Title: Measurement report: Molecular Insights into Organic Aerosol Sources and Formation at a Regional Background Site in South China

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We appreciate the reviewers' helpful comments on our manuscript. We have carefully considered each of their suggestions and questions, and revised the manuscript accordingly. In compliance with the reviewers' detailed comments, We thoroughly reviewed and updated the text, figures and references. All corrections are marked in the revised manuscript. Our responses to the reviewers' comments are provided below, with responses highlighted in **Blue** and revised sections in **Green**.

Response to Anonymous Referee #3

RC: Comments on the manuscript of "Measurement report: Molecular Insights into Organic Aerosol Sources and Formation at a Regional Background Site in South China" by Jiang et al.

High-resolution AMS (HR-AMS), thermal-desorption aerosol gas-chromatography coupled with time-of-flight mass spectrometry (TAG-TOF-MS), and high-performance liquid chromatography with high-resolution mass spectrometry (HPLC-HR-MS) were combined to systematically analyze the composition of organic aerosol (OA) from macroscopic to molecular level, breaking through the limitations of traditional methods only relying on a single technique. Combining the AMS-PMF, tracer based PMF and Non-negative matrix factorization (NMF), the source and molecular composition characteristics of organic aerosols in Hong Kong were explored. The integration of online/offline measurements and multi-method source apportionment provides good insights into OA formation mechanisms under low-PM but high-O_3 conditions. After reading through the whole manuscript, I think several clarifications and expansions are still needed to enhance reproducibility, interpretability, and contextual relevance.

AC: We thank the reviewer for providing a comprehensive summary and positive evaluation of our work. To address the reviewer's concerns, we have carefully revised the manuscript in following her/his comments and suggestions. The point-by-point responses to the reviewer's comments are listed below.

Major comments:

RC: I do not quite understand why the authors present the AMS-data like new results in the manuscript, since the results have already been presented in another independent paper by Huo et al. (2024) in Science of the Total Environment. This is quite unusual and necessary.

AC: Thank you for the reviewer's comments. We appreciate your concern and would like to clarify the following points. Firstly, although Huo et al. (2024) reported AMS results in our previous research, their sampling period (September 29 to November 18, 2020) was longer than the time window used in this study (November 7 to November 16, 2020). This discrepancy in sampling periods may lead to differences in AMS results, making previously reported AMS data and conclusions not directly applicable to the present study. To ensure consistency with the offline $PM_{2.5}$ samples analyzed here, we selected and reanalyzed only the AMS data corresponding to the time window as the offline $PM_{2.5}$ samples. Secondly, AMS data serve as a crucial foundation for our manuscript. Incorporating AMS data is essential for understanding the composition and temporal variations of OA during the sampling period, and it also provides important context for the research objectives and content of this study. Therefore, in Section 3.1 of the original manuscript, we reported AMS results that matched the offline $PM_{2.5}$ samples to maintain consistency and relevance.

In the revised manuscript, to address the reviewer's concerns, we have added detailed explanations in Section 3.1. The following is the revised content:

In Section 2.1, we have added the sampling period for the offline PM2.5 samples as follows:

"……During the campaign, 31 valid daily fine particulate matter ($PM_{2.5}$) samples were collected from November 7 to November 16, 2020, using a high-volume sampler (TE-6070, TISCH, U.S) operating at a flow rate of approximately 1.0 $m^3 \cdot min^{-1}$ (Table S1)……"

In Section 3.1, we have included the following clarification:

"It should be noted that although our previous study (Huo et al., 2024) reported AMS results, the differing time windows between the two studies may lead to different conclusions. To ensure consistency with the offline $PM_{2.5}$ samples analyzed in this study, only the AMS data corresponding to the same sampling period were selected and reanalyzed. Notably, even though the sampling campaign occurred during a period influenced by both continental and costal air masses from mainland China, the observed $PM_{2.5}$ concentrations remained relatively low……"

RC: The authors present PMF results derived from three distinct datasets; however, no definitive conclusions regarding OA sources are drawn from this comparative analysis.

