

We are thankful to the two reviewers for their thoughtful comments that help improve the manuscript significantly. Following the reviewers' suggestions, we have revised the manuscript accordingly. Listed below are our point-by-point responses in blue to each reviewer's comments.

Response to Reviewer #1

The manuscript entitled, "Chemical characterization of submicron aerosol and particle growth events at a National Background Site (3295 m a.s.l.) in the Tibetan Plateau" by W. Du et al., presents non-refractory plus black carbon (BC) aerosol chemical composition and particle size distribution data from a remote location on the northeastern region of the Tibetan Plateau. The observations reported here fill a gap of data from this part of the world. The location is very interesting for readers of Atmospheric Chemistry and Physics. The manuscript is generally well-written, yet needs clarification in some areas and additional analysis. Overall, the data were presented well, but much of the interpretation was left to the reader, which makes it difficult to understand the broader picture of the important findings from this study. I recommend publication after addressing these issues.

Thank the reviewer's comments. The manuscript was significantly revised according to the reviewer's suggestions listed below.

Major comments:

The overriding issue is that while the data are from a remote, "background" site, there needs to be some analysis as to where the episodes with high mass concentrations and new particle formation events are coming from. An analysis of the various wind directions and back trajectories would also be helpful in putting these observations in the context of other nearby measurements (in particular, Bird Island at Qinghai Lake and Mt. Waliguan). Throughout the text, there is mention of regional transport being important to observations. From where? I was surprised to find that there are many urban (prefecture) areas within 200 km of the site with populations greater than 500,000 that may be contributing to the background aerosol. This information was not provided in the manuscript. Also, the infrastructure (railroads, agriculture, power plants, etc.) for supporting these people needs to be considered as potential sources.

Thank the reviewer's comments. The back trajectories for the episodes with high aerosol mass loadings are now shown in Fig. S3 in supplementary in the revised manuscript. The new particle formation is formed locally which is initiated by the formation of gas sulfuric acid. In this study, we focus on the particle growth stage rather than new particle formation due to the limitations of SMPS and ACSM measurements. Additional analysis of winds and back trajectories were added in the revised manuscript. In addition, the reasons we didn't compare with the measurements at Bird Island at Qinghai Lake were detailed below. Regional transport is an important contribution to aerosol particles at the rural site. However, this study was not to quantify the contributions of regional transport from different source areas because such analyses need the involvement of modeling work which is beyond the scope of this study. Similarly, the infrastructure (railroads, agriculture, power plants, etc.) might also have impacts on the sampling site, however, without additional measurements, it is difficult to evaluate and quantify their impacts.

Following the reviewer's suggestions, we revised the manuscript as much as we can. Please see our detailed point-to-point responses below.



Fig. S3. The back trajectories during five episodes marked in Fig. 3. The time for each trajectory was 00:00 on 14 Sep. for Clean 1, 12:00 on 22 Sep. for Ep1, 00:00 on 27 Sep. for Ep2, 00:00 on 9 Oct. for Clean2, 00:00 on 14 Oct. for Ep3. The time is UTC time which is equal to Beijing time minus 8 hours.

It was a bit confusing seeing several comparisons in the text and figures with the other sites that are listed in the Supporting Information (SI) Table S1. Those sites are very far away from the sampling location and this paper is probably not intended to be a review of all aerosol composition measurements in China. It was also misleading that the Aerosol Chemical Speciation Monitor (ACSM) instrument is not sensitive to refractory material, which previously was shown to comprise over 60% of the $PM_{2.5}$ composition for a summertime study at the Bird Island site (Li et al., *Tellus B*, 2013). The Bird Island results are probably the most relevant published data for comparison, yet they were barely mentioned in the paper. It may be more appropriate to limit the other comparisons to a short, stand-alone section.

The main objective of Fig.1 and Table S1 was to have a better understanding of aerosol characteristics at the rural site on the Tibetan Plateau compared to other rural sites in Asia. As shown in Fig. 1, aerosol composition at the national background site in Menyuan was substantially different from that observed in eastern China and also that over the Pacific Ocean, indicating the different impacts of anthropogenic activities on regional background aerosols. Such information is important for readers to have a better knowledge of aerosol chemistry at various rural sites rather than one single site in East Asia. Although ACSM is insensitive to refractory materials, we focus on the comparisons of non-refractory submicron aerosol composition here. In addition, all the data in Fig. 1 and Table S1 were measured by Aerosol Mass Spectrometers, which can be directly compared to the results in our study. The study by Li et al. (2013) at the Bird Island site showed a large fraction of unknown material (61%) in $PM_{2.5}$. Without further analysis of the unknown materials, it is hard to tell that the unknown material was mainly mineral dust. There's also possibility that organic matter (OM) was underestimated when converting OC to OM with a low OM/OC ratio. Unfortunately, we didn't find the OM/OC ratio used in Li et al. (2013).

The Bird Island results were cited in the introduction but not compared in detail in the text. The reasons include: 1) Li et al. (2013) reported $PM_{2.5}$ composition rather than PM_{1} ; 2) different techniques were used (offline filter sampling vs. online real-time measurements). For instance, filter sampling of

ammonium nitrate might have significant loss in summer; 3) the study by Li et al. (2013) was conducted in a different season; 4) Li et al. (2013) focus on the analysis of molecular markers in organic matter.

