

Response to reviewer

We are very grateful for the reviewer's critical comments and suggestions, which have helped us improve the paper quality substantially. We have addressed all of the comments carefully as detailed below in our point-by-point responses. Our responses start with "R:".

This paper reports BC, ISOC, MD concentrations in TP glaciers using an integrating sphere/integrating sandwich spectrophotometer (ISSW). The data are valuable and I am happy to see these can be published.

R: Thanks for all of the comments. We have carefully responded the following questions and concerns.

Issues:

Comment 1: The manuscript seems to be prepared in 2016. There are very few updated literatures were cited. Since 2017, lots of LAIs (BC, OC, MD) data measured by TOR methods (DRI) in TP glaciers have been published, and assessment of LAIs impacts on surface albedo and glacier melt has been also reported. I encourage the authors to check these references and add necessary discussion on BC and MD loadings, sources and impacts on accelerating glacier melt. Here I list some of new literatures in the region.

1. Li X., S Kang, G. Zhang, B. Que, L. Tripathee, R. Paudyal, Z. Jing, Y. Zhang, F. Yan, G. Li, X. Cui, R. Xu, Z. Hu, C. Li. 2017. Light-absorbing impurities in a southern Tibetan Plateau glacier: Variations and potential impact on snow albedo and radiative forcing. *Atmospheric Research*, 200: 77-87. Doi: 10.1016/j.atmosres.2017.10.002.
2. Li X. F., S. Kang, X. He, B. Qu, L. Tripathee, Z. Jing, R. Paudyal, Y. Li, Y. Zhang, F. Yan, G. Li, C. Li. 2017. Light-absorbing impurities accelerate glacier melt in the Central Tibetan Plateau. *Science of the Total Environment*, 587-588: 482-490. Doi: 10.1016/j.scitotenv.2017.02.169.
3. Zhang Y., S Kang, M. Sprenger, Z. Cong, T. Gao, C. Li, S. Tao, X. Li, X. Zhong, M. Xu, W. Meng, B. Neupane, X. Qin, M. Sillanpää. 2018. Black carbon and mineral dust in snow cover on the Tibetan Plateau. *The Cryosphere*, 12: 413-431. Doi: 10.5194/tc-12-413-2018.

4. Zhang Y. L., S. Kang, C. Li, T. Gao, Z. Cong, M. Sprenger, Y. Liu, X. Li, J. Guo, M. Sillanpää, K. Wang, J. Chen, Y. Li, S. Sun. 2017. Characteristics of black carbon in snow from Laohugou No. 12 glacier on the northern Tibetan Plateau. *Science of the Total Environment*: 607-608: 1237-1249. Doi: 10.1016/j.scitotenv.2017.07.100.
5. Zhang Y.L., S. Kang, Z. Cong, J. Schmale, M. Sprenger, C. Li, W. Yang, T. Gao, M. Sillanpää, X. Li, Y. Liu, P. Chen, X. Zhang. 2017. Light-absorbing impurities enhance glacier albedo reduction in the southeastern Tibetan Plateau. *Journal of Geophysical Research - Atmosphere*, 122. Doi: 10.1002/2016JD026397.
6. Zhang Y.L., S. Kang, M. Xu., M. Sprenger, T. Gao, Z. Cong, C. Li, J. Guo, Z. Xu, Y. Li, G. Li, X. Li, Y. Liu, H. Han. 2017. Lightabsorbing impurities on Keqikaer Glacier in western Tien Shan: concentrations and potential impact on albedo reduction. *Sciences in Cold and Arid Regions*, 9(2): 97-111. Doi: 10.3724/SP.J.1226.2017.00097.
7. Schmale J., M. Flanner, S. Kang, M. Sprenger, Q. Zhang, J. Guo, Y. Li, M. Schwikowski, D. Farinotti. 2017. Modulation of snow reflectance and snowmelt from Central Asian glaciers by anthropogenic black carbon. *Scientific Reports*, 7: 40501. Doi: 10.1038/srep40501.
8. Niu H., S. Kang, Y. Zhang, X. Y. Shi, X. F. Shi, S. Wang, G. Li, X. Yan, T. Pu, Y. He. 2017. Distribution of light-absorbing impurities in snow of glacier on Mt. Yulong, southeastern Tibetan Plateau. *Atmospheric Research*, 197: 474-484. Doi: 10.1016/j.atmosres.2017.07.004.
9. Niu H., S. Kang, X. Shi, R. Paudyal, Y. He, G. Li, S. Wang, T. Pu, X. Shi, 2017. In-situ measurements of light-absorbing impurities in snow of glacier on Mt. Yulong and implications for radiative forcing estimates. *Science of the Total Environment*. 581-582: 848-856. Doi: 10.1016/j.scitotenv.2017.01.032.
10. Ji Z., S. Kang, Q. Zhang, Z. Cong, P. Chen, M. Sillanpää. 2016. Investigation of mineral aerosols radiative effects over High Mountain Asia in 1990–2009 using a regional climate model. *Atmospheric Research*, 178-179: 484-496. Doi : 10.1016/j.atmosres.2016.05.003.
11. Jenkins, M., S. Kaspari, S., S. Kang., B. Grigholm, B., P.A. Mayewski. 2016. Tibetan Plateau Geladaindong black carbon ice core record (1843–1982): Recent increases due to higher emissions and lower snow accumulation. *Advances in Climate Change Research*, 7(3): 132-138. Doi: 10.1016/j.accre.2016.07.002.
12. Yang J., S. Kang, Z. Ji, D. Chen. 2018. Modeling the origin of anthropogenic black carbon and its climatic effect over the Tibetan Plateau and surrounding regions. *Journal of Geophysical Research: Atmospheres*, 123. Doi: 10.1002/2017JD027282.

