Response to reviewers' comments on "Chromophores and chemical compositions of brown carbon aerosol before and after photooxidation of combustion emissions" (EGUSPHERE-2026-559)

The authors kindly thank the reviews for the careful review of the manuscript, and the helpful comments and suggestions, which improve the manuscript a lot. All the comments are addressed below point by point, with our responses in blue, and the corresponding revisions to the manuscript in red. All updates of the original manuscript are marked in the revised version.

General Comment:

The authors of the MS titled "Chromophores and chemical compositions of brown carbon aerosol before and after photooxidation of combustion emissions" have made a commendable effort to address my comments and have strengthened the overall quality of the article. However, the small sample size and lack of replicates weaken my confidence in the study findings (considering that the variability of each measurement is largely undefined). I suggest that the authors consider how to address this issue; perhaps a "study limitations" discussion in the conclusion section? After the authors address this primary concern and the specific comments listed below, I would recommend the MS for publication in ACP.

Thank you for pointing this out. We agree that the lack of replicates leads to higher uncertainty of results. We added study limitations in the conclusion section. Though, we note that all experiments from different fuel sources are aggregated together. Note, that the study does not claim to find unique species present in each individual biomass burning source (and its aged counterpart), but rather treats the separate fuel sources together in Figures 4 and 5. Despite these different sources, there are specific common markers for the different chromophores present in all fuel types. To address the reviewers concerns we note in the conclusions:

"Please note that this study lacks in replicating the individual burning experiments. Since individual burning experiments can vary substantially, we have reported relatively high uncertainties for our results. The study of Zhang et all. (2023), employing a very similar methodology, shows repeated burning experiments with a moderate variability for most fuels (beech, spruce, pine, straw, and plastic), but except for cow dung. Therefore, we consider the results given in this work as valid while the data for cow dung may vary substantially with burning conditions. It still needs more studies to investigate the aging process of combustion emission before and after photooxidation for a specific fuel type to ascertain the robust character and inherent variability of the chromophores and their markers. Though, we note that a strength of this study is the comparable common markers for each chromophore type despite the different fuel types."

Specific Comments:

Lines 46-48: I am pleased to see that the authors have provided more context concerning the Feng et al. study; however, some of this information could be condensed. Specifically, I would simplify the statement: "global distributions of aerosol concentrations were simulated with a chemical transport model with a horizontal resolution of 2° × 2.5° and 26 vertical layers from the surface to the top of the atmosphere" and merge it with the following sentence.

We merged the two sentences as follows.

"Global simulations using a chemical transport model (2° × 2.5° horizontal resolution and 26 vertical layers) showed that brown carbon (BrC) absorption at ultraviolet-visible wavelengths contributes 7–19% of total atmospheric aerosol absorption (Feng et al., 2013)."

Line 53: I'm afraid my original question has not been answered. What do they authors mean by this statement? The use of "primarily" suggests that primary and secondary pathways are not the only sources of atmospheric BrC.

We have deleted "primarily".

"The sources of BrC consisted of secondary formation and primary emissions."

Lines 64-65: Needs rephrasing. Perhaps something like "molecules capable of absorbing and reflecting specific wavelengths of electromagnetic radiation".

Changed accordingly.

"molecules capable of absorbing and reflecting specific wavelengths of electromagnetic radiation".

Lines 76-77: Reword. There is an unnecessary "the" in this sentence. Also, "burning combustion" is somewhat redundant. Do the authors mean biomass burning, or combustion in general?

Changed accordingly:

"Many studies have investigated aging processes of BrC from combustion emissions."

Lines 110-115: This section has improved but still requires clarification. I would suggest something like: "In this study, we investigated … secondary BrC. Within a laboratory setting, a selection of fuels (list) was combusted to generate primary particles and gases; these emissions were oxidized within a reaction chamber to simulate the photooxidative aging process. The primary and secondary emissions produced through these experiments were analyzed ….etc."

We agree to modify the sentences as below:

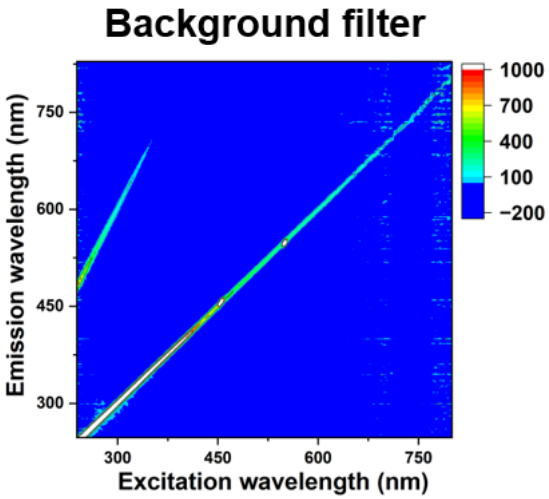
"In this study, we investigated the influence of fuel type and photochemical aging on the optical and chemical properties of primary and secondary BrC. The fuels (wood beech, straw, cow dung, and plastic) were combusted to generate primary particles and gases by using an open stainless-steel cylinder and a holding tank. These emissions were oxidized within an oxidative flow reactor to simulate the photooxidative aging process. The primary and secondary emissions produced through these experiments were analyzed by excitation emission spectroscopy and FIGAERO-CIMS."

