

**Response to anonymous referee #1'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 by Xiao et al. presents a very detailed and comprehensive study of total suspended particulates (TSP) in the South China Sea. TSP was collected for the period of one year, covering all seasons, and analyzed for major ions. A variety of source apportionment methods, such as correlation analysis, principal component analysis, back trajectory analysis and positive matrix factorization, were applied to reveal the regional and source-specific origins of TSP. In addition, results are compared to previous studies from the literature and put into wider context.

Generally, this study is of scientific interest as it provides lots of detailed information on TSP in a region where various types of anthropogenic pollution as well as natural emissions from the sea contribute to the local aerosol load. However, this study shows a lack of methodological detail, the discussion is partly redundant and confusing, and a coherent storyline is missing. While this work is certainly worthwhile to be published, I recommend major revisions as detailed below.

**General comments**

1. The applied analysis methods must be explained in more detail. In particular, there is no information on the methodologies behind concentration weighted trajectories (CWT), the principal component analysis (PCA) and positive matrix factorization (PMF). It is not sufficient to provide references without explaining the methodology in the text. The reader must be able to understand what the authors did, on a general level, without consulting further literature. In addition, there is no information on how many blanks were produced and in which intervals. In the following I will elaborate a bit on how the PMF related part can be improved. I have less expertise for PCA and CWT but would recommend that the authors check very carefully what the standard for reporting is in the literature and include the respective information this in the manuscript. For instance, with which program were the back trajectories run, Hysplit, Flexpart, Lagranto or other.? What are the uncertainties in relation to the covered distance from the receptor?

**Response:** Thank you for your suggestion. We have added more information about the applied analysis methods in Section 2.4 and 2.5 (see detail information in supplementary text S2 and S3).

According to your general comments 3 and 4, we delete the analysis methods of PCA.

2. With regards to PMF, it is well established in the literature which aspects need to be explained at least (e.g., Zhang et al., 2011). In the presented manuscript, the authors do not describe how they prepared the error matrix and especially how they dealt with combining errors from different measurement techniques (i.e. TSP vs major ions). This can be very difficult and has a large effect on the results, please see for example (Crippa et al., 2013) for details. Did the authors downweight any component of the input matrix? In fact, the input matrix is not even described. Furthermore, the authors do not discuss how many solutions they explored (e.g. 1-10 solutions), the number of fpeaks and seeds and their range etc.

With regards to reporting of PMF results, here again a large discrepancy exists between what one would expect to see and what is actually reported (please see again Zhang et al., 2011). For example, as an absolute minimum the time series and profiles of the chosen factors need to be shown and discussed. Based on the presented information, I am unable to review the credibility of the presented results, because in addition to lacking methodological information, I do not know how similar or different the resulting factors are. What are the correlation coefficients between the factor time series and profiles? How do these factors relate to external variables, e.g. meteorological parameters? On the basis of what are the selected factors justified? Etc. All this information needs to be included, before the manuscript can be considered for publication.

Response: Thank you for providing literatures and suggestions. We recalculate the PMF results using PMF5.0 (United States Environmental Protection Agency, EPA) according to the methods of Zhang et al. (2011) and EPA PMF5.0 user guide (Norris et al., 2014), more information was described in Section 2.5 and its supplementary text S3.

In the revised manuscript, all ions were re-calculated without TSP concentrations. The uncertainties by ionic species were provided by the analytical library and we used the uncertainties as error matrix (Norris et al., 2014), and used ionic species and sampling time as input matrix. We downweighted Fe and F<sup>-</sup> since they had low signal-to-noise ratios (S/N), and there were no excluded species and samples.

In addition, we added two figures of profiles and time series.

Figure 9 Profiles of five sources identified from the PMF 5.0 model, including sea salt (two species),

crust, secondary inorganic aerosol and oceanic emission.

Figure 10 Time series contributions from each identified sources, including sea salt (two species), crust, secondary inorganic aerosol, and oceanic emission.

3. The manuscript is lengthy. This is in part due to redundancy in the discussion of results from different source apportionment methods, see specific comments. I suggest shortening the discussion section and focusing on a few findings instead of discussing all details. The manuscript is partly confusing for the reader and in the end it is not clear what the main points are. A consistent story line needs to be crafted.

Response: Yes, the manuscript is lengthy. 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 moved some information about the methods of Sample collection and chemical analyses (S1), Back trajectories and CWTs analysis (S2), and PMF model (S3) into supplementary text.

