Dear Editor and Reviewers,

The authors would like to thank the editor and the reviewers for the constructive and good suggestions to improve our manuscript! We have carefully considered all the review comments and revised the manuscript. Below, we provide responses to the comments, with changes made in the manuscript highlighted in red.

Sincerely

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Response to Reviewer 1:

The authors present data from measurements at Mt Tai focusing on dicarboxylic acids and related compounds. Based on back trajectory and model analysis they conclude that aerosol arriving on Mt Tai has undergone long range transport and has a variety of sources, including anthropogenic emissions and biomass burning. This study is with some new features (back trajectories, WRF model) which, however, have not been really made use of. Overall, it may be an interesting data set, in particular as it is discussed in the context of previous measurements of the same compounds at many different locations and also at Mt Tai in 2006. However, I think the discussion is quite confusing and needs major revision.

Response: We appreciate the reviewer for the comments and suggestions. We have revised the manuscript accordingly and here address individually the review comments. WRF model and back trajectories have been studied throughout the draft. Detailed descriptions about aqueous phase oxidation are also presented. Other content cohesions also have been done. A tight and clear manuscript has been obtained. For clarity, the reviewer's comments are listed below in black italics, while our responses and changes in manuscript are shown in blue and red, respectively. Revised table and figure are in the end.

1. This study is very similar to previous studies from some of the same authors (27% similarity rate), with some new features (back trajectories, WRF model) which, however, have not been really made use of.

Response: About 15% similarity rate was caused by the name "dicarboxylic acids and related compounds" or the name "dicarboxylic acids, oxocarboxylic acids and α-dicarbonyls". We have changed "dicarboxylic acids and related compounds" and "dicarboxylic acids, oxocarboxylic acids and α-dicarbonyls" to "DCRCs". Moreover, we rephrased the similar expressions sentence by sentence according to the similarity report, please see more details in the revised manuscript.

2. Day vs night time samples. The authors find that day and night time samples show almost identical concentrations. However, I am not sure that distinguishing day- and night-time-samples is really meaningful here: If a sample was collected at nighttime (i.e. 6 pm - 6 am), it was likely processed during the day(s) before. The same might be true for day time samples that travelled to the sample location for several days. Thus, I am not surprised that samples collected during day and night show very similar composition and loadings.

Response: DCRCs (see above on this abbreviation) concentrations can be influenced during the day by higher chemical formations due to the occurring photochemistry. On the other hand, DCRCs, such as oxalic acid, can act in the atmosphere as efficient complexing agent forming iron-oxalate complexes which can be effectively photolysis during the day. Thus, the photochemistry may also represent an important sink.

It is expected that DCRCs are secondary products of atmospheric chemistry processes, therefore, the investigation of DCRCs concentration differences during the day and night conditions has been done. The performed day/night sampling is not ideal and has limitations due to the measured DCRCs form the processing during the day (s) before. Therefore, a sentence addressing this issue has been added to the revised manuscript as follows:

In order to identify the impact of atmospheric chemistry processes on DCRCs, $PM_{2.5}$ samples were collected during the day and night, respectively, from 4 June to 4 July 2014.

(Page 5, Line 7-9)

Choosing 06:00-18:00 and 18:00-06:00 local time as day and night sampling period were due to diurnal variation of UV radiation at the ground of Mt. Tai (Fig. S1). Figure S1 shows an increase from 06:00 when UV radiation begin, followed by a sharp increase and a maximum at 12:00, and a sharp decrease until 18:00. From 19:00 to 23:00 and from 0:00 to 5:00, UV radiation is close to zero.

Moreover, a number of published papers, performing day and night sampling between 06:00-18:00 and 18:00-06:00 local time, showed different diurnal variation, for example Pavuluri et al. (2010), Miyazaki et al. (2009) and Fu et al. (2008). Pavuluri et al. (2010) reported that mostly DCRCs presented clear diurnal trend. Miyazaki et al. (2009) reported that higher concentrations of mostly DCRCs were observed in the night samples. Fu et al. (2008) reported that most of organic compound classes showed higher concentrations in the night samples. We have added descriptions about choosing 06:00-18:00 and 18:00-06:00 local time as sampling time in the revised manuscript as follows:

Considering the diurnal variation of ultraviolet radiation around Mt. Tai (Fig. S1), 06:00-18:00 and 18:00-06:00 local time were selected as the sampling times for day and night, respectively.

(Page 5, Line 9-12)

Reference:

Pavuluri, C.M., Kawamura, K., and Swaminathan, T. Water-soluble organic carbon, dicarboxylic

acids, ketoacids, and a-dicarbonyls in the tropical Indian aerosols. Journal of Geophysical Research, 2010, 115.

Miyazaki, Y., Aggarwal, S.G., Singh, K., Gupta, P.K., Kawamura, K. Dicarboxylic acids and water-soluble organic carbon in aerosols in New Delhi, India, in winter: Characteristics and formation processes. Journal of Geophysical Research, 2009, 114.

Fu, P.Q., Kawamura, K., Okuzawa, K., Aggarwal, S.G., Wang, G.H., Kanaya, K., Wang, Z.F. Organic molecular compositions and temporal variations of summertime mountain aerosols over Mt. Tai, North China Plain, 2008. Journal of Geophysical Research, 2008, 113.

Unless I misunderstood the sampling and nomenclature of day/night samples, I suggest removing the discussion of day- versus night-samples. That way, hypotheses such as on night time oxidation (p. 2, l. 12) or less effective loss during night (p. 10, l. 24) could be removed as they do not seem supported.

Response: Our results showed the day concentrations of DCRCs were similar with their night concentrations. The day-night concentration ratios of 79% DCRCs individual species ranged between 0.9 and 1.1. However, 2006 results showed day concentrations of DCRCs were 2-3 times higher than those in the night (Wang et al., 2009). In order to accurately identify major control factor of DCRCs at Mt. Tai, such as boundary layer heights, long-range transport, ground pollutants transport and aqueous phase oxidation, day/night variations in this study were necessary to discuss.

Reference:

Wang, G.H., Kawamura, K., Umemoto, N., Xie, M.J., Hu, S.H., Wang, Z.F. Water-soluble organic compounds in PM2.5 and size-segregated aerosols over Mount Tai in North China Plain. Journal of Geophysical Research, 2009, 114, D19208.

3. Trajectories a) In Figure 1, the authors show 72 h-back trajectories of air masses arriving at Mt Tai. These trajectories are briefly discussed in Section 3.1. However, the authors do not link their later discussion to these trajectories. For example, there seems to be change in conditions (meteorology, emissions, air mass?) after the first half of the sampling period that leads to lower diacid loadings. Could that be linked to different trajectories? I suggest adding somehow the dates to the trajectories in

Figure 1 or adding discussion in the discussion section.

