

General comments:

For this paper, the authors employed a Neutral cluster and Air Ion Spectrometer (NAIS) to investigate the early steps of new-particle formation (NPF) events in Beijing, China, over a period of 3 months. Specifically, observations were made down to particle (or cluster) sizes of about 2 nm. NPF events in large, polluted urban areas, in particular in E Asia, are a current subject of atmospheric research (e.g. Kulmala et al., 2017). To my knowledge, this is the first report on deploying an NAIS in a Chinese megacity for this purpose, and it constitutes one of recent attempts of improving on the observations of NPF in such environments by directly measuring in the sub-3 nm size range (cf. Cai and Jiang, 2017; Yu et al., 2016). As such, the study is timely and of interest to the scientific community engaged in this field, and I recommend its publication in *Atmospheric Chemistry and Physics*. Before that however, I recommend a major revision to take care of some important issues.

My main concern with the study in its present form is the treatment and discussion of the NAIS measurements for the sub-3 nm size range. The treatment, presentation and interpretation of these data need to be brought into a form more rigorously consistent within the paper itself, as well as with best-practices recommended by the community (Manninen et al., 2016) – in particular as the corresponding results are a major selling point here.

Comments regarding sub-3 nm measurements:

Lines 109-110: “The NAIS ... can detect particles down to a size of 0.8 nm”:

My main point is that the NAIS can actually *not* be used to measure *neutral* compounds down to this size, so this statement is misleading in its current form. The NAIS does detect ions with the corresponding mobility, but due to the interference from charger ions it is deemed not possible to determine concentrations of neutral clusters for the smallest size bins. Quoting Manninen et al. (2016), which is cited also in this paper (line 137), “the particles below about 2 nm cannot be reliably distinguished from the corona-generated ions. Typically, the lowest detection limit for the NAIS in the particle mode is between 2 and 3 nm depending on the corona voltage and on the properties and composition of carrier gas (environmental conditions).” Details can be found in their paper and references therein.

At one occasion later, the authors appear to consider this instrumental limitation, e.g. section 2.2.3.

Then, section 3.4 (including Fig. 5 and Table 2) discusses charged vs. neutral “cluster” and “particle” concentrations. Here, the authors need to state what is their definition of “cluster” and “particle”. And in light of the above, they might need to reconsider if total neutral cluster concentrations (as implied in section 3.4) can even be derived from the NAIS measurements! The discussions throughout section 3.4 may have to be revised...

E.g., depending on those definitions, could the observed decreases of “neutral clusters” for NPF days (e.g. Fig. 5b) be explained by instrument response to a change in environmental conditions?

Figure 2, top panel, and Figure 7:

As a consequence, I would argue that particle size distribution data below 2 nm shouldn't even be shown. The concentrations at the size bins <2 nm are subject to instrumental factors, not necessarily resulting from actual variations in the concentrations of sub-2 nm neutral clusters (particles). Hence, their display here could prompt an unaware reader to draw wrong conclusions about the actual population of sub-2 nm neutral clusters.

Other comments:

Line 68: Kulmama should probably be Kulmala – also in later instances for this reference. Speaking of which, the recent paper by Kulmala et al. (2017) is relevant to this study and should be brought to attention in the introduction.

As condensation sinks were calculated for this study, it might be useful even to shortly discuss the authors' findings in light of the conclusions of that paper (see e.g. lines 237-239).

Also, it could be interesting to compare the results here with those in Yu et al. (2016). Therein, they report in particle formation and growth rates during NPF events in Nanjing, also down to sub-3nm sizes.

Lines 277 & Fig. 3, line 287:

“Haze days” seems to be used interchangeably with “no-NPF days”. Are they? If so, that point should be made clearer. If not, it may be feasible to mark them in Fig. 3.

The various types of day are actually defined later on (lines 325-329). I suggest moving this definition to an earlier place, and then shortly mention it again later.

Line 318: “attachment to existing particles”

I would have expected this process be more pronounced on the *no*-NPF days, when condensation sinks were higher.

Line 378: “previous have not been able”

I assume the authors refer to their novel measurement of particles in the 2-3 nm allowing them to more accurately calculating the coagulation sink (CoagS) for particles down to 2 nm. That's technically OK, but one would expect those small particles (i.e. in the 2-3 nm range for instance) to play a minor (negligible?) role in determining CoagS. How much is the value obtained here improved (increased) by the possibility to take the 2-3 nm range into account?

Minor comments:

Abstract, 2nd sentence: The statement should be clarified. From what are the estimated characteristics different in the case of restricted measurements?

Lines 152-153: It may be interesting and instructive for the reader to hear, in short, about the nature of the problems encountered.

Line 263: Does this t-test result apply to the whole measurement campaign, or just the subset shown in Fig. 2? In the latter case, would it change when applied to the whole period?

Line 297: “are more likely ...” than what else?

Lines 329-332, Table 2: The source of the uncertainty of 20% has remained unclear to me. Maybe the authors can rephrase.

Most figures have a gray background and odd dark-gray or blank frames. They would look better without any that.

The text/numbers in the color bar in Figures 2 and 7 are difficult to read and lack units.

References:

- Cai, R. and Jiang, J.: A new balance formula to estimate new particle formation rate: reevaluating the effect of coagulation scavenging, *Atmos. Chem. Phys. Discuss.*, in review, doi:10.5194/acp-2017-199, 2017.
- Kulmala, M., Kerminen, V.-M., Petäjä, T., Ding, A. and Wang, L.: Atmospheric Gas-to-Particle Conversion: why NPF events are observed in megacities?, *Faraday Discuss.*, doi:10.1039/C6FD00257A, 2017.
- Manninen, H. E., Mirme, S., Mirme, A., Petäjä, T. and Kulmala, M.: How to reliably detect molecular clusters and nucleation mode particles with Neutral cluster and Air Ion Spectrometer (NAIS), *Atmos. Meas. Tech.*, 9(8), 3577–3605, doi:10.5194/amt-9-3577-2016, 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(4), 2641–2657, doi:10.5194/acp-16-2641-2016, 2016.