Interactive comment on "Brown carbon's emission factors and optical characteristics in household biomass burning: Developing a novel algorithm for estimating the contribution of brown carbon" by Jianzhong Sun et al.

J. Sun et al.

zhxi.2006@163.com

Reply to Referee 1

General comment:

This is a manuscript that reports on the emission factors and optical characteristics of BB-derived BrC and development of a novel algorithm for estimating the contribution of BrC. The results indicated the mean emission factors of BB-BrC are 0.71 g/kg, which were affected by the plant type and burning styles. The average AAE value was 2.46 ± 0.53, which are much higher than that of coal-chunks combustion smoke. The contribution of absorption by BB-BrC to the total absorption by BC + BrC were also calculated, is 50.8%. Finally, a novel algorithm was developed for estimating the F_{BrC} for any combustion sources. This is an interesting research about the emission factors and light-absorption characteristics of BrC emitted from biomass burning. I think the manuscript can be accepted after the following comments are addressed.

Response:

Thanks a lot for the positive comment. The recommendation for publication in ACP is encouraging. We would further improve our manuscript according to the comments and suggestions below.

Comments 1:

Line 11: what's the meaning of "0.24, 2.18"?

Response:

Thanks. When we initially submitted our manuscript to editorial office, the editor

suggested us not to use arithmetic mean but other forms, such as the median value and/or a range because the values of EF_{BrC} (or $EF_{BC)}$ for different samples differed by more than an order of magnitude. Following the suggestion, we turned to use geomeans instead of arithmetic means. Line 11 in original version described the geomean (i.e., 0.71g/kg) calculated for EF_{BrC} and the (lower, upper) limits (i.e., 0.24, 2.18) calculated via a geomean (i.e., 0.71g/kg) divided/multiplied by the geometric standard deviation (i.e., 2.95). We added a note to Table 1 to show how the geomean and range (lower and upper limits) were obtained. In addition, we changed the upper limit of 2.18 in original version to 2.09 in revised version to correct for wrong calculation in original version. In line 11 (revised version), we changed 0.71 g/kg (0.24, 2.18) to 0.71 g/kg (0.24-2.09).

Comments 2:

Lines 70-76: several important references for the BrC from biomass burning in China were missed, such as Fan et al. (2016) ACP, 16, 13321-13340; Huo et al. (2018) Atmos. Environ., 191, 490-499, etc.

Response:

Thanks for this reminder. We added these citations proposed by the reviewer (revised version, lines 72-73).

Comments 3:

Experimental section: accuracy, precision, and repeatability are not well quantified or discussed in this paper. The 11 biomass fuels are each burned and sampled once. The filter sample for each fire is collected in background air, so ambient aerosol may present in the sample. These may be reasonable experimental procedures, but the following information is missing: i) Blank filter sample for ambient air only to determine the background concentrations; ii) Repetitions of identical sample burns to determine the repeatability of the fires and the analysis procedure.

Response:

So sorry that we didn't completely and clearly describe the accuracy, precision, and repeatability. In the revised version, we added the missing information and bettered the unclear description, particularly on the two key concerns: i) Blank filter sample for ambient air only to determine the background concentrations; ii) Repetitions of identical sample burns to determine the repeatability of the fires and the analysis procedure.

The revised version is as follows: "Each of the 11 biomass fuels was burned for 2-3 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." (lines 107-109, revised version).

Comments 4:

Please reduce the number of significant digits (2-3 is preferred) in Table 1, S1, and possibly in the main text. For example, "7.259" (four significant digits) can be present as "7.26" (maximum three significant digits). Please double check such errors throughout the entire manuscript.

Response:

Thanks for this reminder. We checked throughout the whole manuscript and reduced the number of significant digits after the decimal point to 2 in Table 1 and Table S1 except for EF_{BrC} and EF_{BC} of the rape straw. We actually collected 2 samples for rape straw. The experimental results of the duplicates were extremely close, which made the standard deviations be 0.002 for EF_{BrC} and 0.001 for EF_{BC}. When we initially submitted our manuscript to editorial office, the editor suggested us to increase the number of significant digit after the decimal point from 2 (0.00) to 3 (0.002 and 0.001 specifically for the EF_{BrC} and EF_{BC} of rape straw sample) to avoid the uncertainty value of 0.00. For this reason, we maintained the two data of rape straw with 3 significant digits after the decimal point, as 7.259±0.002 and 2.537±0.001 (See revised version, Table 1 and S1), while the data of other biomass fuels were designated with 2 significant digits after the decimal point.

Comments 5:

Lines 205-208: The ratios of EF_{BrC} to EF_{BC} for different samples were varied with very large range (the highest one is 10.0 and the lowest one is 1.5). Why? Please add some explanation. This is very important for the estimation of the contribution of BB BrC.

Response:

Thanks for this suggestion. Sure the ratios of EF_{BrC} to EF_{BC} for different samples varied with very large ranges; for example the highest one is 10.0 and the lowest one is 1.5. Although the reasons for the large range in $R_{BrC/BC}$ ratio among different biomass cases involves very complicated factors, they are essentially attributed to the differences in chemical composition and physical structure. It is acknowledged that both BrC and BC are products of incomplete combustion of biomass fuels (Andreae and Gelencsér, 2006; Yan et al., 2015). Different biomass fuels are composed by different organics that have different combustion performances (Reid et al., 2005; Saleh et al., 2014); meanwhile, different biomass fuels are also different in densities and moistures (Shen et al., 2014; Jacobson et al., 2015), which are also potential to exert influences on the 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 suggests that the generation processes of BC and BrC are often not synchronous but in opposite trend, which made the values of $R_{BrC/BC}$ vary terribly.

We understand the importance of the $R_{BrC/BC}$ for the estimation of the contribution of biomass BrC and accordingly added some explanations in lines 221-235 in our revised version.

Comments 6:

Figure 2: the data of BrC from BB and coal combustion should be labelled with

different markers.

Response:

Thank you. In our revised version, the data of BrC from BB and from coal combustion in Fig. 2 have been labelled with red and blue markers, respectively.

Comments 7:

Lines 252-258: China's BrC and BC emissions from biomass fuels burned in household stoves were calculated. This section is associated with high uncertainties due to the reliable consumption amounts of different types of biomass fuels and forms, representative BrC emission factors from this study. I'd like to suggest to add discussions on uncertainties and limitations.

Response:

Thanks for this suggestion. Lines 252-258 (previous version) described China's BrC and BC emissions from biomass fuels burned in household stoves. The calculated emissions indeed contained uncertainties resulting from the consumption amounts and forms of different types of biomass fuels as well as the representativity of BrC emission factors measured in this study. We added discussions on the uncertainties and limitations (lines 286-289, revised version).

Comments 8:

Section 3.4, Lines 295-306: To construct the function for F_{BrC}, with AAE as the independent variable, four pairs of F_{BrC} vs AAE values were investigated: one pure BC and three pure BrC. For the three pure BrC, I have two questions: 1) why the average values of F_{BrC} vs AAE rather than the data of each sample were used to construct the function between F_{BrC} and AAE? 2) As shown in Table S3, the AAE values of WSOC or MSOC in the literature were determined in solution. However the AAE values of BrC were determined with the integrating sphere method in this paper and the previous study (Sun et al., 2017). How about the differences of AAE values measured with these two methods. You should add some discussions to interpret that.

Response:

Thanks for this question and suggestion.

