

Review of “Diurnal variability and controlling mechanisms of marine aerosol distributions over the South China Sea: Insights from shipborne observations”
Qiao et al.

This study reports on aerosol properties measured using a TSI aerodynamic particle sizer (APS) and their relationship to environmental parameters measured on a 1-month cruise in the South China Sea and from reanalysis data. The paper aims to characterize the differences in aerosol number concentrations and size distributions between coastal and open-ocean regions and the implications transport has on the variability of modal concentrations. They argue that areas more proximate to the coast have higher aerosol number concentrations. While offshore, wind speed, SST, and the difference between SST and 2-m temperature (SST-T2m) influence aerosol concentration positively (wind speed through mechanical generation) and negatively (SST and SST-T2m through changes in buoyancy and bubble viscosity).

Many of the claims and arguments presented in this work are unsupported or they have been given with vague descriptions and unclear relationships to previous literature. Because this is an interesting dataset in an under-sampled region and some interesting results are obscured within this study, I feel that instead of a full rejection, this paper should undergo substantial major revision before being considered appropriate to be considered for publication. Below I have provided detailed list of major concerns and many technical comments, however many editorial issues persist throughout. I recommend the authors employ these corrections to improve this paper.

Main Comments

- The writing in many places is very hard to read due to grammatical errors and the use of frequent platitudes making the paper very difficult to follow. In some places I noted where these were and made suggestions, but the issues were far too numerous to point out each one. Many details are missing, and the discussion is vague without specific directed attention to the very detailed figures or putting the results in context with the broader literature context or studies within this region. The authors should make a concerted effort to carefully re-read the paper to ensure its clarity.
- In many cases, there is an incorrect or superfluous use of the article “the”.
 - Examples:
 - Line 70: “[the] aerosol generation and transport...”
 - Line 71: “...found that [the] marine aerosol...”
- There was also the use of “meanwhile” and “on the other hand” when the authors were trying to further describe findings or procedures. In many, if not all, of the cases that either of these phrases were used they were unnecessary and confound clarity. I recommend the authors remove these phrases.
- The authors should use words like “the “correlations can explain”, rather than declarative statements on correlations and other relationship because many of

their claims are based on mostly visual comparisons and not substantive correlations or quantitative analyses.

- It is the opinion of this reviewer that the word “production” or “emission” be used in place of “generation” when describing the marine sources of aerosol. In the way “generation” is used in this study, the authors are describing wind-generated aerosol production/emission, a more canonical use of the word. Either choice of terminology should be used consistently throughout the text.
- Citations in the text and references list should be checked for correct formatting. In many cases the authors have only listed the first author of papers rather than conforming to the proper citation style of this journal. This should be corrected.
- Abstract
 - The text in the graphical abstract is very difficult to read because the resolution is very poor.
- Introduction
 - The introduction lacks a clear narrative of the research problem and does not provide proper context that motivates the research questions. The current state of the introduction makes it hard for the reader to follow the motivation of this work. The authors should make sure to highlight what relevant prior has been that either supports or motivates why this work has been done. There is very little attention given to introducing the region, the region’s sources of aerosol, and how this work will effectively fill in the gaps based on substantive research questions.
- Some comments on the methods of this work
 - Were the aerosols dried before being sampled? Are you sampling rain drops thus leading to some of the effects presented? Prior work has often shown (e.g. (Petters et al., 2006; Zheng et al., 2018)) precipitation acts as a large sink for accumulation and large accumulation-mode aerosol. I’m not sure I follow or buy the conclusions about wave droplet induced increases in accumulation-mode particles presented in this work based on the available observations.
 - Do the authors use aerosol composition from MERRA-2 in a similar size range of the APS? Many of the MERRA-2 aerosol species will have some non-negligible contributions from particles <0.5 μm that cannot be explained with changes in the APS alone.
 - Data screening: “Nucleation events” (Line 185 and in other passages) in the marine environment from new particle formation or otherwise are observable typically at sub-100nm sizes (in the nucleation, Aitken-mode range) and stop growing well below 500 nm. These are not measurable with an APS. How are the authors able to justify that such events can be

adequately observed with their measurement limitations? I am not confident that a number concentration criteria for screening ship pollution or continental influence would be meaningful from an APS alone due to it measuring mostly coarse particles. Plots and discussion of such a justification should be provided so that the reader is confident that there is fidelity in such a screening. Further, new particle formation is not discussed as a potential effect or limitation on the findings of this work and this should be included.

