Response to the comments of reviewer #1

(The responses are highlighted in blue)

The authors are very grateful to the editors and reviewers for their valuable comments and constructive suggestions. The reviewers' questions and comments are highlighted in **black font**, and the answers in **blue**. The changes made in the revised manuscript are highlighted in **red.**

Comments: This study calculated the optical properties and radiative effects of black carbon (BC) particles with different absorption Ångström exponent (AAE) methods, and further estimated their effect on brown carbon (BrC). The manuscript explained how the microphysical properties of BC particles determine wrong BC's and BrC's absorption and radiative properties under two AAE methods. The study is interesting and the results are helpful for understanding why the optical and radiative properties of BrC is deviated to BC. Overall, the manuscript can be revised and then may be published in ACP. The problems are addressed as following:

Comments: The author calculated the absorption deviation for BrC by using non-absorbing coating. If these calculations make sense, we must assume that the true AAE of the BC particles with BrC coating is the same with those with non-absorbing coating. However, is that true? And hence I concern whether the "babs_BC_440_Estimated - babs_BC_440" can represent the Δ I'm very confused here.

Reply: thank you for your comment. Here we have only calculated the absorption of black carbon with non-absorbing coating materials, and the BrC absorption is the difference between the total absorption and the black carbon mixed with non-absorbing coating materials. We have assumed that the total absorption at the near-infrared wavelength comes entirely from BC mixed with non-absorbing coatings. Thus, if we know the AAE of BC with non-absorbing coatings, we can calculate the absorption of BC with non-absorbing coating materials, including BC absorption and lensing effect, but excluding the absorption of BrC. Of course, the block effect of BrC would also affect the total absorption, but it is also caused by BrC absorption, so we attribute the effect to BrC absorption. Therefore, to calculate the BrC absorption, we need the AAE of BC mixed with non-absorbing materials, but not the AAE of BC with BrC coating materials. If we know the AAE of BC mixed with BrC, we can only calculate the total absorption, but not the absorption of BC with non-absorbing coatings, and therefore we cannot separate the absorption of BC and BrC.

Comments: Besides, refractory index is one of the factors determining the absorption. The deviation of BC absorption will be also affected by the refractory index. All the discussion in this manuscript is carried out under the 1.95+0.79i for BC and 1.55+0i for coating. The results for "the estimated BrC absorption should be the absorption from BC that is incorrectly attributed to BrC" is not comprehensive because the refractive index varies.

Reply: From the review by Bond and Bergstrom (2006), it appears that the value 1.95+0.79i should be used at 550 wavelengths. While it is true that the BC refractive index can vary with wavelength, it deviates significantly in the UV and NIR. In addition, it is still unclear how the BC refractive index varies with wavelengths, and we do not expect much variation from the UV and NIR, as shown in Bond and Bergstrom (2006),. Therefore, it is often assumed that the BC refractive index is independent of wavelength in the UV and NIR. Furthermore, if we use a spectral dependent refractive index, we cannot see how the microscopic properties affect the measurements, since the BC refractive index would also affect the total absorption. Therefore, we used a fixed refractive index. It is difficult to do all the calculations because the actual refractive index of BC is still an open question. However, since our study focuses on the effects of microscopic properties, we used a fixed refractive index. Even though we only consider a fixed refractive index, we believe that the main conclusions are similar for other refractive indices.

Comments: The study may have an important implication on the estimation of BrC's optical properties and radiative effect. This should be addressed in the manuscript. The author only summarized the main conclusions and did not give impressive implication.

