Dear Editor,

The authors would like to thank you for the good suggestions obtained in the review and for giving us the chance to further improve our manuscript! We have carefully considered all of your comments and revised the manuscript accordingly. Below, we provide the point-to-point response to your comments, with changes made in the manuscript highlighted in red.

Sincerely

Lingxiao Yang

Ph.D., Professor

Environment Research Institute

Shandong University

Jinan 250100

P. R. China

Response to Editor comments on the author's response:

Thank you for your efforts in responding to the reviewer's comments. The responses took care of some of the reviewer's original comments. However, there are a number of places where the responses were not adequate, as described below. The still requires significant revision before it will be acceptable for publication.

Response: We appreciate the editor for the comments and suggestions. We have revised the manuscript accordingly and here address the comments. For clarity, the editor's comments are listed below in black italics, while our responses and changes in the manuscript are shown in blue and red, respectively.

1. Pages 2-3. The author's response to the comment the lack of day/night differences might be due to the fact that transport to the site takes several days is not really

adequate. The authors need to admit the part of the reason for the lack of day/night difference is due to multi-day transport.

Response: We have admitted and added more discussion about multi-day transport being part of the reason for the lack of day/night difference in the revised manuscript as follows:

Furthermore, the predicted BLH (Fig. 3) suggests that the sampling site was mostly above the BLH during the sampling period, thus the impact of uplifted air on the 12h filter measurements should be minor. Moreover, it is noted that the summit of Mt. Tai is about a few hundred meters above other summits in the surrounding region (Fig. 4). Therefore, the airflow at Mt. Tai should be mainly influenced by the synoptic flow rather than drainage flows. Such an isolated mountain peak is often characterized by wind flows around the peak and small amounts of lifting over it. Nevertheless, under light wind conditions, sunlit mountain slopes may be a favored location for thermals lifting up air from lower levels. However, due to the predominant northwesterly winds, this might have only a minor effect on the performed measurements. No day-night variations of the DCRCs were observed, indicating similar air masses throughout the day and night measurement periods. Due to the fact that air masses arriving at Mt. Tai are transported over several days, multi-day transport has to be considered as part of the reason for the similar concentrations of the field samples taken during the day and night.

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

2. Page 3, Figure S1 doesn't add anything to the discussion, you could transmit the same information by noting the times of sunrise and sunset.

Response: We have deleted Fig. S1 and included the information about the time of sunrise and sunset in the revised manuscript as follows:

The time of sunrise and sunset in June at Mt. Tai was around 06:00 and 18:00, respectively. Therefore, 06:00-18:00 and 18:00-06:00 local time have been selected as the sampling times for day and night, respectively.

(Page 5, Line 11-14)

3. Page 3. What was different about the studies that did show day/night differences?

Probably the nearby presence of the sources?

Response: Pavuluri et al. (2010), Miyazaki et al. (2009) and Fu et al. (2008) have performed day and night sampling between 06:00-18:00 and 18:00-06:00 local time and showed different diurnal variations.

The reasons for the different diurnal variations in the three papers were as following:

Pavuluri et al. (2010) reported that due to the sea breeze effect, most of DCRCs presented much higher concentrations during the day. The sea breeze causes onshore flow of marine air masses in daytime, which are enriched with relatively fresh marine aerosols containing unsaturated fatty acids. They should produce diacids ($\geq C_4$) by photochemical oxidation.

Miyazaki et al. (2009) suggested that due to aqueous-phase oxidation and biomass burning, most of DCRCs had higher concentrations in the night samples.

Fu et al. (2008) reported that most of the organic compound classes showed higher concentrations in nighttime samples when organic aerosols can be transported over long distances, passing different source regions to the summit of Mt. Tai above the planetary boundary layer (PBL).

