**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.

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**Reply to Editor,**

EDITORIAL REVIEW:

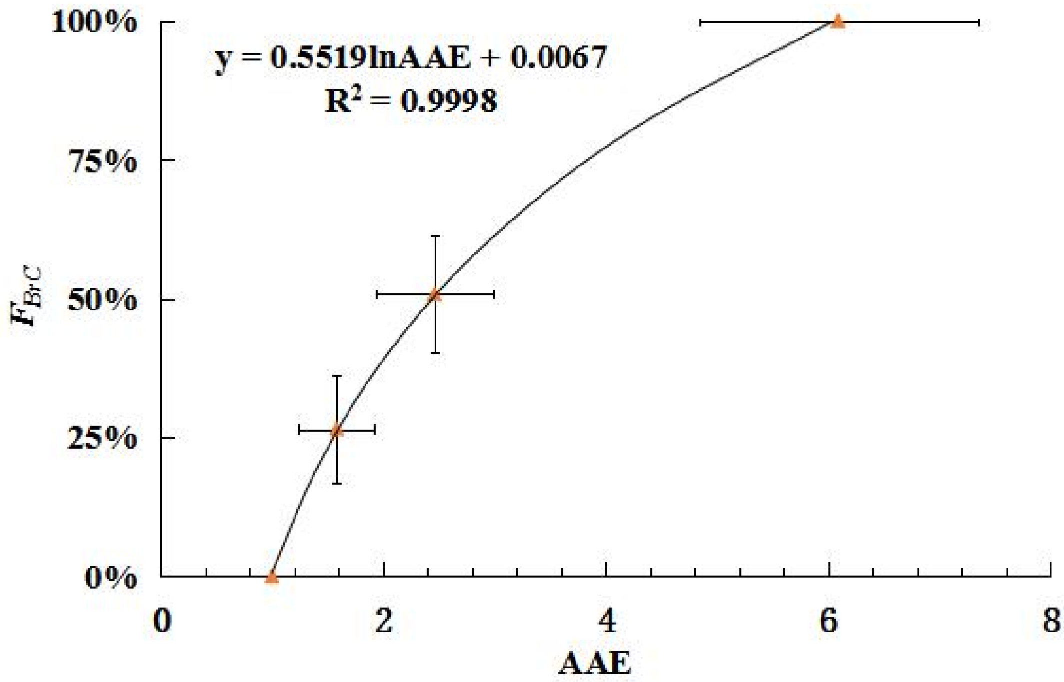
1. L343: the 4 points in figure 5 fall on a straight line. I am curious why you elect to complicate the fit by using a logarithmic function? If you insist on using logarithmic fit you need to provide a theoretical justification for it. If there is no theoretical reason, and the fit is purely empirical, using the simplest possible function is better.

**Response:**

Thanks for this comment. The fit in Figure 5 sure was from the data of our studies and existing literature and is basically empirical. Although the 4 points in the Figure looks like a straight line, it actually is a curve as the x axis is not scaled uniformly but geometrically.

As for why we expressed the empirical relation with a logarithmic function instead of others is (i) when AAE=1 (pure BC), $F_{BrC}$ = 0, which complies with the property of logarithmic function, i.e., log1.0 = 0 and (ii) the fitted logarithmic function displayed a significant correlativity between $F_{BrC}$ and AAE ($F_{BrC} = 0.5519\ln AAE + 0.0067$, $R^2 = 0.9998$).

Given the above facts, in our newest version, we changed geometrically scaled x axis to uniformly scaled x axis while maintained the logarithmic relation in Figure 5, as below:




2. Table 1: Some digits can be cut from numbers without loss of information. For example, 2.50 +/- 3.064 can be replaced by 2.50 ± 3.06 as there is no reason to specify error in the digit that is not even listed. Same for 1.25 +/- 0.074, 1.51 +/- 0.389, etc. As I mentioned in the initial review, your EF error for rape straw is unphysically small leading to unnecessarily precise EFs reported here. I am pretty sure that using more conservative measurements uncertainties would increase the reported error, perhaps from 0.002 to more than 0.01 (allowing you to cut one significant digit out). Are you comfortable reporting an emission factor with 4 significant digits when the rest of them have 2-3?

