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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Abstract. Recent studies have highlighted the importance of brown carbon (BrC) in various fields, particularly relating to climate change. The incomplete combustion of biomass in open and contained burning conditions is believed to be a significant contributor to primary BrC emissions. So far, few studies have reported the emission factors of BrC from biomass burning, and few studies have specifically addressed which form of light absorbing carbon, such as black carbon (BC) or BrC, plays a leading role in the total solar light absorption by biomass burning. In this study, the optical integrating sphere (IS) approach was used, with carbon black and humic acid sodium salt as reference materials for BC and BrC, respectively, to distinguish BrC from BC on the filter samples. Eleven widely used biomass types in China were burned in a typical stove to simulate the real household combustion process. (i) Large differences existed in the emission factors of BrC (EF_{BrC}) among the tested biomass fuels, with a geometric mean EF_{BrC} of 0.71 g/kg (0.24-2.09). Both the plant type (herbaceous or ligneous) and burning style (raw or briquetted biomass) might influence the value of EF_{BrC}. The observed reduction in the emissions of light absorbing carbon (LAC) confirmed an additional benefit of biomass briquetting in climate change mitigation. (ii) The calculated annual BrC emissions from China's household biomass burning amounted to 712 Gg, higher than the contribution from China's household coal combustion (592 Gg). (iii) The average absorption Ångström exponent (AAE) was (2.46 ± 0.53), much higher than that of coal-chunks combustion smoke (AAE = 1.30 ± 0.32). (iv) For biomass smoke, the contribution of absorption by BrC to the total absorption by BC + BrC across the strongest solar spectral range of 350–850 nm (F_{BrC}) was 50.8%. This was nearly twice that for BrC in smoke from household coal combustion (26.5%). (v) Based on this study, a novel algorithm was developed for estimating the F_{BrC} for perhaps any combustion sources ($F_{BrC} = 0.5519\ln AAE + 0.0067$, $R^2 = 0.999$); the F_{BrC} value for global entire biomass burning (open + contained) ($F_{BrC\text{-entire}}$) was 64.5% (58.5–69.9%). This corroborates the dominant role of BrC in total biomass burning absorption. Therefore, an inclusion of BrC is not optional but indispensable when considering the climate energy budget, particularly for biomass burning emissions (contained and open).

1 **Introduction**

Brown carbon (BrC) refers to the fraction of organic carbon (OC) that is light-absorbing, with a pronounced wavelength dependence of absorption (Kirchstetter et al., 2004; Bosch et al., 2014; Chakrabarty et al., 2014; Mo et al., 2017; Jiang et al., 2018; Sun et al., 2018). Recent studies have highlighted the importance of BrC in not only atmospheric chemistry, air quality and human health, but also for climate change (Chakrabarty et al., 2010; Huang et al., 2018; Yan et al., 2018; Han et al., 2020). The light absorption by BrC is more emphasised towards short wavelengths (IPCC, 2014; Pokhrel et al., 2017; Li et al., 2018; Xie et al., 2018; Ferrero et al., 2020). By calculating the radiative forcing (RF) of BrC at the surface and at the top of the atmosphere, Park et al. (2010) found that more than 15% of the total RF caused by light absorbing carbon (LAC, including BrC and BC) could be attributed to BrC. Yao et al. (2017) demonstrated that a positive direct radiative effect (DRE) of absorption (+0.21 $W\cdot m^{-2}$) was caused by BrC-containing organic aerosols from the burning of crop residues in East China during the summer harvest season. This is indicative of the negative effects on not only air quality, but also on climate. Pokhrel et al. (2017) found that the absorption by BrC at shorter visible wavelengths was equal to or greater than that by BC.

The incomplete smouldering combustion of biomass in open environments or contained stoves is a major contributor to primary BrC emissions (Lukács et al., 2007; Chakrabarty et al., 2010; Hecobian et al., 2010; Chakrabarty et al., 2013). High gas and particle emissions have often been observed during these combustion processes (Kirchstetter et al., 2004; Chen and Bond, 2010; Bosch et al., 2014; Budisulistiorini et al., 2017). Ground-based observations and model simulations have revealed that in some regions with high biomass consumption intensities, such as South America, South Asia, Africa, Russia, China, and India, high column concentrations of BrC (10–35 $mg\cdot m^{-2}$) are found in the atmosphere (Arola et al., 2011; Feng et al., 2013; Huang et al., 2018). In these regions, the climatic effects of BrC are expected to be stronger than in other regions.

In China, biomass burning contributes a substantial quantity of carbonaceous particles, along with many other air pollutants. The available emission inventories show that approximately 20% of primary fine particulate matter ($PM_{2.5}$) originates from biomass burning (open and contained) (Yao, 2016). Zong et al. (2017) used the Positive Matrix Factorisation (PMF) method, linked with radiocarbon analysis, to conduct a source apportionment study of $PM_{2.5}$ at a regional background site in northern China. They found that biomass combustion comprised a significant contribution (19.3%) to atmospheric $PM_{2.5}$. Cheng et al. (2013) confirmed the significance of biomass burning in air pollution, finding that approximately 50% of OC and elemental carbon (EC) in Beijing were associated with biomass burning processes. It is also suggested that more biomass is burned in stoves than in open fields, due to China's continued efforts to prevent and control forest fires and the burning of field stalks (Tian et al., 2011; Zhi et al., 2015a; Cheng et al., 2016). Hence, more attention should be paid to the household sector than to open burning, as far as biomass-related emissions are concerned in China. In addition, unlike other regions where firewood often plays a major role as a biomass fuel, China has more access to agricultural waste (e.g. maize straw, wheat straw, and rice straw) for household heating/cooking purposes (Huang et al., 2012; Shen et al., 2013; Chen et al., 2015a). This suggests that studies of BrC originating from China's household biomass fuel combustion should consider as many biomass fuel varieties as possible, so that the actual characteristics of BrC emissions can be comprehensively investigated and represented.

The available literature dealing with BrC from biomass burning in China to date has generally focussed on ambient observation (Arola et al., 2011; Chakrabarty et al., 2014; He et al., 2017; Zhao et al., 2018) and modelling (Gustafsson et al., 2009; Feng et al., 2013) of the basic characteristics of atmospheric BrC, such as the concentrations and temporal and spatial distributions. Even though a few studies have collected emission samples at some sources, the objectives of these studies was to further understand the general properties of water soluble organic carbon (WSOC) or methanol soluble organic carbon (MSOC) (Cheng et al., 2013, 2016; Fan et al., 2016; Lin et al., 2017; Phillips et al., 2017; Huo et al., 2018; Wu et al., 2019; Yan et al., 2020). Consequently, there is a lack of knowledge regarding source emission strengths (emission factors; EFs) and regarding how BrC's role of absorption differs from that of BC (Lack et al., 2012; Healy et al., 2015; Washenfelder et al., 2015; Srinivas, et al., 2016; Zhang et al., 2016) because there is still no standard quantitative method to determine BrC. An intensive study on BrC from China's household biomass emission sources is therefore necessary to provide insight into both the EFs and light absorption properties of particulate emissions.

