

## *Responses to Comments*

We appreciate the constructive comments from the reviewers on this manuscript. We have answered the reviewers' 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.

### *I. Comments from Reviewer 2*

*1. In the methods and conclusions sections, please add a couple sentences referencing companion publications-in-preparation that discuss the speciated gas- and condensed-phase compounds and their individual SOA formation potentials.*

We appreciate the reviewer's comment. We have added necessary description referencing the companionate publications-in-preparation in the Conclusion section.

**“Revised text in Conclusion:** In another companionate publication-in-preparation (Song Kai, Guo Song\*, *et al.*, Cooking emitted S/IVOCs are a large pool of SOA formation precursors, *In preparing*), gas and particle phase VOCs and S/IVOCs from four typical Chinese domestic cuisines are quantified. It is found that 26-78% of cooking SOA could be explained from the oxidation of VOCs and S/IVOCs. Moreover, oxygenated compounds were the most abundant in particle phase, including acids, furans, amides and esters. In contrast, significant differences were found in gas phase among four cuisines, for example, Kung Pao Chicken and shallow-frying Tofu showed larger proportion of aromatics.”

Besides, the mass spectra that we obtained could be used in the ambient air, and we referred it briefly in the Conclusion section.

**“Revised text in Conclusion:** The contribution of vehicle SOA and cooking SOA for OA were estimated by ME-2 model in urban Beijing (Zhang Zirui, Hu Min\*, *et al.* Secondary Organic Aerosol Formation from Urban Lifestyle Sources in Beijing. *In preparing*). It is found that cooking SOA (27-42% of OA) and vehicle SOA (58-73% of OA) presented different diurnal patterns, implying their different formation pathways. Similar features of urban lifestyle SOA were found between laboratory and field results.”

## II. Comments from Reviewer 3

1. The authors have adequately addressed most of the reviewers' comments in this round of revision. However, I disagree with a point in their response to one of my comments (Response 3): High/Low NO<sub>x</sub> conditions should not be distinguished by VOC-to-NO<sub>x</sub> ratio, but by the fate of organic peroxy radicals (see e.g. Wennberg (2013)). Regardless of VOC concentrations, organic peroxy radicals react dominantly with NO as long as the latter's concentration is at ppb level. If the authors intend to show the variability of high/low NO<sub>x</sub> regimes in the ambient air in Beijing, it needs to be based on organic peroxy loss pathways.

Thanks for the reviewer's constructive and professional comments. We agree with the reviewer. It's better to distinguish high/low NO<sub>x</sub> conditions by the fate of organic peroxy radical than VOCs-to-NO<sub>x</sub> ratio. Frankly, high-NO<sub>x</sub> pathways in organic peroxy radical chemistry may allow fragmentation of organics to occur at lower degree of oxidation than low-NO<sub>x</sub> pathways. However, the measurements of RO<sub>2</sub>, VOCs or S/I VOCs components are usually uncomplete and full of uncertainties. Therefore, VOCs-to-NO<sub>x</sub> ratio were widely used to estimate SOA formation condition in laboratory simulation and field studies (Liu, et al., ACP, 2018). Even so, relatively simple laboratory results also have many uncertainties when it was applied in the ambient air, considering the extremely complicated ambient conditions.

In summary, the variability of high/low NO<sub>x</sub> regimes in the ambient air in Beijing were not described in the manuscript. This is actually one of uncertainties of our research. We greatly appreciate the reviewer's comments that have pointed out the uncertainty of our research. We have added the related description of many uncertainties in Conclusion method, and also the potential application of our laboratory results.

**Revised text in Conclusion:** "In the future, it'll be better to strictly control the temperature, RH, high/low NO<sub>x</sub> or SO<sub>2</sub>, 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 Conclusion:** The contribution of vehicle SOA and cooking SOA for OA were estimated by ME-2 model in urban Beijing (Zhang Zirui, Hu Min\*, et al. Secondary Organic Aerosol Formation from Urban Lifestyle Sources in Beijing. *In preparing*). It is found that cooking SOA (27-42% of OA) and vehicle SOA (58-73% of OA) presented different diurnal patterns, implying their different formation pathways. Similar features of urban lifestyle SOA were found between laboratory and field results."

### References:

Liu, T., Wang, Z., Wang, X., and Chan, C. K.: Primary and secondary organic aerosol from heated cooking oil emissions, *Atmospheric Chemistry and Physics*, 18, 11363-11374, 10.5194/acp-18-11363-2018, 2018.

