

Dear Editor,

We have made some revisions to clarify the issues according to reviewer's comments, and resubmitting the revised manuscript for publishing in the Journal.

Thank you and best regards.

Your Sincerely,

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Report #1

Anonymous Referee #3

Suggestions for revision or reasons for rejection (will be published if the paper is accepted for final publication)

The manuscript was improved to some extent in the revision. However, there are some arguments/conclusions that needed to be further clarified.

Response: The authors thank the reviewer's comments and try our best to respond and revise our manuscript accordingly.

1. The authors compared the number of their observations with the case of the gravitational wave in 2016 in response to reviewer 1. I have to say that the comparison is nevertheless meaningless and they cannot be compared in any way. The reviewer 1 raised a very important point that the authors ought to fully address. The authors present three evidences that support their conclusion. However, evidence one is ambiguous, i.e., the authors didn't give any strong evidences that micro-meteorology played a minor role in their case and they just presented a general experienced judgement which was not supported by any data. Evidence two and three are not evidences at all. They are just a representation of the data or to be straight, they are just what the data look like. I have the same concern as reviewer one if a general conclusion can be drawn based on just one or two observations.

Response: Considering the comments, we further improve our manuscript accordingly. We agree more data are needed to draw a general conclusion. In abstract, we added "However, the number of datasets used in this study is relatively small and

larger datasets are essential to draw a general conclusion”.

To address possible influence of micro-meteorology, in page 10, line 228, we added “Micro-meteorology at street sites may cause accumulation or dilution of atmospheric particles.” To better defend our evidences presented to support reduced and enhanced NPF occurring in spring and winter times, respectively. We made a few revisions as listed below:

Page 11, lines 231-244, it was revised as “Evidences 2-3 were obtained by subtracting the PNC of different sized particles at the rooftop site from the corresponding one at the street site and the size-segregated difference of PNC between the two sites in April was thereby calculated (Fig. 4). The difference was largely negative for particles <14 nm during the NPF periods on 25 and 27 April (solid lines) against the positive difference of Aitken mode particles. In contrast, such a difference was slightly positive for particles <14 nm during the non-NPF days and during the morning rush hours on 25 and 27 April prior to the occurrence of NPF events (Fig. 4, dash lines), because of increasing contributions from on-road vehicles at the street site as well as the accumulation effect associated with micro-meteorology at the street site. The same accumulation effect should theoretically exist during the NPF periods on 25 and 27 April, but the observed result showed the reverse. We thus obtained Evidence 2, i.e., the negative difference of nucleation mode particles on NPF days against the positive difference of those on non-NPF days. Considered the positive difference of Aitken mode particles during the NPF periods on 25 and 27 April (solid lines), it can be inferred that micro-meteorology favored an increase of nucleation mode particle number concentration at the street site. However, the observed result was contradictory to the interference. We thus obtained Evidence 3, i.e., the negative difference of nucleation mode particles on NPF days against the positive difference of those Aitken mode particles.”

Page 12, lines 269-270, we added “The different increasing patterns of nucleation mode particles at two sites strongly indicated that they were subject to different NPF mechanisms.”

Page 13, lines 277-278, we added “The measured concentration of BC was thereby tried for deduction and much poor correlation was obtained due to one-minute time resolution can’t allow successfully capturing vehicle spikes varying in a few seconds (see supplementary).”

Page 13, lines 292-293, we added “Theoretically, larger condensation sinks should cause stronger scavenging effects at the street site during the NPF periods in the wintertime than in the springtime. However, the difference of nucleation mode particles in the wintertime was positive.”

Page 13, line 297, we added “The reverse was true for 20-80 nm particles.”

Our Evidences 2-3 are unique and are self-independent on each other. We strongly believe that the revision has been demonstrated this issue clearly.

2. The authors emphasized many times on the importance of the instrument they employed. As far as I know, the fast mobility particle sizer has limited ability and limited applications so far. The data might have high uncertainties when the measurements were taken too fast compared to the traditional SMPS in term of accuracy of the sizes and concentrations.

Response: Over sixty publications related to FMPS measurements can be searched from popular databases, e.g., Web of Science, Google Scholar, etc, and dozens of these were published on international-recognized journals including Atmospheric Chemistry and Physics, Environmental Science and Technology, Journal of Geophysical Research (Atmosphere), Aerosol Science and Technology, Atmospheric Environment, Science of Total Environment and Environmental Pollution, etc. Building on these publications, the measurements made by two FMPS in this study were processed the correction for accuracy in size and concentration using recently developed approaches by Zimmerman et al. (2015).

