RE: A point-to-point response to reviewers' comments

"Strong Atmospheric New Particle Formation in Winter, Urban Shanghai, China" (acp-2014-719) by Shan XIAO, Mingyi WANG, Lei YAO, Markku Kulmala, Bin ZHOU, Xin YANG, Jianmin CHEN, Dongfang WANG, Qingyan FU, Douglas R. Worsnop, and Lin WANG

We are grateful to the helpful comments from the anonymous referees, and have carefully revised our manuscript accordingly. A point-to-point response to the reviewers' comments, which are repeated in italic, is given below.

Anonymous Referee #1

This paper reports on field measurements of new particle formation in the urban atmosphere of Shanghai. The concentrations of clusters and nanoparticles along with concentrations of potential nucleation precursors obtained during the field campaign are used to derive the nucleation and growth rates. Conclusions are made regarding the different factors that govern the nucleation and growth mechanisms. This study provides substantial data that will help to understand mechanistic details and identify the sources of secondary aerosols in China, where aerosol pollution is severe but its causes are not understood. A major deficiency of this paper is that presented data do not always substantiate the conclusions and interpretations; also, some crucial statistical information is missing. These deficiencies need to be addressed before the manuscript can be considered for publication.

Major points:

1. *As presented, the paper lacks clear focus. It will be beneficial to list scientific questions that were to be addressed by the field campaign.*

Reply: A new paragraph has been added to clearly present the scientific goals of this field campaign, which reads (L93-106),

Direct measurements of atmospheric nucleation rates down to 1.5 ± 0.4 nm provide a better and more accurate characterization of atmospheric nucleation, since the indirect calculation of atmospheric nucleation rates from the formation rates of 3 nm particles leads to substantial uncertainty due to our incomplete understanding of condensational growth and coagulation scavenging of particles in the 1.5 to 3 nm range (Anttila et al., 2010; Korhonen et al., 2011). With the growing number of reports of real nucleation rates in clean atmosphere (e.g., Kulmala et al., 2012; Yu et al., 2014), it is ideal to measure nucleation rates in a polluted urban atmosphere to find out how atmospheric nucleation rates vary under different atmospheric conditions. In addition, the nucleation mechanism in a polluted urban atmosphere that is vital to understand atmospheric nucleation at a global scale and for atmospheric model development can be preliminarily investigated by examining the relationship between the measured atmospheric nucleation rates and the well-accepted precursor gases that exist in high concentrations.

In addition, we have revised the abstract to present the main scientific findings of this study (L30-31).

2. *Based on the slope (L359-367), authors conclude that the nucleation of new particles can be explained by the activation theory. However, no standard deviation of the slope is given anywhere in the article. In fact, Figure 4 shows very poor correlation between the nucleation rate and sulfuric acid concentration, so the reported value of 0.64 could be significantly higher or lower. This needs to be addressed. Similarly, the standard deviation must be provided for the slope obtained from Figure 5 for ammonia.*

What is the uncertainty in the calculation of proxy sulfuric acid concentration? Is daytime average sulfuric acid proxy a good metric for producing Figure 4? Maybe better use the peak concentration?

Reply: According to Mikkonen et al. study (2011), the uncertainty for the calculation of sulfuric acid proxy is 42%. We have now added the error bars of the sulfuric acid proxy in Figure 4, and clearly stated in our experimental session that (L231-233) "The relative error between calculated sulfuric acid proxy and measured sulfuric acid concentration is estimated to be 42% (Mikkonen et al., 2011)."

Following Anonymous Referee #1's advice, the peak concentration of sulfuric acid proxy instead of daytime average of sulfuric acid proxy is now used in Figure 4, which gives of a power exponent of 0.65±0.28. To address the correlation between the nucleation rate and sulfuric acid proxy, we have now stated that (L406-414) "Our P of 0.65±0.28 is of a significant uncertainty, which could come from the uncertainty during the calculation of sulfuric acid proxy $[H_2SO_4]$ and the scarcity of our data points. The upper limit of our P indicates that nucleation occurs after activation of clusters containing one molecule of sulfuric acid, with subsequent growth involving other species (Kulmala et al., 2006). The lower limit, on the other hand, suggests that a less important role of sulfuric acid in the critical nucleus during our campaign, which is unlikely to be true according to numerous previous studies (Weber et al., 1996; Sipila et al., 2010; Yu and Hallar, 2014)."

