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 green. 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.

Reviewer #2

This paper presents the effect of the COVID-19 lockdown on atmospheric new particle formation. Indeed, the COVID-19 lockdown provided us a unique opportunity to investigate the effect of reduced anthropogenic emissions (probably similar to pre-industrial conditions) on a variety of atmospheric processes. Shen et al (2021) recently reported enhanced nanoparticle formation and growth during the COVID-19 lockdown in urban Beijing, but without much of the process-level explanation of nanoparticle formation and the role of key vapors. Here, the authors provide a more detailed analysis of nano particles and the role of sulfuric acid and oxygenated organic molecules in particle formation and growth. Authors report that the formation rate of 1.5 nm clusters was unchanged by drastically reduced traffic emissions. However, the cluster's survival probability was increased due to the higher formation of sulfuric acid, oxygenated organic molecules, and other vapors, indicating the enhanced atmospheric oxidative capacity.

Authors conclude that traffic emissions play a limited role in atmospheric NPF as opposed to the previous reports showing traffic as a high source of ultrafine particles such as Rönkkö et al., 2017 (https://doi.org/10.1073/pnas.1700830114), Guo et al., 2020 (https://doi.org/10.1073/pnas.1916366117). While Okuljar et al., 2021 (https://doi.org/10.5194/acp-21-9931-2021) also showed that traffic contribution to sub-3nm particles is lower during NPF events, Gani et al., 2021 (10.1039/D1EA00058F) showed NPF contributions to ultrafine particles in locations with high concentrations of precursors (e.g. traffic) are critical. Another recent study from an Indian urban location Kanawade et al., 2022 (https://doi.org/10.1029/2021JD035392), however, showed that NPF and growth events were suppressed under the reduced anthropogenic emissions during the lockdown. Kanawade et al. also reported an unaltered particle formation rate of 1.5 nm (and number concentrations of sub-3nm particles), but nanoparticle growth was limited by likely lower condensable vapors. This probably hints the role of micro-meteorology is also imperative. I suggest authors discussing all the above papers.

We thank the reviewer for this constructive comment. Indeed, several recent studies have suggested the contribution of traffic emissions to the concentration of 1-3 nm particles, as brought up by the reviewers. These studies were conducted in different locations and environments, and because of this, the reported contribution of traffic emissions differs significantly. A mechanistic understanding of NPF, however, was not reached in these studies, due to either the lack of measurement of the NPF precursors (such as H2SO4, amines, NH3, and organic vapors) and/or the analysis of cluster dynamics vs. particle nucleation rate. Therefore, it is not so straightforward to compare our results to these studies.

For example, Gani et al., (2021) reported the contribution of traffic emissions to the concentration of ultrafine particles (d_p<100nm). In fact, this is consistent with our observations that a burst of ca. 7-30 nm particles can be observed during traffic rush hours at our measurement site (Fig. S3). However, we focus mainly on sub-3 nm particles in the context of particle nucleation, so the scope of these two studies is not entirely the same. Guo et al., (2020) suggested that organic vapors, as the oxidation products of traffic exhaust, are solely important for urban particle nucleation. This study was based on a chamber study, and without the measurement of NPF precursors, it remains unclear how well the chamber condition mimicked the ambient atmosphere. For these reasons, we feel that it is difficult to discuss the similarities and contrasts between our study and these two publications.

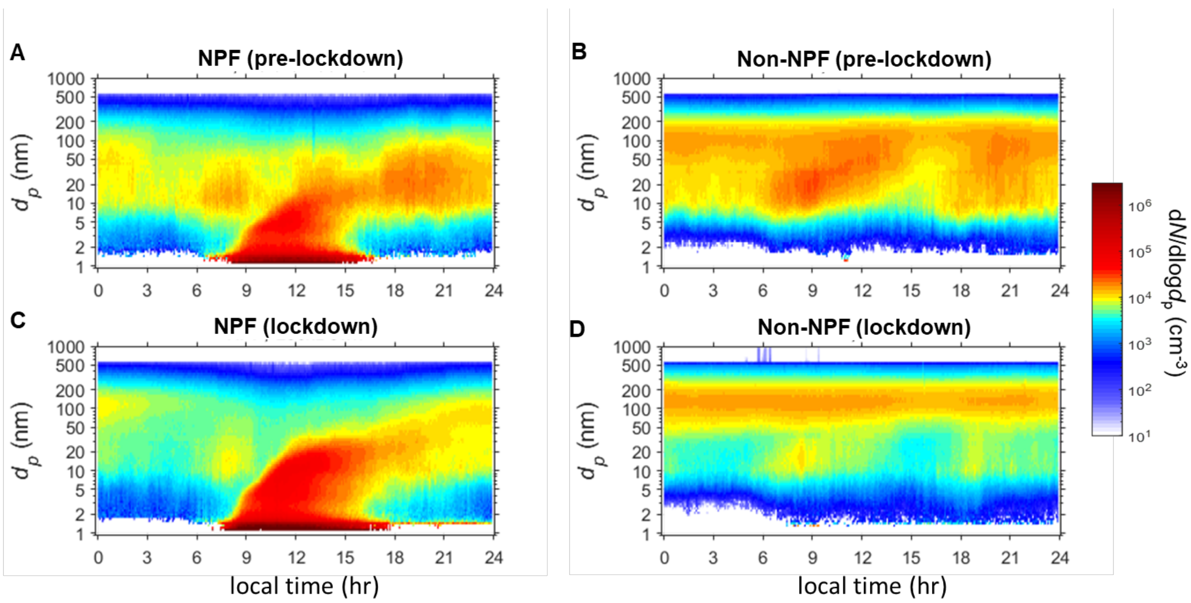

Figure S3. Particle number size distribution in NPF and non-event days during the pre-lockdown and lockdown periods.

