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Atmospheric Chemistry and Physics Discussions

Interactive comment on "New particle formation in marine atmosphere during seven cruise campaigns" *by* Yujiao Zhu et al.

Anonymous Referee #2

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Overall comment: This study includes the observations of nanoparticles in six cruises over the marginal seas of China and one cruise to the Northwest Pacific Ocean. The particle number concentration, size distribution, formation rate and growth rate of new particles are discussed. The authors also try to illustrate the roles of anthropogenic and marine biogenic emissions in new particle formation, through analyses on several specific NPF events. The experiments are interesting, and should be beneficial to advance the knowledge on the impacts of human being activities on NPF and global climate change. However, the experimental design has obvious drawbacks in considering the adequate data to support the analyses in this meaningful research. Nearly no data of the precursors of condensable vapors are available. Though some chemicals, such as the amines and the oxalic acid, in the size-segregated are analyzed, the

sampling period even missed the NPF periods, which led to the inappropriateness of using these data to infer the processes and chemical species dominating NPF. I also have serious concern on the explanations to the different relationships between the formation rate and the net maximum increase in the nucleation mode particle number concentration. Similarly, the conclusion that the NPFs, regardless of which categories, are regional phenomenon cannot convince me, since no solid evidence has been provided. In view of the inadequate discussions, misleading inferences and even wrong interpretations, the paper needs to be revised substantially before being considered to be accepted. Specific comments are also given for the authors' reference. Specific comments: 1. Page 3, "In November 2012, the NO2 column densities were higher in the eastern mainland of China due to the house-heating". House heating is not the sole cause of elevated NO2 in autumn. 2. Page 6, lines 6-10. How do you confirm that these NPF events were the regional NPF events, rather than the local ones that occasionally occurred on the same days? Is there any evidence proving that the air masses were homogeneous on these days, except for the backward trajectories? Since the ship location and the coastal sites were generally in an area influenced by the same monsoon, they always received air masses from the same directions. However, it does not mean that the regional air overrode the properties of local air masses. 3. Page 6, lines 11-13. From the particle number distributions shown in Fig. A1h, i, l, m, I can hardly believe that these are the regional NPF events. Besides, could the delay be caused by the different weather conditions, or downward transport of nanoparticles in the afternoon? 4. Section 3.2. The observational particle number distributions at OUC were not well presented. 5. Page 6, lines 20-23. How did you remove the influence of ship-self emissions? This needs to be demonstrated in methodology. 6. Page 7, lines 13-14. "The increase likely induced by the long-range transport of air pollutants from the continents, inferred from the doubled number concentrations of accumulation mode particles in Category 2 relative to Category 1." This is contradictory to the previous statement that "the concentration increase was limited to particles with the diameter less than 20 nm". 7. Page 7, lines 19-22. The authors should illustrate in more

details the size ceilings that the particles could grow up to. What caused the different ceilings, and what were the implications from the differences in particle size distributions? 8. Page 8, lines 10-15. Condensation sink is an important factor influencing particle formation. Throughout the manuscript, CS has never been presented and has seldom referenced for discussions. The lack of measurements of condensable vapors makes so many inferences in the paper not reliable, not to say some inferences are contradictory to common sense. For example, here I cannot believe that the loadings of precursors favorable for the formation of new particles were higher over the marginal seas than in the coastal area. Evidences need to be provided to support the inferences. 9. Page 8, lines 17-32. I cannot understand why the higher formation rate did not result in larger increase of nucleation mode particles, note that the formation rate is closely related to the increase of nucleation mode particles if looking at the calculation formula of formation rate. All the explanations are based on the assumptions, which cannot convince me. The authors should provide more evidences to validate their assumptions. The authors state that "the NMINP was always determined by the consumed H2SO4 vapor for nucleation". Sorry for that I cannot accept this view. How about the number of nucleation mode particles when the organic vapors facilitated the nucleation and particle growth to the detectable size? The so called threshold of formation rate, i.e. 8 cm-3s-1, was exactly the same as that reported in the study previously published by the same authors. This cannot convince the readers unless the similar phenomenon has been reported by other groups. I tried to understand the authors' view by finding the clues from the paper "Simultaneous measurements of new particle formation at 1 s time resolution at a street site and a rooftop site". However, it is hard for me to follow up the authors in many points. For example, in this paper, the sentence "Supposing that sulfuric acid vapors are completely nucleated, followed by the nucleated particles growing to the detectable size, the yields of newly formed particles are determined mainly by the supply of sulfuric acid vapor and are less affected by the formation rate" is problematic. How could you separate the role of sulfuric acid from the formation rate, as sulfuric acid plays critical role in nucleation? In the sentence "Scenario 1: H2SO4

vapor is relatively sufficient against NucOrg, and J8 is therefore determined mainly by the availability of NucOrg vapor. A good correlation is theoretically expected for J8 and NMINP". To be honest, I do not understand the logics behind. 10. Page 9, lines 29-33. The concurrent occurrences of class II NPF events at the coastal site and over the marginal seas could not be an evidence of the regional characteristics. The particle number distributions at the two sites were quite different on the days specified by the authors (Figure A1). Besides, it is difficult to convince me with the backward trajectories. The two sites were in a same region under the influence of the same monsoon. Even so, the air masses could be totally different in chemical compositions when they passed over the different cities. With no chemical information or mesoscale simulation, it is hard to say the two sites were interacted and the regional NPF events occurred at the two sites. 11. Page 10, lines 1-6. Condensable vapors are of course critical in NPF. However, it is not reasonable to simply attribute the different characteristics of NPF to the abundances of the condensable vapors. Other factors, such as the preexisting particles and the meteorological conditions also influence the NPF. In this case, more preexisting particles with larger diameters existed at the marginal sea site. Could this also account for the insignificant particle growth? 12. Section 4.1. I do not agree that new particle formation occurred in this case, i.e. 30 August 2015. 13. Page 11, lines 12-15. Figure 7c does not show the altitude variation of the backward trajectories. 14. Page 11, lines 16-26. The sampling periods of MOUDI samples were after the NPF events, not including the hours when the new particles were formed and grew up. I would doubt the reasonability of using these data to infer the chemical species dominating NPF. Same for the other similar discussions. 15. Page 12, lines 8-10. I do not understand the logics behind this inference, though it is true that the AR increased after Dpg was higher than 50 nm. Why not present the number concentration of >50 nm particles or its fraction in total particles against the NCCN? It would be a more direct way to link the particles larger than 50 nm to CCN. 16. Caption of Figure 3, what does "exteriors" mean? Why should they be excluded from the regression? Figure 4, what does the black dots represent, same for the other figures? Figure 9, what does the

highlighted area denote for? 17. The manuscript needs to be grammatically checked by an editing company or a native English speaker professor.

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