(A) We described how we constructed the relation in lines 330-341 (revised version). That is, 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 theoretically for pure BC and pure BrC, respectively. For pure BC (free of BrC), the 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, and for samples of pure BrC (free of BC), we averaged over the AAE values in the literature for WSOC or MSOC, 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., 2018) (Part I in Table S3). The other two pairs of the F_{BrC} vs AAE values were obtained from our previous and current measurements. 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).

(B) The question why the average values of F_{BrC} vs AAE rather than the data of each sample were used to construct the function between F_{BrC} and AAE is worth explaining. The same question had actually been raised by the editor and we had explained the reason in advance. On the one hand, we know, each of the latter three points (i.e., 1.58, 0.265; 2.46, 0.508; 6.09, 1.00) in Figure 5 is the average of a number of data, and therefore each of them can be potentially replaced with a cluster of individual dots if we like; yet on the other hand, the first point (0.00,1.00) is not originated from averaging over a cluster of individuals but from theoretical consideration, and thus there are no cluster of individual dots usable to replace this single point. Under the circumstances, replacing each of the latter three points with an individual cluster of dots while leaving the first point with single dot will substantially lower the weight of the first point from 25% to almost being negligible. Given the theoretical significance of the first point, this is not only unfair but also unacceptable. For this consideration, we preferred to the average value for each of the latter three points so that all the four

points in Figure 5 were put weights equally (25%). Additionally, compared with a cluster of individuals, an average is usually closer to or more representative of the true value and hence is more persuasive. We added an explanation in our revised version (lines 340-341).

(C) As regards the need to add some discussion on the differences between AAE values measured with IS method and the AAE values measured through WSOC or MSOC, the former is for the entirety of a sample including BrC+BC whereas the latter is for BrC alone (free of BC).

Comments 9:

Table S1: The abbreviation of "M%, CR, FW, PF" should be illustrated in full name.

Response:

Thanks for reminder. We gave the full names of the abbreviations as a note to Table S1, as follows:

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.

Comments 10:

Is Fig S4 cited from the literature of authors (Sun, J., Zhi, G., et al., Emission factors and light absorption properties of brown carbon from household coal combustion in China, Atmos. Chem. Phys., 17, 4769-4780)? If so, please add references in the caption.

Response:

Thanks for reminder. We added 'Sun et al., 2017' in Fig. S4 accordingly.

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Reply to Referee 2

This paper presents emission factors for brown carbon and black carbon from 11 different biomass fuels in a commonly used cookstove. Most of the paper is focused on the development of an algorithm to convert AAE into the mass ratio of BrC to BC and solar absorption fraction attributed to BrC. The paper makes the important point that BrC absorption needs to be included in assessments of the climate impacts from biomass burning. Given the data presented here (one stove and several biomass fuels), the universality of the algorithm for multiple emission sources is overstated. Also, there is limited comparison of the algorithm with other methods, so it is not clear if it is an improvement over other approaches of estimating the impact of BrC on climate. I recommend major revisions to address the following comments:

Response:

Many thanks for the instructive and constructive comments. We have revised our manuscript according to the questions raised and suggestions made below.

General Comments:

1. The quantification of BrC as a mass emission is a relatively uncommon approach due to the complexity of BrC (i.e. many different chromophores with differing mass absorption efficiencies that are source dependent). A fuller description of the proxies used in this current study and how they compare to other black and brown carbon sources should be outlined (e.g. absorption efficiencies, AAE, primary particle size, etc.) and any shortcomings of using these proxies should be noted. How does this method of estimating BrC mass emissions compare with other approaches used in the literature that were cited for comparison of BrC emission factors (e.g. Aurell and Gullett 2013 and Schmidl et al. 2008)?

Response:

Thanks for this comment. It's true that the quantification of BrC as a mass emission or a mass ambient concentration is a big challenge because BrC is not a pure substance but a mixture of light-absorbing organic compounds that may contain many different chromophores with differing

mass absorption efficiencies dependent on origins.

Two reference materials were designated as proxies respectively for BC and BrC: carbon black (CarB) for BC and humic acid sodium salt (HASS) for BrC. Early studies by other researchers used to designate these two substances as proxies because of their similarities to actually observed BC and BrC in especially optical properties. For example, CarB was used as proxy for BC in diesel exhaust by Medalia et al. (1983) and HASS was used as proxy for BrC in wood combustion by Wonaschütz et al. (2009). In a previous study conducted by ourselves, CarB and HASS were used as proxies for BC and BrC, respectively, from residential coal combustion (Sun et al., 2017). We carried over this philosophy to the current study and thus the reported BC and BrC masses here were essentially CarB-C-equivalent and HASS-C-equivalent in light absorption. This is different from some other approaches in literature reporting BC and/or BrC using 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). For example, some researchers used an aethalometer to quantify BrC, and the reported BrC mass was actually aethalometer-BC-equivalent in light absorption (Aurell and Gullett, 2013).

In one of our previous articles (Sun et al., 2017) (Table S1 and Figure S3 therein), the IS measured BC was significantly correlated with the thermal/optical EC (EC=0.884BC-0.114, R^2=0.927). In addition, with CarB and HASS, researchers can link and compare the emission characteristics of BC and BrC from various sources or various regions. We described above notion in revised version (lines 147-154).

2. More description of the test protocol is needed, e.g. cold start? size of fuel? How was it determined that the test method was relevant for real-world stove emissions?

Response:

Thanks for this suggestion. We optimized the description on the test protocol by accounting for the suggestion (revised version, lines 100-102).

3. Overall, there needs to be an analysis of the uncertainty or the error and potential impacts of the

assumptions in the algorithm? What is the impact of assuming AAE = 1 for BC? How might lensing impact this analysis? What is the impact of measurement limit of detection?

Response: Thanks for the instructive comment. This comment contains 4 questions. We'll deal with them one by one.

(A) Regarding the uncertainty or error and potential impacts of the assumption in the algorithm. The algorithm is a function for F_{BrC}, with AAE as the independent variable, expressed as F_{BrC}=0.5519lnAAE+0.0067. To construct this function, we managed to gather four pairs of F_{BrC} vs AAE values. The first pair is for pure BC (free of BrC), of which AAE and F_{BrC} are 1.0 (Lack and Langridge, 2013; Laskin et al., 2015; Yan et al., 2015; Zhang et al., 2020) and 0.0, respectively. The second and third pairs are for household coal and biomass fuels, respectively, directly measured by our team. Concretely, our previous study on household coal (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. The last pair is assumedly for pure BrC. We averaged over the AAE values in the literature for WSOC or MSOC (free of BC) and obtained 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., 2018) (Table S3 Part I). Uncertainty exists in every pairs and thus in the algorithm. For example, for pure BC (the first pair), Lack and Langridge (2013) estimated that the uncertainty in short wavelength absorption by BC based on an extrapolation using an AAE=1 ranged from +7% to −22%. The other 3 points in Figure 5 are all averages over a cluster of individual dots (samples) and therefore we are able to give error bars for every points (lines 345-348).

(B) Regarding the impact of assuming AAE = 1 for BC. AAE = 1 is actually not an assumption, but derives from Mie Theory – for small particles (the primary particles of BC are of the order of 30 nm) AAE = 1, and this does not change for conglomerates of primary particles (application of Rayleigh-Gans-Debye theory). For this reason, it is no problem to think that the AAE of BC is about 1.0. However it is indeed problematic if we accept that the AAE of BC is a constant of 1.0. Studies show that the AAE values of BC actually vary from 0.8-1.4 depending on BC's source, diameter, and coating manner (Gyawali et al., 2009; Lack and Cappa, 2010; Lack and Langridge, 2013). Here we would like to use the estimate of Lack and Langridge (2013) again: the uncertainty

in short wavelength absorption by BC determined by extrapolation using an AAE=1 ranged from +7% to−22% (lines 347-348).

