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Atmospheric Chemistry and Physics Discussions

Interactive comment on "First comprehensive modelling study on observed new particle formation at the SORPES station in Nanjing, China" *by* X. Huang et al.

Anonymous Referee #2

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New particle formation (NPF) in the atmosphere is a hot topic in recent years. Although there are some studies conducted in China, no studies are reported in Nanjing, the capital of Jiangsu province in Yangtze River Delta region. In particular, this is the first study in China to understand the NPF mechanism using a number of models. In detail, this study utilized a comprehensive modeling system, combining WRF-Chem and MALTE-BOX model to investigate the complex processes of the NPF and subsequent growth in the Yangtze River Delta region. Three typical NPF events, which were probably influenced by distinguished emission sources, were selected for mechanism study. Two kinetic-type nucleation mechanisms including homogenous homomolecular

sulfuric acid vapours and heteromolecular nucleation involving organic vapours were tested. The simulated NPF events were generally in good agreement with the field measurements, providing a possibility to better understand the NPF processes in this region. The paper is well organized and well written. It is worth to be published and will definitely add values to the literature. Nevertheless, more in-depth model simulations and discussion can contribute more to the literature. Hence, this paper can be further strengthened by more comprehensive discussion of the simulated results. The specific comments are shown as follows.

Major comments:

In "Introduction" section, the information about the MALTE -BOX modeling studies on the aerosol formation in recent years should be provided.

In "Data and methodology" section 2.1, has the diffusion loss been considered when sampling the ultrafine particles by DMPS, have you corrected the number concentration of nucleation mode particles (<10 nm) measured by DMPS due to the large diffusion loss of nanoparticles?

In section 2.2.1, the calculation method or equation of sulphuric acid vapor concentration should be provided.

Twenty ELVOCs and seven SVOCs were selected as condensable vapors. Why did you choose these compounds not others? What are the precursors of these condensable organic vapors? More detailed information should be provided.

In section 3.1 lines17-18, the number concentration of 500 000 # cm-3 is extremely higher than other studies. Double check if the number concentration of particles here is in unit particles/cm3 not in the unit dN/dLogDp.

Page 27511 lines 8-9, the contribution of sulphuric acid vapour to the particle growth can be calculated, and also this sentence is not very clear, please rewritten. In section 3.2.1 line 17 what are the major species of alkenes and aromatic compounds simulated

by WRF-Chem? (e.g. were only biogenic terpenes included in the alkenes group?)

Lines 17-19, based on the normalized mean bias, the simulated results of alkenes, aromatic and isoprene concentration should be evaluated separately, such as, which one showed better agreement and whyïij§

Lines 21-26, the simulated varied spatial patterns of biogenic terpenes during the three NPF days could not be explained by the dominant wind directions. Since biogenic emissions are closely related to the weather temperature, as shown in Fig.3, the higher temperature during the second NPF case was observed than that in the other two cases. That would be one important reason causing high biogenic terpenes emission simulated by WRF-chem.

In section 3.2.2, this part should be further improved with more model test and in-depth discussion.

Two kinetic nucleation mechanisms were introduced in this section. However, only homogenous nucleation mechanism of sulfuric acid vapours was tested by the box model. The nucleation mechanism involving organic vapours was only investigated by the relationship of [H2SO4]1.0 [NucOrg]0.8 and J6, and not applied into the model to evaluate whether it improved the simulation results or not.

As shown in Fig.5, in Cases 2 and 3, simulated nucleation mode particle number concentrations were higher than observed values, while in case 1 it was opposite. Also, the simulated results were the highest in case 3, followed by cases 2 and 1. These findings need more explanations and discussions in order to explore the major factors influencing the results during the each event. For example, in case 3, the RH was very high, while the wet deposition was not included in the MALTE model; hence the weather condition may partly influence the modeled results causing a higher simulated result than the observed one.

As described in the paper, the highest condensation sink (CS) and biogenic VOCs

concentration were observed or simulated during the first and second events, while the lowest condensation sink and higher sulphuric acid production were found during the third event. These three events provide a good opportunity to investigate the relative role and sensitivity of CS, BVOCs and sulphuric acid vapour concentrations in the new particle formation and to the growth, respectively.

