

We sincerely thank the referee for the helpful and constructive comments.

### **1. Low concentrations and uncertainty (Lines 225–226)**

The winter concentrations of the three OA fractions are quite small (HP-WSOM 0.13, HULIS 0.31, and WISOM 0.17  $\mu\text{g m}^{-3}$ ). Please clarify the associated uncertainty for such low values, ideally in relative terms or in comparison with the uncertainty of total OA. This would help the reader assess the robustness of the seasonal interpretation.

Two blank filter samples were extracted and analyzed following the same procedures as the ambient samples. Although the absolute signal intensities of the AMS spectra for OA extracts were not used for the quantification of OA masses (it can be affected by nebulizer conditions and the relationship between the size distributions of generated particles and the size window of the AMS inlet), we performed comparison between the absolute OA signal intensities (Hz) of the field blanks and those of the samples. For all samples, the blank-to-sample ratios of mean signal intensities were 20.0% for HP-WSOM, 6.1% for HULIS, and 4.9% for WISOM. In contrast, for winter samples, these ratios increased to 46.3%, 16.6%, and 13.2%, respectively. This indicates that the relative influence of blank signals was larger for low-signal samples, particularly for HP-WSOM in winter. Although not directly about the samples used in this study, further details on blank signal characteristics and potential biases associated with size distribution relative to the HR-AMS detection range can be found in Zhou et al. (2021). Note that we regard the signal intensities of the OA extracts to be too low to support reliable quantification using the phthalic acid internal standard method. The points mentioned above have been added in Lines 197–207.

The blank level associated with the aerosol sampling and the extraction procedure was assessed further. The results indicate a strong correlation of the sum of carbon in WISOM, HULIS and HP-WSOM with the total OC ( $r = 0.987$ ) (Fig. S3a) and the ratio of the latter to the former was  $0.75 \pm 0.12$  (mean  $\pm$  SD). To assess the uncertainty associated with low-concentration samples, we further calculated this ratio for winter, the season with low concentrations. The winter mean ratio was  $0.72 \pm 0.08$ , indicating that the uncertainty associated with WISOM was not significantly high in winter. The WSOC derived from AMS (HP-WSOM + HULIS) was in good agreement with the WSOC measured by the TOC analyzer, which is represented by a high correlation coefficient ( $r = 0.985$ ; Fig. S3b) and the ratio of the former to the latter of  $0.93 \pm 0.10$  (mean  $\pm$  SD). The corresponding winter mean ratio was  $0.92 \pm 0.10$ , suggesting that the uncertainty of AMS measurements for HP-WSOM and HULIS in winter was not noticeably higher. For TOC, the blank-to-sample ratio of mean values was 2.53%, whereas for the OC data, the values of the two blank samples were below the detection limit of 0.8  $\mu\text{g-C}$  per filter, which is much lower than the sample loadings (23.4 to 322  $\mu\text{g-C}$  per filter). Because results above show that the blank level does not have a significant influence, we consider that the uncertainty introduced by the absence of the subtraction of the offline AMS procedural blank is acceptable. This point is now explained in Lines 225–238.

In addition, the standard deviations have now been added to the summer and winter OA concentrations. (Lines 304–306).

### **2. Source-region discussion could be more specific (Lines 236–240; Fig. 3)**

The discussion of the PSCF results remains somewhat broad. The text states that air masses passed over Scandinavia and other parts of Europe, but from Fig. 3 the western-European influence does not appear equally clear for both events. Please specify more precisely which parts of Europe are implicated for P1 and P2. In addition, the map readability in Fig. 3 should be improved because the current figure is difficult to interpret.

The readability of the figure has been improved by adjusting the color scheme, and the corresponding source regions have been clarified in the revised manuscript (Lines 317–320).

### **3. Figure numbering / SI cross-reference inconsistency**

Please carefully check the figure numbering throughout the manuscript and Supplement. In the Methods, all sample trajectories are said to be shown in Figure S4 (Lines 180–181), and the PMF stability analysis is also said to be shown in Figure S4 (Lines 193–194). Later, in the Results, the backward trajectory analysis is instead referred to as Fig. S6 (Line 242). This is confusing and should be corrected throughout the manuscript and SI.

We apologize for the incorrect figure numbering. The numbering and description in text has now been corrected (Lines 247–249, 272, 327).

### **4. Clarify relation to previous SVOOA/LVOOA terminology (Lines 244–245 and Section 3.2.1)**

Since earlier Hyytiälä studies identified SVOOA/LVOOA, it would be useful to briefly explain for readers how these previously reported factors relate to the MO-OOA terminology adopted here. This would make it easier to place the current results in the context of previous source-apportionment work at the site.

MO-OOA in the present study should roughly correspond to LVOOA because it showed a high O/C ratio and strong oxygenated fragment ion signals. The BSOA-like factor may be more closely related to SVOOA in earlier studies because it was less oxidized than MO-OOA and likely represented relatively fresh SOA (Ng et al., 2010). This explanation has been added in Lines 363–367. The term MO-OOA is based on the degree of oxidation/oxygenation, while the latter is defined based on volatility. Since volatility-related data are not available in this work, we adopted the MO-OOA terminology, which is more directly interpretable. We used the term BSOA-like rather than SVOOA because it could be successfully associated with the biogenic source in this study.

### **5. PMF factor selection and apparent typo (Lines 270–272; Fig. 4 caption)**

Please comment briefly on whether a six-factor or higher-factor solution was examined and why it was not retained. Also, there appears to be a typo in the description of the four-factor solution: it is written as “MO-OOA, BSOA-like factor, BSOA-like factor, and HOA,” where one of the repeated BSOA-like entries is presumably intended to be BBOA-like. The same duplication appears in the Fig. 4 caption (“MO-OOA, BSOA-like, BSOA-like, CROA, and HOA”). Please correct this in both the main text and figure caption.

To investigate the potential sources of the three OA fractions (WISOM, HULIS, and HP-WSOM), we tested PMF solutions with different numbers of factors. The  $Q/Q_{\text{expected}}$  values of two and three factor solutions are substantially higher than the cases of four and five factor solutions. The PMF results from a four factor solution provided interpretable source apportionment and were broadly consistent with previous studies summarized in Table 1, such as Crippa et al. (2014). Both the four factor and five factor solutions were considered to provide reasonable source interpretations. The four-factor solution consisted of more-oxidized oxygenated organic

aerosol (MO-OOA), BBOA-like factor, BSOA-like factor, and HOA. In comparison, the five-factor solution further separated an additional combustion-related organic aerosol (CROA) factor characterized by distinct aromatic structure. Although  $Q/Q_{\text{expected}}$  continued to decrease with increasing factor number, the improvement becomes minor after the five factor solution (Fig. S4g, newly added). The five-factor solution was selected as a conservative solution because the number of resolved factors was close to those reported in previous PMF studies at Hyytiälä (Table 1). Although higher-factor solutions may contain more detailed information on submolecular structures or mixed sources, their source interpretation remains a challenge. The five-factor solution was therefore used for further discussion in the present study. The points above are now explained in Text S3.

The duplicated "BSOA-like factor" has been corrected.

**6. Paragraph logic around HOA/BSOA-like discussion (Lines 292–296).**

The paragraph beginning with HOA is somewhat awkward because the section is focused on the BSOA-like factor. I suggest restructuring it so that the paragraph starts from the BSOA-like behavior and then introduces HOA as a secondary observation. As written, the logic is somewhat difficult to follow.

We have added one sentence (Line 384) to improve the logical flow of this paragraph.

**7. Figure guidance for the BSOA-like tracer discussion (Lines 297–307)**

In the paragraph discussing the selected BSOA-related fragment ions, it would help the reader if the relevant figure reference were given at the start of the paragraph. At present, the text discusses the ions first and only later says that the results are shown in Fig. 5b. Since the previous paragraph already refers to Fig. 6, this section becomes harder to follow than necessary.

Fig. 5b is now referred to in Lines 390–391.

**8. Seasonality of  $C_5H_6O^+$  / tracer discussion (Line 314)**

Since  $C_5H_6O^+$  is presented as a particularly source-specific tracer for the BSOA-like factor, please comment more explicitly on its seasonal behavior in the main text. That would further strengthen the interpretation.