In addition, since the diurnal profile of ammonia was irregular, there was no peak concentration for ammonia. In this case, daytime average concentration of ammonia is used to investigate the relation between the nucleation rate and ammonia. In figure 5, the error bar of the ammonia concentration represent the standard deviation of daytime average concentration of ammonia, and a power exponent of 0.57±0.17 was obtained.

3. *Figure 8 shows no clear correlation between GR and $PM_{2.5}$. What are the corresponding correlation coefficient and standard deviation for the slopes? Would aerosol surface provide a better correlation? If not, in my opinion, this fragment and Figure 8 should be removed.*

Reply: The correlations coefficient and standard deviation for the slopes are -0.019 ± 0.022, -0.041±0.071, 0.135±0.054 for $GR_{1.35\sim2.39}$, $GR_{2.39\sim7}$ and $GR_{7\sim20}$ with $PM_{2.5}$, respectively.

We have tested the correlation between GR and aerosol surface, which does not give a better correlation. Hence, as suggested by Anonymous Referee #1 and #2, Figure 8 and corresponding texts have been removed in the revised manuscript.

4. *The abstract gives an excessively detailed description of some of the obtained results, e.g., growth rates binned for many size ranges are given with standard deviations. On the other hand, no standard deviation is provided for the power exponent of the sulfuric acid, although the latter is used to make an important conclusion regarding the nucleation mechanism.*

Reply: The standard deviation for the correlation between the nucleation rate and sulfuric acid proxy is elaborated in the revised manuscript. We have now stated that (L28-31) that "Correlation between nucleation rate ($J_{1.34}$) and sulfuric acid proxy indicates that nucleation rate $J_{1.34}$ was proportional to a 0.65±0.28 power of sulfuric acid proxy, indicating that the nucleation of particles can be explained by the activation theory".

5. *I suggest adding a scatter plot showing the concentration of 1.34-10 nm nanoparticles as a function of the aerosol surface area obtained from SMPS measurements (L439- 452). The scatter plot may show a better dependence. Also, the surface area may be a better parameter than $PM_{2.5}$.*

Reply: As suggested by Anonymous Referee #1, the $PM_{2.5}$ mass plot has been replaced with the surface area plot in Figure 7. It is evident that NPF events in urban Shanghai occurred with low levels of aerosol surface area and that high sulfuric acid favored NPF events when aerosol surface area was low. The text in Section 3.4 has been revised accordingly.

Minor comments and corrections:

6. *L68: add comma after 'air pollution'*

Reply: A comma has been added.

7. *L93,99: replace 'have been' with 'are'*

Reply: As suggested by Anonymous Referee #1, the sentence at L110 has been revised. As suggested by Anonymous Referee #2 (comments #14), the sentence at L99 has been removed in the revised manuscript.

8. *L119: A flow rate of 153 CFM corresponds to more than 4 m^3/min. This does not appear to be a 'low-volume blower'.*

Reply: We have now stated that "Ambient air was drawn into a stainless steel manifold of 5.0 m length and 4 inch inner diameter at a flow rate of 153 CFM using a blower (Model DJT10U- 25M, NUSSUN, China)" (L133-135).

9. *L127: replace 'silica' with 'silicon'*

Reply: We replaced "silica" with "silicon" (L142).

10. *L136: replace 'grow' with 'grows'*

Reply: We replaced "grow" with "grows" (L152).

11. *L170: 'aerosol cutter' is not a good term*

Reply: This sentence has been removed in the revised manuscript because the mass concentrations of $PM_{2.5}$ are not presented in the revised manuscript.

12. *L176: must be 'Data Processing'*

Reply: We replaced "Date Processing" with "Data Processing" (L190).

13. *L182: perhaps a better term can be used instead of 'growth losses'. Unlike coagulation, growth does not reduce the particle number concentration.*

Reply: We used the term "condensational growth out of the considered size range", instead (L196).

14. *L229: rho_d is not defined. Also, is it not necessary to account for the uptake of water by sulfuric acid, e.g., by assuming that sulfuric acid is instantaneously equilibrated with gas-phase water?*

Reply: The definition of rho has been added as a correction factor according to (Nieminen et al., 2010) (L243).

We agree with Anonymous Referee #1 that the particle growth due to the hydration of H_2SO_4 should be taken into account. We have now stated that (L248-253) "The particle growth due to the hydration of H_2SO_4 is taken into account by assuming that sulfuric acid is instantaneously equilibrated with gas-phase water. During our campaign, daily average RH varied between 28.7%~60.0%. Hence, using the H_2SO_4-hydrate distribution data given by Kurtén et al.(2007) , the density and mass of the average hydrated H_2SO_4 molecule at 50% relative humidity is calculated and further used in equation (6)".

