Supplement to "Radiative properties of coated black carbon aggregates: numerical simulations and radiative forcing estimates"

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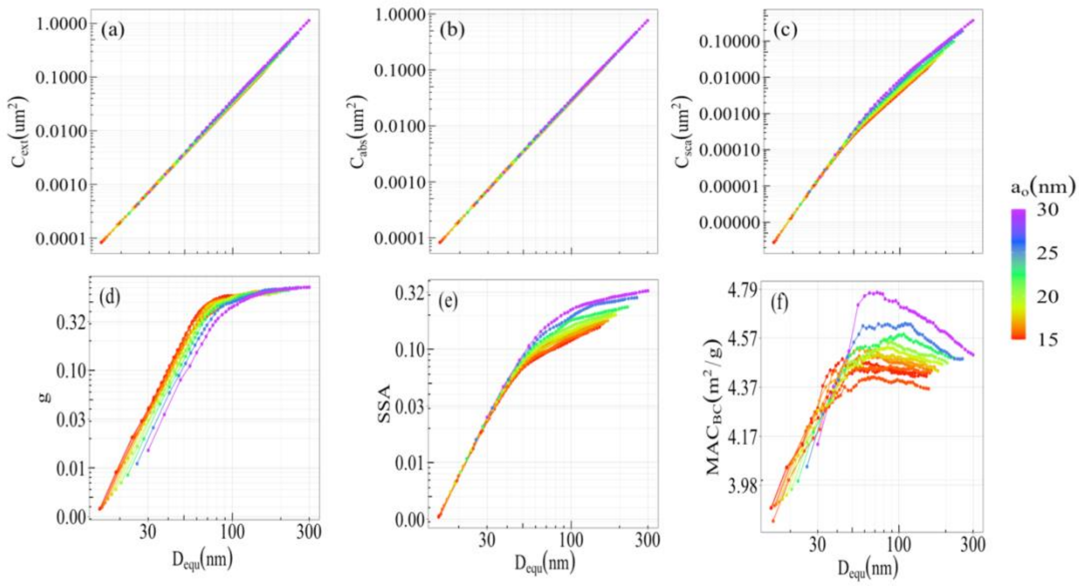

Figure S1. Optical properties of pure BCFAs at various radius of primary particle (a_o) with respect to volume equivalent radius (R_{equ}): extinction cross-section C_{ext} (a), absorption cross-section C_{abs} (b), scattering cross-section C_{sca} (c), asymmetry parameter g (d), single scattering albedo SSA (e), and black carbon mass absorption cross-section MAC_{BC} (f) at λ = 660nm.

