

# ***Interactive comment on “Molecular distribution and stable carbon isotopic compositions of dicarboxylic acids and related SOA from biogenic sources in the summertime atmosphere of Mt. Tai in the North China Plain” by Jingjing Meng et al.***

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Anonymous Referee #4

General comments:

1. Comments: This manuscript summarizes results from a summertime study at Mt. Tai in China where aerosol samples, ozone, and environmental parameters were measured. The authors determine daytime/nighttime concentrations and  $\delta^{13}\text{C}$  of various carboxylic acids in an effort to characterize the role of bVOCs on SOA formation and

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aging. The paper is overall well-written although some parts need improvements. My major concern is about interpretation of the daytime/nighttime data. First of all, the standard deviations of the average values presented throughout the paper are rather large which mean that although the average values may be slightly different during day and night, statistically there's not a significant difference between the observations. These need to be addressed/corrected throughout the paper. Secondly, as indicated in L171-172, nighttime data represent free tropospheric measurements, meaning whatever was left in the residual layer from daytime, so nighttime observations aren't truly representing nighttime emissions/oxidations. The most unique aspect of the paper is the  $\delta^{13}\text{C}$  analysis and interpretation of the results. I therefore support publishing the manuscript with major revisions after the authors have addressed my comments above as well as the other scientific comments and editorial suggestions listed below.

Response: We thank the reviewer for the helpful comments above. In the revised version, we have performed a statistic test (i.e., Student's t-test) to verify if the day and night aerosol chemistry is of significant difference. As shown in the Table S1, the concentrations and compositions of major species in PM<sub>2.5</sub> between day and night show a P value less than 0.005, which clearly demonstrates that the abundances and compositions of the major species during the day and night are statistically different. Figure S1 also shows that during the nighttime sulfate decreased while nitrate and ammonium increases. Such a diurnal change in inorganic ion compositions further suggests the significant difference in aerosol chemistry between day and night. Related statements have been added into the text. Please see Line 211-217 in Page 8 and Line 223-233 in Page 8-9, respectively.

Scientific Comments:

2. Comments: L40: define C6, Ph, Gly, and mGly (also in L249). Also how are the ratios mentioned here expected to behave for anthropogenic precursors?

Response: Suggestion taken. We have defined C6, Ph, Gly, and mGly in the revised

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manuscript. Please see Line 42-43 in Page 2 and Line 310-311 in Page 12, respectively. C6 and Ph are believed to be formed via secondary oxidations of anthropogenic cyclic olefins (e.g., cyclohexene) (Hatakeyama et al., 1987) and aromatic hydrocarbons, respectively. C9 is mainly produced from photochemical oxidation of oleic acid, which is a biogenic unsaturated fatty acid containing a double bond at the C-9 position (Wang et al., 2010). Therefore, both ratios of C9/C6 and C9/Ph are indicative of the source strengths of biogenic versus anthropogenic emissions. Please see Line 362–370 in Page 13-14 of the revised manuscript. Model simulation (Fu et al., 2008) and field observations (Meng et al., 2014) have suggested that the concentration ratio of particulate Gly/mGly is about 1:5 when biogenic sources are predominant and is about 1:1 when anthropogenic sources are predominant. Therefore, the mass ratio of particulate Gly/mGly is also indicative of the source strengths of biogenic versus anthropogenic emissions. Please see Page 14, Line 377-380.

3. Comments: L41: how is ‘related SOA with biogenic precursor’ defined/ determined?

Response: Thanks for your suggestion. “related SOA” is defined as “major dicarboxylic acids and related SOA”, which consists of major dicarboxylic acids, ketocarboxylic acids and  $\alpha$ -dicarbonyls (i.e., C2, C3, C4,  $\omega$ C2, Pyr, Gly and mGly), while “biogenic precursor” is defined as “SOA tracers derived from isoprene-,  $\alpha$ -/ $\beta$ -pinene- and  $\beta$ -caryophyllene”. We have added these definitions into the revised manuscript. Please see Page 2, Line 42-46. As for the analysis method of biogenic precursors, we have added the related information as follows: The analysis method of biogenic precursors has been reported elsewhere (Li et al., 2013). Briefly, one fourth of the filter was cut and extracted with a mixture of dichloromethane and methanol (2:1, v/v) under ultrasonication. The extracts were concentrated using a rotary evaporator under vacuum conditions and then blow down to dryness using pure nitrogen. After reaction with a mixture of N,O-bis-(trimethylsilyl) trifluoroacetamide (BSTFA) and pyridine (5:1, v/v) at 70°C for 3 h. Biogenic secondary organic aerosol (BSOA) tracers in the derivatized samples were determined by GC-MS. These data were used in this study to explore

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the biogenic sources of dicarboxylic acids and related SOA. Please see Page 6-7, Line 165-173.

