Dear editor,

Here we submit a revised manuscript by *Li* et al., entitled "Mixing state and sources of submicron regional background aerosols in the North Qinghai-Tibetan Plateau and the influence of biomass burning". The point-point responses were listed as below.

We carefully revised the manuscript based on the reviewers' comments. Correspondence and phone calls about the paper should be directed to *Weijun Li* at the following address, phone, and e-mail address:

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Best regards,
Sincerely yours,
Weijun Li on behalf of the coauthors

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Thanks for The referee#1's comments. Those comments are all valuable and helpful for improving our paper. We answered the comments carefully and have made corrections in the submitted manuscript. The corrections and the responses are as following:

1. Page 24372, line 10-11, here author said "however, aerosols of the vast grasslands of the northern TP have not been studied".

a) It is better to briefly state why it is necessary to characterize the aerosols from the grassland atmosphere, if the physicochemical properties of the grassland aerosols are different from those in other places of TP?

b) Page 24372, line 18-20, what is the definition of age? Why aged aerosols represent the typical chemical composition of this continental background region? If this means that fresh aerosols are very less at the sampling site and most of airborne particles are long-range transported?

Response 1 a): We added the reasons why we do study aerosol particles in grassland over the TP.

"Grassland is one of the largest geomorphology in the TP. There are only a few herdsmen and farmers living in the vast grasslands of the norther TP. Air pollutants from anthropogenic and natural sources can be easily transported over low bushes in the grasslands under high wind speed in north TP (Figure S1). However, aerosols in the troposphere have not been studied over the vast grassland in the northern TP."

Response 1 b): Here we deleted the sentence based on the logic in the paragraph. We added the definition about the "aged" particles in section 4.3

2. Page 24373, section 2.1,

a) what is the altitude of the sampling site? This information is important.

b) The density of particles is assumed to be 2 g/cm3, what is the rationale?

Response 2 a): We added

Response 2 b): We usually used the value to calculate the size cut off of the impactor. We consider ammonium sulfate (1.77 g/cm^3), mineral dust (2.6 g/cm^3), and organic matter (1.5 g/cm^3) as the major aerosol types. The average density (calculated at 1.96) of the mixed aerosol particles was assumed to 2 g/cm3.

3. Page 24376 and 24391, Table 1, it's better to give more data such as the standard

deviation, minimum, and maximum values, because data here are statistic numbers; the mean value itself does not give enough information.

Response 3: Thank you. We add standard deviation, min, and max value.

Table 1 Concentrations of six air pollutants during the sampling period, two pollution periods, and clean period

Pollutants	All data		polluted period-1		polluted period-2		other period	
	mean ±SD Max, Min	n	mean ±SD Max, Min	n	mean ±SD Max, Min	n	mean ±SD Max, Min	n
$PM_{2.5}$	17.06 ±11.39 68.70, 0.20	715	17.6 ±11.46 59.10, 0.20	152	24.45 ±15.12 68.70, 0.30	99	15.32 ±10.41 62.80, 0.20	464
BC	0.54 ±0.42 3.73, 0.02	805	0.55 ±0.52 3.73, 0.04	176	0.85 ±0.50 2.04, 0.02	119	0.47 ±0.40 3.73, 0.03	510
SO_2	1.27 ±1.34 13.93, 0.02	8822	1.2 ±0.99 8.43, 0.20	1981	2.73 ±3.09 13.93, 1.41	1063	1.03 ±0.65 8.43, 0.02	5778
NO_X	2.05 ±1.96 9.86, 0.31	8842	2.37 ±1.33 9.33, 0.65	2001	3.41 ±1.70 9.59, 0.55	1063	1.69 ±0.97 9.33, 0.31	5778
CO	44.78 ±48.03 318.00, 0.20	7822	63.45 ±55.59 318.00, 0.20	1939	104.23 ±54.69 272.40, 0.60	1030	24.68 ±39.91 318.00, 0.20	4853
O_3	50 ±7.86 98.63, 20.43	8817	47.87 ±7.70 67.70, 26.66	2000	49.01 ±10.00 98.63, 20.43	1039	50.53 ±7.56 96.77, 26.66	5778

All data period: 10 Sept.-15 Oct. 2013; Polluted period-1: 18 Sept.-25 Sept. 2013; Polluted period-2: 11 Oct.-15 Oct. 2013

4. Page 24377, lines 5-9, the method for classification of the aerosol types should be briefly introduced, which would be helpful for readers to understand why the particles are categorized as fly ash and others are classified as mineral dust.

