Comments From Reviewer 2

We appreciate the constructive comments from the reviewer on this manuscript. We have answered the reviewer's questions point to point in the following paragraphs (the texts italicized are the comments, the texts indented are the responses, and the texts in blue are revised parts in the new manuscript). In addition, all changes made are marked in the revised manuscript.

1.The authors' reply to my comment #1 added information about the tubing materials and the Go:PAM reactor geometry, but information about the inlet tubing length, diameter, and residence time were not given, and are still needed.

> Thanks for the reviewer's response. In order to make the article easier to follow, we have supplied the information about the inlet tubing length, diameter, and residence time in SI.
>
> "**Revised text in Section S1:** From the sampling port at the source (cooking and vehicle) to the inlet of HR-ToF-AMS, the 3/8 inch (inner diameter was 6 mm) stainless steel tubes were totally 7 meters long and the corresponding residence time was 4.9 s. There were 5 meters long from the sampling port to the Go: PAM, and the flow rate was 5.5 L/min (HR-Tof-AMS and other instruments jointly determined the flow rate). There were 2 meters long from the Go: PAM to the HR-ToF-AMS, and the flow rate was 1 L/min (HR-ToF-AMS and its drainage system determined the flow rate)."

2.The reply to my comment #2 concedes my point that the different residence times complicate direct comparisons between cooking and vehicle exhaust measurements. This needs to be stated in the methods section, so that readers understand the importance of maintaining a fixed residence time and apply it in their own work. Similarly, the concluding section would benefit from a sentence or two that is more committal to future better-quality OFR experiments than "we really hope we could strictly control the temperature, RH, and other conditions". The authors argue that that the relative humidity, although different between in the two sources, is low enough in both cases to minimally influence heterogenous chemistry. This is a fair point, and one that should be added to the revised manuscript.

> Thanks for the reviewer's suggestion. We have supplied necessary expressions in Method and Conclusion sections.
>
> "**Revised text in Method 2.3.2:** "The Go: PAM conditions for vehicle and cooking experiments could be seen in Table 3 and Table 4, respectively. Their experiment conditions (such as residence time and RH) were not completely the same because of the inherent difference and experimental design between two sources. Whereas, some comparisons could be still analyzed in the similar OH exposure, and their RH conditions were both low where photochemical oxidations instead

of aqueous-phase processing dominated the chemical evolution process (Xu et al., 2017)."

"**Revised text in Conclusion:** "There are some uncertainties of our Go: PAM simulation. We focused more on the photochemical oxidation of SOA under low RH levels, but aqueous-phase processing at high RH levels may also have impacts to SOA production. In the future, it'll be better to strictly control the RH, high/low NO_x or SO_2, additional inorganic seeds, and so forth, to deeply investigate how the aerosol ages as a function of equivalent days of atmospheric oxidation."

Reference:

Xu, W., Han, T., Du, W., Wang, Q., Chen, C., Zhao, J., Zhang, Y., Li, J., Fu, P., Wang, Z., Worsnop, D. R., and Sun, Y.: Effects of Aqueous-Phase and Photochemical Processing on Secondary Organic Aerosol Formation and Evolution in Beijing, China, Environmental science & technology, 51, 762-770, 10.1021/acs.est.6b04498, 2017.

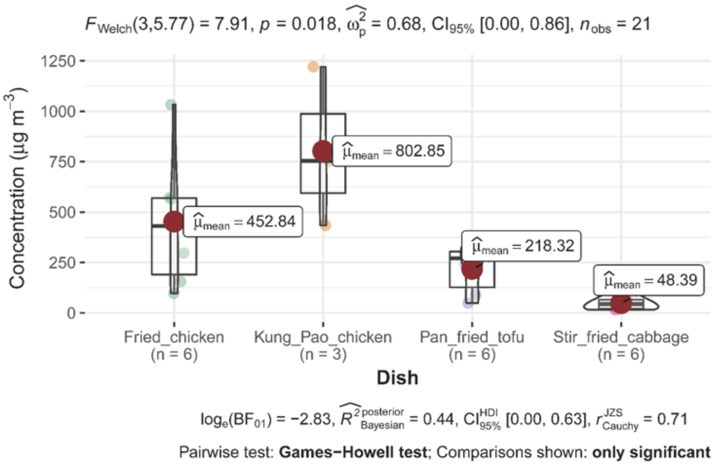
3.In response to my comment #3, the authors added fractional contributions of alkanes, alkenes,aromatics, O-VOCs, and X-VOCs to the measured VOC emissions from each source, along with kOH values for the major components of each of those compound classes. This added information is appreciated and is likely sufficient to give the reader a general idea of which compounds contribute to the calculated OH reactivity values. However, in my opinion, the discussion of which emitted compounds contribute to the measured SOA formation is an important outcome of this study whose discussion remains unclear/incomplete. For example, the SOA/POA ratio was ~100 for GDI emissions and ~2 for cooking emissions, but only 29-35 ppb VOCs were speciated in GDI emissions and 124-189 ppb VOCs were speciated in cooking emissions. It is clear from these numbers that the aggregate SOA yields of cooking VOCs are lower than GDI VOCs. It is not clear how much of the observed SOA can be explained from the speciated VOCs (and laboratory SOA yields obtained in chamber studies), and whether an unresolved complex mixture of (I)VOCs is plausibly an important class of SOA precursors, particularly for the GDI emissions. To increase the scientific significance of this manuscript, I feel strongly that additional analysis needs to be done in this regard, such that we can gain insight into contributions of measured VOCs to observed SOA formation in both sources. See, for example, Figure 3 of Liao et al., ES&T, 2021, reproduced below for reference – it should be possible to perform a similar analysis from the data obtained here.

Thanks for the constructive comments. The reviewer pointed out that "*For example, the SOA/POA ratio was ~100 for GDI emissions and ~2 for cooking emissions, but only 29-35 ppb VOCs were speciated in GDI emissions and 124-189 ppb VOCs were speciated in cooking emissions.*" In fact, The GDI emission underwent a larger dilution ratio than cooking emission did, so that the VOCs from GDI is not far lower than those from cooking.

We agree with the reviewer's comment that the gaseous results are important. In this article, we just used simple VOCs results from GC-MS in order to estimate the OHR. If we need to do a further analysis, more sophisticated results are necessary. To be frank, we have measured the VOCs and S/I VOCs by 2D-GC-MS, VOCUS and CIMS during Go: PAM experiments, but their sampling, quantification and data analysis are so complicated and different from particle phase, that our group hope to write individual articles to show our findings in detail. As following figures show, here are some initial results of cooking experiment (unpublished):