4. Comments: L164: What's the influence of organic acids on pH? Since the AIM model incorporates only the major inorganic ions, how do you think your pH results are affected by the presence of organic acids?

Response: The Extended AIM Thermodynamic Model (E-AIM, Model II, <http://www.aim.env.uea.ac.uk/aim/>) was employed to calculate aerosol liquid water content (LWC) and in-situ particle pH (pHIS). E-AIM II is an equilibrium thermodynamic model that can simulate liquid and solid phase of ionic compositions accurately in the SO<sub>2</sub>–NO<sub>3</sub>–NH<sub>4</sub><sup>+</sup>–H<sup>+</sup> system under certain temperature and relative humidity (Clegg et al., 1998a; Li et al., 2013). Compared with SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup>, organic acid has little influence on estimation of aerosol acidity due to their low abundance in aerosols (Zhou et al., 2018). Organic acids may contribute free H<sup>+</sup> in aerosol aqueous phase and affect partitioning/dissociation of inorganic species in acidic particles. However, Huang et al. (2010) found that oxalic acid, the single most abundant organic acid, contributed little to the free acidity of rain water. This implied that the contribution of organic acids to aerosol free H<sup>+</sup> in the Mt. Tai aerosols was most likely minor. Moreover, quantity of free H<sup>+</sup> released from organosulfates was proved to be small compared with that estimated from inorganic anion and cation balance. Hygroscopicity of organic compounds are significantly weaker than inorganic species such as NH<sub>4</sub>NO<sub>3</sub> and (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> (Ansari and Pandis, 2000). Therefore, organic species generally have minor influence on pHIS. So, we did not take into account the organic species, neither did previous studies about the in-situ acidity of aerosol (Li et al., 2013; Meng et al., 2014; Xue et al., 2011). Both field observation and laboratory simulation suggest that oxalic acid is largely derived from the acid-catalyzed heterogeneous oxidation of glyoxal and related precursors in the aqueous phase. As shown in Fig. 6f in the revised manuscript, oxalic acid exhibits a significant negative correlation with pHIS for the daytime (R<sup>2</sup>=0.68) and nighttime (R<sup>2</sup>=0.75) samples, respectively, suggesting that acidic conditions are favorable for

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the formation of organic acids in the aerosol aqueous phase.

5. Comments: L177: Looking at the observed variability in the values presented in Table 1, there isn't a significant change between daytime and nighttime concentrations although the average for some species is higher during daytime. I think this point needs to be clarified here and throughout the paper.

Response: The variability of the data presented in Table 1 and throughout the paper is large, but our statistic t-test analysis results (Table S1) show that both concentrations and mass ratios between day and night are significantly different. Therefore, our statements, such as "the higher SOA concentrations and mass ratios in daytime are due to the strong photochemical oxidation" and other related discussions, are reasonable. We have also modified the related discussions as follows: OC and WSOC in the PM<sub>2.5</sub> samples in daytime are similar to those in nighttime (Table 1), but OC/EC and WSOC/OC ratios are around 1.4 times higher in daytime than in nighttime (Fig. 4), indicating an enhancing SOA production due to the stronger photochemical oxidation in daytime rather than the changes in the boundary layer heights (Hegde and Kawamura, 2012). Please see Page 8, Line 218-222.

6. Comments: L217: is the vegetation (tree types) also similar at this location and Mt. Fuji? I think that's more important rather than latitude and altitude of the sites.

Response: We have checked the vegetation compositions at Mt. Tai and Mt. Fuji, and found both are dominated by broad-leaved forest. We have modified the related statement. Please see Page 10, Line 269-270.

7. Comments: L243-245: Based on the average values in the table, total  $\alpha$ -dicarbonyls were actually higher in daytime, so I don't think the data support the statement mentioned here. Do you mean only glyoxal and methylglyoxal? Also based on the graph, it seems on most days the difference between daytime and nighttime total conc. of  $\alpha$ -dicarbonyls was minor, so the pattern you're highlighting is not consistent. Please clarify.