Technical/Editorial Comments

- Abstract, Line 17: I believe “increase” would be a more appropriate word here than “elevation”; “...a 120% [increase] in offshore aerosol number concentrations...”
- Abstract, Lines 26-27: This sentence is written vaguely and should be improved for clarification. Are the authors saying that the results of this work provide evidence to support differences in the spatial variability of marine aerosol in the SCS, and further, these results can help improve understanding of production and transport? The authors should rework this sentence to ensure these points are coming across clearly.
- Line 46: I would revise the latter part of this sentence as, “...and climate change, [there has been an increasing research focus on marine aerosols over the last] forty years.”
- Line 47: “NC” is not defined before its first use here.
- Lines 47-49: The summary of findings in the Hoppel (1979, 1985) studies presented here are insufficient and vague. What do the authors mean by “associated with changes in meteorological parameters and oceanic air mass”? What meteorological parameters?
- Lines 49-57: This passage is not written clearly. The authors are attempting to provide a survey of mass concentration differences between different marine regions, but they are listed in a disjointed manner that makes it difficult for the reader to understand. I would recommend that this passage be shortened and combined in a way that illustrates the differences in reported aerosol mass in the different regions. Please also be cognizant of consistent unit usage, e.g. use $\mu\text{g m}^{-3}$ as it is the most common of your reported masses.
- Lines 57: Revise “For the China waters, ...” to “In marine regions off the coast of China,...”
- Lines 60-63: The authors should clarify what is meant by “discrepancies in the marine aerosol concentrations and size distributions.” Do the authors mean “differences”? The ending of this sentence also seems like a bit of a tangent and

is not explained, (“especially from 10degN-20degN...” Why is this included and what is its relevance to the rest of the passage?

- Lines 63-65: I think a more substantive take away from this paragraph is that shipboard measurements provide better spatial (and temporal) context for aerosol measurements in diverse marine regions such as off the coast of China and they can further help improve characterizations by being updated. Can the authors please revise the sentence they have written in these lines to better convey that?
- Line 66: The sentence is written in a way that conveys a finding. If so, the authors need to provide support. I believe the authors are actually saying, “Aerosol generation and transport [can lead] to differences in marine aerosol concentrations and size distributions.” Is that correct?
- Line 67: Clarify “aerosol components.”
- Line 68: Are “weather events” synoptic weather patterns, mesoscale weather events, storms? Please clarify.
- Line 71: The authors have written “et al.” after “relative humidity (RH).” Do they mean “etc.”? “et al.” or “etc.” is not appropriate here. If there are additional pertinent meteorological parameters to include, please list them.
- Lines 71-75: The description of Tang RH effects on marine aerosol and the overall implication from the studies in the following sentence are not connected through the same logic. The authors are correct to note that RH changes can affect aerosol size through deliquescence and efflorescence, however, this is not related to the “wet deposition and dispersion” mentioned later. The RH effects on size come into play when the aerosol are being sampled, so if mentioning this point is to say that the previous work was inconsistent with their sampling procedures (drying, heating) and that makes comparison and aerosol characterization difficult, then that is what should be discussed here. Because the authors are instead discussing effects on generation and transport evidence of that effect should be discussed and not the Tang result.
- The reference for Tang et al. (1997) is not properly cited in the main text or reference list. In the reference list, the authors have only provided the name of the first author.
- Lines 75-78: The authors should specify what the relationships are between aerosol generation and wind speed from the studies cited. Is it the total aerosol concentration, are they size dependent?
- Line 79: “[They] explained that the SST ...” Please clarify what or who is meant by “They.”

- Lines 79-81: This is an inadequate summary of SST effects on aerosol properties. Please be specific based on the studies the authors have cited.
- The authors have made no mention of aerosol generation in the free troposphere (e.g. from new particle formation). This can have important implications for aerosol properties measured in the marine boundary layer and cannot be ignored in the discussion of important generation and transport drivers. Further, do they think, based on previous studies in this region, that new particle formation has any effect on the observed relationships?
- Line 66-88: Although I have provided rather detailed guidance on this entire paragraph, I believe the authors should completely rework this section. Very vague statements and disjointed thoughts persist throughout. The authors cite lots of work but don't use any of these citations for clear contextual support.
- Line 89: Please revise, "...most [observational] data..."
- Line 89-96: I believe the authors should remove this passage up to the start of the motivation sentence, "To address these, ..." Everything prior is not needed and is discussed in the previous paragraphs.
- Line 102: Remove the semicolon after "respectively." This should begin a new sentence.
- APS data: do the authors use all of the channels in 0.5-10 μm diameter range? I believe previous work has shown that the first channel in the APS has issues with counting efficiency and sizing accuracy. Can the authors please provide clarification on if this channel was used and justification for why it is appropriate to use here?
- Line 119: delete "future."
- Reanalysis data: For the atmospheric dynamic/thermodynamic properties (temperature, wind speed, etc...) was the ERA5 reanalysis, MERRA-2, or a combination of ERA5 and MERRA-2 used? The authors discuss ERA5 in the first part of Section 2.3.1., but then say, "Meanwhile, the MERRA-2 was..." Why were two different datasets used for these variables? Why not use only MERRA-2 or ERA5? What motivated using two different datasets? I understand using MERRA-2 for the aerosol mass concentrations. Additionally, the authors should justify why they believe the coarse resolution of these datasets compared to the in situ cruise data are representative of the conditions measured where the ship is. Also, please provide a citation for MERRA-2 and spell out its acronym on the first use.
- Line 203: The authors need to define the size ranges used for their quantification of accumulation and coarse modes.

