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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.

Yujiao Zhu et al.

xhyao@ouc.edu.cn

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Anonymous Referee #2

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

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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.

Response: The authors thank the reviewer's comments. We agree that we have no data of the precursors of condensable vapors. The weakness will be added in the revision. The weakness is quietly common in NPF studies in the literature. We also add the results of condensation sinks to support our analysis in revision. A few more comments are also very constructive for us to improve the quality of this manuscript because the related parts in the origin version are indeed misleading or even wrong. We make a substantial revision accordingly and explain why.

For parts of reviewer's comments, we believe that more clarifications are needed to make the analysis more readable. We revise these parts accordingly and explain why. Moreover, the authors may disagree with a few reviewer's comments. We explain why in this response below.

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.

Response: Agree. In addition to the house heating, poor dispersion conditions (e.g. temperature inversion) and other factors may also lead to the elevated column densities of NO2 in November. In revision, it will be revised as “In November 2012, the elevated NO2 column densities in the eastern mainland of China were likely due to combined factors such as intensive house heating, poor dispersion conditions, etc.”

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.

Response: We are sorry that we cannot agree with the comment. The authors believe that the reviewer may mix up a few concepts.

In the recent highly cited article entitled as “Measurement of the nucleation of atmospheric aerosol particles”, regional NPF events refer to these events occurring in a spatial extent varies from tens to thousands of kilometers (Kulmala et al., 2012, Nat. Protoc.). The reviewer may mix up concepts such as “regional NPF events”, “simultaneous NPF events” and “regional-identical NPF events” (Hussein et al. et al., 2009, Atmos. Chem. Phys.). The authors believe that the reviewer was arguing against “regional-identical NPF events” rather than “regional NPF events”. The regional-identical NPF events are a subset of simultaneous NPF events. The same can be said for simultaneous NPF events against regional NPF events. The authors also have a big concern for occurring regional-identical NPF events in the marine atmosphere over the marginal seas of China and in the continental atmosphere over the eastern part of China. NPF events usually occur in either less polluted or clean atmospheres. Under such condi-

tion, it is hard for the authors to believe that the regional air mass always overwhelms the local air mass in the atmosphere over a large spatial area in the eastern part of China and downwind seas.

The authors agree that measurements at two or even more fixed sites are really difficult to justify NPF events as regional. In fact, mobile measurements over a large spatial scale are well suitable to examine regional NPF events. We will clarify that the on-board observations were made mostly on traveling instead of anchoring at the fixed locations. According to the definition above-mentioned, the NPF events observed over the marginal seas have no doubt to be confirmed as regional events except NPF event on 15 May 2014. The solid evidences include 1) the duration of the NPF events exceeded 3 hours in 22 days out of the total 23 days over the marginal seas of China, 2) on-board observations were made mostly during traveling instead of when anchored at fixed locations. The ship travelled at a speed of 18 km/h. A rough calculation of the spatial span is 18 km/h$\times$NPF time in hours for the NPF events over the sea. The NPF event on 15 May 2014 appeared to last for about one hour due to the ship emissions overwhelming the new particles signal after 09:30, this has been clarified in the revision.

Moreover, simultaneous observations of NPF events at the coastal site on the same day further zoom regional NPF events into simultaneous NPF events, i.e., NPF events occurring on a line over dozen of kilometers in a marginal sea plus at an additional coastal site. The simultaneous NPF events are a subset of regional NPF events and the types of NPF events had been claimed based on several sets of measurements over a large spatial range in literatures (Hussein et al., 2009, Atmos. Chem. Phys., Jeong et al., 2010, Atmos. Chem. Phys., Wang et al., 2013, Atmos. Chem. Phys., Shen et al., 2018, Atmos. Chem. Phys.).

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?

Response: On 17 Nov. 2012, the NPF event lasted for 4 hours over the marginal seas and 3 hours at the OUC site (due to the instrument maintenance after 15:00). Even longer duration for NPF events occurred on 7 Nov. 2013. Referred to our response to Comment 2, the two NPF events should be considered as regional events.

We agree that different weather conditions and downward transport of nanoparticles could be ones of causes for the delay of the NPF observed in the coastal atmosphere. Weather conditions can affect concentrations of precursor vapors and affect the occurrence of NPF. In the revision, the sentence will be revised as "Many other factors, such as weather conditions which can affect the concentrations of precursor vapors and gas-aerosol partitioning, downward transport of nanoparticles, etc., might also contribute to the delay."