AC: Thanks for the reviewer's comments. In lines 229-231 of the original manuscript, we presented PMF results based on three datasets. We emphasized the significant contribution of secondary processes to OA sources at the study site, with the combined contributions of gas-pSOA, SIA-OA, and biogenic SOA approaching 70%. This comprehensive conclusion could not be drawn from any single dataset alone. Furthermore, in the subsequent sections, we detailed the contributions of each source to AMS components and emphasized the importance of biomass combustion and cooking to HOA and LO-OOA1 , which was informed by our analysis of AMS data. We also applied PSCF analysis to identify potential source regions for different OA sources, emphasizing the role of regional trasnsport. Overall, integrating PMF results from the three datasets has substantially enhanced our understanding of OA sources and formation mechanisms at the study site.

RC: For the tracer-based PMF analysis, I would prefer to see total OA mass applied as input rather than the pre-resolved AMS-PMF factors. The findings in Section 3.2 suggest that the AMS-PMF results may be methodologically problematic, which creates confusion. It is wrong. Using total OA mass would at least enable direct comparison between PMF results derived from tracer species and AMS data.

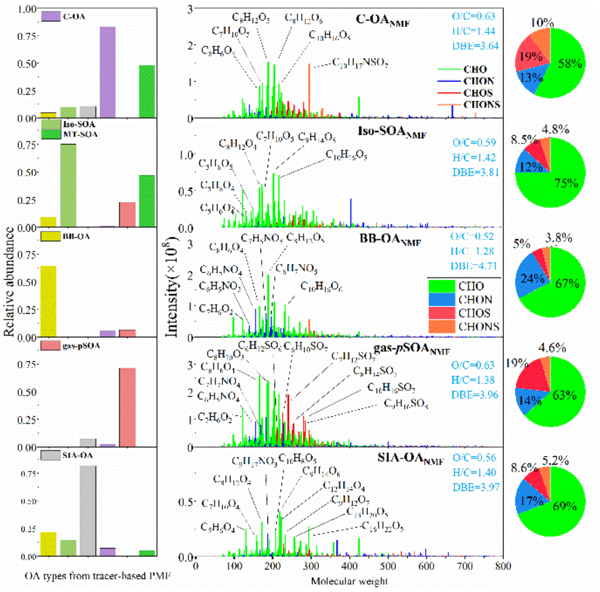
AC: Thank you for your valuable comments. **Firstly,** we agree that including total OA mass as a variable alongside chemical tracers in PMF model analysis can help separate the contributions of different sources to OA. However, doing so would result in the loss of detailed organic fragment information provided by AMS analysis, thereby diminishing the role of AMS in elucidating OA sources and formation mechanisms. **Secondly**, AMS is one of the most important tools for investigating OA sources and formation. By integrating organic fragment data from AMS with PMF analysis, OA can be effectively divided into several components. While these AMS-derived components reflect the impacts of atmospheric environmental changes on OA, the source information they provide remains limited, as noted in Lines 43-47 of the original manuscript. Therefore, a key objective of this study is to identify the sources of these AMS-components, and how atmospheric environments affected the oxidation states of these AMS components and to understand how atmospheric conditions influence their oxidation states (Lines 109-110 in the original manuscript). Without including AMS component data in the PMF model analysis, it would not be possible to obtain detailed source information for these components. **Thirdly,** although AMS components were used in the PMF analysis, we can still compare PMF results with the total OA mass measured by AMS. This does not pose a significant issue. As shown in Figure S2, the simulated concentrations from the PMF model correlate well with the measured values, both for AMS components and total OA concentration. This indicates the validity of using AMS components as input variables in the PMF model.

To address the reviewer's concern, we have added additional notes in Section 2.3. Please find the revised version below:

"The EPA PMF5.0 model was employed to determine the relative contributions of different sources to the four AMS-PMF OAs. The mathematical framework of PMF has been described in detail in previous studies (Hopke, 2016; Norris et al., 2014). In brief, we used hourly concentration data of AMS-PMF OAs and SOA tracers as input for the EPA PMF model. While incorporating total OA mass along with chemical tracers in the PMF analysis can help distinguish the contributions of different sources to OA, it does not provide detailed source information for these AMS components, nor does it reveal how atmospheric conditions influence their oxidation states ……Ultimately, a six-factor solution was adopted, which showed good correlations between modeled and measured concentrations of each AMS-PMF OA (r = 0.70-0.94), supporting the validity of using AMS components as input variables."

