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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Investigating three patterns of new particles growing to cloud condensation nuclei size in Beijing's urban atmosphere" *by* Liya Ma et al.

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Anonymous Referee #2

This study investigated seasonal variations of new particle formation (NPF) events in Beijing by using observations of particle size distributions and chemical compositions of aerosols and numerical model simulations. The authors found no apparent growth of new particles in winter whereas the growth of new particles to CCN size (50 or 75 nm) was often observed in summer. The three patterns of NPF events during the summertime were discussed in terms of secondary aerosol formation, evaporation of semi-volatile species, and spatial heterogeneity of NPF events. The scope of this manuscript

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is well suited to ACP, and the data obtained by the authors are valuable and important to understand the mechanisms of NPF events in urban atmospheres. However, the current manuscript needs substantial revisions before the manuscript is considered as a publication of ACP as shown below.

Response: Thanks for the reviewer's comments. We will try our best to respond and revise our manuscript accordingly.

1) Page 1, Line 17: "11/27" should be revised. For example, "11 new particle formation (NPF) events out of 27 events" may be better. Other parts written similarly in the text should also be revised.

Response: Thanks for the reviewer's suggestion. We will revise this in the manuscript.

2) Page 2, Lines 21-28: The authors described what they did in this study. However, it is not clear to me which parts of this manuscript are scientifically new. There are many previous studies on NPF in Beijing and other urban areas. The authors should summarize these previous studies and describe what are well understood and what are poorly understood in Introduction. Then, the objectives of this study should be described more clearly. The sentence at Lines 18-20 (Thus far, which chemicals drive the growth.) is a point poorly understood, but I don't think the understanding on this point was improved by this study.

Response: Thanks for the reviewer's comment. The NPF events in Beijing have been widely reported (e.g., Wu et al., 2007; Yue et al., 2010; Wang et al., 2013; Kulmala et al., 2016; Chu et al., 2019). Previous studies focus mainly on the nucleation precursors (e.g., sulfuric acid, organic vapors) and the coagulation scavenge under high loadings of pre-existing particles. In this paper, we concentrated on the growth process of newly formed particles, and found that the new particles may encounter the ceiling at 20$\sim$50 nm before they grow to CCN size. The growth mechanisms can improve our understanding of the effects of NPF events on climate. More revisions will be added to better demonstrate the progress on NPF in Beijing. The sentence at Lines 18-20 will

be removed in the revised manuscript.

3) Page 4, Line 3: Equation (2) Please add descriptions on the uncertainty of this equation. Response: The uncertainty of equation (2) has been reported by Lu et al., (2019). The correlation coefficient between measured [H2SO4] and the proxy function was 0.83, and the relative error was less than 20%. These uncertainties will be added in the revised manuscript.

4) Page 4, Line 11: The SPR analysis (section 4.5) is not meaningful. It is hard to quantitatively estimate the survival fraction of new particles from this equation because the SPR values can be greater than 100% in many cases (Table 1). I think the authors may be able to calculate the loss rate of new particles during each NPF event from CS.

Response: Thanks for the comment. In the revised manuscript, we will remove the discussion of SPR into the supporting information.

5) Page 4, Line 13: Please clarify why 3 sigma was chosen.

Response: The particle number concentration follows the lognormal distribution. In the function curve, 1 sigma covers 68.27% area, 2 sigma covers 95.45% area, and 3 sigma covers 99.74% area. In this study, we use 3 sigma to represent almost all particles in this mode.

6) Page 4, Line 20: Massrequired The authors compared Massrequired with the changes in mass concentrations of organic and nitrate aerosols, but the latter is generally controlled by accumulation mode particles, not nucleation mode particles. The comparison between Massrequired (changes in aerosol mass for nucleation or Aitken mode particles) and the changes in mass concentrations of organic and nitrate aerosols (mainly controlled by accumulation mode particles) is therefore not so meaningful (in sections 4.1-4.3).

Response: Ideally, the chemical composition of nucleation mode particles would be the best to explain the growth of new particles. The data was not available in this study

unfortunately and it is still a common challenge in research community. In our previous observation using AMS, the real-time chemical composition of sub-100 nm particles has large artifacts, and AMS measurements for these small particles are not reliable. Thus, in the literature, the PM1.0 chemical composition was usually used to explain the new particles chemical information, e.g., Salimi et al. (2015) and those revised by Chu et al., (2019). In our manuscript, we combined all chemistry information to explain the growth of new particles, and the uncertainties will be added in the revised version.

