Responses to the comments of Anonymous Referee #1

Comment 1 - This study presents multi-year data of diacids and other aerosol tracer compounds collected in Gosan site in East Asia. The authors investigated the seasonal and interannual variations in these compounds as well as their correlations. They concluded that the dominant sources of the diacides vary with season and the intensity of the pollution sources have increased over years. This study is based on a very comprehensive data set that worth documentation. The data analysis is carefully performed and the manuscript well written with few grammar mistakes and language misuse. For these reasons, I recommend publication of this work on ACP after the authors address my following comments.

Response 1 - Thank you so much. We have carefully checked English throughout the manuscript. Also, we have considered the comments of anonymous referee #3 related to English language.

Comment 2 - Page 22190 Line 11-12: The authors concluded here that the sources of the organic acids remain the same from 2001-2008 based on the similarity of the molecular distribution of the observed diacids. However, the back trajectories in Figure 3 clearly show that at least for July the origin of the averaged air masses sampled at Gosan is distinctly different from other months, which suggests different aerosol sources.

Response 2 - Please note that above conclusion is related to the interannual variations, but not seasonal variations. We have discussed the seasonal variation in section 3.1. To avoid any confusion, we have re-phrased the above conclusion as "To explore interannual differences in the molecular distributions, the analysis of variance (ANOVA) was carried out by comparing the median concentrations of the similar months of 2001-2008. Significant ($p < 0.05$) differences in the molecular distributions of diacids were not observed among the studied years. Based on these results and similarity of mean air mass trajectory pathways among the identical months (exception: May, June, August and September) of 2001-2008 (as shown in Fig. 1a-d and Fig. S1a-h), we conclude that the sources of diacids mostly remain identical in East Asia over a sub-decadal scale." Please see lines 177-183 in the revised manuscript.

Comment 3 - Page 22191 Line 17-23: What about the correlation between saturated diacids and CO in spring, which could provide evidence for diacids being from anthropogenic sources?

Response 3 - Thank you so much for this important comment. We did not discuss the correlation between saturated diacids and CO in the previous manuscript as the time resolution of diacid data (2-7 days), and CO (1-hr) data were quite different. According to the comment, we investigated the correlations of diacids with CO data over the study period (2001-2008). The seasonal average, median and percentile (25^{th} and 75^{th}) concentrations of CO do not correlate with those of diacids concentrations. We believe that the lack of correlations is due to the different time resolution between data.

Comment 4 - Page 22195 Line 18-19: This statement is confusing and should be re-worded. As the authors discussed in previous sections, the pollution sources of diacids are not identical but vary with season, i.e. anthropogenic in spring, biogenic in summer and biomass burning in winter, and differ for different types of diacids (saturated vs. unsaturated). Also, I

don't think that the authors have evidence to show that the diacids is "a major surrogate" of SOA in this study. I suggest changing "a major surrogate" to "an important fraction".

Response 4 - To avoid the confusion, we have re-phrased the sentence as "The seasonal variations in the pollution sources of diacids, an important fraction of organic aerosols (OA), remain identical over a sub-decadal scale in East Asia. But, the strength of their pollution sources has increased from 2001 to 2008, which is evidenced by the increases in the loadings of diacids in atmospheric aerosols." Please see lines 323-326 in the revised manuscript. Note that we have replaced the term "a major surrogate" to "an important fraction" in the above discussion.

