Response to Review of "The Driving Factors of New Particle Formation and Growth in the Polluted Boundary Layer" by M. Xiao et al.

This is a well-written and extremely interesting manuscript describing a comprehensive set of experiments on the chemical mechanisms of new particle formation (NPF) in an environment representative of a polluted urban atmosphere. The study provides an explanation for the occurrence of NPF when there is very strong competition for precursor vapors by high concentrations of pre-existing aerosol particles. The successful nucleation and growth of particles in these environments is important, because it distributes aerosol mass to smaller sizes, changing the interaction of these particles with sunlight, the numbers of cloud-nucleating particles, and the fate of the aerosol mass in the human respiratory system. These topics have broad interest, and the manuscript is definitely suitable for publication in ACP.

The investigators demonstrate that sulfuric acid + dimethylamine and other bases like ammonia dominate NPF in polluted atmospheres, even in the presence of high concentrations of oxidation products of aromatics, which can have quite low volatility. The growth of the newly formed particles to larger sizes involves many other compounds, including organics. There is a very strong temperature dependence to the NPF.

This work employs cutting-edge research instruments and the CERN CLOUD chamber to make these measurements. The results are well described and the interpretation is clear. The methods are also well described, and earlier work is appropriately referenced and discussed. The manuscript is very well written. I believe it is suitable for publication with only minor technical changes.

The authors would like to thank the reviewer for the detailed comments, corrections, and suggestions. Here we replied to all the comments and improved the paper following their recommendations.

Comments:

1) Throughout the PDF manuscript, following a capital "A" there is a space and the remainder of the word is shifted to the right. This must be some sort of PDF conversion problem, but the authors and copy editors should carefully proof the online and print versions of the final manuscript to see if this issue still exists.

We did not find this feature in the pdf version downloaded from ACP. In any case we will carefully read the proof and focus on this issue.

2) It's a bit odd in ACP to have the Methods following the Results. This is more common in journals such as Nature and Science, where a wide audience is presumed and the methods are relegated to an appendix-like attachment, with a smaller font. I don't have a problem with this structure for ACP, but wish to call attention to it in the event there are editorial norms that would suggest moving the Methods toward the front.

We submitted the manuscript for consideration as an ACP Letter. This requires the description of the applied methods in the form of an appendix. If the manuscript will be accepted as ACP Letter this formatting does comply with the formatting requirements. Otherwise, it will be changed.

3) Line 134: define "GR"

Added: growth rate (GR)

4) Line 138: For clarity, perhaps say "cluster self-coagulation dominates the growth." It took me a moment to understand what the authors were meaning.

We changed the text as the reviewer suggested.

5) Line 144: "The multicomponent system used in the simulations. . . ." What simulations? Stolzenburg et al. (2020)?

It refers to the multicomponent experiments in this study. We changed “the simulations” to “this study”.

6) Line 168: The chamber simulations do reproduce observed urban GRs; however, it is probably worth noting here that the more complex chemistry of the ambient urban atmosphere may lead to other species contributing to particle growth than are investigated at the CLOUD chamber. One should not presume that the relatively simple mixtures explored here can replicate the full complexity of atmospheric processes.

We agree that the chamber does not represent the full complexity in the atmosphere. However, the multicomponent system we used reproduce the major variabilities in terms of particle production rates, particle growth rates, sulfuric acid and bases concentrations and particle condensation sink. In this regard, in line 164 we state: ‘While the precursor composition in the ambient atmosphere is indeed much more complex than in the chamber’. Details on the selection of organics and oxidation processes are further discussed at line 250-260 and SI. We think this covers the precaution raised by the reviewer.

7) Line 214: In the previous paragraph you state that the experiments were at the CLOUD chamber; no need to repeat that here.

The repetition is removed.

8) Line 240: DMA is already defined.

We removed the repeated definition.

9) Line 244: No mention here of how OH was determined; this appears later in the manuscript (Eq. 5)but would logically be verbally described here.

We think the detailed description of OH and OxOrg determination would break the flow of text. We add (for details see section 4.5).

10) Line 265: Replace "DMA-train" with "differential mobility analyzer train". Too many "DMA"s.

DMA is replaced with the full name “differential mobility analyzer”, also later in the text

11) Lines 266-7: Define PSM, SMPS, and CPC.

PSM and CPC are already defined earlier in this paragraph. Scanning mobility particle sizer is added for SMPS.

12) Eq. 1: Perhaps have a subscript "dp" for the J and N variables, since you calculate these values for different size particles (e.g., 1.7 nm, 3 nm).

"J" changed to "J_{dp}" and "N" changed to 'N_{dp}".

13) Line 305: Provide model number and company name for the H3O+ CIMS and state the method (e.g., cavity ringdown spectrometry), model number, and company name for the NH3 analyser.