In the present study, eleven biomass fuels that are widely used in China were burned in an ordinary stove, to simulate domestic burning practices. Particulate emissions were collected on quartz filters to measure the EFs of BrC (EF_{BrC}) and BC (EF_{BC}) for China's household biomass burning, for investigating the spectral characteristics of absorption by BrC and estimating the contribution of BrC to total light absorption by BC + BrC across a broad solar spectral range (350–850 nm). The integrating sphere (IS) method, which had been refined in a previous study of residential coal combustion (Sun et al., 2017), was used here to simultaneously quantify BrC and BC. Furthermore, based on this intensive study of contained biomass burning (in stoves), we extrapolated the results to develop a novel algorithm for estimating the contribution of solar light absorption by BrC to the sum of BC + BrC for perhaps any combustion source. This will help to gain a clearer idea of whether BC or BrC dominates the light absorption properties of biomass burning (contained plus open) on a global scale.

2 Experimental Section

2.1 Biomass fuels and stove

Eleven biomass fuels tested in this work were classified into three groups: crop residue (CR, nine types), firewood (FW, one type), and pellet (PF, one type) fuels. The details of these fuels are given in Table S1-I. The stove that we used in this study was a natural draft stove developed specifically for biomass fuels (see Figure S1 in the Supplement). It is simple and traditional, accounting for approximately a half of biomass stoves in China (World Bank, China, 2013; Ran et al., 2014).

2.2 Combustion experiment and sample collection

The burning and sampling procedures used in this study were in general similar to those described in a previous coal combustion experiment (Sun et al., 2017). Briefly, each biomass fuel was burned in the most commonly used biomass-burning stove with cold start. The size of a fuel was the same as that used in rural households. The fuels were burned in natural combustion processes and rural operation mode. For each biomass fuel, the first batch (30–300 g) was put into the stove and then ignited with solid alcohol. Sampling and monitoring were immediately initiated. When the combustion began to fade (the first burning cycle, 3–5 min), a second batch of the fuel was added into the stove until it had been burned out (the second burning cycle, 3–5 min). Some biomass fuels (e.g. rice and wheat straws) burned so fast that a third or fourth addition was needed to sustain the combustion for an adequate sampling period. Each of the 11 biomass fuels was burned for 2-3 individual times and the emissions were collected on individual filters. The 2-3 duplicate samples helped check the reproducibility and analysis procedure. Background concentrations in ambient air were obtained separately. The modified combustion efficiency (MCE) ranged from 84.0% (peanut stalk) to almost 100% (Sorghum stalk), with an average of 93.9 ± 5.9% (see Table S4 in the Supplement), generally comparable to the results for residential coal combustion (average MCE values were 88.0 ± 4.0% and 82.5 ± 17.4% for bituminous chunk and anthracite chunk, respectively, and were 90.1 ± 1.3% and 92.8 ± 1.7% for all briquettes tested) (Zhang et al., 2020).

Although usually biomass fuels are ignited by gas lighters by ordinary stove users, there are some difficult-to-ignite biomass fuels (e.g., wood) that need to be kindled by some flammable soft materials (e.g., wheat straw, rice straw, or even leaves). Additional emissions from the flammable soft materials are inevitable. In such situations, using solid alcohol to ignite experimental biomass fuels in this study was important because no pollutants other than CO_2 and H_2O were released from alcohol combustion, though the MCE value of each sample might be a little higher than it would have been without the solid alcohol.

A diversion-dilution-sampling system (the Supplement, Figure S2) was set up to sample and/or monitor the combustion emissions. The dilution factors were set between 3 to 140 to confine the measured BrC of collected samples in the range of linearity (See Table S1- II). It should be pointed out that the sampling concentration is an important factor in the partitioning of semi-volatile species, which, if collected on the filter, may contribute to BrC absorption. The quartz fibre filters used for sampling were pre-baked in a muffle furnace at 450 °C for 6 h to remove carbonaceous substances from the filters. Each combustion experiment was repeated 2–3 times to determine the reproducibility. After sampling, the particle-loaded filters were kept in a freezer at -20 °C until needed for further analysis.

2.3 Measurement of BrC with the integrating sphere method

The differentiation of BrC from BC is a key step toward determining BrC. The mechanism and procedure of the IS method were detailed in a previous study (Sun et al., 2017). Briefly, a 150 mm IS (manufactured by Labsphere, Inc, see Figure S3) was built into a UV-Vis-NIR spectrophotometer (Perkin Elmer Lambda 950). The sphere was internally coated with Polytetrafluoroethylene (PTFE), which can reflect more than 99% of the incident light in the range of 0.2–2.5 μm (Wonaschütz et al., 2009). A specially customized transparent quartz cuvette was placed in the center of the sphere using a specially customized cuvette holder. Inside the cuvette was 3 mL of a 1:1 mixture of acetone and an 80 : 20 mixture of water and isopropanol in which a filter punch (rectangle punch, 30 × 8 mm) could be immersed. With this assembly, we scanned through the wavelength range of 350–850 nm to measure the light absorption by the collected samples. As the samples are immersed in a liquid, the absorption enhancement by possible non-absorbing coatings is negligible (Hitzenberger and Tohno, 2001; Wonaschütz et al., 2009; Sun et al., 2017).

Two reference materials were used as proxies for BC and BrC. They were carbon black (CarB) (e.g. Elftex 570, Cabot Corporation) for BC (Fisher, 1970; Andre et al., 1981; Heintzenberg, 1982; Hitzenberger et al., 1996; Wonaschütz et al., 2009) and humic acid sodium salt (HASS) (Acros Organics, no. 68131-04-4) for BrC (Wonaschütz et al., 2009). CarB had been used as proxy for BC in diesel exhaust by Medalia et al. (1983) and HASS had been used as proxy for BrC from wood combustion by Wonaschütz et al. (2009). In a previous study, CarB and HASS were used as proxies for BC and BrC, respectively, to characterise household coal burning samples, by assuming that BC and BrC in household coal emissions had the same light-absorbing properties as CarB and HASS, respectively (Sun et al., 2017). In the present study, we continued this logic, and assumed that BC and BrC in household biomass smoke have the same light-absorbing properties as CarB and HASS, respectively. In other words, the reported BC and BrC masses here are essentially CarB-C-equivalent and HASS-C-equivalent, respectively, from the perspective of light absorption and are different from those measured by other measurement techniques (e.g., thermal–optical method or aethalometer) (Chen et al., 2006; Zhi et al., 2008, 2009; Shen et al., 2013, 2014; Aurell and Gullett, 2013) or reference materials (e.g., fulvic acid, humic acid, or humic-like substances) (Duarte et al., 2007; Lukács, et al., 2007; Baduel et al., 2009, 2010). Although such an assumption is not perfect, researchers can take advantage of these two reference materials to relatively quantify and assess the features (chemical or optical) of BrC and BC derived from different combustion sources or regions. It should be noted that the IS method does not depend on an actual chemical separation, but on a virtual optical allocation of a mixed absorption signal to BrC and BC, with HASS and CarB used as references, respectively.