For Case 1, the event occurred even under the high level of condensation sink. How high CS would finally inhibit the event on this day? And also how low sulphuric acid vapour concentration could still trigger the nucleation under such high CS? Their contributions and sensitivity tests on new particle formation rate and growth rate can be conducted by increasing the CS value or decreasing the calculated sulphuric acid vapour concentration gradually for box model runs.

Similarly, for Case 3 (actually, it is not a suitable day identified as a NPF event occurred under the low level of condensation sink since it was a rainy or cloudy day, and the RH was very high on this day which enhanced the condensation sink), it would be better to select a sunny NPF day when condensation sink was very low. If not, at least you could investigate the lowest level of sulphuric acid vapour which would induce the initial nucleation during this event.

For Case 2, organic vapours were showing more important role in the particle formation than the other two cases, and the authors also tried to use a heteromolecular nucleation theory involving organic vapours to better explain the observed particle formation rate. However, the focus was lost by combination of all data during the three events as shown in Fig. 7. Based on the distinguished characteristics of these three events, sulfuric acid vapours and organic vapours may play different roles in the formation process, and only using one relationship ([H2SO4]1.0 [NucOrg]0.8) of sulfuric acid and organic vapours for the simulation of three events is not scientific. According to the observation, sulfuric acid vapours seem to involve more significantly into nucleation process during the Case 1 and Case 3 than during Case 2. Here, the importance of sulfuric acid and organic vapours in each event should be investigated individually.

As mentioned above, in Fig.7 (b) and (c) the relationship of sulfuric acid and organic vapours with particle formation rate should be separately investigated during each event. In Fig.7 (b), two lines should be drawn based on the equations provided in the manuscript, i.e., J =6.0×10-19×[H2SO4]2.0 for 10 July (Case 2) and 22 August (Case 3), and J =2.2×10-16×[H2SO4]2.0 for 22 June (Case 1). In Fig.7 (c), a line based on the equation J1.5 = 7.2±1.4×10-13×[H2SO4]1.0 [NucOrg]0.8 should be drawn. It is very noteworthy that J6 in Fig. 7(c) should be J1.5 which is significantly larger than J6, and can be calculated based on J6. Then the correlation coefficients of sulfuric acid and organic vapours with particle formation rate during each event in Fig.7 (b) and (c) can be calculated, respectively, by linear regression analysis. According to these correlation coefficients, you can find out which line fit well with which event and explore the potential formation mechanisms during each NPF event. Page 27508, section "data analysis". What is the difference between coagulation loss and condensation sink. CS was not included in equation (2). Page 27511, section 3.1 "Observations and data analysis". "Along with the active photochemistry and high concentration of O3, rapid oxidation of SO2 and accumulation of gaseous sulphuric acid are expected". The authors should briefly introduce the mechanism of SO2 oxidation by O3 or OH. Page 27514, the explanation of third NPF case (22 August) was not convincing enough. The concentrations of SO2, sulphuric acid, SVOCs and ELVOCs were all pretty lower than those in the other two cases. However, the concentration of OH was remarkably higher. The explanation of "little condensational loss" was a factor causing the third NPF. However, it maybe not enough, the authors need dig depth for better explanations. Page 27515-27516. Can the equation of nucleation rate of 1.5 nm cluster (equation (5)) be directly applied to J6? Page 27515, lines 14-16, as the production of ELVOCs and SVOCs was mainly initialized by the reactions between monoterpene and ozone, the contribution of monoterprene oxidation to the production of ELVOCs and SVOCs should be evaluated by models and provided here. Page 27516 lines 15-21, for the particle growth, the contributions of the OH and O3 oxidation mechanism on the volume concentration of small particles can be further investigated to find out

the dominant precursors and their oxidations involving in the particle growth in these studied events.

P27517 lines 2-5, the sentences "According to the simulation, ... at the experimental site." are difficult to be understood.

Minor comments: It may be inappropriate to use "first" in the title. In Fig. 3, draw diurnal variation of CS. In Fig. 3, check if the diurnal variation of particle size distribution during the first event is in the same value scale range as other two events. In Fig. 4 (a), (b) and (c), the meanings of the dashed lines should be provided. In Fig.5, check if the unit of particle number concentration is not "dN/dLogDp" in right panel.

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