We appreciate the reviewers for their constructive comments and suggestions. The manuscript has been revised accordingly. Our point-by-point responses to the comments are presented below. The comments are in <u>black</u> and responses in <u>blue</u>.

### **Response to reviewer#1**

In this study, a filed study was performed during June 2015 at a site in the central of Tibet Plateau using a HR-AMS and a MAAP with the target to characterize the chemical composition, sources, and transport mechanism of polluted air mass in this pristine area. The aerosol in the free troposphere is very important, but hard to be studied. The data set provided by this work is thus valuable. The authors confirmed a general low PM concentration with the highly oxidized organic aerosol as the dominated composition. The data set provided by this work is thus valuable. The manuscript is overall well written and documented. The topic fits well in the scope of ACP. I recommend this manuscript can be published after some revisions.

### Thank you very much for your positive comments.

1) Page 7, Line 233-237: this classification need to be re-considered. Firstly, the standards of these classifications are different that the pre-monsoon and monsoon periods are distinguished mainly by the meteorological parameters, but P1 and P2 is based on the differences of aerosol composition concentrations. Secondly, the differences of aerosol chemical compositions between P1 and P2 are small. Are these differences statistically significant?

Thanks for the suggestion. Sorry that we didn't describe the classification very clear, in fact the classification for distinguishing different periods is basically consistent in our study for both pre-monsoon and monsoon (P3), and P1 and P2. The classification is mainly based on the pollution levels (PM<sub>1</sub> mass loadings) and meteorological conditions. The differences between pre-monsoon and monsoon were obvious. For P1 and P2 periods, the differences were actually also significant. The P2 period had higher contributions from west and north air masses than those during P1, and it in particular included a relatively high pollution episode, thus with an average higher mass concentration than that of P1 (2.9 vs 2.1 µg m<sup>-3</sup>). In addition, the aerosol chemical compositions during these two periods were also different. For example, the P1 was characterized by higher sulfate concentration, while P2 was characterized by higher nitrate contribution. Furthermore, LO-OOA accounted for 41% of the total OA mass during P1, whereas it was only 24% during P2. All these results verified the significantly different properties of the PM<sub>1</sub> during P1 and P2. In this regard, we believe our classification of P1 and P2 is reasonable and the comparison between aerosol chemical characteristics of P1 and P2 is also important for evaluating the radiative forcing of aerosols in the future. In the updated manuscript, we have added such information correspondingly.

2) Page 8, Line 265-266, air masses from west was confirmed to be favorable to transport polluted air masses. Is there some information to show where these air masses originally come from? Could it be influenced by the local sources, e.g. some small villages and tourists (Page 4, Line 119-121). Is there any signal to show if polluted air mass from east China can influence the site?

The back trajectory analysis of air masses is only a qualitative method which can be used to show some potential source regions for the aerosols. It cannot provide the exact regions where the air pollutants come from. It is true that the air mass may intercept local emissions during transportation and such influences cannot not be excluded, but qualitatively, we believe such contributions might be small as the organic aerosol (OA) was overall highly oxidized, indicating negligible contributions from fresh primary OA. The air mass from east China should be negligible because the dominated climatic system is westerlies in this

season. In fact, although we only conducted three-day trajectory analysis, the trajectories are almost the same as those from seven-day trajectory analysis.

3) Page 8, the 2nd paragraph: the discussion the formation/sources of aerosol chemical compositions is a bit of an oversimplification. I recommended more discussions on how these chemical compositions are formed or transported.

Agree. We have revised this paragraph and made it less speculative. It should be mentioned that a clear understanding of chemical processes of aerosols influenced by long-range transport are very difficult due to the complexity of the conditions and possible chemical processes likely occurred during transport. In addition, as we did not measure the gas-phase species along with the aerosol measurements, so we mostly focused on the temporal and spatial distributions of the aerosols.

"In order to further elucidate the chemical processes and potential sources of the aerosol species, the relationships of chemical species with wind conditions were analyzed based on bivariate polar plot analysis (Figure 6). During P1, sulfate and ammonium had hotspots from all directions across a wide range of wind speed  $(0 - 10 \text{ m s}^{-1})$ ; while OA was prevailing from southeast and northwest winds; nitrate had hotspots when the wind speed was relatively slow  $(0 - 8 \text{ m s}^{-1})$  and from southeast/east mainly; BC had hotspots from south, west and northwest at high wind speeds  $(4 - 12 \text{ m s}^{-1})$ . During P2, all species except nitrate had hotspots from south/southwest at high wind speeds  $(6 - 14 \text{ m s}^{-1})$ ; Nitrate also had hotspots at a relative low wind speed from southwest. These results suggested that the sources and formations of aerosols during P1 and P2 could be different, albeit the diurnal variations of species between these two periods were similar. During monsoon period, all species had similar distributions of the hotspots, which could be from all directions but relatively weak from southeast. "

4) Fig. 9b: it's interesting that the O/C ratio increased accompanied with the increased of OA mass concentration. But more discussion is needed to interprate the reasons. In general, a higher O/C ratio would correlate to a more aged air masses/aerosols, but should be independent to the mass concentrations.