4. The authors use a suite of source apportionment techniques, however it is not clear what the added value is. This is due to the fact that the results are discussed one after another separately per method and no connection between them is established. Often this results in repetitive discussion. Each technique has its strengths and weaknesses that are hardly exploited in this work. When applying so many methods, I would expect that e.g. the CWT are used to supplement PMF results where the PMF results show ambiguities, or that PCA is used in addition to CWT because CWT cannot determine specific source types which PCA can help with. Conversely, CWT are helpful to determine regional provenance of TSP which PCA or PMF cannot provide. Also, in some instances, results are contradictory (see specific comments), this is however not discussed. Such discrepancies need to be addressed rather than focusing only on confirmative results.

Response: Thank you for your suggestion. In the revised manuscript, we combined CWT with PMF to explore source identification, apportionment, and region. It was shown in the new section 3.3.

5. As indicated in the specific comments sections, references are sometimes missing, while in other instances it is not clear what exactly the authors refer to in a study when providing a reference.

Response: Thank you. We have added some references in the manuscript.

**Specific comments:**

l. 30f: It is not clear what you mean by “Na<sup>+</sup> and Cl<sup>-</sup>...made up 74 % and 82%...” These numbers clearly don’t add up and information on the reference is missing.

Response: We revised the sentence in Lines 31-32.

l. 31f: What is marine aerosol in this context? How was it determined?

Response: Marine aerosols in the paper refer to those sampled at Yongxing Island over the South China Sea.

l. 34: Already in the abstract NH<sub>4</sub><sup>+</sup> is claimed to originate from marine biogenic sources. However, throughout the manuscript there is no explanation what these marine biogenic sources are, which seasonality they follow and how the measured ammonium is related to marine biological activity. Without this information, I am not convinced that the ocean is the primary source of ammonium.

Response: We change “marine biogenic sources” to “oceanic emission” in the manuscript. In addition, according the results of CWTs, air masses with high concentrations of ammonium were from the open ocean, so we suggest that ocean may be the primary source of ammonium.

l. 38f: what about the role of climate? This first sentence could use some more references since many factors are mentioned.

Response: Thank you. We have revised the sentence in Lines 39-42.

l. 40: What are “complex sources”? I could imagine that the authors wanted to express that aerosols have many sources which create a complex mixture of aerosol components? What about mineral dust emissions from wind opposed to rock weathering? Also, references are missing.

Response: Yes. We have revised the sentence in Lines 42-47.

l. 44: I find the list of aerosol components random. E.g. organics are not mentioned while it has been shown that they constitute an important fraction of aerosol chemical components. Also BC is not mentioned.

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

l. 45: This statement is not differentiated enough. Some parts of the world have undergone significant socio-economic growth in the past decades, such as East Asia, which has led to much higher emissions. In other parts of the world, emissions have decreased due to stricter air quality legislation. This should be reflected in this sentence or the focus should clearly be on East Asia, the region relevant for the South China Sea (SCS).

Response: Thank you for your suggestion. We have revised in Lines 49-51.

l. 50 – 65: The purpose of this paragraph is not clear. What is the point of discussing aerosol deposition and ocean productivity in the context of this particular manuscript? If the idea was to provide a brief review of particulate pollutants to the ocean atmosphere it is not clear why only nitrogen containing compounds are mentioned? Also I do not see the value of reporting observation from many different locations. I would suggest focusing on what is known about the SCS and report on aspects that are of relevance to TSP observations as presented in this manuscript.

Response: Thank you. We have deleted in this paragraph.

l. 75: What is the difference between “aerosols and pollutants” in this context? Do the authors want to distinguish between natural and anthropogenic sources or particulate and gas phase pollutants?

Response: Yes. Thank you.

l. 79: Is there not a more recent reference for biomass burning emissions and resulting deposition?

Response: We have added more two recent references.

l. 95 f: a reference is missing.

Response: Thank you. We have added them.

l. 97: “In the present study, it was...” What is meant by “it”?

Response: The sampling period. We have revised it.

l. 101: What is the “local southeast”?

Response: Local and short air masses. We have revised it.

l. 101: Since the variations of temperature and the difference between what is called the “cold” and “warm” seasons are very small some more information is needed on how seasons were separated and why. Especially what qualifies as transition season?

Response: In the manuscript, we separated 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. 122f: what do the relative standard deviations refer to? Repeated measurements of a standard, a blank or something else? What about the number of blanks that were generated in the course of the year? Please include more detailed information.

