**Response to the comments of the Reviewer #3**

(The responses are highlighted in blue)

First of all, we would like to thank the three anonymous reviewers for their 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 the revised manuscript in **BLUE** in the revision.

In this response, the questions and comments of reviewers are in black font, and responses are highlighted in **BLUE**. The changes made in the revised manuscript are marked in RED font.

The topic of black carbon (BC) absorption enhancement has been investigated by numerous previous modeling/lab/field studies. The present manuscript systematically quantified the effects of brown carbon coating and associated morphological properties on BC absorption enhancement and proposed a "sunglasses effect", which provides some new understanding in this topic. This study is suitable for ACP and its structure is clear. Before it can be considered for publication, I have a few comments and suggestions to help improve the manuscript.

Abstract: The authors mentioned "thickly-coated" and "thinly-coated" here. How thick is "thickly-coated"? Please quantitatively define it here and in the main text as well

.**Response:** Thanks for your comments and valuable suggestions. In this work, BC is defined as thickly coated when the BC volume fraction is lower than 20%, and other BC is considered to be thickly coated. We have defined it in the abstract and the introduction of the revised manuscript.

Abstract (Lines 11-12): "the uncertainties … have differences of less than 2.6% and 6% …". The expression "uncertainties have differences …" is weird. Please rephrase this sentence.

**Response:** Thanks for your comments. We agree that this sentence would make readers confused. After carefully consideration, we think that this point is not the most important in this work, therefore we have removed this sentence in the revised manuscript, and the abstract is rewritten. They are all marked in blue in the revised manuscript.

Page 1, Line 20: "second contribution" should be "second contributor". The reference for this sentence should also include Bond et al., 2013 (JGR).

**Response:** Thanks for pointing it out. We have added the reference in the revised manuscript.

Introduction: The authors mentioned that BC absorption enhancement varies significantly in different field measurements, which could be due to complex morphology and mixing state during BC aging processes. But one missing part is the evidence for the complex BC morphology and mixing state observed in field measurements (e.g., Y. Wang et al. 2017, doi:10.1021/acs.estlett.7b00418; S. China et al. 2013, doi:10.1038/ncomms3122). I suggest including several sentences in the introduction to point out this aspect.

**Response:** Thanks for your comments and suggestions. We have added it in the introduction of the revised manuscript:

"Freshly emitted BC commonly presents fractal structures. As the BC ages in the atmosphere, BC becomes more compact and OC materials can condense onto the particles. Therefore, BC can be embedded in an OC shell (China et al., 2013a; Wang et al., 2017). When non-BC fraction is low, BC can still present near fractal structure (referred as thinly coated in this study) (Wang et al., 2017). As BC further coated, BC aggregates are collapsed into more compact and spherical clusters when fully engulfed in coating material (referred as thickly-coated BC in this study) (Zhang et al., 2008b)."

Page 4, Lines 8-10: Using $D_f$ values to define "thickly-coated" and "thinly-coated" BC is not straightforward. Why not use the coating thickness or mass directly? There may be some situations where $D_f$ is smaller than 2.6, but the coating is still more than that of BC with a relatively higher *Df*.

Response: We are sorry for not clarifying the definition of "thickly-coated" and "thinly-coated" BC. We don't define "thickly-coated" and "thinly-coated" BC using $D_f$ values. In this work, BC is defined as thickly coated when the BC volume fraction is lower than 0.2.

We do acknowledge that in some cases, $D_f$ of BC is small but the coating fraction is high (such as the partially encapsulated coated BC). In general, however, thickly-coated BC is with compact structures due to squeeze effect. China et al. (2013) reported that atmospheric soot particles with the thickest coating had the highest fractal dimension. Compact structures are commonly observed for aged BC (Moffet and Prather, 2009). Therefore, $D_f$=2.6 was assumed for thickly-coated BC. This aging process is also assumed in other studies (such as the study of Wu el at., 2016; 2018 and Kanngiesser et al., 2018).

Page 5, Lines 6-10: Recently, another important and efficient particle light-scattering method, the geometric-optics surface-wave (GOS) method (Liou et al. 2011, doi.org/10.1016/j.jqsrt.2011.03.007; C. He et al. 2016, doi.org/10.1016/j.jqsrt.2016.08.004), has also been developed and applied to resolve complex BC coating structures and showed consistent results with MSTM, which could be included here

Response: Thanks for your comments. We have included it in the revised manuscript.

