We would like to thank the reviewer for valuable comments and suggestions. We have addressed all raised issues in the revision accordingly. Please kindly find our following point-by-point responses (the reviewer's comments in black, our responses in blue, and relevant changes in red).

Major comments:

1. The authors mention that they removed abnormal measurement spikes from their data set. They attribute these spikes to potential interference from their ship or other ships' emissions. This seems rather arbitrary way of throwing out data without more concrete reasoning. Can the authors provide chemical analysis of particles found in these spikes? Does the composition match what is expected for ship exhaust? Also, does the wind direction correlate with when ship emission would impact the container of instruments? How long did these spikes last? How many spikes in the data were there (hard to tell from the Figure S1)? Were the spikes just removed or averaged out? It would be helpful to actually reference the supplemental figures in the main text so the reader knows what the raw data of NCN looked like. The authors point out that shipping emission likely contribute significantly to particles in the South China Sea (line 120). Why then would the authors throw out observations from shipping emissions? In short, the authors must better justify why these spikes in data were thrown out and to know more definitively what was causing these spikes.

Reply:

We thank the reviewer for commenting on processing the abnormal spikes. We have added the temporal profiles of aerosol chemical composition, $NO/NO_2/NO_X$, along with wind direction and speed in Figure S1. Before removing the abnormal data, the spikes appear simultaneously in N_{CN} (d), organics (f) and $NO/NO_2/NO_X$ (g). There are mainly three potential sources: 1) emissions from our ship and other adjacent ships; 2) emissions from the kitchen in our ship; 3) smoking from sailors in our ship (It is really unfortunate, however, smoking did happen frequently during the measurements). It is difficult to distinguish those sources without detailed source apportionment. As we focus on cloud condensation nuclei (CCN) activity, we do not intend to apportion sources of organic matter in this paper and detailed organic source apportionment will be the focus of our next paper.

We measure NO/NO_2/NO_X every minute in comparison to 5 and 15 min per cycle respectively for the CCN and ToF-ACSM measurements. Hence more spikes could be found in the profiles of NO/NO_2/NO_X than in organic composition from the ToF-ACSM measurements. A total number of about 30, 40, 50 spikes were found in the profiles of organic composition, N_{CN}, and NO/NO_2/NO_X, respectively.

The weather station was placed on the rooftop of the cockpit which was located in the front of the ship. Wind direction and speed measured by the weather station was shown in Figure S1 (a, b). Note that the wind direction and speed are relative to those of the ship because it sailed most of the time during the measurements. Hence corrections are needed for accurate determination of the absolute wind direction and speed, which needs high temporal resolution and precise records on the direction and speed of the ship. Unfortunately, such records are not available during the measurements. Nevertheless, the spikes do not correspond significantly to changes of the wind direction and speed as shown in Fig. S1.

The spikes could last from minutes to hours. If they are from smoking, then they could last a few minutes. In some cases, if other ships sailed adjacently (i.e., within a few hundred meters) in the same direction, emissions from those ships could cause spikes that would last a few hours. When the ship anchored, emissions from the ship itself could cause spikes as well. In this study, the protocol for removal of spikes is to keep as many data as we can while avoiding too complicated data processing. We argue that ship emission could be one of the major particle sources in the South China Sea region. Here ship emission should be regional representative, rather than emissions from the ship itself and emissions from other adjacent ships which are really close to the measurement ship (i.e., within several hundred meters) because those emissions could cause extreme heterogeneity of spatial distributions. Hence those emissions should be excluded from further data analysis. Base on the above rationales, we remove all the spikes associated with smoking, emissions from the ship and from other adjacent ships, and emissions from the kitchen on the ship. We have also included concentrations of chemical composition and NO/NO_2/NO_X before and after removal in Fig. S1 for comparison. We believe that removal of the spikes based on the proposed protocol would not significantly affect our conclusions on the CCN activity in this study.