The H_3O^+ CIMS is an APi-TOF (TOFWERK AG) coupled with a home-made crossflow ionization source. This is further specified as follows:

"Ammonia concentrations were measured with cavity ring-down spectroscopy (G2103, Picarro, Inc) and an H_3O^+ chemical ionisation mass spectrometer (H_3O^+ CIMS) (Pfeifer et al., 2020). The latter was an APi-TOF (TOFWERK AG) coupled with a home-made crossflow ionization source using positively charged water clusters to detect ammonia in real time."

14) Line 311: Is the PTR3 a custom-built instrument? If so, say this and if not, give model number and company name.

PTR3 is custom-made. We add "custom-made" in the description.

15) Line 314: Define STOF and give model number and company name for the "PTRS", which should be "PTRMS".

PTRS is also custom-made. We changed the text as "… a custom-made short TOF proton transfer mass spectrometer was used in CLOUD12".

16) Line 359: Please reference the form of the Cunningham slip correction equation you are using; there are several and they do have some differences.

We add the Cunningham slip correction equation we used as equation 18.

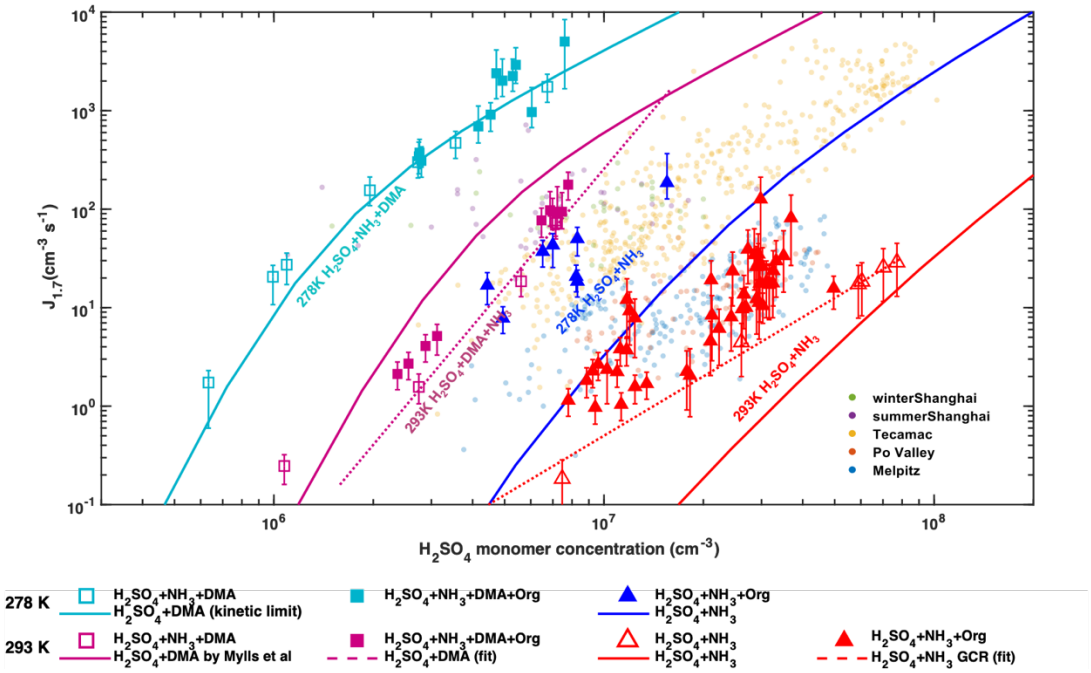
$$C_c = 1 + Kn * (1.142 + 0.558 * exp(-0.999/Kn));$$

17) References: Please review the reference formatting and ensure it follows Copernicus guidelines. EndNote-style reference managers always make mistakes; for example, for Breitenlechner et al. the title of the paper is capitalized, and in Dunne et al. the page range is not completed. If you correct these errors now it will save the copy editors the effort of finding all them and asking you to fix them later.

We thank the reviewer for spotting these errors. We have corrected these references.

18) Fig. 1. I used a pen to label all the lines and symbols because this is such a busy graph. I suggest you go ahead and do that to make it much easier to interpret.

We added the labels to guide the eyes.



19) Fig. 1 caption. The penultimate sentence should say that the nucleation rate of H2SO4+DMA at the kinetic limit is shown by the cyan curve.

The cyan curve shows the H_2SO_4 nucleation rate at the kinetic limit, which matches our H_2SO_4+DMA experiments. The line is not specifically modeled for H_2SO_4+DMA and the kinetic limit is not sensitive to the presence or absence of DMA. We changed the sentence to "The nucleation rate of H_2SO_4 at the kinetic limit is indicated by the solid cyan curve, which matches our H_2SO_4+DMA experiments."