Calibration curves (see Figure S4) were plotted for CarB masses from 1.5–90 μg and HASS masses from 3–240 μg, according to their respective absorption signals as measured by the IS device, at both 650 nm and 365 nm (Sun et al., 2017). The BrC and BC masses of the samples were calculated through an iterative procedure based on the different spectral dependences of absorption by BrC and BC (See methods for the calculation using iteration procedure and Figure S4 in the Supplement). In most cases, 20 iterative calculations will achieve a convergent value for either BrC or BC. Note that carbon accounts only for 47% of the mass of HASS, and therefore all measured HASS equivalent values based on the calibration curves in Figure S4 were multiplied by 0.47 to obtain the mass of pure brown 'carbon' (rather than that of the BrC-containing compounds).

The CarB used in this study was Elftex 570, Cabot Corporation. It had an AAE of 0.91 and mass absorption efficiencies (MAEs) of 27.96 m^2/g and 20.64 m^2/g, respectively, for 365 nm and 650 nm. The HASS used in this study was from Acros Organics. It had an AAE of 1.86 and MAEs of 6.78 m^2/g and 0.57 m^2/g, respectively, for 365 nm and 650 nm. Both of materials are similar to actual BC and BrC in source emissions or ambient particles (Hitzenberger et al., 1996, 2001, 2006; Reisinger et al., 2008; Wonaschütz et al., 2009; Sun et al., 2017).

2.4 Calculation methods

Details of the methods for calculating EF_{BrC}, EF_{BC}, absorption Ångström exponent (AAE), the wavelength-dependent BrC contribution to total light absorption ($f_{BrC}(\lambda)$), and average BrC contribution to total solar light absorption (F_{BrC}) in the range of 350–850 nm are provided in the Supplement.

3 Results and Discussion

3.1 Emission factors of BrC from biomass fuels

The calculated EFs of the 11 biomass fuels are presented in Table 1. EF_{BrC} varied significantly among biomass fuels. Rape straw had the highest EF_{BrC} (7.26 ± 0.01 g/kg), whereas pellet fuel had the lowest (0.13 ± 0.06 g/kg). The observed differences may be related to the type of plant (see Figure 1). We notice that the EFs of BrC for herbaceous plants (HP, the former nine samples in Figure 1) were higher than those for ligneous plants (LP, the latter two samples in Figure 1). This possibly implies that herbaceous plants have a higher potential for forming BrC than ligneous plants. Although the reason underlying this difference is currently unknown, in view of the lower contents of C and H in HPs than in LPs, it seems reasonable to speculate that burning herbaceous plants in household stoves releases less heat than burning ligneous ones, which leads to a lower burning temperature for the former than for the latter, and therefore favours the generation of BrC for the former (Chen et al., 2015b; Wei et al., 2017). In this study, the temperature measured in the stovepipe (50 cm above the stove's upper surface) during HP combustion was 62.9 °C while during LP combustion, increased to 77.1 °C. Another possible explanation is the distinction in the modified combustion efficiency (MCE) values between LPs and HPs. Our measurements show that HPs tended to have lower MCEs ($93.4 \pm 6.49\% < 95.9 \pm 2.05\%$), resulting in a greater chance for the formation of BrC (Shen et al., 2013). In this perspective, greater importance ought to be attached to herbaceous biomass fuels than to ligneous ones as far as BrC emissions are concerned.

The EF_{BC} values for PFs were the lowest among all the tested biomass fuels; the briquetting effect helped to lower the occurrence of incomplete combustion and thus likely decreased the formation of primary carbonaceous particles (including BC and BrC) (Zhi et al., 2008, 2009). This agrees with the findings of Lei et al. (2018a), as the sum of LAC (BrC + BC) was observed to decrease after the maize straw was transformed to a maize briquette. In view of the virtues of biomass briquetting, regarding both air quality (less pollutant emissions) and climate change mitigation (carbon-neutral), the present study identified an additional benefit of biomass briquetting in climate change mitigation, because of the reduction of the emission of LAC (Sun and Xu, 2012; Arshanitsa et al., 2016; Chen et al., 2016).

Geometrically averaging the EF_{BrC} values over all tested biomass fuels yielded a value of 0.71 g/kg. This value was comparable to the obtained EF_{BrC} for forest fires in the south-eastern United States, measured with an aethalometer AE52 (1.0–1.4 g/kg, BC-equivalent) (Aurell and Gullett, 2013). In another study by Schmidl et al. (2008), the IS method was used to measure the BrC and BC emission characteristics of the open fires of three kinds of leaves. As BrC accounted for 18.5% (w/w) of the PM_{10} of leaf smoke (Schmidl et al., 2008) and as the PM_{10} EF for biomass fuel combustion (given by Cao et al. (2011)) is 5.77 g/kg (field burning), the EF_{BrC} can be inferred for the open fires of the three kinds of leaves, i.e. 1.07 g/kg. This value is also comparable to the averaged EF_{BrC} obtained in this study. In addition, the current EF_{BrC} average value, 0.71 g/kg, was closer to the values obtained for the combustion of anthracite-chunks (1.08 ± 0.80 g/kg) and anthracite-briquettes (1.52 ± 0.16 g/kg) than to those obtained for the combustion of bituminous-chunks (8.59 ± 2.70 g/kg) and bituminous-briquettes

(4.01 ± 2.19 g/kg) (Sun et al., 2017). This suggests the specific importance of the residential combustion of bituminous coals in BrC emissions.

Figure 1 compares EF_{BrC} and EF_{BC}. The ratios of EF_{BrC} to EF_{BC} ($R_{BrC/BC}$) varied greatly among various biomass fuels and corncobs and sorghum stalks gave the highest (10.0) and lowest (1.5) $R_{BrC/BC}$ values, respectively. Generally, the large rang of $R_{BrC/BC}$ values among different biomass fuels is attributable to the individual biomass fuels themselves, or more concretely their chemical composition and physical structure. Here both BrC and BC were products of incomplete combustion of biomass fuels (Andreae and Gelencsér, 2006. Yan et al., 2015). Different biomass fuels were composed of different organics that had different combustion performances (Reid et al., 2005; Saleh et al., 2014); meanwhile, different biomass fuels were also different in densities and moistures (Shen et al., 2014; Jacobson et al., 2015), which also have a potential influence on combustion performance. The combustion performance relates to something like the combustion speed and temperature, both of which are important to the formation of BrC and BC. Usually a low combustion temperature is more favorable for BrC formation and a relatively high combustion temperature is more favorable for BC formation (Chen and Bond, 2010; Bond et al., 2013; Shen et al., 2014). This makes the generation processes of BC and BrC often not synchronous but in opposite trend, which may account for wide variations of $R_{BrC/BC}$ for different fuels of combustion conditions.