We have now added several sentences on p14 (L278-282) in the revision, “For consistency, we removed spikes likely associated with smoking, emissions from the ship itself and other adjacent ships and cooking from further data analysis, including either abrupt high number concentrations of particles (measured by SMPS), organics (measured by ToF-ACSM), and NO_X (measured by the NO_X monitor) (Detailed criteria can be referred to descriptions and Fig. S1 in supplementary).”

We have also included a paragraph of detailed descriptions regarding how to remove spikes from the data in supplementary.

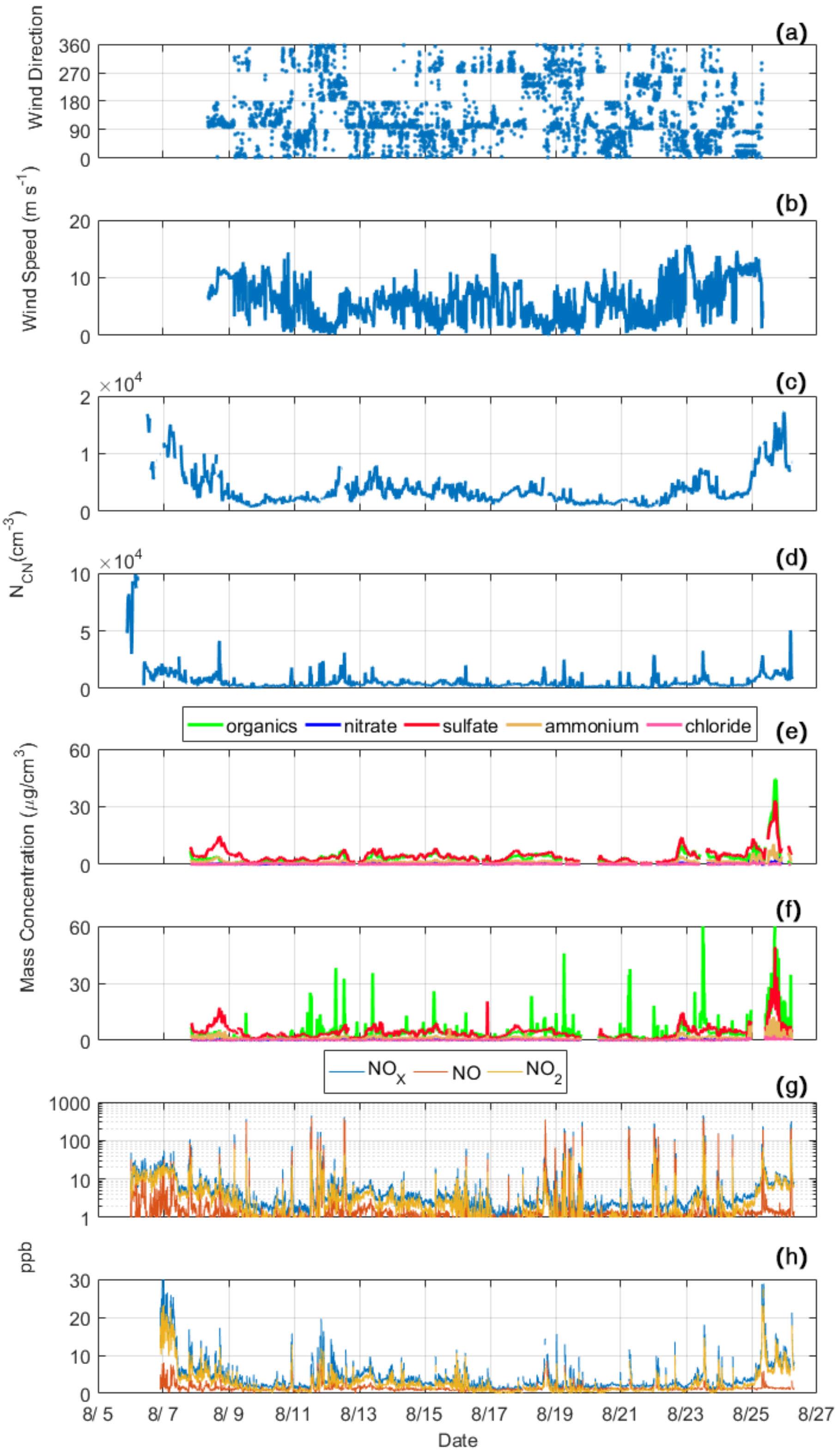


Figure S1. Temporal profile of wind direction (a) and wind speed (b), N_{CN} after (c) and before (d) removal of the abnormal data, chemical composition after (e) and before (f), and $NO/NO_2/NO_X$ before (g) and after (h) removal of the abnormal data.

