Supplement of Atmos. Meas. Tech., 15, 6965–6989, 2022
https://doi.org/10.5194/amt-15-6965-2022-supplement

Atmospheric Measurement Techniques
Open Access

Supplement of

Importance of size representation and morphology in modelling optical properties of black carbon: comparison between laboratory measurements and model simulations

Baseerat Romshoo et al.

Correspondence to: Baseerat Romshoo (baseerat@tropos.de)

1. Transmission electron microscopy (TEM) images of black carbon fractal aggregates from mini-CAST soot generator

Depending on the burning conditions of the flame as described by the flame equivalence ratio, the percentage of organic matter which forms an external layer around the BC may vary (Mamakos et al., 2018). Ouf et al., 2016 showed images of how the structure of the BC aggregate varied with the operating conditions of the mini-CAST burner. They operated the mini-CAST burner on three conditions producing BC aggregates with foc of 4% (CAST1), 47% (CAST2), and 87% (CAST3). The TEM images from the three cases are shown below.

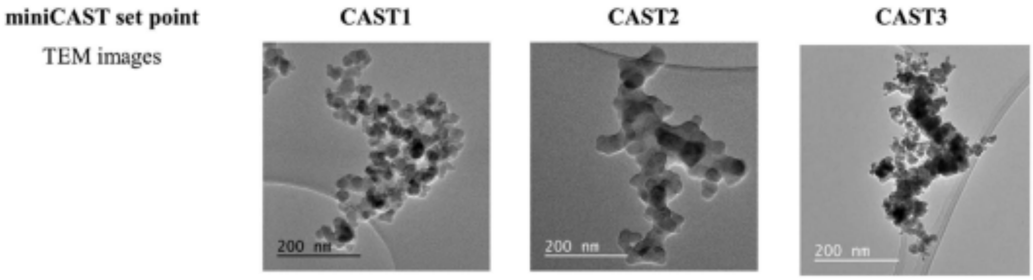


Fig S1. TEM images of soot particles produced by the miniCAST for various set points and analysed off-line on samples deposited on carbon-coated copper grids. Adapted from Ouf et al., 2016.

2. Transmission electron microscopy (TEM) images of ambient aerosols containing black carbon

When we study BC containing aerosols in the atmosphere, we find that, in addition to the nature of the emission source, factors such as the residence time and atmospheric chemistry have an impact on their morphology. During the same period, multiple kinds of BC-containing particles with differing levels of aging can exist simultaneously. Fu et al., 2011 presented the following image which illustrates different types of BC-containing particles collected from Shanghai's atmosphere. As seen in the picture below, BC can maintain fractal aggregate morphology even as it ages.

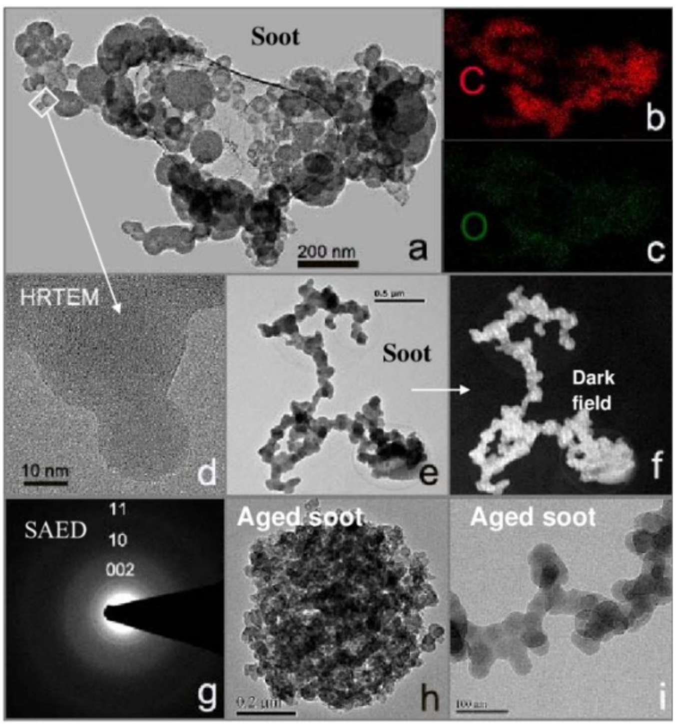


Fig S2. TEM images of soot particles collected from the atmosphere of Shanghai. Aerosols in the atmosphere contain a mixture of fresh, semi-aged, and aged aerosols that will have fractal morphology. Adapted from Fu et al., 2012.