Section 3.1: As mentioned above, it would be useful to have a series of back trajectories for the site – wind-rose plots for the higher wind speed data. It was not clear where the winds were coming from at the various wind speeds. From the back trajectories presented in Figure S1, Clean1 appeared to be near Xining in the past 12 hours whereas Clean2 appeared to be only from desert. Why does the data with the back trajectory from near Xining appear “clean”? The back trajectories for Episodes(EP) 1, 2, and 3 of high mass concentrations were not presented. It would be helpful to see where the potential large sources are – desert, saline lakes, forest, populated areas (density map?), power plants, railroads, etc.

We thank the reviewer’s comments. As shown in Fig. 3, there were no clear wind direction patterns during this study, which was also illustrated by the wind rose plot below (Fig. R1). The wind rose plot showed that high wind speeds were mainly from the north, the northeast, the south, and the southeast.

Although the back trajectory during clean 1 was near Xining, the trajectory height remained at more than 3000 m which was well above the elevation of Xining. This indicated that the air masses could be above the boundary layer height when passing through Xining, which explained the low aerosol loadings.

The typical trajectories during five episodes are now presented in supplementary, and some hot spots, like cities were marked. However, this study was not intending to investigate the impacts of deserts, saline lakes, and power plants to aerosol composition at the sampling site, these potential sources were not marked to avoid misleading readers. Still, the reviewer pointed out a good point, future studies, e.g., modeling work can be used to further investigate the sources and transport of aerosol particles on the Tibetan Plateau.

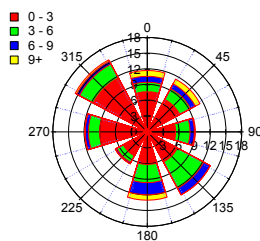


Fig. R1. Wind rose plot of for the entire campaign.

Section 3.2: The diurnal plots are difficult to interpret because all the data are combined and there was no common air mass history selected for this analysis. If biomass burning was a large local source, it should be removed from these plots. An indicator of time since emission could be determined from the fraction of total sulfur as sulfate or $(\text{sulfur from sulfate})/(\text{sulfur from sulfate} + \text{sulfur from sulfur dioxide})$. This would only be relevant for sulfur sources and it is unclear that sulfur sources are coincident with other pollutants (for example, carbon monoxide or CO and BC) in the region.

We thank the reviewer’s comments. The average diurnal plots for the entire study can eliminate the impacts of individual plumes and give the most important and common mechanisms driving the diurnal variations of aerosol species. Such an approach using diurnal plots to explore the formation mechanisms

and boundary layer dynamics has been widely used in the community of atmospheric chemistry. Although biomass burning was a large source of organic aerosol in this study, it won't change the overall diurnal patterns of aerosol species. For example, Fig. S4 shows the diurnal patterns of aerosol species after excluding biomass burning events which were remarkably similar to those with biomass burning events included. The diurnal variation of organics showed the largest difference as biomass burning aerosol was dominated by organic aerosol. Nevertheless, the diurnal cycle of OOA was exactly that of organics without biomass burning impacts, which is shown in Fig. 5 in the revised manuscript.