2. Line 270: authors attribute high sulfate content to ship emissions instead of oxidation of DMS from ocean. Can the authors estimate sulfur emissions from the ocean from previous studies and compare that to what they saw? This would provide more solid evidence that the elevate sulfate content is due to ship emissions than from ocean DMS.

Reply:

We thank the reviewer for valuable suggestions on sulfate formation. Possible sources of sulfate include ship emissions, DMS oxidation, and transport from inland etc. The oxidation of DMS leads to formation of sulfur dioxide and methansulfonic acid (MSA) both of which can be further oxidized to produce non-sea-salt (NSS) sulfate in marine atmosphere. Oxidation of SO_2 from ship emissions or inland transport can also be a major source of NSS sulfate (Savoie et al., 2002). As an intermediate between DMS and sulfate, MSA in principle can be detected by ToF-ACSM, although resolution of the instrument is low. We are currently working on MSA identification and quantification from this cruise measurement. Preliminary results show that the fraction of sulfate from DMS oxidation is far below that from ship emissions. An early study showed that anthropogenic sulfate accounted for about 81-97% of NSS sulfate over China Sea (Guo et al., 1996). A ratio of 15-655 NSS sulfate to MSA in $PM_{2.5}$ was reported in the Northern South China Sea (Zhang et al., 2007), much higher than that (18-20) in the remote marine (Savoie et al., 2002). Here we employed the Modern-Era Retrospective analysis for Research and Applications, Version 2 (MERRA-2) to analyze the distribution of ratio of sulfate to MSA at 925 hPa during the measurement period. The results were shown in Fig. S2 and the ratio ranged from 100 to 10000 over the SCS, much higher than that in the remote Pacific Ocean (1-50). In addition, it also increases with latitude, indicating that the anthropogenic emission is the major source of the total sulfate in the Northern SCS region.

To be clarified, we have added several sentences to discuss the origin of sulfate on p13-14 (L257-270) in the revision, “The oxidation of DMS leads to formation of sulfur dioxide and methansulfonic acid (MSA) both of which can be further oxidized to produce non-sea-salt (NSS) sulfate in marine atmosphere. Oxidation of SO_2 from ship emissions or inland transport can also be a major source of NSS sulfate (Savoie et al., 2002). As an intermediate between DMS and sulfate, MSA in principle can be detected by ToF-ACSM, although resolution of the instrument is low. Preliminary results show that the fraction of sulfate from DMS oxidation is far below that from ship emissions. An early study showed that anthropogenic sulfate accounted for about 81-97% of NSS sulfate over China Sea (Gao et al., 1996). A ratio of 15-655 NSS sulfate to MSA in $PM_{2.5}$ was reported in the northern South China Sea (Zhang et al., 2007), much higher than that (18-20) in the remote marine (Savoie et al., 2002). Here we employed the Modern-Era Retrospective analysis for Research and Applications, Version 2 (MERRA-2) to analyze the distribution of ratio of sulfate to MSA at 925 hPa during the measurement period (GMAO, 2015). The results were shown in Fig. S2 and the ratio ranged from 100 to 10000 over the SCS, much higher than that in the remote Pacific Ocean (1-50). In addition, it also increases with latitude, indicating that the anthropogenic emission is likely the major source of the total sulfate in the northern SCS region."

