A point-by-point response to the reviews:

Response to reviewer 1

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 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. Tripatheea, 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 (1843âAŠ1982): 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±11%) and biomass (54±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 relay 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 $\mu g\, l^{-1}$ 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 (Atn) is expressed as:

$$Atn = \ln [S_0(\lambda)/S(\lambda)]$$

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. You 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., Tripatheea, 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., Joswia, 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.

Response to reviewer 2

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 manuscript presents interesting and valuable measurements of light-absorbing particles (LAP) from Tibetan plateau glaciers. The LAP concentrations are surprisingly high, when compared to other studies from the Tibetan plateau and the Himalaya. A strength of the manuscript is the extent of data presented. The amount of work that goes into collecting samples from such remote and harsh locations as the Tibetan plateau often seems to be neglected in the larger community. Here the authors have performed measurements covering large areas that presumably have different sources of LAP, as well as different meteorology affecting the glaciers. Publishing these measurements would be of benefit to the public, and this should be possible after some structural and interpretational changes are made. Along with the suggested changes, the language needs to be reviewed carefully, in order to make sure that future readers will interpret

the claims and results of the paper correctly. See below for both major and specific comments.

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

Issues:

Comment 1: In the section describing the sampled glaciers (2.1) information should be added. It currently lacks crucial information such as: any estimates on the area or volume of the glaciers? Are they 'typical' valley-type glaciers? or what are the general characteristics of the glaciers? Nearby emission sources? Are the glacier fronts heavily debris covered? (See also additional specific comments given below).

R: We have added some information in this section as follows: The Qiyi glacier (39°14' N, 97°45' E) is located in the eastern part of the TP, with an elevation of 6178 m. It is classified bucket-valley glacier according to its shape, and is classified subcontinental glacier according to the physical characteristics of glacier. The Xiaodongkemadi glacier (33°04' N, 92°04' E) is located in the central Qinghai-Tibetan Plateau. It is 2.8 km in length and the average snowline is 5560 m a.s.l. The annual mean air temperature at the equilibrium line altitude is in the range -5~-7℃. The surrounding region is mainly tundra. The Yuzhufeng glacier (35°38' N, 94°13' E) is the highest peak across the eastern Kunlun Mountains, with an elevation of 6178 m. The Meikuang and Qiumianleiketage glaciers are also located over the Kunlun Mountains, and these glaciers have an average altitude of 5100 m and 5500 m a.s.l, respectively. The Meikuang glacier is located in the eastern Kunlun Mountains, where is characterized by alluvial deposits and sand dunes, the glacier covers an area of 1.1 km2, is 1.8 km in length (Xiao et al., 2002). The Qiumianleiketage glacier is located in the Heyuan District of the Nagora River, the largest river in the Qaidam Basin, which originated in the Kunlun Mountains of the Qinghai-Tibet Plateau. The length of the glacier is 2.6 km, and the area is 1.73 $km^2$. The glaciers of Hariqin and Meikuang have similar altitudes but are from different mountains. The Gurenhekou glacier is located on the south-eastern margin of the Nyenchen Tanglha Mountains, and seated about 90 km northwest of Lhasa, the capital city of Tibet (Liang et al., 1995).

Comment 2: In the same section I would like to see more details on the snow sample collection. For example: what was the elevation of sample locations; which ones were snow? ice? In the supplementary material I see what the cold and warm seasons correspond to, but this information is valuable to have in the manuscript itself also (In table 1 possibly).

R: Parts of the detail information of the snow sample collection were listed in the method section in Page 5, Line 25-Page 6, Line 12 (Also see the reply to comment 1). We have submitted this table as Table 1 in the previous manuscript in this journal, and the former editor suggested that this Table should be moved to the supporting information, because it is too long and not appropriated to show in the main manuscript. However, if all of the reviewers and the editor agreed, we can change the Table S1 to Table 1 in the final manuscript.

Comment 3: Although the measurement method for the filters has been used previously, it would be beneficial to have a few words on the principles of the method (in section 2.2), and how measurements were carried out in practice for this manuscript. After reading Doherty et al. (2010; 2014; 2016) it is evident that the instrument has gone through modifications with time. Please provide information on how the instrument used in this manuscript is similar/different compared to Doherty et al. and if it contains the latest updates or not. Also, how did the authors take into account filter samples with a high mineral dust load? With higher load a bias can be introduced to the data (described well in Doherty et al. 2016, doi/10.1002/2015JD024375, and references therein). Further, information about the filters used should be provided since it can make a significant difference on the undercatch (look for example in Doherty et al. 2014).

R: Yes, we have added information on optical analysis as follows: An updated integrating sphere/integrating sandwich spectrophotometer (ISSW) was used to measure the mass mixing ratio of BC in snow, which is similar with the instrument developed by Grenfell et al. (2011). Compared with the ISSW spectrophotometer developed by Grenfell et al. (2011), the major difference is that we used two integrating sphere to calculate the relative attenuation instead of the integrating sandwich diffuser to reduce the diffuse radiation during the measuring process. 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). Briefly, 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 (Atn) is expressed as:

$$Atn = \ln [S_0(\lambda)/S(\lambda)]$$

The following measured parameters included equivalent BC ($C_{BC}^{equiv}$), maximum BC ($C_{BC}^{max}$), estimated BC ($C_{BC}^{est}$), fraction of light absorption by non-BC ILAPs ($f_{non-BC}^{est}$), the non-BC absorption Ångström exponent ($Å_{non\text{-}BC}$) and the absorption Ångström exponent of all ILAPs ($Å_{tot}$), were calculated by using the wavelength dependence of the measured spectral light absorption and by assuming that the MACs of the BC, OC, and Fe are 6.3, 0.3, and 0.9 $m^2$ $g^{-1}$, respectively, at 550 nm and that the absorption Ångström exponents (Å or AAE) for BC, OC, and Fe are 1.1, 6, and 3, respectively (Doherty et al., 2010, 2014; Grenfell et al., 2011; Wang et al., 2013). These parameters are defined as follows:

1. $C_{BC}^{max}$ (ng $g^{-1}$): maximum BC is the maximum possible BC mixing ratio in snow by assuming all light absorption is due to BC at the wavelengths of 650-700 nm.

2. $C_{BC}^{est}$ (ng $g^{-1}$): estimated BC is the estimated snow BC mixing ratio derived by separating the spectrally resolved total light absorption.

3. $C_{BC}^{equiv}$ (ng $g^{-1}$): equivalent BC is the amount of BC that would be needed to produce absorption of solar energy by all insoluble particles in snow for the wavelength-integrated from 300-750 nm.

4. $Å_{tot}$: absorption Ångström exponent is calculated for all insoluble particles deposited on the filter between 450 and 600 nm.

5. $Å_{non\text{-}BC}$: non-BC absorption Ångström exponent is defined as the light absorption by non-BC components of the insoluble particles in snow between 450-600 nm.

6. $f_{non-BC}^{est}$ (%): fraction of light absorption by non-BC light absorbing particles is the integrated absorption due to non-BC light absorbing particles, which is weighted by the down-welling solar flux from snow at the wavelengths of 300-750 nm.

It is well known that the aerosol composition and the size distribution are key parameters that affect the absorption Ångström exponent. Doherty et al. (2010) reported that the value of the absorption Ångström exponent of OC was close to 5, which is consistent with previous studies with values ranging from 4-6 (Kirchstetter et al., 2004). Several studies indicated that the absorption Ångström exponent of mineral dust ranged from 2 to 5 (Fialho et al., 2005; Lafon et al., 2006). The variation in the absorption Ångström exponents for urban and industrial fossil fuel emissions is typically in the range of 1.0-1.5 (Millikan, 1961; Bergstrom et al., 2007), which is slightly lower than that of biomass burning, which primarily falls in the range of 1.5-2.5 (Kirchstetter et al., 2004; Bergstrom et al., 2007). Although the source attribution of the insoluble light-absorbing particles in the samples is not a dominant determinant of the value of the absorption Ångström exponent, fossil fuel burning may have a lower absorption Ångström exponent (<2) than 2-5 (Millikan, 1961; Fialho et al., 2005). In this study, we noted that the absorption Ångström exponent ($Å_{tot}$) is due to a mix state of BC and non-BC impurities on our filters, and the calculations of $Å_{tot}$ and $Å_{non\text{-}BC}$ could be found in the study of Doherty et al. (2014). The OC mixing ratio was also determined according to Eq. (2) in Wang et al. (2013), and the Fe concentration was determined according to the inductively coupled plasma-mass spectrometry (ICP-MS) measurements.

