Responses to the comments of the reviewers

(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.

Response to the reviewer #1

This is a solid contribution on an important subject. I appreciate the authors' us of state-of-the-art modeling techniques for the study of soot-containing aerosols with highly complex morphologies. The technical content of the paper appears to be correct, and the conclusions are well justified. I have only three minor comments.

1. In Section 2.3, an appropriate generic reference for the DDA would be J. Quant. Spectrosc. Radiat. Transfer 106, 558-589 (2007).

Response: Thanks for pointing it out. We have modified it in the revised manuscript.

2. The authors model randomly oriented non-spherical aerosols. The use of the model of randomly oriented particles has two aspects (see the recent rigorous analysis in Optics Letters 42, 494-497 (2017)). First, the orientation distribution function mush have a specific mathematical form, so I wonder whether this is the case with the computer program used to calculate light scattering. Second, technically speaking, the computation for a non-spherical particle must be supplemented by the computation for its mirror image. I wonder whether this was done, or it was found that the two computations yield very close results. These two issues need to be clarified.

Response: Thanks for your comments. First of all, we agree that the calculations for randomly oriented non-spherical aerosols should be clarified. The non-spherical particles that we considered in this manuscript are atmospheric aerosols. It is reasonable to assume the orientations of black carbon (BC) particles in the atmosphere is completely random, that is to say that the probability of every orientation is identical. As a result, the normalized probability density function of particle directions is nearly a constant. Therefore, Eqs. (3) in the study of Mishchenko and Yurkin, (2017) is satisfied. We have added some text and related reference in the revised manuscript:

"In this study, all the radiative properties of BC were calculated based on the assumption that BC particles and their mirror counterparts are present in equal numbers in ensemble of randomly oriented particles. In the atmosphere, it is reasonable to assume that the possibility of each particle direction is identical, which mathematically satisfies the definition of random orientation (Mishchenko and Yurkin, 2017)."

Rigorously, the computation for a non-spherical particle should indeed be supplemented by the computation for its mirror image. However, Kahnert (2017) has demonstrated that the calculations for closed-cell model calculated using DDA by numerically averaging over each particle direction and those using MSTM don't deviate largely. This indirectly verifies that the results for randomly oriented non-spherical aerosols are close to their mirror counterparts. In addition, we found little changes by altering the option of target_euler_angles_deg in MSTM. Therefore, we didn't provide the computation for their mirror counterparts.

3. The authors' analysis of the differences between the effects of absorbing and non-absorbing shells is quite interesting. It would be instructive to compare their observations with those in Optics Letters 39, 2607-2610 (2014).

Response: Thanks for your suggestion. We have compare the results in present study with the results presented in Mishchenko et al. (2014) in the section 3.1 of revised manuscript:

"For aged BC with thick coatings, BC absorption is underestimated at the UV, visible, and IR wavelengths (Kahnert et al., 2012). Mishchenko et al. (2014) has also demonstrated that the C_{abs} of thickly coated with non-absorbing coatings is significantly underestimated by a core-shell sphere, and investigated the effects of off-center of BC. Their results indicated that the C_{abs} of aged BC covered with thickly non-absorbing coatings are approximately 1.44 times higher than those calculated with a core-shell sphere model. Nevertheless, the effects of coating absorption on the applicability of the core-shell sphere model have not been evaluated".

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.

Mishchenko, M. I., and Yurkin, M. A.: On the concept of random orientation in far-field electromagnetic scattering by nonspherical particles, Opt Lett, 42, 494-497, 2017.

Mishchenko, M. I., Liu, L., Cairns, B., and Mackowski, D. W.: Optics of water cloud droplets mixed with black-carbon aerosols, Opt Lett, 39, 2607-2610, 2014.

Response to the reviewer #2

Black carbon (BC) particles mixing with brown carbon (BrC) coatings are simulated by two morphologies, including: thinly and thickly coated states. Light absorption properties of BC-containing particles are calculated using the superposition T-matrix method. The sensitivity of the imaginary part of BrC refractive index on the light absorption is investigated for the realizations with the aerosol ensembles. The authors showed some interesting results, but the effects they presented are not clear and the simulations are not validated by the measurements. While you revise the paper, please take the following into consideration.

Response: Thanks for your valuable comments. Please see the point-to-point response below to the concerns raised for this manuscript.

1. In this study, the absorption enhancement, lensing effect, blocking effect, sunglasses effect, and strengthening effect are discussed, but they are confused. In the previous studies, e.g. Liu (2017), "lensing effect is that the addition of non-black-carbon materials to black-carbon particles may enhance the particles' light absorption by 50 to 60% by refracting and reflecting light". The clear definitions of these effects are important, because brown coating also absorbs solar radiation itself. In Equation (3), the effect of brown coating on absorption enhancement is not considered, and may generate an unreasonable E_{abs} value, such as 5.4 (Line 5 in abstract). It would more appropriate to compare the absorption of BC coated by BrC with an external mixture of BrC and BC, rather than bare BC alone. In Equation (4), how to calculate far-field results of C_{abs_BrC} (total_size) and C_{abs_BrC} (bare_size) in the BC-BrC mixtures, and do you considered the complex morphologies of BrC in 'total size' cases? In bare BC, the BrC coating may be not exist. The 'lensing effect' is widely used in the climate studies, thus, Equation (5) may be potentially misleading. It is necessary to clearly explain these effects.

Referece: Liu, Dantong, et al. "Black-carbon absorption enhancement in the atmosphere determined by particle mixing state." Nature Geoscience 10.3 (2017): 184.

Response: Thanks for your comments and valuable suggestions. We agree that the definition of $E_{abs\_lensing}$ in the previous version of the manuscript is not clear. The sunglass effect, BC absorption enhancement, BrC enhancement, and the lensing effects should be clearly explained.

Usually, "lensing effect is widely used in the climate studies", and they contribute all absorption enhancement of internally mixed BC to the "lensing effect" of coatings. From a physics point of view, we think lensing effects is suitable for non-absorbing coatings, the definition is as follows:

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

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 we defined.

Liu et al. (2017a) defined the lensing effect as the absorption enhanced by addition of non-black carbon. However, from the 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 agree that it is a valuable suggestion to compare the absorption of BC coated by BrC with an external mixture of BC and BrC. Actually, the $E_{abs\_lensing}$ defined in the previous version of the manuscript has compared the absorption of BC coated by BrC with an external mixture of BC and BrC. However, the comparison is not in a direct manner. Therefore, we define a new parameter, $\boldsymbol{E_{abs\_internal}}$, to represent the ratio between the absorption of BC coated by BrC and an external mixture of BC and BrC. However, in the revised manuscript, the E_{abs} was compared with measurement results in the literatures, while the $E_{abs\_internal}$ was not. It is because the E_{abs} were commonly measured while usually $E_{abs\_internal}$ results were not available by measurements.

The absorption of BrC shell is calculated by the absorption of BrC that is with the same shape as the coated BC subtracting the absorption of BrC that is with the same shape as the bare BC, as shown in Equation (4) of previous version. The calculation of BrC shell is illustrated in Figure 1 in this response (Figure S1 in the revised manuscript). In this process, we assume that the absorption of BrC with the same shape as the coated BC is identical as the external mixture of BrC with the same shape as bare BC and BrC shell. We must clarify that this process neglects the blocking effect and lensing effect of outer BrC shell on the internal BrC. However, as the BrC absorption is significantly less than the BC absorption with identical shape, the absorption caused by the blocking effect and lensing effect of outer BrC on the internal BrC is relative small compared with the BC absorption. Therefore, it is reasonable to make some simplifications.

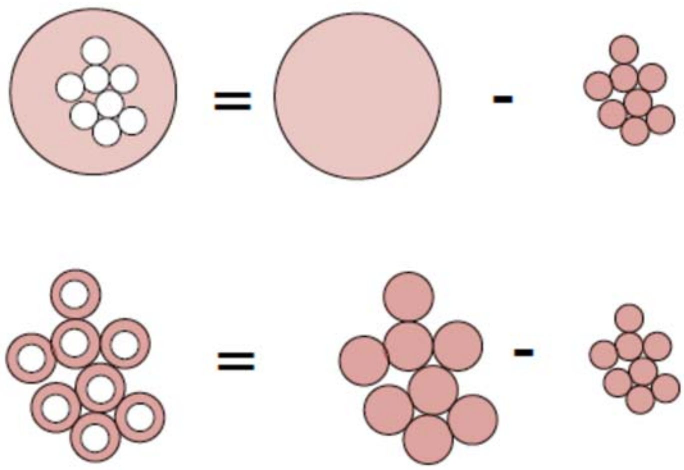


Figure 1 Calculation of the absorption of BrC shell.

Therefore, we indeed considered the complex morphologies of BrC in 'total size' cases. We think "$C_{abs\_BrC\_(total\_size)}$" and "$C_{abs\_BrC\_(bare\_size)}$" may be a little confusing. For this reason, we have changed "$C_{abs\_BrC\_(total\_size)}$" and "$C_{abs\_BrC\_(bare\_size)}$" into "$C_{abs\_BrC\_(coated\ shape)}$" and "$C_{abs\_BrC\_(bare\ shape)}$". It is true that the BrC coating don't exist in the case of bare BC. However, Equation (4) is aimed to calculate the BrC shell of absorption.

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. Combining all the definitions, we can easily obtain the relation that the absorption of BC coated with BrC is less than that of an external mixture of BrC and BC when $E_{Sunglass} > E_{abs\_lensing} - 1$.

One may confused about the calculation of coated BC with the BrC shell. Coated BC was calculated directly using MSTM. The calculation of the absorption caused by the Sunglass effect is shown in Figure 2 of this response.

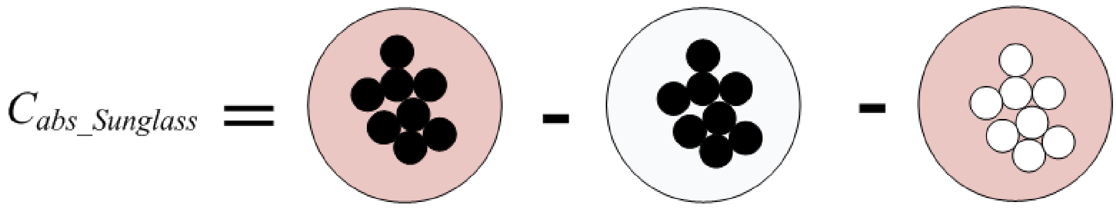
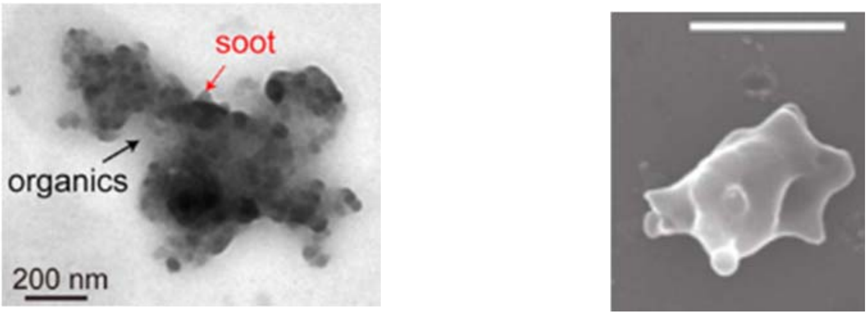


Figure 2 Calculation of the absorption caused by the sunglass effect. $C_{abs\_Sunglass}$ represents the absorption cross-section caused by the sunglass effect.

2. Line25-27, Page 2. "Nevertheless, in the atmosphere, there is a type of organic carbon that absorbs the radiation in the range of the ultraviolet and visible spectra, which is known well as brown carbon (BrC); BC can also be mixed with BrC." Please give the references about the morphologies and mixing states of BC-BrC mixtures to support this simulation.

Response: Thanks for your comments. As the BC ages in the atmosphere, BC becomes more compact and other materials (such as sulfate and organic substances) can condense onto the particles. The organic coating can be POA or SOA. BC can be embedded in a non-BC shell (Wang et al., 2017) (China et al., 2013). When non-BC fraction is low, BC can still present fractal structure, as demonstrated in the Figure 1 (a-3) of Wang et al. (2017) (Figure 3a in this response). As BC is further coated, BC becomes more compact and the coating shell becomes more spherical (Lewis et al., 2009), as shown in the Figure 3 of China et al. (2013) (Figure 3b in this response). Eventually, 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). We added the references about the morphologies and mixing states of BC in the introduction of 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 is 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)."



(a) (b)

Figure 3 Typical morphologies of coated BC. (a) Thinly-coated BC with fractal structure (Wang et al., 2017); (b) Coated BC with more spherical coating (China et al., 2013).

3. In Section 3.2 Bulk radiative properties, it is suggested to estimate the absorption enhancements of BC aerosols by the mass absorption cross section (MAC) rather than the cross section (Cabs), because of the normalization of BC mass. Moreover, the simulations of MAC can be validated by the previous measurements.

Response: Thanks for your valuable suggestions. The calculation of E_{abs} was not estimated by mass absorption cross section (MAC), the reason is that the absorption enhancement is defined as the amplification of total absorption but not the amplification of mass absorption. Although many studies used the MAC to estimate E_{abs}, MAC is calculated as the mass absorption per unit mass of BC but not per unit mass of total BC-containing aerosols. Therefore, the MAC can be calculated using:

$$MAC_{coated/bare} = C_{abs\_coated/bare} / m_{BC}$$

Where m_{BC} represents the mass of BC.

The E_{abs} can be expressed as:

$$E_{abs} = \frac{MAC\_abs\_coated}{MAC\_abs\_bare} = \frac{C_{abs\_coated}}{C_{abs\_bare}}$$

This is consistent with the definition of E_{abs} in our work.

We do agree that our calculations should be validated by measured results. We compared the modeled E_{abs} and MAC with the results of Liu et al. (Liu et al., 2015), and the comparison is supplemented in support information, as shown in the Figure S1. The measured E_{abs} values were estimated from Figure 1 of Liu et al. (Liu et al., 2015). In this work, we assume the E_{abs} and MAC at λ=700 nm and 781 nm do not deviate largely. The comparison of measured and modeled MAC is shown in Figure S7 and Figure S8 in the revised manuscript. When BC mass density is assumed to be 1.5g/cm^3, the calculated MAC and E_{abs} are relatively well in agreement with the measurements (see Figure 5 in this response or Figures S8-S9 in the revised manuscript). As for why assuming BC mass density to be 1.5g/cm^3, we have explained the reason in the revised manuscript (page 11 in the revised manuscript):

"In this work, we assume ambient BC mainly composed of primary organic matter with a low degree of oxidation. Based on the study of Nakao et al. (2013), an OC mass density range of 1-1.2 g/cm^3 has been used by Liu et al. (2017). ρ_{BrC} = 1.1 g/cm^3 is assumed in this work. For the BC mass density, the study of Horvath (1993) gives values of ρ_{BC} = 0.625 g/cm^3 and ρ = 1.125 g/cm^3. However, Fuller et al. (1999a) pointed out that the values may be not representative for BC in the atmosphere. Medalia and Richards (1972) and Janzen (1980) suggested ρ_{BC} in the range of 1.8-1.9 g/cm^3, while Bergstrom (1972) found that the ρ_{BC} value of 1.9-2.1 g/cm^3. Bond and Bergstrom (2006) suggested to use a value of 1.8 g/cm^3. Figure S8 compares the computations with measurements by assuming ρ_{BC} = 1.8 $g/cm^{3.}$ We assume that E_{abs} and MAC at λ = 0.7 *um* do not deviate largely with those at λ = 0.781 *um*. Modeled E_{abs} at λ = 0.7 *um* agrees well with the measurements. Although E_{abs} at λ = 0.404 *um* seems to be relatively higher than the measurements, it dose not deviate largely with the measurements. However, modeled MAC is a little smaller than the measured MAC. Similar results were obtained for bare BC ((Kahnert, 2010b), (Liu and Mishchenko, 2005)) . Therefore, ρ_{BC} = 1.8g/cm^3 may be a little high for estimation of MAC.

Bond and Bergstrom (2006) concluded that MAC value of 7.5 ± 1.2 m^2/g for bare BC can be assumed at λ = 0.55um by reviewing 21 publications of MAC measurements. However, our calculated MAC of 6.02-6.2 m^2/g (see Table 2) at λ =0.532 um lies below the range of MAC values suggested by Bond and Bergstrom (2006). Similar conclusions were drawn by Kahnert (2010b) and Liu and Mishchenko (2005). However, our calculated MAC agrees well with the calculated MAC of 6.0 ± 0.1 m^2/g by Kahnert (2010b) at λ = 0.55 um. As MAC depends significantly on BC mass density, to agree with measurements, Liu and Mishchenko (2005) used ρ_{BC} = 1.0 g/cm^3. However, as pointed by Kahnert (2010b), the measured MAC and modeled MAC were not at the same wavelength, therefore leading to too low retrived ρ_{BC}. To raise the computed MAC values to the average observed value of MAC = (7.5 ± 1.2) m^2/g, ρ_{BC} = 1.3-1.4 g/cm^3 was suggested by Kahnert (2010b). However, this ρ_{BC} value is rather drastic smaller than the value suggested by Bond and Bergstrom (2006). Therefore, Kahnert (2010b) suggested to assume ρ_{BC} = 1.5 ~ 1.7 g/cm^3 to raise the computational MAC results to the lower bound of the observations. By assuming ρ_{BC} = 1.5 g/cm^3, the comparison of modeled MAC and E_{abs} with measurements is shown in Figure S9. Overall, the modeled MAC and E_{abs} agree relatively well with the measurement by assuming ρ_{BC} = 1.5 g/cm^3. Therefore, ρ_{BC} = 1.5 g/cm^3 is assumed in this study."

Some E_{abs} values (eg. E_{abs}=5.4, see Figure 11 in the revised manuscript) are scarcely observed in the atmosphere. The most likely reason is the results for BC with thicker BrC coating is unavailable at ultraviolet wavelength. For sensitivity analysis, the BC volume fraction is independent on the BC size. Therefore, to gain E_{abs}=5.4, the BC volume fraction should be 5% for all BC. However, in the atmosphere, not all BC is thickly coated. Limited measurements were conducted at ultraviolet wavelength, therefore the measurements are not available for all circumstance. In fact, our calculated results are in general agreement with the measurements in the visible wavelengths. The E_{abs} can reach approximately 3.96 at λ =532*nm* when f_{BC}=5% in this work, which is consistent with the reported E_{abs} value of 2.6-4.0 at Beijing for λ =470*nm*, China (Xu et al., 2016).

