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 2:

Zhu et al. discuss trends in concentrations of particle-phase polar oxygenated organic compounds during one month of summer 2014 at Mount Tai. The dataset presented is interesting, particularly in showing daytime versus nighttime measurements, boundary layer height (BLH) estimates, and a broad range of chemical species concentrations with ~high frequency. The use of principal component analysis (PCA) is also an apt way to summarize potential sources. However, the extent of discussion in the current draft is insufficient for these data and results: each data analysis piece is discussed separately, and cohesionis needed between the BLH estimates, back trajectories, PCA factors, and concentration trends. I believe the article therefore requires major revisions before final publication in the form of reorganization of the results and discussion, and additional synthesis of the conclusions.

Response: We thank the reviewer for the helpful comments. Below we address the comments and have revised the manuscript accordingly. For clarity, the reviewer's comments are listed below in black italics, whilst our responses and changes in manuscript are shown in blue and red, respectively. Revised table and figure are in the end.

1. There are several pieces of background information that are missing from the introduction. These include brief discussions (with references) of:

- Boundary layer behavior in complex topography;

- Biomass burning emissions and the new regulations mentioned (Pg. 8, line 26);

and - More about general emissions at Mount Tai.

Response: We have added background information about boundary layer behavior, biomass burning emissions and the new regulations mentioned, and emissions at Mount Tai as follows:

Moreover, it should be noted that mountain areas, with parallel ridges or isolated ridges and peaks, are different from the plain in terms of geometric structures. This has implications for modifying the ambient air flow by this complex terrain, which leads to complexity of the mountain boundary layer structure (Smith et al., 2002). Naturally, the boundary layer structure plays important roles in the transport and dispersal of atmospheric pollutants during long-range transport (Garratt, 1994).

(Page 4, Line 1-7)

In addition, the NCP is one of the most productive agricultural regions in China, and agricultural waste burning occurs frequently during the harvest seasons. Although some management strategies have been implemented by the Chinese government, such as lawful punishment or punishment by a fine, biomass burning still occurs during the harvest seasons (Zhu et al., 2017).

(Page 3, Line 15-20)

There are many tourists at Mt. Tai in summer, so there are some local emissions from small restaurants and temples (Gao et al., 2005). Furthermore, 80% of Mt. Tai is covered by vegetation (mostly bushes).

(Page 3, Line 28-Page 4, Line 1)

2. The methods section is lacking key information. Examples of additional information to be included (can go to supplemental material if desired):

- Details about the VOC concentrations: which species do "VOC concentrations" include?

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.

Several studies have demonstrated that, e.g., ethyne, ethene, isoprene, α-pinene, β-pinene, toluene, m/p-xylene, o-xylene represents potential DCRCs precursors (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 a scatter plot using total concentration of selected DCRCs precursors and DCRCs total concentration. The result was presented in Fig. 4.

We also added descriptions about 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)

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.

- *Discussion of whether the sampling period is representative of Mount Tai during all seasons, years, etc. (concentrations, BLH, and back trajectories).*

Response: In this study, the results were only representative of Mt. Tai in 04 June-04 July 2014. According to straw burning hotspots number from May to November 2014 reported by weather satellites of the Ministry of Environment Protection of the People's Republic of China (Fig. R1), we can see that June can represent the wheat burning season in 2014. Therefore, we changed title of the draft to “Molecular distributions of dicarboxylic acids, oxocarboxylic acids and α-dicarbonyls in $PM_{2.5}$ collected at the top of Mount Tai, in North China during wheat burning season 2014”.

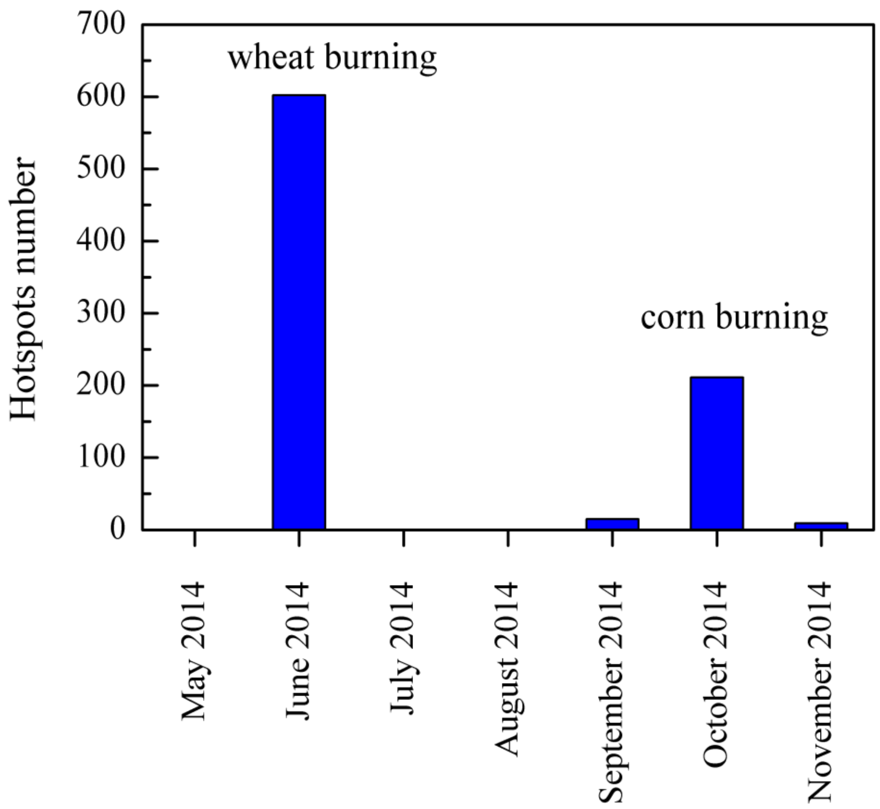

Fig. R1. Straw burning hotspots number in China from May to November 2014.

- *The method for calculating limits of detection for the measured chemical species.*

Response: Detection limits of the measured chemical species were calculated on the basis of minimum areas, which were set at usually 500 counts in Shimadzu CR7 integrator.

We have added method for calculating detection limits as follows:

Detection limits of the measured chemical species were 0.05 to 0.1 ng m^{-3}, which were calculated on the basis of minimum areas.

(Page 6, Line 25-27)

-*Uncertainties (specify type; e.g., standard deviation) about the measurements of each chemical species reported*

Response: Standard deviation is used to quantify the amount of variation of a set of data values. Therefore, we use standard deviation to present measured concentration of each chemical species. The standard deviation of measured DCRCs individual species has been provided in the revised manuscript. Please see Table 1.

- *Meteorological conditions and variations during the study*

Response: We have added descriptions about meteorological conditions and variations during the sampling period and shown in Fig. 1. Moreover, meteorological conditions have been added in the revised manuscript as follows:

The meteorological data during the sampling period are summarized in Fig. 1. The ambient temperatures covered a range of 10-25 ℃ with an average of 17 ℃. Relative humidity (RH) varied between 58 to 100% with an average of 87%. Winds generally came from the northwest, and wind speeds ranged from 1-7 m s^{-1}. Weather conditions during the campaign were mostly cloudy and occasionally foggy. 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 4, Line 24-Page 5, Line 3)

- *Frequency of blanks*

Response: We have added the frequency of blanks as follows:

Blank samples were collected between 06:00-18:00 and 18:00-06:00 local time from 5-7 July 2014, and their sampling manner was similar to the real samples, but without pumping.

(Page 5, Line 12-15)

- *A brief synopsis of data used in this article from Zhu et al., 2017*

Response: We have added a brief synopsis of data used in this study from Zhu et al. (2017) as follows:

In our previous publication (Zhu et al., 2017), we described the meteorological conditions, sampling site, $PM_{2.5}$ sampling, VOCs sampling and analysis from 04 June to 04 July 2014 at Mt. Tai. Therefore, in this study we describe these experimental methods only briefly.

(Page 4, Line 16-19)

- In the PCA analysis: is the replacement of values below detection limit with have the value a common convention? I am not familiar with this technique, and it seems like it may bias the measurements low.

Response: According to Wold et al. (1987), when compound concentration was below the detection limit, the data was replaced by a value half of the corresponding detection limit. The method has been used in van Pinxteren et al. (2014).

Reference:

van Pinxteren, D., Neusüß, C., and Herrmann, H.: On the abundance and source contributions of dicarboxylic acids in size-resolved aerosol particles at continental sites in central Europe, Atmos. Chem. Phys., 14, 3913-3928, 2014.

Wold, S., Esbensen, K., Geladi, P.: Principal component analysis, Chemometr. Intell. Lab., 2, 37-52, 1987.

-Please report a reference for this if possible, and discuss briefly (this can be in the supporting information).

