

MS NO.: acp-2015-437

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:

1. 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.”

2. 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”.

3. 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/cm^3 .

4. p. 24375, line 2-6 “Additionally we know the relation . . . diameter smaller than $1 \mu\text{m}$.” 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 $\text{ESD}=0.64\text{ECD}$.” 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 $1 \mu\text{m}$ ”.

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 $\text{EVD}=0.64\text{ECD}$. 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 $1 \mu\text{m}$ where the correlation between the ECD and EVD is especially good (Fig.2).”

5. 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.

6. 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.”

7. 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 $>1\mu\text{m}$), 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 $>1\mu\text{m}$), it is unlikely that they originated from natural sources such as saline Qinghai Lake and desert.”

8. 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 PM_{2.5} in 11-15 October (Du et al., 2015).”

9. 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.

10. 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.

11. 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.”

12. 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.”

13. 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.

14. 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.”

15. 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.

16. 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).”

17. 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.

18. 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.

19. 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.

20. 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.”