Comment 4: In the current manuscript text the discussion on the Ångström exponent is not sufficient and I'm not entirely sure that the interpretation is correct. For the $Å_{non\text{-}BC}$ fraction, more information in section 2.2 on how it was determined is needed. For the results, I do not think that the differentiation between fossil fuel and biomass burning aerosol can be determined in the way it is done here (combined with the PMF it is possible nonetheless to distinguish fossil vs. biomass burning). I assume the authors already have studied Doherty et al.'s work, but I would urge the authors review it once more (especially Doherty et al. 2014 regarding Ångström) to guide their interpretations and text (and of course reference to what is done the same way).

R: Previous studies indicated that the absorption Ångström exponent is driven primarily by the composition of the aerosol and secondarily by the aerosol size distribution. Therefore, variations in $\mathring{A}_{tot}$ can reflect variations in the source of ILAPs in the snow. Sure, $\mathring{A}_{tot}$ can't distinguish fossil or biomass burning, however, combining with the $C_{BC}^{est}$ and $\mathring{A}_{non\text{-}BC}$, we can get the general status of the BC and non-BC contents in the snow or ice. Therefore, we note the parameter of absorption Ångström exponent is still valuable in this study. But we also reconstructed section 3.1 based on the recent study by Doherty et al. (2014).

Reference:

Doherty, S. J., Dang, C., Hegg, D. A., Zhang, R. D., and Warren, S. G.: Black carbon and other light-absorbing particles in snow of central North America, J. Geophys. Res.-Atmos., 119, 12807-12831, 10.1002/2014JD022350, 2014.

Comment 5: Section 3.1 would benefit from more structure. In its current form, I have a hard time following in a logical order. At the moment it starts with an introduction to the concentrations of LAP in the samples, followed by statements on the Ångström exponent, and then back to more discussion on LAP concentrations. Additionally, it would be valuable to see the results obtained in this study compared in a larger Tibetan/Himalayan perspective with results from other nearby measurements of LAP in snow.

R: In the section 3.1, we firstly analyzed the general description of the total concentration of ILAPs and the absorption Ångström exponent in seven glaciers firstly. Then, we discussed the distribution of ILAPs concentration in each glacier region. We have also reconstructed this section and added more comparison with the other nearby measurements of ILAPs in snow.

Comment 6: Section 3.2 needs to be majorly changed. In the references provided in this manuscript the OC/BC ratio is not used in such a way that the authors here claim. From the ratio it is not (unfortunately) as straight forward to say that a ratio of XX corresponds to the aerosol particles originating from fossil or biomass burning. As an example, what about secondary formation of organics? This section therefore needs further work, or to possibly taken out from the manuscript.

R: We have taken out this section and reconstructed the result section based on the reviewer's suggestion.

Comment 7: Page 3 Lines 4-6: The second sentence of the introduction almost seems contradictory to the following sentence. I believe this contradiction could be removed with careful language editing.

R: Thanks for the reminder. We have converted the relative sentences 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, S., Yao, T., Yang, W., Yu, W., and Zhu, M.: Glacier Energy and Mass Balance in the Inland Tibetan Plateau: Seasonal and Interannual Variability in Relation to Atmospheric Changes, Journal of Geophysical Research: Atmospheres, 10.1029/2017jd028120, 2018.

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, 10.1007/s00376-014-0010-0, 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, Annals of Glaciology, Vol 43, 2006, 43, 257-262, Doi 10.3189/172756406781812122, 2006.

Xu, B. Q., Cao, J. J., Hansen, J., Yao, T. D., Joswia, 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, Proc. Nat. Acad. Sci., 106, 22114-22118, 10.1073/pnas.0910444106, 2009a.

Xu, B. Q., Wang, M., Joswiak, D. R., Cao, J. J., Yao, T. D., Wu, G. J., Yang, W., and Zhao, H. B.: Deposition of anthropogenic aerosols in a southeastern Tibetan glacier, J. Geophys. Res.-Atmos., 114, 10.1029/2008jd011510, 2009b.

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, Nat. Clim. Chang., 2, 663-667, Doi 10.1038/Nclimate1580, 2012.


Comment 8: Page 3 Lines 8-9: This is incorrect use of the Jacobi et al. 2015 reference for that statement.

R: We have deleted the reference.

Comment 9: Page 3 Lines 17-19: Do you mean climate forcing from BC deposition world-wide? Please clarify. Also believe reference should be Bond et al. 2013, not 2014.

R: We have corrected this mistake, and we believed that the climate forcing in here shows the global deposition of black carbon on the snow and sea ice in the industrial era. Therefore, the we have modified the sentence as "Bond et al. (2013) indicated that the best estimate of global climate forcing from BC deposition on snow and sea ice …"

Comment 10: Page 3 Lines 29-2 (page 4): Insoluble organic carbon in previous studies, please provide the references.

R: We have modified the sentence as "Although the mass mixing ratio of insoluble organic carbon in the snow and ice has been widely investigated in previous studies(Xu et al., 2006, 2009; Flanner et al., 2009; Wang et al., 2013), there are still limited studies that measure the mass mixing ratios of both WSOC and ISOC in ice samples, especially across the TP regions (Li et al., 2016; Yan et al., 2016)."

Reference:

Flanner, M. G., Zender, C. S., Hess, P. G., Mahowald, N. M., Painter, T. H., Ramanathan, V., and Rasch, P. J.: Springtime warming and reduced snow cover from carbonaceous particles, Atmos. Chem. Phys., 9, 2481-2497, 2009.

Li, C. L., Chen, P. F., Kang, S. C., Yan, F. P., Li, X. F., Qu, B., and Sillanpaa, M.: Carbonaceous matter deposition in the high glacial regions of the Tibetan Plateau, Atmos. Environ., 141, 203-208, 2016.

Wang, X., Doherty, S. J., and Huang, J. P.: Black carbon and other light-absorbing impurities in snow across Northern China, J. Geophys. Res.-Atmos., 118, 1471-1492, 2013.

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., Joswia, 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, Proc. Nat. Acad. Sci., 106, 22114-22118, 2009.

Yan, F. P., Kang, S. C., Li, C. L., Zhang, Y. L., Qin, X., Li, Y., Zhang, X. P., Hu, Z. F., Chen, P. F., Li, X. F., Qu, B., and Sillanpaa, M.: Concentration, sources and light absorption characteristics of dissolved organic carbon on a medium-sized valley glacier, northern Tibetan Plateau, The Cryosphere, 10, 2611-2621, 2016.

Comment 11: Page 4 Line 3: The abbreviation 'ILAPs' should first be written out.

R: The sentence has been modified as "Due to the importance of the climate effects by insoluble light-absorbing particles (ILAPs), numerous snow surveys have been conducted to investigate the light absorption of ILAPs and their potential source attribution in snow (Clarke and Noone, 1985; Doherty et al., 2010, 2014; Hegg et al., 2010; Huang et al., 2011)".