RC: The authors also applied PMF factors derived from the tracer method to the NMF analysis of molecular composition. The x-axis label for these factors ('AMS-PMF-OA') appears inconsistent with standard terminology – please verify whether this is a typographical error. Additionally, clarification is needed regarding how the percentages of PMF factors contributing to each NMF factor (shown in Figure 4) were quantitatively defined. So, what is the fraction of NMF factor to the total OA?

AC: Thank you for the reviewer's valuable comments and suggestions. To better elucidate the impact of each factor on OA formation at the molecular level, we applied NMF model analysis to integrate the molecular composition data obtained from offline high-resolution mass spectrometry with the factors identified in the tracer-based PMF analysis. By constraining the NMF model with the PMF factors, we were able to determine the molecular composition associated with each factor. In the revised manuscript, we have redrawn Figure 4 accordingly, and updated the x-axis label. Please review the changes.



It should be noted that in the revised manuscript, Section 2.4 now includes two additional sentences clarifying how the percentages of PMF factors contributing to each NMF factor (as shown in Figure 4) were quantitatively defined. However, the fractions of NMF factors relative to the total OA were not determined, as the total OA concentrations were not used as the total variable in the NMF analysis. Instead, we calculated the total intensity fraction of each NMF factor relative to the total molecular intensity, and these results are presented in Figure 5. The detailed changes can be found in Section 2.4. The revised version is provided below:

"……Specifically, the contribution matrix of PMF factors was combined with the intensity-normalized data of HR-MS molecules to form a comprehensive input matrix, **X**.……The function of "nnmf" decomposes the input matrix **X** into two non-negative matrices, **G** and **F**, by minimizing the root-mean-squared (rms) residual (E) between **X** and its approximation, **GF**. Through this approach, both the percentages of PMF factors contributing to each NMF factor and the intensity fraction of each NMF factor relative to the total molecular intensity can be derived from the reconstructed matrix of **GF**."

RC: The analysis relies predominantly on qualitative descriptions without sufficient quantitative validation. For instance, Line 341 states: 'The NMF-IsopreneSOA exhibited an O/C ratio similar to that of NMF-cooking but had the highest effective oxidation number (nOeff) values among all NMF-factors.' However, no actual O/C ratio values are provided to substantiate these claims. This omission undermines the methodological rigor and obscures critical differences between factors.

AC: Thanks for the reviewer's comments. In Line 325 of the original manuscript, we stated that "Figure 4 presents the mass spectra of each factor obtained from the constrained-NMF analysis, while Table S2 displays the average chemical parameters." First, we apologize for the error in the table reference; it should be Table S4, not Table S2. Additionally, please note that the average chemical parameters for each NMF factor included elemental ratios, DBE values, and aromatic index (AI). Other chemical parameters related to oxidation state (nO_{eff}), aromaticity (Xc), and volatility (LogC*) were also presented. Since we did not refer to Table S4 again in subsequent sections, this may have caused confusion regarding the discussion of these parameters. In the revised manuscript, we have corrected the reference from "Table S2" to "Table S4" to address this issue.

"Figure 4 presents the mass spectra for each factor obtained from the constrained-NMF analysis, with several highly intense species also indicated. As shown in Table S4, which summarizes the average chemical parameters of each NMF factor, the C-OA$_{\mathrm{NMF}}$ was featured by the highest O/C ratio and the lowest DBE value……"

We have also added the average O/C and H/C ratios, as well as DBE values, to Figure 4 to help readers better understand the compositional differences among the various NMF factors. Additionally, we have included references to Table S4 in Lines 365 and 371 of the revised manuscript. Please review these changes in the corresponding sections of the revised manuscript.

RC: Line 344"a series of C5 oxygen-containing compounds also displayed high intensity in NMF IsopreneSOA." What is the intensity?

