Responses to the comments & suggestions from the editor what is more the reviewers

Journal: Atmospheric Chemistry and Physics

Manuscript ID: acp-2021-216

Original Title: "Mass spectral characterization of secondary organic aerosol from urban cooking and vehicular sources emissions"

Revised Title: "Mass spectral characterization of secondary organic aerosol from urban cooking and vehicular sources"

Comments from the reviewer:

Reviewer

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.

- Thanks for the reviewer's patience and careful reviews on our manuscript. The comments from the reviewer on this manuscript have helped us organize our article in a more professional way. According to the points mentioned by the reviewer, we mainly supplemented the corresponding limitations in the "limitations and further work" section in the revised manuscript. The texts in bold black are the comments from the reviewer, followed by our responses in blue and/or red. All the changes are marked in the revised manuscript.

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??

- We appreciate the comments from the reviewer. In order to make the kitchen full of cooking fumes, we cook each dish 8 times; each time contains the same dish mass, oil temperature and frequency of stir-frying, etc. Therefore, we express it as "Each dish was continuously carried out 8 times in parallel during the cooking process until the closed kitchen was full of fumes". When the closed kitchen was full of fumes, the fumes were then introduced through the pipeline from the kitchen into the Gothenburg Potential Aerosol Mass (Go:

PAM) reactor. We do three parallel oxidation experiments. It is expressed in the original manuscript as "Each sampling was in parallel three times".

Combined with comment 2, we have deleted the detailed sentence "Each sampling was in parallel three times" in the revised manuscript to make it clear (line 92 in the marked revised manuscript).

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.

- Thanks for the reviewer's comment. There is really only one GO: PAM reactor. The fumes were continuously introduced through the pipeline from the kitchen into the reactor. After finishing an oxidation cycle, we flushed the pipeline and the reactor. Then, do the same oxidation experiment group again, repeat three times.

Combined comment 1 and 2, the sentence has been removed from the manuscript.

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?

- Thanks for the reviewer. The relative standard deviations of OA concentrations were small, which were under 10% in most cases during the experiment. This sentence is followed by the sentence "Each sampling was in parallel three times". Refer to comment 2, we decided to delete the sentence "The relative standard deviations were small, which were under 10% in most cases" (line 92-93 in the marked revised manuscript).

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"

- Thanks for the reviewer's comment. We have modified the description in the revised manuscript (line 100-101 in the marked revised manuscript) as follows:

"the combination of 1500 rpm rotating speed and 16Nm torque and 2000rpm rotating speed and 16Nm torque for the engine in this study reflect the realistic vehicle speed of 20km/h and 40km/h, respectively"

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?

- We appreciate the comments from the reviewer. For each running condition, we conducted five parallel experiments (cycle: EPA from 0 to 4.1 days). In each cycle, each instrument collected three parallel data groups for each EPA. Therefore, we expressed it as "The sampling time with collecting three parallel data groups was about 60 min for each experiment". Indeed, the sentence can easily confuse the reader. We have modified the descriptions in the revised manuscript (line 108 in the marked revised manuscript) as follows:

"The sampling time was about 60 min for each experiment."

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.

- Thanks for the reviewer's constructive comment. We have supplemented the table on POA mass concentration in vehicle experiments in the supplement (Table S4), and added the reference in the revised manuscript (line 193-194; line261-263; line 304-306 in the marked revised manuscript) as follows:

Table S4. The mass concentrations of primary organic aerosol (POA) for all conditions in vehicle experiments

Experiment	POA Mass concentration ($\mu g/m^3$)	
	Average	Standard Deviation
1500rpm_16Nm	1.20	0.30
1750rpm_16Nm	1.26	0.61
2000rpm_16Nm	1.14	0.30
2000rpm_32Nm	1.29	0.62
2000rpm_40Nm	1.23	0.31

*"It was worth noting that the source characteristics of vehicle POA were uncertain due to its low concentration emitted from the engine in this study (**Table S4**)."*

*"Different from the cooking, two-vehicle PMF SOA factors were derived from aged HOA, rather than integrated primary HOA and aged HOA datasets due to the low primary HOA emission (**Table S4**), as described in sect. 3.1."*

*"Considering the actual oxidation conditions, that is the concentration of OH radicals, and the lacking vehicle POA due to its low emission (**Table S4**), and the SOA spectra constraining reasonably"*

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"

- Thanks for the reviewer's comment. The "within" has been replaced by "with an" in the revised manuscript (line 146-147 in the marked revised manuscript) as follows:

"In this study, we adopted the toolkit SoFi (Source Finder) with an a-value approach to perform organic HR-AMS datasets collected in Shanghai."