- Lines 202-203: I don't believe that it is appropriate for the authors to define aerosol number concentration integrated from the APS as the "total marine aerosol" or even the "total aerosol." A large portion of marine aerosol number concentrations come from substantial sub-500 nm particle contribution. As such, it would be much better suited if the authors revise this terminology; e.g. "APS integrated NC", "summed NC".
- Table 2: define the size range for "accumulation mode." Clarify in the caption that these are shipboard measurements or please specify the observational platform.
- Lines 206-207: The sentence "Due to the constraints..." is not necessary and should be removed.
- Lines 207-209: Delete "data recorded and" in the sentence "The shipboard observation data..."
- Lines 211-214: I don't understand how the authors came to these conclusions or what evidence is being used to support these claims of new particle formation being the cause of differences in accumulation-mode number concentrations. As I mentioned in one of my main comments, new particle formation and growth occurs at much smaller sizes than what is measured by the APS. The authors also do not provide any literature support for how they can argue this claim based on their measurements. Additionally, there is no discussion prior to or proceeding this sentence about westerlies and what that would mean for aerosol NC changes.
- Lines 213-216: I don't understand much of the discussion here or how it relates to the citation from the Atlantic. Did the Atlantic study measure the same size range? How can this study be used for comparison without mentioning these specific differences?
- Line 248: replace "region" with "range" or "bin."
- Figure 6: were these plots created using a fixed wind direction, relative humidity, precipitation, or other controlling factor? If not, how can the authors argue, especially based on the apparent low correlation and large scatter of the data, that wind generation is the primary mechanism for driving variability of this mode?
- Figure 7a: What do the different colors of the boxplots represent? If they are offshore and pelagic, you should use the same color scheme throughout the whole figure.
- Figure 7a: are the differences in pelagic and offshore number concentrations statistically significant for each mode and their sum?
- Lines 295-296: How does the bimodality of the distributions reported here compare to previous literature? Given the counting uncertainty in the lower bin of the APS,

I'm not sure I believe true bimodality is being observed here, nor do I believe it will be comparable to prior reports of bimodal marine size distributions such as in Hoppel et al (1986).

- Line 297: Please clarify what is meant by the aerosols were “evenly distributed in the 0.835 to 1.981 μm particle size range.” Later in the text this term “evenly distributed” is mentioned again. It should be replaced with something more specific.
- Line 298: Where do the authors describe a “transport effect” on the size distribution below 1.114 μm ? Please clarify.
- Line 301-305: Is this discussion only about the accumulation mode, coarse mode, or sum? Please specify.
- Lines 303-305: Were only 2 data points used for the offshore correlation? In Figure 7c, there are only 2 dates and two bars pertaining to offshore. 2 data points are not sufficient for a correlation. Did the authors use all data points for those days or just the average in the bar charts? Please clarify (1) what was used for the correlation and (2) what data is being shown in figure 7c; the caption says “diurnal variations” which is very vague.
- Line 310: “meteorological element distributions” is a very confusion description. This should be revised to “meteorological parameters” as in the table header.
- Line 310: The authors say the meteorological parameters are “significantly different” between offshore and pelagic areas. Based on the means and standard deviations this does not appear to be the case as the absolute differences are within only a few percent between the areas. The authors should please explain this claim and provide statistical evidence to support it.
- Lines 312-313: “In addition to the WS influence, the frequency...” Where is this shown?
- Lines 314-325: This passage and its discussion of effects on the aerosol is exceptionally inadequate. Significant jumps to conclusions are made throughout. (1) are the aerosols emitted from Guangzhou and Hainan and the “islands and countries surrounding SCS” expected to be observable in the size range of measurement of the APS? What evidence is there to support this? (2) “...underwent atmospheric transport, transformation, and deposition processes...” is very vague and not an appropriate claim based on the available measurements and analysis of this study. Please provide specific description of processes that the authors think the aerosol experienced that can explain the differences.
- Line 325-327: The authors need to justify that the dust and sulfate aerosol are representative of continental aerosol sources by providing citable studies.

