

We thank the reviewers for taking the time to review our manuscript and for providing constructive advices. We have addressed each of the reviewers' comments individually. Below are our responses to each comment, with the reviewers' comments in black, our responses in red, and the revised manuscript content in italicized orange font.

## #Reviewer 1

The manuscript presents a study on cloud condensation nuclei (CCN) activity in the South China Sea (SCS), examining seasonal variations and anthropogenic influences through two shipborne campaigns in the summer and winter of 2021. The researchers measured aerosol chemical composition, particle number size distribution (PNSD), and CCN, revealing significant seasonal differences in aerosol properties and the impact of anthropogenic emissions on CCN activity.

Despite these insights, the manuscript appears hastily written, lacking novelty, and presents vague data and discussions that are difficult to interpret. Furthermore, the manuscript fails to clearly differentiate these campaigns from previously reported measurements in the SCS, reducing its contribution to the existing body of knowledge. The manuscript is challenging to follow, primarily describing a lengthy dataset with confusing correlations and writing that diminish readability. The main scientific conclusions are unclear, and the text is overly lengthy, obscuring the scientific merits of the data analysis. It is recommended that the authors focus on a few key scientific findings in their revision. Based on the given title, the manuscript should focus solely on reporting the measurements from the cruise campaigns. However, it also delves into scientific analysis, giving the impression that the authors aim to publish their analytical findings under the guise of reporting measurements. This approach detracts from the clarity and purpose of the study, as the title suggests a primary emphasis on measurement data rather than comprehensive scientific interpretation. In its current form, the manuscript is not suitable for publication in ACP.

*Reply: Thanks for the reviewer's comment. According to reviewer's comment, the topic of our article focused on the different effect of anthropogenic influences between summer and winter on CCN activities in SCS. We restructured the manuscript. In the first section of the Results and Discussion (3.1 CCN concentration and aerosol characteristics over SCS in summer and winter), we introduce the basic information of the two cruises conducted in the South China Sea during summer and winter. The second section focuses on the impact of different terrestrial air mass sources on CCN activity in the two seasons (3.2 Anthropogenic influence on CCN concentration in different season). In the last section, we followed reviewer's advice employing the CCN closure method to discuss the effects of aerosol composition and mixing state on CCN activity (3.3 CCN closure analysis).*

### Major comments:

1.Regarding the main scientific findings presented in the manuscript, the discussion does not adequately support these claims. The authors assert that they represent the seasonal variation of aerosol-CCN activity; however, this is not convincingly demonstrated. The winter observations span only 10 days and, unlike the summer campaign, do not spatially cover the entire region. Furthermore, there is no discussion about the potential bias introduced by the limited sampling, which could significantly affect the derived results. Addressing this gap is crucial for validating the study's conclusions.

Reply: Thanks for reviewer's comment. Both the summer and winter cruises are extensive observational missions covering fields such as marine geology, oceanography, and atmospheric science. As a result, the timing and routes of these cruises are planned with an interdisciplinary perspective. The winter cruise has a smaller range and shorter duration (only 10 days) than the summer cruise. This limitation is due to adverse weather conditions, such as strong winter monsoon winds causing poor sea conditions, and the short expedition which considered that it was the first scientific deployment of the research vessel Sun Yat-sen University (lines 145-148).

*Unfortunately, due to adverse weather conditions, such as strong winter monsoon winds causing poor sea conditions, and the fact that it was the first scientific deployment of the research vessel Sun Yat-sen University, the winter cruise had a shorter duration and covered a narrower spatial range compared to the summer cruise.*

In our revised version, we focus on the influence of terrestrial air masses from different sources on CCN activity during the summer and winter seasons. The SCS, characterized by a typical monsoon climate, is predominantly affected by the southwest monsoon in summer and the northeast monsoon in winter. As a result, the terrestrial air masses affecting the South China Sea in winter primarily originate from the Chinese mainland, whereas in summer, they mainly come from the Indochinese Peninsula and the Philippine Islands. Although our winter observations mainly focused on the northern SCS compared to the broader summer observations, they still provide valuable insights into the impact of winter terrestrial air masses on this region.

In the Introduction section, we have added background information on the climate of the South China Sea (Lines 109-122):

*The SCS experiences a typical monsoon climate with distinct seasonal wind direction changes (Wang et al., 2009). The northeast monsoon, occurring from November to March, is characterized by stronger average wind speeds and longer period compared to the southwest monsoon, which dominates from June to August. The transitional periods occur from April to May and September to October. During the northeast monsoon, air pollutants are primarily transported to the SCS by terrestrial air masses from China (Xiao et al., 2017; Liu et al., 2014; Geng et al., 2019). In contrast, during the summer, pollutants mainly originate from terrestrial air masses from the Indochinese Peninsula and Maritime Southeast Asia (Geng et al., 2019; Liang et al., 2021; Sun et al., 2023). These varying sources of anthropogenic emissions exerts different impacts on CCN activity differently across seasons. Additionally, the high cloud fraction over the SCS varies from approximately 0.3 to 0.7 across different months, indicating that aerosol-cloud interactions in the region may differ between seasons (Lu et al., 2022). However, due to limited observational data, our understanding of seasonal variations in CCN activity in the SCS remains incomplete. Conducting comprehensive observational studies on CCN activity across different seasons is essential for improving our understanding of aerosol-cloud interactions in the SCS.*