3. Does Catalytic Stripper change the morphology of heated particles?

Our partners from the project at the Federal Institute of Metrology METAS, Switzerland took some TEM measurements of BC particles from the mini-CAST before and after the Catalytic Stripper. The tests were performed with 60 nm particles which typically have 20-25% OC/TC mass fraction. The images of the BC particles before and after the Catalytic Stripper are given in Fig S3 and S4, respectively. It was shown that there is no major morphological transformation after the BC particles are treated by the Catalytic Stripper. The BC particles remain fractal-like, and do not collapse to a compact structure.

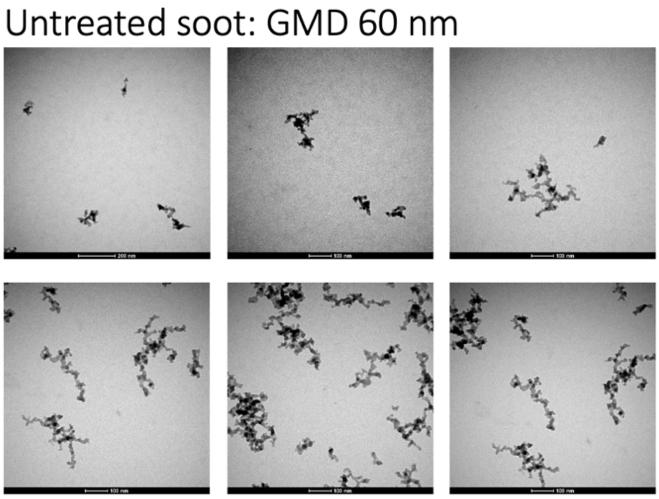


Fig S3. TEM images of black carbon particles collected before operating the Catalytic Stripper.

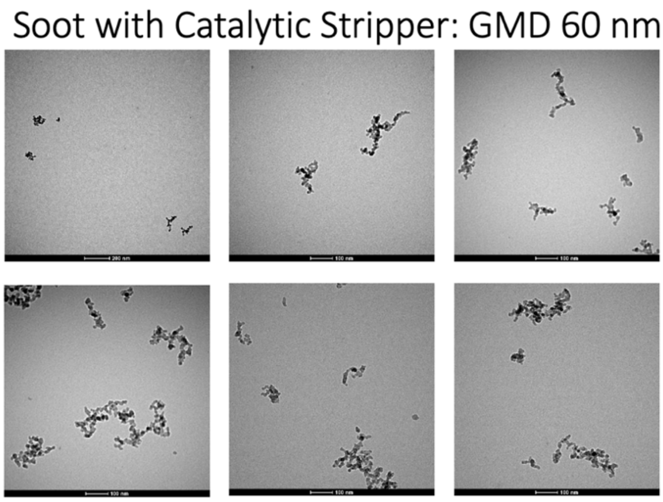


Fig S4. TEM images of black carbon particles collected after operating the Catalytic Stripper.

4. Comparison of absorption from different instruments

The absorption was also calculated using extinction minus scattering (ems), and compared to the MAAP and AE33. The extinction minus scattering (ems) shows better comparability to the MAAP.

