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Brown carbon's emission factors and optical characteristics in household biomass burning: developing a novel algorithm for estimating the contribution of brown carbon

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Table S1-I. Elemental analysis of biomass fuels used in this study

Biomass fuels	Classify	M%	C%	H%	N%
Rape straw	CR	10.8	43.8	5.80	0.80
Peanut stalk	CR	10.2	36.0	5.07	1.31
Rice straw	CR	7.29	37.0	5.31	0.39
Wheat straw	CR	8.79	36.7	5.22	0.34
Bean straw	CR	10.8	39.5	5.87	1.19
Corncob	CR	12.8	43.3	5.93	0.52
Sorghum stalk	CR	11.9	41.1	5.66	0.55
Maize straw	CR	6.56	43.8	5.96	0.67
Cotton straw	CR	7.66	41.8	5.75	1.19
Pine	FW	4.91	46.5	6.21	0.06
Pellet fuel	PF	9.14	46.7	6.70	1.31

Note: M% - moisture on air-dry basis (%); 11 biomass fuels used in this study were divided into 3 categories: CR - crop residue; FW - fire wood; PF - pellet fuel.

Table S1--II. The combustion parameters of biomass fuels used in this study

Biomass fuels	weight burned (g)	dilution ratio
Rape straw	275	100
Peanut stalk	300	35
Rice straw	185	50
Wheat straw	250	40
Bean straw	360	35
Corncob	220	40
Sorghum stalk	80	140
Maize straw	205	40
Cotton straw	240	40
Pine	600	40
Pellet fuel	1000	3

Table S2-I. AAE values based on IS method (365 and 650 nm) for household biomass fuel burning

Biomass fuels	**AAE**	**SD**
Rape straw	1.84	0.029
Rice straw	2.98	0.43
Wheat straw	2.92	0.42
Cotton straw	2.77	0.36
Bean straw	2.59	0.31
Corncob	2.88	0.55
Peanut stalk	2.21	0.26
Sorghum stalk	1.38	0.15
Maize straw	2.67	0.54
Pine	2.82	0.57
Pellet fuels	1.95	0.65
Average	2.46	0.53

Table S2-II. f_{BrC} of biomass fuel and coal (Sun et al., 2017) values based on IS method for household biomass fuel burning and coal combustion in China

nm	f_{BrC} of Biomass	f_{BrC} of Coal	nm	f_{BrC} of Biomass	f_{BrC} of Coal
350	0.7957	0.4327	**605**	0.4813	0.2281
355	0.7911	0.4697	**610**	0.4744	0.2282
360	0.7845	0.4659	**615**	0.4633	0.2264
365	0.7903	0.4633	**620**	0.4575	0.2263
370	0.7798	0.4616	**625**	0.4478	0.2266
375	0.7684	0.4239	**630**	0.4384	0.2251
380	0.7626	0.4106	**635**	0.4308	0.2214
385	0.7596	0.4071	**640**	0.4172	0.2212
390	0.7570	0.4039	**645**	0.3944	0.2197
395	0.7532	0.4021	**650**	0.4221	0.2200
400	0.7480	0.3973	**655**	0.4282	0.2181
405	0.7416	0.3938	**660**	0.4095	0.2170
410	0.7365	0.3901	**665**	0.4204	0.2140
415	0.7320	0.3856	**670**	0.4062	0.2131
420	0.7275	0.3812	**675**	0.4056	0.2106
425	0.7223	0.3764	**680**	0.4118	0.2097
430	0.7172	0.3720	**685**	0.4003	0.2068
435	0.7110	0.3670	**690**	0.3879	0.2071
440	0.7070	0.3634	**695**	0.3798	0.2046
445	0.7011	0.3592	**700**	0.3686	0.2004
450	0.6962	0.3570	**705**	0.3697	0.1984
455	0.6911	0.3557	**710**	0.3836	0.1960
460	0.6863	0.3539	**715**	0.3632	0.1939
465	0.6825	0.3529	**720**	0.3448	0.1901
470	0.6764	0.3492	**725**	0.3224	0.1819
475	0.6721	0.3443	**730**	0.3068	0.1758
480	0.6654	0.3350	**735**	0.3115	0.1749
485	0.6563	0.3190	**740**	0.2924	0.1721
490	0.6480	0.3075	**745**	0.1703	0.1652
495	0.6419	0.3016	**750**	0.2068	0.1640
500	0.6372	0.3011	**755**	0.1837	0.1629
505	0.6344	0.3031	**760**	0.2522	0.1596
510	0.6306	0.3050	**765**	0.2360	0.1568
515	0.6264	0.3087	**770**	0.0000	0.1543
520	0.6228	0.3088	**775**	0.2246	0.1506
525	0.6185	0.3072	**780**	0.1082	0.1521
530	0.6130	0.3023	**785**	0.0000	0.1464
535	0.6062	0.2966	**790**	0.1279	0.1390
540	0.5998	0.2894	**795**	0.2240	0.1424
545	0.5941	0.2835	**800**	0.2487	0.1370