Response: 4-19 June was the first half sampling period, and 20 June-04 July was the second half sampling period. Sampling dates in different clusters as follows:

Cluster 1: 4-5 June, 29 June

Cluster 2: 6 June, 11-13 June, 22-24 June, 28 June, 4 July

Cluster 3: 7-8 June

Cluster 4: 9-10 June, 14-21 June, 25-27 June, 30 June, 1-3 July.

We have added sampling dates in 3.1 discussion part as follows:

Sampling dates in cluster 2 included 6 June, 11-13 June, 22-24 June, 28 June and 4 July, while 9-10 June, 14-21 June, 25-27 June, 30 June and 1-3 July belonged to cluster 4.

(Page 8, Line 23-25)

Trajectories on 4-5 June and 29 June were grouped into cluster 1, while trajectories on 7-8 June were grouped into cluster 3.

(Page 9, Line 1-2)

Impact of air mass and meteorology on DCRCs concentrations in the first and second half sampling periods have been discussed and added in the revised manuscript as follows:

From the trajectory analysis, we can see that during the first and second half of the sampling periods, 4 and 5 days, respectively, belonged to cluster 2. In addition, 8 and 9 days belonged to cluster 4, respectively. Therefore, the dominant air masses in the first and second half of the sampling periods were similar, and thus had a low impact on DCRC concentrations in the two periods. Figure S2 shows meteorological data in the different backward trajectory clusters during the sampling period at Mt. Tai. We can see that pressure, temperature and RH didn’t change much in clusters 2 and 4. Moreover, as shown in Fig. 1, meteorological data at Mt. Tai site also didn’t change much between the first and second half of the sampling periods. Therefore, the quite stable meteorological conditions may have had a low impact on the DCRC concentrations between the first and second half of the sampling periods.

(Page 13, Line 29-Page 14, Line 10)

Higher DCRCs concentrations in the first half sampling period were most likely caused by biomass burning. Dicarboxylic acids and K^+ showed strong correlation in the first half, while in the second half, dicarboxylic acids and K^+ exhibited no correlation (Fig. 8). Moreover, as shown in Fig. 9, straw burning hotspots mainly distributed in the first half sampling period, while in the second half sampling period, hotspots almost disappeared. Detailed descriptions about biomass burning have been showed in the revised manuscript as follows:

Dicarboxylic acids and K^+ exhibited a strong correlation during the first half, while during the second half, dicarboxylic acids and K^+ exhibited no correlation (Fig. 8). The peaks of dicarboxylic acids and K^+ appeared almost simultaneously (Fig. 7). It is also clear that when the K^+ concentration increased, dicarboxylic acids correspondingly increased during the first half (Fig. 8). These results imply that biomass burning was an important contributor to DCRCs during the first half of the measurement period. Moreover, according to reports by weather satellites of the Ministry of Environment Protection of the People's Republic of China (http://www.zhb.gov.cn/), straw burning hotspots in air masses that passed over key areas (Anhui, Hebei and Shandong province) were mainly distributed in the first half of the sampling period (Fig. 9). This result further supports that biomass burning was more important in the first half of the sampling period.

(Page 14, Line 11-22)

b) The average RH on Mt Tai was low (17%). If indeed aqueous phase processing was a major contributor to the target compounds, RH would have needed to be high during the transport. Can the trajectories tell anything about clouds and/or high RH fields the air masses experienced during transport? Could any precipitation during transport explain an observed decrease in concentrations?

Response: Please note in 2.1 part, we reported that “During sampling period, average values of temperatures, relative humidity (RH) and winds were 17 ℃, 87% and 2.1 m/s.” Average RH values at Mt. Tai were 87%, not 17%.

As shown in Fig. 1, average RH values during the sampling period at Mt. Tai were 87%, up to 100%. Moreover, as mentioned above, average RH values were 67% and 72% in clusters 2 and 4 (they were dominant back-trajectory clusters) (Fig. S2), respectively. However, due to the coarse resolution of HYSPLIT, it was difficult to judge whether clouds occurred. Therefore, MODIS satellite pictures were investigated, and the results showed sometimes clouds occurred in the region of Mt. Tai and in the areas of trajectories passed over during the sampling period. But MODIS satellite pictures limited information about the cloud base and top heights, thus it cannot exactly explore whether there were clouds in the height of the trajectories.

As shown in Fig. S2, we can see that precipitation during the back-trajectory clusters was low, the maximum of precipitation was just 0.11 mm, almost zero. During the campaign at Mt. Tai, minor rain events occurred on 15, 16 June and 3 July, major rain events occurred on 24 June and 4 July. Therefore, precipitation was also likely not the reason for the decrease of DCRCs concentrations in the second half sampling period.

We have added some descriptions about RH in 3.4 discussion part as follows:

Average RH values during the sampling period at Mt. Tai were 87%, up to 100% (Fig. 1), and higher on average during the night (Fig. 5). In addition, average RH values along the dominant back-trajectory clusters (clusters 2 and 4) were about 70% (Fig. S2). However, due to the coarse resolution of HYSPLIT, it was difficult to judge whether clouds occurred. Therefore, MODIS satellite pictures were investigated, and the results showed that clouds sometimes occurred in the region of Mt. Tai and in the areas that the trajectories passed over during the sampling period. But MODIS satellite pictures have limited information about the cloud base and top heights, and thus it cannot exactly explore whether there were clouds at the height of the trajectories.

(Page 12, Line 27-Page 13, Line 7)

We have added descriptions about precipitation as follows:

Minor rain events occurred on 15, 16 June and 3 July, and major rain events occurred on 24 June and 4 July. The sample collection was ended just before the major rain.

(Page 5, Line 2-3)

c) The discussion of the concentrations of diacids in the various clusters is not clear. How was the number of 73% of total dicarboxylic acids and related compounds in clusters 2 and 4 determined (p. 7, l. 8)?

Response: We have revised the sentence as follows:

The sum of DCRC concentrations in clusters 2 and 4 contributed 73% of the total concentration of DCRCs during the sampling period.

(Page 8, Line 21-22)

4. Mass fraction of total and individual dicarboxylic acids etc a)-1 The authors point out in the introduction that dicarboxylic acids are 'significant constituents in PM2.5' and ': : :impact on air quality' (p. 3, l. 4ff). However, later in the text the author quantify that these compounds contribute on average < 3% of OC (p. 7, l. 26), and related compounds even less (< 1%). What is the overall fraction of these compounds in the aerosol, i.e. not only related to organic mass but to total mass?