Thank you for this suggestion. The seasonal variation of  $C_5H_6O^+$  has been added to Figure 5a, and a brief explanation on the variation has been added in Lines 402–404.

**9. Tree species / regional vegetation context (Line 335)**

When using the lodgepole pine AMS spectrum and discussing MBO emissions from coniferous trees such as pines, please clarify whether this is representative of the dominant tree species in the region surrounding the site.

Lodgepole pine differs from the dominant species at SMEAR II, i.e., Scots pine (Heikkinen et al., 2021), which may cause confusion. However, as a coniferous species similar to Scots pine, one of the main forest-forming tree species in Fennoscandia (Heikkinen et al., 2021; Kuuluvainen and Aakala, 2011), lodgepole pine provides a relevant reference for comparison. We have clarified this in Lines 435–438.

**10. P1 and P2 should be labeled more consistently (Line 341 and related figures)**

P1 and P2 are introduced clearly in Fig. 2 and in the text, but it would help the reader if these periods were identified consistently throughout all relevant time-series figures, especially where their interpretation is

discussed later.

We have added P1 and P2 labels consistently in Figs. 5, 7, and 9.

#### 11. Sulfate associated with BBOA-like periods (Lines 347, 354–355)

The co-variation between  $\text{nss-SO}_4^{2-}$  and the BBOA-like factor is interesting, but the interpretation remains somewhat underdeveloped. Since the reported correlation is only moderate ( $r = 0.45$ ), please discuss more explicitly what processes or source regions might explain this association and how selective this relationship really is relative to the other PMF factors.

We apologize for the confusion caused by an error in our original description. We previously stated that  $\text{nss-SO}_4^{2-}$  showed peaks during P1 and P2 while  $\text{K}^+$  only slightly increased. However, the actual characteristics are the opposite. The correlation of  $\text{nss-SO}_4^{2-}$  with the BBOA-like factor was modest ( $r = 0.45$ ), and  $\text{nss-SO}_4^{2-}$  increased during P1 but did not show a clear increase during P2. Therefore, the association between the BBOA-like factor and  $\text{nss-SO}_4^{2-}$  was not particularly strong. In contrast,  $\text{K}^+$ , which is more strongly linked to biomass burning, shows clear peaks during both P1 and P2, consistent with the influence of BBOA during the periods. The points above have now been explained appropriately in section 3.2.4, Lines 440–450.

#### 12. Formatting consistency (Line 371 and elsewhere)

Please check the formatting of  $m/z$  and ion notation throughout the manuscript for consistency.

The expressions  $\text{C}_2\text{H}_4\text{O}_2^+$  and  $m/z$  60 were used inconsistently and without clarification. Whereas only unit mass  $m/z$  60 is often used to discuss the contribution of levoglucosan or BBOA in AMS studies, in high-resolution AMS studies, unit mass  $m/z$  60 can be resolved into  $\text{C}_2\text{H}_4\text{O}_2^+$  and other ions. We have clarified the distinction in the last paragraph of Section 3.2.4, Lines 476–484.

#### 13. CROA interpretation / other combustion sources (Line 394)

The discussion of CROA is interesting. In addition to aged fossil-fuel emissions and aged biomass-combustion material, could the authors briefly comment on whether coal combustion may also contribute to this aromatic factor, or explain why that possibility is unlikely in this setting?

This study did not explicitly distinguish OA from coal combustion and that from fossil fuel emissions. The sampling site is located in a sparsely populated boreal forest area, and OA at this site should have been strongly influenced by emissions from Eastern Europe and Russia, as discussed in Section 3.1.2. According to data from the International Energy Agency, coal and coal products still account for substantial fractions of the energy supply in some Eastern European countries, including Poland (32.7% in 2024, <https://www.iea.org/countries/poland>) and Estonia (56.6% in 2024, <https://www.iea.org/countries/estonia>). Therefore, the possible contribution of coal combustion is included in the fossil fuel emissions in this study.

#### 14. Interesting polarity/solubility anomalies (Lines 451, 455, 490)

There are a few periods in the later polarity-distribution discussion that appear noteworthy, including elevated MO-OOA in the water-insoluble fraction in late July, the small winter increase in BSOA-like material, and the behavior of HP-WSOM during the same period. Please comment on whether these features reflect genuine compositional changes or could partly result from factor mixing/rotational ambiguity and larger uncertainty under low-concentration conditions.

We apologize for the confusion, which was caused by an error in the figure legend. The purple color represents CROA, not MO-OOA, which is now correctly presented in the legend of Fig. 12.

The data points for MO-OOA during 15–22 December, CROA during 7–14 July, and BSOA-like in winter showed abrupt increases. As shown in Fig. 4b, these factors exhibited very low concentrations in all three fractions during these periods. Therefore, these apparent increases in water-insoluble fraction should be interpreted with caution. Given the complexity of ambient OA sources and formation and aging pathways, and also the uncertainty of OA mass spectra, representing the spectra by PMF inevitably involves some ambiguity, which may be more pronounced if factor concentrations are very low. This point is now addressed in Section 3.3.2, Lines 572–576.

Crippa, M., Canonaco, F., Lanz, V. A., Äijälä, M., Allan, J. D., Carbone, S., Capes, G., Ceburnis, D., Dall'Osto, M., Day, D. A., DeCarlo, P. F., Ehn, M., Eriksson, A., Freney, E., Hildebrandt Ruiz, L., Hillamo, R., Jimenez, J. L., Junninen, H., Kiendler-Scharr, A., Kortelainen, A. M., Kulmala, M., Laaksonen, A., Mensah, A. A., Mohr, C., Nemitz, E., O'Dowd, C., Ovadnevaite, J., Pandis, S. N., Petäjä, T., Poulain, L., Saarikoski, S., Sellegri, K., Swietlicki, E., Tiitta, P., Worsnop, D. R., Baltensperger, U., and Prévôt, A. S. H.: Organic aerosol components derived from 25 AMS data sets across Europe using a consistent ME-2 based source apportionment approach, *Atmos. Chem. Phys.*, 14, 6159-6176, <https://doi.org/10.5194/acp-14-6159-2014>, 2014.

Heikkinen, L., Äijälä, M., Daellenbach, K. R., Chen, G., Garmash, O., Aliaga, D., Graeffe, F., Rätty, M., Luoma, K., Aalto, P., Kulmala, M., Petäjä, T., Worsnop, D., and Ehn, M.: Eight years of sub-micrometre organic aerosol composition data from the boreal forest characterized using a machine-learning approach, *Atmospheric Chemistry and Physics*, 21, 10081-10109, <https://doi.org/10.5194/acp-21-10081-2021>, 2021.

Zhou, R., Chen, Q., Chen, J., Ren, L., Deng, Y., Vodicka, P., Deshmukh, D. K., Kawamura, K., Fu, P., and Mochida, M.: Distinctive Sources Govern Organic Aerosol Fractions with Different Degrees of Oxygenation in the Urban Atmosphere, *Environ Sci Technol*, 55, 4494-4503, <https://doi.org/10.1021/acs.est.0c08604>, 2021.

RC2: 'Comment on egusphere-2026-1023', Anonymous Referee #2

We sincerely thank the referee for the helpful and constructive comments.

- 1. Section 1, lines 80-83:** The AMS fragment ions  $m/z$  60 ( $C_2H_4O_2^+$ ) and  $m/z$  73 ( $C_3H_5O_2^+$ ) are commonly associated with levoglucosan and have been widely used as markers of fresh BBOA in previous studies. Although the diagnostic value of these two ions is weaker for aged BBOA, they are still important reference markers and should be acknowledged in the introduction.

An explanation about  $m/z$  60 ( $C_2H_4O_2^+$ ) and  $m/z$  73 ( $C_3H_5O_2^+$ ) has been added in Lines 82–83.

- 2. Section 2.2, lines 126-129:** Please clarify the extraction conditions in more detail. Was the extraction bottle capped and sealed during sonication? Was the sonication done in an ice bath or under other temperature-controlled conditions?