Response: We have added reference in the revised manuscript as follows:

If the compound concentration was below the detection limit, the data were replaced by a value half of the corresponding detection limit (Wold et al., 1987).

(Page 15, Line 28-Page 16, Line 1)

3. The results and discussion section should be reorganized to offer a more cohesive analysis of all analytical tools/results. Some specific examples include the following.

- A relationship can be drawn between back trajectory clusters and the chemical concentrations/PCA factors. Do dates of influence of particular source regions align with sources/PCA factors? Do the dominant back trajectory clusters change between the first and second halves of the study, which seem to have different chemical features?

(1) A relationship can be drawn between back trajectory clusters and the chemical concentrations.

Response: We have connected DCRCs concentrations with back-trajectory clusters as follows:

According to back-trajectories and classification results of DCRCs in different back-trajectory clusters in Fig. 2, we can see that DCRCs concentrations were mostly higher in air masses that originated from north of Mt. Tai (northern Hebei province) (31%) and south of Mt. Tai (northern Anhui province) (48%), but lower in air masses derived from the ocean (11%) and Siberia (10%).

(Page 9, Line 20-25)

(2) A relationship can be drawn between back trajectory clusters and the PCA factors.

Response: We have added back-trajectory parameters (mean trajectory length, solar flux along trajectory, mixing depth along trajectory) in PCA analysis as follows:

The negative loading of mean trajectory length and mixing depth along the trajectory to PC1 suggested high residence times of trajectories above continental areas.

(Page 16, Line 9-11)

The low loading of solar flux in PC2 indicated that secondary sources were not primarily driven by radiation.

(Page 16, Line 13-14)

The negative loading of mean trajectory length and mixing depth along the trajectory in PC1, PC2 and PC3 indicates long residence times above the continental areas.

(Page 17, Line 3-5)

(3) Do dates of influence of particular source regions align with sources/PCA factors?

Response: Using PCA method, DCRCs sources in the first and second half sampling periods were identified, respectively. As shown in Table R1, sources in principal components (PCs) in the two periods were as follows:

In the first half sampling period:

PC 1: anthropogenic activities (such as fossil fuel and biomass burning) followed by photochemical aging

PC 2: secondary sources

PC 3: fuel combustion, solid wastes/plastic polymers burning

PC 4: photooxidation of unsaturated fatty acids emitted from the sea surface together with sea salt particles

In the second half sampling period:

PC 1: anthropogenic activities (such as fossil fuel combustion) followed by photochemical aging

PC 2: secondary sources

PC 3: fuel combustion, solid wastes/plastic polymers burning

PC 4: photooxidation of unsaturated fatty acids emitted from the sea surface together with sea salt particles

We can see that only biomass burning was different in the two periods, other sources were the same in the two periods. Biomass burning was important in the first half sampling period, while in the second half sampling period, biomass burning had no impact. 3.5 discussion part has presented detailed descriptions about the importance of biomass burning during the first half sampling period.

Table R1. PCA analysis results for DCRCs in the first and second half sampling periods, respectively.

Compounds	The first half sampling period				The second half sampling period			
C_2	**0.765**	0.339	0.409		**0.734**	0.367	0.411	
C_3	**0.879**	0.318			**0.882**	0.302		
C_4	**0.725**	0.402	0.512		**0.697**	0.382	0.523	
C_5	**0.798**		0.590		0.484		**0.605**	
C_6	0.570				0.371			
C_9				**0.772**				**0.792**
iC_5	**0.683**		0.619		**0.637**		0.652	
M			**0.824**				**0.847**	
F	**0.592**	0.207	**0.607**		**0.627**		**0.613**	
hC_4	**0.609**	0.197			0.502			
Ph	**0.616**		0.398		**0.639**		0.351	

tPh			**0.626**				**0.661**	
kC_3	**0.670**				**0.606**			
Pyr	**0.768**	0.332			**0.731**	0.393		
ωC_2	**0.812**	0.397			**0.797**	0.342		
ωC_4	**0.809**				**0.693**			
Gly	**0.813**	0.396			**0.761**	0.304		
MGly	**0.618**	0.429			0.582	0.471		
OC	**0.728**			0.470	**0.717**			0.528
EC	0.428	0.186	**0.679**		0.457		**0.702**	
Na^+		0.339		**0.832**		0.359		**0.857**
NH_4^+	0.365	**0.827**			0.380	**0.892**		
K^+	**0.709**	0.253						
NO_3^-	0.239	**0.792**	0.397		0.253	**0.778**	0.278	
SO_4^{2-}	0.272	**0.819**			0.277	**0.719**		
Mean trajectory length	**-0.645**	-0.589	-0.535		**-0.683**	-0.602	-0.589	
Solar flux along trajectory	0.328	0.389		0.272	0.356	0.210		
Mixing depth along trajectory	-0.478	-0.492	-0.361		-0.519	-0.418	-0.432	
Variance (%)	68%	10%	9%	6%	57%	18%	11%	7%

(1) Do the dominant back trajectory clusters change between the first and second halves of the study, which seem to have different chemical features?

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)

- *The authors note that there are relationships between the VOC concentrations and those of the polar organic species measured. Please provide an explanation of what this relationship might be: are the higher concentrations of polar organic species at Mount Tai in 2014 a result of the aging of the measured VOCs? Could they have been directly emitted together as primary aerosol particles? Both? Please support with references. If possible and relevant, please also consider individual VOCs.*

Response: When aerosols are aged, C_4 will be oxidized to C_3. However, in this study, C_4 concentrations were much higher than C_3. Therefore, it was possible that polar organic compounds together with VOCs originated from primary sources.

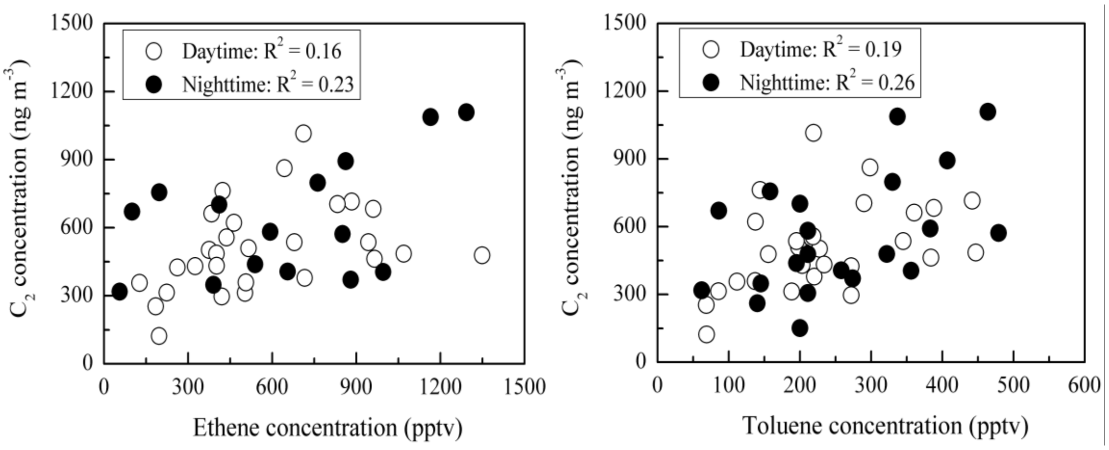

Fig. R2. Scatter plot of the day and night concentration between ethene/toluene and oxalic acid (C_2).

As shown in Fig. R2, ethene or toluene presented correlation with oxalic acid (C_2), but not strong. When ethene or toluene concentration increased, C_2 concentration also increased. These results suggested the higher concentrations of polar organic species at Mt. Tai in 2014 were likely caused by the aging of the measured VOCs.

- *A relationship could be drawn between how BLH estimates might alter the effect of long-range transport (back trajectories) on concentrations. Even if the BLH is only above the sampling location during some sampling times, these could be interesting.*

Response: We have made a graph about relationship between BLHs and total concentration of DCRCs and shown in Fig. 3.

We have added descriptions about relationship between BLHs and total concentration of DCRCs as follows:

The BLHs were higher during the day, peaking near noontime. The boundary layer occasionally extended high enough during the day to approach the sampling site (Fig. 3). However, the maximum BLH was only ~ 600 m during the night, which was much lower than the sampling site height. As shown in Fig. 3, the total concentration of DCRCs increased when BLHs were higher than the site elevation. These results suggest that mountain/valley breezes may bring ground-level pollutants to the summit of Mt. Tai during the day when the BLHs were above the sampling site height.

(Page 12, Line 8-14)

4. The MGly recovery is estimated to be~50%. Do the authors expect trends in concentration of MGly, then, to be meaningful? Why is the recovery of Gly expected to be so different?

Response: MGly recovery was ca. 50% using authentic standard. The reproducibility of MGly is good enough (analytical errors < 30%) to discuss the changes in the concentrations and trends.

