Response to comments on " The effect of COVID-19 restrictions on atmospheric new particle formation in Beijing"

We thank the reviewers for their time, efforts, and constructive comments. We provide our point-to-point replies to these comments below. The comments by reviewers are in black, and the replies to the comments are in blue. The corresponding changes are noted in the manuscript and Supplementary Data with the same color code. All references are provided at the end of the replies.

Referee 1

There is a current debate on the driving mechanism(s) of NPF in urban environments, more specifically, sulfuric acid-amine clustering or oxidized organics originating from traffic emissions. The lockdown during COVID-19 pandemic provided a unique chance to definitively resolve this issue. Very simply put, if the strength of NPF reduced significantly during the lockdown, the dominant role of traffic emissions can be confirmed. A recent paper by Shen et al., (2021), that is also cited in this study, showed that NPF was stronger during the lockdown, which may suggest the less important role of traffic emissions in NPF in Beijing. In this study, the authors show consistent observational results with what have been reported by Shen et al., (2021), and further extended the mechanistic understanding of such NPF enhancement by performing detailed molecule-level analyses on NPF precursors, i.e., sulfuric acid and oxygenated organics. The authors found that the enhanced NPF were an overall result of two facts: first, the sulfuric acid-amine clustering remained as the driving mechanism and led to a similar $J_{1.5}$; second, the growth and survival of very small particles were enhanced by the elevated abundance of condensable oxidized organics.

Overall, I think this paper presents a significant advance in the understanding of NPF in urban environments, and thus I recommend accepting it for publication with a few minor comments/questions that I hope the authors can answer:

1. Line 216 - 218 "In addition, the concentration of OOMs increased by about 50% during the lockdown. This is because the concentration of volatile organic compounds (VOCs) only declined slightly in the lockdown period (Shen et al., 2021b), but the photochemistry was much more enhanced."
The overall pollution level was more serve during lockdown period. So will some of the OOMs be able to transport from other region(s) to the measurement site along with $PM_{2.5}$, and thus leading to the enhancement of OOM concentration?
Response: Thanks for the comments. The transport of OOMs depends on their atmospheric lifetimes. Firstly, OOMs with very low volatility are not expected to transport across long distances, because they would be lost on particles via condensation during the transport. In

Fig.R1, we show the volatility distribution of OOMs in this study, where ULVOCs, ELVOCs, LVOCs accounted for 5.2 %, 25.1 %, and 35.8 %, respectively. Secondly, the rest, relatively highly volatile OOMs (i.e., SVOCs, IVOCs, and VOCs) could partially survive from long-range transport. Yet, they may also lose via multi-generation of oxidations. Currently, limited information is available for the oxidation rates between these OOMs and oxidants, and therefore, it is hard to evaluate the fraction of OOMs lost via further oxidation during the transport. In summary, we can conclude that a minimum of ~ 2/3 of OOMs are formed locally. In addition, it is worth mentioning that the precursor of OOMs, i.e., various VOCs, are most likely transported from other regions. Therefore, the abundance of OOMs depends on both on the strength of photochemistry and air mass origins.
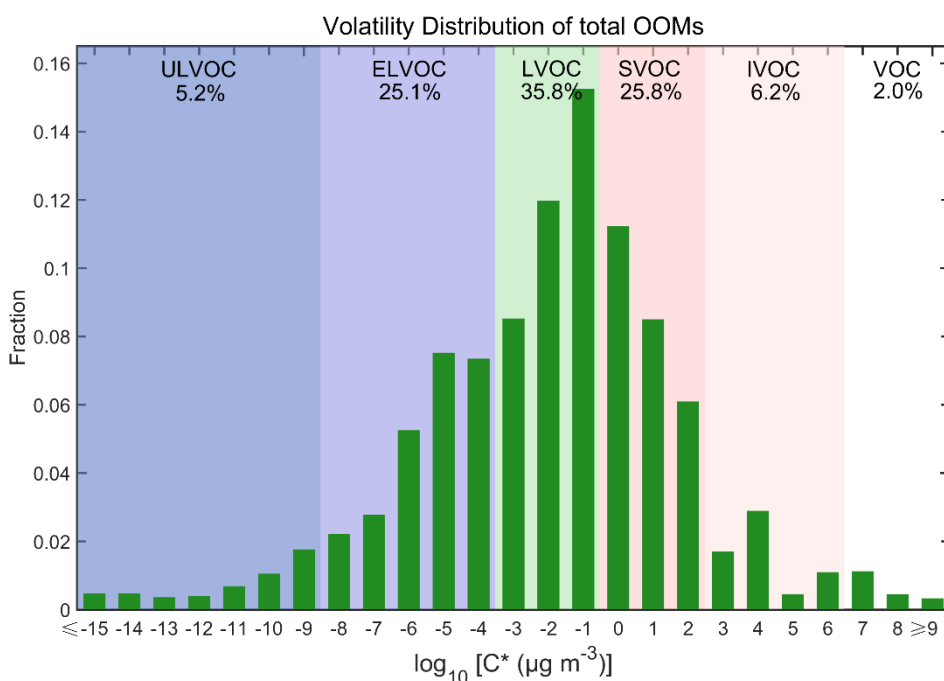

