Interactive comment on “Mixing state, composition, and sources of fine aerosol particles in the Qinghai-Tibetan Plateau and the influence of agricultural biomass burning” by W. J. Li et al.

Anonymous Referee #2

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General comments:

This paper presents detailed characteristics of fine (<1 µm) aerosol particles collected at a background site (grassland) in the Tibetan Plateau (TP). As the authors mention, information of atmospheric aerosol particles in the remote inland at high elevation has been scarce. In my opinion, the size distributions and particle compositions obtained by analyzing thousands of particles are significant results and worth publishing. The present paper is a revised version of the first manuscript to which I requested major reorganization and rewriting. It has been significantly improved, but I still find points to be corrected and clarified as listed below. Although the points are mostly minor (mainly about writing), a few are related with the authors’ interpretations and may require significant revisions.

Specific comments:

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. 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. On the contrary, “medium size” in the line 9 of p. 24385 should be “median size”.

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.

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.” Are the pressure and temperature typical at the sampling site? Also, what kind of particle is assumed that has density of 2 g/cm3?

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”.

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).

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.

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 µm), it is unlikely that they originated from natural sources such as saline Qinghai Lake and desert.”

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.

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?

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?

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”?

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.

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?

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

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?
Thirdly, the dominant organics, ... in fine particles.” I don’t get the meaning of this sentence. Please rewrite.

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.

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?

Some of the letters in the figure would be difficult to be read when printed on paper. Enlarge.

Technical corrections:

the brightening and ‘dimming’ phenomenon

that is ...

slightly’ lower than ...

by 36-42 % (Fig. ‘7’).

Figure ‘7’ shows that ...

particle ‘aging’ and formation ...

and ‘aging’ processes of ...

Delete the second “the”.

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