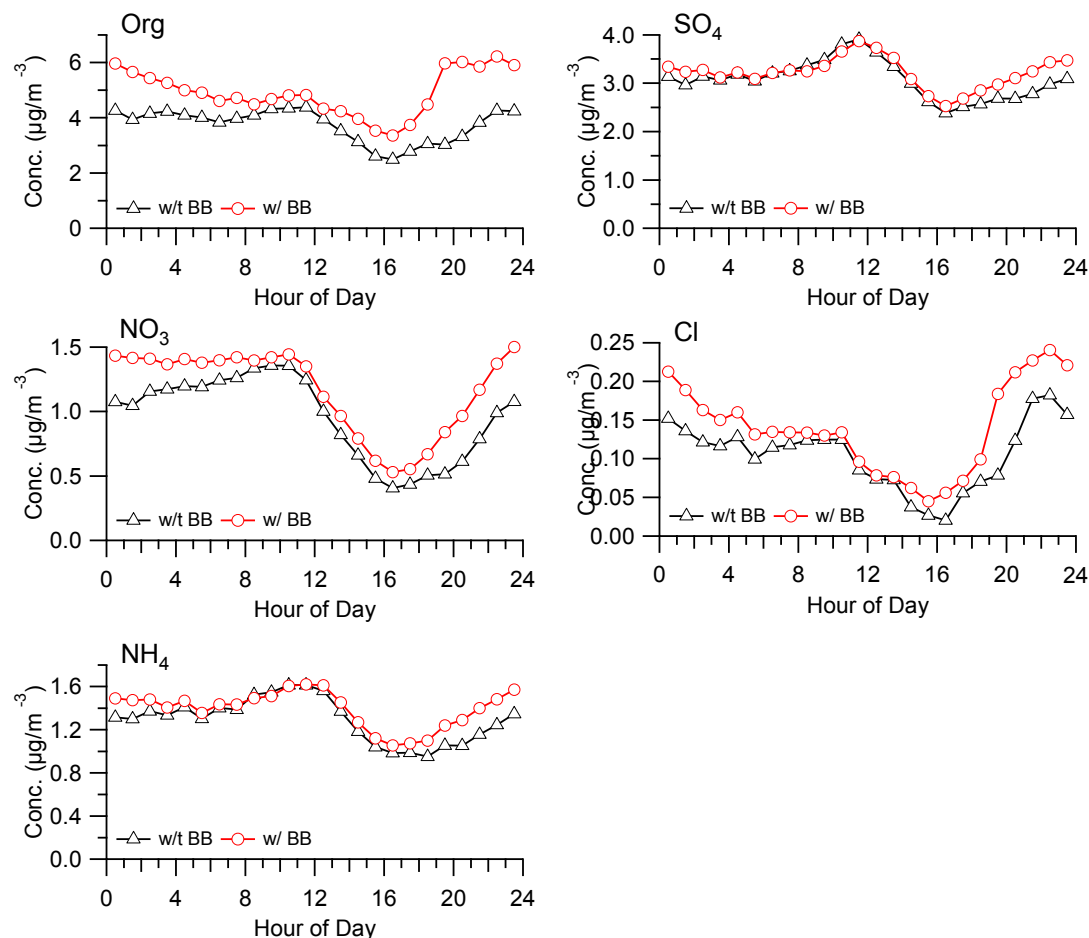


Fig. S4. Diurnal profiles of NR-PM₁ species with (w/) and without (w/t) biomass burning (BB) events.

The fraction of sulfur as sulfate in total sulfur, which is also known as sulfur oxidation ratio (SOR), can be used to indicate how much SO₂ is oxidized to sulfate (Fig. R2). The SOR is subject to multiple influences, for instance, gas-phase photochemical production and aqueous-phase processing. Considering that our sampling site is far away from point sources, it is difficult to use this ratio to evaluate the emission sources because of oxidation processing of SO₂ during the transport. But still, we observed some correlations between SO₂ and CO (see Fig. S5 for detail) except biomass burning events. It's likely that they were from the same sources. Modeling work is needed for further investigation which is beyond the scope of this study.

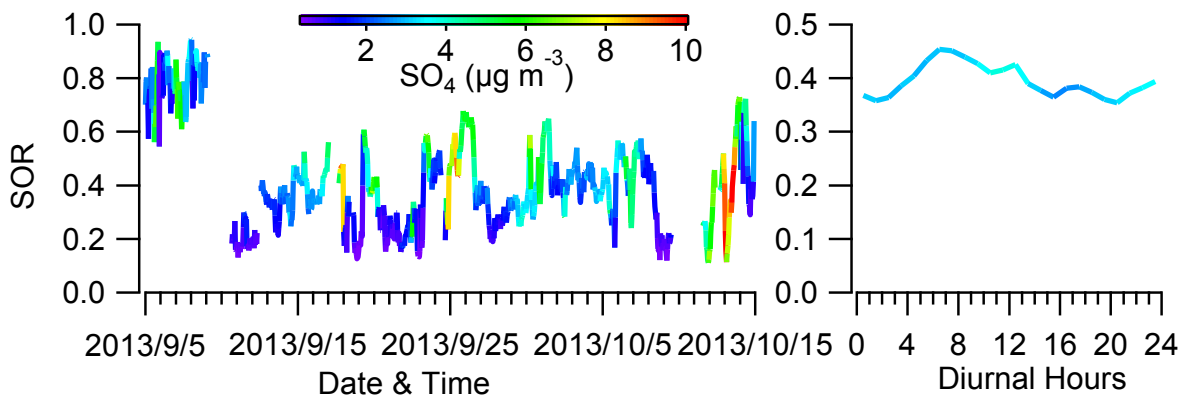


Fig. R2. The time series and diurnal variation of the fraction of S in total sulfur, which was calculated by (sulfur from sulfate)/(sulfur from sulfur dioxide + sulfur from sulfate).

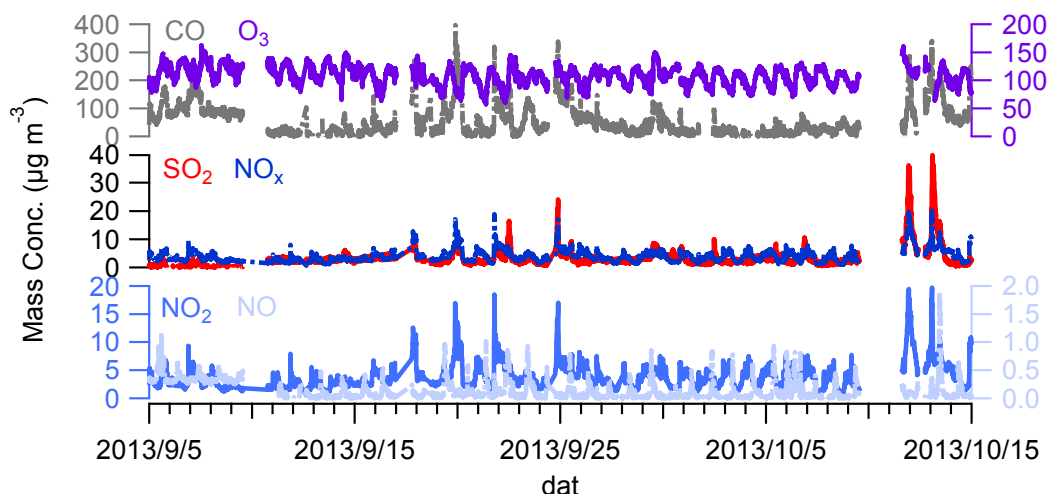


Fig. S5. Time series of gas phase species including CO, O₃, SO₂, NO_x, NO₂, NO during this study.

