

We thank the reviewers for the patient and meticulous review of the manuscript. Following those comments, the manuscript has been carefully revised. We have addressed each of the reviewers' comments. All the modifications and corrections are marked in red in the manuscript text.

Below are our point-to-point responses to the reviewers' comments and suggestions, with the reviewers' comments (RC) in black, **our response in red**, and *the revised manuscript content in italicized blue font*.

## Answer to Reviewer 1

RC1.1: Graphical abstract: make the y-axis on the right same limits for the size distributions and what's the unit? Is the right y-axis same as left y-axis?

**Response:** We confirm that the right y-axis has the same unit as the left y-axis, both representing aerosol NC ( $\text{cm}^{-3}$ ). For the initial manuscript version, we make the y-axis on the right same limits for the size distributions. However, using these same limits failed to clearly demonstrate the characteristics of coarse mode particles; therefore, we adopt the present limits based on the editor's suggestion.

RC1.2: The second panel of graphical abstract: are those correlations for pelagic or offshore? Not clear from the figure

**Response:** These correlations are specific to the pelagic regions. We have explicitly added "pelagic regions" in the second panel of graphical abstract to eliminate ambiguity.

RC1.3: Introduction: line 45: "Hoppel (1979, 1985) studied the aerosol NC and the particle size distribution on the east coast of the United States, and the significant changes in the particle size distribution can be associated with the changes in meteorological parameters and oceanic air mass." . When you say significant changes in PSD, my question is change from what? To what? Did he compare spatial distribution of aerosols? Or what you mean? Please clarify

**Response:** We appreciate the reviewer's request for clarification regarding Hoppel's work (Hoppel, 1979; 1985). In these studies, Hoppel observed increased aerosol NC and the number size distribution with rising wind speeds off the U.S. East Coast. The term "significant changes in particle size distribution (PSD)" specifically refers to shifts toward higher concentrations across the size spectrum under high-wind conditions, compared to low-wind periods.

Meanwhile, we consider that this citation is inconsistent with the theme of this section - "Researchers' reports on aerosol mass concentration and number concentration". Therefore, we have removed this sentence and will cite it in the next section that discusses the influence of meteorological factors on aerosols.

RC1.4: Line 50: "In the Arctic, Leck (1996) reported that the submicrometer aerosol ( $D_p \leq 1000 \text{ nm}$ ) mass concentrations during the International Arctic Ocean

Expedition (IAOE-91) cruise; for instance, the average mass concentration was  $0.76 \mu\text{g m}^{-3}$  over the ocean.:" rewrite the sentence. "reported that" doesn't make sense if you don't follow it with result.

**Response:** We have revised this sentence. Meanwhile, as requested by Reviewer 3, we have streamlined and integrated these paragraphs. The final version is as follows:

*Early observations by Prospero (1979) across multiple marine areas showed notable variations in marine aerosol concentrations, ranging from  $3.34$  to  $8.71 \mu\text{g m}^{-3}$ . Subsequent reported measured data verify substantial regional marine aerosol concentration differences between different ocean areas. In polar regions, submicrometer aerosol ( $D_p \leq 1000 \text{ nm}$ ) mass concentrations averaged  $0.76 \mu\text{g m}^{-3}$  in the Arctic (Leck & Persson, 1996) versus  $3.15 \mu\text{g m}^{-3}$  in the Antarctic (Savoie et al., 1993). In the Pacific Ocean, the  $\text{PM}_{2.5}$  ( $D_p \leq 2500 \text{ nm}$ ) concentration averaged  $12.3 \pm 9.1 \mu\text{g m}^{-3}$  in the Western Pacific (Ma et al., 2022) versus  $140 \pm 48.1 \mu\text{g m}^{-3}$  in the Bohai Sea (Han et al., 2019). In the Indian Ocean, Pant et al. (2009) observed that the average micrometer aerosols ( $500 \text{ nm} \leq D_p \leq 10000 \text{ nm}$ ) mass concentrations were  $8.89 \mu\text{g m}^{-3}$ .*

RC1.5: Rewrite this too for clarity: ". In terms of the Antarctic, Savoie (1993) reported the submicrometer aerosol ( $D_p \leq 1000 \text{ nm}$ ) concentrations, and the mean concentrations were  $3.15 \mu\text{g m}^{-3}$  at Marsh."

**Response:** Please see response to RC1.4.

RC1.6: All your references measure in  $\mu\text{g}/\text{m}^3$ , and Sakerin (2015) 's value is in  $\text{ng}/\text{m}^3$ . I suggest use same units so its easy for the reader to compare

**Response:** Please see response to RC1.4.

RC1.7: ". For the China waters, Kim (2009) found that the average submicrometer aerosol particle ( $10 \text{ nm} \leq D_p \leq 300 \text{ nm}$ ) concentrations were  $4335 \pm 2736 \text{ cm}^{-3}$  over the East China Sea and  $5972 \pm 2736 \text{ cm}^{-3}$  over the Yellow Sea." This reference feels out of place specially when the previous and following references mention units in  $\mu\text{g}/\text{m}^3$ .

**Response:** We appreciate the reviewer's remark on the unit inconsistency when citing Kim (2009). To enhance coherence and clarity, we have added a transition sentence and repositioned the original sentence to prevent potential confusion among readers.

*In the Indian Ocean, Pant et al. (2009) observed that the average micrometer aerosols ( $500 \text{ nm} \leq D_p \leq 10000 \text{ nm}$ ) mass concentrations were  $8.89 \mu\text{g m}^{-3}$ . In addition to aerosol mass concentrations, researchers have also observed aerosol number concentrations (NCs) differences. For instance, in marine regions off the coast of China, Kim et al. (2009) found that the average submicrometer aerosol particle ( $10 \text{ nm} \leq D_p \leq 300 \text{ nm}$ ) concentrations were  $4335 \pm 2736 \text{ cm}^{-3}$  over the East China Sea and  $5972 \pm 2736 \text{ cm}^{-3}$  over the Yellow Sea.*

RC1.8: "All in all, there were some discrepancies in the marine aerosol concentrations and size distributions between the different ocean areas;" what discrepancy? That they

aren't able to come to a common consensus for mass concentration? Or what you mean here?

Response: We sincerely appreciate this insightful comment. The term "discrepancies" specifically refers to discrepancies in aerosol concentrations (both number concentrations and mass concentrations) and size distributions (e.g., particle size distributions) across ocean areas. This is demonstrated by variations between two open oceans (the Arctic and the Southern Oceans), as well as variations between inland seas (e.g., the Yellow Sea) and continental marginal seas (e.g., the East China Sea). These differences are caused by the differences in aerosol production and transport.

These regional differences also highlight the urgent need for targeted studies in specific marine areas - particularly in data-sparse regions such as the South China Sea, which further underscores the value of our present study in this region.

RC1.9: "the major measurement data are relatively outdated and need to be updated." What you mean by outdated? Like previous measured values have changed or science behind them changed? What exactly and why outdated?

Response: We sincerely appreciate this valuable comment. The "outdated" does not refer to changes in the scientific principles behind aerosol measurements, but to the previous measured values.

First, over the past two decades, remarkable changes in aerosol concentrations and compositions have been observed worldwide (Fioletov et al., 2023; Gupta et al., 2022). Zhang et al. (2025) reported pronounced discrepancies in annual mean aerosol concentrations across regions such as Europe, North America, and Southeast Asia using AERONET station data. Critically, over the past three decades, Aerosol Optical Depth has increased substantially in Southeast Asia and Chinese coastal regions (Zhao et al., 2016). However, most of the publicly available in-situ measurement results of marine aerosols in the South China Sea can only be traced back to before 2020 (Kong et al., 2016; Su et al., 2022), and there is a lack of newly updated observational data about recent five years. This long time span makes the existing data unable to reflect the current status of aerosol properties in the region.

Second, rapid urbanization, industrialization, and population growth in Asia have significantly increased anthropogenic aerosol emissions in recent years; these aerosols are transported to the South China Sea, altering the region's aerosol concentration and composition. Global warming has caused variations in wind speed (Zheng et al., 2016) and sea surface temperature (Forestieri et al., 2018; Paulot et al., 2020): wind speed (a key driver of marine aerosol production) and sea surface temperature (which affects sea surface tension, sea surface density, and bubble breaking processes) both influence the marine aerosol generation efficiency and aerosol particle size. Additionally, climate change has led to geographical and annual variations in marine phytoplankton and biological content (Asch et al., 2019), further causing differences in primary and secondary organic aerosols (Chevassus et al., 2025).

