***Supplement of***

# 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. Elemental analysis of biomass fuels used in this study**

| Biomass fuels | Classify | M% | C% | H% | N% |
|---|---|---|---|---|---|
| Rape straw | CR | 10.78 | 43.80 | 5.80 | 0.80 |
| Peanut stalk | CR | 10.23 | 35.95 | 5.07 | 1.31 |
| Rice straw | CR | 7.29 | 37.01 | 5.31 | 0.39 |
| Wheat straw | CR | 8.79 | 36.65 | 5.22 | 0.34 |
| Bean straw | CR | 10.85 | 39.45 | 5.87 | 1.19 |
| Corncob | CR | 12.82 | 43.28 | 5.93 | 0.52 |
| Sorghum stalk | CR | 11.91 | 41.06 | 5.66 | 0.55 |
| Maize straw | CR | 6.56 | 43.82 | 5.96 | 0.67 |
| Cotton straw | CR | 7.66 | 41.79 | 5.75 | 1.19 |
| Pine | FW | 4.91 | 46.51 | 6.21 | 0.06 |
| Pellet fuel | PF | 9.14 | 46.69 | 6.70 | 1.31 |

**Table S2. 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 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,<br>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,<br>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,<br>4.6±0.8 night,（330-500 nm) | 4.75 | Kirillova et al., 2016 |
| | Methanol extract | 4.0±1.0 afternoon,<br>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 |

| | | | | |
|---|---|---|---|---|
| | | Mean | 6.09 | |
| | | SD | 1.25 | |
| | | SD of means | 0.27 | |

**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<br>Alta Floresta, Brazil<br>Jaru Reserve, Brazil<br>Rio Branco, Brazil | Modeling | 440-870 nm | 1.78 | Matichuk, et al., 2008 |
| | Mean | | 3.44 | |
| | SD | | 1.75 | |
| | SD of means | | 0.42 | |

**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:

$$EF = \frac{CF \times \rho \times A \times 10^{-6}}{(M1 - M2) \times f} \text{……..……….........…………(1)}$$

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 coal before combustion (kg)

M2—the mass of coal 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):

$$AAE = \frac{-\ln(A_{650} / A_{365})}{\ln(650/365)} \text{…………………… ……(2)}$$

**(C) $f_{BrC}$(λ) 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).

$$ABS(\lambda) = -\ln[I(\lambda) / I_0(\lambda)] \text{…………………… ……(3)}$$

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):

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

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:

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

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：

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

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

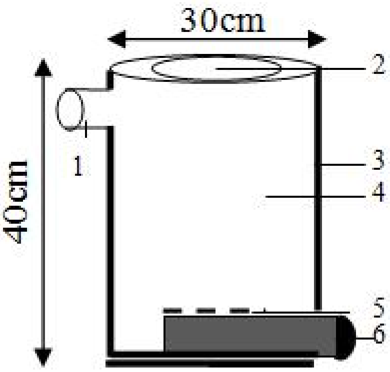


**Figure S1. Cross sections of the selected Chinese household biomass burning stove**

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.