(C) Regarding how lensing might impact the analysis. As the IS technique uses samples suspended in liquid, the lensing effect is not applicable – if the non-absorbing coatings are soluble in the suspension liquid, they no longer coat the BC particles, and if they are not, the *relative* refractive index of the suspension fluid relative to the typical coating materials is quite near to 1, so the light does not "see" the coating material. A detailed discussion of this feature or the IS method is given by Hitzenberger and tohno (2001). In the revised version, lines 132-135 described the mechanism of the IS method, and lines 136-137 pointed out that the absorption enhancement by the coating is negligible.

(D) Regarding what about the impact of measurement limit of detection (LOD). In this study, we plotted the calibration curves (see Figure S4) for CarB masses from 1.5–90 μg and HASS masses from 3–240 μg according to their respective absorption signals 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 calculation of iteration procedure in subsection 2.4 and Figure S4 in Supporting Information). Although the LODs were not tested when we prepared the calibration curves, all measured concentrations of our samples fell in the ranges: CarB masses from 1.5–90 μg and HASS masses from 3–240 μg. This anyhow reminds us of the importance of LOD.

4. How does the novel algorithm presented here compare with other approaches to quantifying the fractional contribution of BrC and BC to absorption? Is there a benefit to calculating a BrC mass emission factor over other approaches based on AAE? A few studies that may pertain might be Corbin et al. 2018 (https://doi.org/10.1029/2017JD027818), Tian et al. 2019 (https://doi.org/10.1029/2018JD029352), or Zhang et al. 2018 (https://aaqr.org/articles/aaqr-17-12-ac3-0566.pdf) among many others.

Response:

Thanks for this comment. The novel algorithm used in this study can estimate the absorption contribution of BrC in the wide range of solar spectrum from 350-850 nm. Yet other approaches

(e.g., based on AAE method) usually quantify the fractional absorption contribution of BrC in low wavelengths. For example, Corbin et al. (2018), Zhang et al. (2018) and Tian et al. (2019) found that the contributions of BrC to total light absorption were 50%, 37.4% and 41-85%, respectively, at the wavelength of 370 nm. Note that the values of BrC contributions in those examples are not F_{BrC} (integrated over 350-850 nm of solar radiation) but $f_{BrC}(370)$ (the absorption in 370 nm by BrC relative to that by BC+BrC, without accounting for solar spectrum). The f_{BrC} values for coal and biomass fuels in 370 nm were 46.2% (Sun et al., 2017) and 77.9% (this study), respectively, whereas the F_{BrC} values for coal and biomass fuels were 26.5% (Sun et al., 2017) and 50.8% (this study), respectively. Apparently the absorption contribution measured at a single wavelength cannot represent the actual absorption contribution associated with the solar spectrum. This is the biggest advantage of the novel algorithm over some other approaches. The definitions of F_{BrC} and $f_{BrC}(\lambda)$ can be found in subsection 2.4 and the Supporting Information.

5. There is no validation that this algorithm works for sources other than the cookstove samples measured in this study and in Sun et al. 2017. Unless the authors can include some additional data points from some other sources in their algorithm development the statements made throughout the paper about the wide applicability of the algorithm for 'any combustion sources' are unsupported and should be removed.

Response:

We agree to this comment. There is no validation that this algorithm works for sources other than the cookstove samples measured in this study and in Sun et al. (2017). To assure the solidness and rigorousness of our statement, we have revised "in any combustion process" to "perhaps many combustion process" (revised version, line 389).

6. The manuscript needs to be edited for language, see minor comments for specific examples. But generally, if you are writing 'in other words' it means your first explanation should be simplified and stated only once.

Response:

Thanks for this comment. We'll improve our text following the specific comments below.

Specific Comments:

Line 30-31: The sentence needs to be rewritten since the two clauses of this sentence are saying the same thing, BrC absorbs more at shorter wavelengths.

Response:

Thanks. We have deleted "particularly in the ultraviolet (UV) range, on account of there being a larger spectral dependence for BrC than for BC". The remained sentence now is "The light absorption by BrC is more emphasised towards short wavelengths (IPCC, 2014; Pokhrel et al., 2017; Li et al., 2018; Ferrero et al., 2020)" (lines 30-31, revised version).

Line 46: Why are the units in mg/m^2? Most species in the atmosphere are reported in terms of concentration. Is this a typo?

Response:

Thanks for this question. The units in "mg/m^{-2}" here are for column concentration instead of volume concentration. We have changed "high levels of BrC" to "high column concentrations of BrC". (line 45, revised version).

Line71: Need to be clearer about what characteristics are being referred to here. There are many references published on emissions which measure chemical composition, size distribution, and some even quantify optical properties. Most do not report a BrC emission factor because there is no standard for quantifying BrC mass.

Response:

Thanks for this reminder. After a comprehensive consideration of the context of this paragraph, we have decided to delete the sentence "Few studies have addressed the typical sources of emission characteristics (Fan et al., 2016; Lin et al., 2017; Mo et al., 2017; Phillips et al., 2017; Huo et al., 2018; Rawad et al., 2018; Sumlin et al., 2018; Xu et al., 2018; Zhang et al., 2018)" without affecting our intent.

Line 114: These 'soft materials' are usually referred to as kindling and is commonly used when

igniting wood and the emissions from a kindling ignition should be included in this analysis since they are representative of real-world use.

Response:

Thanks for this comment. The paragraph (lines 114-118 in original version and lines 115-120 in revised version) intends to show: (i) in ordinary practice, 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) and therefore additional emissions from the flammable soft materials must be considered; (ii) however in our study, only solid alcohol was used to ignite experimental biomass fuels and almost no pollutants other than CO_2 and H_2O were released from alcohol combustion.

Line 120: What is meant by 'envisaged emission intensity'? How was this determined? Is this just the concentration in the sampling duct?

Response:

Thanks for this question. The emission intensities of different biomass fuels varied greatly, so we have to properly set appropriate dilution ratios for different biomass fuels to meet the experimental needs. The 'envisaged emission intensity' was obtained from two approaches, one from our experiences in household solid fuel combustion experiments, and the other from sufficient pre-experiments.

A stream of flue gas was ducted from the stovepipe into the diluter. That is, the concentration before the diluter was the same as in the stovepipe and the concentration after the diluter was lower than in the stovepipe. The 'envisaged emission intensity' mentioned in this study refers to the concentration inside the stovepipe or before the diluter. The dilution ratios were preset depending on the envisaged emission intensity. Please see the description in lines 119-125.

Line 172: Please cite a reference and quantify how much lower the burning temperature or heat release is for herbaceous fuels to support this speculation.

Response:

Thanks for this suggestion. In lines 189-191, we added a sentence "In this study, the temperature tested in the stovepipe (50 cm above the stove upper surface) for HPs was 62.9 °C

while for LPs, was 77.1 °C ".

Lines 175 – 181: Were no other measurements made during the tests (e.g. CO, CO_2, PM, EC, OC)? These other measurements would greatly support some of the speculation in this section. I am not sure the speculation is justified without measurements from actual study here.

Response:

Thanks for the suggestion. We do have got some data during the tests, including organic carbon (OC), elemental carbon (EC), and modified combustion efficiency (MCE) of every combustion experiment (Table S4 here). OC and EC values were extracted from our previous publication (Sun et al., 2018). These data favor our speculation mentioned in this section.

