Responses to the Reviewer #2' Comments

"Measurement report: Size distributions of urban aerosols down to 1 nm from long-term measurements" by Deng et al.

We appreciate valuable comments from the reviewer, which helped to improve this manuscript. We have addressed them in the following paragraphs (the text in italics is the comments, followed by our response). Additionally, all changes made are highlighted in the revised manuscript.

I recommend the publication of this manuscript, but suggest the authors to address the following minor concerns.

1. line 120.

Your NPF definition looks like the classification of size distribution, not NPF. It's quite different from conventional NPF definition, like Dal Maso et al. Please describe the difference and how this will affect your conclusions.

Response: We identified NPF and non-NPF using the classification method reported in Deng et al. (2020). According to this classification method, we identified a day with a burst of sub-3 nm particles and subsequent growth for hours as a NPF day and a day without burst of sub-3 nm particles and subsequent growth as a non-NPF day. A typical NPF and non-NPF day was shown in Fig. R2.

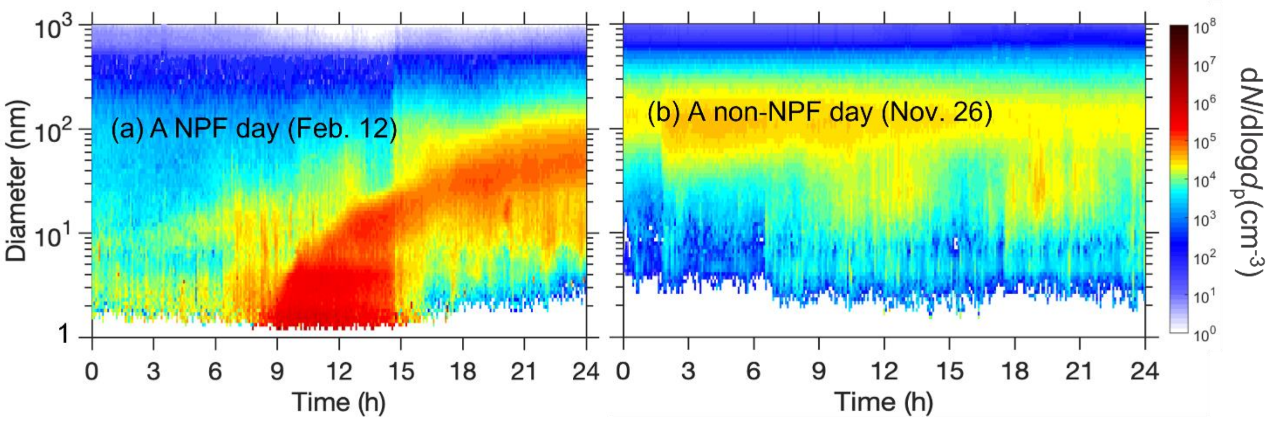

Figure R2. Typical particle size distributions of (a) a NPF day (Feb. 12, 2018) and (b) a non-NPF day (Nov. 26, 2018).

In Dal Maso et al. (2005), a day is identified into a NPF day if the nucleation mode (~3-25 nm) appears and those newly formed particles continue to grow over a time span of hours. The classification method used in our study is on the basis of the classification method in Dal Maso et al. (2005), but it can determine the starting point of the occurrence of NPF events more accurately with the information of sub-3 nm aerosols. Therefore, the difference between classification results by these two classification methods would be relatively small and would not affect the reported findings.

2. line 129. why was power law function used? not other function? did you try any others? What information or contribution does the function provide on nucleation mechanistic, regional, and global atmospheric models?

Response: We did try other function, e.g., the log-normal distribution function. As shown in Fig. R3, the size distribution of H_2SO_4 clusters and aerosols was fitted using the combination of four lognormal distributions. In the sub-3 nm size range, the fitting is not so good and it fails to catch the rapid decrease of the distribution function from H_2SO_4 monomer to dimer and then to aerosol size because the lognormal distribution decreases slowly from the peak diameter to larger sizes. Furthermore, we would not think there is a "mode" with a peak diameter of ~0.4 nm existing in the sub-3 nm size range. Instead, the concentration decreases monotonically from gaseous precursors to aerosol size. This characteristic is well captured by the power law function; therefore, we choose it to fit the size distributions in the sub-3 nm size range.