5. Daytime/nighttime differences:

- The daytime/nighttime analysis gives a summary of the results, but provides little explanation for the observations. How are these trends informative? Please explain the hypothesis about aqueous photochemical reactions (pg. 10, line 22) more thoroughly and with references. Can the similarities between daytime/nighttime concentrations be supported by looking at diurnal changes in relative humidity, or contrast between high elevation/summit and low-elevation/base measurements of any kind at Mount Tai?

Response: Average diurnal change of RH was presented in Fig. 5. And we have added description about average diurnal variation of RH 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)

During the campaign (04 June-04 July 2014) at Mt. Tai, measured ozone (O_3) concentration was 85 ± 21 ppb. However, at the same period (04 June-04 July 2014),

O_3 concentration at the foot of Mt. Tai was 123 ± 18 ppb (https://www.zq12369.com/environment.php?city). The large difference of O_3 concentration between high elevation and low elevation measurements suggested that anthropogenic sources in the valleys had low impact on pollutant concentrations at the top of Mt. Tai. As a consequence of the low BLH and mixing layer height, air masses in the low and high altitude might be separated.

-The authors suggest that the strengths of nighttime vs. daytime correlations in Figures 2 and 3 explain daytime/nighttime ratios reported (although, confusingly, these ratios are ~1 for most species). However, the correlations are not clearly different (daytime/nighttime) in either figure. Please find agreement between the daytime/nighttime ratios, Figures 2 and 3 correlations, and the hypotheses about diurnal variations in concentrations/atmospheric processes.

Response: We have changed Fig. 2, and now shown in Fig. 4. Ethyne, ethene, isoprene, α-pinene, β-pinene, toluene, m/p-xylene and o-xylene were used as DCRCs precursors-VOCs, which were pointed out in published papers as sources of dicarboxylic acids (Warneck, 2003; Ervens et al., 2004; Bikkina et al., 2014; Tilgner and Herrmann, 2010). Then we made scatter plot using total concentration of selected DCRCs precursors and DCRCs concentration. The result was presented in Fig. 4. The moderate correlations suggested that the difference in the day and night measurements was possibly dependent on the amount of precursor emissions.

We added descriptions about 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)

We have changed Fig. 3 by deleting outlying point at ~1800 ng m^{-3} C_2 and 35 µg m^{-3} SO_4^{2-}, and now shown in Fig. 6. Higher correlation at the night suggested more important of aqueous phase oxidations at the night.

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.

-Figure 3 includes one outlying point at ~1800 ng m-3 C2 and~35ð˙IIJ˘Ggm-3 SO42-; what is the result of removing this point? This looks to me to be driving the daytime/nighttime difference. There is certainly a relationship between these two chemical species, but this may not be easily related to the iron-oxalate hypothesis drawn.

Response: We have deleted the point at ~1800 ng m^{-3} C_2 and 35 μg m^{-3} SO_4^{2-}. The result was presented in Fig. 6, which was similar with the result including the point.

According to Ervens et al. (2003), Tilgner and Herrmann (2010), Pavuluri and Kawamura (2012), we proposed hypothesis that photolysis of iron-oxalate complexes can be an important sink of C_2 in the aqueous phase. Moreover, Weller et al. (2014) reported iron complex photolysis was an important sink for oxalate, with a complex photolysis contribution to overall degradation of 99%, compared to all possible sink reactions with atmospheric aqueous phase radicals, such as •OH, NO_3• and SO_4^-•.

We have changed the old sentence about photolysis of iron-oxalate complexes as follows:

In addition, photolysis of iron-oxalate complexes is considered as an important sink of C_2, which is effective under clear-sky sunlight conditions (Ervens et al., 2003; Pavuluri and Kawamura, 2012; Weller et al., 2014). Therefore, the removal of C_2 was lower during the night than during the day.

(Page 13, Line 15-19)

Reference:

Ervens, B., George, C., Williams, J., Buxton, G., Salmon, G., Bydder, M., Wilkinson, F., Dentener, F., Mirabel, P., and Wolke, R.: CAPRAM 2.4 (MODAC mechanism): An extended and condensed tropospheric aqueous phase mechanism and its application, J. Geophys. Res. Atmos., 108, 2003.

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.

Pavuluri, C. M., and Kawamura, K.: Evidence for 13 - carbon enrichment in oxalic acid via iron catalyzed photolysis in aqueous phase, Geophys. Res. Lett., 39, 2012.

Weller, C., Tilgner, A., Bräuer, P., Herrmann, H.: Modeling the Impact of Iron−Carboxylate Photochemistry on Radical Budget and Carboxylate Degradation in Cloud Droplets and Particles, Environ. Sci. Technol., 48: 5652−5659, 2014.

6. Although the BLH discussion is essential to this analysis, uncertainty in estimating the BLH using a model at a mountaintop should be discussed briefly. In addition, results from the Mount Tai Experiment (Kanaya et al., 2013) showed that their sampling site was above the BLH during many days, and within the residual layer some nights. Please contrast the estimates of these two studies briefly.

Response: We have added descriptions about uncertainty as follows:

In YSU scheme, the boundary layer approaches its top when the critical bulk Richardson number is zero (Hong et al., 2006). The YSU scheme simulates deep vertical mixing accurately in buoyancy driven BLHs (Hong et al., 2006), and reasonably captures the diurnal cycle of BLHs and thermodynamic vertical structure of atmosphere (Hu et al., 2010). The daytime boundary layer structure is well represented by the WRF model with the YSU BLH scheme (Hu et al., 2013). Previous studies also reported that the WRF model can capture the boundary layer structure and local circulation over the NCP mountainous region during summer (Chen et al., 2009), and well captures the vertical structure of potential temperature (Hu et al., 2014). Although the model may have lower confidence for the night BLH estimation (Hu et al., 2013), the larger uncertainty at night would not significantly influence our analysis and conclusions because the nighttime boundary layer was always lower than the measurement site.

(Page 6, Line 3-15)

The comparison with Kanaya et al. (2013) was as follows:

The result was different from a previous study at Mt. Tai in 2006 (Kanaya et al., 2013), which reported that the daytime BLHs during the campaign were mostly higher than the observation site, and the night BLHs were generally within the residual layer and occasionally in the free troposphere. Different meteorological conditions between 2006 and 2014 at Mt. Tai, such as higher air pressure conditions but quite low wind speeds in 2014, were the probably reasons for the BLH differences.

(Page 9, Line 10-15)

7. Could any of the back trajectories suggest that regional emissions from the previous day or two impacted the measured concentrations? Long range transport is suggested to be dominant throughout the study, but perhaps regional emissions have been transported aloft due to topography and/or convection.

Response: As shown in Fig. 2, clusters 1 and 3 contained the highest altitudes, and their heights were all higher than 2000 m. The result suggested all heights of clusters 1 and 3 were higher than the maximum of BLHs. Therefore, clusters 1 and 3 can’t bring ground pollutants to Mt. Tai elevation.

Only 5% of cluster 2 and 9% of cluster 4 back-trajectories were firstly lower than

BLHs, then higher than the site elevation and possibly brought ground pollutants to Mt. Tai top. Therefore, pollutant concentrations at Mt. Tai were largely controlled by non-local transport.

8. Contrast with other studies:

- The contrast between this summer 2014 study at Mount Tai and others is informative. However, the ratio used for conversion of TSP to PM2.5 likely introduces large uncertainties. Is this ratio relevant for Mount Tai or the North China Plain? For summer? For 2014? Specify briefly, and consider the degree of confidence that the reader can have in these estimated concentrations, including significant digits of concentrations reported. Please note that composition is size-dependent for aerosol particles.

(1) Is this ratio relevant for Mount Tai or the North China Plain? For summer? For 2014?

Response: Using the ratio of $PM_{2.5}$/TSP = 0.91 was reasonable for calculating DCRCs concentrations in $PM_{2.5}$ in June 2006 at Mt. Tai.

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 from June 2006 at Mt. Tai. Therefore, using the ratio of $PM_{2.5}$/TSP = 0.91 was reasonable to obtain DCRCs and levoglucosan concentrations in $PM_{2.5}$ in June 2006 at Mt. Tai.

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.

Due to the reviewer comment, the text on $PM_{2.5}$/TSP has been extended more details 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)

(2) Specify briefly, and consider the degree of confidence that the reader can have in these estimated concentrations, including significant digits of concentrations reported.

Response: As above descriptions, using the ratio of $PM_{2.5}$/TSP = 0.91 can obtain reasonable DCRCs concentrations in $PM_{2.5}$ in June 2006 at Mt. Tai. Significant digits of these estimated concentrations were consistent with measured concentrations of DCRCs in this study.

When compared with other studies, values reported in the published papers were used. We didn't make any changes. Therefore, significant digits of concentrations were not consistence.