We agreed that the winter cruise's temporal and spatial limitations present certain constraints to our study. Firstly, the spatial limitations hinder our ability to accurately observe the impact of air masses from the Mainland China on the remote SCS. Secondly, the combined temporal and spatial constraints suggest that other terrestrial air masses, such as those from the Indochinese Peninsula, may also influence the remote SCS.

Additionally, we discuss the limitations of our observations in lines 434 to 437:

*Additionally, our observation in winter focused on the CCN activity over the northern SCS, while the influence of air masses from Mainland China in remote SCS was still unclear. Further observations in remote SCS areas could help clarify the anthropogenic*

*influence during winter under the effect of the winter monsoon.*

2. While the title effectively highlights the focus on seasonal variations in CCN activity in the South China Sea (SCS), the Introduction needs further development for full contextualization. It should provide detailed background on the SCS's unique meteorological and environmental characteristics, such as meteorology and anthropogenic influences, to explain its significance for aerosol studies. Explicitly identifying specific research gaps will underscore the necessity of the study. Additionally, clearly stating the objectives of study and hypotheses will provide a precise research roadmap. Integrating a comprehensive review of recent, relevant literature will position the research within the broader scientific context, highlighting its novelty and relevance. Does author think that focusing solely on regional measurements can effectively reduce uncertainties related to aerosol-cloud interactions and radiative forcing? Emphasizing the potential significance and broader impact of the findings, such as advancements in scientific understanding and improvements in climate modeling, will underline the study's importance. Addressing these aspects will enhance the clarity, relevance, and impact of the Introduction, setting a solid foundation for the manuscript.

**Reply:** We appreciate the reviewer for this valuable suggestion. To clarify our research gap, we have added background information on the climate of the South China Sea and discussed the different impacts of terrestrial air masses under the influence of various monsoons in the Introduction section (Lines 110-123):

*The SCS experiences a typical monsoon climate with distinct seasonal wind direction changes (Wang et al., 2009). The northeast monsoon, occurring from November to March, is characterized by stronger average wind speeds and longer period compared to the southwest monsoon, which dominates from June to August. The transitional periods occur from April to May and September to October. During the northeast monsoon, air pollutants are primarily transported to the SCS by terrestrial air masses from China (Xiao et al., 2017; Liu et al., 2014; Geng et al., 2019). In contrast, during the summer, pollutants mainly originate from terrestrial air masses from the Indochinese Peninsula and Maritime Southeast Asia (Geng et al., 2019; Liang et al., 2021; Sun et al., 2023). These varying sources of anthropogenic emissions exerts different impacts on CCN activity differently across seasons. Additionally, the high cloud fraction over the SCS varies from approximately 0.3 to 0.7 across different months, indicating that aerosol-cloud interactions in the region may differ between seasons (Lu et al., 2022). However, due to limited observational data, our understanding of seasonal variations in CCN activity in the SCS remains incomplete. Conducting comprehensive observational studies on CCN activity across different seasons is essential for improving our understanding of aerosol-cloud interactions on the SCS.*

**This emphasizes the purpose for our study, which focuses on the influence of terrestrial air masses on CCN activity in the South China Sea during the winter and summer seasons under the respective monsoon influences. Besides, we stated the focus of our study at the end of the introduction (lines 126-128).**

*Our results provide valuable insights into the differences in CCN activity between winter and summer, as well as the influence of different types of terrestrial air masses on CCN activity in the SCS across different seasons.*

3. Although the methodology section comprehensively details the measurements and data analysis, some critical information is missing.

Firstly, the authors need to provide a rationale for selecting the specific periods in summer and winter for the cruise measurements. The inconsistency in measurement

periods between summer and winter requires clarification, along with the reasoning behind the chosen cruise routes. This scientific justification is crucial for understanding the relevance to the study's objectives.