4. Section 3.2. The observational particle number distributions at OUC were not well presented.

Response: This study focuses on NPF events in marine atmospheres. The authors prefer to revise the title of Section 3.2 as: "Particle number concentrations and size distributions in presence of NPF events against the background in marine atmospheres". As a comparison, we agree that a short summary of particle number size distribution in the coastal atmosphere should be included. For Category 1, the authors will add a short summary: "In the Category 1 data observed at the OUC site, the size distribution of the average particle number concentration was similar to that in the atmosphere over the marginal seas of China. For example, the Aitken mode and accumulation mode particles accounted for approximately 80% of the total particle number concentration. However, the average particle number concentration of $1.4\pm0.8\times104$ particles cm-3 observed at the OUC site increased by one-fold compared to that over the marginal seas of China and by approximately four-fold compared to that over the NWPO."

For Category 2, more information of particle number size distributions at OUC will be

added: “Over the NWPO, the increase in concentration of newly formed particles was limited to particles with diameter lower than 30 nm, possibly because of the growth pathways of newly formed particles being different from those at the OUC site and over the marginal seas, where newly formed particles can grow to diameters up to 60 nm.” “For example, Fig. A1a, b showed ceilings of approximately 50 nm, and Fig. A1c, d showed ceilings of approximately 20 nm during the events over the marginal seas and at the OUC site.”

5. Page 6, lines 20-23. How did you remove the influence of ship-self emissions? This needs to be demonstrated in methodology.

Response: The ship-emitted particles can be clearly identified in the high-time resolution measurements. First, ship-emitted particles exhibit a uni-modal size distribution at 10-60 nm with a peak at 20-30 nm. There is only a small variation in the particle number size distribution, depending on weather conditions. Second, the number concentration of the ship-emitted particles is an order of magnitude higher than that of the background particles as well as new particles. Third, there are dozens to hundreds of spikes in the particle number concentration when the ship-emitted particle signal is detected. For example, Fig. 1 at the end of this response showed the ship plumes (from 12:32 to 13:01, from 14:57 to 15:37 and from 17:33 to 18:10) with the high particle number concentration ($7.3\times104\pm2.5\times104$ cm-3). When we do the calculation for FR, GR and NMINP, the three features above-mentioned were used to remove the ship emission periods. We will add the part in the revision.

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".

Response: What we exactly want to say are different from those shown in the original

version. Thank for the comment help us realize this. In the revision, we rewrite the part. It is "Compared to Category 1, NPF events greatly enhance the total particle number concentrations (Fig. 2, solid lines) in Category 2 over three regions including the NWPO, the marginal seas of China and OUC, mostly because of a large increase in the number concentration of newly formed particles. Over the NWPO, the concentration increase of newly formed particles was limited to particles with diameters lower than 30 nm, possibly because of the growth pathways of newly formed particles being different from those at the OUC site and over the marginal seas, where newly formed particles can grow to diameters up to 60 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?

Response: Agree. The part will be revised as "The results were caused by varying size ceilings in the growth of newly formed particles, i.e., the growth of newly formed particles apparently stopped when they grew to the maximum sizes during these events. For example, Fig. A1a, b show ceilings of approximately 50 nm, and Fig. A1c, d show ceilings of approximately 20 nm during the events over the marginal seas and at the OUC site. In fact, a size ceiling is a common phenomenon during NPF events occurring in various urban or coastal atmospheres, as highlighted by Zhu et al. (2014, 2017) and Man et al. (2015). They also proposed that the size ceiling is associated with the thermodynamic partitioning of semi-volatile species in growing newly formed particles."

We also agree with this reviewer, i.e., it is important to ask "What caused the different ceilings, and what were the implications from the differences in particle size distributions". Theoretically, which semi-volatile species dominate the growth of newly formed particles and what their vapor concentrations are in the atmosphere during NPF events are critical to fully answer the question. In absence of the two results, we cannot speculate more from the differences in particle size distributions. Honestly, we have no breakthrough progress on determining these semi-volatile species in the last decade.

However, we believe that the ceiling phenomenon would stimulate more future studies in research community.

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.