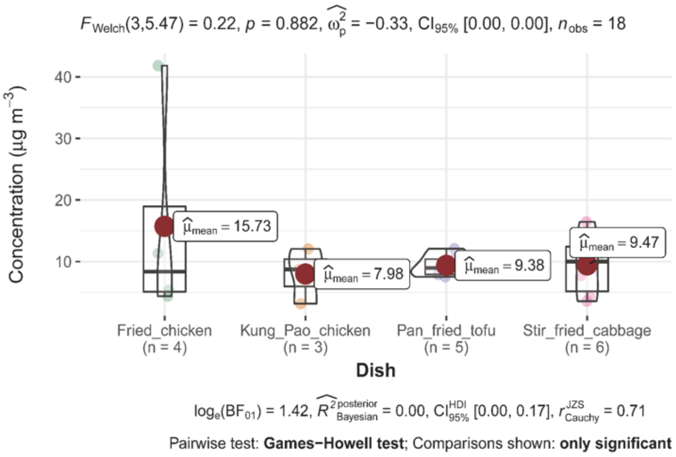
Gas-phase

- Significant difference between four dishes in gas-phase total S/I VOCs ($p < 0.05$)
- Kung Pao chicken & Pan-fried tofu: large proportion of aromatics
- Furans were abundant in gas-phase oxygenated compounds (> 6%)



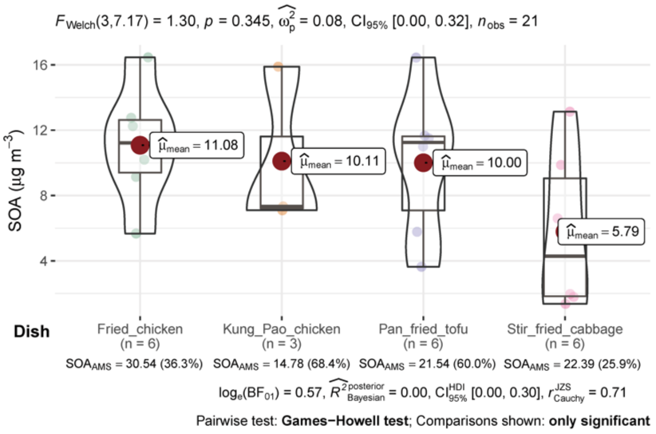
Particle-phase

- NO significant difference between four dishes in particle-phase total S/IVOCs ($p > 0.8$)
- Oxygenated compounds were the most abundant (>40%), including acids, furans, amides and esters

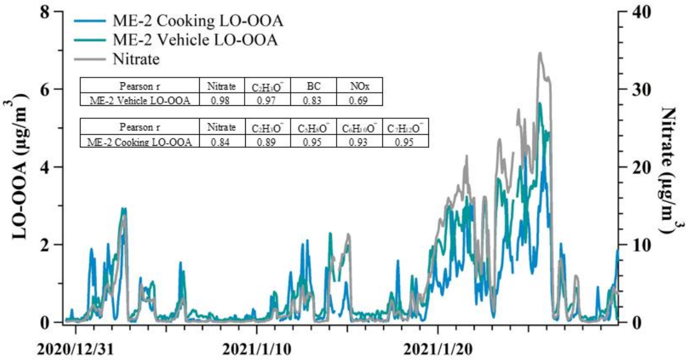


SOA formation

- 26% - 68% SOA could be explained from oxidation of VOCs, IVOCs, SVOCs precursors



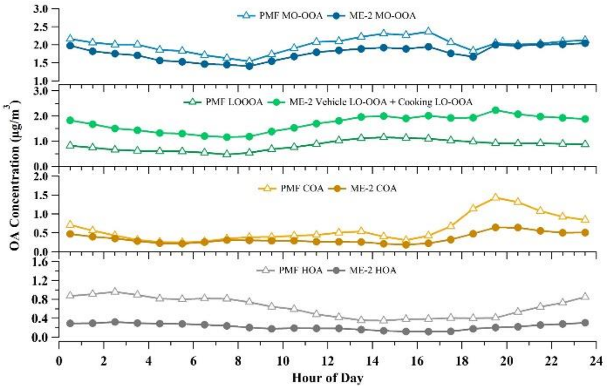
This resubmitted manuscript focused more on particle phase (based on HR-ToF-AMS), especially its SOA growth factor, O/C, oxidation pathway and mass spectra. We have compared cooking and vehicle SOA mass spectra with ambient SOA spectra in Beijing (Discussion 3.4). In the future, the cooking and vehicle SOA mass spectra could be used as the input mass spectra in ME-2, in order to estimate the mass concentration of cooking SOA and vehicle SOA, and then study their chemical evolutions in ambient. We are designing another article to study the application of our SOA mass spectra (cooking and vehicle) in urban areas. As following figures show, here is one practice result in Beijing (unpublished).



Pearson r	Nitrate	$C_2H_3O^-$	BC	NOx
ME-2 Vehicle LO-OOA	0.98	0.97	0.83	0.69

Pearson r	Nitrate	$C_2H_3O^-$	$C_5H_8O^-$	$C_6H_{10}O^-$	$C_7H_{12}O^-$
ME-2 Cooking LO-OOA	0.84	0.89	0.95	0.93	0.95

Timeseries of OA factors and good correlations with relevant species



Reasonable diurnal variations of OA factors

Therefore, the results in this article are valuable and could be used to do further estimation in ambient. We regarded this article as a beginning of our “source secondary simulation” research. We will try our best to give more informative results in other new articles as soon as possible. Thanks for the reviewer’s helpful advice again.

4.OK

Thanks for the helpful advice.

5.The addition of Table S7 is useful and appreciated. However, the discussion about non-OH artifacts misses the point : ozonolysis is (likely) important for unsaturated fatty acids that were probably emitted from the *cooking sources, and perhaps measured (e.g. oleic acid parent and dehydration peaks at AMS m/z = 282 and 264), but have not been analyzed. At the least, a couple sentences should be added to the discussion indicating that contributions from ozonolysis of unsaturated fatty acids to SOA/OPOA are not constrained in the data that has been analyzed but may be important.*

We agree with the reviewer that ozonolysis is (likely) important for unsaturated fatty acids that were probably emitted from the cooking sources, but we didn't find the significant effect of ozonolysis in our experiment. This may be because the residence time is too short for ozonolysis reactions. Besides, Vesna *et al.* found that the heterogeneous reaction of ozone with oleic acid aerosol particles was influenced by humidity and reaction time in an aerosol flow reactor. We have added necessary expressions in Method and Conclusion section.

"**Revised text in Method 2.3.2: "**It is found that the heterogeneous reaction of ozone with oleic acid aerosol particles was influenced by humidity and reaction time in an aerosol flow reactor (Vesna et al., 2009). Therefore, non-OH reactions, such as the ozonolysis of unsaturated fatty acids, may also be important in forming SOA, which missed specific designs in our experiment."

"**Revised text in Conclusion:** "Moreover, contribution of ozonolysis to SOA formation should be individually studied in further research."

6.I suggest paraphrasing this response and adding it to the revised manuscript.

We have added this response in Method section as the review suggested.