(3) Please note that composition is size-dependent for aerosol particles.

Response: When compared with other studies, we have added size data as follows:

Compared with other Asian urban sites, the total concentration of DCRCs reported here was about 1-2 times higher when compared with those reported in PM_{10} in Chennai, India in 2007 (Pavuluri et al., 2010b), and in TSP in Tokyo, Japan in 1989 (Kawamura and Yasui, 2005) and Sapporo, Japan in 2005 (Aggarwal and Kawamura, 2008), but lower than that in $PM_{2.1}$ in Raipur, India in 2012-2013 (Deshmukh et al., 2016).

(Page 11, Line 9-14)

Meanwhile, the result reported in this study was about 5 and 13 times higher than those in PM_{10} in Leipzig, Germany in 2003-2005 (van Pinxteren et al., 2014) and in TSP in Zurich, Switzerland in 2002 (Fisseha et al., 2006), respectively.

(Page 11, Line 16-18)

- *Please consider the season, year, and mountainous/urban/rural category of these studies (include study year in the comparison table as well).*

Response: We have added year in the Table 2. And we have added "year" in the manuscript as follows:

Compared with the Chinese megacities, such as Guangzhou in 2007 and Beijing in 2006 (Ho et al., 2010; Ho et al., 2011), the total concentration of DCRCs at Mt. Tai in 2014 was about 1-2 times higher. The concentration of dicarboxylic acids at Mt. Tai in 2014 was similar to the concentration reported in 14 Chinese cities in 2003 (Ho et al., 2007), while oxocarboxylic acids and α-dicarbonyls were more than three times higher. Compared with other Asian urban sites, the total concentration of DCRCs reported here was about 1-2 times higher when compared with those reported in PM_{10} in Chennai, India in 2007 (Pavuluri et al., 2010b), and in TSP in Tokyo, Japan in 1989 (Kawamura and Yasui, 2005) and Sapporo, Japan in 2005 (Aggarwal and Kawamura, 2008), but lower than that in $PM_{2.1}$ in Raipur, India in 2012-2013 (Deshmukh et al., 2016). Furthermore, the Mt. Tai DCRC total concentration in 2014 was approximately 13 times higher compared with Houston, USA in 2000 (Yue and Fraser, 2004). Meanwhile, the result reported in this study was about 5 and 13 times higher than those in PM_{10} in Leipzig, Germany in 2003-2005 (van Pinxteren et al., 2014) and in

TSP in Zurich, Switzerland in 2002 (Fisseha et al., 2006), respectively.

(Page 11, Line 4-18)

9. Biomass burning discussion:

- The biomass burning discussion is interesting, but incomplete. The authors draw the conclusion that, "...from 2006 to 2014, biomass burning decreased by about 80%." This conclusion cannot be drawn from the estimated concentration of a single species (levoglucosan). Many factors could confound this relationship, such as atmospheric oxidant concentrations, or meteorology during the study. Please rephrase and support with additional observations.

Response: Only based on decrease of levoglucosan concentrations, we can't conclude that "from 2006 to 2014, biomass burning decreased by about 80%". Atmospheric oxidant concentrations and meteorological parameters at Mt. Tai between 2014 and 2006 were different. According to Kanaya et al. (2009), the simulated maximum of OH concentrations over Mt. Tai in June 2006 was 6×10^6 molecules cm^{-3}. However, in our follow-up study about "Modelling multiphase chemistry of secondary aerosol obtained at Mt. Tai in 2014", maximum OH concentration in 04 June-04 July 2014 at Mt. Tai was about 3×10^6 molecules cm^{-3}, which was half of 2006 result. Moreover, compared meteorological parameters in June 2006 (Kawamura et al., 2013) with our study, we also found some differences. However, we can't deeply study impact of atmospheric oxidants and meteorological parameters.

We have changed the old sentence as follows:

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 11, Line 1-3)

Reference:

Kanaya, Y., Pochanart, P., Liu, Y., Li, J., Tanimoto, H., Kato, S., Suthawaree, J., Inomata, S., Taketani, F., Okuzawa, K., Kawamura, K., Akimoto, H., and Wang, Z. F.: Rates and regimes of

photochemical ozone production over Central East China in June 2006: a box model analysis using comprehensive measurements of ozone precursors, Atmos. Chem. Phys., 9, 7711–7723, 2009.

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.

- *Please include more information about the "emission hotspots" mentioned on pg. 11, line 27, along with references. Are these the locations of biomass burning events? On a related note, please discuss whether there is any indication that biomass burning events decreased between the first and second halves of the study (satellite data, perhaps). Do trends in concentrations match observations in biomass burning events?*

Response: We have shown straw burning hotspots in Fig. 9 and added more information about emission hotspots as follows:

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 17-22)

10. PCA analysis:

- *Please be more explicit about the methods and the vocabulary used to describe the results. Specifically, in the methods section, the authors should include not only the information at the beginning of section 3.6, but also whether the data were standardized or mean-centered. Are the "weighting factors" the same as the "factor-loadings"? Please label which values are reported in the table.*

(1) Specifically, in the methods section, the authors should include not only the information at the beginning of section 3.6, but also whether the data were standardized or mean-centered.

Response: We have added more descriptions about data standardization and PCA method in the methods section as follows:

2.6 The principal component analysis (PCA) method

Principal component analysis (PCA) is a multivariate analytical tool. It starts with a great many correlated variables and attempts to find a smaller number of independent factors, which can explain the variance in data. Here, the compound concentrations should firstly be transformed into standardized form using the following formula:

$$Z_{ij} = \frac{C_{ij}-\overline{C_j}}{\sigma_j}$$

where i = 1, ⋯, n sample; j = 1, ⋯, m compound; C_{ij} is the concentration of compound j in sample i; and $\overline{C_j}$ and σ_j are the arithmetic mean concentration and the standard deviation for compound j, respectively. The derived variables are linear combinations of original variables (Callén et al., 2009). In order to better identify the influence of the original variables, varimax rotation is used to obtain the rotated factor loadings that reflect the contribution of each variable to its principal component (PC) (Almeida et al. 2005; Viana et al., 2006). Factor loadings reveal how much a variable contributes to the corresponding PC and how well a variable differs from others. Only factors with eigenvalues greater than 1 are extracted based on Kaiser-Meyer-Olkin (KMO) and the Bartlett's test of sphericity.

(Page 7, Line 22-Page 8, Line 11)

(2) Are the "weighting factors" the same as the "factor-loadings"?

Response: The "weighting factors" is the same as the "factor-loadings". In this study, only the term "factor-loadings" is used. We have changed "weighting factors" to "factor-loadings".

(3) Please label which values are reported in the table.

Response: We have added descriptions about values reported in the table as follows:

In this study, PCA was employed to identify the DCRC sources in $PM_{2.5}$. Concentrations of C_2, C_3, C_4, C_5, C_6, C_9, iC_5, hC_4, M, F, Ph, tPh, kC_3, Pyr, ωC_2, ωC_4, Gly, MGly (compound abbreviation in Table 1), OC, EC, Na^+, NH_4^+, K^+, NO_3^- and SO_4^{2-} as well as mean trajectory length, solar flux along the trajectory and mixing depth along the trajectory were used for PCA using IBM SPSS Statistics 21.0, and the results are presented in Table 3 and Table 4. If the compound concentration was below the detection limit, the data were replaced by a value half of the corresponding detection limit (Wold et al., 1987). Only factor loadings $|x| > 0.2$ were considered, and $|x| > 0.6$ were considered high loading and are depicted in bold.

(Page 15, Line 23-Page 16, Line 2)

- A distinction is made between daytime and nighttime concentrations in the PCA analysis, and slightly different factors are identified. Please provide explanations for differences between all of the daytime and nighttime factors. (In the case of the nighttime factor 4, mixed marine and plastic burning emissions are suggested—please explain further and cite references.) Please consider agricultural activities as a possible emissions source.

(1) Please provide explanations for differences between all of the daytime and nighttime factors.

Response: We have added explanations for differences between day and night factors as follows:

Day and night sources of DCRCs were similar, but there were some differences in source order and contribution. Anthropogenic activities followed by photochemical aging had a higher contribution during the day, which was probably related with higher BLHs during the day. Fuel combustion was the second most important source during the night, and its contribution was also higher during the night. Although secondary processing was the third most important source at night, its contribution was higher than that during the day, which may be related to more effective aqueous oxidation during the night. The daytime sources in PC4 and PC5 were not separated during the night.

(Page 17, Line 11-19)

(2) In the case of the nighttime factor 4, mixed marine and plastic burning emissions are suggested—please explain further and cite references.