Response: The authors agree that it is worthy of the inclusion of condensation sink. Condensation sink prior to or during NPF event plays an important role in removing condensation vapors, although it may or may not dominantly determine concentrations of condensation vapors. In this study, the CS over the marginal seas were 1.1±1.0 (10-2 s-1), and much lower than that at OUC site of 4.1±2.0 (10-2 s-1) during simultaneous NPF events. However, no significant negative correlation between the FR/GR and CS was observed (Fig. 2 at the end of this response). The results will be added in the revision and Supporting Information (Fig. S4 in revision).

In addition, it is hard to say that the cleaner atmosphere should have fewer loadings of precursors, e.g., the FR is lower than that in the clean atmosphere than in polluted atmospheres. In our previous studies to compare NPF events in the atmospheres at different pollution levels (Qingdao, Hong Kong, Toronto), we did not find a clear relationship between the degree of air pollution and FR (Zhu et al., 2014, Man et al., 2015). So does in a number of investigations summarized by Kulmala et al., 2004. Kulmala et al. (2005) claimed that the larger CS in the polluted atmospheres can be compensated by a larger vapor source rate, which can up to four orders of magnitude larger than in the clean atmospheres.

Larger FR should be an important evidence for higher concentrations of condensation

vapors based on the nucleation theory. We agree that the measurement of condensation vapors are more direct evidences than FR. The weakness of lack of measurements of condensation vapors will be added in the revision.

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.

Response: In the revision, we add more clarification to better defense our arguments. We also try our best to explain the difference between our analysis and the reviewer's

thoughts.

We assume that NPF rapidly stops after dozens of minutes bursting. This is consistent with huge measurements of banana-shaped NPF events reported in literature (Kulmala et al., 2004). If NPF continuously occurs during the whole event, a fan-shaped NPF event would be detected instead of banana-shaped NPF event. This is because of continuous formation and growth of new particles. In fact, a fan-shaped NPF event was hardly observed. Moreover, in Fig. 1 published by Yue et al, 2010 (Atmos. Environ.) and Fig. 3-4 published by Wang et al., 2011 (Atmos. Chem. Phys.), NPF rapidly stops after dozens of minutes bursting with rapid consumption of H2SO4 vapor. These studies also directly supports our assumption. We will add the part of analysis in the revision.

The reviewer commented "note that the formation rate is closely related to the increase of nucleation mode particles if looking at the calculation formula of formation rate." The comment does not sound scientific. This is no doubt that FR is determined mainly by the nucleation mechanism (i.e., nucleation of sulfuric acid vapor enhanced by organics). The equation is used to measure the apparent formation rate and has nothing to do with nucleation mechanisms. Technically, we can measure the vehicle speed on basis of vehicle traveling mileage in a fixed time. We clearly know that a vehicle speed depend mainly on engines and fuels, etc., but has nothing to do with vehicle traveling mileage. Moreover, the largest mileage of a vehicle is mainly determined by the used liters of fuel in vehicle tank. Engines and other factors can greatly affect vehicle speed, but the influence on the largest traveling mileage is not comparable to that of the liters of fuel in vehicle tank. The same can be said for FR and NMINP. In NPF events, the authors technically consider sulfuric acid vapor as fuel while organic as engine and other factors affecting vehicle speeds.

The reviewer commented "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 authors may disagree with the comments. Regarded much low nucleation rates of inorganic vapors reported so far, the authors strongly believe that all NPF events observed in the atmospheric boundary layer on the earth were facilitated by organic vapors to some extent. When the organic vapors don't facilitate the nucleation and particle growth to the detectable size, there are no NPF events to be observed in the atmospheric boundary layer and the NMIMP is zero.

We also agree that the threshold of formation rate, i.e. 8 cm-3s-1, may be coincidentally consistent with our previous study. This needs more work to be confirmed. The part will be added in the revision.

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.

Response: Referred to our response to Comment 2, the reviewer may mix up a few concepts. The class II NPF events lasted for 3-5 hours and these NPF events should be considered as regional events.

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

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for the insignificant particle growth?

Response: We agree that weather conditions can affect the growth of newly formed particles by changing gas-aerosol partitioning. We will revise the part accordingly: "Theoretically, higher CS can remove more condensable vapors and consequently reduce the vapor pressure of precursors. In this case, the apparent particle growth was undetectable in the marine atmosphere with the smaller CS of 0.6±0.1 (10-2 s-1) against the value of 5.3±0.2 (10-2 s-1) in the coastal atmosphere. However, the apparent growth of new particles observed at the OUC site indicates that 1) the concentrations of condensable vapors are higher than the required value to support the growth; 2) CS is not the dominant factor to determine the growth. Apart from the condensation vapor, weather conditions can also affect the growth of newly formed particles by changing gas-aerosol partitioning."