R: We have updated more relative literatures in the manuscript (included all of the references suggested by the reviewer), which were published since 2017.

Comment 2: Geographical information is poor. Some glacier names are not correct. And sampling method and site should be represented clearly. In a glacier, you can collect samples from surface snow, surface ice and snowpit. This information are absent. I emphasize this because LAIs concentration mainly depends on which kind of

samples collected in the glacier. Usually old snow and ice have higher LAIs concentrations than fresh snow and snowpit (with one or two magnitudes).

R: We checked the glacier names in this study very carefully, and we have corrected the “Yangbajing and Tanggula glaciers” as “Gurenhekou and Xiaodongkemadi glaciers”. In order to better illustrate the glaciers, we added a photograph as revised Figure 2 to show the major feature of the glaciers. Due to the samples were only collected approximately for each six months in the TP glaciers, we note that most of the collecting samples were ice samples, which were less than 1 m from the glacier surface (Details could be found in Table S1). We also agreed that the LAIs concentrations in the surface layer were relatively higher than that in the subsurface layers shown in Figure S2-S6, only except some dirty layer in the subsurface layer in this study.

Comment 3: About the phrases in this manuscript. The author use the term of “Light-absorbing particles” in the title, while in the main text, “particulates” were used. What are the differences between these two phrases? I suggest to use the same one in the whole manuscript. In the main text, the author use the term of “High glacier”, it is not a proper phrase. The author should revise these words. “Cold season and warm season” in the Tibetan Plateau which should be “monsoon season and non-monsoon season”. “soil dust” should be “mineral dust”.

R: We have modified “particulates” as “particles”, and removed the term of “High glacier” throughout the manuscript. We have modified the cold and warm season as “monsoon and non-monsoon season”. We have also changed “soil dust” to “mineral dust”.

Comment 4: Introduction: The authors mentioned “BC, OC and MD contribute to spring snowmelt and surface warming through snow darkening effects (Page 3 Line 19-21)”. Then the authors give the research progress of BC and OC, what is the role of MD in this study? The authors should give their points on MD. Page 4 Line 17-18: Check the recent literatures and the authors should point out the differences/advantages between previous studies and this study. In the Tibetan Plateau, Li et al. (2016) use the dual-carbon isotopes to distinguish the different sources of BC, which is helpful for interpretation of BC sources.

R: We have added the recent literatures in analyzing the properties of MD in the TP glaciers as follows: It is well known that the light absorption by MD is mostly related to iron oxides. For instance, the increased radiation forcing by MD in snow has affected the timing and magnitude of runoff from the Upper Colorado River Basin (Painter et al., 2007, 2010). In addition, we have added the literature of Li et al. (2016) in the introduction suggested by the reviewer as follows.