We add some sentences to further interpret this relationship. In our opinion,  $PM_1$  during long-range transport could experience many chemical and physical processes. We think our results are closely related with the scavenging effects during transportation of the aerosols. Some  $PM_1$ , usually highly oxidized aerosol, is easy to be scavenged by cloud droplets due to its high hygroscopicity, whereas some  $PM_1$  is not easy to be scavenged and can sustain during long-range transport. In this case, the scavenging effect can decrease the aerosol loadings and the left  $PM_1$  are typically less oxidized, and correspondingly, the O/C ratio may positively correlate with the OA mass loadings. We indeed observed the lower concentration and less oxidized OA at the NamCo station during the monsoon period. On the other hand, if there were not significant wet scavenging processes during pre-monsoon period, then the OA concentration could be relatively high, while the O/C ratio could be high as well as it experienced long-time atmospheric oxidation processing.

"We examined the variation of elemental ratios with OA mass concentrations, and found that the O/C increase was accompanied with the increase of OA mass concentration (Figure 9b). This relationship could relate with the different influences of wet scavenging on more oxidized and less oxidized OA. In addition, this result likely suggested the importance of transportation on the oxidized OA during afternoon due to the higher mass concentration occurring frequently on afternoon time."

**Response to reviewer#2** 

In this study, the authors report inorganic salts, BC, and carbonaceous components in submicron aerosol particles at a remote elevated site in the Tibet Plateau in a period including pre-monsoon and monsoon weather conditions. The data in the manuscript are original and in good quality. In particular, the long-term trend and the diurnal variation of the components are very meaningful for understanding the origins and the effects of aerosol particles due to the long-range transport from the outside of the plateau and the local or regional emissions. I suggest the accept of the publication in the journal after a number of minor revisions.

We thank the reviewer for his/her careful review of the manuscript.

### Major comments:

Most of the species measured in this study had a clear diurnal variation, which was closely dependent on the meteorological conditions, i.e. wind direction, wind speed and likely solar radiation. Regarding the distinctive terrain around the observation site, the variation of wind direction and wind speed on clear days is naturally expected and is a local and terrain-dependent phenomenon. As the authors attributed the diurnal variation of the chemical composition of aerosol particles to the diurnal variation of meteorological conditions, they also emphasized the predominance of the long-range transport. This makes me confused that which part of the variations of aerosol particles were caused by long-range transport and which part were directly or indirectly due to the diurnal variation of meteorological factors or local emissions. According to the description of the observation site, there were some anthropogenic sources near the site which might have constant or episodic influence.

Due to the challenges associated with separating the sources of aerosol, we cannot clearly quantify the contributions from local sources and those from long-range transportation for the measured aerosol in our study, but the long-range transportation is believed to play a major role in such remote site. For example, previous studies focusing on mineral dust using CALIPSO lidar measurement have observed the longrange transport process (Huang et al., 2007; Liu et al., 2008). For our data, in general, we observed significantly lower mass loadings of PM<sub>1</sub> during monsoon period than those during pre-monsoon period. As it is expected that due to strong scavenging effect during monsoon, the aerosols are likely dominated by local/regional emissions, this on the other hand, indicates that the relatively high PM<sub>1</sub> concentrations are significantly contributed by long-range transportation. Our discussion is in two-fold, first, we mainly focused on the characteristics of PM<sub>1</sub> chemical components influenced by long-range transport processes. The diurnal variation of chemical species of course can be influenced by the local meteorological conditions, but such PM<sub>1</sub> was likely in a large part originated from the southern and western Asia as illustrated from the back trajectory analysis. The local primary sources indeed had a negligible contribution at least based on the PMF results - unable to directly separate a primary OA factor. Nevertheless, we agree with the reviewer that our title could be a little bit confused, and it cannot cover what we presented in the manuscript. We thus revise the title to "Chemical characteristics of submicron particles at the central Tibet Plateau: insights from aerosol mass spectrometry".