Additionally, I recommend incorporating the detailed data quality control procedures (currently in Text S1) into the methodology section, as this is crucial for a measurement report. The authors should clarify why wind direction data from other platforms was not utilized for quality control during winter, particularly in the absence of onboard wind measurements, and explain how ship emissions were prevented from being considered in the measurements. Additionally, the justification for the wind direction ranges used to filter out ship emissions during the summer campaign needs to be provided. Including wind rose plots, similar to those in Hung et al. (2018), would enhance this section. The current data filtering approach, adopted from previous studies, lacks sufficient justification, especially given the differences in cruise measurements and periods. The choice of wind direction ranges and data filtering criteria needs a more logical and scientific basis specific to this study. Furthermore, the rationale for calibrating the ACSM only at the start and end of the cruise, particularly for the longer summer campaign, requires clarification. Additionally, please provide an abbreviation for SMCA. Moreover, The authors mentioned removing abnormal measurement spikes ( $> \pm 3 \sigma$ ), attributing them to potential ship emissions. This approach appears arbitrary without concrete reasoning. Providing chemical analyses of particles in these spikes would help determine if they match expected ship exhaust compositions.

Additionally, correlating these spikes with wind direction data and specifying their duration, frequency, and handling (whether removed or averaged out) is essential.

**Reply:** Thanks for the reviewer's suggestion. Our two cruises were conducted during the prevailing summer monsoon and winter monsoon periods, respectively. We have added the objectives of these two cruises in lines 133-136:

*These two cruises were interdisciplinary scientific expeditions, integrating fields such as marine geology, oceanography, and atmospheric environment. The primary objective in atmospheric environment was to investigate the impact of different monsoons on the atmospheric environment of the South China Sea (SCS).*

Due to the fact that the December 2021 cruise was the first observation mission conducted by the "Sun Yat-sen University" vessel, the meteorological station installed from December 19 to December 22 was under calibration, resulting in a lack of meteorological data. Consequently, in the previous version, we did not use meteorological data for data screening for this cruise. In the revised version, we have included a screening criterion based on relative wind direction and relative wind speed for the period after December 22 in winter. Referring to other literature (Huang et al., 2018; Cai et al., 2020; Liang et al., 2021), we adopted two criteria for screening: the first is based on the components associated with ship emissions such as organic matter, black carbon, and fine particles—if these components showed sudden peaks, they were considered influenced by ship emissions; the second criterion is based on relative wind direction and relative wind speed. We have added a new section in the methods part to describe the data exclusion method in detail (lines 252-266):

*To ensure reliable atmospheric samples in the SCS and mitigate the influence of research vessel emissions, we applied the following data processing procedures (Huang et al., 2018; Cai et al., 2020; Liang et al., 2021).*

*Firstly, we identified organic compounds, black carbon (BC), and small particulate matter (41.4 nm particles) as indicators of ship emissions, recognizing their sudden peak values as indicative of the ship's own emissions.*

Secondly, we accounted for the relative positions of the ship's chimney and the sampling tube. During the summer cruise, we excluded data corresponding to a relative wind direction (with respect to the ship's bow) between  $150^\circ$  and  $270^\circ$  and a relative wind speed (with respect to the ship's speed) of less than  $2.5 \text{ m s}^{-1}$  (Fig. S4a, Fig. S5a1, and Fig. S6a-c). During the winter cruise, we excluded data for a relative wind direction between  $150^\circ$  and  $220^\circ$  and a relative wind speed of less than  $2.5 \text{ m s}^{-1}$  (Fig. S4b, Fig. S5b1, and Fig. S6d-f).

Applying these criteria, 74.8% of the data in summer and 92.2% in winter (both at 10-minute resolution) were classified as "clean" and retained for analysis. The timeseries of data before and after quality control is shown in Fig. S7.

We have provided the criteria for screening based on relative wind direction and relative wind speed in the supplement, along with wind rose diagrams for the relevant substances and time series graphs before and after data exclusion.

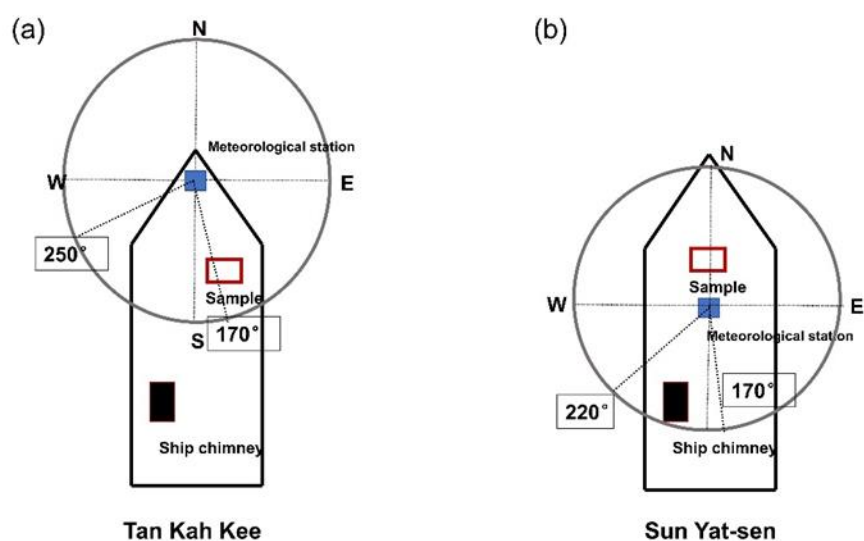


Figure S4. Instrument and ship chimney location in two cruises.

