



Supplement of

Optical properties of coated black carbon aggregates: numerical simulations, radiative forcing estimates, and size-resolved parameterization scheme

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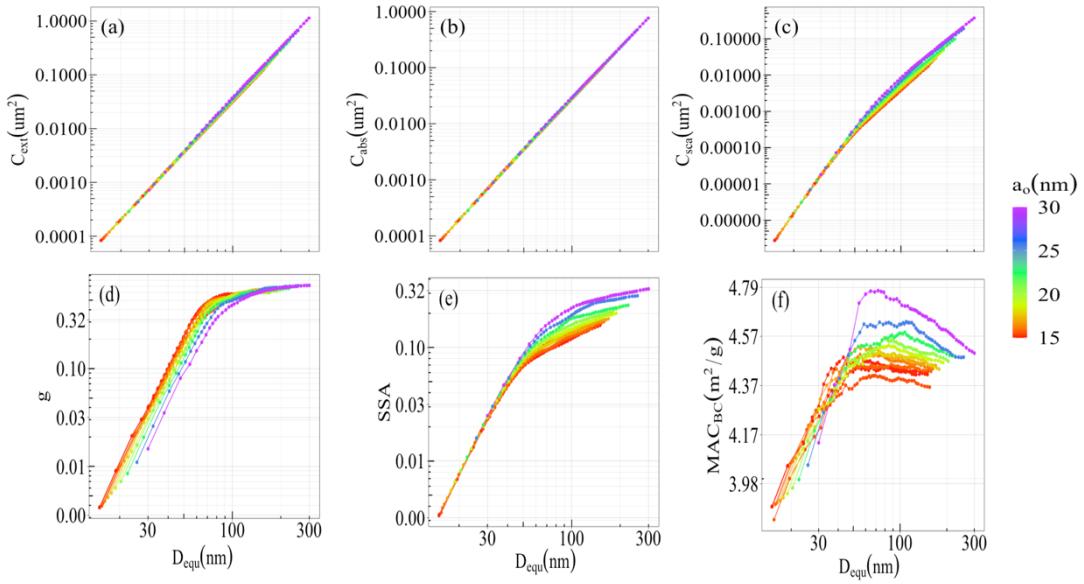


Figure S1. Optical properties of pure BCFAs for various radii of primary particles (a_o) as a function of the volume equivalent radius (R_{equ}): (a) extinction cross-section C_{ext} , (b) absorption cross-section C_{abs} , (c) scattering cross-section C_{sca} , (d) asymmetry parameter g , (e) single scattering albedo SSA , and (f) black carbon mass absorption cross-section MAC_{BC} at $\lambda = 660\text{nm}$.