Reference:

Clarke, A. D., and Noone, K. J.: Soot in the Arctic Snowpack - a Cause for Perturbations in Radiative-Transfer, Atmos. Environ., 19, 2045-2053, 1985.
Doherty, S. J., Dang, C., Hegg, D. A., Zhang, R. D., and Warren, S. G.: Black carbon and other light-absorbing particles in snow of central North America, J. Geophys. Res.-Atmos., 119, 12807-12831, 2014.
Doherty, S. J., Warren, S. G., Grenfell, T. C., Clarke, A. D., and Brandt, R. E.: Light-absorbing impurities in Arctic snow, Atmos. Chem. Phys., 10, 11647-11680, 2010.
Hegg, D. A., Warren, S. G., Grenfell, T. C., Doherty, S. J., and Clarke, A. D.: Sources of light-absorbing aerosol in arctic snow and their seasonal variation, Atmos. Chem. Phys., 10, 10923-10938, 2010.
Huang, J. P., Fu, Q. A., Zhang, W., Wang, X., Zhang, R. D., Ye, H., and Warren, S. G.: Dust and Black Carbon in Seasonal Snow across Northern China, Bull. Amer. Meteor. Soc., 92, 175-181, 2011.

Comment 12: Page 4 Lines 3-6: Did all of these references actually perform source attribution of the ILAP in the snow? If not, please adjust references referred to. Lines 6-9: This sentence needs to be reworked, confusing at the moment.

R: We have carefully checked the references throughout the manuscript, and rewritten the sentence as "Due to the importance of the climate effects by insoluble light-absorbing particles (ILAPs), numerous snow surveys have been conducted to investigate the light absorption of ILAPs and their potential source attribution in snow (Xu et al., 2009a, b; Doherty et al., 2010, 2014; Hegg et al., 2010; Wang et al., 2015; Wang et al., 2017). For instance, Hegg et al. (2009) found out that the light absorption in Arctic snow by ILAPs is mainly originated from two distinct biomass burning sources, a pollution source, and a marine source based on the EPA PMF receptor model."

Reference:

Doherty, S. J., Warren, S. G., Grenfell, T. C., Clarke, A. D., and Brandt, R. E.: Light-absorbing impurities in Arctic snow, Atmos. Chem. Phys., 10, 11647-11680, 2010.
Doherty, S. J., Dang, C., Hegg, D. A., Zhang, R. D., and Warren, S. G.: Black carbon and other light-absorbing particles in snow of central North America, J. Geophys. Res.-Atmos., 119, 12807-12831, 2014.
Hegg, D. A., Warren, S. G., Grenfell, T. C., Doherty, S. J., Larson, T. V., and Clarke, A. D.: Source Attribution of Black Carbon in Arctic Snow, Environ. Sci. Technol., 43, 4016-4021, 2009.
Hegg, D. A., Warren, S. G., Grenfell, T. C., Doherty, S. J., and Clarke, A. D.: Sources of light-absorbing aerosol in arctic snow and their seasonal variation, Atmos. Chem. Phys., 10, 10923-10938, 2010.
Wang, M., Xu, B., Cao, J., Tie, X., Wang, H., Zhang, R., Qian, Y., Rasch, P. J., Zhao, S., Wu, G., Zhao, H., Joswiak, D. R., Li, J., and Xie, Y.: Carbonaceous aerosols recorded in a southeastern Tibetan glacier: analysis of temporal variations and model estimates of sources and radiative forcing, Atmos. Chem. Phys., 15, 1191-1204, 2015.
Wang, X., Pu, W., Ren, Y., Zhang, X., Zhang, X., Shi, J., Jin, H., Dai, M., and Chen, Q.: Observations and model simulations of snow albedo reduction in seasonal snow due to insoluble light-absorbing particles during 2014 Chinese survey, Atmos. Chem. Phys., 17, 2279-2296, 2017.
Xu, B. Q., Cao, J. J., Hansen, J., Yao, T. D., Joswia, 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,

Proc. Nat. Acad. Sci., 106, 22114-22118, 2009a.
Xu, B. Q., Wang, M., Joswiak, D. R., Cao, J. J., Yao, T. D., Wu, G. J., Yang, W., and Zhao, H. B.: Deposition of anthropogenic aerosols in a southeastern Tibetan glacier, J. Geophys. Res.-Atmos., 114, 2009b.

Comment 13: Page 4 Lines 14-16: What did the results of Doherty et al. 2014 show? In the previous sentences you provide some highlight from each study, but not for Doherty et al. Could be useful for readers.

R: Sorry, we have added the results of Doherty et al. (2014), and the sentence has been rewritten as "Recently, vertical profiles of ILAPs in seasonal snow were performed from 67 North American sites, and biomass/biofuel burning, soil and fossil fuel pollution are explored as the major sources of particulate light absorption based on the chemical and optical data, and these were (Doherty et al., 2014)".

Comment 14: Page 4 Lines 17-18: At this time there has been an increasing number of observations on ILAP in Tibetan snow. The other referee provided a comprehensive list on this.

R: We have corrected this mistake.

Comment 15: Page 4 Lines 29-30: Units for the AOD numbers? And for what time period is this? Also, it would be good to include some information on what an AOD number of XX means (e.g. 0.4 meaning high optical depth, etc.)

R: Aerosol optical depth (AOD) is a description of the attenuation of light by aerosol, and it is a dimensionless quantity. Figure 1 showed the spatial distribution of the averaged AOD retrieved from Aqua-MODIS over Tibetan Plateau from 2013 to 2015. According to the reviewer's suggestion, we have added this information to the caption in Figure 1.

Comment 16: Page 5 Lines 1-2: How is the glacier located above the ELA? Lines 2-3: The glacier must encompass an elevation range, and not only located at 5743 m a.s.l. The average snowline is based on what? Reference on this number ? Lines 4-5: Is the Yuzhufeng glacier part of the highest peak? Please clarify.

R: Page 5 Lines 1-2: The sentence has been rewritten as "The Qiyi glacier (39°14' N, 97°45' E) is located in the eastern part of TP, with an elevation of 4850 m".

Lines 2-3: The average snowline is just a general background of Xiaodongkemadi glacier, and doesn't correlate with our study. Therefore, we deleted the sentence.

Lines 4-5: The sentence has been rewritten as "The Yuzhufeng glacier (35°38' N, 94°13' E) is the highest peak across the eastern Kunlun Mountains, with an elevation of 6178 m".

Comment 17: Page 5 Lines 8-9: The surrounding areas characteristics, why is this information important? Is such information available for other glaciers? Line 12: What is the point of the Liang et al. 1995 reference?

R: Page 5 Lines 8-9: this information was used to describe the topography of the Meikuang glacier. The Meikuang glacier is located in the eastern Kunlun Mountains, where is characterized by alluvial deposits and sand dunes (Xiao et al., 2002), meanwhile, ~47.9% of the sources of ILAPs in the Meikuang glacier was from soil dust in the PMF analysis, so there was a good correspondence.

For line 12: It should be a general description to illustrate Yangbajing glacier and we deleted the citation of Liang et al. (1995).

Comment 18: Page 5 Line 15: I don't see in table S1 (or anywhere else) if the sample is ice or snow. Please provide this information. Line 16: What was the volume of the tubes? And what kind of tubes were they?

R: 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).

Line 16: We have modified the sentence as "The collected snow/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 19: Page 5 Lines 18-20: Please provide more information on how the snow/ice samples were cut. After this (and elsewhere in the manuscript) I'm missing some description on the filtering of snow samples. Lines 24-28: This sentence is confusing and needs to be

reworked. I do not find the argument made in the references given, but once this sentence is reorganized it may be more clearly.

R: We collected 67 ice samples in seven glaciers on the Tibetan Plateau, and cut each sample from top to bottom at a length of approximately 10 cm. Finally, we obtained a total of 189 small samples. For the filtration process, we have added the description in section 2.2.

Lines 24-28: The sentence has been rewritten as "The OC mixing ratio was also determined according to Eq. (2) in Wang et al. (2013), and the Fe concentration was determined according to the inductively coupled plasma-mass spectrometry (ICP-MS) measurements".

Comment 20: Page 6 Lines 19-20: What does this opening sentence have to do with the chemical analysis?

R: We have reconstruction the method section, and the sentence in Lines 19-20 has been deleted.