The vials for the extraction were capped and sealed with PTFE liners during sonication, and the sonication bath was not temperature-controlled, which are now explained in Lines 153-154. Regarding the concern about degradation, our group previously compared online AMS results and the results from the offline analysis using ultrasonication (Deng et al., 2022), which demonstrated that the offline AMS analysis with ultrasonication-based extraction did not cause apparent degradation detectable by the quantification and the elemental analysis. This point and the reference has also been added to Section 2.2, Lines 156–157.

- 3. Section 2.2, lines 131-135:** Please provide more details on the SPE procedure. Was the SPE cartridge conditioned and equilibrated before sample loading? Why was the aqueous extract acidified to pH 2 before SPE? Were the filter extraction and SPE procedures performed in the dark? What was the approximate duration of the full extraction and SPE workflow? Prolonged light exposure could potentially alter the OA composition through photooxidation.

Please justify the definitions of the two eluted fractions. Why is the first portion assigned as high polarity-WSOM and the retained portion as HULIS? A brief explanation supported by relevant literature would be helpful.

The conditioning and equilibration procedures are now explained in Lines 159–160. The SPE cartridge was conditioned with methanol to allow the solvent to efficiently access the stationary-phase pores and surface, thereby facilitating uniform interaction between the organic compounds in the sample and the active retention sites. The cartridge was then equilibrated with acidic 0.01 M HCl before sample loading in order to pre-adjust the system to pH 2 conditions before SPE according to the method originally proposed by Varga et al. (2001). This reference has already been cited in Section 2.2. Under neutral pH conditions, some organic compounds exist in ionic forms and are relatively hydrophilic, making them difficult to retain on reversed-phase SPE cartridges. At pH 2, acidic functional groups become protonated, increasing the hydrophobicity of the molecules and thus improving their retention on the cartridge. The acidification was performed immediately before sample loading, which minimizes possible acid hydrolysis.

The samples were stored in the dark, which mitigates possible changes in OA composition caused by light exposure. The filter extraction and SPE procedures were conducted under normal laboratory lighting conditions without light shielding. This point has been added to Lines 126–128. We have also added an explanation that minor degradation during the procedure is not fully ruled out to Lines 165–167.

Regarding the definitions of HP-WSOM and HULIS, a brief introduction has been provided in the Introduction section of the original manuscript (Lines 65-69 in the revised manuscript). According to the manufacturer's instructions, under acidic conditions, relatively hydrophobic organic compounds that can be retained by the HLB sorbent are adsorbed onto the cartridge and subsequently eluted with methanol. In contrast, more polar water-soluble organic compounds that are not effectively retained under these conditions pass through the cartridge first. Therefore, the first effluent fraction was operationally assigned as HP-WSOM, as described earlier by Chen et al. (2016). Our results also support this classification: HP-WSOM showed the highest O/C ratio among the fractions (Fig. 1b), indicating its higher oxidation level and hydrophilicity. Lin et al. (2010) was among the earlier studies to characterize aerosol HULIS using water extraction combined with SPE separation, and noted that HULIS is operationally defined by the isolation and detection protocol used for its determination. This clarification has also been added to the second paragraph of Section 2.2 (Lines 162–164).

**4. Section 2.3:** Please clarify whether blanks were analyzed, for example, by subjecting a blank filter to the same extraction, SPE, and AMS workflow as that used for the ambient samples. These blank measurements can provide background OM concentrations and AMS mass spectra and would help readers estimate potential contaminations from filter material and sample-processing steps. These data are particularly important when examining low-concentration samples.

Two blank filter samples were extracted and analyzed following the same procedures as the ambient samples. Although the absolute signal intensities of the AMS spectra for OA extracts were not used for the quantification of OA masses (it can be affected by nebulizer conditions and the relationship between the size distributions of generated particles and the size window of the AMS inlet), we performed comparison between the absolute OA signal intensities (Hz) of the field blanks and those of the samples. For all samples, the blank-to-sample ratios of mean signal intensities were 20.0% for HP-WSOM, 6.1% for HULIS, and 4.9% for WISOM. In contrast, for winter samples, these ratios increased to 46.3%, 16.6%, and 13.2%, respectively. This indicates that the relative influence of blank signals was larger for low-signal samples, particularly for HP-WSOM in winter. Although not directly about the samples used in this study, further details on blank signal characteristics and potential biases associated with size distribution relative to the HR-AMS detection range can be found in Zhou et al. (2021). Note that we regard the signal intensities of the OA extracts to be too low to support reliable quantification using the phthalic acid internal standard method. The points mentioned above have been added in Lines 197–207.

We added the HR-AMS spectra of two field blank samples, FIN20Q008 and FIN20Q020, to the dataset file to be published in Zenodo data repository. However, these data are provided for reference only.

Regarding the reproducibility of the experimental procedure, Table S3 summarizes the results of duplicate analyses for samples with moderate concentrations. The same extraction procedure, including the SPE step, was applied to evaluate uncertainties arising from sample handling and instrumental analysis. The maximum relative difference was 17.0%. For these two samples, the results from the duplicate analyses were averaged and were used in this study. This is now explained in Lines 208–211.

In addition, regarding the uncertainty of the quantification of low-concentration samples, we also assessed the quantifications of different methods for WSOC and total OC (Lines 225–238).

**5. Section 2.4, lines 181-182:** Please justify the use of 10-day back trajectories. Although HYSPLIT can be run over this duration, trajectory uncertainty increases with transport time, which may affect the

interpretation of the trajectory and PSCF results. Most receptor-oriented trajectory studies usually use shorter durations (e.g., 48-120 h) for source-region analysis. Please explain why a 10-day duration is used in this study.

SMEAR II is a site that is frequently influenced by long-range transport (Riuttanen et al., 2013), and therefore a 10-day back-trajectory duration was initially selected in this study. However, we agree that such long trajectories may be largely influenced by the increasing uncertainty associated with transport time. To improve the robustness and credibility of the manuscript, we have replaced the trajectories to the ones for 5-days, and the corresponding description in Section 2.4 has been revised accordingly (Line 248, Line 254).

**6. Section 2.5:** Please clarify the PMF input and preprocessing in more detail. How many variables were included? Were UMR data, HR data, or both used? Only organic ions or also any inorganic species were included? Please also describe any pretreatment applied to the error matrix before PMF, such as downweighing of noisy ions or  $\text{CO}_2^+$ -related ions.

We used the PMF Evaluation Toolkit v3.08C for PMF analysis, and only high-resolution organic ion data were included as input, comprising 1279 ions. The data were pretreated following the PET template and the procedure described by Ulbrich et al. (2009). Specifically, NaN and zero-only columns were removed before PMF analysis. For the error matrix, ions with  $\text{SNR} < 0.2$  were treated as bad variables and excluded, while ions with  $0.2 \leq \text{SNR} < 2$  were treated as weak variables and downweighted by doubling their uncertainties. In addition, duplicated  $\text{CO}_2^+$ -related ions ( $\text{CO}_2^+$ ,  $\text{CO}^+$ ,  $\text{H}_2\text{O}^+$ ,  $\text{HO}^+$ , and  $\text{O}^+$ ) were downweighted to avoid overweighting of duplicated information. These details are now explained in section 2.5 (Lines 263–270).

**7. Section 2.5, lines 191-192:** The author states that "HP-WSOM, HULIS, WISOM, and WSOM were processed together." If these four datasets were merged into a single PMF input matrix, please describe exactly how this was done. Were the matrices combined vertically or horizontally? These two approaches can lead to different types of PMF solutions and provide different scientific information.

Well, based on the PMF results shown in Figure 4, it appears that the datasets may have been combined vertically. If that is correct, please state this explicitly in the Methods section. The author should also briefly discuss why a "vertical combination" approach was used in this PMF analysis.

In addition, I'm curious about the PMF results that could be obtained if the input matrices were combined horizontally. In that setup, distinct MS profiles for the HP, HULIS, and WISOM fractions can be resolved for each PMF factor.

The spectra from different filter samples and different OA fractions (HP-WSOM, HULIS, WISOM, and WSOM) were treated independently in a single PMF input matrix. This has now been clarified in section 2.5 (Lines 263–266).