In summary, updating in-situ aerosol measurement data for the South China Sea is necessary to obtain accurate and timely insights into the region's aerosol properties.

#### Reference:

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- Zheng, C. W., Pan, J., and Li, C. Y.: Global oceanic wind speed trends, *Ocean Coast. Manag.*, 129, 15-24, <https://doi.org/10.1016/j.ocecoaman.2016.05.001>, 2016.

RC1.10: Line 77: are generation functions and source functions same? In that case use source function. May be cite Gong et al for source function involving wind speed?

Response: The generation functions are same with source functions. We have changed generation functions to source functions. We sincerely appreciate your recommendation of Gong's work. This paper has now cited it in the revised version.

RC1.11: Is SCS analogous to any other place on earth in terms of meteorology? I ask this because studying marine aerosols at a single location which isn't representative of similar areas would make little less sense but if it were to explain (or represent) marine aerosols for larger areas, it would make more sense. What I mean to ask is why is this location important to study?

Response: Thank you for raising this important point. The South China Sea is analogous to some continental marginal seas in terms of geography and meteorology, such as the Coral Sea, the Arabian Sea, and the Gulf of Mexico.

As a continental marginal sea, the South China Sea is located on the continental margin and separated from the open ocean by peninsulas, islands, or island arcs. The aerosol measurement results can clearly reflect the influence of continental air masses in the offshore regions. The South China Sea has large latitudinal and longitudinal range and area (01°12'N-23°24'N, 99°00'E-122°08'E, covering 3.5 million square kilometers). In the pelagic regions, the aerosol measurement results can clearly reflect the distribution characteristics of background aerosols.

Compared with the open ocean, continental marginal seas are affected by topography and land, resulting in clearer changes in wind speed, sea surface temperature, and air temperature difference. This makes it easier for us to identify the relationships between these meteorological factors and aerosol production.

In summary, we can analyze the influences of continental air masses on marine aerosols and the effects of meteorological factors on aerosol production. Most importantly, in-situ aerosol measurement data in the South China Sea region (especially between 10°N and 20°N) are very scarce, and the major measurement data are relatively outdated. Therefore, we have selected this region for our study.

RC1.12: Line 94. It is mentioned that diurnal variations are lacking. My question is why is it important? Add a hypothesis or reasoning why we want to learn about diurnal variations of marine aerosols and what impacts do diurnal variations have?

Response: Thank you for raising this important point. Aerosols are crucial for radiation transfer, cloud microphysical processes, and the climate system. If the diurnal variation of aerosols follows a certain pattern, it can more clearly and directly indicate that aerosol generation and transport are influenced by specific parameters. The discovery of the diurnal variation pattern opens many new questions for future research to elucidate the mechanism underlying this phenomenon and the direct impact of diel cycle of marine aerosol on the radiation balance. Furthermore, on a larger scale, it also involves links to cloud microphysical processes, which in turn relate to energy fluxes and the climate. Ultimately, these insights contribute to the improvement of atmospheric models.

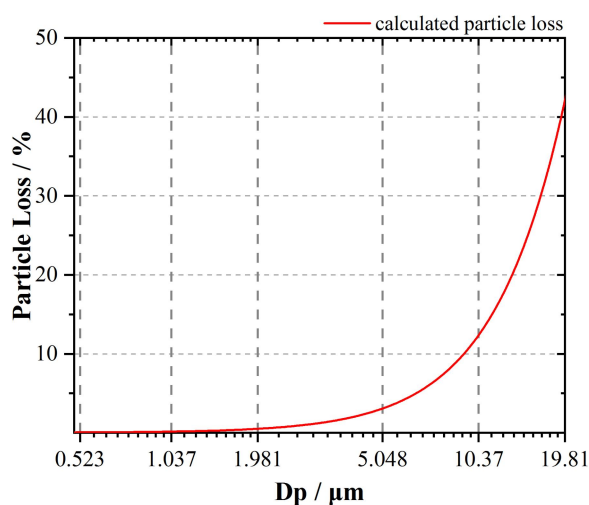
We have revised this sentence to highlight the importance of aerosol diurnal variation.

*The diurnal scale of marine aerosol variation can provide valuable information about their production and transport (Flores et al., 2021), and how these processes are influenced by meteorological parameters. Understanding the diurnal variation is also crucial for improving atmospheric models. Studies on the scale of diurnal variation in marine aerosol remain scarce, and there is an urgent need to clarify the specific connection between these diurnal variation and meteorological parameters to better understand aerosol production and transport.*

RC1.13: Line 119: ". Fig. 1 showed that the particle losses were small in the size range from 0.5  $\mu\text{m}$  and 10  $\mu\text{m}$ . Thereby, aerosol data within the size range of 0.5 to 10  $\mu\text{m}$  were selected for future analysis in this study." You only measured this size range and calculated loss for this range, which is understandable. But saying that this range was selected for future analysis due to a small loss raises a question of whether you know the losses of other size ranges?

**Response:** We appreciate your constructive suggestions. We have redrawn the particle losses for the Model 3321 APS spectrometer across the full size range (0.5-20  $\mu\text{m}$ ) in Fig. 1. The figure revealed a dramatic increase in the particle loss at particle diameters exceeding 10  $\mu\text{m}$ , accordingly we excluded data for particles >10  $\mu\text{m}$  in subsequent analyses. Concurrently, we have revised this sentence to eliminate potential ambiguities.

*We used the Particle Loss Calculator (PLC) to calculate the particle losses for the Model 3321 APS spectrometer in this cruise (Fig. S1) (Von Der Weiden et al., 2009). Fig. S1 revealed a dramatic increase in aerosol particle loss at particle diameters exceeding 10.37  $\mu\text{m}$ . Meanwhile, the accuracy of the aerosol data for particle diameters between 0.5  $\mu\text{m}$  and 10  $\mu\text{m}$ , as measured by the Model 3321 APS spectrometer, had been fully validated in previous studies (Pagels et al., 2005; Peters et al., 2003; Peters, 2006). Thereby, aerosol data within the size range of 0.5 to 10  $\mu\text{m}$  were selected for analysis in this study.*



**Fig. S1** *The calculated particle losses for the Model 3321 APS spectrometer in this cruise.*

RC1.14: I don't think equation 1 is readable. Its all symbols and I think format needs to be changed

Response: Thanks very much for your suggestion. We have revised this equation.

RC1.15: Line 204; avoid using words like drastically. If you use, give a quantitative measure of it'

Response: Thanks very much for your suggestion. We have deleted "drastically".

RC1.16: Line 203: During the shipboard observation period, the average total marine aerosol NC was  $54.01 \pm 35.37 \text{ cm}^{-3}$ , the NC of aerosol accumulation mode was  $52.35 \pm 34.96 \text{ cm}^{-3}$ , and the NC of aerosol coarse mode was  $1.66 \pm 0.83 \text{ cm}^{-3}$ ; do the mean values include/exclude measurements during data events?

Response: The mean values excluded measurements during data events.

RC1.17: Line 205: unnecessary use of 'meanwhile'

Response: Thanks very much for your suggestion. We have deleted "meanwhile".

RC1.18: Rephrase this sentence for clarity: Line 207 ". The shipboard observation data recorded and showed the overall average values and standard deviations of marine aerosol NCs under different temporal and geographical conditions, which were used to compare with the marine aerosol NCs observed."

Response: Thanks very much for your suggestion. We have revised this sentence.

*The shipboard observational data showed overall average values and standard deviations of maritime aerosol NCs under different temporal and geographical conditions. We used these data to compare with the marine aerosol NCs during this cruise period.*

RC1.19: Line 212: "This suggested that the NC of aerosol accumulation mode in the East China Sea might affected by the higher frequency of the new marine aerosol particle" can you cite some studies? Do newly formed particle have high growth rate that they can impact accumulation mode aerosols so much? Cite some studies which have shown the growth of small aerosols in the mentioned region. And studies which show npf is sparse in SCS.