Response 4: We briefly introduced the classification.

"For example, mineral dust particles normally display irregular shapes and fly ash particles are spherical, although they both have similar compositions such as Si and Al."

5. Page 24377, line 6 KCl-NaCl particle. The particle should contain K, Na, and Cl in Figure 4.

a) The authors didn't show the crystalline of the particle. The name should be changed to K-Na-Cl.

b) And line 17 organic carbon should be changed to organic.

Response 5 a): We changed the "KCl-NaCl" to K-Na-Cl particle

Response 5 b): We changed the "organic carbon" to "OC".

6. Page 24378, line 25, 33-36 and 34-48 of what? These percentage numbers are in

mass or particle numbers?

Response 6: These percentage numbers are particle numbers.

7. Page 24379, line14 Yak dung,

Response 7: We revised this.

8. Page 24380, line 18, what the regional property is? Please give more specific descriptions.

Response 8: We revised the sentence.

"For example, Du *et al.* (2015) suggested that oxygenated organic aerosols from anthropogenic sources and biomass burning transported over a long distance to the sampling site in the QTP."

9. Page 24380, line 19-20, I do not think primary organic aerosols are refractory. In fact, unlike mineral dust and soot, both are refractory, organic compounds in airborne particles can be completely measured by aerosol mass spectrometer and OC/EC carbon analyzer via heating evaporation, although both instruments can not give molecular information.

Response 9: Thank you. We revised the part.

10. Page 24381, line 10-14, it's better to specifically mark the particles in figures 6, 9 and 10 in order to let readers easily recognize which particle is heterogeneously mixed and which is homogeneously mixed.

Response 10: We specifically point out the heterogeneous and homogeneous mixture. We also added description in each figure caption.

11. Page 24383, line 9-11, this sentence is confusing to me.

Response 11: We revised the sentence as follows:

"However, there is no any report say that the emissions of coal combustion from power plants or other industrial sources have critical regional influence."

12. Page 24383, line 4 and other places throughout the paper, the authors emphasized many times that aerosols in TP are highly aged.

a) What does the age mean?
b) Aerosols in TP are highly aged, if this statement means that aerosols in other East Asia regions are less aged?

Response 12 a): The reviewer 1 also raised this question. We made one definition to explain the "aged" particle in Section 4.3

Response 12 b): We added one sentence here which can make readers to

understand our true meaning. We just pointed out the aged particles in the TP and don't extend to other East Asia regions. Because aging processes of aerosol particles during their transports can significantly change particle hygroscopic and optical properties, we need to pay attention to the issue. In the study we didn't expect the aged particles (SIA associated with fly ash, spherical organic, soot, and mineral) in the remote site. Obviously, the findings in our study in the remote site are different from one recent result in remote Siberia site (Mikhailov et al., 2015).

"Because the complex aerosol particles from different anthropogenic sources intruded into pristine background air, the suspended aerosols became highly aged."

13. In the Figure 4 and Figure 5, EDS spectra were obtained from the individual particles or their part. The measured part on the individual particles should be marked. Otherwise, it's hard for readers to know the details.

Response 13: Thanks. We added markers.

14. In Figure 2/7, equivalent spherical diameter should be equivalent volume diameter.

Response 14: We revised those "spherical" to "volume" in section 2.3 and Figure 2/7.