This approach can examine whether similar chemical structures appear across different fractions. A common PMF analysis for aerosol composition probably assumes that aerosols from respective sources have fixed composition. This assumption is, however, not necessarily reasonable in the case of AMS-based PMF analysis for OA because atmospheric aging of OA would change a part of the structure within molecules while keeping the structure of the remaining part. To be robust against such aging processes, consideration of the mass spectra

from different OA fractions without constraints is a reasonable approach. This view is consistent with the explanation in the Summary: “*The PMF factors identified in this study are better interpreted as dynamic mixtures of submolecular structures rather than fixed sets of organic compounds. This is because OAs from respective sources would not have nearly-fixed mass spectra, given the aging of OA during transport. This may also be the case for PMF factors reported in previous conventional online studies.*” Combining the mass spectra from different OA fractions into a single spectra is an interesting approach, but it is not adopted in this study.

Our group previously compared different PMF input strategies (Zhou et al., 2021). In addition to the method used in the present study, another method was tested: the mass spectra of different OA fractions from each sample were first summed into a single bulk-like OA spectrum before PMF analysis. This method is closer to the conventional PMF approach used for online AMS data. The similarity of PMF profiles and the contributions of some factors to total OA between the two different methods supports that our PMF analysis is not substantially different from conventional PMF analysis of online AMS data. Another point is that the number of samples could affect the robustness of the PMF solution, and the PMF results from the concatenation of the spectra has an disadvantage because of the decrease in the number of inputs.

**8. Section 3.1.3, lines 243:** The author states that "The backward trajectory analysis (Fig. S6) reveals the possibility of long-range transport for P1." This interpretation would benefit from more careful justification. With a 10-day trajectory length, the older portions of the trajectories are subject to large uncertainties, and longer trajectories typically span much broader spatial domains. These factors may bias the PSCF analysis towards emphasizing remote source regions.

We have updated the analysis by using 5-day trajectories for the PSCF calculation. Accordingly, Fig. S7 and S8 in the Supporting Information have also been revised to present the 5-day trajectory results. The 5-day and 10-day PSCF were generally similar, and the related discussion remained unchanged.

**9. Section 3.2.3:** When evaluating whether an AMS fragment ion can serve as a marker for BSOA, it's better to consider not only its correlations with the factor and external references, but also its relative abundance in the factor MS profile. I encourage the author to also report the relative abundances of the proposed marker ions (e.g.,  $C_5H_6O^+$ ,  $C_7H_9O_3^+$ , and  $C_9H_{13}O_4^+$ ) in the factor MS.

We examined the relative abundances of the three proposed marker ions, and the results are shown in Tables S6 and S7. Taking  $C_5H_6O^+$  as an example, both the CROA and BSOA-like factors exhibited higher relative abundances than the other factors (Table S6). However, because of the large abundance, the factor contribution was dominated by the BSOA-like factor (Table S7). It should also be noted that relative abundance of  $C_5H_6O^+$  in the BSOA-like factor (0.35%) falls within the range ( $0.55 \pm 0.20\%$ ) reported for monoterpene-derived SOA by Hu et al. (2015). Explanations on the points above have been added to Section 3.2.3, Lines 401–410.

Both the CROA and BSOA-like factors showed high relative abundances of  $C_5H_6O^+$ , showing the limited specificity of  $C_5H_6O^+$ . Its relative abundance is only about one quarter of that observed in the spectrum of IEPOX-SOA (Hu et al., 2015), making it difficult to use as a standalone marker. To our knowledge, an independent BSOA tracer suitable for monoterpene-dominated environments have not been found. The differences in the

mass spectral characteristics of BBOA-like, BSOA-like, and CROA factors are as follows. The BSOA-like factor was enriched in oxygenated fragment ions, especially  $C_xH_yO_{>1}$  ions, whereas CROA showed stronger aromatic fragment ion characteristics, and BBOA-like was characterized by CHN-family structures and a distinctly higher N/C ratio. The explanations on the association of sources to the PMF factors in Section 3.2.3 have been revised.

**10. Section 3.3.3, line 467-470:** When interpreting seasonality and source influences, both the absolute concentration and the fractional contribution of WISOM should be considered. Although the fractional contribution of WISOM increased (Figure 12), the absolute concentration of WISOM was lower in winter (Figure 2). The statement "This seasonality likely reflects increased inputs of fossil fuel-derived aerosols in winter" may need more careful justification.

We agree that the increased fractional contribution of WISOM in winter requires a more careful interpretation and should not be linked to enhanced fossil fuel emissions alone. We have substantially revised the discussion in Section 3.3.3 and have softened the tone about the association between the wintertime increase in the WISOM fraction and fossil fuel-derived aerosols.

#### **Minor points:**

1. Line 22: Please spell out CROA at its first occurrence.

Revised.

2. Line 65: Change "isolated by using SPE" to "isolated using SPE".

Revised.

3. Lines 75-77: This sentence doesn't make sense to me. I think fragmentation in AMS breaks molecules into smaller ions, which leads to loss of much of the original molecular structure and therefore reduces molecular specificity.

Our previous wording was not fully appropriate. We have revised the part as: "Compared with the molecular-tracer-based approach, fragment ions from AMS analysis may retain structural information of compounds including those are not identified and quantified in molecular-based approach." (Lines 76-77)

4. General comment on terminology: Throughout the manuscript, the authors use terms such as "chemical structural information" and "chemical structures" to describe the information obtained from AMS. In my view, this wording may be somewhat too strong. AMS provides valuable information on elemental composition, fragment ion patterns, and broad chemical composition, but it does not directly resolve molecular structures in a strict sense. I suggest using more cautious language, such as "chemical composition" and "chemical information".

We agree that the term “chemical structures” may be too strong, because it should generally mean the arrangement of atoms and chemical bonds. However, we are concerned that the term “chemical composition” may also be misleading, as it could be interpreted as referring to the composition of individual compounds. The information provided by AMS includes structural information from fragment ion patterns. Some ion families such as CHN and CHON and characteristic ions like  $C_5H_6O^+$  can be associated with potential structural motifs. Therefore, we regard “chemical structural information” and “chemical structural characteristics” to be appropriate for describing the structural features inferred from the fragmentation patterns. We have replaced “chemical structures” throughout the manuscript with “chemical structural characteristics” or “chemical structural information” depending on the context.

5. Table S2: Please specify in the table note which instrument was used to determine the WSOM, HULIS, and HP-WSOM concentrations.

The table caption has been revised accordingly.

6. Line 139: Please provide the manufacturer and model information for the gas exchange device.

The manufacturer and model information for the gas exchange is now provided.

7. Please check the figure numbering and cross-references throughout the manuscript and supplement.

Line 181: The sample trajectories are cited as Figure S4, but they appear in Figure S7.

Line 242: The back trajectories are cited as Figure S6, but they appear in Figure S8.

Line 219: "Fig. 2a" should be changed to "Fig. 2"

Corrected.

8. The section numbering skips from 3.1.1 to 3.1.3.

Section numbering has been corrected.

9. Figure 4 caption repeats BSOA-like and omits BBOA-like.

Corrected.

10. Line 316: "BBOA" should be corrected to "BSOA".

Corrected.

11. Figure 5b: Please clarify how the correlations were calculated. Were they based on the summed concentrations across HP-WSOM, HULIS, and WISOM?

The correlations were calculated by the sum of fragment ions in HP-WSOM, HULIS, and WISOM. The Figure 5b. caption has been revised.

12. Figure 6: In the figure caption, please clarify that the correlations are of different types. Some are based on mass spectral similarity and others are based on time series relationships.

The figure caption has been revised according to the comment.

13. Line 338: How were  $\text{nss-K}^+$  and  $\text{nss-SO}_4^{2-}$  calculated?

The calculation methods for  $\text{nss-K}^+$  and  $\text{nss-SO}_4^{2-}$  have been added (in Line 440–442).

14. Figure 11: It may be helpful to add a summary panel showing the seasonal average contributions of the factors.

A summary panel has now been added to Figure 11.

Chen, Q., Ikemori, F., and Mochida, M.: Light Absorption and Excitation-Emission Fluorescence of Urban Organic Aerosol Components and Their Relationship to Chemical Structure, *Environ Sci Technol*, 50, 10859-10868, 10.1021/acs.est.6b02541, 2016.