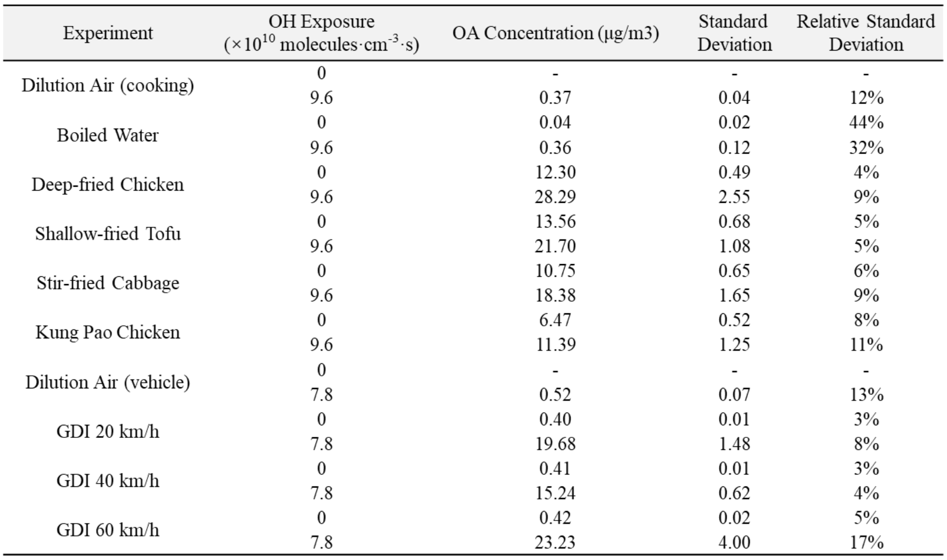
"**Revised text in Method 2.1:** Vehicle exhaust from tailpipe was first diluted by a gradient heated dilution system (6 fold) and then diluted by an unheated dilution system (5 fold). The temperature of sample flow was near indoor temperature (20-25°C) after secondary dilution systems. The cooking fumes was collected through the kitchen ventilator, where the temperature was similar to that of indoor air."

7.This response does not address my comment. Cleaning the (dark) Go:PAM by flowing dry air and ozone through it (very likely) does not remove all of the residual organics that are reactive towards OH but not O3. Thus, the next time the lights are turned on to make OH, some of the SOA formed is almost certainly associated with residual organics that were not sufficiently removed from the previous experiment by only flowing dry carrier gas + ozone through the dark OFR between experiments. As far as I can tell, and based on my own experience, it is impossible to rule this out the way the experiments were performed. This is a flaw in the cleaning protocol that has to be expressed in the methods section - i.e. that the SOA that is formed in any experiment with OH as the oxidant represents an upper limit because the background is not well constrained. This should be directly addressed in future studies by adding humidity to the carrier gas and turning on the *lights during the OFR cleanout stage.*

Thanks for the reviewer's careful reviews on technical details of our experiment. As reviewer suggested, it is recommended to add humidity to the carrier gas and turn on the lights during the OFR cleanout stage, in order to minimize the background concentration in Go: PAM. We feel grateful and will remember this important point in the future.

As for our experiments, before each experiment, all pipelines and the Go: PAM chamber were continuously flushed with purified dry air, until the concentrations were minimal (just like blank groups in Table S1) when the UV was on or off. We agree with that the SOA formed represented an upper limit because the background was not totally clean, but its influence is minimal according to the comparison of blank and experimental groups in Table S1. We have supplied necessary expressions in Method and Conclusion section.

Table S1. Comparison of results between blank and experimental groups (Dilution air and boiled water are two kinds of blank groups. The others are experimental groups).



Experiment	OH Exposure (×10^{10} molecules·cm^{-3}·s)	OA Concentration (μg/m3)	Standard Deviation	Relative Standard Deviation
Dilution Air (cooking)	0	-	-	-
	9.6	0.37	0.04	12%
Boiled Water	0	0.04	0.02	44%
	9.6	0.36	0.12	32%
Deep-fried Chicken	0	12.30	0.49	4%
	9.6	28.29	2.55	9%
Shallow-fried Tofu	0	13.56	0.68	5%
	9.6	21.70	1.08	5%
Stir-fried Cabbage	0	10.75	0.65	6%
	9.6	18.38	1.65	9%
Kung Pao Chicken	0	6.47	0.52	8%
	9.6	11.39	1.25	11%
Dilution Air (vehicle)	0	-	-	-
	7.8	0.52	0.07	13%
GDI 20 km/h	0	0.40	0.01	3%
	7.8	19.68	1.48	8%
GDI 40 km/h	0	0.41	0.01	3%
	7.8	15.24	0.62	4%
GDI 60 km/h	0	0.42	0.02	5%
	7.8	23.23	4.00	17%

"**Revised text in Method 2.2:** "Before each experiment, all pipelines and the Go: PAM chamber were continuously flushed with purified dry air, until the concentrations were minimal (just like blank groups in Table S1) when the UV was on or off. The SOA formed in each experiment represented the upper limit due to the presence of background concentration."

"**Revised text in Conclusion:** "Besides, it is recommended to add humidity to the carrier gas and turn on the lights during the OFR cleanout stage, in order to minimize the background concentration in the Go: PAM."

8.Thank you.

Thanks for the careful review.

9.I disagree that discussion of acetylacetone is useful when there is no acetylacetone in the emissions. Please focus the discussion of non-OH artifacts on the alkanes, alkenes, aromatics, OVOCs, and X-VOCs that were measured, and the co-emitted compounds that were emitted and (likely) measured but not analyzed (e.g. fatty acids).

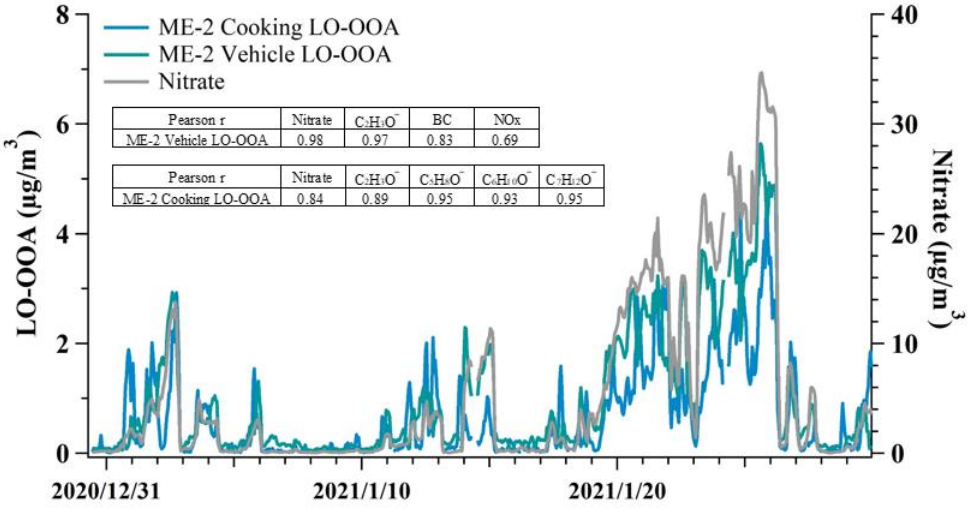
Thanks for the reviewer’s comments. We have removed the discussion of acetylacetone.

10.No additional comments

Thanks for the constructive comments.

11.See response to comment #3. The authors argue that it is beyond the scope of this paper to discuss the SOA formation potential of individual measured VOCs. I disagree – in my opinion, that is one of the potentially most novel results for which data is already available. Further analysis and discussion is required (e.g. Liao et al., 2021).