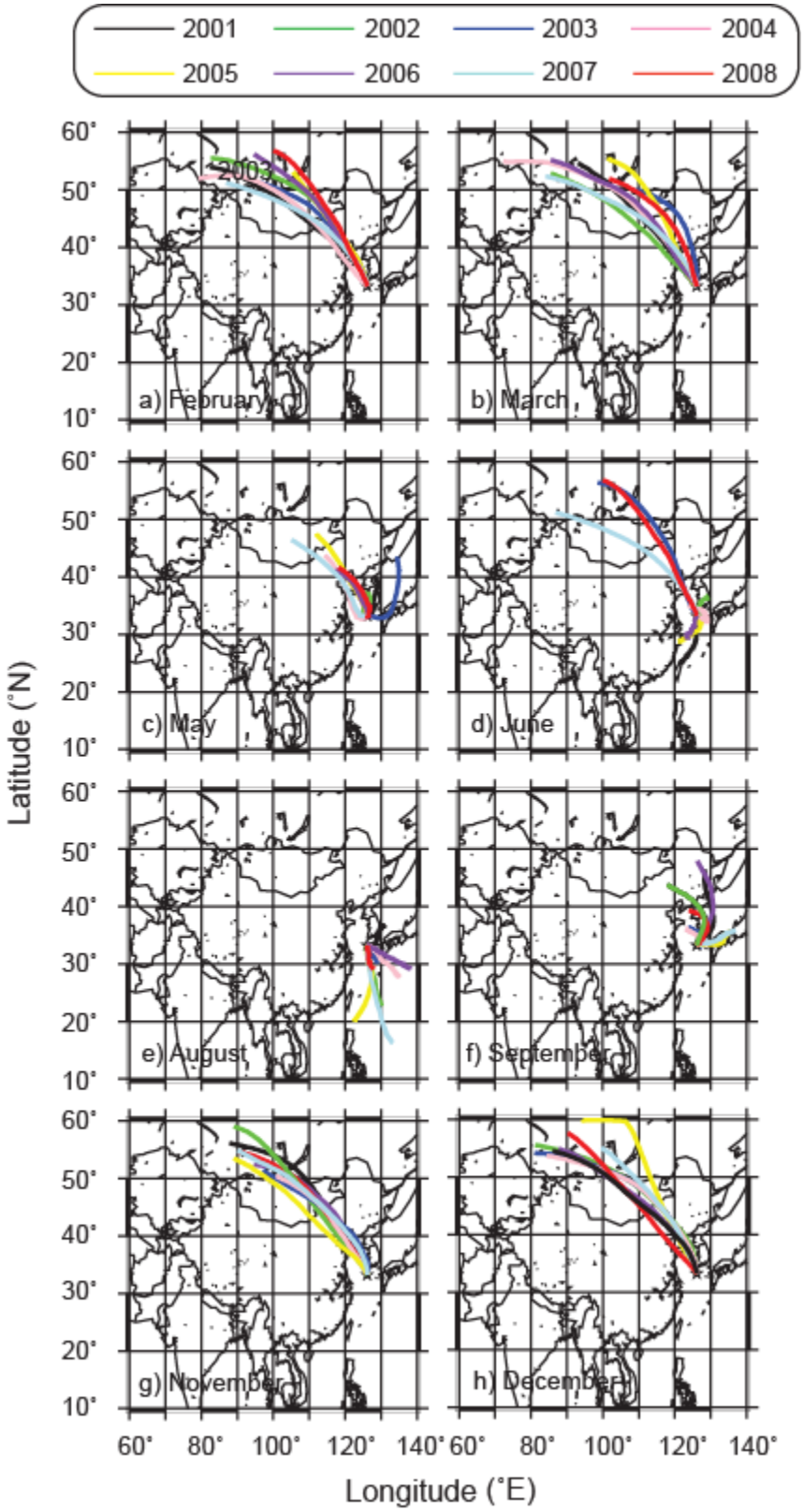
Comment 5 - Page 22195 Line 25-26 (and abstract): Do the authors have any measurement of SOA mass? If not, I don't think that it is appropriate to extrapolate the results with diacids to the total SOA budget in East Asia. It is completely plausible that although the absolute concentration of diacids were increasing over years, their fractions in total SOA and hence the total SOA mass were not.

Response 5 - Unfortunately, we do not have the estimation of the SOA mass. We agree that the increase of diacid fractions may not enhance the SOA mass. Hence, we are not going to extrapolate the diacids increases to the increases of total SOA budget in the abstract, conclusions and other parts of the manuscript. In the revised manuscript, we have deleted the related sentences.

Comment 6 - Figure 3. Is the back trajectory of July representative of all the summer months (i.e. June- August)?