Deng, Y., Fujinari, H., Yai, H., Shimada, K., Miyazaki, Y., Tachibana, E., Deshmukh, D. K., Kawamura, K., Nakayama, T., Tatsuta, S., Cai, M., Xu, H., Li, F., Tan, H., Ohata, S., Kondo, Y., Takami, A., Hatakeyama, S., and Mochida, M.: Offline analysis of the chemical composition and hygroscopicity of submicrometer aerosol at an Asian outflow receptor site and comparison with online measurements, *Atmos. Chem. Phys.*, 22, 5515-5533, <https://doi.org/10.5194/acp-22-5515-2022>, 2022.

Hu, W. W., Campuzano-Jost, P., Palm, B. B., Day, D. A., Ortega, A. M., Hayes, P. L., Krechmer, J. E., Chen, Q., Kuwata, M., Liu, Y. J., de Sá, S. S., McKinney, K., Martin, S. T., Hu, M., Budisulistiorini, S. H., Riva, M., Surratt, J. D., St. Clair, J. M., Isaacman-Van Wertz, G., Yee, L. D., Goldstein, A. H., Carbone, S., Brito, J., Artaxo, P., de Gouw, J. A., Koss, A., Wisthaler, A., Mikoviny, T., Karl, T., Kaser, L., Jud, W., Hansel, A., Docherty, K. S., Alexander, M. L., Robinson, N. H., Coe, H., Allan, J. D., Canagaratna, M. R., Paulot, F., and Jimenez, J. L.: Characterization of a real-time tracer for isoprene epoxydiols-derived secondary organic aerosol (IEPOX-SOA) from aerosol mass spectrometer measurements, *Atmos. Chem. Phys.*, 15, 11807-11833, <https://doi.org/10.5194/acp-15-11807-2015>, 2015.

Lin, P., Huang, X.-F., He, L.-Y., and Zhen Yu, J.: Abundance and size distribution of HULIS in ambient aerosols at a rural site in South China, *Journal of Aerosol Science*, 41, 74-87, <https://doi.org/10.1016/j.jaerosci.2009.09.001>, 2010.

Riuttanen, L., Hulkkonen, M., Dal Maso, M., Junninen, H., and Kulmala, M.: Trajectory analysis of atmospheric transport of fine particles, SO<sub>2</sub>, NO<sub>x</sub> and O<sub>3</sub> to the SMEAR II station in Finland in 1996–2008, *Atmospheric Chemistry and Physics*, 13, 2153-2164, <https://doi.org/10.5194/acp-13-2153-2013>, 2013.

Ulbrich, I., Canagaratna, M., Zhang, Q., Worsnop, D., and Jimenez, J.: Interpretation of organic components from Positive Matrix Factorization of aerosol mass spectrometric data, *Atmospheric Chemistry and Physics*, 9, 2891-2918, <https://doi.org/10.5194/acp-9-2891-2009>, 2009.

Varga, B., Kiss, G., Ganszky, I., Gelencsér, A., and Krivácsy, Z.: Isolation of water-soluble organic matter from atmospheric aerosol, *Talanta*, 55, 561-572, [https://doi.org/10.1016/S0039-9140\(01\)00446-5](https://doi.org/10.1016/S0039-9140(01)00446-5), 2001.

Zhou, R., Chen, Q., Chen, J., Ren, L., Deng, Y., Vodicka, P., Deshmukh, D. K., Kawamura, K., Fu, P., and Mochida, M.: Distinctive Sources Govern Organic Aerosol Fractions with Different Degrees of Oxygenation in the Urban Atmosphere, *Environ Sci Technol*, 55, 4494-4503, <https://doi.org/10.1021/acs.est.0c08604>, 2021.

We sincerely thank the referee for the helpful and constructive comments.

**Major comments:**

- 1. Please provide more details on the analysis method in the Method section. For example, how were the one-year aerosol samples stored? Were any background measurements taken, and are the results presented background-subtracted?**

How the one-year aerosol samples were stored is now explained in Lines 126–128.

Two blank filter samples were extracted and analyzed following the same procedures as the ambient samples. Although the absolute signal intensities of the AMS spectra for OA extracts were not used for the quantification of OA masses (it can be affected by nebulizer conditions and the relationship between the size distributions of generated particles and the size window of the AMS inlet), we performed comparison between the absolute OA signal intensities (Hz) of the field blanks and those of the samples. For all samples, the blank-to-sample ratios of mean signal intensities were 20.0% for HP-WSOM, 6.1% for HULIS, and 4.9% for WISOM. In contrast, for winter samples, these ratios increased to 46.3%, 16.6%, and 13.2%, respectively. This indicates that the relative influence of blank signals was larger for low-signal samples, particularly for HP-WSOM in winter. Although not directly about the samples used in this study, further details on blank signal characteristics and potential biases associated with size distribution relative to the HR-AMS detection range can be found in Zhou et al. (2021). Note that we regard the signal intensities of the OA extracts to be too low to support reliable quantification using the phthalic acid internal standard method. The points mentioned above have been added in Lines 197–207.

The blank level associated with the aerosol sampling and the extraction procedure was assessed further. The results indicate a strong correlation of the sum of carbon in WISOM, HULIS and HP-WSOM with the total OC ( $r = 0.987$ ) (Fig. S3a) and the ratio of the latter to the former was  $0.75 \pm 0.12$  (mean  $\pm$  SD). To assess the uncertainty associated with low-concentration samples, we further calculated this ratio for winter, the season with low concentrations. The winter mean ratio was  $0.72 \pm 0.08$ , indicating that the uncertainty associated with WISOM was not significantly high in winter. The WSOC derived from AMS (HP-WSOM + HULIS) was in good agreement with the WSOC measured by the TOC analyzer, which is represented by a high correlation coefficient ( $r = 0.985$ ; Fig. S3b) and the ratio of the former to the latter of  $0.93 \pm 0.10$  (mean  $\pm$  SD). The corresponding winter mean ratio was  $0.92 \pm 0.10$ , suggesting that the uncertainty of AMS measurements for HP-WSOM and HULIS in winter was not noticeably higher. For TOC, the blank-to-sample ratio of mean values was 2.53%, whereas for the OC data, the values of the two blank samples were below the detection limit of  $0.8 \mu\text{g-C}$  per filter, which is much lower than the sample loadings (23.4 to  $322 \mu\text{g-C}$  per filter). Because results above show that the blank level does not have a significant influence, we consider that the uncertainty introduced by the absence of the subtraction of the offline AMS procedural blank is acceptable. This point is now explained in Lines 225–238.

Regarding the reproducibility of the experimental procedure, Table S3 summarizes the results of duplicate analyses for samples with moderate concentrations. The same extraction procedure, including the SPE step, was applied to evaluate uncertainties arising from sample handling and instrumental analysis. The maximum relative difference was 17.0%. For these two samples, the results from the duplicate analyses were averaged and were

used in this study. This is now explained in Lines 208–211.

- 2. WISOM was extracted using several organic solvents, which could contribute fragmentation ions in the following AMS measurement. Could the authors comment on whether these organic solvents could contribute signals to the five PMF factors? Similarly, in the extraction of WSOM, were the HCl and methanol removed before the OA was analyzed by AMS? How can we evaluate the effect of these solvents on the AMS mass spectra and the resulted PMF factors?**

As described in Section 2.3, the generated aerosols were passed through diffusion driers with silica gel and activated carbon to remove solvent vapors, and the carrier gas was subsequently replaced with high-purity Ar using a gas exchange device. After these two steps, the majority of HCl and solvents should have been removed before the AMS analysis, considering their high volatility. Therefore, they were unlikely to enter the AMS and contribute significantly to the measured signals.

In addition, this study mainly focused on OA, and only the organic ion signals were used for the PMF analysis. Even if small amounts of chloride related signals were present, they were not included in Org or in the PMF input matrix. Therefore, they would not significantly affect the results. For solvents, even if a small amount of the solvent vapor enters the AMS, their molecular sizes are too small to be efficiently focused and transmitted by the aerodynamic lens like aerosol particles. Therefore, the interference is considered very low.

The agreement between OC measured by the carbon analyzer and the sum of the three fractions measured by AMS also suggests that the influence of HCl and solvents on the experimental results was small (Lines 225–238). Furthermore, as mentioned in the first comment, the AMS analysis of the field blanks also provides additional support (Lines 197–207).