15. *L238-242: 'In this study, ……event' – this sentence is very heavy. Consider rewriting.*

Reply: We have now stated that (L264-268) “In this study, we define an observation day with appearances of sub-3 nm clusters/particles over a time span of hours and subsequent growth to larger sizes for a few hours as a NPF event day. In this case, a NPF day will present a banana-shaped contour plot of particle size distributions obtained from SMPS (Dal Maso et al., 2005). We focus on characteristics and potential mechanisms of these events”.

16. *L273-276: Elaborate explicitly on why similar size distributions ‘suggest’ that photochemical products contribute to the formation of smallest particles. It is not clear as written.*

Reply: We have now stated that (L296-302) “On the NPF day, 1.34-3 nm particles appeared as early as 7 am in the morning that was right after sunrise (6:42 am on Dec. 11th, 2013), reached its maximum just before noontime, and spanned for almost the whole daytime (sunset at 4:52 pm, Dec. 11th, 2013), suggesting that photochemistry products likely contribute to formation of smallest particles. This size distribution of atmospheric neutral and charged clusters/particles by a scanning PSM is identical to that measured at Hyytiälä, Finland (Kulmala et al., 2013)”.

17. *Section 3.2 and Table 1: It would be beneficial to add and discuss the data obtained in other sulfur-rich locations, e.g., Atlanta*

Reply: Results from two Atlanta studies (Stolzenburg et al., 2005; Kuang et al., 2012) have been added into Table 1 and texts of session 3.2. We have now stated that (L354-360) “ $GR_{1.35\sim2.39}$, $GR_{2.39\sim7}$, and $GR_{7\sim20}$ were in the range of 0.49-8.1, 3.1-35.7, 4.5-38.3 nm h^{-1}, respectively. The arithmetic average values of $GR_{1.35\sim2.39}$, $GR_{2.39\sim7}$, and $GR_{7\sim20}$ were 2.0±2.7 (one standard deviation), 10.9±9.8 and 11.4±9.7 nm h^{-1}, respectively, which are comparable to 3-20 nm h^{-1} for nucleation mode particles in another sulfur-rich city, Atlanta, GA (Stolzenburg et al., 2005). In addition, $GR_{1.35\sim2.39}$ at Shanghai is smaller than the growth rates (5.5-7.6 nm h^{-1}) for particles in 1~3 nm geometric diameter range in Atlanta (Kuang et al., 2012).”

18. *L331-336: This sentence is very heavy. Consider adding a small table and revising the sentence.*

Reply: We have now stated that (L360-366): “A closer examination of growth rates was performed by dividing $GR_{1.35\sim2.39}$ into growth of clusters/particles from one bin to another, i.e., $GR_{1.35\sim1.39}$ (1.6±1.0 nm h-1 from the bin of 1.34-1.37 nm to the bin of 1.37-1.41 nm), $GR_{1.39\sim1.46}$ (1.4±2.2 nm h-1 from 1.37-1.41 nm to 1.41-1.52 nm), $GR_{1.46\sim1.70}$ (7.2±7.1 nm h-1 from 1.41-1.52 nm to 1.52-1.89 nm), and $GR_{1.70\sim2.39}$ (9.0±11.4 nm h-1 from 1.52-1.89 nm to 1.89-3.0 nm). These growth rates show a clear size-dependent particle growth (Fig.3), owing to…”.

19. *L336: Replace ‘size depend’ with ‘size-dependent’*

Reply: We replaced “size-depend” with “size-dependent” (L366).

20. *L336: Elaborate explicitly why size-dependent growth owes to nano-Kohler activation. What about size-dependence of the accommodation coefficient of sulfuric acid on clusters/nanoparticles? Isn’t it supposed to decrease for with decreasing size?*

Reply: We meant to suggest that “size-dependent growth owes to the combined effect of nano-Köhler activation, Kelvin effect and surface or volume-controlled reaction corrected for the Kelvin effect on surface or volume concentrations, not only owes to nano-Köhler activation”. According to the nano-Köhler activation theory, inorganic stable nano-clusters will be activated into aerosol particles in a supersaturated organic vapor which initiates spontaneous and rapid growth of clusters. After activation, nanoparticles are expected to grow faster due to a decreasing Kelvin effect and, thus, an enhanced condensation flux. We have now stated that (L365-370) “These growth rates show a clear size-dependent particle growth (Fig.3), owing to the nano-Köhler activation that suggests a faster growth for activated nanoparticles due to a decreasing Kelvin effect and, thus, an enhanced condensation flux (Kulmala et al., 2004), Kelvin effect, and surface or volume-controlled reaction corrected for the Kelvin effect on surface or volume concentrations (Kuang et al., 2012)".