Sub section 3.6 has little contribution to the major conclusions. Simplifying largely the descriptions into a few sentences and moving Figure 13 into supplementary materials are likely a good choice.

Thanks for the suggestion. This part tried to analyze the climate conditions for the transportation of polluted air mass. We here observed events of pollutants into inland of the Tibet Plateau, but the mechanism of this transport is not clear. In order to give a full story from our paper, we think such discussion is useful and adds the values of the paper, thus we prefer to keep this part in the main text.

In addition, adding a brief discussion on the implication of the present results in understanding the air pollution in the Tibetan atmosphere will increase largely the significance of this paper.

We add a brief discussion on the implication of the present results in the updated manuscript.

# **"3.7 Atmospheric implications**

Our results have several potential implications to the atmospheric studies in the TP and Himalayas. Firstly, it is useful for the accurate estimation of the radiative forcing of aerosols in this region and validation of current model simulation results based on our observed chemical composition and mass loadings of fine aerosols. Ji et al. (2015) estimate the radiative forcing from aerosols over the TP and Himalayas at the surface level using a regional climate model (RegCM4.3); for carbonaceous aerosols, there are several literatures that tested the model results, but all other species were referred to data available in the published inventory with a coarse spatial resolution. Secondly, our findings are implicate to the estimation of aerosol deposition on the glacier of this region and evaluation of subsequent impacts on the melting of snow/ice (Yasunari et al., 2010). Thirdly, highly-time resolved aerosol data is very scarce in this remote plateau, thus our data are valuable to validate modeling results regarding the transport of polluted air mass as demonstrated in section 3.6. At last, the transport mechanism of aerosol to the inland of TP is less understood so far. Hindman and Upadhyay (2002) suggested that the vertical lifting due to convection and subsequent horizontal mountain-valley wind could lead to the transport of aerosol from Nepal to Tibet. Dumka et al. (2010) also highlighted the important role of mountain-valley wind in the aerosol transport in the central Himalayas. The dynamic variations of aerosol chemical species measured here, are likely helpful to elucidate the transport mechanism of polluted air mass. Nevertheless, this scientific issue required further detailed investigations in the future."

Minor comments:

1. Both "O/C" and "O:C" are used to describe the ratios of O to C. Unify them please, unless they have different meaning.

All the ratios are unified in the updated manuscript.

2. Lines 24-27: Remove "The average ambient : : :worldwide". The results are dependent on the length of the pre-monsoon period and the monsoon period used for the average. In addition, the results in the abstract, i.e. the relative contribution of each species, were different from those shown in Figure 2 and described in the text in Line 213. The authors are requested to carefully check all figures in the whole manuscript with authors' records.

Thanks for the suggestion. However, we think such descriptions which show the average mass loading and composition are useful. It is indeed relevant with the sampling time and location, but this is true for any kind of filed measurements (you cannot have a field measurement covering all times and environments). The sampling length was one month which should be representative for the atmospheric pollution for our sampling site, so we keep this sentence. We believe as long as we stated clearly the sampling site, time and duration of our measurement, then these average values will be useful to other future measurements. In addition, we have carefully checked the numbers in the manuscript and the abstract, and make them consistent throughout the manuscript. Thank you for pointing out our typos.

3. Line 34: the ratio of O/C 0.48 was not found in the manuscript. From Fig.8, the ratio was between 0.7-0.8. Could this value mean a significant difference of the oxidation state of OA in comparison to the OA with the ratio 0.88?

We made a mistake for this number which should be 0.72. The content in the manuscript are revised accordingly. We performed chi-square test for these two groups of datasets and they presented statistically different behaviors (p < 0.05).

4. Line 62-64: After this description, a brief introduction of the difference of large-scale circulation between pre-monsoon periods and monsoon periods helps readers to easily understand the results of the present study.

Agree. We have added a few sentences to describe the variation of climatic systems during pre-monsoon and monsoon as follows.

"The distinct seasonal variation of aerosol loading is mainly attributed to the change of the dominated climatic systems and then weather conditions. During pre-monsoon, the cold and dry southern Westliers dominated the southern TP and Himalayas, while South Asia Monsoon covers most of South Asia, Himalayas, and the southern TP during summer period."

5. Line 133: Shown in Figure 1 are different from the description here.

### We made a mistake here and this sentence has been removed.

6. Line 212-224 "The average mass concentration : : :high sulfate": These descriptions and comparisons are unnecessary. As mentioned above, the average results of per-monsoon and monsoon periods here are not referentially meaningful due to the dependence on the length of the periods.