AC: Thanks for the reviewer's comments. In Figure 4, we presented the mass spectra for each factor obtained from the constrained-NMF analysis. Also, Figure 4 shows the percentages of different compound groups, as well as several of the highest-intensity species for each NMF factor. Thus, the intensity of the C5 oxygen-containing compounds displayed in Iso-SOA$_{\mathrm{NMF}}$ factor can be found in Figure 4.

In the revised manuscript, we have updated this sentence and added a reference to Figure 4:

"……In addition to several highly intense monoterpene SOA species, a series of C_5 oxygen-containing compounds, such as $C_5H_{6\text{-}8}O_4$ and $C_5H_{6\text{-}8}O_5$, also displayed high intensity in the Iso-SOA$_{NMF}$ factor (Figure 4)……"

RC: Line 355 "The NMF-gas-pSOA factor contained a high abundance of aromatic-CHO compounds" What is the high abundance?

AC: Thank you for the reviewer's comments. As mentioned above, we have presented several of the highest-intensity species for each NMF factor in Figure 4. Some aromatic-CHO compounds with high intensity in the gas-pSOA$_{NMF}$ factor, including $C_7H_6O_2$ and $C_8H_6O_4$, are highlighted in the figure. We did not include the detailed abundance data for these compounds in the main text, as reporting these specific values would not provide additional useful information. However, in the revised manuscript, we have added a reference to Figure 4 in the relevant section.

"The gas-*p*SOA$_{NMF}$ factor contained a high abundance of aromatic-CHO compounds, nitroaromatic compounds, and CHOS compounds (Figure 4)……"

RC: The constrained NMF approach (Section 2.5) is innovative but requires clearer justification for selecting 5 factors. Elaborate on how "99% variance explained"

AC: Thank you for the reviewer's comments and suggestions. In this study, the details of the constrained NMF approach are provided in Section 2.4. In Lines 136-140 of the original manuscript, we described the method used to determine the optimal number of factors. In the revised manuscript, we have further clarified this section. The revised version is as follows:

"……To determine the optimal number of factors, we plotted the variation of RMSE against the number of factors and selected the optimal value based on the Elbow rule (i.e., the point at which the RMSE reaches a minimum). We also calculated the global relevance between X and GF, following the method described by Rund et al. (2023), to identify the number of factors that best explain the variance. Furthermore, consensus cluster analysis was performed using the R package "ConsensusClusterPlus" (Monti et al., 2003) to further support the selection of the appropriate number of factors. By integrating all these evaluation criteria, we determined that five-factor was the optimal choice, as this was the smallest number that explained 99% of the variance in the input data matrix (Figure S3)……"

Minor comments

RC: The full name of the instrument should be provided upon its first mention, such as EESI- and CI-TOF-MS and BVOCs……..

AC: Thank you for the reviewer's reminder. In the revised manuscript, we have provided the full names for EESI- and CI-TOF-MS in line 39. Additionally, we have thoroughly checked the manuscript to ensure that the full terms are given when abbreviations first appear.

RC: Line 116: For the input matrix of NMF, is it the intensity matrix detected by HRorbitrap-MS combined with the mass concentration matrix of six factors? Whether the order of magnitude difference between the two values affects the NMF results?

AC: Thanks for the reviewer's comments. As mentioned above, the contribution matrix of the PMF factors was combined with the intensity-normalized data of the HR-MS molecules to form the complete input matrix X. Using the normalized data matrix does not affect the NMF results; rather, it improves the stability of the model analysis.

RC: Line 160: Whether the PM1 mass concentration measured by AMS is accurate, whether it has been calibrated by pure ammonium nitrate, ammonium sulfate and other pure particulate matter samples, as well as IE and RIE correction. As you write that "We applied relative ionization efficiencies (RIE) of 1.4, 1.1, 1.2, 4.0, and 1.3 to calculate the concentrations of total organics, nitrate, sulfate, ammonium and chloride, respectively.", you used default IE and RIE. Please verified the PM1 mass concentration and the OA contribution.