20) Fig. 2 caption: In the last sentence there needs to be a space in bicycloalkylradicals.

We added the space, now it is "bicycloalkyl radicals".

21) Fig. 4 caption: What do you mean by, "At larger particle sizes, the contribution of organics will increase further."? What's the basis for this statement.

As particle size increases, more volatile organics can also participate in particle growth. We added Tröstl et al., 2016 here.

22) Fig. 4b: It's hard to distinguish the green and blue curves (biogenic with and without NO) from each other. Can you use a different line type for each?

We changed biogenic without NO to a dashed line.

23) Fig. 5: In the caption please list the name and country of each location indicated in the condensation sink box in the graph, as you did for Fig. 7.

We added the following text to the caption: "Also shown are typical CS from observations in the polluted boundary layer at the following locations: Po Valley regional (San Pietro Capofiume,

SPC) (Kontkanen et al., 2017), Madrid (Carnerero et al., 2018), Tecamac (Kuang et al., 2010), Nanjing (Yu et al., 2016) and Beijing (clean, transition and haze) (Cai et al., 2017)."

24) Fig. 6. What information is conveyed by the size of the symbols?

The size of the symbols is proportional to their intensity in the mass spectrum. We added this information in the caption.

Reference:

Cai, R., Yang, D., Fu, Y., Wang, X., Li, X., Ma, Y., Hao, J., Zheng, J. and Jiang, J.: Aerosol surface area concentration: a governing factor in new particle formation in Beijing, Atmos. Chem. Phys., 17(20), 12327–12340, doi:10.5194/acp-17-12327-2017, 2017.

Carnerero, C., Pérez, N., Reche, C., Ealo, M., Titos, G., Lee, H. K., Eun, H. R., Park, Y. H., Dada, L., Paasonen, P., Kerminen, V. M., Mantilla, E., Escudero, M., Gómez-Moreno, F. J., Alonso-Blanco, E., Coz, E., Saiz-Lopez, A., Temime-Roussel, B., Marchand, N., Beddows, D. C. S., Harrison, R. M., Petäjä, T., Kulmala, M., Ahn, K. H., Alastuey, A. and Querol, X.: Vertical and horizontal distribution of regional new particle formation events in Madrid, Atmos. Chem. Phys., 18(22), 16601–16618, doi:10.5194/acp-18-16601-2018, 2018.

Kontkanen, J., Lehtipalo, K., Ahonen, L., Kangasluoma, J., Manninen, H. E., Hakala, J., Rose, C., Sellegri, K., Xiao, S., Wang, L., Qi, X., Nie, W., Ding, A., Yu, H., Lee, S., Kerminen, V.-M., Petäjä, T. and Kulmala, M.: Measurements of sub-3 nm particles using a particle size magnifier in different environments: from clean mountain top to polluted megacities, Atmos. Chem. Phys., 17, 2163–2187, doi:10.5194/acp-17-2163-2017, 2017.

Kuang, C., Riipinen, I., Sihto, S.-L., Kulmala, M., Mccormick, A. V and Mcmurry, P. H.: An improved criterion for new particle formation in diverse atmospheric environments, Atmos. Chem. Phys., 10, 8469–8480, doi:10.5194/acp-10-8469-2010, 2010.

Pfeifer, J., Simon, M., Heinritzi, M., Piel, F., Weitz, L., Wang, D., Granzin, M., Müller, T., Bräkling, S., Kirkby, J., Curtius, J. and Kürten, A.: Measurement of ammonia, amines and iodine compounds using protonated water cluster chemical ionization mass spectrometry, Atmos. Meas. Tech., 13(5), 2501–2522, doi:10.5194/amt-13-2501-2020, 2020.

Tröstl, J., Chuang, W. K., Gordon, H., Heinritzi, M., Yan, C., Molteni, U., Ahlm, L., Frege, C., Bianchi, F., Wagner, R., Simon, M., Lehtipalo, K., Williamson, C., Craven, J. S., Duplissy, J., Adamov, A., Almeida, J., Flagan, R. C., Franchin, A., Fuchs, C., Guida, R., Gysel, M., Pet, T. and Steiner, G.: The role of low-volatility organic compounds for initial particle growth in the atmosphere, Nature, 533, 527–531, doi:doi:10.1038/nature18271, 2016.

Yu, H., Zhou, L., Dai, L., Shen, W., Dai, W., Zheng, J., Ma, Y. and Chen, M.: Nucleation and growth of sub-3 nm particles in the polluted urban atmosphere of a megacity in China, Atmos. Chem. Phys., 16, 2641–2657, doi:10.5194/acp-16-2641-2016, 2016.