Response to Reviewer 1's comments

This manuscript by Basnet et al. reports results of emission factors and light-absorption properties of carbonaceous aerosol emissions from residential biomass combustion appliances. The experiments involved an extensive set of fuels (7) and appliances (15). The major measurements involved (i) offline thermal-optical analysis to determine emission factors of elemental carbon (EC) and organic carbon (OC), (ii) online measurements of light-absorption at 7 wavelengths using an aethalometer, and (iii) online measurements of size distributions using a low-pressure impactor. The major analysis involved apportioning absorption to either black carbon (BC) or brown carbon (BrC) based on the assumptions that (i) only BC absorbs at 880 nm and (ii) BC absorptions exhibits a wavelength dependence with AAE = 1. The results show variable contributions to absorption by BC and BrC, with fuel moisture content playing an important role.

We thank Reviewer 1's positive comments and constructive suggestions for improving the manuscript. The responses to the comments are addressed in blue text. The line number refers to the lines in the revised manuscript.

Major comments:

1) There are a lot of previous studies that quantified BrC and BC absorption from residential wood burning. It is not clear if/how this study provides any new insights beyond what is already in the literature. For this paper to be suitable for publication in ACP, it needs to clearly identify the new knowledge generated from the experiments. Given the large data set in this study, there is probably potential for deriving new useful knowledge. However, this is not clear in the current version of the paper. A couple of examples of how the paper could potentially highlight new/important/useful results:

While numerous investigations have quantified the absorption of BrC and BC arising from residential wood burning, the majority of these investigations primarily concentrate on assessing ambient air quality for which the residential wood burning emissions occur as mixed with other air pollutants. Thus, there has been a lack of measurement data on wavelength-dependent absorption for residential wood combustion emissions without any mixing factors (such as other pollutants, photochemical/dark atmospheric transformation, etc.). There has been specifically limited information about BrC emissions and light absorption wavelength dependency from European residential wood combustion appliances. As aethalometers are widely used for, e.g., air quality monitoring, our extensive dataset of the wavelength-dependent absorptions of emissions originating from European residential wood combustion can be used for improved source apportionment.

We thank the reviewer for these suggestions, which we in the following respond to point-by-point.

1.1) Discuss how emission factors from European residential combustion is currently quantified in emission inventories. Can the results obtained here (Figure 1) help improve these emission inventories?

Figure 1 illustrates the emission factors for organic carbon and elemental carbon, derived from the offline thermal-optical analysis along with the equivalent black carbon data obtained from the aethalometer. These emission components are widely covered in emission inventories. However, this study provides useful emission factors for the latest modern northern European appliances. Further, BrC is presently absent from the European emission inventories. The results of this paper enable the inclusion of BrC-induced absorption in emission inventory assessments. The contribution of BrC to the total light absorption for the studied wood combustion appliances is shown in Figure 5 (previous Figure 6).

We have improved the manuscript by adding discussion on this to the introduction (lines 49-55) and conclusions (lines 528-530) sections:

page 3, lines 49-55: "The accurate quantifying of the amount and impacts of the absorbing aerosols emitted from RWC is challenged by the gaps in knowledge regarding the particle optical properties and potential variance in emission factors (EFs). However, only a few RWC appliance types and fuels have been studied (e.g., Fang et al., 2022; Martinsson et al., 2015; Zhang et al., 2020; Saleh et al., 2013; Kumar et al., 2018; Saliba et al., 2018; Olsen et al., 2020; Li et al., 2022; Leskinen et al., 2023; Tissari et al., 2019; Hartikainen et al., 2020; Savolahti et al., 2016, 2019; Sun et al., 2021), which causes uncertainties in assessing the direct radiative forcing of RWC emissions. Inclusion of BrC in emission inventories is further constrained by the lack of characterization and quantification of BrC emissions."

page 18, lines 528-530: "In addition to providing BC emission factors for the modern RWC appliances, these findings can also aid in integrating the BrC-induced absorption in emission inventory assessments using the multi-wavelength aethalometer data from air quality monitoring networks."