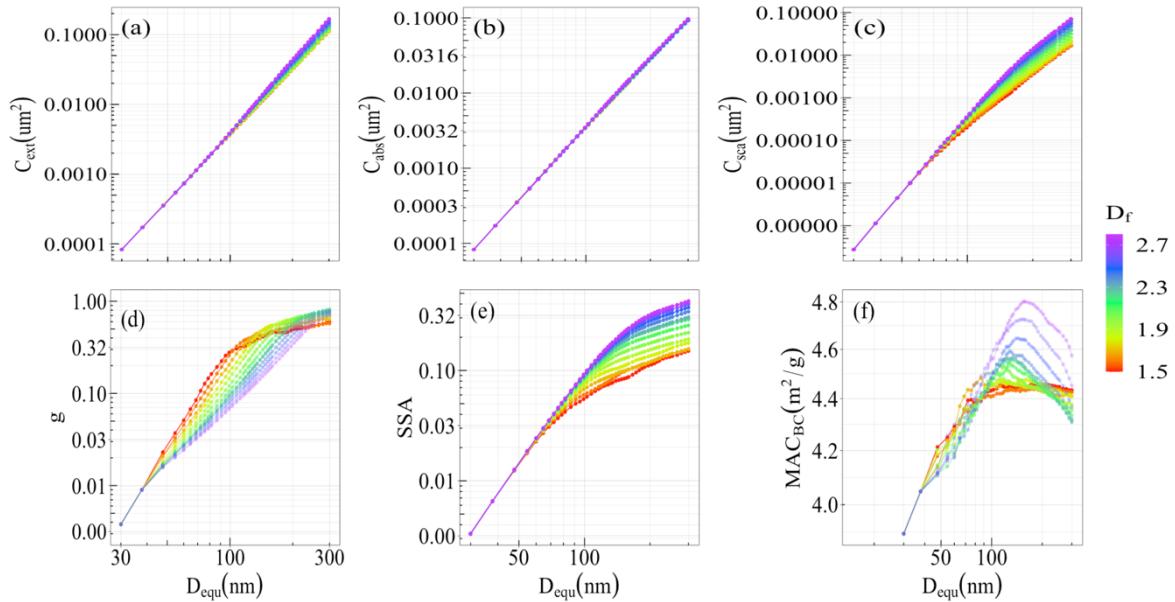
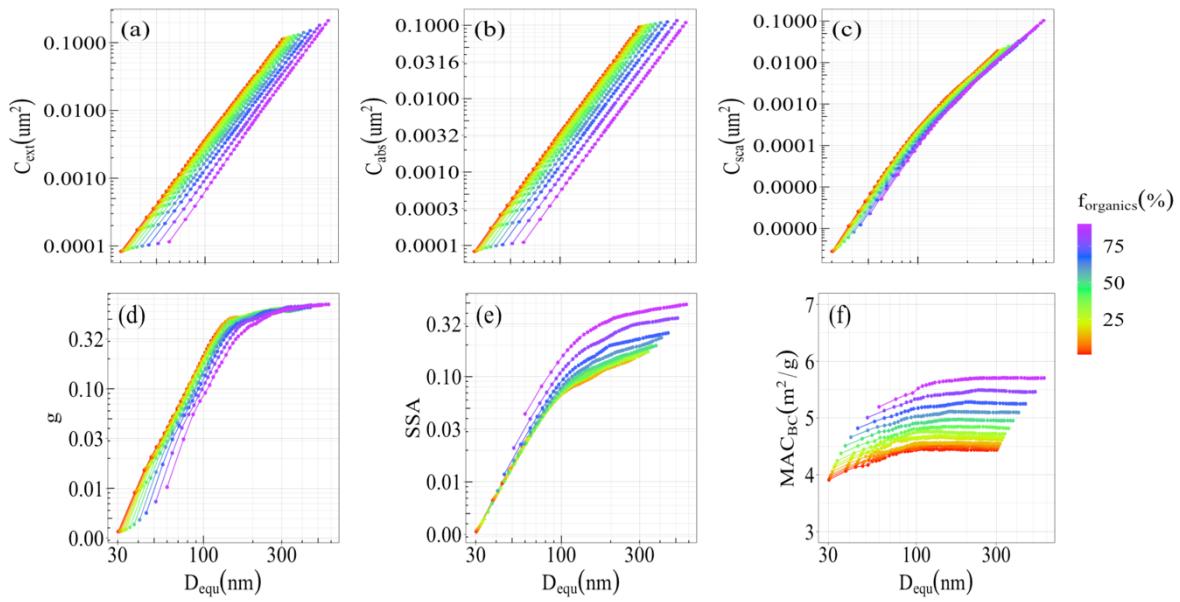
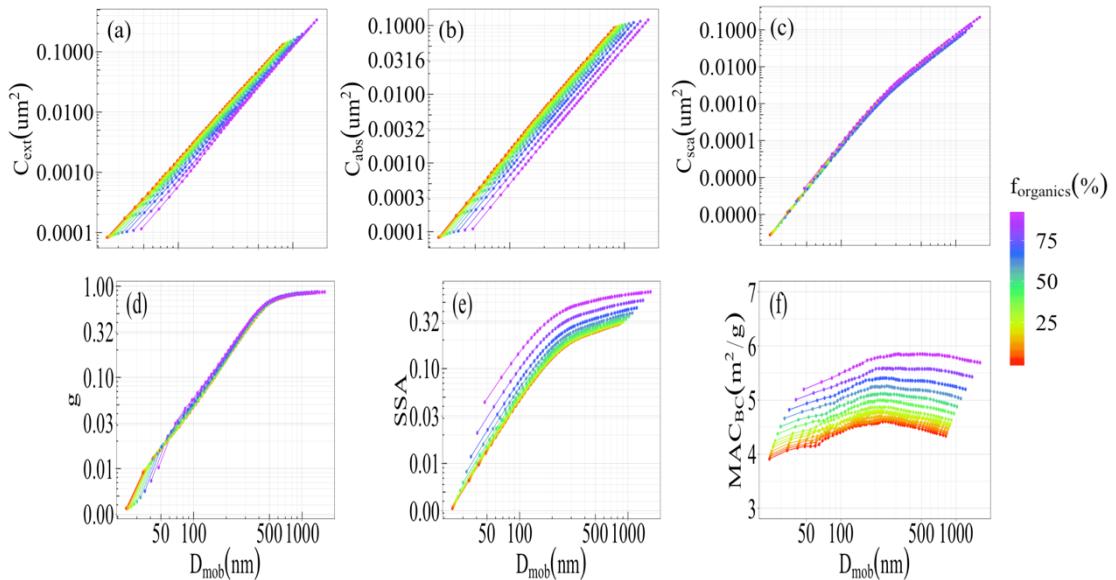