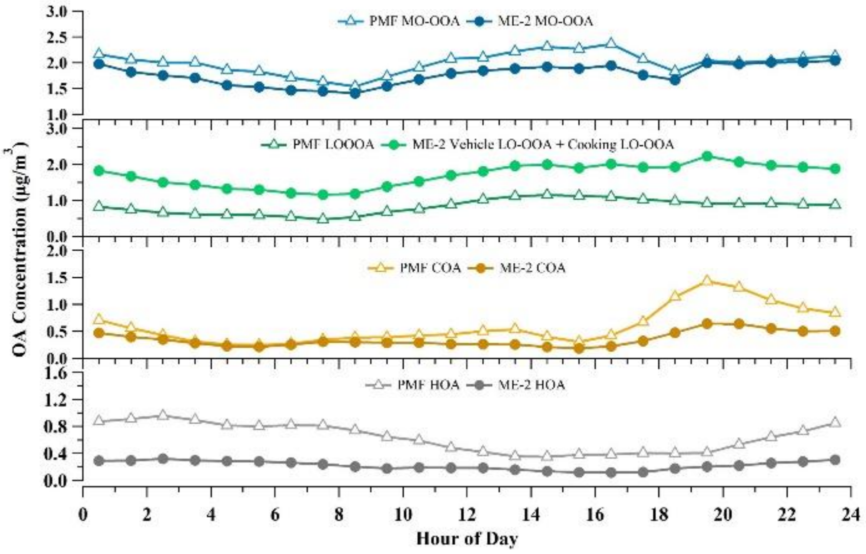
Thanks for the constructive comments. This resubmitted manuscript focused more on particle phase, especially its SOA growth factor, O/C, oxidation pathway and mass spectra. We have compared cooking and vehicle SOA mass spectra with ambient SOA spectra in Beijing (Discussion 3.4). In the future, the cooking and vehicle SOA mass spectra could be used as the input mass spectra in ME-2, in order to estimate the mass concentration of cooking SOA and vehicle SOA, and then study their chemical evolutions in ambient. We are designing another article to study the application of our SOA mass spectra (cooking and vehicle) in urban areas. As following figures show, here is one practice in Beijing (unpublished):



Pearson r	Nitrate	$C_2H_3O^-$	BC	NOx
ME-2 Vehicle LO-OOA	0.98	0.97	0.83	0.69

Pearson r	Nitrate	$C_2H_3O^-$	$C_5H_8O^-$	$C_6H_{10}O^-$	$C_7H_{12}O^-$
ME-2 Cooking LO-OOA	0.84	0.89	0.95	0.93	0.95

Timeseries of OA factors and good correlations with relevant species



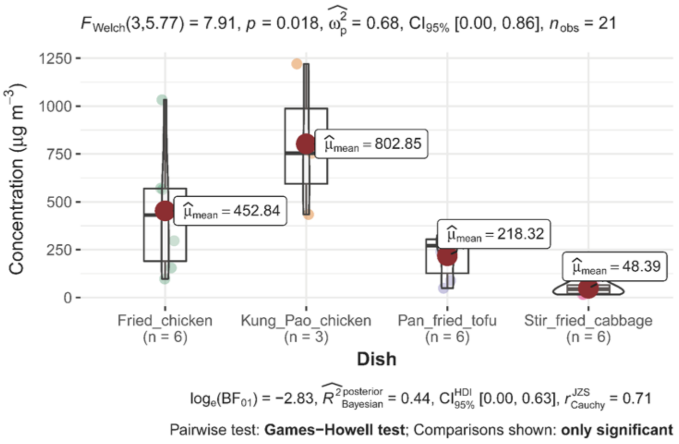
Reasonable diurnal variations of OA factors

Therefore, the results in this article are valuable and could be used to do further estimation in ambient. We regarded this article as a beginning part of our "source secondary simulation" research. We will try our best to give more informative results in other new articles as soon as possible.

We agree with the reviewer's comment that the gaseous results are important. In this article, we just use simple VOCs results from GC-MS in order to estimate the OHR. As for the further measurement and analysis results of gaseous phase, we have regarded it as another independent and attractive part. In fact, we have measured the VOCs and S/I VOCs by 2D-GC-MS, VOCUS and CIMS during Go: PAM experiments, but their sampling, quantification and data analysis are so complicated and different from particle phase, that we need to write individual articles to show our findings. Thanks for the reviewer's helpful advice again. As the following figures show, here are some initial results of cooking experiment (unpublished). We really hope our findings would benefit other researchers' studies in the future.

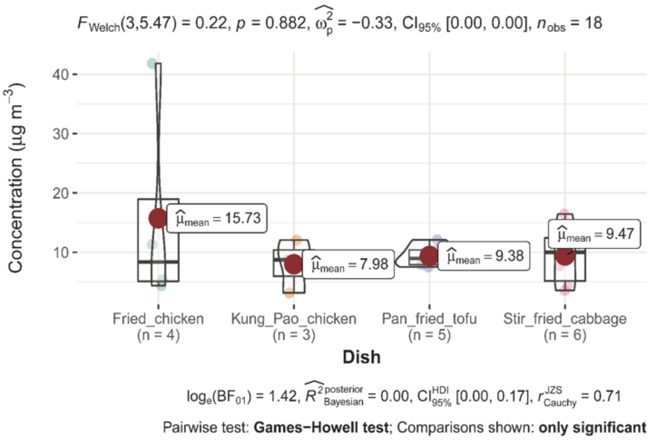
Gas-phase

- Significant difference between four dishes in gas-phase total S/I VOCs ($p < 0.05$)
- Kung Pao chicken & Pan-fried tofu: large proportion of aromatics
- Furans were abundant in gas-phase oxygenated compounds (> 6%)



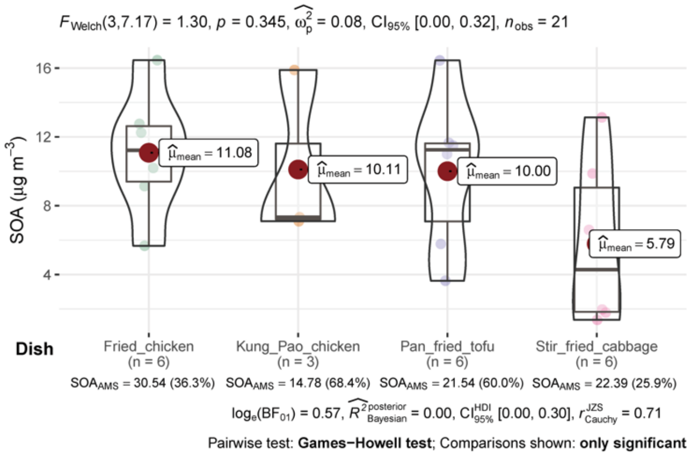
Particle-phase

- NO significant difference between four dishes in particle-phase total S/IVOCs (p > 0.8)
- Oxygenated compounds were the most abundant (>40%), including acids, furans, amides and esters



SOA formation

- 26% - 68% SOA could be explained from oxidation of VOCs, IVOCs, SVOCs precursors



12.Thank you.

Thanks for the correction.

13.Figure 2 would be much easier (for me) to read if it were split into two panels or a single "split" yaxis, one with an SOA/POA axis ranging from 0.4 to 4, the other with an SOA/POA axis ranging from 40 to 200. The maximum photochemical ages (~3 and 6 days) are close enough that, in my opinion, they can both be plotted satisfactorily on the same x-axis, here and also in Figure 3.

