

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

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Editor's comments

I have taken into consideration all of the reviews and the authors' response. Two of the reviews were quite supportive, whereas one was more critical of the work, requesting major corrections. However while I feel that some of the concerns would be considered objectively valid for a research article, because this is a measurement report, there is less of a requirement for novel and evidenced scientific insight. In the spirit of this paper presenting novel molecular data in a chemically rich environment, I am satisfied that this is publishable as a measurement report in its current form.

The only modification I ask for is to the abstract because it current conform to ACP's current style guides. I would ask that the authors, where possible, identify a knowledge gap that this dataset addresses, and/or explicitly state the novel aspects of this dataset.

Response: Thank you very much for your work on our manuscript. We greatly appreciate the editor for recognizing the merits of our work and satisfying with the modifications we made. We also thank the editor for providing valuable suggestions on the abstract.

In the revised manuscript, we have modified the abstract as possible to identify the knowledge gap, and to state the novelty of this work. For example, in the first line of abstract, we have added a sentence to state the shortcoming of current OA analysis using a single mass spectrometer. Then, to stress the novelty of this work, we state that this study combined the several mass spectrometers on OA characterization for the first time. In the following parts, we have changed some expressions of the major finding to make them as concise as possible. In the last sentence, we have added two short sentences to emphasize the significance of this research and highlight the novelty of this work. The updated abstract is:

“Understanding the chemical composition and quantifying the sources of organic aerosols (OA) are crucial for assessing their formation and human-related effects, yet individual mass spectrometry techniques still struggle to reveal the effects of sources and atmospheric processes on OA composition at the molecular level. In this study, we combined for the first time a high-resolution time-of-flight aerosol mass spectrometer (AMS), a thermal-desorption aerosol gas-chromatograph-mass spectrometer (TAG-MS), and an electrospray ionization high-resolution orbitrap mass spectrometer (HR-MS) to analyze OA at a background site in South China from bulk to molecular levels. Positive matrix factorization (PMF) analysis based on AMS data and organic tracers from TAG-MS showed that the low-oxidized oxygenated OA (LO-OOA1) was mainly contributed by biomass burning-related OA (BB-OA) and

gas-phase secondary OA (gas-*p*SOA), while the high-oxidization degree of more-oxidized oxygenated OA (MO-OOA) was mainly due to isoprene-derived secondary OA. Using a non-negative matrix factorization (NMF) approach constrained by PMF source contributions to offline HR-MS data, we identified molecules associated with each PMF-resolved factor. The NMF resolved cooking-related OA (C-OA_{NMF}) factor exhibited the highest O/C ratio but the lowest double bond equivalent (DBE) value, whereas the BB-OA_{NMF} factor was characterized by the greatest aromaticity and a high abundance of nitroaromatics. For secondary processes, sulfur additions played a more significant role in gas-*p*SOA than in secondary inorganic aerosol-related OA (SIA-OA). Overall, this study enhances our understanding of the sources and formation of different AMS components, reveals the impact of different sources on the molecular composition of OA, and underscore the prominent impact of anthropogenic emissions and their photo-oxidation on ambient OA in areas with low particulate matter pollution but high O₃ levels.”

Please noted that in this revision, we have also checked our manuscript carefully for typos, missing co-authors and their affiliations, terminology, updates of data in tables, or updates of variables in equations. All the modifications can be found in the track-changed version, please check it.