However, in this study and as noted above, the predicted BLH (Fig. 3) suggests that the sampling site was mostly above the BLH during the sampling period, thus the impact of uplifted air on the 12h filter measurements should be minor. Moreover, it is noted that the summit of Mt. Tai is about a few hundred meters above other summits in the surrounding region (Fig. 4). Therefore, the airflow at Mt. Tai should be mainly influenced by the synoptic flow rather than drainage flows. Such an isolated mountain peak is often characterized by wind flows around the peak and small amounts of lifting over it. Nevertheless, under light wind conditions, sunlit mountain slopes may be a favored location for thermals lifting up air from lower levels. However, due to the predominant northwesterly winds, this might have only a minor effect on the performed measurements. No day-night variations of the DCRCs were observed, indicating similar air masses throughout the day and night measurement periods. Due to the fact that air masses arriving at Mt. Tai are transported over several days,

multi-day transport has to be considered as important reason for the similar concentrations of the field samples taken during the day and night.

References:

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.

4. Page 5. When you say "pressure, temperature and RH didn't change much in clusters 2 and 4, are you saying between clusters 2 and 4, or in each cluster 2, and 4, over the timescale of the back trajectory?

Response: We have changed the sentence in the revised manuscript as follows:

We can see that the pressure, temperature and RH didn't change much in each of cluster 2 and cluster 4 over the timescale of the mean trajectories.

(Page 15, Line 6-7)

We also changed Fig. S1 caption as follows:

Fig. S1. Mean meteorological parameters along the mean trajectories in clusters 1 to 4.

5. Page 6. Yes, the reviewer was mistaken about the average RH.

Response: We corrected it as suggested, thanks a lot.

6. Page 12. To use the ratio of PM2.5 to TSP to scale DCRCs and Levoglucosan, the way the authors have done requires the assumption that there is no size-dependent composition differences, so this needs to be stated as an assumption. Your response did not answer the reviewer's question about measurements from other locations.

Response: We have added the statement of assumption as follows:

Deng et al. (2011) also showed that most of the water-soluble ions presented similar concentrations in $PM_{2.5}$ and TSP, and the ratios of their concentrations in $PM_{2.5}$ and TSP were more than 0.9. Therefore, we assumed there were small contributions of DCRCs from coarse mode particles.

(Page 10, Line 21-24)

We have added a reference from other locations in the revised manuscript as follows:

The low impact of particle size on particle composition has been reported at Mt. Gongga in China (Yang et al., 2009).

(Page 10, Line 24-26)

7. Pages 14 and 15. The reviewers' comment that you have included too many significant figures in many places is correct, and your response is not correct. The appropriate number of significant figures should be based on the uncertainties of your measurements, which is a combination of the propagated errors, and detect limit. So, for example, your stated detection limits are between 0.05 and 0.1 ng/m3, and you do not specify what your propagated errors are, but let's assume they are ±10%, your uncertainty then would be ± the sum of 10% + the detection limit. So, in that case 3 significant figures are not justified. For numbers below 1 ng/m3, you can't justify more that 1 significant figure, since your detection limit is 0.1 ng/m3. Likewise, numbers in the 0.01 place are not significant, so those numbers in Tables 1 and 2 should be rounded to the nearest 0.1 place.

Response: We have corrected the significant figures in Table 1 and 2, which were rounded to 0.1 place in the revised Table 1 and 2.

8. Page 15. The correct terms are "VOC sampling" and "VOC samples", using two plural terms (e.g. VOCs samples) is not correct, please those changes.

Response: We have corrected the two plural terms throughout the manuscript, and changed "VOCs samples" to "VOC samples".

9. Pages 19 and 20. You did not describe the overall uncertainties in the measurements as requested. This needs to be done and then reflected in the reported

data, i.e. significant figures.

Response: We have added description about overall uncertainties in the revised manuscript as follows:

Overall uncertainties for DCRC species were about 15% (see Boreddy et al., 2017 for details).

(Page 7, Line 7-8)

10. Page 28. Point (5). Your answer does not really answer the question. The answer may be that transport to the site takes place over several day/night cycles.

Response: We have admitted and added more discussion about multi-day transport being part of the reason for the lack of day/night difference in the revised manuscript as follows:

Furthermore, the predicted BLH (Fig. 3) suggests that the sampling site was mostly above the BLH during the sampling period, thus the impact of uplifted air on the 12h filter measurements should be minor. Moreover, it is noted that the summit of Mt. Tai is about a few hundred meters above other summits in the surrounding region (Fig. 4). Therefore, the airflow at Mt. Tai should be mainly influenced by the synoptic flow rather than drainage flows. Such an isolated mountain peak is often characterized by wind flows around the peak and small amounts of lifting over it. Nevertheless, under light wind conditions, sunlit mountain slopes may be a favored location for thermals lifting up air from lower levels. However, due to the predominant northwesterly winds, this might have only a minor effect on the performed measurements. No day-night variations of the DCRCs were observed, indicating similar air masses throughout the day and night measurement periods. Due to the fact that air masses arriving at Mt. Tai are transported over several days, multi-day transport has to be considered as part of the reason for the similar concentrations of the field samples taken during the day and night.