As the reviewer advised, we have modified the Figure 2 and Figure 3 into the same x-axis.

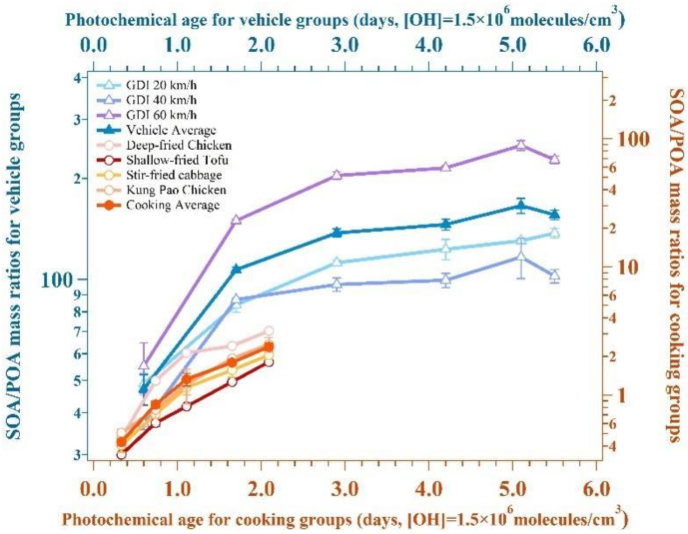


Figure 2. Secondary mass growth potentials for two urban lifestyle SOA. The SMPS-1 determined the mass concentration of POA, while the SMPS-2 determined the mass concentration of aged OA, and their mass difference could be regarded as the SOA. The average data and standard deviation bars are shown in the figure.

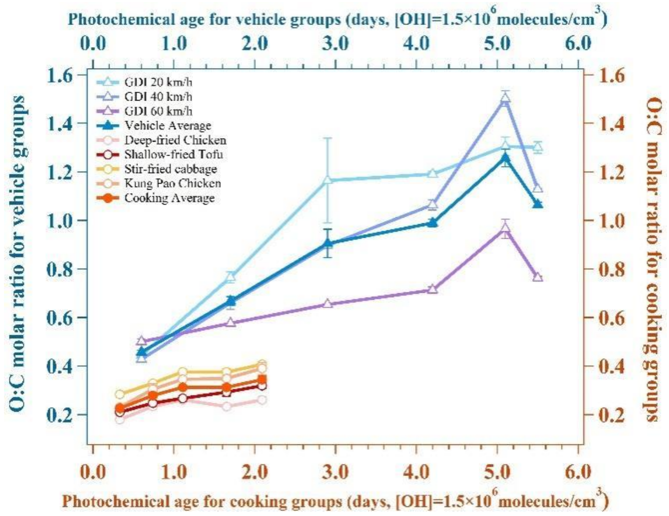


Figure 3. Evolution of O:C molar ratio for two urban lifestyle OA. The O:C molar ratios are determined by HR-Tof-AMS. The average data and standard deviation bars at each gradient are shown in the figure.

14.Thank you.

Thanks for the positive reply.

References

Liao, K., Q. Chen, Y. Liu, Y. J. Li, A. T. Lambe, T. Zhu, R.-J. Huang, Y. Zheng, X. Cheng, R. Miao, G. Huang, R. B. Khuzestani and T. Jia. Secondary Organic Aerosol Formation of Fleet Vehicle Emissions in China: Potential Seasonality of Spatial Distributions. Environmental Science & Technology, https://doi.org/10.1021/acs.est.0c08591, 2021.

We have added this article in Introduction section. Thanks for the positive reply.

"**Revised text in Introduction:** "NO_x levels may greatly influence the chemical evolution of vehicle SOA, and its NO_x/VOCs values are often strongly dependent on the sampling time and place in urban areas (Zhan et al., 2021;Wei et al., 2014). It is found that the photochemical ages for maximum SOA production under high-NO_x levels were lower than those under low-NO_x levels among OFR simulations (Liao et al., 2021)."

Comments From Reviewer 3

We appreciate the constructive comments from the reviewer on this manuscript. We have answered the reviewer's questions point to point in the following paragraphs (the texts italicized are the comments, the texts indented are the responses, and the texts in blue are revised parts in the new manuscript). In addition, all changes made are marked in the revised manuscript.

1.Responses to Comment 2-1 and 2-6: first I do not understand the response to Comment 2-6. Did the authors mean that because the fume was only a little warmer than indoor air, it had already cooled down and condensable gases condensed onto particles? If so, it is unclear to me what role pipe heating really played and the phrase "prevent freshly warmed gas from condensing on the pipe wall" does not seem to be appropriate; otherwise, I am not convinced that pipe heating was sufficient to prevent wall losses/tubing delay of S/IVOCs in the pipe. Obviously, pipe heating could help substantially. It is however unclear whether this measure can reduce wall/tubing effects in the pipe to minor/negligible level unless the authors perform a more quantitative analysis e.g. based on Pagonis et al. (2017).

> Thanks for the reviewer's patience on our manuscript. To be exact, the fume was hot after emission, but was only a bit warmer than the indoor air after our dilution system. As for vehicle experiment, vehicle exhaust from tailpipe was first diluted by a gradient heated dilution system (6 fold) and then diluted by a unheated dilution system (5 fold). The temperature of sample flow was near indoor temperature (20-25℃) after secondary dilution systems. As for cooking experiment, the cooking fumes was collected through the kitchen ventilator, where the temperature was just similar to that of indoor temperature (20-25℃). Compared to the condensation caused by instantaneous temperature difference, our gradient heated dilution system and slight temperature difference (between emission and measurement) could decrease the loss of gaseous compounds. In this article, the main instrument is HR-Tof-AMS which focused on the particle phase, so our sampling tubes were made of stainless steel rather than Teflon tubes (Pagonis et al discussed the wall loss of Teflon tubes). The reviewer 2 has accepted our response and suggested us to supply necessary description in the Method section as following. Thanks for the reviewers again.
>
> "**Revised text in Method 2.1:** "Vehicle exhaust from tailpipe was first diluted by a gradient heated dilution system (6 fold) and then diluted by an unheated dilution system (5 fold). The temperature of sample flow was near indoor temperature (20-25°C) after secondary dilution systems. The cooking fumes was collected through the kitchen ventilator, where the temperature was similar to that of indoor air."

S/I VOCs were indeed partly lost in pipelines, and its sampling and quantification are really hard and challenging, which was actually one of uncertainties of our study. We have added this uncertainty in the Conclusion section.

"**Revised text in Conclusion:** "S/I VOCs may play important roles in forming SOA but were indeed partly lost in pipelines, and its sampling and quantification are really hard and challenging, which needs more sophisticated experimental design."

2.Response to Comment 2-9: the observation that oleic acid ozonolysis made little SOA in Go:PAM does not imply that this reaction cannot produce SOA significantly in the atmosphere. Ozonolysis of oleic acid breaks its long chain and produces higher-volatility products than its oxidation by OH does. It does not surprise me that the ozonolysis products did not condense onto particles during the very short residence time of Go:PAM. However, in the atmosphere, the O3:OH ratio is even higher than in Go:PAM and the products of e.g. oleic acid ozonolysis, particularly organic peroxy radicals, have enough time to undergo several generations of autoxidation to become of sufficiently low volatility and condense. Therefore, the (lack of) contribution of ozonolysis to SOA formation in Go:PAM should be seriously discussed.