Figure S2. Optical properties of pure BCFAs as a function of volume equivalent radius (R_{equ}) for various fractal dimension (D_f): (a) extinction cross-section C_{ext} , (b) absorption cross-section C_{abs} , (c) scattering cross-section C_{sca} , (d) asymmetry parameter g , (e) single scattering albedo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



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

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

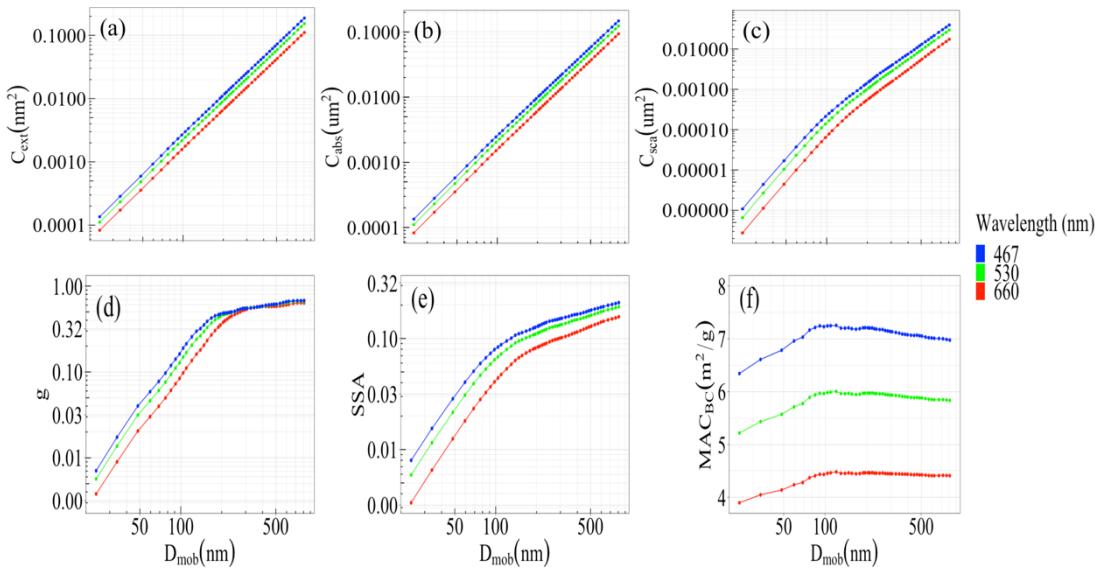


Figure S5. Optical properties of pure BCFAs ($D_f = 1.7$) as a function of D_{mob} for various wavelengths (λ): (a) extinction cross-section C_{ext} , (b) absorption cross-section C_{abs} , (c) scattering cross-section C_{sca} , (d) asymmetry parameter g , (e) single scattering albedo SSA , and (f) black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

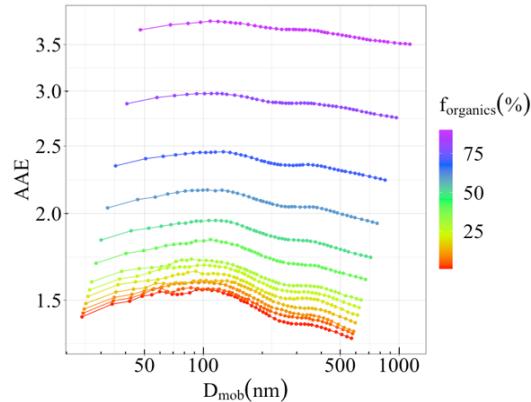


Figure S6. Ångström Absorption Exponent (AAE) of slightly compact BCFAs ($D_f = 2.2$) with changing fraction of organics ($f_{organics}$) and mobility diameter (D_{mob}).

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 mobility diameter D_{mob} of BCFAs follow a lognormal size distribution:

$$n(D_{mob}) = \frac{1}{\sqrt{\pi D_{mob}} \ln(\sigma)} \exp \left[- \left(\frac{\ln(D_{mob}/2) - \ln(D_{mob_0}/2)}{\sqrt{2} \ln(\sigma)} \right)^2 \right], \quad (1)$$

where D_{mob_0} is the geometric mean mobility diameter, and σ is the standard deviation in $\ln(D_{mob})$.