Response: Thank you for your constructive comments. As Reviewer 3 pointed out, the size range (nanometer scale) where new particle formation and growth processes occur is far smaller than the measurement range of the APS ( $0.5\text{--}30 \mu\text{m}$ ). Therefore, the claim that new particle formation causes differences in accumulation mode ( $0.5\text{--}1.981 \mu\text{m}$ ) NC is unreliable and cannot be supported by the measured data. Current studies focus on new particles growing and forming new cloud condensation nuclei (CCN) that are typically 50 to 100 nm across. No study clearly demonstrates that new particles exhibit a sufficiently high growth rate to exert a significant influence on accumulation mode aerosols. Nor is there any study clearly indicating that new particle formation is sparse in the central region of the SCS. Therefore, we



have removed the claim that "new particle formation is the cause of differences in accumulation-mode number concentrations".

Instead, we have provided the reason for the difference in aerosol NCs between the East China Sea and the South China Sea: Aerosol emissions from the Yangtze River Delta region are higher than those from the Pearl River Delta region (Li et al., 2017). Due to the influence of aerosol transport, a greater amount of continental and anthropogenic aerosols from the Yangtze River Delta are delivered to the East China Sea compared to the amount transported from the Pearl River Delta to the South China Sea.

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RC1.20: Line 214: please rephrase this for clarity "Meanwhile, the total marine aerosol NC observed in this study contained the aerosol coarse mode ( $2\ \mu\text{m} \leq D_p \leq 10\ \mu\text{m}$ ) and the part of aerosol accumulation mode ( $500\ \text{nm} \leq D_p \leq 2000\ \text{nm}$ ), and the NC was slightly lower than the marine aerosol NC in the Atlantic by Flores et al. (2020)". Why sudden comparison with Atlantic? Are SCS and Atlantic supposed to have similar concentrations? Or why just compare with Atlantic?

Response: We have revised this sentence.

*The SCS is one of the marginal seas of the Western Pacific. The summed NC observed in this study ( $54\ \text{cm}^{-3}$ ) was slightly lower than NC in the Western Pacific ( $83\ \text{cm}^{-3}$ ) by Flores et al. (2020).*

We have reselected the measurement data from the Flores et al. (2020) experiment, which were collected in the Western Pacific. The South China Sea is one of the marginal seas of the Western Pacific; meanwhile, both our experiment and the Western Pacific experiment in Flores were conducted in spring and located in the tropical zone and westerlies. Therefore, we initially hypothesize that the aerosol number concentrations from the two measurements should be similar.

RC1.21: What seasons did Cai et al. (2020) and Kong et al. (2016) carry their study in? easier to mention, it leaves the reader well informed. Did you compare the rain events between their study and your? is it possible they had less rain which kept the concentrations higher?

Response: We appreciate the reviewer for this useful suggestion. We have added the specific months and seasons of all cruises in Table 2. Specifically, Kong et al. (2016) conducted their observations in Autumn (2012.09 - 2012.10), while Cai et al. (2020) carried out their study in Summer (2018.08). This allows readers to gain a more comprehensive understanding of the research background.

**Table 2**



*Summary of the available study results on the shipboard observation of marine aerosol NC ( $\text{cm}^{-3}$ )*

Region	Time	Season	Latitude	Longitude	Parameter	Value	Parameter	Value	Reference
South China Sea	2023.05 - 2023.06	Spring	21°N - 8°N	115°E - 110°E	Accumulation mode ( $n_{500-2000}$ )	$52.4 \pm 35.0$	$n_{500-10000}$	$54.0 \pm 35.3$	This Study
South China Sea	2018.08	Summer	23°N - 19°N	118°E - 108°E	$n_{400-32000}$	61			Cai et al., 2020
South China Sea	2012.09 - 2012.10	Autumn	21°N - 20°N	118°E - 113°E	$n_{120-10000}$	175			Kong et al., 2016
South China Sea	2005.05	Spring	20°N - 18°N	118°E - 113°E	Accumulation mode ( $n_{50-2000}$ )	$50.3 \pm 19.5$			Lin et al., 2007
East China Sea	2005.05	Spring	30°N - 26°N	122°E - 117°E	Accumulation mode ( $n_{50-2000}$ )	$109.2 \pm 51.8$			Lin et al., 2007
East China Sea	2017.04 - 2017.05	Winter	28°N - 20°N	130°E - 120°E	$n_{250-2500}$	$57.4 \pm 40.9$	$n_{2500-10000}$	$57.5 \pm 41.3$	Ma et al., 2022
Western Pacific	2017.04 - 2017.05	Spring	20°N - 0°N	180°E - 130°E	$n_{100-19800}$	$83 \pm 30$			Flores et al., 2020

*Note.* In the column of the "Parameter", "n" indicated the NC and the subscripts indicated the particle size (nm); in the column of the "Latitude", "N" represented north latitude. The results of this study and these references were the overall average aerosol NCs.

We compared the rainfall events in their studies with those in ours. Kong et al. (2016) reported a conclusion consistent with ours: the marine aerosol NCs increased significantly during rainfall periods. Cai et al. (2020) observed a decrease in marine aerosol NCs during rainfall; the rainfall events were triggered by Tropical Storm Bebinca. During this rainfall periods, the air mass shifted from continental polluted air masses to marine clean air masses; moreover, the typhoon resulted in the removal of air pollutant in Huizhou and in Hong Kong, ultimately led to lower aerosol NCs in their study.

Regarding whether "they have less rain kept the concentrations higher", we cannot directly confirm this conclusion. The key reason is that the average aerosol NCs in our study were specifically calculated by excluding all rainfall periods. However, neither Cai et al. (2020) nor Kong et al. (2016) explicitly stated in their manuscripts whether their reported average aerosol NCs excluded rainfall events or included the entire observation period.

RC1.22: Line 218: "Although the differences in the observation seasons, the study region, and the particle size might influenced the average NC observations, it can still show that the marine aerosol was significantly affected by the continental transport

and the anthropogenic activity in the offshore areas according to the latitude and longitude." Why did you mention differences in study region? The line before this says Cai and Kong did their study in the same region as yours. Next line says differences in study region. Its confusing.

Response: We sincerely appreciate your detailed question. We apologize for the confusing expressions "differences in the study region". Inconsistent expressions between the manuscript can indeed easily cause misunderstanding and confusion for readers. To resolve this confusion, we have deleted the confusing expressions "differences in the study region".

We initially intended to use "the study region" to refer to different marine sub-regions of the South China Sea. For instance, the studies of Cai et al. (23°N - 19°N) and Kong et al. (21°N - 20°N) were conducted in the northern South China Sea, while our study (21°N - 8°N) almost covers the entire South China Sea (including its northern, central, and southern parts). Rather than suggesting that Cai, Kong, and we measured marine aerosols in different ocean areas. Different marine sub-regions of the South China Sea can also be expressed using distinct latitude and longitude ranges.

*Regarding the marine aerosol NCs in the same ocean area, the observations of Cai et al. (2020) and Kong et al. (2016) were significantly higher than the observations in this study. Although the differences in the observation seasons and the particle size might influenced the average NC observations, it can still show that the marine aerosol was significantly affected by the continental transport and the anthropogenic activity in the offshore regions according to the latitude and longitude.*

RC1.23: Line 222: "However, some studies found that the aerosols might be generated on the porous surface when impinged by liquid droplets" porous surface of what? I know the next lines make it clear but it would be better if you introduce the concept when you first mention it.

Response: Thanks very much for your suggestion. We have add the explanation.

*However, intense precipitation events can paradoxically elevate aerosol Ns. Some studies indicate that impaction of liquid droplets on porous surfaces (e.g., the ocean and ship surfaces) may generate aerosol particles (Bird et al., 2010; Joung & Buie, 2015; Zhou et al., 2020).*

RC1.24: Line 229: you mention only one size 4068nm. It's hard to say if ambient aerosols are just of one particular size. Please give a range.

Response: Thanks for the reviewer's comment. The specific range has been added as suggested.

*3786 to 4371 nm*

RC1.25: Line 243: ". High NCs ( $\geq 150 \text{ cm}^{-3}$ ) were observed almost entirely in which the WD were between NW and N that were caused by the high RH accompanied by the rainfall events, and the distributions of NCs were uniform when the wind was blowing in the other directions." So this high NC is the artificial aerosols created by porous surface of the ship and not natural aerosols? Because rainfall is supposed to

cause wet removal or deposition of aerosols and thus decreased concentrations

Response: These high concentrations of aerosol particles include both the artificial aerosols created by droplets impinging on the porous surface of ships and the natural aerosols created by droplets striking the ocean surface.