We agree with the reviewer that ozonolysis is (likely) important for unsaturated fatty acids that were probably emitted from the cooking sources, but we didn't find the significant effect of ozonolysis in our experiment. This may be because the residence time is too short for ozonolysis reactions. Besides, Vesna et al. found that the heterogeneous reaction of ozone with oleic acid aerosol particles was influenced by humidity and reaction time in an aerosol flow reactor. We have added necessary expressions in Method and Conclusion section.

"**Revised text in Method 2.3.2: "**It is found that the heterogeneous reaction of ozone with oleic acid aerosol particles was influenced by humidity and reaction time in an aerosol flow reactor (Vesna et al., 2009). Therefore, non-OH reactions, such as the ozonolysis of unsaturated fatty acids, may also be important in forming SOA, which missed specific designs in our experiment."

"**Revised text in Conclusion:** "Moreover, contribution of ozonolysis to SOA formation should be individually studied in further research."

References:

Vesna, O., Sax, M., Kalberer, M., Gaschen, A., and Ammann, M.: Product study of oleic acid ozonolysis as function of humidity, Atmospheric Environment, 43, 3662-3669, 10.1016/j.atmosenv.2009.04.047, 2009.

3.In addition, I have 2 major concerns about this paper:- Liao et al. (2021) did OFR experiments of SOA formation from on-road vehicle emissions in Beijing. They kept their experimental conditions high-NOx and *an earlier and relatively low peak of SOA formation (as a function of photochemical age), compared to literature studies of this kind, which were all conducted under low-NOx conditions in OFRs. The peak SOA age in Liao et al. (2021) was only ~1 d, significantly lower than in this study (~2 d). High-NOx pathways in organic peroxy radical chemistry allow fragmentation of organics to occur at lower degree of oxidation than low-NOx pathways. As a result, the compositions of SOA formed through these 2 types of pathways may be significantly different. I thus have doubts whether the SOA formed in Go:PAM in this study is sufficiently representative of SOA formed in urban areas, where NOx is usually high. If the SOA was not representative, the analysis in the triangle plot and the VK diagram, PMF etc. in the paper did not bring much insight. I believe that the authors should justify that the SOA in their experiments were representative of SOA in urban atmospheres.*

We appreciate the reviewer's reasonable concern and careful review. We agree with that different NO_x/VOCs conditions may have significant influence on SOA formation. In urban areas, high/low NO_x conditions may appear alternately, it strongly depends on the place and time of observation. As the review mentioned, Liao et al (2021) conducted the OFR simulation on a busy road of Beijing under high-NO_x condition. However, Wei et al (2014) found that low-NO_x (High VOCs) phenomenon around a refinery in Beijing (as shown in the following figure). A busy road may emit more NO_x, and a refinery may emit more VOCs. High/low NO_x (VOCs/NO_x) greatly depends on the measurement place.

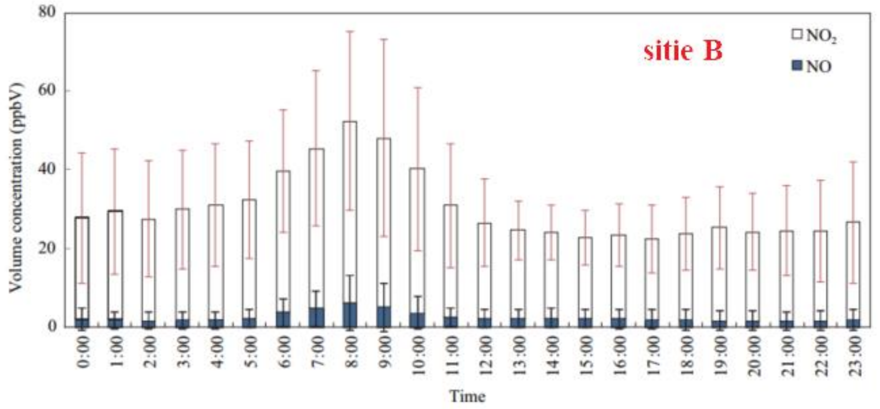


Fig. 5 Hourly mean NOx concentrations in daytime around the refinery.

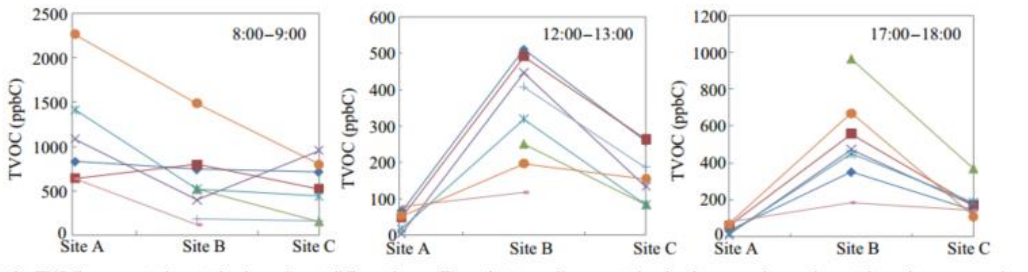
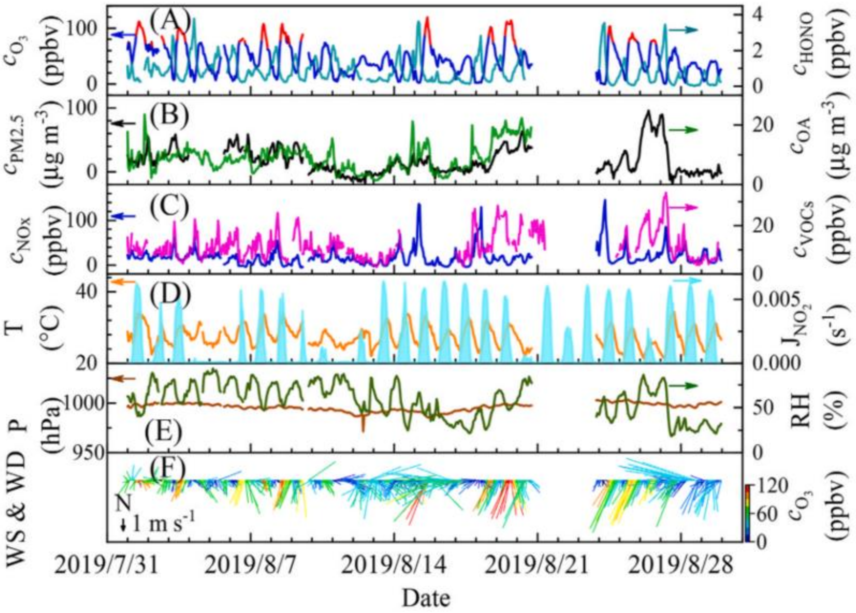


Fig. 6 TVOCs concentrations at the three sites at different hours. The point at one line meant the simultaneous observations at three sites on a monitor city.

