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).

1. The manuscript is quite difficult to follow as it is just describing a lengthy dataset by correlating each other. It is highly unclear what is the main scientific conclusions of the data analysis. This issue is well represented in the lengthy abstract of the manuscript. It is just way too long, which makes difficult to grasp the scientific merits of data analysis. I strongly recommend the authors to remind themselves a couple of main scientific findings that they hope to come across in the manuscript for the revision.

Reply:

We thank the reviewer for valuable suggestions. As we state in the abstract, aerosol particles in marine atmosphere can significantly affect cloud formation, atmospheric optical properties, and climate change. Currently, high temporally and spatially resolved atmospheric measurements over sea are sparse, limiting our knowledge on understanding of aerosol properties in marine atmosphere. We utilized measurement data from a recent ship-based cruise campaign to analyze the CCN activity over the northern South China Sea and to investigate the effects of continental emissions on the CCN activity. We agree with the reviewer that the abstract should be concise and we have hence 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. For example, the analysis for the different air masses, came across during the cruise, should be developed further more thorough fashion. I would present available ground data either concentrations of emissions from the different region to discuss their characteristics to elucidate how the chemical evolution affects the outflow to evaluate whether the observational result makes sense or not.

Reply:

We thank the reviewer for valuable suggestions on the air masses analysis and ground data for chemical evolution analysis. We admit that currently physical and chemical properties of aerosol particles based on ground measurements (i.e., chemical composition and particle size distribution) are still largely lacking, especially for long-term measurements. In addition, such nearshore ground measurements and cruise-based measurements are very also scarce, making it hard to evaluate detailed chemical processes for aerosol particles over the SCS region. Besides, we focus on the CCN activity rather than chemical process of aerosol particles over the northern SCS in this study. Based on the above reasons, we analyzed the MODIS fire spots to discuss potential impacts of biomass burning on the air masses which might affect our measurements as shown in Fig. 7 (please refer to answer #5). In addition, we also discuss the contribution of anthropogenic emission to sulfate over the South China Sea based on the MERRA-2 data as shown in Fig. S2. (please refer to answer #3).

3. For example discussion about the presence of sulfate over the South China Sea (line numbers between 269 to 271) can certainly go further by discussing upper end DMS emission rates and whether the assumption can account the observed SO2.

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."

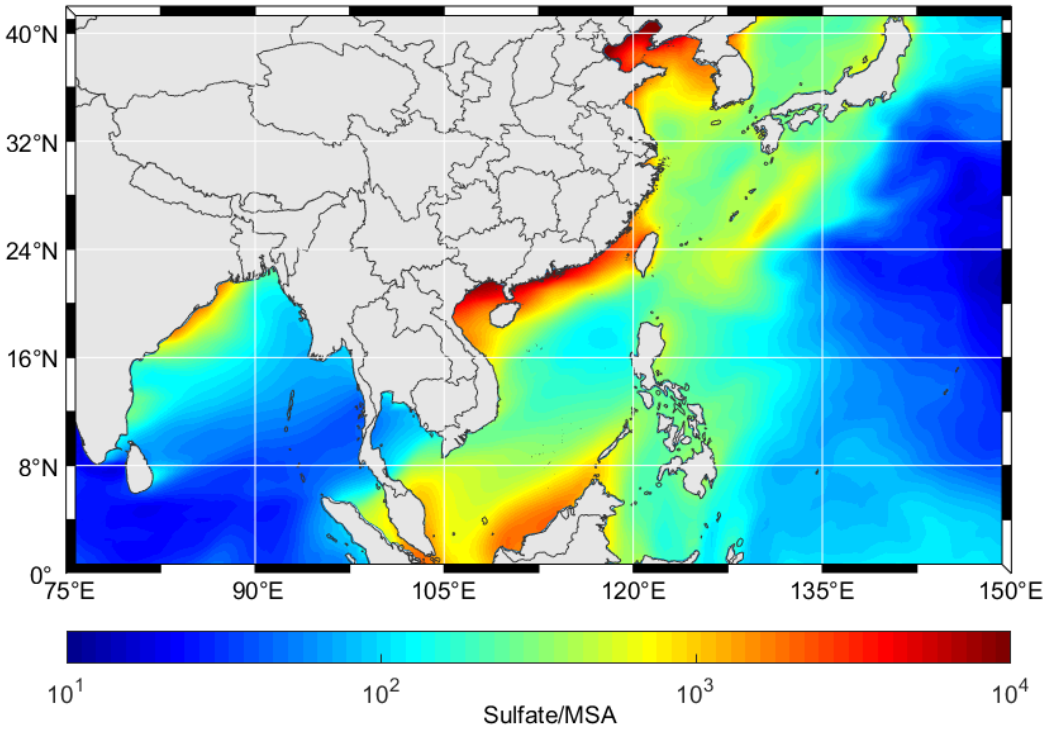

Figure S2. The ratio of sulfate to MSA at 925 hPa from MERRA-2 reanalysis dataset (GMAO, 2015).

