

This manuscript shows an improved method for estimating the optical properties of BrC and their size distribution, which are one of the biggest challenges in estimating their climate effects. This study will be an interesting contribution to our understanding of BrC's climate effects. However, I still have some comments to help the author improve this manuscript. In general, I recommend that the editor consider this manuscript as a major revision.

#### **Major comments:**

1. Acronyms and abbreviations need to be well defined the first time they appear in the manuscript. Please check that.
2. I am not a big fan of calculating AAE by only using two wavelengths. It can increase the influence of systematic errors due to measurement. Moreover, usually, AAE calculated using short-wavelength will be larger since they are more dominating than longer wavelengths. In that case, I suggest using power law fitting since you have more absorption measurements at 7 wavelengths. Please consider that in future studies and add some discussion in the manuscript.
3. I did not see a particle scattering correction for aethalometer measurement, although you have particle scattering measurement. The correct factor you used might not work well for your sample. I want to see some discussion of this issue.
4. Correct me if I am wrong. What wavelengths do you use to calculate BrC absorption? I might miss that in your manuscript.
5. I did not see any comparison between modeled results and standards such as direct absorption measurements of BC (or any BC surrogate such as cab-o-jet, see "Characterization of light-absorbing aerosols from a laboratory combustion source with two different photoacoustic techniques") and coated. Without these experiments, it is not easy to validate your method. Have you considered performing these types of experiments?
6. BrC can also absorb light near-IR (see "Characterization of light-absorbing aerosols from a laboratory combustion source with two different photoacoustic techniques"; "Investigating the dependence of light absorption properties of combustion carbonaceous aerosols on combustion conditions"). Thus, I suggest you should also add some discussion about BrC absorption at near-IR

#### **Specific comments**

7. L141-143, "including multi-wavelength ... 2021b)." This is not clear to me. Could you provide more details about your scattering measurements? I want to know what type of instrument you used and the wavelength of that instrument.
8. L171-173, "The mass concentrations ... components." This part is also not clear to me. How do you do offline filter measurements and online inorganic aerosol component measurements? If you have any data, I suggest including them in the SI to support your argument.
9. L175-186, "Six-factors ... (2021b)." This part is a little bit confusing to me. Are these thresholds developed before, or are these just the average value of each class? Also, in Fig. S3, you showed more element ratios such as N:C, H:X, and OM:OC. I am also not clear on how did you get these values. I suggest putting these parameters in a table.
10. L186-188, "On the basis ... this study." With your current setup, it should be able to retrieve the density of BBOA and HOA by using SP-AMS. I am curious how close these values are to the literature values.
11. Saleh, 2020a and Saleh, 2020b are the same. Please correct that.
12. L231-235, "These parameters ... mass size distributions." This part is not clear to me. Did you use ranges of these parameters to calculate the AAE? Then what are the ranges you used? How did you decide on the ranges?

13. L254-257, "The average ... respectively." First, the first one and the last one are the same. Please check and correct that. Second, what is the uncertainty range of these ratios? Are they more significant than the uncertainty? These ratios are very close and increase with increasing of  $\lambda$ . This might be due to the increased weight of absorption at short wavelengths.
14. L296-298. "During the ... occurred." It is not clear to me how you chose spikes. In Fig. S7, the Shaded areas are very difficult to see. Please consider using a different color. Some BBOA spikes are not highlighted (e.g., beginning of Oct 19 and end of Oct 23). Is there any reason for that?
15. L314-316, "The average ... 2020)." How do you calculate  $\Delta$  for all parameters you show in this manuscript? This is not clear to me. I assume the  $\Delta$  you used in the manuscript is the difference between that variable before and after the BBOA spike. Then, my question is, what are the start and end times you used to get the average before and during the spike? This is not clear to me and can significantly affect your results.
16. L329-333, "The  $D_{gv}$  ... 2000)." Please check these two sentences carefully. In L329, you subscripted  $D_a$ . In L 330, you used  $D_{va}$  instead of  $D_a$ , which  $D_{va}$  should be more suitable. In L332, you used C as the factor. You need to use a different letter since you previously defined C as the Multiple-scattering correction factor.
17. L345-347, "BC/BBOA ratio ...  $R=0.84$ )." Is CO in L346 carbon monoxide? If yes, do you also have CO<sub>2</sub> measurements? Then you can use modified combustion efficiency (MCE) to estimate combustion efficiency. Also, please describe CO and CO<sub>2</sub> measurements in your Method section.
18. L466-468, "The average ... respectively." This is not clear to me. How did you do that? How do you get absorption for HOA, aBBOA, and MOOA?
19. L470, "suggesting significant changes of  $MAE_{BBOA}$ ." This is not clear to me. What are the significant changes you mentioned here, and why are there significant changes? Please explain that to me.
20. L474-475, The results in this section can also be supported by "Investigating the dependence of light-absorption properties of combustion carbonaceous aerosols on combustion conditions", "Brownness of organics in aerosols from biomass burning linked to their black carbon content", "Light-Absorbing organic carbon from prescribed and laboratory biomass burning and gasoline vehicle emissions", and "Parameterization of single-scattering albedo (SSA) and absorption Ångström exponent (AAE) with EC / OC for aerosol emissions from biomass burning". Please consider adding these references.
21. In SI, L73, How do you calculate the mass fraction of pure externally mixed BC?
22. In SI, L74,  $R_{NBC}$  is not well defined. Please add details like how you retrieve it.
23. In SI, L87, the density of BC should use  $1.8 \text{ g cm}^{-3}$  from "Bounding the role of black carbon in the climate system: A scientific assessment", unless you measured the BC density or have other references.
24. In SI, L116, what is Fig. Sx? I did not see it.