Lines 134 – 135: The phrasing in this opening sentence is confusing. Are the authors stating that straw and beech burned in the open cylinder were representative of waste burning and forest fires, respectively? I would also suggest moving this sentence below the following sentence.

Thank you for pointing this out. We have deleted this sentence to avoid any misunderstanding.

Lines 154-155: Were these blanks used to correct the sample data? I'm assuming the blanks were low?

We used the blank samples to correct the samples. For example, the fluorescence intensity of blank filter was relatively low, as shown in the below figure.



The excitation emission spectrum of methanol soluble organic carbon from a blank filter sample.

Lines 157-159: "However, due to sufficient particle mass loading on the filters, the potential contribution of this effect can be considered as minor". This assumption makes me a little nervous. The (S)VOCs co-emitted with BB aerosols are similarly high in concentration – and could impart a high loading to the gas-adsorbing filter as well.

Sorry for the unclear explanation. FIGAERO-CIMS technique typically uses Teflon filters to reduce interferences from the gas phase. The quartz fiber filters are widely used for offline sampling of organic aerosol due to their high melting point and insolubility in water and typical organic solvents. In addition, it is more easier to punch out parts of quartz filters comparing with Teflon filters. Sardena et al. (2026) also used quartz filter to collect particles for analyzing the singlet oxygen and Teflon filters for FIGAERO-CIMS analysis during the same campaign.

"The partitioning of the VOC into the aerosol particles loaded on the filters may have a larger impact than the adsorption on the filter surface and this effect is expected to be similar for both filter types."

There are other experimental conditions to consider regarding the SVOC from the gas-phase being adsorbed to the filter. The mass concentrations present in the chamber were 1-5 mg m^{-3} meaning that most of the SVOCs will already be within the aerosol phase. In addition, to mitigate gaseous uptake onto the filters, the sampling time for fresh particles was only 5 minutes. The sampling flow was 8 lpm. Therefore, the trace gases only had a short interaction time with the filters. The short collection times will mitigate the high loading effect from gas phase, especially S-VOCs. Blank filters were also collected during the campaign. We used the blank samples to correct the sample filter. To avoid any misunderstanding, we added the following sentence:

"However, the sampling time for fresh particles was 5 minutes and the sampling flow for fresh and aged particles was 8 lpm, which will help mitigate uptake of gases onto the filters. The overall adsorption of SVOC on the filter was small compared to the typical aerosol particle mass loading."

Lines 211-212: The authors mention high methanol extraction efficiency but provide no further elaboration. Was this tested in the lab? A brief quantification of extraction efficiency would be helpful.

Thank you for pointing this out. Sorry we did not directly measure methanol extraction efficiency. However, according to previous studies, the methanol extraction efficiency for organic carbon was around 85% (Cheng et al., 2016). To avoid this misunderstanding, we added sentence as below:

"The methanol extraction efficiency for organic carbon was around 85% (Cheng et al., 2016)."

Line 231: If the authors measured the UV-Vis absorbance of the filter extracts, why was this information not included in the MS? These data can be very useful when characterising brown carbon.

Thank you for pointing this out. We measured the light absorption of the filter extraction solution. However, we did not measure the total organic carbon on the filter

and cannot provide mass absorption efficiencies. Therefore, the light absorption of filter extraction solution was show as Figure S5. We added ass the following sentence to the manuscript pointing to this:

"After photooxidation, the light absorption and Absorption Ångström Exponent were also significantly deceased (Figure S5)."

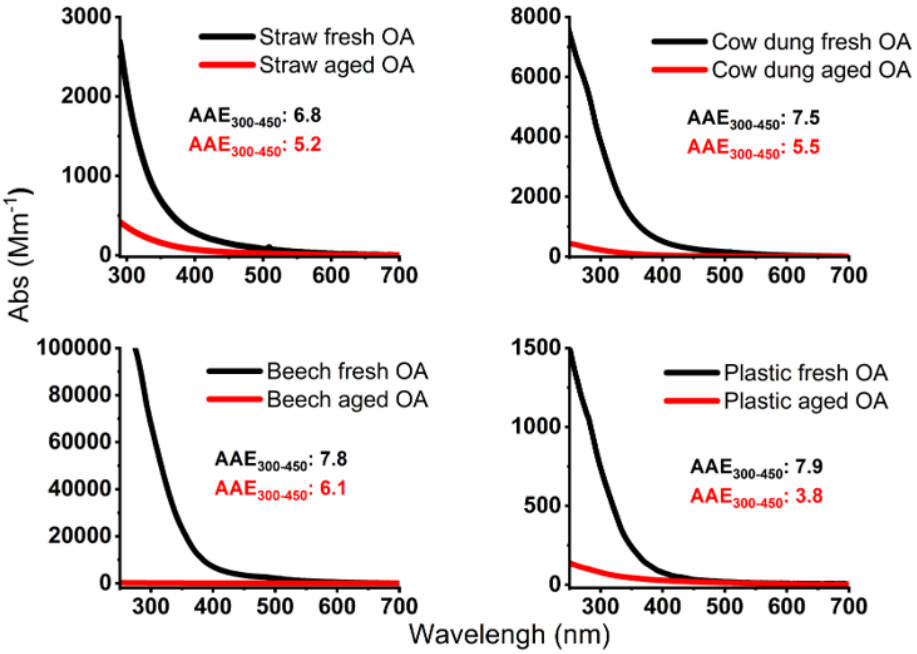


Figure S5. The light absorption of methanol soluble organic carbon from all 8 samples.