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.

- We appreciate the comments from the reviewer. The m/z 67, 69, and the word "respectively" have been added in the sentence in the revised manuscript (line 253-255 in the marked revised manuscript) as follows:

"The cooking PMF POA of four Chinese dishes all showed obvious hydrocarbon-like signals at m/z 41, 43, 55, 57, 67, and 69 with ion fragments of $C_3H_5^+$, $C_3H_7^+$, $C_4H_7^+$, $C_4H_9^+$, $C_5H_7^+$, and $C_5H_9^+$, respectively."

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".

- We agree with the reviewer. The "oxygen-oxidation" has been replaced with "oxidized" in the revised manuscript (line 257-258 in the marked revised manuscript) as follows:

"For mass spectra of cooking PMF SOA, the oxidized ion fragments had higher signals than those of hydrocarbon-like ion fragments."

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".

- Thanks for the reviewer's comment. We have checked the manuscript, and replaced "LO-SOA; MO-SOA" with "vehicle PMF LO-SOA; vehicle PMF MO-SOA". All the changes are documented in the revised manuscript (line 265-268; line 268-270; line 271-273; line 277-285) as follows:

*"As indicated in Fig.3 and Table S12, the prominent m/z 28 (average $f_{28}=0.045$), 41 (average $f_{41}=0.046$), 43 (average $f_{43}=0.158$),44 (average $f_{44}=0.054$), 55 (average $f_{55}=0.039$), 57 (average $f_{57}=0.027$) of **vehicle PMF LO-SOA** were comparable with those of cooking PMF SOA. The fraction of m/z 43 of **vehicle PMF LO-SOA** was higher than that in cooking SOA by a factor of 2. The abundant m/z 28 and 44 (mainly generated from CO_2^+) are widely used as the ambient MO-OOA markers. (Sun et al., 2018; Xu et al., 2017). We observed high factions of m/z 28 ($f_{28}=0.110\sim0.214$) and m/z 44 ($f_{44}=0.121\sim0.224$) in **vehicle PMF MO-SOA** (Table S13) and high O/C ratios (0.88~1.33), which were much higher than those of **vehicle PMF LO-SOA** (O/C=0.37~0.53) and cooking SOA (O/C=0.29~0.41)."*

*"Similarly, for the resolved SOA factors, the correlation of mass spectra among cooking groups under different cooking methods ($\theta = 8\sim21°$) was worse than that of vehicle groups (**vehicle PMF LO-SOA**; $\theta = 3\sim19°$) under different running conditions (Table S14 and Table S16). The mass spectra of the PMF POA factors for deep-frying chicken exhibited poor agreement with those of stir-frying cabbage, Kung Pao chicken, and shallow-frying tofu (Table S15). In addition, we also found that the θ angles between **vehicle PMF LO-SOA** and **vehicle PMF MO-SOA** under five GDI running conditions were ranged from 36° to 50° (Fig.S11), indicating that the mass spectra profiles of **vehicle PMF LO-SOA** are poor consistency with those of **vehicle PMF MO-SOA**, consistent with the changes in the mass spectra characteristics of vehicles, under the same emission conditions and different oxidation conditions."*

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.

- We appreciate the constructive comment from the reviewer. We have added the related description of issue in the revised manuscript (line 263-264;269-270). Meanwhile, the limitation that the inability to separate POA in vehicle experiments has been added in the "Limitations and future work" section of the revised manuscript (line 400-404 in the marked revised manuscript) as follows:

"Unfortunately, vehicle PMF POA factor cannot be separated from aged HOA due to higher OH exposure."

"The fraction of m/z 43 of vehicle PMF LO-SOA was higher than that in cooking SOA by a factor of 2, which may be caused by the inability to separate vehicle PMF POA factor in the PMF analysis."

"Especially, the absence of primary HOA due to low emissions of engine and the inability to separate vehicle PMF POA from aged HOA in the PMF analysis were major limitations of this study. In addition to obtaining pure vehicle POA through source experiments, further work can apply ME-2 model for constraining pure SOA profiles from experimental datasets to obtain the vehicle POA profiles."

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?