1.2) The paper mentions that moisture content is more important than the type of appliance in dictating BrC emissions. However, the results (Figures) are not formulated in a way to make use of this finding, or to clearly show that this assertion is valid to begin with. I think that the paper can possibly be restructured to focus on the effect of moisture content versus appliance type. In order to do that, the paper needs to establish the significance of the ranges of moisture contents used in the experiments: Is this variability typical in residential appliances? Data (e.g. BrC absorption) needs to be plotted as a function of moisture content to actually show that moisture content is indeed important and that the data does not cluster based on appliance type.

Fuel moisture content may indeed be one of the factors influencing the brown carbon content in the exhaust. Unfortunately, the impact of fuel moisture content was only studied in detail for two of the sauna stoves, and restructuring around moisture content might conceal other influencing factors. In fact, fuel moisture content alone does not correlate well with BrC contribution. We find the best correlation between particulate OC/EC and BrC contribution, as depicted in Figure 3. Both OC/EC and BrC contribution seems to be influenced by several factors, including fuel moisture, appliance type, wood fuel, and modified combustion efficiency. We have revised the discussion and conclusions texts in the manuscript to better reflect this complexity (i.e., fuel moisture is not a single major parameter influencing the BrC contribution).

We have revised lines 22-23 in the abstract: "Additionally, $BrC_{370\text{-}950}$ was clearly influenced by the fuel moisture content and the combustion efficiency, while the effect of combustion appliance type was less prominent."

A discussion of the influencing factors was added on lines 507-510. "The $BrC_{370\text{-}950}$ varied greatly (ranging from 1.28 % to 20.8 %) for wood log combustion events and was primarily influenced by fuel moisture content and modified combustion efficiency but also by the combustion appliance type. The highest $BrC_{370\text{-}950}$ contributions were observed for the fuels with the highest fuel moisture contents due to the decreased combustion efficiency."

2) The figures are often hard to follow and are not discussed well in the text. For example:

We have updated the figures and the discussion on their implications in the accompanying text. The changes are described in the following answers.

2.1) Most of the details in Figure 4 are not discussed in the text. Why were these specific 8 experiments chosen?

Figure 4 provides a comprehensive overview of the temporal changes in total absorption by BrC and $AAE_{470/950}$ during the progression of the experiment. Given the diverse set of experimental data involving various combustion appliances, the selection of these eight figures aimed to encompass at least one experiment from each appliance.

The updated version of this manuscript has moved this figure to the supplementary material (Figure S12, previously Figure 4) while explaining it in the main text (line 410). "The temporal diversity in the $AAE_{470/950}$

is illustrated for all the different fuel and combustion appliance combinations used in this study in the exemplary time series of data in Figure S12."

2.2) It is not clear what the purpose of Figure 5 is. Figure 5, which has 9 panels, is referred to only once in the text, rather in passing.

Figure 4 (previously Figure 5) summarizes the key findings of this manuscript by illustrating the total BrC absorption within the 370-950 nm range. This is presented as the difference between the overall absorption provided by the aethalometer and the absorption by BC when $AAE_{BC} = 1$.

We recognize the need for detailed referencing of figures in the text and we have provided more thorough explanations in the updated version of the manuscript (line 444 onwards). "Figure 4 summarizes the total absorption coefficients by BrC for the different combustion appliances, with the absorption by BrC in MMH and MCS (Figure 4a and 4c respectively) given as the combinations of the different fuel types (beech, spruce, and birch for MMH, and pine, spruce, and beech for MCS). The average $BrC_{370\text{-}950}$ is given for all the individual fuel-appliance combinations in Figure 5."

2.3) What does Figure 7 signify? And what are the data points?

The data points in Figure 6 (previously Figure 7) present the contributions of BrC to the absorption at 470 nm, which is a commonly used single wavelength for BrC detection, versus the contribution of BrC over the total wavelength range of 370–950 nm. The values are averages for each fuel- and appliance-type combination. This is now clarified in the figure title (line 481). Given that a majority of studies on BrC absorption tend to concentrate on a specific wavelength, such as 370 nm or 470 nm, Figure 6 (previous Figure 7) concisely demonstrates the relation of BrC contribution if considering the total absorption instead of only the commonly used wavelength (470nm) to detect BrC. This observation highlights the importance of comprehensively assessing BrC absorption across a broader spectrum. This is discussed in the text from line 466 onwards.