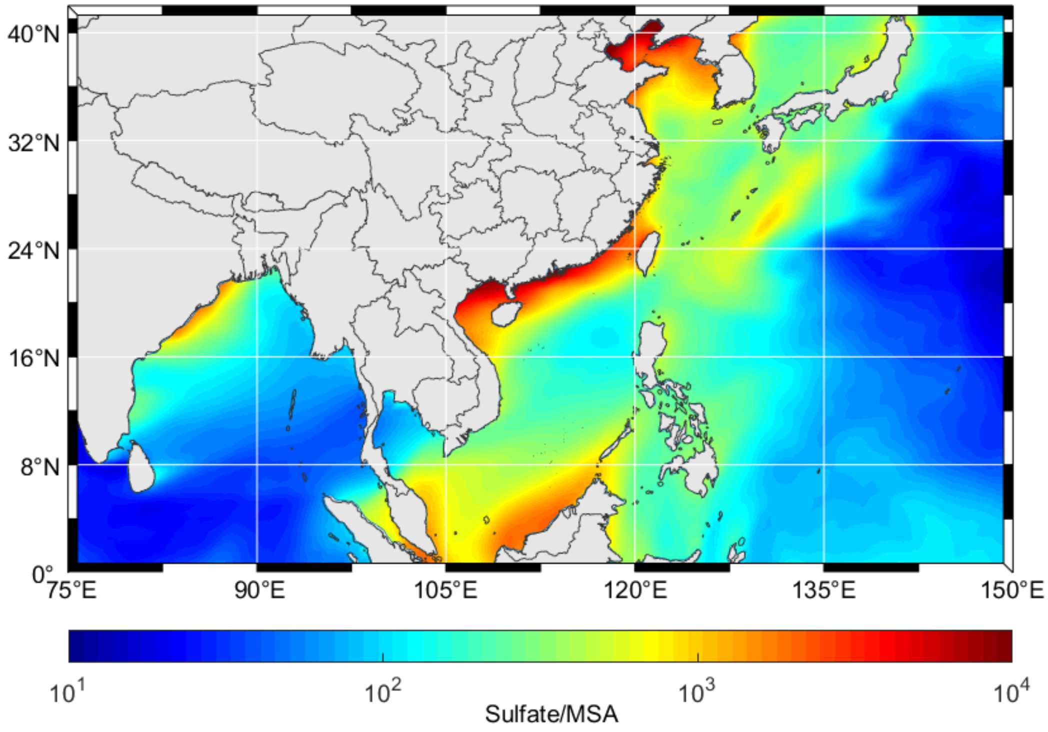


Figure S2. The ratio of sulfate to MSA at 925 hPa from MERRA-2 reanalysis dataset (GMAO, 2015).

Minor Comments:

1. Abstract seems unnecessarily long. Would be more readable if it were shortened to include the main point of the paper.

Reply:

We have modified the abstract as follows, "Aerosol particles in marine atmosphere have been shown to significantly affect cloud formation, atmospheric optical properties, and climate change. However, high temporally and spatially resolved atmospheric measurements over sea are currently sparse, limiting our understanding of aerosol properties in marine atmosphere. In this study, a ship-based cruise campaign was conducted over northern South China Sea (SCS) region during summertime 2018. Chemical composition of non-refractory PM_1 (NR-PM_1), particle number size distribution (PNSD) and size-resolved cloud condensation nuclei (CCN) activity were measured by a time-of-flight aerosol chemical speciation monitor (ToF-ACSM), and the combination of a cloud condensation nuclei counter (CCNc) and a scanning mobility particle sizer (SMPS), respectively. Overall, aerosol particles exhibited a unimodal distribution centering at 60~80 nm and chemical composition of the NR-PM_1 was dominated by sulfate (~46%) which likely originated from anthropogenic emissions rather than dimethyl sulfide (DMS) oxidation. Two polluted episodes were respectively observed at the beginning (P1) and at the end (P2) of the campaign and both were characterized by high particle number concentrations (N_{CN}) which originated respectively from local emissions and from emissions in inland China via long range transport as shown by back trajectory analysis. The concentrations of trace gases (i.e., O_3, CO, NO_X) and particles (N_{CN} and N_{CCN} at ss=0.34%) were elevated during P2 and decrease with the offshore distance, further suggesting important impacts of anthropogenic emissions from the inland Pearl River Delta (PRD) region on the northern SCS. Two relatively clean periods (C1 and C2) prior to and after tropical storm Bebinca were classified due to substantial removal of pollutants by strong winds and rainfalls accompanying with the storm. During C1 and C2 periods, the air was affected by air masses from southwest and from Indo-China Peninsula, respectively. Chemical composition measurements showed an increase of organic mass fraction during P2 compared to C2; however, no obviously different κ values were obtained from the CCNc measurements, implying that the air masses carried pollutants from local sources during long range transport. We report an average value of about 0.4 for aerosol hygroscopicity parameter κ which falls within the literature values (i.e., 0.2-1.0) for urban and remote marine atmosphere. In addition, our results showed that the CCN fraction ($N_{CCN}/N_{CN,tot}$) and the κ values obtained from the CCNc measurements (ss=0.34%) had no clear correlation either with the offshore distance or with concentrations of the particles. Our study highlights dynamical variations of particle properties and the impact of long range transport from the China continent and Indo-China Peninsula on the northern SCS region during summertime."

2. Line 21: high temporally and spatially resolved

Reply: The sentence has been revised to "…high temporally and spatially resolved" in the abstract.

3. Line 82: how was mixing state important for CCN? If it's important, why did the authors not address how mixing state may impact their measurements conclusions?

Reply:

Mixing state of particles has important impact on CCN prediction based on chemical composition and D_{50} if the measurements were significantly affected by primary emissions, since the above methods for CCN prediction assume particles to be internally mixed. Freshly emitted black carbon (BC) particles are externally mixed and hydrophobic, hence they are hard to be activated as CCN. However, aged BC particles coated with inorganic matter will become internally mixed and CCN activated. In addition, mixing state of particles can also affect the shape of activation curves. For externally mixed particles, their activation curves are usually smooth (Cai et al., 2018).

We have plotted the activation curves at 0.18% ss during the four periods (P1, P2, C1, and C2) as shown in Fig. S4 in supplementary and have added a paragraph to discuss mixing state on p21-22 (L429-441) in the revision, "The mixing state and heterogeneity of particles can affect the steepness of the activation curves (Cai et al., 2018). A steeper curve indicates that particles intend to be internally mixed and have a higher similarity in hygroscopicity. The average activation curves at 0.18% SS during the P1, C1, C2 and P2 periods are shown in Fig. S4. The parameter C (in Eq. 1) can be used to present the steepness of activation curve. A small C value indicates a steep activation curve. The C values during P1, C1, C2 and P2 periods were -8.5, -14.3, -13.7 and -10.6, respectively. The smooth curve and the largest C value during P1 suggest that particles had a higher degree of external mixing and higher heterogeneity, owing to the local fresh emissions. The C values during C1 and C2 periods were close and smaller than those in pollution periods, implying particles during clean periods were more aged and tend to be more internally mixed. The backward trajectories show that the air masses during clean periods were less affected by fresh emissions. The activation curve during P2 period was smoother than C1 and C2 but steeper than P1, indicating that the particles during this period could be a mixture of aged particles from China inland and fresh particles from onshore emissions."