Recently, Li et al. (2016) exhibited that similar contributions from fossil fuel ($46\pm 11\%$) and biomass ($54\pm 11\%$) combustion of the BC sources based on the dual-carbon isotopes technique from aerosol and snowpit samples in the TP regions.

Reference

- Painter, T. H., Barrett, A. P., Landry, C. C., Neff, J. C., Cassidy, M. P., Lawrence, C. R., McBride, K. E., and Farmer, G. L.: Impact of disturbed desert soils on duration of mountain snow cover, *Geophys. Res. Lett.*, 34, L12502, doi: 10.1029/2007gl030284, 2007.
- Painter, T. H., Deems, J. S., Belnap, J., Hamlet, A. F., Landry, C. C., and Udall, B.: Response of Colorado River runoff to dust radiative forcing in snow, *P. Natl. Acad. Sci. USA*, 107, 17125-17130, 2010.

Comment 5: For the sampling: In the abstract, the author used “~67 snow/ice samples” (Page 2 Line 3), do the authors mean about 67 snow/ice samples or more/less than 67 snow/ice samples? The author can give the exact number of samples. But then in the section 2.5 the author mentioned “189 samples”. Do you mean 67 snowpits? Is these snow samples collect from the accumulation zone of the glaciers? What is the “sites” mean in the main text? (for example in Page 10 Line 11 “site65”). For the same glacier, for example Qiyi glacier, as shown in Table S1, I can’t find any information for the site 1 or site 2? What are the differences between them (which part of the glacier)? And in the main text, did the authors also show the results from surface snow samples? The author needs to clarify type of samples in the section 2.

R: We have revised the sentence more clearly as “We collected 67 ice samples in seven glaciers on the Tibetan Plateau. Then each ice sample was cut vertically into small pieces from the surface to the bottom. Therefore, 189 pieces of the ice samples were analyzed in this study. All of the parameters for each ice samples were listed in Table S1. In order to analyze light absorption of ILAPs in the ice samples more clearly, we arranged seven glaciers from north to south according to their latitude and longitude, and samples in each glacier were sorted by sampling time. Therefore, the

ice samples numbered in chronological order from 1 to 19 was in the Qiyi glacier, while sites 20-22, 23-32, 33-44, 45-49, 50-60, and 61-67 in the Qiumianleiketage, Meikuang, Yuzhufeng, Hariqin, Xiaodongkemadi, and Gurenhekou glaciers, respectively.”

Comment 6: Section 2.1: What is the glacier name at Tanggula Mountains? “Tanggula glacier” is not the exact name of the glacier. The same question for “Yangbajing glacier” (I think it is Gurenhekou glacier).

R: Also see our reply to comment 2. We agree with the reviewer. Therefore, we have updated all of the glacier name very carefully throughout the manuscript, such as changing the “Yangbajing and Tanggula glaciers” to “Gurenhekou and “Xiaodongkemadi glaciers”, respectively.

Comment 7: Section 2.1 and 2.2: When the snow/ice samples were prepared for analysis, what is the procedure on how to get the samples for ISSW analysis and WSOC analysis? What kind of filters and vials you used? Do you have blank samples? And duplicate samples?

R: An integrating sphere/integrating sandwich spectrophotometer (ISSW) instrument that developed by Grenfell et al. (2011) was used to measure the mass mixing ratio of BC in snow by Doherty et al. (2010, 2014) and Wang et al. (2013a). This ISSW spectrophotometer measures the light attenuation spectrum from 400 to 700 nm. The total light attenuation spectrum is extended over the full spectral range by linear extrapolation from 400 to 300 and from 700 to 750 nm (Grenfell et al., 2011). Light attenuation is nominally only sensitive to ILAPs on the filter because of the diffuse radiation field and the sandwich structure of two integrated spheres in the ISSW (Doherty et al., 2014). Meanwhile, to quantify WSOC, about 10 ml of the filter liquor was injected into a total carbon analyzer (TOC-V, Shimadzu) and the method detection limit (MDL) used was $4 \mu\text{g l}^{-1}$ with a precision of $\pm 5\%$ (Cong et al., 2015). For filtrate processing, we used 0.2- μm nuclepore filters, as were used in Doherty et al., (2010, 2014), and Wang et al., (2013). In this study, the transmitted light detected by the system for an ice sample, $S(\lambda)$, are compared with the signal detected for a blank filter, $S_0(\lambda)$, and the relative attenuation (A_{tn}) is expressed as:

$$A_{tn} = \ln [S_0(\lambda)/S(\lambda)]$$

Comment 8: Page 5 Line 16: “0.5-m pure, clean tubes”, what is the material of this tube? What is the diameter of this tube? Do you have any photos provided in the SI?

