

**Response to anonymous referee #2's interactive comment on the manuscript "Atmospheric aerosol compositions over the South China Sea: Temporal variability and source apportionment" point by point below.**

The manuscript presents results from observations of Total Suspended Particulate matter (TSP) from an approximately year long study in a marine region of the northern South China Sea (SCS). Four-day filters were collected and analyzed for major inorganic chemical ionic concentration observed in the marine boundary layer. Several source apportionment methods were then used to both differentiate between aerosol types that contributed to measured values and link them with potential sources. These included correlation between various factors, principal component analysis, positive matrix factorization, and backtrajectory analysis by means of concentration weighted trajectories for various identified sources. The results are interesting and provide new findings that contribute to knowledge of aerosol impacts on the northern SCS. However, the manuscript lacks a clear justification of the basis for the study or description of the implications of its findings. In addition, further information on the methods used is needed in order to fully ascertain how the study was conducted and if the findings are justified from the measurements and information described. I therefore recommend that the authors conduct major revisions to the manuscript to focus on providing more reasoning behind why the study was conducted and what conclusions regarding marine aerosol impacts are justified from TSP observations. In addition, the manuscript should be reorganized to more clearly link each part of the methods, results, and conclusions to the overall purpose of the study.

**General Comments:**

1. The authors utilized only measurements of ionic concentrations of TSP and meteorological information to investigate aerosol sources and impacts on a remote marine region. This provides interesting information, but as filter collections of TSP can be dominated by larger aerosol particles, the authors should briefly discuss the limitations of such measurements in comparison to size resolved measurements.

**Response:** Thank you. The term "total suspended particulate" (TSP) has referred in principle to the mass concentration of all particles considered to be airborne. Samplers for "total aerosol"

first emerges during the time when it is widely thought that it is sufficient to simply draw an aerosol sample through an inlet and to collect the particles on a filter. When sampling is required for health-related purposes, we now know that such sampling needs to be carried out with respect to specific particle size fractions.

2. The methods section 2 did not contain enough information to fully describe how the study was conducted. While the cited studies are helpful and needed, at least a basic description of each method, along with the specific parameters of the method used in this study are needed. Specific examples are included in the specific comments.

**Response:** Thank you for your suggestion. We have added more information in section 2, and supplementary text S2 and S3.

3. Several sections of the results are repetitive, and some of the sections contain information that would be more helpful to the reader by including it in the methodology section before the results are presented. Streamlining the results section to more clearly describe the results first, followed by a discussion of their implications might allow the reader to follow the logic of the study better. The results section in particular could be better organized in a way such that the results directly lead to descriptions of the findings. Specific examples are included in the specific comments.

**Response:** Thank you for your suggestion.

We have streamlined the manuscript from 19 pages to 15 pages, excluding references. We have deleted Lines 50-65, section 3.3.1 and 3.3.2, and other sentences in pre-revised manuscript. At the same time, we reorganized the manuscript in some sections and described the results first, followed by a discussion, and moved some information into supplementary text.

4. The authors might consider a more thorough description of the justification for the study, and specifically why TSP measurements are appropriate for identification of source types impacting remote marine regions of the SCS. The reason for inclusion of some parts of the manuscript was not immediately evident, and did not always lead to a coherent storyline or scientific narrative. A more concise description of what was conducted and why would alleviate

many of these concerns.

Response: Thank you for your suggestion. We have revised them in manuscript. Such as Lines 19-21 and Lines 65-69:

In order to evaluate impacts of different source emission on marine atmospheric particles over the South China Sea (SCS), major inorganic ionic concentrations ( $\text{Na}^+$ ,  $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{K}^+$ ,  $\text{NH}_4^+$ ,  $\text{NO}_3^-$ ) were determined in total suspended particulates (TSP) at Yongxing Island from March 2014 to February 2015.

However, the observational data on aerosol chemistry over the SCS are very sparse (Xiao et al., 2015). To get better understanding of potential sources, source contributions, and spatio-temporal variations of marine aerosols over the SCS, total suspended particulate (TSP) were continuously collected at Yongxing Island from March 2014 to February 2015.

**Specific Comments:**

Line 30. It is not clear what “74% and 82%” are referring to in the abstract. Similarly, percentages in the rest of the abstract should be clearly described.

Response: Because we re-calculate the PMF model, some values have been changed. We revised the sentence in Lines 31-32.

L 43-45. Organics also constitute an important source, and should be mentioned even if measurements were not conducted.

Response: Yes. Thank you. We have added these information to the revised manuscript in Lines 47-49.

L 70. The description of “warm” and “cool” periods of the monsoon are not clearly linked to the description of the Boreal seasons.

Response: Yes.

In the manuscript, we separate the seasons based on the primarily air masses directions. In generally, the temperature was lower when air masses were primarily from northeast; while the temperature was higher when air masses were primarily from southwest. The air masses of transition season were changed from northeast to southwest when cool season changed to cool season. In some year,

the air masses of transition season were changed from southwest to northeast when warm season is changing to cool season. But it was not found in our sampling period.

