

Response to the comments of Anonymous Referee #3

Comment 1 - This paper presents results from analysis of filter samples collected over the past decade at the Gosan site on Jeju Island, South Korea. The filter samples were analyzed for diacids. The diacids are used as surrogate compounds to examine the trend of secondary organic aerosols (SOA) in atmospheric aerosols over the past decade. The seasonality, distributions, temporal variations, and intensity increases of the diacids are explored.

The change in SOA has been predicted to be highly uncertain in the future atmosphere in Asia. This work aims to try to better understand this by examining the trends in diacids. Given that air quality regulations are becoming stricter and the results from this work could have implications on control strategies for Asia, many in the atmospheric community would be interested in this work.

Overall, this is a good paper. The methods and analyses presented seem scientifically sound. The one main comment I have is that it can be a bit confusing using the words saturated and unsaturated acids in the text, but then naming the specific acids in the figures. Maybe it would be helpful to add the words saturated and unsaturated into the figure captions. Otherwise, I really just have a number of comments to help with the flow of the paper, which are outlined below and need to be addressed before the paper can be considered for publication

Response 1 - Thank you so much. We have added the terms “saturated” and “unsaturated” in the captions of Figures 1 -2 and Figures 5 - 8. Please see below point-by-point responses. Please see lines 554-564 and 576-601 in the revised manuscript.

Comment 2 - General Comments: -It is not clear what order is being used for the reference citations throughout the text. Sometimes alphabetical order is used, other times chronological order is used. Other way is technically fine, but it should be consistent throughout the text. I have noted below the ones I noticed.

Response 2 - The in-text citations have been arranged chronologically throughout the manuscript now. Thank you so much.

Comment 3 - Specific Comments:

1) Abstract Page 22184, Lines 3-4 – Suggest changing we study a sub-decadal to we examine the sub-decadal

Response - Done. Please see line 16 in the revised manuscript.

2) Page 22184, Line 5 – Suggest changing from Gosan site in Jeju to from the Gosan site on Jeju. Also suggest adding a The before Gosan site

Response – Corrected. Please see line 17 in the revised manuscript.

3) Page 22184, Line 6 – Suggest removing the the before pollution-outflows

Response - Fixed. Please see line 18 in the revised manuscript.

4) Page 22184, Line 14 – The chemical abbreviation is not defined

Response - Done. Please see line 26 in the revised manuscript.

5) Page 22184, Line 16 - Suggest removing the an before enhanced

Response - Corrected. Please see line 28 in the revised manuscript.

6) Introduction Page 22185, Line 3 – Suggest removing the The before source

Response - Fixed. Please see line 42 in the revised manuscript.

7) Page 22185, Lines 4-5 – It is not clear the order that is being used for the references

Response - Done. Please see line 43-44 in the revised manuscript.

8) Page 22185, Line 9 - It is not clear the order that is being used for the references

Response - Corrected. Please see line 47 in the revised manuscript.

9) Page 22185, Line 10 - It is not clear the order that is being used for the references

Response - Modified. Please see line 48 in the revised manuscript.

10) Page 22185, Lines 13-15 - It is not clear the order that is being used for the references

Response - Done. Please see line 51-52 in the revised manuscript.

