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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Source apportionment of the submicron organic aerosols over the Atlantic Ocean from 53° N to 53° S using HR-ToF-AMS" *by* Shan Huang et al.

Anonymous Referee #1

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(My ratings for both scientific and presentation quality are between "Good" and "Fair", so I give one "Good" and one "Fair".)

General Comments:

The authors performed 4 month-long field measurements across 53°N-53°S over the Atlantic Ocean from 2011-2012 and reported convincing source apportionment based on adequate and well-processed datasets obtained from HR-ToF-AMS and other techniques. Due to few number of similar studies that covered long time series and/or oceanic regions, the findings in this paper certainly provide valuable insights into the sources and origins of marine submicron atmospheric aerosols. Overall, the paper dis-

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Discussion paper

cussed relevant scientific questions within the scope of ACP journal with novel methods and datasets, and the results are generally (but not all) supportive to the interpretations and conclusions, in spite of some technical concerns and vague presentation or expression that need to be further supported, examined, or re-phrased. Therefore, I would recommend this paper be accepted for publication once the following specific comments are completely addressed.

Suggestions for major improvements and revision:

1. The author(s) should have made best use of their valuable datasets as well as the previously published studies, and emphasized the significance of their findings, if they also agree that they haven't done this enough in the abstract and the in the introduction. Besides, the author(s) should also, on one hand, carefully refer to previous studies that used similar techniques for marine aerosol, and on the other hand, include necessary comparisons (if available) in their own discussions. For example: The authors should add proper references to the sentences ended in: Page 10 Line 5; Page 11, Line 25; Page 12, Line 27; Page 12, Line 34; Page 13, Line 6; Page 16, Line 24 (Might be useful: Charlson et al., 1987, Nature; Bonsang et al. 1992, GRL; Yassaa et al., 2008, Env. Chem.; Shaw, Gantt, and Meskhidze, 2010, Advances in Meteorology).

2. Conclusions discussing causality or reasoning must be carefully examined. Just give a few examples: Page 9, Line 5: The authors attributed "insufficient offline samples" to the weak correlation between AMS and offline sea salt. Actually this might not be a reasonable explanation especially if they used AMS data collected from the exactly same periods of time during the offline filter sampling. The data size itself should not affect the R2, and the authors should also examine p-value of correlation for "meaningfulness". Furthermore, in this case, the authors should also clarify how they measured sea salts using the individual techniques and why they applied the method from Ovadnevaite et al. (2012). For example, what ions were included as sea salts? Did they count Na+, Cl-, SO42-, K+, Mg2+, etc. in both? If NaCl accounted for different fraction from that in Ovadnevaite et al. (2012), was the scaling factor of 51 still suitable? Other-

wise, the “therefore” in Line 6, did not explain why the same scaling factor was applied, considering the correlation and the coverage of time (“full year measurements in the reference”) discussed above was not supportive, or not relevant. Page 10, Line 18: The author stated “The ammonium concentrations didn’t follow a clear seasonal trend, although its precursor ammonia could be emitted from ocean (Ikeda, 2014; Johnson et al., 2008). The absence in seasonality suggests particulate ammonium during Polarstern cruises was contributed by both anthropogenic and biogenic sources.” The deduction did not support their conclusion. Page 13, Line 29: “The diurnal variation of NOA shows clear peak in the afternoon, reaching the maximum while the global radiation starts decreasing (Figure 4), indicating that the NOA factor is certainly composed of secondary organic products.” The evidence is weak, and the authors should specify “global radiation” and cite papers that observed the similar diurnal trends of such a NOA factor, if available. Page 13, Line 20: The authors suggested “This can be useful for better estimation of marine DMS related SOA both in field measurements and in models”. However, MSA as a fraction of SOA can vary largely and different from time to time (especially between summer and winter). In addition, MOA in this case might not be equivalent to SOA.

3. For better presentation quality and reading experience, the English language and scientific writing in this paper can be more precise and largely improved. Just give a few examples: Page 11, Line 9: “These S/C ratios derived from the PMF analysis tool contain however certain estimation uncertainties and have therefore to be used with caution.” This seems to be a grammatically wrong sentence. Page 12, Line 24: “The minimum of the diurnal variation (0.04 μg m-3) appears around 09:00, probably linking to the increase of mixing layer in morning.” This sentence needs to be re-phrased and also supported with references. Page 10, Line 27: I think it is more precise to say “57 hours” rather than “about 2 consecutive days”, unless there was an interruption. Page 13, Line 6: “the this OA component”. Despite the grammatical error and lack of references, “OA component” was vague in the context. Page 17, Line 17: In this paragraph, the author said “still questionable” and then “This suggests. . . could be not

correlated". This led to confusion due to the inappropriate English or logical expression.

Other technical and specific comments to be addressed:

1. Generally when discussing seasonality, the difference between "spring & autumn" might not be as distinct as that between "summer & winter", in term of many factors such as meteorological parameters and marine bioactivity. Besides the "spring vs. autumn" comparison, the authors may also want to look into "spring/autumn vs. tropic". In addition, their measurements on board was changing with time and location at the same time, so this will be different from those studies took place at a ground site over seasons. I wonder if the authors would like to make some comments on these.

2. The authors should try to clarify the influences from the "open oceans", "marine", and "coastal" when interpreting results in the discussions, even though the boundaries might be blurry. For example, on Page 11, Line 30, the author stated "The S/C ratio of the MOA factor is also over twice that of marine factor observed in Paris (0.013, Crippa et al., 2013b), implying a stronger influence from marine phytoplankton on aerosol particles over the ocean than those in the coast city.", but actually the abundance of phytoplankton can be much higher in the coastal areas. See https://earthobservatory.nasa.gov/GlobalMaps/view.php?d1=MY1DMM_CHLORA

3. The authors are suggested to add discussions for organosulfates, since they can make a considerable contribution to continental SOA masses at certain locations, and also derived from the same biogenic precursors over the oceans. For example, how is this class of compounds measured using AMS? Was it included in organics or sulfate, or neither?

4. Last but not least, the authors should revise the manuscript carefully by their own. Just give a few examples: 1) Page 12, Line 34: "Figure 4" – should this be Figure 5? 2) Acronym: define before use. For example, "SOA" was not defined but used in the abstract; "OA" was firstly defined on Page 10, Line 30 in the main text; "biomass burning" was defined but not used in many places. 3) Please be consistent when using

terms such as “fPeak” or “fpeak”, “CxHyO” or “CxHyO1”. 4) Please be consistent about adding a “_” between numerical values and their units. 5) Please specify “CxSj+” on Page 12, Line 21.

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