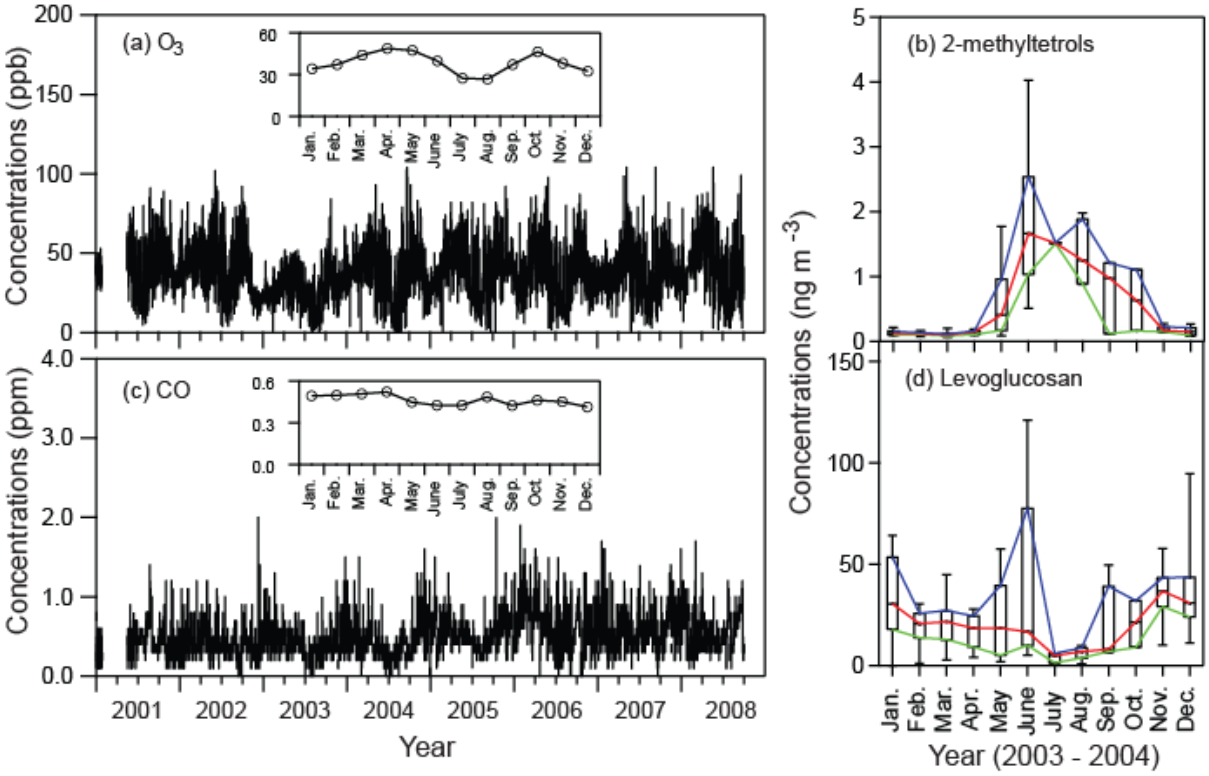
Response 6 - The air masses are oceanic in June (exception 2003, 2007 and 2008) and August, similar to those in July (see the figure below).

According to this comment, we have added the air mass history for these months as Fig. S1 in the supplementary materials. We have also described the results of Fig. S1 in the text. Please see lines 180-183 and 195-196 in the revised manuscript.



Comment 7 - Figure 4. I suggest making monthly average plots (same as those for 2-methyltetrols and levoglucosan) for O3 and CO as well for better comparison with other diacids and tracer compounds

Response 7 - We have modified the Figure 4 as below.


(a) O_3
Concentrations (ppb)
(c) CO
Concentrations (ppm)
Year
(b) 2-methyltetrols
(d) Levoglucosan
Concentrations (ng m $^{-3}$)
Year (2003 - 2004)
Jan.
Feb.
Mar.
Apr.
May
June
July
Aug.
Sep.
Oct.
Nov.
Dec.

Responses to the comments of Anonymous Referee #2

Comment 1 - In this article authors represent extensive and very impressive dataset, that is definitely worth of publishing. Article is easy to read and understand. Topic, SOA concentrations in East Asia is very important and requires attention. I will recommend accepting this paper with a few minor comments.

Response 1 - Thank you so much. Please see bellow point-by-point responses to the comments.

Comment 2 - Chapter 2. - Include number of samples /year to give reader an idea on how many samples conclusions are based on - CO, O3 data are used in results. Please add information about these instruments also.