AC: Thanks for the reviewer's comments. We would like to clarify that both at the beginning and throughout the sampling process, the instruments were regularly calibrated using pure chemical standards, such as ammonium nitrate, as recommended by the manufacturer. Typically, calibration of the ionization efficiency (IE) for m/z 30 and m/z 46 was performed using pure ammonium nitrate particles (350 nm in diameter). Based on weekly IE calibrations, the relative ionization efficiency (RIE) value for ammonium was determined to be 4.0, which was subsequently applied in data processing. For other species, default RIE values were used according to the recommendations on the Jimenez group's webpage. In addition, particle velocity during sampling was calibrated using Nanosphere PSL particles of various sizes (50, 100, 200, 300, 400, 500, and 600nm; Duke Scientific, Palo Alto, CA, USA). The collection efficiency (CE) of NR-PM1 was determined by comparing AMS NR-PM1 measurements with PM1 values from HKEPD (after subtracting black carbon). The online instruments used by HKEPD for PM_1 and black carbon measurements were a Tapered Element Oscillating Microbalance (TEOM) and a black carbon analyzer (BC, model AE16, Magee, USA), respectively. A CE coefficient of 0.73 was determined and applied to all measured NR-PM_1 components. Through these calibration procedures, we are confident that the PM1 mass concentrations and OA contributions measured by AMS are accurate.

In the revised manuscript, we have added further details on these calibration processes in the supplemental text (**HR-TOF-AMS analysis section**) to address this concern.

"……To ensure the accuracy of NR-PM_1 measurements, the instruments were regularly calibrated with pure chemical standards, such as ammonium nitrate, both before and during the sampling period. Pure ammonium nitrate particles (350nm in diameter) were applied to calibrate the ionization efficiency (IE) for m/z 30 and m/z 46. Based on weekly calibrations, the relative ionization efficiency (RIE) value for ammonium was determined to be 4.0. In addition, particle velocity during sampling was calibrated using Nanosphere PSL particles of various sizes (50, 100, 200, 300, 400, 500, and 600nm;

Duke Scientific, Palo Alto, CA, USA). The collection efficiency (CE) of NR-PM1 was determined by comparing AMS NR-PM1 measurements with PM_1 concentrations measured by HKEPD, after subtracting black carbon at the same location. PM1 and black carbon concentrations from HKEPD were obtained using a Tapered Element Oscillating Microbalance (TEOM) and a black carbon analyzer (BC, model AE16, Magee, USA), respectively. The final CE value for this study was determined to be 0.73 and was applied to all measured NR-PM_1 components."

RC: What is the basis for the error score set in the PMF model (0.2 for OA and 0.3 for n-alkanes)? If the error parameter is adjusted, what is the impact on the source analysis result?

AC: Thank you for your good questions. As noted in a previous study (Wang et al., 2017), PMF modeling typically adopts a fixed error fraction for all data points from a given species and employs the default robust mode. In robust mode, PMF treats occasional high concentration data as "outliers" (defined as scaled residuals >4) and down-weights them during the iteration process by replacing their scaled residuals with a value of 4. This approach can prevent the model from accurately reproducing extremely high concentrations in otherwise low-variance time series. Furthermore, the concentrations of species used in PMF analysis often vary by orders of magnitude, especially when organic molecular markers are analyzed alongside inorganic ions and other substances. Such differences can compromise model robustness and increase uncertainties. To address this issue, Wang et al. (2017) proposed an equation to determine appropriate error fractions for different types of species in PMF analysis. In this study, we applied error fractions of 0.2 for OA and 0.3 for n-alkanes, following the approach of (Wang et al., 2019). These values have also been validated in our previous research to improve the robustness and accuracy of PMF results (Jiang et al., 2021).

To address this concern, we have added explanatory notes to the relevant section in the revised manuscript. The revised text is as follows:

"……Given that the concentrations of species used in PMF analysis can vary by orders of magnitude, applying the default error fraction may lead PMF to treat high concentration data as outliers, thereby reducing model robustness and increasing uncertainty in the results (Wang et al., 2017a). To address this issue, we adopted the error fraction of 0.2 for OA components and polar SOA tracers, and 0.3 for n-alkanes (Wang et al., 2019)……"

RC: Figure 1&2: Please unify the number of decimal places in the pie chart

AC: Thank you for your reminder. In the pie charts of Figures 1 and 2, all values are presented with two significant digits, rather than being rounded to a specific number of decimal places.

RC: Line 205: The instructions for the figures in the manuscript are not clear enough.