Response: DCRCs are not only oxygenated organic compounds but water-soluble components of SOA (Kawamura and Sakaguchi, 1999; Kawamura and Yasui, 2005; Pavuluri et al., 2010). Until now only a small fraction of water-soluble and oxygenated SOA can be identified in a compound specific manner. Zhu et al. (2017) reported that only 18.1-49.1% of SOA can be detected, a large fraction can't be identified. Therefore, DCRCs are important constituents in the water soluble and oxygenated matter of $PM_{2.5}$. We have changed related descriptions in the revised manuscript as follows:

Dicarboxylic acids and related compounds (oxocarboxylic acids and α-dicarbonyls) (DCRCs) are important constituents in $PM_{2.5}$, and mainly produced by secondary processes (Kawamura and Yasui, 2005; Pavuluri et al., 2010a). Due to their high water solubility, DCRCs contribute to the water soluble organic fraction of $PM_{2.5}$, which can have an impact on air quality (van Pinxteren et al., 2009; Kawamura and Bikkina, 2016; Kundu et al., 2010b).

(Page 3, Line 2-7)

$PM_{2.5}$ mass concentration at Mt. Tai during the campaign was 98.2 ± 29.2, 98.6 ± 25.3 μg m^{-3} in the day and night, respectively. DCRCs concentration contributed 1.2% and 1.1% to $PM_{2.5}$ in the day and night, respectively. The DCRCs-C at Mt. Tai in 2014 accounted for 3.24% and 3.20% of OC in the day and night, respectively. We have added these results in the revised manuscript as follows:

$PM_{2.5}$ mass concentration at Mt. Tai during the campaign was 98.2 ± 29.2 and 98.6 ± 25.3 μg m^{-3} during the day and night, respectively. DCRC total concentration contributed about 1.2% and 1.1% to $PM_{2.5}$ in the day and night, respectively. In addition, the DCRCs-C accounted for 3.3% and 3.2% of OC in the day and night, respectively.

(Page 9, Line 25-29)

Reference:

Kawamura, K. and Sakaguchi, F.: Molecular distributions of water soluble dicarboxylic acids in marine aerosols over the Pacific Ocean including tropic, J. Geophys. Res., 104, 3501–3509, 1999.

Kawamura, K. and Yasui, O.: Diurnal changes in the distribution of dicarboxylic acids, ketocarboxylic acids and dicarbonyls in the urban Tokyo atmosphere, Atmos. Environ., 39, 1945-1960, 2005.

Pavuluri, C. M., Kawamura, K., and Swaminathan, T.: water-soluble organic carbon, dicarboxylic acids, ketoacids, and a-dicarbonyls in the tropical Indian aerosols, J. Geophys. Res., 115, D11302, doi:10.1029/2009JD012661, 2010.

Zhu, Y., Yang, L., Kawamura, K., Chen, J., Ono, K., Wang, X., Xue, L., and Wang, W.: Contributions and source identification of biogenic and anthropogenic hydrocarbons to secondary organic aerosols at Mt. Tai in 2014, Environ. Pollut., 220, 863-872,2017.

a)-2 The total (organic + inorganic) mass should be also reported in Table 1.

Response: $PM_{2.5}$ mass concentration at Mt. Tai during the campaign was 98.2 ± 29.2, 98.6 ± 25.3 μg m^{-3} in the day and night, respectively. We have added $PM_{2.5}$ mass concentration in the Table 1.

b) The authors continue using ‘significant’ for contributions of > 2% within the *diacid mass (p. 8, l. 5). I would not call such masses 'significant' given that these species contribute overall < 0.1% tot the total organic mass in the aerosol.*

Response: We have changed "significant" to "some" as follows:

Ph (day: 4%, night: 3%), glutaric acid (C_5) (day: 3%, night: 3%) and azelaic acid (C_9) (day: 2%, night: 2%) also exhibited some contributions.

(Page 10, Line 7-8)

5. VOC measurements a) Only the total VOC mixing ratios are reported, assuming that all of them could be precursors for the target species. Figure 2 might be more meaningful if only a few selected VOCs are shown that have been shown to act as precursors for the identified compounds in Table 1. b) I think it is rather unusual that VOC measurements were performed at University of Irvine but neither in the acknowledgement nor in the author list anyone from this place is listed.

Response: Detailed descriptions about VOCs concentrations at Mt. Tai in 04 June-04 July 2014 have been given in our previous study-Zhu et al. (2017). Therefore, in this study, we briefly described the measured VOCs.

DCRCs precursors clearly pointed out by published papers included ethyne, ethene, isoprene, α-pinene, β-pinene, toluene, m/p-xylene, o-xylene (Warneck, 2003; Ervens et al., 2004; Bikkina et al., 2014; Tilgner and Herrmann, 2010). Therefore, in this study, we selected ethyne, ethene, isoprene, α-pinene, β-pinene, toluene, m/p-xylene, o-xylene as DCRCs precursors. Then we made scatter plot using total concentration of these selected DCRCs precursors and DCRCs total concentration. The result was presented in Fig. 4, which was more meaningful relative to using total VOCs concentration.

We also added descriptions about selected DCRCs precursors as follows:

As shown in Fig. 4, DCRC concentrations exhibited weak and moderate correlation with total concentration of selected DCRC precursors in the day and night, respectively, where selected DCRCs precursors included ethyne, ethene, isoprene, α-pinene, β-pinene, toluene, m/p-xylene and o-xylene (Warneck, 2003; Ervens et al., 2004; Bikkina et al., 2014; Tilgner and Herrmann, 2010).

(Page 12, Line 18-22)

Thanks for your reminding, Donald R. Blake from University of Irvine was added in the author list as follows:

Yanhong Zhu [1], Lingxiao Yang [1,7*], Jianmin Chen [1,6,7], Kimitaka Kawamura [3,a], Mamiko Sato [3], Andreas Tilgner [4], Dominik van Pinxteren [4], Ying Chen [4,b], Likun Xue [1], Xinfeng Wang [1], Isobel J. Simpson [5], Hartmut Herrmann [4,2,1], Donald R. Blake [5], Wenxing Wang [1]

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[7] Jiangsu Collaborative Innovation Center for Climate Change, China

Reference:

Zhu, Y., Yang, L., Kawamura, K., Chen, J., Ono, K., Wang, X., Xue, L., and Wang, W.: Contributions and source identification of biogenic and anthropogenic hydrocarbons to secondary organic aerosols at Mt. Tai in 2014, Environ. Pollut., 220, 863-872,2017.

Warneck, P.: In-cloud chemistry opens pathway to the formation of oxalic acid in the marine atmosphere, Atmos. Environ., 37, 2423–2427, 2003.

Ervens, B., Feingold, G., Frost, G, J., and Kreidenweis, S. M.: A modeling study of aqueous production of dicarboxylic acids: 1. Chemical pathways and speciated organic mass production, J.

