

Interactive comment on “Simultaneous measurements of new particle formation in 1-second time resolution at a street site and a rooftop site” by Yujiao Zhu et al.

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Received and published: 7 April 2017

The manuscript presents results from measurements of new particle formation (NPF) in Beijing during Spring and Winter using high time resolution particle sizers. The characteristics of new particle formation at two adjacent sites were compared to assess the impacts of local traffic emissions on NPF. Traffic exhausts might emit primary particles or gas phase precursors that contribute significantly to secondary particle formation and hence traffic emissions play important roles in severe haze formation in megacities such as Beijing in China. While the topic is important in atmospheric chemistry and is of interest to the general readers of this journal, the manuscript in general is yet to be improved and several major issues need to be resolved before the manuscript

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can be publishable in the journal.

Response: The authors thank the reviewer's comments and try our best to address the issues point-by point.

Major comments:

1. The rational of selecting plume events rather than regional events as examples must be clearly persuasive. Apparently, good “Banana shape” regional events were measured during the campaigns. Comparison of the differences of new particle formation between the two sites is of great interest. It will be clearer to see the impacts of traffic emissions on the new particle formation processes if those well-defined events were used as examples. For example, how particle number size distribution and particle composition might be affected by local emissions. The plume events are rather not well defined in term of the formation rates and the growth rates which will need to be resolved in the next comment.

Response: All simultaneous observations at two sites have been included and presented in the original version. During the two periods of this study, the authors didn't observe “banana shape” regional events simultaneously occurring at two sites. Therefore, the authors had no way to use them for discussion. This will be clarified in revision.

Similar to Class II NPF events with the particle growth to be undetectable presented in this study, extremely low growth rate of newly formed particles ($\sim 1 \text{ nm h}^{-1}$) in Beijing was also previously reported by Wehner et al. (2004). In our unpublished data, the authors simultaneously observed Class II NPF event and NPF event with extremely low growth rate at $\sim 240 \text{ km}$ distance (Fig. 1, the case will also be presented in Supplementary). In the last three years (data unpublished), we had simultaneous observations of NPF events at 100-500 km distance. The authors obtained six cases based on simultaneous observations at two locations, i.e., one case featured by Class II NPF vs Class II NPF, four cases featured by Class II NPF vs NPF with an extremely low growth rate, one case featured by Class II NPF vs NPF with “banana shape” particle growth.

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The authors strongly believed that Class II NPF events lasted for 4-8 hours should be considered as regional NPF events. “Banana shape” particle growth, extremely low particle growth and no detectable growth are probably related to spatial heterogeneity of gaseous precursors supporting the growth of > 10 nm particles, but NPF events indeed occurred regionally.

FMPS measured different sized particle number concentration in one second. The high time-resolution data can allow calculating the apparent formation rate of new particles such as J8 almost in any complicated situations. The reviewer's comments may be related to the low time-resolution data such as in 5-10 minutes time resolution, but it should not be the problem for FMPS measurements. This is also probably why FMPS measurements can show a few unique findings like in this study, but it is impossible for the 5-10 minutes time-resolution measurement previously conducted.

2. Particle formation rates and growth rates. First, the formation rate should be stick to J8 instead of new particle formation rate since particles in the range of 8-20 nm are rather too big to be called new particles. Also why a size range of 8-20nm is selected? Why is not 8-30 or 40 nm or others? The determination of the formation time (not the nucleation time) used in this paper seems to be objective and is of the authors' preferences, profoundly affecting the formation rate calculations. The width of the size range also affects the determination of formation time which will need to be clarified. The formation rate of nanoparticles in a plume event is difficult to calculate and needs to use a more sophisticate method than the simple one used in this paper.

Response: The authors agree that 8-20 nm particles should be called as grown new particles rather than new particles. In revision, we will add “Noted that J8 reflects a combination of nucleation and subsequent initial growth of new particles.” in the explanation of new particle formation rate.

