Supplement for

Determination of black carbon mass concentration from aerosol light absorption using variable mass absorption cross-section

Weilun Zhao[1], Wangshu Tan[1], Gang Zhao[2], Chuanyang Shen[1], Yingli Yu[1], Chunsheng Zhao[1]

[1]Department of Atmospheric and Oceanic Sciences, School of Physics, Peking University, Beijing 100871, China

[2]State Key Joint Laboratory of Environmental Simulation and Pollution Control, College of Environmental Sciences and Engineering, Peking University, Beijing 100871, China

1 The schematic representation of the three different measurement systems

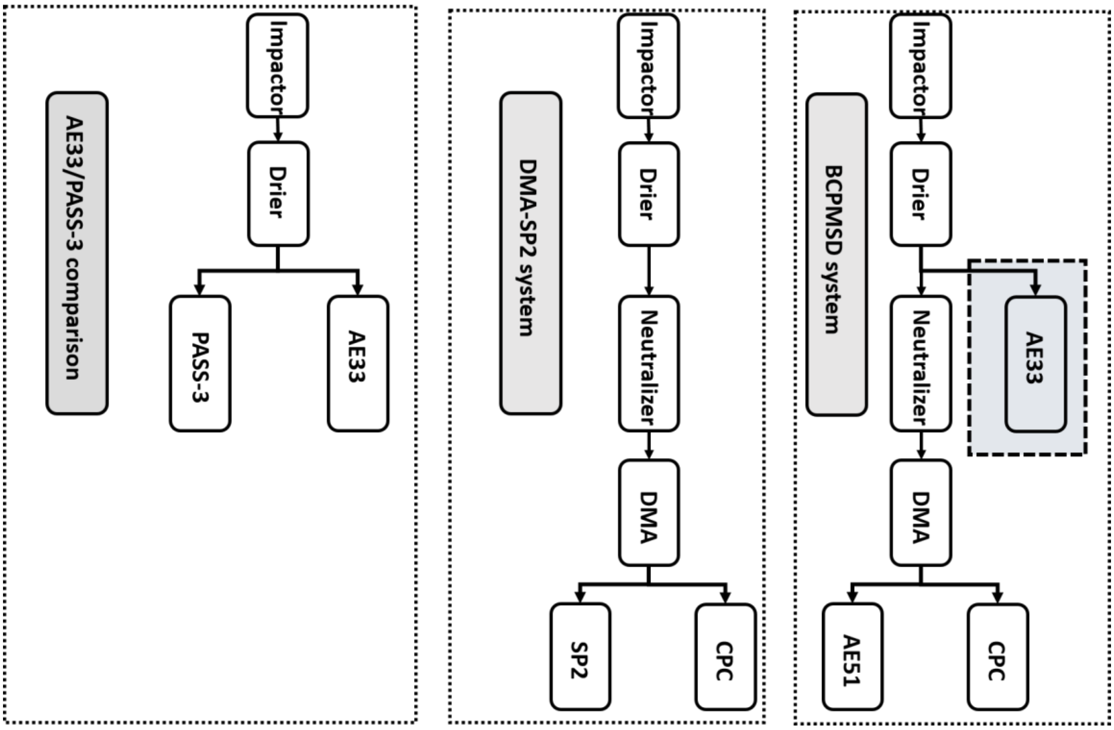

Figure S1. The schematic diagram of the AE33/PASS-3 comparison system, DMA-SP2 system and BCPMSD measurement system.

2 The uncertainties of derived BC mass concentration caused by using a constant BC-containing particle fraction

Figure.S2 shows the deviation of BC particle mass size distribution (BCPMSD) calculated from different BC-containing particle fractions (8.5%, 17%, 34%). We can see that for our newly proposed method, using a constant BC-containing particle fraction does not change the size-resolved distribution mode. There is still a finer mode and coarser mode with a boundary of 240 nm. Besides, the influence of using different BC-containing particle fractions to derived BC mass concentration (m_{BC}) is very limited when particles are larger than 200 nm. However, the deviations beween the m_{BC} derived from different fraction values are large when particles diameters are smaller than 200 nm. At this range, if the BC-containing particle fraction is underestimated, the m_{BC} will be underestimated. On the contrary, the m_{BC} is overeatimated if the fraction is overestimated.