2. Regarding Response 4, while it is fine to focus on the particle phase, the manuscript should not leave to readers an impression that cooking SOA is mainly from heterogeneous oxidation of POA unless the authors show the evidence.

Thanks for the reviewer's helpful advice. We are sorry to cause this misunderstanding. The simulated SOA could be generated by the photochemical oxidation from gaseous precursors and the heterogeneous oxidation from POA. In this work, we couldn't clearly analyze the relative strength of gaseous photochemistry or that of the heterogeneous chemistry. We have added necessary expressions in the start of Discussion section and the uncertainty part of Conclusion section.

**“Revised text in Discussion 3.1:** The simulated SOA could be generated by the photochemical oxidation from gaseous precursors and the heterogeneous oxidation from POA.”

**“Revised text in Conclusion:** Furthermore, the relative strength of the photochemical oxidation from gaseous precursors and the heterogeneous oxidation from POA were not deeply distinguished in this work.”

3. For Response 1, I think that Deming et al. (2020) is an appropriate reference for the discussion about stainless steel tubing loss/delay.

Thanks for the careful review. We agree with the reviewer that it's important to evaluate the pipe loss in order to ensure the preciseness of experimental design. The reviewer has recommended an article for us (Deming et al., 2012). They expand recent results by comparing different types of Teflon and other polymer tubing, as well as glass, uncoated and coated stainless steel and aluminum, and other tubing materials by measuring the response to step increases and decreases in organic compound concentrations. It is found that conductive PFA tubing and silonite are the best choices for simultaneous gas and particle sampling. In our work, all stainless steel pipes are silanized, which fully meet the needs of particulate and gas sampling. In order to eliminate misunderstandings, we have added the word “silanized” before the stainless steel in the manuscript. In addition, we have added the evaluation of particle penetrating fraction according to the equivalent pipe length method in SI (Wiedensohler et al., 2012).

**“Revised text in Method section:** All the sampling tubes are made of silanized stainless steel which is appropriate for a simultaneous gas and particle sampling (Deming et al., 2019; Wiedensohler et al., 2012).”

**“Revised text in SI Section S1:** There were 5 meters long from the sampling port to the Go: PAM, the flow rate was 5.5 L/min (HR-ToF-AMS and other instruments jointly determined the flow rate), **and the penetrating fraction was more than 90% for those particles whose diameter was larger than 10 nm (equivalent pipe length method) (Wiedensohler et al., 2012).** There were 2 meters long from the Go: PAM to the HR-ToF-AMS, the flow rate was 1

L/min (HR-ToF-AMS and its drainage system determined the flow rate), **and the penetrating fraction was more than 88% for those particles whose diameter was larger than 10 nm (equivalent pipe length method) (Wiedensohler et al., 2012).**

#### References:

Wiedensohler, A., Birmili, W., Nowak, A., Sonntag, A., Weinhold, K., Merkel, M., Wehner, B., Tuch, T., Pfeifer, S., Fiebig, M., Fjåraa, A. M., Asmi, E., Sellegri, K., Depuy, R., Venzac, H., Villani, P., Laj, P., Aalto, P., Ogren, J. A., Swietlicki, E., Williams, P., Roldin, P., Quincey, P., Hüglin, C., Fierz-Schmidhauser, R., Gysel, M., Weingartner, E., Riccobono, F., Santos, S., Grüning, C., Faloon, K., Beddows, D., Harrison, R., Monahan, C., Jennings, S. G., O'Dowd, C. D., Marinoni, A., Horn, H. G., Keck, L., Jiang, J., Scheckman, J., McMurry, P. H., Deng, Z., Zhao, C. S., Moerman, M., Henzing, B., de Leeuw, G., Löschau, G., and Bastian, S.: Mobility particle size spectrometers: harmonization of technical standards and data structure to facilitate high quality long-term observations of atmospheric particle number size distributions, *Atmospheric Measurement Techniques*, 5, 657-685, 10.5194/amt-5-657-2012, 2012.

References given by Reviewer 3: Deming, B. L., Pagonis, D., Liu, X., Day, D. A., Talukdar, R., Krechmer, J. E., de Gouw, J. A., Jimenez, J. L., and Ziemann, P. J.: Measurements of delays of gas-phase compounds in a wide variety of tubing materials due to gas-wall interactions, *Atmos. Meas. Tech.*, 12, 3453–3461, 2019.

Wennberg, P. O.: Let's abandon the “high NO<sub>x</sub>” and “low NO<sub>x</sub>” terminology, *IGAC News*, 50, 3–4, 2013.