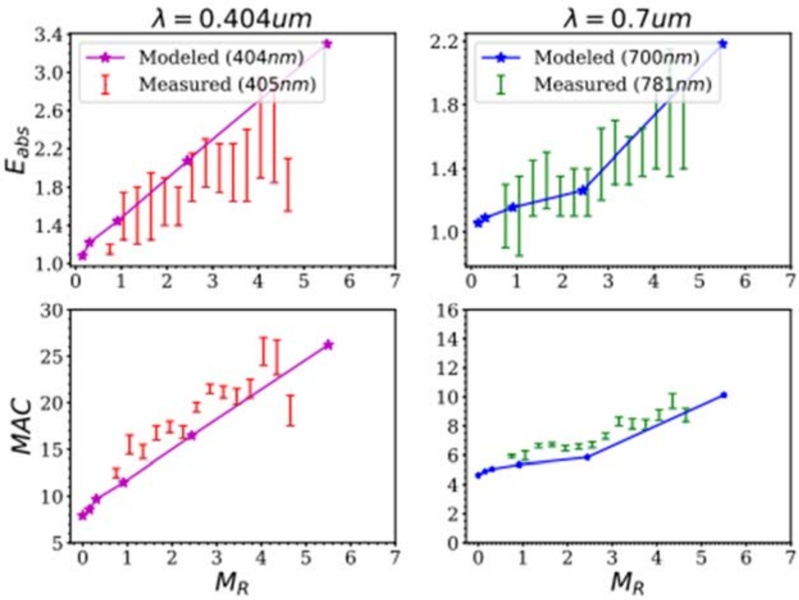


Figure 4 (Figure S8 in the revised manuscript) Comparison of modeled E_{abs} and MAC with measurements, BC mass density is assumed to be 1.8g/cm3, and the measured results are derived from the study of Liu et al. (2015).

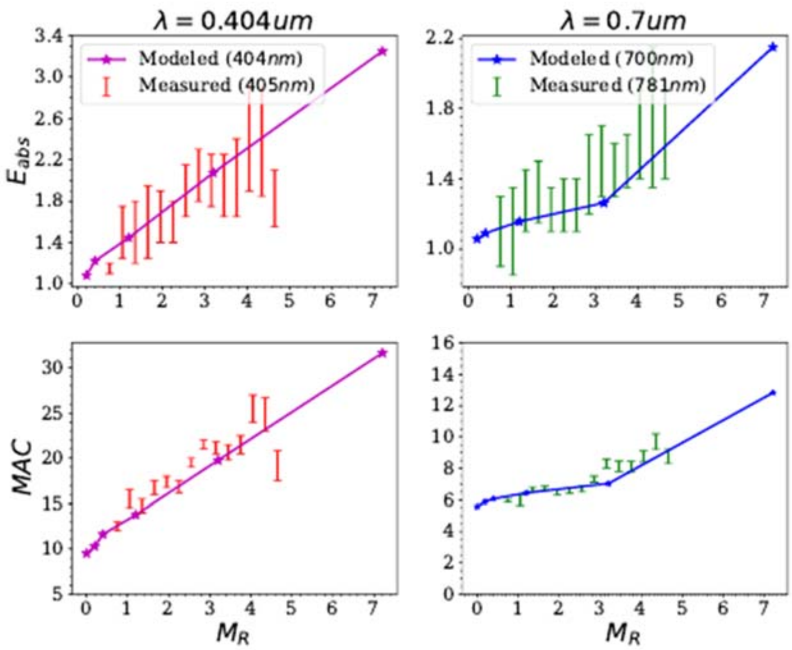


Figure 5 (Figure S9 in the revised manuscript) Comparison of modeled Eabs and MAC with measurements, BC mass density is assumed to be 1.5g/cm3, and the measured results are derived from the study of Liu et al. (2015).

Table 1 (Table 2 in the revised manuscript) MAC (m^2/g) for bare BC at different D_f (r_g = 0.06 um, σ_g = 1.5)

λ(nm)	D_f=1.8	D_f=2.2	D_f=2.6
350	9.30	9.03	8.48
404	8.14	7/95	7.60
532	6.20	6.11	6.02
700	4.68	4.64	4.65

4. In the abstract, please define the 'C_{abs}', 'K_{BrC}' before use them.

Response: Thanks for pointing it out. We have corrected it in the revised manuscript.

5. In Figure 6, 7 and 11, the range of color bar is suggested to be unified.

Response: Thanks for your suggestions. The range of color bar has been unified.

References

Adachi, K., Chung, S. H., and Buseck, P. R.: Shapes of soot aerosol particles and implications for their effects on climate, J Geophys Res-Atmos, 115, 2010.
Bergstrom, R. W.: Predictions of the spectral absorption and extinction coefficients of an urban air pollution aerosol model, Atmospheric Environment (1967), 6, 247 – 258, 1972.
Bond, T. C. and Bergstrom, R. W.: Light absorption by carbonaceous particles: An investigative review, Aerosol Science and Technology, 40, 27–67, 2006.
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.
Fuller, K. A., Malm, W. C., and Kreidenweis, S. M.: Effects of mixing on extinction by carbonaceous particles, Journal of Geophysical Research-Atmospheres, 104, 15 941–15 954, 1999a.
Horvath, H.: Atmospheric Light-Absorption - a Review, Atmospheric Environment Part a-General Topics, 27, 293–317, 1993.
Janzen, J.: Extinction of Light by Highly Nonspherical Strongly Absorbing Colloidal Particles - Spectrophotometric Determination of Volume Distributions for Carbon-Blacks, Applied Optics, 19, 2977–2985, 1980.
Kahnert, M.: On the Discrepancy between Modeled and Measured Mass Absorption Cross Sections of Light Absorbing Carbon Aerosols, Aerosol Science and Technology,

44, 453–460, <GotoISI>://WOS:000277436300006, 2010b.
Lewis, K. A., Arnott, W. P., Moosmuller, H., Chakrabarty, R. K., Carrico, C. M., Kreidenweis, S. M., Day, D. E., Malm, W. C., Laskin, A., Jimenez, J. L., Ulbrich, I. M., Huffman, J. A., Onasch, T. B., Trimborn, A., Liu, L., and Mishchenko, M. I.: Reduction in biomass burning aerosol light absorption upon humidification: roles of inorganically-induced hygroscopicity, particle collapse, and photoacoustic heat and mass transfer, Atmos Chem Phys, 9, 8949-8966, 2009.
Liu, D. T., Whitehead, J., Alfarra, M. R., Reyes-Villegas, E., Spracklen, D. V., Reddington, C. L., Kong, S. F., Williams, P. I., Ting, Y. C., Haslett, S., Taylor, J. W., Flynn, M. J., Morgan, W. T., McFiggans, G., Coe, H., and Allan, J. D.: Black-carbon absorption enhancement in the atmosphere determined by particle mixing state, Nature Geoscience, 10, 184–U132, 2017.
Liu, L. and Mishchenko, M. I.: Effects of aggregation on scattering and radiative properties of soot aerosols, Journal of Geophysical ResearchAtmospheres, 110, 2005.
Medalia, A. I. and Richards, L. W.: Tinting Strength of Carbon-Black, Journal of Colloid and Interface Science, 40, 233–&, 1972.
Nakao, S., Tang, P., Tang, X. C., Clark, C. H., Qi, L., Seo, E., Asa-Awuku, A., and Cocker, D.: Density and elemental ratios of secondary organic aerosol: Application of a density prediction method, Atmospheric Environment, 68, 273–277, 2013.
Wang, Y. Y., Liu, F. S., He, C. L., Bi, L., Cheng, T. H., Wang, Z. L., Zhang, H., Zhang, X. Y., Shi, Z. B., and Li, W. J.: Fractal Dimensions and Mixing Structures of Soot Particles during Atmospheric Processing, Environ Sci Tech Let, 4, 487-493, 2017.
Xu, X. Z., Zhao, W. X., Zhang, Q. L., Wang, S., Fang, B., Chen, W. D., Venables, D. S., Wang, X. F., Pu, W., Wang, X., Gao, X. M., and Zhang, W. J.: Optical properties of atmospheric fine particles near Beijing during the HOPE-J(3)A campaign, Atmos Chem Phys, 16, 6421-6439, 2016.

Response to the reviewer #3

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.

Effects of brown coatings on the absorption enhancement of black carbon: a numerical investigation

Jie Luo, Yongming Zhang, Feng Wang, and Qixing Zhang

State Key Laboratory of Fire Science, University of Science and Technology of China, Hefei, Anhui 230026, China

Correspondence: Qixing Zhang (qixing@ustc.edu.cn)

Abstract. Using the numerically exact multiple sphere T-matrix (MSTM) method, we explored the effects of brown coatings on absorption enhancement (E_{abs}) [..[1]]of black carbon (BC) at different wavelengths (λ). In addition, the ratio of the absorption of BC coated by brown carbon (BrC) to an external mixture of BrC and BC ($E_{abs\_internal}$) is also investigated. In this work, thinly-coated BC is defined as those with BC volume fraction over 20%, and other BC is considered to be thickly-coated. E_{abs} increases with the absorption of coatings, while an opposite trend is observed for [..[2]]$E_{abs\_internal}$. A much wider range of E_{abs} is observed for BC with brown coatings compared to that with non-absorbing coatings. E_{abs} can reach approximately 5.4 for BC with brown coatings at $\lambda = 0.35\ um$ [..[3]]under a typical size distribution. [..[4]]Previous studies have focused on the lensing effects of coatings but neglected the blocking effects of absorbing coatings. [..[5]]$E_{abs\_internal}$ can be below [..[6]]1 at ultraviolet spectral region for BC with brown coatings, which indicates that the absorption of [..[7]]internally mixed BC is less than that of an external mixture of BrC and BC due to the blocking effects of outer coatings, and we named the blocking effect of absorbing coatings "sunglasses effect". [..[8]]In addition, the applicability of core-shell sphere model are also evaluated for BC with brown coatings. Absorption cross-section (C_{abs}) of thickly-coated BC is underestimated by core-shell sphere model for all wavelengths while the underestimation becomes negligible as the imaginary part of the refractive index of brown carbon (k_{BrC}) turns very large. [..[9]]The lensing effect and the sunglass effect is clearly defined. Moreover, the effects of [..[10]]composition ratios, the size distribution are explored at different wavelengths. Our findings can improve the understanding of the absorption enhancement of BC with brown coatings.

---

[1]removed: and the lensing effects ($E_{abs\_lensing}$)

[2]removed: $E_{abs\_lensing}$

[3]removed: given

[4]removed: In addition, previous

[5]removed: $E_{abs\_lensing}$

[6]removed: 0.5

[7]removed: internal BC is diluted by approximately 0.5 times

[8]removed: Hence, it is imperative to take the blocking effects of outer coatings into consideration for estimation of total aerosol absorption.

[9]removed: E_{abs} and $E_{abs\_lensing}$ are insensitive to the size distribution for thinly-coated BC, and the uncertainties caused by the size distribution have differences of less than 2.6% and 6%, respectively. However

[10]removed: size distribution on E_{abs} and $E_{abs\_lensing}$ are rather obvious for thickly-coated BC. In addition, the uncertainties in E_{abs} and $E_{abs\_lensing}$ caused by D_f are less than 9%. E_{abs} seems to be essentially wavelength-independent for thickly-coated BC with non-absorbing coatings but rather wavelength-dependent for thickly-coated BC with

1 Introduction

Recent modeling and field studies have indicated that aerosol light absorption is an important contributor to climate forcing [..[11]](Jacobson, 2001; Krishnan and Ramanathan, 2002; Bond et al., 2013). Black carbon (BC) , which is a product of incomplete combustion, is the strongest solar-absorbing aerosol in the atmosphere (Lack et al., 2009; Zhang et al., 2008b). BC radiative forcing from fossil fuels and biomass burning have been estimated to be approximately $0.4W/m^2$, as the second [..[12]]contributor (after CO_2) to climate forcing due to their strong absorption of solar radiation (Forster et al., 2007; Schwarz et al., 2008). Sensitivity tests suggest that the mixing state and morphology of BC aerosols can largely affect the absorption of BC (Ma et al., 2012; Zhang et al., 2018). Due to the large uncertainties of BC morphologies and mixing states, the understanding of BC absorption is still limited. Even when coated with non-absorbing materials, the BC absorption can be enhanced (Liu et al., 2017; Cappa et al., 2012). Many studies mainly attribute the absorption enhancements (E_{abs}) to the lensing effect (Bond et al., 2006; Fuller et al., 1999b).

For the estimation of BC absorption enhancements, many field measurements have been conducted. Naoe et al. (2009) presented factors of 1.1-1.4 for BC absorption enhancement at a suburban site in Japan, while Cui et al. (2016) indicated that the absorption enhancement factors increase from 1.4 ± 0.3 during fresh combustions to ~ 3 for aged BC in a rural site over the North China Plain (NCP). Liu et al. (2017) found that BC absorption enhancement is significantly influenced by the particle mixing state. The measured range of E_{abs} is approximately $1 \sim 1.5$. You et al. (2016) observed the wavelength-dependent absorption enhancement of coated BC. In their measurements, E_{abs} increased up to 3 at the shortest measured wavelengths, while it was approximately 1.6 in the near-IR wavelength. A negligible absorption enhancement of only 6% for ambient BC particles was reported by Cappa et al. (2012) based on direct measurements over California (USA). Chen et al. (2017) reported an average E_{abs} of 2.07 ± 0.72 for the urban haze in winter in northern China. However, this result was time-dependent. The absorption enhancement of BC during the urban PM2.5 pollution was 1.31 ± 0.29 in the morning, while in the afternoon, it increased to approximately 2.23 ± 1.05; then, it decreased to 1.52 ± 0.75 in the evening. Recently, larger E_{abs} value of 2.6-4.0 at Beijing, China was reported by Xu et al. (2016). In summary, the reported E_{abs} values are not consistent in different studies due to the complex aging statuses.

Although the field measurements can provide referential absorption enhancement values for different aging statuses and regions, causes of these enhancements are not clear. For example, what is the main factor that causes the complex absorption enhancements: morphology, the mixing states or the types of coatings? To our best knowledge, field measurements have difficulty answering these questions currently. Numerical simulation is a strong tool that reveals the mechanism responsible for the complex absorption enhancements. To improve the understanding of the complex absorption enhancements of BC, numerical studies have also been conducted. For instance, based on the core-shell Mie theory, the absorption enhancement factors have been estimated up to 3 (Bond et al., 2006). By the numerically exact multiple sphere T-matrix (MSTM) method, Zhang et al. (2017) presented the absorption enhancements of non-absorbing coatings for aged BC ranging from 1.1-2.4, and they were significantly influenced by the morphology and aging statuses but insensitive to the BC refractive index. However,

[11] removed: (Jacobson, 2001; Krishnan and Ramanathan, 2002)
[12] removed: contribution

previous studies have failed to uncover the effects of coating absorption. In their studies, coatings were considered as non-absorbtive, and BC absorption enhancements were completely caused by lensing effects. Nevertheless, in the atmosphere, there is a type of organic carbon (OC) that absorbs the radiation in the range of the ultraviolet and visible spectra, which is known well as brown carbon (BrC); BC can also be mixed with BrC. Compared with non-absorbing [..[13]]materials, the absorption of BrC is significantly wavelength-dependent, and the imaginary part of the refractive index for BrC has a wide range (Kirchstetter et al., 2004), which results in large uncertainties for the estimation of aerosol absorption. Therefore, the absorption of BrC has gained increasing interests (Kirchstetter et al., 2004; Shamjad et al., 2018).

Many studies have been conducted to evaluate the absorption of BrC. One typical method for the determination of BrC absorption is isolating BrC by extracting filtered samples (Cheng et al., 2017). This method can be used to determine the imaginary part of the BrC refractive index. However, it is difficult to understand the effects of BrC on the total aerosol absorption, as BrC is commonly mixed with other chemical compositions. The assumption of externally mixing can be used to evaluate the absorption of BrC and BC separately. Nevertheless, in many cases, BC is internally mixed with other materials. It is widely accepted that the absorption is underestimated by the external mixing assumption when BC is coated with non-absorbing materials due to lensing effects. However, whether this is true for BC with BrC coatings is not clear. To understand the effects of BrC coatings, the contributions of "lensing effects" and the total absorption enhancement of BC with BrC coatings should be analyzed individually.

Cheng et al. (2017) has conducted a numerical investigation on BC absorption enhancement, BrC absorption enhancement and lensing effects on BC mixed BrC by assuming a core-shell structure. While the internal mixing of BC is widely accepted, the core-shell structure is debated (Adachi et al., 2010; Cappa et al., 2012; Bond et al., 2013). He et al. (2015) developed a theoretical BC aging model and concluded that the evolution of coating thickness, morphology, and composition during the aging process could have significant impacts on BC absorption. [..[14]]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 BC [..[15]]in this study) (Wang et al., 2017). As BC is 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) (Coz and Leck, 2011a; Zhang et al., 2008a).

In this study, a numerical investigation was conducted [..[16]]to explore the factors that contribute the complex absorption enhancement of BC with BrC coatings for different mixing states. Two types of mixing states were considered: thinly-coated BC and thickly-coated BC. Thinly-coated BC is assumed as those with BC volume fraction over 20%, and other BC is considered to be thickly-coated. The results would give further understanding for the causes of BC absorption enhancements and suggestion for the inferred BC mixing states.

---

[13] removed: OC

[14] removed: To investigate the lensing effects and total absorption enhancements of BrC coatings throughout the whole aging process, two types of mixing states were considered:

[15] removed: and

[16] removed: for the evaluation of the lensing effects and the

2 Methodology

2.1 Geometric properties of BC aerosols

In climate modeling, a spherical shape is commonly assumed for aerosols and can be calculated with high efficiency using the Mie theory (Mie, 1908). However, in many cases, this shape can introduce large errors compared with the measurements due to the oversimplification of the shape. Recently, the nonsphericity of aerosols has gained increasing interests (Yang et al., 2003; Bi and Yang, 2016). Specifically, observations have indicated that uncoated BC particles are commonly composed of numerous small spherical particles. Fractal aggregates can be greatly used to describe their geometric properties. Mathematically, the structure satisfies the well-known fractal law (Mishchenko et al., 2002):

$$n_s = k_0 \left(\frac{R_g}{R}\right)^{D_f} \tag{1}$$

$$R_g^2 = \frac{1}{n_s} \sum_{i=1}^{n_s} l_i^2 \tag{2}$$

where n_s represents the number of the monomers in the cluster, R represents the mean radius of the monomers, k_0 represents the fractal prefactor, D_f represents the fractal dimension, R_g represents the radius of gyration, and l_i represents the distance from the ith monomer to the center of the cluster.

The fractal dimension is a key parameter that describes the compactness of BC aggregates (Sorensen, 2001; Sorensen and Roberts, 1997; Luo et al., 2018a). Generally, aggregates tend to be more compact with the increase in D_f. A D_f of 1 can describe an open-chain-type shape, while the aggregates tend to be spherical as D_f approaches 3. Numerous experimental studies have been carried out to evaluate the D_f of BC aggregates. Immediately after they are emitted, BC aggregates generally exhibit fluffy structures with a small fractal dimension (D_f), that is normally less than 2, such as the D_f of BC aggregates from biomass burnings (1.67-1.83) (Chakrabarty et al., 2006), the D_f of BC from vehicle emissions (1.52-1.94) (China et al., 2014), and the D_f of BC from diesel combustion (1.6-1.9) (Wentzel et al., 2003).

However, under the effects of atmospheric aging, the structures and chemical compositions of BC may change. Aged BC tends to be mixed with other chemical components, and the shape becomes more compact. Therefore, in the atmosphere, aggregates can have fractal dimensions up to 2.6 (Chakrabarty et al., 2006). In some cases, BC aggregates are thinly coated with other materials, and still exhibit a fractal structure. However, different from freshly emitted BC aggregates, both lacy and compact structures can exist. Therefore, for thinly-coated BC, the D_f was assumed to be in the range from $1.8-2.6$. As BC becomes increasingly coated, BC aggregates may transform from highly agglomerated to nearly spherical particles. A $D_f = 2.6$ was assumed for thickly-coated BC. Even though a fractal prefactor can also vary under different combustion and aging statuses, it has less significant effects on the absorption of BC compared to the D_f. When fixing D_f to be 1.82, Liu and Mishchenko (2005) demonstrated that the absorption cross-section of BC aggregates does not change substantially as fractal prefactor varies from 0.9 to 2.1. Therefore, a fixed fractal prefactor of 1.2 was assumed in this work.