Comment 21: Page 6 Lines 20-23: What does the MACs have to do with the chemical analysis? This information should instead be included in the optical analysis section (2.2.). Line 24: Do you mean 10 mL of water from the filtered meltwater? Please clarify. In addition, a few more words describing the total carbon analyzer would be beneficial. Lines 29-30: Was it the filtered meltwater again that was analyzed for major metallic elements?

R: Page 6 Line 20-23: We have moved this information into section 2.2.

Line 24: 10 ml refers to the amount of sample solution after filtration that used to measure the WSOC concentration, and more descriptions of the total carbon analyzer were described by Cong et al. (2015).

Lines 29-30: We used the sample solution before filtration when analyzed the major metallic elements.

Comment 22: Page 7 Line 3-4: Did you acidify all samples or not? I find it confusing with the opening of the sentence 'generally speaking'. Line 5: Measurement precision

range, do you mean ±?

R: No, for each ice sample, we acidified all samples before filtration and then used ICP-MS to measure the trace element contents. Meanwhile, we deleted "generally speaking".

Line 5: The relative deviation between most of the measured values of the elements and the standard reference values is within ±10%.

Comment 23: Page 8 Line 6: What are EFc ? Please write out the abbreviation. Line 25: What does *Q* values stand for?

R: We have changed "EFc" to "EF", which is consistent with the caption of section 2.4. EF is the Enrichment factor defined as the concentration ratio of a given metal to that of Al. It is a reliable measure of crustal dust, normalized to the same concentration ratio characteristic of the upper continental crust (Wedepohl, 1995), calculated with the following equation:

$$EF=\frac{(X/Al)_{snow}}{(X/Al)_{crust}}$$

Line 25: Q is the goodness of fit parameter calculated excluding points not fit by the model, which is defined as samples for which the uncertainty-scaled residual is greater than 4. The best solution in PMF is typically identified by the lowest Q value along with the path and may be imagined as the bottom of a trough in the multidimensional space.

Comment 24: Page 9 Line 1: Should not this section be referred to as Results and Discussion? And not only Results as it is now. Line 4: I'm confused with the number of samples. Here you state over 67 and in section 2.1 ~67. Please clarify. Line 5: The second sentence of section 3.1 is vague and not necessary for this section. As mentioned previously, please elaborate on this in the methods section.

R: We have changed "Results" to "Results and Discussion".

Line 4: The number of samples is 67 and we have modified them.

Line 5: We moved the sentence into the method section, and reconstructed the method section.

Comment 25: Page 9 Lines 7-10: Here the authors provide a range for the higher values, what is the range for the lower values? Lines 15-16: This is interesting that there is no difference between seasons since several other authors have observed the contrary. This should be elaborated on in the revised manuscript. Lines 17-29: These sentences should rather be included in the methods section, assisting with data interpretation.

R: According to the Table 1, the range for the lower median values of $C_{BC}^{est}$ is 23-53 ng $g^{-1}$ in the Xiaodongkemadi, Hariqin, and Gurenhekou glaciers.

Yes, we found that there is no apparent difference of ILAPs in each glacier between monsoon and non-monsoon seasons in these glacier regions. We noted several reasons could lead this discrepancy.

First of all, all of the ice samples were collected in an individual time. Another major issue is that except the long-range transport of ILAPs, local air pollutants can also affect the ILAPs in the glacier via wet and dry deposition. For instance, Li et al. (2016) indicated that the Fossil fuel contributions of BC is much higher in the Laohugou No. 12 glacier (close to Qiyi glacier) due to human activities. Finally, based the study by Xu et al. (2009), the ILAPs in the TP glacier also can be affected by the European air to reach that location on the eastern plateau, and thus lead the greater proportion of the heavy leading of the BC in the glacier. For instance, the mass concentration of BC in the Muztagh Ata and Xiaodongkemadi glaciers in northern part of Tibetan Plateau is much higher than that in the East Rongbuk, Noijin Kangsang and Zuoqiupo glacier, which are located in the southern part of Tibetan Plateau. Above all, we noted that the reasons could be very complicated, and the possible explanation has been added in the result section in Page 13, Line 12-23.

Lines 17-29：We have moved the sentences into the method section based on the reviewer's suggestion.

Comment 26: Page 10 Lines 2-5: Low $Å_{tot}$ means that the LAP originated from combustion sources? Lines 16-21: I find it confusing with these couple of sentences here as the previous sentences discusses $Å_{non\text{-}BC}$, and the sentences before that $Å_{tot}$. First discussion on $Å_{tot}$, and then into $Å_{non\text{-}BC}$. Lines 27-29: I do not understand this reasoning and how the sentences leading up to this reasoning support it. Line 29-30: Section 3.1

started with introducing the LAP concentrations and now they come back continued in the following sentences. The structure of this section would be better by having the concentration discussion intact.

R: Based on previous studies, the variation in the absorption Ångström exponents for urban and industrial fossil fuel emissions is typically in the range of 1.0-1.5 (Millikan, 1961; Bergstrom et al., 2007), which is slightly lower than that of biomass burning ranging from 1.5 to 2.5 (Kirchstetter et al., 2004; Bergstrom et al., 2007). Meanwhile, the lower median values of the absorption Ångström exponents in Xiaodongkemadi glacier is about 2.1, so the results indicated that the emission of the ILAPs in Xiaodongkemadi glacier likely originated from the combustion sources.

Lines 27-29: We have deleted the sentence.

Lines 29-30: See our reply to comment 5.

Comment 27: Page 11 Line 2: What do you mean by individual period? Lines 4-5: How is a decreasing trend observed? Please include information in the manuscript that show the trend. Line 7: How is that due to heavy human activities? Lines 5-8: Please check this sentence structure. Line 8-9: In what sense is there good agreement with Ming et al. 2013? Line 16: What does the sample depth range presented here correspond to? Line 20: In table S1 I find ISOC to be 9.16 ppm, not 8600 ng $g^{-1}$.

R: As shown in Table S1, the sampling time for each sample in seven glaciers was different, so we noted that all of the ice samples were collected in the individual period from 2013 to 2015.

Lines 4-5: The sentences have been modified as "Based on our sampling locations shown in Figure 1, the Qiyi, Qiumianleiketage, Meikuang, Yuzhufeng glaciers were located in the northern part of Tibetan Plateau, while Xiaodongkemadi, Hariqin, Gurenhekou glaciers were located in the southern part of Tibetan Plateau. As shown in Figure 4, the median values of the $C_{BC}^{est}$ and $C_{ISOC}$ obviously showed a significant decreasing trend from the northern TP to the southern TP."

Lines 5-8: The sentence has been rewritten as "Comparing with Hariqin, Xiaodongkemadi and Gurenhekou glaciers, the relative higher values of the $C_{BC}^{est}$ and

$C_{ISOC}$ in Qiyi, Qiumianleiketage, Yuzhufeng, and Meikuang glaciers are mainly influenced by human activities."

Lines 8-9: We have modified the sentence as "The mass concentration of BC in northern TP glaciers was higher than that in southern TP glaciers, which shows a good agreement with Ming et al. (2013)."

Line 16: We have rewritten this sentence more clearly as "The depth of these ice samples collected in Yuzhufeng glacier is ranging from 15 to 45 cm as Table S1 shown."

Line 20: We have corrected this mistake.

Reference:

Ming, J., Xiao, C. D., Du, Z. C., and Yang, X. G.: An overview of black carbon deposition in High Asia glaciers and its impacts on radiation balance, Adv. Water Resour., 55, 80-87, 2013.

Comment 28: Page 12 Lines 5-6: The previous sentences suggest that it is not similar emission sources. Please clarify. Lines 6-8: A similar statement has been made earlier in the manuscript. Please combine these observations. Line 17-18: Do you mean increased from the bottom to the top? Lines 20-21: How were the samples more complicated? Line 26-28: Would it not lead to lower LAP concentrations at the surface the way this sentence describes it now? with LAP being scavenged with meltwater?