7) Page 5, Lines 1-2: Please provide some brief descriptions on model setups.

Response: Thanks for the comment. We will provide brief descriptions on model setups in the revised manuscript.

8) Pages 5, Lines 8-9: Please describe on model evaluations more clearly (e.g., the degree of agreement with observations, chemical species evaluated).

Response: Thanks for the comment. We will add the model evaluations as follows: "The model results generally could meet the benchmark criteria of above four species (US-EPA, 2007), with correlations of higher than 0.51 (Table S1). The concentrations of SO42- and NH4+ had been slightly overestimated (with Normalized Mean Bia (NMBs) and 12%, 6%), while the concentrations of NO3- and OOA were underestimated (with NMBs of -29% and -39%). Detailed evaluation results of this study could be found in Supporting Information."

9) Page 5, Lines 23-27: The unit of number concentrations in this paragraph is probably not correct.

Response: We will correct the unit in this paragraph.

10) Page 6, Lines 2-19: Please clarify why Class II was subclassified to 4 scenarios. What is the purpose of this?

Response: In Class II, we would like to distinguish scenario 1 from other types. New particles in Scenario 1 can grow to 27-48 nm and then stop growing. The stop lasted

for a few hours until the new particle signal dropped to a negligible level. This growth pattern means the new particles encounter the ceiling in the growth, and we try to interpret these events in terms of meteorology, physical and chemical properties of particulate matter. However, for other scenarios, no continuous observations can allow us to justify whether the ceiling exist or not.

11) Page 8, Line 3: the contribution of <2% Please clarify how the authors estimated this contribution. I think the authors have sulfate data observed by AMS. The data can be shown like OM and nitrate in Figures 2-4.

Response: We estimated the contribution of H2SO4 vapor to the particle growth followed the equation of Kulmala et al. (2001), i.e., R = ([H2SO4]avg/C) $\times$ 100%. where [H2SO4]avg is the average concentration of H2SO4 during the particle growth period, and C is the total concentration of condensable vapor for the particle growth, which can be calculated as described by Kulmala et al. (2001). We will add this equation in the revised manuscript. The calculation may be more accurate to reflect the contribution than the use of sulfate in PM1.0.

12) Page 8, Line 7: 13 ug m-3 Please clarify how the authors estimated this value. Did the authors consider the spread of particle size distributions? (like 3 sigma in equation (3)).

Response: We calculated the required mass follow equation (4). The averaged integral value of particle number concentration also consider the 3 sigma in the lognormal distribution.

13) Page 8, Lines 6-8: As I described above, the comparison between the required mass (13 ug m-3) and PM1.0 enhancement (15 ug m-3) is not so meaningful because the former focuses on nucleation/Aitken mode particles but the latter is usually dominated by accumulation mode particles. I think what the authors can do here is to calculate mass concentration changes for sulfate, nitrate, ammonium, and SOA and to discuss which changes are the largest during the growth periods of new particles.

Response: Thanks for the question. In the revised manuscript, we will use the change of sulfate, nitrate and OOA instead of OM, to discuss which chemical contributes largest to the particle growth. Besides, the uncertainties will be added in section 2.1.

14) Page 8, Line 7: OM can be divided into HOA (POA like) and OOA (SOA like) by using m44 and m57 signals. Only OOA can contribute to the growth of particles.

Response: Thanks for the comments. In the revised manuscript, we use the OOA data to discuss the growth of new particles.

15) Page 9, Lines 3-4: I don't agree with this authors' description. The simulated OA and nitrate cannot be used to interpret the data unless the authors evaluate the simulations with observations.

Response: We will add the model evaluations as follows: "The model results generally could meet the benchmark criteria of above four species (US-EPA, 2007), with correlations of higher than 0.51 (Table S1). The concentrations of SO42- and NH4+ had been slightly overestimated (with NMBs and 12%, 6%), while the concentrations of NO3- and OOA were underestimated (with Normalized Mean Bia (NMBs) of -29% and -39%). Detailed evaluation results of this study could be found in Supporting Information."

16) Page 10, Line 16 Delete "(ON)".

Response: Corrected.

17) Page 10, Line 28: OM can be divided to HOA and OOA as I described above.

Response: We will change OM to OOA in the main text and figures.