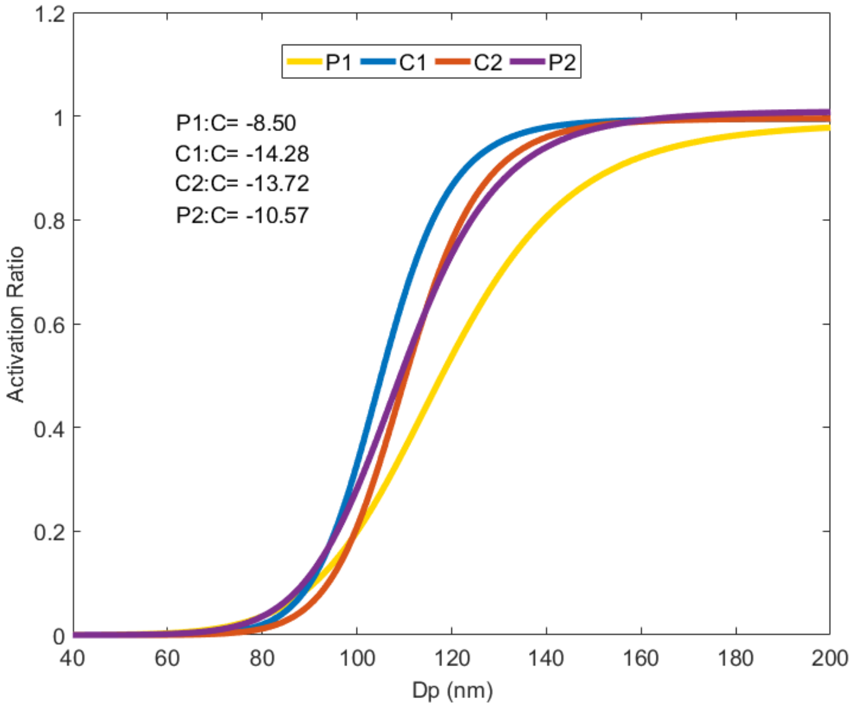


Figure S4. The activation curve at 0.18% ss during the P1, C1, C2 and P2 periods. Different colors represent different periods.

4.Line 102: unclear the sentence starting with Furthermore.

Reply: "Furthermore" has been replaced by "Meanwhile" on p6 (L95)

5. Line 148: delete both

Reply: deleted.

6. Line 149: comma before which

Reply: A comma has been added.

7. Line 161: ", which is listed in Table 1,"

Reply: The sentence has been revised.

8. Line 213: remove firstly

Reply: Removed.

9. Line 299: please use parallel phrasing for this sentence.

Reply: This sentence has been revised on p15 (L299-301) in the revision to "Comparison of the hygroscopicity parameter κ obtained from this study, urban Guangzhou, remote marine Okinawa, remote South China Sea, and mountain Goldlauter was shown in Fig. 3."

10. Line 385: was likely

Reply: It has been revised.

11. Line 405: if the particles are from biomass burning, do the authors observe any of the classic biomass burning tracers (such as K)?

Reply:

The K can be a tracer of biomass burning and sea salt. However, the vaporizer of ACSM and AMS can produce a large amount of K^+, which will interfere the ambient K signal. To our knowledge, there could be a large uncertainty of the K concentration measured by ACSM. The levoglucosan can also be the tracer of biomass burning. Nevertheless, the mass resolution of our ACSM is too low to distinguish levoglucosan from other species. Currently, there are no direct evidences indicating the impact of biomass burning. We analyzed fire detection data during the measurements from MODIS in combination with back trajectories during C2 which clearly show that the air parcels pass through the fire region. We have modified Fig. 7 and added a discussion on p20 (L407-410), "The backward trajectories during C2 pass through the burning regions in Southeast Asia (e.g., Viet Nam, Laos, Cambodia etc.), also supporting this conjecture.

However, more solid evidences are needed since the observation of biomass burning tracers (such as K and levoglucosan) is missing in this campaign.”