On the other hand, mass accommodation coefficient is the probability that a molecule that strikes the liquid/solid surface and then enters the bulk. The value of the mass accommodation coefficient depends on the system parameters and can either increase or decrease with a reduction in the nanoparticle size (Levdansky, 2012).

21. *L344: 'was intense' – consider rewording*

Reply: We have now stated that (L375-376) "indicating that high concentrations of condensable vapors existed".

22. *L346: replace 'will grow' with 'would grow'*

Reply: We replaced "will grow" with "would grow" (L377).

23. *L360,362: replace 'have been' with 'were'*

Reply: We replaced "have been" with "were" and "was", respectively (L391,393).

24. *L362: replace 'the data points on both figures are' with 'the number of data points on both figures is'*

Reply: We replaced "the data points on both figures are' with 'the number of data points on both figures was" (L393).

25. *L363-364: why proxy for ammonia? Was it not measured directly?*

Reply: The concentration of ammonia was measured directly. We have now stated that (L394-396) "Daily peak concentration of sulfuric acid proxy and daytime (6 am – 6 pm) averages of ammonia were used as approximations for their effective concentrations on a NPF day since there was no peak concentration for ammonia".

26. *L391: elaborate explicitly on the 'co-occurring sulfuric acid concentration'*

Reply: "Co-occurring sulfuric acid concentration" means when both sulfuric acid and ammonia concentration was high enough, new particle formation occurs with ternary nucleation. We have now used the term "co-existing" instead (L426).

Anonymous Referee #2

The paper by Xiao et al. reports field observations of new particle formation events in a polluted environment and is trying to discern potential formation mechanisms. The paper is presented fairly well but lacks clear focus, good articulation of goals and more in-depth analysis of data, especially when it comes to unexpected findings. The paper may become suitable for publication pending consideration of comments and appropriate revisions.

Major Comments:

1. *The goals of the study are presented poorly and it looks like a repetition of other similar studies in just a different area. The whole paper is presented like a report, seeking similarity with other studies. Why nucleation would be different at a Chinese location as is implied in line 14, P26658? At the moment I don't see one unique finding enhancing the understanding of new particle formation.*

Reply: A new paragraph has been added to clearly present the scientific goals of this field campaign, which reads (L93-106),

Direct measurements of atmospheric nucleation rates down to 1.5 ± 0.4 nm provide a better and more accurate characterization of atmospheric nucleation, since the indirect calculation of atmospheric nucleation rates from the formation rates of 3 nm particles leads to substantial uncertainty due to our incomplete understanding of condensational growth and coagulation scavenging of particles in the 1.5 to 3 nm range (Anttila et al., 2010; Korhonen et al., 2011). With the growing number of reports of real nucleation rates in clean atmosphere (e.g., Kulmala et al., 2012; Yu et al., 2014), it is ideal to measure nucleation rates in a polluted urban atmosphere to find out how atmospheric nucleation rates vary under different atmospheric conditions. In addition, the nucleation mechanism in a polluted urban atmosphere that is vital to understand atmospheric nucleation at a global scale and for atmospheric model development can be preliminarily investigated by examining the relationship between the measured atmospheric nucleation rates and the well-accepted precursor gases that exist in high concentrations.

Regarding the Nucleation mechanisms in China, we have now stated that (L88-91) "where concentrations of sulfuric acid and basic gases including ammonia and amines are high (Zheng et al., 2011; Zheng et al., 2015) but concentrations of extremely low volatility organic compounds formed from biogenic emissions are yet to be determined.".