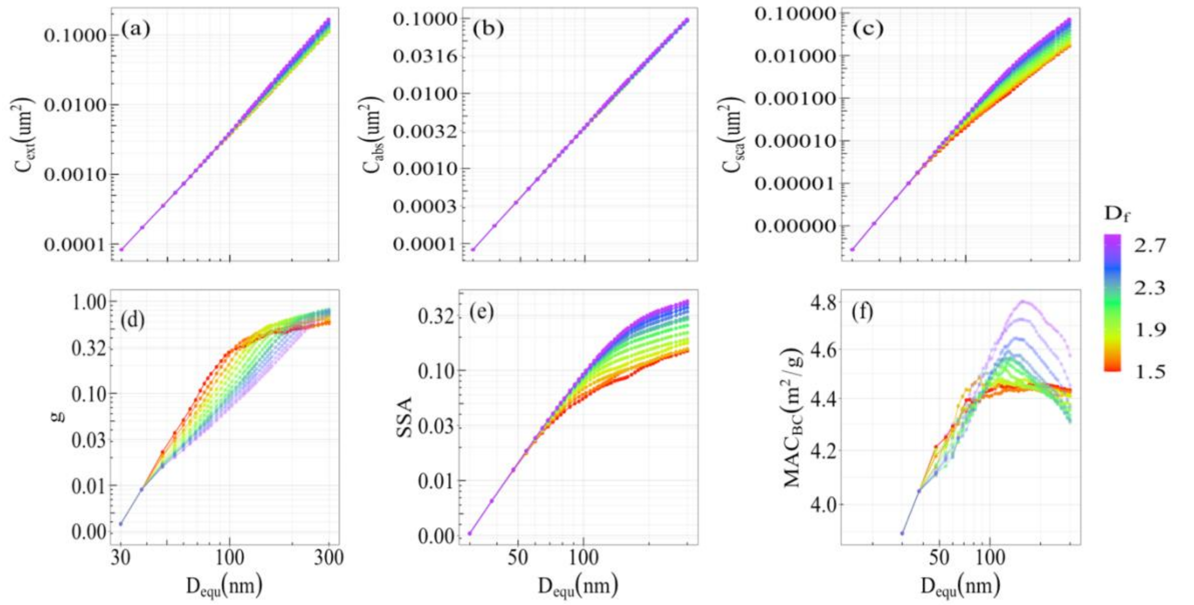

Figure S2. Optical properties of pure BCFAs as a function of volume equivalent radius (R_{equ}) at various fractal dimension (D_f): extinction cross-section C_{ext} (a), absorption cross-section C_{abs} (b), scattering cross-section C_{sca} (c), asymmetry parameter g (d), single scattering albedo SSA (e), and black carbon mass absorption cross-section MAC_{BC} (f) at λ = 660nm.

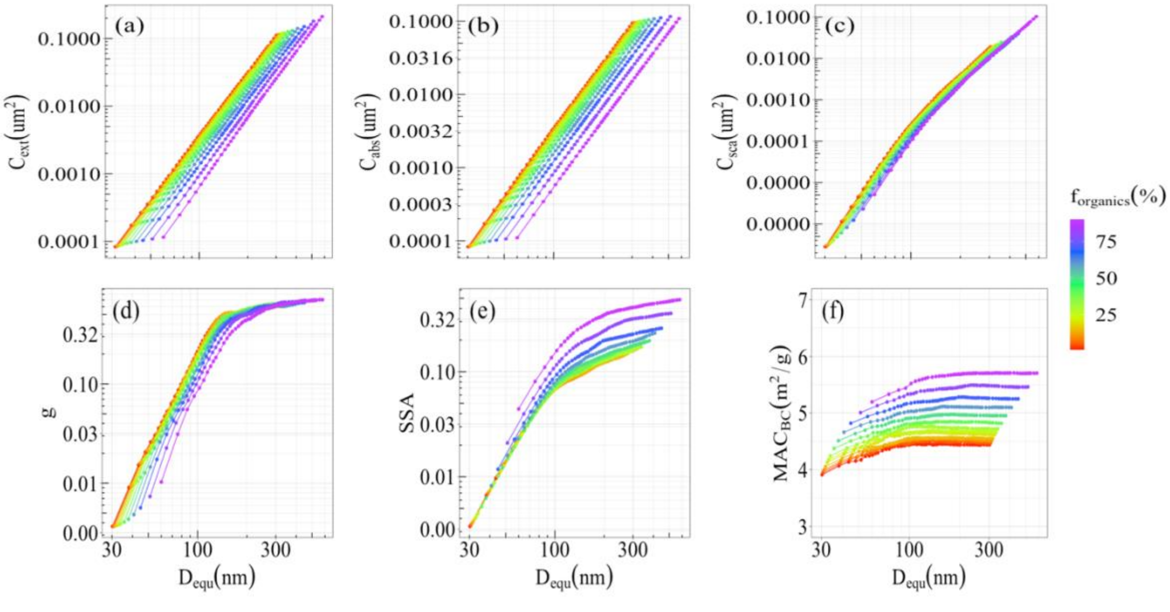

Figure S3. Optical properties of BCFAs (D_f = 1.7) as a function of volume equivalent radius (R_{equ}) at various fraction of organics ($f_{organics}$): extinction cross-section C_{ext} (a), absorption cross-section C_{abs} (b), scattering cross-section C_{sca} (c), asymmetry parameter g (d), single scattering albedo SSA (e), and black carbon mass absorption cross-section MAC_{BC} (f) at λ = 660nm.