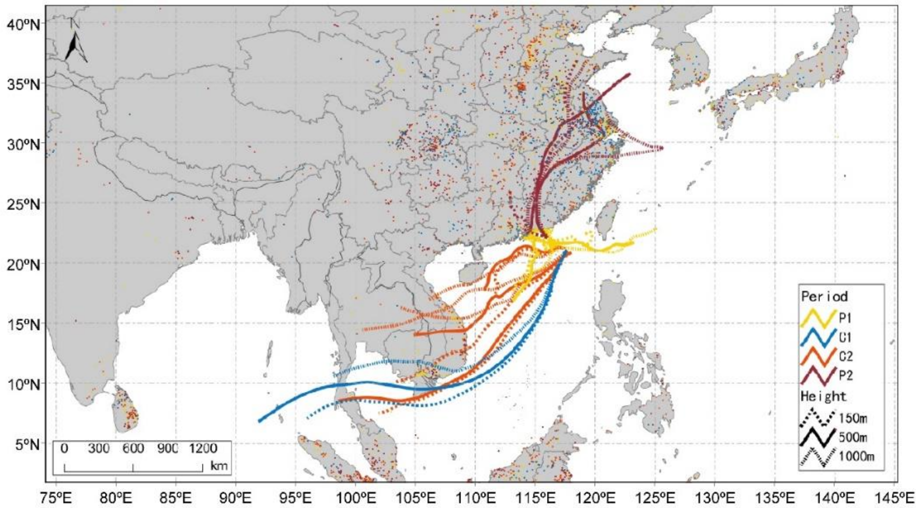


Figure 7. The 72 h backward trajectories arriving at the location of the vessel with three heights (150 m, 500 m, and 1000 m) during P1, C1, C2, and P2, respectively. The dots represent the fire spots detected by MODIS.

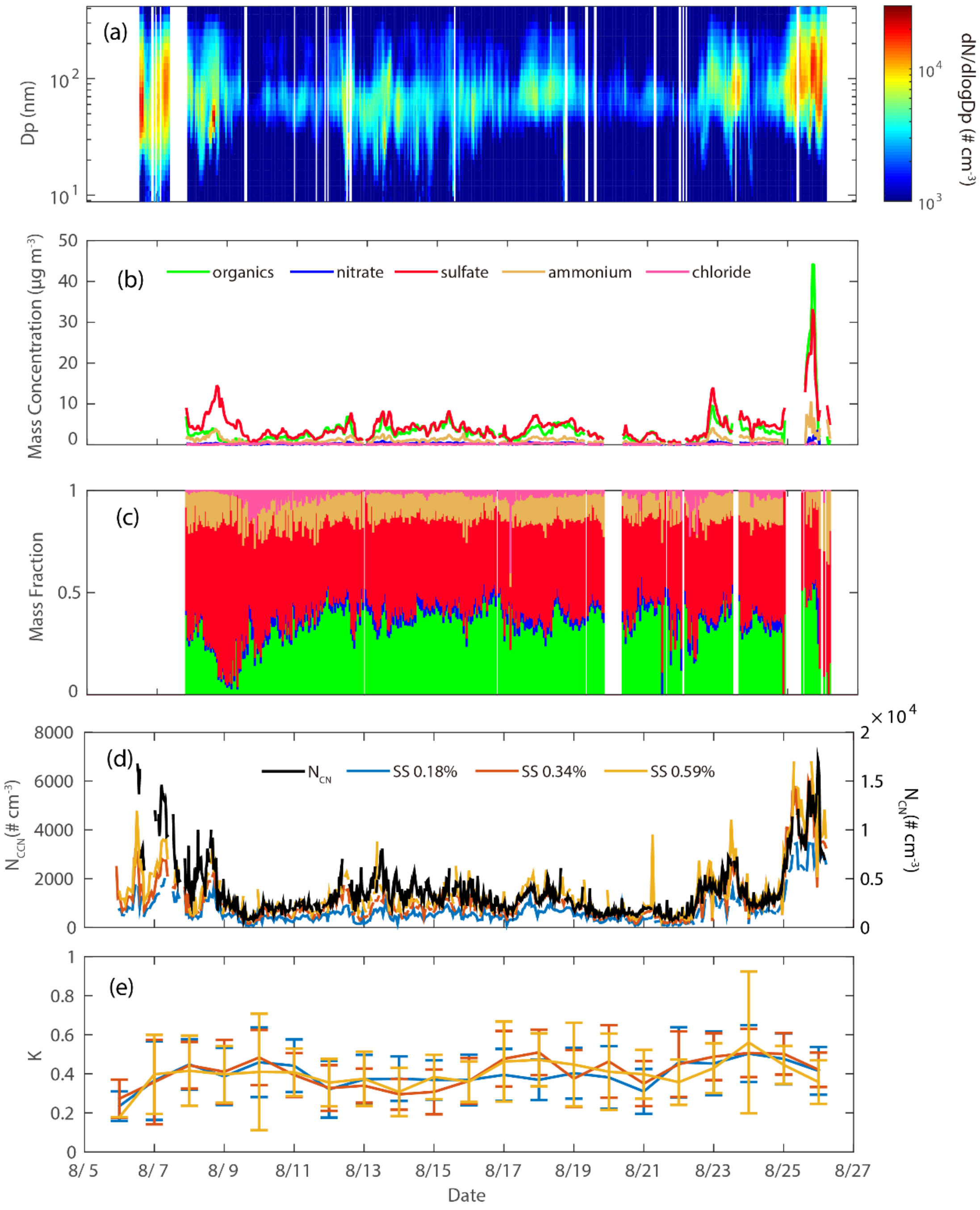
12. Table 2: the values have too many significant figures given the uncertainties of the measurement