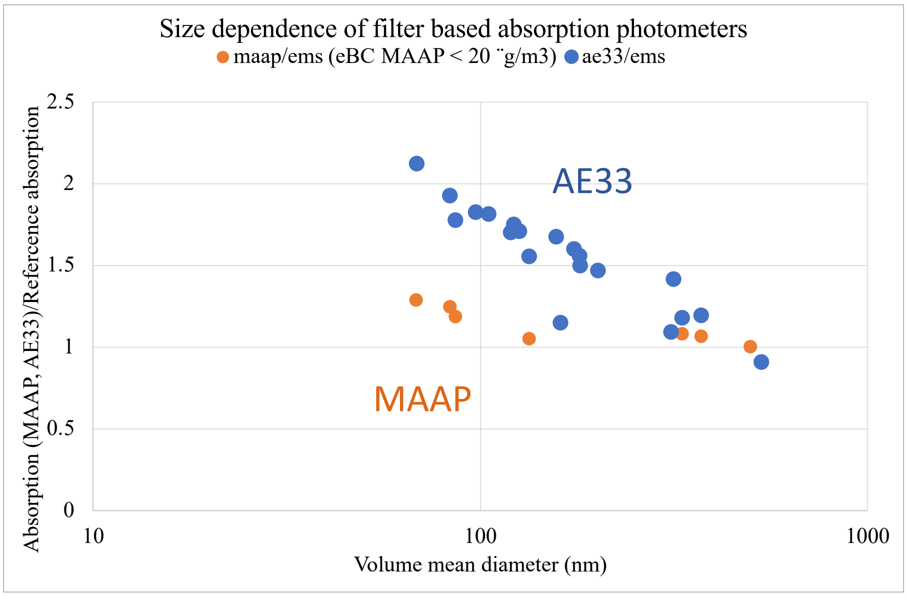
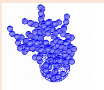
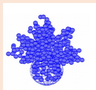
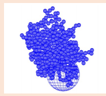


Fig S5. Absorption calculated using extinction minus scattering (ems) compared to the MAAP and AE33.

5. Case study of intersecting spheres in MSTM code

He et al. (2015) studied various morphologies of black carbon aggregates with sulfuric acid (H_2SO_4) coating using the Geometric Optics Surface-wave (GOS) approach. In our manuscript, the case of 'aggregate and sphere' representation (Fig. 3f) matches morphologically to the case of BC aggregates partially embedded in a sulfuric acid (H_2SO_4) coating of the study by He et al. (2015). In order to test the results from the MSTM, the BC aggregates partially embedded in a sulfuric acid (H_2SO_4) coating were regenerated and their optical properties were calculated using MSTM. The specifications of the particles regenerated from the study of He at al. (2015) are summarized in Table S1. In the Fig. S1, the results from the MSTM for the particles in Table S1 are plotted over the figure copied from He at al. (2015). The results from the MSTM for the all the three cases of BC aggregates partially embedded in a sulfuric acid (H_2SO_4) coating closely match the GOS approach values. Therefore, the MSTM code was considered good enough for modeling the optical properties of aggregates with few intersecting spheres. It must be noted that if the number of intersecting spheres increase significantly, the difference between the MSTM and GOS approach might change.

Table S1. Details of the aggregates with spherical sulfuric acid coating modelled by He at al. (2015). In the case of the coated aggregate: values of mobility diameter of the core (BC D_p), number of primary particles (N_{pp}), radius of the primary particle (a_{pp}), and radius of the spherical coating. He et al. (2015) normalized each calculation with the results from heir equivalent core-shell model. In the case of core-shell model: outer radius and inner radius.

Sphere embedded in aggregate	BC D_p *(nm)*	155	245	320
	N_{pp}	164	416	651
	a_{pp} *(nm)*	7.5	7.5	7.5
	D_f	2.1	2.1	2.1
	Coating sphere radius (nm)	3.5	55.5	65
	Image			
Core – shell model	Outer Radius (nm)	80.8	126.18	163.49
	Inner Radius (nm)	77.5	122.5	160