We agree that these kinds of comparisons are dependent on the length of the covered periods. However, this is in fact true for any other field measurements. As we pointed out in our manuscript (line 225-227) and our response in comment 2, this kind of comparison is still valuable and other researchers can compare their results to our values fairly as long as the measurement periods are stated clearly. In addition, these values are also used to elucidate the influences of pre-monsoon and monsoon periods on the aerosol properties, so the average values and their comparisons are of course valuable. Scientific findings derived from such values and comparisons will not limit to our certain site and sampling time, but useful to reveal the role of monsoon influences on the aerosol properties in general. So we keep these sentences.

7. Line 229: The description of "suggesting high efficiency of wet scavenging for …" lacks evidence. The authors did not give any data on the wet scavenging. If all pollutants were consistently from same sources and experienced similar history, this statement might be acceptable.

#### Agree. We delete this sentence.

8. Line 230. "The contribution of OA was thus : : :": Do the authors mean wet removal of salts such as sulfate was more efficient than the removal of OA, and this difference resulted in the present result? Is the result the matter of origins, transport and in-air variations?

We totally agree this comment. The scavenging efficiencies of sulfate and ammonium are indeed higher than that of OA as illustrated from the mass ratios between pre-monsoon and monsoon since sulfate and ammonium had higher water uptake abilities. In addition, the source, transport and evolution of aerosol during pre-monsoon and monsoon were indeed different. We add a sentence to describe this uncertainty.

"Apart from the potential scavenging effect, these results could also be influenced by the sources, transport route, and chemical processes during different periods."

9. Line 251-253 "This high : : : for ammonium": (1) explain RIE; (2) evidence for this explanation is necessary.

The settings of RIE for different species during the campaign are added in the section 2.3, and the evidence for variation of RIE for ammonium is also added in the manuscript as follows.

"Default relative ionization efficiencies (RIEs) were used for organics (1.4), nitrate (1.1), and chloride (1.3), while a RIE value of 3.8 was determined for ammonium and 1.1 for sulfate based on the calibrations for pure  $NH_4NO_3$  and  $(NH_4)_2SO_4$ , respectively."

"This excess ammonium determined by the neutralization might be related to the presence of significant amounts of organic anions in aerosol (such as carboxylic acids) or variation of RIE for ammonium which could be higher in the mixed acidic particles."

10. Line 257-273: Observational data, including meteorological data, from activities of this study are necessary to support the discussion in this paragraph. My feeling is that the long-range transport provided the background level of pollutants, and the diurnal variation was driven by the meteorological conditions on the basis of the long-range transported and locally-originating background level of pollutants. Please notice that soot particles in the Tibetan atmosphere could be aged rapidly (Zhang et al. 2001. Atmos. Environ. 35, 5883-5894). In addition, please check carefully the gramma and words.

Agree. The logistic of this paragraph has been adjusted following the comments. The reference which mention the age of soot particles has been cited in this paragraph. The grammar and words are also improved.

"The diurnal cycles of OA, sulfate, nitrate, ammonium, and BC during different periods are shown in Figure 5. All these species unexpectedly present dramatic diurnal variations, especially during P2. OA, sulfate, ammonium, and BC showed a similar pattern with low values during nighttime to early morning and high values during afternoon which suggest their common sources or similar transport pathways (Zhang et al., 2001). We checked the diurnal variation of the origination of air masses and found that there were increased air masses from south during nighttime and from west during afternoon (Figure S6), which could be related to the plateau monsoon during summer (Tang and Reiter, 1984). The enhanced air mass from west during afternoon could favor the transportation of polluted air mass. The enhanced WS during afternoon was also observed with the increase of air temperature (Figure 5). The diurnal variations of chemical species during monsoon period were relatively flat comparing with those during P1 and P2 which may relate with the relatively consistent air mass origination during monsoon. Nitrate presented a significant different diurnal variation with high values during nighttime to early morning and low values during afternoon. These features were highly correlated with that of the RH and air temperature (Figure 5) suggesting the importance of thermodynamically-driven gas/particle partitioning of ammonium nitrate and heterogeneous production of nitrate due to hydrolysis of N<sub>2</sub>O<sub>5</sub>. In addition, during the early morning time (6:00 - 8:00), there was a peak for most species, which was accompanied with the lowest air temperature and the highest RH, and the lower plenary boundary layer (PBL), which could concentrate all the air pollutants (Yanai and Li, 1994). Overall, the diurnal variations of aerosol species at Nam Co may be dominated by the variabilities of both long-range transport air mass and local meteorological conditions."