More importantly, each of the 11 biomass fuels tested in this study had a higher EF_{BrC} than EF_{BC}; that is, the ratios of EF_{BrC} to EF_{BC} ($R_{BrC/BC}$) were all >1.The average $R_{BrC/BC}$ over all biomass fuels was 6.7 ± 2.7. Kirchstetter et al. (2004) measured the light absorption by filter-based aerosol samples from biomass burning before and after acetone treatment (which removed OC). They found that 50% of total light absorption was attributable to OC. In view of the much smaller average absorption efficiency of BrC relative to that of BC (for example, Yang et al. (2009) reported that the MAEs at 550 nm were 9.5, 0.5, and 0.03 m^2/g, respectively, for BC, BrC, and dust), the contribution of BrC to the mass of total LAC is undoubtedly far higher than that of BC, an inference which is consistent with the present study.

3.2 Spectral dependence of absorption

AAE represents the spectral dependence of the light absorption efficiency (Martinsson et al., 2015; Washenfelder et al., 2015; Yan et al., 2015). Usually, the AAE is close to 1.0 (Lack and Langridge, 2013; Laskin et al., 2015) for BC that is pronounced by a graphitic structure. This has been demonstrated by several studies of diesel exhaust or urban particulate matter (Rosen et al., 1978; Horvath, 1997). However, the existence of BrC in aerosols makes the mass absorption efficiency (MAE) increase more strongly towards shorter wavelengths, due to a larger AAE for BrC than for BC, which makes the AAEs of BrC-containing carbonaceous aerosols larger than 1 (Chakrabarty et al., 2013; Yan et al., 2015).

In this study, the measured AAE values for smoke from the combustion of the 11 biomass fuels (see Table S2-I) ranged from 1.38 (sorghum stalk) to 2.98 (rice straw), with an average of 2.46 ± 0.53. This suggests the existence of BrC in the particulate emissions. As a comparison, in a previous study that used the IS method for household coal combustion (Sun et al., 2017), average AAE values of 2.55 ± 0.44 for coal-briquettes and 1.30 ± 0.32 for coal-chunks were obtained (Sun et al., 2017). Cai et al. (2014) observed an AAE value of 3.02 ± 0.18 for the open burning of wheat straw, and of 1.43 ± 0.26 for household coal burning, using an aethalometer (AE31). Other studies have reported a wide range of AAE values, dependent on fuels, combustion conditions, aging effects after emission, the wavelengths covered and the pre-treatment experienced. (see Table S3 in the Supplement).

However, as AAE >1 for aerosol samples theoretically results from BrC instead of BC (Martinsson et al., 2015; Washenfelder et al., 2015; Zhi et al., 2015b; Yuan et al., 2016), the wide range of AAE literature values is believed to be linked to variation in the ratio of BrC to BC ($R_{BrC/BC}$). That is, the increase in $R_{BrC/BC}$ theoretically leads to an increase in AAE (Lack and Langridge, 2013). Indirect support for this interpretation can be inferred from the existing literature. For example, Saleh et al. (2014) noticed that the effective absorptivity of organic aerosol in biomass burning emissions could be parameterised as a function of the ratio of BC to OC (an umbrella term that also includes BrC). Costabile et al. (2017) found that the AAE (467–660 nm) in the atmosphere of the urban Po-Valley was positively correlated with the ratio of organic aerosol (OA) to BC ($R^2 = 0.78$), rather than to OA concentrations alone. The more persuasive scenario concerns WSOC, which is free of BC ($R_{BrC/BC} = +\infty$), for this scenario the AAE reaches its maximum (also see Table S3).

The EFs and AAEs of 11 biomass fuels used in this study and the EFs and AAEs of seven coals used in a previous study (Sun et al., 2017) are collated and arranged in a scatter plot in Figure 2. Obviously the AAE values are positively correlated with $R_{BrC/BC}$ values. Considering that the AAE for pure BC (i.e., $R_{BrC/BC}$ = 0) is conventionally accepted as 1.0, we set the intercept to 1.0 to comply with the theoretical constraint. The relation between AAE and $R_{BrC/BC}$ can be expressed in Equation (1).

$$AAE = 0.199R_{BrC/BC} + 1.00 \qquad (R^2 = 0.7527) \qquad (1)$$

Equation (1) supports the AAE-$R_{BrC/BC}$ relation in a quantitative way.

3.3 Light absorption by BrC from household biomass combustion in household stoves

With the EF_{BrC} and EF_{BC} obtained in the present study, as well as publicly available consumption data of household biomass fuels, China's BrC and BC emissions from biomass fuels burned in household stoves can be calculated, following the method described in the Supplement. In 2013, the biomass fuels consumed in China comprised 695 Tg (1 Tg = 10^{12} g) for household cooking/heating purposes (Lu et al., 2011; Tian et al., 2011; NBSC, 2014). The calculated BrC emissions were as high as 712 Gg. We acknowledge that the calculated emissions contained large uncertainties resulting from the amounts and forms of different types of biomass fuels and the representativity of BrC EFs measured in this study. Improved fuel consumption data and EFs will lead to better future emission estimates. South Asia funeral pyres release 92 Gg of BrC in 2011 (calculated with the double IS system method) (Chakrabarty et al., 2014), which is much less than that from China's household biomass combustion. This implies a clear need to control BrC emissions from household biomass burning in China.

Figure 3 compares the emissions of BrC and BC from biomass fuels in this study, and from coals as reported in a previous study (Sun et al., 2017). It is obvious that BrC emissions were always higher than BC emissions for both household biomass fuels and coals, which is attributable to the higher EF_{BrC} than EF_{BC} for both biomass fuels and coals. It is also interesting to note that, for BrC, biomass fuel dominated, whereas for BC, coal was more important. This suggests the relative importance of biomass fuels in controlling BrC.

The calculated huge emissions of BrC for China's household biomass-fuel combustion represent a strong argument for including BrC in estimating the total light absorption by emissions from burning biomass. Here, we used $f_{BrC}(\lambda)$ to represent the fraction of BrC absorption in the sum of light absorption by BrC + BC at individual wavelengths of the scanned spectral ranges (350–850 nm), measured with the IS. A detailed description of the theory and method for calculating $f_{\mathrm{BrC}}(\lambda)$ is given in the Supplement. The detailed values of f_{BrC} for biomass fuel and coal (Sun et al., 2017) from 350-850 nm were given in Table S2-II in the Supplement. The results of $f_{\mathrm{BrC}}(\lambda)$ for biomass fuels in this study are plotted in Figure 4 (blue line).