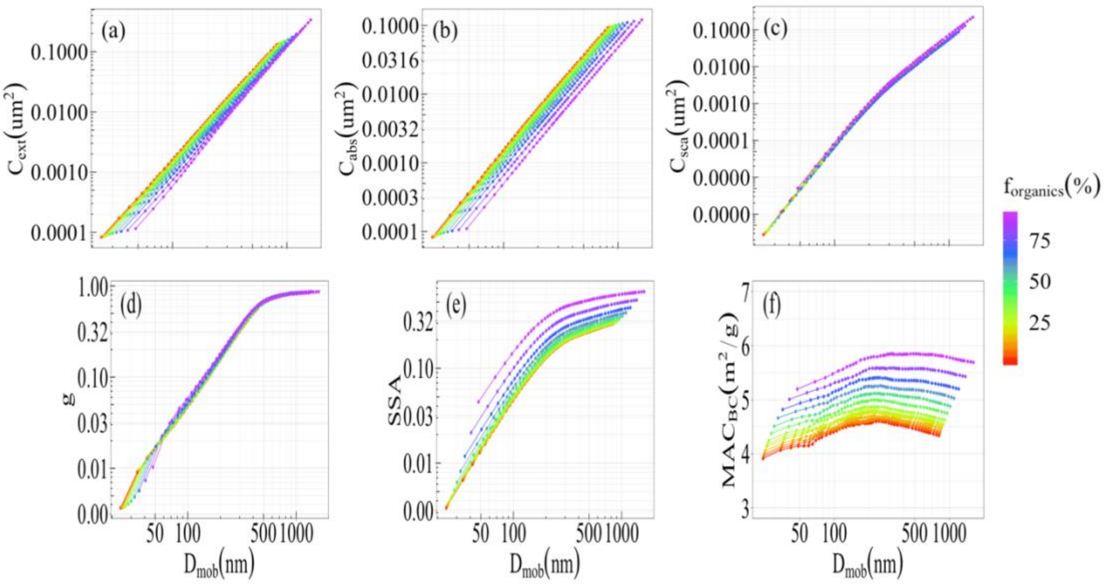

Figure S4. Optical properties of BCFAs (D_f = 2.2) as a function of D_{mob} at various fraction of organics ($f_{organics}$): extinction cross-section C_{ext} (a), absorption cross-section C_{abs} (b), scattering cross-section C_{sca} (c), asymmetry parameter g (d), single scattering albedo *SSA* (e), and black carbon mass absorption cross-section MAC_{BC} (f) at λ = 660nm.