Table S4 The values of MCEs 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

Lines 205: This paragraph needs to be revised for language usage.

Response:

Thanks for this comment. We have carefully read this paragraph and have tried to improve the language. Particularly, the next two comments of this reviewer and a comment of another reviewer are all regarding this paragraph and have incurred immense changes in the text. The paragraph in our original version has now even been expanded into two paragraphs (lines 221-235 and lines 236-243). We paid great attention to language usage when constructing these two paragraphs.

Line 208: What is meant by 'the significant potential of BrC emissions than BC emissions'? Does this mean larger emissions? Larger mass fractions? Larger BrC/BC ratios? Larger impact? Be specific about what quantity is of BrC emissions is significant and by what amount.

Response:

Sorry for the ambiguity. This sentence has been deleted in our revised version without affecting our intent.

Line 212: Please provide the average absorption efficiencies of BrC and BC that are being referenced for this statement.

Response:

Thanks. We have provided a set of MAE values for BC, BrC, and dust in lines 241-242 (revised version) to show the huge difference between the MAEs of BC and BrC. In Yang et al.(2009), the MAEs at 550 nm were estimated to be 9.5, 0.5, and 0.03 m^2/g, respectively, for BC, BrC, and dust.

Line 255-57: Why are funeral pyres used as an emissions comparison? It seems like an odd source to include and to leave out any mention of open burning (e.g. ag residues, forest fires) or coal for cookstoves. Is coal included in the 'biomass fuels' in mentioned in line 255?

Response:

Indeed, funeral pyres combustion is an odd source for comparison. However the studies regarding the emission factors of biomass BrC were so scarce, we had to mention funeral pyres combustion as one source of information.

In line 255 (original version), coal is not included in the 'biomass fuels' (line 284 in revised version).

Line280: What was the source of the uncertainties in the Lack and Langridge analysis? Do they apply in this study?

Response:

The uncertainty analysis in Lack and Langridge (2013) includes the uncertainty of AAE allocation method (the uncertainty of AAE of BC) and the uncertainty of experiment (the

uncertainty of instrument measurement). We quoted them here just for knowing the potential of uncertainty subject to AAE.

Line 328-331: How does this compare to the direct radiative forcing attributed to BrC referred to in the introduction?

Response:

Thanks for this comment. The F_{BrC} and radiative forcing (RF) are of different concepts. The former refers to "the contribution of absorption by BrC to the total absorption by BC + BrC across the strongest solar spectral range of 350–850 nm" (see lines 16-17 in Abstract), while the latter refers to the difference of insolation (sunlight) absorbed by the Earth and energy radiated back to space (https://encyclopedia.thefreedictionary.com/Radiative+forcing). There is no fixed relation between them. However, the knowledge of F_{BrC} helps identify which one of BC and BrC dominates the light absorption of solar radiation.

Figure 1: Please include error bars to show the uncertainty in the measurement. Presumabley repeat measurements were made because there are standard deviations (standard error?) provided in Table 1.

Response:

Thanks for this suggestion. We have done accordingly.

Table 1: Please include all the quantities measured and calculated for each sample (e.g. AAE, $R_{BrC/BC}$, f_{BrC}, F_{BrC}) along with propagated uncertainties.

Response:

Thanks for this suggestion. The AAE data can be found in Table S2-I, and the $R_{BrC/BC}$ data were added in Table 1. The data of f_{BrC} were both sample-specific (11 biomass fuels and more than 20 coals) and wavelength-specific (stepwise from 350 nm to 850 nm) and therefore were too many; we had to arrange them (for biomass fuels and coals (Sun et al., 2017)) in Table S2-II (Supporting Information). The plots of f_{BrC} for biomass fuel and coal can be seen in Figure 4.

F_{BrC} is just sample-specific and can't be given in every single wavelength like f_{BrC}.

Figure 4: What is the impact of limit of detection on this plot? The data > 750 nm is very noisy, and I wonder if that is not due to limitations of the measurement? If this data is below the limit of detection it should not be used in the calculation of f_{BrC}.

Response:

Thanks for this reminder. The samples of coals and biomass fuels were actually analyzed with the same instrument (Perkin Elmer Lambda 950) during the same period. The status of the instrument was normal and stable then. In Figure 4, although the f_{BrC} at wavelength >750 nm for biomass fuels looked very "noisy", the f_{BrC} at wavelength >750 nm for coals fluctuated very gently. This implies that the larger fluctuation for biomass fuels than for coals in Figure 4 resulted unlikely from the limitation of measurement (instrumental detection limit) but very likely from samples themselves (e.g, chemical composition). Sure it deserves further study in future. Again thanks.

Figure 5 and line 303: Why only use the mean (median?) f_{BrC} from these current study and Sun et al. 2017? Although the regression is strongly correlated here, the scatter in the data is covered up by using the mean value instead of every measured data point.

Response:

The question why only the mean values of F_{BrC} vs AAE rather than the data of each sample were used to construct the function between F_{BrC} and AAE is really worth explaining. The same question had actually been raised by the editor and we had listed the reasons. On the one hand, we know, each of the latter three points (1.58, 0.265; 2.46, 0.508; 6.09, 1.00) in Figure 5 is the average of a number of data, and therefore each of them can be replaced with a cluster of individual dots if we like; yet on the other hand, the first point (0.00, 1.00) is not originated from averaging over a cluster of individuals but from theoretical consideration, and hence there are no cluster of individual dots to replace this single point. Under the circumstances, replacing each of the latter three points with a cluster of individual dots will substantially lower the weight of the first point from 25% to almost being negligible. Given the theoretical significance of the first point, this is not only unfair but also unacceptable. For this consideration, we prefer to use the average value for each of the latter three points so that all the four points in Figure 5 are put equal weight (25%). Additionally,

compared with a cluster of individuals, an average is usually more representative of the true value and hence is more persuasive. We added an explanation in our revised version (lines 340-341).

Uncertainty exists in every pairs and thus in the algorithm. For example, for pure BC (the first pair), 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%. The other 3 points in Figure 5 are all averages over a cluster of individual dots and therefore we are able to give error bars for every points (Figure 5).

SI:

Tables S3 Part I: Extracts are dominated by ambient aerosols, what about source? E.g. fossil fuel combustion, woodstoves, open burning? (Just a few examples are: Xie et al. 2017 https://doi.org/10.1038/s41598-017-06981-8 for open burning and gasoline exhaust; Xie et al. 2018 https://doi.org/10.1016/j.envpol.2018.04.085 for cookstoves using wood, kerosene and charcoal; Corbin et al. 2018 https://doi.org/10.1029/2017JD027818 for marine diesel engines). Since this paper is focused on emissions it would be good to have a more exhaust list of emissions AAE measurements. Calculations: should 'coal' here be 'biomass fuel'?

Response:

Thanks for this reminder. We agree that "Since this paper is focused on emissions it would be good to have a more exhaust list of emissions AAE measurements". In our revised version, the suggested AAEs have been added to Table S3 part I.

We are grateful for the reviewer's carefulness in finding our miswording and have changed 'coal' to 'biomass fuel'.

Figures S2-S4: Appear to be identical to those in Sun et al. 2017, should the reference be noted in the caption?

Response:

Thanks. We have added the reference of 'Sun et al., 2017' in Figure S2-S4.

Figure S4: Hard to follow the text here, would be easier to understand in equation form or even a

diagram.

Response:

Thanks for this suggestion. We'd like to add a flow chart in Supporting Information (Figure S5), so that readers could understand the mechanism of iterative process used in this study more easily.

