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## Comment on acp-2021-23

Anonymous Referee #2

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Referee comment on "Evaluation of the contribution of new particle formation to cloud droplet number concentration in the urban atmosphere" by Sihui Jiang et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2021-23-RC1>, 2021

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The manuscript presents observations on the effects of new particle formation (NPF) on cloud condensation nuclei (CCN) concentrations in urban Beijing. These observations are used to quantify the limiting effects of extremely high CCN concentrations on cloud activation. The topic is scientifically relevant and suitable for ACP. However, there are some issues that need to be solved before the manuscript can be accepted for publication.

### General comments

The paper focuses only on 7 "typical" NPF events, which is a small data set. Potential issues related to the small sample size should be discussed and limitations should be noted when drawing conclusions. Regarding the small sample size, it would be important to justify why these seven days were selected and what makes them typical. There are also case studies focusing on "typical" single days, which is generally fine, but again these should be justified.

Contrasting cloud droplet number concentrations based on a constant maximum supersaturation and updraft velocity seems like comparing apples and oranges. It is well known CCN compete for water vapor during cloud activation and for adiabatically rising air parcels this is one of the main factors determining the maximum supersaturation. Why would using a prescribed updraft velocity be better than prescribed maximum supersaturation in calculating CDNC? Surely, both updraft velocity and cloud supersaturation are parameters that are difficult to measure in the actual environment. While updraft velocity has its use in the modeling community, CDNC at a fixed maximum supersaturation is a useful parameter to compare with observations.

Some calculations and choices require better explanations. Specific comments related to this and other issues are given below.

## Specific comments

Lines 48-49: contributions of NPF on aerosol (number concentration) needs a better explanation. Are the numbers from the reference (Merikanto et al., 2010)? Is this for boundary layer? Are there any other references (e.g., urban or China)?

Lines 112-116 and 187-191: were there specific conditions for identifying/classifying NPF events? Specifically, how the seven NPF cases (Fig. 1; line 190) were selected? The authors write that "While non-NPF events may also have sudden increases of nucleated particles at a short time scale, but they do not show further growth" (lines 115-116), so does this mean that the focus is on particle growth rather than NPF?

Section 2.2: this section could be clarified. Mixing the growth of particles (dry size) and droplets (wet size) is confusing. This includes also the critical droplet/dry size. There is also minimum particle size, which is somehow mixed with critical diameter (Eqs 1 and 2). Eqs 1 and 2 have also unexplainable variables. Which species are "organic and inorganic" (line 145) and what are their hygroscopic parameters?

Section 2.3: this section could be clarified.  $dw/dt$  in Eq. 4 needs to be explained. In addition, it is not clear how maximum supersaturation is solved from this equation. Supersaturation has different symbols ( $s$  and  $S$ ), and Eq. 5 has unexplainable variables and index  $i$ . It would be useful to give more details about "empirical values of cloud updraft velocity" (line 170) to justify the selected updraft velocity range.

Figure 1: it looks like the smallest particles (below 20 nm) are missing; is this a measurement artefact or a real observation? Does this cause difficulties in identifying NPF events? At least nucleation mode (10-25 nm) particle concentration would be underestimated.

On line 195 it is stated that "The variation of  $N_{CCN}$  seems to be more consistent with that of  $N_{CN}$  than  $N_d$ ", but is this true? Please clarify the text and also show some evidence (for example correlation coefficients) to support this.

In general, it would be better to write that  $N_{CCN}$  is based on a constant maximum supersaturation rather than refer to the availability of water vapor (lines 196-197).

Calculation of the effect of NPF on CCN and clouds should be clarified. Please explain "decoupling time" (line 202): how is it defined and what does it mean in practice. Is there

a specific definition for  $t_{\text{start}}$  (line 203) and how is it related to the start of the period from 12:00 to 16:00 to estimate the change of CCN during NPF events (line 204)? Also, using the constant time period (12:00-16:00) should be justified; this works only if the events start at the same time. What are the averaging time periods for calculating CCN concentrations before and during NPF events (line 207)?

Calculation of the effect of NPF on CCN and clouds does not account for the changes in background conditions (except the evening traffic emissions at 16:00). This should be examined and explained in the manuscript. For example, would there be a change in CCN concentration during non-event days?

Where is it shown that NPF drives the variance of  $N_{\text{CCN}}$  (line 210)? What is the role of background particles and primary emissions (Sect. 3.5) for variance of CCN concentration?