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

11. Page 29 and 30. Removing a data point doesn't any more valid, and in fact could be interpreted as deceptive and misleading, and therefore, highly inappropriate.

Response: The coeditor is right and, therefore, we have corrected this issue and have

added the point at ~1800 ng m^{-3} C_2 and 35 μg m^{-3} SO_4^{2-} in Fig. 7. Moreover, we have changed the corresponding data in the revised manuscript.

The correlation coefficient was 0.28 for all daytime C_2 and SO_4^{2-} concentrations. If we delete the point at ~1800 ng m^{-3} C_2 and 35 μg m^{-3} SO_4^{2-}, the correlation coefficient becomes 0.26. Therefore, the influence of this point was low.

12. Page 30, bottom. Do you have any iron measurements to back up your supposition about ironoxalate photolysis?

Response: We haven't performed iron measurements during the sampling period. So unfortunately, we can't back up the supposition about iron-oxalate photolysis. However, as a supplementary solution, we searched for measurements at Mt. Tai in the literature reporting TMI concentrations. Based on the literature, we have extended the related descriptions in the revised manuscript as follows:

Deng et al. (2011) and Shen et al. (2012) reported that Mt. Tai aerosol particles and cloud droplets include a substantial amount of transition metal ions, such as iron. Deng et al. (2011) reported that iron concentration was 0.71 μg m^{-3} in $PM_{2.5}$ and 1.69 μg m^{-3} in TSP during summer 2006. Moreover, Shen et al. (2012) reported that the average bulk cloud water concentration of iron was 44 μg L^{-1} and 416 μg L^{-1} during summer 2007 and 2008, respectively. Thus, iron-oxalate complex formation and photolysis might be possible chemical pathways occurring in Mt. Tai aerosols.

(Page 14, Line 11-18)

13. Page 34. Your response to the reviewer's concern about significant figures is not acceptable. Your significant figures need to be based on measurement or estimate uncertainties. For example, 387 ng/m3 should be 390, due to both kinds of uncertainties.

Response: We have corrected the significant figures in the revised manuscript as follows:

Using the ratio of $PM_{2.5}$/TSP ($PM_{2.5}$/TSP = 0.91) and DCRC concentrations in TSP at Mt. Tai in June 2006 (Kawamura et al., 2013), we have estimated the corresponding DCRC concentrations in $PM_{2.5}$ at Mt. Tai in June 2006 (1550, 220, 62 ng m^{-3} for dicarboxylic acids, oxocarboxylic acids and α-dicarbonyls, respectively).

(Page 10, Line 26-Page 11, Line 1)

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 390 ng m^{-3}. The result was more than five times higher than that in 2014 (levoglucosan: 70 ng m^{-3}) (Zhu et al., 2017), which suggests that biomass burning may have decreased from 2006 to 2014, or Mt. Tai was less influenced by emissions from lower altitudes during summer 2014.

(Page 11, Line 5-11)

14. Page 35. When you say "more than three times higher" Do you mean three times higher at Mt Tai?

Response: Thanks for the carefulness and your understanding is correct, we have corrected the expression as shown following:

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 at Mt. Tai.

(Page 11, Line 14-17)

15. Page 38 Point #3 at the bottom the reviewer wants to know what the values in the tables are. This should be in the title of the table. Also, you should note that the term "factor loading" means the correlation coefficient (r) between the variable (e.g. Zj) and the principal component (PC#).

Response: According to the comment, we have changed the title of Table 3 and Table 4 as follows:

Table 3. PCA factor loadings for daytime DCRCs, OC, EC and inorganic ions as well as mean trajectory length, solar flux along trajectory and mixing depth along trajectory.