Evidently, the $f_{\mathrm{BrC}}(\lambda)$ increased towards shorter wavelengths: the $f_{\mathrm{BrC}}(\lambda)$ at 850 nm was 0.25, whereas the $f_{\mathrm{BrC}}(\lambda)$ at 350 nm increased to 0.8. In addition to the spectrally-dependent $f_{\mathrm{BrC}}(\lambda)$ for biomass fuels, Figure 4 also presents the spectrally dependent $f_{\mathrm{BrC}}(\lambda)$ values for coal (red line) as obtained in a previous study (Sun et al., 2017). The lowest value of $f_{\mathrm{BrC}}(\lambda)$ for coal occurred at 0.061 (850 nm), and the highest value occurred at 0.47 (355 nm). The average $f_{\mathrm{BrC}}(\lambda)$ for coal was 0.26, which is distinctly lower than that for biomass fuels. This difference in f_{BrC} between coal and biomass smoke can be explained by the difference in $R_{\mathrm{BrC/BC}}$ between coal and biomass smoke. It is necessary to exercise caution when attributing the absorption to BrC vs BC based on wavelength dependence (expressed as AAE). For example, Lack and Langridge (2013) found that the uncertainties in attributed BrC absorption might be ±33 % when BrC contributed 23% to 41% to total absorption (assuming an absorption measurement uncertainty of ±5 %).

Integrating $f_{\mathrm{BrC}}(\lambda)$ over the solar spectrum results in F_{BrC}, which represents the fraction of solar radiance absorbed by BrC relative to the total absorption by BC + BrC (refer to the Supplement for the method for the calculation of F_{BrC}). The standard solar spectrum is also plotted in Figure 4 (yellow line) as a contrast and reference. A value of 0.508 (0.471–0.542) was obtained for the F_{BrC} of household biomass fuels across the wavelength range of 350–850 nm, which was nearly twice that of household coal combustion (0.265) in China (Sun et al., 2017).

3.4 Extrapolation towards a novel algorithm for estimating the relative contribution of BrC

As F_{BrC} is defined as the ratio of the solar light absorption by BrC to that by (BrC + BC) across 350–850 nm, it is physically dependent on $R_{\mathrm{BrC/BC}}$. There is a scarcity of reported $R_{\mathrm{BrC/BC}}$ values, whereas conversely AAE is frequently reported in the existing literature. Therefore, the logarithmical function that can be fitted to the relationship between $R_{\mathrm{BrC/BC}}$ and AAE (Figure 2) can be used for the practical application of expressing F_{BrC} as a function of AAE.

To construct the function for F_{BrC}, with AAE as the independent variable, we managed to gather four pairs of F_{BrC} vs AAE values. Two of these pairs were based on theory. For pure BC (free of BrC), AAE and F_{BrC} were 1.0 (Lack and Langridge, 2013; Laskin et al., 2015; Yan et al., 2015; Zhang et al., 2020) and 0.0, respectively; whereas for samples of pure BrC (free of BC), we averaged over the AAE values in the literature for WSOC or MSOC (free of BC), thus obtaining an AAE value of 6.09 ± 1.45 (Hoffer et al., 2006; Hecobian et al., 2010; Voisin et al., 2012; Srinivas and Sarin, 2013, 2014; Srinivas et al., 2016; Lei et al., 2018b) (Table S3 Part I). The other two pairs of the F_{BrC} vs AAE values were obtained from our previous and current studies. The previous study (Sun et al., 2017) demonstrated that, when AAE was 1.58, F_{BrC} was 0.265. In the present study, as mentioned in Section 3.3, an AAE of 2.46 led to an F_{BrC} of 0.508. These four F_{BrC} vs AAE pairs were used to construct the relationship between F_{BrC} and AAE (Figure 5). It should be noted that we used the average value for each of the latter three points so that all the four points in Figure 5 were given equal weight (25%). A logarithmical equation was established between F_{BrC} and AAE, with a very high correlation coefficient.

$$F_{BrC} = 0.5519 \ln \mathrm{AAE} + 0.0067 \quad (R^2 = 0.999,\ 1 \leqslant \mathrm{AAE} \leqslant 6.09) \qquad (2)$$

Equation (2) provides a novel algorithm for deriving F_{BrC} from AAE, without consideration of the process details for perhaps any kinds of combustion sources. Uncertainties are unavoidable due to the uncertainties of each of the points (Lack and Langridge, 2013; Sun et al., 2017; references in Part I of Table S3). For example, Lack and Langridge (2013) estimated that the uncertainty in short wavelength absorption by BC determined by extrapolation using an AAE=1, ranged from +7% to −22%. Equation (2) helps to broaden insight into biomass burning issues from contained conditions to open conditions. The results of F_{BrC} for open fresh emissions from open biomass burning ($F_{BrC\text{-open}}$) vary in the literature, and most have values below 0.50 (or 50%) (Lack et al., 2012; Healy et al., 2015; Washenfelder et al., 2015; Srinivas, et al., 2016). We collected $\mathrm{AAE}_{\text{-open}}$ data from available journal articles and included them in Table S3 (Part II). The calculated average $\mathrm{AAE}_{\text{-open}}$ value was 3.44 ± 1.75, which was larger than the $\mathrm{AAE}_{\text{-contained}}$ value obtained in this study (2.46 ± 0.53). Substitution of the $\mathrm{AAE}_{\text{-open}}$ value (3.44 ± 1.75) into Equation (2) leads to a value of 0.685 for $F_{BrC\text{-open}}$, which is higher than the F_{BrC} for contained combustion ($F_{BrC\text{ -contained}}$) (0.508), indicating that BrC's light absorption was more dominant in open biomass burning emissions than in contained biomass burning emissions.

Assuming that the $\mathrm{AAE}_{\text{-contained}}$ and $\mathrm{AAE}_{\text{-open}}$ identified above apply to global biomass burning, we can now assess BrC's role in the biomass burning globally (contained + open) ($F_{\mathrm{BrC\text{-}entire}}$), in combination with the respective shares of open and contained burning. Previous studies show that the annual open and contained biomass burning amounts are 5953 Tg (Wiedinmyer et al., 2011) and 2457 Tg (Fernandes et al., 2007), respectively. This implies that open biomass burning represents 71% of total biomass burning and contained biomass burning represents 29%. Subsequently, the $F_{\mathrm{BrC\text{-}entire}}$ can be calculated according to the following equation:

$$F_{\mathrm{BrC\text{-}entire}} = 0.29 \times (0.5519\ln\mathrm{AAE}_{\mathrm{-contained}} + 0.0067) + 0.71 \times (0.5519\ln\mathrm{AAE}_{\mathrm{-open}} + 0.0067) \quad (3)$$

With Equation (3), the distribution of $F_{\mathrm{BrC\text{-}entire}}$ was simulated through the Monte Carlo approach, as shown in Figure 6. The $F_{\mathrm{BrC\text{-}entire}}$ was 0.644 on average, and with an 80% probability range it lay between 0.585–0.699. Particularly, the probability of $F_{\mathrm{BrC\text{-}entire}}$ being larger than 0.500 was higher than 99%, corroborating the leading role of BrC in the absorption by solar light for total biomass burning emissions. Kirchstetter and Thatcher (2012), calculate that OC from wood smoke would account for 14% of solar radiation absorbed by wood smoke in the atmosphere (integrated over the solar spectrum from 300 to 2500 nm). 14% is much smaller than our data $F_{\mathrm{BrC\text{-}entire}}$= 64.4% because Kirchstetter and Thatcher (2012) only focus on rural California wintertime wood combustion but we calculated the global contribution to absorption by BrC originating from biomass combustion.