Calculation of the effect of NPF on cloud droplet number should be clarified. The explanation in lines 237-246 is confusing. Adiabatic rise does not produce water vapor (line 239) but has an effect on vapor pressure (or saturation). In don't think that cloud droplets "feel" particles (line 244), but they will compete with other cloud droplets about the available water vapor. Terminology could be similar with the section focusing on the effect of NPF on CCN. Is  $t_{\text{start}}$  the same in all cases (lines 203, 247 and 333)? Is  $t_{\text{Nd}}$  always 12:00 (line 246)? Is the end time case-dependent, why? If the event end times are different, this will have an effect on CCN and cloud droplet enhancements.

Variance analysis (line 261-265) needs a brief explanation in the main text (now there is nothing in the main text and the equations in the supplementary material seem to be incorrect). I suggest either presenting the method in the main text or removing this part from the manuscript.

Section 3.3 focuses on the effect of water vapor competition (or depletion), which is a known phenomenon related to cloud activation. The first paragraph (lines 271-287) could be reformulated for clarity (technical corrections listed below). Terms "with and without the water vapor depletion" (line 294) are misleading, because these droplet numbers are based on fixed updraft velocity and constant supersaturation, respectively. The depletion effect is included also in the latter approach because its constant supersaturation is taken from calculations "with water vapor depletion". Another reason for not to compare these two approaches is that the constant updraft velocity method accounts also for hygroscopicity variations. For these reasons I would not use term water vapor depletion/competition in this context.

Section 3.5 focuses on primary emissions, but a few things need to be clarified. First, please explain how these were identified as primary particles? Secondary (SOA) and primary (POA) organic aerosols are shown in Fig. 9, and there are high concentrations of SOA. How were SOA and POA calculated? This information is currently completely missing and must be added. Isn't there any background aerosol between 18:00 and 21:30 (lines

370-371) or at any other time? How was the growth rate of the newly formed particles calculated (line 385)? The method for separating PNSD of primary emissions (lines 386-390) needs a better explanation. It seems to be based on the fitted modes, so please explain why the three modes were selected (there could be more than three) and justify their origin (no background aerosol and no effect from boundary layer dynamics?). Why weren't the modes tracked from the beginning of NPF event? This could have shown their origin. Better justification is needed to convince readers that Figs 9e and 9f show PNSDs of NPF and primary emissions, respectively.

### **Technical corrections**

Below are line numbers and the part of the text that could be improved/fixed/clarified is quoted. Some of these appeared in the text more than once, but only the first line is listed here.

1: "cloud droplet" or maybe cloud droplet [number] concentration?

19: "markedly reduction"?

28: "particles are very low"

39: "a lot of researchers"

39: "parametric model"

62: "response to aerosol particle increases"

63-64: "as is different from"

75-76: "and its characteristics like nucleation, ..."

101: "catering"

103: “. And”

103: “the radiation in summer is relatively strong”

104-105: “contribute many CCN size particles”

109: “) .”

110: “particle sizes”

119-120: “measurements were deployed at ground level and at the 260 m level”

120: “2017), Before”

124: “Black carbon (BC), the BC”

128-129: “their surface water vapor phase equilibrium (supersaturation ratio) will gradually increase”

141: “formula (5),”

145: “the next calculation rule”

152: “cloud parametric model”

168: “(8)”

174: “field observation or empirical”

Figure 1: the x-axis is shows day and not time. It looks like some vertical axis have been cut, which should be explained in the text.

187: "As a typical NPF event..." is a long and confusing sentence

195: "The variation of  $N_{CCN}$  seems to be more consistent with that of  $N_{CN}$  than  $N_d$ "

199: "time node"

200: "we evaluating"

273: "linear correlated"

203: "hours later after"

217: "2017, respectively"

238-240: "The rise of environmental supersaturation..."

250: "define this impact continues"

276: "was thought caused"

277: "updraft velocities generate more water vapor"

278 "form more  $N_d$ "

279-281: "This fully suggests the difference between the fixed..."

281: "Because in the actual environment, it is often unable"

285: "The latter"

315: "nucleation particles condense and coagulate"

330: "accumulated mode particles"

342: "due to that there"

343: "pre-exist"

353: "Chang"

354: "get the this proportion"

363: "demonstrated related"

364: "Brines, M et al."

364: "primarily"

367: "after one hour of the eruption of newly formed"

376: "Mass fraction of aerosol composition"

395: "taking as a background"

409: "CCN and cloud droplet."

414: "V that can provide more sufficient water vapor"

418: "As a result, although 418 a larger increase..."

425: "variance of CDNC"

427: "leading to ~50% enhancement in the year from 1850 to 2000 change in cloud albedo."

429: "related cloud microphysical progress research"

433, 436: Code/data availability and Competing interests should be separate sections

504: "droplets[J]. Transactions"

516: "Zhang F\*, Li"

613: "Zhang, R., ... Li"