Lines 264-266: The mass fraction of C6H10O5 was ~35% in the plastic burn sample - why is this not mentioned/explored here?

Thank you for pointing this out. Sorry for not exploring the mass fraction of $C_6H_{10}O_5$ with ~35%. Even though we detected $C_6H_{10}O_5$ in the plastic samples, those compounds could be isomers compared to the same formulas in wood fuels. The FIGAERO-CIMS only can provide sum formulas but not the molecular structure of isomers. This does not mean that plastic burning can emit levoglucosan. Also, this could be a function of the FIGAERO-CIMS not being sensitive to the majority of species emitted by plastic combustion (namely non-oxygenated organics)(Shen et al., 2025; Zhang et al., 2023). To avoid this misunderstanding, we added the following sentences:

"The plastic burning also had high $C_6H_{10}O_5$ signal with ~34% as detected by FIGAERO-CIMS. However, this could be a function of the FIGAERO-CIMS not being sensitive to non-oxygenated components emitted resulting in an amplified signal for species that are present. Additionally, this compound is not necessarily levoglucosan but could be isomers with the same sum formulas."

Lines 267-268: Can you confidently state that these mass fractions are substantially different? These values are fairly close - and we do not know how much these measurements vary, due to the lack of replicate samples.

We agree that a lack of replicate samples limits our ability to state if the mass fractions are significantly different. To avoid any misunderstanding, we modified the sentences as given below:

"In natural burning, the mass fraction of sugar derivatives ($C_5H_8O_4$, $C_8H_{12}O_6$, $C_6H_{12}O_5$) was slightly higher (6.7 ± 3.5%) than that in plastic burning (4.9%). Therefore, natural burning could produce more sugar derivatives than plastic burning on average. However, these compounds are not a definitive tracer for distinguishing them due to uncertainties from combustion emissions."

Line 335: Since there are only 8 samples in total, I would suggest presenting the individual EEM scans in the supporting information section. Direct visual inspection may reveal subtle differences between the fuel types – differences that may not be observable in a small sample size PARAFAC model.

We agree to add 8 individual EEM scans of each sample, as shown in Figure S4.

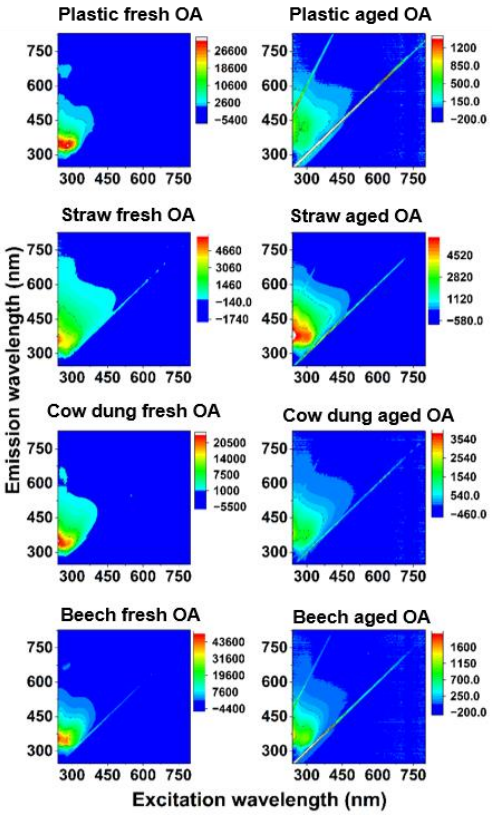


Figure S4. The excitation emission spectrum of methanol soluble organic carbon from all 8 samples.

We also added the following sentence to the main text:

"The excitation emission spectrum of each sample is shown in Figure S4."

Reference

Cheng, Y., He, K.-b., Du, Z.-y., Engling, G., Liu, J.-m., Ma, Y.-l., Zheng, M., and Weber, R. J.: The characteristics of brown carbon aerosol during winter in Beijing, Atmospheric Environment, 127, 355–364, https://doi.org/10.1016/j.atmosenv.2015.12.035, 2016.

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Zhang, J., Li, K., Wang, T., Gammelsæter, E., Cheung, R. K. Y., Surdu, M., Bogler, S., Bhattu, D., Wang, D. S., Cui, T., Qi, L., Lamkaddam, H., El Haddad, I., Slowik, J. G., Prevot, A. S. H., and Bell, D. M.: Bulk and molecular-level composition of primary organic aerosol from wood, straw, cow dung, and plastic burning, Atmos. Chem. Phys., 23, 14561–14576, https://doi.org/10.5194/acp-23-14561-2023, 2023.