The monomer radius and monomers number are two key parameters that determine the particle size. Even though the monomers radii are polydispersed in the atmosphere, they vary within a narrow range. Monomer radii are commonly observed within $\sim$ 10-25 nm (Bond and Bergstrom, 2006). In addition, Kahnert (2010b) demonstrated that C_{abs} is insensitive to monomer radii when the monomer radii are within $\sim$ 10-25 nm. As a result, for convenient application, a fixed monomer radius of $R=$20 nm was assumed in this work. Based on TEM/SEM image, the monomer number n_s can reach approximately 800 (Adachi and Buseck, 2008). Values of $1 \leq n_s \leq 1000$ were considered in this work. For an aggregate with n_s monomers, the equivalent radius was given by the equivalent volume sphere radius $R\sqrt[3]{n_s}$. The morphological parameters considered in this work are shown in Table 1.

2.2 Generation of BC aerosols

The morphologies of coated BC considered in this work are classified into two categories: thinly-coated BC and thickly-coated BC. The closed-cell structure, which is an example of where coating material that not only covers the outer layers of BC aggregates but also fills the internal voids among primary spherules, can be used to represent the thinly-coated BC (Liou et al., 2011; Strawa et al., 1999). In addition, Kahnert (2017) demonstrated that the absorption of closed-cell structures and more realistic morphologies do not have large deviations. Therefore, it is reasonable to use the closed-cell model for calculating the absorption of thinly-coated BC, while the thickly-coated BC are commonly represented by a structure where BC aggregates are encapsulated in a sphere (Zhang et al., 2017; Cheng et al., 2014). The typical morphologies are shown in [..[17]]Figure 1.

Diffusion-limited algorithms (DLA), including the particle-cluster aggregation (PCA) (Hentschel, 1984) and the cluster-cluster aggregation (CCA) methods (Thouy and Jullien, 1994), have been developed for the generation of aggregates. However, adjustable DLA codes are commonly applied due to its quick implementation and adjustable fractal parameters (Koylu et al., 1995). In this work, an adjustable DLA code developed by Woźniak (2012) was used. Compared with ordinary DLA codes, this code preserves fractal parameters during each step of the aggregation, which avoids the generation of multifractal aggregates (Jensen et al., 2002). After the generation of the aggregates, the coatings were added. More specifically, for thinly-coated BC, the BrC shells were generated by the adjustable algorithm, and then the BC cores were added; the details are shown in previous studies (Luo et al., 2018c; Wu et al., 2014). The thickly-coated BC is generated by covering the BrC spherical coatings on the BC aggregates, as shown in the study of Cheng et al. (2015).

2.3 Light scattering method

To calculate the radiative properties of BC in this work, numerical solution methods from Maxwell's equations, including the finite-difference time-domain (FDTD) method (Yee, 1966; Taflove and Hagness, 2005), generalized multiparticle Mie (GMM) method (Xu, 1997; Xu and Gustafson, 2001), numerically exact multiple sphere T-matrix (MSTM) method (Mackowski and Mishchenko, 2011; Mishchenko et al., 2004), the geometric-optics surface-wave (GOS) method (Liou et al., 2011; He et al., 2016) and discrete-dipole approximation (DDA) method [..[18]](Draine and Flatau, 1994; Laczik, 1996; Yurkin and Hoekstra,

[17] removed: Fig.

[18] removed: (Draine and Flatau, 1994; Laczik, 1996; Smith and Stokes, 2006)

2007), can all be used. However, compared with other numerical methods, the MSTM has an advantage for the calculation of optical properties for randomly oriented particles analytically without numerically averaging over particle orientations. Therefore, this method has high efficiency to calculate optical properties of BC. In this work, the latest MSTM code, MSTM version 3.0 (Mackowski, 2013), was applied.

In this study, all the radiative properties of BC were calculated based on the assumption that BC particles and their mirror counterparts are present in equal numbers in ensemble of randomly oriented particles. In the atmosphere, it is reasonable to assume that the possibility of each particle direction is identical, which mathematically satisfies the definition of random orientation (Mishchenko and Yurkin, 2017).

2.4 Calculating absorption enhancement [..[19]]of BC

The presence of non-BC coated materials can result in the enhancement of BC absorption[..[20]], referred as BC absorption enhancement (E_{abs}). Therefore, E_{abs} can be defined as the amplification of BC absorption after BC being coated:

$$\mathrm{E_{abs}} = \frac{\mathrm{C_{abs\_coated}}}{\mathrm{C_{abs\_bare}}} \tag{3}$$

where C_{abs_coated} and [..[21]]C_{abs_bare} represent the absorption cross-sections of coated BC and bare BC respectively.

As BrC also absorbs solar radiation, it is also desirable to compare the absorption of BC coated by BrC coatings with BC and an external mixture of BrC and BC. The absorption of BrC shell is calculated as:

$$\mathrm{C_{abs\_BrC\_shell}} = \mathrm{C_{abs\_BrC(\ coated\ shape)}} - \mathrm{C_{abs\_BrC(\ bare\ shape)}} \tag{4}$$

where [..[22]]$C_{abs\_BrC(\ coated\ shape)}$ and [..[23]]$C_{abs\_BrC(\ bare\ shape)}$ represent the absorption [..[24]]cross-sections of BrC [..[25]]with morphologies that are identical to coated BC and bare BC, respectively. The calculation of the absorption of BrC shell is shown in Figure S1. In this process, we assume that the absorption of BrC with the same shape as the coated BC is identical to the external mixture of BrC that has the same shape as bare BC and BrC shell. We must clarify that this disposal method neglects the blocking effect and lensing effect of outer BrC shell on the internal BrC. However, as the BrC absorption is significantly less than the BC absorption with identical shape, the absorption caused by the blocking

[19] removed: and lensing effects

[20] removed: . However, it is difficult to distinguish how these enhancements are caused (i. e., by "lensing effect" or by the absorption of BrC). Based on Cheng et al. (2017), we attribute the BC absorption enhancements to two factors: (1) the effect of BrC absorption (E_{abs_BrC}); (2) the effects of lensing ($E_{abs\_lensing}$), which indicates that the shell can act as a lens that changes the absorption enhancement. It can be used to evaluate the relation between the absorption of a total particle and the sum of the coating and BC separately. When $E_{abs\_lensing} > 1$, the total absorption is enhanced by internally mixing, while the total absorption is weakened when $E_{abs\_lensing} < 1$. Here, $E_{abs\_lensing}$ characterizes the comprehensive outcome of lensing effects and blocking effects caused by coatings. To evaluate the effects of BrC coatings on the absorption enhancenment of BC

[21] removed: $E_{abs\_lensing}$ were calculated using:

[22] removed: $C_{abs\_\ whole\ particle}$

[23] removed: C_{abs_bare} represent the

[24] removed: cross-sections of coated BC and bare BC respectively. $C_{abs\_BrC(\ toal\ size)}$ and $C_{abs\_BrC(\ bare\ size)}$ represent the absorption

[25] removed: with morphologies that are identical for coated BC and bare BC, respectively.

effect and lensing effect of outer BrC on the internal BrC is relative small compared with the BC absorption. Therefore, it is reasonable to make some simplifications.

In this work, we defined an parameter ($E_{abs\_internal}$) to represent the ratio between the absorption of BC coated by BrC coatings and an external mixture of BrC and BC:

$$E_{abs\_internal} = \frac{C_{abs\_coated}}{C_{abs\_BrC\_shell} + C_{abs\_bare}} \quad (5)$$

2.5 Size distribution

The absorption of BC is significantly affected by the particle size (Kahnert, 2010b; Luo et al., 2018b). Therefore, the effects of the size distribution on BC absorption enhancement should be considered carefully. The shape of BC particles is commonly irregular. To describe the size of each BC particle, the radius of the corresponding equivalent volume sphere is typically used. Based on numerous measurements, a lognormal size distribution is observed to fit the realistic BC size distributions well (Bond et al., 2002; Chakrabarty et al., 2006; Wang et al., 2015), and it is widely used in climate models for the estimation of BC radiative forcing (Moffet and Prather, 2009; Chung et al., 2012). However, the mean size and standard deviation vary with the combustion status and aging status. In the atmosphere, geometric mean radii (r_g) between 0.05 um and 0.06 um for BC are widely accepted (Alexander et al., 2008; Coz and Leck, 2011b; Liu et al., 2018; Li et al., 2016). The geometric standard deviation (σ_g) varies within a relatively narrow range. Consequently, bare BC with [..[26]]r_g between 0.03 um and 0.1 um is considered for sensitivity analysis, an σ_g [..[27]]from 1.15-1.75. The minimum and maximum equivalent volume radii are $r_{min} = 0.02\ um$ and $r_{max} = 0.2\ um$, respectively.

To estimate the effects of coating thickness on the absorption properties of BC, we assumed that BrC coatings [..[28]]ratio are independent of BC size. The difference between the size distributions of bare BC and coated BC is attributed to the coatings thickness. The size distribution of bare and coated BC is shown in [..[29]]Figure S2. Even though the assumption does not completely agree with the real cases, it is reasonable to make some simplifications [..[30]]for the sensitivity analysis. Here, we must [..[31]]clarify that the size distribution parameters (r_g and σ_g) mentioned in this work are applied for the bare BC, and the overall effective volume radius of coated BC is equal to the sum of coatings thickness and radius of bare BC.

[26] removed: an
[27] removed: varies
[28] removed: are uniformly coated on the BC, and the mixing states are
[29] removed: Fig. **??**.
[30] removed: to
[31] removed: classify

2.6 Calculation of bulk radiative properties of BC

To make our work more consistent with real circumstance, bulk optical properties are considered. These properties are calculated by averaging over a certain particle size distribution. In application, the equivalent volume radii of BC are commonly assumed to follow a lognormal size distribution :

$$\mathrm{n(r)} = \frac{1}{\sqrt{2\pi}\mathrm{r}\ln(\sigma_\mathrm{g})}\exp\left[-\left(\frac{\ln(\mathrm{r})-\ln(\mathrm{r_g})}{\sqrt{2}\ln(\sigma_\mathrm{g})}\right)^2\right] \qquad (6)$$

where r_g and σ_g represent the geometric mean radius and geometric standard deviation, respectively. Given the size distribution, the bulk C_{abs} can be obtained by the following equation:

$$< \mathrm{C_{abs}} >= \int_{\mathrm{r_{min}}}^{\mathrm{r_{max}}} \mathrm{C_{abs}(r)n(r)dr} \qquad (7)$$

The bulk E_{abs} [..[32]]and $E_{abs\_internal}$ are calculated as those in equations 3-5. The only difference is that the absorption cross-section is now bulk absorption cross-section.

3 Results

3.1 Effects of the imaginary part of the BrC refractive index: lensing effect and sunglasses effect

The refractive index of BC is commonly assumed to be wavelength-independent over the visible and near-visible spectral regions, and the imaginary part $k_{BC} \approx 0.79$ (Moosmuller et al., 2009; Bond and Bergstrom, 2006). In addition, Zhang et al. (2017) has demonstrated that the uncertainties of the BC refractive index have little impact on the absorption enhancement of coated BC aggregates. Therefore, a typical refractive index $m = 1.95 + 0.79i$ of BC, was adopted in this study.

[..[33]]The real parts of the BrC refractive indices were assumed to have a constant value of 1.5 (Schnaiter et al., 2005), while the imaginary part of the refractive index (k_{BrC}) was significantly dependent on wavelength at shorter visible and ultraviolet (UV) wavelengths (Moosmuller et al., 2009; Andreae and Gelencser, 2006; Alexander et al., 2008). [..[34]]Figure 2 shows the effects of k_{BrC} on E_{abs} and [..[35]]$E_{abs\_internal}$, where f_{BC} represents the BC volume fraction. Large deviations in E_{abs} and [..[36]]$E_{abs\_internal}$ can be observed given different values of k_{BrC}. Generally, E_{abs} increases with k_{BrC}, while [..[37]]$E_{abs\_internal}$ decreases with increasing k_{BrC}. Therefore, it is desirable to evaluate the effects of absorbing coatings on BC absorption enhancement. Given identical k_{BrC} values, the absorption enhancements of thickly-coated BC [..[38]]increase with wavelength.

[32]removed: , E_{abs_BrC} and $E_{abs\_lengsing}$ are similar to
[33]removed: For BrC, the
[34]removed: Fig. **??**
[35]removed: $E_{abs\_lensing}$
[36]removed: $E_{abs\_lensing}$
[37]removed: $E_{abs\_lensing}$
[38]removed: increases

However, for BC that is internally mixed with BrC, wavelength-dependent absorption enhancements are measured to decrease with λ (You et al., 2016). This may be due to the wavelength-dependent k_{BrC}. For [..[39]]thickly-coated BC, $E_{abs\_internal}$ and E_{abs} decrease with wavelength, but they are not a strong function of λ for thinly-coated BC. In addition, compared with BC with non-absorbing coatings, E_{abs} for thinly-coated BC with absorbing coatings seems to be less wavelength-dependent, while E_{abs} for thickly-coated BC with absorbing materials is more sensitive to wavelength.

Many studies have noticed that the lensing effect can greatly enhance the absorption of BC. However, there is also an opposite effect, which is commonly neglected. As shown in [..[40]]Figure 2, as k_{BrC} increases, the value of [..[41]]$E_{abs\_internal}$ of thickly-coated BC can be below 1. This indicates that the absorption of BC internally mixed with BrC coatings may be less than the sum of the absorption of an external mixture of BrC coatings and BC when k_{BrC} is large. This phenomenon can be explained from physical insights. When the absorption of the coatings is weak, the light can penetrate the coatings into the BC materials, and the absorption of BC is significantly enhanced by the lensing effect. However, as the coating absorption increases, the light is blocked by the outer coatings. Therefore, the light can not fully and deeply penetrate the absorbing coatings on BC. As a result, the total absorption is less than the sum of the absorption of coatings and BC that are calculated separately. Therefore, there is a need to classify the [..[42]]BrC coating effect into lensing effect ($E_{abs\_lensing}$) and sunglasses effect ($E_{Sunglass}$), which represents the absorption enhancements and blocking effects of coatings, respectively.

Liu et al. (2017) defined the lensing effect as the absorption enhanced by addition of non-black carbon. However, from physical point, for BC with BrC coatings, the definition may be not clear as BrC also absorbs solar radiation, and it can be confused with E_{abs}. Therefore, here we redefine the lensing effect as the absorption enhanced by addition of non-absorbing materials. In addition, we assume that the lensing effect of BC with absorbing coatings is the same as those with non-absorbing coatings. Accordingly, $E_{abs\_lensing}$ can calulated using:

$$\mathsf{E_{abs\_lensing}} = \frac{\mathsf{C_{abs\_non-absorbing}}}{\mathsf{C_{abs\_bare}}} \tag{8}$$

where $C_{abs\_non-absorbing}$ represents the absorption cross-section of BC with non-absorbing coatings. The total E_{abs} should be contributed to the lensing effect, absorption of BrC shell and the sunglass effect. Therefore, E_{abs} can be expressed by:

$$\mathsf{E_{Sunglass}} = -\frac{\mathsf{C_{abs\_coated}} - \mathsf{C_{abs\_BrC\_shell}} - \mathsf{C_{abs\_non-absorbing}}}{\mathsf{C_{abs\_bare}}} \tag{9}$$

Combining Equations 3-9, we can obtain $E_{Sunglass}$, and the negative sign represents that the sunglass effect can cause the decrease of total absorption. According to definition of $E_{Sunglass}$, we can easily obtain the relation that the absorption of BC coated with BrC is less than that of an external mixture of BrC and BC when $E_{Sunglass} > E_{abs\_lensing} - 1$.

The sensitivity of $E_{Sunglass}$ to k_{BrC} is shown in Figure 2. For both thinly- and thickly-coated BC, $E_{Sunglass}$ increases with k_{BrC}. Fixing k_{BrC}, $E_{Sunglass}$ of thinly-coated BC decreases with wavelength. However, for thickly-coated BC,

[39]removed: aged BC, $E_{abs\_lensing}$
[40]removed: of Fig. **??**(d)
[41]removed: $E_{abs\_lensing}$
[42]removed: lensing effect into a convex lensing effect

$E_{Sunglass}$ can increase with wavelength at large k_{BrC} (such as $k_{BrC} = 0.16$). For the thinly-coated BC, the blocking of $E_{Sunglass}$ is less than the enhancement of $E_{abs\_lensing}$ (see Figure 5 and Figure 10), therefore, $E_{abs\_internal}$ of thinly-coated BC is larger than 1. For thickly-coated BC, the blocking of $E_{Sunglass}$ can be larger than the enhancement of $E_{abs\_lensing}$ as k_{BrC} is larger, which leads to $E_{abs\_internal}$ of less than 1. The threshold value of k_{BrC} is dependent on particle size and mixing states. Generally, the threshold k_{BrC} value decreases with particle size and coatings thickness, as $E_{abs\_internal}$ of BC thickly-coated with BrC coatings decreases with particle size and coatings thickness in the ultraviolet region (see Figure 6 and Figure 9).

Although core-shell sphere model has been debated for a long time, it is still widely used in climate models. By Combining the electron tomography (ET) and discrete dipole approximation (DDA) method, Adachi et al. (2010) found that the absorption of BC with fluffy structures is significantly enhanced by a core-shell structure at $\lambda = 0.55\ um$. However, for [..[43]]thickly-coated BC, BC absorption is underestimated at the UV, visible, and IR wavelengths (Kahnert et al., 2012). Mishchenko et al. (2014) has also demonstrated that the C_{abs} of thickly-coated BC with non-absorbing coatings is significantly underestimated by a core-shell sphere, and investigated the effects of off-center of BC. Their results indicated that the C_{abs} of aged BC covered with thick non-absorbing coatings are approximately 1.44 times higher than those calculated with a core-shell sphere model. Nevertheless, the effects of coating absorption on the applicability of the core-shell sphere model have not been evaluated. As shown in [..[44]]Figure 3, C_{abs} for thinly-coated BC is enhanced by a core-shell sphere structure in the visible spectral region, which agrees with the study of Adachi et al. (2010), while it is underestimated in the ultraviolet region. In addition, the ratio of C_{abs} of thinly-coated BC to core-shell sphere model increases with k_{BrC}. However, the applicability of the core-shell sphere model to thickly-coated BC is diverse. Consistent with Kahnert et al. (2012), thickly-coated BC absorption is underestimated by core-shell sphere model when coated with non-absorbing materials. Nevertheless, as k_{BrC} increases, the underestimation becomes insignificant. The reason may be that less light can penetrate deeply into the BC as the k_{BrC} increases, which leads to less variation in absorption. Therefore, the morphological effects of BC are relatively small.

