Responses to Reviewers' Comments on Manuscript egusphere-2023-809

(Seasonal variations in composition and sources of atmospheric ultrafine particles in urban Beijing based on near-continuous measurements)

We are grateful for the reviewers' comments and we feel that our responses to these have improved this manuscript. We have addressed the comments in the following paragraphs and made corresponding changes in the revised manuscript. Comments are shown as *blue italic text* followed by our responses. Changes are highlighted in the revised manuscript and shown as "quoted underlined text" in our responses.

Reviewer #1:

This is a well written paper on the sources and growth of ultrafine particles in urban air. Chemical composition measurements assist the identification of UFP sources, which include both primary emissions and new particle formation. Aqueous/heterogeneous growth of preexisting particles is also indicated from chemical composition data. Measured particle growth rates (3-50 nm) during new particle formation are consistent with theoretical growth rates estimated from condensation of gas-phase sulfuric acid and low-volatility organic compounds.

Specific comments:

In the next to the last sentence of Section 3.3, the authors estimate the times needed for sub-3 nm particles to grow to above 50 and 100 nm. The authors do not explicitly state how these times are calculated, but I am assuming that they are based on the measured and/or estimated 3-50 nm growth rates. If this is the case, then the times they give to grow above 50 nm are accurate, but the times they give to grow above 100 nm are likely to be a substantial overestimate. Figure S12 shows that the CHON/S composition factor associated with aqueous/heterogeneous chemistry becomes very large above 50 nm, roughly equaling that of the CHO-rich factor which presumably includes (butis not limited to) condensation of low volatility organics. Therefore, it is likely that particle growth due to aqueous/heterogeneous chemistry equals or exceeds particle growth due to condensation alone. If both growth channels are taken into account, the actual time to grow to 100 nm is likely to be much shorter than what the authors report. I encourage the authors to consider this possibility (or to clarify how the times were calculated) and discuss appropriately in the body of the paper.

Response: We agree with the reviewer that the time estimated for <3 nm particles to grow above 100 nm was possibly overestimated. The time was estimated from the measured average $GR_{3\text{-}50}$, the latter of which is derived from the observed particle size distribution, and we used $GR_{3\text{-}50}$ for the estimation of the size range between 3 and 100 nm. This would probably overestimate the time needed to grow above 100 nm, as our data have suggested growing GR with increasing d_p. The increasing contribution from aqueous/heterogeneous chemistry above 50 nm also suggests that $GR_{50\text{-}100}$ are larger than $GR_{3\text{-}50}$. However, it is difficult to determine $GR_{50\text{-}100}$ from the measured size distribution for most of the observed NPF events. The main reason is that the new particle formation events was usually disturbed during ~16:00-20:00 by primary particle emission (cooking and vehicle emission) and the decrease of boundary layer. Before 16:00-20:00, the particle mode diameters usually cannot grow above ~50 nm. Furthermore, when particles grow to ~70-80 nm, they start to mix with the background particles. These influence associated with an inhomogeneous environment made it difficult to retrieve $GR_{50\text{-}100}$ from the measured particle size distributions.

In the revised manuscript, we removed the time needed to grow above 100 nm (as it is hard to estimate). The sentence is revised as:

[Line 383-385] Under the observed average $GR_{3\text{-}50}$, the time needed for sub-3 nm to grow above 50 nm is the shortest in summer (~9 h, respectively) and the longest in winter (~24 h).

A related question: What is the approximate survival probability of a sub-3 nm particle growing to 50 or 100 nm without being removed by a typical loss process? It seems the authors dataset is robust enough to provide an estimate.

Response: We thank the reviewer for this inspiring comment. Based on case-by-case analysis of previous dataset from the same measurement site, we estimated that the median survival probability of a 3 nm particle growing to 100 nm was ~1 %, with good consistency between the measurement and theory of growth and coagulation scavenging (Cai et al., 2022; Li et al., 2022). For example, the survival probability of the 8 nm particles to 50 nm is ~0.8% during the new particle formation and growth events in Oct. 2nd, 2020. However, not every NPF event has such a clear growth shape. The calculation of survival probability of new particles in each NPF events needs to carelly isolate the newly formed particles from the background particles, and batch calculations require plenty of surpervised work. The seansonal variations of CS and GR shown in Fig. 5 in this manuscript indicate that the survival probability may vary with seasons. Combining careful analysis of every NPF events and our robust dataset, we think it will be an interesting future work to provide an estimate of survival probability under different environmental conditions (e.g. seasons).

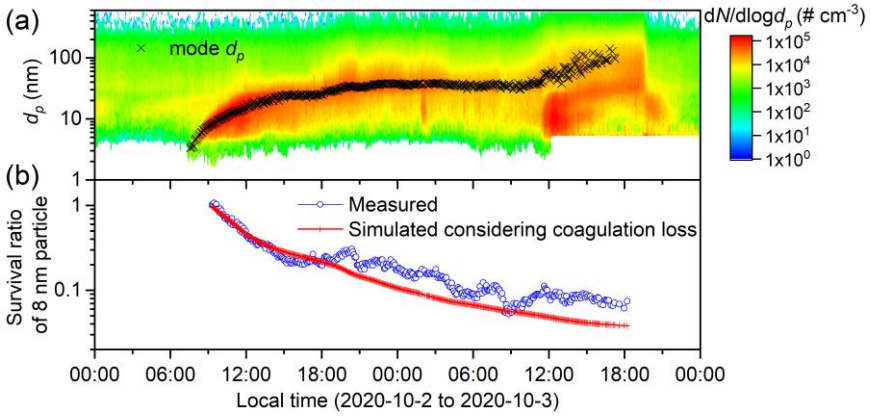

Figure R1. The growth and survival of new particles in Beijing, October 2, 2020. (a) Size distributions of particles and the mode diameters of new particles from October 2, 2020, to October 3, 2020. (b) The survival ratios of new particles. The measured survival ratio of the 8 nm particles to 50 nm was ~0.8%, close to the theoretical survival ratio (~0.5%) due to coagulation loss. (Li et al., 2022)

References

Li, X., Li, Y., Cai, R., Yan, C., Qiao, X., Guo, Y., Deng, C., Yin, R., Chen, Y., Li, Y., Yao, L., Sarnela, N., Zhang, Y., Petäjä, T., Bianchi, F., Liu, Y., Kulmala, M., Hao, J., Smith, J. N., and Jiang, J.: Insufficient Condensable Organic Vapors Lead to Slow Growth of New Particles in an Urban Environment, Environ. Sci. Technol., 55, 9936-9946, 10.1021/acs.est.2c01566, 2022.

Cai, R., Deng, C., Stolzenburg, D., Li, C., Guo, J., Kerminen, V., Jiang, J., Kulmala, M., Kangasluoma, J.: Survival probability of new atmospheric particles: closure between theory and measurements from 1.4 to 100 nm, Atmos. Chem. Phys., 22, 14571-14587, 10.5194/acp-22-14571-2022, 2022.