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.

Reviewer #1

The manuscript titled “Chromophores and chemical compositions of brown carbon aerosol before and after photooxidation of combustion emissions” investigates the influence of fuel-source and photooxidative aging over the optical and chemical properties of combustion-based brown carbon (BrC). The authors generated fresh and aged brown carbon from a variety of fuels within an oxidative flow reactor and analyzed these aerosols using fluorescence spectroscopy and mass spectrometry. These experiments demonstrated that photooxidation impacted the chemical and fluorescent structures of brown carbon aerosols; where compared to primary aerosols, aged brown carbon displayed a higher fraction of oxygen and nitrogen containing organic matter. The observations from this study may be of interest to the broader atmospheric science community; however, certain elements of the paper (including the written structure, methodology, and discussion) require considerable improvement. I will expand on these issues in the “specific comments” section. As a final note, I encourage the authors to more effectively communicate the scientific significance and implications of their study.

General comments

1. Lines 20-22: I suggest mentioning that the key analytical methods used in the study (fluorescence, MS). Without this context, the remaining abstract will confuse some readers.

We agree to add key analytical methods in the abstract. The contents were shown as the following:

“To address this gap, we studied emissions of burning beech wood, straw, plastics, and cow dung in an oxidative flow reactor allowing to conduct photooxidation aging of fresh emissions by using mass spectrometry and excitation–emission spectroscopy with a parallel factor analysis.”

2. Lines 38-42: The content in this first paragraph is useful; however, I would suggest some restructuring to include a brief statement clearly defining brown carbon. Ie., “diverse range of aerosols consisting of organic compounds capable of absorbing UV-Vis light etc., etc.”

We agree to add a statement that defines the brown carbon in the first paragraph.

"Organic aerosols have a profound impact on air quality, human health and climate. Typically, organic aerosol (OA) is considered to be non-light-absorbing, hence only contributing to scattering of solar radiation that leads to atmospheric cooling (Shrivastava et al., 2017). However, some OA compounds, known as Brown Carbon (BrC), can absorb solar radiation in the near-ultraviolet and visible spectral range contributing to atmospheric warming (Moise et al., 2015; Laskin et al., 2015)."

3. Line 39: "The global simulation showed that…" This phasing is confusing. You must first introduce/establish the simulation, as this will contextualize the following discussion.

We agree to introduce the global simulation model.

"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 (Feng et al., 2013)."

4. Line 43: The phrasing in this sentence is somewhat confusing; I would suggest rewording to state:"The sources of BrC primarily consisted of…" Perhaps I am misinterpreting the phrasing, but I'm not sure what the authors mean by this statement – considering that primary emission and secondary formation are effectively the only two pathways for contaminants to enter the atmosphere.

Changed accordingly.

5. Lines 44-45: Correct to "… in the presence of NOx…".

Changed accordingly.

6. Line 47: I would suggest rewording: "Field-based measurements have shown that…"

Changed accordingly.

7. Lines 47-51: This is important context for the study; however, it would be helpful to provide some additional data/stats to help emphasize the relative size of biomass burning emissions? i.e. what percent of BrC was linked to burning in the papers cited?

We agree to describe the percentage of brown carbon absorption from biomass burning.

"Field-based measurements have shown that a majority of BrC aerosol mass was associated with biomass burning dominating BrC absorption in different areas, e.g, more than 90% in rural southeastern United States (Washenfelder et al., 2015), ~ 60% in Arctic region (Yue et al., 2022), 29%–35% in Himalayas and Tibetan Plateau (Zhu et al., 2024), 50−80% in Israel (Lin et al., 2017), and ~60% in central Europe (Moschos et al., 2018)."

8. 52: Briefly define the term, "chromophore".

We defined chromophore as below:

"molecules responsible for absorbing light and reflecting specific electromagnetic radiation which leading to a color."

9. Lines 54-62: I suggest starting this section with a brief sentence establishing to the concept of BrC aging its influence over aerosol fate. I.e., what is BrC aging and why do we care? Then expand onthe literature.

We have added more contents to discribe the concept of BrC aging and aerosol fate. The following sentences were added:

"BrC aging refers to the chemical and photochemical transformation of light-absorbing organic aerosols, a process that is critical because it governs their optical properties, reactivity, and overall atmospheric fate (Laskin et al., 2025)."

10. Line 63: Reword: "…spectroscopy has been widely used…"

Changed accordingly.

11. Line 66: Considering that this term is only used twice in the MS, I don't think the acronym "PLOM" is needed here.

Changed accordingly.

12. Lines 71-72: This sentence is somewhat confusing – please rephrase.

Changed accordingly.

"However, the chemcial composition of BrC chromophores are still not well known, especailly for particles from biomass burning."

13. Lines 83-84: It is established here that the study measured BrC emissions from a range of fuel types. This is a good element of the study – but I suggest that the authors establish earlier in the introduction why we are interested in different fuel types. Cite the literature that highlights the influence of burning material over BrC composition.

We agree to add introduction why we are interested in different fuel types.

"In India, more than half of households use inefficient stoves for cooking, burning solid fuels such as cow dung, firewood, crop residues, and charcoal (Census of India, 2011). This contributes to poor household air quality, chronic and acute respiratory diseases, and even premature death (Smith et al., 2014). The burning of plastic has been estimated to contribute 13.4% of $PM_{2.5}$ in India, and 6.8% in China (Haque et al., 2019). Previous studies have shown that the BrC chromophores and compositions were different from different biofuels and burning conditions (Huang et al., 2022; Song et al., 2022). Therefore, it is essential to investigate the chromophores and compositions of aerosol particles emitted from burning different fuels."