**Response:**

Thanks for this suggestion. We followed the suggestion and replaced 2.50 ± 3.064 with 2.50 ±3.06, replaced 1.25 ± 0.074 with 1.25 ± 0.07, and replaced 1.51 ± 0.389 with 1.51 ± 0.39. Actually we examined all the data in Table 1 and cut the digits after the decimal point to two. This includes the data for the rape straw from 7.259 ± 0.002 to 7.26 ± 0.01 ($EF_{BrC}$), from 2.537 ± 0.001 to 2.54 ± 0.01 ($EF_{BC}$), and from 2.86 ± 0.018 to 2.86 ± 0.02 ($R_{BrC/BC}$).

3. L118: Use of alcohol presumably makes MCE higher than it would have been without it. Should it be mentioned?

**Response:**

We agree to this comment and mentioned it in our newest version (lines 121-123).

4. L206: This is an important practical recommendation from your study, and if this is not widely known it may be worth inserting a sentence about the benefits of biomass briquetting in the abstract.

**Response:**

Thanks for this suggestion. In the Abstract we have inserted a sentence about the benefits of biomass briquetting (lines 12-14).

TECHNICAL CORRECTIONS:

5. L11: geomean -> geometric mean (also make this change in other places in the text such as Table 1)

**Response:**

Thanks for this reminder. We checked throughout the manuscript and changed "geomean" to "geometric mean".

6. L92: Eleven biomass fuels were tested: they were classified into three groups, i.e. crop -> Eleven biomass fuels tested in this work were classified into three groups: crop

**Response:**

Thanks for this suggestion. We have done accordingly in line 94-95.

7. L111: 93.86 ± 5.93% -> 93.9 ± 5.9%,

**Response:**

Thanks for the suggestion. All similar cases have been updated accordingly, as follows:

- 83.95% (line 110 in previous version) was changed to 84.0% (line 112 in newest version)
- 93.86 ± 5.93% (line 111 in previous version) to '93.9 ± 5.9% (line 113 in newest version)

8. L141: (e.g. Acros -> (Acros

**Response:**

Thanks for this suggestion. We have done accordingly (newest version, line 147)

9. L154: not fully perfect -> not perfect

**Response:**

Thanks for this suggestion. We have deleted ‘fully’ in the sentence (newest version, line 160).

10. L221: aids to compare -> compares

**Response:**

We have changed ‘aids to compare’ to ‘compares’ (newest version, line 224).

11. L274: are collated and arranged in a scatter plot (Figure 2) -> are collated and arranged in a scatter plot in Figure 2

**Response:**

Thanks for this suggestion. We have done accordingly (newest version, line 277).

12. L327: logarithmical -> linear

**Response:**

Thanks for this suggestion. We actually kept the logarithmical relation unchanged (newest version, line 344). In response to editor’s comment 1, we have explained why we maintained the logarithmic relation for Figure 5.

Again we thank the editor for above suggestions, which are very helpful to further improve our manuscript.

**Reply to Reviewer 1,**

**Additional minor comments:**

Many thanks for recommending publication of our manuscript after the following minor comments are addressed. We have accordingly revised our manuscript, as follows.

1. Line 121 – Instead of "envisaged emissions intensity of each combination process" state "desired PM2.5 concentration in the dilution system" that way the reader can understand what criteria was used and present the target concentration as well. The sampling concentration is an important factor in the partitioning of semi-volatile species, which may be contributing to BrC absorption that was collected on the filter. There is semi-volatile BrC – see Xie et al. 2020 https://doi.org/10.5194/acp-20-14077-2020 -which may end up being collected on the filter depending upon the sampling concentrations.

**Response:**

Thanks for this insightful comment. It does be important to arrange an appropriate dilution factor for each biomass fuel so that the light absorbance of absorbing aerosols (BrC+BC) deposited on a filter falls in the linear range of integrating sphere (IS) approach. Pre-experiments were carried out in advance to decide on the weight of a biomass fuel to be burned and the ratio of a dilution to be set. For each biomass fuel, the weight burned and the ratio diluted are both presented in Table S1-II in the newest version of Supplement. We also gave a brief description of the principle and specific arrangement of flue gas diluting in lines 125-126 (newest version).