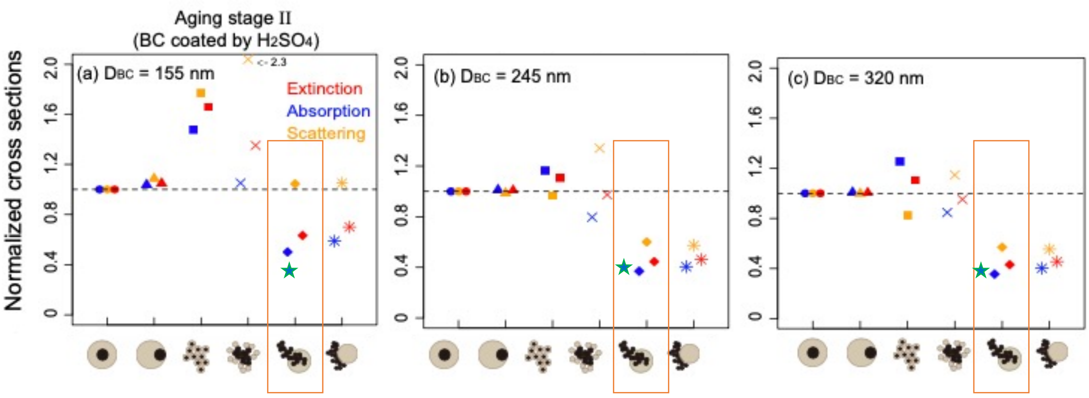


Figure S6. Comparison of the absorption results using MSTM for the three cases from He at al. (2015). The case for which the comparison is made is highlighted in a orange box. The blue star point represents the calculation using the MSTM code which should be compared to the blue diamond point of He at al. (2015). Adapted from He at al. (2015).

6. Modelled optical properties from denuding experiment E1 – bare black carbon

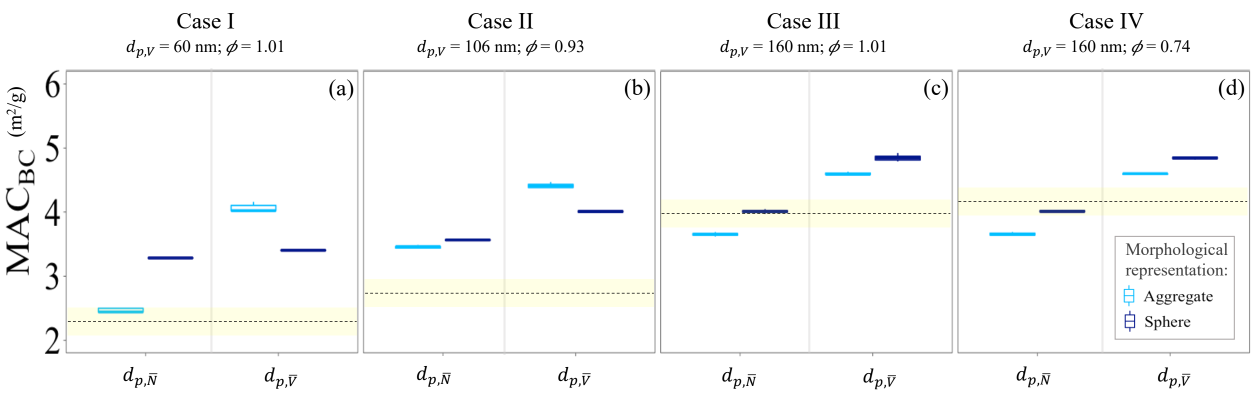


Figure S7. The modelled BC mass absorption cross-section (MAC_{BC}) compared to the measured MAC_{BC} for the four experiments of E1.