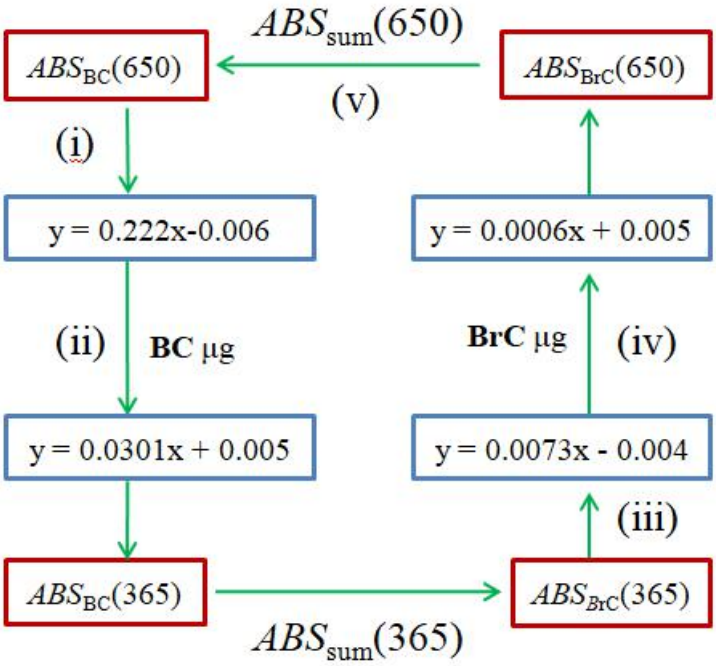

Figure S5 Calculation of BC and BrC with iterative process

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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

Jianzhong Sun[1, 2, #], Yuzhe Zhang[1#], Guorui Zhi[1, *], Regina. Hitzenberger[3], Wenjing Jin[1], Yingjun Chen[4, *], Lei Wang[1], Chongguo Tian[5], Zhengying Li[1], Rong Chen[6], Wen Xiao[7], Yuan Cheng[8],Wei Yang[2], Liying Yao[2], Yang Cao[2], Duo Huang[2], Yueyuan Qiu[2], Jiali Xu[2], Xiaofei Xia[2], Xin Yang[2], Xi Zhang[2], Zheng Zong[5], Yuchun Song[9], Changdong Wu[9]

[1]State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy of Environmental Sciences, Beijing 100012, China

[2]Shangrao Normal University, Shangrao 334001, China

[3]University of Vienna, Faculty of Physics, Boltzmanngasse 5, 1090 Vienna, Austria

[4]Shanghai Key Laboratory of Atmospheric Particle Pollution and Prevention (LAP3), Department of Environmental Science & Engineering, Institute of Atmospheric Sciences, Fudan University, Shanghai 200433, China

[5]Key Laboratory of Coastal Environmental Processes and Ecological Remediation, Yantai Institute of Coastal Zone Research, Chinese Academy of Sciences, Yantai 264003, China

[6]College of Physical and Health Education, East China Jiaotong University, Nanchang 330006, China

[7]School of Physical Education, Jiangxi Science & Technology Normal University, Nanchang 330006, China

[8]State Key Laboratory of Urban Water Resource and Environment, School of Environment, Harbin Institute of Technology, Harbin 150001, China

[9]Jiangxi Sports Hospital, Nanchang 330006, China

*_Correspondences to_: Guorui Zhi (zhigr@craes.org.cn) and Yingjun Chen (yjchenfd@fudan.edu.cn)

[#]These authors contributed equally to this work

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 geomean 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}. (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 any combustion sources (F_{BrC} = 0.5519ln(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 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 were tested: they were classified into three groups, i.e. 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. The stove that we used in this study was a natural draft stove developed specifically for biomass fuels (see Figure S1 in Supporting Information). 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 (30–50 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 83.95% (peanut stalk) to almost 100% (Sorghum stalk), with an average of 93.86 ± 5.93%, 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 is important because no pollutants other than CO_2 and H_2O are released from alcohol combustion.

A diversion-dilution-sampling system (Supporting Information, Figure S2) was set up to sample and/or monitor the combustion emissions. The dilution ratios were 20:1 to 80:1, depending on the envisaged emission intensity of each combination process, as well as on the burning conditions. 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

136 the light absorption by the collected samples. As the samples are immersed in a liquid, the absorption
137 enhancement by possible non-absorbing coatings is negligible (Hitzenberger and Tohno, 2001;
138 Wonaschütz et al., 2009; Sun et al., 2017).

139 Two reference materials were used as proxies for BC and BrC. They were carbon black (CarB) (e.g.
140 Elftex 570, Cabot Corporation) for BC (Fisher, 1970; Andre et al., 1981; Heintzenberg, 1982;
141 Hitzenberger et al., 1996; Wonaschütz et al., 2009) and humic acid sodium salt (HASS) (e.g. Acros
142 Organics, no. 68131-04-4) for BrC (Wonaschütz et al., 2009). CarB had been used as proxy for BC in
143 diesel exhaust by Medalia et al. (1983) and HASS had been used as proxy for BrC from wood
144 combustion by Wonaschütz et al. (2009). In a previous study, CarB and HASS were used as proxies for
145 BC and BrC, respectively, to characterise household coal burning samples, by assuming that BC and
146 BrC in household coal emissions had the same light-absorbing properties as CarB and HASS,
147 respectively (Sun et al., 2017). In the present study, we continued this logic, and assumed that BC and
148 BrC in household biomass smoke have the same light-absorbing properties as CarB and HASS,
149 respectively. In other words, the reported BC and BrC masses here are essentially CarB-C-equivalent
150 and HASS-C-equivalent, respectively, from the perspective of light absorption and are different from
151 those measured by other measurement techniques (e.g., thermal–optical method or aethalometer) (Chen
152 et al., 2006; Zhi et al., 2008, 2009; Shen et al., 2013, 2014; Aurell and Gullett, 2013) or reference
153 materials (e.g., fulvic acid, humic acid, or humic-like substances) (Duarte et al., 2007; Lukács, et al.,
154 2007; Baduel et al., 2009, 2010). Although such an assumption is not fully perfect, researchers can take
155 advantage of these two reference materials to relatively assess the features (chemical or optical) of BrC
156 and BC derived from different combustion sources or regions. It should be noted that the IS method
157 does not depend on an actual chemical separation, but on a virtual optical allocation of a mixed
158 absorption signal to BrC and BC, with HASS and CarB used as references, respectively.

159 Calibration curves (see Figure S4) were plotted for CarB masses from 1.5–90 μg and HASS masses
160 from 3–240 μg, according to their respective absorption signals as measured by the IS device, at both
161 650 nm and 365 nm (Sun et al., 2017). The BrC and BC masses of the samples were calculated through
162 an iterative procedure based on the different spectral dependences of absorption by BrC and BC (See
163 methods for the calculation using iteration procedure and Figure S4 in Supporting Information). 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. The HASS used in this study was from Acros Organics. It had an AAE of 1.86. 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 Supporting Information.

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} (7.259 ± 0.002 g/kg), whereas pellet fuel had the lowest (0.13 ± 0.061 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 the 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 ± 6.49% < 95.9 ± 2.05%), resulting in a greater chance for the formation of BrC (Shen et al., 2013). A similar phenomenon was also observed by Shen et al. (2013), who carried out a systematic measurement of PM, OC, and EC released from various solid fuels burned in residential stoves; these authors found that crop residues, which were composed of herbaceous plants, were more likely to have higher BrC EFs than wood fuels, which were composed of ligneous plants. 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

220 combustion of bituminous coals in BrC emissions.