18) Page 11, Lines 5-6: "Repartition of the. . ." This part should be removed because no data can support this sentence.

Response: Corrected.

19) Page 12, Line 19 “then in”, “transience”: they should be corrected.

Response: We will remove “then in”, and change “transience” to “the changes of season”.

20) Page 12, Line 26: Section 4.5 As I described above, this section is not so meaningful and should be removed. How did the author consider the contribution of primary particles in this analysis?

Response: We removed section 4.5 into the supporting information. The contribution of primary particles, such as vehicles particles, restaurants or factories emissions will be added in the supporting information (Figure 1-2). In this study, we use a high-time resolution particle sizer, i.e., FMPS, to measure the particle number size distribution (PNSD) in 1 s time resolution. The high time resolution of FMPS can allow clearly identify the signals of newly formed particles from preexisting ambient particles, e.g., freshly emitted particles from combustion, as well as the mixing process of the different types of particles (Liu et al., 2014; Man et al., 2015; Zhu et al., 2017, 2019). During the NPF events (e.g. Figure 3), COA occasionally influences the new particles signal, and the growth of new particles was consistent with the increase in LO-OOA and MO-OOA. Therefore, we argued that we can distinguish the primary particles from new particles signal, and the growth of new particles depends largely on the condensational growth.

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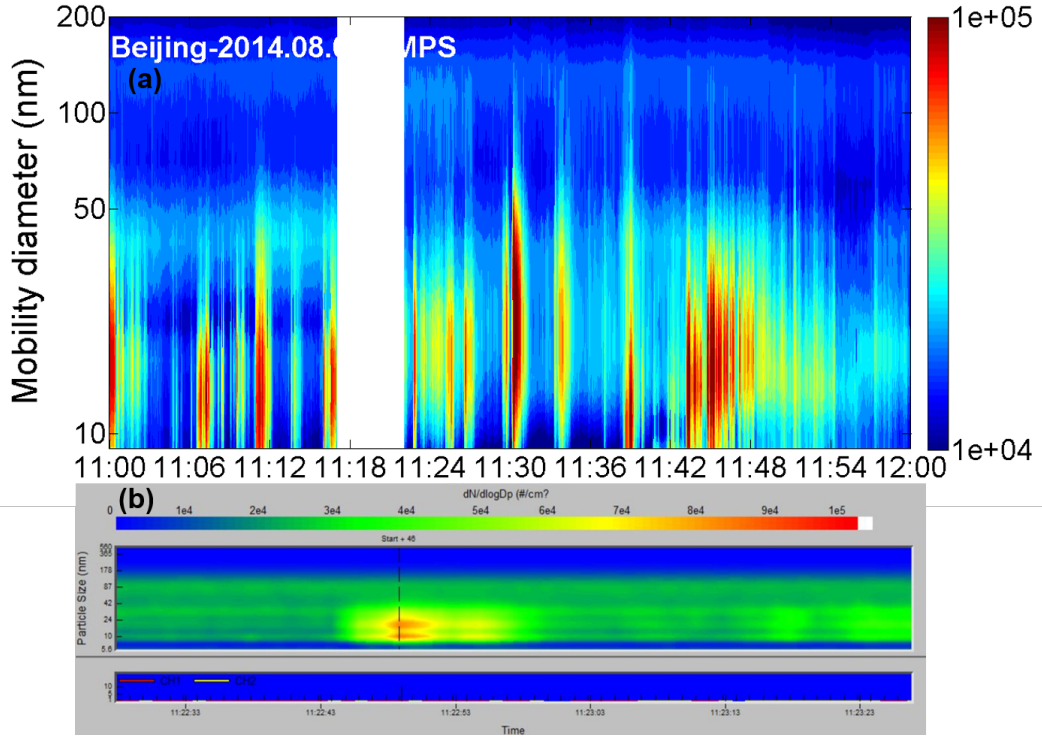

Fig. 1. Contour plot of traffic emissions (a) and the raw FMPS data of the vehicle spikes (b)