We may disagree with other parts of reviewer's comments. Preexisting particles can remove condensable vapors, nucleating clusters and newly formed particles from the atmosphere and then affect NPF and the growth of new particles. In addition to affecting condensable vapors, the authors cannot figure out other pathways for preexisting particles to affect particle growing larger than 10 nm.

12. Section 4.1. I do not agree that new particle formation occurred in this case, i.e. 30 August 2015.

Response: The NPF event on 30 August 2015 followed the definition proposed by Dal Maso et al. (2005), Hirsikko et al. (2007) and Kulmala et al. (2012), i.e., the nucleation mode of newly formed particle was observed for about 6 hours, and newly formed particles grew up to approximately 20 nm. In the revision, the time series of N<30 nm and CS (Fig. 3 at the end of this response) will be added and discussed. "To delve into the characteristics and evidence of oceanic precursors related NPF event on 30 August 2015, the transport pathway on that day was first zoomed in Fig. 6a. As is illustrated in Fig. 6b, the NPF event started to be observed at 09:40 under meteorological conditions

with ambient temperature of 26âĎČ, high relative humidity of 74%, and low wind speed of 1.5 m s-1 (not shown). During the first hour, the N<30nm increased from 0.6×103 cm-3 to 1.7×103 cm-3. The weaker NPF was associated with higher CS (2×10-2 s-1). When CS decreasing to approximately 1×10-2 s-1 after 11:00, the N<30nm sharply increased to 3×103 cm-3, and Dpg increase from 13 nm to 18 nm during the following 3 hours with the growth rate of 1.7 nm h-1. The signal of new particles disappeared at approximately 16:00. The overall NMINP was 5-20 times lower than all the other NPF events over the marginal seas, and the overall FR of 0.3 cm-3 s-1 was the minimum in this study."

We are sorry for color bar used in Fig. 6b (original version), which may mislead the reviewer. To make the weak signal of new particles to be visible, the scale of color bar in Fig. 6b was one fifth of other contour figures. The choice also makes the signal of pre-existing particles darker in Fig 6b in comparison with other contour figures.

13. Page 11, lines 12-15. Figure 7c does not show the altitude variation of the backward trajectories.

Response: We will add the altitudes in the supplementary as Fig. S7 (as shown in Fig. 4 at the end of this response).

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.

Response: We thank the comment. In the original version, the inclusion of MOUDI data on those two days are not well justified. The analysis is also too speculative to be convincing. The part will be revised as below:

"One set of MOUDI samples was collected during the period from 11:12 to 23:33. Although the sampling period had several hours delay against the NPF period on that

day, the air mass back trajectories swept the oceanic zone were highly consistent between the two periods (Fig. S8, as shown in Fig. 5 at the end of this response). The concentrations of particulate chemical species were thereby used to argue the polluted extent of air mass at these periods. The mass concentration of nss-sulfate and oxalate in particles less than 10 μm was 1.9 μg m-3 and 0.12 μg m-3 (derived from Fig. 7d), respectively, higher than in other non-NPF days in this study. Previous studies, e.g., Mukai et al., (1995), Matsumoto et al. (1997), and Jung et al. (2014), reported the mass concentration of nss-sulfate was approximately 0.5 μg m-3 in the clean background over the NWPO. The elevated concentration of nss-sulfate and oxalate on 8 April suggested the enhanced anthropogenic precursors input which was very likely from the continent of Japan based on the calculated air mass back trajectories (Fig. 7c,d). The MOUDI's data implied that the NPF event likely occurred in the air masses rich in anthropogenic precursors.