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

236 More importantly, each of the 11 biomass fuels tested in this study had a higher EF_{BrC} than EF_{BC};
237 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
238 6.7 ± 2.7. Kirchstetter et al. (2004) measured the light absorption by filter-based aerosol samples from
239 biomass burning before and after acetone treatment (which removed OC). They found that 50% of total
240 light absorption was attributable to OC. In view of the much smaller average absorption efficiency of
241 BrC relative to that of BC (for example, Yang et al. (2009) reported that the MAEs at 550 nm were 9.5,
242 0.5, and 0.03 m^2/g, respectively, for BC, BrC, and dust), the contribution of BrC to the mass of total
243 LAC is undoubtedly far higher than that of BC, an inference which is consistent with the present study.

244 **3.2 Spectral dependence of absorption**

245 AAE represents the spectral dependence of the light absorption efficiency (Martinsson et al., 2015;
246 Washenfelder et al., 2015; Yan et al., 2015). Usually, the AAE is close to 1.0 (Lack and Langridge,
247 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 Supporting Information).

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 ($R_{BrC/BC}$ = + ∞); 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 (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).

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

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 Supporting Information. 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), 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 $f_{BrC}(\lambda)$ is given in the Supporting

Information. 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 Supporting Information. The results of $f_{BrC}(\lambda)$ for biomass fuels in this study are plotted in Figure 4 (blue line).

Evidently, the $f_{BrC}(\lambda)$ increased towards shorter wavelengths: the $f_{BrC}(\lambda)$ at 850 nm was 0.25, whereas the $f_{BrC}(\lambda)$ at 350 nm increased to 0.8. In addition to the spectrally-dependent $f_{BrC}(\lambda)$ for biomass fuels, Figure 4 also presents the spectrally dependent $f_{BrC}(\lambda)$ values for coal (red line) as obtained in a previous study (Sun et al., 2017). The lowest value of $f_{BrC}(\lambda)$ for coal occurred at 0.061 (850 nm), and the highest value occurred at 0.47 (355 nm). The average $f_{BrC}(\lambda)$ 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 $R_{BrC/BC}$ 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 $f_{BrC}(\lambda)$ 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 Supplementary Information 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 $R_{BrC/BC}$. There is a scarcity of reported $R_{BrC/BC}$ values, whereas conversely AAE is frequently reported in the existing literature. Therefore, the logarithmical function that can be fitted to the relationship between $R_{BrC/BC}$ 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.

$$F_{BrC} = 0.5519 \ln AAE + 0.0067 \qquad (R^2 = 0.999) \qquad (2)$$

Equation (2) provides a novel algorithm for deriving F_{BrC} from AAE, without consideration of the process details for 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 $AAE_{\text{-open}}$ data from available journal articles and included them in Table S3 (Part II). The calculated average $AAE_{\text{-open}}$ value was 3.44 ± 1.75, which was larger than the $AAE_{\text{-contained}}$ value obtained in this study (2.46 ± 0.53). Substitution of the $AAE_{\text{-open}}$ 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 $AAE_{\text{-contained}}$ and $AAE_{\text{-open}}$ identified above apply to global biomass burning, we can now assess BrC's role in the biomass burning globally (contained + open) ($F_{BrC\text{-entire}}$), 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 $F_{BrC\text{-}entire}$ can be calculated according to the following equation:

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

With Equation (2), the distribution of $F_{BrC\text{-}entire}$ was simulated through the Monte Carlo approach, as shown in Figure 6. The $F_{BrC\text{-}entire}$ was 0.644 on average, and with an 80% probability range it lay between 0.585–0.699. Particularly, the probability of $F_{BrC\text{-}entire}$ 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 out data $F_{BrC\text{-}entire}$= 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 ($F_{BrC} = 0.5519\ln(AAE) + 0.0067$, $R^2 = 0.999$), 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 ($F_{BrC\text{-}open}$ = 70.1%) and entire biomass burning ($F_{BrC\text{-}entire}$ = 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	7.259 ± 0.002	2.537 ± 0.001	2.86 ± 0.018
Rice straw	2.50 ± 3.064	0.31 ± 0.25	8.06 ± 6.67
Wheat straw	1.25 ± 0.074	0.13 ± 0.039	9.62 ± 5.17
Cotton straw	0.89 ± 0.51	0.10 ± 0.019	8.91 ± 2.99
Bean straw	0.57 ± 0.12	0.089 ± 0.035	6.41 ± 2.21
Corncob	0.56 ± 0.55	0.056 ± 0.017	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 ± 0.389
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 ± 0.061	0.023 ± 0.037	5.65 ± 2.58
Geomean	0.71 (0.24, 2.09)	0.12 (0.033, 0.436)	5.90 (3.26, 10.68)

Note: The last row for geomean is expressed as geomean (lower limit, upper limit). The lower/upper limits are calculated via geomean 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.

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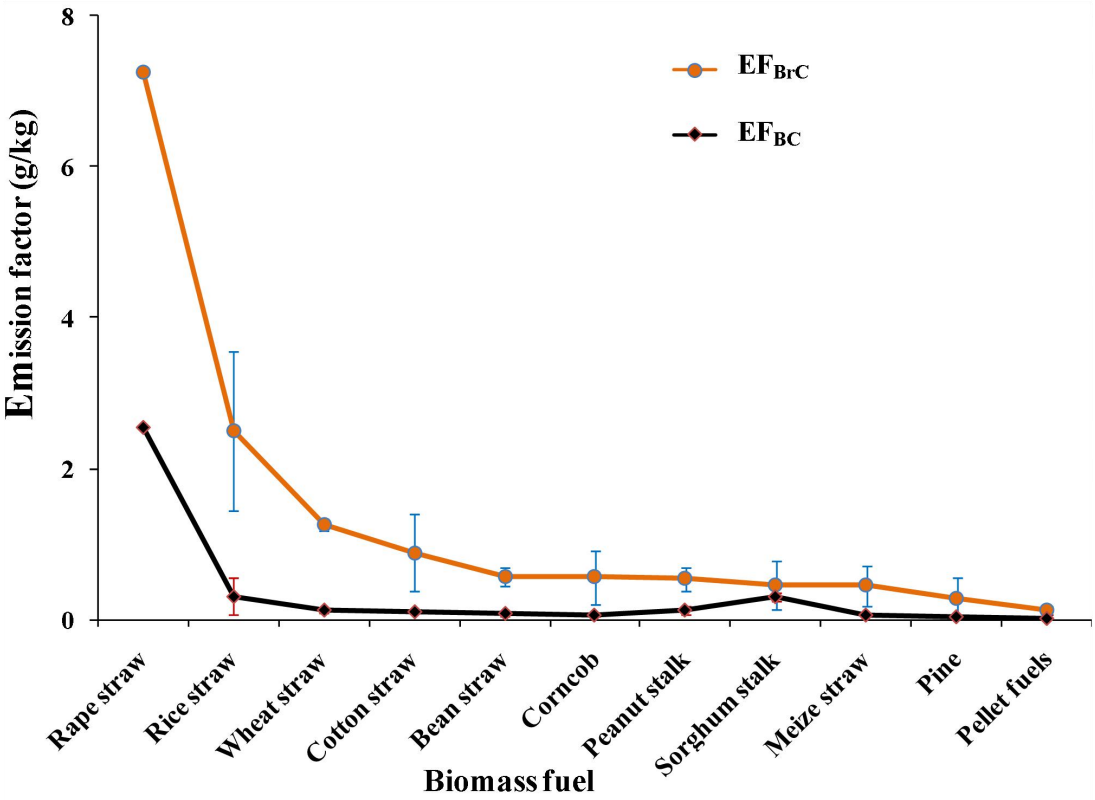