Response: We have further explained night PC4 and cited references as follows:

The correlation between C_9 and Na^+ (R^2 = 0.51) suggests photooxidation of unsaturated fatty acids emitted from sea surface together with sea salt. As mentioned above, tPh is produced by wastes burning. High correlations of C_9, tPh, OC and Na^+ in PC4 may reveal a mixed aerosol source related to waste burning and photooxidation of unsaturated fatty acids emitted from the sea surface together with sea salt.

(Page 17, Line 5-10)

(3) Please consider agricultural activities as a possible emissions source.

Response: We have added agricultural activities as follows:

As shown in Table 3, PC1 was dominated by high loadings of C_2-C_6, iC_5, F, hC_4, Ph, kC_3, Pyr, ωC_2, ωC_4, Gly, MGly, OC and K^+, which were associated with anthropogenic activities (such as agricultural activities) followed by photochemical aging.

(Page 16, Line 4-7)

11. The phrase "dicarboxylic acids and related compounds" is overused in the paper, and must be abbreviated for clarity. Please find an appropriate way to do so. An example might be "polar organic compounds (POCs)".

Response: We have defined an abbreviation "DCRCs" for "dicarboxylic acids and related compounds", and have changed it throughout the draft.

12. When reporting values summarizing the campaign data, be clear about whether the value is a mean, etc., in every case.

Response: We have added "mean" or "total" when summary the campaign data. And have checked them throughout the manuscript.

13. Throughout the document, please revise for grammar and accuracy of the wording. For example, on pg. 7, line 29, "trends" should be "concentrations".

Response: We have changed "trends" to "concentrations", and revised grammar and accuracy of the wording throughout the manuscript.

14. Please choose a consistent spelling and format for the following terms: "airmass", "daytime" vs. "day", "nighttime" vs. "night", "back trajectory" vs. "back-trajectory".

Response: We have changed "air mass" to "airmass", "daytime" to "day", "nighttime" to "night", "back trajectory" to "back-trajectory" throughout the manuscript.

15. Pg. 3, line 8: Is this really true that dicarboxylic acids and related compounds are typically studied in TSP rather than PM2.5? Please revisit.

Response: We have deleted the old sentence and changed Pg. 3, line 8 in revised manuscript as follows:

Therefore, it is necessary to study the DCRC characteristics in $PM_{2.5}$.

(Page 3, Line 7-8)

16. Section 3.2 (and throughout): The discussion of the contributors to "dicarboxylic acids and related compounds" would be much stronger with some context (rather than an empirical grouping of chemicals based on methods). What does this category of chemicals represent in the atmosphere? Could it be representative of water soluble organic carbon? Oxygenated organic species in general? Please support this with references.

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.

17. Top of pg. 8: Please clarify the definition of each of the percentages reported here. Are these all percentages of the total dicarboxylic acids concentration?

Response: We have added descriptions about percentages as follows:

C_2 was found to be the most abundant dicarboxylic acid compound. The relative abundance of C_2 in the total concentration of dicarboxylic acids was 57% during the day and 60% during the night, followed by C_4 (day: 14%, night: 14%) and C_3 (day: 10%, night: 9%).

(Page 10, Line 2-5)

18. There are several scientific language choices that should be reconsidered: "significant" should be used only when statistical significance is demonstrated; "levels" of chemicals is not precise - please use "concentrations"; "considerable amount" is not precise – please use "substantial concentration", for example.

Response: We have changed "significant" to "important", "levels" to "concentrations", "considerable amount" to "substantial concentration" through the manuscript.

19. Where coefficients of determination are discussed, please also report the values within the text.

Response: Coefficients of determination have shown in Table 3 and Table 4. Discussions about coefficients of determination have presented in 3.6 part.

20. Please introduce each chemical abbreviation in the article body (e.g., "C2" for oxalic acid, "VOCs" for volatile organic compounds).

Response: We have introduced each chemical abbreviation through the draft.

21. Note that phthalic acid and azelaic acid both have primary as well as secondary atmospheric sources.

Response: Phthalic acid (Ph) is produced in the atmosphere by photochemical oxidation of naphthalene and its alkylated forms, which are derived from incomplete combustion of fossil fuels (Kawamura and Yasui, 2005). In PC1, Ph showed high correlations with F and K^+, which suggested Ph originated from primary sources. In addition, strong correlations between Ph and C_2, C_3, C_4, hC_4, kC_3, Pyr and ωC_2 indicated Ph originated from secondary sources. Therefore, PC1 source (anthropogenic activities followed by photochemical aging) was reasonable for Ph.

C_9 is a photooxidation product of biogenic unsaturated fatty acids (Kawamura and Gagosian, 1987). Unsaturated fatty acids can be emitted from sea surface microlayers (Kunwar and Kawamura, 2014). Moreover, a high correlation was found between C_9 and Na^+ ($R^2 = 0.71$). Therefore, we think photooxidation of unsaturated fatty acids emitted from the sea surface together with sea salt particles could be the source for

PC4.

Reference:

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.

Kawamura, K. and Gagosian, R. B.: Implications of a-oxocarboxylic acids in the remote marine atmosphere for photooxidation of unsaturated fatty acids, Nature, 325, 330–332, 1987.

Kunwar, B. and Kawamura, K.: Seasonal distribution and sources of low molecular weight dicarboxylic acids, w-oxocarboxylic acids, pyruvic acid, a-dicarbonyls and fatty acids in ambient aerosols from subtropical Okinawa in the western Pacific Rim, Environ. Chem., 11, 673–689, 2014.

22. "Boundary layer height" should be consistently abbreviated to "BLH".

Response: "Boundary layer height" has been abbreviated to "BLH" throughout the manuscript.

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.

Table 2. DCRCs concentrations reported in this study and literature data from the previous measurements at Mt. Tai in 2006 and other urban sites in the world (unit: ng m^{-3}).

Location	Type	Year	Season	Size	Total dicarboxylic acids	Total oxocarboxylic acids	Total α-dicarbonyls	Major Species
This study (day)	mountain	2014	summer	PM$_{2.5}$	893 ± 479	128 ± 88.9	24.4 ± 20.5	$C_2 > C_4 > C_3 > \omega C_2$
This study (night)	mountain	2014	summer	PM$_{2.5}$	892 ± 402	122 ± 73.6	25.9 ± 21.6	$C_2 > C_4 > C_3 > \omega C_2$
Mt. Tai, China [a]	mountain	2006	summer	TSP	1702 ± 1385	242 ± 210	68.3 ± 64.1	$C_2 > C_4 > C_3 > \omega C_2$
14 Chinese cities [b]	urban	2003	summer	PM$_{2.5}$	892 ± 457	36.7 ± 23.7	5.17 ± 4.14	$C_2 >$ Ph $> C_4 > C_3$
Guangzhou, China [c]	urban	2007	summer	PM$_{2.5}$	523 ± 134	19.5 ± 9.59	5.11 ± 2.13	$C_2 >$ Ph > tPh $> C_3$
Beijing, China [d]	urban	2006	autumn	PM$_{2.5}$	760 ± 369	44.7 ± 26.6	9.05 ± 4.90	$C_2 >$ Ph $> C_4 > C_9$
Chennai, India [e]	urban	2007	summer	PM$_{10}$	502.9 ± 117.9	31.7 ± 11.2	7.1 ± 2.0	$C_2 >$ tPh $> C_3 > C_9$
Raipur, India [f]	urban	2012-2013	winter	PM$_{2.1}$	1072	90.9	30.2	$C_2 > C_4 > C_9 >$ Ph
Tokyo, Japan [g]	urban	1989	summer	TSP	726 ± 636	117 ± 95	46 ± 39	$C_2 > C_4 > C_3 >$ Pyr
Sapporo, Japan [h]	urban	2005	summer	TSP	406	35	9.7	$C_2 > C_3 > C_4 > \omega C_2$
Leipzig, Germany [i]	urban	2003-2005	summer/winter	PM$_{10}$	175 [l]			$C_2 > C_3 > C_5 > hC_4$
Zurich, Switzerland [j]	urban	2002	summer	TSP	66.9 [l]			$C_2 > C_3 > hC_4 > C_4$
Houston, USA [k]	urban	2000	summer	PM$_{2.5}$	67.7 [l]			$C_4 > C_3 > C_9 > C_5$

[a] Kawamura et al. (2013).

[b] Ho et al. (2007).

[c] Ho et al. (2011).

[d] Ho et al. (2010).

[e] Pavuluri et al. (2010a).

[f] Deshmukh et al. (2016).

[g] Kawamura and Yasui (2005).

[h] Aggarwal and Kawamura (2008).

[i] van Pinxteren et al. (2014).

[j] Fisseha et al. (2006).

[k] Yue and Fraser (2004).

[l] did not include all dicarboxylic acid species.

Table 3. PCA analysis results for day DCRCs in PM$_{2.5}$ aerosols collected at Mt. Tai in 2014.