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We are grateful for the referee#2' comments. Those comments are all valuable and helpful for improving our paper. We answered the comments carefully and have made corrections in the submitted manuscript. The corrections and the responses are as following:

15. p. 24370, line 11 ". . . at the median pollution level . . ." Both "medium" and "median" are used to indicate the pollution level throughout the text. And "median" also appears as a statistical term in the sections 3.2 and 4.3.

a) To prevent confusion, I suggest using only "medium" for the pollution level. There are other "medians" to be corrected in the line 11 of p. 24378, line 1 of p. 24380, line 2 of p. 24385, and caption of Figure 7.

b) On the contrary, "medium size" in the line 9 of p. 24385 should be "median size".

Response 1 a): We appreciated your comments. We revised the "median" to "medium" in the line 11 of p. 24370, the line 11 of p. 24378, line 1 of p. 24380, line 2 of p. 24385, and caption of Figure 7.

Response 1 b): We revised the "medium size" in the line 9 of p. 24385 to "median size."

16. p. 24371, line 10 "Few aerosol measurements have been conducted in the TP." I do not think the number of the references following this line "few". "Quite a few" sounds more appropriate.

Response 2: Yes, we fully agree with the reviewer. We have revised this sentence as follows:

"Quite a few aerosol measurements have been conducted in the TP".

17. p. 24373, line 14 ". . ., with an atmospheric pressure of 69 kPa, a temperature of 283.5 K, and an assumed particle density of 2 g/cm3."

a) Are the pressure and temperature typical at the sampling site?

b) Also, what kind of particle is assumed that has density of 2 g/cm3?

Response 3 a): The pressure is lower at high altitude but the temperature is

normal in summer.

Response 3 b): We usually used the value to calculate the size cut off of the impactor. We consider ammonium sulfate (1.77 g/cm^3), mineral dust (2.6 g/cm^3), and organic matter (1.5 g/cm^3) as the major aerosol types. The average density (calculated at 1.96) of the mixed aerosol particles was assumed to 2 g/cm3.

18. p. 24375, line 2-6 "Additionally we know the relation . . . diameter smaller than 1 μm." The first sentence is awkward and not grammatically right. How about writing like this? "By plotting the ECD against the ESD (Fig. 2), we also obtain the relationship between them as ESD=0.64ECD." In the following sentence, use the abbreviations (ECD, ESD) provided above. Also, I suggest adding a line like "where the correlation between the ECD and ESD is especially good (Fig.2)." after "diameter smaller than 1 μm".

Response 4: We have revised the sentence as follows:

"By plotting the ECD against the EVD (Fig. 2), we also obtain the relationship between them as EVD=0.64ECD. As a result, ECD (d) of individual aerosol particles measured from the iTEM software can be further converted into EVD (D) based on this relationship. In this study, we only considered fine aerosol particles with equivalent volume diameter smaller than 1 μm where the correlation between the ECD and EVD is especially good (Fig.2)."

19. p. 24377, line 12-14 I suggest deleting the line "because understanding their mixing state enables one to determine their sources,. . ., and potential health effects". This is already mentioned in "Introduction" (p. 24372, line 30).

Response 5: Yes, this is already mentioned in "Introduction". We deleted this sentence.

20. p. 24377, line 14 "TEM observations indicate that SIA and organics . . . normally coated these SIA particles (e.g., Figs. 4d, 5a, and 6)." For readers not familiar with TEM analysis, it would be helpful to briefly explain how the features in the figures can be recognized as SIA particles coated with OC.

Response 6: We added more description.

"In other words, OC occurred on surfaces of the SIA particles."

21. p. 24379, line 8-11 "Because KCl-NaCl particles associated with organic matter . . . saline Qinghai Lake and desert." Here the authors present the reasons why they interpret the KCl-NaCl particles to have resulted from biomass burning. In fact, sea-salt particles (similar to particles from saline lake water) smaller than 1 μm do occur at certain conditions. I prefer a milder expression than "should be excluded", like; "Because the KCl-NaCl particles associated with organic matter occurred only in the short pollution periods and are smaller than typical sea-salt or soil particles (mostly >1μm), it is unlikely that they originated from natural sources such as saline Qinghai Lake and desert."