Compared to the event above on 8 April, the event on 13 April showed a longer NPF duration, i.e., the NPF event lasted from 07:50 to approximately 08:50 (Fig. 7b). The new particles signal was intermittently observed and the FR was difficult to calculate. The total particle number concentrations increased from 0.3$\times$104 cm-3 to the maximum of 2.6$\times$104 cm-3 during the NPF event, and the NMINP was 1.4$\times$104 cm-3. The Dpg increased from 8 nm to 14 nm in one hour, and the estimated GR was 3.6 nm h-1. One set of MOUDI samples was collected immediately after the event during the period from 09:10 to 21:05. Again, the calculated air mass back trajectories were consistent between the NPF period and the MOUDI's sampling period (Fig. S8, as shown in Fig. 5 at the end of this response). The mass concentration of nss-sulfate and oxalate in particles less than 10 μm was only 0.6 μg m-3 and 0.05 μg m-3. The values were close to the clean background of NWPO, indicating a much low anthropogenic input on 13 April (Fig. 7e). It is interesting that the NMINP was similar to each other during the two NPF events, although the air mass on 8 April was slightly polluted by anthropogenic inputs. However, due to lack of the measurements of precursor vapors, what caused NPF events needs further study."

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.

Response: "threshold" indeed causes misleading. Not all particles larger than 50 nm can be activated as CCN. In the revision, the part has been revised as "At SS of 0.4%, the Dpg increased from 19 nm to 50 nm during 10:40-13:10 (black circles in Fig. 8a) with AR fluctuating at 0.1-0.2 (Fig. 8c). After 13:10, the Dpg increased from 50 nm to 77 nm with increasing AR from $\sim$0.2 to $\sim$0.4. The results are consistent with those reported in the literature, i.e., particles smaller than 50 nm are unlikely activated as CCN at SS=0.4% (Dusek et al., 2006; Petters and Kreidenweis, 2007).

Following the reviewer's comments, we plotted time series of the number concentration of >50 nm particles (N>50nm) and the NCCN (as shown in Fig. 6 at the end of this response). Variations between N>50 nm and AR are clearly inconsistent. For example, AR showed an increasing trend from 0.2 to 0.4 during 13:10-15:00. N>50 nm decreased from 1.8$\times$104 cm-3 to 1.1$\times$104 cm-3 during the period of 13:10-13:30, then increasing to 2.4$\times$104 cm-3 at 14:30, followed a decreasing trend after 14:30. Atmospheric particles with the diameter larger than 50 nm include not only the grown new particles, but also preexisting particles. The inconsistency is not very surprised. The reviewer's comment is valid only when the number concentration of preexisting particles >50 nm was either near constant or was negligible relative to grown new particles during the growth period. This is not the fact. Therefore, we disagree with the reviewer on this point.

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?

Response: It should be outlier rather than exterior. We are sorry for our language

problem. In Figure 3a, there was a moderately good linear correlation at FRs $\leq$ 8 cm-3 s-1. The data points with FRs larger than 10 cm-3 s-1 are deviated largely from the regression curve obtained from the data with FRs $\leq$ 8 cm-3 s-1 and are thereby treated as outliers. For example, in the linear regression question of [NMINP]=3.9$\times$103$\times$FR, r=0.83 P<0.01, we consider three times of standard deviation for the slope. At the FR of 11.8 cm-3 s-1, the range of NMINP is predicted from 3.56$\times$104 to 5.64$\times$104 particles cm-3. The observed NMINP was only 1.17$\times$104 particles cm-3 and largely deviated from the range. In Figure 3b, the black triangle represents the GR of 26.3 nm h-1 and also deviated largely from the regression curve obtained from other data. The point is also treated as an outlier. This will be added in the figure caption.

The black dots in the contour plot of NPF events (Fig. 4, Fig. 6b, Fig. 7a, b, Fig. 8a and Fig. A1 in original version) represent the fitted geometric median diameter of new particles (Dpg) in 1-minute time resolution. The clarification will be added in the revised caption of Fig. 4.

In the revision, the shading in Fig 9 will be removed to avoid any misleading.

17. The manuscript needs to be grammatically checked by an editing company or a native English speaker professor.

Response: Thanks. The revised version will be language-edited.