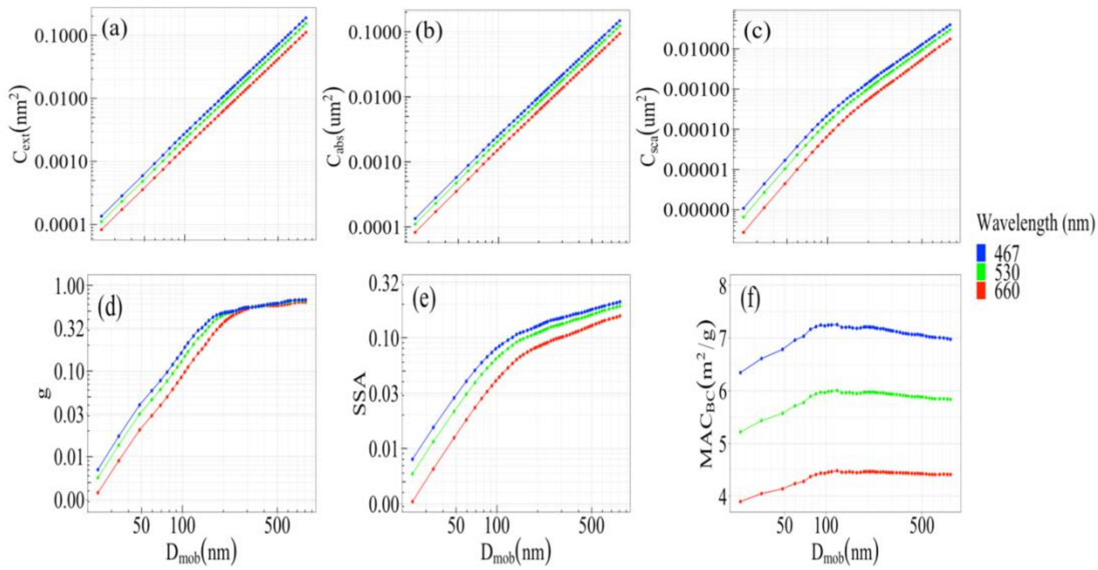

Figure S5. Optical properties of pure BCFAs (D_f = 1.7) as a function of D_{mob} at various wavelengths (λ): extinction cross-section C_{ext} (a), absorption cross-section C_{abs} (b), scattering cross-section C_{sca} (c), asymmetry parameter g (d), single scattering albedo *SSA* (e), and black carbon mass absorption cross-section MAC_{BC} (f).

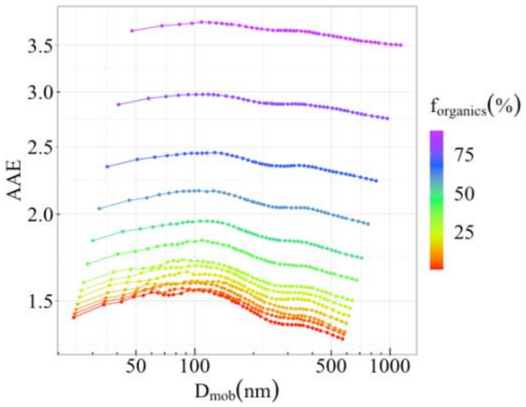

Figure S6. Ångström Absorption Exponent (AAE) of slightly compact BCFAs (D_f = 2.2) with changing fraction of organics ($f_{organics}$) and mobility diameter (D_{mob}).

Bulk optical properties of BCFAs

For application over the atmosphere, it is more meaningful to know the averaged optical properties over a certain particle size distribution, i.e. the bulk optical properties. Bulk optical properties of BC are useful since they can be used directly for simulations of radiative forcing. The bulk optical properties of BCFAs are calculated with an assumption that the radii of the volume equivalent spheres of BCFAs follow a lognormal size distribution:

$$n(r) = \frac{1}{\sqrt{2\pi r}\ \ln(\sigma)} exp\left[-\left[\frac{\ln(r)-\ln(r_o)}{\sqrt{2}\ln(\sigma)}\right]^2\right], \quad (1)$$

where r is the radius of a volume equivalent sphere corresponding to an aggregate, r_o is the geometric mean radius, and σ is the standard deviation in $\ln(r)$. Following a typical lognormal size distribution, the values of r_o and σ are fixed to 0.12µm and 1.5, respectively (Chung et al., 2011).

Table 4 shows the bulk optical properties of BCFAs for various compositions and morphologies at a wavelength of 530 nm. For each case, the bulk optical properties are calculated as integrals over the lognormal size distribution $n(r)$ with limits of r varying from 0.015 to 0.30µm, and the corresponding MSTM calculated values. The calculations are done following the mathematical formulas summarised by Li et al., 2016.

Table S1. Bulk optical properties of black carbon for different fraction of organics ($f_{organics}$) and fractal dimension (D_f) at a wavelength of 530nm.