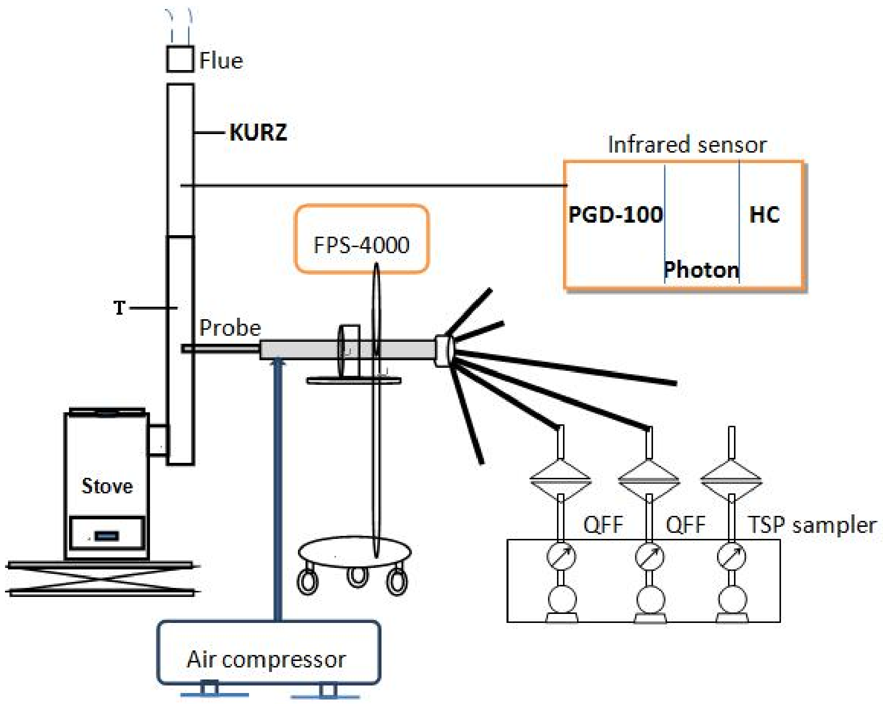


**Figure S2. Diversion-dilution-sampling system**

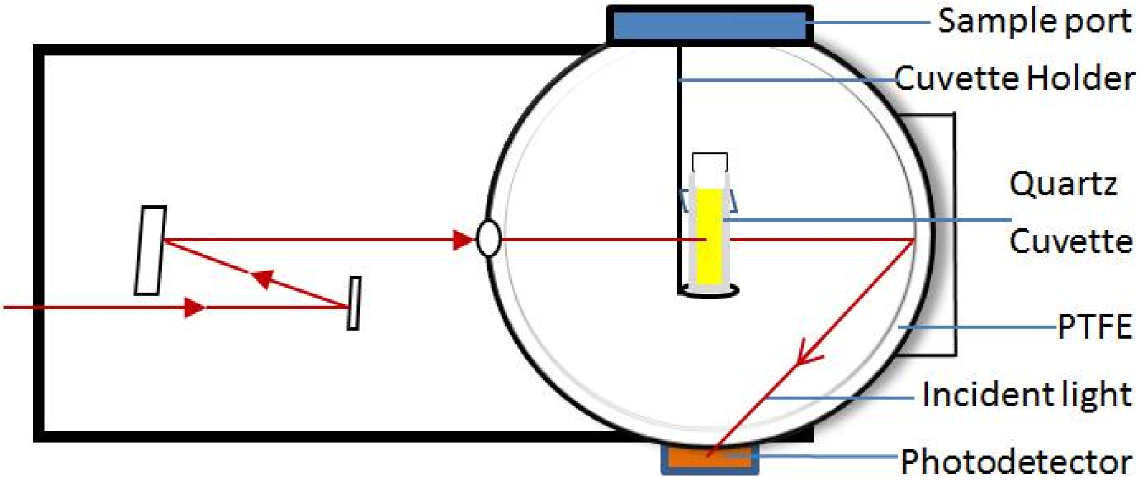


**Figure S3. The sketch of integrating sphere method**

Note: PTFE is the Polytetrafluoroethylene reflective coating.

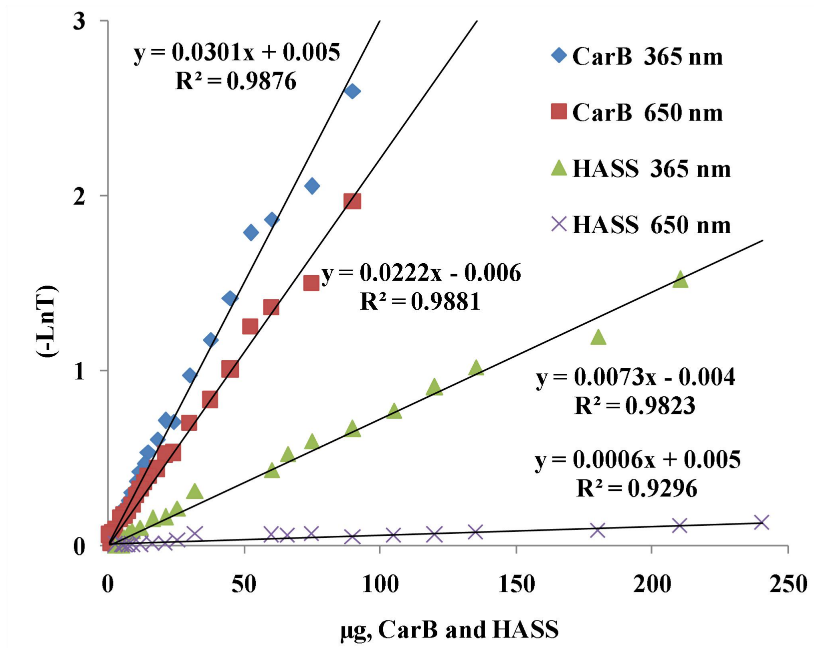


**Figure S4. Calibration curves for CarB (diamonds, squares) and HASS (crosses, triangles) at 365 and 650 nm.**

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

(i) Assuming the measured absorbance at 650nm ($ABS_{sum}$(650)) totally comes from BC ($ABS_{BC}$(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 ($ABS_{BC}$(365)).

(iii) Subtracting $ABS_{BC}$(365) from the total absorbance at 365nm ($ABS_{sum}$(365)) leads to the absorbance of BrC at 365nm ($ABS_{BrC}$(365)), and then the first mass value of BrC can be calculated by using the standard curve (y =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 ($ABS_{BrC}$(650)) can be obtained.

(v) Subtracting $ABS_{BrC}$(650) from the total absorbance at 650nm ($ABS_{sum}$(650)) leads to the updated absorbance of BC at 650nm ($ABS_{BC}$(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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