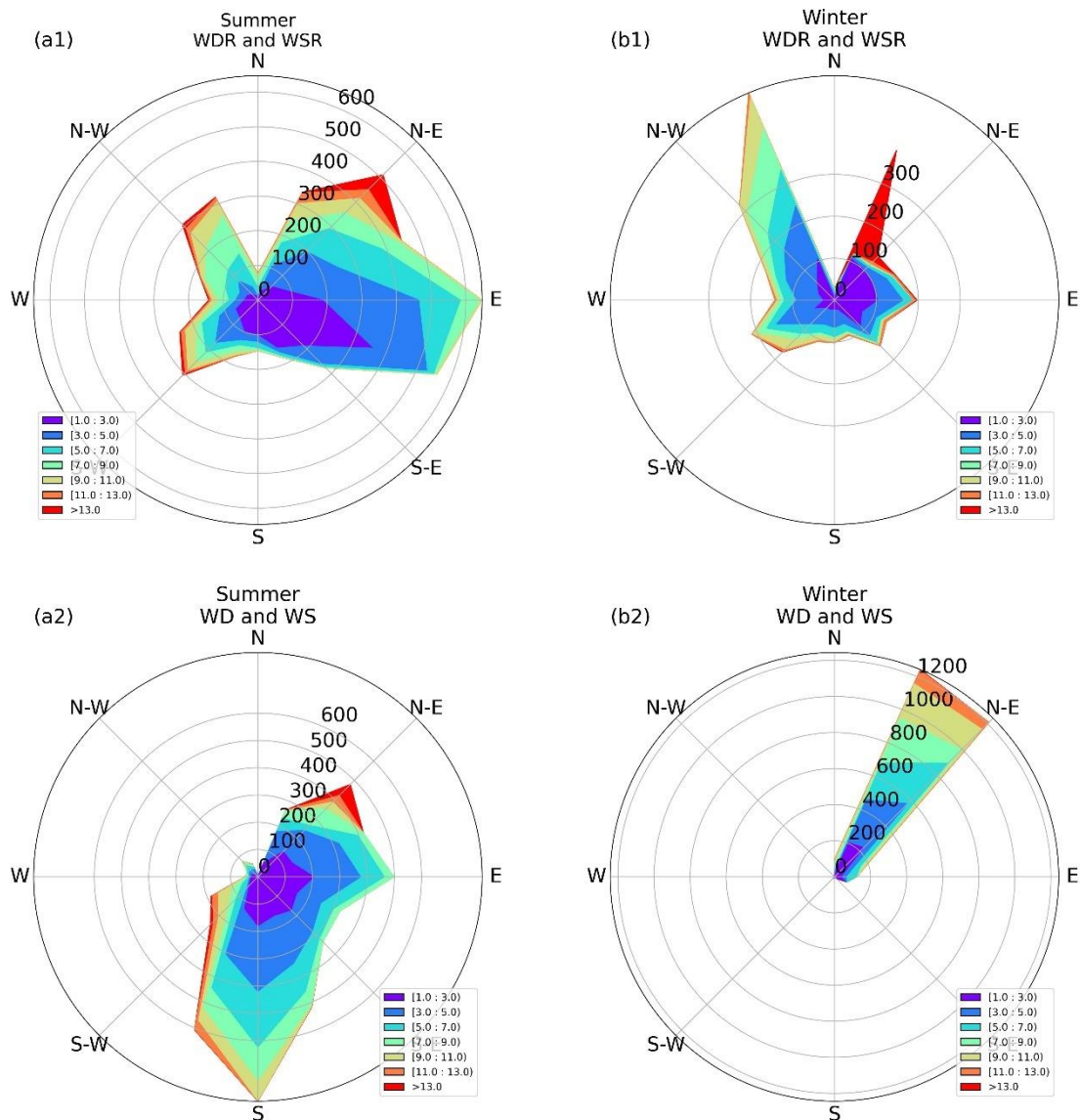


Figure S5. Wind rose of the relative wind direction (with respect to the bow) and relative wind speed (with respect to the ship speed) in summer and winter cruises; The radius represents the frequency of wind direction occurrences, and the shaded areas indicate wind speed (a1) and (b1); Wind rose of the wind direction and wind speed in summer and winter (a2) and (b2).