Geophys. Res., 109, D15205, doi:10.1029/2003JD004387, 2004.

Bikkina, S., Kawamura, K., Miyazaki, Y., Fu, P., 2014. High abundances of oxalic, azelaic, and glyoxylic acids andmethylglyoxal in the open oceanwith high biological activity: implication for secondary OA formation from isoprene. Geophys. Res. Lett. 41, 3649–3657. http://dx.doi.org/10.1002/2014GL059913.

Tilgner, A., and Herrmann, H.: Radical-driven carbonyl-to-acid conversion and acid degradation in tropospheric aqueous systems studied by CAPRAM, Atmos. Environ., 44, 5415-5422, 2010.

6. Comparison to previous studies How much of diacids and related compounds is expected to be in the size range of PM2.5 to PM10? I.e. is it reasonable to assume that the same scaling factor (0.91) for the total PM2.5/TSP mass can be applied to the diacids and levoglucosan? Are there any measurements (from other locations) that support this assumption?

Response: Studies carried out in June 2006 at Mt. Tai only showed mass concentrations of $PM_{2.5}$ and TSP, no PM_{10}. So, $PM_{2.5}/PM_{10}$ ratio was not obtained in June 2006 at Mt. Tai.

Using the ratio of $PM_{2.5}$/TSP = 0.91 is reasonable for calculating DCRCs concentrations in $PM_{2.5}$ in June 2006 at Mt. Tai based on the results of previous field studies.

Deng et al. (2011) reported the chemical characterization of aerosols in June 2006 at Mt. Tai. The paper clearly pointed out that the ratio of $PM_{2.5}$/TSP in June 2006 at Mt. Tai was 0.91. Moreover, DCRCs (Kawamura et al., 2013) and levoglucosan (Fu et al., 2008) concentrations in TSP were also obtained in June 2006 at Mt. Tai. Therefore, using the ratio of $PM_{2.5}$/TSP = 0.91 is reasonable to obtain DCRCs and levoglucosan concentrations in $PM_{2.5}$ in June 2006 at Mt. Tai.

We have provided more details on $PM_{2.5}$/TSP in the revised manuscript as follows:

Deng et al. (2011) reported that the ratio of $PM_{2.5}$/TSP in June 2006 at Mt. Tai was 0.91. Using the ratio and DCRCs concentrations in TSP at Mt. Tai in June 2006 (Kawamura et al., 2013), we estimated the corresponding DCRC concentrations in

$PM_{2.5}$ at Mt. Tai in June 2006 (1550, 220, 62.2 ng m^{-3} for dicarboxylic acids, oxocarboxylic acids and α-dicarbonyls, respectively).

(Page 10, Line 18-23)

In addition, using the ratio of $PM_{2.5}$/TSP and the levoglucosan concentration in TSP at Mt. Tai in June 2006 (Fu et al., 2008), the estimated levoglucosan concentration in $PM_{2.5}$ at Mt. Tai in June 2006 was 387 ng m^{-3}. The result was more than five times higher than that in 2014 (levoglucosan: 70.4 ng m^{-3}) (Zhu et al., 2017), which suggests that biomass burning may have decreased from 2006 to 2014.

(Page 10, Line 27-Page 11, Line 3)

Reference:

Deng, C., Zhuang, G., Huang, K., Li, J., Zhang, R., Wang, Q., Liu, T., Sun, Y., Guo, Z., Fu, J.S., Wang, Z. Chemical characterization of aerosols at the summit of Mountain Tai in Central East China, 2011. Atmospheric Chemistry and Physics, 11, 7319-7332.

Kawamura, K., Tachibana, E., Okuzawa, K., Aggarwal, S., Kanaya, Y., and Wang, Z.: High abundances of water-soluble dicarboxylic acids, ketocarboxylic acids and α-dicarbonyls in the mountaintop aerosols over the North China Plain during wheat burning season, Atmos. Chem. Phys., 13, 8285-8302, 2013.

Fu, P., Kawamura, K., Okuzawa, K., Aggarwal, S. G., Wang, G., Kanaya, Y., and Wang, Z.: Organic molecular compositions and temporal variations of summertime mountain aerosols over Mt. Tai, North China Plain, J. Geophys. Res. Atmos., 113, 2008.

7. *p. 3, l. 28: 'secondary oxidation' seems redundant*

Response: deleted.

8. *p. 3, l. 15: Are these considered primary sources? In my understanding, combustion of fossil fuel or biomass is an oxidative process and thus the products are secondary.*

Response: Fossil fuel or biomass combustion can directly emit aerosol particles into the atmosphere, so they were considered as primary sources. However, partly primary pollutants emitted from fossil fuel or biomass combustion subsequently were oxidized then produced secondary aerosol. Therefore, someone argued that fossil fuel or biomass combustion also can be regarded as secondary sources.

Dicarboxylic acids and related compounds (DCRCs) can be directly emitted into the atmosphere from fossil fuel or biomass combustion (Gao et al., 2003; Falkovich et al., 2005; Kundu et al., 2010; Kawamura and Kaplan, 1987; Ho et al., 2006). Of course they can be produced during chemical ageing of combustion plumes. Given Mt Tai is quite away from combustion sources I would expect the observed acids to be a mixture from primary emission and secondary formation.

Reference:

Gao, S., Hegg, D. A., Hobbs, P. V., Kirchstetter, T. W., Magi, B. I., and Sadilek, M.: Water-soluble organic components in aerosols associated with savanna fires in southern Africa: Identification, evolution, and distribution, J. Geophys. Res., 108, 8491, doi:10.1029/2002JD002324, 2003.

Falkovich, A. H., Graber, E. R., Schkolnik, G., Rudich, Y., Maenhaut, W., and Artaxo, P.: Low molecular weight organic acids in aerosol particles from Rondˆonia, Brazil, during the biomass-burning, transition and wet periods, Atmos. Chem. Phys., 5, 781–797, doi:10.5194/acp-5-781-2005, 2005.

Kundu, S., Kawamura, K., Andreae, T. W., Hoffer, A., and Andreae, M. O.: Molecular distributions of dicarboxylic acids, ketocarboxylic acids and a-dicarbonyls in biomass burning aerosols: implications for photochemical production and degradation in smoke layers, Atmos. Chem. Phys., 10, 2209–2225, doi:10.5194/acp-10-2209-2010, 2010.

Kawamura, K. and Kaplan, I. R.: Motor exhaust emission as a primary source of dicarboxylic acids in Los Angeles ambient air, Environ. Sci. Technol., 21, 105–110, 1987.

