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' valuable comments on our manuscript. We have carefully considered their detailed comments and addressed all suggestions and questions. In accordance with the reviewers' recommendations, we have thoroughly revised the manuscript, including the text, figures, and references. All corrections are clearly marked in the revised manuscript, with responses highlighted in **Blue** and revised parts in **Green**. Our point-by-point responses are provided below.

Response to Anonymous Referee #2

RC: In this study, Jiang et al. utilized advanced analytical techniques, including high-resolution AMS, TAG-TOF-MS, and HPLC-HR-MS, to investigate the composition, sources, and evolution of OA in Hong Kong. Their findings reveal that during the COVID-19 lockdown, biomass burning and gas-phase secondary organic aerosols (gas-pSOA) were the primary contributors to low-oxidation OA, influenced by air mass transport from mainland China. In contrast, secondary inorganic aerosols (SIA), gas-pSOA, and biogenic SOA accounted for over 90% of high-oxidation OA, with isoprene-derived SOA emerging as the dominant contributor to the high oxidation levels. HR-MS analysis showed that CHO and CHON compounds were key components of OA, accounting for more than 60% of the total intensity and over 40% of molecular formulas, respectively. This indicates that atmospheric oxidation processes led to the accumulation of specific CHO compounds, while nitrogen addition reactions increased the diversity of the molecular composition.

A key innovation of this study lies in addressing the challenges of interpreting factors in PMF analysis due to the complexity of atmospheric chemical processes and the diverse sources of OA. By integrating molecular composition data into a non-negative matrix factorization (NMF) model constrained by PMF factors, the study improved the interpretability of these factors and shed light on the molecular composition differences among sources and formation pathways. Overall, this research employed robust analytical methods, provided comprehensive discussions, and delivered novel insights into the sources, formation mechanisms, and regional air pollution associated with OA. The manuscript aligns well with the journal's scope, and I recommend its acceptance following minor revisions.

AC: We appreciate the reviewer for his/her positive feedback and constructive comments, which have greatly helped to improve the quality of our manuscript.

Specific comments

RC: Line 43: This sentence is not well organized. Please rephrase them.

AC: Thanks for the reviewer's suggestion. We have rephrased this sentence as follows:

"……The tracer-based PMF often identifies ambiguous SOA factors (e.g., secondary inorganic aerosols) with uncertain chemical mechanisms and source contributions (Reff et al., 2007; Wang et al., 2017a; Lyu et al., 2020)……"

RC: Line 159: Capitalize the first letter "yet".

AC: We have corrected the word "yet" into "Yet" in the revised manuscript. Please verify this change in the updated version.

RC: Line 176-178: This long sentence is suggested to be broken down into multiple short sentences to enhance the logic of the sentence ".

AC: Thanks for the reviewer's comment. We have rephrased this sentence as follows:

"……In addition, photochemical reactions involving anthropogenic emissions have been shown to play a particularly important role in the formation of LO-OOA1, whereas MO-OOA is more influenced by biogenic emissions. During the focused sampling period in this study, LO-OOA1 exhibited strong associations with gas-phase photochemical reactions involving both biogenic and anthropogenic VOCs species, as well as NOx (Figure S5)……"

RC: Line 179: "Good correlations between MO-OOA and O3…", please list what is their correlation coefficient (R2).

AC: Thank you for the reminder. We have added p and r values to this sentence as follows:

"……In contrast, MO-OOA was more closely associated with SOA formation processes driven. by ozone oxidation of BVOCs, as evidenced by strong correlations between MO-OOA and both O_3 and biogenic SOA tracers (r>0.50, p<0.01)……"

RC: Line 259-260: The subject of the sentence is long and begins with "Although", making the logic seem complicated. Please simplify it.

AC: Thanks for pointing out this. We have rephrased this sentence as follows:

"The integration of molecular tracers with AMS analysis provides an effective approach for elucidating the sources of OA, emphasizing the significant role of SOA processes in OA formation in South China. However, the molecular transformation mechanisms underlying the PMF-resolved factors, particularly for SOA, remain unclear……"

RC: Line 263: The word "formular" is misspelled; it should be "formula".

AC: Thank you for the reviewer's reminder. In this line, we stated that "CHO compound accounted for 30 ± 2.7% of formular number", where the word should function as an adjective. Therefore, "formular" is the appropriate term in this context.

RC: Line 316: "might not entirely belong" This phrase is vague.

AC: Thank you for the reviewer's suggestion. We have revised this sentence for clarity. The updated version is as follows:

"……The compounds identified in each NMF factor may not fully correspond to the names of the PMF factors, as PMF factors represent not only specific sources but also indicate that certain compounds may share similar formation pathways with the atmospheric processes implied by the factor names. For example, in the Iso-SOA$_{NMF}$ factor, many anthropogenic-related compounds might also be present alongside biogenic SOA, suggesting that these anthropogenic compounds could have similar atmospheric formation pathways as biogenic SOA."

RC: Line 370: Please specify what constraints were applied in the NMF model.

AC: Thank you for the reviewer's suggestion. In fact, as stated in Line 130 of the original manuscript, we emphasized that the daily concentration data for the six factors obtained from the tracer-based PMF model were incorporated into the NMF analysis as constraints. This approach greatly improved the interpretability of NMF factors, which included thousands of formulas, and helped elucidate the molecular formation mechanisms underlying the PMF factors. In the revised manuscript, following the reviewer's suggestion, we have added a note to enhance clarity for readers:

"Figure 5 illustrates the relative abundance of molecules identified by the PMF factor-constrained NMF method as a proportion of the total identified compounds……"

RC: Line 391-392: The phrase "IsopreneSOA being dominant contributor" is missing the article "a" before "dominant contributor." It should read: "IsopreneSOA being a dominant contributor..." for grammatical accuracy.

AC: Thank you for the reviewer's reminder. We have added the article "a" to the phrase, so it now reads: "……with Iso-SOA being a dominant contributor to the high oxidation level of OA."