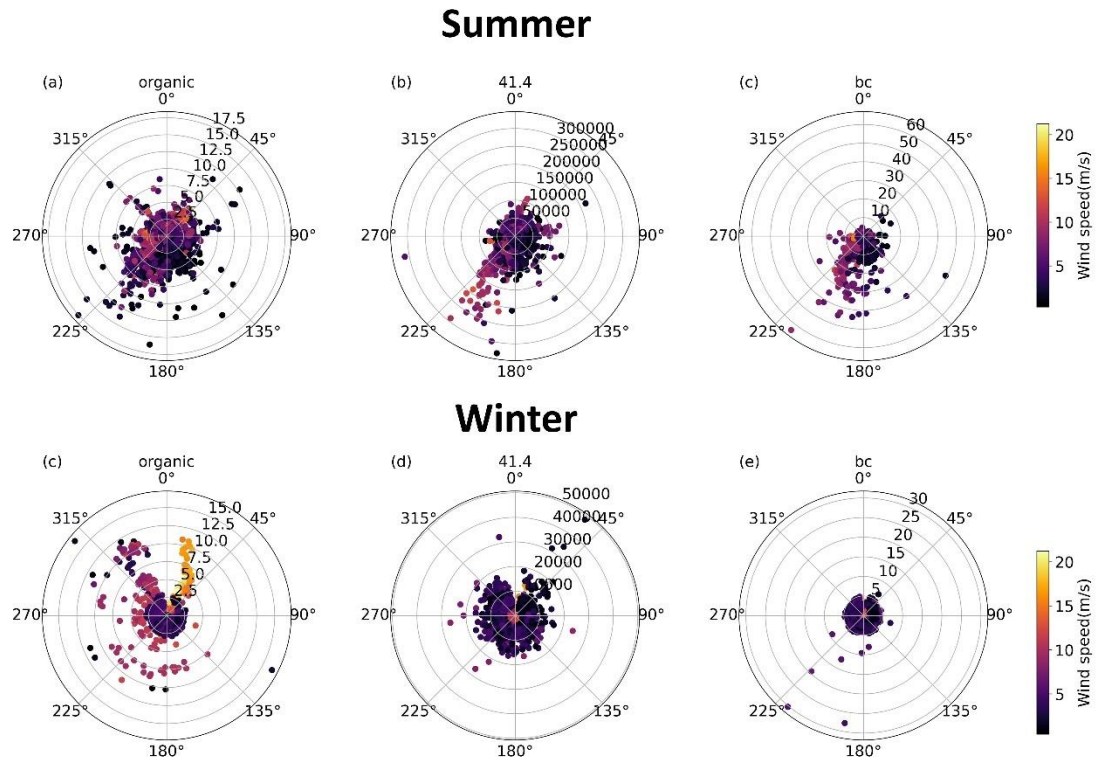


Figure S6. Wind rose of the organic, particle in 41.4 nm, and black carbon (BC) in summer (a-c) and winter (c-e) measurements; The radius represents the organic and BC mass concentration and number concentration ( $dN/d\log D_p$ ) of particle in 41.4 nm, and the color indicate wind speed.

