Responses to Reviewer's comments

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

Reviewer #2 (Formal Review for Author (shown to author)):

The manuscript studied the distributions of dicarboxylic acids, oxocarboxylic acids and α-dicarbonyls in marine aerosols during a cruise from 10 March to 26 April 2015. The cruise area is over South China Sea to East Indian Ocean. There were many samples collected, and the analysis were based on four regions, SCS, EIO-WI, EIO-SL, and SLDP. Through the different concentrations and ratios of dicarboxylic acids, oxoacids and α-dicarbonyls, their sources and possible formation pathways in each studied region were discussed. The work in this manuscript is very important. However, the article still needs to be major revised and then can be published on Atmos. Chem. Phys.

Major Comments:

1. The main meaningful was not very clear during the part of Introduction. The author should give more discussion about this cruise especially the important of this studied area in the Introduction.

Response: Thanks for the reviewer's suggestions. We added the following sentences in the revised manuscript in the section of Introduction.

"Land-sea-air interaction is one of the most important issues in earth system science. Atmospheric aerosol is the major component in the Earth's atmosphere and is one of the key carriers of the global biogeochemical cycle of nutrients (Zhuang et al., 1992;Li et al., 2017;Tan et al., 2011)." (see Page 2, Line 35–37)

"The oceans account for more than 70% of the earth's surface, and marine aerosols are important components of the global aerosol system of natural sources. However, current knowledge about the biogeochemical cycles of organic matters in the tropical marine atmosphere is very limited." (see Page 3, Line 79–81)

"Studies on the molecular composition and distribution of dicarboxylic acids and related compounds can provide useful information for source analysis, secondary formation and photochemical transformation processes of atmospheric organic aerosols." (see Page 3, Line 97–99)

"The South China Sea (SCS) is a large semi-closed marginal basin and one of the largest marginal seas in the world (Liu et al., 2002). The seasonal division of prevailing winds in the South China Sea is mainly influenced by the northeast monsoon roughly from mid-October to mid-March of the following year and by the southwest monsoon from mid-May to mid-September; while from mid-March to mid-May is the spring transition period, during which the wind direction is variable. The climate of the South China Sea is part of the East Asian monsoon system (Lau et al., 1998). The Indian Ocean is the third largest ocean in the world, with distinct tropical maritime and monsoon climate characteristics. The prevailing wind over the Indian Ocean in summer is the southwest monsoon, while the prevailing wind in winter is the northeast monsoon (Fu et al., 2016;Ramanathan et al., 2005). The Indian Ocean is warmer than the Pacific and Atlantic at the same latitude, so it is called the tropical ocean. The tropical East Indian Ocean (10°S–15°N, 65°E–100°E), including the southern bay of Bengal, southeastern Arabian Sea, eastern equatorial Indian Ocean and parts of the southern Indian Ocean, is one of the key regions affecting climate change such as drought and flood in China (Yao et al., 2015)." (see Page 3–4, Line 81–93)

2. There are many analyses and data in the manuscript. It is very difficult for readers to understand the information present in the article because of the illogical.

Part 3.1.1 Dicarboxylic acids:

There are many discussions about C_9 in paragraph 4, 5, 6, 7, and discussions about Ph in paragraph 5 and 8. The whole part of 3.1.1 is very illogical, and I can't catch the important point and main results. The author should analysis the main connection between these dicarboxylic acids and different areas, give more clearly analysis. For example, form the analysis in the manuscript, the most important dicarboxylic acid is the C_9 which have relation with C_2–C_4, C_6, and Ph. The author can put these results together and give the discussion, then give the main point of these results.

Response: Thanks for the reviewer's suggestions. The revised manuscript have moved the overall description of dicarboxylic acids and related compounds to the front of Section 3.2.1 (original section 3.1.1), and have reorganized the contents of Section 3.2.1 (original Section 3.1.1) into four paragraphs, i.e. the first paragraph introduces C_2–C_4, the second paragraph introduces C_9, the third paragraph introduces C_6, and the fourth paragraph introduces Ph. The logic of the revised manuscript should be clear enough for reader to follow. And at the beginning of Section 3.2.1, a general sentence has been added:

"C_2–C_4, C_6, C_9 and Ph are important species of dicarboxylic acids; this section briefly summarizes the main sources and/or formation processes of these organic acids." (see Page 9, Line 238–239)

Part 3.1.2 Oxocarboxylic acids:

The meaning of oxocarboxylic acids in the secondary paragraph should be discussed firstly in this part.

Response: Thanks for the reviewer's suggestions. At the beginning of Part 3.2.2 (original Part 3.1.2), this manuscript has noted that oxocarboxylic acids mainly contain ωC_2−ωC_9 and pyruvic acid, and have added the following sentence:

"Being similar to diacids, oxocarboxylic acids are mainly derived from combustion sources, but can also be produced by photooxidation of various organic precursors in the atmosphere from anthropogenic and biological sources (Kawamura and Bikkina, 2016)." (see Page 11, Line 297–300)

Part 3.3.1 Diagnostic mass ratios:

The data of C_3/C_4, C_2/C_4, $C_2/\Sigma(C_2-C_{12})$, and M/F can give the information of organic aerosols aging. And they can be put together to give the discussion which can be more clearly.