AC: Thank you for your question. To address this, we have added a reference to Figure

2 in the relevant section of the revised manuscript:

"……Figure 2c-i also presents the diurnal variations of OA concentrations contributed by each PMF factor……The OA concentrations from these factors exhibited similar diurnal patterns, peaking in the morning, declining during the daytime, and rising again in the evening (Figure 2c-d)……"

RC: Line 202&224: Does "secondary inorganic aerosols (SIA)" represent the SIA-derived OA or secondary inorganic aerosols, and if the latter, how to understand the contribution of SIA to OA concentration. It is suggested to rename the tracer-PMF factors, such as Biomass-burning derived OA (or BB-related OA as you write in Line 246), cooking derived (or related) OA, SIA derived OA.........., to better understand. Also, please distinguish between AMS-PMF factors and tracer-based PMF factors in the manuscript, otherwise it is easy to confuse the meaning.

AC: Thanks for the reviewer's reminder. The phrase "secondary inorganic aerosols (SIA)" refers to the association between the formation of certain SOA and the generation of SIA through gas-phase oxidation, or to cases where SIA directly participates in SOA formation. In accordance with the reviewer's suggestion, we have renamed the tracer-PMF factors as follows: "BB-related OA (BB-OA), cooking-related OA (C-OA), isoprene-derived SOA (Iso-SOA), terpene-derived SOA (MT-SOA), secondary inorganic aerosol-related OA (SIA-OA), and gas-phase photochemical SOA (gas-pSOA)."

In this study, we prefer to the factors derived from AMS-PMF as AMS-components to distinguish them from the tracer-based PMF factors. We have recognized and carefully addressed this distinction throughout the manuscript.

RC: Figure 2b: How is the proportion of tracer-based PMF factors (EPA) in the AMS-PMF factor calculated?

AC: Thanks for your valuable comments. As described in Section 2.3 of the original manuscript, we used hourly concentration data of AMS-PMF OAs and SOA tracers as input for the EPA PMF model. For each tracer-based PMF factor, the concentrations of AMS-PMF-derived OAs were obtained from the EPA PMF analysis. Thus, the proportion of each tracer-based PMF factor within an AMS-PMF factor was calculated by dividing the concentration of the tracer-based PMF factor by the corresponding total concentration of the respective AMS-PMF OA.

RC: Line 246: The backward trajectory of the air mass at 120h shows that BB-related OA comes from Henan and Shanxi provinces, as well as Korean Peninsula, then what is the corresponding contribution approximately and whether it is meaningful to discuss. Such a long transport distance, whether BB-related OA has been aged during long-range transport, but the result shows BB-related OA has little contribution to MO-OOA but contributes most to HOA (Figure 2b).

AC: Thank you for your excellent comments. In this study, we assessed the potential influence of regional transport on OA composition during the sampling campaign using backward trajectory analysis, and evaluated possible source regions of PMF-derived factors through potential source contribution function (PSCF) analysis. In Figure S8, the contributions of source regions to total BB-related OA are indicated by gradient colors. Based on the PSCF results, we identified Henan and Shanxi provinces, as well as the Korean Peninsula, as major source regions for BB-related OA. While further discussion on this topic would be interesting and meaningful, it is beyond the scope of this manuscript. Therefore, we have limited our discussion to noting that straw burning was an important source of PM during the harvest season in these regions, as supported by satellite fire maps showing intense fire activity.

We agree with the reviewer that long-term transport can lead to the aging of BBOA. Nonetheless, our results suggested that BB-related OA contributed little to MO-OOA, but was a major contributor to HOA. Several factors may explain this observation. First, the sources and secondary atmospheric processes affecting OA at the sampling site were complex, with biomass burning accounting for only about 20% of total OA concentrations. In addition to long-range transport from northern China, nearby biomass burning in the PRD region might also contribute to OA. Since long-range transport could lead to the loss of BBOA, its contribution at the sampling site might not be substantial. Second, although BB-related OA had limited contribution to MO-OOA, it was the main contributor to LO-OOA. The aging of BBOA during transport might not lead to a high degree of oxidation compared to locally formed SOA, the high oxidation capacity of the local atmosphere. Third, BBOA concentrations were determined using levoglucosan, a relatively stable organic tracer with a long atmospheric lifetime, indicating that the measured BBOA was primarily associated with stable primary OAs. More highly oxidized compounds resulting from atmospheric processing might have been assigned to other PMF factors, such as SIA-OA or gas-pSOA.