14. Lines 81-88: I recommend that the authors restructure this final paragraph. The first sentence should clearly outline the study objectives: "In this study, we investigated the influence of fuel type and photochemical aging over the optical and chemical properties

of primary and secondary BrC."… or something along that line. Once the former is established, you can briefly outline how the study design will achieve these objectives.

We changed the final paragraph according to the suggestions.

"In this study, we investigated the influence of fuel type and photochemical aging on the optical and chemical properties of primary and secondary BrC. Burning different fuels generated primary particles and gases which were subsequently oxidized forming secondary organic aerosol mass."

15. Lines 92-99: The authors should provide some additional details justifying the selection of these specific burning materials. Some statistics / data describing the prevalence of these fuel types would help.

The beech, straw, and cow dung are representative of residential wood burning. The plastic is representative for waste burning.

"We categorized four burning types for this experiment: beech, cow dung, straw, and plastic. We selected these four solid fuels and conducted emissions tests to simulate certain types of burning in the atmosphere. The beech, straw, and cow dung are representative of residential wood burning, which is consistent with the materials used in previous articles (Zhang et al., 2023; Li et al., 2024; Wang et al., 2025). The polyethylene plastic materials are representative of plastic burning in India and China (Haque et al., 2019)."

16. Lines 96-99: These sentences are somewhat confusing. Do the authors mean to say, "Straw and beech materials were burned in an open stainless-steel cylinder…". Moreover, this is the first time the steel-cylinder has been mentioned in the MS – without initial context these statements will confuse the reader, as it is not clear why the straw and beech were burned in the cylinder, and the dung/plastic on top.

The stainless steel cylinder is 65 cm in diameter and 35 cm in height as shown in Figure S 1a. the straw and beech burning in cylinder was presented as agricultural waste combustion and forest fires. The cow dung and plasctic can be burned effecency on the top of stainless-steel cylinder. To avoid confuse, we added description about the cylinder and why the straw and beech were burned in the cylinder and the cow dung/plastic on the top of tylinder.

"The stainless-steel cylinder is 62 cm in diameter and 35 cm in height. This cylinder has been used to investigate beech wood burning emissions in previous studies (Sardena et al., 2026; Bogler et al., 2025)."

"In addition, the cow dung and plastic burning on the top of stainless-steel cylinder was more efficiency than in the stainless-steel cylinder."

17. Line 108: Correct: "The particles in the holding tank…". In addition, merge this sentence with the following one.

Changed accordingly.

“The particles in the holding tank were collected by quartz and Teflon filters and samples can be considered as primary organic aerosol”

18. Lines 108-109: I am curious regarding the QA/QC methods applied in these experiments. For instance, were blank and replicate samples collected for analysis? This information should be available in the MS.

During the campaign, the blank samples were collected.

“Blank quartz and Teflon filters were collected during the campaign and analyzed in the same way as the other samples.”

19. Lines 125-133: It is well established that sample fluorescence can be highly pH-dependant. Did the authors account for this factor? Was sample pH measured? Was sample pH adjustment considered?

someone else tested PH for the exact same samples after extraction and PH was around 7

Previous studies have shown that the pH is a crucial factor to affect the light absorption and fluorescence efficiencies of water-soluble organic compounds (Qin et al., 2022). However, since the organic compounds dissolved by methanol in this studies, we did not measure the sample pH. In addition, Sardena et al. (2026) found the pH of same samples after extraction was around 7.

20. Line 134: Could the authors name the software used for EEM / PARAFAC processing?

We added more content to explain the software used for PARAFAC proceessing.

“We used the StaRdom package for RStudio version 2026.04.0 of PARAFAC model (Pucher et al., 2019), which was downloaded from https://cran.r-project.org/web//packages/staRdom/vignettes/PARAFAC_analysis_of_EEM.html (last access: 19 February 2026).”

21. Lines 142-143: Split-half validation is a useful tool for PARAFAC model selection. However, I have noticed that Fig. S2 mentions the Core consistency and eemqual metrics. These are also valuable tools for model validation; however, I do not see these methods described in the MS or SI files. Could the authors provide brief descriptions of these techniques?

We agree to add contents about the Core consistency and eemqual metrics in the supplement.

The additional information was added into the supplement content:

“A new diagnostic called the core consistency diagnostic (CORCONDIA) is suggested for determining the proper number of components for a PARAFAC models. Models specified with an appropriate number of components should have high core consistencies (near 100%), whereas low core consistencies indicate that too many components were specified. The core consistency diagnostic should protect against over-fitting but rigid adherence to guidelines can lead to under-fitted models. Core consistency in this study is 0.954. It indicates that the 3 components are appropriate numbers for the PARAFAC model. The EEMqual is a quality parameter integrating the model fit. It was calculated with the following equation:

$$\text{EEMqual} = \text{Fit} * \text{Core consistency} * \text{Split half analysis}$$

Thus, when EEMqual is close to one, all three measures are high and the model analysis successful."

We also modified the line 144 as the following:

"The corresponding model parameters and detailed information of the Split-half validation are shown in Table S2 and following contents."