- We appreciate the constructive comment from the reviewer. Hu et al., 2016a checked through all correlations between time series of organic ions in AMS and COA, and found that $C_6H_{10}O^+$ ion correlates best with COA and thus suggest it is best tracer ion for COA. The correlation between COA and $C_6H_{10}O^+$ tracer ion was also analyzed in other previous studies, e.g., Ge et al., 2012; Sun et al., 2011; Xu et al., 2016.

The time series of mass concentration of NR-PM$_1$ species and the ion-speciated mass e.g., $C_6H_{10}O^+$ were analyzed with the ToF-AMS standard data analysis software (SQUIRREL version 1.57 and PIKA version 1.16). The time series of OA factors can be obtained in PMF analysis. As shown in Fig.S17, Fig.S18, and Fig.S21, we provided time series of OA factors and other relevant tracer species and ions. The time-series correlations of COA with $C_6H_{10}O^+$ were presented in these figures. In addition, we analyzed the correlation coefficient (Pearson r) between the OA factors and the corresponding tracer during the winter and summer observations, which is presented in Table S22 and Table S23.

We have added the related references in the revised manuscript (line 355-357 in the marked revised manuscript) as follows:

"However, COA_ME-2 correlated better with $C_6H_{10}O^+$ than COA_PMF, which was considered a fragment tracer mainly from cooking emissions (Ge et al., 2012; Hu et al., 2016a; Sun et al., 2011; Xu et al., 2016)."

Ge, X., Setyan, A., Sun, Y., Zhang, Q., 2012. Primary and secondary organic aerosols in Fresno, California during wintertime: Results from high resolution aerosol mass spectrometry. Journal of Geophysical Research-Atmospheres 117.

Hu, W., Hu, M., Hu, W., Jimenez, J.L., Yuan, B., Chen, W., Wang, M., Wu, Y., Chen, C., Wang, Z., 2016a. Chemical composition, sources, and aging process of submicron aerosols in Beijing: Contrast between summer and winter. Journal of Geophysical Research Atmospheres 121, 1955-1977.

Sun, Y.L., Zhang, Q., Schwab, J.J., Demerjian, K.L., Chen, W.N., Bae, M.S., Hung, H.M., Hogrefe, O., Frank, B., Rattigan, O.V., Lin, Y.C., 2011. Characterization of the sources and processes of organic and inorganic aerosols in New York city with a high-resolution time-of-flight aerosol mass apectrometer. Atmospheric Chemistry and Physics 11, 1581-1602.

Xu, J., Shi, J., Zhang, Q., Ge, X., Canonaco, F., Prévôt, A.S., Vonwiller, M., Szidat, S., Ge, J., Ma, J., 2016. Wintertime organic and inorganic aerosols in Lanzhou, China: sources, processes, and comparison with the results during summer. Atmospheric Chemistry and Physics 16, 14937-14957.

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 addresal of this limitation (lack of external tracers) would be needed.

- We appreciate the comments from the reviewer. Figure S21 shows the time series of OA factors and the tracers in summertime. During the summer observation period, neither the ME-2 nor the PMF model has resolved other-POA factor. Therefore, there is no time series tracer for other-POA. The time-series correlations of the other-POA factor identified in wintertime with external tracers displayed in Fig.S17. The Sandradewi model can well extract the fossil fuel and wood burning components of BC. It is a better way to use the extraction as tracers for other-POA and HOA factors. Undeniably, the perform on Sandradewi model is difficult for me. In this study, we used the primary emission tracers (i.e., BC and NOx) to compare with the time-series correlation of HOA. The correlation analysis between primary emission tracers and HOA has been widely used in the previous studies (Duan et al., 2020; Hu et al., 2016a). m/z 60 in AMS OA spectra is a tracer of BBOA (Alfarra et al., 2007; Lee et al., 2010). The previous studies also analyzed the correlation between BBOA and m/z 60 tracer ($C_2H_4O_2^+$) (Duan et al., 2020 and Hu et al., 2016a), as shown in Fig.5 and Fig.S17 in this study.

Duan, J., Huang, R.-J., Li, Y., Chen, Q., Zheng, Y., Chen, Y., Lin, C., Ni, H., Wang, M., Ovadnevaite, J., Ceburnis, D., Chen, C., Worsnop, D.R., Hoffmann, T., O'Dowd, C., Cao, J., 2020. Summertime and wintertime atmospheric processes of secondary aerosol in Beijing. Atmospheric Chemistry and Physics 20, 3793-3807.