R: We have corrected the sentences as “The collected ice samples were preserved in 0.5-m pure clean plastic bag with a diameter of 20 cm, and kept frozen at the State Key Laboratory of Cryospheric Sciences, Cold and Arid Regions Environmental and Engineering Research Institute in Lanzhou.”

Comment 9: Page 5 Line 18-20: “was cut vertically into small pieces from”, please indicate a resolution.

R: See our reply to comment 5.

Comment 10: Section 2.3: equation 2 and equation 6, there are many saline lakes on the Tibetan Plateau, Na and K may be affected by the moisture evaporated from these saline lakes. How the authors to eliminate this effects when to discuss the NaSs?

R: Actually, there are several saline lakes on the Tibetan Plateau. In order to avoid the moisture evaporation, all of the ice samples were collected far from the saline lakes and the industrial areas to eliminate this effect.

Comment 11: Section3: The author used “site 65” (Page 10 Line 11) confused me. Do the authors mean for the same glacier, the ILAPs were affected by different sources? (Page10 Line 9-13)

R: We have changed the site number as the ice sample number to illustrated our result more clearly, which is similar with the above comment 5 (Table S1). The ice samples collected in each glacier were marked with same color or labeled with the glacier names in all of the Figures. The sentence has been deleted due to the reconstruction of this manuscript.

Comment 12: Page 11 Line 19-20, this is not a complete sentence.

R: This sentence has been rewritten as “One notable feature is that the highest concentrations of C_{BC}^{max} and ISOC for the surface layer are 1600 ng g⁻¹ and 9160 ng g⁻¹ at site 41.”.

Comment 13: Page 12 Line 25-26: this sentence is contradict with the next sentence

R: The sentence has been rewritten as “At sites 52-54, a notable feature is that the surface mixing ratios of C_{BC}^{est} are significantly larger than those in the sub-surface layers, possibly because of the accumulation of BC via dry/wet deposition on the surface samples.”

Comment 14: Page 14 Line 7-8, biomass burning? Do you mean the agricultural/straw burning?

R: Biomass burning represents an important source of atmospheric air pollutants, and it is mainly the burning of agriculture/straw.

Comment 15: Section 3.6: lack of several important references to discuss the potential sources of BC. Yang et al., 2018, JGR; Li et al., 2016, Nature Communications; Zhang et al., 2018, TC.

R: We have added the recent references in section 3.5 based on the reviewer’s suggestions.

Reference:

- Li, C. L., Bosch, C., Kang, S. C., Andersson, A., Chen, P. F., Zhang, Q. G., Cong, Z. Y., Chen, B., Qin, D. H., and Gustafsson, O.: Sources of black carbon to the Himalayan-Tibetan Plateau glaciers, *Nat. Commun.*, 7, 12574, doi: 10.1038/ncomms12574, 2016.
- Yang J., Kang, S., Ji, Z., Chen, D.: Modeling the origin of anthropogenic black carbon and its climatic effect over the Tibetan Plateau and surrounding regions, *J. Geophys. Res.-Atmos.*, 123, doi: 10.1002/2017JD027282, 2018.
- Zhang Y., Kang, S., Sprenger, M., Cong, Z., Gao, T., Li, C., Tao, S., Li, X., Zhong, X., Xu, M., Meng, W., Neupane, B., Qin, X., Sillanpää, M.: Black carbon and mineral dust in snow cover on the Tibetan Plateau, *The Cryosphere*, 12, 413-431, 2018.

Comment 16: Page17 Line12-14: “originated from the local soil source instead of the biomass burning and industrial pollution than previous studies”. This sentence is contradict with the authors stated above.