R: We agreed with the reviewer, and deleted the sentences in Page 12 Lines 5-6.

Lines 6-8: The similar statement in the earlier manuscript has been deleted.

Lines 17-18: Except sites 7, 8, and 9, the mixing ratios of ILAPs in the ice samples increased remarkably from the top to the bottom from the Figure S4.

Lines 20-21: Comparing with the other glaciers, there is no significant trend of the vertical profile in the mixing ratios of ILAPs in Xiaodongkemadi glacier (see Figure S5), so we indicated that the vertical profiles of the mass mixing ratios of BC, ISOC, and Fe for the ice samples in Xiaodongkemadi glacier were more complicated than those in the other regions.

Lines 26-28: During snow melting process, black carbon (BC) and other insoluble light-absorbing particulate impurities (ILAPs) were retained at the snow surface, because

their scavenging efficiency with meltwater was <100%. Therefore, the mass concentrations of ILAPs in surface snow increased with snow melting (Doherty et al., 2013).

Reference:

Doherty, S. J., Grenfell, T. C., Forsström, S., Hegg, D. L., Brandt, R. E., and Warren, S. G.: Observed vertical redistribution of black carbon and other insoluble light‐absorbing particles in melting snow, J. Geophys. Res.-Atmos., 118, 5553-5569, 2013.

Comment 29: Page 13 Line 6-7: This is the wrong Conway et al reference, should be 1996 instead of 2002. Please check this also in the reference list.

R: Sorry, we have modified the reference as follows: "Previous studies have also illustrated that the ILAPs could become trapped and integrated at the surface of the snowpack due to melting and sublimation to enrich the surface concentrations (Conway et al., 1996; Painter et al., 2012; Doherty et al., 2013)".

Reference:

Conway, J. H., Hardin, R. H., and Sloane, N. J. A.: Packings in Grassmannian spaces, Experimental mathematics, 5, 139-159, 1996.
Doherty, S. J., Grenfell, T. C., Forsström, S., Hegg, D. L., Brandt, R. E., and Warren, S. G.: Observed vertical redistribution of black carbon and other insoluble light‐absorbing particles in melting snow, J. Geophys. Res.-Atmos., 118, 5553-5569, 2013.
Painter, T. H., Bryant, A. C., and Skiles, S. M.: Radiative forcing by light absorbing impurities in snow from MODIS surface reflectance data, Geophys. Res. Lett., 39, L17502, doi: 10.1029/2012gl052457, 2012.

Comment 30: Page 14 Lines 15-17: Please provide a reference for this. Lines 17-19: I do not find this to be the case in given reference. Lines 23-27: How is that?

R: We have provided a reference in Lines 15-17, and modified the reference in Lines 17-19. Then, the sentences have been rewritten as "There is a very large fraction representing the average WSOC (>80%) to TOC, which can absorb solar light and enhance cloud formation through their direct and indirect climate effects (Ram et al., 2010). These results are highly consistent with previous study showing that fossil fuel combustion plays a key role in leading to the higher fraction of WSOC to total organic carbon (TOC) (Zhang et al., 2012)".

Reference:

Ram, K., Sarin, M. M., Strawa, A. W., Kirchstetter, T. W., and Puxbaum, H.: Spatio-temporal variability in atmospheric abundances of ec, oc and wsoc over northern india, J. Aerosol Sci., 41, 88-98, 2010.
Zhang, X., Liu, Z., Hecobian, A., Zheng, M., Frank, N. H., and Edgerton, E. S.: Spatial and seasonal variations of fine particle water-soluble organic carbon (wsoc) over the southeastern united states: implications for secondary organic aerosol formation, Atmos. Chem. Phys., 12, 6593-6607, 2012.

Comment 31: Page 15 Lines 1-2: Is this a general statement or results of this paper? My guess is the former, and if it is that, I would place this statement in the introduction of this paper. Line 13-15: That there is a small contribution from dust is, to my knowledge, not consistent with that results presented earlier in the manuscript. Please clarify this.

R: Yes, it's just a general statement, and we have moved this sentence into the introduction section. In the former section, we analyzed the fraction of BC and non-BC contents in the ice samples. We should note that the non-BC contents are not only include OC, but also include MD. However, we demonstrated the median fraction of light absorption by ILAPs is only ~13% due to MD based on the PMF receptor model in section 3.4. As a result, we indicated that there is no conflict between our major results.

Comment 32: Page 16 Lines 11-12: What do you mean 'for the anthropogenic emission source'? I find this sentence confusing.

R: Except the natural dust source, we referred to attribute the major industrial pollution and biomass burning sources to the anthropogenic emission sources, so we noted that the natural dust source and anthropogenic emission source are both non-negligible to the light absorption by ILAPs in the TP glaciers according to the previous analysis.

As a last note, I wanted to comment on the in-text citations, are they done by year or alphabetical order? I did not find an order to this. Please check this throughout your manuscript.

R: We have arranged the in-text citations in chronological order throughout the manuscript.

# Quantifying light absorption and its source attribution of insoluble light-absorbing particles in Tibetan Plateau glaciers from 2013-2015

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**Abstract.** Amounts of insoluble light-absorbing particles (ILAPs) deposited on the surface of snow and ice can significantly reduce the snow albedo and accelerate the snow melting process. In this study, 67 ice samples were collected in 7 high mountain glaciers over the Tibetan Plateau (TP) regions from May 2013 to October 2015. The mixing ratio of black carbon (BC), organic carbon (OC), and mineral dust (MD) was measured using an integrating sphere/integrating sandwich spectrophotometer (ISSW) system associated with the chemical analysis by assuming the light absorption of mineral dust due to iron oxide. The results indicate that mass mixing ratios of BC, ISOC, and MD show a large variation of 10-3100 ng $g^{-1}$, 10-17000 ng $g^{-1}$, 10-3500 ng $g^{-1}$, with a mean value of 218±397 ng $g^{-1}$, 1357±2417 ng $g^{-1}$, 241±452 ng $g^{-1}$ on TP glaciers during the entire ice field campaign, respectively. The chemical elements and the selected carbonaceous particles were also analyzed of the attributions of the particulate light absorption based on a positive matrix factorization (PMF) receptor model. On average, the industrial pollution (33.1%), biomass/biofuel burning (29.4%), and mineral dust (37.5%) were the major sources of the ILAPs in TP glaciers. Although the mineral dust assumed to be the highest contributor to the mass loading of ILAPs, we noted that the averaged light absorption of BC (50.7%) and ISOC (33.2%) was largely responsible for the measured light absorption in the high mountain glaciers at the wavelengths of 450-600 nm.

## 1 Introduction

The Tibetan Plateau (TP), known as the highest plateau in the world, and its surrounding areas contain the largest snow and ice mass outside the polar regions (Qin et al., 2006). 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). The unusual increase in temperature over the TP is now considered one of the major contributors to glacial shrinkage (Ding et al., 2006). Climate models indicated that black carbon (BC) heats the troposphere by absorbing solar radiation (Jacobson, 2001; Ji et al., 2016; Yang et al., 2017), and BC reduces snow and ice albedos when it is deposited on their surface, thus leading to the acceleration of snowmelt ( Hansen and Nazarenko, 2004; Flanner et al., 2007, 2009; Hadley and Kirchstetter, 2012; Yasunari et al., 2015; Schmale et al., 2017; Zhang et al., 2017, 2018). For example, a mixing ratio of 10 ng $g^{-1}$ of BC in snow can reduce snow albedo by 1%, which has a similar effect to that of 500 ng $g^{-1}$ of mineral dust on the albedo of snow and ice at 500 nm wavelength (Warren and Wiscombe, 1980; Warren, 1982; Wang et al., 2017).