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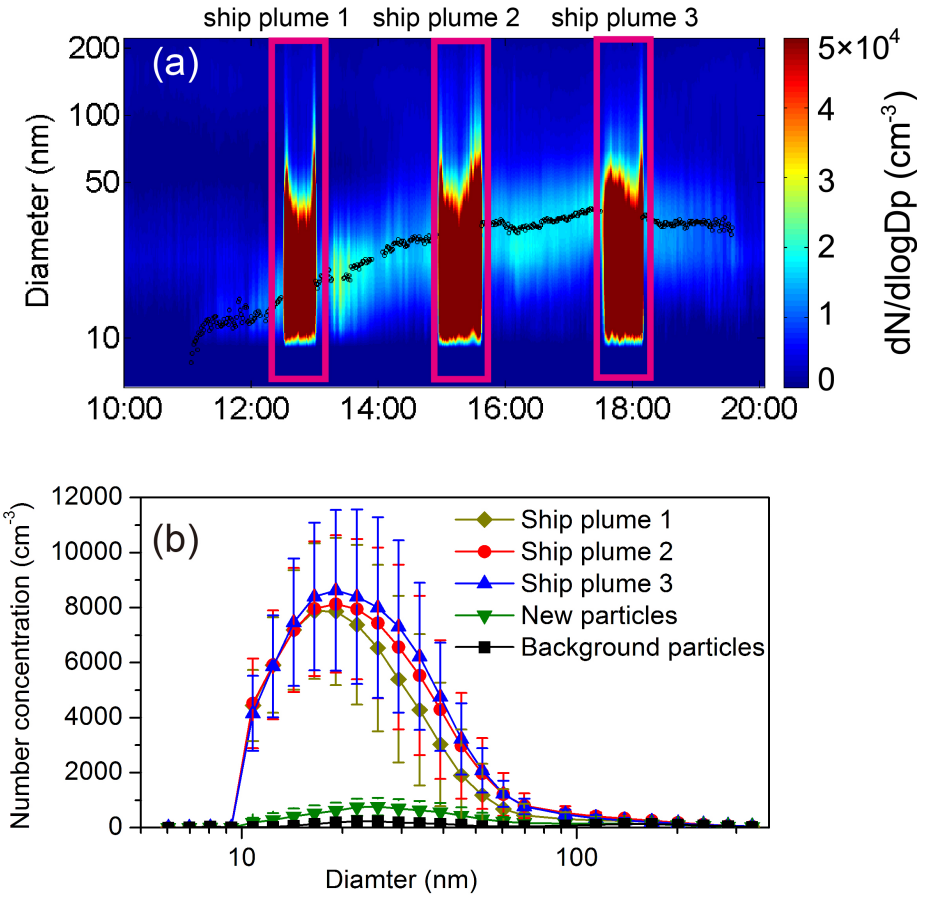

Fig. 1. Size distribution of ship emitted particles, new particles and background particles on 14 Nov. 2012.

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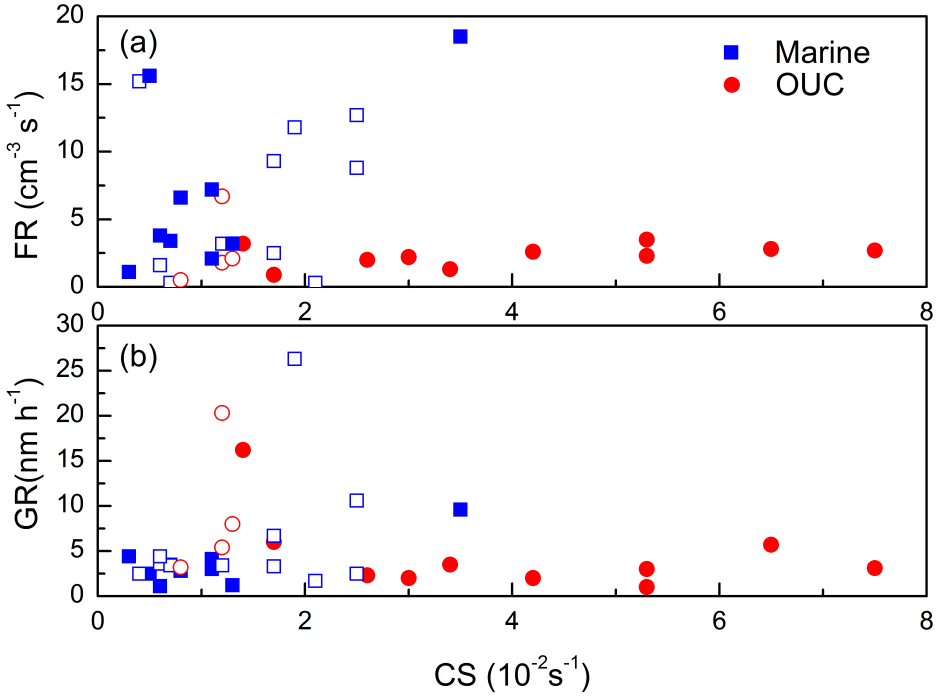

Fig. 2. Relationship of condensation sink (CS) with formation rate (FR) and growth rate (GR) over the marine (NWPO and marginal seas) and at OUC site (Solid markers represent the simultaneous NPF events).