22. Lines 108-109 & 115: I am somewhat concerned by these methods. Particles generated during these experiments were collected on quartz and Teflon filters - these filter samples were thenallocated for EEM and FIGAERO-CIMS analysis, respectively. The parallel EEM and chemical datasets underwent statistical comparison (e.g. Spearman correlation) under the general assumption that the particulates collected on the quartz and Teflon filter were identical. Quartz and Teflon filters have fundamentally different structures and particle retention capacity – not to mention that quartz filters can retain volatile organics that can lead to an overestimation of aerosol brown carbon. As such, I am worried that the quartz and Teflon filter samples are not as chemically similar as the authors might assume, and this dissimilarity could weaken the comparison of EEM and FIGAERO-CIMS.

Thank you for pointing this out. We agree that the different filter materials have different absorption characteristics for VOC. However, the retention capacity of the filter types used are sufficiently high for the particle sizes of this study to ensure good comparability. The potential adsorption of e.g. semi-volatile or volatile compounds in typically higher for quartz fibre filters. However, the typical particle mass loading were so high that partial adsorption of VOC on the filter material should be negligeble. However, to avoid misunderstanding, we have added the following sentences to the method section 2.2:

"Please note that quartz filters and Teflon filters have different adsorption behaviour of gases which could lead to different filter loading and hence differences between EEM and mass spectrical analysis. However, due to sufficient particle mass loading on the filters, the potential contribution of this effect can be considered as minor."

23. Lines 175-176: I'm curious why a Spearman r of 0.643 was selected as the cut-off point? This value seems somewhat arbitrary (and low); why not let the p-value of the Spearman test (at higher significance levels) inform the pairing of FIGAERO-CIMS

and PARAFAC components? What methods have previous studies used? I would suggest citing them here.

The Spearman r of 0.643 was calculated by the following equation:

$$t = \frac{r\sqrt{n-2}}{\sqrt{1-r^2}}$$

Where t = 1.943 and n =8, a rs value of 0.643 was calculated as significant at the 95% confidence limit (one tailed test for Spearman) (Stubbins et al., 2014; Tang et al., 2024). We added more content about the Spearman correlation in the supplement section 2.

We also added more discription about the Spearman's analysis in the main manuscript:

"The molecules correlated to PARAFAC component intensities with Spearman ≥ 0.643 (one tailed test, see the detailed information in the supplement section 2) were assigned to each PARAFAC component (Table S5, S6, and S7) (Tang et al., 2024; Stubbins et al., 2014)"

In the supplemental section 2

"For our samples size of 8, the 8 raw scores X_i (i.e. PARAFAC component intensities), Y_i (i.e. FIGAERO-CIMS peak intensities) were converted to ranks X_i, Y_i, and r was computed (Tang et al., 2024; Stubbins et al., 2014). To assess the significance of Spearman's correlation coefficients, one tailed test was performed. For a sample size of 8, a threshold for Spearman's coefficient (r_s) was calculated to be significant at the $\alpha = 0.05$ confidence level, using the following equation:

$$t = \frac{r\sqrt{n-2}}{\sqrt{1-r^2}}$$

Where t = 1.943 and n =8, A r_s value of 0.643 was calculated as significant at the 95% confidence limit (one tailed test value for Spearman) (Stubbins et al., 2014)."

24. Line 182: Section 3 first covers the chemical characteristics of sampled aerosols, followed by discussion regarding EEM-PARAFAC. However, section 2 discusses the corresponding methods (CIMS, EEM-PARAFAC) in the reverse order. This is a somewhat pedantic critique, but I would suggest reordering either the methods or results/discussion sections to improve the written flow of the MS.

Thank you for pointing this out. We agree and reversed the order of CIMS and PARAFAC.

25. Line 214: remove "also"

Changed accordingly.

26. Line 230: Correct phrasing. Perhaps "A PARAFAC model was…".

Changed accordingly.

27. Lines 232-233: The phrasing in this sentence is somewhat confusing. Do the values "239, 300, and 372 nm represent three separate peaks associated with the C1 fluorophore? Are they representative of the excitation or emission wavelengths? Please clarify.

The 239 nm and 300 nm were representative as the peak wavelengths of excitation. The 372 nm was representative as the peak wavelength of emission.

"The peaks of excitation/emission (Ex/Em) values for C1 were at 239, 300 nm for excitation and at 372 nm for emission."

28. Line 233-234: Please define "AMS". The phrasing here is somewhat confusing – make sure that it is clear you are comparing C1 to fluorophores from an external study.

Changed accordingly.

29. Line 236: I'm apprehensive about the decision to compare the methanol-soluble PARAFAC components to water-soluble fluorophores from other studies. I would suggest comparing against other methanol-soluble fluorophores.

Thank you for pointing this out. Tang et al. (2020) used water to extract the filters and then freeze-dried and extracted the same filter with methanol. This way brown carbon can be separated as water-soluble BrC and methanol-soluble (water-insoluble) BrC. Therefore, the fluorophores characteristics show clear differences. However, during our measurement, we directly used methanol to extract the filters. This way we had methanol soluble but also water-soluble components in our solutions. Therefore, we can compare our methanol-soluble BrC also with other water-soluble BrC, because the solution contains water-soluble and water-insoluble organic carbon. In addition, Chen et al. (2020) also determined PARAFAC components of methanol-soluble organic carbon according to fluorophores in water-soluble organic carbon. However, to avoid misunderstandings we have added the following sentences to the method section 2.3:

"Please note that the MSOC contains methanol-soluble but also water-soluble compounds since also water-soluble compounds are partially soluble in methanol. We did this direct methanol extraction in order to dissolve a maximum number of compounds facilitating a good comparison with the mass spectrometric analysis in which there is no differentiation between different solubilities. However, this has to be kept in mind when comparing our results with studies that separated the water and methanol soluble fractions."