L 100. Consider changing “major” to another term such as “primarily”.

Response: Thank you. We have revised them.

L 106; Section 2.2. Additional description on how samples were collected is needed. What height was the inlet? What were the inlet dimensions and type? How was representative sampling of the aerosol assured? Importantly: Were there any local sources at the island that could contribute to TSP and skew results in comparison to background SCS marine boundary layer conditions? How were they identified and/or removed from the results? If other studies have considered this for the location, please note this and cite the study.

Response: Thank you for your suggestion. We have added the description in the manuscript (supplementary text S1). We think the samples were representative, because the sampling station is far away from continent at the island with few population and no industry.

L 123 and 124. Should be relative standard deviation?

Response: Yes and thank you.

L 127; Section 2.3. Additional information is needed on the backtrajectory model used, the version, the meteorological dataset source used, the receptor height, and how often backtrajectories were run. A brief description of why 10 day backtrajectories were selected would be helpful as well. Detailed information is not required and can be referred to the cited works, but a brief description would be helpful. Similarly, a brief description of how CWTs were used would be helpful.

Response: Thank you for your suggestion. We have revised this section 2.4 and supplementary text S2.

L 136; Section 2.4. Similarly, more detail on the PMF model setup is needed. For instance, a brief description of how and why five sources were selected by the methodology would be

helpful for the reader.

Response: Thank you. We also have revised this section 2.5 and supplementary text S3.

L 153. Consider more clearly specifying that percentages are on a mass basis throughout the manuscript. This would assist the reader in more clearly describing what percentages are referring to and how they should be interpreted.

Response: Thank you for your suggestion. We have added the information into the manuscript in Lines 128-129.

L 160; Section 3.1.2. Results from this section, section 3.1.1., and section 3.2 all contain somewhat repetitive, though slightly different descriptions, of similar results. Consider reorganizing the results in a way that presents this information, then discusses different relevant findings in a more ordered manner that leads to the study conclusions.

Response: Thank you. We have revised the sections 3.1.1.

L 223. The lack of correlation between rainfall observed at the receptor and TSP may not be sufficient to warrant the finding that “rainfall is not a major factor controlling seasonal variation of that concentration”. Precipitation is a complex process that can lead to both increases in TSP (e.g. via gust fronts etc.) and decreases through wet deposition, among other processes. In addition, relevant processes can occur on time scales below the four-day sample period of the filter samples. A more nuanced discussion of the relationship between precipitation and TSP should be included.

Response: We agree and have deleted this sentence.

L 226. State the hypothesized mechanism that links decreased TSP to higher temperatures and RHs via particle hygroscopic growth and interactions. Is this finding justified by the available data or are there better source to support this finding? A significant correlation is not sufficient to justify this statement.

Response: Thank you for your suggestion. We have added a reference in the manuscript.

We discovered negative correlations between TSP concentration and temperature ( $p < 0.01$ ) and

relatively humidity ( $p < 0.01$ ) (Table 2), indicating that warm temperatures and high relatively humidity enhance particle activation and scavenging is happening (Liu et al., 2011).

L 371; Section 3.3.1. Methods for this section could be included earlier in the methods section 2. It may help to better understand these results when they are first discussed in earlier results sections.

Response: We agree and have deleted this section. But we have reorganized them into other sections.

L 414. It can be very difficult to form valid conclusions on the composition of various particle sizes or modes from TSP observations on their own. These statements are somewhat speculative in nature, even with other studies to cite. Additional evidence or discussion should be included to support any contentions based on distinct sources associated with size distribution differences or size segregation.

Response: Thank you for your suggestion. We have deleted section 3.3.2 “Principal component analysis and classical multidimensional scaling”.

L 418; Section 3.3.3. More discussion on how PMF results were linked to the identified source types would be helpful. Was this solely based on relative ion concentration? This could be added to the methods section as well.

Response: Thank you for your suggestion. We have added more information to the methods section.

L 450. Smoke can be an important source of aerosol into the southern SCS during the “warm” monsoonal season as extensive burning can occur in Borneo and Sumatra. That less evidence of this impact is found (due to the noted potassium ratios) in the study’s more northern SCS marine region is interesting. Similar impacts from anthropogenic pollution are likewise noteworthy. The authors may wish to spend some time in the discussion emphasizing that there are important sources of aerosol throughout SE Asia and the maritime continent, while the CWT and ionic ratios indicate that sources important to the southern parts of the SCS may be removed or less important to northern SCS regions than those of regions around SE Asia and China.

Response: Thank you. We have re-calculated the PMF model and combined the PMF results with

CWTs results to discuss. In the revised manuscript, we found that K from biomass burning would loss its information when it transport to open ocean, because it can react with  $\text{H}_2\text{SO}_4$  or  $\text{HNO}_3$  in the atmosphere to form secondary aerosols.