

Major comments:

1) A new method by considering the variation in MAC is developed to obtain BC mass size distribution and then bulk BC mass concentration from size-resolved light absorption measurements. Size-resolved MAC calculated on the basis of core-shell Mie model is mainly discussed, which is determined by D_p -dependent D_{BC} and coating thickness. However, there are many assumptions in calculation processes, e.g., same D_{BC} and coating thickness at each selected mobility size, a constant number fraction of BC-containing particles, etc. Meanwhile, measurements were not described clearly.

Response: Thanks for your comments. The size resolved MAC in this study was based on core-shell Mie model. The influence of the BC aggregates on the MAC as well as the relative deviation between the core-shell model and BC aggregates were discussed in section 5.1 of the revised manuscript to evaluate the effects of the morphology on MAC. With respect to the assumptions used in this study, their uncertainties were discussed in section 5, such as the uncertainties caused by using idealized core-shell model (section 5.1), by using a constant BC-containing particle fraction (section 5.2) and by variation of refractive index (section 5.3). With respect to description of measurements, a more detailed description of our measurement was added in section 2.

2) The significance of this study should be also strengthened. In my point of view, compared to BC mass loading, the light absorption measurements are more required to evaluate the influences of BC particles on solar radiation. Thus, MAC is likely to be more important for converting bulk BC mass loading, which can be directly measured by using chemical method (e.g., Thermo Optical Reflection-EC) or laser-induced incandescence techniques (e.g., SP2-rBC), to light absorption in climate research. The current study is more important for obtaining BC mass size distribution from size-resolved absorption measurement. BC mass size distribution obtained from the DMA-AE51 measurement based on the new method is also suggested to compare

with that obtained from the direct measurement from DMA-SP2 system, which has used in the field campaign.

Response: Thanks and we agree with your comments. More sentences were added in this text to stress the significance. The main goal of this study was to derive equivalent BC mass concentration (EBC, after Petzold et al. (2013)) more precisely and obtain BC particle mass size distribution (BCPMSD) from size-resolved absorption measurement. MAC is an important variable that has to be discussed in the process. Derivation of the EBC and related uncertainties were more discussed to emphasize that our goal was to determine EBC more precisely.

3) The Mie model is likely to not suitable for the calculation of BC aggregates with large sizes. For a small BC particle (core), the mass equivalent diameter of the assumed BC sphere is much smaller than the wavelength (880 nm) resulting in a less effect of morphology to absorption. In this case, the Mie model is somewhat feasible for absorption estimation. However, for a large BC particle (core), its mass equivalent diameter is close to the wavelength (i.e., large size parameter); thus, the absorption is largely influenced by the morphology. Moreover, large BC particles are more likely to exhibit loose fractal aggregates with thin coating, thus, is likely much different from core-shell structure. MAC in this case cannot be well depicted by using Mie model.

Response: Thank you for your comments. In section 5.1 of our revised manuscript, the uncertainty caused by using idealized core-shell model was discussed by replacing the BC core with cluster-like aggregates calculated with multiple sphere T-matrix (MSTM) method. The relative deviation between MAC calculated by MSTM model and by core-shell Mie model was investigated. The results showed that when the size of BC core was smaller than 150 nm, the overall deviation was within 4 %, which indicated that Mie theory was a good approximation to the BC aggregates even when BC core reached 200 nm. When BC core was larger than 200 nm, MAC calculated by MSTM model increased with increasing thickness of shell. The deviations between MAC calculated by the idealized concentric core-shell model and letting BC particles be in the form of

cluster-like aggregates were overall within 15%.

Specific comments:

1) Wavelength should be addressed when the absolute value of MAC is mentioned.

Response: Thank you for your recommendation. Wavelength was addressed when the absolute value of MAC was mentioned.

2) Line 13, what do the ‘different core-shell structures’ mean? Different core size and shell thickness?

Response: Yes, ‘different core-shell structures’ meant different core sizes and shell thicknesses in this study. ‘Different core-shell structures’ was changed into ‘different core sizes and shell thicknesses’ in the revised manuscript to avoid ambiguity.