Reply: Thanks for your comments. We added a section to emphase the implication：

AAE-based methods have been widely used to estimate the absorption of BrC, while they are subject to large uncertainties due to the properties of BC. We quantify the effects of the microphysical properties of BC based on numerical simulations and investigate how the applicability of AAE-based methods varies at different aging conditions. From the above, it is clear that using a BC AAE of 1 can provide reasonable estimates for BrC absorption, while the deviation from the "true" BrC absorption becomes significant as the particles age. This means that the AAE = 1 method can provide inaccurate estimates when aged BC is present. In general, regions near emission sources, such as urban traffic areas, contain mainly freshly emitted BC. In this case, it is reasonable to use the AAE = 1 method. With atmospheric aging, we should adjust the AAE values because both the AAE = 1 and WDA methods can sometimes result in misallocations of tens of percent of BrC absorption. However, the adjustments should differ depending on the aging condition. As shown in Figure 3 – 4, for fluffy BC partially mixed with coating materials 440 (Df = 1.8 and 0 < F < 1 in this work), ABSBrC = 0 occurs in most cases when AAE > 1. Therefore, we generally propose a relatively larger AAE, while a smaller AAE is recommended for compact BCs, including coated and uncoated compact BCs. Recent observations have shown that the average D_f is often small even for coated BCs in regions far from emission sources (Wang et al., 2017; Yuan et al., 2019). Therefore, we prefer larger AAEs. However, there are also more compact BC aerosols in the atmosphere, and we should also consider the uncertainties when the real BC aerosols have a compact structure. In addition, the WDA methods does not improve the estimation. Therefore, we should carefully consider the uncertanties caused by the microphysical properties of BC when estimating the BrC absorption and DRF based on the AAE based methods."

Besides, we have also written the conclusion and summary:

“The AAE-based method is commonly used to estimate the absorption of BrC, but may provide inaccurate estimates due to the effects of the microphysical properties of BC. The goal of this work is not to discuss the use of the AAE-based method, but to assess the uncertainties of the AAE-based method. We find that an AAE of 1 can provide a reasonable estimate when BC is freshly emitted. Therefore, an AAE of 1 is suggested for regions close to the emission source, such as vehicle emission region. However, we should also note the uncertainties associated with using an AAE of 1. We estimate an ABS_{BrC} range of about -4.8% to 2.7% when using an AAE of 1 for freshly emitted BC. However, the ABS_{BrC} range becomes broader when BC is aged, and sometimes ABS_{BrC} can be varied in the range of about -34.5% – 38.7%, depending on the aging status and morphologies. Therefore, we need to adjust the AAE value when the fixed AAE method is applicable to the region consisting of aged BC, such as regions far from the emission source. However, even for aged BC, different AAE values should be used for different aging conditions, since we show that no fixed AAE is applicable for all cases.

This work represents the aging condition by assuming a more compact structure, more coating materials and a larger F. For different aging processes, the adjustment of AAE values should be different. For fluffy BC partially mixed with coating materials (Df = 1.8 and 0<F<1 in this work), we generally propose a larger AAE, while a smaller AAE is recommended for the compact BC. Our results also show that the Mie theory-based WDA method does not necessarily improve the estimate, with a corresponding ABSBrC range of about -40.8% – 35.7% in our simulation cases, due to the substantial WDA deviation between the morphologically realistic BC and the spherical BC.

At the global level, the use of BC AAE of 1 can lead to a global mean misassigned AAOD of about -0.43 – 0.46 × 10^{-3} resulting in a corresponding global mean mis-assigned DRF of -0.073 ± 0.0185 to +0.078 ± 0.0198 W/m2. However, for the freshly emitted BC, an AAE of 1 does not lead to a significant misestimation of the AAOD. At the regional level, for an AAE of 1, the mean mis-assigned AAOD can vary in the range of -7.3 to 5.7 × 10^{-3} in some regions, leading to a mis-assigned DRF of about -1.24 ± 0.314 W/m^2 to +0.97 ± 0.245 W/m^2. The WDA method can provide a less accurate estimate for BrC absorption, and sometimes in some regions we can see a mean mis-assigned AAOD of about -22 × 10^{-3}, leading toa mis-assigned DRF of about -3.74 ± 0.946 W/m^2. Therefore, the effects of the microscopic properties of BC should be carefully considered when estimating BrC absorption and its direct radiative forcing based on the measurements at multiple wavelengths."

Comments: The author gave the results for F<=0.3. However, the aged BC particles have F ranges in 0-1 in the atmosphere. It seems the present results in this study are not complete. I think it is impossible to construct a shape model with large F and small coating thickness due to the limitation of the MSTM method (The coating must be sphere). If this is the situation, then why the author did not use DDA to calculate the results? If the data for F > 0.3 can not be supplemented, please clarify this in the main context.

Reply: Thanks for your comments. In this work, we just considered a spherical coating. Therefore, for fluffy BC, larger F may be not found. However, we expect that larger F may not modify the main conclusion of this work. In addition, for compact BC (D_f = 2.6), we have considered an F of 1 for comparison (please see Figure S1 – S2). In the future, more complex coating structures should be considered. We have clarified that in the revised manuscript:

"Since we consider only spherical coating structures in this work, a large F for completely BC may not be found for a BC volume fraction. Therefore, we only consider an F range from 0 to 0.3 for fluffy BC. However, we assume that BC with large F would not change the main results of this work."

