Response to Referee #1

This manuscript "First comprehensive modeling study on observed new particle formation at the SORPES station in Nanjing, China" presents measurements and modeling of new particle formation (NPF) events with the intention to investigate the contribution of different chemical compounds and aerosol properties on the formation and growth to 6 nm aerosol particles. It is fairly well written and the modeling tools used in the study are of good quality. However, some of the details of the methods are missing. It is also unclear, what is the main outcome of this study and how the scientific community would benefit from it. This should be clarified by the authors. In addition, Abstract and Conclusions do not include any quantification of the results. For example, it is said that "simulated NPF events were generally in good agreement with the corresponding measurements" but it is not explained which parameters are in good agreement and what qualifies as "good agreement".

Response: We would like to thank the referee for providing the insightful suggestions, which indeed help us reconsider and further explore the underlying problems in these NPF events and their simulations. In the revised manuscript, we will add more descriptions on the method of measurement and modeling, as well as in-depth discussions concerning model performance.

Major comments:

• It is unclear how the nucleation coefficient k1 is determined for Equation (1). On Page 27506 it is said they were chosen "after comparing the simulations and DMPS measurements". This should be explained better. If the nucleation coefficient is tuned to match the model to measurements, wouldn't it be obvious that the model is in good agreement with the measurements? In addition to my previous point, the values of k1 are extremely low. For example, Pietikäinen et al. 2014, use k1 = 1.4×10 ^{15} and that is for the formation rate of 3 nm particles. For 1.5 nm particles it would be even higher. Could these extremely low values of k1 explain why the nucleation events start too late in the model?

Response: Here we have to state that it is important to have reviewers who read the manuscript very carefully and find mistakes, which should normally not happen. After checking the numbers in the model again we realized that the units of k1 used here are m^3/# and not as mentioned in other publications like e.g. Boy et al., 2008 (Atmos. Chem. Phys., 8, 1577–1590, 2008) cm^3/#. Taking this into account we end up with values of $6x10^{-13}$ cm^3/# for 10 of July and 22 of August and $2.2x10^{-10}$ cm^3/# for 22 of June. How could this happen? UHMA the aerosol dynamic code used in MALTE-BOX was developed more than 10 years ago at the University of Helsinki and was written in SI units, so it uses for gases and particles #/m^3 and not #/cm^3 as we do in the other MALTE-

BOX code. Normally we change all the units when they are transferred to UHMA but the nucleation coefficients are set direct in UHMA and this caused the problem. So in practice the values were not wrong but compared to earlier values with different units.

The new or related to the units corrected values are now in full agreement with values published in Boy et al., 2008 for Hyytiälä with $5.7x10^{-13}$ cm^3/# to $5.5x10^{-14}$ cm^3/# or Heidelberg, Germany (more polluted) with $2.3x10^{-11}$ cm^3/#. Obviously, the most polluted day the 22 of June requires a higher (about 400 times higher compared to the other two days) kinetic nucleation coefficient, which could be related to more anthropogenic compounds in the nucleation mechanism similar to the comparison of Hyytiälä and Heidelberg.

Concerning the first statement of the referee how we achieved the values of the kinetic nucleation coefficient this was done similar as in other manuscripts like Boy et al., 2008 by running the model with different values until the best fit between measured and modeled number concentrations in the lowest available bins were achieved. We agree with the referee that this could be seen as tuned but it also provides us input how important sulfuric acid or other unidentified molecules are when comparing the required kinetic nucleation coefficient. And by comparing the values of these values with different stations it gives us some idea how important sulfuric acid is in the nucleation compared to other unidentified compounds

We will add all this information in the final version of the manuscript and of course correct the k1 values with units.