- 11) Page 22185, Line 15 - Suggest changing emit from to be emitted from
Response - Fixed. Please see line 53 in the revised manuscript.
- 12) Page 22185, Line 17 - Suggest removing the the before precursors
Response - Done. Please see line 54 in the revised manuscript.
- 13) Page 22185, Line 19 - Suggest adding a the before diacids and a for before as much
Response - Done. Please see line 57 in the revised manuscript.
- 14) Page 22185, Line 22 - Suggest adding a the before water-soluble
Response - Corrected. Please see line 59 in the revised manuscript.
- 15) Page 22186, Line 3 – Suggest removing the The before chemistry-transport
Response - Done. Please see line 68 in the revised manuscript.
- 16) Page 22186, Lines 5-6 - It is not clear the order that is being used for the references
Response - Done. Please see line 69-70 in the revised manuscript.
- 17) Page 22186, Line 10 - Suggest changing emission has been to emissions have been
Response - Corrected. Please see line 75 in the revised manuscript.
- 18) Page 22186, Lines 11-12 - It is not clear the order that is being used for the references
Response - Edited. Please see line 75-76 in the revised manuscript.
- 19) Page 22186, Line 15 – Suggest changing an importance to the importance
Response - Edited. Please see line 79 in the revised manuscript.
- 20) Page 22186, Line 17 – Suggest removing the the after influenced by
Response - Done. Please see line 81 in the revised manuscript.
- 21) Page 22186, Line 20 – Suggest removing the The before East
Response - Done. Please see line 83 in the revised manuscript.
- 22) Experimental section 2.1.Site description Page 22187, Line 3 – Suggest changing Gosan site in Jeju to The Gosan site on Jeju
Response - Done. Please see line 90 in the revised manuscript.
- 23) Page 22187, Line 4 - Figure 3 is referred to before Figures 1 and 2
Response - The issue has been fixed by referring Fig. 3 as Fig. 1 and the Figs. 1- 2 have been referred as Figs. 2- 3.
- 24) Page 22187, Line 4 – Suggest removing the off the before south and adding a the before Korean
Response - Done. Please see line 91 in the revised manuscript.
- 25) Page 22187, Line 5 – Suggest removing the off the before east
Response - Done. Please see line 92 in the revised manuscript.
- 26) Page 22187, Line 6 – Suggest removing the off the before west and before northeast
Response - Done. Please see line 92 in the revised manuscript.
- 27) 2.2. Aerosol sampling Page 22187, Line 15 – Suggest changing cannot be collected to could not be collected
Response - Done. Please see line 103 in the revised manuscript.
- 28) Page 22187, Line 20 – Suggest changing jar to jars
Response - Done. Please see line 108 in the revised manuscript.
- 29) Page 22187, Line 21 – Suggest changing was used to were used
Response - Done. Please see line 108 in the revised manuscript.
- 30) 2.3. Analytical method Page 22188, Line10 – What does the phrase dissolved into n-hexane layer mean? It is not clear from the text.
Response - We have re-written “n-hexane layer” as n-hexane to avoid any confusion. Please see line 123.
- 31) Page 22188, Line 11 – When does the water get added to the extraction? Is it before the derivatization?
Response - To avoid the confusion, we have re-phrased the content as “Water and n-hexane were simultaneously added to isolate derivatized esters and acetals from inorganic

materials such as boric and fluoric acids. Esters and acetals are transferred into n-hexane whereas inorganic acids are dissolved into water. The hexane layer was separated and completely dried using a rotary evaporator under vacuum and nitrogen (N₂) blow-down technique.” Please see lines 121-126 in the revised MS.

- 32) Page 22188, Line 11 – The chemical abbreviation is not defined
Response - Done. Please see line 125 in the revised manuscript.
- 33) Page 22188, Line 12 – Suggest adding a the before esters
Response - Done. Please see line 125 in the revised manuscript.
- 34) Page 22188, Line 14 – The abbreviation GC is not defined
Response - Done. Please see line 127 in the revised manuscript.
- 35) Page 22188, Line 16 – The abbreviation FID is not defined
Response - Done. Please see line 129 in the revised manuscript.
- 36) Page 22188, Line 20 – The abbreviation MS is not defined
Response - Done. Please see line 133 in the revised manuscript.
- 37) Page 22189, Line 13 – The chemical abbreviation is not defined
Response - Done. Please see line 153 in the revised manuscript.
- 38) 3. Results and discussion 3.1. Interannual variations in the molecular distribution of diacids Page 22189, Line 18 – Suggest removing the an before important
Response - Done. Please see line 163 in the revised manuscript.
- 39) Page 22189, Line 26 – Suggest changing of individual year to of the individual years
Response - Done. Please see line 170 in the revised manuscript.
- 40) Page 22190, Lines 6-7 - It is not clear the order that is being used for the references
Response - Done. Please see line 175-177 in the revised manuscript.
- 41) 3.2. Seasonal variations of diacids Page 22190, Line 22 - The chemical abbreviation is not defined
Response - Done. Please see line 192 in the revised manuscript.
- 42) Page 22190, Line 26 – The chemical abbreviation is not defined
Response - Done. Please see line 198 in the revised manuscript.
- 43) Page 22190, Line 27 – Suggest removing the before satellite
Response - Done. Please see line 199 in the revised manuscript.
- 44) Page 22191, Line 3 – Suggest changing least to low
Response - Done. Please see line 202 in the revised manuscript.
- 45) Page 22191, Line 5 – Suggest adding a the before Gosan
Response - Done. Please see line 204 in the revised manuscript.
- 46) Page 22191, Lines 10-11 - It is not clear the order that is being used for the references
Response - Done. Please see line 209-210 in the revised manuscript.
- 47) Page 22191, Line 12 – Suggest removing the before atmospheric
Response - Done. Please see line 210 in the revised manuscript.
- 48) Page 22192, Line 5 – Suggest adding a the before major
Response - Done. Please see line 231 in the revised manuscript.
- 49) Page 22192, Line 6 – Suggest adding a the before 95%
Response - Done. Please see line 232 in the revised manuscript.
- 50) Page 22192, Line 18 – Suggest changing alike to like
Response - Done. Please see line 243 in the revised manuscript.
- 51) Page 22192, Line 19 – Suggest changing the peaks were emerged to a peak emerged
Response - Done. Please see line 243-244 in the revised manuscript.
- 52) Page 22192, Line 21 – Suggest removing the the before atmospheric
Response - Done. Please see line 245 in the revised manuscript.
- 53) Page 22192, Line 26 – Suggest adding a the before seasonality
Response - Done. Please see line 250-251 in the revised manuscript.