Response 7: We agreed with your advice, and revised this as follows:

"Because the K-Na-Cl particles associated with organic matter occurred only in the short pollution periods and are smaller than typical sea-salt or soil particles (mostly >1μm), it is unlikely that they originated from natural sources such as saline Qinghai Lake and desert."

22. p. 24379, line 11-15 "In addition, our field experimental investigations . . . in 11-15 October (Du et al., 2015)." The phrase "in addition" repeats in the two successive sentences. The first one had better be deleted.

Response 8: We have revised it as follows:

"Our field experimental investigations showed that a few farmers burned cole flowers and highland barley during the autumn harvest season, which are main season crops in the QTP. In addition, the burning of cow dung for residential heating likely caused the high PM2.5 in 11-15 October (Du et al., 2015)."

23. p. 24379, line 5-15 One thing I'm wondering about the KCl-NaCl particles is that, according to Li et al. (2003), KCl in biomass burning smoke can be converted to K2SO4 or KNO3 pretty rapidly. Li et al. (2003) showed that particles in the smoke 16 km downwind included K2SO4 and KNO3 but not KCl. In the present study, the EDS spectra of the KCl-NaCl particles don't show significant

peak of S (Figure 4), suggesting that the particles are "fresh". Doesn't this mean that the particles came from an area relatively close to the sampling site, rather than were transported for distance?

Response 9: Thank you for your good comments. We also considered the question. First, the area is very clean and the smog plume can be spread quickly following the high wind speed. Therefore, the heterogeneous reactions of KCl with acidic gases could be slow. Second, the agricultural biomass burning spots could exist in large area but different time period. It could be long and short transport distance. In cases, we slightly modified the sentences.

24. p. 24379, line 20-24 "The fly ash-containing particles . . . the background air quality." This part sounds rather enigmatic. Coal combustion emits both fly ash and soot. Why do the proportions of fly ash- and soot-containing particles have a reverse relationship between the high and medium pollution levels? To me, the result seems to indicate that the air at the medium pollution level was more affected by coal combustion than at the high pollution level, and that soot-containing particles at the high pollution level were more from biomass burning than coal combustion. Is this consistent with the authors' other observations?

Response 10: Yes, your understanding is right. Coal combustion should emit more fly ash and organics than soot particles (from our recent results). The coal combustion emissions should constantly influence air quality at the medium level and at the high pollution level. Because biomass burning emissions increase during the high pollution period, coal combustion emissions relatively became smaller.

25. p. 24381, line 4 "The results show that more than 90 % of particles at the background site were highly aged." What kinds of particles are defined as "aged"?

Response 11: Individual particle clearly contained more than two types of components which have been defined above. We added one sentence to define the

aged particles.

"In this study, individual particle clearly containing more than two types of aerosol components (e.g., mineral dust, K-Na-Cl, fly ash, SIA, organics, and soot) has been defined as aged particle. More than 90% of particles at the background site were highly aged."

26. p. 24381, line 16 "Figure 7 shows that SIA with OC coating . . . total individual particles." In Figure 7, "particles with coating" are not shown. So comparison between "coated" and "uncoated" particles cannot be done from the figure.

Response 12: We added explanation. SIA with OC coating represent the particle without inclusions in Figure 7. In Figure 7, we only compared the particle with inclusion and without inclusion.

"SIA with OC coating (i.e., particle without inclusions in Figure 7) shift to one smaller size than the total individual particles."

27. p. 24381, line 25 - p. 24382, line 10 "In addition, Figs. 9 and 10 show . . . within sulfate particles (Adachi et al., 2010)." Here the authors discuss the occurrence of soot inclusions at the surface of SIA particles and their effects on optical absorption. This is one of the most interesting parts of this paper, but I would like to point out that a similar occurrence of soot and sulfate was reported in Posfai et al. (1999) (JGR 104, pages 21685 – 21693). Posfai et al. (1999) suggested that the soot at the edges of sulfate particles is a result of crystallization of the sulfates from droplets on the TEM grids, and that the spatial relationship of soot and sulfates observed on the TEM grids is not the same as that in the original airborne particles (page 21689 of JGR 104). Is there any evidence that can disprove this interpretation?