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Response: In the current work, total  $\alpha$ -dicarbonyls means only glyoxal and methylglyoxal. As seen in Table 2, concentrations of  $\alpha$ -dicarbonyls were  $19 \pm 11$  ng/m<sup>3</sup> in daytime, which were lower than that ( $27 \pm 17$  ng/m<sup>3</sup>) in nighttime. As we mentioned before, we have done a t-test analysis to check if the diurnal difference is of a statistic meaning (Table S1). The t-test results show that concentrations and mass ratios of major species between day and night are of a P-value less than 0.005, which clearly suggests that the diurnal differences are statistically significant.

8. Comments: L265-267: I'm not understanding the difference between the beginning statement and the second part of the sentence. C4 is still a longer-chain diacid compared to C3. Please clarify and be more precise in what constitutes a longer-chain diacid.

Response: Suggestion taken. The longer-chain diacids are defined as the number of carbons contained in the diacids is larger than four such as C5-C11. To clarify this, we have revised the sentence as follows: Previous studies have proposed that the hydroxylation of C4 can be further oxidized into C2 and C3, and C3 can also be oxidized into C2 through intermediate compounds such as hydroxymalonic acid or ketomalonic acid. Please see Page 12, Line 328-330.

9. Comments: L269-275: Again it seems that given the variability of the observed diacid ratios in this study and those of previous studies, there's no significant difference between observations at different photochemical ages. I'm not convinced the conclusions regarding age are strongly supported by the data.

Response: As seen in our previous response, we did a statistic analysis, which shows that the diurnal variation is statistically significant.

10. Comments: L275-277: Photochemical oxidation is stronger compared to what? Nighttime or aqueous oxidation? Please clarify.

Response: Thanks for your suggestion. Photochemical oxidation is stronger in the

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daytime than that in the nighttime. We have revised the old sentence as follows: Compared with those in the nighttime, the higher ratios of C2/C4 and C3/C4 (Fig. 4) in the daytime again indicated that the photochemical modification of aerosols is stronger. Please see Page 13, Line 339-341.

11. Comments: L298-299: again given the variability observed in the daytime/nighttime data, the difference in the average values isn't significant.

Response: Here we agree with the comments above. The t-test analysis shows that differences in ratios of C9/C6 and C9/Ph between day and night are not significant. Thus, we have revised the related discussions as follows: As shown in Fig. 4, both ratios of C9/C6 and C9/Ph are similar in the daytime to those in the nighttime. Please see Page 14, Line 370-371.

12. Comments: L328: what are the SOA tracers from these compounds? It will be useful to add to the legend in Table 4 what parent hydrocarbon is associated with each tracer.

Response: Suggestion taken. The SOA tracers from these compounds consist of the secondary organic aerosols (SOA) derived from isoprene,  $\alpha$ -/ $\beta$ -pinene and  $\beta$ -caryophyllene. We have added this explanation to the legend in Table 4. Please see the red words in Page 31, Table 4.

13. Comments: L339: some of the acids related to  $\alpha$ -pinene and  $\beta$ -caryophyllene oxidation also correlate well with the diacids, so why only highlight isoprene contribution to SOA? In fact, the following sentence indicates that bSOA in general control production of the diacids, so perhaps it's better to combine these few sentences together.

Response: Suggestion taken, we have combined these sentences together. Please see Page 15, Line 412-419.

14. Comments: L360: I disagree with the statement that daytime ozone oxidation of isoprene and  $\alpha$ -pinene was more important than OH oxidation of these compounds.

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Lifetime of these bVOCs even at background OH levels is a lot shorter than with respect to ozone oxidation. The observed correlation is just a correlation and not a causality. Related to this, I think the conclusion in the sentence starting in L454-455 needs to be removed.

Response: We agree with the reviewer on the comments above. We have revised the discussions about the reason why the lack of correlation was observed between BSOA tracers and O<sub>3</sub> at night as follows: These results suggest that the daytime oxalic acid and related SOA in the mountaintop of Mt. Tai are largely derived from O<sub>3</sub> and OH radical oxidation of BVOCs such as isoprene and  $\alpha$ -pinene, while the nighttime oxalic acid and related SOA might be mostly produced by NO<sub>3</sub> radical and other oxidizing agents such as H<sub>2</sub>O<sub>2</sub> (Claeys et al., 2004; Herrmann et al., 1999). In addition, the titration of O<sub>3</sub> by the residual NO in the nighttime atmosphere could also be responsible for the lack of the correlation between BSOA tracers and O<sub>3</sub>. Please see Page 16, Line 434-441. We have deleted the conclusion in the sentence starting in L454-455 in the old version manuscript.