The E_{abs}, compared with that for core-shell sphere model, is also calculated. For thinly-coated BC, the E_{abs} is significantly overestimated by core-shell sphere model. However, this overestimation is alleviated by an increasing k_{BrC}. For BC that is thickly-coated with non-absorbing materials, the E_{abs} is underestimated by core-shell sphere model at all wavelengths, while it decreases as k_{BrC} becomes larger. The E_{abs} can be overestimated by core-shell sphere model in the ultraviolet spectral region when k_{BrC} is large. Therefore, the absorption characteristics of BC are significantly affected by the absorption of coatings. To agree with the measurements, typical k_{BrC} values are assumed according to Kirchstetter et al. (2004), as shown in [..[45]]Figure S2. In this work, k_{BrC} values of 0.168, 0.114, 0.0354 and 0.001 were assumed for 4 typical wavelengths (λ =350 nm, 404 nm , 532 nm and 700 nm, respectively) via interpolation.

[43] removed: aged BCwith thick coatings

[44] removed: Fig. **??**

[45] removed: Fig. **??**.

3.2 Bulk radiative properties: effects of the size distribution

The sensitivity study conducted by Zhang et al. (2017) showed that the E_{abs} for aged BC was significantly affected by the size distribution. They reported different E_{abs} values of ∼1.7 - 2.4 and ∼ 2.0 - 2.7 for accumulated and coarse modes, respectively. By setting the fractal dimension to be 2.2 and f_{BC} to be 40%, the variations in BC absorption enhancements for different particle size distributions are shown in [..[46]]Figure 4 .Generally, weaker absorption enhancement can be observed [..[47]]by increasing λ from ultraviolet region to visible region, which is in agreement with the study of You et al. (2016). By defining the monomers radii, Kahnert (2010a) demonstrated that the absorption cross-section is significantly affected by the particle size, and the cubic fit can greatly describe the relations among equivalent volume radii for freshly emitted BC. However, for the absorption enhancement of thinly-coated BC, the effects of size distribution are not obvious. With variations in r_g and σ_g, the absorption enhancement changes at ranges of ∼1.563 - 1.603, ∼1.427 - 1.465, ∼1.2440 - 1.275 and ∼1.146 - 1.169 [..[48]]at $\lambda = 0.35\ um$, 0.404 um, 0.532 um and 0.7 um, respectively. The relative [..[49]]uncertainty in the absorption enhancements caused by the size distribution are 2.56%, 2.66%, 2.81% and 2.01%, respectively. The effects of the size distribution on the absorption enhancement of thinly-coated BC are similar at different wavelengths. Generally, E_{abs} has the largest value when both r_g and σ_g are extremely small or extremely large.

The absorption of BrC and BC are considered separately in most cases. To investigate the difference between the absorption of internally mixed BC and the total absorption of BrC and BC (calculated separately), [..[50]]$E_{abs\_internal}$ is also calculated. $E_{abs\_internal}$ of thinly-coated BC is greater in the visible region due to the insignificant sunglass effects. The senstivity of $E_{abs\_internal}$ is also not obvious to the size distribution. With the size distribution varying, $E_{abs\_internal}$ changes in the range of $\sim 1.055 - 1.099$, $\sim 1.081 - 1.112$, $\sim 1.132 - 1.147$ and $\sim 1.140 - 1.165$ for $\lambda = 0.35um, 0.404um, 0.532um$ and $0.7um$, respectively, and the relative uncertainties all are below 2%. In addition, $E_{abs\_lengsing}$ shares a similar dependence on size distribution as E_{abs} [..[51]]in the visible wavelengths. The reason is that the E_{abs} mainly derives from lensing effects due to the weak absorption of coatings. However, for ultraviolet wavelengths, there is a completely different pattern [..[52]]due to the sunglass effect.

As both the lensing effect and sunglass effect may affect the $E_{abs\_internal}$, $E_{abs\_lensing}$ [..[53]]and $E_{Sunglass}$ are also investigated, and the results are shown in Figure 5. Here $E_{abs\_lensing} - 1$ represents the E_{abs} enhancement caused by the lensing effect, and $E_{Sunglass}$ is the E_{abs} decrease caused by the sunglass effect. For thinly-coated BC, although both $E_{abs\_lensing}$ and $E_{Sunglass}$ decrease with increasing λ, compared with $E_{Sunglass}$, $E_{abs\_lensing}$ has less spectral-dependence. $E_{abs\_lensing} - 1$ is in the range of $\sim 0.205 - 0.283$, $\sim 0.186 - 0.251$, $\sim 0.163 - 0.2$ and $\sim 0.147 - 0.171$ for

---

[46] removed: Fig.**??**. Generally, increased

[47] removed: at shorter wavelengths

[48] removed: for

[49] removed: errors

[50] removed: $E_{abs\_lengsing}$

[51] removed: does for

[52] removed: . The largest

[53] removed: occurs when both r_g and σ_g are extremely small. Given the understanding of the values, the effects of size distribution are also not obvious. $E_{abs\_lengsing}$ changes in the ranges of ∼1.080 - 1.146, ∼1.107 - 1.148, ∼1.145 - 1.164 and ∼1.144 - 1.1652 for λ =0.35

$\lambda = 0.35\ um$, 0.404 um, 0.532 um and 0.7 um, respectively. [..[54]]However, $E_{Sunglss}$ can reach approximately 0.2 at $\lambda = 0.35\ um$, but is about 0 at $\lambda = 0.7\ um$. In addition, for thinly-coated BC, [..[55]]the enhancements of the lensing effect is stronger than the blocking of the sunglass effect. Therefore, $E_{abs\_internal}$ is above 1 [..[56]]for thinly-coated BC (as shown in Figure 2 and Figure 4).

Figure [..[57]]6 illustrates the effects of size distribution [..[58]]on E_{abs} and $E_{abs\_internal}$ of thickly-coated BC. Compared with thinly-coated BC, there is a different effect pattern for thickly-coated BC. For ultraviolet wavelengths (e.g., λ =0.35 um and 0.404 um), absorption enhancements decrease as r_g or σ_g increases. This indicates that as the particle becomes larger or the size distribution becomes wider, the absorption enhancements become weaker. However, for the visible wavelengths, the effects of the size distribution is quite complicated. The absorption enhancements are relatively small when both r_g and σ_g are extremely large or small. The peak value commonly occurs when σ_g is extremely small. Zhang et al. (2017) concluded that the E_{abs} of aged BC is more sensitive to the size distribution in the accumulation mode (where the σ_g is relatively small), while the E_{abs} of coarsely coated BC aggregates (i.e., with large σ_g) show little variation with r_g. This is precisely true for BC with weak absorbing coatings, as shown in the results for λ =0.7 um. However, for BC with absorbing coatings, E_{abs} is sensitive to the size distribution for both modes. When fixing the f_{BC} to be 6%[..[59]], as r_g and σ_g vary, the absorption enhancements change in the ranges of ~3.7 - 7.1, ~3.85 - 5.80, ~3.06 - 3.74 and ~1.63 - 2.59 for λ =0.35 um, 0.404 um, 0.532 um and 0.7 um, respectively, and the uncertainties in E_{abs} can reach up to 91.9%, 50.7%, 22.2% and 60.7%, respectively. [..[60]]

[54] removed: The relative errors of the different size distributions are less than 6.11%, 3.70%, 1.66% and 1.89%, respectively

[55] removed: $E_{abs\_lensing}$ is greater than

[56] removed: , and the values are in the range of $\sim$ 1.08 - 1.166. This indicates that the blocking effects of BrC coatings are weaker than the strengthening effects caused by lensing effects, as the coatings are thin

[57] removed: **??**

[58] removed: for

[59] removed: ($D_p/D_c = 2.5544$)

[60] removed: The calculated results are greater than the measured absorption enhancements of $\sim$ 1.05 - 3.5 in the visible region (Khalizov et al., 2009; Xue et al., 2009; Bond et al., 2013). Our calculations indicate that the absorption enhancements can reach approximately 3.7 for $\lambda = 0.532 um$. The reason is mainly due to three aspects: (1) all coatings were assumed to be BrC coatings in this work, and the absorption of coatings was obvious in the ultraviolet band and near the ultraviolet spectral regions. However, in the atmosphere, the coatings are not only BrC coatings but also inorganic material coatings (e.g., sulfates), which present weak absorption in the ultraviolet to near-infrared regions (Aouizerats et al., 2010). (2) The results in Fig. **??** are obtained by assuming that all the BC are thickly coated, while in the atmosphere, not all BC are thickly-coated. According to China et al. (2013b), approximately 50% of freshly emitted BC particles are thickly-coated. In many cases, BC is thinly-coated. Therefore, the absorption enhancement in this work is relatively higher than that of the measurements. (3) For the sensitivity analysis, the size distributions cover a broaden range in this work, while the real size distributions in the atmosphere are in a narrower range; therefore, the range of E_{asb} observed in the atmosphere is relatively narrow. Even though the calculated results do not completely reflect the real absorption enhancements, based on the sensitivity analysis, they can give the upper values of the absorption enhancements and improve the understanding on the effects of BrC coatings. In addition, they can provide theoretical explanations for the high absorption enhancements. For example, the measured absorption enhancements can reach approximately 3.5, while the calculated results based on non-absorbing coatings are commonly less than 3.0. To better correspond to the measurements, we discussed the influences of mixing states for a typical size distribution in the next section (see Fig. **??** and Fig. **??**).

[..[61]]$E_{abs\_internal}$ of thickly-coated [..[62]]BC is also significantly affected by the size distribution. With the r_g [..[63]]varying in the range of 0.03-0.1 um and σ_g [..[64]]varying in the range of 1.15-1.75, $E_{abs\_internal}$ varies in the range of 0.871-1.053, 0.891-1.121, 1.115-1.383, 1.615-2.442 for λ =0.35 um[..[65]], 0.404 um, 0.532 um and 0.7 um, respectively. In addition, [..[66]]

[..[67]]effects of the size distribution on $E_{abs\_internal}$ and E_{abs} [..[68]]are related to wavelength. $E_{abs\_internal}$ decreases with particle size (i.e, increasing r_g[..[69]]

[..[70]]) at ultraviolet wavelengths[..[71]], [..[72]]while it increases as the particles become [..[73]]larger at visible wavelengths (also see Figure S6). Based on physical insights, the reason may be due to two aspects. When the wavelength is [..[74]]in the ultraviolet region, the absorption of the coatings is large, therefore, the blocking effects of the coatings is obvious. Given identical f_{BC} values, the superficial area of the outer coating becomes lager as [..[75]]the particle size increases. As a result, the blocking effects of the outer coatings increases. Therefore, the [..[76]]$E_{abs\_internal}$ decreases. At visible wavelengths, the absorption of the coatings is negligible, and the light can penetrate deeply into BC. At that point, the main factor is the enhancement of the lensing [..[77]]effect, and the larger particles may cause a larger superficial area, which leads to [..[78]]

[61]removed: The $E_{abs\_lensing}$ value for

[62]removed: BC has the similar dependences on the size distribution compared to E_{abs}. $E_{abs\_lensing}$ changes substantially as

[63]removed: or

[64]removed: varies. As r_g or σ_g decreases, $E_{abs\_lensing}$ increases from 0.45 to 1.37 and from 0.51 to 1.61

[65]removed: and 0.404 um respectively. For visible wavelengths, the effects of size distribution are rather complicated. As r_g and σ_g vary, $E_{abs\_lensing}$ can change from 1.455 to 2.025 and from 1.63 to 2.52 for λ =0.532 um and 0.7 um, respectively. Variation in the size distribution can lead to $E_{abs\_lensing}$ differences of 204.4%, 215.7%, 39.2% and 56.4% for λ =0.35 um

[66]removed: for the ultraviolet wavelengths, $E_{abs\_lensing}$ can be less than 1. This phenomenon is caused by the large absorption of coatings, which blocks the light penetrating deeply in to the inner materials. As a result, the total absorption is less than the sum of the absorption of coatings and BC.

[67]removed: In climate models, the σ_g is commonly assumed to be a fixed value, and the BC size distribution is commonly assumed to be in accumulation mode (Liu et al., 2012). To further understand of the effects of size distribution, we fixed σ_g to be 1.5, and the variations in

[68]removed: and $E_{abs\_lensing}$ with different f_{BC} values at various r_g are shown in Figs. **??-??**. For thinly-coated BC, E_{abs} seems to be insensitive to r_g. As

[69]removed: changes, the variations of E_{abs} are below 2% for different f_{BC} and λ. However, the effects of r_g cannot be neglected for thickly-coated BC. Assuming f_{BC} is 0.06, as r_g decreases from 0.1 to 0.03, E_{abs} increases from 4.25 to 7.11 and from 4.44 to 6.28 for λ =0.35 um and λ =0.404 um, respectively, which leads to approximate 1.673 and 1.414 times increments, respectively. The effects of r_g are relatively small for visible wavelengths. However, it can still introduce a relative difference of approximately 23% for λ =0.7 um as r_g varies. In addition, contrary to thickly-coated BC with absorbing coatings, E_{abs} increases with r_g for BC with non-absorbing coatings, which is consistent with the results reported by Zhang et al. (2017).

[70]removed: $E_{abs\_lensing}$ is in the range of $\sim$1.03 - 1.27 for thinly-coated BC. Generally, the effects of r_g are more sensitive

[71]removed: and for BC with thicker coatings. By fixing f_{BC} to be 20%, as r_g increases from 0.03 to 0.1

[72]removed: $E_{abs\_lensing}$ decreases from 1.18 to 1.1 when λ =0.35 um, which results in an approximate 7.3% variation. However, the relative deviations are within 2% for λ =0.532 um and 0.7 um. Nevertheless, the effects of r_g are rather obvious for thickly-coated BC. The effects of r_g are related to wavelength. $E_{abs\_lensing}$ decreases with r_g at ultraviolet wavelengths,

[73]removed: large for visible wavelengths

[74]removed: at

[75]removed: r_g

[76]removed: $E_{abs\_lensing}$ decreases. For

[77]removed: effects

[78]removed: enhanced $E_{abs\_lensing}$.

[..[79]]the enhanced $E_{abs\_internal}$. E_{abs} shares similar dependences on the size distribution for different wavelengths. [..[80]]In addition, $E_{abs\_internal}$ can be less than 1. This means that the enhancement of the lensing effect is less than the blocking of the sunglass effect. In climate models, the [..[81]]σ_g is commonly assumed to be a fixed value , and the BC size distribution is commonly assumed to be in accumulation mode (Liu et al., 2012). The effects of the size distribution at fixed $\sigma_g = 1.5$ are supplemented in Figure S4-S7. [..[82]]

The effects of the size distribution on the lensing effect and sunglass effect of thickly-coated BC are shown in Figure 7. $E_{abs\_lensing}$ [..[83]]is in the range of 2.197-2.514, [..[84]]2.045-2.486, [..[85]]1.844-2.526, 1.6147-2.568 at $\lambda = 0.35$ um, 0.404 um, 0.532 um and 0.7 um, [..[86]]respectively. It seems that $E_{abs\_lensing}$ [..[87]]is more sensitive to the size distribution in the visible region compared with ultraviolet region. However, $E_{abs\_lensing}$ at different wavelengths does not deviate largely, and the uncertainty is within 25%. However, effects of the size distribution on $E_{Sunglass}$ largely depend on the wavelength. Fixing $f_{BC} = 6\%$, $E_{Sunglass}$ is in the range between 1.586-2.062 at $\lambda = 0.35$ um, while in the range between 0.001-0.027 at $\lambda = 0.7$ um. In addition, from Figure 7, we can also see that the enhancement of the lensing effect (represented by $E_{abs\_lensing} - 1$) is less than the blocking of sunglass effect in the ultraviolet region for thickly-coated BC, [..[88]]while the opposite phenomenon is observed in the visible region.

3.3 Bulk radiative properties: effects of the composition ratio

To make our calculation meaningful, we compare the calculated E_{abs} and mass absorption cross-section (MAC) with the measurements of Liu et al. (2015). The measurement results for E_{abs} and MAC are estimated from Figure 1 and supplementary Figure 2 of Liu et al. (2015). MAC is calculated using:

$$\mathrm{MAC} = \mathrm{C_{abs\_coated}}/\mathrm{m_{BC}} \tag{10}$$

$$\mathrm{m_{BC}} = \int_{\mathrm{r_{min}}}^{\mathrm{r_{max}}} \rho_{\mathrm{BC}} \frac{4}{3}\pi \mathrm{r}^3 \mathrm{n(r)dr} \tag{11}$$

[79]removed: Figure ?? also demonstrates that there are different dependences on f_{BC}

[80]removed: As the coatings become thicker (i. e., f_{BC} decreases), $E_{abs\_lensing}$ becomes weaker for λ =0.35 um and 0.404 um, while when λ =0.7 um, $E_{abs\_lensing}$ is enhanced as f_{BC} decreases. This phenomenon can also be explained from physical insights. When the wavelength is short, increased thickness of the coatings may lead to a greater blocking effect, which weakens the total absorption of

[81]removed: coatings and BC

[82]removed: However, for visible wavelengths, enhanced

[83]removed: can be obtained by increasing the coatings due to the negligible blocking effects of the coatings. In addition, $E_{abs\_lensing}$ increases with wavelength due to the decrease in coating absorption (see Fig. ??). When setting σ_g to be 1.5, for different r_g

[84]removed: $E_{abs\_lensing}$ is in the ranges of ∼0.42 - 1.1, ∼0.45 - 1.35

[85]removed: ∼1.41 - 1.95 and ∼1.75 - 2.5 for λ =0.35

[86]removed: respectively. For ultraviolet wavelengths, the

[87]removed: can be less than 1. This indicates that the blocking effects of absorbing coatings may be greater than the enhancements of the lensing effects. Therefore, when BC coated with BrC, we should not only focus on the enhancements of the lensing effects but also carefully consider the blocking effects of the coatings. In addition, for both thinly-coated and

[88]removed: $E_{abs\_lensing}$ is quiet sensitive to the composition ratio of the coatings.

where m_{BC} and ρ_{BC} represent the mass and mass density of BC. To agree with measurements, the coating thickness is determined by the mass ratio of BrC and BC components M_R. In this study, M_R is calculated by:

$$\mathsf{M_R} = \rho_{\mathsf{BC}}.\mathsf{f_{BC}}/(\rho_{\mathsf{BrC}}.(1-\mathsf{f_{BC}})) \tag{12}$$

where ρ_{BrC} represents the mass density of BrC. In this work, we assume ambient BC mainly composed of primary organic matter with a low degree of oxidation. Based on the study of Nakao et al. (2013), an OC mass density range of 1-1.2 g/cm^3 has been used by Liu et al. (2017). $\rho_{BrC} = 1.1\ g/cm^3$ is assumed in this work. For the BC mass density, the study of Horvath (1993) gives values of $\rho_{BC} = 0.625\ g/cm^3$ and $\rho_{BC} = 1.125\ g/cm^3$. However, Fuller et al. (1999a) pointed out that the values may be not representative for BC in the atmosphere. Medalia and Richards (1972) and Janzen (1980) suggested ρ_{BC} in the range of 1.8-1.9 g/cm^3, while Bergstrom (1972) found that the ρ_{BC} value of 1.9-2.1 g/cm^3. Bond and Bergstrom (2006) suggested to use a value of 1.8 g/cm^3. Figure S8 compares the computations with measurements by assuming $\rho_{BC} = 1.8\ g/cm^3$. We assume that E_{abs} and MAC at $\lambda = 0.7\ um$ do not deviate largely with those at $\lambda = 0.781\ um$. Modeled E_{abs} at $\lambda = 0.7\ um$ agrees well with the measurements. Although E_{abs} at $\lambda = 0.404\ um$ seems to be relatively higher than the measurements, it dose not deviate largely with the measurements. However, modeled MAC is a little smaller than the measured MAC. Similar results were obtained for bare BC ((Kahnert, 2010b), (Liu and Mishchenko, 2005)) . Therefore, $\rho_{BC} = 1.8\ g/cm^3$ may be a little high for estimation of MAC.