- 3. A nebulizer was used to generate aerosols. Please add more details on the nebulizer, e.g., operation flow rate and inlet pressure. Since the compounds in OA may have different volatilities, will there be potential loss of semi-volatile compounds in the setup or different efficiencies in generating the aerosols? Could the authors comment on the effect of size-dependent transmission efficiency of AMS on the quantification of aerosols generated from the nebulizer? Why did the authors generate aerosols using compressed air and then transfer to Ar?**

Aerosols were generated using a home built atomizer equipped with an electrospray nebulizer assembly (G1946-60098 and/or equivalent model; Agilent Technologies, Inc.), to nebulize the solution delivered from a syringe pump without applying a voltage. Approximately 1 mL of the extract solution was loaded into a 5 mL syringe. This information has been added to Lines 169–175.

The inlet pressure was not recorded. It should be noted that the quantification method does not rely on the absolute aerosol mass concentration generated by the nebulizer. Instead, the quantification is based on the relative abundance of the extract and phthalic acid in the extract–phthalic acid mixture, rather than on the absolute amount of aerosol mass produced by the nebulizer (Mihara and Mochida, 2011). Therefore, the nebulizer condition is expected to have a very limited influence on the final quantification.

Because nebulization of the extract solutions and passage of the generated aerosols through diffusion dryers filled with silica gel and activated carbon required a large and stable gas flow to ensure effective removal of potential contaminants, compressed air was first used for aerosol generation and drying. Therefore, generating and drying the aerosols with compressed air first, followed by replacement of the carrier gas with Ar before introduction into the AMS, is considered a more practical and stable experimental approach.

The main advantage of using Ar is that the  $\text{N}_2^+$  signal at  $m/z$  28 can be greatly reduced, thereby suppressing its interference with the quantification of  $\text{CO}^+$  and  $\text{CHO}^+$ . In the case of the analysis in unit mass resolution,  $\text{CO}^+$  at  $m/z$  28 was still assumed to be equal to  $\text{CO}_2^+$  at  $m/z$  44, following the common treatment used in online AMS analysis.

**4. The calculation of SPE extraction efficiency: Does the mass concentration in  $\mu\text{g m}^{-3}$  in Table S2 refer to the concentration of the organic matters in the aqueous solution or in the aerosols? If it is the concentration in aerosols, how was the extraction efficiency estimated? More detailed information would be helpful.**

An explanation of the calculation and the meaning of the concentrations has been added to the caption of Table S2.

**Minor comments:**

1. Please add the full name of CROA in the abstract.

The full name has been added (Line 23).

2. The abbreviation for "OAs" is duplicated in lines 30 and 39.

The latter description about the abbreviation has been deleted (Line 39).

3. Line 57: What do the "additional tools" refer to?

The additional tools such as FT-IR or NMR are now explained in line 57.

4. Line 71: What does "each fraction" refer to?

The OA fractions (HP-WSOM, HULIS and WISOM) have been added in lines 71-72.

5. Section 3.1.2 is missing. Section 3.1.3: it would be helpful to add a sentence to summarize the possible OA sources of P1 and P2.

"Section 3.1.3" has been corrected to "Section 3.1.2". More descriptions regarding the possible OA sources have also been added in the second paragraph of Section 3.1.2.

6. Is the quantification of OA based on V-mode or W-mode AMS results? Are the mass spectra used for PMF analysis based on V-mode or W-mode results? Please clarify.

The V-mode data were used only for the quantification of extracted OAs. The W-mode data were used for other mass spectral analyses. This information has been added to Lines 175–178.

7. Line 232: What is the PSCF analysis? Similar comment on line 234: “DMPS measurement”. Please add more information on them to the Method section. Additionally, why did the authors choose the 75th percentile instead of the 90th or 60th percentile in the PSCF analysis?

An explanation of the PSCF analysis has been added to Section 2.4 (Lines 250-258). The description of the DMPS measurements has also been added to Text S2.

The 75th percentile is widely used in previous studies (Deng et al., 2020; Zhou et al., 2018; Xu and Akhtar, 2010). The figure clearly shows that the thresholds select characteristic high-concentration data points. We regard that the 75<sup>th</sup> percentile threshold provides a balance between selectivity and statistical robustness. The choice of the 75th percentile is now explained in Lines 257-260, where a new figure (Fig. S9) is referred.

8. Line 273: A potential typo: two “BSOA-like factors” in the four-factor solution. Or is it one BSOA-like factor and one BBOA-like factor?

The first “BSOA-like factor” should be “BBOA-like factor.” This has been corrected in Section 3.2.2, Line 357.

9. Line 294-296: Not sure I agree with the interpretation, because the concentrations of most PMF factors increased in P2.

There is a study claiming that regional scale anthropogenic pollution conditions, such as anthropogenic emissions of SO<sub>2</sub> and NO<sub>x</sub>, can affect the concentration levels and composition of BSOA in the Nordic region (Nguyen et al., 2014). In the revised manuscript, we no longer claim this possibility strongly, but describe it as one of the possible reasons (Lines 387-388).

10. Figure 10: The y-axis scaling cuts off some prominent peaks. Either use broken axes or state the scaling explicitly in the caption.

We have added a note in the caption stating that the full-scale spectra are shown in Figure 4a.

Deng, J., Guo, H., Zhang, H., Zhu, J., Wang, X., and Fu, P.: Source apportionment of black carbon aerosols from light absorption observation and source-oriented modeling: an implication in a coastal city in China, *Atmos. Chem. Phys.*, 20, 14419-14435, <https://doi.org/10.5194/acp-20-14419-2020>, 2020.

Mihara, T. and Mochida, M.: Characterization of Solvent-Extractable Organics in Urban Aerosols Based on Mass Spectrum Analysis and Hygroscopic Growth Measurement, *Environmental Science & Technology*, 45, 9168-9174, <https://doi.org/10.1021/es201271w>, 2011.

Nguyen, Q. T., Christensen, M. K., Cozzi, F., Zare, A., Hansen, A. M. K., Kristensen, K., Tulinius, T. E., Madsen, H. H., Christensen, J. H., Brandt, J., Massling, A., Nøjgaard, J. K., and Glasius, M.: Understanding the anthropogenic influence on formation of biogenic secondary organic aerosols in Denmark via analysis of organosulfates and related oxidation products, *Atmos. Chem. Phys.*, 14, 8961-8981, <https://doi.org/10.5194/acp-14-8961-2014>, 2014.

Xu, X. and Akhtar, U. S.: Identification of potential regional sources of atmospheric total gaseous mercury in Windsor, Ontario, Canada using hybrid receptor modeling, *Atmos. Chem. Phys.*, 10, 7073-7083, <https://doi.org/10.5194/acp-10-7073-2010>, 2010.

Zhou, B., Wang, Q., Zhou, Q., Zhang, Z., Wang, G., Fang, N., Li, M., and Cao, J.: Seasonal Characteristics of Black Carbon Aerosol and its Potential Source Regions in Baoji, China, *Aerosol and Air Quality Research*, 18, 397-406, <https://doi.org/10.4209/aaqr.2017.02.0070>, 2018.

Zhou, R., Chen, Q., Chen, J., Ren, L., Deng, Y., Vodicka, P., Deshmukh, D. K., Kawamura, K., Fu, P., and Mochida, M.: Distinctive Sources Govern Organic Aerosol Fractions with Different Degrees of Oxygenation in the Urban Atmosphere, *Environ Sci Technol*, 55, 4494-4503, <https://doi.org/10.1021/acs.est.0c08604>, 2021.

We sincerely thank the referee for the helpful and constructive comments.

### Major comments:

#### 1. Physical Assumption of PMF Matrix Concatenation:

In Section 2.5, the authors state that the mass spectra of HP-WSOM, HULIS, and WISOM were "processed together". It is fundamentally unclear how the data matrices were combined (e.g., were they appended into a single concatenated matrix?). More importantly, concatenating chemically fractionated samples into a single 2D PMF model inherently assumes that a resolved source factor (e.g., BBOA-like) possesses the exact same mass spectral profile regardless of the polarity fraction it resides in. Chemically, this is highly unlikely. Since fractionation physically separates molecules by polarity, the BBOA components in the HP-WSOM fraction should have a higher degree of oxygenation (higher O/C) and a different fragmentation pattern than the BBOA components in the non-polar WISOM fraction. Forcing a static factor profile across fractions of varying polarities may lead to significant mathematical distortion. The authors must explicitly justify the validity of this physical assumption and explain how ion contributions were handled across fractions.