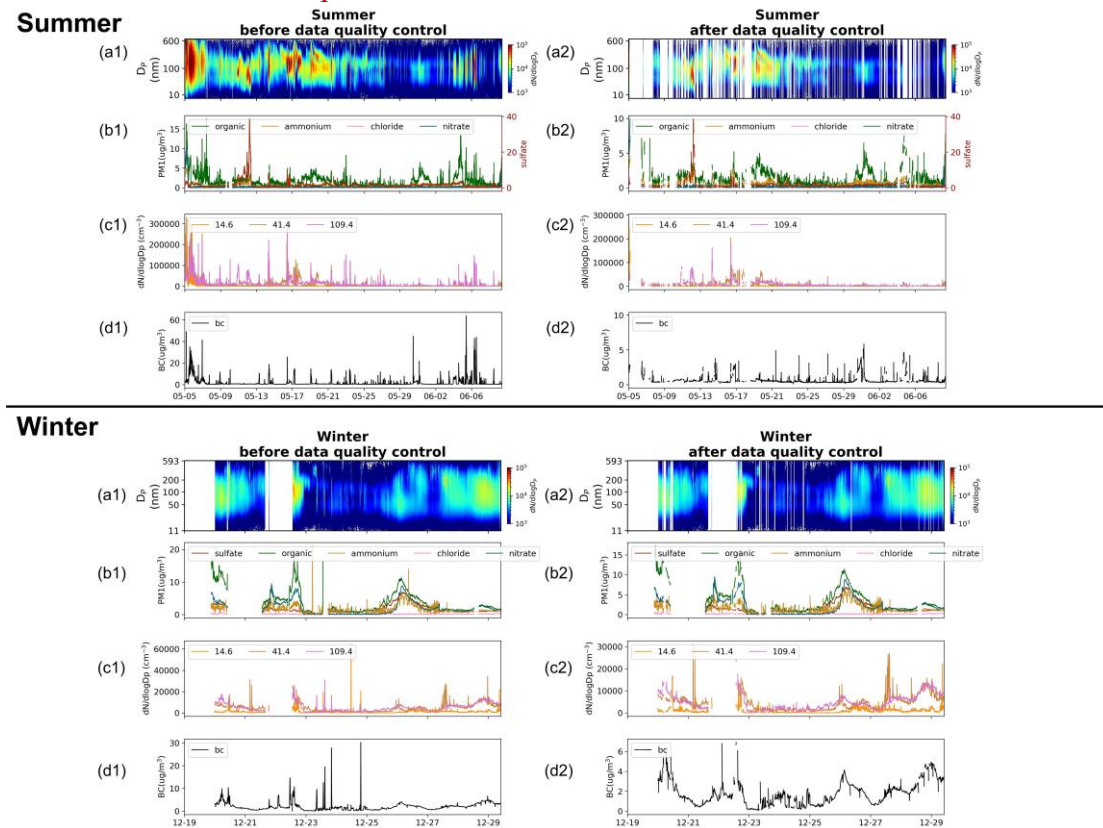


Figure S7. Timeseries of particle number size distribution (a) and (e), mass

concentration of NR-PM<sub>1</sub> (b) and (f), particle number concentration in 14.6, 41.4, and 109.4 nm (c) and (g), mass concentration of black carbon (d) and (h); The figure number from (a) to (d) means the data in summer, and the figure number from (e) to (h) means the data in winter; The number 1 represented the data before data quality control and the number 2 represent the data after data quality control.

Reference:

1. Huang, S., Wu, Z. J., 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 degrees N to 53 degrees S: significant contributions from marine emissions and long-range transport, *Atmos. Chem. Phys.*, 18, 18043-18062, doi: 10.5194/acp-18-18043-2018, 2018
2. Cai, M. F., Liang, B. L., Sun, Q. B., Zhou, S. Z., Chen, X. Y., Yuan, B., Shao, M., Tan, H. B., and Zhao, J.: Effects of continental emissions on cloud condensation nuclei (CCN) activity in the northern South China Sea during summertime 2018, *Atmos. Chem. Phys.*, 20, 9153-9167, doi:https://doi.org/10.5194/acp-20-9153-2020, 2020.
3. Liang, B., Cai, M., Sun, Q., Zhou, S., and Zhao, J.: Source apportionment of marine atmospheric aerosols in northern South China Sea during summertime 2018, *Environ. Pollut.*, 289, 117948, doi:https://doi.org/10.1016/j.envpol.2021.117948, 2021.

4. Regarding the back trajectory analysis, there is confusion, particularly in winter. Unlike in summer, distinguishing terrestrial and mixed air masses in winter is challenging despite notable differences in particle concentrations and chemical compositions. The short winter cruise period and limited sampling frequency complicate the analysis. To improve clarity, a cluster analysis of back trajectories is recommended to identify distinct air mass origins and pathways, as suggested by Patel et al., 2021, ERC. Including pathway altitudes would enhance understanding of air mass transport. The authors should justify the 48-hour period for back trajectory calculations and consider whether extending this period would provide more comprehensive information. Conducting cluster analysis for distinct source region identification is strongly encouraged.

Reply: Thank you for the suggestion. We selected midpoints of the ship trajectories for two periods in summer and winter, and conducted 72-hour back trajectories at 500m hourly, followed by cluster analysis. During the summer cruise, we conducted cluster analysis at two key locations: the midpoint of the ship's track before the outbreak of the summer monsoon (May 5-23) and the midpoint of the track after the summer monsoon began (May 24-June 9). In the winter cruise, cluster analysis was performed at two specific locations: the ship's anchorage near Big Ten-thousand Mountain Island (December 19-22 and December 27-29) and the midpoint between Dawan Mountain Island and Yongxing Island (December 23-26). In summer, we identified three periods: those influenced by terrestrial air masses from Luzon Island ("Luzon" period), from the Indochinese Peninsula ("Indochinese Peninsula" period), and by marine air masses ("Marine-s" period). Due to the small fraction of air masses from Palawan Island, we did not consider them in this study. In winter, we identified periods influenced by terrestrial air masses from Mainland China ("Mainland China" period), Mainland China-marine mixed air masses ("Mixed" period), and marine air masses (Marine-w period).

The result of cluster analysis result is shown in Fig. 4.