550	0.5869	0.2776	**805**	0.1295	0.1389
555	0.5793	0.2715	**810**	0.3461	0.1456
560	0.5539	0.2544	**815**	0.3587	0.1324
565	0.5415	0.2478	**820**	0.3523	0.1218
570	0.5345	0.2428	**825**	0.3644	0.1225
575	0.5270	0.2395	**830**	0.0659	0.1124
580	0.5227	0.2384	**835**	0.1829	0.1174
585	0.5169	0.2364	**840**	0.2823	0.1087
590	0.5062	0.2330	**845**	0.4437	0.2130
595	0.4951	0.2298	**850**	0.2488	0.0608
600	0.4938	0.2303			

Table S3. AAE values reported by literature

Part I: AAE for pure BrC (Water or methanol extracts)

Aerosol type	Method	AAE range	AAE average	Reference
Ambient air	water extract	6.9 (365-550 nm)	6.9	Zhu et al., 2017
Biomass burning	Water extract	6.4-6.8 (300-700 nm)	6.6	Hoffer et al., 2006
Ambient air	Water extract	6-8 (300-500 nm)	7	Liu et al., 2013
Ambient air	Water extract	5.8-11.7 (300-500 nm)	7.5	Du et al., 2014
Ambient air	Water extract	6.2-8.3 (365 nm)	7.25	Hecobian et al., 2010
Ambient air	Water extract	7.6 ± 0.5 (300-600 nm)	7.6	Zhang et al., 2013
Ambient air	Water extract	3.4 ± 0.7 (300-550 nm)	3.4	Zhang et al., 2011
Ambient air	Water extract	Summer 7.0 ± 0.8, Winnter 7.5 ± 0.9, (330-480 nm)	7.25	Cheng et al., 2011
Ambient air	Water extract	5.6-7.7 (330-400 nm)\	6.4	Kirillova et al., 2014a
Ambient air	Water extract	6.1 (300-500 nm)	6.1	Voisin et al., 2012
Ambient air	Water extract	5.1 ± 2.0 (330-400 nm)	5.1	Kirillova et al., 2014b
Ambient air	Water extract	5.1 ± 1.9 day, 5.3 ± 2.0 night, (300–700 nm)	5.2	Srinivas et al., 2016
Ambient air	Water extract	6.0 ± 1.1 (300-700 nm)	6	Srinivas and Sarin, 2014
Ambient air	Water extract	5.8 ± 1.5 (300–700 nm)	5.8	Srinivas and Sarin, 2013
Ambient air	Water extract	6.9 ± 1.9 (300-700 nm)	6.9	
Ambient air	Water extract	7.2 ± 0.7 (330-400 nm)	7.2	Bosch et al., 2014
Ambient air	Water extract	4.9 ± 0.7 afternoon, 4.6 ± 0.8 night,（330-500 nm)	4.75	Kirillova et al., 2016
	Methanol extract	4.0 ± 1.0 afternoon, 3.7 ± 1.3 night, （330-500 nm)	3.85	
Ambient air	Methanol extract	4-6 (300-500 nm)	5	Liu et al., 2013
Ambient air	Methanol extract	8.2 (365-550 nm)	8.2	Zhu et al., 2017
Ambient air	Methanol extract	4.82 ± 0.49 (300-600 nm)	4.82	Zhang et al., 2013
Ambient air	Methanol extract	5.2 (330-400 nm)	5.2	Lei et al., 2018