Fig. R1. Volatility distribution of total OOMs. ULVOC, ELVOC, LVOC, SVOC, IVOC and VOC denote ultra-low volatility organic compound, extremely low volatility organic compound, low volatility organic compound, semi-volatile organic compounds, intermediate volatility organic compound, and volatile organic compound respectively.

2. Line 285 "… range of our observations (Fig. S6)." Is this Fig. S6 should be Fig. S5?
Thanks. This has been corrected.
3. Line 291 "periods (Fig. S7). This is less than …" Is this Fig. S7 should be Fig. S6?
Thanks. This has been corrected.
4. Line 334-336 "When the NO concentration declined from the pre-lockdown period to the lockdown period, the ratio of $C_{6-9}H_{7,9,11,13}O_6N$ concentration to $C_{6-9}H_{7,9,11,13}O_5$ concentration decreased as well." Will the photolysis of nitrogen-containing aromatic OOMs influence the ratio

of $C_{6-9}H_{7,9,11,13}O_6N$ concentration to $C_{6-9}H_{7,9,11,13}O_5$ Concentration? And what will happen if color Fig. 5 (A) with UVB?

Response: Thanks for the comment. Photolysis is a loss pathway of some organic nitrates. As suggested, we colored Fig. 5 by UVB and the results are shown in Fig. R2.

It can be found that in all four panels, data points with different UVB values are well mixed, showing no clear dependence on the UVB level. This suggests that the UVB in wintertime Beijing is not a controlling factor that can substantially influence the ratio between nitrogen-containing and nitrogen-free OOMs.
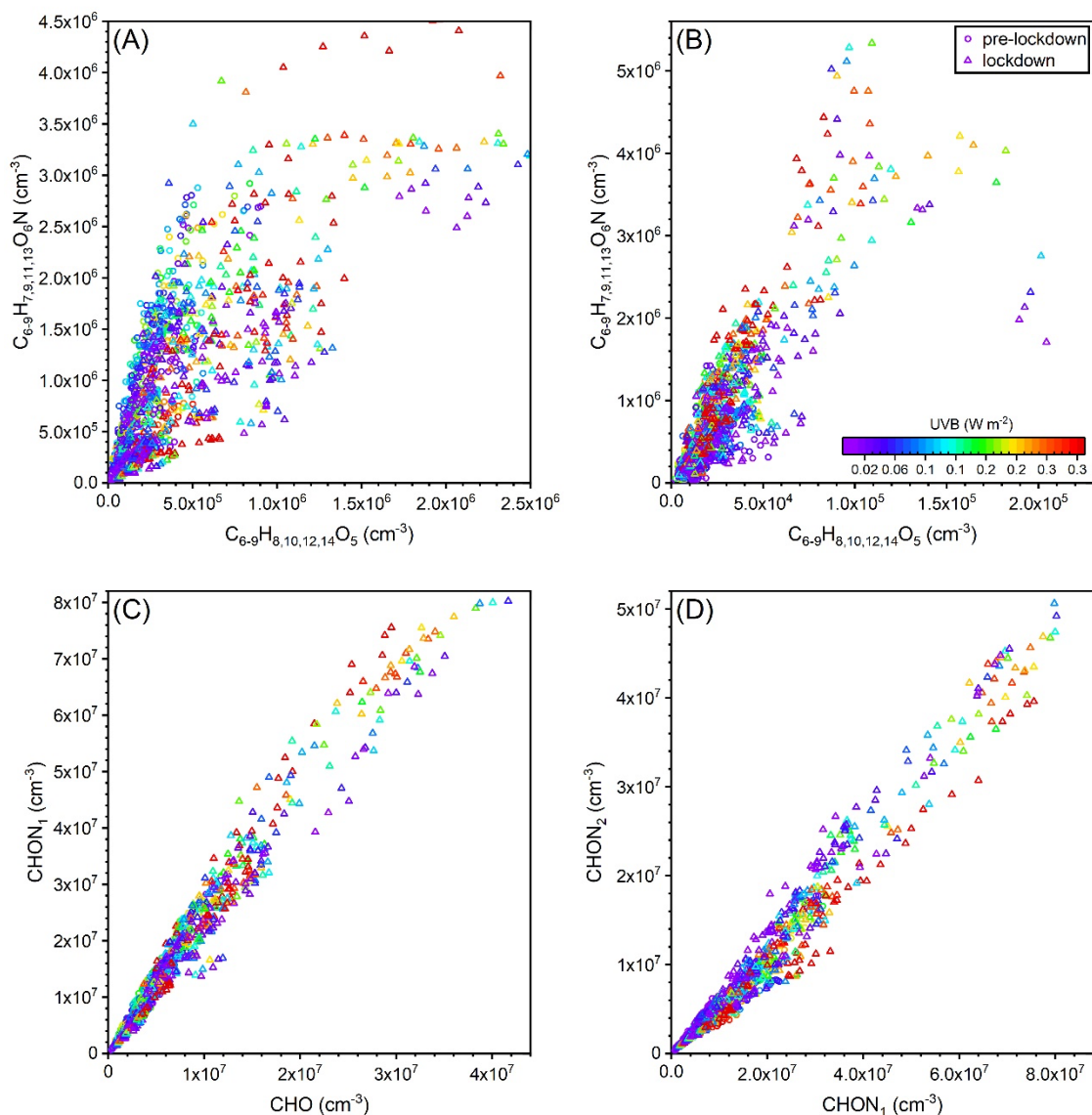