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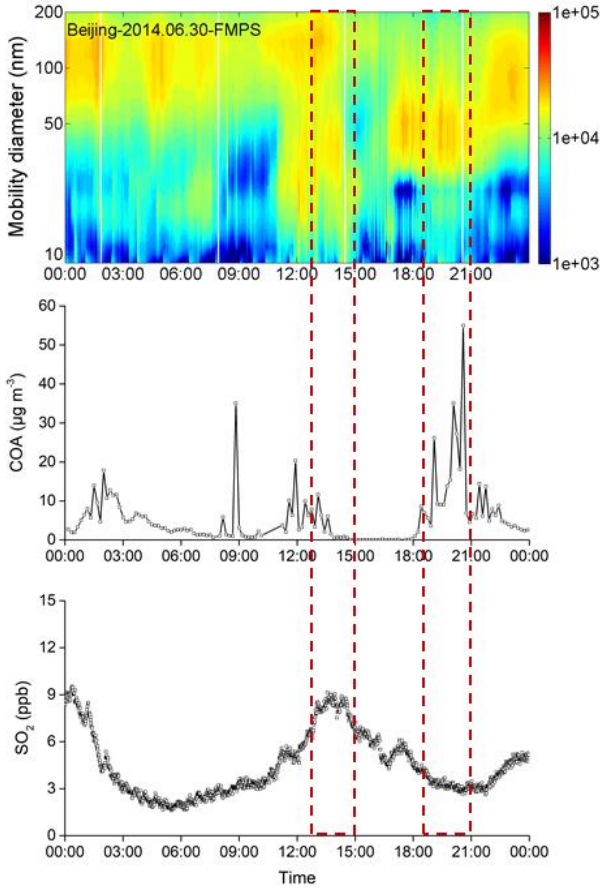

Fig. 2. Fresh industrial emissions with high SO2 (12:30-15:00) and the cooking emissions with increased cooking OA (COA, 18:20-21:00)

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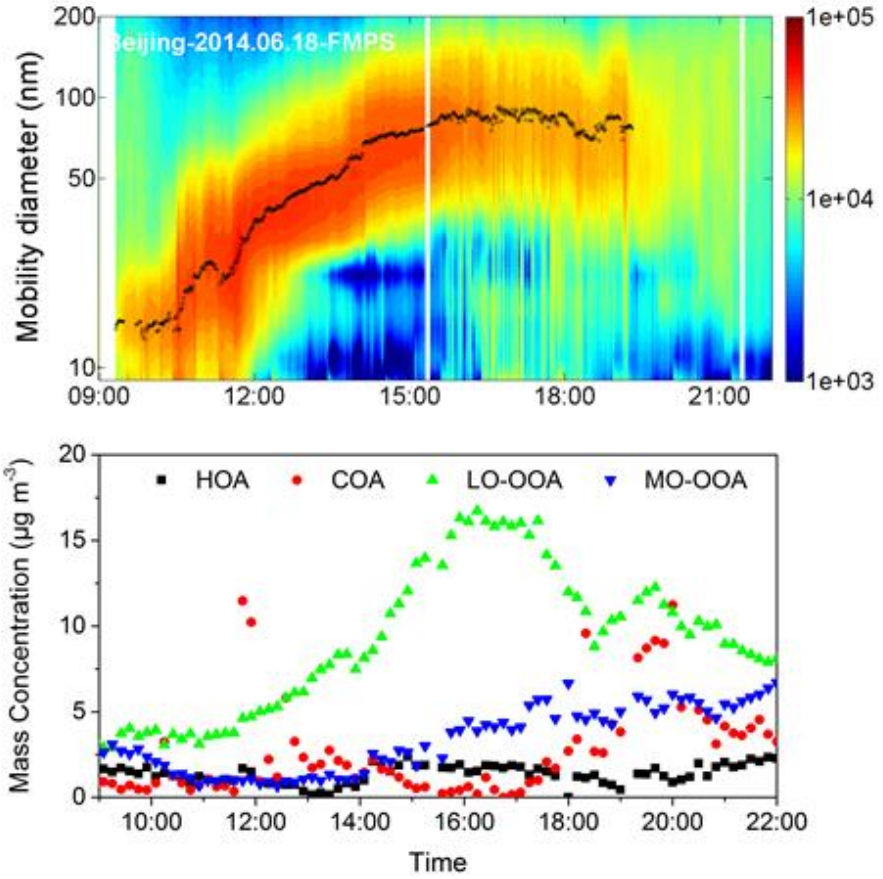

Fig. 3. NPF event and the variations of hydrocarbon-like OA (HOA), cooking OA (COA), less oxidized oxygenated OA (LO-OOA) and more oxidized oxygenated OA (MO-OOA) on June 18, 2014

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