11. Line 277-284: The discussion lacks evidence. The authors used several "suggesting", but did not show evidence. In addition, the sentence is too long, tedious, and includes "suggesting" repeatedly.

Agree. We rewrite these sentences and removed the "suggested" parts and now it reads as follows.

"During P1, sulfate and ammonium had hotspots from all directions across a wide range of wind speed  $(0 - 10 \text{ m s}^{-1})$ ; while OA was prevailing from southeast and northwest winds; nitrate had hotspots when the wind speed was relatively slow  $(0 - 8 \text{ m s}^{-1})$  and from southeast/east mainly; BC had hotspots from south, west and northwest at high wind speeds  $(4 - 12 \text{ m s}^{-1})$ ."

12. Line 291-306: As mentioned in the major comments, the discussion is too qualitative.

Agree. But unfortunately, we did not measure the gas phase species in our study, thus it is not easy to elucidate quantitatively the chemical processes of sulfate and nitrate. We have carefully written this part and try my best to present some useful insights. If allows, we will elaborate this issue in more details in our future study.

13. Line 316-317 ": : : O:C ratio (0.90 vs 0.98) and : : : ": Is this correct?

We made a mistake and have corrected it as follows.

"The OA was more oxidized during pre-monsoon than monsoon with higher O/C ratio (0.94 vs. 0.72) and lower H/C ratio (1.28 vs. 1.44)"

14. Line 337-340: "deep convection over the Tibet Plateau" usually causes upward mixing. Evidence from the present study for the conclusion of "Thus the : : : layer." (Line 339-340) is necessary.

The deep convection can also mix down the pollutants as illustrated by the enhanced  $O_3$  and sulfur isotope from the ground measurement in the Tibet Plateau during summer (Lin et al., 2016). As mentioned in the manuscript, the mechanism of mixing down of pollutants is a hypothesis and needs further validation. We were puzzled by the afternoon peak of chemical species which are unlikely due to long-range transportation directly from South Asia. The long-range transport of pollutants cannot arrive at the inland of the Tibet Plateau along the surface level since there are many ridges of mountains. The distance of the long-range transport should positively relate with the lift height by convection at the source region. To clarify this, we rewrite this sentence as follows.

"Recently, Gu et al. (2016) examined the aerosol compositions using the global three-dimensional Goddard Earth Observing System chemical transport model (GEOS-Chem) and found elevated concentrations of sulfate, nitrate, ammonium, BC, and organic carbon over the TP. Further, observational and modeling studies have also shown that deep convection over the TP during daytime is one of the important routes for tropospheric and stratospheric exchange of aerosols (Cristofanelli et al., 2009; Cristofanelli et al., 2010; Lin et al., 2016). Thus the enhanced aerosol concentrations during afternoon could be possibly attributed to the mixed downward of aerosol layer at 16 - 18 km altitude during the growth of TP boundary layer. This type of transportation could not be captured by re-analysis of data used in the back-trajectory analysis likely due to the low time and spatial resolution. Nevertheless, this hypothesis needs further validation in the future in this region."

15. Line 365-366: The ratios separately in pre-monsoon and monsoon periods are more referentially meaningful. Average over the whole period should be avoided.

This issue is related with the assumption of PMF algorithms. PMF assumes the profile of a fixed factor is unchanged throughout the study period, thus the O/C ratios of PMF factors are the same for different

periods. While the O/C ratios of total OA can be different as the contributions from different factors vary. We compare the O/C ratios of OA in section 3.4 during pre-monsoon and monsoon periods.

16. Line 396: I cannot find results on PM1 from REAM in the manuscript.

This headline has been revised to "Sensitivity of the transport of pollutants to synoptic process".

17. Line 398-424: The linkage between the simulated results and the observational facts is weak. The point in the last sentence (Line 423-424) is important, but there is no discussion on its confidence.

Thank you very much for your suggestion. In the updated manuscript, we rewrite the sentences of the linkage between the simulated and observational results. A figure of  $PM_1$  species corresponding with the simulated period is added in the Figure S9.

"Although these trends are basically consistent with our AMS results, there were also significant differences (Figure S9). The possible reason was that the weak trough during P2 intensified the wind from west other than south where a lot of biomass burning emission sources locate (Figure 5b). Zhang et al., (2017) suggested that a low-cut system from stratosphere could be an important driver for pollution transport into the TP. In our study, the trough/ridge system seems to be also an important factor affecting the transport of air pollution from south and west, although this effect tends to be weaker in summer than in the other seasons because the tropopause is higher and stratospheric wave activity is weaker in summer."