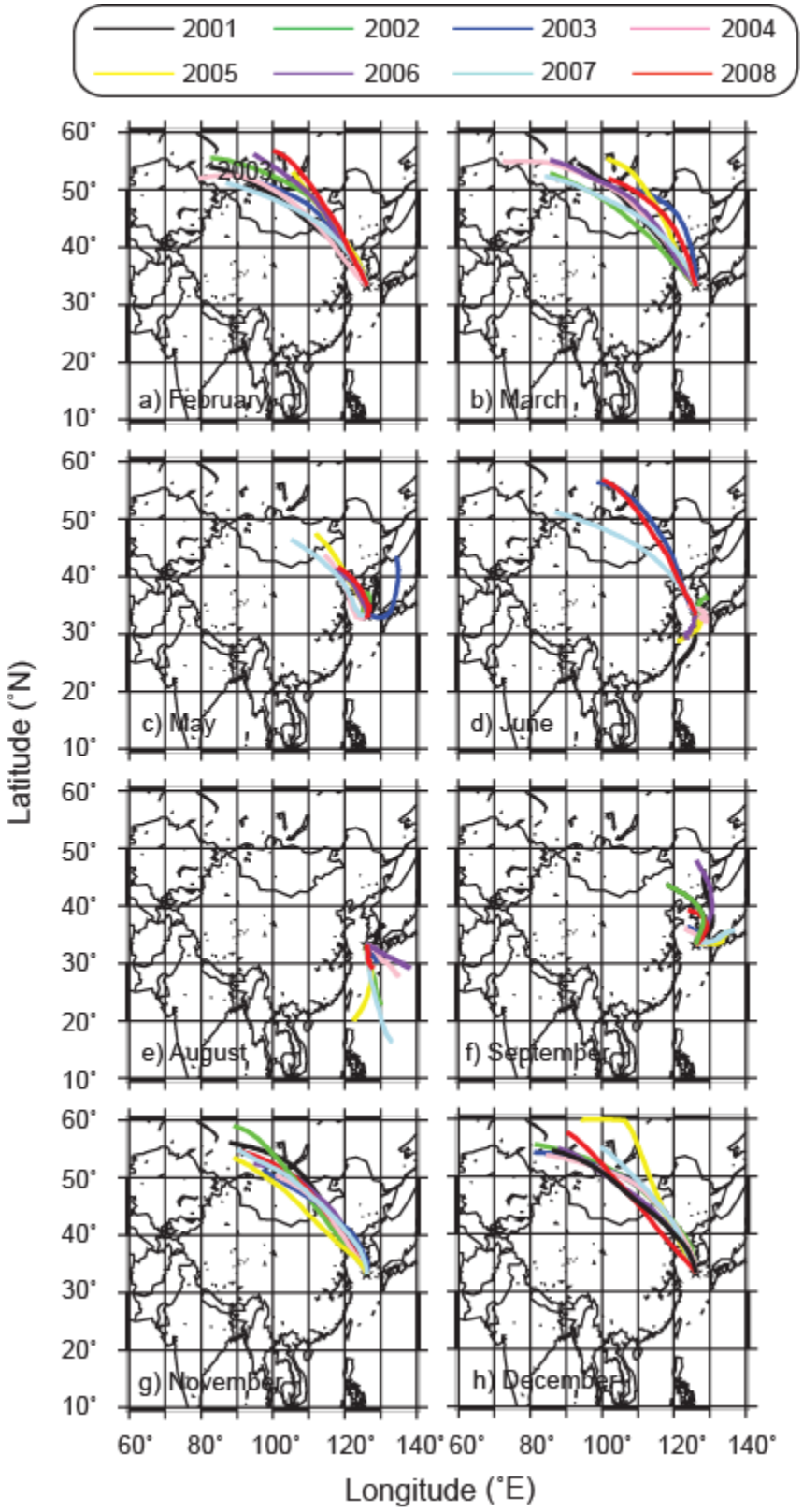
Response 2 - The samples collected per year have written in the revised manuscript as "The total numbers of samples collected were 71 in 2001, 25 in 2002, 48 in 2003, 98 in 2004, 123 in 2005, 116 in 2006, 142 in 2007, and 91 in 2008." Please see lines 100-102 in the revised manuscript. The information related to the O_3 and CO monitor has been provided as "The concentrations of O_3 and CO were measured by the Korean Meteorological Administration (KMA) using Thermo Environmental Instrument 49C and 48C, respectively (Thermo Inc., USA). Please see lines 154-157 in the revised manuscript.