Ho, K. F., Lee, S. C., Cao, J. J., Kawamura, K., Watanabe, T., Cheng, Y., and Chow, J. C.: Dicarboxylic acids, ketocarboxylic acids and dicarbonyls in the urban roadside area of Hong Kong, Atmos. Environ., 40, 3030–3040, 2006.

9. p. 11, l. 3: What is the meaning of the slope of the correlation?

Response: In this study, the slope of the correlation between C_2 and SO_4^{2-} can be used to judge aqueous phase oxidation. When the slope is higher, the aqueous phase oxidation is easier to occur.

10. Table 1: All values should be rounded to significant digits, e.g. 86 +/- 33 instead *of 86.2+/- 33.8.*

Response: In order to keep 3 significant digits, we described malonic (C_3) concentration using 86.2 ± 33.8 ng m^{-3}. In addition, due to the analysis precision, when compound concentration less than 1 ng m^{-3}, we kept 2 significant digits. For example, concentration of suberic (C_8) was described using 0.33 ± 0.71 ng m^{-3}.

The uncertainties in the measurements don't justify the significant figures, so when concentration large than 1000 ng m^{-3}, significant digit in ones place was replaced by zero. Such as total concentration of dicarboxylic acids and related compounds in the day was described using 1050 ± 580 ng m^{-3}.

11. Figure 4: The x-axis is very blurry. I suggest using fewer tick marks.

Response: We have changed Figure 4 using fewer tick marks in the x-axis.

12. p. 2, l. 6: 'measure' should be 'measured'.

Response: changed.

(Page 2, Line 7)

13. p. 4, l. 22: blank samples.

Response: changed.

(Page 5, Line 13)

14. p. 4, l. 24, and remainder of the manuscript: VOC sampling (and VOC samples etc).

Response: Section 2.2 outlines the collection of VOCs samples, so we used "VOCs sampling". VOCs samples suggested type of collected sample. We have checked "VOC sampling" and "VOC samples" throughout the manuscript.

15. p. 6, l. 21: remove 'the trajectories'.

Response: removed.

(Page 7, Line 19)

16. p. 6, l. 22: have been given.

Response: changed.

(Page 7, Line 20)

17. p. 13, l. 20: 'related' should be 'correlated'.

Response: changed.

(Page 16, Line 17)

Table 1. Measured concentrations of DCRCs, $PM_{2.5}$, OC and EC at the top of Mt. Tai from 4 June to 4 July 2014.

	Day (n = 32)				Night (n = 27)				Day-Night Ratio
Components, abbreviation	Min.[a]	Max.[b]	Mean	SD [c]	Min.	Max.	Mean	SD	Mean
I. Dicarboxylic acids (ng m^{-3})									
Oxalic, C_2	122	1790	512	304	151	1280	534	272	1.0
Malonic, C_3	23.7	195	86.2	33.8	17.9	141	78.4	32.6	1.1
Succinic, C_4	25.8	485	126	81.1	62.3	227	121	53.7	1.0
Glutaric, C_5	6.88	99.3	26.3	15.9	10.5	53.7	24.9	11.3	1.1
Adipic, C_6	4.69	46.4	12.6	7.46	3.97	31.8	12.7	7.45	1.0
Pimelic, C_7	0.69	22.6	4.14	4.32	BDL	19.1	4.38	4.12	0.9
Suberic, C_8	BDL[d]	2.85	0.34	0.71	BDL	1.85	0.36	0.48	0.9
Azelaic, C_9	3.42	95.1	16.2	20.6	2.30	68.1	19.4	17.6	0.8
Sebabic, C_{10}	BDL	8.95	1.18	2.14	BDL	13.2	1.84	3.01	0.6
Undecanedioic, C_{11}	BDL	5.99	1.67	1.91	BDL	7.56	1.73	1.99	1.0
Dodecanedioc, C_{12}	BDL	1.33	0.20	0.33	BDL	1.83	0.29	0.46	0.7
Methylmalonic, iC_4	BDL	7.92	3.43	1.56	0.66	7.59	3.33	1.54	1.0
Methylsuccinic, iC_5	2.85	32.7	8.03	5.47	3.01	18.3	7.98	4.53	1.0
2-methylglutaric, iC_6	BDL	6.78	1.82	1.17	BDL	4.23	1.64	1.09	1.1
Maleic, M	BDL	25.7	7.44	7.23	2.19	23.7	6.56	5.03	1.1
Fumaric, F	0.99	15.1	3.90	2.73	BDL	7.13	3.15	1.85	1.2
Methylmaleic, mM	1.25	13.1	3.52	2.53	1.05	6.42	2.90	1.40	1.2
Malic, hC_4	0.18	5.15	1.19	1.04	0.29	3.27	1.37	0.75	0.9

Phthalic, Ph	19.3	99.4	36.8	16.1	16.3	53.9	29.3	10.2	1.3
Isophthalic, iPh	BDL	13.8	1.78	2.55	BDL	9.06	1.57	2.22	1.1
Terephthalic, tPh	0.88	130	13.6	24.9	0.58	155	12.9	32.2	1.1
Oxomalonic, kC_3	3.66	31.5	12.6	7.04	2.06	29.5	11.3	6.83	1.1
4-oxopimelic, kC_7	2.93	29.3	11.6	6.56	2.12	27.8	11.6	5.71	1.0
Subtotal	239	2950	893	479	358	1970	892	402	1.0
II. Oxocarboxylic acids (ng m^{-3})									
Pyruvic, Pyr	6.34	124	23.6	22.9	7.07	54.8	19.7	11.0	1.2
Glyoxylic, ωC_2	8.84	241	54.7	45.2	11.9	166	55.9	41.4	1.0
3-oxopropanoic, ωC_3	1.99	24.3	8.85	4.99	1.60	24.6	8.31	4.93	1.1
4-oxobutanoic, ωC_4	5.54	52.4	16.0	10.1	5.48	54.2	15.3	10.6	1.0
5-oxopentanoic, ωC_5	1.03	12.2	3.78	2.21	1.29	10.7	3.53	2.08	1.1
7-oxoheptanoic, ωC_7	2.09	17.8	7.31	3.33	1.93	13.7	6.67	2.95	1.1
8-oxooctanoic, ωC_8	1.13	29.9	9.40	5.66	0.86	18.3	8.78	4.43	1.1
9-oxononanoic, ωC_9	BDL	10.8	3.93	2.96	BDL	13.2	3.64	3.69	1.1
Subtotal	26.9	496	128	88.9	49.0	344	122	73.6	1.0
III.α-dicarbonyls (ng m^{-3})									
Glyoxal, Gly	1.82	59.6	12.3	10.3	3.20	39.3	12.4	9.68	1.0
Methylglyoxal, MGly	BDL	45.2	12.1	11.2	BDL	59.9	13.6	13.1	0.9
Subtotal	5.07	105	24.4	20.5	8.04	94.9	25.9	21.6	0.9
Total (all detected organics)	271	3550	1050	580	429	2380	1040	490	1.0
IV. Carbonaceous aerosols (μg m^{-3})									
$PM_{2.5}$	37.0	193	98.2	29.2	55.7	143	98.6	25.3	1.0
OC	4.42	30.7	11.6	5.77	4.02	32.9	11.7	7.75	1.0
EC	0.46	3.31	1.34	0.67	0.40	4.69	1.50	0.90	0.9

[a] Minimum. [b] Maximum. [c] Standard deviation. [d] BDL: Below detection limit.