Any instruments had advantages and disadvantages. This study fully took advantage of the high time resolution instrument and minimized the uncertainty on the measured number concentration and size. However, we delete a few statements on the importance of the instrument in revision.

3. The regional nucleation event by its definition is an event representing regional phenomenon. I am quite doubtful that a lot of such events are observed in Beijing at the sites as used in this study. Otherwise the measurements only represent nucleation events in a very local scale, say the environment around the site.

Response: NPF events were widely studied in Beijing since 2004. Previous studies showed a high frequency (~40%) of NPF events during spring and winter in the urban air of Beijing (e.g., Wu et al., 2007; Wang et al., 2013; Wang et al., 2017). In this study, the NPF occurrence frequency was 44% in the springtime and 50% in the wintertime, which was consistent with previous studies. Class II NPF events were previously assumed to occur in a very local scale because of short databases, particularly lack of simultaneous observations in polluted marine and coastal atmospheres. The data presented in the supplementary clearly showed that Class II NPF events can occur regionally.

Class II NPF events were confirm to occur regionally because 1) the duration period of NPF events lasted for 4-8 hours with the wind speed >4m/s, 2) the initial diameter of observed new particles was about 10 nm, needing at least dozens of minutes or 1-2 hours growing from the sub-3 nm clusters (Kulmala et al., 2007, 2013). The long

lasting time indicated the NPF events occur in a region range over tens to hundreds of kilometers. In revision, Class II NPF events to occur regionally has been well defended.

4. The authors used the proxy to calculate sulfuric acid concentrations and discussed a little bit the role of sulfuric acid. However I didn't see any presentation of the concentration or the detailed application of sulfuric acid concentrations. As discussed on p17, it is not clear when sulfuric acid should play a role quantitatively in Fig.9. The arguments seem to be speculative but I believe the authors should present a clearer picture to illustrate such an important point.

Response: In revision, the authors add some detail descriptions on the sulfuric acid, e.g., Page 14, lines 314-315, “the calculated concentrations of sulfuric acid were between 2×10^6 and 2×10^7 cm⁻³ during NPF periods, which was comparable with previous observations in Beijing (Yue et al., 2010; Zheng et al., 2011; Wang et al., 2017).” Lines 320-322, “However, the number of datasets in this study was too small to gain a reasonable equation to link the calculated concentrations of sulfuric acid with FRs.”

In addition, we added a new Fig 9b. Fig 9b showed that NMINP was determined by SO₂ to some extent, which further supported our argument on the importance of sulfuric acid on NMINP.

5. There are still lots of typos and ill-sentences that need to be proofread. A few examples:

1) Some of the picture captions are still not well written. For example, “the difference in number concentration” in Fig. 4 should be clearly defined in the caption. The readers don't have to refer to the text when read the figure; wrong link provides in Fig. 1.

Response: The authors have rewritten the caption of Fig.4 and check the other captions. The 3D-view figure in Fig. 1 was downloaded in the year of 2012. Now the map has been updated in revision.

2) P5 and others, a lot of times “on day-day” was used, while “during day-day” was also used. I think it might be better to be consistent and “during day-day” might be better here.

Response: Corrected.

3) P6: we write “cut a length of 2.8 m” not “cut as 2.8m in length; “in the supplementary” is enough to use, “document” in the “supplementary document” seems to be redundant. I believe there are still a lot in the text that the authors need to carefully edit.

Response: The authors corrected the errors and carefully edit the text accordingly.

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Report #2

Anonymous Referee #2

Suggestions for revision or reasons for rejection (will be published if the paper is accepted for final publication)

In the revised manuscript, the authors have done a good job to fully address the reviewer's concerns. Especially, they have provided additional analyses to validate the NPF events observed at both the street and rooftop levels. The reviewer indeed agree with the authors that amines, HOMs or other organics may play important roles in the new particle formation events, during both nucleation and the following growth processes. However, the reviewer must point out that no direct observations of amines and HOMs were provided in this study. Therefore, the reviewer suggests the authors insert a sentence in the conclusion (end of L416) to state that "although further validations including direct measurements of amines and HOMs in the newly formed particles are still needed." After addressing the above issue, the reviewer will recommend the manuscript for publication in ACP.

Response: The authors thank the reviewer's comment and have added the statement in the conclusion section.