Section 3.3: It appears that biomass burning was a large local source of PM₁ during the study, however, this point was not made clear in this section.

The fresh and aged BBOA together accounted for 33% of the total OA, which is ~14% of PM₁. Following the reviewer's suggestion, we claimed the importance of BBOA as a local source in the revised manuscript, which is "The fresh and aged BBOA together accounted for 33% of the total OA suggesting that BBOA was a large local source of OA during the observational period."

Section 3.4: It would be helpful to know a bit more about the meteorology, wind speed and direction as a function of the time of day. Back trajectories would also be useful to interpret the data, especially since particle nucleation was previously observed at Mt. Waliguan for air masses originating from the western sector of that site. There is no physical basis for changes in the smallest particle diameter (Equation 2) to be correlated to bulk particle composition (Figure 9), especially since the size of particles measured by the ACSM is much larger than detected by the Scanning Mobility Particle Sizer (SMPS).

during new particle events. Suggest converting the growth rates from a diameter to volume unit for comparison and seeing if the volume increases match the mass increases.

Thank the reviewer's comments. The diurnal variations of temperature (T), relative humidity (RH), wind speed (WS) and wind direct (WD) are shown in supplementary as Fig. S1. Different from Mt. Waliguan, we observed particle nucleation event for almost every day (~80% of the time). We agree with the reviewer that there was no direct relationship between bulk particle composition and particle growth at small sizes. The best way is to measure the chemical composition of nano particles which was unfortunately not available in this study. Therefore, we used the bulk composition change to demonstrate the potential role of aerosol species in particle growth. In fact, the particles with mobility diameter larger than 20 nm (approximately 30 nm in vacuum aerodynamic diameter) can be detected by the ACSM. The aerosol composition change could indicate, at least to a certain degree, their roles in particle growth.

The time series of SMPS volume concentration would be very similar to that of PM₁ since the estimated particle density was relatively constant (1.5 g cm⁻³) and the SMPS mass (volume times density) agreed well with PM₁. Therefore, the diurnal cycles of particle volume during NPE and non-NPE would be similar to those of PM₁ in Fig. 8. No doubt, the volume increases would match the mass increases.

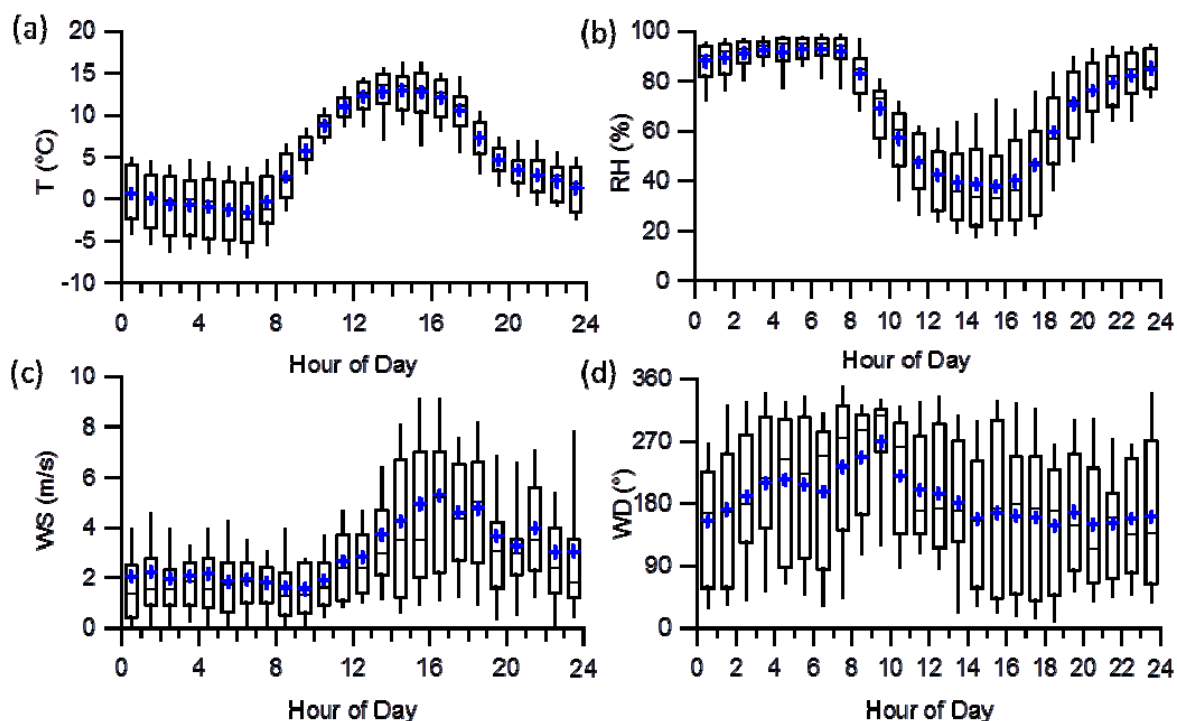


Fig. S1. The diurnal variation of meteorology including (a) temperature (T), (b) relative humidity (RH), (c) wind speed (WS), and (d) wind direction (WD).

Other comments:

Title: change "in" to "on"

Changed.

Line 142: change “plotentilla” to “potentilla”

Changed.

Line 144: It seems that there were no other urban areas nearby. How big is Menyuan? Datong? Also mention the population of Xining. Are there railroads or power plants impacting the site? Note that the comparisons of city size and population need to be more quantitative, to be put into context with other locations around the world.