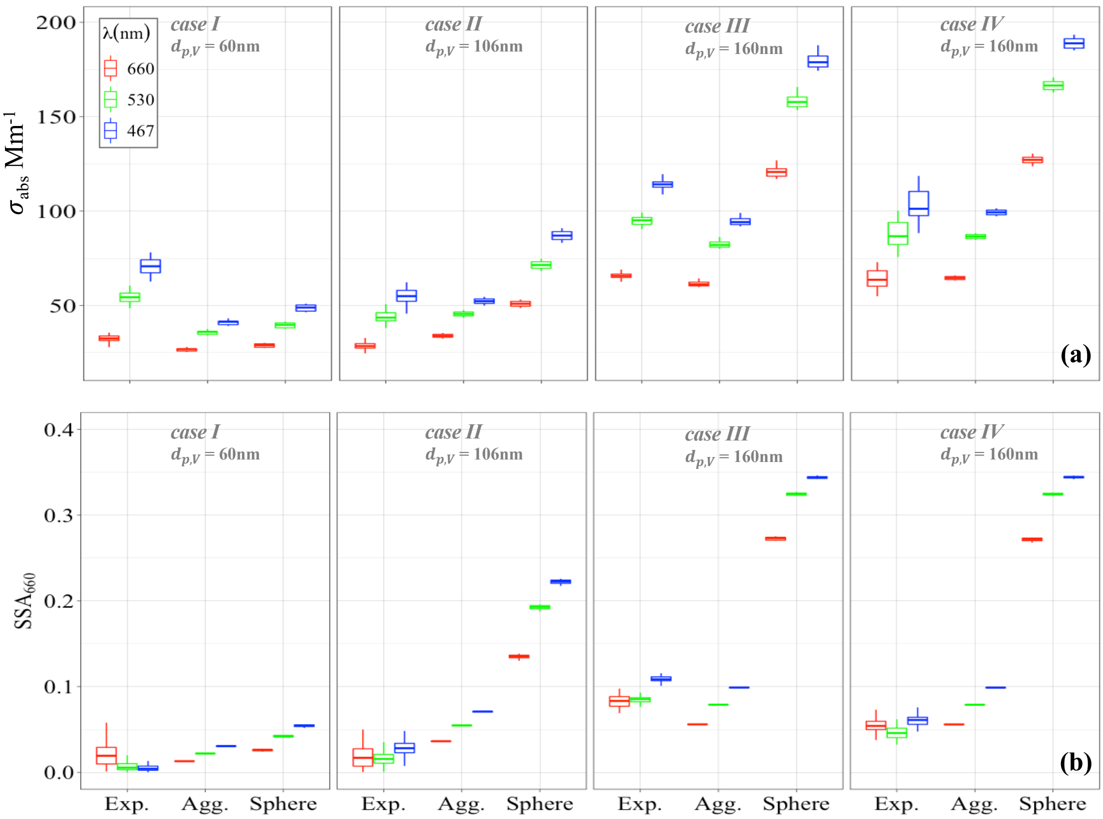


Figure S8. Modelled optical properties for three wavelengths in the visible range (467, 530, and 660 nm) using the polydisperse method. Panels **(a)** and **(b)** shows the results of absorption coefficient σ_{abs} and single scattering albedo *SSA,* respectively, for the cases *I* – *IV* of E1 using the "aggregate" and "sphere" representation. For each case, the modelled optical property is compared to the experimentally measured values marked as "Exp." on the X-axis.