4 Conclusions

The optical IS approach was used to distinguish BrC from BC in filter samples of the emissions of 11 types of biomass after burning in a typical stove. The measured average EF of household biomass fuels for BrC was 0.71 g/kg, and the calculated annual BrC emissions from China's household biomass burning amounted to 712 Gg. This is higher than the emissions from China's household coal combustion (592 Gg). Moreover, it was observed that BrC contributed to approximately half of all light absorption by BC + BrC across the strongest solar spectral range (350–850 nm; F_{BrC} = 50.8%). Furthermore, a novel relationship was constructed ($F_{\mathrm{BrC}} = 0.5519\ln(\mathrm{AAE}) + 0.0067$, $R^2 = 0.999$), which can simplify the calculation of F_{BrC} by using AAE. With this mathematical relationship, we calculated the F_{BrC} values for open biomass burning ($F_{\mathrm{BrC\text{-}open}}$ = 70.1%) and entire biomass burning ($F_{\mathrm{BrC\text{-}entire}}$ = 64.4%), thereby establishing the dominant role of BrC in biomass burning absorption. From this perspective, we recommend that it is necessary to include BrC in the climate discussion, particularly concerning biomass burning (contained and open). The algorithm developed here omits the long procedures of chemical treatment, optical measurement and tedious calculations, and provides a scheme for estimating the contribution of BrC relative to BC in perhaps any combustion process with LAC emissions.

Data availability

The research data can be accessed, on request, from the corresponding author (zhigr@craes.org.cn).

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Competing interests. The authors declare that they have no conflicts of interest.

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Table 1. Measured EF_{BrC} and EF_{BC} (g/kg) values for household biomass burning

Biomass fuels	EF_{BrC}	EF_{BC}	$R_{BrC/BC}$
Rape straw	7.26 ± 0.01	2.54 ± 0.01	2.86 ± 0.02
Rice straw	2.50 ± 3.06	0.31 ± 0.25	8.06 ± 6.67
Wheat straw	1.25 ± 0.07	0.13 ± 0.04	9.62 ± 5.17
Cotton straw	0.89 ± 0.51	0.10 ± 0.02	8.91 ± 2.99
Bean straw	0.57 ± 0.12	0.09 ± 0.04	6.41 ± 2.21
Corncob	0.56 ± 0.55	0.056 ± 0.02	10.01 ± 8.77
Peanut stalk	0.54 ± 0.15	0.13 ± 0.054	4.15 ± 1.42
Sorghum stalk	0.45 ± 0.32	0.30 ± 0.054	1.51 ± 0.39
Maize straw	0.45 ± 0.76	0.053 ± 0.014	8.49 ± 4.97
Pine	0.27 ± 0.29	0.034 ± 0.017	7.94 ± 3.41
Pellet fuels	0.13 ± 0.06	0.023 ± 0.037	5.65 ± 2.58
Geometric mean	0.71 (0.24, 2.09)	0.12 (0.033, 0.436)	5.90 (3.26, 10.68)

Note: The last row for geometric mean is expressed as geometric mean (lower limit, upper limit). The lower/upper limits are calculated via geometric mean divided/multiplied by the geometric standard deviation (GSD). The GSDs for EF_{BrC}, EF_{BC}, and $R_{BrC/BC}$ are 2.95, 3.63, and 1.81, respectively.