Page 6, Lines 13-14: The authors assumed BrC coatings are uniformly distributed over the BC surface, but they also argued that the blocking effect of coating is important, which could be affected by how coating materials are distributed over BC particle surface. Thus, assuming the uniform distribution of BrC coating may lead to nontrivial biases in calculations. Could the authors comment or add some discussions on this?

Response: We are sorry for not clarifying the means of the sentence in Page 6, Lines 13-14, and it doubts you. What we really want to express is that the composition ratio of BC to coating is independent to size. It is reasonable to make the simplification for easily understanding the effects of brown coatings.

The coatings are believed to be uniformly distributed over the BC surface for thinly coated BC (closed cell). Moreover, Kahnert (2017) has demonstrated the differences between closed cell model and more morphologically realistic model are not large for calculation of absorption of thinly-coated BC. For thickly coated BC, the relative position of BC to coatings can also affect the absorption, while one should not expect large difference in absorption (Liu et al. 2017, He et al. 2015). In this work, the geometric center of black carbon are located in the center of BrC sphere.

Page 7, Lines 11-12: "Generally, $E_{abs}$ increases … with increasing $k_{BrC}$." Is this true for all wavelengths? Please clarify here.

Response: Thanks for your comments. It is true that $E_{abs}$ increases with increasing $k_{BrC}$ for all wavelengths selected in this work. The reason is that the absorption of BrC increases with $k_{BrC}$, which contributes the increase of $E_{abs}$. Even though the sunglass effect also increase with $k_{BrC}$, the increase in sunglass effects is relative small, compared with the increase caused by BrC absorption, therefore leads to increase in $E_{abs}$.

Page 7, Lines 16-17: "… compared with BC with non-absorbing coatings, $E_{abs}$ for thinly-coated BC with absorbing coatings seems to be less wavelength-dependent, …" This is interesting but a little counter-intuitive. Could the authors provide some explanations?

**Response:** Thanks for your comments. The wavelength-dependence of $E_{abs}$ for absorbing coatings is at fixed $k_{BrC}$. The results is the interaction of the lensing effect and BrC absorption. At fixed $k_{BrC}$, lensing effect commonly decrease with wavelength for thinly-coated BC, while BrC absorption can increase with wavelength at fixed $k_{BrC}$. Therefore, the two effects are counteracted for thinly-coated BC. Therefore, $E_{abs}$ wavelength–dependences of thinly-coated BC decrease with wavelength. However, for thickly coated BC, the lensing effect also increases with wavelength. Therefore, the effect of lensing effect and BrC absorption are superimposed, which leads to larger wavelength-dependence of $E_{abs}$.

Section 3: The authors highlighted two important but opposite effects: conventional lensing effect and sunglasses effect. It is interesting to see how these two effects vary with $k_{BrC}$, $D_f$, and wavelength. Since the authors already calculated the absorption due to these two effects, it is straightforward to calculate the contributions of these two effects to the total absorption enhancement. This would be very informative and worth discussing. Also, according to the authors' arguments, there should be one critical point (or critical $k_{BrC}$ value) for the two effects to be the same. It would be very interesting to see what this point/value is.

**Response:** Thanks for your comments. We agree that it is straightforward to calculate the contributions of these two effects to the total absorption enhancement.

After carefully consideration, we think lensing effects is for non-absorbing coatings, the definition is as follows:

$$E_{lensing} = \frac{C_{abs\_coated\_non\text{-}absorbing}}{C_{abs\_bare}}$$

Combining the comments of other reviewers, we think that the lensing effect defined in Equation (5) of the previous version of the manuscript is a little misleading. The lensing effect defined in Equation (5) of previous version is the comparison of the absorption of BC coated by BrC with an external mixture of BrC and BC. It is resulted from the interaction of lensing effect and sunglass effect.

Liu et al. (2017a) defined the lensing effect as the absorption enhanced by addition of non-black carbon. However, from a physical point of view, for BC with BrC coatings, the definition may be not clear, and it can be confused with $E_{abs}$. Therefore, we redefined the lensing effect as the absorption enhanced by addition of non-absorbing coatings in the revised manuscript. In addition, we assume that the lensing effect of BC with absorbing coatings is the same as those with non-absorbing coatings. We believe this is a reasonable assumption since the BrC and nonabsorbing coating have a similar value of real part of refractive index..