While wet deposition is indeed the dominant aerosol removal mechanism under most precipitation scenarios, some observations (Bird et al., 2010; Joung & Buie, 2014; Zhou et al., 2020) reveal a different regime where heavy rainfall episodes temporarily enhance aerosol concentrations through secondary production mechanisms. This phenomenon is specifically attributable to the fact that the droplets can release aerosols when they influence porous surfaces (e.g., the ocean surface and ship superstructure), and these aerosols can deliver elements of the porous media to the environment. Hence, after accounting for the observation environment and rainfall intensity, it is evident that short-duration heavy rainfall resulted in numerous raindrops impacting the ocean and ship surfaces, generating aerosol particles. Subsequently, the monitoring instrument captured some of these aerosol particles, ultimately contributing to the increased aerosol particle concentration, which can also be observed in Fig. 3 (the blue-shaded region). Consolidated evidence from prior research and current findings suggests that short-duration heavy rainfall may lead to a transient increase in accumulation mode particles.

We do not deny the mechanism by which rainfall can cause wet removal or deposition of aerosols. What is mentioned in this paper may merely be a special phenomenon, and there is no conflict between the two.

Reference:

Bird, J. C., de Ruiter, R., Courbin, L., and Stone, H. A.: Daughter bubble cascades produced by folding of ruptured thin films, *Nature*, 465,759-762, <https://doi.org/10.1038/nature09069>, 2010.

Joung, Y., Buie, C.: Aerosol generation by raindrop impact on soil, *Nat. Commun.*, 6, 6083, <https://doi.org/10.1038/ncomms7083>, 2015.

Zhou, K., Wang, S., Lu, X., Chen, H., Wang, L., Chen, J., Yang, X., Wang, X.: Production flux and chemical characteristics of spray aerosol generated from raindrop impact on seawater and soil, *J. Geophys. Res.-Atmos.*, 125, e2019JD032052, <https://doi.org/10.1029/2019JD032052>, 2020.

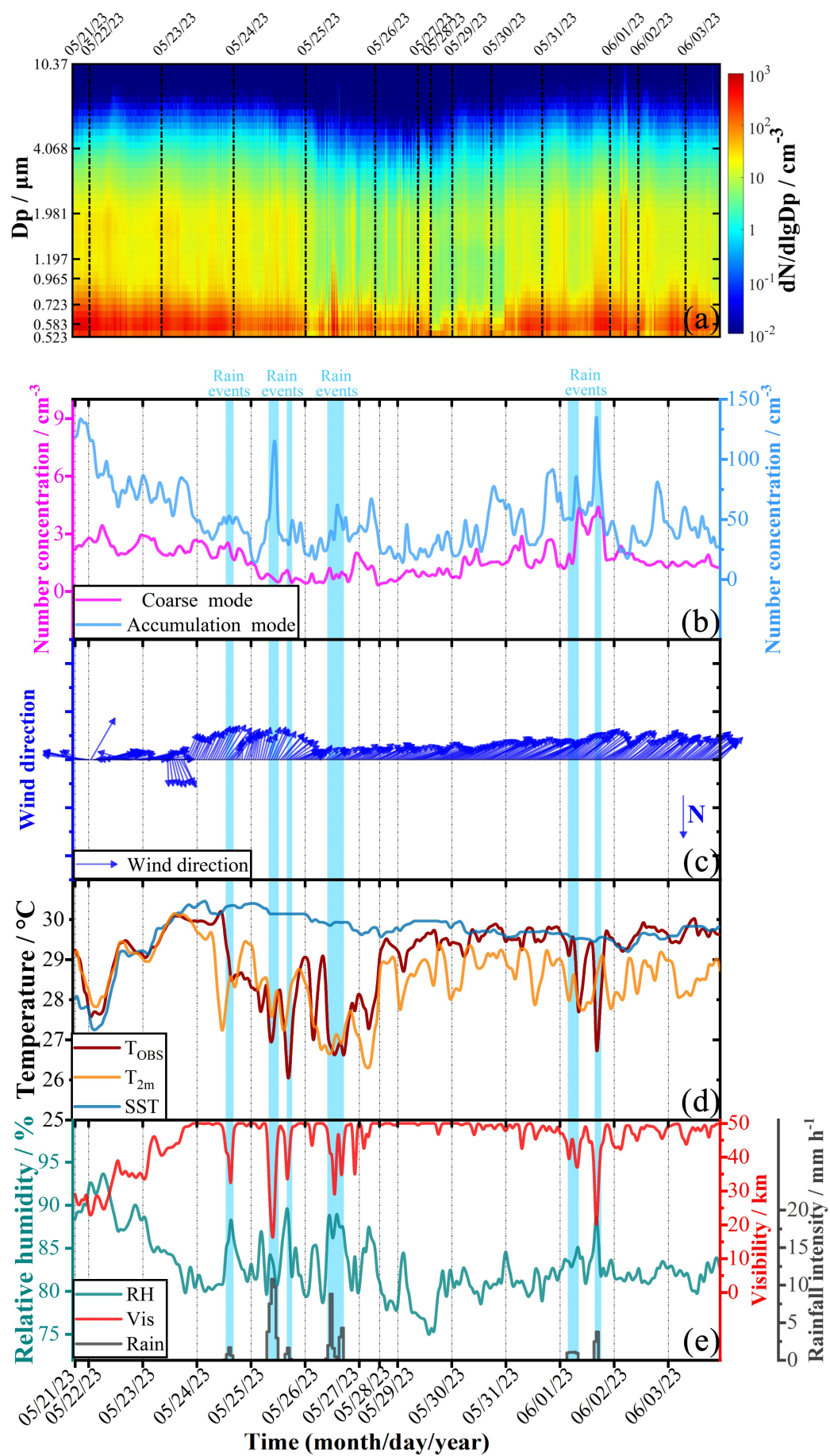


Fig. 3 The time series of the shipboard observations in the SCS from 21 May to 3 June 2023. The blue-shaded regions represented periods affected by rain events. (a) Trend of the aerosol size distributions. (b) Trends of NCs of the two aerosol particle modes (black solid line represented the NC of the coarse mode, and red solid line represented the NC of the accumulation mode). (c) Trend of the WD. (d) Trends of the TOBS (dark orange solid line), T2m (light orange solid line), and SST (blue solid line). (e) Trends in the RH (gray solid line), the VIS (red solid line), and the rainfall intensity (dark blue solid line).

RC1.26: Table 2: when you write accumulation mode, please mention the size, I don't think there's any use of comparing with  $n_{10-400}$  because your measurement starts from 500nm.

Response: We appreciate your suggestion. All marine aerosol NC measurements now specify the particle size ranges covered, and data for the  $n_{10-400}$  category have been removed as suggested.

RC1.27: Line 274:284: include this is methods instead of results

Response: We have relocated the offshore distance calculations to the Section 2.3.3.

### 2.3.3 Distances from the coast

*The ArcGIS path distance method was used to calculate distances from the coast. In equidistant projection, ship positions were used as input data, and coastline position data were used as reference lines for distance analyses. Considering the actual surface distance as well as horizontal and vertical factors, the shortest distance from the ship to the coastline can be calculated.*

The justification for selecting 50 km as the critical threshold for dividing the offshore and pelagic regions in the SCS falls within the scope of the Discussion section; so we have retained this part.