The area of Menyuan autonomous county is 6896 km² with a population of 155,800. Note that our sampling site is approximately 45 km from the town of Menyuan. In comparison, Datong autonomous county is 3090 km² with a population of 453,000. The population of Xining is 2,290,000 until 2013. The closest road near the sampling site is the national road G227. There are power plants in the city of Xining. As shown in our data, several SO₂ plumes might be impacted by the emissions from power plants. However, this study is not intending to investigate the impact of railway roads and power plants to aerosol chemistry at the national background site, therefore, we did not discuss such details in the text to avoid misleading the readers.

Following the reviewer’s suggestions, some quantitative information on the sampling site was added in the revised manuscript.

Line 146: define “few”

We thank the reviewer’s comment. During this study, we only saw sporadic vehicles on G227. Because we didn’t count the number vehicles passing through the national road G227, we cannot give the exact number here, so “few” was used here. The vehicle emissions could not make a large contribution to the aerosols at our sampling site because the concentration of traffic-related NO was very low, < 1 ppb for most of time (see Fig. S5 for detail).

Section 2.2: How high were the sampling inlets? How was black carbon measured?

The sampling height is approximately 2 m, and the particle residence time in the sampling tube is ~ 5 s. Black carbon was measured by an Aethalometer (AE31, Magee Scientific Corp.). Such information was added in section 2.2 in the revised manuscript.

Lines 205-207: Need to mention the particle size range that is transmitted into the ACSM.

ACSM measures aerosol particles in the size range of 10 nm – 1 μm (vacuum aerodynamic diameter), and SMPS measures aerosol particles in the size range of 12 – 478 nm. The size information on ACSM measurement was added in section 2.2.

Lines 273-279: These sentences imply that air is transported from Lanzhou to the site. Is that what was intended here? Perhaps it needs to be reworded.

Thank the reviewer’s comments. To avoid confusion, we deleted this sentence in the revised manuscript.

Section 3.2: This is the first place where the gas phase measurements are discussed. It would be helpful to have a time series of them in the SI.

Good suggestion. The time series of gas phase species including CO, O₃, SO₂, NO_x, NO₂, NO are now shown in the Fig. S5.

Line 369: change “rationale” to “rational”

Changed.

Line 403: change “bio-modal” to “bi-modal”

Corrected.

Line 448-449: How long would it take for urban air to get to the site?

It may take hours depending on wind speed and also the vertical convection.

Line 485: delete the word “ubiquitously”

Deleted.

Many of the figures do not include the units of measurements. Since the sampling site is at a high altitude, the units of everything should be converted to standard conditions (273.15 K temperature and 1013 hPa pressure). Units for gas phase data should be in mixing ratio (ppbv or pptv) instead of micrograms per cubic meter.

We thank the reviewer’s comments. The ambient mass concentrations can be easily converted to those under standard conditions using ideal gas law based on the measurements of pressure and temperature. However, we kept the ambient concentrations here to be consistent with previous studies in Qianghai e.g., (Li et al., 2013) and also the gas-phase species measurements. This was clearly stated in section 2.2 as “All the data are reported with ambient conditions at Beijing Standard Time”.

The concentrations of gaseous species can be reported either in mixing ratio with the units of ppbv or ppmv, or in micrograms per cubic meter that has been widely used in the community.

Figure 1: It should be noted that the pie-charts do not include dust and salts, which could be important for the total PM₁ at the National Background Site. Suggest making the inset satellite image larger than the pie-chart map (moving the pie chart map to SI). There should be a scale on the image, along with markers for other locations such as Bird Island, Mt. Waliguan, Xining, and Wuwei (another potentially significant “nearby” urban source). Consider making it a slightly larger scale to show the location of Lanzhou, too.

We thank the reviewer’s comments. Mineral dusts and salts have the dominant fraction in coarse particles and a small fraction in PM_{2.5}, for example, the water-soluble Ca²⁺ and Mg²⁺ together accounted for < 2% of PM_{2.5} in Qianghai (Li et al., 2013). Considering that we measured PM₁ in this study, mineral dust and salts are expected to contribute a much smaller fraction in PM₁. As a result, we don’t think that refractory dust and salts can make an important contribution to PM₁. More important, all the

comparisons in Fig. 1 are from AMS and BC measurements none of which included mineral dust and salts.

Fig. 1 is important for readers to understand the aerosol chemistry at the rural site on the Tibetan Plateau compared to other rural sites in Asia. It is of particular interest that aerosol composition at rural sites showed clear differences from the west to the east in Asia, which was strongly associated with the influences of anthropogenic activities. Therefore, we kept this figure in the revised manuscript. In addition, although previous measurements were conducted at various sites near Menyuan, e.g., Bird Island, Mt. Waliguan, Xining, and Wuwei, this study was not intended to compare with them since the measurements were different. To address the reviewer's comments, a large satellite image plot with the marked locations is given in Fig. S3

Figure 2: The correlation seems to change with time, where the SMPS is lower in the later part of the study by a larger amount than in the beginning. Is there a reason for this?

Right, we also noticed these differences. One of the reasons was likely due to the size differences between 12 – 478 nm and 478 nm – 1 μ m during these periods. Unfortunately, we don't have the size measurements above 478 nm to investigate such differences.