Compounds	PC1	PC2	PC3	PC4	PC5
C_2	**0.854**	0.382	0.203		
C_3	**0.832**	0.277			
C_4	**0.751**	0.353	0.407		
C_5	**0.764**	0.267	0.437		
C_6	**0.697**	0.222	0.322		
C_9		-0.256	0.294	**0.756**	0.389
iC_5	**0.762**		0.523		
M			**0.885**		
F	**0.630**	0.288	**0.635**		
hC_4	**0.794**	0.205			
Ph	**0.693**		0.431		0.313
tPh					**0.904**
kC_3	**0.716**		0.285		-0.202
Pyr	**0.823**	0.353	0.218		
ωC_2	**0.854**	0.406			
ωC_4	**0.881**				
Gly	**0.834**	0.396	0.248		
MGly	**0.687**	0.540			
OC	**0.787**			0.559	
EC	0.411	0.226	**0.632**		-0.337

Na^+	0.241	0.314		**0.862**	
NH_4^+	0.315	**0.938**			
K^+	**0.875**	0.289		0.293	
NO_3^-	0.355	**0.814**	0.302		
SO_4^{2-}	0.279	**0.895**			
Mean trajectory length	**-0.629**	**-0.627**	-0.255		
Solar flux along trajectory	-0.401	0.380			
Mixing depth along trajectory	-0.507	0.393	-0.302		
Variance (%)	64%	9%	7%	6%	4%

Extraction method: Principal Component Analysis (PCA).

Rotation method: varimax with Kaiser normalization.

Table 4. PCA analysis results for night DCRCs in $PM_{2.5}$ aerosols collected at Mt. Tai in 2014.

Compounds	PC1	PC2	PC3	PC4
C_2	**0.674**	0.504	0.464	
C_3	0.341	**0.728**	0.436	
C_4	0.356	**0.678**	0.506	
C_5	0.578	**0.699**	0.285	
C_6	**0.661**	0.400		0.516
C_9		0.531		**0.726**
iC_5	0.407	**0.657**		0.585
M		**0.870**		0.239
F	0.538	**0.642**	0.334	
hC_4	**0.735**		0.364	
Ph	**0.610**	0.478	0.305	0.467
tPh				**0.953**

kC_3	0.514	**0.779**		
Pyr	**0.834**	0.293	0.356	
ωC_2	**0.823**	0.312	0.435	
ωC_4	**0.893**	0.261		0.283
Gly	**0.819**	0.352	0.378	
MGly	0.568		**0.671**	
OC	**0.674**	0.223		**0.660**
EC		**0.770**		
Na^+	0.374			**0.865**
NH_4^+	0.273		**0.921**	
K^+	**0.894**	0.248		
NO_3^-	0.540	-0.206	**0.684**	
SO_4^{2-}		0.365	**0.887**	
Mean trajectory length	-0.564	-0.408	-0.531	
Solar flux along trajectory				
Mixing depth along trajectory	-0.522	-0.427	0.293	
Variance (%)	56%	14%	13%	6%

Extraction method: Principal Component Analysis (PCA).

Rotation method: varimax with Kaiser normalization.

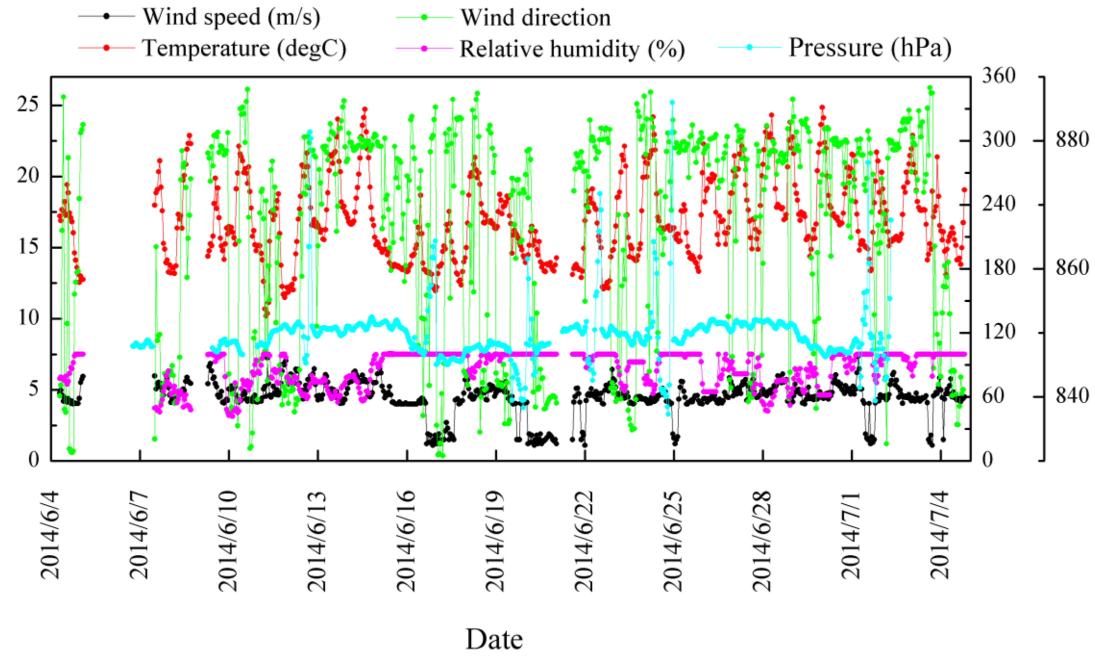

Fig. 1. Summary of meteorological data during the sampling period at Mt. Tai. The left y axis is for wind speed and temperature, whereas the right y1 axis (0-360) shows wind direction (degree) and relative humidity, right y2 axis (830-890) shows pressure.

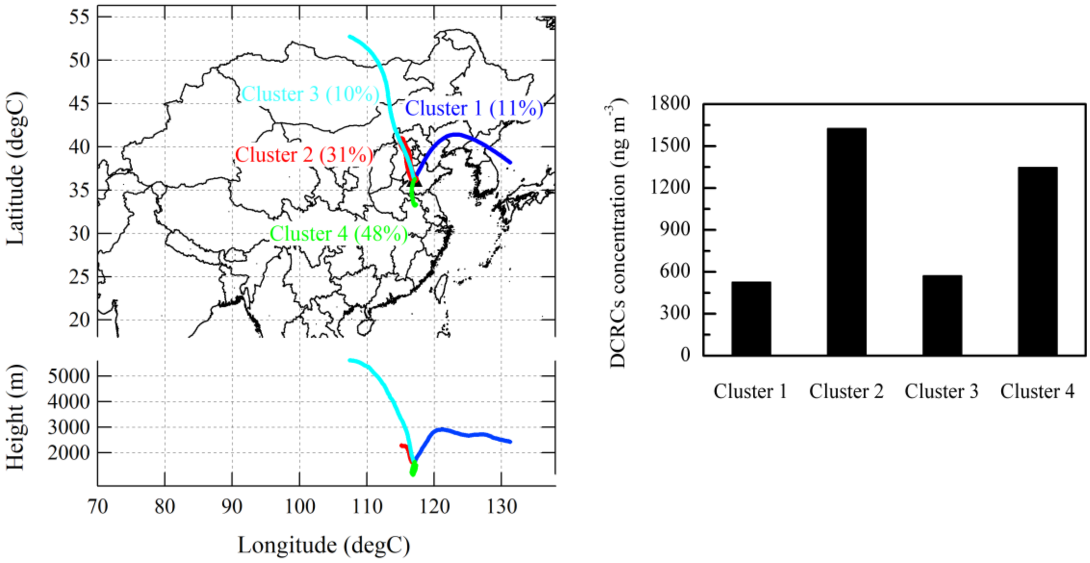

Fig. 2. Three-day backward trajectories for Mt. Tai during the study period (since cluster 2 was covered by cluster 3, the width of the cluster 2 has been increased).