30. Lines 235-236, 238: why not state the specific wavelength here?

We added more contents about C2 chrmomophore.

"The peak of excitation/emission (Ex/Em) values for C2 was at 245 nm for excitation and at 372 nm for emissions."

31. Line 302: The header for Table 1 would benefit from some additional detail. For instance, what does “average properties” refer to?

Thank you for pointing this out. We added more information to explain the header of Table 1.

“The average properties of molecules were associated to three characteristic chromophores. The number of average molecular mass, averages molecular properties, and structural grouping of molecular formulas as determined by FIGAERO-CIMS in 8 samples. These were founded to correlate with each of the 3 fluorescence PARAFAC components identified (LO-HULIS, HO-HULIS, and PLS); and those which did not correlate with any the PARAFAC component (Not included)”

32. Line 330: The scale of the O/C axis is different between the three plots shown in Figure 4. I would suggest making note of this discrepancy in the figure header (or unify the axis scaling).

Thank you for pointing this out. We unified the axis scaling.

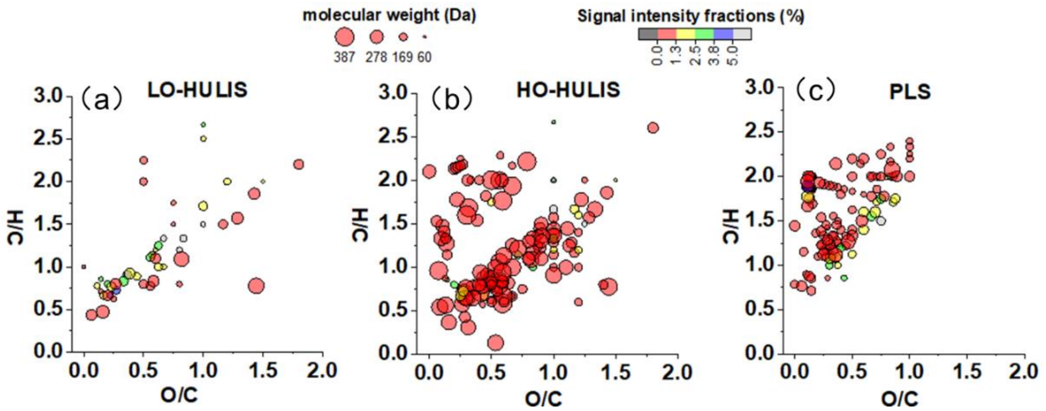

Figure 4. Van Krevelen diagrams of FIGAERO-CIMS identified compounds assigned to PARAFAC components (LO-HULIS, HO-HULIS, and PLS).

33. Lines 333-334: The authors discuss the possible influence of NOx over PLS oxidation. Does this imply that NOx was included in the flow reactor experiments? Was NOx injected into the reactor? At what concentration? Perhaps I am missing something, but this was not disclosed in the methods section. Please elaborate.

Thank you for pointing this out. The NOx was generated during burning of 4 fuels (beech, cow dung, straw, and plastic) (Jin et al., 2021). The concentrations of NOx were variable but around a few hundred ppm. To clarify, we added a description of NOx concentrations as the following:

In the main manuscript section 2.1:

“Please note that the NOx was generated during the burning. There were no instruments to measure the NOx concentration. However, according to previous studies, the concentrations of NOx were around a few hundred ppm (Winter et al., 1999;

Courtemanche and Levendis, 1998). Furthermore, the volatile organic compounds were generated from the combustion (Li et al., 2024)."

In the main manuscript section 3.5:

"Please note that the NOx was generated during the burning. According to previous studies, the concentrations of NOx were around a few hundred ppm (Winter et al., 1999; Courtemanche and Levendis, 1998)."

34. Line 366: Correct to "..component, and a phenolic-like substance…"

Changed accordingly.

35. Lines 355-383: This section reads more like a summary than a discussion regarding atmospheric implications. In particular, the first two paragraphs repeat key findings reported earlier in the MS.I strongly suggest the authors revisit this section to consider what their observations tell us regarding the atmospheric chemistry, fate, and source-dependence of combustion based BrC.

We changed the section of atmopheric implication as conclusion. In addition, we added some content as the following:

"Assuming typical ambient OH concentrations (~1.5×10^6 molec cm^{-3}) and the laboratory photooxidation timescale (5.4×10^{11} molecules cm^{-3} s in experiment, equivalent to 4 days of atmospheric aging), the implied atmospheric lifetime of PLS is on the order of a few days. PLS can serve as a source-specific tracer only for very fresh combustion plumes."

"This points to photooxidation as a net formation pathway for HO-HULIS formation."

36. Lines 374-376: "The oxidation of volatile organic compounds and degradation of large molecules were smaller organic compounds with higher oxidation state and the high fractions of nitrogen containing chromophore." I'm not sure I understand this sentence. Please reword.

I reformulated this sentence.