*This marine scientific research campaign started southward from the harbor of Zhanjiang (21°16'21.12" N, 110°23'45.17" E) on 21 May and reached up to the southernmost (8°5' N) point of this cruise on 3 June. In different latitudes of the SCS, there were vastly different marine aerosol distribution characteristics, meteorological parameters, and marine aerosol transport sources. Therefore, we assessed features of marine aerosol distribution at various distances from coast. We conducted real-time analysis of the 72-hour backward trajectories of air masses at the ship's location (Fig. 6a, b). The backward trajectory analysis indicated that the air masses had last passed over continental areas on 22 May 2023, 11:00 local time (LT), at a point 50 km from the coast (the red solid lines in Fig. 6b). Consequently, for all sampling locations within this 50 km boundary, the air masses had directly passed over mainland areas. This meant they carried continental and anthropogenic aerosols that ultimately influenced the aerosol distributions (Braun et al., 2020; Wu & Boor, 2021). For regions more than 50 km from the coast, the backward trajectory results consistently showed that the air masses did not pass over any mainland areas before reaching the sampling site (the blue solid lines in Fig. 6a). The prevailing wind direction was*

*primarily from the southwest (Fig. 3c) in these regions, so aerosols could not be directly transported from the continent to the ship's location. Additionally, continental and anthropogenic aerosols, which were emitted from islands and countries surrounding the SCS, lost their original characteristics through the long-duration (over 72 hours) transport. These aerosols underwent atmospheric long-range transport, dry deposition, wet deposition, and aging processes. Such processes led to the removal of continental aerosols or their gradual dilution and mixing with natural aerosols (Hodshire et al., 2019; Ohata et al., 2016; Xu et al., 2021). Over time, the continental and anthropogenic aerosols transformed or integrated into the background aerosols. Hence, 50 km from the coast was taken as the boundary distance to distinguish offshore and pelagic regions in this study.*

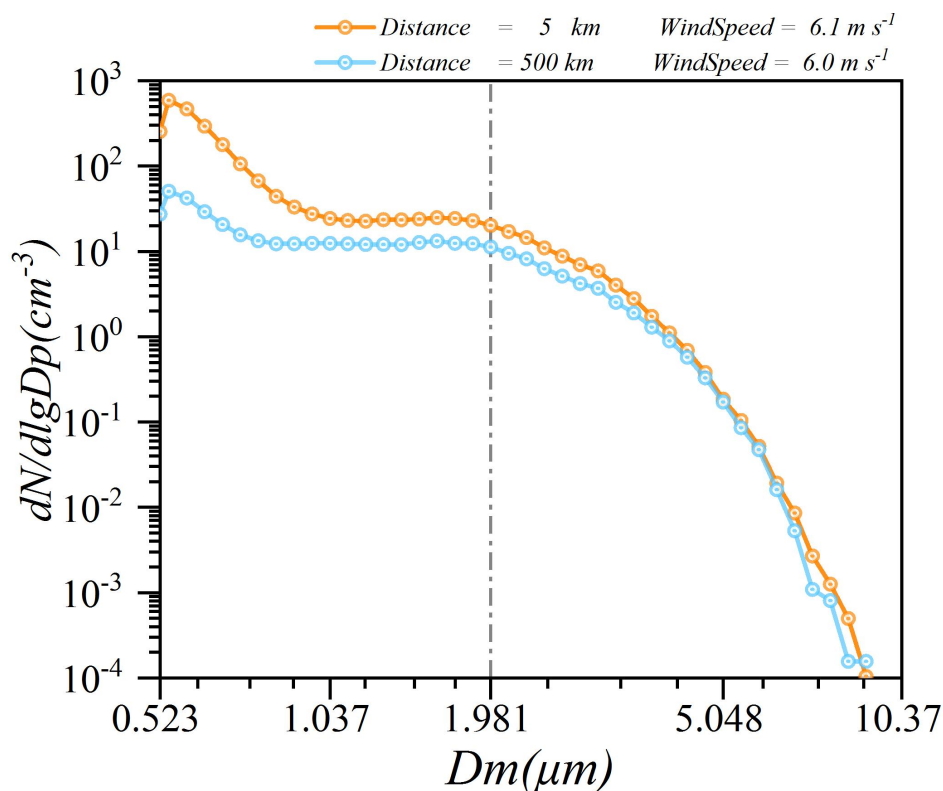
RC1.28: Figure 7b shows same shape of size distribution, of course, the concentrations are different. But doesn't same shape of distribution say same sources for the two regions? If one were affected by continental sources, wouldn't the distributions be slightly different?

Response: We sincerely appreciate the reviewer for the insightful suggestion. The same distribution shape does not indicate that the two regions have the same aerosol sources.

We plotted the number size distributions at the two time points with the largest difference in the distance from the coast (New Fig. 1). When the distance from the coast was 5 km, the measured aerosols were significantly affected by continental sources. When the distance from the coast was 500 km, the measured aerosols were barely influenced by continental sources. Notably, despite the distinct differences in continental source influence between these two regions, the aerosols still exhibited the same number size distribution shape, with only a more obvious difference in concentrations (higher in the offshore regions and lower in the pelagic regions). This directly demonstrates that similar distribution shapes do not imply the same aerosol sources.

Meanwhile, our finding is consistent with previous studies. Specifically, Ma et al. (2022) and Kong et al. (2016), who also conducted observations in the SCS, showed that the influence of continental sources mainly alters the concentration of marine aerosols (by adding anthropogenic particles) but does not lead to significant differences in aerosol distribution shape (within the range of 0.5–10  $\mu\text{m}$ ).





New Fig. 1 The NCs of average size distributions for marine aerosols in different distances.

Reference:

Kong, Y. W., Sheng, L. F., Liu, Q., and Li, X. Z.: Impact of marine atmospheric process on aerosol number size distribution in the South China Sea, (in Chinese), Environ. Sci., 37, 2443-2452, 10.13227/j.hjx.2016.07.005, 2016.

Ma, X., Jing, Z., Chang, P., Liu, X., Montuoro, R., Small, R. J., Bryan, F. O., Greatbatch, R. J., Brandt, P., Wu, D., Lin, X., and Wu, L.: Western boundary currents regulated by interaction between ocean eddies and the atmosphere, Nature, 535, 533-537, <https://doi.org/10.1038/nature18640>, 2016.

RC1.29: Line 298: Can you elaborate this more? ". The marine aerosols decreased slowly with the increasing particle diameters below 1.114 μm due to the transport effect." You didn't discuss the 'transport effect' before, so its difficult for the reader to associate this.

Response: Thanks very much for your suggestion. We have changed "transport effect" to "the influence of aerosol transport", and have also added discussion on "the influence of aerosol transport".

*Due to the influence of aerosol transport, the continental air masses carried continental and anthropogenic aerosols, which ultimately affected aerosol distributions in the 0.5-5.0 μm particle size range. The number size distributions in the offshore regions were obviously higher than in the pelagic regions in the 0.5-5.0*



*μm particle size range. The findings were consistent with the previous studies (Braun et al., 2020; Lorenzo et al., 2023).*

RC1.30: In figure 7c, where is the correlation ( $r=-0.87$ ) shown between Acc NC and distance? All is see is boxplot and distance points in red. Where's the correlation? The distance on 5/31 was higher than 5/27 but still number concentrations were higher on 5/31. How do you explain this?

Response: Thank you for your constructive comments.

As mentioned in the manuscript, the NC of the accumulation mode (indicated by the height of the blue box) showed a decreasing trend with the increase distance from the coast (marked by red dots), which reflected a negative correlation between the NC and the distance. The correlation coefficient was obtained through our calculations. Therefore, in the revised manuscript, we clearly state that the corresponding correlation coefficient between the NC of the accumulation mode and the distance from the coast are derived from calculations, rather than directly observed from the graph.

*It was obvious from Fig. 7c that the NC of the accumulation mode showed a decreasing trend with the increasing distance from the coast, and the correlation coefficient between the NC of the accumulation mode and the distance from the coast was calculated as  $R = -0.87$ .*

On May 31 and May 27, the research vessel was located in the pelagic ocean regions. In the pelagic ocean region, the correlation coefficient between the NC of the accumulation mode and the distance from the coast was very low ( $R = -0.28$ ), indicating that aerosol NCs were less affected by the distance in this regions. Compared with May 27 ( $5 \text{ m s}^{-1}$ ,  $2.9 \text{ }^{\circ}\text{C}$ ), the higher wind speed ( $10 \text{ m s}^{-1}$ ) and lower sea-air temperature difference ( $0.9 \text{ }^{\circ}\text{C}$ ) on May 31 contributed to the higher aerosol NCs.

RC1.31: Your accumulation mode starts from 500nm, how can you show that transport brings in particles which are atleast 500nm for contributing to increase in NC of accumulation mode? Can you cite someone who has shown the size of transport particles in this range? You also mentioned higher wind speed in offshore compared to pelagic areas. How can you say the higher NC at offshore was due to transport and not high WS?