4. Another example is in line 342. CO is an obvious long lived tracer for pollution, therefore the correlation of CO with parcels # is not surprising. I would recommend the authors to discuss further more process level aerosol chemistry evolution than these rather one dimensional comparisons of observables.

Reply:

We agree with the reviewer that CO is a long lived tracer for pollution and its correlation with particles might be expected. However, in a recent study, CO was used as a wildfire tracer to study the impact on aerosol properties in the marine atmosphere and it was found that CO concentration had a strong correlation with non-volatile particle concentration (R^2=0.70) (Zheng et al., 2020). Here we used CO as a tracer for biomass burning or anthropogenic emissions. We found that CO is not always correlated with particles, for example, it is indeed correlated with particles during the second half of the cruise while is not correlated at all during the first half. Hence CO can be used as a useful tracer for source identification. In this paper, we focus on the effects of pollution on CCN activity. Detailed source apportionment and chemical processes will be the focus of our next paper.

We have added several sentences on p17 (L338-344) in the revision, "The variation of SO_2 concentration was independent of N_{CN}, suggesting that SO_2 did not share the same source with particles. The CO concentration is positively correlated with N_{CN} during the second half of the cruise, while no obvious correlation is observed during the first half, implying that sources of particles could be different during the two periods. The correlation during the second half of the cruise indicates that the particles might share the same source with CO which was attributed to biomass burning or anthropogenic emissions."

5. It is even more troubling by attributing biomass burning sources as presented in lines between 403 to 405. I would recommend to take full advantage of your wealthy dataset and back trajectory analysis to solidly argue the origin of the observed airmass of bio mass burning.

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."

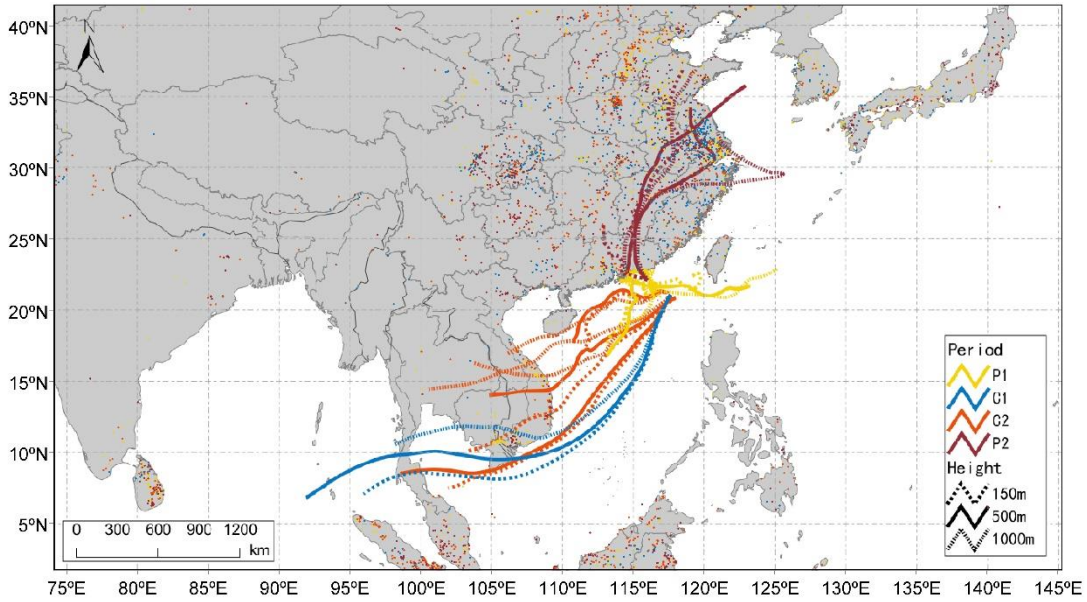

Figure 7. The 72 h backward trajectories arriving at the location of the vessel with three heights (150 m, 500 m, and 1000 m) during P1, C1, C2, and P2, respectively. The dots represent the fire spots detected by MODIS.

Reference:

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

Geng, X., Mo, Y., Li, J., Zhong, G., Tang, J., Jiang, H., Ding, X., Malik, R. N., and Zhang, G.: Source apportionment of water-soluble brown carbon in aerosols over the northern South China Sea: Influence from land outflow, SOA formation and marine emission, Atmospheric Environment, 229, 117484, ttps://doi.org/10.1016/j.atmosenv.2020.117484, 2020.

Huang, S., Wu, Z., Poulain, L., van Pinxteren, M., Merkel, M., Assmann, D., Herrmann, H., and Wiedensohler, A.: Source apportionment of the organic aerosol over the Atlantic Ocean from 53°N to 53°S: significant contributions from marine emissions and long-range transport, Atmos. Chem. Phys., 18, 18043-18062, 10.5194/acp-18-18043-2018, 2018.

Zheng, G., Sedlacek, A. J., Aiken, A. C., Feng, Y., Watson, T. B., Raveh-Rubin, S., Uin, J., Lewis, E. R., and Wang, J.: Long-range transported North American wildfire aerosols observed in marine boundary layer of eastern North Atlantic, Environ. Int., 139, 105680, https://doi.org/10.1016/j.envint.2020.105680, 2020.