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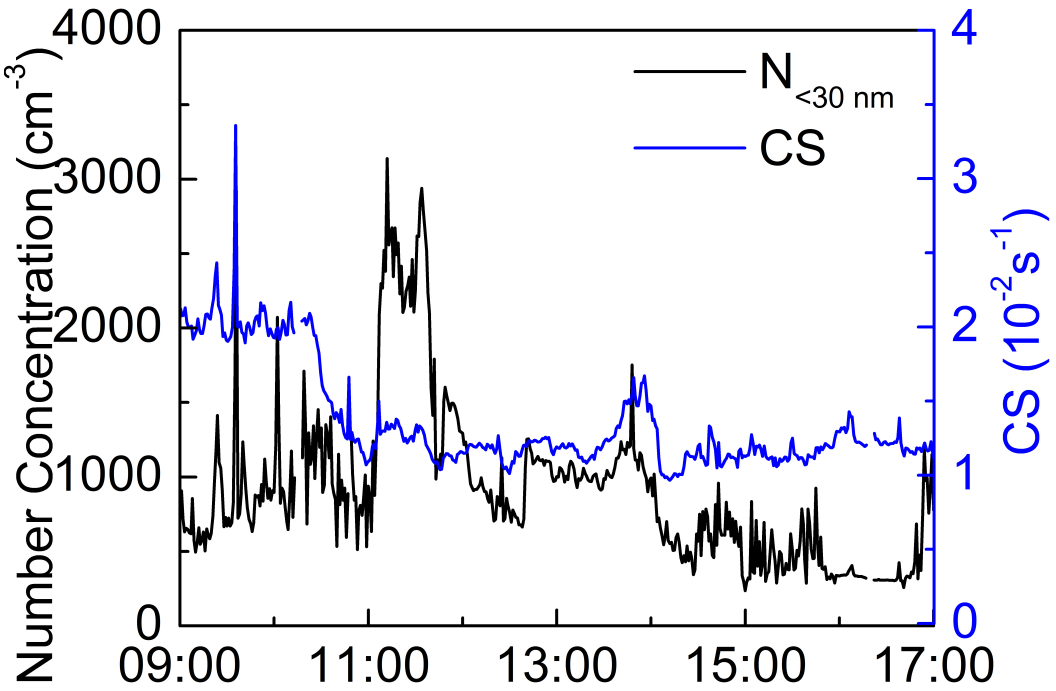

Fig. 3. Time series of N<30 nm and CS on 30 August 2015.

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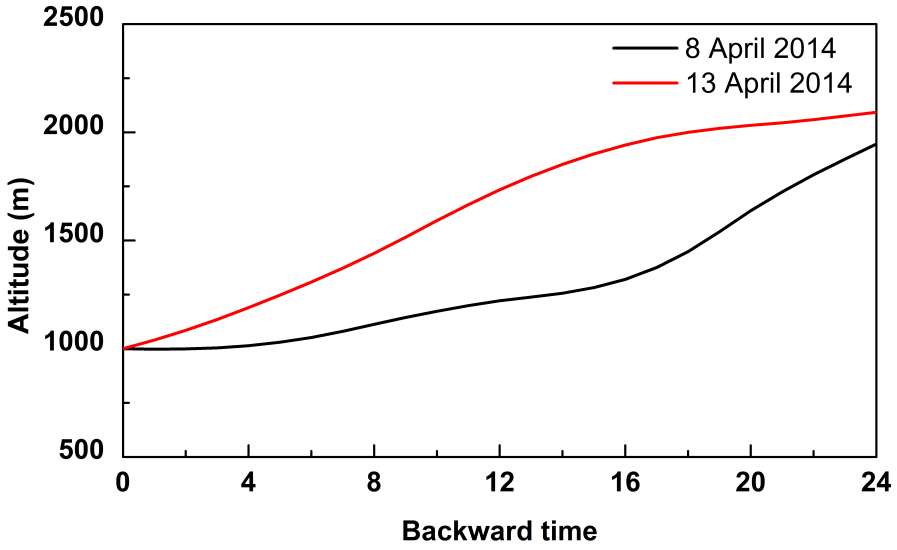

Fig. 4. Height of the air mass back trajectories on 8 and 13 April 2014 over the NWPO.