15. Comments: L366-368: SOA formation potential under different oxidants are also different, and so can contribute to the higher observed tracers during daytime.

Response: We agree with the comments above that SOA formation potential under different oxidants are different, which could contribute to the higher observed tracers during daytime. As seen in Table 1, O<sub>3</sub> concentration during the sampling period is 50% higher in daytime than in nighttime, clearly indicating that oxidation potential in daytime at the Mt. Tai site is stronger. Moreover, isoprene is only emitted by trees during daytime. Thus, we think the higher loadings of BSOA tracers in daytime are caused not only by stronger photochemical oxidation but also by enhanced emissions of BVOCs. We have revised the statement. Please see Page 16-17, Line 446-452.

16. Comments: L381-382: In addition to the similar formation pathways (aqueous oxidation) for sulfate and oxalic acid, since oxalic acid formation is acid-catalyzed, one

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expects to have a good correlation with sulfate and oxalic acid (since the site is far from agricultural sources, I'm assuming most of the sulfate is acidic). Please add this discussion as a contributor to the good correlation as well.

Response: Suggestion taken. Please see Page 17, Line See 462-463.

17. Comments: L393: aerosol composition is also very important for determining LWC of aerosols at a given RH.

Response: We agree with the comments above. Aerosol LWC in this study was calculated by using AIM-II model, which considered a  $\text{SO}_4^{2-}\text{NO}_3^- \text{NH}_4^+\text{H}^+$  system and allowed variable temperature and relative humidity (Clegg et al., 1998b; Li et al., 2013), therefore aerosol LWC is controlled by the ambient relative humidity (Clegg et al., 1998b) and the concentrations of inorganic salt (Fountoukis and Nenes, 2007). As shown in Table 1 of the revised manuscript, the total concentration ( $21.7 \pm 11.5 \mu\text{g m}^{-3}$ ) of  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$  and  $\text{NH}_4^+$  during the daytime was almost equivalent to that ( $20.4 \pm 8.2 \mu\text{g m}^{-3}$ ) during the nighttime. However, the relative humidity ( $92 \pm 5.0\%$ ) in daytime was higher than that in nighttime ( $77 \pm 8.2\%$ ). Therefore, we only discussed the effect of RH on aerosol LWC. We have changed old sentence as follows: Both RH and aerosol composition are key factors controlling the aerosol LWC. Please see Page 18, Line 480-481.

18. Comments: L428-429: Please indicate here specifically what trends in the ratios would suggest aging.

Response: Suggestion taken. The higher the mass ratios of  $\text{C}_2/\omega\text{C}_2$ ,  $\text{C}_2/\text{Gly}$  and  $\text{C}_2/\text{mGly}$ , the more aged the organic aerosol. To express clearly, we have revised the old sentence as follows: Thus, the higher mass ratios of  $\text{C}_2/\omega\text{C}_2$ ,  $\text{C}_2/\text{Gly}$  and  $\text{C}_2/\text{mGly}$  indicate that organic aerosols are more aged (Wang et al., 2017). Please see Page 19, Line 518-519.

19. Comments: L440-442: I'm a bit confused about this sentence. Higher values of

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glyoxal and methyl glyoxal relative to what? Please clarify. Also, from sentence above, I was under the impression that freshly emitted BVOCs are depleted in  $^{13}\text{C}$ , so why do the authors indicate that Gly and mGly are formed from oxidation of bVOCs enriched in  $^{13}\text{C}$ ?

Response: Suggestion taken. The  $\delta^{13}\text{C}$  values of Gly ( $-17.4\pm 1.9$ ) and mGly ( $-18.8\pm 2.1$ ) were relatively higher than fresh BVOCs such as isoprene ( $-32\text{‰}$  –  $-27\text{‰}$  emitted directly from vegetation. To express this point clearly, we have revised the related sentence as follows: Therefore, the  $\delta^{13}\text{C}$  values of Gly and mGly are relatively higher than fresh BVOCs such as isoprene, largely attributed to the secondary formation from the oxidation of isoprene and other biogenic precursors. Please see Page 19-20, Line 531-533.

20. Comments: L448 (also in the abstract): Indicate that ‘average’ concentration of some species are higher in the day compared to night since as mentioned above, the variability in the measured data was too high to conclude beyond the average.

Response: As mentioned above, our t-test analysis showed that the diurnal difference is significant, although the variability in the measured data was very high.

Minor Comments:

21. Comments: L56: consider changing to “... of which up to 80% are water soluble”.

Response: Suggestion taken. Please see Page 3, Line 59.