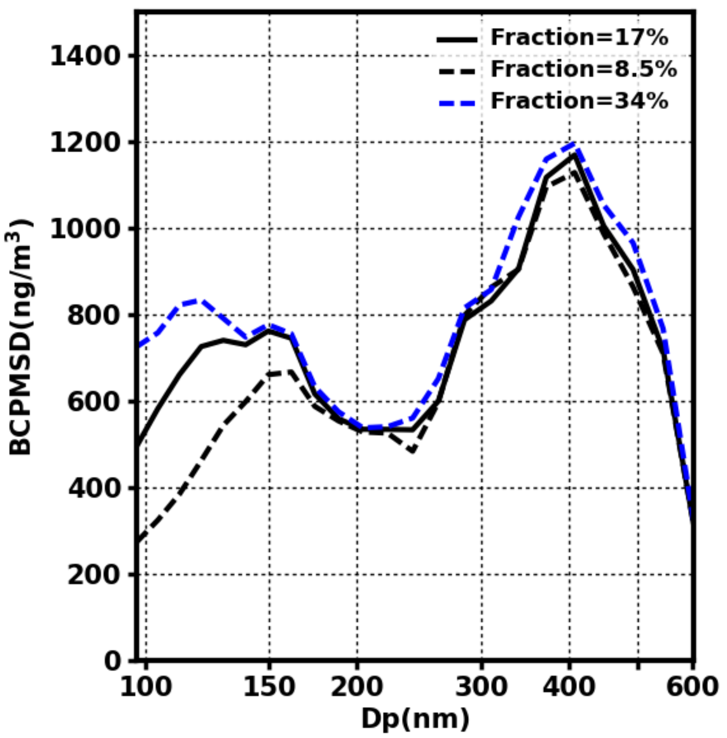

Figure S2. The derived BCPMSD by using different constant BC-containing particle fraction. The solid black line represents the result derived from a fraction of 17%. The dashed black line and blue line show the results derived from a fraction of half of 8.5% and double of 34%.

3 The uncertainties of MAC caused by using idealized core-shell model

3.1 The formation of BC aggregates with a determied morphology

The fractal aggregates of BC have been well described by fractal geometries through the well-known statistical scaling law (Sorensen, 2001):

$$\mathrm{N} = k_f \left(\frac{R_g}{a}\right)^{D_f}, \qquad (1)$$

where N is the number of “same-sized” monomers in the cluster, a is the monomer radius, D_f and k_f are known as the fractal dimension and fractal prefactor respectively, determining the morphology of BC cluster. The compactness of a fractal aggregate increses as the increase of D_f or k_f. R_g is the gyration radius, infering the overall aggregate radius, determined by :

$$\mathrm{R}_g = \sqrt{\frac{1}{N}\sum_{i=1}^{N} {r_i}^2}, \qquad (2)$$

where r_i represents the distance of the ith monomer from the center of mass of BC cluster.

In order to generate fractal-like aggregates with given N, R_g, a, D_f and k_f, the sequential algorithm proposed in Filippov et al. (2000) is introduced in this paper to add the primary monomers one by one. On condition that there is an aggregate including N-1 monomers, the Nth monomer is constantly placed randomly until it has at least one contact point with the previously attached N-1 monomers with no overlapping. Besides, the mass center of the next Nth monomer must obey the rule as follows:

$$(r_N - r_{N-1})^2 = \frac{N^2a^2}{N-1}\left(\frac{N}{k_f}\right)^{2/D_f} - \frac{Na^2}{N-1} - Na^2\left(\frac{N-1}{k_f}\right)^{2/D_f}, \qquad (3)$$

where r_{N-1} and r_N are the mass center of the first N-1 monomers and the Nth monomer, respectively. After the fractual configuration of BC aggregates, the absorption properties of BC containing particles need to be evaluate.

The fractal dimensions for aged BC aggregates are generally close to 3 (Kahnert et al., 2012). The aim of this study is to evaluate the effects of aerosol microphysics on the absorption enhancement of fully coated BC particles, which can be regarded as the aged BC aerosols. Therefore, the fractal dimension D_f is set to be 2.8 and k_f is generally set to be 1.2. The diameter of the primary monomers are usually between 20-50 nm and the number of the primary monomers for an aggregates is between 50-300. The size of BC core calcuated by the new method is smaller than 300 nm most of the time during Zhangqiu campaign. The diameter of primary monomers is set to be 50 nm and the number of the primary monomers for an aggregates ranges from 2 to 200, leading to the largest size of volume equivalent BC core close to 300 nm. Summary results indicate that the real part of BC is generally in the range of 1.5 to 2.0 while the imaginary part usually varies from 0.5 to 1.1 (Liu et al., 2018). Therefore, the mean value 1.75 for BC real part and 0.8 for BC imaginary part are adopted here to calculate MAC values for BC/sulfate mixtures at the wavelength of 880 nm.