Response: Repeated measurements of a standard. Three blank filters were taken from each package (25 filters).

We have added the detailed information in the revised manuscript (supplementary text S1).

l. 139: In how far do these references reflect what the authors did? These references point towards different tools for running PMF.

Response: We recalculate the PMF results using PMF5.0 (United States Environmental Protection Agency, EPA) according to the methods of Zhang et al. (2011) and EPA PMF5.0 user guide (Norris et al., 2014), more information was described in Section 2.5 and supplementary text S3.

l. 149: TSP mass concentrations are compared to those in other cities. The authors write “around the world”, however the references point only towards Asian cities. It is fine to compare to Asian cities only, but this should be made explicit, i.e. state the locations and reference TSP concentrations there.

Response: Thank you. We have deleted this comparison.

l. 151f: Again provide numbers for reference. Where are those places, why are they comparable?

Response: Thank you. We have deleted those places.

l. 155f: This paragraph is not readable. A table is preferable.

Response: Thank you. We have added a table.

**Table 1** Annual average, minimum and maximum mass concentrations ( $\mu\text{g}/\text{m}^3$ ) of TSP and aerosol chemical species at Yongxing Island.

	TSP	Na <sup>+</sup>	Cl <sup>-</sup>	K <sup>+</sup>	Ca <sup>2+</sup>	Mg <sup>2+</sup>	SO <sub>4</sub> <sup>2-</sup>	NO <sub>3</sub> <sup>-</sup>	NH <sub>4</sub> <sup>+</sup>
Annual	89.6 ± 68.0	4.00 ±	7.73 ± 5.99	0.33 ±	2.15 ±	0.44 ± 0.33	5.54 ± 3.65	1.95 ± 1.34	0.07 ± 0.07
		1.88		0.22	1.54				
Minimum	16.4	0.90	0.39	0.06	0.17	0.02	0.52	0.10	0.01
Maximum	440.1	8.86	36.47	1.13	9.65	1.55	23.34	10.05	0.32

l. 162: What does the value in parenthesis represent? An annual average? What is the standard deviation?

Response: An annual average. However, we did not found the standard deviation in their papers.

l. 164: Again providing numbers for references is needed.

Response: OK. The revised sentence was shown in Lines 138-142.

l. 166: Here Fig. 5 is mentioned, while Fig. 4 has not yet been referred to. Please check the order of the figures.

Response: Thank you. We have re-checked the order.

l. 172: What is the “global ocean”? This expression is used various times. Please replace it by a

more accurate description of what is meant, e.g. “among all locations”

Response: Thank you. We revised “global ocean” as “among all locations” in the manuscript.

l. 174: Are the dead corals under water or exposed to the atmosphere? If they are not exposed, I don't understand how they can contribute to the measured calcium.

Response: Thank you. We have deleted this.

l. 175: Starting from here, the authors refer to some major ions as non-sea salt ions. However, it is not explained in the manuscript how sea salt and non-sea salt contributions to ions were determined. Please include this information in the methods section.

Response: Thank you for your suggestion. We have added this information in section 2.3.

l. 179: How can the authors show that the Sahara Desert is a source of dust for the measurement location? The way the information is provided is not convincing.

Response: Sorry to make a misunderstanding about this. We revised this sentence in Lines 158-159.

l. 179 f; l. 202: What do the authors want to say with “average  $Mg^{2+}$  concentrations” being “nearly consistent with... $Na^+$ ”? Or  $NO_3^-$  concentrations “were often consistent with those of  $nss-SO_4^{2-}$ ”. Do the authors refer to the ion balance? Please explain and change the formulation in the manuscript.

Response: We have revised them in Lines 159-161 and Lines 187-189.

l. 201: references are missing.

Response: We have added them.

l. 216f: “most other studies”. Are there only the four that are cited or more? What are their locations? What are those studies about?

Response: Thank you. We have revised them in Lines 202-204.

l. 222: Why “many other studies” when only two are cited. Again, which locations do these studies refer to?



Response: Thank you. We have revised it in Lines 208-209.

l. 223: How can TSP and rainfall not be related if some major ions are influenced by TSP?

Response: Precipitation is a complex process that can lead to both increases in TSP (e.g. via gust fronts etc.) and decreases through wet deposition. In addition, relevant processes can occur on time scales below the four-day sample period of the filter samples (anonymous referee #2's interactive comment).