Figure 1. EFs of tested biomass fuels

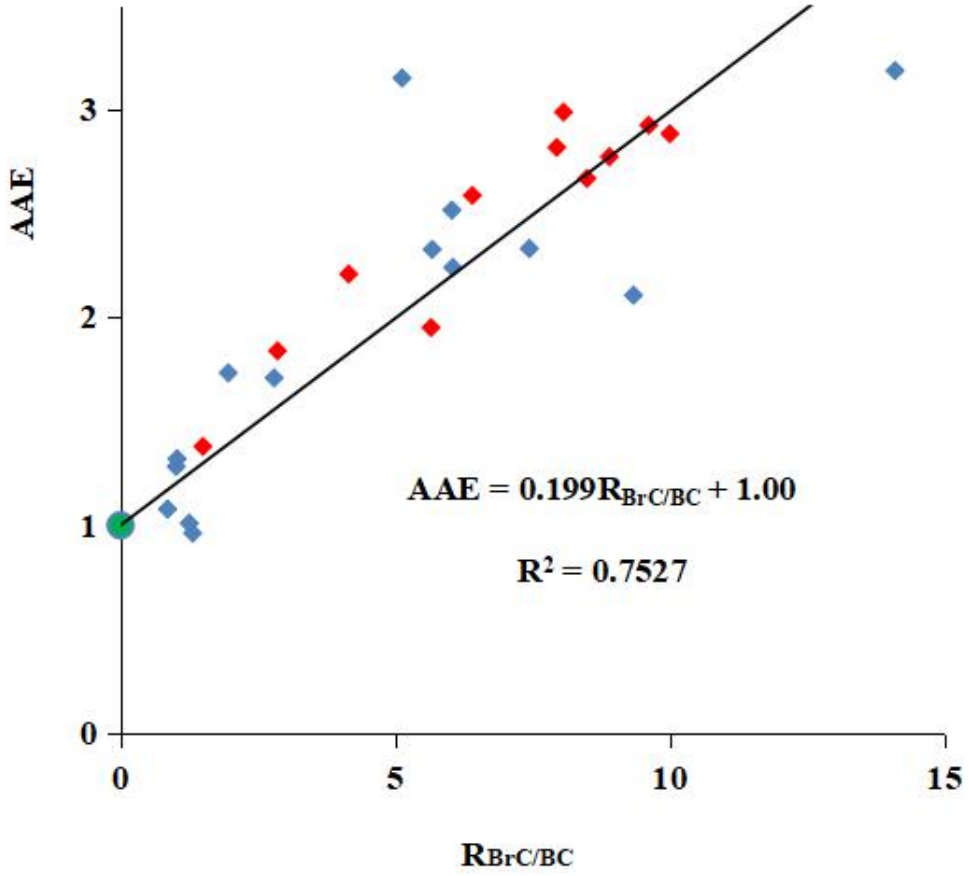

Figure 2. Relationship between AAE and EF_{BrC}/EF_{BC} ratio ($R_{BrC/BC}$) for both biomass fuel (red) and coal (blue). The intercept is designated as 1.0 to echo the conventionally accepted notion that the AAE for pure BC (i.e., $R_{BrC/BC} = 0$) is 1.0.

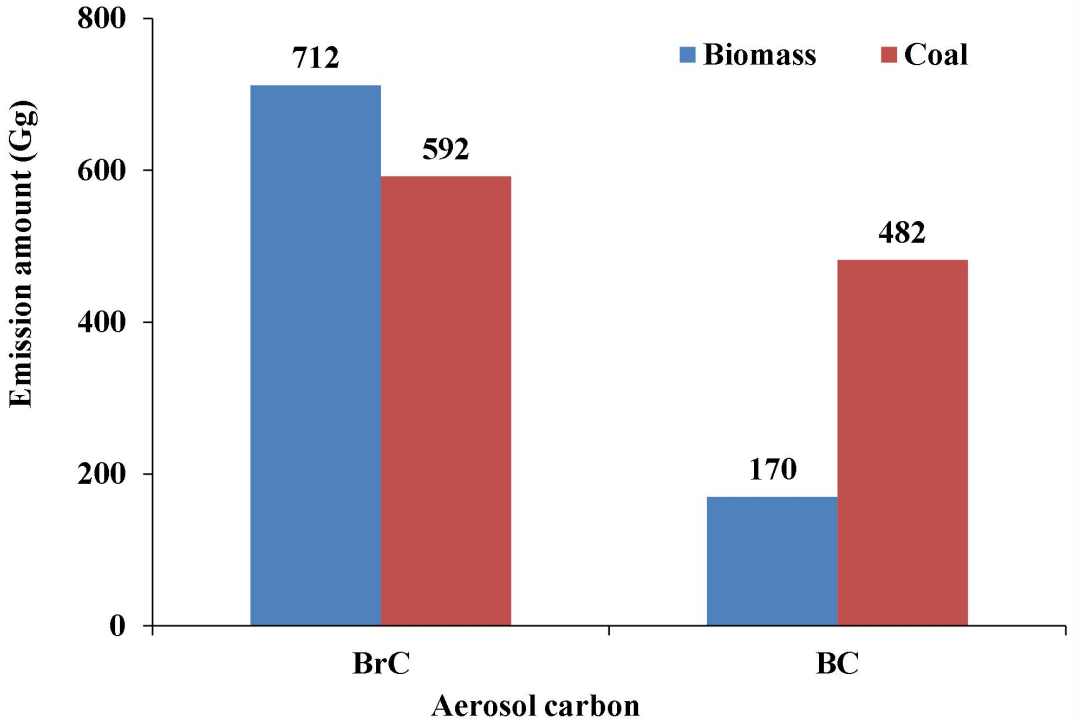

Figure 3. Comparison of BrC and BC emissions between biomass burning and coal combustion in China's household sector of 2013