Figure 3: It is difficult to see several traces on the figure, especially the wind direction, black carbon, and PM₁. Why are there gaps in the data? There is a clear diurnal pattern in wind speed. Is there a diurnal pattern in wind direction too?

We tried our best to make Fig. 3 clear to readers. The gaps in the Figure were caused by the unavailable BC data due to malfunction of the instrument. It seems that there was no clear pattern for wind direction (Fig. R1). As shown in Fig. S1, the wind direction varied largely throughout the day (see 25 – 75th percentiles).

Figure 4: It would be helpful in the caption to point out local sunrise and sunset. Note the legend should indicate that lines with symbols in Part (a) correspond to PM₁.

Good point, the time of sunrise and sunset were added in the caption in the revised manuscript. In addition, the legend for PM₁ was added in (a).

"Figure 4. Average diurnal cycles of (a) mass concentration; (b) mass fraction of PM₁ species; (c) ratios of aerosol species to CO, and (d) gaseous species. The local sunrise and sunset was around 7:00 and 19:00."

Figure 5: There should be tick marks on the top x-axis of Part (a) or perhaps a marker indicating where m/z 60 is located. It is difficult to distinguish the different colors in these plots.

The markers of m/z 60 and 73 were added in the figure. Because BBOA highly correlated with m/z 60, it's a bit difficult to distinguish these two species in the figure.

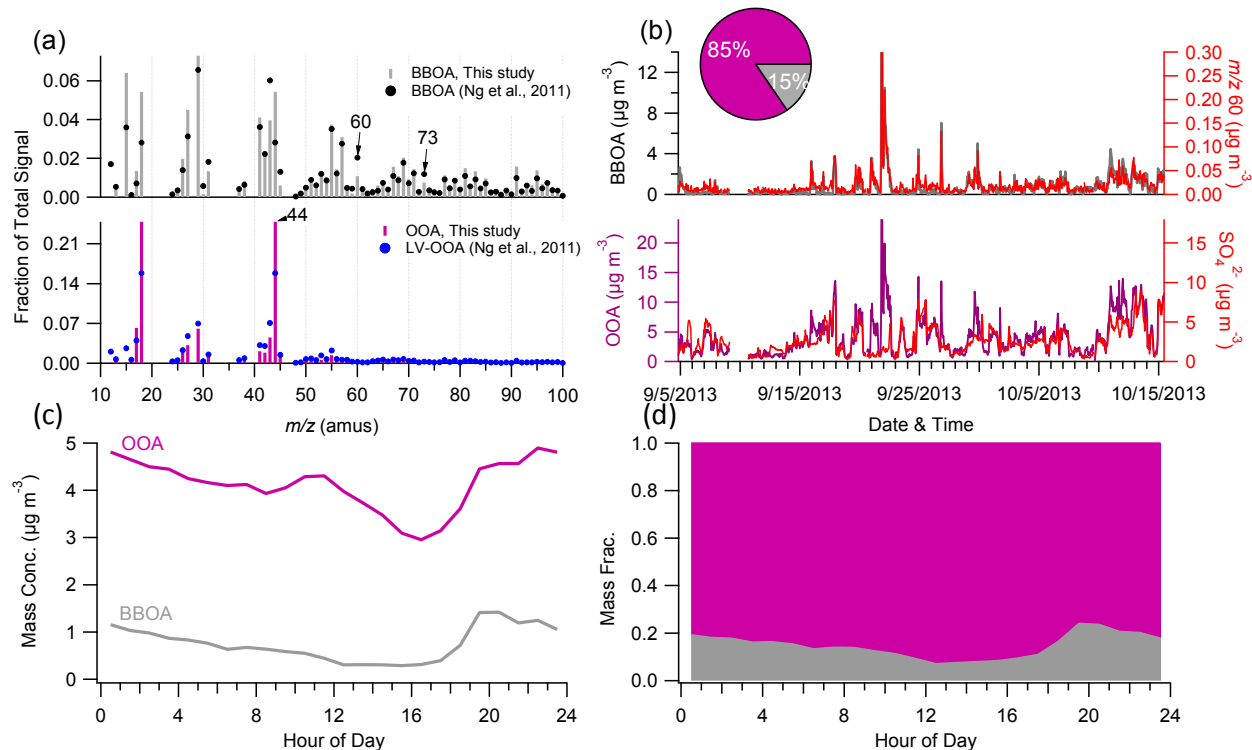


Fig. 5. (a) Mass spectra and (b) time series of mass concentrations of BBOA and OOA. (c) and (d) show the average diurnal cycles of BBOA and OOA. In addition, the standard average mass spectra of BBOA and OOA in Ng et al. (2011) are also shown in (a) for the comparison. The pie chart in (b) shows the average composition of OA for the entire study.

Figure 6: Should define in the caption what “post-processed OOA” means.

The post-processed OOA that is “ $OOA - SO_4 \times [OOA/SO_4]_{NBB}$ ” was added in the caption.

Figure 7: For Part (b), need to label the y-axis and include text about the dotted lines in the caption. Should also note when it rained in Part (a) and the time of local sunrise in Part (d).

Fig. 7 was revised according to the reviewer’s suggestions. Note that it’s difficult to read if adding precipitation in (a). The readers can find precipitation information in Fig. 3.

Figure 8: Remove irrelevant plots (top mass concentration, mass fraction) or put into SI. Units for CO and PM_{10}/CO are missing. Again, note the time of local sunrise in the caption.

