

Mass spectral characterization of secondary organic aerosol from urban cooking and vehicular sources emissions by Zhu et al.

This research work has been submitted for consideration as a “research article”. In an earlier review cycle, I summarized the importance of the work, and identified several areas for the authors to improve on. The authors have made significant improvements to the quality of the manuscript. They have also supplied additional information that needs some minor revisions. Most importantly, I suggest the authors acknowledge limitations with regards to their analytical approach (lack of separation of POA in HOA experiments, lack of measurement/analysis of time series tracers). **I recommend publication after minor revisions.**

1. Lines 85-95: “Each dish was continuously carried out 8 times in parallel during the cooking process until the closed kitchen was full of fumes... Each sampling was in parallel three times.” If eight parallel experiments were conducted, why was sampling conducted in parallel only thrice?
2. Lines 94-95: “Each sampling was in parallel three times “. What does this sentence mean? The setup in Zhang et al., 2020 suggests there is only one Go: PAM so, how are the authors doing parallel sampling? Address this issue by either removing the sentence or explaining this sentence in more detail here.
3. Line 95: “The relative standard deviations were small, which were under 10% in most cases.” What is this relative standard deviation of? SOA yield? Either remove this sentence or clarify what specifically are you referring to? Also, add a table in the supplement showing these results. If the table is already present, add a reference to that table here.
4. Lines 102-103: “the combination of 1500 rpm rotating speed and 16Nm torque, 2000rpm, and 16Nm torque for the engine in this study reflect the realistic vehicle speed of 20km/h and 40km/h” Remove comma before and after 2000 rpm and add an “and” before “2000 rpm”.
5. Lines 109-111: “For each running condition, five parallel experiments were conducted (**Table S2**). The sampling time with collecting three parallel data groups was about 60 min for each experiment.” If five parallel experiments were conducted, why were only three parallel data groups collected?
6. In author response lines 7-8, the authors state: “The source characteristics of POA were uncertain due to the low concentration of particulate matter emitted from the engine in this study.” Can you add a table on the yields in all experiments to the supplement? And reference it wherever you must justify not using the POA from engine experiments.
7. Lines 150-151: “In this study, we adopted the toolkit SoFi (Source Finder) within a-value approach to perform organic HR-AMS datasets collected in Shanghai.” “within” should be replaced by “with an”.
8. Lines 257-259: “The cooking PMF POA of four Chinese dishes all showed 258 obvious hydrocarbon-like signals at m/z 41, 43, 55, and 57 with ion fragments of C₃H₅⁺, C₃H₇⁺, C₄H₇⁺, C₄H₉⁺, C₅H₇⁺, and C₅H₉⁺.” Add in m/zs for C₅H₇ and C₅H₉ and the word “respectively” at the end of the sentence.

9. Lines 261-262: “For mass spectra of cooking PMF SOA, the oxygen-oxidation ion fragments had higher signals than those of hydrocarbon-like ion fragments.” The phrase “oxygen-oxidation” should be replaced by “oxidized”.
10. Lines 265-274: The authors repeatedly refer to PMF LO-SOA in this paragraph. I think that factor should be referred to as “vehicle PMF LO-SOA” everywhere for clarity. Similarly, PMF MO-SOA should be referred to as “vehicle PMF MO-SOA”.
11. Line 270: “The fraction of m/z 43 of PMF LO-SOA was higher than that in cooking SOA by a factor of 2.” Could this not be a consequence of the inability to separate a POA factor in this analysis? While I do not suggest authors conduct another analysis (constrained PMF for vehicle experiments by constraining the presence of POA) at this point, I recommend that the authors point out that this observation “points to the issues caused by the inability to separate vehicle POA profile in the PMF analysis. Future work could address this limitation by applying constrained PMF techniques such as ME-2 for extraction of SOA profiles from experimental data (instead of only applying constrained techniques on ambient data.” The inability to separate POA in vehicle experiments is a major limitation of this work and should be added to the “Limitations and future work” section.
12. Lines 350-351: When asked to provide information on external tracers (since lack of external tracers can generate factor mixing/lead to spurious factors (Ulbrich et al., 2009)), the authors have provided mass spectral tracer for cooking (m/z 98) (Table S21). Can you provide references for this tracer you have used? Why is this mass spectral tracer sufficient to justify cooking POA in the absence of time series tracers? Additionally, there are no time series tracers for other-POA either. The authors report a factor associated with biomass burning (other POA), and report BC measurements (Fig. S21). However, the authors do not apply the Sandradewi model to extract the fossil fuel and wood burning components of BC to use as a tracer for this other POA factor or the HOA factors. I suggest incorporating these components of BC in the analysis. The authors may choose to not perform additional analysis, but explicit addressal of this limitation (lack of external tracers) would be needed. While the authors tangentially touch on the associated factor mixing in lines 325-327, I suggest adding this as a limitation in the limitations section, clearly stating that “measurements of time series tracers for all PMF factors should be conducted in future work to avoid factor mixing. For example, significant mixing can be expected in the factors of COA, HOA, and Other POA in this work. This will affect absolute percentages of factor fractions reported in this work”.
13. Fig. S21: The authors report BC measurements but do not report instrumentation to measure BC in the Methods section. This should be corrected.
14. In Tables S19, S21, and S22, I suggest authors show correlations of all time series tracers with all PMF factors. Correlations are relative, not absolute, and in the current presentation of these tables, there is no way to check that aspect. Correlations with all external tracers used (CO, SO₂, NO_x, sulfate, nitrate, chloride, ammonium, and BC) should be reported here. The authors should not selectively present “good” data.
15. Lines 302-303: “Constraining many SOA factors could be over-constraining the ME-2 runs, which leads to factor mixing and reduces the number of factors.” Move this sentence to the section on limitations. Link it to sentences related to discussion based on (12) above.

16. Change in title: In the earlier iteration, I had suggested that “lifestyle sources emissions” be replaced with “urban cooking and vehicular sources” in the title and throughout the revised manuscript. The authors mistakenly replaced “lifestyle sources” only. Please make the correction. The word “emissions” is not necessary here.
17. In minor comments 12, 17, and 20, the authors obtained two vehicle SOA factors and attributed that observation “to higher OH exposure” (compared to cooking experiments, I am assuming). However, the EPA and OH exposures for vehicle experiments are not that different from cooking experiments (Table S3). Thus, higher OH exposure is not a sufficient explanation for lack of separation of a POA factor for vehicular experiments. I suggest removing the following sentence from the text on lines 265-266, “Different from the cooking, two-vehicle PMF SOA factors were derived from aged HOA due to higher OH exposure”. In addition, refer to discussion in (11) above to add this absence of a POA factor to the limitations section. Again, this absence is not a limitation of PMF itself, but the choice of the authors to not use constraints on experimental data.

References:

- Ulbrich, I.M., Canagaratna, M.R., Zhang, Q., Worsnop, D.R. and Jimenez, J.L., 2009. Interpretation of organic components from Positive Matrix Factorization of aerosol mass spectrometric data. *Atmospheric Chemistry and Physics*, 9(9), pp.2891-2918.
- Zhang, Z., Zhu, W., Hu, M., Wang, H., Chen, Z., Shen, R., Yu, Y., Tan, R. and Guo, S., 2020. Secondary Organic Aerosol from Typical Chinese Domestic Cooking Emissions. *Environmental Science & Technology Letters*.