Response 13: It is very good comment for us to further do more test. Here I would like to answer it. If you noticed that there are not droplets for all the particles on the TEM grids in Figures 9 and 10. Our samples are different from Posfai et al., (1999)' samples collected over ocean. We indeed found the difference using the

same sampler in East China and QTP.

28. p. 24383, line 9-11 “However, the emissions . . . has not been reported.” This sentence is not grammatically right. Please rewrite.

Response 14: We rewrote this sentence as follows:

“However, the emissions of coal combustion from power plants or other industrial sources have a decided regional influence. The statement has not been reported.”

29. p. 24383, line 19-24 “Interestingly, we found that . . . the current climate models.” The same question as I already mentioned for the part in p. 24381-24382. I suppose that the difference in spatial relationship of soot and SIA may be due to relative size of the soot inclusions to the SIA particles. Soot particles observed in polluted areas are much larger than those in remote areas, thus appear to be embedded in sulfates on TEM grids. Isn’t this the case?

Response 15: Here, we try to raise one question based on our study. Soot size is a possible reason for the case. However, the details are beyond this study. We may have one systemic study about soot particles in near future. Indeed, the soot particles have different mixing structure with SIA.

30. p. 24383, line 24-27 “Thirdly, the dominant organics, . . . in fine particles.” I don’t get the meaning of this sentence. Please rewrite.

Response 16: We rewrote this sentence.

“fine aerosol particles in the TP mainly contain organics and sulfates with minor nitrates. The result is largely different from fine particles with high nitrate in more polluted areas (Li et al., 2013a;Du et al., 2015;Xu et al., 2015).”

31. p. 24384, line 3-13 “Fourthly, the high-elevation . . . particle aging and formation in the TP.” Indeed, the atmospheric chemistry and processes in the TP are likely to differ from those in the polluted area. But what kind of differences the present study has revealed? Without discussing the findings from the present study, this part is unnecessary and had better be omitted.

Response 17: We received your advice and deleted this part.

32. p. 24393, Figure 2 caption In the text, the number of the particles analyzed by both AFM and TEM is 194 (p. 24374, line 20). Why is the number in the caption is 157?

Response 18: We made a mistake and revised the 157 to 194.

33. p. 24399, Figure 8 Some of the letters in the figure would be difficult to be read when printed on paper. Enlarge.

Response 19: We enlarge the letters in the figure.

34. Technical corrections:

a) p. 24371, line 3 the brightening and 'dimming' phenomenon

b) p. 24376, line 10 at Waliguan in the summer of 2006, 'that' is . . .

c) p. 24376, line 23 is 'slightly' lower than. . .

d) p. 24381, line 21 by 36-42 % (Fig. '7').

e) p. 24381, line 24 Figure '7' shows that . . .

f) p. 24384, line 13 particle 'aging' and formation . . .

g) p. 24385, line 15 and 'aging' processes of. . .

h) p.24392, Figure 1 caption "Topographical map showing the the sampling location . . ." Delete the second "the".

Response 20: We have revised as follows:

a) Revised "diming" to "dimming" in line 3 of p. 24371

b) We added "that" in the sentence as follows:

That is the site of the observation station of the World Meteorological Organization's (WMO) Global Atmospheric Watch (GAW) (Xue et al., 2011)

c) We revised "slighter" to "slightly".

d) We revised it like this: "Inclusions within SIA particles increase their size by 36-42% (Figure 7)."

e) We revised the number of Figure as follows:

"Figure 7 shows that the number of particles with inclusions increases substantially with diameters above 200 nm."

f) We revised "ageing" to "aging" in the line 13 of p. 24384.

g) We revised “ageing” to “aging” in the line 15 of p. 24385.

h) We deleted the second “the”, the sentence is as follows:

“Topographical map showing the sampling location and surrounding regions in the Tibetan Plateau.”