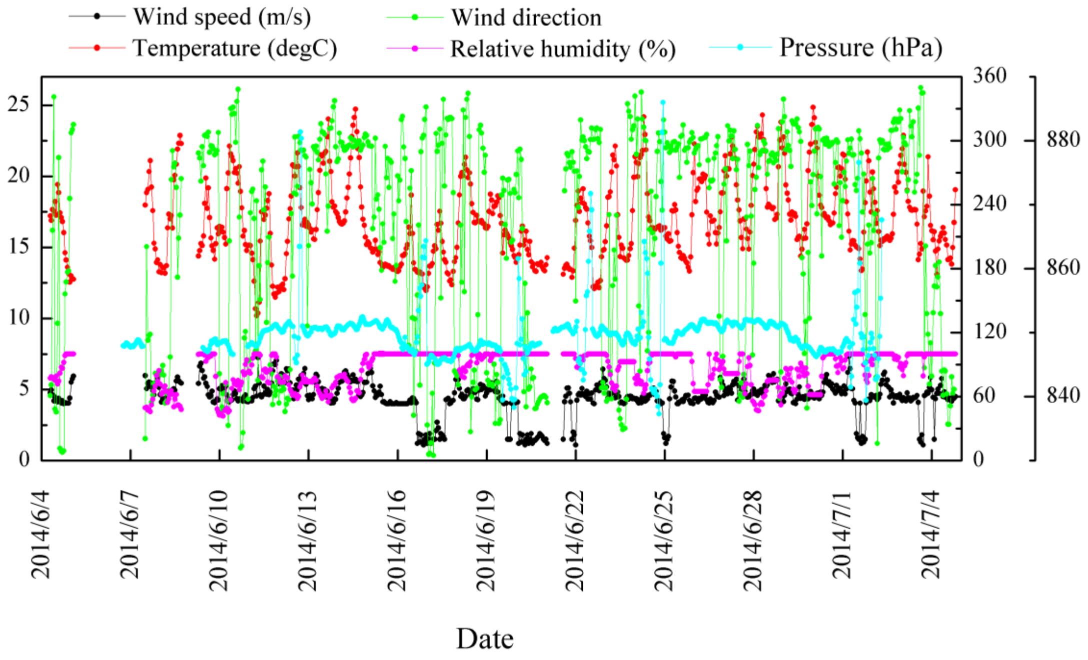

Fig. 1. Summary of meteorological data during the sampling period at Mt. Tai. The left y axis is for wind speed and temperature, whereas the right y1 axis (0-360) shows wind direction (degree) and relative humidity, right y2 axis (830-890) shows pressure.

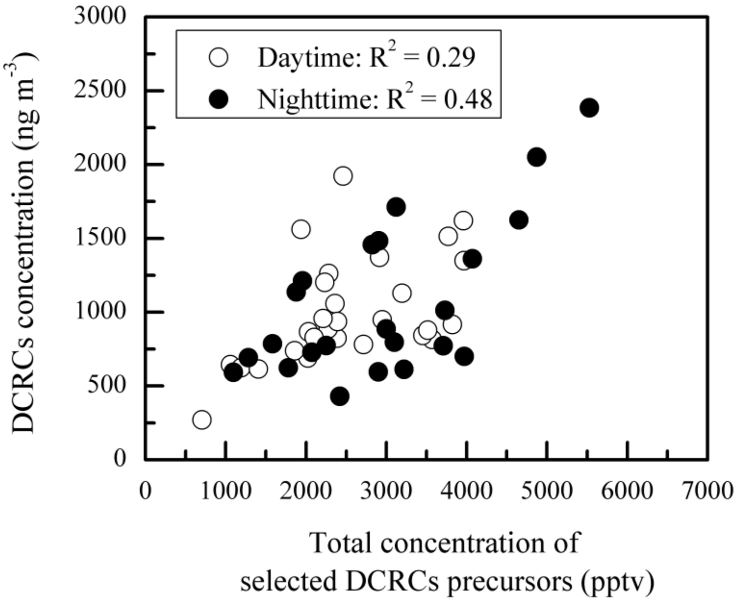

Fig. 4. Scatter plot of the day and night concentration of DCRCs and selected DCRCs precursors. Here, the total concentration of selected DCRCs precursors is the summed concentration of ethyne, ethene, isoprene, α-pinene, β-pinene, toluene, m/p-xylene and o-xylene.

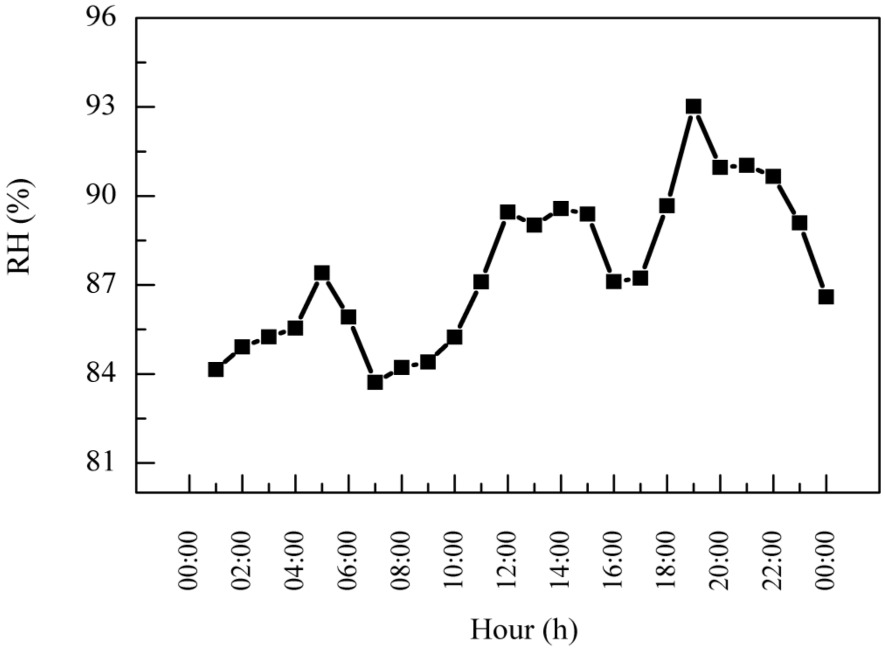

Fig. 5. Average diurnal variation of RH during the sampling period at the top of Mt. Tai.