Thank the reviewer’s comments. The diurnal plots of mass concentrations and mass fractions are important for readers to know the variations of aerosol species and composition during NPE and non-NPE events. Therefore we kept them in the revised manuscript. The unit of CO, and the sunrise time were added. Note that PM_{10}/CO has no unit since both PM_{10} and CO were reported in $\mu g m^{-3}$.

Figure 9: The bottom plot in Part (a) is probably not relevant and could be removed. Suggest creating a new plot for Part (b) with growth rate in units of volume change per unit time and aerosol mass change (the difference in mass normalized to CO) per unit time. Still may not be correlated if the size ranges are not overlapping.

We thank the reviewer's comments. Fig. 9a is important for readers to know the variations of particle growth rates and also chemical composition, thus we kept it in the revised manuscript. The time series of SMPS volume concentration would be very similar to that of PM₁ since the estimated particle density was relatively constant (1.5 g cm⁻³) and the SMPS mass agreed well with PM₁. Therefore, the diurnal cycles of particle volume during NPE and non-NPE would be similar to those of PM₁ in Fig. 8. The volume concentration would show decreases during NPE. Even considering the dilution by planetary boundary layer height using CO, the variation of the volume concentration would be also small (see Fig. 4c for PM₁/CO). In addition, the diurnal variation of OOA was similar to most of aerosol species in this study. As a result, we don't think it can tell us much information by linking particle volume with OOA. Here, we focus on the relationship between the growth of particle sizes and aerosol composition. Such an approach was also used in many previous studies on new particle formation. However, we agree with the reviewer that our understanding of NPE and aerosol composition were still limited because no size-resolved composition was available in this study. Future studies are absolutely needed for further investigations.

Table S1: Make headers match the site location names on the map from Figure 1.

Following the reviewer's suggestion, we corrected the headers of Table S1.

"Table S1 A summary of mass concentration and composition of PM₁ species measured by AMS at different locations in East Asia."

Location	Okinawa Japan	Fukue Japan	Jeju Korea	Jiaying Shanghai	Changdao Island	Mount.Tai	Kaiping Shenzhen	Lanzhou	
Time	10/3/2003	3/20/2003	4/13/2001	6/29/2013	3/21/2011	2011	10/12/2008	7/11/2012	
	10/28/2003	4/18/2003	4/30/2001	7/15/2013	4/24/2011		11/18/2008	8/7/2012	
Org	Mass	3.1	5.0	3.5	10.6	13.4	11.2	11.2	11.5
	Frac.	21.4	41.7	40.7	32.1	28.8	32.6	33.9	47
SO ₄	Mass	9.2	4.8	3.1	8.2	8.3	9.2	11.1	3.9
	Frac.	63.4	40.0	36.0	25.2	17.8	26.7	33.6	16
NO ₃	Mass	0.19	0.56	0.51	5.9	12.2	7.2	3.5	2.5
	Frac.	1.3	4.7	5.9	18.0	26.1	20.9	10.7	10
NH ₄	Mass	1.9	1.6	1.5	4.2	6.5	5.8	4.6	2.7
	Frac.	13.1	13.3	17.4	12.6	13.9	16.9	14.0	11
Chl	Mass.	0.06	0.07		1.0	1.3	0.95	0.36	1.0

Frac.	0.4	0.6		3.0	2.8	2.8	1.1	4
BC	Mass			3.0	2.5		2.2	2.9
	Frac.			9.1	5.4		6.7	12
NR-PM ₁	14.5	12.0	8.6	29.9	44.1	34.4	30.8	21.6
PM ₁				32.9	46.6		33.1	24.5
References	(Zhang et al., 2007)	(Takami et al., 2005)	(Topping et al., 2004)	(Huang et al., 2013)	(Hu et al., 2013)	(Zhang et al., 2014a)	(Huang et al., 2011)	(Xu et al., 2014)

Figure S2: Add the outline of Qinghai Lake and the locations of Qilian Shan Station, Bird Island, Mt. Waliguan, and Xining, Maybe Wuwei. Is the black curve on the bottom plot indicating the ground level? Please note that in the caption. Also add the time difference between UTC and the sampling site.

Thank the reviewer's suggestions. A new figure with the back trajectories of five episodes is presented in supplementary. The locations of various sites mentioned by the reviewer were marked in the figure, and the back trajectory time was also included in the caption.

Response to Reviewer #2

The paper entitled of “Chemical characterization of submicron aerosol and particle growth events at a national background site (3295 m a.s.l) in the Tibetan Plateau” present the first high-time resolution chemical composition measurement in the Tibetan Plateau where the chemical composition and distribution are important factors for evaluating the climate forcing of aerosol. PMF analysis is applied on organic mass spectra and obtains two factors and the results show biomass burning emitted aerosol is an important source for primary and secondary OA. In addition, at this remote area, the paper also shows new particle formation (NPF) events are an important chemical process and aerosol source. The topic of this paper is interesting and suitable for publication in ACP. I agree this paper for publication after revising following comments and suggestions below.

We thank the reviewer’s positive comments.

Specific comments:

Section 2.1: add several sentences for describing the meteorological conditions of study period, such as air temperature, precipitation, and wind condition.

Following the reviewer’s suggestions, we expanded the discussions on meteorological conditions in the revised the manuscript.