Figure S9. The mass concentration of (a) AMS results and (b) REAM-simulated reactive aromatics and the averaged wind data.

18. Line 416: What does "this difference" mean here?

This sentence has been revised as presented in response to comment 17.

19. Line 694 Figure 2: wind direction is hard understood.

This figure has been improved as follows.



Figure 2. The combo plot of the data of the Nam Co study including (a) the meteorological conditions (T: air temperature; RH: relative humidity; Precip.: precipitation), (b) the variation of WS (wind speed) colored by WD (wind direction), (c) the temporal variation of mass concentration of PM1 species and the average contribution each species (pie chart), (d) the mass contribution of each PM1 species and the total mass concentration of PM1, and (e) the mass contribution of PMF results (section 3.5). Three periods based on the meteorological conditions were marked.

### **Syntax corrections:**

I found inaccurate words and sentences, and syntax errors. I list some of them here. I am not a native speaker, but feeling the authors need to carefully check words and gramma in the whole manuscript, including figure captions, in the revision.

Thank you very much for your suggestions. The manuscript has been improved and we hope this new version satisfy the reviewer and is easy to read and understand.

1. Line 20: replace "mechanism" with "processes". This study discusses processes, does not mechanism.

#### Corrected.

2. Line 20: remove "pristine": The area was polluted according to the content of the paper.

## Corrected.

3. Line 20: replace "performed" with a suitable word, such as "did" or "carried out".

## Corrected.

4. Line 28: replace "while" with a suitable word. Similarly, in Line 38.

## Corrected.

5. Line 86-88: remove "Therefore", and rephrase this sentence properly.

This sentence has been revised as follows.

"High-time resolution measurement is thus necessary in this region for detecting short-term events and the evolution of pollutants."

6. Line 104-105: remove "However, : : : so far".

Corrected.

7. Line 107: remove "comprehensive".

Corrected.

8. Line 119: "This" is vague.

This sentence has been revised as follows.

"The Nam Co station and its surrounding is a pristine region except for a small county for local people that is about 10 km west to the station."

9. Line 133: replace "is" with "was"; replace "conditions" with "conditioners".

Corrected.

10. Line 134: "total inlet"?

Change to "inlet" and this sentence now read as follows.

"The inlet was maintained by a vacuum pump with a flowrate of 10 L/s.....".

11. Line 139: "this inlet"?

Change to "the inlet".

12. Line 139-140: Check the gramma.

This sentence has been revised as follows.

"The total flowrate of the inlet was maintained at ~16 m/s and a PM<sub>2.5</sub> cyclone was used at the front of the inlet (model URG-2000-30EH, URG Corp., Chapel Hill, NC, USA)."

13. Line 181: replace "decomposition" with "apportionment". Similar in Line 344.

Corrected.

14. Line 245: remove "chemical"; replace "from" with "in"; and replace "between" with "of".

Corrected.

15. Line 249: remove "concentration of".

## Corrected.

16. Line 250: replace ", however, the" with ". The".

### Corrected.

17. Line 250: rephrase "The ratios of : : : and : : : were : : :" into "The ratios of : : : and : : : to : : : were : : :".

### Corrected.

18. Line 279: replace "to" with "on".

# Corrected.

19. Line 296: rephrase "all periods during the : : : period of this study".

This sentence has been improved as follows.

## "Figure 7 showed all periods with high nitrate during the study...".

20. Line 298: remove "were".

#### Corrected.

21. Line 301: remove "While".

Corrected.

22. Line 319-320: rephrase the sentence.

This sentence has been improved as follows.

"Correspondingly, the OA during P2 contained higher contribution of  $C_xH_yO_1^+$  (40.3 vs. 39.1%) and  $C_xH_yO_2^+$  (25.2 vs. 23.6%) ions than those during P1 (Figure 8c)."

23. Line 325: replace "check" with "examined" or a proper word.

Corrected.

24. Line 325-326: "at the function"?

Changed as follows.

"We examined the variation of elemental ratios with OA mass concentrations, and found that the O/C increase was accompanied with the increase of OA mass concentration (Figure 9b)."

25. Line 342: "The" is likely a typo.

Corrected.

26. Line 355: remove "was".

Corrected.

27. There are too many "in this study" or "during this study" in the text.

In the updated manuscript, these phrases have been replaced or deleted.

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