Bond and Bergstrom (2006) concluded that MAC value of $7.5 \pm 1.2\ m^2/g$ for bare BC can be assumed at $\lambda = 0.55um$ by reviewing 21 publications of MAC measurements. However, our calculated MAC of 6.02-6.2 m^2/g (see Table 2) at $\lambda = 0.532um$ lies below the range of MAC values suggested by Bond and Bergstrom (2006). Similar conclusions were drawn by Kahnert (2010b) and Liu and Mishchenko (2005). However, our calculated MAC agrees well with the calculated MAC of $6.0 \pm 0.1\ m^2/g$ by Kahnert (2010b) at $\lambda = 0.55\ um$. As MAC depends significantly on BC mass density, to agree with measurements, Liu and Mishchenko (2005) used $\rho_{BC} = 1.0\ g/cm^3$. However, as pointed by Kahnert (2010b), the measured MAC and modeled MAC were not at the same wavelength, therefore leading to too low retrieved ρ_{BC}. To raise the computed MAC values to the average observed value of MAC = $(7.5 \pm 1.2)\ m^2/g$, $\rho_{BC} = 1.3\text{-}1.4\ g/cm^3$ was suggested by Kahnert (2010b). However, this ρ_{BC} value is rather drastic smaller than the value suggested by Bond and Bergstrom (2006). Therefore, Kahnert (2010b) suggested to assume $\rho_{BC} = 1.5 \sim 1.7\ g/cm^3$ to raise the computational MAC results to the lower bound of the observations. By assuming $\rho_{BC} = 1.5\ g/cm^3$, the comparison of modeled MAC and E_{abs} with measurements is shown in Figure S9. Overall, the modeled MAC and E_{abs} agree relatively well with the measurement by assuming $\rho_{BC} = 1.5\ g/cm^3$. Therefore, $\rho_{BC} = 1.5\ g/cm^3$ is assumed in this study.

Figure [..[89]]8 compares the E_{abs} and [..[90]]$E_{abs\_internal}$ for thinly-coated BC with different fractal dimensions at different composition ratios. [..[91]]Following Liu et al. (2018), a r_g of 0.06 um and [..[92]]a σ of 1.5 are assumed to reflect the real size

[89]removed: **??**

[90]removed: $E_{abs\_lensing}$

[91]removed: D_p/D_c represents the shell-core ratio (i.e., the equivalent particle diameter divided by equivalent BC diameter). Followling Liu et al. (2018), an

[92]removed: an

distribution of BC. It is expected that as the coatings increase, E_{abs} becomes much stronger. With [..[93]]M_R varying from 0 to 2.93, E_{abs} variations of ∼1 - 2.5, ∼1 - 2.2, ∼1 - 1.6 and ∼1 - 1.285 are obtained for $\lambda = 0.35\ um$, 0.404 um, 0.532 um and 0.7 um, respectively. The E_{abs} for thinly-coated BC with weakly-absorbing materials (i.e., $\lambda = 0.7\ um$) is significantly lower than that for core-shell sphere, as reported by Zhang et al. (2017), where E_{abs} can reach approximately 1.5 when [..[94]]the shell-core ratio is 1.6 ($M_R = 2.2709$) at $\lambda = 0.55\ um$. Even though the results are gained at two different wavelengths, the E_{abs} for BC that is coated with weakly absorption coatings should not [..[95]]deviate substantially between $\lambda = 0.55\ um$ and $\lambda = 0.7\ um$ (see [..[96]]Figure 12). Therefore, the differences from the previous study are mainly caused by the BC shape, as demonstrated in [..[97]]Figure 3. When the relative contents of BC vary, substantial variations in [..[98]]$E_{abs\_internal}$ can also be observed. As [..[99]]M_R varying in the range of 0-2.93, $E_{abs\_internal}$ increases from 1 to [..[100]]1.07, 1 to [..[101]]1.1, 1 to [..[102]]1.22 and 1 to 1.285 for $\lambda = 0.35\ um$, 0.404 um, 0.532 um and 0.7 um, respectively[..[103]]. $E_{abs\_internal}$ of thinly-coated BC increases with M_R in the visible spectral region, while a little decrease in $E_{abs\_internal}$ can be observed in the ultraviolet region as M_R increases when M_R is larger than a value. This is mainly caused by the blocking of the sunglass effect.

At different wavelengths, the effects of D_f may vary. [..[104]]$E_{abs\_internal}$ increases with D_f [..[105]]in the visible wavelengths, as the more compact structure can lead to a greater lensing interaction. [..[106]]While in the ultraviolet region, as the structure becomes more compact, the interaction of absorbing coatings also increases; therefore, the blocking effects of outer coatings are greater. Therefore, the [..[107]]$E_{abs\_internal}$ can decrease with D_f when D_f is greater than a value.

Even though Cheng et al. (2014) and Luo et al. (2018b) showed that the effects of D_f on C_{abs} are not obvious for thinly-coated BC, for E_{abs} of thinly-coated BC, the sensitivity of D_f has not been investigated. To quantify the effects of D_f, the relative deviations between $D_f = 1.8$ and $D_f = 2.6$ are also calculated for thinly-coated BC. From [..[108]]Figure 8, we found that the differences in E_{abs} and [..[109]]$E_{abs\_internal}$ among different values of D_f are larger for thicker coatings. Therefore, to evaluate the maximum [..[110]]uncertainty, the f_{BC} is fixed to be 20%. As shown in [..[111]]Figure 9, the differences in E_{abs} and

[93]removed: D_p/D_c varying from 1.0 to 1.71
[94]removed: $D_p/D_c = 1.6$
[95]removed: diverge
[96]removed: Fig. **??**
[97]removed: Fig. **??**.
[98]removed: $E_{abs\_lensing}$
[99]removed: D_p/D_c varies from 1.0 to 1.71, $E_{abs\_lensing}$
[100]removed: 1.13
[101]removed: 1.18
[102]removed: 1.27
[103]removed: , and enhancements of approximately 13%, 18%, 27% and 28.5% can be obtained. For
[104]removed: $E_{abs\_lensing}$
[105]removed: at
[106]removed: For ultraviolet wavelengths
[107]removed: $E_{abs\_lensing}$
[108]removed: Fig. **??**
[109]removed: $E_{abs\_lensing}$
[110]removed: error
[111]removed: Fig. **??**, except for $\lambda = 0.35\ um$,

[..[112]]$E_{abs\_internal}$ between $D_f = 1.8$ and $D_f = 2.6$ are all below 5%. [..[113]]E_{abs} of BC thinly coated with non-absorbing coatings is more obviously affected by D_f. However, the relative deviations between $D_f = 1.8$ and $D_f = 2.6$ [..[114]]are not exceed 12% (as shown in Figure S10.).

To reveal the factors that comtributes to the complex $E_{abs\_internal}$, the effects of M_R on $E_{abs\_lensing}$ [..[115]]and $E_{Sunglass}$ of thinly-coated BC are investigated at different wavelengths. $E_{abs\_lensing}$ increases with M_R for all wavelengths. It seems that the sensitivity of $E_{abs\_lensing}$ to M_R are more obvious in ultraviolet region compared with visible region. Fixing $D_f = 2.2$, with M_R varying from 0 to 2.93, $E_{abs\_lensing}$ increases from 1 to 1.46, 1.4, 1.32, 1.25 for for λ =0.35 um, [..[116]]0.404 um, [..[117]]0.532 um and 0.7 um, respectively. In addition, more compact structure can result in stronger lensing interaction between monomers, so leads to an $E_{abs\_lensing}$ [..[118]]increase with D_f[..[119]]. Moreover, compared with visible region, the effects of D_f are more obvious at ultraviolet region. $E_{Sunglass}$ also increases with D_f, as more compact structure may lead to stronger blocking interaction between BC monomers. As expected, $E_{Sunglass}$ is stronger in the ultraviolet region, while tends to be 0 in the visible region. As M_R reaches 2.93, $E_{Sunglass}$ can reach approximately 0.46 at $\lambda = 0.35$ um, [..[120]]while $E_{Sunglass}$ is below 0.02 at $\lambda = 0.7$ um.

Figure [..[121]]11 demonstrates the absorption enhancements of thickly-coated BC at different wavelengths for different composition ratios. Similar to thinly-coated BC, E_{abs} increases with increasing [..[122]]M_R or decreasing λ. When setting $r_g = 0.06$ um and $\sigma_g = 1.5$, as [..[123]]M_R varies from 6.6 to 13.9, E_{abs} increases from 3.4 to 5.4 and 3.25 to 5.2 for $\lambda = 0.35$ um and $\lambda = 0.404$ um, respectively, [..[124]]while E_{abs} varies from 2.78 to 3.96 and 2.2 to 2.4 for $\lambda = 0.532$ um and 0.7 um, respectively. In addition, the E_{abs} seems to be more sensitive to the composition ratios [..[125]]in the ultraviolet wavelengths. This may be caused by the absorption of coatings, which can substantially enhance the total absorption. In addition, combined of E_{abs} values of thinly-coated and thickly-coated BC, E_{abs} range for BC with BrC coatings is much wider than that for BC with non-absorbing coatings (E_{abs} of ~1 - 2.4) (Zhang et al., 2017, 2018)

[112]removed: $E_{abs\_lensing}$

[113]removed: These errors are much less than that for thickly-coated BC , where the difference can reach approximately 20% for coarse mode BC (Zhang et al., 2017). In addition, the difference

[114]removed: seems to be larger for large particles. Although the differences in

[115]removed: can reach approximately 9% for $\lambda = 0.35$

[116]removed: when r_g is below 0.07

[117]removed: the relative deviation between $D_f = 1.8$ and $D_f = 2.6$ is within 5%. As a result , E_{abs} and

[118]removed: do not change substantially

[119]removed: when r_g is in the range of $0.05 \sim 0.06$

[120]removed: which is widely accepted for bare BC (Alexander et al., 2008; Coz and Leck, 2011b)

[121]removed: **??**

[122]removed: D_p/D_c

[123]removed: D_p/D_c varies from 2.15 to 2.71

[124]removed: which results in an approximate 1.6 times E_{abs} enhancements,

[125]removed: at

[..[126]]At visible wavelengths, [..[127]]the $E_{abs\_internal}$ is greater than 1 due to the small blocking effects of BrC. [..[128]]Defining r_g to be 0.06 um and σ_g to be 1.5, as M_R varies from 6.6 to 13.9, $E_{abs\_lensing}$ ranges from ∼[..[129]]1.222 - [..[130]]1.337 and ∼[..[131]]2.115 - [..[132]]2.357 for $\lambda = 0.532$ um and 0.7 um, respectively. This indicates that the total absorption of BC and BrC can be substantially enhanced by the lensing effects. However, for ultraviolet wavelengths, the [..[133]]$E_{abs\_internal}$ is less than 1. [..[134]]$E_{abs\_internal}$ is within ∼[..[135]]0.913 - [..[136]]0.924 and ∼[..[137]]0.956 - [..[138]]0.974 for $\lambda = 0.35$ um and $\lambda = 0.404$ um, respectively. This demonstrates the absorbing coatings can significantly block the light into BC. Therefore, the total absorption is less than the sum of BrC absorption and BC absorption. In recent studies, the enhancements of lensing effects has gained increasing attention. However, few studies have investigated the blocking effects of absorbing coatings. As a matter of fact, the blocking effect of absorbing coatings is also a significant factor that affects the total absorption, as the [..[139]]$E_{abs\_internal}$ can be below [..[140]]1. This indicates that the blocking effects of absorbing coatings may be greater than the enhancements of the lensing effects. Therefore, when BC coated with BrC, we should not only focus on the enhancements of the lensing effects but also carefully consider the blocking effects of the coatings.

There is a different dependence on M_R for $E_{abs\_internal}$ at different wavelengths. $E_{abs\_internal}$ increases with M_R at relative long wavelengths (eg. $\lambda =$0.7 um), while decreases as the coatings become thicker at relative short wavelengths (0.404 um and 0.532 um). This phenomenon can also be explained from physical insights. When the wavelength is short, increased thickness of the coatings may lead to a greater sunglass effect, which weakens the total absorption of the coatings and BC. However, at $\lambda = 0.7$ um, enhanced $E_{abs\_internal}$ can be obtained by increasing the coatings due to the negligible blocking effects of the coatings. In addition, $E_{abs\_internal}$ increases with wavelength due to the decrease in coating absorption (see Figure 2). $E_{abs\_internal}$ of [..[141]]thickly-coated BC is insensitive to M_R at $\lambda = 0.35$ due to the similar variations of $E_{abs\_lensing}$ and $E_{Sunglass}$ with M_R. As M_R varies from 6.6 to 13.9, E_{abs_lesnig} increases from 2.204 to 2.363, 2.214 to 2.390, 2.216 to 2.473, 2.165 to 2.509 at $\lambda = 0.35$ um, 0.404 um, 0.532 um, and 0.7 um, respectively. While $E_{Sunglass}$ largely affected by wavelengths. At $\lambda = 0.35$ um, $E_{Sunglass}$ is in the range from 1.523 to 1.807, while $E_{Sunglass}$ approaches 0 at $\lambda = 0.7$ um. It is also can be seen from Figure 11 that $E_{Sunglass} > E_{abs\_lensing} - 1$ at $\lambda = 0.35$ um and 0.404 um, therefore, $E_{abs\_internal}$ <1.

[126] removed: There is a different dependence on D_p/D_c for $E_{abs\_lensing}$. $E_{abs\_lensing}$ increases with D_p/D_c at

[127] removed: while decreases as the coatings become thicker at ultraviolet wavelengths. For visible wavelengths, the $E_{abs\_lensing}$

[128] removed: By setting

[129] removed: 1.75

[130] removed: 1.9

[131] removed: 2.05

[132] removed: 2.4

[133] removed: $E_{abs\_lensing}$

[134] removed: $E_{abs\_lensing}$

[135] removed: 0.65

[136] removed: 0.76

[137] removed: 0.88

[138] removed: 1.02

[139] removed: $E_{abs\_lensing}$

[140] removed: 0.5 (see Fig. **??**), which indicates that the absorption

[141] removed: BC can be reduced by 0.5 times. As a result, the blocking effect of absorbing coatingsshould also be taken into consideration in climate studies.

You et al. (2016) demonstrated that there are different wavelength dependencies for BC that is coated with absorbing and weak absorbing materials. E_{abs} for BC coated with humic acid was observed to vary from 3.0 to approximately 1.6 as λ increased from 0.554 um to 0.84 um, while it seemed to be essentially wavelength-independent for BC that is coated with sodium chloride. Figure [..[142]]12 compares the wavelength dependencies of BC coated with non-absorbing materials and BrC. For thinly-coated BC, there are substantial wavelength dependencies for BC coated with BrC. By setting f_{BC} to be 40%, E_{abs} increases from 1.15 to 1.57 with λ varying from 0.7 um to 0.35 um, which results in approximately 49.6% increase. However, when coated with non-absorbing materials, E_{abs} exhibits small wavelength-dependences. This leads to approximate 8.7% increases as λ decreases from 0.7 um to 0.35 um. Furthermore, for thickly-coated BC, E_{abs} is significantly wavelength-dependent for BC with BrC coatings. The decrease in λ from 0.7 um to 0.35 um would result in approximate 100% increase in E_{abs}, while E_{abs} seems to be essentially wavelength-independent for BC with non-absorbing coatings ($E_{abs\_lensing}$); it is approximately 2.4 when $f_{BC} = 6\%$, which is consistent with the value reported by Zhang et al. (2017). The differences of $E_{abs\_lensing}$ of thickly-coated BC between $\lambda = 0.35\ um$ and 0.7 um are below 6.2%. Therefore, the variation in k_{BrC} should be mainly responsible for the significant wavelength dependencies of E_{abs} for BC with BrC coatings when the wavelength is long. For ultraviolet wavelengths (λ from 0.35 [..[143]]um to 0.404 [..[144]]um), wavelength dependence of E_{abs} is relatively small, as the E_{abs} may increase with wavelength when k_{BrC} is fixed at a large value (see [..[145]]Figure 2), which can reduce the wavelength dependence. Therefore, the contribution of k_{BrC} to the wavelength dependence should be further analyzed [..[146]]in the ultraviolet wavelengths in the future.

In addition, the [..[147]]$E_{abs\_internal}$ of BC coated with BrC is also significantly wavelength-dependent. [..[148]]Fixing $f_{BC} = 40\%$ and 6%, respectively, with λ [..[149]]varying form 0.35 um to 0.7 um, $E_{abs\_internal}$ increases from 1.05 to 1.18 and from approximately 0.92 to 2.3, respectively. $E_{abs\_lensing} - 1$ and $E_{Sunglass}$ are also compared in Figure 12. [..[150]]$E_{Sunglass}$ decreases significantly with λ for both thinly- and thickly-coated BC[..[151]]. For thinly-coated BC, $E_{abs\_lensing} - 1$ is larger than $E_{Sunglass}$ for all wavelengths. However, $E_{Sunglass}$ can be stronger than $E_{abs\_lensing} - 1$ in ultraviolet region for thickly-coated BC. This indicates that the total absorption of BC and BrC is weakened by internal mixing. Therefore, the sunglass effect should also be noticed for the estimation of aerosol absorption.

[142] removed: **??**
[143] removed: um
[144] removed: um
[145] removed: Fig. **??**
[146] removed: at
[147] removed: $E_{abs\_lensing}$
[148] removed: There is an approximately 7.5%increase in $E_{abs\_lensing}$ as
[149] removed: increases from
[150] removed: For
[151] removed: , $E_{abs\_lensing}$ is more significantly affected by wavelength, and varies from approximately 0.6 to approximately 2.4 when $f_{soot} = 6\%$.

4 Summary and Discussion

Using MSTM method, the E_{abs} and $E_{abs\_lensing}$ of BC with BrC coatings were investigated at $\lambda = 0.35um, 0.404um, 0.532um$ and $0.7um$, respectively. The main findings of this work are as follows:

1. Generally, E_{abs} increases with k_{BrC} while [..[152]]$E_{abs\_interanl}$ decreases as k_{BrC} becomes larger. For the thinly-coated BC, [..[153]]$E_{abs\_internal}$ is greater that 1 due to the enhancements of the lensing effects. However, for thickly-coated BC, the [..[154]]$E_{abs\_internal}$ can be less than 1. It indicates the total absorption of BrC and BC is less than sum of BrC and BC absorption individually, which is opposite to BC that is coated with weakly-absorbing coatings. This phenomenon may be caused by the blocking effects of outer coatings. As the absorption of coatings increases, less light can penetrate into BC materials. Therefore, the total absorption of BrC and BC is weakened, resulting in [..[155]]$E_{abs\_internal}$ of less than 1. This effect is named "sunglasses effect" in this study.