We clearly defined the sunglass effect in the revised manuscript. We contribute $E_{abs}$ of BC with BrC coatings to lensing effect, BrC absorption enhancement and sunglass effect. Therefore:

$$E_{Sunglass} = -\frac{C_{abs\_\mathrm{coated}} - C_{abs\_BrC\_shell} - C_{abs\_\mathrm{non-absorbing}}}{C_{abs\_bare}}$$

The negative sign represents that the sunglass effect can cause the decrease of total absorption. There is indeed be one critical point (or critical $k_{BrC}$ value) for the two effects to be the same ($E_{abs\_internal}$<1 in the revised manuscript). However, the critical $k_{BrC}$ value is dependent on the mixing states and particle size. Therefore, it is difficult to give the critical $k_{BrC}$ value for each case. Nevertheless, we have investigated the effects of mixing states and particle size in the revised manuscript:

" Generally, the threshold $k_{BrC}$ value decreases with particle size and coatings thickness, as $E_{abs\_internal}$ of BC thickly-coated with absorbing coatings decreases with particle size and coatings thickness (see Figure 6 and Figure 9)."

Page 8, Line 19: "shorter wavelength". Please give a more quantitative wavelength range

**Response**: Thanks for your comments. This sentence has been corrected in to "larger absorption enhancement can be observed by increasing λ from ultraviolet region to visible region".

Page 8, Line 15 (and elsewhere): "relative errors". I suggest using "relative uncertainty" instead of "error".

**Response:** Thanks for your suggestions. We have corrected it in the revised manuscript.

Page 12, Line 1: "combined of $E_{abs}$ …". Should it be "combining $E_{abs}$ …"?

**Response:** Thanks for your suggestions. We have corrected it in the revised manuscript.

Section 4: Could the authors add some discussions on how to apply their results in this study to climate models? Current climate models do not simulate any morphological information of aerosols and generally assume a core-shell structure or external mixing for aerosols.

**Response:** Thanks for your comments. In the revised manuscript, we have added the calculation of mass absorption cross section (MAC). The MAC of bare BC can be estimated by measurements or parameterization. After investigating the mechanism of absorption enhancement, we can understand and simulated the MAC for coated BC in various circumstances. If validated by measurements, we can incorporated the results into the CTMs. We have added some discussions on this aspect in the section 4:

"In this work, complex morphologies and mixing states are considered. Although current climate models do not simulate any morphological information of aerosols, many laboratory studies has been conducted to investigate the BC morphologies in different mixing states and in different regions. Therefore, our calculations can be applied according to specific mixing states (such as composition ratios) and regions. However, we acknowledge that the understanding of the relation between BC morphology and the composition ratio is still limited. Therefore, further laboratory investigations for the coated BC morphologies should be conducted in the future."

**Reference**

China, S., Mazzoleni, C., Gorkowski, K., Aiken, A. C., and Dubey, M. K.: Morphology and mixing state of individual freshly emitted wildfire carbonaceous particles, Nat Commun, 4, 2013.

Cheng, T. H., Wu, Y., and Chen, H.: Effects of morphology on the radiative properties of internally mixed light absorbing carbon aerosols with different aging status, Opt Express, 22, 15904-15917, 2014.

He, C., Liou, K. N., Takano, Y., Zhang, R., Zamora, M. L., Yang, P., Li, Q., and Leung, L. R.: Variation of the radiative properties during black carbon aging: theoretical and experimental intercomparison, Atmos Chem Phys, 15, 11967-11980, 2015.

Kahnert, M.: Optical properties of black carbon aerosols encapsulated in a shell of sulfate: comparison of the closed cell model with a coated aggregate model, Opt Express, 25, 24579-24593, 2017.

Kanngiesser, F., and Kahnert, M.: Calculation of optical properties of light-absorbing carbon with weakly absorbing coating: A model with tunable transition from film-coating to spherical-shell coating, J Quant Spectrosc Ra, 216, 17-36, 2018.

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

Wu, Y., Cheng, T. H., Zheng, L. J., and Chen, H.: Black carbon radiative forcing at TOA decreased during aging, Sci Rep-Uk, 6, 2016.

Wu, Y., Cheng, T. H., Liu, D. T., Allan, J. D., Zheng, L. J., and Chen, H.: Light Absorption Enhancement of Black Carbon Aerosol Constrained by Particle Morphology, Environ Sci Technol, 52, 6912-6919, 2018.