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 $n(D_{mob})$ with limits of D_{mob} varying from 0.030 to 0.60 μm, and the corresponding MSTM calculated values. The calculations are done following the mathematical formulas summarised in Li et al (2016).

55 **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.

	$f_{\text{organics}} (\%)$													
	0	1	5	10	15	20	25	30	40	50	60	70	80	90
$D_f = 1.7$														
$C_{\text{ext}} (\times 10^{-2} \mu\text{m}^{-2})$	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
$C_{\text{abs}} (\times 10^{-2} \mu\text{m}^{-2})$	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
$C_{\text{sca}} (\times 10^{-2} \mu\text{m}^{-2})$	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
$D_f = 2.2$														
$C_{\text{ext}} (\times 10^{-2} \mu\text{m}^{-2})$	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
$C_{\text{abs}} (\times 10^{-2} \mu\text{m}^{-2})$	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
$C_{\text{sca}} (\times 10^{-2} \mu\text{m}^{-2})$	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

60 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:

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

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

70 The bulk extinction cross-section is given by:

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

using the parametrization scheme for extinction cross-section,

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

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.

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

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

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

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

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

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

Parametrization scheme for optical properties of BCFAs

$$\ln C_{ext} = c_0 + c_1 \ln D_{mob}$$

105 $\ln C_{abs} = g_0 + g_1 \ln D_{mob}$

$$\ln C_{sca} = H_0 + H_1 \ln D_{mob} + H_2 \ln (\ln D_{mob})$$

$$\ln SSA = k_0 + k_1 \ln D_{mob} + k_2 \ln (\ln D_{mob})$$

110 $\ln g = \sum_{n=0}^3 s_n \ln D_{mob}^n$

Table S2. Parameterization scheme for coated and non-coated black carbon fractal aggregates (BCFA).