Kentucky Bluegrass	Methanol extract	6.8 ± 0.16 (300-500 nm)	6.8	Xie et al., 2017
Wheat		7.06 ± 0.32 (300-500 nm)	7.06	
Wheat+ Herbicide		7.18 ± 0.61 (300-500 nm)	7.18	
Forest burn		7.27 ± 0.22 (300-500 nm)	7.27	
Grass burn		6.68 (300-500 nm)	6.68	
Heavy fuel oil	extinction-minus-scattering technique	1.7 ± 0.2 (370-950 nm)	1.7	Corbin et al., 2018
		Mean	6.09	
		SD	1.45	
		SD of means	0.28	

Part II: AAE_{-open}

Source	Method	AAE range	AAE average	Reference
Wheat straw	AE31 method	3.016 ± 0.181 (370-880 nm)	3.02	Cai et al., 2014
Oak	AE31 method	1.38 (370-950 nm)	1.37	Saleh et al., 2013
Pocosin pine		1.48 (370-950 nm)		
Gallberry		1.25 (370-950 nm)		
Forest fire	A photo-acoustic aerosol absorption spectrometer (PAS)	2.3 (658-404 nm)	2.30	Lack et al., 2012
Wood combustion	a spectrometer in transmission mode for QFF	3-7.4 (300-2500 nm)	5.00	Kirchstetter and Thatcher, 2012
Wood combustion (Smog chamber)	Perkin-Elmer Lambda 35 UV-visible spectrophotometer	400-700 nm	4.74	Zhong et al., 2013
Biomass smoldering combustion	integrated photoacoustic-nephelometer (IPNs)	2.3、2.4、2.8 (532-780 nm)	2.50	Chakrabarty et al., 2010
Summer, southern California forest fires	AERONET	4.55 ± 2.01 (440-675 nm)	4.55	Bahadur et al., 2012
Biomass (birch)	AE33 method	2.5-2.7 (370-950 nm)	2.60	Martinsson et al., 2015

Incense smoke	3-wave length integrated photoacoustic-nephelometer (IPN)	8.32 (405-532 nm); 6.48 (532-781 nm)	7.40	Chakrabarty et al., 2013
Funeral wood combustion	Maya-2000 spectrophotometer	2.8-4 (450-880 nm)	3.40	Chakrabarty et al., 2014
Biomass burning	AE31 method	1.49 ± 0.08 (370-950 nm), 1.53 ± 0.1 (470-660 nm)	1.43	Yang et al., 2009
Fresh plume		1.35 ± 0.1 (370-950 nm), 1.35 ± 0.12 (470-660 nm)		
Amazonian Forest: Brazil (1993–1994); Bolivia(1998-1999)	AERONET network	1.2-2.1 (440-780 nm)	2.14	Reid, et al., 2005
South American Cerrado: Brazil (1993-1995)		1.2-2.1 (440-780 nm)		
African Savanna: Zambia (1995-2000)		1.4-2.2 (440-780 nm)		
Boreal Forest: USA, Canada (1994-1998)		1.0-2.3 (440-780 nm)		
Bonfire festival in Israel	UV-Vis spectrometer (USB 2000+, Ocean Optics)	300-650 nm	3.50	Bluvshtein, et al., 2017
Leaf litter	AE31	532-870 nm	5.92	Olson et al., 2015
Abracos Hill, Brazil Alta Floresta, Brazil Jaru Reserve, Brazil Rio Branco, Brazil	Modeling	440-870 nm	1.78	Matichuk, et al., 2008
	Mean		3.44	
	SD		1.75	
	SD of means		0.42	