The authors followed the conventional approach to calculate the apparent formation rate of new particles. In literature, the upper limits of the particle size was reportedly

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set at 20 nm, 25 nm or 30 nm (Kulmala et al., 2004; Kulmala and Kerminen, 2008; Yao et al., 2010; Kulmala et al., 2012). 20 nm was set as the upper limit in this study, following the suggestions by Kulmala et al. (2012), i.e., 1) newly formed particles rarely grow over 20 nm during the initial nucleation time, 2) nucleation may occur at some distances from the measurement point and the particles have grown a few nanometers before they were observed. In addition, the upper limit of 20 nm can minimize possible interferences from primary traffic particles. This will be clarified in revision.

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The authors used two methods to minimize the interference from primary traffic particles and the calculated apparent formation rates were consistent. Again, the high time-resolution data indeed allows calculating the apparent formation rate of new particles such as J8 in presence of strong interferences.

3. The classification of the nucleation events. It is very awkward to denote a nucleation event longer than 1 hour “a long term event”. It might sound better if “a long lasting event” or another name is adopted. Similarly, please change the notation of “a short term event” to another proper name. In addition, the two types of events are the well-known “regional events” and “plume events” in atmospheric aerosol sciences. It is not necessary to create new names for them.

Response: The authors would like to correct “short-term NPF event, long-term NPF event” to “short-lived NPF event, regional NPF event” (Stanier et al., 2010; Jeong et al., 2010).

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The authors strongly believed that Class II NPF events lasted for 4-8 hours should be considered as regional NPF events. Details can be found in the response to major comment 1.

4. Heterogeneity of NPF in Section 3.2. Quite a few “heterogeneity” were mentioned for NPF in both horizontal and vertical directions. It is really not that meaningful to emphasize the spatial inhomogeneity of particle formation because in the urban atmosphere, gas phase precursors are inhomogeneous and particle formation is also greatly constrained by local emissions and meteorological conditions.

Response: The authors thank and fully agree “the spatial inhomogeneity of particle formation because in the urban atmosphere, gas phase precursors are inhomogeneous and particle formation is also greatly constrained by local emissions and meteorological conditions”. A question is then automatically raised, i.e., what does it mean “regional NPF events”? It is just because it regionally occurred, but could be in different formation rates, growth rates, different initial bursting times? Most potential readers may disagree.

Most readers may also disagree with the reviewer and the authors at this point, i.e., the spatial inhomogeneity of particle formation very commonly occur in regional NPF events. However, the authors agree to cut a few details of “heterogeneity” in revision.

5. Reasons for the reduced NPF and the enhanced NPF at the street site respectively in the springtime and in the wintertime. It is very interesting to figure out the reasons behind those observed phenomena. First, the authors need to confirm that particle formation is always reduced in the springtime and always enhanced in the springtime at the street site. That will exclude the possibility of dominant effects from the meteorological conditions e.g. differences in wind directions or mixing heights, temperature, humidities etc. Second, the authors need to present more other companion measurements of gas phase precursors and chemical composition of nanoparticles in order to elucidate the mechanisms of reduced or enhanced NPF at the street site. Without the

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information, the proposed explanation for the observed opposite effects on NPF during springtime and wintertime is only speculative.

Response: At the street site, the reduced NPF events always occurred in the springtime while the enhanced NPF events always occurred in the wintertime. This will be highlighted in revision.

The authors provided three types of evidences from different angles to confirm the reduced NPF rather than simple comparison between rooftop site and street site measurements, i.e., Evidence 1: The lower particle number concentration (PNC) of nucleation mode particles at the street site mainly because of a shorter initial burst time. Evidence 2: The authors used the PNC at the street site subtracting the corresponding PNC at the rooftop site to calculate the difference. The authors then obtained the second evidence: the negative difference of nucleation mode particles against the positive difference of Aitken mode particles on NPF days. Evidence 3: Using the same approach, the authors obtained the third evidence: the negative difference of nucleation mode particles on NPF days against the positive difference of that on non-NPF days (Figs. 3 and 4 in the origin version). The second and third evidences are not affected by these factors suggested by the reviewer. All these will be better clarified in revision.