So, we have deleted this sentence.

l. 228: What is meant by “particle wetting and interaction”? From the previous paragraphs I understand that there is more rain during the warm season. So my guess would be that particle activation and scavenging is happening. Do the authors refer to aerosol cloud interactions?

Response: Thank you. We have revised them in Lines 212-214.

l. 241: This suggests, it doesn't show.

Response: OK.

l. 255 before and after: It is not clear to me, why the authors do not discuss the concentrations and ratios of major ions that may originate from sea salt in the context of their ratios in sea salt. The authors even provide a table with typical major ion ratios in sea salt but do not refer to it. The discussion could highly benefit from this addition at this point.

Response: Thank you for your suggestion. We have added the discussions of the concentrations and ratios in the manuscript. For examples:

As shown in Fig. 5, Tables 1 and 2, similar trends and strong correlation were observed among  $\text{Na}^+$ ,  $\text{Cl}^-$  and  $\text{Mg}^{2+}$ , and the ratios of  $\text{Mg}^{2+}$  to  $\text{Na}^+$  in aerosols were close to that in seawater, suggesting that  $\text{Mg}^{2+}$  may mainly derive from sea salt rather than continental sources.

l. 257: What do the authors mean with “complex”?

Response: We revised “complex” to “different”.

l. 258: Please specify what is meant with “phenomenon”.

Response: It mean the same with rainwater at Yongxing Island. We revised it in Lines 234-235.

l. 259: How does the study of Moody et al. compare with this work? Why is it comparable?

Response: We have deleted it.

l. 269: Biomass burning is not a major source of sulfur containing species compared to other sources.

Why do the authors refer several times to biomass burning as source of SO<sub>2</sub> in the manuscript?

Response: Thank you for your suggestion. We have revised them.

l. 276: I suggest reformulating this sentence: “Lawrence and Leliveld (2010) attributed x % of NO<sub>x</sub> emissions to...” In the current form it sounds like these values were recently measured.

Response: Thank you for suggestion. We have revised and moved to section 3.3, in Lines 375-376.

l. 291: which time period is reflected?

Response: 2011. We have added it.

l. 296: What about the influence of anthropogenic activities?

Response: Thank you. We have added the information in the manuscript in Lines 265-266..

l. 330: What are “dynamic” smoke surface concentrations?

Response: We deleted “dynamic”.

l. 352: “Figure S2 confirms these findings” by showing and proving what?

Response: Thank you. We have revised them in Lines 319-319..

l. 363: Do the authors mean “accumulation mode” aerosol?

Response: Yes.

l. 369: After reading this long description I lost track of what the main message is. This needs to be

written much more concisely by focusing on the most important findings.

Response: Thank you for your suggestion. We have deleted section 3.3.1 and reorganized them to other paragraph.

l. 381: “depletion probably occurred”. There is no evidence for it.

Response: Thank you. We have deleted it.

l. 384f: This conclusion is not evident. How can Cl<sup>-</sup> from KCl be more dominant than Cl<sup>-</sup> from sea salt? Furthermore, I do not understand what the difference is to what has been discussed before with regards to K in l. 330-338 (K as marker for biomass burning). This is confusing.

Response: Thank you. We have deleted it and reorganized them.

l. 387: I am not convinced that SO<sub>4</sub><sup>2-</sup> is a biomass burning marker. The relation between potassium and sulfate might result from the transport of air masses from the same source region with different source types.

Response: Yes. Thank you. We have revised it.

Section 3.3.1: I suggest integrating the findings from the correlation analysis into the other sections. This section is very redundant and makes the manuscript unnecessarily long.

Response: Thank you for your suggestion. We have reorganized them to other sections.

l. 404: This statement is disconnected from the previous analysis. What has been mentioned that relates to this section?

Response: We have deleted section 3.3.2 in pre-revised manuscript.

l. 408: An explanation for what CMDS is, is needed.

Response: We have deleted section 3.3.2 in pre-revised manuscript.

Section 3.3.3: Please see comments above. The lack of information and figures is not acceptable.

Response: Thank you for your suggestion. We have added two figures in this section (Figure 9 and

Figure 10).

l. 439: Why 50 % now, in l. 422 it was 58 %.

Response: We have revised them.

l. 441: I do not understand this sentence “CaSO<sub>4</sub> and sulfate containing both K and Ca...”

Response: Thank you. We have deleted it.

l. 449f: Do the authors say that 41 % of potassium comes from biomass burning?