Table S4. The values of MCEs and EF for OC/EC of every samples

Sample ID	Biomass fuels	MCE (%)	$EF_{OC}^{\#}$ (g/kg)	$EF_{EC}^{\#}$ (g/kg)
1	rape straw	88.12	15.46	3.43
2	peanut stalk	83.95	0.53	0.05
3	rice straw	93.40	2.76	0.35
4	wheat straw	84.83	0.82	0.10
5	bean straw	92.70	0.67	0.081
6	corncob	99.21	1.15	0.12
7	sorghum stalk	~100.00	0.28	0.08
8	maize straw	99.86	0.76	0.086
9	cotton straw	98.63	0.91	0.16
10	pine	97.34	0.37	0.063
11	pellet fuel	94.45	0.05	0.016
	Mean	93.86	2.16	0.42

[#]Reference from Sun et al., 2018.

Methods for calculation of EFs (BrC and BC), AAEs, $f_{BrC(\lambda)}$, and F_{BrC}.

(A) EFs.

Each EF (g/kg) of BrC or BC can be calculated as follows:

$$\mathrm{EF} = \frac{CF \times \rho \times A \times 10^{-6}}{(M1 - M2) \times \mathrm{f}} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots(1)$$

Where,

CF—conversion factor from measured equivalent of carbon black (CarB) to BC or from measured equivalent of humic acid sodium salt (HASS) to BrC. As described in our manuscript, CF is 1 for the former and is 0.47 for the latter

ρ—the mass of CarB equivalent or HASS equivalent per unit area of sampling filter (μg/cm^2)

A—the area of sampling filter (cm^2)

M1—the mass of biomass before combustion (kg)

M2—the mass of biomass after combustion (kg)

f—the fraction of sampled flue gas in total flue gas

(B) Absorption Ångström Exponents (AAEs).

Based on the light absorption at the wavelength pair of 365 and 650 nm measured by the IS method, AAEs are calculated as follows (Krivácsy et al., 2001; Chen an Bond, 2010; Sun et al., 2007; Lukács et al., 2007; Lack et al., 2013; Forrister at al., 2015; Yuan et al., 2016):

$$\mathrm{AAE} = \frac{-\ln(A_{650} / A_{365})}{\ln(650/365)} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ \ldots\ldots(2)$$

(C) $f_{BrC}(\lambda)$ and F_{BrC}.

The attenuated signal is measured using an Ocean Optics model Maya- 2000 spectrophotometer in the 300-1000 nm spectral range. If an absorbing substance is present in the sample, the signal attenuates, which is given by (Kirchstetter, and Thatcher, 2012).

$$\mathrm{ABS}(\lambda) = -\ln[\mathrm{I}(\lambda)/\mathrm{I}_0(\lambda)] \quad \ldots\ldots\ldots\ldots\ldots\ldots\ \ldots\ldots(3)$$

where I(λ) and I_0(λ) are the intensities measured with a sample filter and a blank filter, respectively, for each wavelength, λ.

The spectrally dependent absorbance by BrC (ABS_{BrC}(λ)) is obtained by subtracting the BC absorbance from the total absorbance(Chakrabarty et al., 2014; Kirchstetter and Thatcher, 2012; Sun et al., 2017):

$$ABS_{BrC}(\lambda) = ABS_{sum}(\lambda) - ABS_{BC}(\lambda) \quad \ldots\ldots\ldots\ldots\ldots (4)$$

The acquisition of $ABS_{BrC}(\lambda)$ and $ABS_{BC}(\lambda)$ depends on an iterative process. The detail of the iteration process is provided in a note to Figure S4.

