Response to reviewer's comments

Editor Decision: Publish subject to minor revisions (review by editor) (22 Oct 2020) by Stefania Gilardoni

Comments to the Author:

One of the reviewers suggested further revision of the manuscript. Here is the list of the changes suggested by the reviewer.

1) Page 2, line 24-

The authors summarized previous NPF studies in Beijing well in this paragraph. However, it is not clear yet in the revised manuscript what are different from previous studies and what are scientifically new. For example, in Page 3, lines 3-6, the authors described as follows: "sulfuric acid and/or organic vapors have been proposed to drive particle growth in different NPF events (Wiedensohler et al., 2009; Yue et al., 2010; Wu et al., 2016). Recently, the formation of NH_4NO_3 has been proposed as a driver of the rapid growth of newly formed particles in field studies and chamber experiments (Zhu et al., 2104; Man et al., 2015; Wang et al., 2020)." Which parts of this study are different from these previous studies? How was the understanding of NPF in Beijing advanced in this study? Because the importance of NH_4NO_3 and organics has already been found in previous studies, the sentence at Page 3, lines 6-7 ("The role of NH_4NO_3 in the growth of newly formed particles in Beijing remains poorly understood.") is not enough.

Response: In the revision, page 3, the top paragraph, after "The patterns of particle growth have not been well characterized.", we add "For example, the occurrence frequency of NPF events in which newly formed particles can grow to CCN size is virtually unknown. The same can be said to season trends in the occurrence frequency.". After "The role of NH_4NO_3 in the growth of newly formed particles in Beijing remains poorly understood." we add "The contributions of sulfuric acid, organic vapors and NH_4NO_3 to the growth of newly formed particles at different sizes are also non-existent."

2) Page 5, lines 23-

The SP analysis (this part and section 4.5) should be removed. I cannot understand why the authors can say that the calculated SP is valid for most of NPF events. This analysis implicitly assumes that all atmospheric particles in Beijing are formed during a new particle formation event in Beijing. I think this assumption is unrealistic. Some aerosol particles in Beijing may be preexisting particles (e.g., primary particles, new particles formed a few days ago), transported from other cities, and/or transported from higher altitudes. Although fresh primary particles may be small enough, many particles in the atmosphere (>50 nm or >70 nm) are probably produced by non-NPF or non-local NPF processes. This analysis is therefore not valid and should be excluded from the manuscript.

Response: We thank very much for the reviewer fastening upon this point. This helps us to eventually find the right way to calculate the net maximum number concentration of grown new particles beyond 50 nm or 70 nm. The same can be said to the calculated SP. In the revision, page 5, we revised "To calculate the survival probability (SP) of grown new particles beyond 50 nm or 70 nm, temporal variations in $N_{50\text{-}200\ nm}$ and $N_{70\text{-}200\ nm}$ were plotted, e.g., Fig. S2a-b showed the temporal results on 25 August 2014. The stable minimum $N_{50\text{-}200\ nm}$ or $N_{70\text{-}200\ nm}$ can be clearly identified approximately 2-3 hours after the NPF event to be observed. The stable maximum $N_{50\text{-}200\ nm}$ or $N_{70\text{-}200\ nm}$ can be also clearly identified approximately 11-13 hours later. The difference between the two values was used to estimate the net maximum number concentration of grown new particles beyond 50 nm or 70 nm, i.e., $N_{50\text{-}200\ nm}$ (*net*) and $N_{70\text{-}200\ nm}$ (*net*). Thus, $SP_{50\ nm}$ and $SP_{70\ nm}$ were estimated as:

$$SP_{50\ nm} = \frac{N_{50-200\ nm}\ (net)}{NMINP} \times 100\% \quad (5)$$

$$SP_{70\ nm} = \frac{N_{70-200\ nm}\ (net)}{NMINP} \times 100\% \quad (6)$$

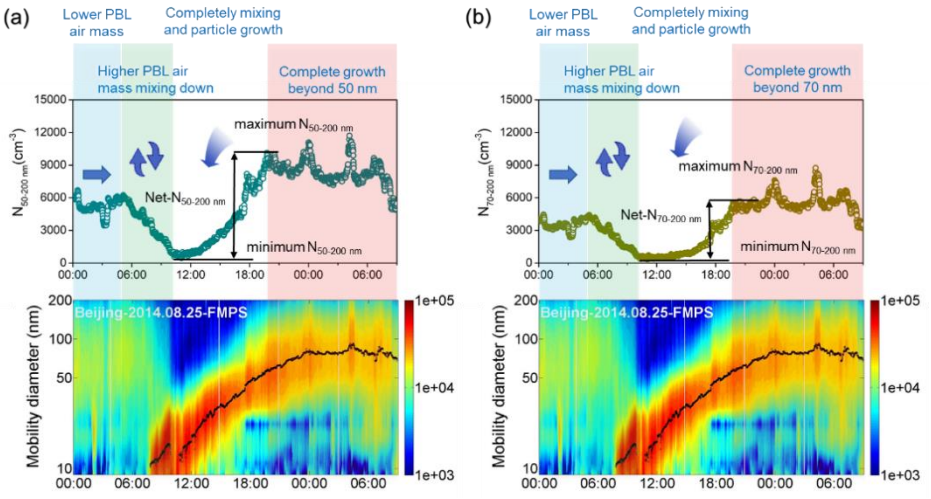

Fig. S2 Schematic diagram of $N_{50\text{-}200\ nm}$ (*net*) (a) and $N_{70\text{-}200\ nm}$ (*net*) (b) on 25 August 2014"

Using the right approach, the calculated $SP_{50\ nm}$ and $SP_{70\ nm}$ have been updated in the revision. So does the analysis.

3) Page 6, line 9, equation (5)

Please clarify why D_{pg1} and D_{pg2} (median diameters) were used instead of observed particle diameters. It is more straightforward to use the observed diameters of the size distribution observations directly rather than using the D_{pg1} and D_{pg2}.

Response: In the revision, page 6, line 4, we add "Followed Equation (4) proposed by Kulmala et al., (2001), in which D_{pg1} and D_{pg2} are used to calculate C,".

4) Page 6, lines 16-, Section 2.3

The model simulations cannot be used to interpret the relative importance of NH_4NO_3 and SOA because SOA is severely underestimated. Model simulations and their interpretation should be removed from the manuscript. The main results of this study do not change even if the model results were excluded. I think the discussion using the model simulations decreases the reliability of the whole results shown in this manuscript.

Response: We agree the model simulations cannot be used to interpret the relative importance of NH_4NO_3 and SOA. However, the modeled results can be used to argue the importance of NH_4NO_3 based on the reasonable performance in comparison with the observations. In the revision, quantitative analysis of simulated ammonium nitrate and semi-quantitative analysis of simulated SOA were used. Please see the revision.

5) Page 7, line 8

NMB of -39% is probably wrong for SOA when I see Figure S7. Simulated SOA is severely (more than an order of magnitude?) underestimated.

Response: The NMB was mistakenly calculated by using the simulated SOA against the measured OOA. The mistake has been corrected in the revised Table S1. This does not affect the Fig. S7. Sorry for this error and thank very much for the important comments.

6) Page 7, line 15

Figure S2 can be used in the main manuscript.

Response: Agree. Done.

7) Page 8, line 8-

Line 10: Fig S3b -> S3c, line 16: Fig S3c -> S3e, line 18: Fig S3d -> S3g, line 20 Fig. S3e -> S3i

Response: Done.

8) Page 10, line 26

Fig 2b does not show that sulfuric acid contribution is small.

Response: The sentence has been revised as "Based on the observed mixing ratio of SO_2 shown in Fig. 3b and Equations 2–4, sulfuric acid was estimated to contribute < 2% to particle growth during the whole NPF period"

9) Page 11, line 19-Page 12, line 5
These three paragraphs should be removed as I described in the comment 4.

Response: See our response above.

10) Page 13, lines 5-8
This paragraph should be removed also.

Response: See our response above.

11) Page 14, lines 4-12
Authors explain what was obtained but do not explain why this result was obtained.

Response: The sentences have been revised as "The growth rate of newly formed particles is an intensity quantity and mainly determined by the concentrations of condensable vapors such as sulfuric acid, organics of various volatilities, nitric acid, and ammonia (Zhang et al., 2012; Ehn et al., 2014; Man et al., 2015; Lee et al., 2019). In contrast, D_{pgmax} values are determined by the total amount of vapors condensed on grown new particles (an extensive quantity), which may or may not have a positive correlation with the concentrations of these vapors (Zhu et al., 2019)."

12) Section 4.5
This section should be removed as I commented before.

Response: See our response above.