On the other hand, other studies are relevant to this study (Ronkko et al., 2017; Okuljar et al., 2021; Kanawade et al., 2022). We added the following discussion to our manuscript (line 311-334) and updated the references.

"Ronkko et al., (2017) and Okuljar et al., (2021) both showed that in traffic-dense areas, the concentration of sub-3nm particles is obviously higher than in background areas. Kanawade et al., (2022) conducted measurement of sub-3nm particles at a site that is ~ 1km away from traffic emission and found an insignificant influence of traffic emission on the particle concentration. These studies suggest that the distance between the measurement site and the traffic emission source is crucial for the observation of the emitted sub-3 nm particles, likely due to the dilution and coagulation loss of these nano-particles. However, it is probably not the same reason for our study, because the measurement site of this study is very close to an arterial road with heavy traffic. One possibility of the discrepancy is that the emission factor of sub-3nm particles is significantly lower for vehicles in Beijing. As shown in the laboratory study by Ronkko et al., (2017), the emission factor can vary by up to three orders of magnitude, being the highest for heavy-duty vehicles (e.g., diesel vehicles) and the lowest for light-duty cars. In Beijing, diesel vehicles are forbidden in downtown areas during traffic rush hours, so it is likely that the emission of sub-3nm particles is weak. Also, the high coagulation sink in Beijing and India might be another reason for the small contribution of traffic emissions. Another possibility that cannot be fully ruled out is the potential biases due to different detection methods of sub-3nm particles. The aforementioned studies utilized the PSM to detect sub-3nm particles, for which the size-classification of particles is based on the saturation ratio of diethylene glycol (DEG), while we use the soft Xray neutralizer and a DMA to classify particle size. The intrinsic difference between these two methods is not well quantified. It is also possible that the sub-3nm particles by vehicles are not efficiently charged by the soft Xray, and/or can be more efficiently activated by highly saturated DEG. Future research on the comparison between the PSM and SMPS is highly desired."

Overall Recommendation: The paper presents detailed analyses using new techniques that can characterize nanoparticles and provide new insights into the response of NPF to drastic changes in the atmospheric chemical cocktail. The manuscript should be published after the authors' elaborate discussion as indicated above and the following minor issues are addressed.

The pre-lockdown period falls during the peak winter season, followed by the lockdown during early spring, the temperature is expected to increase as the season progresses. The role of different micro-meteorological conditions should be highlighted between the time periods considered in this study. Or is it the critical factor for more occurrence of NPF and growth during lockdown with elevated temperature (more active photochemistry) rather than reduced anthropogenic emissions as background concentrations are on the higher side in urban areas.

Thanks for the comment. Indeed, the changes in temperature and solar radiation have multiple influences on particle formation and growth, which have been demonstrated in the manuscript. The increased UV radiation and atmospheric oxidative capacity (Fig. S4) outset the decreased SO2 concentration (Fig.2C), which results in a similar H2SO4 concentration (Fig. 2E). This is explained in the manuscript (Line 208-210 of the revised manuscript): "The median SA_1 and SA_2 concentrations were also stable between the two periods. This is because the decline of the sulfuric acid precursor (i.e., SO_2, Figure 2C) was completely compensated by the enhanced photochemistry, as indicated by the variation of UVB (Fig. S4B)."

If the lockdown had not been imposed and SO_2 was not reduced, we would expect a higher SA_1 concentration in Feb than in Jan, due to the stronger UV radiation. However, a higher SA_1 concentration does not necessarily cause more frequent or stronger NPF. We found that the non-NPF days have a higher SA_1 concentration than that of NPF days due to the higher SO_2 concentration (Yan et al., 2021).

Our previous studies have shown that the CS is the governing parameter of the occurrence of NPF (Deng et al., 2021). Thus, the frequency of NPF in Beijing is primarily influenced by the origin of air masses. If the air mass came from the clean north area, the low CS due to the low concentration of pre-existing particles would cause more NPF events; and oppositely, if the air mass came from the polluted south area, there would be fewer NPF events.

Besides, high temperature can reduce the stability of SA clusters, as we show in Fig.4A, and ultimately weaken the strength of NPF (Deng et al., 2020). The high temperature should facilitate the formation of highly oxygenated organic molecules (HOM), but HOM are not the main precursor of initial particle formation (quantified by $J_{1.5}$), so this does not have a direct influence on the occurrence and strength of NPF.

Lines 85-90: there are laboratory studies showing clustering between sulfuric acid and organic acids e.g. Schobesberger et al. (https://doi.org/10.1073/pnas.130697311) or multi-component nucleation of sulfuric acid, ammonia, and organics (10.1126/sciadv.aau5363), and traffic is not the only source of organic acids to the atmosphere. For better readability, remove "on one hand" and "on other hand".

Agreed. We have removed "on one hand" and "on the other hand", as suggested.

Line 185: Fig. S4 cited for particles in the size range of 10-30 nm, but Fig. S4 in the supplementary shows diel patterns of temperature and UVB

Thanks for pointing out the error, Fig.S4 should be Fig.S3.

Lines 202-203: Correct as Fig. S4

Yes, this has been corrected.

Supplementary figures are incorrectly cited in the main text at most places. Please check carefully.

Yes, these have been checked and corrected.

Line 292: you mean to say "i.e., 1.3 pptv"?

Yes, we use "ppb" or "ppt" throughout the manuscript. To avoid confusion, we added "a volume mixing ratio of" in front of "1.3 ppt".

References

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