The authors also provided three types of evidences from different angles, rather than simple comparison between rooftop site and street site to confirm the enhanced NPF in the wintertime, i.e., Evidence 1: The significantly larger PNC of nucleation mode particles at the street site and a larger apparent formation rate of new particles mainly because of a shorter initial burst time. Evidence 2: The positive difference of nucleation mode particles in the wintertime against the negative difference of nucleation mode particles in the springtime on NPF days. Evidence 3: The larger positive difference of nucleation mode particles on NPF days against that on non-NPF days in the wintertime (Figs. 5 and 7 in the origin version). Again, the second and third evidences are not affected by these factors suggested by the reviewer. All these will be better clarified in revision.

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To confirm what the reviewer suggested requires simultaneous measurements of vapor precursors such as H₂SO₄, HOM, etc, and chemical composition of ~10 nm particles at the rooftop site and street site. This is indeed beyond the capacity of the whole research community, but not only for the authors. The authors tried the best and used all data we had. Other indirect evidences such as chemical composition of >50 nm particles were reportedly used to argue NPF in literatures. Theoretically, the arguments could be true, but also could be misleading by considering largely varying chemical composition in different sized atmospheric nanoparticles. The authors hope that the reviewer can agree this.

Minor comments:

1. There are a lot of typos, ill-sentences all over the manuscript. It is recommended that the manuscript should be carefully edited prior to submission. Below are a few examples: L31, specie; L208, a several minutes; L254, didn't detail; L270, didn't detail description; L292, "were available currently", . . .

Response: The authors are sorry for this and will correct the errors through the manuscript before re-submitting.

2. Rewrite all the figure captions clearly as those captions are hard to read and understand.

Response: Thank for the comment, the authors will rewrite all the figure captions.

3. L225, a few more sentences might be needed to explain why NPF inside the street canyon was reduced.

Response: The authors will revise L225 as: "Several factors can lead to the reduced NPF events at the street site, i.e., 1) a larger condensation sink because of more pre-existing atmospheric particles from primary emissions; 2) tall buildings along both the sides of urban streets can provide additional surface areas to scavenge gases and atmospheric particles (Yao et al., 2011); 3) vehicle-emitted NO reacting with RO₂ and

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suppressing NPF (Wildt et al., 2014).

4. L189, "estimating that the NPF possibly occurred in cleaner atmospheres over the region scale of ~120 km", "suggesting that the NPF likely occurred in cleaner atmospheres over the region scale of ~140 km in different NPF rates.", "The NPF was roughly estimated to occur in a semi-regional scale over Δ Lij50 km". How do you know the scales of those events?

Response: The authors agree that our estimation indeed suffered from uncertainty. In revision, it will revise as "the long lasting time for NPF implied that the event may occur in regional or semi-regional scale." Again, the authors believed that the long lasting Class II NPF events may occur regionally and the argument has presented above.

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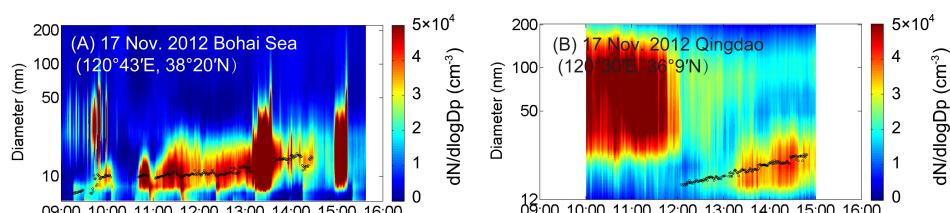


Fig. 1. Simultaneous observed Class II NPF event and NPF event with extremely low growth rate at ~240 km distance.

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