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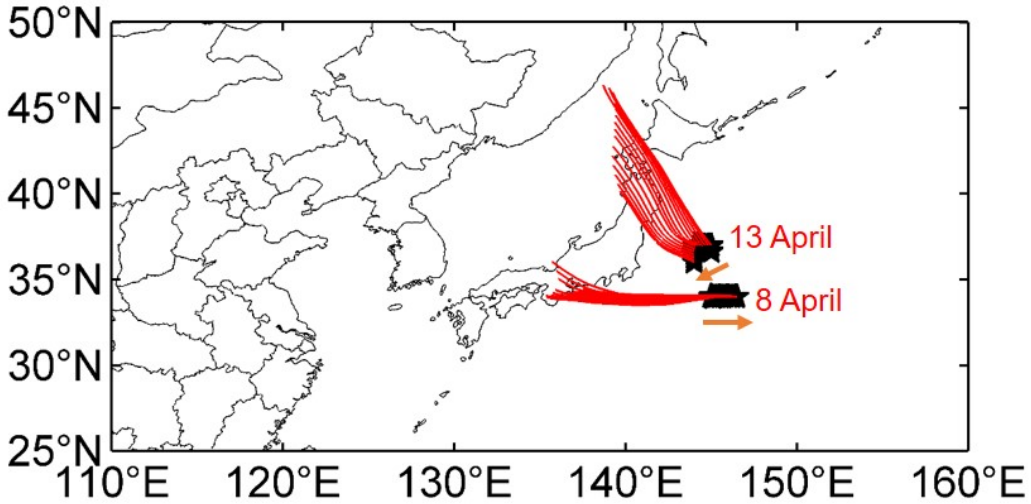

Fig. 5. 24-h air mass back trajectory throughout the NPF event and sampling periods (From 7:00 to 24:00 on 8 April, from 7:00 to 21:00 on 13 April).

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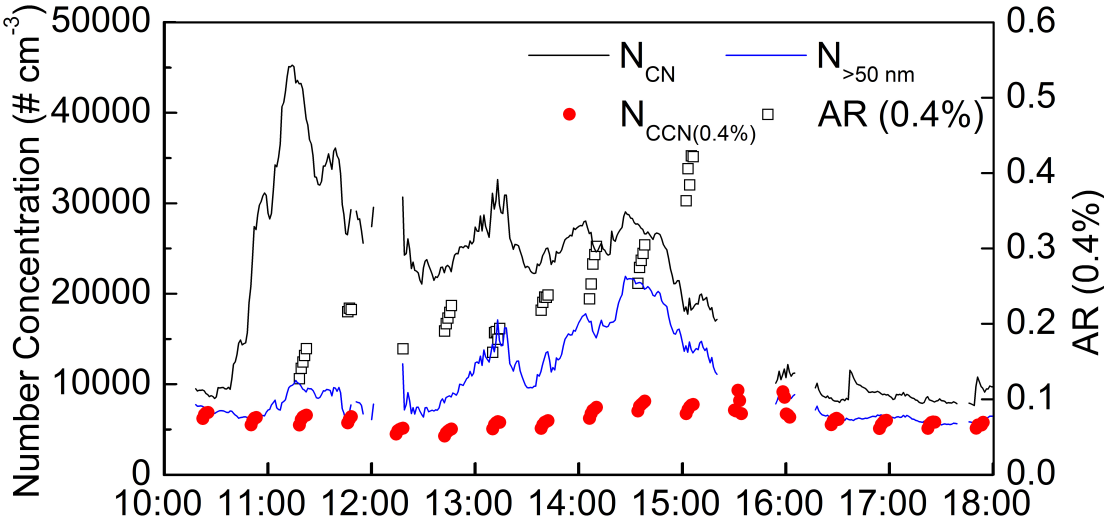

Fig. 6. Time series of total particle number concentration (NCN), number concentration of >50 nm particles (N>50 nm), CCN number concentration at the SS=0.4% (NCCN(0.4)) and AR on 4 September 2015 in BS.

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