"The oxidation of volatile organic compounds and degradation of large molecules generated smaller organic compounds with higher oxidation state and relatively high fractions of nitrogen containing molecules."

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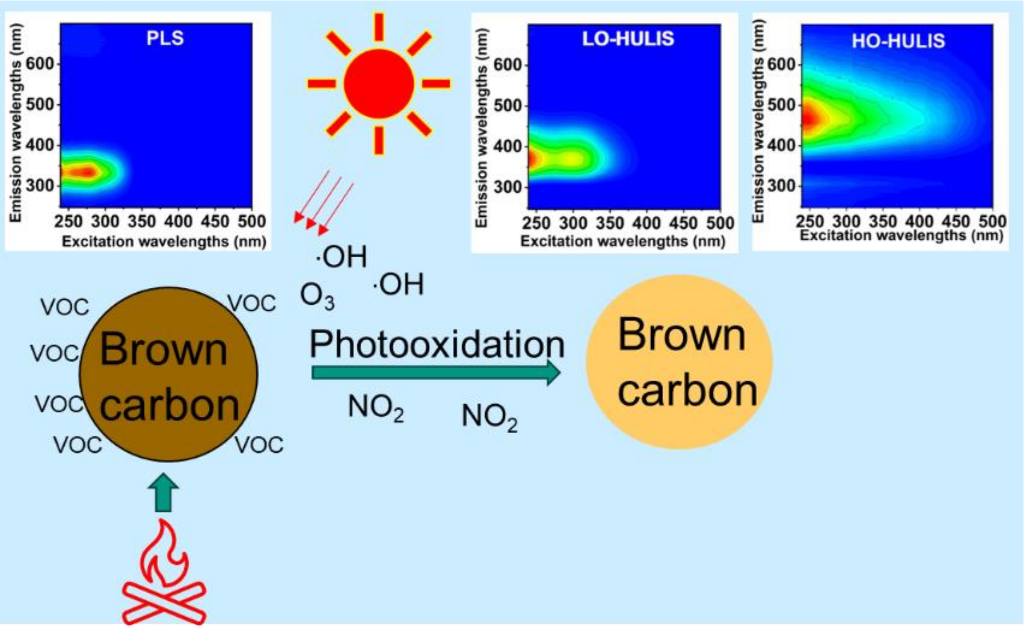
Reviewer #2

In this manuscript, the authors report on the fluorescence and composition of primary and secondary organic aerosol from the combustion emissions of a range of fuels, including wood, grass, dung, and plastic. The selection of fuels is broad but would benefit from more clearly written rationale. In the experiments, the authors measured excitation-emission matrix spectra and chemical ionization mass spectra for filter-collected samples upstream and downstream of an oxidative flow reactor. The combination of EES and CIMS is strong, leading to new insights regarding the fractions of species in these organic aerosols before and after photochemical aging. The structure of the manuscript and the presentation of the results in figures are generally clear. However, there are many typos throughout the manuscript - I do not list them out below, but those found on the following line numbers are just a selection: 11, 20, 32, 105, 213, 295, 315, 337, and 348. In addition to these corrections, a thorough language revision is required - those found on the following line numbers are examples: 18, 21, 38, 43, 48, and 84. With revisions, the manuscript should be suitable for publication in ACP.

General comments

1. 15: The graphical abstract should be revised since many of the elements (like tick labels) are too small to be legible at the size they will appear after publication. If EES spectra are shown, I recommend including axes labels.

We agree and increased the size and axes labels of EEM in the graphical abstract.



We also changed Figure 2 in main manuscript as the following:

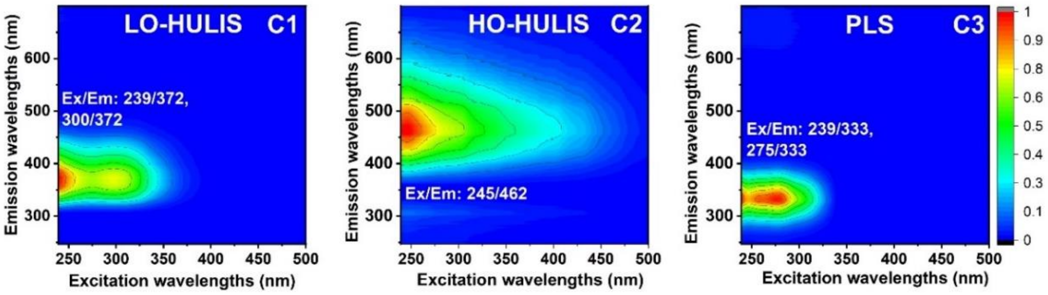


Figure 2. The three chromophores were identified by the PARAFAC model analysis of the excitation–emission spectra from all filter extracts collected from combustion of beech wood, straw, cow dung, and plastic.

2. 54: I suggest revising this paragraph on past studies of aging. I think it is more accurate to say many rather than several studies have investigated aging. I understand there is not room to explore all aspects of these studies here, but the selection of features is too limited even for this space. For example, absorption enhancement is a common feature of OH radical oxidation, so it may justify mentioning briefly.

We agree to modify this paragraph. In addition, we have added more content to describe the concept of BrC aging and aerosol fate as below:

"BrC aging refers to the chemical and photochemical transformation of light-absorbing organic aerosols, a process that is critical because it governs their optical properties, reactivity, and overall atmospheric fates (Laskin et al., 2025)."