RC: Line 255: Please add reference to support the explanation.

AC: Thank you for your reminder. We have added a reference to support our explanation, as suggested. Please see the corresponding sections in the revised manuscript for details.

RC: Are 10,012 unique molecular formulas detected in daily samples or after screening under other conditions.

AC: Thanks for the good comment. For high-resolution mass spectrometry data, after background noise subtraction and blank correction, we detected thousands of compounds in each daily sample. The 10,012 unique molecular formulas reported here represent the total number of distinct molecular formulas identified across all samples in our dataset; that is, the cumulative number of unique formulas detected in all samples does not exceed 10,012.

In the revised manuscript, we have rephrased this sentence for clarity as follows:

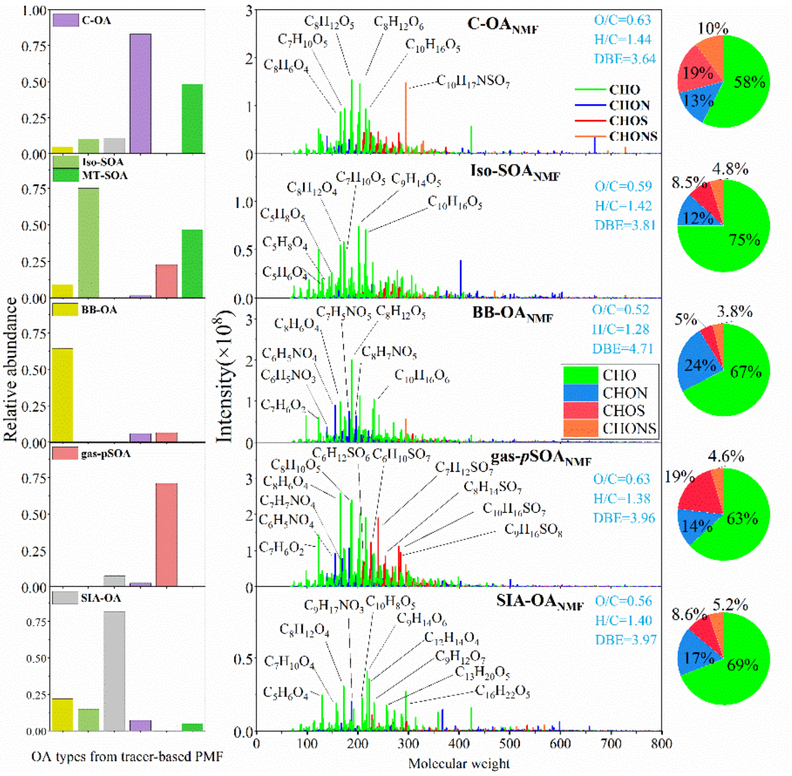
"……A total of 10,012 unique molecular formulas were identified across all samples, among ……"

RC: Line 339: "C10H16NSO7" does not match the C10H17NSO7 in the Figure 4. Please check it.

AC: Thank you for your careful review. We have corrected the typo in Line 339 and it now reads "$C_{10}H_{17}NSO_7$".

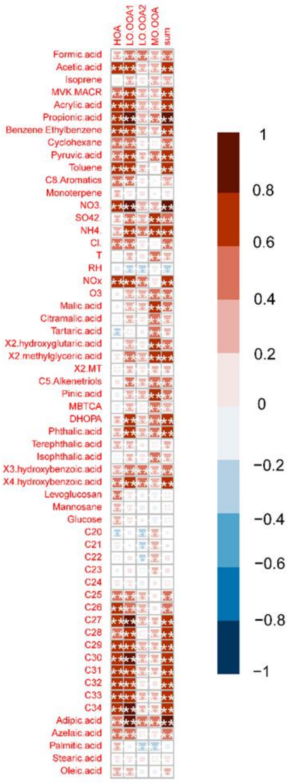
RC: It is suggested that the average O/C ratio, H/C ratio, and DBE can be annotated on the mass spectrogram in the Figure 4.