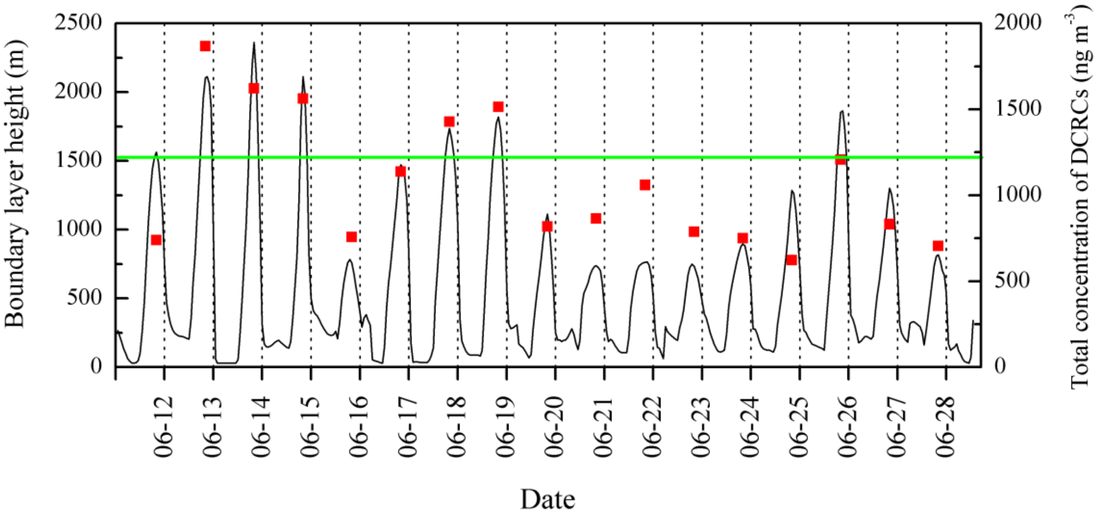

Fig. 3. Boundary layer height at Mt. Tai area during selected sampling period modeled with WRF model (red square: DCRCs concentration; green line: height of sampling site).

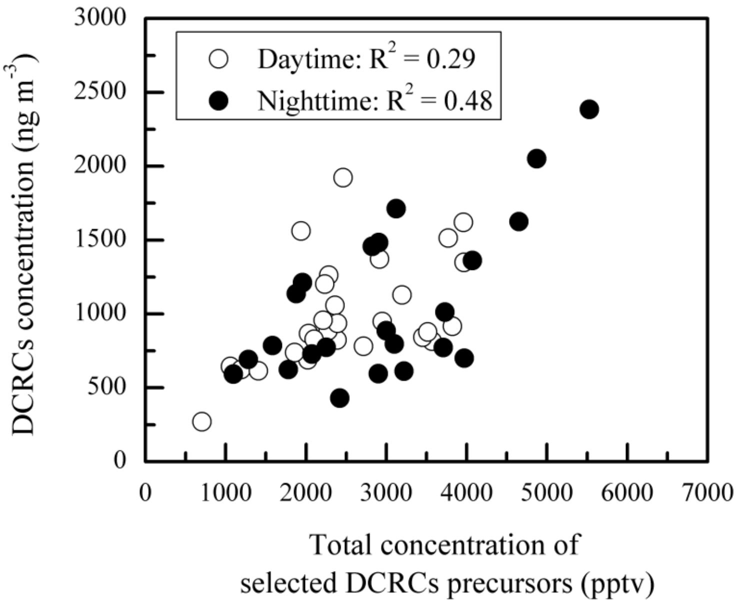

Fig. 4. Scatter plot of the day and night concentration of DCRCs and selected DCRCs precursors. Here, the total concentration of selected DCRCs precursors is the summed concentration of ethyne, ethene, isoprene, α-pinene, β-pinene, toluene, m/p-xylene and o-xylene.

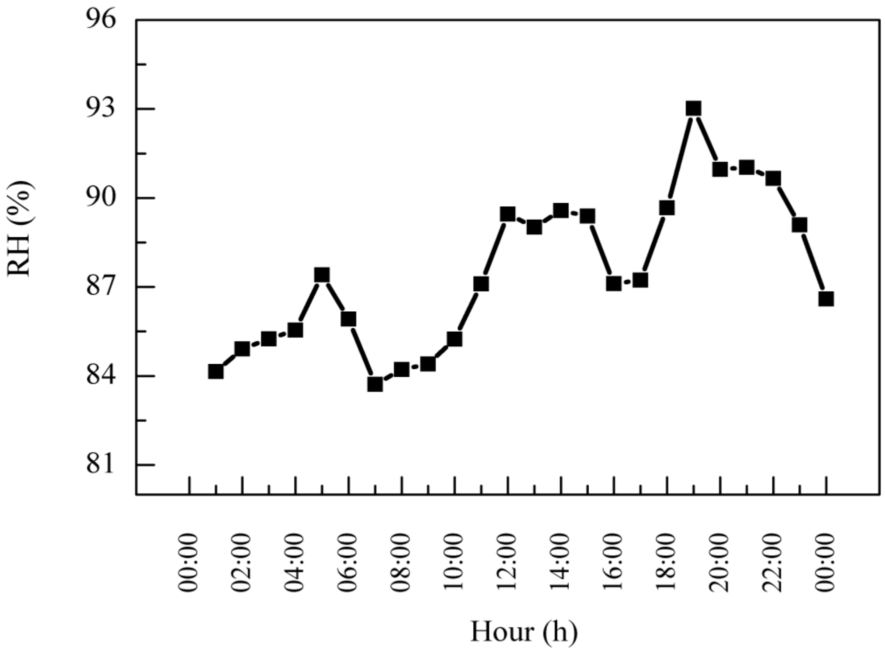

Fig. 5. Average diurnal variation of RH during the sampling period at the top of Mt. Tai.

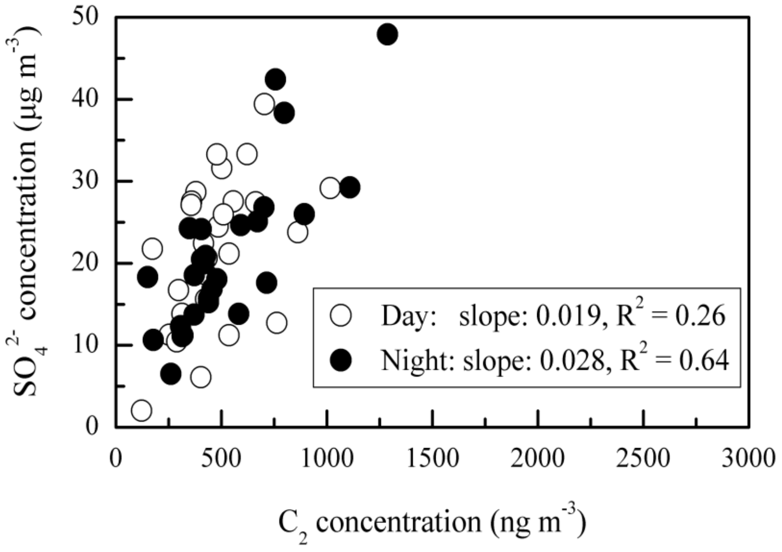

Fig. 6. Day and night scatter plot between C_2 and SO_4^{2-}.

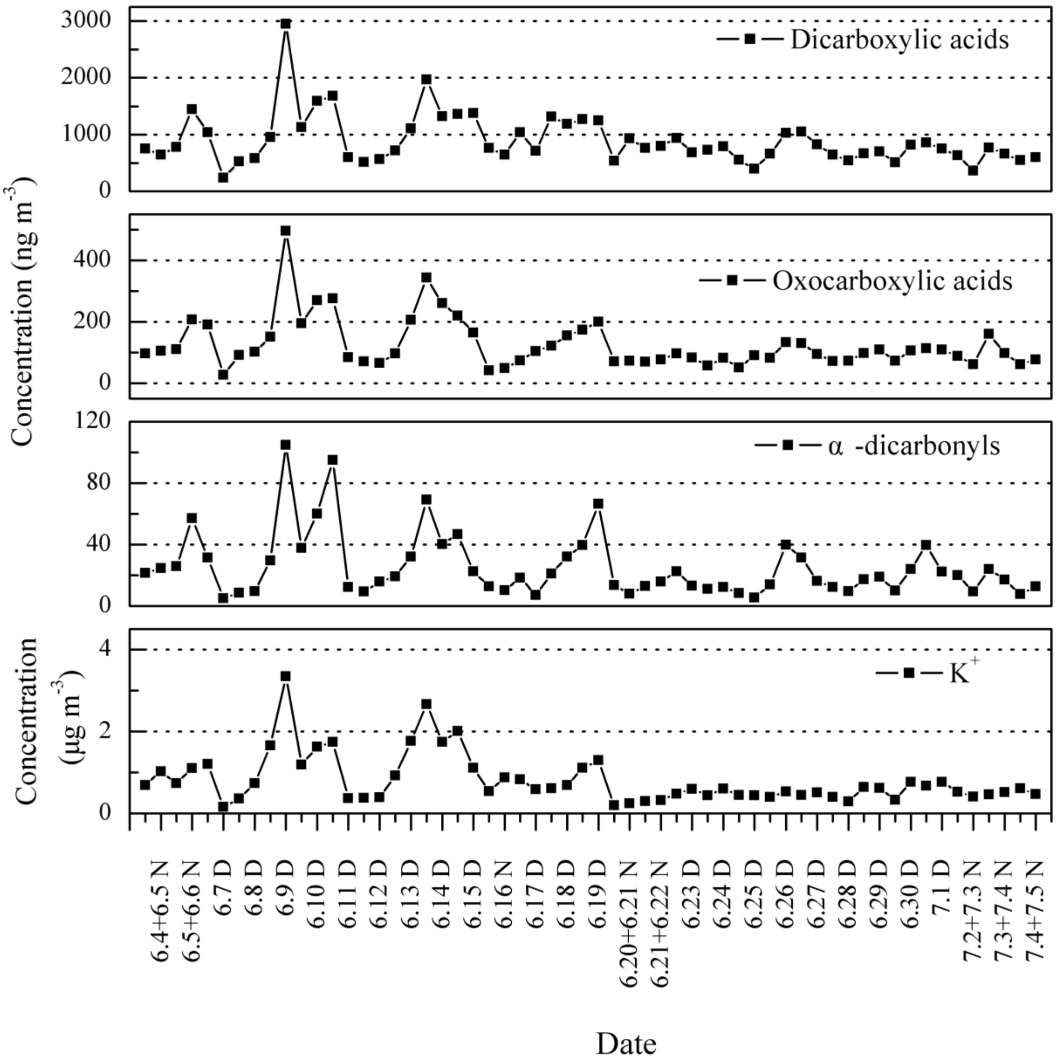

Fig. 7. Temporal variation of DCRCs and K^+ in $PM_{2.5}$ aerosols collected at Mt. Tai during the day (D) and night (N) in 2014.