Recently, Li et al. (2016) exhibited that similar contributions from fossil fuel (46±11%) and biomass (54±11%) combustion of the BC sources based on the dual-carbon isotopes technique from aerosol and snowpit samples in the TP regions. Bond et al. (2013) indicated that the best estimate of climate forcing from BC deposition on snow and sea ice in the industrial era is +0.13 W $m^{-2}$ with 90% uncertainty bounds of +0.04 to +0.33 W $m^{-2}$. In addition to BC, organic carbon (OC) and mineral dust (MD) also substantially contribute to springtime snowmelt and surface warming through snow darkening effects (Painter et al., 2010; Huang et al., 2011; Painter et al., 2012; Wang et al., 2013;Kaspari et al., 2014; Wang et al., 2014; Yasunari et al., 2015). Water-soluble organic carbon (WSOC) and insoluble organic carbon (ISOC) are the major components of organic carbon in the atmosphere, snow and sea ice. In addition to the strong light absorption of ISOC and WSOC may also influence the regional and global climate through the heating and evaporating of clouds and by acting as cloud condensation nuclei (CCN) (Chen and Bond,

2010; Alexander et al., 2012; Witkowska and Lewandowska, 2016). Moreover, WSOC also plays a key role in affecting human health due to its toxic effects (McConnell and Edwards, 2008; Wang et al., 2015). Although the mass mixing ratio of insoluble organic carbon in the snow and ice has been widely investigated in previous studies (Xu et al., 2006, 2009; Flanner et al., 2009; Wang et al., 2013), there are still limited studies that measure the mass mixing ratios of both WSOC and ISOC in ice samples, especially across the TP regions (Li et al., 2016; Yan et al., 2016). 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).Due to the importance of the climate effects by insoluble light-absorbing particles (ILAPs), numerous snow surveys have been conducted to investigate the light absorption of ILAPs and their potential source attribution in snow (Xu et al., 2009a, b; Doherty et al., 2010, 2014; Hegg et al., 2010; Wang et al., 2015; Jenkins et al., 2016; Niu et al., 2017; Wang et al., 2017). For instance, Hegg et al. (2009) found out that the light absorption in Arctic snow by ILAPs is mainly originated from two distinct biomass burning sources, a pollution source, and a marine source based on the EPA PMF receptor model. Huang et al. (2011) conducted the first snow survey over northern China, and the sources of ILAPs in seasonal snow in the region were explored based on a positive matrix factorization (PMF) with backward trajectory cluster analysis (Zhang et al., 2013a). Wang et al. (2013) indicated that soil dust was found to be the major contributor to snow particulate absorption in Inner Mongolia regions and Qilian mountains over northern China. Recently, vertical profiles of ILAPs in seasonal snow were performed from 67 North American sites, and biomass/biofuel burning, soil and fossil fuel pollution are explored as the major sources of particulate light absorption based on the chemical and optical data, and these were (Doherty et al., 2014). However, the assessments of the light absorption and its emission sources of ILAPs on the TP glaciers are sparse due to limited observations. Here, we present a snow survey on collecting the ice samples on 7 high mountain glaciers on the TP regions from 2013-2015. By using an integrating sphere/integrating sandwich spectrophotometer (ISSW) system associated with the chemical analysis, the particulate light absorption of

BC, ISOC, and MD in TP glaciers was evaluated. Finally, the relative attribution of emission sources of the ILAPs in these regions was explored based on a positive matrix factorization (PMF) receptor model.

## 2 Site description and methods

### 2.1 Site description and sample collection

As shown in Fig. 1, the spatial distribution of aerosol optical depth (AOD) retrieved from Moderate-resolution Imaging Spectrometer (MODIS) sensors are ranging from 0.1 to 0.4 from south to north near the high mountain glacier regions over TP regions from 2013-2015. The Qiyi glacier (39°14' N, 97°45' E) is located in the eastern part of the TP, with an elevation of 6178 m. It is classified bucket-valley glaciers according to its shape, and is classified subcontinental glacier according to the physical characteristics of glacier. The Xiaodongkemadi glacier (33°04' N, 92°04' E) is located in the central Qinghai-Tibetan Plateau. It is 2.8 km in length and the average snowline is 5560 m a.s.l. The annual mean air temperature at the equilibrium line altitude is in the range -5~-7℃. The surrounding region is mainly tundra. The Yuzhufeng glacier (35°38' N, 94°13' E) is the highest peak across the eastern Kunlun Mountains, with an elevation of 6178 m. The Meikuang and Qiumianleiketage glaciers are also located over the Kunlun Mountains, and these glaciers have an average altitude of 5100 m and 5500 m a.s.l, respectively. The Meikuang glacier is located in the eastern Kunlun Mountains, where is characterized by alluvial deposits and sand dunes, the glacier covers an area of 1.1 $km^2$, is 1.8 km in length (Xiao et al., 2002). The Qiumianleiketage glacier is located in the Heyuan District of the Nagora River, the largest river in the Qaidam Basin, which originated in the Kunlun Mountains of the Qinghai-Tibet Plateau. The length of the glacier is 2.6 km, the area is 1.73 $km^2$. The glaciers of Hariqin and Meikuang have similar altitudes but are from different mountains. The Gurenhekou glacier is located on the south-eastern margin of the Nyenchen Tanglha Mountains, and seated about 90 km northwest of Lhasa, the capital city of Tibet (Liang et al., 1995). To investigate the enrichment of ILAPs via wet and dry deposition on glaciers, we collected 67 ice samples in seven glaciers on the Tibetan Plateau from May 2013 to October 2015 (Fig. 2). 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. 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. In order to analyze light absorption by ILAPs in the ice samples 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. The ice samples were filtered at laboratories in Lanzhou University. The samples were quickly melted and then immediately filtered through a 0.2-μm Nuclepore filter. Additional details on the ice filtration processes have been previously reported (Doherty et al., 2010, 2014; Wang et al., 2013).

## 2.2 Optical analysis

An updated integrating sphere/integrating sandwich spectrophotometer (ISSW) is used to calculate the mass mixing ratio of BC in snow, which is similar with the instrument developed by Grenfell et al. (2011). Compared with the ISSW spectrophotometer developed by Grenfell et al. (2011), the major difference is that we used two integrating sphere to calculate the relative attenuation instead of the integrating sandwich diffuser to reduce the diffuse radiation during the measuring process. 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). Briefly, 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 (Atn) is expressed as:

$$Atn=\ln[S_0(\lambda)/S(\lambda)] \quad (1)$$

The following measured parameters included equivalent BC ($C_{BC}^{equiv}$), maximum BC ($C_{BC}^{max}$), estimated BC ($C_{BC}^{est}$), fraction of light absorption by non-BC ILAPs ($f_{non-BC}^{est}$), the non-BC absorption Ångström exponent ($Å_{non-BC}$) and the absorption Ångström exponent of all ILAPs ($Å_{tot}$), were calculated by using the wavelength dependence of the measured spectral light absorption and by assuming that the MACs of the BC, OC, and Fe are 6.3, 0.3, and 0.9 $m^2\ g^{-1}$, respectively, at 550 nm and that the absorption Ångström exponents (Å or AAE) for BC, OC, and Fe are 1.1, 6, and 3, respectively (Doherty et al., 2010, 2014; Grenfell et al., 2011; Wang et al., 2013). These parameters are defined as follows:

1. $C_{BC}^{max}$ (ng $g^{-1}$): *maximum BC* is the maximum possible BC mixing ratio in snow by assuming all light absorption is due to BC at the wavelengths of 650-700 nm.

2. $C_{BC}^{est}$ (ng $g^{-1}$): *estimated BC* is the estimated snow BC mixing ratio derived by separating the spectrally resolved total light absorption.

3. $C_{BC}^{equiv}$ (ng $g^{-1}$): *equivalent BC* is the amount of BC that would be needed to produce absorption of solar energy by all insoluble particles in snow for the wavelength-integrated from 300-750 nm.

4. $Å_{tot}$: *absorption Ångström exponent* is calculated for all insoluble particles deposited on the filter between 450 and 600 nm.