Fig. R2. The influence of UVB (given by symbol color) on the composition of OOMs, indicated by the ratio between nitrogen-containing and nitrogen-free OOMs. (A) Selected OOMs with a double-bond-equivalent (DBE) of 3, which are usually products from the oxidation of aromatic compounds (Molteni et al., 2018; Wang et al., 2017; Garmash et al., 2020). (B) Selected OOMs

with a DBE of 1, which are more likely formed from the oxidation of aliphatic compounds, such as alkenes and alkanes. (C) OOMs containing 0 (CHO) and 1 (CHON$_1$) nitrogen atom. (D) OOMs containing 1 and 2 (CHON$_2$) nitrogen atoms. In all panels, only daytime data (7:00 – 18:00) with UVB higher than 0.001 W m^{-2} were included. Circles and triangles represent data in pre-lockdown and lockdown periods, respectively.

5. Line 340 – 342 "They have a double bond equivalent (DBE) of 1, suggesting that they originate from aliphatic rather than aromatic precursors"
It seems that the authors have some criteria to infer the VOC precursor of OOMs. Is this based on some published results? I would like the authors to reply with more details.
Response: Yes, this is based on our previous studies (Nie et al., 2022; Guo et al., 2022). Nie et al. (2022) developed a novel workflow to retrieve the sources of OOMs. This workflow takes into account the composition of precursors and the current knowledge of atmospheric reaction pathways, which uses carbon number, oxygen number, nitrogen number, and the value of double-bond-equivalence (DBE) for the classification. This workflow classifies all OOMs with DBE≤1 and some OOMs with DBE=2 as aliphatic OOMs. This classification is also justified by various laboratory studies: neither monoterpenes (Ehn et al., 2012; Jokinen et al., 2014; Praplan et al., 2015; Boyd et al., 2015; Berndt et al., 2016) nor aromatics (Molteni et al., 2018; Wang et al., 2017; Garmash et al., 2020) could produce OOMs with DBE≤1. Thus, precursor VOCs with low DBE values are reasonable candidates, such as alkanes and alkenes.