3) Line 57–58, Bond and Bergstrom (2006) just suggested a consistent MAC for fresh (uncoated) BC particles.

Response: This sentence was removed in the revised manuscript.

4) Line 73, a more detailed but clear description of BCPMSD measurement should be addressed. From my understanding, major results and discussion presented in this study are based on the BCPMSD measurements (using DMA-AE51?) at Zhangqiu site. DMA-SP2 measurements at Taizhou, and comparisons of AE33 with PASS-3 at Taizhou and Beijing are mostly used to provide essential parameters (e.g., number fraction of BC-containing particles, multi-scattering correction factor for AE33, etc.) for the BCPMSD retrieval.

Response: Thanks for your comments. More detailed description of BCPMSD measurement was addressed in section 2.2 in our revised manuscript.

Yes, the major results and discussion in this study were based on BCPMSD measurements using DMA-AE51 at Zhangqiu site. The SP2 measurements at Taizhou

as well as comparison between AE33 and PASS-3 were used to provide number fraction of BC-containing particles as well as multi-scattering correction factor for AE33.

5) Line 112–115, the method to determine the size-resolved number fraction of BC-containing particles should be introduced briefly. How to deal with the effect of multicharged particles in the DMA-SP2 system? Why the number fraction of BC-containing particles at Taizhou can be used to represent that at Zhangqiu?

Response: Thanks for your recommendation. The determination of the number fraction of BC-containing particle was introduced briefly in the text.

According to the study of Zhao et al. (2019), The peak height (H) of the aerosol scattering signal could be used to deal with multicharged particle. The probability distribution of H at a given selected mobility diameter had multiple modes, as Fig. 1 showed. The multiple modes corresponded to signals of multicharged particles and could be calculated with theory of DMA.

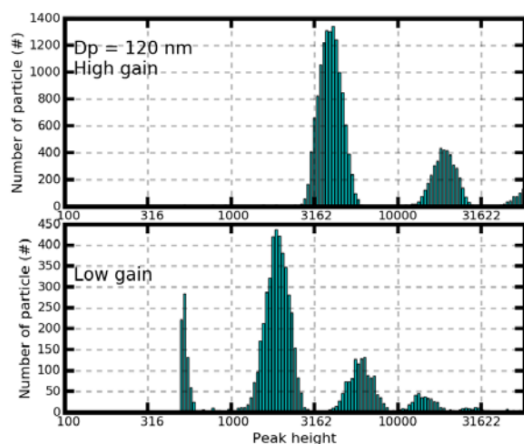


Figure 1. Figure S1 of the study by Zhao et al. (2019). The measured scattering signal distribution at diameter of 120 nm using ammonium sulfate.

Both Zhangqiu site (36°42'N, 117°30'E) and Taizhou site (32°35'N, 119°57'E) are in the east of China. They both experienced pollutions caused by industrialization and urbanization in the past several decades. Hence, the number fraction of BC-containing particle measured at Taizhou was representative and could be used as reference value for Zhangqiu.

6) Line 120, why absorption coefficients measured by AE33 are 2.9 times those measured by PASS-3? Does this ratio mean the multi-scattering effect of the filter loading method? However, as mention in line 106, a compensation factor of 2.6 has been introduced to mitigate multiple scattering effect. Was the PASS-3 well calibrated before the measurement?

Response: 2.9 was from the study by Zhao et al. (2020).

Yes, this ratio, namely the scattering correction factor, was used to correct multi-scattering effect.

In line 106, the factor of 2.6 was the scattering correction factor for AE51. And for AE33 was 2.9. We specified that 2.6 was for AE51.

7) Line 147, although the mantle chemical species would not influence largely the results presented in this study, BC/OM mixtures are more likely existed in the atmosphere of studied regions.

Response: Thanks for the comments. The wavelength used in this study was 880 nm. Previous study indicates aerosol absorption at 880 nm is mainly from BC (Ramachandran and Rajesh, 2007). Therefore, the influence of organic matter was neglected in this study.

References

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