3.2 Multiple Sphere T-matrix (MSTM) method

As the traditional Mie model is not available for the fractal aggregates, the widely used MSTM method is employed here to quantify the absorption properties of BC clusters (Mackowski and Mishchenko, 1996;Mackowski, 2014). The addition theorem of vector spherical wave functions is used in MSTM method to describe the mutual interactions among the system. The T-matrix of aggregates used to derive particle optical properties can be obtained from these individual monomers. MSTM method can calculate light scattering and absorption properties of the randomly oriented aggregates without numerical averaging over particle orientations if the position, size and refractive index of every spherical element are given. However, the MSTM method is only applicable to evaluate the aggregates of spheres without overlapping and it is carried out with high computational demand.

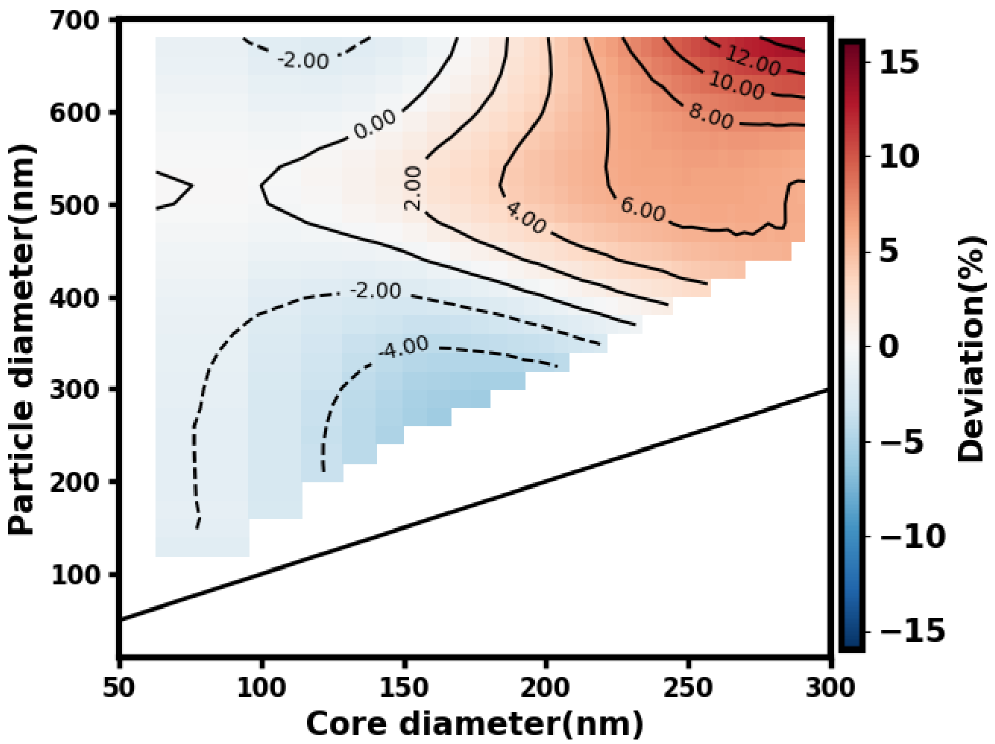

Figure S3. Relative deviations of MAC values calculated by idealized concentric core-shell model and letting BC particles be in the form of cluster-like aggregates. The solid line is 1:1 line.

The deviations showed in Fig.S3 are derived by subtracting MAC values calculated by MSTM model by those calculated by Mie model. The results indicate that most of the MAC values calculated by assuming BC particles are in the form of cluster-like aggregates are smaller when the size of BC core is smaller than 150 nm. When BC core is larger than 150 nm, the MAC values calculated by MSTM model increase with the thickness of shell and will be larger than those derived from concentric core-shell model. As we can see from Fig.S3, the deviations between MAC calculated by the idealized concentric core-shell model and letting BC particles be in the form of cluster-like aggregates are within 15%.

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