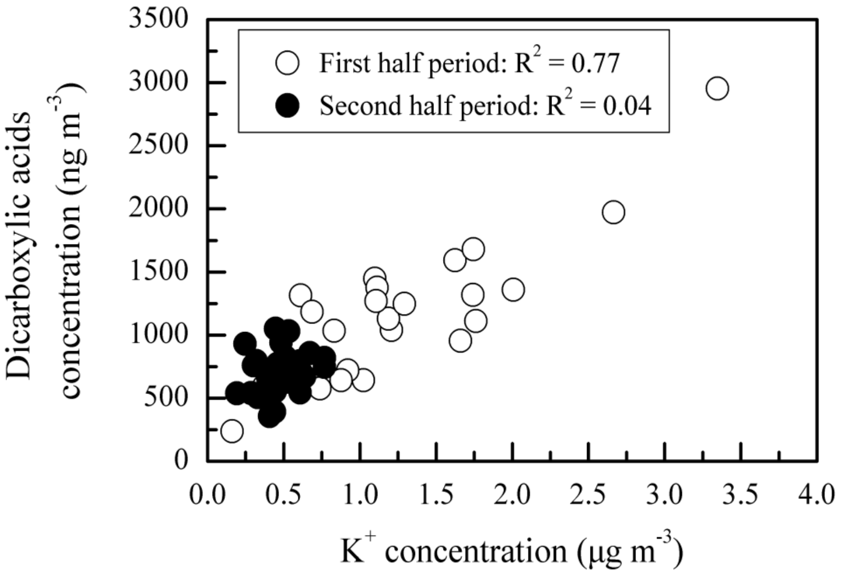

Fig. 8. Scatter plot of concentration between K^+ and dicarboxylic acids during the first and second half of the campaign.

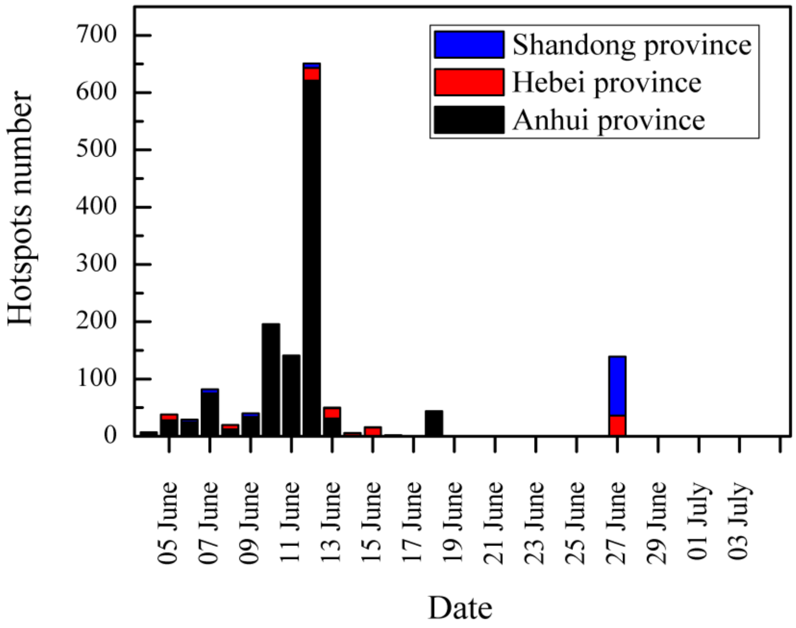

Fig. 9. Straw burning hotspots number in air masses passed over key areas during the sampling period reported by weather satellite of the Ministry of Environment Protection of the People's Republic of China.

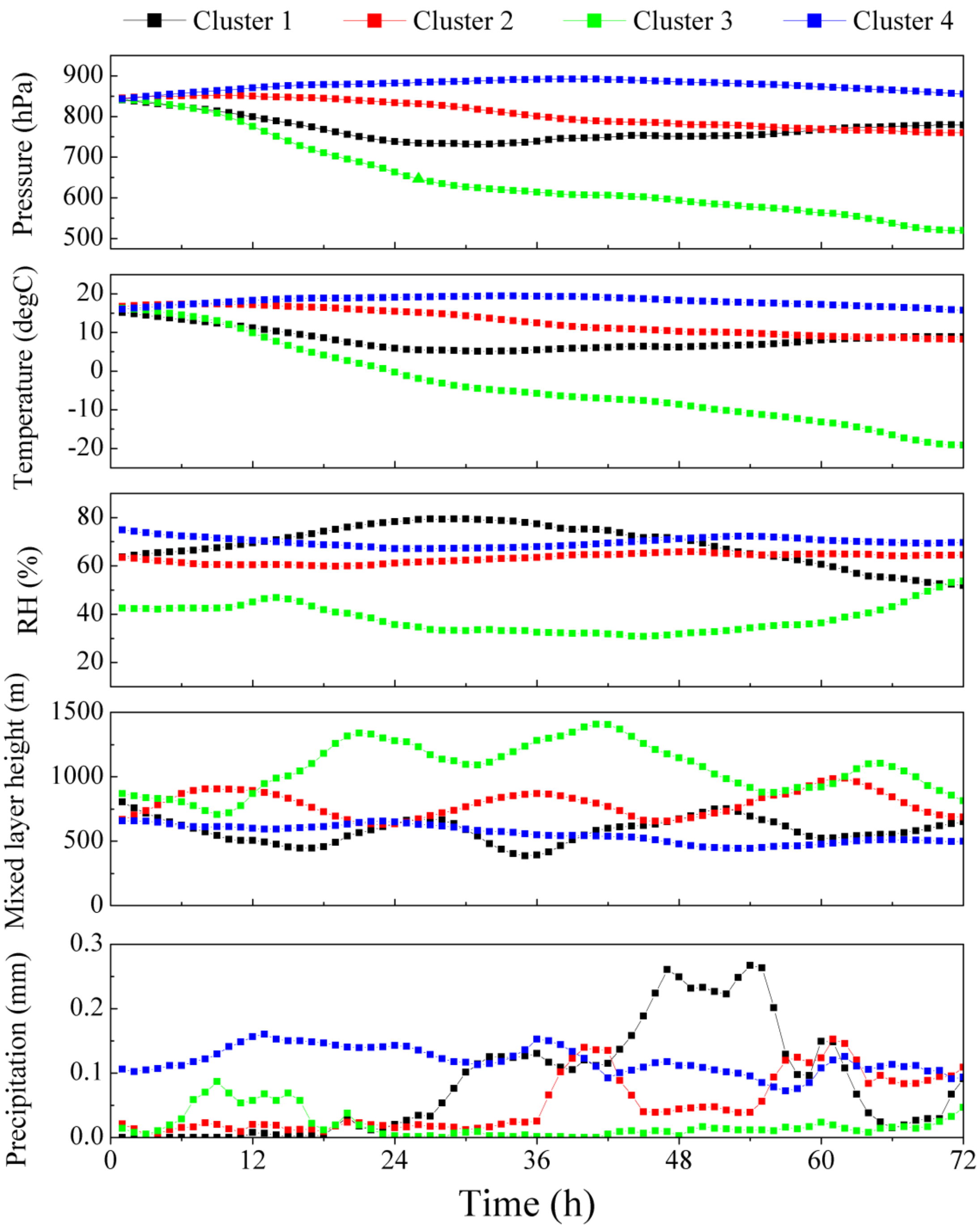

Fig. S2. Meteorological data in the different clusters during the sampling period at Mt. Tai.