References
Berndt, T., Richters, S., Jokinen, T., Hyttinen, N., Kurtén, T., Otkjær, R. V., Kjaergaard, H. G., Stratmann, F., Herrmann, H., Sipilä, M., Kulmala, M., and Ehn, M.: Hydroxyl radical-induced formation of highly oxidized organic compounds, Nature Communications, 7, 13677, 10.1038/ncomms13677, 2016.
Boyd, C. M., Sanchez, J., Xu, L., Eugene, A. J., Nah, T., Tuet, W. Y., Guzman, M. I., and Ng, N. L.: Secondary organic aerosol formation from the beta-pinene+NO3 system: effect of humidity and peroxy radical fate, Atmospheric Chemistry and Physics, 15, 7497-7522, 10.5194/acp-15-7497-2015, 2015.
Ehn, M., Kleist, E., Junninen, H., Petäjä, T., Lönn, G., Schobesberger, S., Dal Maso, M., Trimborn, A., Kulmala, M., Worsnop, D. R., Wahner, A., Wildt, J., and Mentel, T. F.: Gas phase formation of extremely oxidized pinene reaction products in chamber and ambient air, Atmospheric Chemistry and Physics, 12, 5113-5127, 10.5194/acp-12-5113-2012, 2012.
Garmash, O., Rissanen, M. P., Pullinen, I., Schmitt, S., Kausiala, O., Tillmann, R., Zhao, D., Percival, C., Bannan, T. J., Priestley, M., Hallquist, A. M., Kleist, E., Kiendler-Scharr, A., Hallquist, M., Berndt, T., McFiggans, G., Wildt, J., Mentel, T., and Ehn, M.: Multi-generation OH oxidation as a source for highly oxygenated organic molecules from aromatics, Atmospheric Chemistry and Physics, 20, 515-537, 10.5194/acp-20-515-2020, 2020.

Guo, Y., Yan, C., Liu, Y., Qiao, X., Zheng, F., Zhang, Y., Zhou, Y., Li, C., Fan, X., Lin, Z., Feng, Z., Zhang, Y., Zheng, P., Tian, L., Nie, W., Wang, Z., Huang, D., Daellenbach, K. R., Yao, L., Dada, L., Bianchi, F., Jiang, J., Liu, Y., Kerminen, V. M., and Kulmala, M.: Seasonal Variation of Oxygenated Organic Molecules in Urban Beijing and their Contribution to Secondary Organic Aerosol, Atmos. Chem. Phys. Discuss., 2022, 1-33, 10.5194/acp-2022-181, 2022.

Jokinen, T., Sipilä, M., Richters, S., Kerminen, V.-M., Paasonen, P., Stratmann, F., Worsnop, D., Kulmala, M., Ehn, M., Herrmann, H., and Berndt, T.: Rapid Autoxidation Forms Highly Oxidized RO2 Radicals in the Atmosphere, Angewandte Chemie International Edition, 53, 14596-14600, https://doi.org/10.1002/anie.201408566, 2014.

Molteni, U., Bianchi, F., Klein, F., El Haddad, I., Frege, C., Rossi, M. J., Dommen, J., and Baltensperger, U.: Formation of highly oxygenated organic molecules from aromatic compounds, Atmospheric Chemistry and Physics, 18, 1909-1921, 10.5194/acp-18-1909-2018, 2018.

Nie, W., Yan, C., Huang, D. D., Wang, Z., Liu, Y., Qiao, X., Guo, Y., Tian, L., Zheng, P., Xu, Z., Li, Y., Xu, Z., Qi, X., Sun, P., Wang, J., Zheng, F., Li, X., Yin, R., Dallenbach, K. R., Bianchi, F., Petäjä, T., Zhang, Y., Wang, M., Schervish, M., Wang, S., Qiao, L., Wang, Q., Zhou, M., Wang, H., Yu, C., Yao, D., Guo, H., Ye, P., Lee, S., Li, Y. J., Liu, Y., Chi, X., Kerminen, V.-M., Ehn, M., Donahue, N. M., Wang, T., Huang, C., Kulmala, M., Worsnop, D., Jiang, J., and Ding, A.: Secondary organic aerosol formed by condensing anthropogenic vapours over China's megacities, Nature Geoscience, 15, 255-261, 10.1038/s41561-022-00922-5, 2022.

Praplan, A. P., Schobesberger, S., Bianchi, F., Rissanen, M. P., Ehn, M., Jokinen, T., Junninen, H., Adamov, A., Amorim, A., Dommen, J., Duplissy, J., Hakala, J., Hansel, A., Heinritzi, M., Kangasluoma, J., Kirkby, J., Krapf, M., Kürten, A., Lehtipalo, K., Riccobono, F., Rondo, L., Sarnela, N., Simon, M., Tomé, A., Tröstl, J., Winkler, P. M., Williamson, C., Ye, P., Curtius, J., Baltensperger, U., Donahue, N. M., Kulmala, M., and Worsnop, D. R.: Elemental composition and clustering behaviour of α-pinene oxidation products for different oxidation conditions, Atmos. Chem. Phys., 15, 4145-4159, 10.5194/acp-15-4145-2015, 2015.

Wang, S., Wu, R., Berndt, T., Ehn, M., and Wang, L.: Formation of Highly Oxidized Radicals and Multifunctional Products from the Atmospheric Oxidation of Alkylbenzenes, Environmental Science & Technology, 51, 8442-8449, 10.1021/acs.est.7b02374, 2017.