetter et al. [2004] are considered due to a range of BrC refractive indices being provided [Schuster et al., 2016]."

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 as the following: "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."

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

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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.

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 following: "Our results with more realistic geometries also indicate that occurrence of BrC may not be made confidently unless  $AAE > 1.4$ ."

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

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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 in the revision as: "where VBC inside and VBC are the volume of BC monomers encapsulated in coating and overall BC volume, respectively (see Fig. 1)."

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  $k_0$ .

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 following: "We investigate absorption properties of coated BC particles at multiple incident wavelengths between 350 nm and 700 nm in steps of 50 nm. We consider a typical BC refractive index of 1.85-0.71i [Bond and Bergstrom, 2006], as it is normally assumed as wavelength independent in near-visible and visible spectral regions [Moosmuller et al., 2009; Luo et al., 2018]. For the refractive index of coating of absorbing organics (i.e., brown carbon), this study assumes its real part to be a constant of 1.55 [Chakrabarty et al., 2010], whereas its imaginary part is substantially dependent on incident wavelength over shorter visible and ultraviolet regions [e.g., Moosmuller et al., 2009; Alexander et al., 2008]. The imaginary parts of BrC refractive indices at different wavelengths assumed in this study follow Kirchstetter et al. [2004], and are shown in Fig. S1."

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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 deleted "bulk" in the revision accordingly.

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 as: "Given that bulk absorption cross sections at various wavelengths are obtained, we calculate the absorption Angstrom exponent of coated BC, a microphysical parameter describing the wavelength variation in particle absorption."

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 following: "the negative of the line slope".

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  $D_p/D_c > 1.5$  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 as the following: "The AAE of coated BC aggregates is also slightly sensitive to BC  $D_f$ , and the sensitivity shows weaker as  $D_p/D_c$  or  $F$  become larger. The AAEs of compact BC coated by BrC (i.e., BC  $D_f=2.8$ ) are generally smaller than those of lacy coated BC (i.e., BC  $D_f=1.8$ ) with differences less than 0.3, and there is almost no sensitivity of AAE to BC  $D_f$  for  $F > 0$ ."

P6, L22 and elsewhere: The authors frequently discuss the difference between compact and lacy BC aggregates, but they never tell the reader which  $D_f$  is more compact (i.e.,  $D_f=1.8$  or  $D_f=2.8$ ).

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Response: We have revised it accordingly following: "The AAE of coated BC aggregates is also slightly sensitive to BC  $D_f$ , and the sensitivity shows weaker as  $D_p/D_c$  or  $F$  become larger. The AAEs of compact BC coated by BrC (i.e., BC  $D_f=2.8$ ) are generally smaller than those of lacy coated BC (i.e., BC  $D_f=1.8$ ) with differences less than 0.3, and there is almost no sensitivity of AAE to BC  $D_f$  for  $F > 0$ ."

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 as: "In general, the absorbing volume fraction of coating plays a more substantial role in the AAE determination than other sensitive microphysical parameters."

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 inconsistencies 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

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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).

Please also note the supplement to this comment:

<https://www.atmos-chem-phys-discuss.net/acp-2020-224/acp-2020-224-AC2-supplement.pdf>

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Interactive comment on Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2020-224>, 2020.