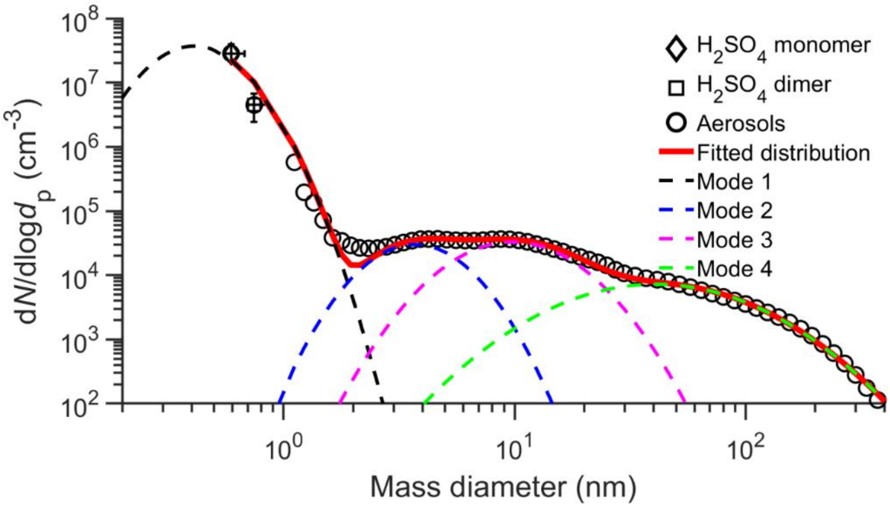

Figure R3. The median daytime NPF type number size distributions from H_2SO_4 monomer to larger aerosols and the fitted size distributions using lognormal distributions.

We added the discussion on the reasons why we chose the power law function to fit the aerosol size distribution in sub-3 nm in the revised manuscript (lines 134-137 and Fig. S4):

"Note that the power law function was chosen to fit in the sub-3 nm size range because it can well capture the monotonic decrease from H_2SO_4 monomer to dimer and then to the sizes representative of aerosol particles. The log-normal distribution function is not a good fit in the sub-3 nm size range (Fig. S4), and especially a "mode" with a peak diameter of ~0.4 nm resulting from such a fit seems not to be reasonable."

As aforementioned, this power law function describes the pattern of size distributions in the sub-3 nm size range in a clear way and can contribute to modeling work. We also added the discussion on the contribution of the power law function to modeling studies (lines 300-306):

"Additionally, the power law function can be readily incorporated in models. For instance, in the global climate models, the observed aerosol size distributions are used to compare with simulated ones (Bergman et al., 2012) and the sub-3 nm size range is the key to simulate nucleation process accurately. However, due to relative scarcity of measured sub-3 nm particle size distributions around the world, the comparison between observed and simulated results is usually lacking this key size range. Our power law function, as the simplified representation of sub-3 nm size distributions, can extend the observed particle size distributions from above 3 nm to sub-3 nm, thus helping to constrain the aerosol module in global models."

3. line 150-165. Is the PNSD for the time slot before, during or after NPF? how did you pick up the PNSD that you named "typical" from a large set of data? The PNSD is always changing even during a single NPF event.

Response: Sorry for the confusion. The daytime NPF, daytime non-NPF and nighttime PNSD is selected during the NPF period on a NPF day (Feb. 16 11:10, 2018), during the daytime on a non-NPF day (Feb. 25 12:25, 2018) and during the nighttime (Apr. 4 00:35, 2018). We added this information in the caption of Fig. 2 (lines 541-543):

"The daytime NPF, daytime non-NPF and nighttime PNSD is selected during the NPF period on a NPF day (Feb. 16 11:10, 2018), during the daytime on a non-NPF day (Feb. 25 12:25, 2018) and during the nighttime (Apr. 4 00:35, 2018)."

The cluster analysis helped us to identify three types of PNSDs based on a large set of data, and then we picked up the PNSDs in Fig. 2 as typical types because they show pretty similar characteristics recognized by the cluster analysis. We agree that PNSD is always changing even during a single NPF event and we can never find two exactly same PNSDs. However, what we are trying to achieve is to reveal the common characteristics of the PNSDs based on a long-term dataset.

4. figure 4b where is Nsub-2 curve?

Response: In Fig. 4b, the median concentration of sub-2 nm aerosols is constantly near zero during the whole day. We added the notation about this issue in the caption of Fig. 4 (lines 564-565):

"Note that the median diurnal variations of $N_{\text{sub-2}}$ are near zero on non-NPF days in (b)."

5. line 245-255. is there any evidence from nanoparticle/cluster chemical composition (e.g., H_2SO_4, amines, organics) measurement to support the source of these sub 3 nm particles were from traffic emission? for example, how were H_2SO_4 monomer and dimer observed in these traffic events?

Response: It would be great if we could be able to identify the chemical composition of sub-3 nm particles. Unfortunately, to date, the lowest size limit of measuring chemical composition of atmospheric particles is ~5 nm in diameter by very few instruments, e.g., a thermal desorption chemical ionization mass spectrometer (TDCIMS) (Smith et al., 2004; Li et al., 2022). Further efforts are needed to extend the detection size limit for measuring chemical composition of atmospheric sub-3 nm particles.

Regarding the clusters we measured, they are majorly formed through secondary processes and are relatively abundant in the atmosphere even on non-NPF days, so the impacts of vehicles are difficult to identify. For example, on non-NPF days, the H_2SO_4 monomer and dimer concentration showed as similar diurnal variations as on NPF days, which peaked around noon (Fig. 4(c) and (d)). There was no exceptional increase of H_2SO_4 monomer and dimer concentration during traffic rush hours even though previous studies show that vehicles can emit primary H_2SO_4 that can be further converted to new particles through nucleation (Arnold et al., 2012; Ronkko et al., 2013). This indicates that H_2SO_4 monomers and dimers emitted by vehicles are negligible compared to those converted from photochemical oxidation in the large-scale atmosphere.

Reference:

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