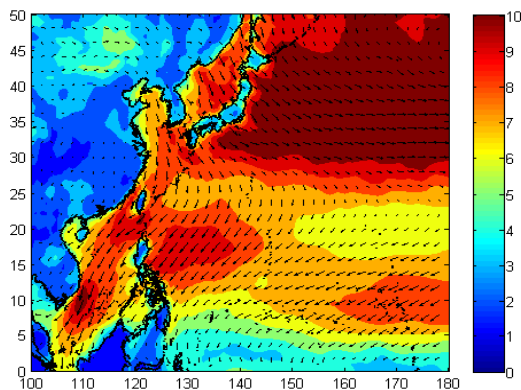
Response: Because we re-calculate PMF model, biomass burning is not a primary source and K<sup>+</sup> is existed the form of secondary inorganic aerosol.

l. 455: “In addition, biomass burning produces SO<sub>2</sub> and NO<sub>x</sub>...” has been mentioned at least for the third time. Again, there are too man repetitions in this manuscript.

Response: Yes, we have reorganized them.

l. 460: What is the reason for it, a meteorological situation that favor southward transport of air masses? Again, in many cases more precise information is needed what the authors refer to exactly in the given literature.

Response: In cool season, the wind direct is from northeast. We have revised them (see below figure: wind direct and wind intensity over the northwest Pacific in Jan. 2015)



l. 485: I do not see the point of “a major discovery”. An explanation is needed why the authors think this is new knowledge.

Response: We agree. We have deleted “a major discovery”.

l. 515-520: The origin of ammonium and ammonia is discussed again here. This is repetitive and it is not clear to me, why the authors reveal the information on the nutrient situation in the marine water only at this very late point in the manuscript?

Response: We reorganize this section with PMF model and delete the discussion about nutrient situation.

**Technical comments:**

l. 19: “major inorganic ion concentrations” instead of “inorganic chemical ionic concentrations”

Response: Accepted.

l. 25: insert “which were” before “higher in the cool season...” and remove the “,”

Response: Accepted.

l. 26: finish the sentence after “seasons” and start a new one with “Factors of influence were...”

Response: Accepted.

l. 33: write “was the dominant source of...”

Response: Accepted.

l. 40 f: write “Aerosols have many sources. Primary aerosols, emitted directly from...”

Response: Accepted.

l. 73: remove “SCS” behind “northeast”.

Response: Accepted.

l. 94: replace “such” with “the high”

Response: Accepted.

l. 95: replace “be” with “arrive”

Response: Accepted.

l. 103: “Accumulated annual rainfall...”

Response: Accepted.

l. 113: remove “an” before “another”

Response: Accepted.

l. 120, 123, 124: “relative” instead of “relatively”

Response: Accepted.

l. 145, 160: “over the SCS”

Response: Accepted.

l. 147: remove “aerosols” behind “TSP”.

Response: Accepted.

l. 165: “annual average TSP and ionic concentration are comparable to...”

Response: Accepted.

l. 172: replace “composed” by “contributes”

Response: Accepted.

l. 205: remove “that”

Response: Accepted.

l. 215: “distinct”

Response: Accepted.

l. 221: I suggest to write: "... because 70 % of rainfall at...happens during the warm seasons..

Response: Accepted.

l. 224: replace "that" by "mass".

Response: Accepted.

l. 225: replace "strong" by "high"

Response: Accepted.

l. 247: "in contrast to"

Response: Accepted.

l. 254: "correlation" instead of "correction"

Response: Accepted.

l. 264: "suggest" instead of "show".

Response: Accepted.

l. 267: insert "were observed" after (Wang et al. 2006).

Response: Accepted.

l. 276: "emissions" instead of "emission", twice

Response: Accepted.

l. 285: replace "difficult" by "limited"

Response: Accepted.

l. 291: insert "the" before "Acid Deposition..."

Response: Accepted.

l. 313: “Excess Cl- in January has been observed by...”

Response: Accepted.

l. 316: insert “for almost all stations” at the end of the sentence.

Response: Accepted.

l. 318: no “s” in “oceans”

Response: Accepted.

l. 340: remove “the reported by”

Response: Accepted.

l. 383: insert “fuel” after “fossil”

Response: Accepted.

l. 395: Table 2, I believe. Delete “that”.

Response: Accepted.

l. 438: Remove “absolutely”

Response: Accepted.

l. 491: replace “as that” by “compared to”

Response: Accepted.

l. 532: “to help better understand their chemical...”

Response: Accepted.

l. 535: “with higher concentrations in the...”



Response: Accepted.