Comment 3 - Chapter 3.1. -Please define molecular distribution

Response 3 - We have described the term molecular distributions in the revised manuscript as "Molecular distributions, related to the presence/absence and abundance of organic compounds in atmospheric aerosols, provide important information about their sources, formation, chemical evolution and physical properties." Please see lines 162-164 in the revised manuscript.

Comment 4 - Chapter 3.2 -Authors state that "Typical air mass transport patterns at Gosan have been shown with reference to the mid-month of each season over the time period of 2001–2008 (Fig. 3)". Have authors checked that these mid-month values represent the whole dataset by calculating daily trajectories? I think so-called trajectory density plot (describing by color how often a trajectory intercepts a given locationbox) would describe situation better, but might be time-consuming to make.

Response 4 - According to this comment, we have added the air mass history for the other months as Fig. S1 in the supplementary materials. We have also described the results of Fig. S1 in the text. Please see lines 180-183 and 196-198 in the revised manuscript.



Comment 5 - Authors state that: “In spring, major saturated diacids did not correlate or loosely correlated with 2 -methyltetrols (isoprene-SOA tracers, r2=0.001–0.05) (Fig. 5a–e), pinic acid (an-pinene- SOA tracer, 0.10–0.39) (Fig. 6a–e) and levoglucosan (a biomass burning tracer, 0.001– 0.07) (Fig. 7a–e) (e.g., Fu et al., 2014). “. However, 2-Methyltetrol concentration were only measured for 2003-2004. Please add this to text.

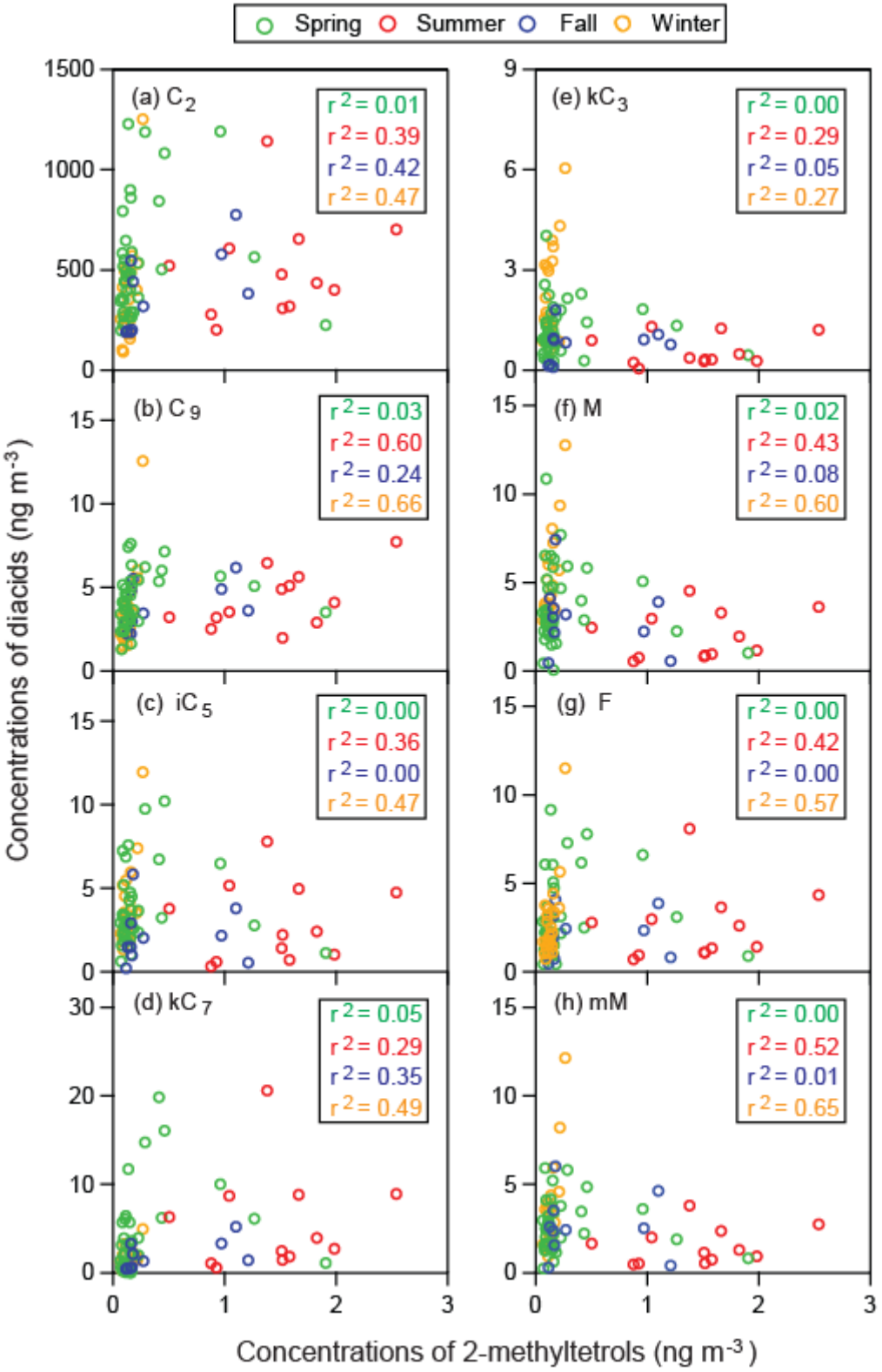
Response 5 - We have provided that information as "In 2003-2004 spring, major saturated diacids did not correlate or loosely correlated with 2-methyltetrols (isoprene-SOA tracers, r^2 = 0.001-0.05) (Fig. 5a-e), pinic acid (α-pinene-SOA tracer, 0.10-0.39) (Fig. 6a-e) and levoglucosan (biomass burning tracer, 0.001-0.07) (Fig. 7a-e)". Please see lines 215-219 in the revised manuscript.