• *Since only three NPF events are investigated, it is unclear how well the model configuration reproduces observed NPF events in general. For example, has it been tested if the modeled frequency of NPF events is similar to observations?*

Response: 2-year DMPS measurements at the SORPES site have detected over 200 NPF events (Qi et al. 2015). It is time consuming and computationally demanding to simulate all these events using this comprehensive modeling method. Thus, the modeling of NPF frequency has not been tested yet. It will be addressed in our future work, but cannot be accommodated in this study. In this paper, we mainly focus on analysis and numerical simulation of the several typical NPF events, which were identified by the distinctions in the potential source regions of air masses.

• *One of the main results of the study is that biogenic organic compounds play an essential role in condensational growth of newly formed clusters. Model results supporting this finding are missing. Have you, for example, investigated the modeled fraction of these biogenic organics in 6 nm particles? If the fraction is significantly high in these particles, it would strengthen your case.*

Response: Accepted. To support the importance of biogenic organic compounds in condensational growth of newly formed clusters, we will add a new figure to show

contributions from different kinds of condensing vapors to initial particle growth (particles with diameter less than 10 nm) in the revised manuscript.

• *Page 27516: It is said that the better correlation of using organic nucleation scheme is evidence for the involvement of ELVOC in NPF. Visual inspection of this data suggest that this increase in the correlation comes only from the fact that [H2SO4]2 has smaller variability than* $[H2SO4]^{1.0}[ELVOC]^{0.8}$*. Based on this approach activation type nucleation mechanism might have even higher correlation coefficient. In addition, the correlation coefficient seems to be calculated from the actual values while the x-data varies over 8 orders of magnitude, so it would have been more appropriate to calculate the correlation coefficients for the logarithm of the values. Please see the Referee #2 comments to improve this analysis.*

Response: Dashed lines showing $J=2.2\times10^{-10}\times[H_2SO_4]^{2.0}$ and $J=6.0\times10^{-13}\times[H_2SO_4]^{2.0}$ will be added for reference in Fig.8 (b). The reason why we included this figure is to provide some hints if ELVOCs are crucial in the formation and growth of particles up to 6 nm. There was no aim from us to state that ELVOCs are important in the formation of clusters which is still open (see discussions on Referee #2's comments).

Minor comments:

• *Please check the grammar.*

Response: The grammar will be thoroughly checked.

• *How do you determine the OH radical concentration?*

Response: In the MALTE-BOX model, the chemical mechanism scheme is produced by selecting chemical reactions primarily from the Master Chemical Mechanism (MCM). The kinetic preprocessor (KPP) was applied to numerically solve for the concentrations of each compound, including OH radical. To make it clearer, we will add more relevant descriptions and references in Section 2.2.1.

• *Photochemistry can also play a significant role in NPF and the model's cloud cover can affect that significantly. How well does WRF-Chem reproduce the cloud cover during these event days?*

Response: As mentioned, cloud cover exerts significant impacts on photochemistry and also NPF processes in the atmosphere. Comparison between WRF-Chem reproduced cloud fraction and corresponding satellite detections from OMI instrument on AQUA is shown in the following figure (Fig. R1). WRF-Chem model displayed similar spatial patterns to OMI observations on the NPF days. On 22 June, dense cloud covered Shandong and Zhejiang province. During the second case, little cloud was

found in areas south of the Yangtze River Delta according to both observation and simulation. While on 22 August, most parts of South China were covered by thick clouds.