Response: The revised manuscript has moved the content of M/F in Section 3.4 (original Section 3.3.1), and discussed it with C_3/C_4, C_2/C_4, $C_2/\Sigma(C_2-C_{12})$, to explain the aging of organic aerosols.

Part 3.3.2 Linear correlations:

This part can be put in the supporting information, the authors just give the result when other parts need to be supported. For example, line 443-446.

Response: This manuscript has merged the content of the original "Section 3.3.1 Diagnostic mass ratios" and the original "Section 3.3.2 Linear correlations", so there is not subtitle in Part 3.3. That is to say, the correlation of C_2, C_3, and C_4, the correlation of C_2% and C_2/C_4, the correlation of Pyr, ωC_2, and MeGly, and the relevant description of chlorophyll a in the original "Section 3.3.2 Linear correlations" has been moved into the "Section 3.3.1 Diagnostic mass ratios", and the rest of original "Section 3.3.2 Linear correlations" has been moved to the supporting information.

3. The authors just compare their data with references, but the discussions are not enough.

Line 158-159, Line 161-164, Line 220 “Oxoacids showed a predominance of ωC2 or ωC3 in five sampling areas (Fig. 3).” Line 221-224.

Response: Thanks for the reviewer for pointing this deficiency. Regarding original “Line 158-159”, the revised manuscript has added the following discussion after it:

“Therefore, combined with the geographical location of SLDP and the fact that the samples were collected in the ship's docking port, it is speculated that the organic aerosol samples of SLDP may be affected by local coastal and terrestrial biological sources and anthropogenic emissions (especially fossil fuel combustion).” (see Page 8, Line 233–236)

Regarding original “Line 161-164”, the revised manuscript has added the following discussion after it: “It can be clearly seen that the concentrations of C_2 in SCS and Malacca were higher than those in EIO-SL and EIO-WI (Table 1). The large amount of C_2 can be generated from the following sources: fossil fuel combustion (Kawamura and Kaplan, 1987;Donnelly et al., 2010), biomass combustion (Schauer et al., 2001;Narukawa et al., 1999), cooking emissions (Kawamura and Kaplan, 1987;Rogge et al., 1993), photooxidation of VOCs and other precursors (Kawamura and Yasui, 2005;Kundu et al., 2010). The diurnal variation trend of C_2 was similar to that of C_3 and C_4, indicating that these compounds may have similar photochemical oxidation pathways or emission sources in the atmosphere.” (see Page 9, Line 242–248)

Regarding original “Line 220-224”, the phrase “Oxoacids showed a predominance of ωC_2 or ωC_3 in five sampling areas (Fig. 3)” has been deleted from the revised manuscript because it overlaps with the next paragraph. And, we have added the following discussion after original “Line 221-224”: “The spatial distributions of ω-oxoacids showed a pattern of SLDP > SCS > Malacca > EIO-SL > EIO-WI, being consistent with those of major diacids (C_2, C_3, and C_4), which indicated that these oxoacids were potential precursors of dicarboxylic acids (Sempéré and Kawamura, 1994).” (see Page 11, Line 305–307)

4. There are many repetitions in the article not only the example below.

The sources of C_9 and the relation of C_9 with other carboxylic acids were discussed repeatedly in part 3.1.1.

Part 3.3.1, the meaning of C_6, Ph, MeGly, Gly, Pyr, Isoprene has been given in the former part, delete the repetitions.

Response: We have reorganized the contents of Section 3.2.1 (original Section 3.1.1) and merged them into four paragraphs. That is, the first paragraph introduces C_2–C_4, the second paragraph introduces C_9, the third paragraph introduces C_6, and the fourth paragraph introduces

Ph. The logic of the revised manuscript is clearer. In addition, we have deleted the meanings of C_6, Ph, MeGly, Gly, Pyr and isoprene in Section 3.4 (original Section 3.3.1).

Minor comments:

Line 254 "C_{10}" to "C_{12}".

Response: Thanks. We have corrected the mistake in the revised manuscript: "Interestingly, the relative abundances of C_2 to total mass concentrations of C_2 to C_{12} diacids (ΣC_2–C_{12}) were similar in four regions (Fig. 5)." (see Page 12, Line 343–344)

Line 386 delete "isoprene and/or aromatic hydrocarbon oxidation products".

Response: Corrected.

Line 287, "It is worth noting that both C3 and C4 acids show a net loss…"

Response: Thanks. This sentence has been deleted from the revised manuscript because it does not make sense to place it there.

Line 202, delete one "that".

Response: We have corrected the mistake in the revised manuscript: "Temporal variations in C_9, C_6, and C_2−C_4 diacids were also similar (Fig. 3), suggesting that photochemical breakdown of C_9 might be the major formation pathway of short-chain diacids such as C_6, C_5, and C_4 diacids." (see Page 10, Line 280–282)

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