Reply: Table 2 has been revised.

Period	ss	0.18%	0.34%	0.59%
P1	N_{CCN} (# cm^{-3})	1825	3969	7198
	D_{50} (nm)	132	96	65
	$N_{CCN}/N_{CN,tot}$	0.19	0.34	0.49
C1	N_{CCN} (# cm^{-3})	566	978	1330
	D_{50} (nm)	105	67	49
	$N_{CCN}/N_{CN,tot}$	0.31	0.54	0.71
C2	N_{CCN} (# cm^{-3})	536	844	1183
	D_{50} (nm)	108	68	48
	$N_{CCN}/N_{CN,tot}$	0.32	0.55	0.73
P2	N_{CCN} (# cm^{-3})	4969	7140	8679
	D_{50} (nm)	101	65	49
	$N_{CCN}/N_{CN,tot}$	0.49	0.74	0.85

13. Figure 2d) what is the black line? And which lines go to which y axis?

Reply: The black line represents the N_{CN}, corresponding to the y axis on the right. The rest represents the N_{CCN} at different ss, corresponding to the left axis. Figure 2 has been modified.



References:

Cai, M., Tan, H., Chan, C. K., Qin, Y., Xu, H., Li, F., Schurman, M. I., Liu, L., and Zhao, J.: The size-resolved cloud condensation nuclei (CCN) activity and its prediction based on aerosol hygroscopicity and composition in the Pearl Delta River (PRD) region during wintertime 2014, Atmos. Chem. Phys., 18, 16419-16437, 2018.

Gao, Y., Arimoto, R., Duce, R. A., Chen, L. Q., Zhou, M. Y., and Gu, D. Y.: Atmospheric non-sea-salt sulfate, nitrate and methanesulfonate over the China Sea, J. Geophy. Res. Atmos., 101, 12601-12611, 10.1029/96jd00866, 1996.

Global Modeling and Assimilation Office (GMAO) (2015), MERRA-2 inst3_3d_aer_Nv: 3d,3-Hourly,Instantaneous,Model-Level,Assimilation,Aerosol Mixing Ratio V5.12.4, Greenbelt, MD, USA, Goddard Earth Sciences Data and Information Services Center (GES DISC), Accessed: 8, 2018, 10.5067/LTVB4GPCOTK2.

Savoie, D. L., Arimoto, R., Keene, W. C., Prospero, J. M., Duce, R. A., and Galloway, J. N.: Marine biogenic and anthropogenic contributions to non-sea-salt sulfate in the marine boundary layer over the North Atlantic Ocean, J. Geophy. Res. Atmos., 107, AAC 3-1-AAC 3-21, 10.1029/2001JD000970, 2002.

Zhang, X., Zhuang, G., Guo, J., Yin, K., and Zhang, P.: Characterization of aerosol over the Northern South China Sea during two cruises in 2003, Atmos. Environ., 41, 7821-7836, 2007.