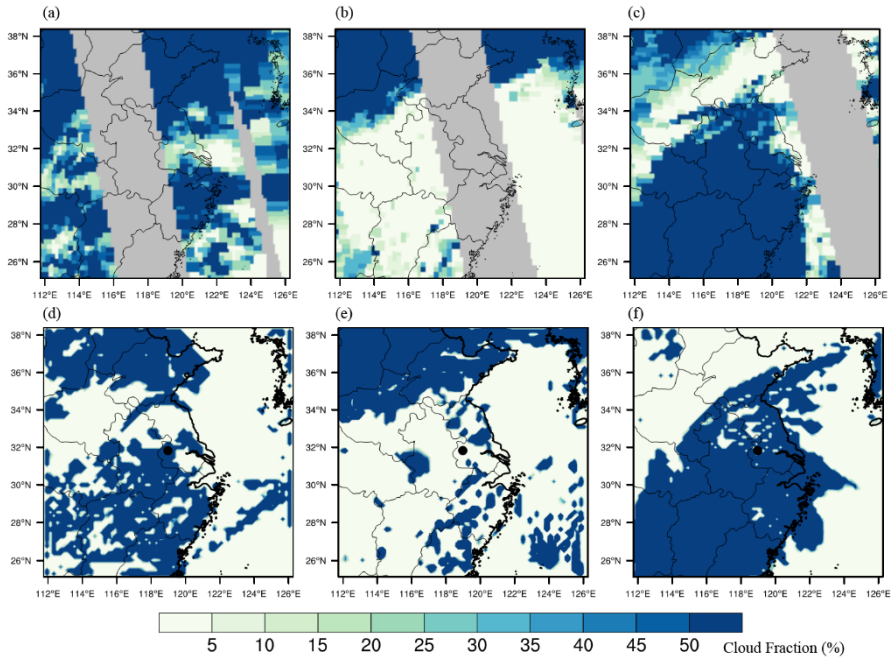

Fig. R1 Comparison of satellite-retrieved and model predicted (d-f) cloud fraction during these three NPF days. (a-c) are OMI detected cloud fraction for 22 June, 10 July and 22 August, respectively. Corresponding simulations are displayed in (d-f).

• *The formation rate of 6 nm particles is not nucleation rate. A preferred term would be "new particle formation rate". Please correct this on Page 27515, Line 24, 25, and in the caption for Fig 7*

Response: Corrected. The term "nucleation rate" for 6 nm particle will be replaced with "new particle formation rate" throughout this manuscript.

• *Page 27506, Line 10: What do you mean by "good"?*

Response: Thanks. Here the words "good cluster formation rate" was unclear for readers. We will rewrite this sentence as "Kinetic nucleation theory has been shown to have good performance in simulating cluster formation in various environments…" in the revised manuscript.

• *Page 27508, Line 20: Have you checked if this distribution is equal also in observations?*

Response: The distribution was not compared with observations. During the summer of 2014, GC-MS-FID (Gas Chromatography-Mass Spectrometry- Flame Ionization Detector) was used to measure concentrations of VOCs at the SORPES station. Technically, all the kinds of biogenic VOCs that listed in Table 1 can be measured. However, during the calibration, retention times were determined for a limited number of compounds. Among species in Table 1, only isoprene was included to generate the calibration curve. Thus, it is infeasible to get the profile of biogenic VOC distribution and compare it with model results in this work. We plan to include more VOC species, particularly biogenic ones, in future VOC measurements.

• *Page 27513, Line 18: This wording "succeeds, on average, to generally reproduce" is very ambiguous. Please rephrase this.*

Response: This sentence will be rephrased in the revised manuscript.

Page 27507, Line 25: What does "for further analysis and box modeling" mean in this context?

Response: In this context, by "further analysis and box modeling", we mean that only the last 48-h simulation results of WRF-Chem were used to provide input to the box model and also data for some figures shown by us. It might be not so clear to use the word "further" and we will change it to "following" in the revision.

• *Page 27510, Line 18: 500 000 #cm3 cannot be the correct value as it is extremely high.*

Response: It will be corrected to 10,000 # cm^{-3} in the revised manuscript.

• *Page 27512, Lines 17-18: It is said, that "On one hand, humid air mass transported from the ocean might have favored the particle growth". Please explain the reasoning behind this sentence.*

Response: Accepted. We will add more explanations in Section 3.1.

• *Page 27514, Line 17: The word "concentration" is missing?*

Response: The word "concentration" will be added.

• *Figure 5: Having different y-scales in the right column plots make the comparison of the events difficult.*

Response: The y-scales of the right subplots in Fig. 5 will be unified in the revised figure.

Reference

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