

Thanks for your comments! The point-by-point responses are listed below.

Comment: The manuscript has been largely improved since the previous few versions. It emphasizes an important issue when converting the bulk MR to the absorbing properties of individual particles. The authors claim that by introducing the parameterization of the mixing state index, the variation of Eabs at the same MR could be explained. I recommend publication after a few issues addressed.

Reply: Thanks for the comments.

Comment: (1) I think it is necessary to show a few plots in the main text to demonstrate how you have measured the mobility size-resolved BC core size (Dc) distribution, e.g. a few Dp-Dc matrix for the cases in Fig. 3.

Reply: Thanks for the comment. The measured mean value of BC-containing number size distributions under different Dp and Dc between the day of 27 and 28, May, 28 and 29, May, 29 and 30, May are shown in Fig.R1. It is obvious that the BC-containing number and coating thickness increase with the pollution levels.

We added the Dp-Dc matrix for the cases of Fig. 3 in the manuscript and the supplementary materials.

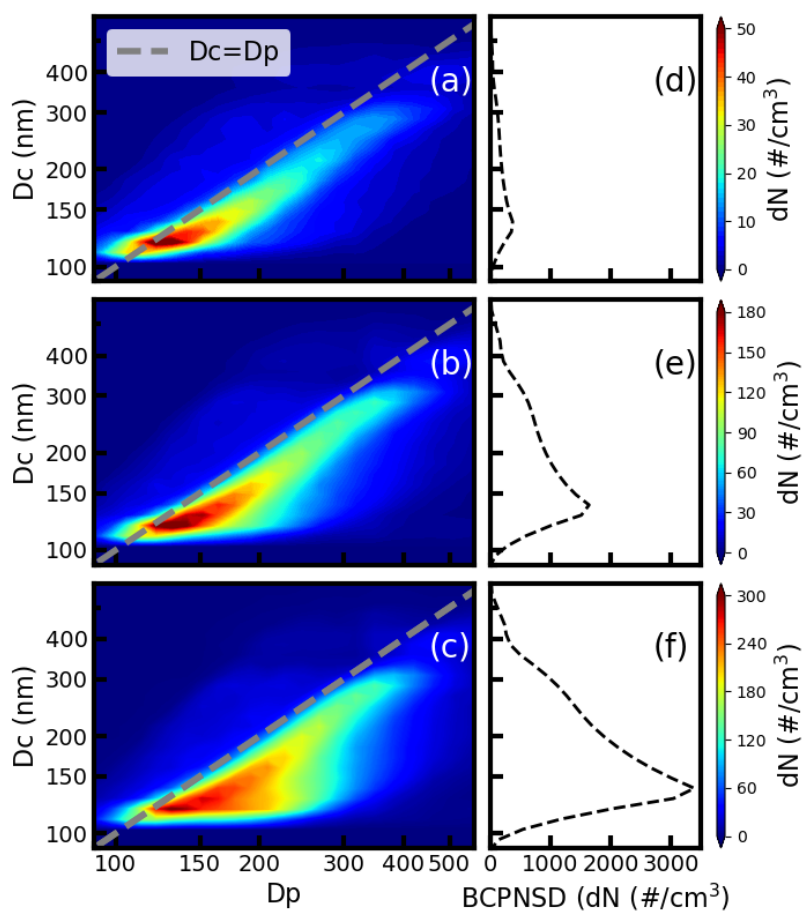


Figure R1. The measured BC-containing aerosols under different D_p and D_c conditions during the period of (a) 27, May and 28, May, (b) 28, May and 29, May, and (c) 29, May, and 30, May. The panels (d), (e), and (f) are the corresponding BC core number size distributions of (a), (b), and (c), respectively.

Comment: (2) One thing still not quite clear is how you have converted the mixing state index to the absorption in bulk.

Reply: When the ambient aerosol χ and MR were measured, the corresponding E_{abs} can be estimated from Fig. 7(a) in the manuscript.

The main purpose of our manuscript is better to constrain the difference between the measured and calculated BC-containing aerosol E_{abs} . We added some descriptions to the manuscript.

Comment: A higher Chi means the coatings were more homogenously distributed on the rBC, rather than the population with lower Chi containing a fraction of BC without apparent Eabs.

Reply: Thanks for the comment.

Comment: Section 3.4 is a bit too simplified for readers to obtain the necessary information.

Reply: Thanks for the comment. We added some descriptions in section 3.4 to make clear the Mont-Carlo simulations.

During the simulation, a group of the BC-containing aerosols was generated with the D_p and D_c meet the following conditions and the number of BC-containing particles was assumed to be 30. For each of the BC-containing particles, the core diameter of the BC particle was randomly generated with a geometric mean diameter of 130.7 nm and a geometric standard deviation of 1.5, which is the mean measurement results of the BC core distribution during the field measurement (Zhao et al., 2020). The corresponding MR of the BC particle is assumed to be randomly distributed in the range between 0.0 (pure BC particles without coating)

and 78.0 (particles with a core diameter of 130 nm and a total diameter of 560 nm). For each group particle, the corresponding aerosol bulk MR, E_{abs} and χ can be calculated using the core-shell Mie scattering model. The simulations were conducted for 10^7 times, and the calculated mean and standard deviation of E_{abs} under different MR and χ are summarized for further analysis.

Comment: Line 241 says you used a constant rBC core size distribution, but Fig. 2 gives a few examples of different chi, which contain apparently different rBC core size distributions.