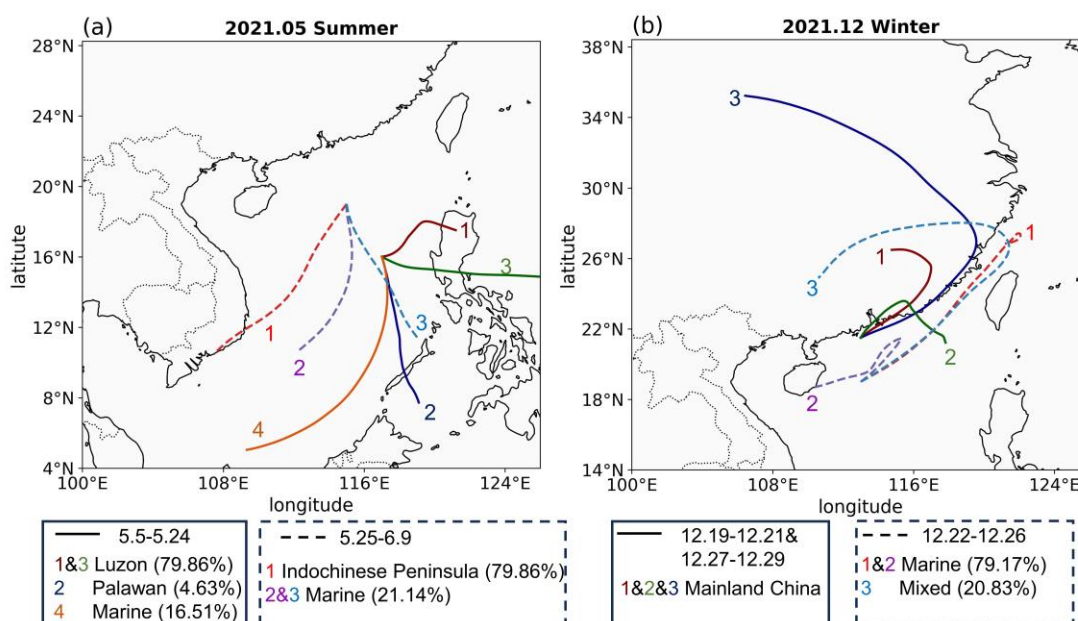


Figure 4. The cluster analysis result in summer (a), and winter (b). The solid line in summer means cluster analysis from May 5 to May 24 and the dash line in summer means cluster analysis from May 25 to June 9; The solid line in winter means cluster analysis from Dec 19 to Dec 21 and Dec 27 to Dec 29, and the dash line in winter means cluster analysis from Dec 22 to Dec 26.

5. In several instances, the authors present global statements based on regional studies without clarifying that the findings are specific to particular regions. For example, the statement in lines 71-72: "Ajith et al. (2022) showed that 64% of particles..." does not universally apply, as the referenced study is not global. It is essential to specify the region where the study was conducted to ensure the accuracy and relevance of the statement.

Additionally, numerous instances in the manuscript show incorrect citation of equation and figure numbers, which need to be addressed for clarity and precision.

Reply: We have specified the regions in this sentence (lines 71-73):

*Ajith et al. (2022) showed that 64% of particles can be activated as CCN when  $\kappa$  is equal to 0.37, whereas when  $\kappa$  decreases to 0.23, only 48% of particles can be activated in the tropical coastal area.*

Additionally, we have checked the figure and equation numbers throughout the manuscript to ensure correct referencing.

6. The manuscript lacks a scientific discussion on how the significantly different chemical compositions in terrestrial and mixed air masses during winter, characterized by high inorganic and low organic concentrations, impact hygroscopicity and CCN activity. It is highly recommended to create a plot showing the contributions of inorganic and organic components for various air masses, further dividing them into specific species (refer to Patel et al., 2021). This approach will provide more comprehensive insights than species plots alone.

Reply: Thanks for reviewer' s valuable suggestion. We have replotted the figure and present in Fig. 5:

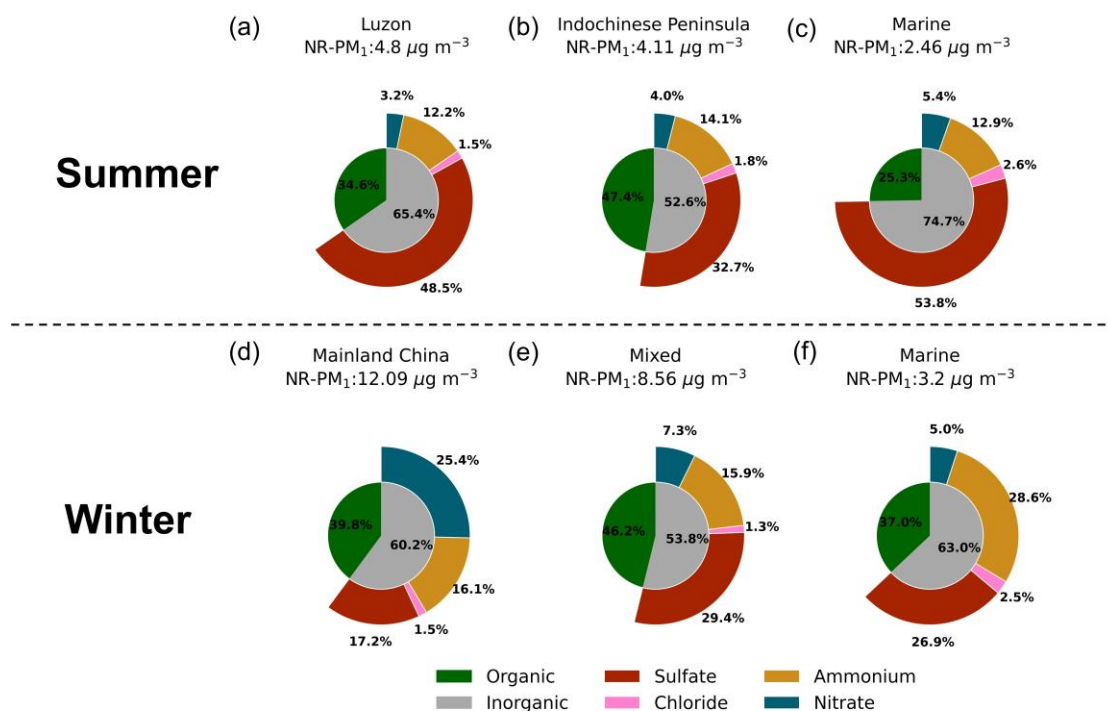


Figure 5. The fraction of NR-PM<sub>1</sub> in “Luzon” period (a), “Indochinese Peninsula” period (b), and “Marine-s” period (c) in summer. The fraction of NR-PM<sub>1</sub> in “Mainland China” period (d), “Mixed” period (e), and “Marine-w” period (f) in winter.

7. Lastly, the discussion on D50 and PNSD for AR and N<sub>CCN</sub> calculations lacks clarity and coherence. Begin by clearly explaining the D50 and PNSD methods used for calculating AR and N<sub>CCN</sub>, detailing how D50 depends on PNSD. Discuss the variations between these parameters comprehensively. Instead of using a single approach, employ various methods to calculate hygroscopicity based on D50, based on the chemical composition, considering both internal and external mixtures. Calculate N<sub>CCN</sub> and AR accordingly and compare these with observations. Refer to previous studies on CCN closure analysis to provide a clear understanding of whether particle concentrations or chemical composition have a greater impact on CCN activity.

**Reply:** We appreciated reviewer's useful advice. In the revision, we use CCN closure method considering aerosol composition and mixing state instead of our origin method to explore the influence of aerosol hygroscopicity in CCN activity (lines 358-388):

*CCN closure study was widely applied to investigate the impacts of different factors on the CCN activity (Patel et al., 2021; Cai et al., 2018; Meng et al., 2014; Deng et al., 2013). In this study, two schemes considering aerosol composition and mixing state based on CCN closure method mentioned in 2.2.3 were applied. The fitting parameter and coefficient of determination ( $R^2$ ) was shown in Table 3 and the fitting plots from two schemes were shown in Fig. S8 and Fig. S9. Besides, the NMB from these two schemes was presented in Fig. 8.*

*In summer, the NMB always lower than 0, which indicated that simulated aerosol hygroscopicity was lower than observed value (Fig. 8). Sea salt which cannot be detected by the ToF-ACSM may account for higher fraction in summer due to low aerosol concentration in summer (Fig. 3c), resulting in the underestimation of aerosol hygroscopicity. The NMB exhibits different trends with changes in SS in “Luzon” and “Indochinese Peninsula” period. Better fitting result appeared in high SS in “Indochinese Peninsula” period, while it appeared in low SS in “Luzon” period (Fig. 8),*

which indicated that aerosol fraction had different trend as particle size increased in these two periods. Besides, "Internal-mixed" scheme had more precious result than it in "External-mixed" scheme in summer (Fig. 8), suggesting the aerosol was primary internally mixed in summer.

In winter, the "External-mixed" scheme always showed a better result than "Internal-mixed" scheme at high SS (0.4% SS and 0.7% SS), indicating that particles in small size were mainly externally mixed. Considering the low hygroscopicity of small-sized particles in winter, it is likely that a significant fraction of these particles consists of externally mixed BC, which probably originated from fresh anthropogenic emissions and remains unmixed with other inorganic salts and organics. As BC ages, inorganic and organic components adhere to it, which would lead to the increase of diameter and particles tended to be internally mixed (Sarangi et al., 2019). This transition resulted in higher hygroscopicity in large-sized particle compared to the smaller-sized particles. Besides, overestimation of aerosol hygroscopicity at high SS could be owing to a higher fraction of non- or less- hygroscopic component (such as organic and BC) at small particle sizes. The predicted  $N_{CCN}$  at 0.1% SS are 10%-20% lower than the observed concentrations, whereas the predicted value at 0.2% SS more closely aligns with the observed concentrations (Fig. 8). It could be owing to the higher fraction of sea salt at larger particle size. However, due to instrument limitations, black carbon and sea salt cannot be detected by the ToF-ACSM. More observations containing sea salt and black carbon are needed in the future to better assess their effects on aerosol hygroscopicity in SCS. In addition, further study size-resolved aerosol composition can also enhance the understanding on CCN activity in the SCS.