The Figure 6 title is changed to "The average contributions of BrC to the absorption at 470 nm versus the total contribution over the wavelength range of 370nm–950 nm for all the fuel and appliance type combinations."

3) What is the physical significance of the dimensionless integrated absorption? If the goal is to quantify the overall contribution to absorption in the atmosphere, the integration has to be performed with respect to the wavelength-dependent intensity of solar radiation. Otherwise, the integration artificially skews the contribution to absorption to shorter wavelengths (because AAE > 0). Usually, experimental results provide wavelength-dependent optical properties, with the understanding that those can be used within radiative transfer calculations that account for the wavelength-dependent solar spectrum. If the authors wish to estimate the integrated contribution to radiative effect by BC and BrC, they could perform 'simple forcing efficiency' calculations (e.g. Chen, Y. & Bond, T. C. Light absorption by organic carbon from wood combustion. Atmos. Chem. Phys. 10, 1773–1787 (2010)).

The dimensionless integrated absorption (DIA) serves as a straightforward parameter aimed at estimating the contribution of BrC to light absorption. This concept revolves around the utilization of a singular parameter to characterize light absorption across the entire measured wavelength interval, as opposed to calculating it individually for each measured wavelength. The adoption of DIA streamlines the assessment of BrC's impact on light absorption, simplifying the analytical approach by condensing the information into a comprehensive parameter. A similar approach has been used previously (e.g., Massabó et al., 2015) for the quantification of the contribution of BrC to the total absorption within the visible wavelength range.

Unfortunately, we cannot reliably perform the proposed 'simple forcing efficiency' (SFE) calculations. The equation for the SFE includes both the total scattering coefficient and the backscattering coefficient. Usually, these are measured with an integrating nephelometer, which was not available for our experiments. Particle number size distributions could in principle be used for calculating both total and backscattering coefficients. However, our particle size distributions were measured using an ELPI, which gives the distributions as the function of aerodynamic diameter with a relatively low size resolution. These size distribution data are not good enough for calculating the SFE reliably enough. The need for the inclusion of scattering measurements is now also stated in line 480.

Instead of the SFE, we enhanced the estimation on the atmospheric importance of the BrC across the solar spectra by weighting the absorptions by BC and BrC by the standard solar spectra (ASTM G173-03). Due to the parabola-like shape of the standard spectra over the considered wavelength range, the resulting estimate is very close to the reported $BrC_{370-950}$ percentages.

This is now also noted in the manuscript, lines 475-479: “The relative contribution of BrC to the absorptivity in the visible wavelength range remains close to that of $BrC_{370-950}$ also if the absorptions illustrated in Fig. 4 are weighted by the standard solar irradiance spectra (ASTM G173-03; Figure S13). However, in order to comprehensively describe the brown carbon’s impact on radiative forcing, future studies should include a description of the BrC’s contribution to the absorption across the visible wavelength range as well as the light scattering by the aerosol.”

Reference:

Massabò, D., Caponi, L., Bernardoni, V., Bove, M. C., Brotto, P., Calzolai, G., Cassola, F., Chiari, M., Fedi, M. E., Fermo, P., Giannoni, M., Lucarelli, F., Nava, S., Piazzalunga, A., Valli, G., Vecchi, R., and Prati, P.: Multi-wavelength optical determination of black and brown carbon in atmospheric aerosols, Atmos. Environ., 108, 1–12, 10.1016/J.ATMOSENV.2015.02.058, 2015.

Minor comments:

1) Why not use absorption at 950 nm instead of 880 nm to estimate BC absorption?

The observed distinctions between these two wavelengths are relatively small. It is also noteworthy that within aethalometers, the wavelength at 880 nm is most frequently employed for the determination of BC, hence reflecting the common practice in assessing BC levels.

2) Line 395-396: The study defines BrC based on the assumption that absorption above the extrapolated BC absorption with AAE = 1 is attributed to BrC. Therefore, it is no surprise that BrC absorption is correlated with AAE. This statement is circular.

Indeed, this is true. The notion that this relation is expected is now added to the line 402.