Response: We have added relevant research literature. Braun et al. (2020) and Lorenzo et al. (2023) demonstrated that in the South China Sea, continental air masses (carrying continental and anthropogenic aerosols) significantly influences aerosol concentration distribution within the  $0.5\text{--}5 \text{ } \mu\text{m}$  (i.e.,  $500 \text{ nm--}5 \text{ } \mu\text{m}$ ) size range. Consistent with these findings, our results also show that continental air masses affected the aerosol distributions in the same  $0.5\text{--}5 \text{ } \mu\text{m}$  range, leading to notably higher number size distributions in offshore regions than in pelagic regions.

*Due to the influence of aerosol transport, the continental air masses carried continental and anthropogenic aerosols, which ultimately affected aerosol distributions in the  $0.5\text{--}5.0 \text{ } \mu\text{m}$  particle size range. The number size distributions in*

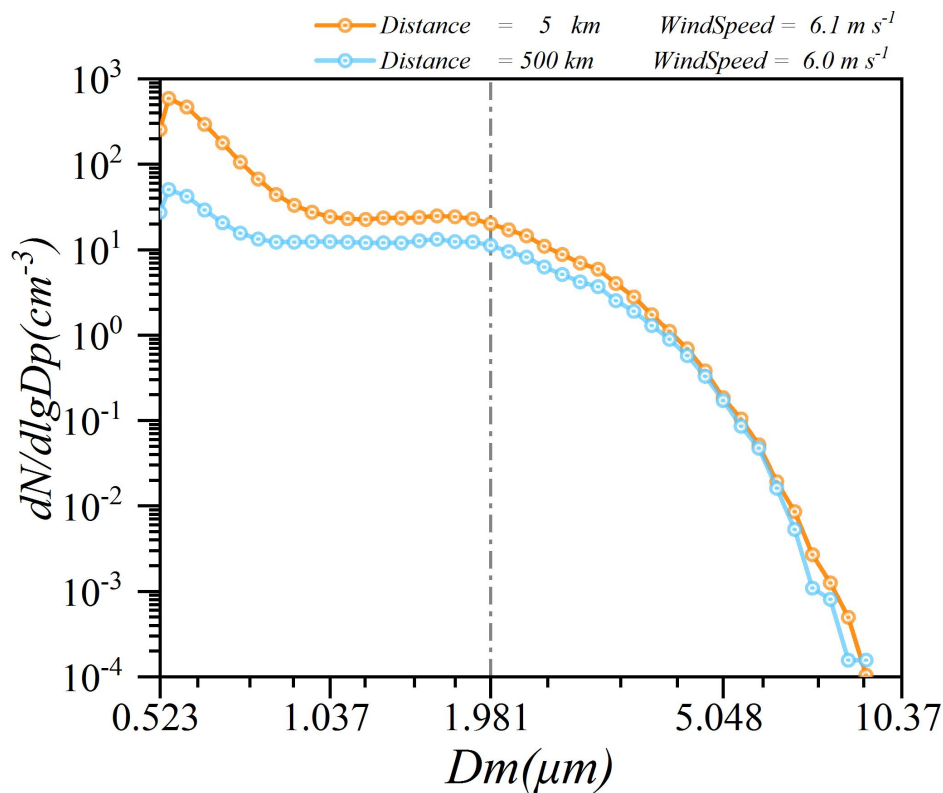
the offshore regions were obviously higher than in the pelagic regions in the 0.5–5.0  $\mu\text{m}$  particle size range. The findings were consistent with the previous studies (Braun et al., 2020; Lorenzo et al., 2023).

#### Reference:

Braun, R. A., Aghdam, M. A., Bañaga, P. A., Betito, G., Cambaliza, M. O., Cruz, M. T., Lorenzo, G. R., MacDonald, A. B., Simpas, J. B., Stahl, C., and Sorooshian, A.: Long-range aerosol transport and impacts on size-resolved aerosol composition in Metro Manila, Philippines, *Atmos. Chem. Phys.*, 20, 2387–2405, <https://doi.org/10.5194/acp-20-2387-2020>, 2020.

Lorenzo, G. R., Arellano, A. F., Cambaliza, M. O., Castro, C., Cruz, M. T., Di Girolamo, L., Gacal, G. F., Hilario, M. R. A., Lagrosas, N., Ong, H. J., Simpas, J. B., Uy, S. N., and Sorooshian, A.: An emerging aerosol climatology via remote sensing over Metro Manila, the Philippines, *Atmos. Chem. Phys.*, 23, 10579–10608, <https://doi.org/10.5194/acp-23-10579-2023>, 2023.

The wind speed in the offshore regions ( $10.7 \text{ m s}^{-1}$ ) registered a 0.2-fold increase compared to that in the pelagic regions ( $8.6 \text{ m s}^{-1}$ ); such a small difference in wind speed (a 0.2-fold increase) could not account for the large difference in NCs (a 1.2-fold increase). Meanwhile, as shown in New Fig. 1, the wind speed was  $6.0 \text{ m s}^{-1}$  at an offshore distance of 5 km and  $6.1 \text{ m s}^{-1}$  at an offshore distance of 5 km. Although the difference in wind speed was extremely small, there were significant differences in aerosol NCs and size distribution. This further indicates that the difference in aerosol NCs between offshore and pelagic regions is more significantly affected by continental transport.



New Fig. 1 The NCs of average size distributions for marine aerosols in different distances.

RC1.32: In Figure 8, please add the arrows showing direction of air

Response: We have added the arrows in Fig. 6.

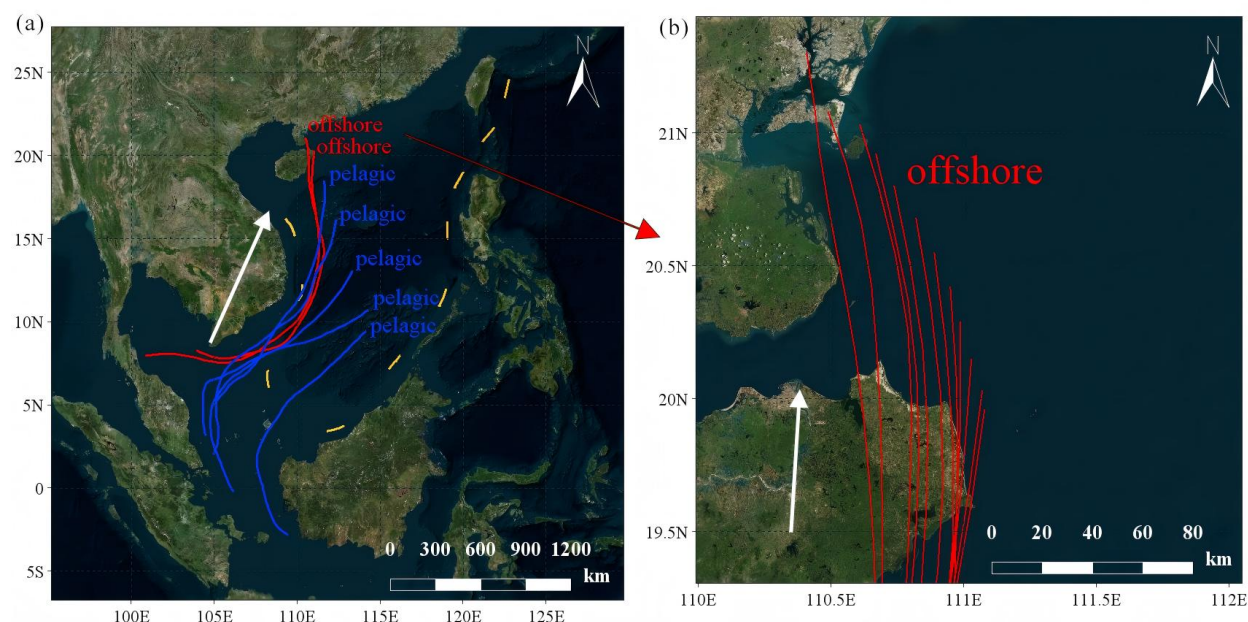


Fig. 6 (a) The 72-h backward trajectory air mass source traces in the offshore (red solid lines) and pelagic (blue solid lines) regions. (b) Detailed map of the backward trajectory air mass source traces passing through the mainland areas (© Google Earth). The white arrows represented the direction of air mass transport.