Hu, W., Hu, M., Hu, W., Jimenez, J.L., Yuan, B., Chen, W., Wang, M., Wu, Y., Chen, C., Wang, Z., 2016a. Chemical composition, sources, and aging process of submicron aerosols in Beijing: Contrast between summer and winter. Journal of Geophysical Research Atmospheres 121, 1955-1977.

Alfarra, M.R., Prevot, A.S.H., Szidat, S., Sandradewi, J., Weimer, S., Lanz, V.A., Schreiber, D., Mohr, M., Baltensperger, U., 2007. Identification of the mass spectral signature of organic aerosols from wood burning emissions. Environmental Science & Technology 41, 5770-5777.

Lee, T., Sullivan, A.P., Mack, L., Jimenez, J.L., Kreidenweis, S.M., Onasch, T.B., Worsnop, D.R., Malm, W., Wold, C.E., Hao, W.M., Collett, J.L., Jr., 2010. Chemical Smoke Marker Emissions During Flaming and Smoldering Phases of Laboratory Open Burning of Wildland Fuels. Aerosol Science and Technology 44, I-V.

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".

- We appreciate the constructive comment from the reviewer. Unconstrained PMF analyses of OA data suffer from rotational ambiguity when sources show similar profiles and temporal covariation (Canonaco et al., 2013; Huang et al., 2019). As we stated "Besides, we observed that HOA and COA profiles (**provided via PMF during the wintertime**) contained high signals at the biomass burning tracer ion (m/z 73), and m/z 91 (PAH-related m/z), indicating that the mixing among HOA, COA, and other source emissions (e.g., BBOA)", mixing can be exhibited in the factors of COA, HOA, and other source emissions, that is, other source cannot be identified by using PMF. However, by introducing a priori information as additional model input and constraining one or more output factor profiles to a predetermined range, ME-2 can overcome such difficulties and provide more environmentally meaningful solutions. As described in our study, other primary emission sources (other-POA) mixed in HOA and COA can be effectively resolved in ME-2 model during the winter observation periods in Shanghai by constraining source profiles of the primary and secondary of cooking and vehicles that obtained in our experiment work. The other-POA factor correlated with m/z 60 (a tracer of BBOA).

Canonaco, F., Crippa, M., Slowik, J. G., Baltensperger, U., and Prévôt, A. S. H.: SoFi, an IGOR-based interface for the efficient use of the generalized multilinear engine (ME-2) for the source apportionment: ME-2 application to aerosol mass spectrometer data, Atmos. Meas. Tech., 6, 3649-3661.

Huang, R.-J., Wang, Y., Cao, J., Lin, C., Duan, J., Chen, Q., Li, Y., Gu, Y., Yan, J., Xu, W., Fröhlich, R., Canonaco, F., Bozzetti, C., Ovadnevaite, J., Ceburnis, D., Canagaratna, M. R., Jayne, J., Worsnop, D. R., El-Haddad, I., Prévôt, A. S. H., and O'Dowd, C. D.: Primary emissions versus secondary formation of fifine particulate matter in the most polluted city (Shijiazhuang) in North China, Atmos. Chem. Phys., 19, 2283-2298.

At present, m/z60, BC (NOx), nitrate, and sulfate has been widely used as the tracer of biomass combustion sources, vehicle sources, LO-OOA and MO-OOA factors in many previous studies, but there are

currently no measured tracer species, which can indicate the secondary sources of vehicles and cooking. The measurement of these tracers will provide a good support for the OA source apportionment. This study was to combine vehicle LO-SOA and cooking SOA as LO-OOA. We analyzed the time series-correlation of LO-OOA with nitrate and other tracer ions. Indeed, it is a limitation in our work. According to the reviewer's comment, we have added the limitation in the revised manuscript as follows:

"Besides, measurements of accurate tracers for all factors that resolved by PMF or ME-2 model should be conducted in future work to improve source apportionment verification. For example, we had to combine vehicle LO-SOA and cooking SOA as LO-OOA due to the lack of the measurement tracers for vehicle and cooking SOA factor, and then we analyzed the time series-correlation of LO-OOA with nitrate and other tracer ions."

13. Fig. S21: The authors report BC measurements but do not report instrumentation to measure BC in the Methods section. This should be corrected.

