

Supporting Information

Light Absorption Properties and Radiative Effects of Primary Organic Aerosol Emissions

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S1. $k_{\text{OA},550}$ and w Corrections for Kirchstetter et al.'s Results

We made special corrections to widely-cited wavelength-dependent k_{OA} values of African savanna fire and firewood smoke given by Kirchstetter et al.,¹ although their estimates were prevalently used in climate models to evaluate the radiative impacts of light-absorbing organic aerosol (OA).^{2,3} First, there were important caveats in determination of the magnitude of aerosol light absorption with the optical transmission method used by Kirchstetter et al.¹ Second, they calculated k_{OA} from mass absorption cross-sections (MAC) using Eq (2) of the main text which is an equation for organic liquid extracts of OA.⁴ Since their OA MAC numbers were computed by measuring the changes in filter-based samples' attenuation before and after the acetone treatment, not by measuring the acetone extracts directly, k_{OA} values would be better not calculated from Eq (2) of the main text but from the Mie theory which is a function of the refractive index, density, and size distribution of the OA particles. Third, compared to the AERONET (Aerosol RObotic NETwork) retrieved data, aerosol absorption Ångström exponent (AAE) values have been reported to be significantly over-predicted over all world regions in model simulations using the k_{OA} datasets estimated by Kirchstetter et al.³ For the reasons above, we made corrections to Kirchstetter et al.'s results. We accepted their estimations of OA MAC at different wavelengths, but used the Mie calculations to determine the corresponding k_{OA} . For a typical size distribution of African savanna fire with a count mean diameter (CMD) of 150 nm and geometric standard deviation (GSD) of 1.45,⁵ assuming the density and the real refractive index of OA was 1.2 g cm^{-3} and 1.7,⁶⁻⁸ respectively, we estimated the $k_{\text{OA},550}$ and w were 0.022 and 3.3, respectively, both of which were lower than the original estimates.

References

- (1) Kirchstetter, T. W.; Novakov, T.; Hobbs, P. V. Evidence that the spectral dependence of light absorption by aerosols is affected by organic carbon. *J. Geophys. Res.* **2004**, *109*, D21208, DOI: 10.1029/2004jd004999.
- (2) Feng, Y.; Ramanathan, V.; Kotamarthi, V. R. Brown carbon: A significant atmospheric absorber of solar radiation? *Atmos. Chem. Phys.* **2013**, *13*, 8607-8621.
- (3) Lin, G. X.; Penner, J. E.; Flanner, M. G.; Sillman, S.; Xu, L.; Zhou, C. Radiative forcing of organic aerosol in the atmosphere and on snow: Effects of soa and brown carbon. *J. Geophys. Res.* **2014**, *119*, 7453-7476.
- (4) Sun, H. L.; Biedermann, L.; Bond, T. C. Color of brown carbon: A model for ultraviolet and visible light absorption by organic carbon aerosol. *Geophys. Res. Lett.* **2007**, *34*, L17813, DOI: 10.1029/2007gl029797.
- (5) LeCanut, P.; Andreae, M. O.; Harris, G. W.; Wienhold, F. G.; Zenker, T. Airborne studies of emissions from savanna fires in southern africa .1. Aerosol emissions measured with a laser optical particle counter. *J. Geophys. Res.* **1996**, *101*, 23615-23630.
- (6) Dinar, E.; Riziq, A. A.; Spindler, C.; Erlick, C.; Kiss, G.; Rudich, Y. The complex refractive index of atmospheric and model humic-like substances (hulis) retrieved by a cavity ring down aerosol spectrometer (crd-as). *Faraday Discuss.* **2008**, *137*, 279-295.
- (7) Hoffer, A.; Gelencser, A.; Guyon, P.; Kiss, G.; Schmid, O.; Frank, G. P.; Artaxo, P.; Andreae, M. O. Optical properties of humic-like substances (hulis) in biomass-burning aerosols. *Atmos. Chem. Phys.* **2006**, *6*, 3563-3570.
- (8) Turpin, B. J.; Lim, H. J. Species contributions to pm_{2.5} mass concentrations: Revisiting common assumptions for estimating organic mass. *Aerosol Sci. Tech.* **2001**, *35*, 602-610.