	$f_{organics}$ (%)													
	0	1	5	10	15	20	25	30	40	50	60	70	80	90
D_f = 1.7														
C_{ext} (×10^{-2}µm^{-2})	2.94	2.96	3.02	3.03	3.04	3.12	3.12	3.14	3.18	3.14	3.08	2.92	2.74	2.35
C_{abs} (×10^{-2}µm^{-2})	2.54	2.55	2.58	2.60	2.62	2.65	2.66	2.67	2.66	2.60	2.48	2.25	1.86	1.35
C_{sca} (×10^{-2}µm^{-2})	0.40	0.41	0.44	0.43	0.42	0.47	0.47	0.47	0.52	0.54	0.61	0.67	0.88	1.00
SSA	0.14	0.14	0.15	0.14	0.14	0.15	0.15	0.15	0.16	0.17	0.20	0.23	0.32	0.43
g	0.60	0.58	0.57	0.60	0.59	0.59	0.61	0.57	0.62	0.60	0.60	0.62	0.64	0.63
D_f =2.2														
C_{ext} (×10^{-2}µm^{-2})	0.03	0.03	0.03	0.04	0.04	0.04	0.04	0.04	0.04	0.04	0.04	0.04	0.03	0.03
C_{abs} (×10^{-2}µm^{-2})	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.02	0.02	0.02	0.01
C_{sca} (×10^{-2}µm^{-2})	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.02	0.02
SSA	0.25	0.26	0.26	0.26	0.26	0.27	0.27	0.27	0.29	0.31	0.33	0.37	0.45	0.57
G	0.70	0.69	0.70	0.70	0.71	0.71	0.72	0.72	0.73	0.73	0.75	0.76	0.77	0.75

Parametrization for bulk optical properties

An important application of the optical parametrization scheme would be calculation of the bulk optical properties of atmospheric BC aerosols. The bulk optical properties can be calculated between any limits of mobility diameter

(D_{mob}), for the cases of fractal dimension (D_f), fraction of organics ($f_{organics}$), and wavelength (λ) provided in the parametrization scheme. Following Eq. (1), it must be assumed that the mobility diameter (D_{mob}) of the BCFAs follow a log normal distribution:

$$n(D_{mob}) = \frac{1}{\sqrt{2\pi D_{mob}}\ln(\sigma)} exp\left[-\left[\frac{\ln(D_{mob})-\ln(\overline{D_{mob}})}{\sqrt{2}\ln(\sigma)}\right]^2\right], \quad (2)$$

where $\overline{D_{mob}}$ is the arithmetic mean diameter, and σ is the standard deviation in $\ln(D_{mob})$.

The bulk extinction cross-section is given by:

$$\langle C_{ext}\rangle = \int_{D_{mob}^{min}}^{D_{mob}^{max}} C_{ext}(D_{mob}, D_f, f_{organics}, \lambda)\ n(D_{mob})\ d(D_{mob}), \quad (3)$$

using the parametrisation scheme for extinction cross-section,

$$\langle C_{ext}\rangle = \int_{D_{mob}^{min}}^{D_{mob}^{max}} (e^{c_0} + e^{c_1} D_{mob})\ n(D_{mob})\ d(D_{mob}). \quad (4)$$

The values of the coefficients C_0 and C_1 must be chosen from their tabulated values for various cases of fractal dimension (D_f), fraction of organics ($f_{organics}$), and wavelength (λ) provided in the section of supplementary below.

Similarly, using the parametrization schemes provided, the corresponding bulk optical properties can be calculated as:

$$\langle C_{abs}\rangle = \int_{D_{mob}^{min}}^{D_{mob}^{max}} (e^{g_0} + e^{g_1} D_{mob})\ n(D_{mob})\ d(D_{mob}), \quad (5)$$

$$\langle C_{sca}\rangle = \int_{D_{mob}^{min}}^{D_{mob}^{max}} (e^{H_0} + e^{H_1} D_{mob} + e^{H_2} lnD_{mob})\ n(D_{mob})\ d(D_{mob}), \quad (6)$$

$$\langle SSA\rangle = \int_{D_{mob}^{min}}^{D_{mob}^{max}} (e^{k_0} + e^{k_1} D_{mob} + e^{k_2} lnD_{mob})\ n(D_{mob})\ d(D_{mob}), \quad (7)$$

$$\langle g\rangle = \int_{D_{mob}^{min}}^{D_{mob}^{max}} (e^{s_0} + e^{s_1} D_{mob} + e^{s_2} D_{mob}^2 + e^{s_3} D_{mob}^3)\ n(D_{mob})\ d(D_{mob}). \quad (8)$$

The relative errors between the bulk optical properties from the MSTM calculations and the parametrisation scheme are comparable to that in the case of single sized BCFAs shown in the section 3.7.1.