2. *The relationship between $J_{1.34}$ nucleation rate and sulphuric acid proxy is poorly investigated and the conclusions poorly supported. The correlation between the two parameters is poor and above all not statistically significant. Note that R2 is not a correlation, but the variance. $R2=0.23$ suggests that only 23% of the total covariance can be explained by the relationship and owing the number of points (8) is not statistically significant even at $P<0.05$ level ($r=0.71$ at $P<0.05$ or $r=0.83$ at $P<0.01$ is required instead of obtained $r=0.48$). That is suggesting that either sulphuric acid plays insignificant role or the scarcity of data prevent statistical robustness. By contrast correlation with ammonia is much stronger and statistically significant, but it is played down to secondary role. What about considering organic species in forming new particles, like amines? The power of sulphuric acid proxy is also presented without error bars or standard deviation and considering the above is highly questionable. At the questionable. At the end there a big question mark whether sulphuric acid proxy was adequately derived.*

Reply: We agree that the correlation between the two parameters is not quite satisfactory. A better R^2 for ammonia and $J_{1.34}$ than that for sulfuric acid and $J_{1.34}$ does not necessarily mean that sulfuric acid plays a secondary role, since (1) sulfuric acid proxy was calculated with a 42% uncertainty whereas ammonia was directly measured with more confidence; (2) the daily peak concentration of sulfuric acid proxy was used whereas daily average of ammonia was used; and (3) the scarcity of data prevent statistical robustness. We have now stated that (L405-407) “Our P of 0.65±0.28 is of a significant uncertainty, which could come from the uncertainty during the calculation of sulfuric acid proxy and the scarcity of our data points”.

At the moment, we cannot rule out potential contributions from organic species including amines. A recent measurement of ammonia and amines at Nanjing, China by Zheng J. et al (2015) observed good correlations between NH_3 and amines, indicating similar emission sources. We have now stated that (L428-430) “A recent CIMS study (Zheng et al., 2015) observed good correlations between NH_3 and amines at an urban site of Nanjing, China. Hence, it is plausible that amines may contribute to nucleation in our site at Shanghai, too”.

According to Mikkonen et al. study (2011), the uncertainty for the calculation of sulfuric acid proxy was 42%. We have now added the error bars of the sulfuric acid proxy in Figure 4, and clearly stated in our experimental session (L231) that “The relative error between calculated sulfuric acid proxy and measured sulfuric acid concentration is estimated to be 42% (Mikkonen et al., 2011).” In addition, following Anonymous Referee #1’s advice, the peak concentration of sulfuric acid proxy instead of daytime average of sulfuric acid proxy is now used in Figure 4, which gives of a power exponent of 0.65±0.28.

3. *The relationship with $PM_{2.5}$ concentration is not appropriate because new particle formation is unrelated to particle mass, but rather its surface. $PM_{2.5}$ mass is a qualifier of air quality and only remotely linked to condensation sink. Why not to correlate to particle surface and also to examine any relationship between surface and mass?*

Reply: As suggested, the $PM_{2.5}$ mass plot has been replaced with the surface area plot in Figure 7. It is evident that NPF events in urban Shanghai occurred with low levels of aerosol surface area and that high sulfuric acid favored NPF events when aerosol surface area was low. The text in Section 3.4 has been revised accordingly.

4. *In a highly dynamic and polluted environment such as urban one should be very careful when it comes to averages. Strictly speaking average only applies to normally distributed values while generally aerosol parameters are log-normally distributed and, consequently, geometric average (median) and ranges should be used instead. Even more so that the authors presented parameters varying by more than one order of magnitude.*

Reply: In this manuscript, arithmetic average of the number concentration of 55~63nm particles and arithmetic average of size-dependent growth rates have been included.

The arithmetic averages of number concentration of 55~63nm particles are averages of those simultaneously measured by a long-SMPS and a nano-SMPS, respectively. The purpose of this averaging is to have a better match for the results between the two instruments. The difference between the geometric mean and the arithmetic mean of 55~63nm particles number concentrations was 0.83±4.55%. Hence, we conclude that there is no significant difference between the geometric mean and the arithmetic mean of 55~63nm particles number concentrations.

The arithmetic averages of size-dependent growth rates are not averages of the growth rates over a number of particle size bins, but averages of the growth rate for a certain particle size bin over a number of NPF event days. We don’t believe that growth rates for a certain size bin on different NPF event days were normally or log-normally distributed. Hence, the arithmetic averages have been used.

Minor comments:

5. *Please tidy up numbers throughout the paper: report particle sizes to three digits consistently;*

Reply: We have reported particle sizes to three digits consistently throughout the manuscript.

6. *remove technical information about size bins in the abstract;*

Reply: We have removed technical information about size bins in the abstract.

7. *growth rates present with one number in the subscript;*

Reply: Growth rates with one number in the subscript is not enough to elaborate explicitly on those of, for example, particles from 1.35nm to 1.59nm ($GR_{1.35\sim1.39}$). Hence, growth rates with three numbers is used in the subscript.