22. Comments: L73: “... C2 is largely produced...”.

Response: Suggestion taken. Please see Page 3, Line 75.

23. Comments: L80: change “liquid water content-enriched aerosol” to “aqueous aerosol”.

Response: Suggestion taken. Please see Page 3, Line 81-82.

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24. Comments: L87: change “independent” to “isolated”.

Response: Suggestion taken. Please see Page 4, Line 89.

25. Comments: L88: change “One of the severest air-polluted regions” to “one of the regions with worst air pollution in the world”.

Response: Suggestion taken. Please see Page 4, Line 90.

26. Comments: L90: change “few information” to “little information”.

Response: Suggestion taken. Please see Page 4, Line 95-96

27. Comments: L109: indicate also the altitude of the sampling site in the main text.

Response: Thanks for your suggestion. We have added the altitude of the sampling site in the revised manuscript. Please see Page 5, Line 128-130.

28. Comments: L121: “site”.

Response: Suggestion taken. We corrected the typo, see Page 4, Line 142.

29. Comments: L152 ad L163: “in-situ particle pH”.

Response: Suggestion taken. Please see Page 7, Line 186-187.

30. Comments: L159: add “. . . to remove insoluble particles. . .”.

Response: Suggestion taken. Please see Page 7, Line 193-194.

31. Comments: L222: replace “difference” with “pattern”.

Response: Suggestion taken. Please see Page 10, Line 276.

32. Comments: L238: The sentence is too long. Consider starting a new sentence after the references.

Response: Thanks for your suggestion. We have changed the old sentences as follows: Ketocarboxylic acids are the major intermediates of aqueous phase photochemi-

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cal oxidation producing dicarboxylic acids in the atmosphere (Kawamura and Ikushima, 1993; Pavuluri and Kawamura, 2016). The concentrations of ketocarboxylic acids are  $43 \pm 28$  ng m<sup>-3</sup> in the daytime and  $37 \pm 19$  ng m<sup>-3</sup> in the nighttime, respectively, with glyoxylic acid ( $\omega$ C2) being the dominant  $\omega$ -oxoacid, followed by pyruvic acid (Pyr) and 3-oxobutanoic acid ( $\omega$ C3) (Table 2 and Fig. 2). Please see Page 11, Line 291-296.

33. Comments: L246: consider replacing “precursors” to “compounds”.

Response: Suggestion taken. We have rephrased the sentences, please see Page 11, Line 302-303.

34. Comments: L248: consider replacing “impressed” with “more significant”.

Response: Suggestion taken. Please see Page 11-12, Line 308-310.

35. Comments: L258: “by wet deposition”.

Response: Suggestion taken. Please see Page 12, Line 320-321

36. Comments: L265: “by photochemical degradation”.(?)

Response: We have rephrased the expressions as follows: Previous studies have proposed that the hydroxylation of C4 can be further oxidized into C2 and C3, and C3 can also be oxidized into C2 through intermediate compounds such as hydroxymalonic acid or ketomalonic acid Please see Page 12, Line 328-330.

37. Comments: L278: delete ‘would’.

Response: Suggestion taken. Please see Page 13, Line 345.

38. Comments: L282: “at Mt. Tai”.

Response: Suggestion taken. Please see Page 13, Line 353.

39. Comments: L289: consider changing “troposphere” to “atmosphere”.

Response: Suggestion taken. Please see Page 13, Line 361.

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40. Comments: L377: “linear”.

Response: Suggestion taken. Please see Page 17, Line 463.

41. Comments: L447: either “ground” or “surface”; probably don’t need to have both words.

Response: Thanks for your suggestion. We have deleted “surface”. Please see Page 20, Line 538-539.

42. Comments: Table1: is the upper end of RH at night 93% rather than 193%?

Response: Sorry for the mistake, we had corrected it. Please see Table 1 in Page 29.

43. Comments: Change the order of Fig. 6 and Fig. 5 as Fig. 6 is referred to before Fig. 5. Also it seems the next Figure that authors refer to is Fig. 8. Please use the figures in the same order they appear in the text.

Response: We disagree on the comments above. Fig. 5 was used to elucidate the dicarboxylic acids and related SOA are mostly derived from the local sources rather than long-range transport, which was discussed in Part 3.3. Fig. 6 was used to investigate the effects of temperature, relative humidity, and O<sub>3</sub> concentrations on the formation of oxalic acid and related SOA, which was discussed in Part 3.5. Thus, Fig. 5 should be shown before Fig. 6.

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