(Page 32)

Table 4. PCA factor loadings for nighttime DCRCs, OC, EC and inorganic ions as well as mean trajectory length, solar flux along trajectory and mixing depth along trajectory.

(Page 33)

Furthermore, factor loading means the correlation coefficient between the variable and the principal component (PC). We have added descriptions about factor loading in the revised manuscript as follows:

Factor loading means the correlation coefficient between the variable and the PC, which reveals how much a variable contributes to the corresponding PC and how much a variable differs from others.

(Page 8, Line 9-11)

16. Page 39. The discussion of PCA results needs to be changed so that the PC numbers are connected to the names they have been given in the text. So Tables 3 and 4 the PC numbers have the names associated with them, and the numbers are given (e.g. PC2) after the name is given in the text.

Response: We have added the names of source types in Tables 3 and 4 as follows:

Table 3. PCA factor loadings for daytime DCRCs, OC, EC and inorganic ions as well as mean trajectory length, solar flux along trajectory and mixing depth along trajectory.

Compounds	PC1[a]	PC2[b]	PC3[c]	PC4[d]	PC5[e]
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.

[a] anthropogenic activities followed by photochemical aging

[b] secondary sources

[c] fuel combustion

[d] photooxidation of unsaturated fatty acids emitted from the sea surface together with sea salt

[e] waste burning

Table 4. PCA factor loadings for nighttime DCRCs, OC, EC and inorganic ions as well as mean trajectory length, solar flux along trajectory and mixing depth along trajectory.

Compounds	PC1[a]	PC2[b]	PC3[c]	PC4[d]

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.

[a] anthropogenic activities followed by photochemical aging

[b] fuel combustion and photochemical reaction

[c] secondary processing

[d] a mixed aerosol source related to waste burning and photooxidation of unsaturated fatty acids emitted from the sea surface together with sea salt

17. Page 43. Aren't "coefficients of determination" and "factor loadings" the same thing?

Response: "coefficients of determination" is not the same as "factor loadings". Factor loading means the correlation coefficient between the variable and the PC, which reveals how much a variable contributes to the corresponding PC and how much a variable differs from others.

Coefficient of determination is R^2, for example from linear regression. We have reported coefficients of determination in the text except in the Figures as follows:

As shown in Fig. 5, DCRC concentrations exhibited weak and moderate correlations with total the concentration of selected DCRC precursors during the day (R^2 = 0.29) and night (R^2 = 0.48), respectively, where selected DCRC 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 13, Line 13-18)

As shown in Fig. 7, C_2 and SO_4^{2-} exhibited a higher correlation during the night (R^2 = 0.64) than that during the day (R^2 = 0.28), and the linear regression slope during the night (0.028) was also higher than that during the day (0.016).

(Page 14, Line 3-6)

Dicarboxylic acids and K^+ exhibited a strong correlation during the first half of the measurement (R^2 = 0.77), while during the second half, dicarboxylic acids and K^+ exhibited no correlation (R^2 = 0.04) (Fig. 9).

(Page 15, Line 12-14)

18. Page 52, Figure 3. Is the boundary layer height above ground, or above sea level? If it is above ground, what site is the reference?

Response: In Fig. 3, the boundary layer height is above sea level, and the reference is sea level. We have added description in the revised manuscript as follows:

The reference height of the calculated BLH was sea level.

(Page 6, Line 17-18)

Response to Editor comments on the corrected manuscript:

19. Page 7, Line 5. Please give the overall uncertainties here.

Response: We have added a description about overall uncertainties in the revised manuscript as follows:

Overall uncertainties for DCRC species were about 15% (see Boreddy et al., 2017 for details).

(Page 7, Line 7-8)

20. Page 8, Line 8. Here you should explain what "factor loading" is.

Response: Factor loading means the correlation coefficient between the variable and the principal component (PC). We have explained "factor loading" in the revised manuscript as follows:

Factor loading means the correlation coefficient between the variable and the PC, which reveals how much a variable contributes to the corresponding PC and how much a variable differs from others.

(Page 8, Line 9-11)

21. Page 10, Line 20. This assumes aerosol composition is not size-dependent, please note that and discuss how reasonable that assumption is.