Comment 6 - Chapter 4-Authors state "The increases of diacids derived from anthropogenic VOCs are more prominent than those of diacids generated from biogenic VOCs in East Asia. If the current rate of increases continued, the SOA budget would increase significantly in the future atmosphere in East Asia.". I am not sure if you can conclude that increase in diacids, directly means increase in SOA. Maybe reformulate this and also same statement/conclusion in other chapters.

Response 6 - Unfortunately, we do not have the estimation of the SOA mass. We agree that the increase of diacid fractions may not enhance the SOA mass. Hence, we are not going to extrapolate the diacids increases to the increases of total SOA budget in the abstract, conclusions and other parts of the manuscript. In the revised manuscript, we have deleted the related sentences.

Comment 7 - Figures - Figure 5, 6, 7 have a very low information value. If there is no correlation, just note that in text. Please consider improving these pictures. Maybe show only those plots that are necessary. Others can be shown in the supplement.

Response 7 - We have improved the color of this picture in the revised manuscript. For example, see the revised Fig. 5 as below. We have decided to keep those figures in the manuscript because diacid concentrations are well-correlated with tracers in other seasons, although the concentrations of diacids are not well-correlated with tracers in spring.


Spring
Summer
Fall
Winter
(a) C2
r2 = 0.01
r2 = 0.39
r2 = 0.42
r2 = 0.47
(b) C9
r2 = 0.03
r2 = 0.60
r2 = 0.24
r2 = 0.66
(c) iC5
r2 = 0.00
r2 = 0.36
r2 = 0.00
r2 = 0.47
(d) kC7
r2 = 0.05
r2 = 0.29
r2 = 0.35
r2 = 0.49
(e) kC3
r2 = 0.00
r2 = 0.29
r2 = 0.05
r2 = 0.27
(f) M
r2 = 0.02
r2 = 0.43
r2 = 0.08
r2 = 0.60
(g) F
r2 = 0.00
r2 = 0.42
r2 = 0.00
r2 = 0.57
(h) mM
r2 = 0.00
r2 = 0.52
r2 = 0.01
r2 = 0.65
Concentrations of diacids (ng m-3)
Concentrations of 2-methyltetrols (ng m-3)

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 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 (N_2) 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 line163 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, Line18 – 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.