"Sumlin et al. (2017) found that OH radical oxidation can initially enhance brown carbon absorption, followed by bleaching upon further oxidation."

3. 84: Based on the preceding material in the Introduction, it is surprising to see plastic included in this list of fuels. Plastic combustion should be introduced and rationalized earlier - why is it studied here?

We agree to add introduction why we are interested in different fuel types. The plasitc was used to simulate the plastic waste burning in China and India (Haque et al., 2019).

"In India, more than half of households use inefficient stoves for cooking, burning solid fuels such as cow dung, firewood, crop residues, and charcoal (Census of India, 2011). This contributes to poor household air quality, chronic and acute respiratory diseases, and even premature death (Smith et al., 2014). The burning of plastic has been estimated to contribute 13.4% of $PM_{2.5}$ in India, and 6.8% in China (Haque et al., 2019). Previous studies have shown that the BrC chromophores and compositions were different from different biofuels and burning conditions (Huang et al., 2022; Song et al., 2022). Therefore, it is essential to investigate the chromophores and compositions of aerosol particles emitted from burning different fuels."

4. 92: Does this total of four include all replicates or just all conditions? If there are no replicate experiments, some acknowledgement of the inherent variability of biomass burning experiments – and consequent limitations – should be provided here.

We did not do replicate experiments. We agree to point out the limitations.

"Please note that each burning experiment had no replicate experiment. This could lead to uncertainties in the results since the biomass burning experiments involve inherent variability."

5. 96: What is the rationale for selecting polyethylene here?

The plastic burning has been estimated to contribute 13.4% of $PM_{2.5}$ in India, and 6.8% in China (Haque et al., 2019). Polyethylene plastic materials were representative of residential plastic burning in India and China (Haque et al., 2019).

6. 105: Was the holding tank also washed or heated between experiments with different fuels? I would be concerned about cross-contamination, e.g., SVOCs from one fuel being released later during an experiment with another. Were control experiments performed to rule out this potential contamination? For example, if no SOA were observed when clean air was passed to it through the holding tank, then the flushing may be adequate.

We agree that the potential for cross-contamination from residual SVOCs in the holding tank is indeed an important consideration when conducting sequential experiments with different fuels. However, before each experiment, the holding tank was flushed overnight with clean air, ensuring the background concentrations were less than 10 particles cm^{-3}. In addition, during the measurement, the holding tank contained high concentration of particles and VOC. The residual SVOCs did not significantly affect subsequent measurements (Li et al., 2024). We added the following text to explain this issue.

"To avoid cross-contamination from residual SVOCs in the holding tank, it was flushed overnight with clean air before each experiment, ensuring the background particle concentrations were less than 10 particles cm^{-3}. This way residual SVOCs did not significantly affect subsequent measurements (Li et al., 2024)."

7. 114: Do you have a rough estimate of the ratio of ozone to OH?

According to previous experiments, the OH exposure is set to 5.4×10^{11} molecules cm^{-3} s during the experiment. We added this information as the following:

"The OH exposure was set to 5.4×10^{11} molecules cm^{-3} s during the experiment (Li et al., 2024)."

8. 119: I wonder if the POA and SOA labels are the best way to describe these materials, considering the POA was not filtered out before the OFR. Perhaps fresh and aged would

be more accurate. In the future, strictly SOA could be accessed by filtering the POA before the OFR.

We agreed and changed POA and SOA label into fresh OA and aged OA though the manuscript.

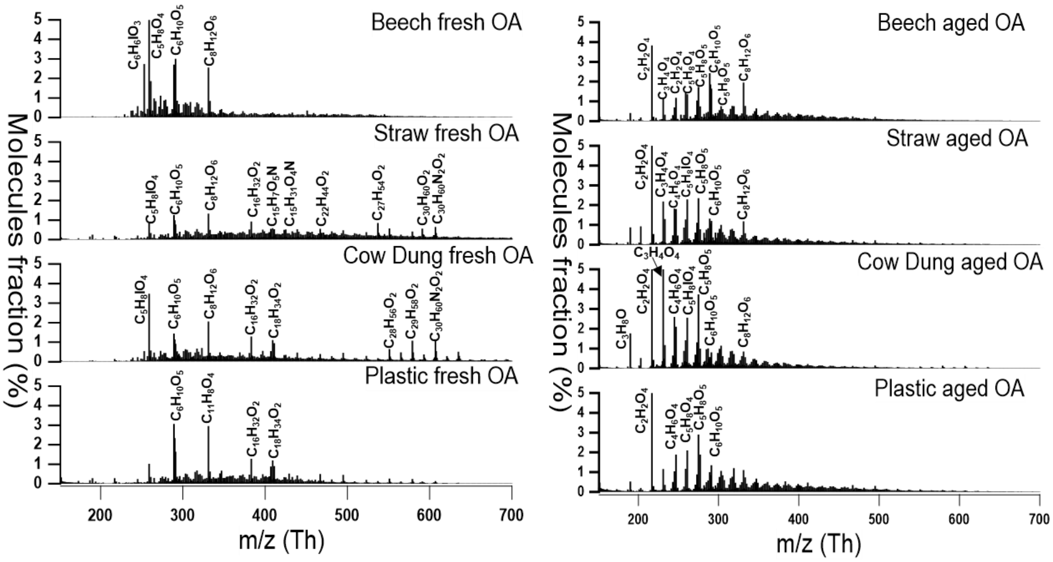


Figure 1. Mass spectra with iodide of particles from fresh OA and aged OA combustion emissions from FIGAERO-CIMS measurements. Particles from combustion/oxidation of beech wood; straw; plastics; cow dung. Please note that the signal intensity of levoglucosan ($C_6H_{10}O_5$) was multiplied by 0.1. Please note that the samples contain both primary emissions and secondary aerosol after photooxidation, we named these samples as aged organic aerosol.