RC1.33: Line 324: again words like significant doesn't make much sense. Talk in numbers. Out of all components dust shows the maximum change between pelagic and offshore, the difference in the rest of the components do not look 'significant'

Response: Thank you for your helpful comments. We have removed words such as "significant" and instead used specific numerical values for illustration.

Fig. 7d indicated a difference in the distribution of marine aerosol components between offshore and pelagic regions. In the offshore regions, the proportions of dust ( $DUST_{10}$ ;  $D_p \leq 10 \mu m$ ) and sulfate aerosols ( $SO_4^{2-}$ ) were 5.04 % and 1.36 %, which were higher than those in the pelagic regions (1.45 % and 0.97 %, respectively). The higher concentrations of dust and sulfate aerosols further indicate that continental aerosols have influenced the aerosol components in the offshore regions. Meanwhile, in the pelagic regions, the proportions of dimethyl sulfide (DMS), organic carbon (OC), and sulfur dioxide ( $SO_2$ ) were 0.15 %, 1.2 %, and 1.7 %. These proportions were higher than those in the offshore regions (0.1 %, 0.84 %, and 0.56 %, respectively).

RC1.34: Rephrase line 367: Fig 9a showed a clear diurnal variation emerged

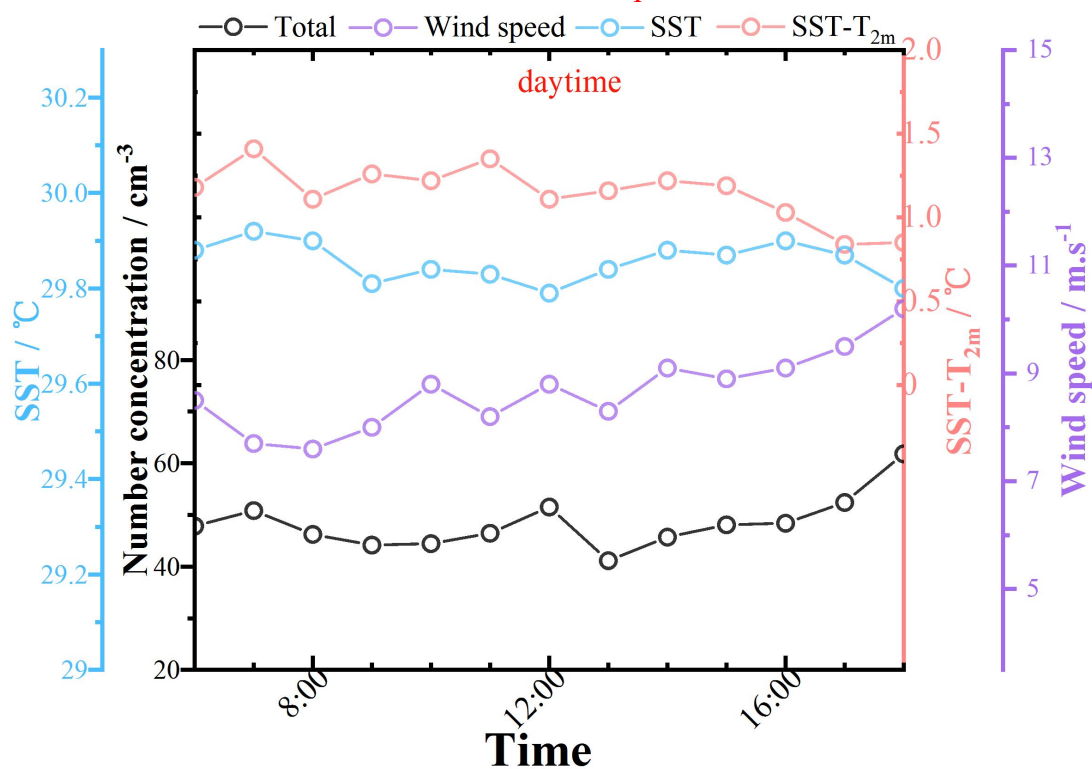
Response: Thanks very much for your suggestion. We have revised this sentence.

*Fig. 8a showed a clear diurnal variation. For the accumulation mode, the variations became readily visible and followed a definite pattern:*

RC1.35: During the daytime, WS increases and SST and difference in temperature between surface and 2m also decreases; so shouldn't the daytime NC be increasing because all the factors you mentioned for the transition period aligns for daytime, yet there's no clear trend in daytime. Why is that?

Response: The aerosol NC, WS, SST, and SST-T<sub>2m</sub> at the end of the daytime were 62.1 cm<sup>-3</sup>, 10.0 m s<sup>-1</sup>, 29.8 °C, and 0.8 °C, respectively. In contrast, the values of these variables at the start of the daytime were 47.8 cm<sup>-3</sup>, 8.5 m s<sup>-1</sup>, 29.9 °C, and 1.2 °C, respectively. As noted by the reviewer, the WS increased during the daytime, while both SST and SST-T<sub>2m</sub> decreased. Notably, the NC exhibited a distinct increase (from 47.8 cm<sup>-3</sup> to 62.1 cm<sup>-3</sup>, a 0.3-fold increase). This increasing trend in aerosol number concentration during the daytime is also observable in New Fig. 2. Furthermore, as shown in Fig. S2 between 14:00 and 18:00, the increase in WS and the decreases in both SST and SST-T<sub>2m</sub> contributed to the rise in aerosol NCs.

The difference in aerosol NC (14.3 cm<sup>-3</sup>) may be difficult to perceive due to the relatively large vertical axis range, which likely led to the reviewer's misunderstanding. However, we chose this vertical axis range primarily to prevent overlapping of the multiple lines (for WS, SST, SST-T<sub>2m</sub>, and NC), thereby making it easier for readers to observe the variation of each parameter.



New Fig. 2 The variations of total NCs, SST, SST-T<sub>2m</sub>, and WS in daytime.

RC1.36: Line 433, add references of studies that showed entrained air decreased with increasing SST. Entrained where? sea surface or boundary layer? clarify

**Response:** We have revised this sentence.

*In the pelagic region of the SCS, consolidated evidence from prior research and current findings suggests that elevated SST likely suppress near-surface air entrainment volumes, consequently decreasing the plunging jets.*

RC1.37: Instead of holding either WS or SST constant for SST-T<sub>2m</sub> correlations, I suggest you perform multi linear regression or lasso regression or any suitable regression analysis to study the impact of these factors on NC variability. When you hold WS const for Fig 13, SST is still varying and when you hold SST const for Fig 14, WS is still varying.

**Response:** Thanks very much for the valuable and insightful comment. We fully acknowledge the limitation pointed out by the reviewer: when we held either WS or SST constant in Figures 13 and 14, the other variable (SST or WS) was still varying. This single-factor control approach cannot fully eliminate the influence of the other variable (SST or WS) on NC. The varying SST or WS may potentially obscure the independent relationship between SST-T<sub>2m</sub> and NC.

However, we would like to emphasize that:

The correlation coefficient between SST-T<sub>2m</sub> and NC reaches as high as  $R = -0.9$  with a very significant p-value ( $p < 0.001$ ) (Fig. 12), and the negative trend is visually striking even with the scattering caused by WS/SST fluctuations. This suggests that despite the variability of WS and SST, the dominant negative relationship between SST-T<sub>2m</sub> and NC is still robustly captured.

The current figures (Fig. 13 and 14) are designed to intuitively illustrate the qualitative trend between SST-T<sub>2m</sub> and NC under WS and SST intervals conditions. We believe that the current figures can already clearly present the preliminary connection between SST-T<sub>2m</sub> and aerosol NC.

We completely agree that multi-factor regression (e.g., multiple linear regression or lasso regression) is essential for quantitatively disentangling the individual and combined effects of WS, SST, and SST-T<sub>2m</sub> on NC variability. Therefore, in the subsequent parameterization modeling, we will prioritize the adoption of these methods to derive a more precise and reliable formula describing the relationship between SST-T<sub>2m</sub> and NC, while accounting for the interference of WS and SST fluctuations. As mentioned in our manuscript, we plan to incorporate SST-T<sub>2m</sub> into the source function in subsequent studies to improve the accuracy of existing source functions.