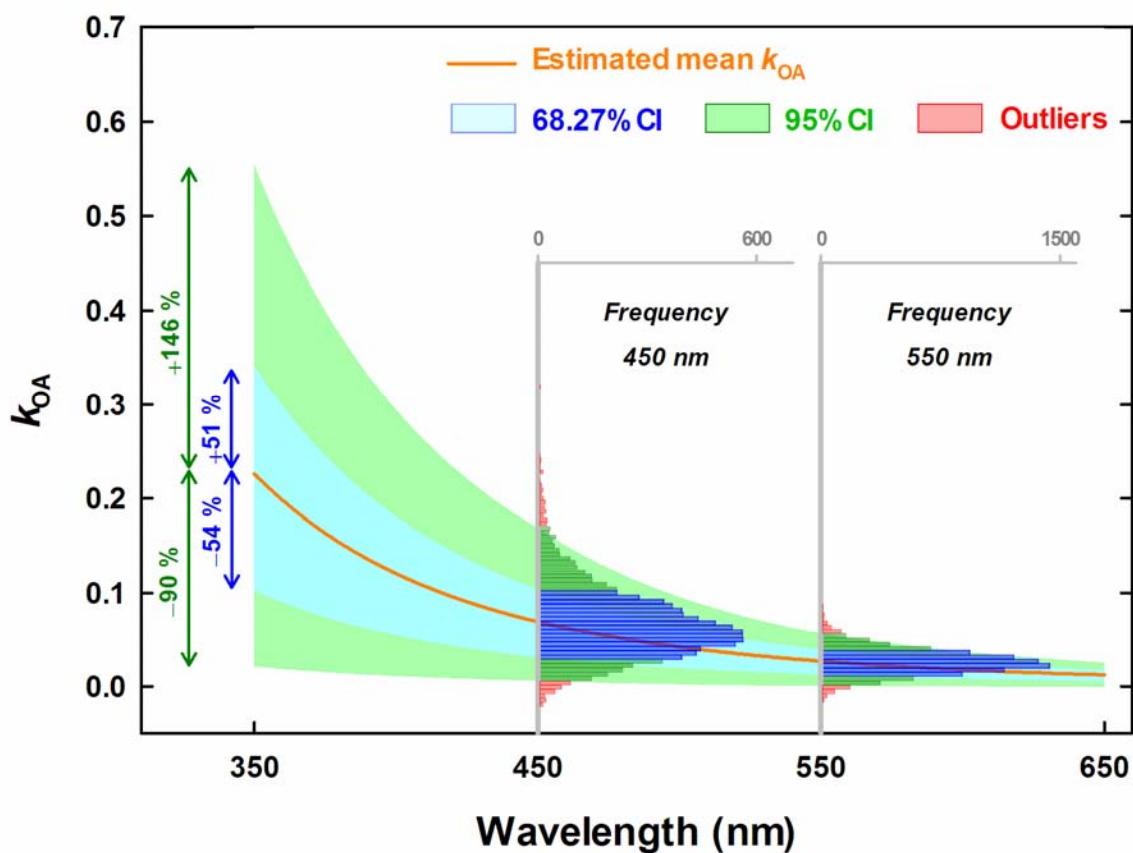


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Table S1. Global BC emissions in 2010 by sector and region (Gg yr⁻¹)

Region	Residential	Industry	Power	Transport	Open biomass burning	Total
Canada	10	8	0	13	154	186
USA	53	53	2	98	151	357
Central America	99	21	1	55	105	282
South America	93	133	1	156	1240	1623
Northern Africa	25	22	0	78	18	143
Western Africa	357	28	0	16	1026	1428
Eastern Africa	180	8	0	14	352	555
Southern Africa	136	20	2	31	1078	1266
OECD Europe	92	24	3	143	25	287
Eastern Europe	64	11	3	35	9	122
Former USSR	38	56	3	65	437	599
Middle East	33	54	4	144	49	284
South Asia	986	175	1	91	177	1431
East Asia	1095	364	20	230	191	1899
Southeast Asia	326	86	1	158	431	1003
Oceania	12	11	1	15	233	271
Japan	5	18	1	15	4	43
World	3604	1093	43	1358	5682	11780

Table S2. Global OC emissions in 2010 by sector and region (Gg yr⁻¹)

Region	Residential	Industry	Power	Transport	Open biomass burning	Total
Canada	42	3	0	8	2210	2264
USA	203	20	5	77	1805	2109
Central America	332	57	1	36	1083	1508
South America	341	595	1	68	9455	10459
Northern Africa	28	49	0	82	144	304
Western Africa	1349	146	0	49	7536	9080
Eastern Africa	635	32	0	18	2480	3165
Southern Africa	476	72	3	66	8377	8993
OECD Europe	245	14	6	122	236	623
Eastern Europe	242	17	7	13	56	334
Former USSR	113	113	4	56	4525	4811
Middle East	121	61	9	96	349	636
South Asia	3087	224	3	67	1424	4805
East Asia	2700	430	11	307	1463	4910
Southeast Asia	1210	200	2	152	4932	6496
Oceania	38	4	2	10	1726	1780
Japan	9	8	1	20	33	70
World	11169	2044	54	1246	47834	62347