9. 122: I would prefer to write EEM here rather than Aqualog, to focus on the technique rather than the model of instrument.

Changed accordingly.

10. 125: What was the rationale for using the methanol-soluble fraction (e.g., the high extraction efficiency in terms of mass)?

Thank you for pointing this out. During our measurement, we directly used methanol to extract the filters. Methanol extraction was indeed efficient. To classify the methanol extraction, we added content as follows:

"The methanol extraction had high extraction efficiency for organic compounds on the filters."

11. 129: Why was the organic mass loading not clear? Were filter masses not measured/measurable? More broadly, I think it is worthwhile determining mass

concentration for fluorescence, too (not just for absorbance), since it can influence quenching, etc.

We did not measure the organic mass in the filter. However, during the fluorescence measurement, the light absorption of the samples was mostly lower than 0.5. This indicates that the inner-filter effect or quenching was relatively low.

12. 189: Here, I was expecting a brief discussion of the contrast between the natural and manufactured (i.e., plastic) fuels.

Thank you for pointing this out. We added a brief discussion about chemical composition of aerosol between natural and manufactured fuels.

"In natural burning, the mass fraction of sugar derivatives ($C_5H_8O_4$, $C_8H_{12}O_6$, $C_6H_{12}O_5$) was higher (6.7 ± 3.5%) than that in plastic burning (4.9%). It indicates that natural burning produces more sugar derivatives than plastic burning on average, but these compounds are not a definitive tracer for distinguishing them."

13. 192: It is surprising to me to see such similar emissions for wood and plastic fuels - see my comment on line 105.

Thank you for pointing this out. Even though the plastic samples can detect $C_8H_{12}O_6$ and $C_5H_8O_4$, those compounds could be isomers compared to the same formulas in wood fuels. The FIGAERO-CIMS only can provide the molecular formulas. Therefore, we cannot name $C_8H_{12}O_6$ as Methyl-butanetricarboxylic acid and $C_5H_8O_4$ as glutaric acid. We changed the sentences as the following:

"$C_8H_{12}O_6$ and $C_5H_8O_4$ had also significant contributions of 1%-6% to the total signals of particles from combustion of beech wood, straw, and plastic fresh OA."

14. 210: I recommend reorganizing the panels here - POA could be aligned more clearly in the left column, and SOA in the right column.

We changed the figure 1 according to this comment.

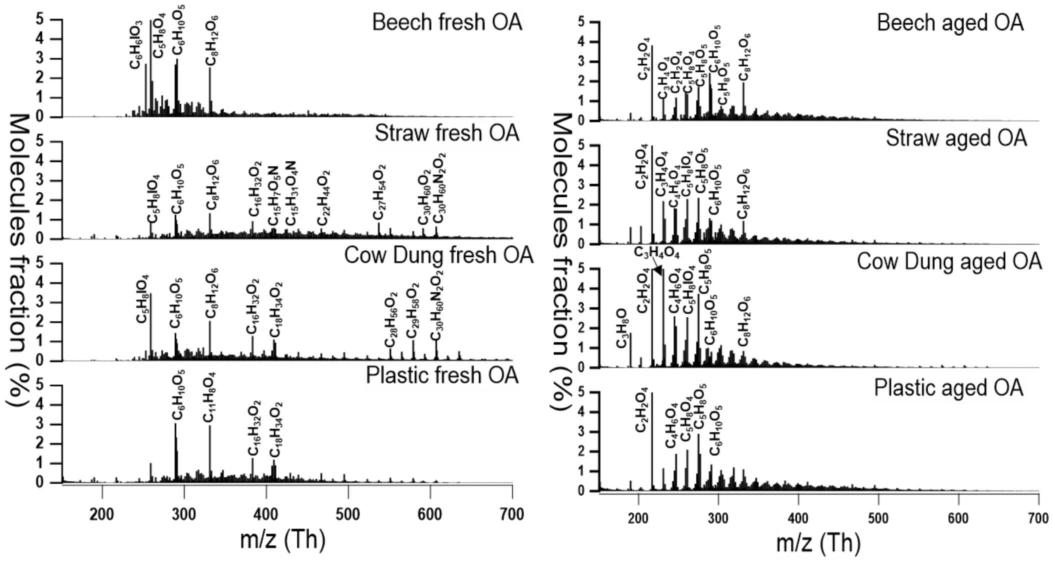


Figure 2. Mass spectra with iodide of particles from fresh OA and aged OA combustion emissions from FIGAERO-CIMS measurements. Particles from combustion/oxidation of beech wood; straw; plastics; cow dung. Please note that the signal intensity of levoglucosan ($C_6H_{10}O_5$) was multiplied by 0.1. Please note that the samples contain both primary emissions and secondary aerosol after photooxidation, we named these samples as aged organic aerosol.

15. 215: See my comment on line 189.

Thank you for pointing this out. We added a brief discussion about chemical composition of aerosol between natural and manufactured fuels

"After photooxidation, $C_6H_{10}O_5$ in plastic fuels showed a 66% decrease, which was larger than that in straw, Cow dung and beech fuels."