The spectra from different filter samples and different OA fractions (HP-WSOM, HULIS, WISOM, and WSOM) were treated independently in a single PMF input matrix. This has now been clarified in section 2.5 (Lines 262–265).

This approach can examine whether similar chemical structures appear across different fractions. A common PMF analysis for aerosol composition probably assumes that aerosols from respective sources have fixed composition. This assumption is, however, not necessarily reasonable in the case of AMS-based PMF analysis for OA because atmospheric aging of OA would change a part of the structure within molecules while keeping the structure of the remaining part. To be robust against such aging processes, consideration of the mass spectra from different OA fractions without constraints is a reasonable approach. Therefore, we did not assume that factor profiles remain statically identical across fractions with different polarities. Instead, we emphasize their dynamic characteristics. In our case, since the relative contributions of HP-WSOM, HULIS, and WISOM differed among individual samples, the chemical structures represented by a given factor may also vary dynamically at the sample level (This point now added to Line 110–117).

This view is consistent with the explanation in the Summary: *“The PMF factors identified in this study are better interpreted as dynamic mixtures of submolecular structures rather than fixed sets of organic compounds. This is because OAs from respective sources would not have nearly-fixed mass spectra, given the aging of OA during transport. This may also be the case for PMF factors reported in previous conventional online studies.”* Combining the mass spectra from different OA fractions into a single spectra is an interesting approach, but it is not adopted in this study. The summary part is also revised (lines 643–650).

In addition, our group previously compared different PMF input strategies (Zhou et al., 2021). In addition to the method used in the present study, another method was tested: the mass spectra of different OA fractions from each sample were first summed into a single bulk-like OA spectrum before PMF analysis. This method is closer to the conventional PMF approach used for online AMS data. The similarity of PMF profiles and the contributions of some factors to total OA between the two different methods supports that our PMF analysis is

not substantially different from conventional PMF analysis of online AMS data. Another point is that the number of samples could affect the robustness of the PMF solution, and the PMF results from the concatenation of the spectra has an disadvantage because of the decrease in the number of inputs.

## 2. The Discrepancy Regarding the C<sub>2</sub>H<sub>4</sub>O<sub>2</sub> (m/z 60) Tracer:

In Figures 9 and 10, the authors use C<sub>2</sub>H<sub>4</sub>O<sub>2</sub><sup>+</sup> as a marker for BBOA based on its strong temporal correlation. However, it is highly contradictory that the corresponding BBOA-like mass spectrum (Figure 10) shows little to no contribution at m/z 60. If m/z 60 perfectly tracks BBOA temporally, why did the PMF algorithm fail to allocate this ion into the BBOA factor profile? This is likely a direct consequence of the PMF concatenation issue mentioned above: levoglucosan is highly water-soluble and may have partitioned exclusively into the HP-WSOM fraction, while the bulk mass of the BBOA factor was driven by HULIS/WISOM. This glaring contradiction between temporal correlation and spectral absence must be critically explained.

We appreciate the reviewer's comment, which is important for a reasonable explanation of the results. Although C<sub>2</sub>H<sub>4</sub>O<sub>2</sub><sup>+</sup> showed a relatively strong temporal correlation with the BBOA-like factor, the contribution of BBOA-like factor to C<sub>2</sub>H<sub>4</sub>O<sub>2</sub><sup>+</sup> is negligible. As shown in Fig. 1a, the relative abundance of m/z 60 was very low, accounting for only approximately 0.23%, 0.086%, and 0.094% of the total signal in HP-WSOM, HULIS, and WISOM, respectively. The presence of BBOA-like signals in HP-WSOM may reflect aged or secondary biomass-burning-related products, even though levoglucosan may have been depleted. The background level of m/z 60 for SOA-dominated ambient OA has been reported to be 0.3% ± 0.06% (Cubison et al., 2011). The relative abundances of m/z 60 in the three fractions were all lower than or close to this background level. We have therefore revised the discussion substantially, as shown in Lines 480–490.

To further validate the assignment of the BBOA-like factor, we revised Fig. 9 and added an explanation based on the EC/OC ratio in the beginning of Section 3.2.5 (Lines 493–501). The EC/OC ratio was used as an auxiliary indicator of combustion-related aerosol influence. The strong consistency between the seasonal variations of the BBOA-like and CROA factors and that of the EC/OC ratio, together with their higher contributions in winter, supports the association of these two factors with combustion-related sources. In addition to this, as we have explained in the original manuscript, the CHN ion family and the comparison with a reference BBOA spectrum provide additional support for the BBOA-like factor assignment. Taken together, these lines of evidence support that the assignment of the BBOA-like factor is reasonable.

## 3. Chemical Artifacts during Offline Extraction (pH Adjustment):

In Section 2.2, the authors describe acidifying the WSOM to pH 2 using 1 M HCl before Oasis HLB solid-phase extraction, followed by elution with methanol. Subjecting a complex mixture of organic aerosols to highly acidic conditions combined with methanol can induce significant chemical alterations, such as acid-catalyzed esterification, hydrolysis, or the ring-opening of biogenic epoxides (e.g., IEPOX-derived SOA). Furthermore, at pH 2, basic compounds like amines (proposed as CHN tracers for BBOA) will be heavily protonated, drastically altering their retention on a reverse-phase SPE column. Since this study heavily relies on specific HR AMS structural fragments to identify sources, the authors must explicitly discuss the potential that some structural features or O/C ratios are artifacts of the aggressive sample preparation.

To assess whether the chemical structure changed by the SPE, we reconstructed the WSOM spectra from the corresponding HP-WSOM and HULIS spectra using their measured mass ratios, and reconstructed spectra were

compared with the directly measured WSOM spectra. For both samples, the reconstructed WSOM spectra showed high correlations with the measured WSOM spectra, with correlation coefficients of 0.996 for FIN20Q002 and 0.989 for FIN20Q018. In addition, the family level distributions of the measured and reconstructed WSOM spectra were highly consistent. Considering possible operational uncertainties during extraction and SPE fractionation, the observed differences were considered small. Although transformations of individual compounds cannot be fully excluded, these comparisons suggest that such effects did not substantially modify the bulk AMS spectral features. An explanation about this point has been added to Lines 192–195, and the table for comparison has been added as Table S8.

This study emphasizes chemical structure-related information (such as CHN ion families and groups with different O/C ratios) rather than specific chemical compounds themselves. Therefore, even if the chemical transformations mentioned by the reviewer occurred during the extraction procedure, their influence could be limited because AMS measurements are based on fragment ions rather than intact molecular components.

#### **4. Filter Sampling Artifacts for Semi-Volatile OAs:**

The aerosol samples were collected on filters over prolonged periods of one week. A 7-day sampling duration is highly susceptible to severe sampling artifacts, particularly the evaporative loss (negative artifact) of semi-volatile organic compounds (SVOCs) and the adsorption of gas phase VOCs (positive artifact). Prolonged sampling likely volatilizes a large portion of fresh BSOA and BBOA, leaving behind only the highly aged, low-volatility OAs. The impact of this extended sampling duration on the reported PMF factor distributions and O/C ratios must be acknowledged as a limitation.

Prolonged filter sampling may introduce some sampling artifacts. For semi-volatile organic compounds, if their atmospheric composition and gas-particle partitioning conditions changed during the one-week sampling period, the compounds already collected on the filter may have re-equilibrated with the air subsequently passing through the filter. As a result, the collected sample may have been influenced, to some extent, more strongly by atmospheric conditions closer to the end of the sampling period. In addition, oxidation or other chemical processes may have occurred on the filter during sampling, potentially altering some reactive organic components. Therefore, compared with results obtained from shorter sampling durations, the OA composition, O/C ratios, and PMF factor distributions reported in this study may have been affected more by the sampling artifacts. This potential effect was not further evaluated in this study. This point has been added in the first paragraph of Section 2.1, Lines 131–138.