Then, in each wavelength, the fraction of BrC absorbance in total absorbance ($f_{BrC}(\lambda)$) is calculated as:

$$f_{BrC}(\lambda) = \frac{ABS_{BrC}(\lambda)}{ABS_{sum}(\lambda)} \quad \ldots\ldots\ldots\ldots\ldots (5)$$

Finally, solar spectrum is considered. The average fraction of absorbed solar radiation by BrC relative to the combined absorption by BrC+BC over the wavelength range from 350 to 850 nm：

$$F_{BrC} = \frac{\int_{350}^{850} f_{BrC}(\lambda) k(\lambda) d\lambda}{\int_{350}^{850} k(\lambda) d\lambda} \quad \ldots\ldots\ldots\ldots\ldots (6)$$

where $k(\lambda)$ is the clear sky air mass one global horizontal solar spectrum at the earth's surface.

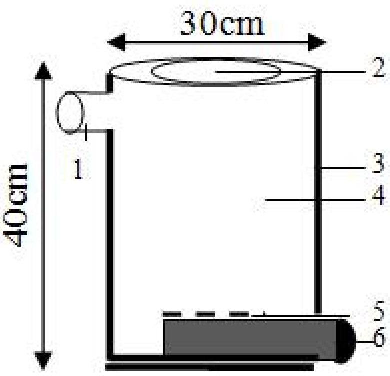

Figure S1. Cross sections of the selected Chinese household biomass burning stove

1, flue; 2, fuel inlet and removable lid; 3, iron casting; 4, biomass burning area; 5, steel grates; 6, air inlet and/or dust bin.The biomass burning stove is 40 cm high by 30 cm wide and has an upper lid and an outlet to flue pipe.

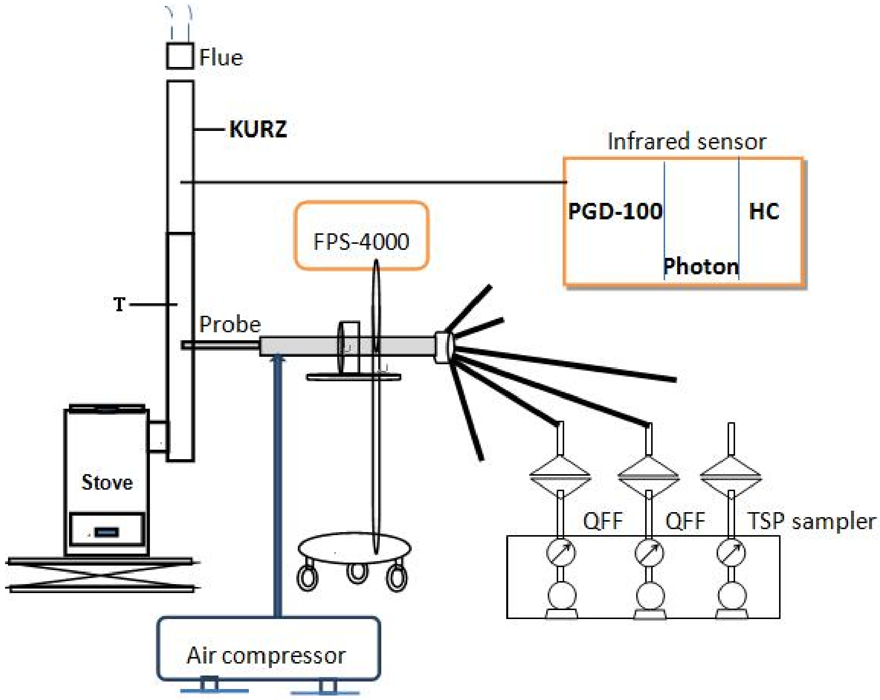

Figure S2. Diversion-dilution-sampling system (Sun et al., 2017)