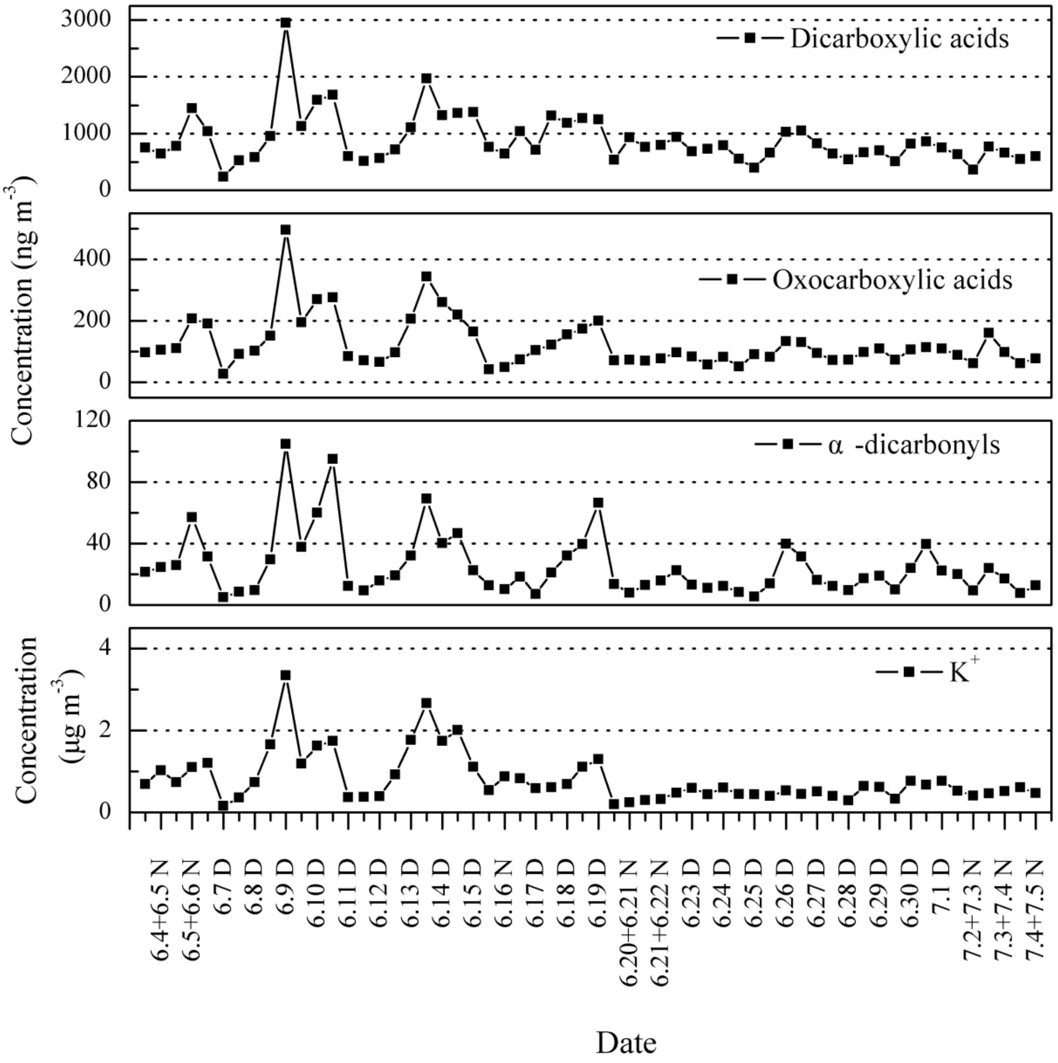

Fig. 7. Temporal variation of DCRCs and K$^+$ in $PM_{2.5}$ aerosols collected at Mt. Tai during the day (D) and night (N) in 2014.

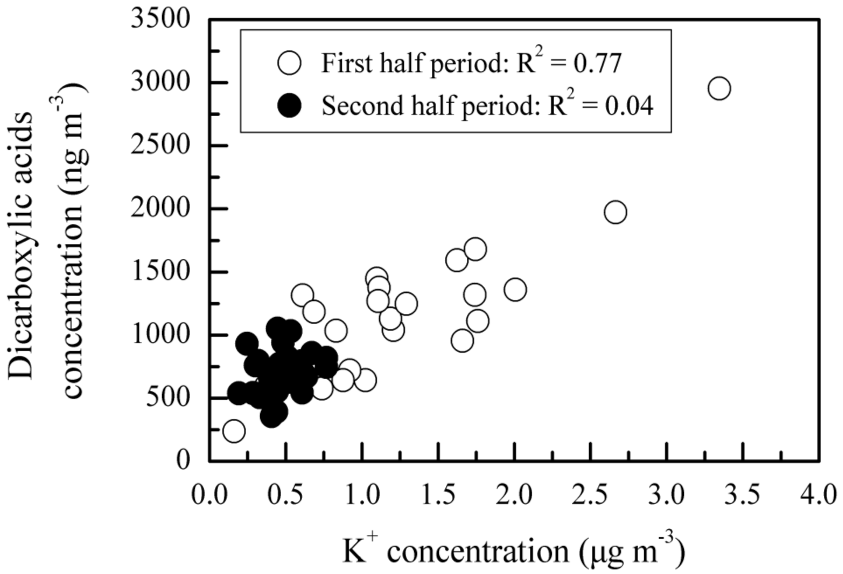

Fig. 8. Scatter plot of concentration between K^+ and dicarboxylic acids during the first and second half of the campaign.