5. $Å_{non-BC}$: *non-BC absorption Ångström exponent* is defined as the light absorption by non-BC components of the insoluble particles in snow between 450-600 nm.

6. $f_{non-BC}^{est}$ (%): *fraction of light absorption by non-BC light absorbing particles* is the integrated absorption due to non-BC light absorbing particles, which is weighted by the down-welling solar flux from snow at the wavelengths of 300-750 nm.

It is well known that the aerosol composition and the size distribution are key parameters that affect the absorption Ångström exponent. Doherty et al. (2010) reported that the value of the absorption Ångström exponent of OC was close to 5, which is consistent with previous studies with values ranging from 4-6 (Kirchstetter et al., 2004). Several studies indicated that the absorption Ångström exponent of mineral dust ranged from 2 to 5 (Fialho et al., 2005; Lafon et al., 2006). The variation in the absorption Ångström exponents for urban and industrial fossil fuel emissions is typically in the range of 1.0-1.5

(Millikan, 1961; Bergstrom et al., 2007), which is slightly lower than that of biomass burning, which primarily falls in the range of 1.5-2.5 (Kirchstetter et al., 2004; Bergstrom et al., 2007). Although the source attribution of the insoluble light-absorbing particles in the samples is not a dominant determinant of the value of the absorption Ångström exponent, fossil fuel burning may have a lower absorption Ångström exponent (<2) than 2-5 (Millikan, 1961; Fialho et al., 2005). In this study, we noted that the absorption Ångström exponent ($Å_{tot}$) is due to a mix state of BC and non-BC impurities on our filters, and the calculations of $Å_{tot}$ and $Å_{non\text{-}BC}$ could be found in the study of Doherty et al. (2014). The OC mixing ratio was also determined according to Eq. (2) in Wang et al. (2013), and the Fe concentration was determined according to the inductively coupled plasma-mass spectrometry (ICP-MS) measurements.

### 2.3 Chemical analysis

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). Meanwhile, to quantify WSOC, about 10 ml of the filter liquor was injected into a total carbon analyzer (TOC-V, Shimadzu). The method detection limit (MDL) used was 4 $\mu g\, l^{-1}$ with a precision of ±5% (Cong et al., 2015). The definition of TOC (Total Organic Carbon) in this study is calculated as the total WSOC measured by carbon analyzer and the ISOC calculated by ISSW instrument.

The major metallic elements (Al, Cr, Mn, Fe, Ni, Cu, Zn, Cd, Pb) were analyzed by an inductively coupled plasma-mass spectrometry (ICP-MS, X-7 Thermo Elemental) at the Institute of Tibetan Plateau Research in Beijing. The detection limits are Al, 0.238 ng $ml^{-1}$; Cr, 0.075 ng $ml^{-1}$; Mn, 0.006 ng $ml^{-1}$; Fe, 4.146 ng $ml^{-1}$; Ni, 0.049 ng $ml^{-1}$; Cu, 0.054 ng $ml^{-1}$; Zn, 0.049 ng $ml^{-1}$; Cd, 0.002 ng $ml^{-1}$; Pb, 0.002 ng $ml^{-1}$. Generally speaking, we acidified all ice samples to pH<2 with ultra-pure $HNO_3$, then let settle for 48h. The instrument adopted national standard materials and standard materials of the United States as the quality monitoring samples. The relative deviation between most of the measured values of the elements and the standard reference values is within 10%. Details on these procedures are given in Li et al. (2009) and Cong et al. (2010).

Meanwhile, for the filtrated ice samples, we measured the major anions ($Cl^-$,

$NO_2^-$, $NO_3^-$, $SO_4^{2-}$) and cations ($Na^+$, $NH_4^+$, $K^+$, $Mg^{2+}$, $Ca^{2+}$) with an ion chromatograph using a CS12 column for cations and an AS11 column for anions at the Institute of Tibetan Plateau Research in Beijing. All the detection limit of the ions was 1 $\mu g\,l^{-1}$. In addition, except for the anions and cations and trace elements, $CL_{salt}$, MD and biosmoke K ($K_{Biosmoke}$) were determined to assess the mass contributions of the major components in the ice samples. $CL_{salt}$ was estimated as follows in accordance with Pio et al. (2007), by adding to sodium, chloride, and sea-salt contributions of magnesium, calcium, potassium, and sulfate, as follows:

$$CL_{salt}=Na^+_{Ss}+Cl^-+Mg^{2+}_{S_s}+Ca^{2+}_{S_s}+K^+_{S_s}+SO^{2-}_{4S_s}$$

$$=Na^+_{Ss}+Cl^-+0.12Na^+_{Ss}+0.038Na^+_{Ss}+0.038Na^+_{Ss}+0.25Na^+_{Ss} \quad (2)$$

$$Na_{Ss}= Na_{Total}-Al\cdot(Na/Al)_{Crust} \quad (3)$$

Where $(Na/Al)_{Crust}=0.33$, and represents the Na/Al ratio in the dust materials (Wedepohl, 1995). With 0.12, 0.038, 0.038, and 0.25 being the mass rations in seawater of magnesium to sodium, calcium to sodium, as well as potassium to sodium and sulfate to sodium, respectively.

The MD content was calculated by a straightforward method, and the Al concentration in dust was estimated at 7% (Zhang et al., 2013b):

$$MD=Al/0.07 \quad (4)$$

We determined $K_{Biosmoke}$ as follows (Pu et al., 2017):

$$K_{Biosmoke}=K_{Total}-K_{Dust}-K_{Ss} \quad (5)$$

$$K_{Dust}=Al\cdot(K/Al)_{Crust} \quad (6)$$

$$K_{Ss}=Na_{Ss}\cdot 0.038 \quad (7)$$

Where $(K/Al)_{Crust}$ is 0.37 and represents the K/Al ratio in the dust materials (Wedepohl, 1995) and $Na_{Ss}$ is estimated by Eq. (3).

## 2.4 Enrichment factor (EF)

To evaluate the relative contributions of trace elements from natural (e.g., mineral and soil dust) versus anthropogenic sources, an inter-annual comparison of $EF_c$ values, which represent the enrichment of a given element relative to its concentration in the

crust of the earth. The primary uncertainty in these calculations is attributed to the differences between chemical compositions in the snow and the reference crustal composition. The EF is defined as the concentration ratio of a given metal to that of Al, which is a reliable measure of crustal dust, normalized to the same concentration ratio characteristic of the upper continental crust (Wedepohl, 1995), calculated with the following equation:

$$EF = \frac{(X/Al)_{snow}}{(X/Al)_{crust}} \quad (8)$$

### 2.5 Source apportionment

The Positive Matrix Factorization (PMF 5.0) is considered as a generally accepted receptor model to determine source apportionment of the ILAPs when source emission profiles are unknown (Paatero and Tapper, 1994). Details of the PMF procedure used in this study are also similar to the previous work as discussed in Hegg et al. (2009, 2010). Generally, the mass concentration of the chemical species and the uncertainty were used as the input. The final data set used for the PMF analysis contained 189 samples with 18 elements whereby only elements that have high recovery were used. The uncertainty value of each variable in each sample estimated from an empirical equation. The PMF model was run for 3 to 6 factors with 6 random seeds, but only a three-factor solution of the ILAPs in TP glaciers could provide the most meaningful results. $Q$ values (modified values) for the 3-factor solution (both robust and true) were closest to the theoretical $Q$ value of any of the factor numbers for which the model was run, suggesting that the 3-factor solution was optimal.