Response: We have added the statement of assumption as shown following:

Deng et al. (2011) also showed that most of the water-soluble ions presented similar concentrations in $PM_{2.5}$ and TSP, and the ratios of their concentrations in $PM_{2.5}$ and TSP were more than 0.9. Therefore, we assumed there were small contributions of DCRCs from coarse mode particles.

(Page 10, Line 21-24)

22. Page 12, Lines 10-14. Mountain top sites are subject to "drainage flow" due to cooling of the ground surface and subsidence of the cooler air, that serves to pull air from above to the surface from above. Please consider what this might mean to your observations.

Response: We have added a related discussion in the revised manuscript as follows:

During the day when the BLHs can be above the sampling site height, more polluted air can be transported from the lower (ground) levels to Mt. Tai top, while during the night, cooling of the ground surface and subsidence of cool air may pull down clean air masses from the free troposphere to the top of Mt. Tai (Fu et al., 2014). However, clear diurnal variations were not found in the DCRC concentrations. Furthermore, the predicted BLH (Fig. 3) suggests that the sampling site was mostly above the BLH during the sampling period, and thus the impact of uplifted air on the 12 h filter measurements should be minor. Moreover, it is noted that the summit of Mt. Tai is about a few hundred meters above other summits in the surrounding region (Fig. 4). Therefore, the airflow at Mt. Tai should be mainly influenced by the synoptic flow rather than drainage flows. Such an isolated mountain peak is often characterized by wind flows around the peak and small amounts of lifting over it. Nevertheless, under light wind conditions, sunlit mountain slopes may be a favored location for thermals lifting up air from lower levels. However, due to the predominant northwesterly winds, this might have only a minor effect on the performed measurements. No day-night variations of the DCRCs were observed, indicating similar air masses throughout the day and night measurement periods.

(Page 12, Line 22-Page 13, Line 12)

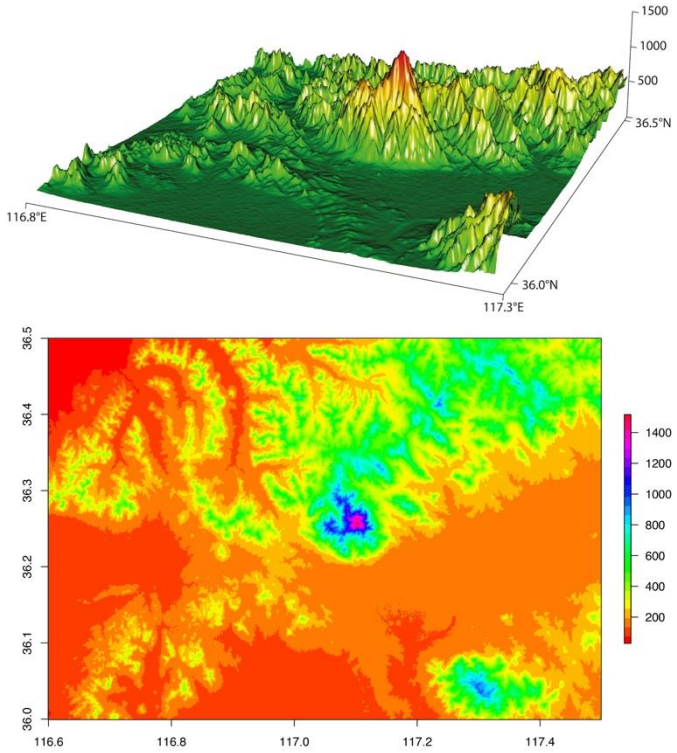

Fig. 4. Topographic map of Mt. Tai and the surrounding region. In the top and bottom panels the altitude is shown by the z-axis and by the color-map, respectively, both with units of meters. The digital SRTM (NASA's Shuttle Radar Topography Mission) elevation data are provided by the CIAT-CSI SRTM website (http://srtm.csi.cgiar.org).

23. Page 17, line 2. Don't you mean that the "contribution of this source to the variance" was 13%?

Response: We have changed the sentence in the revised manuscript as follows:

Moreover, the contribution of this source to the variance was higher during the night (13%) than that during the day (9%) suggesting that secondary processing was more important during the night.

(Page 18, Line 2-5)