2. C_{abs} of thinly-coated BC is underestimated by core-shell sphere model in the ultraviolet region while overestimated in the visible region. In addition, the ratio of C_{abs} of thinly-coated BC to that of core-shell sphere model increases with k_{BrC}. E_{abs} of thinly-coated BC is enhanced by core-shell sphere while the enhancements are alleviated by increasing k_{BrC}. There are different dependencies for thickly-coated BC. C_{abs} of thickly-coated BC is underestimated by core-shell sphere model for all wavelengths while the underestimation becomes negligible as k_{BrC} turns very large. E_{abs} of thickly-coated BC with non-absorbing materials is underestimated by core-shell assumption. However, the ratio of E_{abs} of thickly-coated BC to core-shell sphere model decreases with increasing k_{BrC}, and E_{abs} is enhanced by core-shell sphere in the visible region, when the absorption of coatings is large.

3. To make our calculation more consistent with real circumstance, the bulk absorption was calculated and the k_{BrC} is selected by interpolation based on the study of Kirchstetter et al. (2004). For thinly-coated BC, the effects of size distribution on E_{abs} are not obvious. The uncertainties of size distribution result in E_{abs} differences of less than 2.56%, 2.52%, 2.32% and 2.16% for $\lambda = 0.35\ um$, 0.404 um, 0.532 um and 0.7 um, respectively. However, E_{abs} of thickly-coated BC is quite sensitive to the size distribution. E_{abs} differences of approximately 92% can be obtained as r_g and σ_g vary for $\lambda = 0.35um$. In addition, different from E_{abs} of $2.2 \sim 2.4$ for thickly-coated BC with weak absorbing coatings, E_{abs} of $3.4 \sim 5.4$ is observed for BC with BrC coatings at $\lambda = 0.35um$. For thinly coated BC, E_{abs} of BC with weak absorbing coatings is in the range of approximately $\sim$1 - 1.3 for $\lambda = 0.7\ um$ (i.e. BC with weakly-absorbing coatings) while a wider range of $\sim$1 - 2.5 is obtained for $\lambda = 0.35\ um$. In sumarry, E_{abs} range of BC with BrC coatings is much wider than that of BC with non-absorbing coatings.

4. The suglass effect and lensing effect are compared at different wavelengths. $E_{sunglass}$ is less than $E_{abs\_lensing} - 1$ for thinly-coated BC. This indicates the blocking of the sunglass effect is less than the enhancement of the lensing

[152]removed: $E_{abs\_lensing}$
[153]removed: $E_{abs\_lensing}$
[154]removed: $E_{abs\_lensing}$
[155]removed: $E_{abs\_lensing}$

effect, so the $E_{internal} > 1$ for thinly-coated BC. However, $E_{sunglass}$ can be larger than $E_{abs\_lensing} - 1$ in ultraviolet region for thickly-coated BC, which leads to $E_{internal} < 1$. Therefore, the absorption of BC thickly-coated with BrC can less than an external mixture of BC and BrC. In visible region, $E_{sunglass}$ is less than $E_{abs\_lensing} - 1$ due to the small sunglass effect.

5. E_{abs} of BC with BrC coatings is more wavelength-dependent than those with non-absorbing coatings. For thinly coated BC, E_{abs} of BC with non-absorbing coatings leads to approximately 8.7% increase as λ decreases from 0.7 um to 0.35 um while the difference can reach approximately 50% for BC with BrC coatings. For thickly coated BC, the decrease of λ from 0.7 um to 0.35 um would result in approximately 100% increase of E_{abs} for BC with BrC coatings. However, E_{abs} of BC with non-absorbing coatings seems to be to be essentially wavelength-independent. In addition, for thinly coated BC, the effects of D_f are not obvious for E_{abs} and $E_{abs\_lensing}$. The uncertainties of [..[156]]E_{abs} and [..[157]]$E_{abs\_internal}$ caused by D_f all are less than 5%.

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.

5 Acknowledgments

This work was financially supported by the National Key Research and Development Plan (Grant No. 2016YFC0800100 and 2017YFC0805100); National Natural Science Foundation of China (Grant No. 41675024 and U1733126); Fundamental Research Funds for the Central Universities (Grant No. WK2320000035). We particularly thank Dr. D. W. Mackowski and Dr. M. I. Mishchenko for the MSTM code. We also acknowledge the support of supercomputing center of USTC. We particularly thank the constructive suggestions of the three anonymous reviewers.

[156]removed: D_f lead to

[157]removed: $E_{abs\_lensing}$ differences of less than 7.4%and 9%, respectively.

References

Adachi, K. and Buseck, P. R.: Internally mixed soot, sulfates, and organic matter in aerosol particles from Mexico City, Atmospheric Chemistry and Physics, 8, 6469–6481, <GotoISI>://WOS:000260927800011, 2008.

Adachi, K., Chung, S. H., and Buseck, P. R.: Shapes of soot aerosol particles and implications for their effects on climate, Journal of Geophysical Research: Atmospheres, 115, 2010.

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, <GotoISI>://WOS:000258261000045, 2008.

Andreae, M. O. and Gelencser, A.: Black carbon or brown carbon? The nature of light-absorbing carbonaceous aerosols, Atmospheric Chemistry and Physics, 6, 3131–3148, <GotoISI>://WOS:000239346600001, 2006.

Aouizerats, B., Thouron, O., Tulet, P., Mallet, M., Gomes, L., and Henzing, J. S.: Development of an online radiative module for the computation of aerosol optical properties in 3-D atmospheric models: validation during the EUCAARI campaign, Geoscientific Model Development, 3, 553–564, <GotoISI>://WOS:000285965100012, 2010.

Bergstrom, R. W.: Predictions of the spectral absorption and extinction coefficients of an urban air pollution aerosol model, Atmospheric Environment (1967), 6, 247 – 258, https://doi.org/https://doi.org/10.1016/0004-6981(72)90083-2, http://www.sciencedirect.com/science/article/pii/0004698172900832, 1972.

Bi, L. and Yang, P.: Tunneling effects in electromagnetic wave scattering by nonspherical particles: A comparison of the Debye series and physical-geometric optics approximations, Journal of Quantitative Spectroscopy and Radiative Transfer, 178, 93–107, <GotoISI>://WOS:000376705900009, 2016.

Bond, T. C. and Bergstrom, R. W.: Light absorption by carbonaceous particles: An investigative review, Aerosol Science and Technology, 40, 27–67, <GotoISI>://WOS:000233906000001, 2006.

Bond, T. C., Covert, D. S., Kramlich, J. C., Larson, T. V., and Charlson, R. J.: Primary particle emissions from residential coal burning: Optical properties and size distributions, Journal of Geophysical Research-Atmospheres, 107, <GotoISI>://WOS:000180485800015, 2002.

Bond, T. C., Habib, G., and Bergstrom, R. W.: Limitations in the enhancement of visible light absorption due to mixing state, Journal of Geophysical Research-Atmospheres, 111, <GotoISI>://WOS:000241866200010, 2006.

Bond, T. C., Doherty, S. J., Fahey, D. W., Forster, P. M., Berntsen, T., DeAngelo, B. J., Flanner, M. G., Ghan, S., Karcher, B., Koch, D., Kinne, S., Kondo, Y., Quinn, P. K., Sarofim, M. C., Schultz, M. G., Schulz, M., Venkataraman, C., Zhang, H., Zhang, S., Bellouin, N., Guttikunda, S. K., Hopke, P. K., Jacobson, M. Z., Kaiser, J. W., Klimont, Z., Lohmann, U., Schwarz, J. P., Shindell, D., Storelvmo, T., Warren, S. G., and Zender, C. S.: Bounding the role of black carbon in the climate system: A scientific assessment, Journal of Geophysical Research-Atmospheres, 118, 5380–5552, <GotoISI>://WOS:000325212600025, 2013.

Cappa, C. D., Onasch, T. B., Massoli, P., Worsnop, D. R., Bates, T. S., Cross, E. S., Davidovits, P., Hakala, J., Hayden, K. L., Jobson, B. T., Kolesar, K. R., Lack, D. A., Lerner, B. M., Li, S. M., Mellon, D., Nuaaman, I., Olfert, J. S., Petaja, T., Quinn, P. K., Song, C., Subramanian, R., Williams, E. J., and Zaveri, R. A.: Radiative Absorption Enhancements Due to the Mixing State of Atmospheric Black Carbon, Science, 337, 1078–1081, <GotoISI>://WOS:000308125800042, 2012.

Chakrabarty, R. K., Moosmuller, H., Garro, M. A., Arnott, W. P., Walker, J., Susott, R. A., Babbitt, R. E., Wold, C. E., Lincoln, E. N., and Hao, W. M.: Emissions from the laboratory combustion of wildland fuels: Particle morphology and size, Journal of Geophysical Research-Atmospheres, 111, <GotoISI>://WOS:000236880400003, 2006.

Chen, B., Bai, Z., Cui, X. J., Chen, J. M., Andersson, A., and Gustafsson, O.: Light absorption enhancement of black carbon from urban haze in Northern China winter, Environmental Pollution, 221, 418–426, <GotoISI>://WOS:000392767900044, 2017.

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, Optics Express, 22, 15 904–15 917, <GotoISI>://WOS:000338055900072, 2014.

Cheng, T. H., Wu, Y., Gu, X. F., and Chen, H.: Effects of mixing states on the multiple-scattering properties of soot aerosols, Optics Express, 23, 10 808–10 821, <GotoISI>://WOS:000353299300126, 2015.

Cheng, Y., He, K. B., Engling, G., Weber, R., Liu, J. M., Du, Z. Y., and Dong, S. P.: Brown and black carbon in Beijing aerosol: Implications for the effects of brown coating on light absorption by black carbon, Science of the Total Environment, 599, 1047–1055, <GotoISI>://WOS:000405253500001, 2017.

China, S., Mazzoleni, C., Gorkowski, K., Aiken, A. C., and Dubey, M. K.: Morphology and mixing state of individual freshly emitted wildfire carbonaceous particles, Nature Communications, 4, <GotoISI>://WOS:000323715900002, 2013a.

China, S., Mazzoleni, C., Gorkowski, K., Aiken, A. C., and Dubey, M. K.: Morphology and mixing state of individual freshly emitted wildfire carbonaceous particles, Nature Communications, 4, <GotoISI>://WOS:000323715900002, 2013b.

China, S., Salvadori, N., and Mazzoleni, C.: Effect of Traffic and Driving Characteristics on Morphology of Atmospheric Soot Particles at Freeway On-Ramps, Environmental Science and Technology, 48, 3128–3135, <GotoISI>://WOS:000333776000007, 2014.

Chung, C. E., Ramanathan, V., and Decremer, D.: Observationally constrained estimates of carbonaceous aerosol radiative forcing, Proceedings of the National Academy of Sciences of the United States of America, 109, 11 624–11 629, <GotoISI>://WOS:000306837100034, 2012.

Coz, E. and Leck, C.: Morphology and state of mixture of atmospheric soot aggregates during the winter season over Southern Asia-a quantitative approach, Tellus Series B-Chemical and Physical Meteorology, 63, 107–116, <GotoISI>://WOS:000286001900009, 2011a.

Coz, E. and Leck, C.: Morphology and state of mixture of atmospheric soot aggregates during the winter season over Southern Asia-a quantitative approach, Tellus Series B-Chemical and Physical Meteorology, 63, 107–116, <GotoISI>://WOS:000286001900009, 2011b.

Cui, X. J., Wang, X. F., Yang, L. X., Chen, B., Chen, J. M., Andersson, A., and Gustafsson, O.: Radiative absorption enhancement from coatings on black carbon aerosols, Science of the Total Environment, 551, 51–56, <GotoISI>://WOS:000372589800007, 2016.

Draine, B. T. and Flatau, P. J.: Discrete-Dipole Approximation for Scattering Calculations, Journal of the Optical Society of America a-Optics Image Science and Vision, 11, 1491–1499, <GotoISI>://WOS:A1994NE24300032, 1994.

Forster, P., Ramaswamy, V., Artaxo, P., Berntsen, T., Betts, R., Fahey, D. W., Haywood, J., Lean, J., Lowe, D. C., Myhre, G., et al.: Changes in atmospheric constituents and in radiative forcing. Chapter 2, in: Climate Change 2007. The Physical Science Basis, 2007.

Fuller, K. A., Malm, W. C., and Kreidenweis, S. M.: Effects of mixing on extinction by carbonaceous particles, Journal of Geophysical Research-Atmospheres, 104, 15 941–15 954, <GotoISI>://WOS:000081535000004, 1999a.

Fuller, K. A., Malm, W. C., and Kreidenweis, S. M.: Effects of mixing on extinction by carbonaceous particles, Journal of Geophysical Research-Atmospheres, 104, 15 941–15 954, <GotoISI>://WOS:000081535000004, 1999b.

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, Atmospheric Chemistry and Physics, 15, 11 967–11 980, <GotoISI>://WOS:000364316800029, 2015.

He, C. L., Takano, Y., Liou, K. N., Yang, P., Li, Q. B., and Mackowski, D. W.: Intercomparison of the GOS approach, superposition T-matrix method, and laboratory measurements for black carbon optical properties during aging, Journal of Quantitative Spectroscopy and Radiative Transfer, 184, 287–296, <GotoISI>://WOS:000386982300027, 2016.

Hentschel, H. G. E.: Fractal Dimension of Generalized Diffusion-Limited Aggregates, Physical Review Letters, 52, 212–215, <GotoISI>://WOS:A1984RY26700013, 1984.

Horvath, H.: Atmospheric Light-Absorption - a Review, Atmospheric Environment Part a-General Topics, 27, 293–317, <GotoISI>://WOS:A1993KQ75200002, 1993.

Jacobson, M. Z.: Strong radiative heating due to the mixing state of black carbon in atmospheric aerosols, Nature, 409, 695–697, <GotoISI>://WOS:000166816400037, 2001.

Janzen, J.: Extinction of Light by Highly Nonspherical Strongly Absorbing Colloidal Particles - Spectrophotometric Determination of Volume Distributions for Carbon-Blacks, Applied Optics, 19, 2977–2985, <GotoISI>://WOS:A1980KF80000023, 1980.

Jensen, M. H., Levermann, A., Mathiesen, J., and Procaccia, I.: Multifractal structure of the harmonic measure of diffusion-limited aggregates, Physical Review E, 65, <GotoISI>://WOS:000175146500028, 2002.

Kahnert, M.: Modelling the optical and radiative properties of freshly emitted light absorbing carbon within an atmospheric chemical transport model, Atmospheric Chemistry and Physics, 10, 1403–1416, <GotoISI>://WOS:000274410000036, 2010a.

Kahnert, M.: On the Discrepancy between Modeled and Measured Mass Absorption Cross Sections of Light Absorbing Carbon Aerosols, Aerosol Science and Technology, 44, 453–460, <GotoISI>://WOS:000277436300006, 2010b.

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, Optics Express, 25, 24 579–24 593, <GotoISI>://WOS:000412048500107, 2017.

Kahnert, M., Nousiainen, T., Lindqvist, H., and Ebert, M.: Optical properties of light absorbing carbon aggregates mixed with sulfate: assessment of different model geometries for climate forcing calculations, Optics Express, 20, <GotoISI>://WOS:000303989300074, 2012.

Khalizov, A. F., Xue, H. X., Wang, L., Zheng, J., and Zhang, R. Y.: Enhanced Light Absorption and Scattering by Carbon Soot Aerosol Internally Mixed with Sulfuric Acid, Journal of Physical Chemistry A, 113, 1066–1074, <GotoISI>://WOS:000263134900017, 2009.

Kirchstetter, T. W., Novakov, T., and Hobbs, P. V.: Evidence that the spectral dependence of light absorption by aerosols is affected by organic carbon, Journal of Geophysical Research-Atmospheres, 109, <GotoISI>://WOS:000225190500010, 2004.

Koylu, U. O., Faeth, G. M., Farias, T. L., and Carvalho, M. G.: Fractal and Projected Structure Properties of Soot Aggregates, Combustion and Flame, 100, 621–633, <GotoISI>://WOS:A1995QP52600009, 1995.

Krishnan, R. and Ramanathan, V.: Evidence of surface cooling from absorbing aerosols, Geophysical Research Letters, 29, <GotoISI>://WOS:000178888000023, 2002.

Lack, D. A., Cappa, C. D., Cross, E. S., Massoli, P., Ahern, A. T., Davidovits, P., and Onasch, T. B.: Absorption Enhancement of Coated Absorbing Aerosols: Validation of the Photo-Acoustic Technique for Measuring the Enhancement, Aerosol Science and Technology, 43, 1006–1012, <GotoISI>://WOS:000269731800002, 2009.

Laczik, Z.: Discrete-dipole-approximation-based light-scattering calculations for particles with a real refractive index smaller than unity, Applied Optics, 35, 3736–3745, <GotoISI>://WOS:A1996UW46000056, 1996.

Li, J., Liu, C., Yin, Y., and Kumar, K. R.: Numerical investigation on the Ångström Exponent of black carbon aerosol, Journal of Geophysical Research-Atmospheres, 121, 3506–3518, <GotoISI>://WOS:000375120200027, 2016.

Liou, K. N., Takano, Y., and Yang, P.: Light absorption and scattering by aggregates: Application to black carbon and snow grains, Journal of Quantitative Spectroscopy and Radiative Transfer, 112, 1581–1594, <GotoISI>://WOS:000291714600009, 2011.

Liu, C., Chung, C. E., Yin, Y., and Schnaiter, M.: The absorption Ångström exponent of black carbon: from numerical aspects, Atmospheric Chemistry and Physics, 18, 6259–6273, https://doi.org/10.5194/acp-18-6259-2018, https://www.atmos-chem-phys.net/18/6259/2018/, 2018.

Liu, D. T., Whitehead, J., Alfarra, M. R., Reyes-Villegas, E., Spracklen, D. V., Reddington, C. L., Kong, S. F., Williams, P. I., Ting, Y. C., Haslett, S., Taylor, J. W., Flynn, M. J., Morgan, W. T., McFiggans, G., Coe, H., and Allan, J. D.: Black-carbon absorption enhancement in the atmosphere determined by particle mixing state, Nature Geoscience, 10, 184–U132, <GotoISI>://WOS:000395791400009, 2017.

Liu, L. and Mishchenko, M. I.: Effects of aggregation on scattering and radiative properties of soot aerosols, Journal of Geophysical Research-Atmospheres, 110, <GotoISI>://WOS:000229988800009, 2005.

Liu, S., Aiken, A. C., Gorkowski, K., Dubey, M. K., Cappa, C. D., Williams, L. R., Herndon, S. C., Massoli, P., Fortner, E. C., Chhabra, P. S., Brooks, W. A., Onasch, T. B., Jayne, J. T., Worsnop, D. R., China, S., Sharma, N., Mazzoleni, C., Xu, L., Ng, N. L., Liu, D., Allan, J. D., Lee, J. D., Fleming, Z. L., Mohr, C., Zotter, P., Szidat, S., and Prevot, A. S. H.: Enhanced light absorption by mixed source black and brown carbon particles in UK winter, Nature Communications, 6, <GotoISI>://WOS:000363141700001, 2015.

