

## 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 diacids 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<sup>th</sup> and 75<sup>th</sup>) 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.

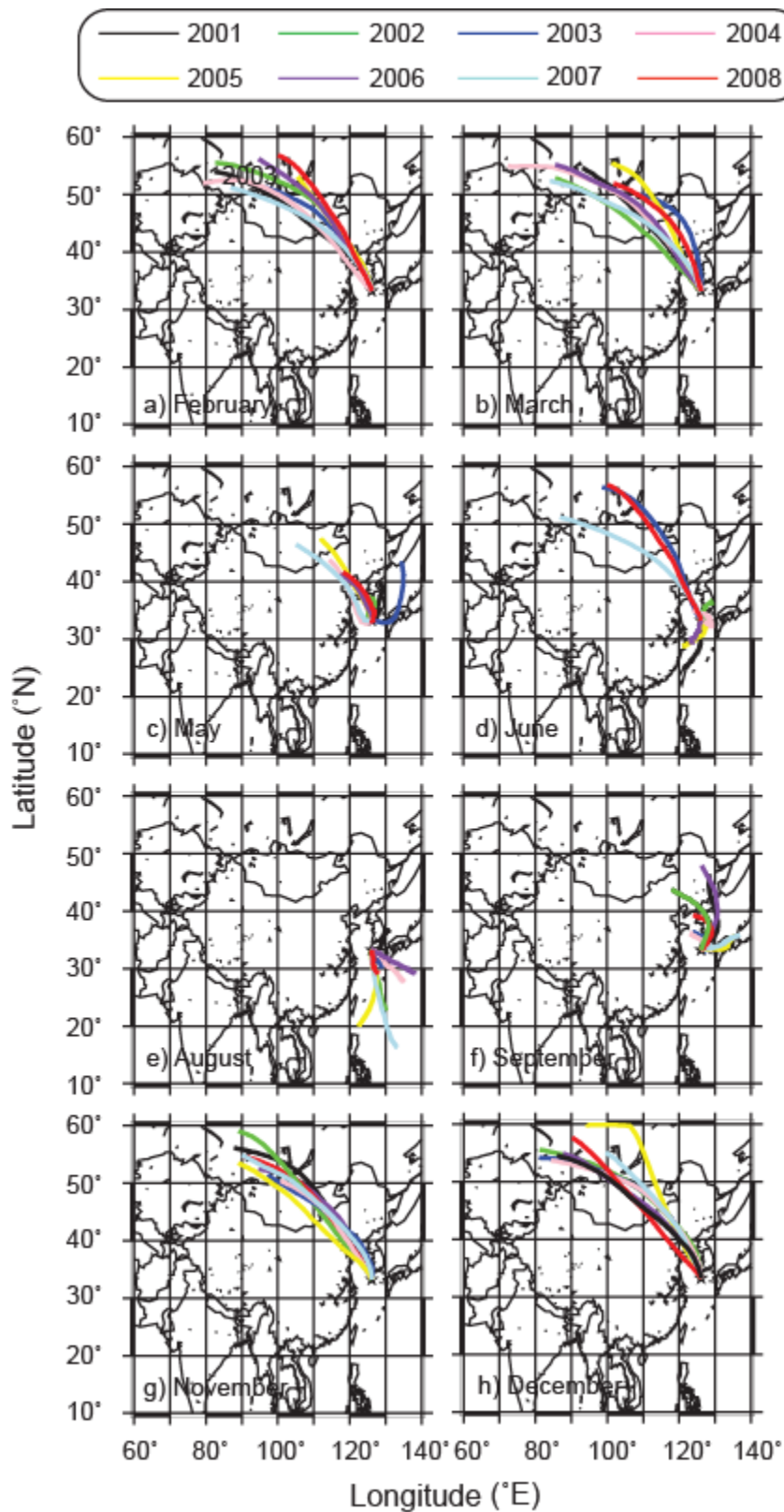
**Comment 5** - Page 22195 Line 25-26 (and abstract): Do the authors have any measurement of SOA mass? If not, I don't think that it is appropriate to extrapolate the results with diacids to the total SOA budget in East Asia. It is completely plausible that although the absolute concentration of diacids 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 O<sub>3</sub> and CO as well for better comparison with other diacids and tracer compounds

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