- Lines 327-330: Have DMS, OC, and SO₂ been shown to be in high concentrations in pelagic regions of the SCS? The “degree of [...] marine biological activity” is alluded to in the following section (Lines 355), but nothing related to this is discussed and how it might explain the differences between pelagic and offshore regions.
- Line 357: “...the meteorological parameters had obvious day-night differences.” The word “obvious” should be removed here and replaced with “it is expected that there are diurnal differences.” The differences are not “obvious” because they have not yet been shown.
- Line 359-363: The threshold of 120 cm⁻³ is not comparable to the Saliba et al. (2019). In that study the condensation nuclei concentration (particles >10 nm) was used, while this study is using mostly large accumulation and coarse mode aerosol. Please clarify the discrepancy and justify this choice of threshold. Were other thresholds tested and what support is available to make this choice?
- Lines 364-367: Please specify the hours used for each time.
- Lines 367-383: I see no “clear diurnal variation” in Figure 9. Figure 9a shows a very minimal increase in mean accumulation mode aerosol and no change in coarse mode. The plots in 9c show basically similar medians with interquartile ranges that are nearly identical for each mode and their respective time periods. Have the authors tested if these differences are statistically significant? Again, differences in the mean concentrations here seem to vary by only 1-5%. What are the differences observed in the size distributions of Figure 9b? These are not discussed clearly in the text and as a reader I see no real changes. These should be quantified as a change in peak diameter, width, number, etc.
- Lines 428: the values are “more negative” not “smaller than” -0.75.
- Section 3.3.2. First Paragraph (SST influence): the authors should comment on the fact that the correlation found here for aerosol concentrations and SST occur for a very small range of SST of about 1-2 deg C. This is likely much smaller than the field and laboratory studies used for comparison. Do the authors think this has any effect on the observed correlations/slopes and the claims the authors make about entrainment and density changes that influence aerosol number concentrations? Terms like “daughter bubbles” are not described and make this discussion confusing. Please clarify this discussion for readability.
- Lines 432-433: Please clarify what is meant by “the influences of the SST on the NCs might be different in different seas due to the different components of the seawater.”
- Line 433: “according to the results of the previous studies” What studies?

- Section 3.3.2. Second Paragraph (SST-T2m): The authors spend quite a lot of time making declarative statements about what's influencing the SST-T2m relationship to the aerosol concentration based on previous work. For such declarative statements, similar analysis exercises need to be carried out. They declare that SST-T2m was the "major determinant of atmospheric stability" which led to the "upward transport" of marine aerosol in the boundary layer. Other such declarative statements are made further in the paragraph, but no such results are shown. If the authors don't mean to declare such factors definitively describe their observations, they should be careful to instead place their findings in context with prior work rather than discuss with certainty.
- Line 496: Did the authors use an anomaly for SST-T2m or is it the difference between SST and T2m. Please clarify.
- Line 501-502: The authors mention "rapid solar radiation shifts" that drive changes in the aerosol concentrations. What is meant by this? Do they mean just day night differences? Please clarify as this is not discussed prior.

References

HOPPEL, W., FRICK, G., and LARSON, R.: EFFECT OF NONPRECIPITATING CLOUDS ON THE AEROSOL SIZE DISTRIBUTION IN THE MARINE BOUNDARY-LAYER, *Geophysical Research Letters*, 13, 125-128, 10.1029/GL013i002p00125, 1986.

Petters, M., Snider, J., Stevens, B., Vali, G., Faloona, I., and Russell, L.: Accumulation mode aerosol, pockets of open cells, and particle nucleation in the remote subtropical Pacific marine boundary layer, *Journal of Geophysical Research-Atmospheres*, 111, 10.1029/2004JD005694, 2006.

Zheng, G., Wang, Y., Aiken, A., Gallo, F., Jensen, M., Kollias, P., Kuang, C., Luke, E., Springston, S., Uin, J., Wood, R., and Wang, J.: Marine boundary layer aerosol in the eastern North Atlantic: seasonal variations and key controlling processes, *Atmospheric Chemistry and Physics*, 18, 17615-17635, 10.5194/acp-18-17615-2018, 2018.