54. Page 22192, Line 28 – Suggest changing generate onto aqueous to be generated in Aqueous

Response - Done. Please see line 252 in the revised manuscript.

55. Page 22193, Line 1 - Suggest adding a the before global

Response - Done. Please see line 253 in the revised manuscript.

56. 3.3. Interannual variability of diacids Page 22193, Line 18 - Suggest adding a the before Major

Response - Done. Please see line 269 in the revised manuscript.

57) Page 22194, Line 3 – Suggest adding a the before diacids

Response - Done. Please see line 281 in the revised manuscript.

58) Page 22194, Line 4 – Suggest changing The increase to An increase

Response - Done. Please see line 282 in the revised manuscript.

59) Page 22194, Line 7 – Suggest removing the the before combustion

Response - Done. Please see line 285 in the revised manuscript.

60) Page 22194, Lines 10-11 - It is not clear the order that is being used for the references

Response - Done. Please see line 288-289 in the revised manuscript.

61) Page 22194, Line 15 – Suggest removing the the after via

Response - Done. Please see line 293 in the revised manuscript.

62) Page 22194, Line 23 – Suggest changing of the cold to in the cold

Response - Done. Please see line 299 in the revised manuscript.

63) Page 22195, Line 1 - It is not clear the order that is being used for the references

Response - Done. Please see line 306 in the revised manuscript.

64) Page 22195, Line 14 – Suggest changing to generate from to to be generated from

Response - Done. Please see line 318-319 in the revised manuscript.

65) 4. Conclusions and atmospheric implications Page 22195, Line 18 – Suggest removing the a before major

Response - The word major has been replaced. Please see line 328 in the revised manuscript.

66) Page 22195, Line 21 – Suggest changing that it is required to that this required

Response - Done. Please see line 327 in the revised manuscript.

67) Page 22196, Line 1 – Suggest changing diacid increases to diacids increasing

Response – Those phrases have been eliminated due to a previous comment.

68) Page 22196, Line 5 – Suggest removing the the before recent

Response - Done. Please see line 335 in the revised manuscript.

69) Acknowledgements Page 22196, Line 18 – Suggest changing available to obtained

Response - Done. Please see line 347 in the revised manuscript.

70) References Page 22197, Lines 16-21 – This reference is the same as the one above it.

Response – The problem has been solved. Please see line 365-370 in the revised manuscript.

71. Tables Table 1 -Sept. is the only month with a period after it

Response - Done. Please see Table 1 in the revised manuscript.

72. Figures Figure 1 -The x-axis for the plots on the left is not labeled

Response - We have fixed it. The Figure 1 has been referred as Figure 2 in the revised manuscript.

73. Figure 2 -The label for the x-axis for the plots on the left is cut-off

Response - We have fixed it. The Figure 1 has been referred as Figure 3 in the revised manuscript.

74. Figure 3 -In caption, suggest adding a the before Gosan and changing in Jeju to on Jeju

Response - We have fixed it. Please see line 554 in the revised manuscript.

75. Figures 5 and 6 -In last line of caption, the acids mentioned for plots g and h do not match the legends. -Plot h is labeled as g -Suggest adding to the caption that the data is segregated by season

Response - The issues have been fixed. Thank you so much. Please see line 579-580 and 585-586 in the revised manuscript.

76. Figure 7 - Suggest adding to the caption that the data is segregated by season

Response - We have fixed it. Please see line 591-592 in the revised manuscript.

77. Figure 8 -In last line of caption, suggest changing the linear to a linear

Response - We have fixed it. Please see line 601 in the revised manuscript.

78. Figure 9 -In last line of caption, suggest changing the linear to a linear

Response - Done. Please see line 608 in the revised manuscript.

79. Supporting Information Tables Table S1 -In first line of caption, suggest changing diacids with to diacids within -In second line of caption, suggest changing the linear regression using least squares to a linear regression using a least squares fit

Response - Done. Please see Table S1 in the revised manuscript.

80) Tables S2 -In third line of caption, suggest changing oxalic acid with to oxalic acid within -In fourth and fifth lines of caption, suggest changing the linear regression using least squares to a linear regression using a least squares fit

Response - Done. Please see Table S2 in the revised manuscript.

81) Figure Captions Figure S5 Caption -In third line of caption, have 10th written with a superscript. Although this is not wrong, nowhere else in the text is in written this way.

Response - Done. Please see line 79 in the revised supplementary materials.