- Thanks for the reviewer's comment. The measurements of BC, NOx, sulfate, and nitrate during the winter or summer observations in Shanghai have been described in our previous study, which investigate the seasonal variation of aerosol compositions in Shanghai (Zhu et al., 2021). We have added the related description and reference in the revised manuscript (line 313-316 in the marked revised manuscript) to make the reader clear.

"For the tracers described below, the mass concentration of chemical compositions e.g., sulfate, nitrate, and ion-speciated fragment were detected by HR-ToF-AMS, as shown in Zhu et al., 2021b. The detail measurements of black carbon (BC) and nitrogen oxides (NOx) can also be found in Zhu et al., 2021b."

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, NOx, sulfate, nitrate, chloride, ammonium, and BC) should be reported here. The authors should not selectively present "good" data.

- Thanks for the reviewer's constructive comment. The final results for source apportionment by using PMF or ME-2 model are verified based on the rationality of unconstrained factors, distinct mass spectra, time series and good correlations with external tracers for all factors. In many previous studies (Ge et al., 2012; Hu et al., 2016a; Sun et al., 2011; Xu et al., 2016; Duan et al., 2020), the time series consistency and high correlation between the resolved factors and the external tracer are considered as an important criterion for source verification. They used m/z60, BC (NOx), nitrate, and sulfate as the tracer of biomass combustion sources, vehicle sources, LO-OOA and MO-OOA factors, respectively. In our work, we investigated the time

series of all resolved factors and corresponding widely used tracers (Fig.S17; Fig.S18; Fig.S21), and analyzed the correlation between all factors and these tracers (Table S20; Table S22; Table S23 in the revised supplement material). Unfortunately, we lack the measurement of SOA tracers for vehicles and cooking. We have added the limitation in the revised manuscript as in the reply of comment 12.

"Besides, measurements of accurate tracers for all factors that resolved by PMF or ME-2 model should be conducted in future work to improve source apportionment verification. For example, we had to combine vehicle LO-SOA and cooking SOA as LO-OOA due to the lack of the measurement tracers for vehicle and cooking SOA factor, and then we analyzed the time series-correlation of LO-OOA with nitrate and other tracer ions."

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.

- We appreciate the comments from the reviewer. The issue "Constraining many SOA factors could be over-constraining the ME-2 runs, which leads to factor mixing and reduces the number of factors." were reported here to explain the reasons for the selection of ME-2 inputting profiles (excluded vehicle MO-SOA). We agree with the review. We have moved the sentence to the "limitation and future work" section in the revised manuscript (line 404-410 in the marked revised manuscript) as follows:

"Constraining many SOA factors could be over-constraining the ME-2 runs, which leads to factor mixing and reduces the number of factors. Therefore, SOA source spectra can only be appropriately and reasonably limited in ME-2 model. Besides, measurements of accurate tracers for all factors that resolved by PMF or ME-2 model should be conducted in future work to improve source apportionment verification. For example, we had to combine vehicle LO-SOA and cooking SOA as LO-OOA due to the lack of the measurement tracers for vehicle and cooking SOA factor, and then we analyzed the time series-correlation of LO-OOA with nitrate and other tracer ions."

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.

- Thanks for the reviewer's constructive comment. We have checked the manuscript carefully and corrected in the title and throughout the revised manuscript.

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.

- We appreciate the comments from the reviewer. Combined with comment 11 and 15, we have modified the related sentence in the revised manuscript and added the limitation that the inability to separate POA in the "Limitations and future work" section of the revised manuscript (line 261-264; line 400-406 in the marked revised manuscript) as follows:

*"Different from the cooking, two-vehicle PMF SOA factors were derived from aged HOA, rather than integrated primary HOA and aged HOA datasets due to the low primary HOA emission (**Table S4**), as described in sect. 3.1. Unfortunately, vehicle PMF POA factor cannot be separated from aged HOA due to higher OH exposure."*

"Especially, the absence of primary HOA due to low emissions of engine, and the inability to separate vehicle PMF POA from aged HOA in the PMF analysis were major limitations of this study. In addition to obtaining pure vehicle POA through source experiments, further work can apply ME-2 model for constraining pure SOA profiles from experimental datasets to obtain the vehicle POA profiles. Constraining many SOA factors could be over-constraining the ME-2 runs, which leads to factor mixing and reduces the number of factors. Therefore, SOA source spectra can only be appropriately and reasonably limited in ME-2 model."