R: According to the PMF results, we summarized the second factor as a natural mineral dust, but a notable feature is that the relative high mass loading of C_{BC}^{max} (76.4%) to that of the previously identified mineral dust source as reported by Pu et al. (2017) and Zhang et al. (2013). Indeed, the C_{BC}^{max} is not only attributed to the biomass burning emission, but also associated with the industrial activities associated with the local mineral dust (Bond et al., 2006). Compared with the previous studies, the BC emission in studied TP glaciers are also associated with the local mineral dust source instead of the biomass burning and industrial pollution.

Comment 17: Page17 Lines 20-22: “Qiyi glacier” is located in the northeast TP. In this region (Laohugou glacier), Li et al. (2016) indicated that 67% of BC was from fossil fuel combustion. Your result is different. Why? Potential reasons? I am inclined to the dual carbon isotopes results.

R: Our result is much different with the previous study by Li et al. (2016). The most important reason is that we only separate the possible emission sources of the ILAPs (not only include BC, but also include OC and MD) in seven glaciers (such as biomass burning, industrial pollution and the mineral dust) by using the PMF receptor model. But Li et al. (2016) used the dual-carbon isotopes technique to calculate the attribution of BC concentration by fossil fuel or biomass combustion. However, we thank the reviewer to provide a very useful technique to analyze the fraction of the BC emission sources for the collected glacier samples in our further study.

Comment 18: Page 17 Line 16 to Page 18 Line 11: the authors only give the specific result of each glacier, what are the general characteristics of sources for glacier in south or north TP? The author should supplement the related references to discuss the sources.

R: We have added several related references to compare with our major findings.

Comment 19: Conclusions: in Page 18 Line 22, Page 18 Line 29-30, Page 19 Line 8, the authors mentioned the sources of ILAPs repeat.

R: We reconstructed the conclusion section, and deleted the sentences in Page 18 Line 22, and Page 18 Lines 29-30.

Comment 20: Unit used in this manuscript: Page 7 Lines 1-3, the unit should be “ng ml⁻¹”

R: We have modified “ng/ml” as “ng ml⁻¹”.

Comment 21: Figure 4: Please clear indicate the mean of red line, the blue box, and the upper and bottom black line. Figure 5: the author should define the meaning of QY QM MK YZF TGL HRQ YBJ in the figure. In this figure, the authors can show the average ratios of ISOC to BC. The caption should change Figure 6: the relationships between ISOC and BC rather than ratios?

R: We have defined the error bars shown in Figure 4 are 10th, 25th, median, 75th, and 90th percentiles of the data, while the dot symbol represents the average concentrations of the ILAPs in each glacier. We have adjusted the structure of the article and Figure 5 & 6 has been deleted.

Comment 22: Other comments: Page 7 Line 6: Gao et al. (2003) is not the proper references here. Cong et al., 2010 or Li et al., 2009 may be better. These data were measured used the same equipment in the same institute.

R: We have updated the sentence as “Details on these procedures are given in Li et al. (2009) and Cong et al. (2010)”.

Reference:

Cong, Z. Y., Kang, S. C., Zhang, Y. L., and Li, X. D.: Atmospheric wet deposition of trace elements to central Tibetan Plateau, *Appl. Geochem.*, 25, 1415-1421, 2010.
Li C. L., Kang S. C., Zhang Q.: Elemental composition of Tibetan Plateau top soils and its effect on evaluating atmospheric pollution transport, *Environ. Pollut.*, 157, 8-9, 2009.

Comment 23: Page 3 Lines 4-6, “most negative mass balance ““with the deposition of black carbon”, I don’t believe. Estimates from related studies and simulations, the contribution from BC, OC, MD can reach to about 30%.

R: The sentence has been rewritten as “Ample evidence has indicated that the deposition of insoluble light-absorbing particles (ILAPs) was one of the major factors (up to 30%) to lead the greatest decrease in length and area of negative mass balance in the TP glaciers over the past decade (Xu et al., 2006, 2009a; Yao et al., 2012; Qian et al., 2015; Li et al., 2017)”.