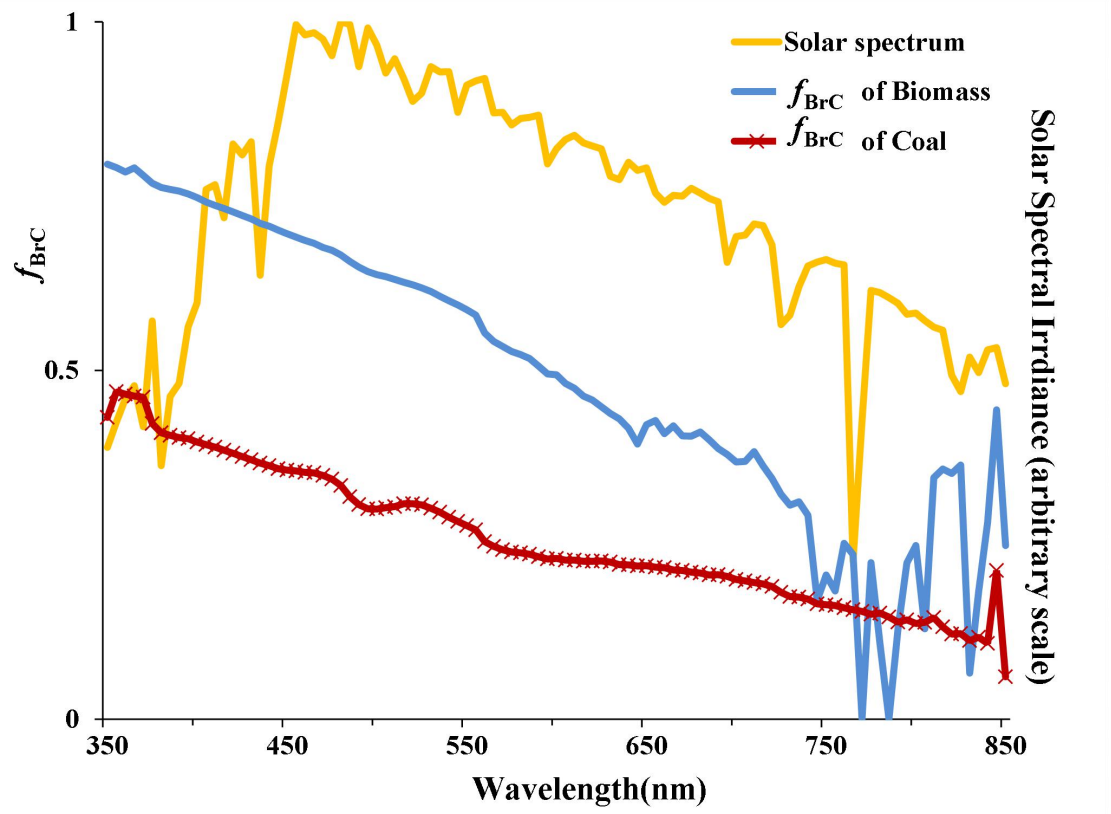

Figure 4. Ratios of light absorption by BrC to total absorption by total mass with respect to China's household biomass and coal burning

Note: The ratio is expressed as f_{BrC} and was calculated in accordance with the method described in the Supporting Information. 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)

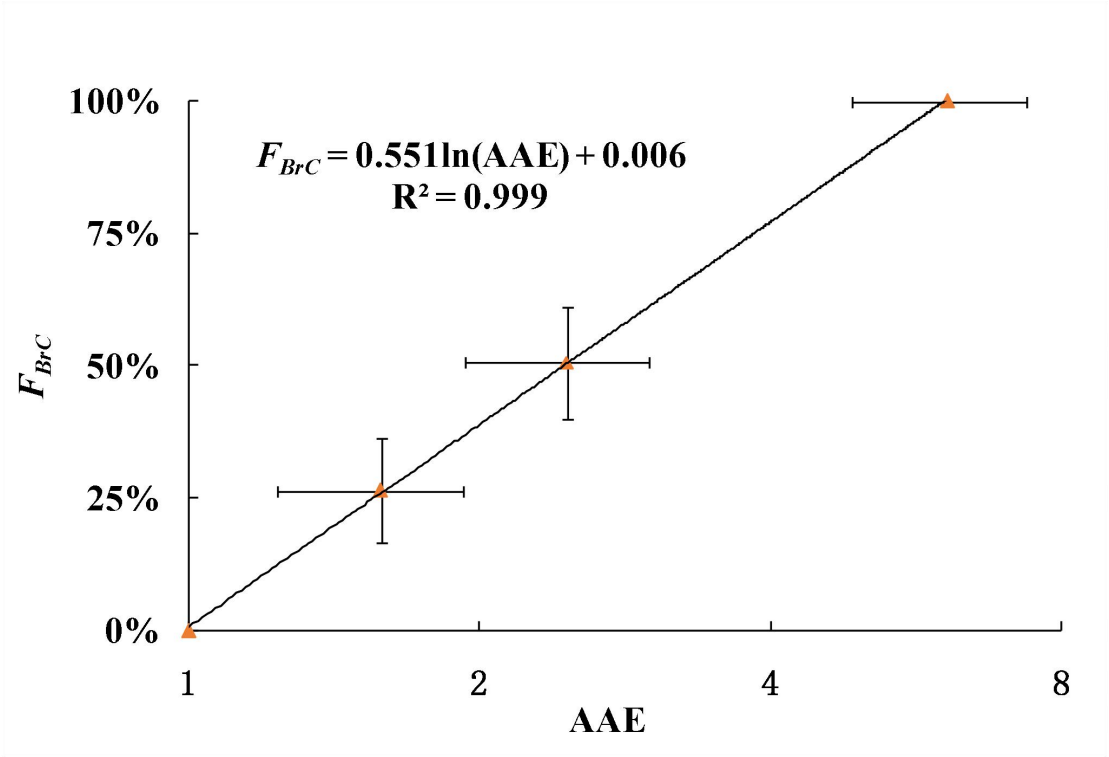

Figure 5. Relationship between F_{BrC} and AAE

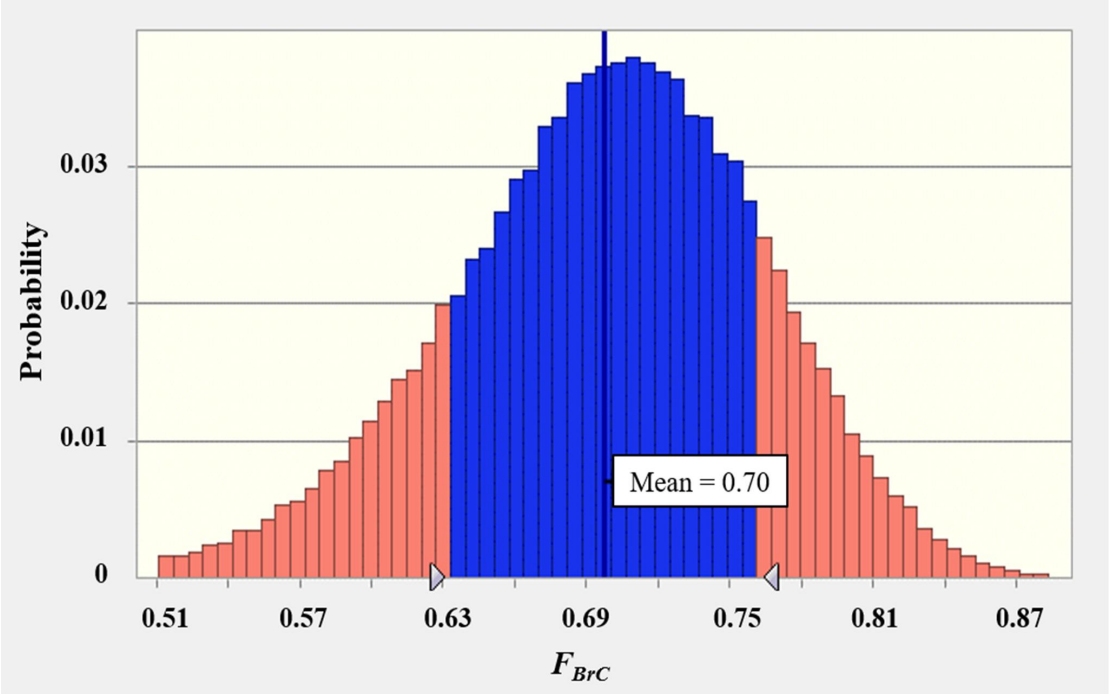

Figure 6. The probability distribution of calculated $F_{BrC\text{-}entire}$. Assuming the $AAE_{-contained}$ value of 2.46 ± 0.16 (mean ± SD of the means) and AAE_{-open} value of 3.44 ± 0.42 (mean ± SD of the means) apply to whole world biomass burning, the combined value for entire biomass burning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