8. *Present error bar for sulphuric acid proxy power.*

Reply: According to Mikkonen et al. study (2011), the uncertainty for the calculation of sulfuric acid proxy is estimated to be 42%. We have now added the error bars of the sulfuric acid proxy in Figure 4.

9. *Revise abstract according to major comments.*

Reply: We have revised the abstract according to major comments.

10. *P26657, Line 17. "Very recently progress has been made by the use of PSM, etc: : :"*

Reply: We have now stated that (L59-61) "Very recently progress has been made by the use of Particle Size Magnifier (PSM) and Chemical Ionization Atmospheric Pressure interface Time-of-Flight (CI-APi-ToF) mass spectrometer".

11. *P26658, line 2. Populated region, not area.*

Reply: We have replaced "area" with "region" (L76).

12. *Line 13. "the use" not "employment".*

Reply: We have replaced "employment" with "use"(L85).

13. *Line 13. Measurement down to the certain size is not the measurement of the nucleation which has to be worked out from data.*

Reply: We have replaced "measure atmospheric nucleation" with "study atmospheric nucleation" (L86).

14. *Line 24. Competition between available surface and condensable vapour is a fundamental process and is very well known.*

Reply: This sentence has been removed in the revised manuscript.

15. *The sampling manifold was poorly designed. First, the flow in the main duct was highly turbulent. Second, various instruments were non-isokinetically subsampling as the air velocity in the duct was 9.1m/s versus subsampling velocity is 0.75m/s. That created a two-fold problem. Particles of different sizes were not sampled at the same efficiency leading to under-sampling of small particles and over-sampling of larger ones (it was like in a virtual impactor). While under-sampling was probably a minor problem, oversampling was rather detrimental leading to overestimated condensation sink. Was the diffusion loss taken into account when processing data?*

Reply: The diffusion loss of sampling lines for particles at each size was taken into account during data processing, which gives a "true" number of particles at each size. We have now stated that (L145-

147) “The calculated diffusion loss is up to 29% for 1.35 nm particles with the above setup, and has been taken into account in the entire size range during the data processing”.

16. *Please report time resolution of SO_2 measurements as it is important in drawing some of the key conclusions.*

Reply: The time resolution of SO_2 measurements was 5 min. Since the time resolution of temperature and relative humidity was 1 hour, the time resolution of sulfuric acid proxy is 1 hour.

We have now stated that (L175-177) “At the same site, sulfur dioxide (SO_2) was measured by an SO_2 analyzer with pulsed UV fluorescence technique (Model 43i, Thermo, USA) with a time resolution of 5 min”, and that (L234-235) “The time resolution of calculated sulfuric acid proxy was 1 hour since those of temperature and relative humidity was 1 hour”.

17. *P26661, line 8. $PM_{2.5}$ aerosol inlet, not cutter.*

Reply: This sentence has been removed in the revised manuscript because aerosol surface area instead of $PM_{2.5}$ is used.

18. *P26665, line 15. Reduction of particles, not loss.*

Reply: We have replaced the “loss” with “reduction” (L307).

19. *P26668. More thorough study is required about sulphuric acid proxy. Present and study not only average concentrations. But also 25/75 percentiles and perhaps min/max too.*

Reply: According to Mikkonen et al. study (2011), the uncertainty for the calculation of sulfuric acid proxy is estimated to be 42%. We have now added the error bars of the sulfuric acid proxy in Figure 4. In addition, following Anonymous Referee #1’s advice, the peak concentration of sulfuric acid proxy instead of daytime average of sulfuric acid proxy is now used in Figure 4, which gives of a power exponent of 0.65±0.28.

20. *Remove Figure 8 as it is conceptually wrong and the graph explains nothing.*

Reply: As suggested by Anonymous Referee #1 and #2, Figure 8 and corresponding texts have been removed in the revised manuscript.

Reference:

Anttila, T., Kerminen, V.-M., and Lehtinen, K. E. J.: Parameterizing the formation rate of new particles: The effect of nuclei self-coagulation, Journal of Aerosol Science, 41, 621-636, 10.1016/j.jaerosci.2010.04.008, 2010.

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Weber, R., Marti, J., McMurry, P., Eisele, F., Tanner, D., and Jefferson, A.: Measured atmospheric new particle formation rates: Implications for nucleation mechanisms, Chemical Engineering Communications, 151, 53-64, 1996.

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