16. 229: Was each condition investigated once? See my comment on line 92.

We investigate each condition just once. There are no replicate experiments.

17. 285: Again, there are surprising similarities between plastic and biomass here. These similarities are a strong feature (e.g., we may be able to treat emissions of such diverse fuels similarly), and they justify discussion and/or rationalization.

The excitation emission spectroscopy has no molecular resolution. Even though the chromophores were classified as LO-HULIS, HO-HULIS and PLS also for plastics, the underlying molecular concentrations in different fuels could be different (Chen et al., 2020). Therefore, using EEM-based classifications alone as source tracers without molecular confirmation may lead to ambiguous source apportionment.

18. 308: The average molecular mass of the PLS fraction is interesting. Can you comment on whether it is consistent with, for example, monomers, dimers, oligomers, etc.?

The molecules of PLS chromophore had high molecular mass with 206.7 Da. Associated molecules could be monomer or dimmer, e.g. $C_{16}H_{14}O_2$, $C_{18}H_{32}O_2$, $C_{18}H_{34}O_2$, $C_{18}H_{35}O_2N_3$, etc. We added more content in the main manuscript.

"In addition, the associated molecules of PLS chromophores had some monomers or dimmers, e.g., $C_{16}H_{14}O_2$, $C_{18}H_{32}O_2$, $C_{18}H_{34}O_2$, $C_{18}H_{35}O_2N_3$, etc."

19. 309: Is this 5.9% by number or mass? Currently, it reads like number, but that would be inconsistent with the next paragraph, I believe.

The 5.9% is molecular intensities fraction detected by FIGAERO-CIMS.

"The 143 molecules associated with HO-HULIS chromophore accounted for 5.9 ± 4.4% of total organic molecular signals detected by FIGAERO-CIMS."

20. 329: This is an interesting combination of EES and CIMS, and a nice comparison.

Thank you.

21. 334: This figure is not a detailed mechanism - more a conceptual illustration.

We agree this Figure 5 more likely as a conceptual illustration. We changed the sentence as follows:

"The conceptual illustration is shown as Figure 5."

22. 350: Is the placement of O3 and OH versus NO2 meaningful? Can you briefly comment on why N is lower for the NO2 branch?

Sorry for the unclear information. The fresh organic aerosol and volatile organic compounds were oxidized and formed HO-HULIS and LO-HULIS in the presence of O_3, OH radicals and NO_2. To avoid misunderstandings, we modified Figure 5 as follows:

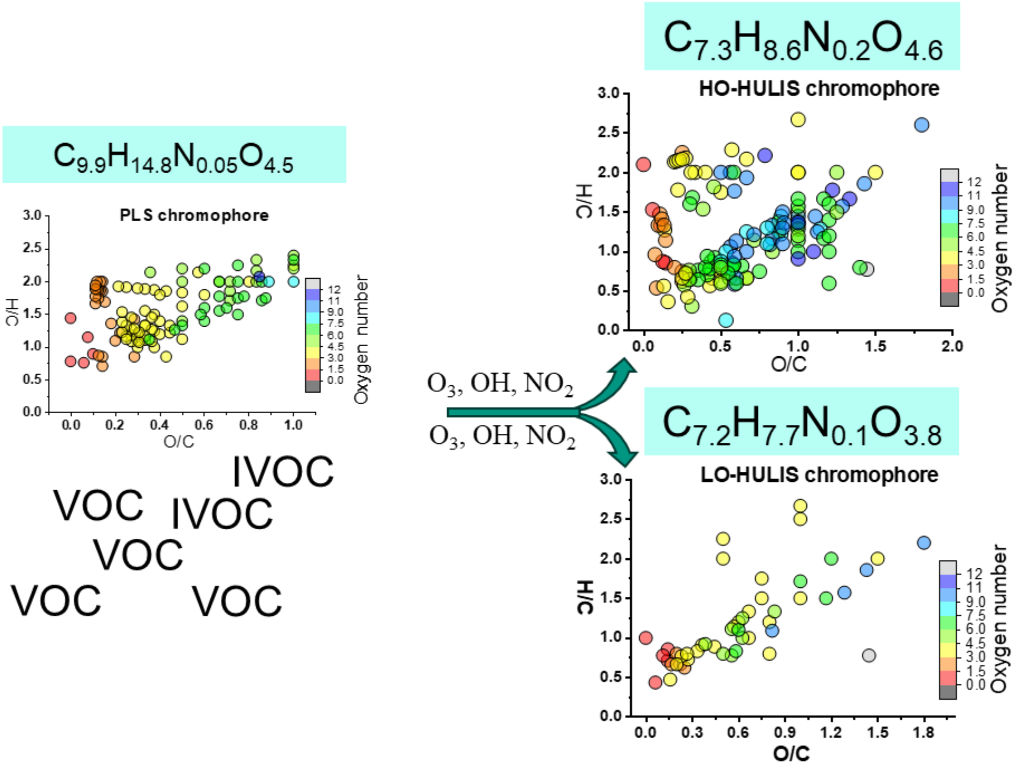


Figure 5. The PLS associated molecules and VOCs were oxidized into HO-HULIS associated molecules and LO-HULIS associated molecules during the photooxidation in the presence of NOx

23. 355: Please number this section.

Changed accordingly.

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