AC: Thanks for the reviewer's suggestion. In the revised manuscript, we have added the average O/C ratio, H/C ratio, and DBE values for each NMF factor to the mass spectra in Figure 4. The revised version is as follows:



RC: Figure S5, the legend for the correlation coefficient was missing.

AC: Thank you for your careful review. We have added the missing legend for the correlation coefficient in Figure S5. Please refer to the revised figure in the revised manuscript.

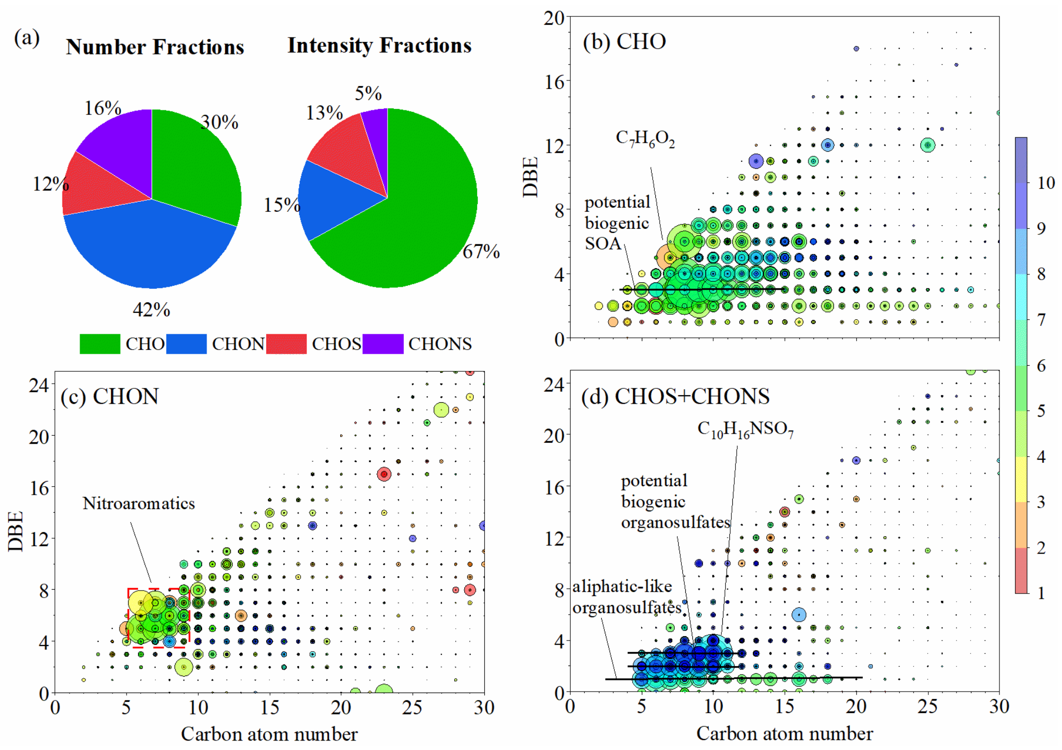


RC: Does the inclusion of PMF factors as constraints in NMF analysis potentially lead to misjudgments of some molecular sources?

AC: Thank you for your valuable comments. I believe that including PMF factors as constraints in the NMF analysis will not lead to misclassification of molecular sources, provided that the PMF factors themselves are accurate. In fact, as demonstrated in Section 3.4, we have presented multiple results supporting the validity of this approach. For example, in the BB-OA$_{\mathrm{NMF}}$ factor, many nitroaromatic compounds previously observed in BBOA samples were successfully assigned to this factor. However, since NMF, like PMF, is fundamentally a mathematical allocation method, there is possibility of misallocation under certain circumstances. To address this, we employed several approaches to verify the reliability of the NMF model results in our study. Therefore, we consider the results from the constrained NMF model analysis to be robust and acceptable.

RC: Legend for Fig. 3 needs to be revised.

AC: Thank you for your careful review. We have corrected the legend for Figure 3. Please see the revised figure below.



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