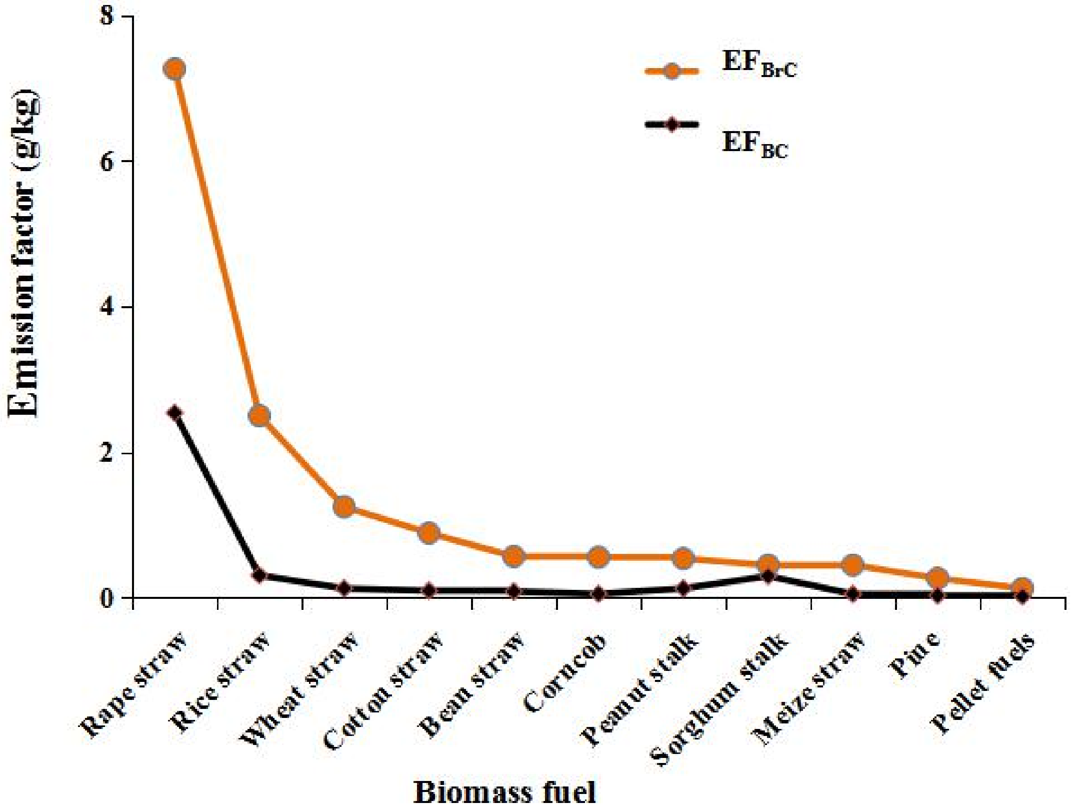

Figure 1. EFs of tested biomass fuels

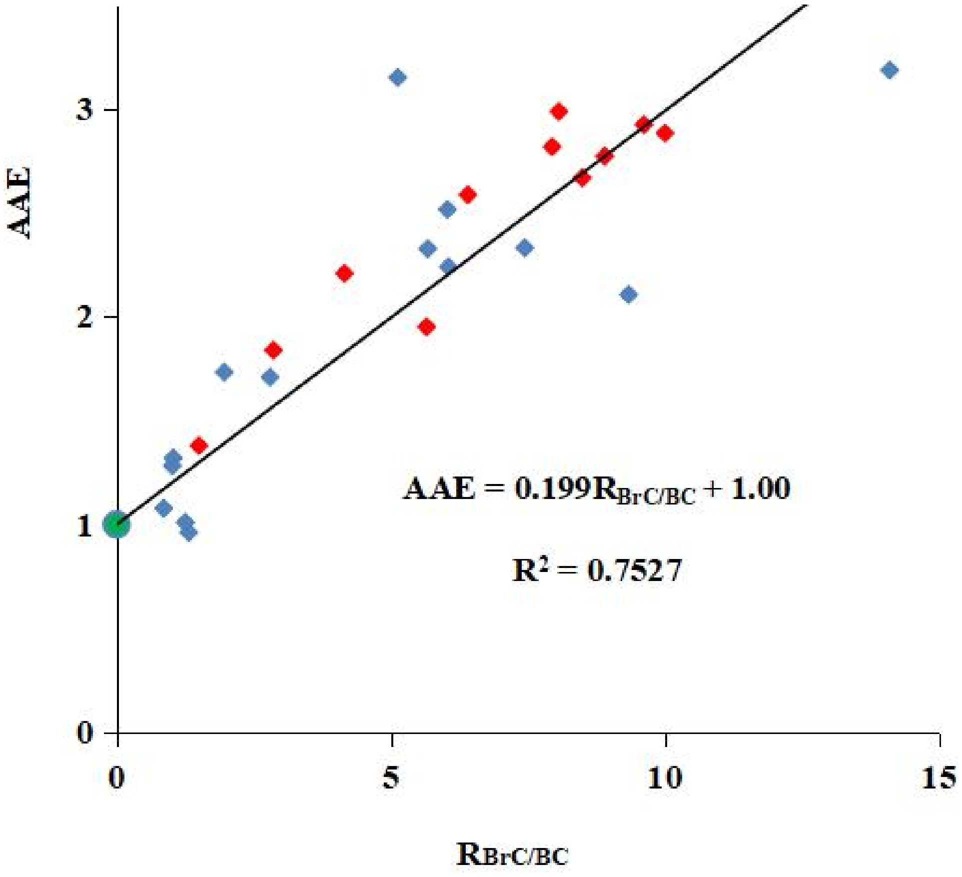

Figure 2. Relationship between AAE and EF_{BrC}/EF_{BC} ratio ($R_{BrC/BC}$) for both biomass fuel (red) and coal (blue). The intercept is designated as 1.0 to echo the conventionally accepted notion that the AAE for pure BC (i.e., $R_{BrC/BC} = 0$) is 1.0.

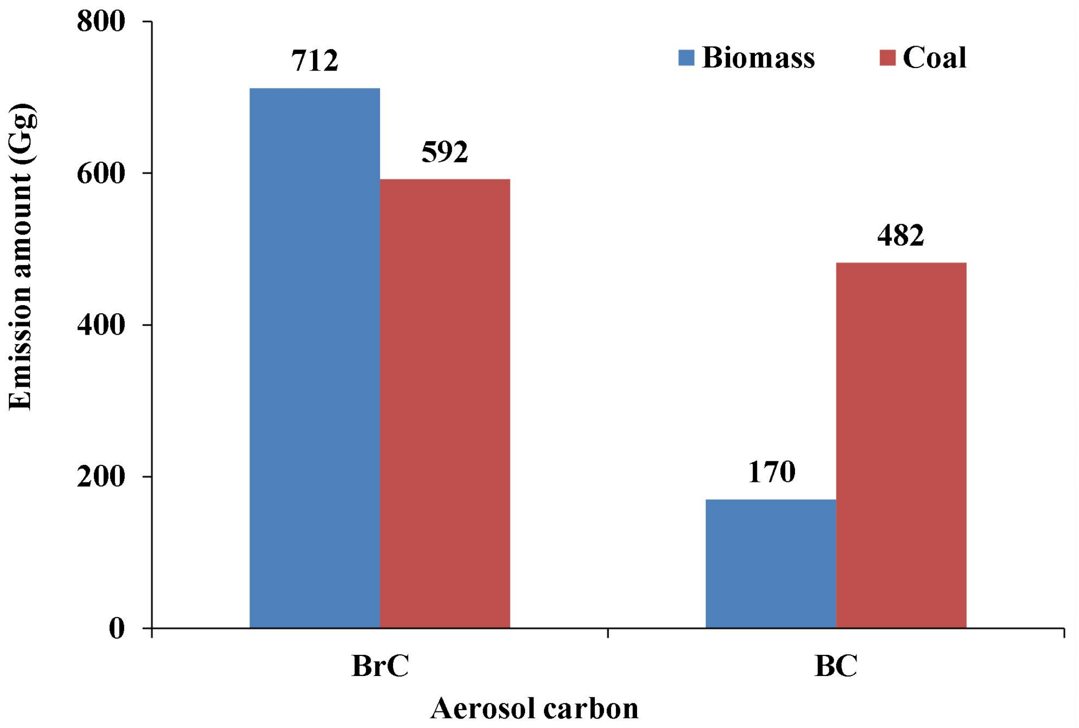

Figure 3. Comparison of BrC and BC emissions between biomass burning and coal combustion in China's household sector of 2013

