Responses to Reviewer' comments

We appreciate the reviewers for their thorough reading and thoughtful comments and suggestions, which greatly improve the quality of the manuscript. We revised the MS accordingly. The point-to-point responses to all the comments are given below in blue.

Reviewer #1 (Formal Review for Author (shown to author)):

This observation-base study presents the data of organic aerosol species from a cruise campaign from the South China Sea to the eastern Indian Ocean. It shows the spatial variations of dicarboxylic acids, oxocarboxylic acids and α-dicarbonyls in the marine aerosols in the investigated oceanic areas. It also discussed their sources and major influence by the oceanic emissions and long-range transport. It could be accepted for publication in ACP after revision.

Response: We thank the reviewer for the comments and the recommendation.

Major comments:

1. I would suggest the authors to re-organize the manuscript in order the present their findings in a clearer way. In the current version, it was not easy for me to follow and to understand the major findings. Some part(s), e.g. their fraction in water-soluble organic carbon, can be moved above to a part maybe mainly describing the overview of data. If possible, the data of ions as well as organic carbon, elemental carbon may be shown there to let the readers quickly get an overview of the data. The authors should also re-organize the discussions for a better presentation of their results and conclusions. For example, the ratios of C_3/C_4 dicarboxylic acids and their correlations are separated into two different parts, which should be merged. These discussions are highly related.

Response: Following the reviewer's suggestion, we have added the description of OC and EC in the revised MS as a sub-section 3.1. Also, the description of WSOC data has been moved to sub-section 3.1 from the 3.5. In addition, the WSOC data has been given in Table 1. The correlations of C_2, C_3, and C_4, and those of C_2% and C_2/C_4, as well as Pyr, ωC_2, and MeGly, and the relevant description of chlorophyll-a in the original sub-section "3.3.2 Linear correlations" have been combined and placed in original sub-section "3.3.1 Diagnostic mass ratios". The rest of original sub-section "3.3.2 Linear correlations" has been moved to the supporting information.

2. I would suggest the authors to polish their findings. What are the major findings in this observation? What information would they like to bring to the readers?

Response: We have improved the discussion of our results to make our findings such as the origins and formation processes (mainly marine BVOCs and *in-situ* secondary formation) over the SCS-EIO region clear to the readers. Please see the abstract and conclusions in the revised MS.

3. Be very careful to deal with the correlations and ratios, I found they may suggest different/contradictory conclusions in the discussion.

Response: We have carefully interpreted the mass ratios and correlations between selected species in assessing the origins and secondary formation/transformations of diacids and related compounds and discussed in detail, without leaving any contradictory view. Please see the sub-section 3.4 in the revised MS.

3. The authors should pay attention to their citations. In these oceanic areas, there are some other observations on organic aerosols which should be inspirational for the authors when undergoing their discussions.

Response: We have tried our best to make modifications to the original text and have added some discussions in the revision. The revised content is as followed:

"The dominance of C_2 followed by C_3 and C_4 diacids is consistent with those coastal marine aerosols (Kundu et al., 2010;Kunwar and Kawamura, 2014) and remote marine aerosols from the western North Pacific (Bikkina et al., 2015), suggesting similar formation processes of dicarboxylic acids in the atmosphere." (see Page 8, Line 213–216).

We also added the following sentences in the revised manuscript.

"For the aerosol samples of SCS, the 5-day back trajectories (Fig. 1b) showed that the air masses were originated from East Asia, whereas the air masses of Malacca were delivered from Southeast Asia. The concentration of diacids and related compounds were highest in SCS and second highest in Malacca, due to increased anthropogenic activities through long-range atmospheric transport in East Asia and Southeast Asia, respectively." (see Page 8, Line 206–210)