Moreover, Zhan et al (2021)'s field study in Beijing showed the timeseries of VOCs and NO_x. The concentration of VOCs was underestimated, because only 57 NMHCs standard materials were used to quantify the C2-C12 NMHC measured by GC-FID. As the figure below shows, the concentration of NO_x was not obviously higher than VOCs all the time, and the concentration of VOCs was not obviously higher than NO_x all the time as well. The NO_x/VOCs condition is dynamic, it depends greatly on the measurement time.



Overall, high/low NO_x condition or other complicated atmospheric conditions are all likely to occur in urban areas, and it greatly depends on the place and time of observation.

There are indeed uncertainties in our laboratory simulation, when we only conducted vehicle experiment under moderate-NO_x condition (the concentration of NO_x is similar to that of VOCs) and cooking experiment under low-NO_x condition (the concentration of NO_x is far lower than that of VOCs). Hence, further researches are still needed. We have added the expression of this uncertainties in Conclusion and Introduction section.

"**Revised text in Conclusion:** "In the future, it'll be better to strictly control the temperature, RH, high/low NO_x or SO_2, additional inorganic seeds, and so forth, to deeply investigate how the aerosol ages as a function of equivalent days of atmospheric oxidation."

"**Revised text in Introduction:** "NO_x levels may greatly influence the chemical evolution of vehicle SOA, and its NO_x/VOCs values are often strongly dependent on the sampling time and place in urban areas (Zhan et al., 2021;Wei et al., 2014). It is found that the photochemical ages for maximum SOA production under high-NO_x levels were lower than those under low-NO_x levels among OFR simulations (Liao et al., 2021)."

References:

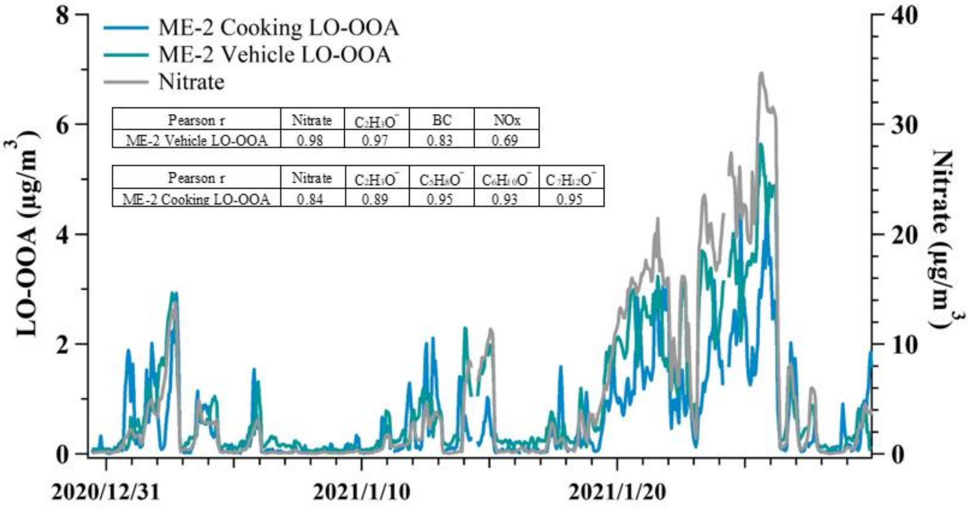
Wei, W., Cheng, S., Li, G., Wang, G., and Wang, H.: Characteristics of ozone and ozone precursors (VOCs and NOx) around a petroleum refinery in Beijing, China, Journal of Environmental Sciences, 26, 332-342, 10.1016/s1001-0742(13)60412-x, 2014.

Zhan, J., Feng, Z., Liu, P., He, X., He, Z., Chen, T., Wang, Y., He, H., Mu, Y., and Liu, Y.: Ozone and SOA formation potential based on photochemical loss of VOCs during the Beijing summer, Environmental pollution, 285, 117444, 10.1016/j.envpol.2021.117444, 2021.

Liao, K., Chen, Q., Liu, Y., Li, Y. J., Lambe, A. T., Zhu, T., Huang, R.-J., Zheng, Y., Cheng, X., Miao, R., Huang, G., Khuzestani, R. B., and Jia, T.: Secondary Organic Aerosol Formation of Fleet Vehicle Emissions in China: Potential Seasonality of Spatial Distributions, Environ. Sci. Technol., 55, 7276–7286, 2021.

4.In a number of places in the paper, the discussions about SOA formation from cooking emissions seemed to be based on heterogeneous oxidation of POA. While I understand that this pathway may play some role, it is slow enough to be minor at photochemical ages of 1-3 days compared to gas-phase oxidation of cooking emissions, a large fraction of which is S/IVOC with C=C bonds. The authors should discuss how cooking SOA is formed from S/IVOC oxidation in the gas phase.

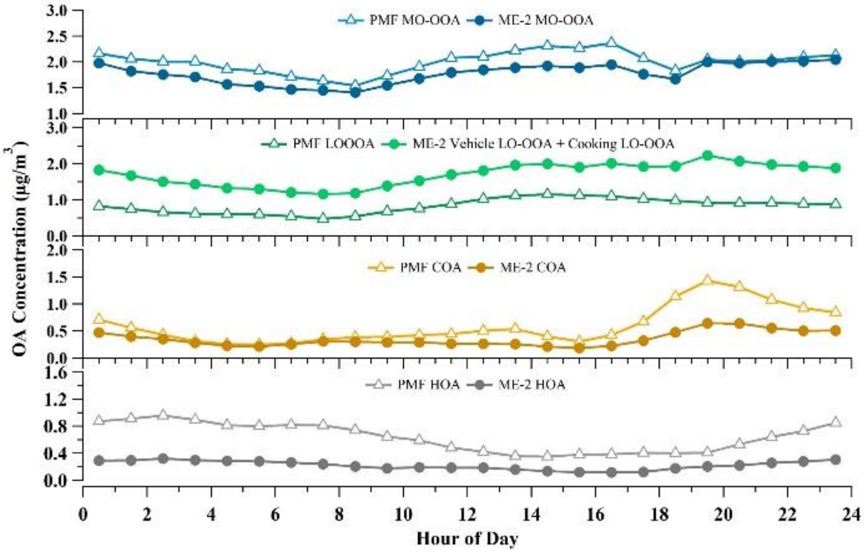
Thanks for the constructive comments. This resubmitted manuscript focused more on particle phase, especially its SOA growth factor, O/C, oxidation pathway and mass spectra. We have compared cooking and vehicle SOA mass spectra with ambient SOA spectra in Beijing (Discussion 3.4). In the future, the cooking and vehicle SOA mass spectra could be used as the input mass spectra in ME-2, in order to estimate the mass concentration of cooking SOA and vehicle SOA, and then study their chemical evolution in ambient. We are designing another article to study the application of our SOA mass spectra (cooking and vehicle) in urban areas. As following figures show, here is one practice in Beijing(unpublished):



Pearson r	Nitrate	$C_2H_3O^-$	BC	NOx
ME-2 Vehicle LO-OOA	0.98	0.97	0.83	0.69

Pearson r	Nitrate	$C_2H_3O^-$	$C_5H_8O^-$	$C_6H_{10}O^-$	$C_7H_{12}O^-$
ME-2 Cooking LO-OOA	0.84	0.89	0.95	0.93	0.95