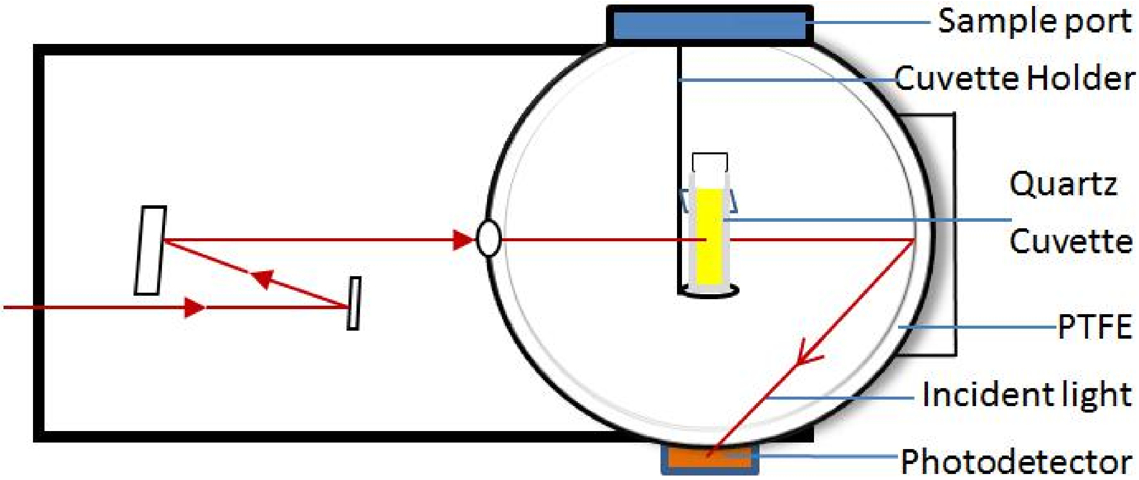

Figure S3. The sketch of integrating sphere method (Sun et al., 2107)

Note: PTFE is the Polytetrafluoroethylene reflective coating.

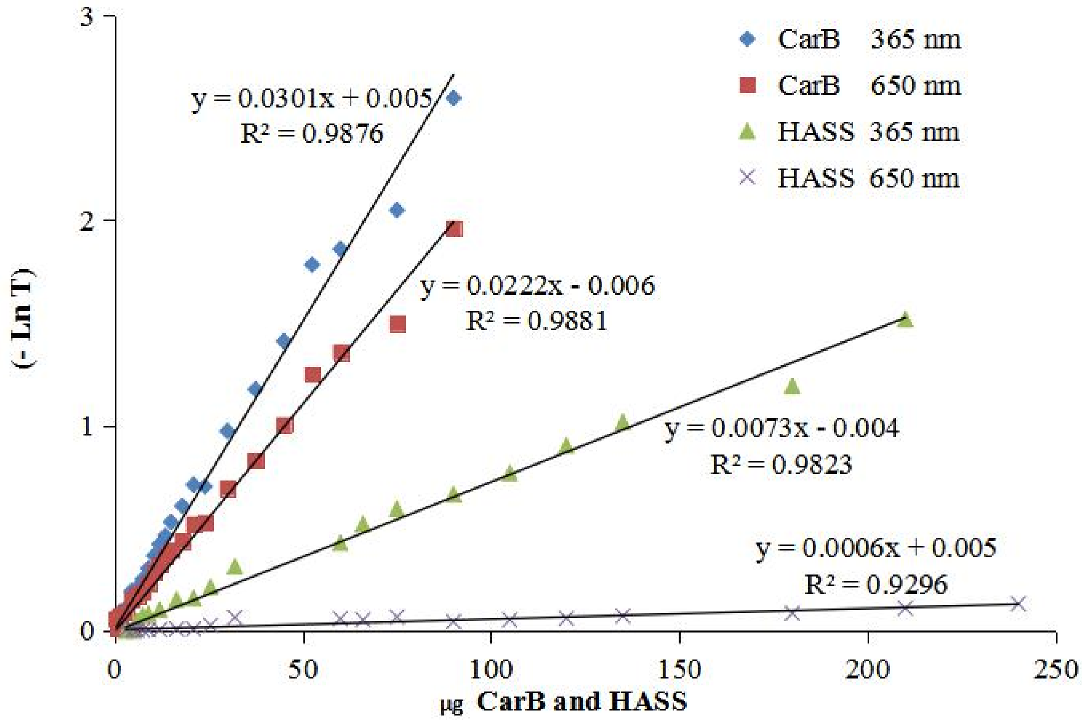

Figure S4. Calibration curves for CarB (diamonds, squares) and HASS (crosses, triangles) at 365 and 650 nm (Sun et al., 2017).