In this study, ambient temperature averaged 4.9 °C (-8.7 - 17.9 °C) and wind speed varied largely with an average value of 3 m s⁻¹. In addition, several precipitation events were also observed, particularly during the first half period of this study (Fig. 3).

Section 2.2: What is the environment condition for instruments? What is the length of inlet and the size of critical orifice? Because the particle loss can be a problem at the high elevation sampling site, so these issue should be considered. In addition, does the SMPS work well at such high elevation site?

All the instruments were placed in an air-conditioned room with the temperature maintaining at ~23°C.

The length of the sampling line (1/2 inch stainless steel) is approximately 2 m. The residence time of aerosol particles in the sampling tube is estimated to be ~ 5 s and the size of critical orifice for ACSM inlet is 100 μm. Therefore, the particle loss for ACSM measurements should be small. The butanol-based SMPS worked well at such a high elevation site (3295 m). This is also supported by the good agreements between ACSM and SMPS (Fig. 2). We agree with the reviewer that SMPS might have problems at a higher elevation site due to the limitation of vapor pressure of butanol.

Section 3.1, p13524, line9-14: the chemical composition of PM1 during the two clean periods may reflect the aerosol composition at the free troposphere which the contribution of sulfate were much higher than those non-clean periods, can the author give some explanations for this phenomenon.

There’s possibility that the aerosol composition during clean periods were affected by free troposphere. According to previous studies (Zhang et al., 2014b), the nighttime would be the period with the largest influence from free troposphere when planetary boundary layer (PBL) height is low. Considering both clean period and pollution events generally lasted more than one day, both of them could be affected by the free troposphere. Unfortunately, we were unable to quantify the chemical composition of aerosol from free troposphere based on the measurements in this study.

However, we noticed that the chemical composition during clean periods showed much lower contribution of nitrate that was dominantly from the oxidation of anthropogenic NO_x . Therefore, the higher sulfate and lower nitrate during clean periods might suggest that the air masses during clean periods were either from a longer transport when ammonium nitrate was deposited or evaporated due to dilution processes, or from less anthropogenic influenced regions with low nitrate.

Following the reviewer's suggestion, we expanded the interpretation of the chemical differences between clean periods and pollution episodes.

"We also noticed that the two clean periods showed overall higher contribution of sulfate and lower contribution of nitrate compared to the three pollution episodes. The possible reasons were likely due to that the air masses during clean periods were either from a longer transport when ammonium nitrate was deposited or evaporated due to dilution processes, or from less anthropogenic influenced regions with low NO_x emissions."

Section 3.1, P13524 line 25 to P13525 line 4: As illustrated at section 3.4, sulfate can be from the new particle formation (NPF) as sulfuric acid is an important component for NPF. Acidic PM₁ particle was also observed at QSS at the Qilianshan Mountain at the northeastern TP which use filter measurement by MOUDI.

It's a good point. We added such an explanation in the revised manuscript. "Also note that the newly formed sulfate particles during the frequent NPF events might also have played a role". In addition, the work conducted at the Qilainshan Mountain (Xu et al., 2015a) was cited.

Section 3.3: Are the diurnal pattern of these two factors evidently? It is better to show diurnal variations in the Fig.5 which is useful to support the results of PMF analysis. In addition, it is interesting that the OOA is highly oxidized, but the possible explanations are not given by the authors. The aqueous processes may be an important factor for this highly oxidized OOA because the extremely high RH (seems more than 95% in Fig.1) during night-time every day.

Yes, the diurnal patterns of OOA and BBOA were evident (Fig. 5). While OOA showed similar diurnal variations to that of PM₁, BBOA was characterized by higher concentration at nighttime. Following the reviewer's suggestions, the diurnal patterns of OOA and BBOA were added in Fig. 5 in the revised manuscript, and the related discussions were expanded in section 3.3.

OOA is highly oxidized suggesting that OOA was well processed at the national background site. As the reviewer mentioned that aqueous-phase processing at nighttime might also play a role. Such explanations were added in the revised manuscript.

Section 3.4: It is not easy to say which species is the major contribution for NPF using the whole size range aerosol composition. But it is not other better way to get the small size range chemical composition using ACSM. One suggestion is to get some information from single particle composition if these data are available.

This is a good suggestion. Unfortunately, the single particle composition data was not available in this study.

Technical comments:

P13517, line 21: $\sim 200000 \text{ km}^2 \rightarrow \sim 2000000 \text{ km}^2$

Corrected.

P13525, line 20: decreases \rightarrow decreased to

Changed.

P13526, line 22: large plumes \rightarrow large peaks

Changed.

P13527, line 25: Figs. 5b and 6 \rightarrow Fig. 5b and 6; the different correlation \rightarrow the weak correlation

Changed.

P13528, line 10: rationale \rightarrow rasionable

We changed "rationale" to "rational".

P13528, line 25: check the number of $3.9 \mu\text{g m}^{-3}$

Thank the reviewer's carefulness. It was corrected as " $0.82 (\pm 2.65) \mu\text{g m}^{-3}$ ".

P13530, line 15-19: "the results this period" is a repeat information.

It was deleted in the revised manuscript.

P13531, line 7: change \rightarrow variation

Changed.

P13532, line 2: Y. M. Zhang et al. (2011) \rightarrow Zhang et al. (2011)

Corrected.

P13532: line 10: 5 September or 4 September

Thank the reviewer's carefulness. It was 5 September. All the date was corrected in the revised manuscript.

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