7. Modelled optical properties from experiment E2 – black carbon containing organic

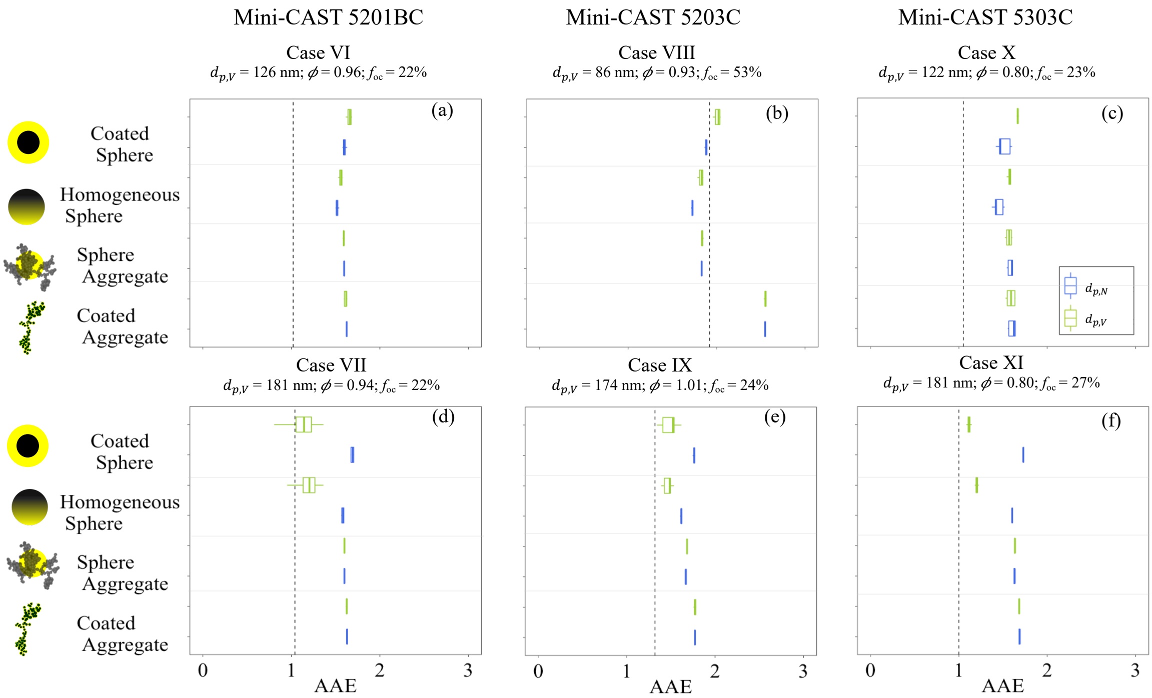


Figure S9. Modelled AAE from various cases of mini-CAST generators in E2 summarized in Table 1. The results are shown for: mini-CAST 5201BC $d_{p,\overline{V}}$ = 126 nm **(a);** mini-CAST 5201BC $d_{p,\overline{V}}$ = 181nm **(d);** mini-CAST 5203C $d_{p,\overline{V}}$ = 86 nm **(b);** mini-CAST 5203C $d_{p,\overline{V}}$ = 174 nm **(e);** mini-CAST 5303C $d_{p,\overline{V}}$ = 122 nm **(c);** mini-CAST 5303C $d_{p,\overline{V}}$ = 181 nm **(f)**. In each panel, the AAE is modelled using four coated BC representations 'coated sphere', 'homogeneously mixed sphere', 'coated aggregate', and 'aggregate and sphere'. The mean of the experimentally measured AAE is shown by the black dashed line in each panel.

References

Mamakos, A., Khalek, I., Giannelli, R. and Spears, M.: Characterization of combustion aerosol produced by a mini-CAST and treated in a catalytic stripper, Aerosol Science and Technology, 47(8), 927–936, doi:10.1080/02786826.2013.802762, 2013.

Ouf, F. X., Parent, P., Laffon, C., Marhaba, I., Ferry, D., Marcillaud, B., Antonsson, E., Benkoula, S., Liu, X. J., Nicolas, C., Robert, E., Patanen, M., Barreda, F. A., Sublemontier, O., Coppalle, A., Yon, J., Miserque, F., Mostefaoui, T., Regier, T. Z., Mitchell, J. B. A. and Miron, C.: First in-flight synchrotron X-ray absorption and photoemission study of carbon soot nanoparticles, Scientific Reports, 6, doi:10.1038/srep36495, 2016.

Fu, H., Zhang, M., Li, W., Chen, J., Wang, L., Quan, X., and Wang, W.: Morphology, composition and mixing state of individual carbonaceous aerosol in urban Shanghai, Atmos. Chem. Phys., 12, 693–707, https://doi.org/10.5194/acp-12-693-2012, 2012.

He, C., Liou, K. N., Takano, Y., Zhang, R., Levy Zamora, M., 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(20), 11967–11980, doi:10.5194/acp-15-11967-2015, 2015.