Specific:

Comments: Line 6: The term ABS_{BrC} was not well explained. It is not "the estimated BrC absorption". The ABS_{BrC} seems to be a critical parameter to understand the whole manuscript. In the Abstract, the meaning of ABS_{BrC} should be clearly explained to help readers to understand the results mentioned in the Abstract.

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

Comments: Line 9: The full name of “WDA” was not mentioned before using this abbreviation.

Reply: Thanks for your comments. We have given the definition in the revised manuscript.

Comments: Lines 181-182: “the corresponding r_{max} and r_{min} are 0.0342 μm and 0.2 μm” seems a wrong sequence for r_{max} and r_{min}.

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

Comments: Lines 235-237 and 238-239: It seems that the AAE_{440_870} = 1 method often has larger ABS_{BrC} than AAE_{440_675} = 1 method. Why this happens? The results should be tried to explain here.

Reply: Thanks for your comments. The reason may the that the gap between 440 and 870nm is further than the gap between 440 and 675nm, which leads to larger ABS_{BrC}.We have clarified it in the revised manuscript:

“The AAE_{40_870} = 1 method generally shows a larger range of ABS_{BrC} than the AAE_{440_675}= 1. This could be due to the larger wavelength gap between 440 and 870 nm. ”

Comments: Lines 243-244: The sentence is too complicated. Maybe the author miss some punctuation marks.

Reply: Thanks for your comments. We have modified the sentence as “With D_f of 1.8, F of 0.1, and D_p/D_c of less than 2.71, ABS_{BrC} varies in the range of about -6% - 18%

and -12% - 9% when $AAE_{440\_675} = 1$ and $AAE_{440\_870} = 1$. The ranges become -18% - 3% and -21% - 4.\% when D_p/D_c is 4.64.”.

Comments: Line 246: “can be observed” I don’t think “observed” is a suitable word here.

Reply: Thanks for your comments. We modified “observed” as “found” in the revised manuscript.

Comments: “On the other hand, the AAE increases with Dp/Dc when BC has a fluffy structure. Thus, the AAE can be greater than 1 when the fluffy BC is partially coated with a thick coating (Zhang et al., 2020b; Luo et al., 2023), resulting in ABS_{BrC} of less than 0.” The author tried to explain why the $ABS_{BrC} < 0$, but there is no direct causal relationship between the former and the latter sentence. The author needs to give more reasonable explanation.

Reply: Thanks for your comments. We are very sorry for without clarifying clearly the reason. As shown in the manuscript, the absorption coefficient of BC, which is incorrectly attributed to BrC, can be calculated as follows:

$$\Delta_{BrC} = b_{abs\_BC\_440\_Estmiated} - b_{abs\_BC\_440}$$

$$b_{abs\_BC\_440\_Estmiated} = b_{abs_{BC_\lambda}} \left(\frac{440}{\lambda}\right)^{-1}$$

$$b_{abs\_BC\_440} = b_{abs_{BC_\lambda}} \left(\frac{440}{\lambda}\right)^{-AAE\_true}$$

Thus, if the AAE of partially coated BC is greater than 1, as λ>440, so $b_{abs\_BC\_440} > b_{abs\_BC\_440\_Estmiated}$. Therefore, $ABS_{BrC} < 0$. We have added clarifications in the revised manuscript:

“On the other hand, the AAE increases with Dp/Dc when BC has a fluffy structure. Thus, the AAE can be greater than 1 when the fluffy BC is partially coated with a thick coating (Zhang et al., 2020b; Luo et al., 2023). This would result in the predicted black carbon absorption coefficient being larger than the true black carbon absorption coefficient, so resulting in ABS_{BrC} of less than 0."

Comments: Line 256: "The more compact structure can also represent another process of atmospheric aging". The compact structure was caused by many aging processes. I wonder the author means which process?

Reply: Thanks for your comments. We have re-written the sentences:

"With atmospheric aging, the BC cores are reconstructed to be a more compact structure. We used a larger Df (D_f =2.6) to represent the compact BC. Even with F = 0, a Df of 2.6 represents the highly aged BC. By comparing BC with fluffy and compact structrues, we can see more deeply from the effects of atmospheric aging on the estimations of BrC absorption."