Reply: Thanks for the comment. The main purpose of the diagram in Fig. 2 is to illustrate the relationship between D_α , D_γ , and χ . For better display the condition with D_α , D_γ , and χ values equaling 1, 1, and 1 respectively, we need to display the BC particles in Fig. 2 with different core distributions. However, it is not related to the rBC core size distributions in Line 241.

Comment: I would suggest giving more details about how the absorption has been calculated, to explain it is the Chi but not the rBC core size distribution causing the variation of resultant Eabs.

Reply: The details of calculating the single particle and bulk absorption are shown in section 2.3.1 and 2.3.2 in the manuscript. Some more detailed

descriptions were added in the manuscript.

We calculated the measured mean BC core distributions under different pollution conditions corresponding to Fig. R1 and the results are shown in Fig. R2. From Fig. R2, the normalized BC-core distributions under different pollutions are almost the same for different pollution levels as shown in Fig. R2. Thus, it is the χ that mainly causes the variation of resultant Eabs.

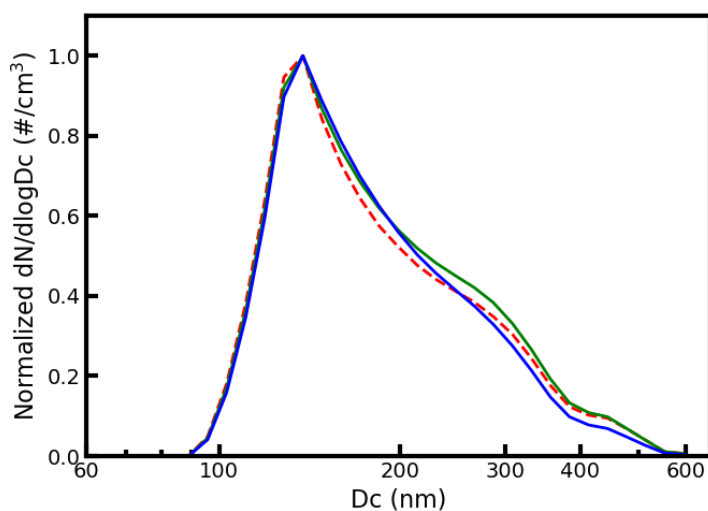


Figure R2. Normalized BC PNSD under different pollution conditions corresponding to fig. S7. a (red), b (green), and c (blue).

Comment: (3) Based on comment (1), it would be useful to see how the ambient chi varies and what is the essential reason Chi has caused different Eabs. Is low Chi because there was a large fraction of uncoated/less coated large rBC?

Reply: Thanks for the comment. The reviewer gives a very perspective view about the research we are going to carry out in our future work as it is very important to see how the ambient χ varies with the ambient conditions. The χ is low partially because there was a large fraction of uncoated/less coated large rBC. From the definition of the χ , the χ is low due to the high particle-to-particle heterogeneity. In our future work, the characteristic of the χ due to ambient processing such as BC emission, aging, and boundary development.

Comment: (4) Also, it may not be appropriate for all to use the core-shell model when BC was thinly coated, has this been considered.

Reply: Thanks for the comment. It is always overestimated when the core-shell model is used to calculating the ambient BC-containing light absorptions. The overestimation was accounted for using the correction coefficient suggested by Wu et al. (2018).

Comment: (5) Has the particle shape been considered regarding the measurement of DMA, given the electrical mobility sizing is sensitive to the particle shape [Hu et al. 2021]. Particles especially at larger D_m may require consideration for the particle non-sphericity, while using mobility size may overestimate the total particle mass. This discussion may be included.

Reply: Thanks for the comment. We agree with the reviewer's opinion that the particle shape should be considered when calculating the BC light absorption. It is always overestimated when the core-shell model is used to calculating the ambient BC-containing light absorptions. The overestimation was accounted for using the correction coefficient suggested by Wu et al. (2018).

Comment: (6) Fig. 3 only shows a very narrow range of χ , would be possible to show a longer time series and cover the range of ambient measured χ .

Reply: Thanks for the comment. We add the time series of the measured χ time series in Fig. S6 (d) in the supplementary material. The χ ranges between 0.6 and 0.83. For a better understanding of the characteristics of the above parameters, we only present the time series of these parameters during a pollution period between 27, May and 30, May in Fig. 3 in the manuscript.

Comment: Other technical comments:

The font size of last paragraph should be consistent with the others. There are many places in the texts having inconsistent format, such as font size and line space.

Reply: Thanks for the comment. We checked the format of the manuscript again.

Comment: Line 164, typo “ss”.

Reply: We have deleted the ‘ss’ at line 164.

Comment: There are grammatical errors in line 278 and line 281 “by simply introducing”, line 279, duplicated “then”. Please carefully check through the whole texts.

Reply: Thanks for the comments. We have checked the grammatical errors of our texts.

References

Hu, K., et al, Measurements of the Diversity of Shape and Mixing State for Ambient Black Carbon Particles: *Geophysical Research Letters*, 48 (17), 10.1029/2021GL094522,2021.

Wu, Y., Cheng, T., Liu, D., Allan, J. D., Zheng, L., and Chen, H.: Light Absorption Enhancement of Black Carbon Aerosol Constrained by Particle Morphology, *Environ Sci Technol*, 52, 6912-6919, 10.1021/acs.est.8b00636, 2018.

Zhao, G., Shen, C., and Zhao, C.: Technical note: Mismeasurement of the core-shell structure of black carbon-containing ambient aerosols by SP2

measurements, Atmospheric Environment, 243, 117885,
10.1016/j.atmosenv.2020.117885, 2020.