λ	D _f	f _{organics}	c0	c1	g0	g1	H0	H1	H2	s0	s1	s2	s3	k0	k1	k2
660	1.5	0	-15.7525	2.0244	-15.4847	1.9605	-24.2159	1.9921	3.5801	-46.0080	21.3332	-3.3873	0.1816	-8.8739	-0.1242	4.1179
660	1.5	5	-15.7550	2.0214	-15.5059	1.9597	-31.3601	0.2872	13.3115	0.6842	-2.4337	0.6085	-0.0406	-14.9613	-1.5899	12.4673
660	1.5	25	-15.8438	2.0177	-15.6260	1.9616	-24.6220	1.6677	4.8432	9.8861	-7.6666	1.5889	-0.1010	-8.5946	-0.3101	4.6065
660	1.5	50	-16.1005	2.0298	-15.8183	1.9591	-21.4000	2.3674	0.6833	-33.6550	14.4609	-2.1386	0.1072	-6.1086	0.1704	1.6966
660	1.5	70	-16.4923	2.0594	-16.1146	1.9624	-26.7081	1.2486	7.4745	-22.5955	8.5255	-1.0988	0.0475	-10.3414	-0.8352	7.6265
660	1.6	0	-15.7675	2.0278	-15.4998	1.9626	-30.8563	0.4502	12.5063	-51.4395	23.8510	-3.7532	0.1980	-14.7285	-1.4969	12.0341
660	1.6	5	-15.7611	2.0214	-15.5484	1.9657	-26.0321	1.3267	6.8446	-67.6214	31.3213	-4.9038	0.2572	-10.3641	-0.7155	6.9666
660	1.6	25	-15.8199	2.0167	-15.5835	1.9543	-29.1829	0.5997	11.0671	15.0126	-11.1566	2.3105	-0.1478	-13.2172	-1.3853	10.8791
660	1.6	50	-16.0327	2.0207	-15.7920	1.9548	-23.5291	1.7441	4.0211	-28.7900	10.8963	-1.3692	0.0558	-7.9102	-0.3621	4.5393
660	1.6	70	-16.3849	2.0426	-16.0953	1.9595	-23.9888	1.6021	4.7863	-52.7974	22.7540	-3.3130	0.1616	-7.2572	-0.3732	4.3668
660	1.7	0	-15.6665	2.0113	-15.4404	1.9526	-22.7664	2.0195	2.7341	-135.7843	65.3114	-10.5239	0.5659	-7.4564	-0.0716	3.2013
660	1.7	1	-15.7172	2.0183	-15.4884	1.9588	-25.8963	1.3457	6.7473	-122.5711	58.9261	-9.5008	0.5114	-10.3542	-0.7121	6.9776
660	1.7	5	-15.7239	2.0186	-15.4929	1.9569	-31.7647	-0.0265	14.6745	-50.8588	22.2507	-3.3079	0.1656	-15.2113	-1.8591	13.5868
660	1.7	10	-15.7602	2.0205	-15.5022	1.9557	-23.8127	1.8851	3.7649	-159.7634	77.9977	-12.7479	0.6950	-8.0993	-0.1458	3.8261
660	1.7	15	-15.7845	2.0191	-15.5576	1.9598	-23.2942	1.9081	3.3738	-133.6831	64.5100	-10.4305	0.5627	-7.8428	-0.1847	3.8075
660	1.7	20	-15.7825	2.0157	-15.5614	1.9552	-32.8077	-0.2625	16.0185	-36.1842	14.4067	-1.9131	0.0833	-16.1224	-2.0803	14.8498
660	1.7	25	-15.8531	2.0220	-15.6139	1.9591	-28.1449	0.8408	9.6764	-100.6791	47.1922	-7.4289	0.3908	-11.9529	-1.1075	9.2395
660	1.7	30	-15.8519	2.0163	-15.6197	1.9546	-23.9360	1.7146	4.3721	-129.5501	61.9130	-9.9117	0.5292	-8.5159	-0.3948	4.9260
660	1.7	40	-16.0243	2.0333	-15.7297	1.9594	-25.4089	1.5384	5.7834	-86.7096	40.5283	-6.3706	0.3349	-9.4150	-0.5013	5.8219
660	1.7	50	-16.0639	2.0259	-15.8239	1.9597	-25.9052	1.2578	6.9899	-80.3566	36.9175	-5.7005	0.2940	-9.5129	-0.7003	6.5787
660	1.7	60	-16.2345	2.0381	-15.9518	1.9608	-26.0324	1.2540	7.0923	-52.0232	23.3486	-3.5458	0.1804	-9.6134	-0.7470	6.8648
660	1.7	70	-16.3536	2.0390	-16.1041	1.9606	-32.1653	-0.0616	14.9674	-21.9360	8.4632	-1.1023	0.0475	-13.6578	-1.6823	12.3611
660	1.7	80	-16.7395	2.0821	-16.3304	1.9612	-38.0200	-0.9987	21.4747	3.1163	-3.3064	0.7257	-0.0464	-16.6107	-2.2156	15.9668
660	1.7	90	-16.9556	2.1004	-16.6020	1.9615	-23.2061	1.5596	4.7601	-47.3551	19.7729	-2.7880	0.1317	-5.4062	-0.3913	3.7885
660	1.8	0	-15.7101	2.0220	-15.4458	1.9541	-23.8616	1.7819	4.2225	-137.0045	64.2293	-10.0778	0.5276	-8.1299	-0.2353	4.1941

λ	D _f	f_{organics}	c0	c1	g0	g1	H0	H1	H2	s0	s1	s2	s3	k0	k1	k2
467	2.3	0	-15.2744	2.0552	-14.6573	1.8961	-29.1260	0.2391	13.1161	-11.3711	-1.2798	1.2579	-0.1228	-12.8534	-1.5781	11.7616
467	2.4	0	-15.3828	2.0789	-14.6223	1.8898	-31.8896	-0.3003	16.5330	20.8495	-18.1860	4.1848	-0.2904	-14.8163	-1.9761	14.2390
467	2.5	0	-15.4665	2.0997	-14.5745	1.8832	-34.9085	-0.9250	20.3731	52.4292	-34.6302	7.0106	-0.4510	-16.7000	-2.3711	16.6524
467	2.6	0	-15.5159	2.1151	-14.4968	1.8723	-37.2465	-1.4072	23.3510	74.5120	-46.0470	8.9528	-0.5601	-18.7793	-2.8188	19.3463
467	2.7	0	-15.6455	2.1437	-14.5196	1.8785	-40.5182	-2.0967	27.5370	96.3327	-57.1947	10.8299	-0.6646	-20.4577	-3.1879	21.5461
467	2.8	0	-15.6455	2.1437	-14.5196	1.8785	-40.5182	-2.0967	27.5370	96.3327	-57.1947	10.8299	-0.6646	-20.4577	-3.1879	21.5461

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