#### **5. Ambiguity of CHN Family Ions as Exclusive BBOA Tracers:**

The authors assign CHN family ions as exclusive tracers for fresh BBOA (protein-rich biomass combustion). However, the boreal forest emits significant amounts of Primary Biological Aerosol Particles and biogenic aliphatic amines, which are heavily involved in local new particle formation and also yield prominent CHN fragments. Assigning these nitrogen-containing ions purely to biomass burning without ruling out PBAP/biogenic amine partitioning, or without providing correlations with independent combustion tracers (like BC or CO), is a significant interpretative leap.

Regarding PBAPs, the present study focused on PM<sub>0.95</sub> samples, which are expected to be less influenced by larger biological particles such as bacteria, fungal spores, and pollen. This clarification has been added in Lines 464–469.

Regarding biogenic aliphatic amines, measurements at SMEAR II have shown that gas-phase organic amines

are extremely low (Hemmilä et al., 2018; Sipilä et al., 2015). Previous BVOC flux studies conducted at Hyytiälä generally reported that the dominant emitted species were terpenes (65%–75%, mainly monoterpenes with smaller contributions from isoprene), followed by oxygenated VOCs such as methanol, acetone, and acetaldehyde (Rantala et al., 2015; Petersen et al., 2023).

As discussed in the manuscript, CHN-related ions, especially  $\text{CH}_4\text{N}^+$ , can serve as a tracer ion for amines, while  $\text{C}_n\text{H}_{2n-1}\text{N}^+$  and  $\text{C}_n\text{H}_{2n-2}\text{N}^+$  are likely associated with nitriles (Ge et al., 2024). For this reason, we did not rely solely on  $\text{CH}_4\text{N}^+$ , but instead examined the relationship between an entire CHN ion family and the BBOA-like factor.

Nevertheless, we admit that the current understanding of CHN family ions remains limited. Whether they can truly serve as reliable tracers requires further validation.

### **Minor & Specific Comments:**

#### **6. Motivation for Polarity-Based Fractionation:**

The motivation for applying polarity-based fractionation would benefit from further clarification in the Introduction. While it is described as a means to improve source apportionment, it is not fully clear what additional insights this approach provides beyond conventional online AMS PMF analysis. Please explicitly explain how polarity-resolved measurements contribute to understanding the chemical processing, structure–property relationships, or atmospheric behavior of OA.

Polarity-based fractionation retains the physicochemical differences among HP-WSOM, HULIS, and WISOM, allowing PMF factors to reflect not only information related to sources but also structural features associated with fractions of different polarity. Conventional PMF analysis generally assumes that aerosols from different sources have relatively fixed compositions. However, this assumption may not necessarily apply to AMS-based PMF analysis of OA, because atmospheric aging can modify some molecular structures while preserving other structural features. Therefore, we adopted polarity-based fractionation and applied PMF independently to the fraction-resolved spectra. This approach allowed us to consider the mass spectra from different OA fractions without imposing additional constraints and to obtain PMF factors that reflect dynamic mixtures of source-related submolecular structures rather than fixed sets of organic compounds. This point has now been emphasized and clarified in the last paragraph of the Introduction (Lines 105–117).

This view is consistent with the explanation in the Summary: *“The PMF factors identified in this study are better interpreted as dynamic mixtures of submolecular structures rather than fixed sets of organic compounds. This is because OAs from respective sources would not have nearly-fixed mass spectra, given the aging of OA during transport. This may also be the case for PMF factors reported in previous conventional online studies.”* The summary part is also revised (lines 643–650).

#### **7. Lines 221-222 (P1 and P2 events):**

The manuscript highlights P1 and P2 as distinct periods because of their high HULIS concentrations. However, it is not entirely clear what makes these periods fundamentally different from the rest of the dataset. While P2 is associated with elevated absolute concentrations, the relative mass contributions of WISOM, HULIS, and HP-WSOM do not seem substantially different from adjacent periods. A more explicit discussion of both the compositional uniqueness during P1/P2 and the overall seasonal trends would improve clarity.

Although these two periods were not characterized by distinct compositions, their elevated OA concentrations deviated from the seasonal pattern. Therefore, we briefly described these periods in the context of seasonal variations. In the following section (Section 3.1.2), we mainly discuss that the increases in OA concentrations were likely influenced by the transport of air masses affected by relatively strong emission sources in Eastern Europe and western Russia.

The relative contributions of the three fractions during P1 and P2 did not show particularly distinct changes compared with adjacent periods (Fig 13a). It is now addressed in Lines 300–301.

**8. Figure S6 (Back trajectories):**

It would be highly beneficial to include time markers (e.g., 12-hour or 24-hour intervals) along the backward trajectories to better indicate transport timescales. Based on the current figure, it is difficult to clearly distinguish whether P1 reflects long-range transport and whether P2 is influenced by more local/regional stagnant air masses.

The time segments have been added to the trajectory lines in Figure S8, with a color gradient to indicate the number of days before arrival.

**9. Lines 302-320 (High-mass BSOA tracers):**

While the authors acknowledge the low abundance and potential fragmentation of these high-mass ions, it remains unclear whether the observed correlations alone are sufficient to support their use as robust characteristic signatures. Given their relatively weak signal intensity in W-mode HR-AMS, further clarification on their signal-to-noise ratios and stability across samples is needed. Providing representative HR peak-fitting plots for these ions in the Supplement would strengthen this claim.

$C_7H_9O_3^+$  and  $C_9H_{13}O_4^+$  indeed showed relatively weak signal intensities in the W-mode HR-AMS measurements. We have added examples of HR peak-fitting plots in the Supporting Information Figure S10.

**10. Line 316:** Please check whether “BBOA” should be corrected to “BSOA” in this context regarding  $C_5H_7^+$  and  $C_6H_9^+$ .

Corrected.

**11. Lines 425-430 (Temperature dependence of BSOA):**

The interpretation of the temperature dependence of BSOA requires further clarification. While BVOC emissions are generally temperature-dependent, monoterpenes (the dominant emissions in boreal pine forests) typically have a different temperature-emission relationship compared to isoprene. It would be helpful to clarify whether the observed BSOA trend is primarily driven by isoprene-derived SOA or if monoterpene-derived SOA also contributes significantly, contextualized by the dominant vegetation types at the site.

As pointed out by the reviewer, isoprene and monoterpenes have different emission mechanisms and temperature dependences. Isoprene is generally produced *de novo* in chloroplasts and emitted almost immediately, making its emission strongly dependent on light and temperature. In contrast, monoterpenes in coniferous trees can originate from both *de novo* synthesis and volatilization or diffusion from plant storage pools, allowing emissions to continue during nighttime (Niinemets et al., 2010; Tarvainen et al., 2005). However,

because our samples were collected on a weekly basis, the short-term differences in the temporal emission patterns between isoprene and monoterpenes are not expected to strongly affect the interpretation of our integrated BSOA signals. We have added a description in the final paragraph of the introduction section clarifying that monoterpenes dominate the emissions at the site.

**12. Line 452 (Date Verification):**

Please verify whether “15-22 December” should actually be “21-28 July.” Looking at Figure 12, the WISOM fraction of MO-OOA does not appear particularly elevated in December, whereas there is a massive spike during the July period.

We apologize that the legend was incorrect. It has now been corrected.

**13. Formatting:** There are inconsistencies in the formatting of subscripts and superscripts throughout the manuscript (e.g., PM<sub>0.95</sub> vs PM<sub>0.95</sub>, C<sub>5</sub>H<sub>6</sub>O<sup>+</sup> vs C<sub>5</sub>H<sub>6</sub>O<sup>+</sup>). A careful proofread is recommended to ensure consistency in their presentation.

The format has been corrected.

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## **Other changes**

In addition to the revisions made in response to the reviewers' comments, we made several minor corrections and clarifications throughout the manuscript. For example, we added a brief description of the ion chromatography method in the Methodology section, because  $K^+$  and  $SO_4^{2-}$  data were used in the discussion but the corresponding analytical procedure had not been described in the original manuscript. All the changes are indicated in the tracked-changes version of the manuscript.