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.

Major comments:

1 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.

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

3 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.

4 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

5 So, what is the fraction of NMF factor to the total OA?

6 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.

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

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

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

Minor comments

8. The full name of the instrument should be provided upon its first mention, such as EESI- and CI-TOF-MS and BVOCs……..
9. Line 116: For the input matrix of NMF, is it the intensity matrix detected by HR-orbitrap-MS combined with the mass concentration matrix of six factors? Whether the order of magnitude difference between the two values affects the NMF results?
10. 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.
11. 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
12. Figure 1&2: Please unify the number of decimal places in the pie chart
13. Line 205: The instructions for the figures in the manuscript are not clear enough.
14. 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.
15. Please distinguish between AMS-PMF factors and tracer-based PMF factors in the manuscript, otherwise it is easy to confuse the meaning.
16. Figure 2b: How is the proportion of tracer-based PMF factors (EPA) in the AMS-PMF factor calculated?
17. 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).
18. Line 255: Please add reference to support the explanation.
19. Are 10,012 unique molecular formulas detected in daily samples or after screening under other conditions.
20. Line 339: "C10H16NSO7" does not match the C10H17NSO7 in the Figure 4. Please check it.
21. 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.
22. Figure S5, the legend for the correlation coefficient was missing.
23. Does the inclusion of PMF factors as constraints in NMF analysis potentially lead to misjudgments of some molecular sources?
24. Legend for Fig. 3 needs to be revised.