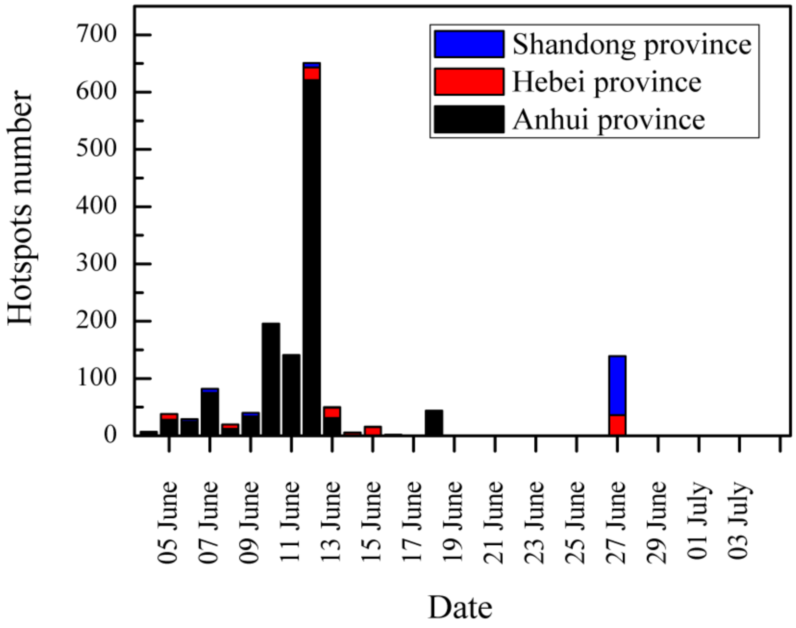

Fig. 9. Straw burning hotspots number in air masses passed over key areas during the sampling period reported by weather satellite of the Ministry of Environment Protection of the People's Republic of China.

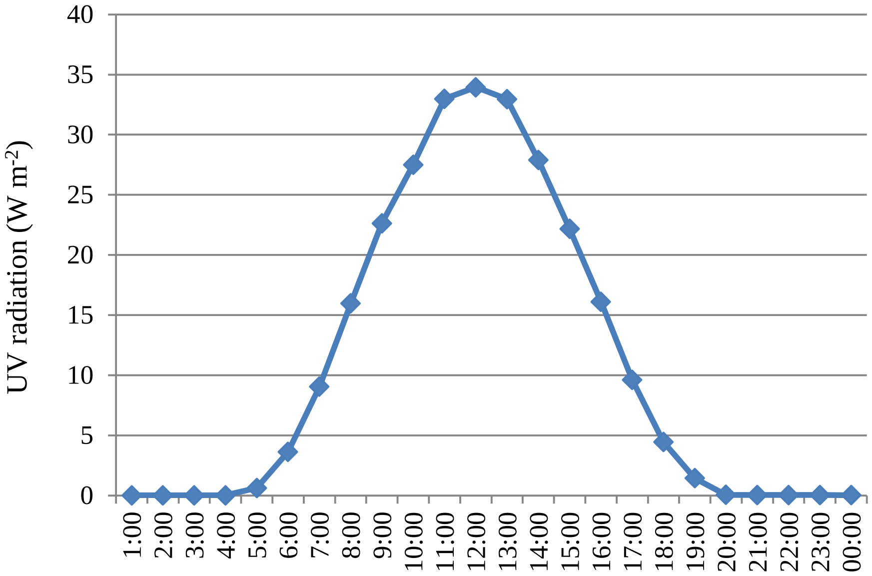

Fig. S1. The diurnal variation of ultraviolet (UV) radiation (W m^{-2}) during 04 June-04 July 2014 around Mt. Tai.

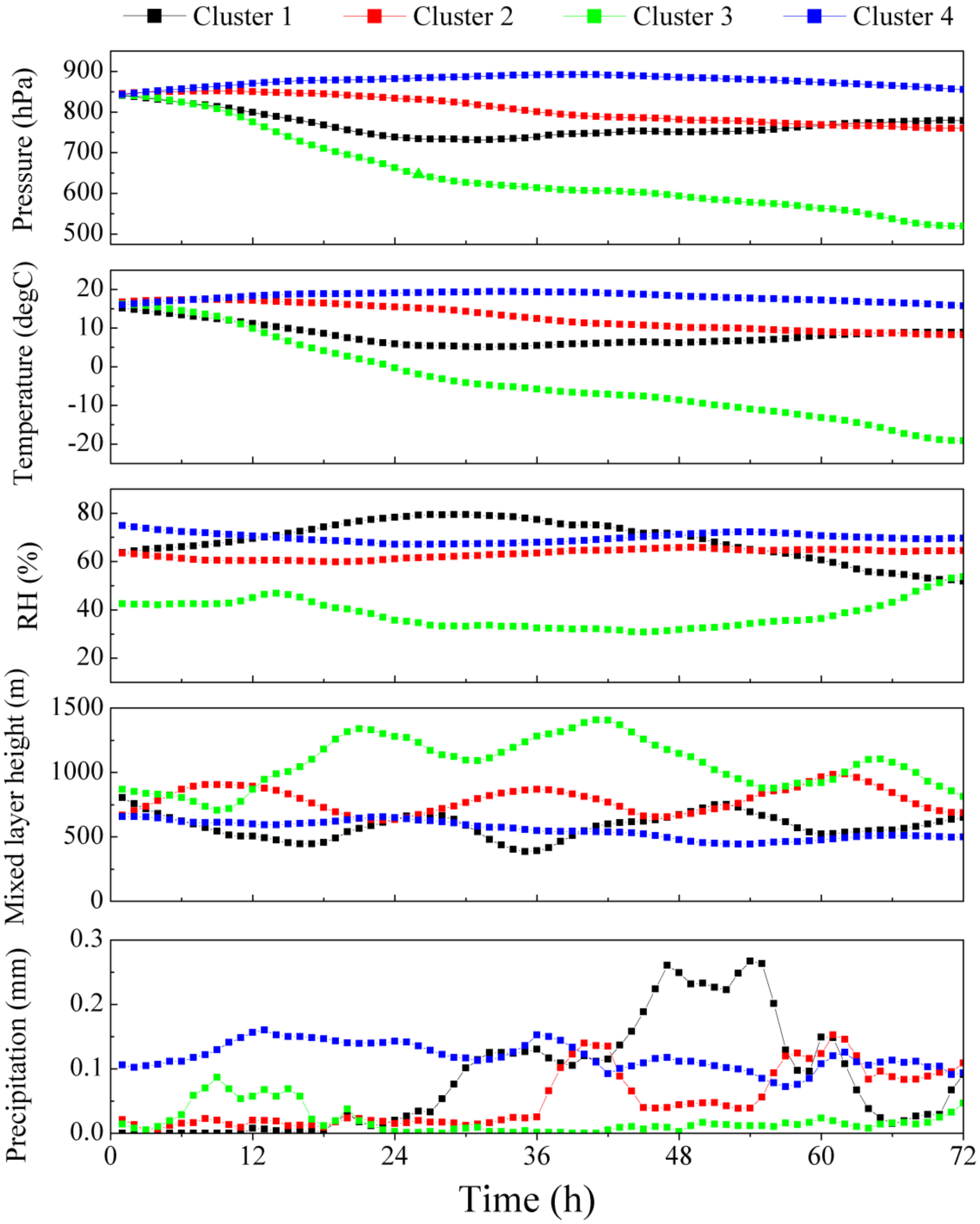

Fig. S2. Meteorological data in the different clusters during the sampling period at Mt. Tai.