We agree to the effect of the sampling concentration on the partitioning of semi-volatile species and added a new sentence in lines 127-128 (newest version) to highlight the implications.

2. Line 168 – 171 – The authors note that their reference material CarB has an AAE of 0.91 and HASS has an AAE of 1.86. It is problematic that the HASS reference material has an AAE that is far lower than the BrC values listed in table S3. The authors need to discuss the implication of using a reference material with a much lower AAE than other BrC sources. The authors should also report the MAE for each of the reference materials to facilitate comparisons with other approaches to quantify BrC mass.

**Response:**

Thanks for this meaningful comment. We also have noticed that the HASS reference material used in our study had an AAE that was far lower than the BrC values listed in table S3.

Since BrC is not a pure substance but a collection of light-absorbing organic substances, the AAE values of BrC of individual samples vary a lot depending on what light absorbing organic substances comprise the BrC. In this sense, it's actually impossible to identify a substance that can precisely represent all BrC substances. However, the need for quantification of BrC implies a need for such a material that is close in some aspects to ordinary BrC species and can be used as a reference or standard in some degree. To this end, the past decades saw HASS being chosen as the reference of BrC (e.g., Wonaschütz et al., 2009; Sun et al., 2017) just as the carbon black (CarB) was chosen as the reference of BC (e.g., Medalia et al., 1983; Hitzenberger et al., 1996; Hitzenberger et al., 2001, 2006; Reisinger et al., 2008 ). HASS is an organic substance that has some similar properties to BrC (e.g., brown color, with an AAE value apparently higher than 1.0). Moreover HASS is chemically stable and water-soluble, and can be conveniently used for the preparation of a standard solution for calibration purpose.

The AAE of HASS is more than twice that of CarB, which suffices the iterative calculation between two designated wavelengths: 365 nm and 650 nm. In this study (newest version, lines 153-162), we stated that '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'.

The MAE values of HASS and CarB were given in lines 174-177, respectively (newest version).

3. Lines 200 – 220 – The authors should present the BrC EFs from Shen et al. 2013, that was referenced on line 194-197.

**Response:**

Thanks for this suggestion. We checked the paper published by Shen et al. (2013) and found no BrC EFs explicitly or implied. Very sorry for the mis-citing. We have deleted the text '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'(lines 193-197 in previous version).

4. Line 289 – The authors should include a reference for the source of the funeral pyre emissions estimate.

**Response:**

Thanks for the suggestion. We added a reference for the source of the funeral pyre emissions estimate (newest version, line 293).

5. Figure 4 – How can you state that the absorption of the samples was above the limit of detection at wavelengths in the 750 – 850 nm range when absorption for the reference material was only quantified at 650 nm? For combustion generated PM the absorption generally decreases as the wavelength increases so being above the limit of detection at 650 nm does not imply that the measurement is above the limit of detection in the range of 750 – 850 nm. Also, there should be error bars on this figure since this is representing multiple samples.

**Response:**

Thanks a lot for this comment. We acknowledge that being above the limit of detection at 650 nm does not necessarily imply that the measurements of combustion samples are above the limit of detection in the range of 750 – 850 nm because the absorption of combustion generated PM generally decreases as the wavelength increases. This didn't influence the results of emission factors and the overall profiles of wavelength dependent $f_{BrC}$ (The emission factors obtained through iterative calculation are only relevant to the wavelengths of 365 nm and 650 nm).

In the newest version, we added error bars not only on the blue line ($f_{BrC}$ for biomass in this study) but also on the red line ($f_{BrC}$ for household coal in previous study [Sun et al., 2017]) in Figure 4.

6. Equation 2 and Figure 5 – It should be noted that this equation only applies to a range of AAEs, since AAE's larger than their max value will result in fractions greater than 1.

**Response:**

Thanks for this suggestion. The paragraph of lines 333-345 describes how Equation 2 is derived, which implies the domain of the function ranges from AAE = 1.0 (first point) to AAE = 6.09 (last point). We gave explanatory note after the expression of Equation 2 (see newest version, line 346).

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