Liu, X., Easter, R. C., Ghan, S. J., Zaveri, R., Rasch, P., Shi, X., Lamarque, J. F., Gettelman, A., Morrison, H., Vitt, F., Conley, A., Park, S., Neale, R., Hannay, C., Ekman, A. M. L., Hess, P., Mahowald, N., Collins, W., Iacono, M. J., Bretherton, C. S., Flanner, M. G., and Mitchell, D.: Toward a minimal representation of aerosols in climate models: description and evaluation in the Community Atmosphere Model CAM5, Geoscientific Model Development, 5, 709–739, <GotoISI>://WOS:000305964700001, 2012.

Luo, J., Zhang, Y., Wang, F., Wang, J., and Zhang, Q.: Applying machine learning to estimate the optical properties of black carbon fractal aggregates, Journal of Quantitative Spectroscopy and Radiative Transfer, 215, 1 – 8, https://doi.org/https://doi.org/10.1016/j.jqsrt.2018.05.002, https://www.sciencedirect.com/science/article/pii/S0022407317309238, 2018a.

Luo, J., Zhang, Y., Zhang, Q., Wang, F., Liu, J., and Wang, J.: Sensitivity analysis of morphology on radiative properties of soot aerosols, Optics Express, 26, A420–A432, https://doi.org/10.1364/OE.26.00A420, http://www.opticsexpress.org/abstract.cfm?URI=oe-26-10-A420, 2018b.

Luo, J., Zhang, Y. M., and Zhang, Q. X.: A model study of aggregates composed of spherical soot monomers with an acentric carbon shell, Journal of Quantitative Spectroscopy and Radiative Transfer, 205, 184–195, <GotoISI>://WOS:000417665000021, 2018c.

Ma, X., Yu, F., and Luo, G.: Aerosol direct radiative forcing based on GEOS-Chem-APM and uncertainties, Atmospheric Chemistry and Physics, 12, 5563–5581, <GotoISI>://WOS:000305835900016, 2012.

Mackowski, D. W.: MSTM Version 3.0: April 2013, http://www.eng.auburn.edu/~dmckwski/scatcodes/, 2013.

Mackowski, D. W. and Mishchenko, M. I.: A multiple sphere T-matrix Fortran code for use on parallel computer clusters, Journal of Quantitative Spectroscopy and Radiative Transfer, 112, 2182–2192, <GotoISI>://WOS:000294518300013, 2011.

Medalia, A. I. and Richards, L. W.: Tinting Strength of Carbon-Black, Journal of Colloid and Interface Science, 40, 233–&, <GotoISI>://WOS:A1972N213300013, 1972.

Mie, G.: Beiträge zur Optik trüber Medien, speziell kolloidaler Metallösungen, Annalen Der Physik, 330, 377–445, https://doi.org/10.1002/andp.19083300302, 1908.

Mishchenko, M. I. and Yurkin, M. A.: On the concept of random orientation in far-field electromagnetic scattering by nonspherical particles, Optics Letters, 42, 494–497, <GotoISI>://WOS:000393307500032, 2017.

Mishchenko, M. I., Travis, L. D., and Lacis, A. A.: Scattering, absorption, and emission of light by small particles, Cambridge university press, 2002.

Mishchenko, M. I., Liu, L., Travis, L. D., and Lacis, A. A.: Scattering and radiative properties of semi-external versus external mixtures of different aerosol types, Journal of Quantitative Spectroscopy and Radiative Transfer, 88, 139–147, <GotoISI>://WOS:000223415600015, 2004.

Mishchenko, M. I., Liu, L., Cairns, B., and Mackowski, D. W.: Optics of water cloud droplets mixed with black-carbon aerosols, Optics Letters, 39, 2607–2610, <GotoISI>://WOS:000335496400020, 2014.

Moffet, R. C. and Prather, K. A.: In-situ measurements of the mixing state and optical properties of soot with implications for radiative forcing estimates, Proceedings of the National Academy of Sciences of the United States of America, 106, 11 872–11 877, <GotoISI>://WOS:000268178400013, 2009.

Moosmuller, H., Chakrabarty, R. K., and Arnott, W. P.: Aerosol light absorption and its measurement: A review, Journal of Quantitative Spectroscopy and Radiative Transfer, 110, 844–878, <GotoISI>://WOS:000267082200006, 2009.

Nakao, S., Tang, P., Tang, X. C., Clark, C. H., Qi, L., Seo, E., Asa-Awuku, A., and Cocker, D.: Density and elemental ratios of secondary organic aerosol: Application of a density prediction method, Atmospheric Environment, 68, 273–277, <GotoISI>://WOS:000315241100031, 2013.

Naoe, H., Hasegawa, S., Heintzenberg, J., Okada, K., Uchiyama, A., Zaizen, Y., Kobayashi, E., and Yamazaki, A.: State of mixture of atmospheric submicrometer black carbon particles and its effect on particulate light absorption, Atmospheric Environment, 43, 1296–1301, <GotoISI>://WOS:000263426600016, 2009.

Schnaiter, M., Linke, C., Mohler, O., Naumann, K. H., Saathoff, H., Wagner, R., Schurath, U., and Wehner, B.: Absorption amplification of black carbon internally mixed with secondary organic aerosol, Journal of Geophysical Research-Atmospheres, 110, <GotoISI>://WOS:000232553600003, 2005.

Schwarz, J. P., Spackman, J. R., Fahey, D. W., Gao, R. S., Lohmann, U., Stier, P., Watts, L. A., Thomson, D. S., Lack, D. A., Pfister, L., Mahoney, M. J., Baumgardner, D., Wilson, J. C., and Reeves, J. M.: Coatings and their enhancement of black carbon light absorption in the tropical atmosphere, Journal of Geophysical Research-Atmospheres, 113, <GotoISI>://WOS:000253233700002, 2008.

Shamjad, P. M., Satish, R. V., Thamban, N. M., Rastogi, N., and Tripathi, S. N.: Absorbing Refractive Index and Direct Radiative Forcing of Atmospheric Brown Carbon over Gangetic Plain, Acs Earth and Space Chemistry, 2, 31–37, <GotoISI>://WOS:000423141600004, 2018.

Smith, D. A. and Stokes, K. L.: Discrete dipole approximation for magneto-optical scattering calculations, Optics Express, 14, 5746–5754, <GotoISI>://WOS:000238437800090, 2006.

Sorensen, C. M.: Light scattering by fractal aggregates: A review, Aerosol Science and Technology, 35, 648–687, <GotoISI>://WOS:000170467100003, 2001.

Sorensen, C. M. and Roberts, G. C.: The prefactor of fractal aggregates, Journal of Colloid and Interface Science, 186, 447–452, <GotoISI>://WOS:A1997WK06000026, 1997.

Strawa, A. W., Drdla, K., Ferry, G. V., Verma, S., Pueschel, R. F., Yasuda, M., Salawitch, R. J., Gao, R. S., Howard, S. D., Bui, P. T., Loewenstein, M., Elkins, J. W., Perkins, K. K., and Cohen, R.: Carbonaceous aerosol (Soot) measured in the lower stratosphere during POLARIS and its role in stratospheric photochemistry, Journal of Geophysical Research-Atmospheres, 104, 26 753–26 766, <GotoISI>://WOS:000083781200045, 1999.

Taflove, A. and Hagness, S. C.: Computational electrodynamics: the finite-difference time-domain method, Artech house, 2005.

Thouy, R. and Jullien, R.: A Cluster-Cluster Aggregation Model with Tunable Fractal Dimension, Journal of Physics a-Mathematical and General, 27, 2953–2963, <GotoISI>://WOS:A1994NM53200012, 1994.

Wang, Q. Y., Huang, R. J., Cao, J. J., Tie, X. X., Ni, H. Y., Zhou, Y. Q., Han, Y. M., Hu, T. F., Zhu, C. S., Feng, T., Li, N., and Li, J. D.: Black carbon aerosol in winter northeastern Qinghai-Tibetan Plateau, China: the source, mixing state and optical property, Atmospheric Chemistry and Physics, 15, 13 059–13 069, <GotoISI>://WOS:000365977100021, 2015.

Wang, Y. Y., Liu, F. S., He, C. L., Bi, L., Cheng, T. H., Wang, Z. L., Zhang, H., Zhang, X. Y., Shi, Z. B., and Li, W. J.: Fractal Dimensions and Mixing Structures of Soot Particles during Atmospheric Processing, Environmental Science and Technology Letters, 4, 487–493, <GotoISI>://WOS:000415913800008, 2017.

Wentzel, M., Gorzawski, H., Naumann, K. H., Saathoff, H., and Weinbruch, S.: Transmission electron microscopical and aerosol dynamical characterization of soot aerosols, Journal of Aerosol Science, 34, 1347–1370, <GotoISI>://WOS:000185856300004, 2003.

Woźniak, M.: Characterization of nanoparticle aggregates with light scattering techniques, Thesis, 2012.

Wu, Y., Cheng, T. H., Gu, X. F., Zheng, L. J., Chen, H., and Xu, H.: The single scattering properties of soot aggregates with concentric core-shell spherical monomers, Journal of Quantitative Spectroscopy and Radiative Transfer, 135, 9–19, <GotoISI>://WOS:000331680400002, 2014.

Xu, X. Z., Zhao, W. X., Zhang, Q. L., Wang, S., Fang, B., Chen, W. D., Venables, D. S., Wang, X. F., Pu, W., Wang, X., Gao, X. M., and Zhang, W. J.: Optical properties of atmospheric fine particles near Beijing during the HOPE-J(3)A campaign, Atmospheric Chemistry and Physics, 16, 6421–6439, <GotoISI>://WOS:000378354100024, 2016.

Xu, Y. L.: Calculation of the addition coefficients in electromagnetic multisphere-scattering theory (vol 127, pg 285, 1996), Journal of Computational Physics, 134, 200–200, <GotoISI>://WOS:A1997XC98800014, 1997.

Xu, Y. L. and Gustafson, B. A. S.: A generalized multiparticle Mie-solution: further experimental verification, Journal of Quantitative Spectroscopy and Radiative Transfer, 70, 395–419, <GotoISI>://WOS:000169975700004, 2001.

Xue, H. X., Khalizov, A. F., Wang, L., Zheng, J., and Zhang, R. Y.: Effects of dicarboxylic acid coating on the optical properties of soot, Physical Chemistry Chemical Physics, 11, 7869–7875, <GotoISI>://WOS:000269548300013, 2009.

Yang, P., Wei, H. L., Kattawar, G. W., Hu, Y. X., Winker, D. M., Hostetler, C. A., and Baum, B. A.: Sensitivity of the backscattering Mueller matrix to particle shape and thermodynamic phase, Applied Optics, 42, 4389–4395, <GotoISI>://WOS:000184252300020, 2003.

Yee, K.: Numerical solution of initial boundary value problems involving Maxwell's equations in isotropic media, IEEE Transactions on antennas and propagation, 14, 302–307, 1966.

You, R., Radney, J. G., Zachariah, M. R., and Zangmeister, C. D.: Measured Wavelength-Dependent Absorption Enhancement of Internally Mixed Black Carbon with Absorbing and Nonabsorbing Materials, Environmental Science and Technology, 50, 7982–7990, <GotoISI>://WOS:000381063200006, 2016.

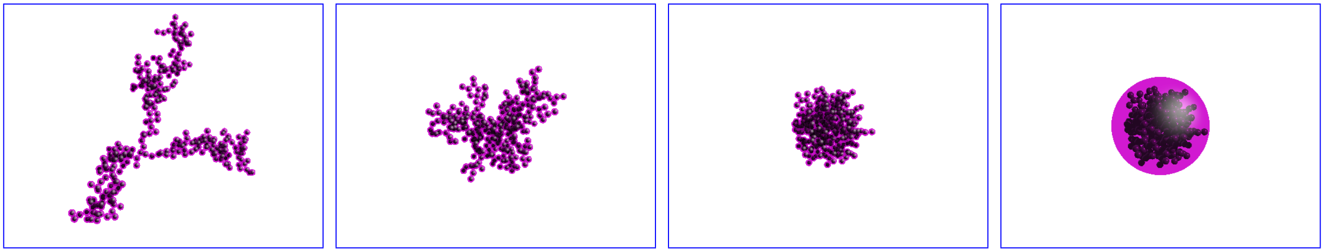
Yurkin, M. A. and Hoekstra, A. G.: The discrete dipole approximation: An overview and recent developments, Journal of Quantitative Spectroscopy and Radiative Transfer, 106, 558–589, <GotoISI>://WOS:000247510300044, 2007.

Zhang, R. Y., Khalizov, A. F., Pagels, J., Zhang, D., Xue, H. X., and McMurry, P. H.: Variability in morphology, hygroscopicity, and optical properties of soot aerosols during atmospheric processing, Proceedings of the National Academy of Sciences of the United States of America, 105, 10 291–10 296, <GotoISI>://WOS:000258211600006, 2008a.

Zhang, R. Y., Khalizov, A. F., Pagels, J., Zhang, D., Xue, H. X., and McMurry, P. H.: Variability in morphology, hygroscopicity, and optical properties of soot aerosols during atmospheric processing, Proceedings of the National Academy of Sciences of the United States of America, 105, 10 291–10 296, <GotoISI>://WOS:000258211600006, 2008b.

Zhang, X. L., Mao, M., Yin, Y., and Wang, B.: Absorption enhancement of aged black carbon aerosols affected by their microphysics: A numerical investigation, Journal of Quantitative Spectroscopy and Radiative Transfer, 202, 90–97, <GotoISI>://WOS:000412962100014, 2017.

Zhang, X. L., Mao, M., Yin, Y., and Wang, B.: Numerical Investigation on Absorption Enhancement of Black Carbon Aerosols Partially Coated With Nonabsorbing Organics, Journal of Geophysical Research-Atmospheres, 123, 1297–1308, <GotoISI>://WOS: 000425520200039, 2018.



(a) Thinly coated, $D_f = 1.8$ (b) Thinly coated, $D_f = 2.2$ (c) Thinly coated, $D_f = 2.6$ (d) Thickly coated, $D_f = 2.6$

Figure 1. Typical morphologies of BC, $n_s = 300, k_0 = 1.2$.

Table 1. Morphological parameters of BC aerosols

Parameters	Thinly-coated BC	Thickly-coated BC
k_o	1.2	1.2
n_s	1-1000	1-1000
D_f	1.8,2.2,2.6	2.6
f_{soot}	0.2,0.4,0.6,0.8,1.0	0.05, 0.06, 0.075, 0.1

Table 2. MAC (m^2/g) for bare BC at different D_f $(r_g = 0.06\ um, \sigma_g = 1.5)$.

$\lambda\ (nm)$	$D_f = 1.8$	$D_f = 2.2$	$D_f = 2.6$
350	9.30	9.03	8.48
404	8.14	7.95	7.60
532	6.20	6.11	6.02
700	4.68	4.64	4.65

[..[158]]

[..[165]]

[158] removed: Size distribution of bare and coated BC.

[165] removed: Imaginary part of the refractive index of BrC based on the study of Kirchstetter et al. (2004).

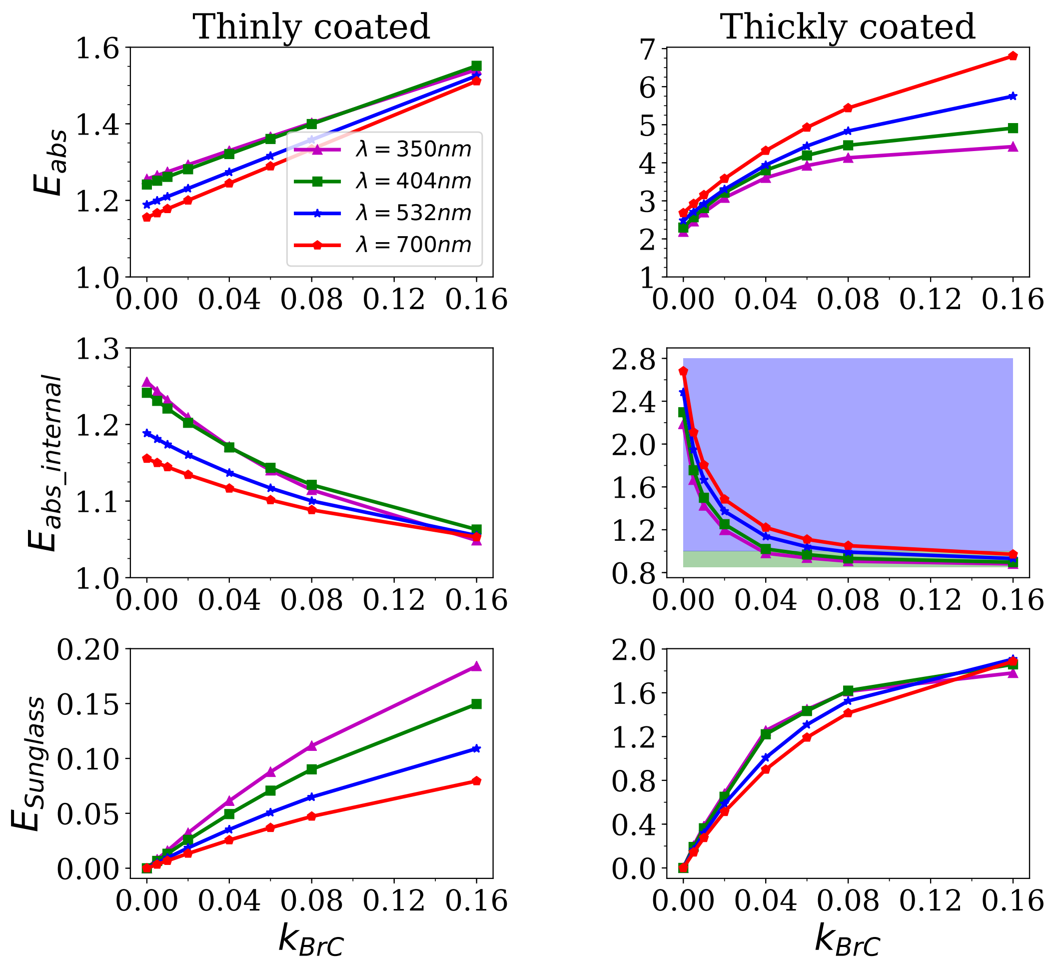


Figure 2. Effects of K_{BrC} on [..[159]]specific enhancement ($n_s = 200$). [..[160]]For thinly-coated BC[..[161]], $D_f = 2.2$, and $f_{BC} = 40\%$[..[162]]; [..[163]]for thickly-coated BC[..[164]], $D_f = 2.6$, and $f_{BC} = 5\%$. The blue shading represents the $E_{abs\_internal}$ of larger than 1, while the green shading describe the range of $E_{abs\_internal}$ of less than 1.

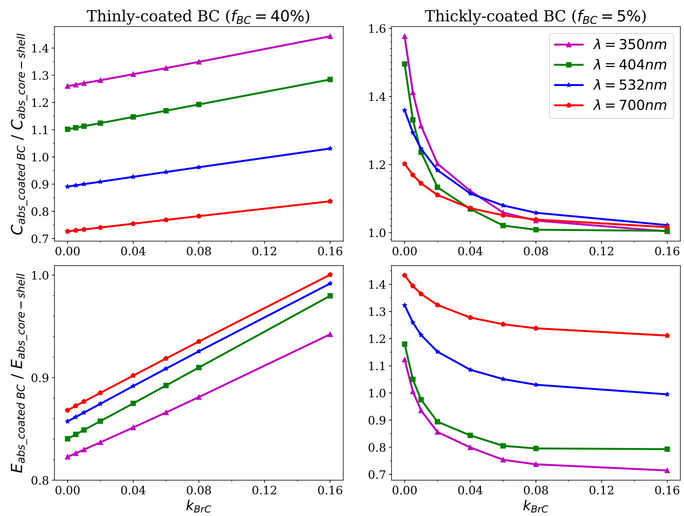


Figure 3. Effects of k_{BrC} on the applicability of core-shell sphere ($n_s = 200$).