Reference:

- Li X., Kang, S., Zhang, G., Que, B., Tripathee, L., Paudyal, R., Jing, Z., Zhang, Y., Yan, F., Li, G., Cui, X., Xu, R., Hu, Z., Li. C.: Light-absorbing impurities in a southern Tibetan Plateau glacier: Variations and potential impact on snow albedo and radiative forcing, *Atmos., Res.*, 200, 77-87, 2017.
- Qian, Y., Yasunari, T. J., Doherty, S. J., Flanner, M. G., Lau, W. K. M., & Jing, M.: Light-absorbing particles in snow and ice: measurement and modeling of climatic and hydrological impact, *Adv. Atmos. Sci.*, 32, 64-91, 2015.
- Xu, B. Q., Yao, T. D., Liu, X. Q., and Wang, N. L.: Elemental and organic carbon measurements with a two-step heating-gas chromatography system in snow samples from the Tibetan Plateau, *Ann. Glaciol.*, 43, 257-262, 2006.
- Xu, B. Q., Cao, J. J., Hansen, J., Yao, T. D., Joswita, D. R., Wang, N. L., Wu, G. J., Wang, M., Zhao, H. B., Yang, W., Liu, X. Q., and He, J. Q.: Black soot and the survival of Tibetan glaciers, *P. Natl. Acad. Sci. USA*, 106, 22114-22118, 2009a.
- Yao, T. D., Thompson, L., Yang, W., Yu, W. S., Gao, Y., Guo, X. J., Yang, X. X., Duan, K. Q., Zhao, H. B., Xu, B. Q., Pu, J. C., Lu, A. X., Xiang, Y., Kattel, D. B., and Joswiak, D.: Different glacier status with atmospheric circulations in Tibetan Plateau and surroundings, *Nature Climate Change*, 2, 663-667, 2012.

Comment 24: Page 6 Lines 19-20, “Previous from BC, OC and Fe” needs references.

R: The sentence has been rewritten as “Previous studies on these parameters have concluded that ILAPs are primarily derived from BC, OC, and Fe (Qian et al., 2015; Wang et al., 2015; Yasunari et al., 2015; Pu et al., 2017)”.

Reference:

- Pu, W., Wang, X., Wei, H. L., Zhou, Y., Shi, J. S., Hu, Z. Y., Jin, H. C., and Chen, Q. L.: Properties of black carbon and other insoluble light-absorbing particles in seasonal snow of northwestern China, *The Cryosphere*, 11, 1213-1233, 2017.
- Qian, Y., Yasunari, T. J., Doherty, S. J., Flanner, M. G., Lau, W. K. M., Ming, J., Wang, H. L., Wang, M., Warren, S. G., and Zhang, R. D.: Light-absorbing Particles in Snow and Ice: Measurement and Modeling of Climatic and Hydrological impact, *Adv. Atmos. Sci.*, 32, 64-91, 2015.
- Wang, X., Pu, W., Zhang, X. Y., Ren, Y., and Huang, J. P.: Water-soluble ions and trace elements in surface snow and their potential source regions across northeastern China, *Atmos. Environ.*, 114, 57-65, 2015.
- Yasunari, T. J., Koster, R. D., Lau, W. K. M., and Kim, K. M.: Impact of snow darkening via dust, black carbon, and organic carbon on boreal spring climate in the Earth system, *J. Geophys. Res.-Atmos.*, 120, 5485-5503, 2015.

Comment 25: Page 8 Line 23, “elements”? Parameters?

R: The major chemical elements used in this study were shown in Figure 9.

Comment 26: Page 7 Line 5, delete the sentence.

R: We have deleted the sentence.

Comment 27: Page 7 Lines 10-11, the sentence should be insert into the section 2.

R: The sentence has been moved into the section 2.

Comment 28: Page7 Lines 15-16, The samples were collected in “warm season”, then the results were attributed to the warm season? For snowpit, it can contain the cold season and warm season snow (non-monsoon and monsoon snow).

R: Sorry for the misleading, we note that all of the samples collected in Qiyi glacier were collected in the monsoon season. Therefore, we have modified the sentence as “Compared with the other TP glaciers, we noted that the vertical profiles of ILAPs in the Qiyi glacier were collected in the monsoon season from 2014 to 2015 (Table S1).”

Comment 29: Page 18 Line 2-4, “with previous studies”. The reference here is not related to the study area (See Li et al. 2016 NC).

R: We have updated the sentence as “This result is highly consistent with the previous study (Andersson et al., 2015)”.

Reference:

Andersson, A., Deng, J., Du, K., Zheng, M., Yan, C., and Sköld, M.: Regionally-varying combustion sources of the January 2013 severe haze events over eastern China, *Environ. Sci. Technol.*, 49, 2038-2043, 2015.