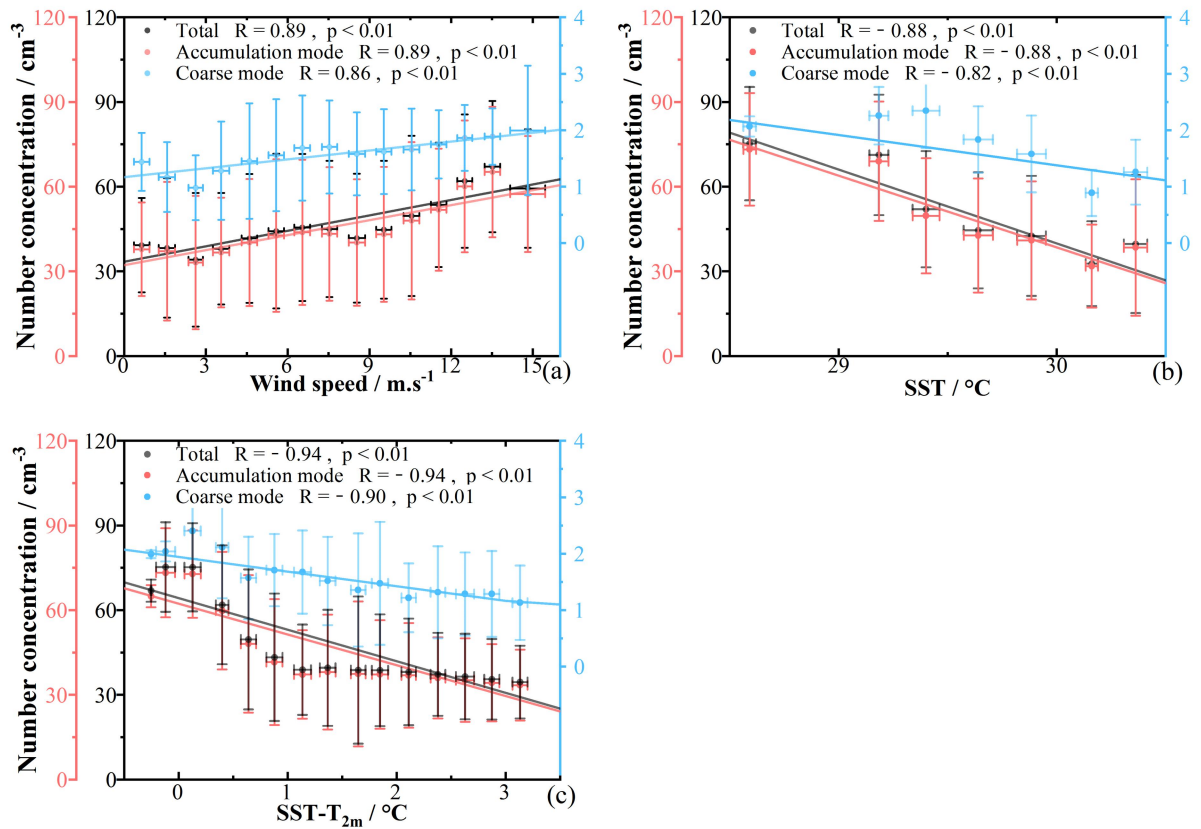


Fig. 12 The relationship between the NC of all aerosol particle modes and WS (a), SST (b), and SST- $T_{2m}$  (c). The error bars represented the standard deviations. The R represented the Pearson correlation coefficients, and the p values were performed to test whether the correlations were significant.

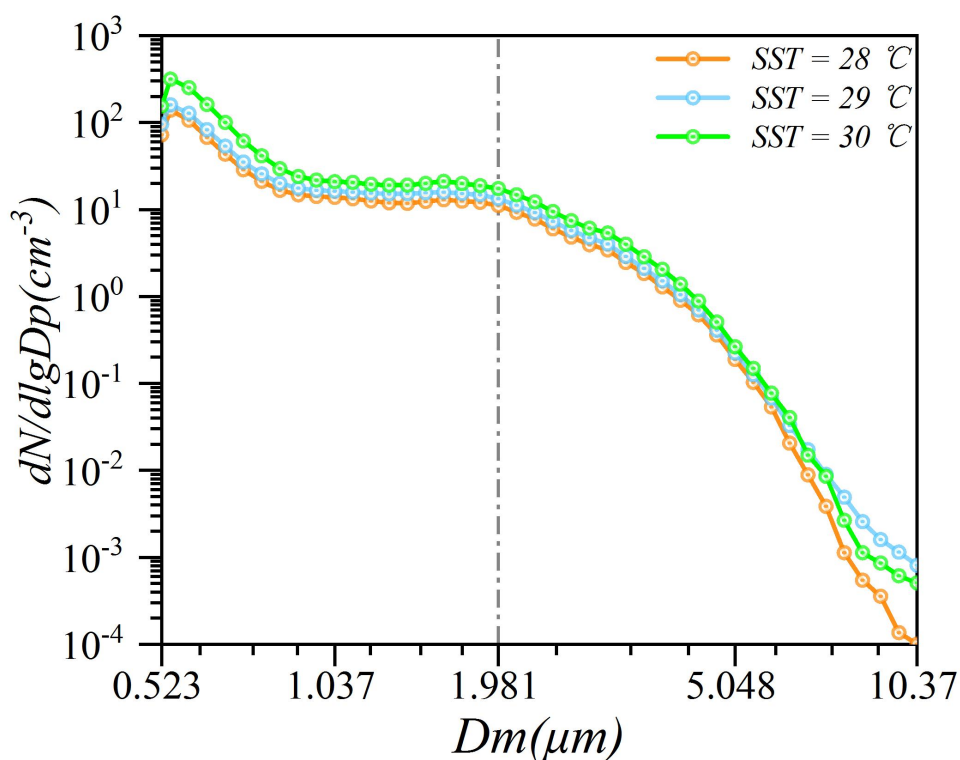
RC1.38: Does the size distribution remain the same with increasing SST? What would you comment on the increase of diameter with warming SST as observed by Saliba (2019)?

Response: Thanks very much for your valuable comments. New Fig. 3 shows the number size distributions of marine aerosols at different sea surface temperatures (SST). Qualitatively, the size distribution almost maintains the same shape with warming SST. Quantitatively, the geometric standard deviation (GSD) shows no significant variation. The mode diameter ( $d_m$ ) slightly increases as the sea surface temperature rises.

We calculated  $d_m$  and GSD of marine aerosol number size distribution under different sea surface temperatures (New Table 1). The  $d_m$  and GSD were obtained by fitting the 0.5 to 5.0  $\mu m$  range of the measured size distribution with a single lognormal mode. The selected size range and calculation method are consistent with those used by Saliba et al. (2019). Importantly, in the South China Sea, we also observed the phenomenon that the diameter increases as the sea surface temperature rises (from 0.564  $\mu m$  at SST = 28 °C to 0.582  $\mu m$  at SST = 30 °C), which is consistent with the findings of Saliba et al. (2019).



This consistency verifies the broad applicability of the SST-dm relationship across different ocean areas, particularly in the tropical marginal sea (South China Sea) that was less studied previously. Moreover, their research conclusions have great significance for understanding marine aerosol production and its radiative impacts in the marine boundary layer. They provide constructive insights for our future research. Specifically, we will optimize the expression of the SST-dm relationship for the South China Sea to better predict marine aerosol concentrations. Ultimately, this regionally optimized expression will be incorporated into the parameterization of future climate models, aiming to improve the prediction accuracy in the western Pacific marginal seas.



New Fig. 3 The NCs of average size distributions for marine aerosols in different sea surface temperatures.

**New Table 1** Summary of mode diameter of the marine aerosol number size distribution ( $d_m$ ) and geometric standard deviation (GSD) within different sea surface temperatures.

SST (°C)	$d_m$ ( $\mu\text{m}$ )	GSD
28 °C	0.564	1.137
29 °C	0.570	1.145
30 °C	0.582	1.266



Reference:

Saliba, G., Chen, C.-L., Lewis, S., Russell, L. M., Rivellini, L.-H., Lee, A. K. Y., Quinn, P. K., Bates, T. S., Haëntjens, N., Boss, E. S., Karp-Boss, L., Baetge, N., Carlson, C. A., and Behrenfeld, M. J.: Factors driving the seasonal and hourly variability of sea-spray aerosol number in the North Atlantic, *P. Natl. Acad. Sci. USA.*, 116, 20309-20314, <https://doi.org/10.1073/pnas.1907574116>, 2019.