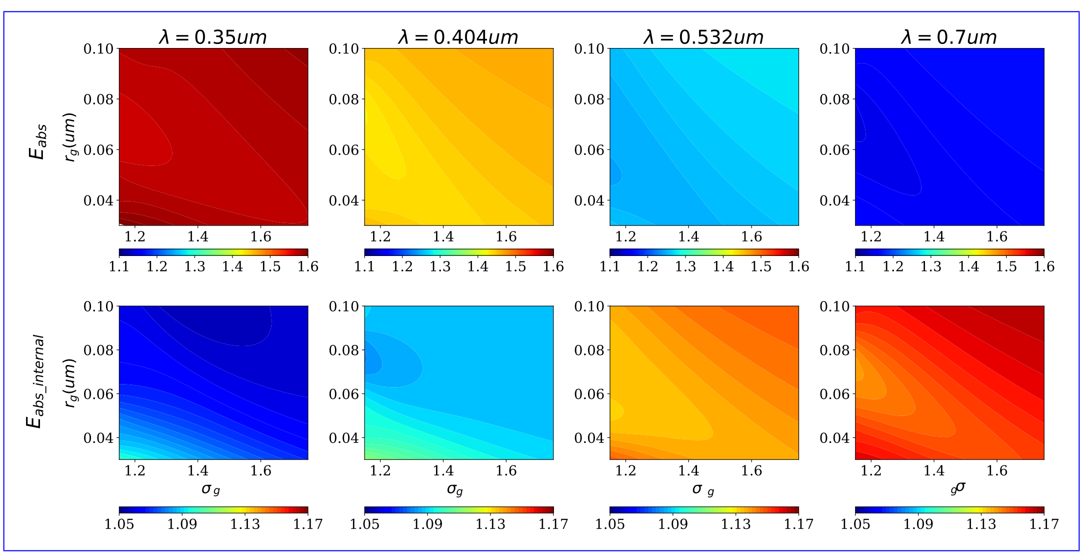


Figure 4. E_{abs} and [..166]$E_{abs\_internal}$ of thinly-coated BC with BrC coatings at different size distributions ($D_f = 2.2, f_{BC} = 40\%$).

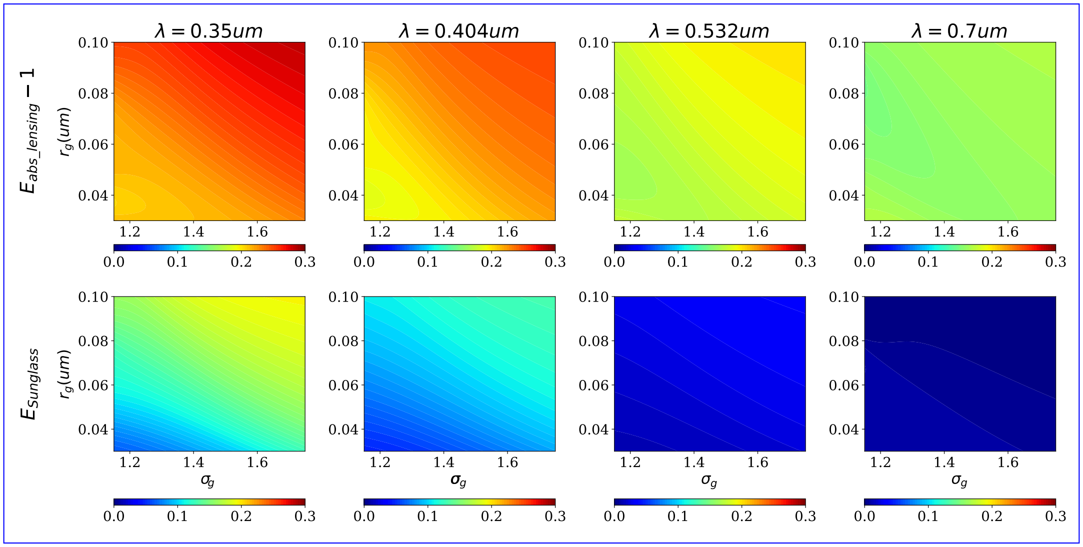


Figure 5. [..[167]]Similar as Figure 4, but for $E_{abs\_lensing}$ [..[168]]and $E_{Sunglass}$.

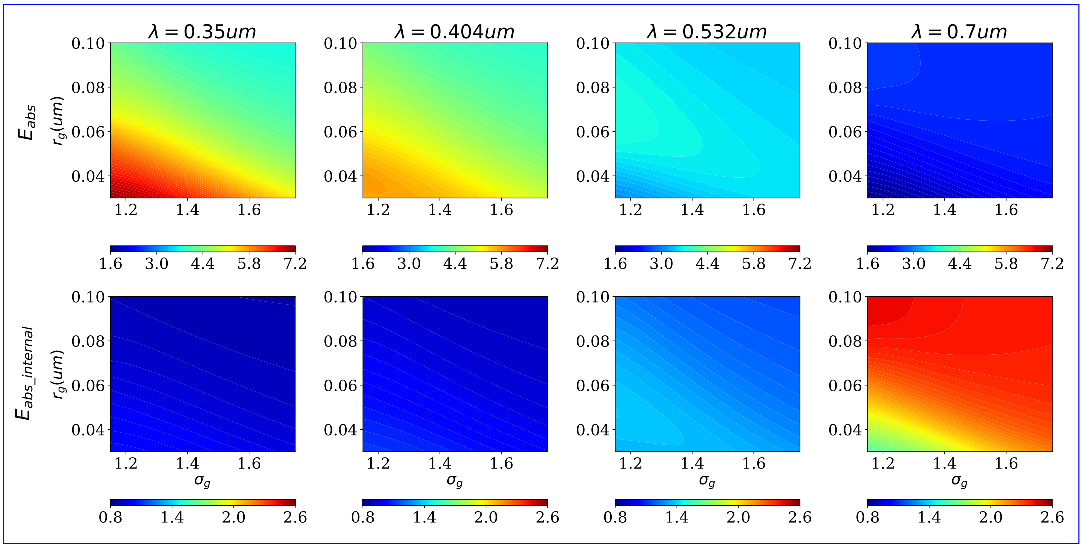


Figure 6. E_{abs} and [..[169]]$E_{abs\_internal}$ of [..[170]]BC thickly-coated with BrC [..[171]]at different [..[172]]size distributions ([..[173]]$D_f = 2.6$, [..[174]]$f_{BC} = 6\%$).

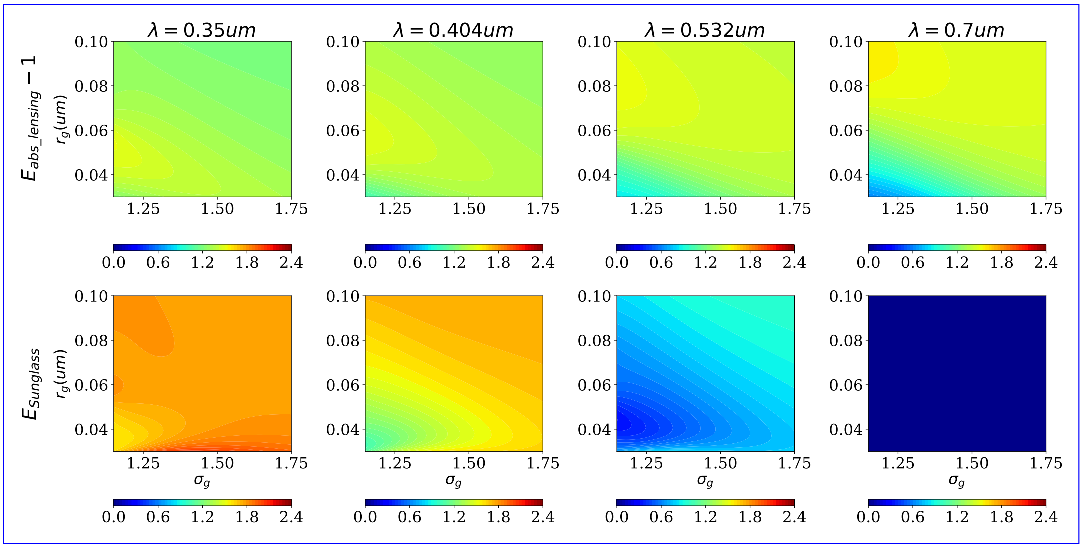


Figure 7. [..[175]]Similar as Figure 6, [..[176]]but for $E_{abs\_lensing}$ and $E_{Sunglass}$.

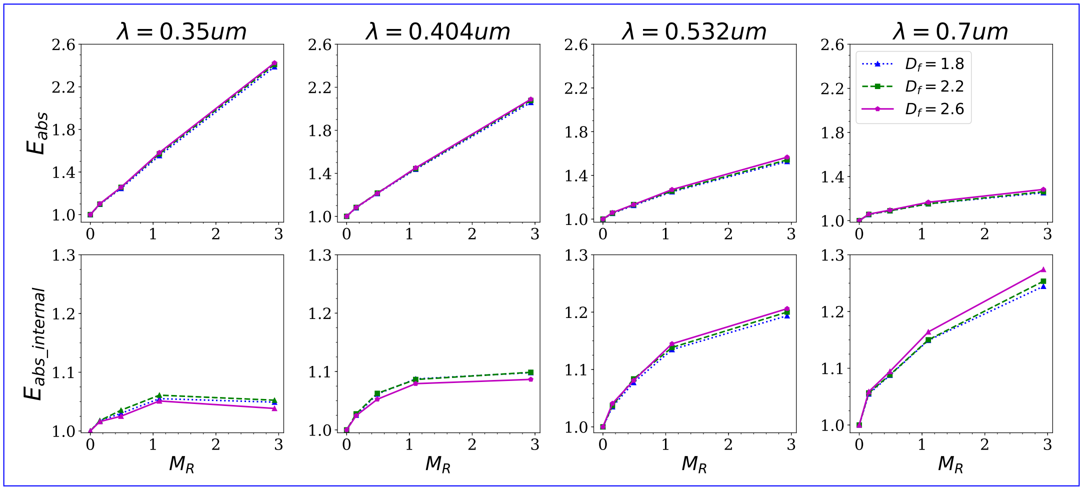


Figure 8. E_{abs} and [..[177]]$E_{abs\_internal}$ of thinly-coated BC with BrC coatings varying with [..[178]]M_R for different $D_f (r_g = 0.06um, \sigma_g = 1.5)$.

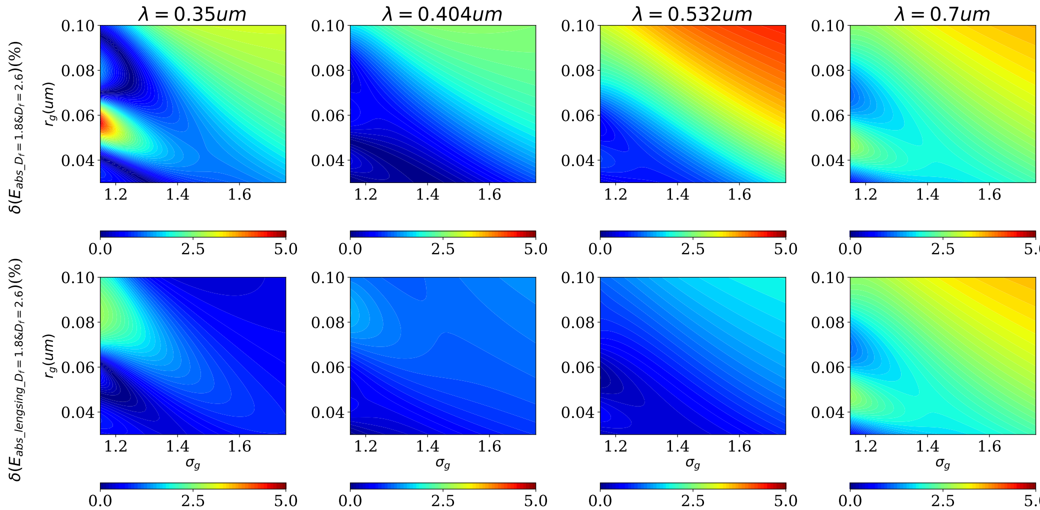


Figure 9. The relative deviations of absorption properties between $D_f = 1.8$ and $D_f = 2.6$ for thinly-coated BC with BrC coating ($f_{BC} = 20\%$).

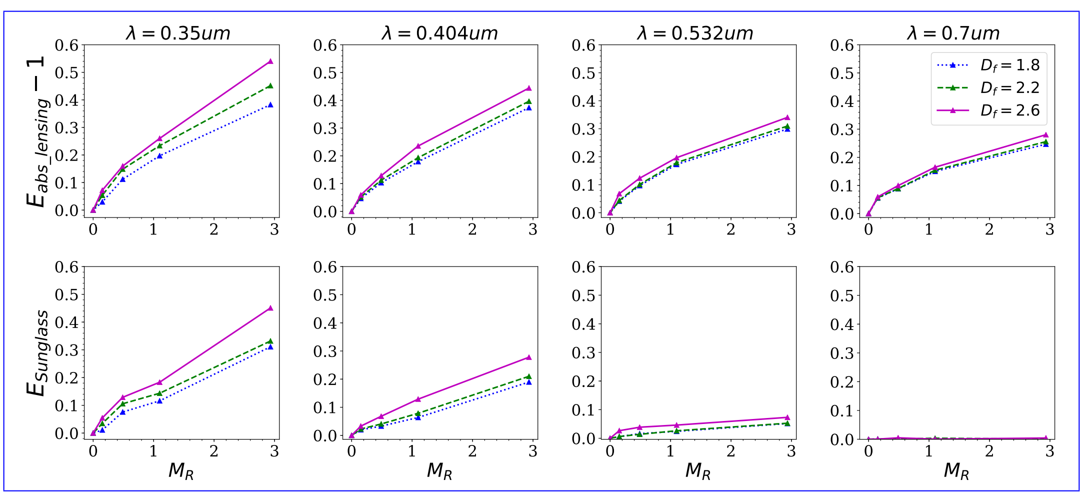


Figure 10. Similar as Figure 8, but for $E_{abs\_lensing}$ and $E_{Sunglass}$.

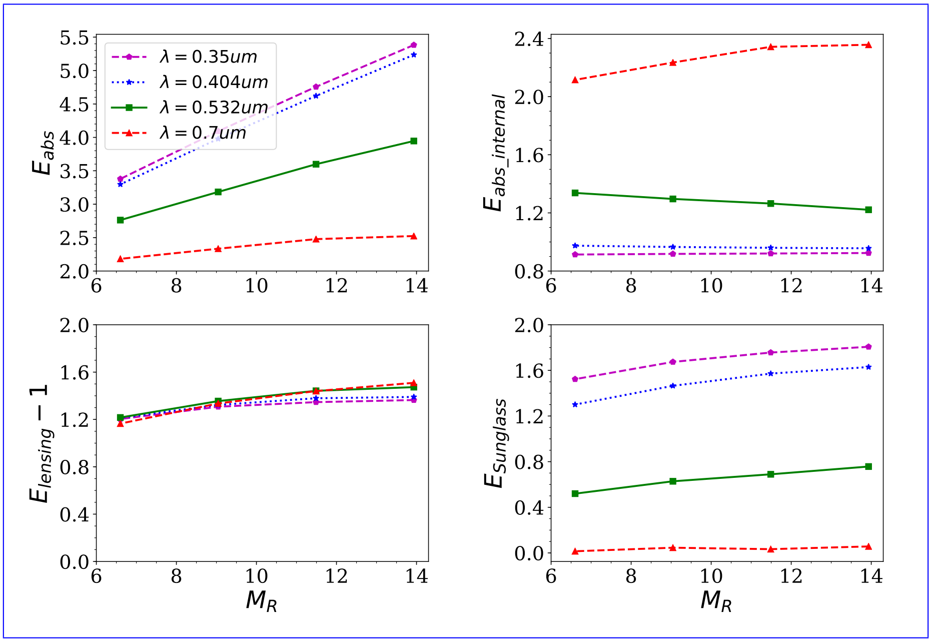


Figure 11. E_{abs} and $E_{abs\_lensing}$ of thickly-coated BC with BrC [..[179]]coatings varying with [..[180]]M_R ([..[181]]$r_g = 0.06um, \sigma_g = 1.5$).

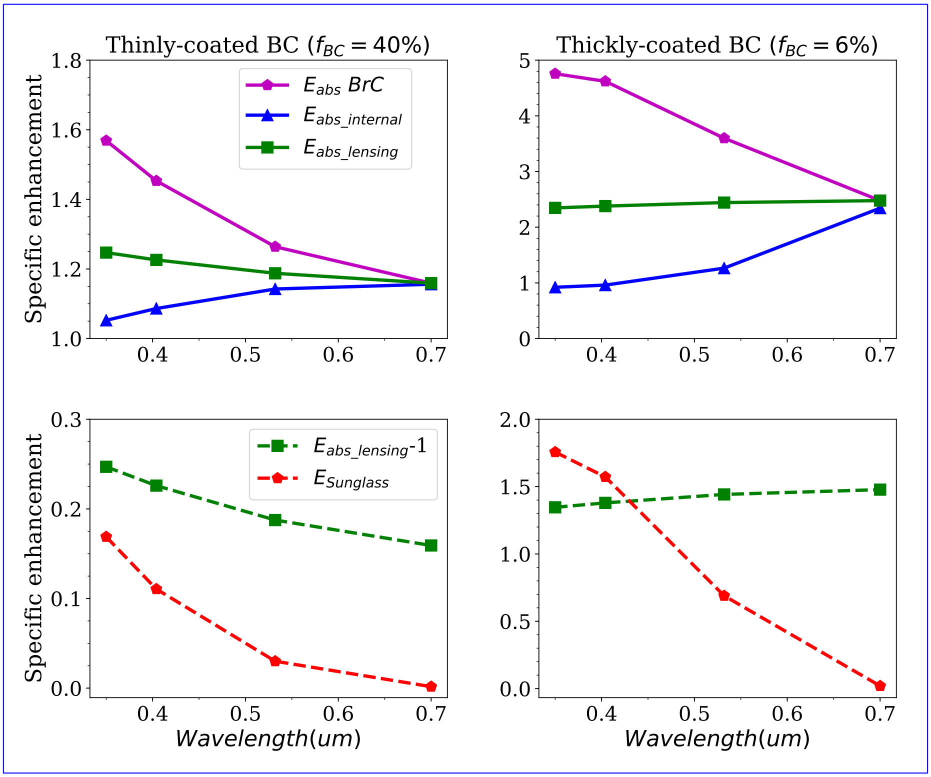


Figure 12. Comparison of BC coated with non-absorbing materials and that coated with BrC (r_g=0.06um, $\sigma_g = 1.5$). $D_f = 2.2$ and $D_f = 2.6$ were assumed for thinly-coated and thickly-coated BC, respectively.