Timeseries of OA factors and good correlations with relevant species



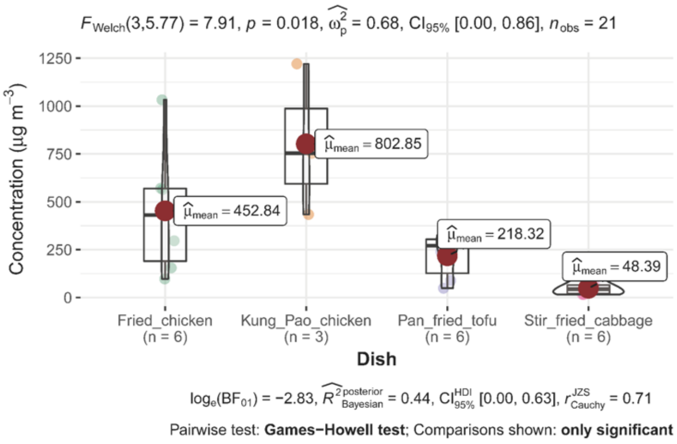
Reasonable diurnal variations of OA factors

Therefore, the results in this article are valuable and could be used to do further estimation in ambient. We regarded this article as a beginning of our "source secondary simulation" research. We will try our best to give more informative results in other new articles as soon as possible.

We agree with the reviewer's comment that the gaseous results are important. In this article, we just use simple VOCs results from GC-MS in order to estimate the OHR. As for the further measurement and analysis results of gaseous phase, we have regarded it as another independent and attractive part. In fact, we have measured the VOCs and S/I VOCs by 2D-GC-MS, VOCUS and CIMS during Go: PAM experiments, but their sampling, quantification and data analysis are so complicated and different from particle phase, that we need to write individual articles to show our findings. Thanks for the reviewer's helpful advice again. As the following figures show, here are some initial results of cooking experiment (unpublished). We really hope our findings would benefit other researcher's studies in the future.

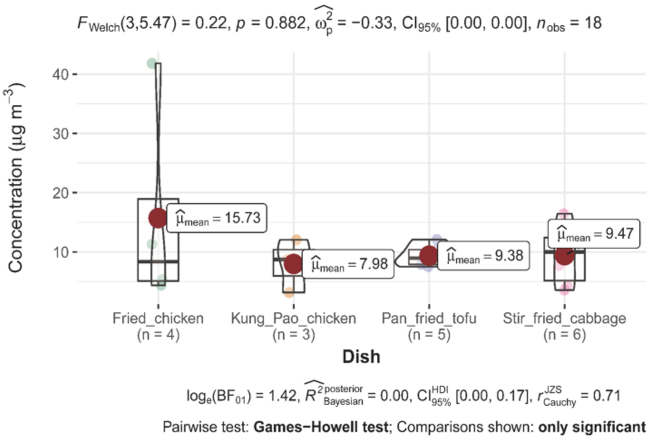
Gas-phase

- Significant difference between four dishes in gas-phase total S/I VOCs ($p < 0.05$)
- Kung Pao chicken & Pan-fried tofu: large proportion of aromatics
- Furans were abundant in gas-phase oxygenated compounds (> 6%)



Particle-phase

- NO significant difference between four dishes in particle-phase total S/IVOCs ($p > 0.8$)
- Oxygenated compounds were the most abundant (>40%), including acids, furans, amides and esters



5.Response to Comment 1-9: I agree that Manchester is not a megacity. But this would be better determined by the total population of its metro area (~3 M) than by that of the city proper (~0.5 M).

We agree with this determination. Thanks for the suggestion.

“**Revised text in Introduction:** “take the megacity (total population of its metro area is more than 3 M) for example”

6.Response to Comment 2-2: to me, the authors seemed to misunderstand the Reviewer's point here. The Reviewer likely meant that low humidity would lead to highly viscous particles, which usually undergo slower heterogeneous oxidation than less viscous particles.

We agree with that low humidity would lead to highly viscous particles, which usually undergo slower heterogeneous oxidation than less viscous particles. This is the uncertainty of our experiment. Therefore, it's better to control the RH in the same level in the future studies.

Relative humidity can influence the photochemical or aqueous-phase processing of SOA, it is stated that the aerosol liquid water content may show a linear increase as a function of RH (at RH > 60%) during the three seasons in Beijing, indicating the potential impacts of aqueous-phase processing at high RH levels (Xu et al., 2017). Therefore, RH 18-23% or RH 44-49% are both relatively low, and photochemical oxidation may still play the leading role in these two experiments. In the future, we really hope we could strictly control the temperature, RH, and other conditions to deeply investigate how the aerosol ages as a function of equivalent days of atmospheric oxidation, under certain gas-phase and heterogeneous oxidations.

According to the reviewers' comments, We have added the necessary description in the Method and Conclusion section as the following text shows:

“**Revised text in Method 2.3.2:** “The Go: PAM conditions for vehicle and cooking experiments could be seen in Tables 3 and Table 4, respectively. Their experiment conditions (such as residence time and RH ranges) were not completely the same because of the inherent difference and experimental design between two sources. Whereas, some comparisons could be still analyzed in the similar OH exposure, and their RH conditions were both low where photochemical oxidations instead of aqueous-phase processing still dominated the chemical evolution process (Xu et al., 2017)."

"**Revised text in Conclusion:** "There are some uncertainties of our Go: PAM simulation. We focused more on the photochemical oxidation of SOA under low RH levels, but aqueous-phase processing at high RH levels may also have impacts to SOA production. In the future, it'll be better to strictly control the RH, high/low NO_x or SO_2, additional inorganic seeds, and so forth, to deeply investigate how the aerosol ages as a function of equivalent days of atmospheric oxidation."

References:

Xu, W., Han, T., Du, W., Wang, Q., Chen, C., Zhao, J., Zhang, Y., Li, J., Fu, P., Wang, Z., Worsnop, D. R., and Sun, Y.: Effects of Aqueous-Phase and Photochemical Processing on Secondary Organic Aerosol Formation and Evolution in Beijing, China, Environmental science & technology, 51, 762-770, 10.1021/acs.est.6b04498, 2017.

7.A lot of text in Sections 2.3.2 and S2 looks too similar. Some of it may be cut to reduce redundancy.

Thanks for the reviewer's comment. Some of the contents in S2 have been cut to reduce redundancy.

8.Line 98: a dash is needed between "Beijing" and "Chengde".

Thanks for the correction. We have revised it to "Beijing-Chengde".

9.Ling 196: "Hongkong" -> "Hong Kong".

Thanks for the correction. We have revised it to "Hong Kong".

References:

Liao, K., Chen, Q., Liu, Y., Li, Y. J., Lambe, A. T., Zhu, T., Huang, R.-J., Zheng, Y., Cheng, X., Miao, R., Huang, G., Khuzestani, R. B., and Jia, T.: Secondary Organic Aerosol Formation of Fleet Vehicle Emissions in China: Potential Seasonality of Spatial Distributions, Environ. Sci. Technol., 55, 7276–7286, 2021.

Pagonis, D., Krechmer, J. E., de Gouw, J., Jimenez, J. L., and Ziemann, P. J.: Effects of gas–wall partitioning in Teflon tubing and instrumentation on time-resolved measurements of gas-phase organic compounds, Atmos. Meas. Tech., 10, 4687–4696, 2017.