Note 1: T is the transmittance of incident light through calibration solution.

Note 2: Iteration process used in this study. The absorbances of BC (ABS_{BC}) and BrC (ABS_{BrC}) and the mass values of BC and BrC are iterated between the wavelengths 650nm and 365nm. The calibration curves in this Figure serve as：

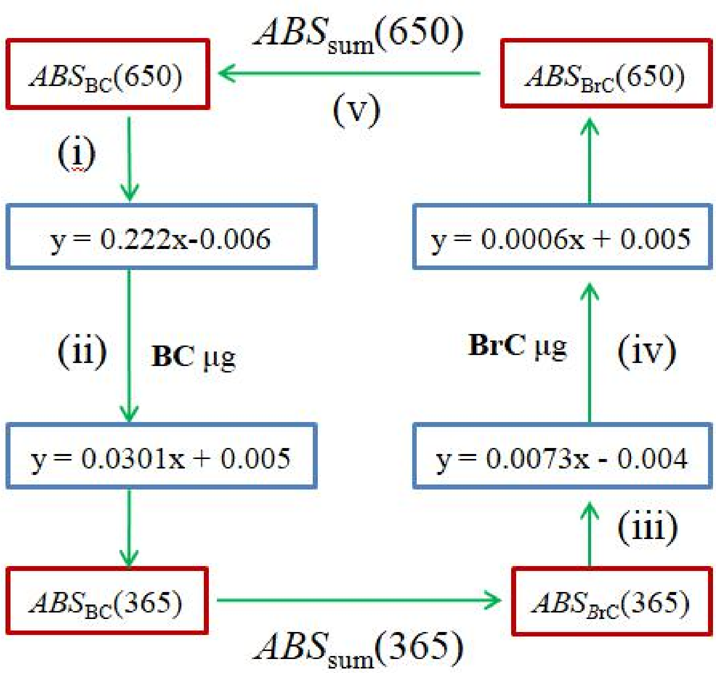

Figure S5 Calculation of BC and BrC with iterative process

(i) Assuming the measured absorbance at 650nm ($ABS_{\mathrm{sum}}(650)$) totally comes from BC ($ABS_{\mathrm{BC}}(650)$), the first mass value of BC can be calculated according to the standard curve ($y = 0.222x-0.006$).

(ii) Bring the first mass value of BC into the standard curve ($y = 0.0301x + 0.005$) to calculate the absorbance of BC at 365 nm ($ABS_{\mathrm{BC}}(365)$).

(iii) Subtracting $ABS_{\mathrm{BC}}(365)$ from the total absorbance at 365nm ($ABS_{\mathrm{sum}}(365)$) leads to the absorbance of BrC at 365nm ($ABS_{\mathrm{BrC}}(365)$), and then the first mass value of BrC can be calculated by using the standard curve ($y =0.0073x - 0.004$).

(iv) Bring the first mass value of BrC into the standard curve ($y = 0.0006x + 0.005$) to calculate the absorbance of BrC at 650 nm ($ABS_{\mathrm{BrC}}(650)$) can be obtained.

(v) Subtracting $ABS_{\mathrm{BrC}}(650)$ from the total absorbance at 650nm ($ABS_{\mathrm{sum}}(650)$) leads to the updated absorbance of BC at 650nm ($ABS_{\mathrm{BC}}(650)$), and then the second mass value of BC can be calculated by using the standard curve ($y = 0.222x-0.006$) again.

(vi) In this way, the mass values of BC and BrC are worked out again and again until the data converges enough.

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