"The spatial distributions of total oxoacids and α-dicarbonyls were similar to those of total diacids, indicating that they were similar in origin or formation mechanism (Kunwar et al., 2017). Based on the backward trajectory analysis, we identified four source regions of diacids and related compounds: (a) East Asia, (b) Southeast Asia, (c) the Bay of Bengal, and (d) the East Indian Ocean. When air masses originated from East Asia and Southeast Asia and their coastal areas (i.e. SCS and Malacca), the concentrations of these compounds were high; when air masses were mainly derived from the East Indian Ocean (i.e. EIO-WI), their concentrations was the lowest; and when the air masses originated from the Bay of Bengal and the East Indian Ocean (i.e. EIO-SL), the concentration of these compounds was between the former two." (see Page 12, Line 327–335)

4. Some figures can be moved to the supporting information. For example, Figure 2 only shows the chemical structures of the diacids. It is hard to get information from Figure 9 efficiently. Figure 8 could also be moved to the SI.

Response: In the revision, we have moved Figure 2, Figure 8 and Figure 9 to the supporting information (SI).

5. L301-305: I do not understand why? From the ratios, we could say these aerosol particles are aged but it is hard to know if they are influenced by marine biota or continental anthropogenic emissions.

Response: To avoid such confusion/misunderstanding to the reader, we modified the discussion by removing the following sentence: "Further ---- emissions." and by detailing the interpretations of the ratios and correlations of the selected species. In fact, the air masses transported from the continental regions are aged (confirmed by C_2%) and also mainly influenced by the marine biota emissions (confirmed by the mass ratios and correlations between species, e.g., C_6 and C_9 diacids). We detailed these points in the revised MS (please see sub-section 3.4).

6. L361-365: The sentence is too long and hard to be followed. Please rephrase it.

Response: Now the revised sentences are as follows:

"In addition, there were good correlations among Pyr, ωC_2 and MeGly in the SCS and Malacca, while the correlations were poor in the samples from EIO-WI and EIO-SL (except for Pyr and MeGly in EIO-WI, and ωC_2 and Pyr in EIO-SL). The results showed that organic aerosols produced by BVOCs (e.g. isoprene) emitted from the ocean surface were more aged in the EIO-WI and EIO-SL than the SCS and Malacca where were affected by anthropogenic emissions." (see Page 15, Line 437–441)

7. Line 394-396: I do not see the high Chlorophyall-a concentrations in the SCS in the satellite image (Figure 1a). A close look at a special case of some samples (e.g. 55-60) would be necessary.

Response: The manuscript shows the time series diagram of chlorophyll-a in Figure 3k. It can be seen that the concentration of chlorophyll-a in SCS and Malacca is significantly higher. In addition, the concentration of chlorophyll-a in samples No. 55-60 was also higher. Chlorophyll-a is a measure of phytoplankton, or algal, biomass (Quinn et al., 2014) and currently most widely used proxy for predicting isoprene concentrations in water (Hackenberg et al., 2017). Numerous studies reported the positive relationship between isoprene emission and chlorophyll-a in the surface seawater (Zhu et al., 2016; Hackenberg et al., 2017). Spatial distributions of marine VOCs are expected to be linked to the distributions of photosynthetic pigments in seawater, such as chlorophyll-a (Ooki et al., 2015). The higher concentrations of chlorophyll-a in the coastal regions stand for higher biological activities and more active to the emission of VOCs (Kang et al., 2018).

8. Minor errors:

L210: it should be "lower than"

Response: We have corrected the mistake in the revised manuscript. "The concentrations of total oxoacids are lower than those from Gosan, Jeju Island, South Korea (average 53 ng m^{-3}) (Kawamura et al., 2004) and urban sites in China (45 ng m^{-3}) (Ho et al., 2007)." (see Page 11, Line 303)

L298-299: should the sentence be "the more the aerosol particles are aged, the higher the ratios are"?

Response: We have corrected the mistake in the revised manuscript. "In general, the more the aerosol particles are aged, the higher the $C_2/\Sigma(C_2–C_{12})$ ratios are (Kawamura and Sakaguchi, 1999)." (see Page 14, Line 398–399)

L311: "attribute for" should be "attribute to"

Response: We have corrected the mistake in the revised manuscript. (see Page 15, Line 419)

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