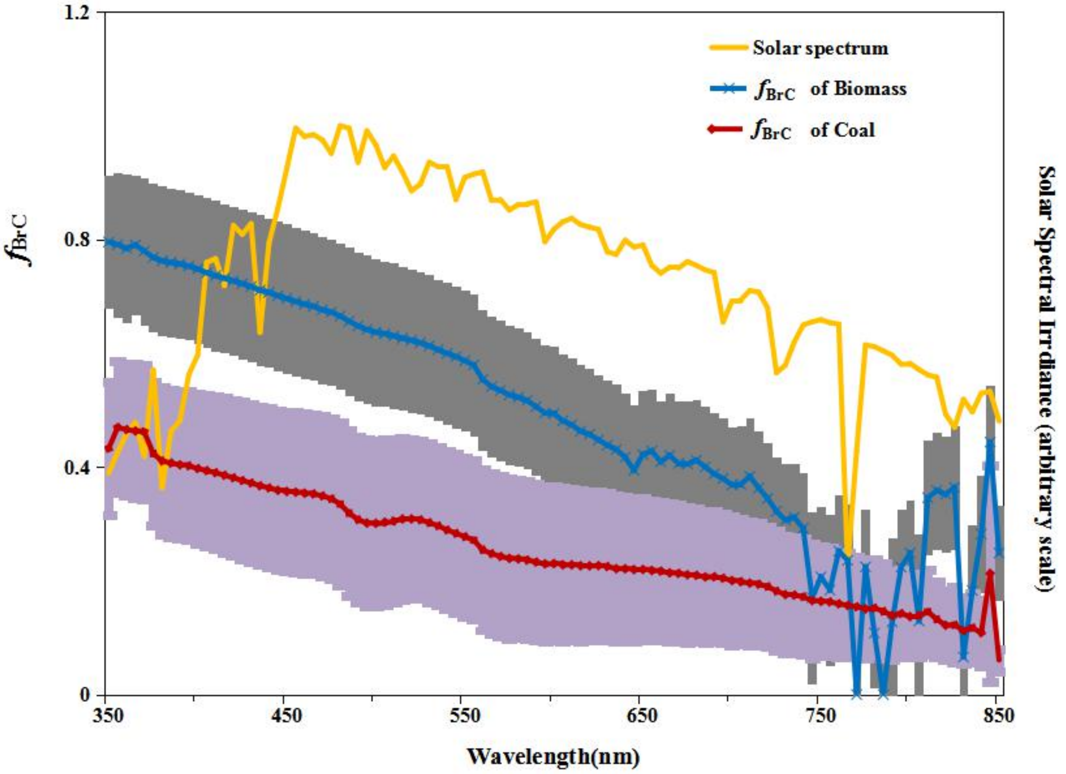

Figure 4. Ratios of light absorption by BrC to total absorption by total mass with respect to China's household biomass and coal burning

Note: The ratio is expressed as f_{BrC} and was calculated in accordance with the method described in the Supplement. The yellow line is the clear sky global horizontal solar spectrum at the earth's surface for one optical air mass in relative units (Levinson et al., 2010; Chakrabarty et al., 2014).

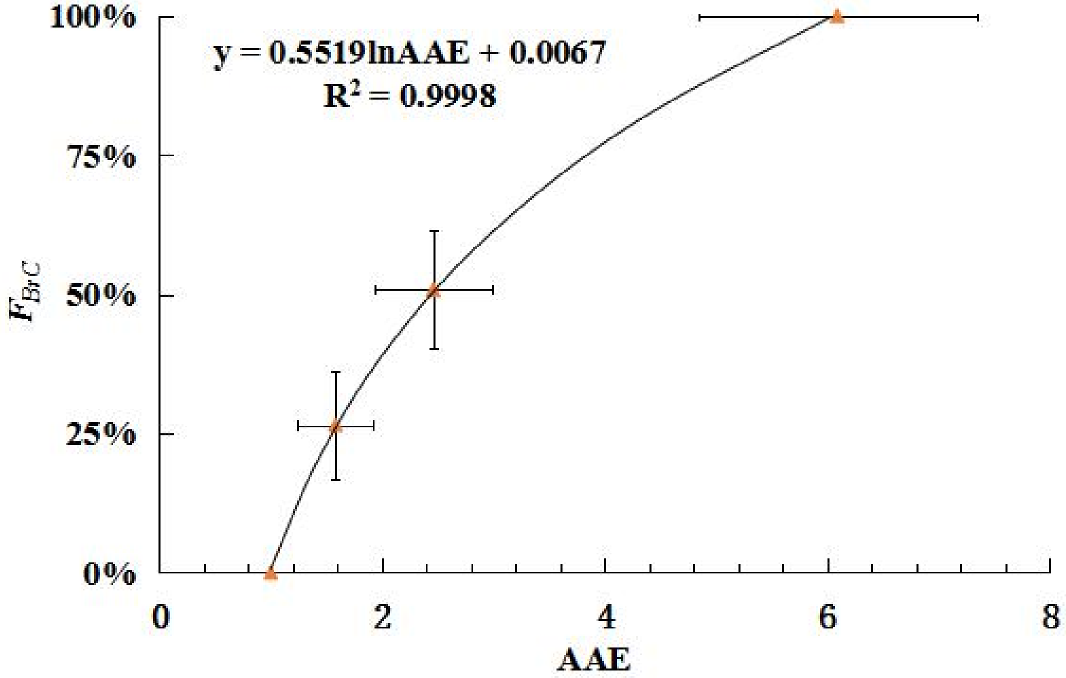

Figure 5. Relationship between F_{BrC} and AAE

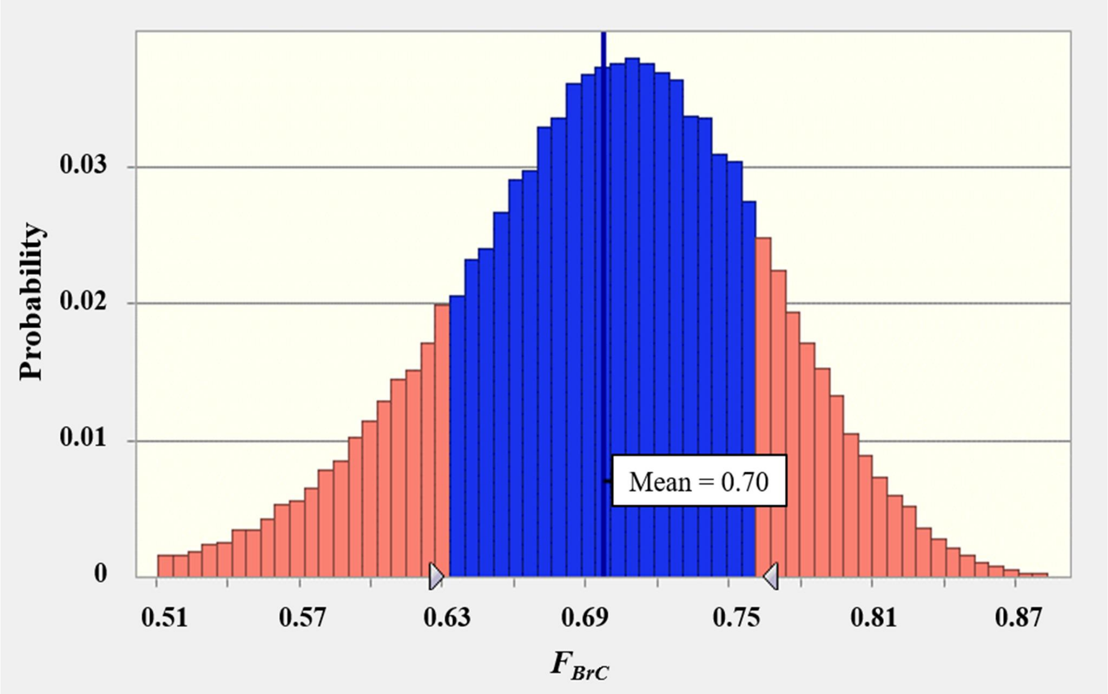

Figure 6. The probability distribution of calculated $F_{\text{BrC-entire}}$. Assuming the AAE$_{\text{-contained}}$ value of 2.46 ± 0.16 (mean ± SD of the means) and AAE$_{\text{-open}}$ value of 3.44 ± 0.42 (mean ± SD of the means) apply to whole world biomass burning, the combined value for entire biomass burning ($F_{\text{BrC-entire}}$) can be calculated as: $F_{\text{BrC-entire}} = 0.71\times (0.5519\ln\text{AAE}_{\text{-open}} + 0.0067) + 0.29 \times (0.5519\ln\text{AAE}_{\text{-contained}} + 0.0067)$