## 3. Results and Discussion

### 3.1 Regional averages

67 ice samples were collected at 7 sites from 2013 to 2015 across the Tibetan plateau field campaign. The general information of $C_{BC}^{est}$, $C_{BC}^{max}$, $C_{BC}^{equiv}$, $f_{non\text{-}BC}^{est}$, $\mathring{A}_{tot}$, and $\mathring{A}_{non\text{-}BC}$ of the ice samples are given in Table 1 for each glacier. The lower median values of $C_{BC}^{est}$ could be found in the Xiaodongkemadi, Hariqin, and Gurenhekou glaciers on the south of

the TP regions, while the other glaciers show a relative higher range (94-172 ng $g^{-1}$) on the north edge of the TP regions. Details of the vertical profiles of all ice samples collected in each site could also be found in Table S1. Based on the parameters listed in Table 1, we can analyze the major characteristics of the BC and non-BC components in the samples during these field campaigns from 2013-2015 in the TP glaciers. We found that the lowest concentration of BC in the ice samples is found in the Xiaodongkemadi glacier, with a value of $C_{BC}^{est}$ ~10 ng $g^{-1}$. In contrast, the highest values of $C_{BC}^{est}$, $C_{BC}^{max}$, and $C_{BC}^{equiv}$ are 3100 ng $g^{-1}$, 3600 ng $g^{-1}$, and 4700 ng $g^{-1}$, respectively, taken in the Gurenhekou region. Generally, the median of the absorption Ångström exponent for total particulate constituents ($Å_{tot}$) exceeds 1.0 at all locations (Fig. 3). As shown in Fig. 3a, the lowest median value of $Å_{tot}$ (~2.1) is found in the Xiaodongkemadi glacier, while the other glaciers exhibit much higher values (2.5-2.9). The results indicated that the emission of the ILAPs in the Xiaodongkemadi glacier likely originated from the combustion sources, which is also consistent with the previous studies (Bond et al., 1999, 2001; Schnaiter et al., 2003, 2005; Bergstrom et al., 2007; Clarke et al., 2007). Except the Hariqin glacier, the other glaciers show an increased trend of the absorption Ångström exponent for non-BC particulate constituents ($Å_{non\text{-}BC}$) from the south to north regions in the TP regions (Fig. 3b). $Å_{tot}$ and $Å_{non\text{-}BC}$ for all ice samples were in the range of 1.4-3.7 and 1.9-5.8, respectively (Table S1). Histograms of the absorption Ångström exponent by region are shown in Fig. 4. In the Yuzhufeng and Xiaodongkemadi glaciers, there is a large variation of the absorption Ångström exponent (~1-4), reflecting that the ILAPs are not only dominated by BC in these regions but also influenced by non-BC absorbers such as OC and mineral dust. In contrast, a common feature in the other regions is that they show a less variable of the absorption Ångström exponent, ranging from 2.5-3.

BC and other ILAPs are integrated into the snowpack and ice surface by dry and wet deposition, such as gravity, turbulence, and precipitation. For instance, Flanner et al. (2012) indicated that BC nucleates ice very poorly via direct deposition of vapor, so most relevant mechanisms involve liquid water. Therefore, the investigation of the mixing ratios of ILAPs in each glacier could be useful to analyze the emission sources of the air pollutants. As shown in Fig. 5, a notable feather is that there are large biases between the

median and the average values of the concentration of ILAPs in ice in each glacier. Due to all of the ice samples were collected in the individual period from 2013-2015, there are large variations of the concentrations of the ILAPs in ice samples. Therefore, we note that median values are more representative in each glacier. Based on our sampling locations shown in Fig. 1, the Qiyi, Qiumianleiketage, Meikuang, Yuzhufeng glaciers were located in the northern part of Tibetan Plateau, while Xiaodongkemadi, Hariqin, Gurenhekou glaciers were located in the southern part of Tibetan Plateau. As shown in Fig. 5, the median values of the $C_{BC}^{est}$ and $C_{ISOC}$ obviously showed a significant decreasing trend from the northern TP to the southern TP. The mass concentration of BC in northern TP glaciers was higher than that in southern TP glaciers, which shows a good agreement with Ming et al. (2013). Comparing with the Hariqin, Xiaodongkemadi and Gurenhekou glaciers, the relative higher values of the $C_{BC}^{est}$ and$C_{ISOC}$ in Qiyi, Qiumianleiketage, Yuzhufeng, and Meikuang glaciers are possibly influenced by human activities. Doherty et al. (2010, 2014) and Wang et al. (2013) indicated that the light absorption of Fe across the Northern Hemisphere is mainly originated from the local mineral dust. Therefore, the concentrations of Fe in ice samples in each glacier are exhibited in Fig. 5c.

The Yuzhufeng glacier is located in the northern part of the Loess Plateau, close to the Meikuang glacier. Twelve ice samples were collected in the Yuzhufeng glacier. The depth of these ice samples collected in Yuzhufeng glacier are ranging from 15 to 45 cm as Table S1 shown. As shown in Fig. S2, most values of $C_{BC}^{est}$ in this region range from ~100-1000 ng g$^{-1}$, with a few values lower than 100 ng g$^{-1}$. One notable feature is that the highest concentrations of $C_{BC}^{equiv}$ and $C_{BC}^{max}$ for the surface layer are 2600 ng g$^{-1}$ and 1600 ng g$^{-1}$, respectively, at site 41. While the concentration of ISOC and $\mathring{A}_{tot}$ are ~9160 ng g$^{-1}$ and 3.41. Due to the non-BC fraction of the light absorption ($f_{non-BC}^{est}$) is 0.56, we pointed out that the light absorption in the surface glacier at site 41 wasn't only influenced by BC, but also possibly related to the ISOC, and MD. The large variation of the absorption Ångström exponent distribution is an indication of the complicated emission sources of ILAPs in the ice samples. In the Yuzhufeng glacier, $\mathring{A}_{tot}$ generally varied between ~2 and 3.7, and the average value of $f_{non-BC}^{est}$ is close to 50%, so these results also reveal that the ILAPs in ice samples are heavily influenced by anthropogenic air pollutants. Large

variations of ISOC (measured by ISSW) and WSOC are also observed, with values ranging from ~10-17000 ng $g^{-1}$ and ~410-43000 ng $g^{-1}$, respectively. Except for site 23, the values of $C_{BC}^{est}$ in the Meikuang glacier are much lower than those in the Yuzhufeng glacier, with values ranging from ~20-670 ng $g^{-1}$ and with a median value of 130 ng $g^{-1}$. The median value of the mass concentration of ISOC is ~600 ng $g^{-1}$ in the Meikuang glacier. The fraction of total particulate light absorption due to non-BC constituents is typically ~16-62%, and $\AA_{non\text{-}BC}$ (5.12) in this region is highly similar to that found in the Yuzhufeng glacier (5.06). In addition, there appears to be no significant difference in the mixing ratios of ILAPs in the ice samples via dry and wet deposition in these glaciers between the monsoon and non-monsoon seasons (Fig. S2, S3, S4, S5, S6). We noted that several reasons could lead this discrepancy. First of all, all of the ice samples were collected in an individual time. Another major issue is that except the long-range transport of ILAPs, local air pollutants can also affect the ILAPs in the glacier via wet and dry deposition. For instance, Li et al. (2016) indicated that the Fossil fuel contributions of BC is much higher in the Laohugou No. 12 glacier (close to Qiyi glacier) due to human activities. Finally, based the study by Xu et al. (2009), the ILAPs in the TP glacier also can be affected by the European air to reach that location on the eastern plateau, and thus lead the greater proportion of the heavy leading of the BC in the glacier. For instance, the mass concentration of BC in the Muztagh Ata and Xiaodongkemadi glaciers in northern part of Tibetan Plateau is much higher than that in the East Rongbuk, Noijin Kangsang and Zuoqiupo glacier, which are located in the southern part of Tibetan Plateau.

In the Qiyi glacier (Fig. S4), the $C_{BC}^{est}$ are much similar than those in the Meikuang glacier, with values ranging from ~20-720 ng $g^{-1}$, which does not include the highest value of 1900 ng $g^{-1}$ at site 13. The fraction of total particulate light absorption due to non-BC constituent $f_{non\text{-}BC}^{est}$ is typically ~20-70%, with a median value of 41%. This information along with the lower $\AA_{tot}$ (2.6) indicates that BC plays the dominant role in influencing the light absorption in this region. 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). In the monsoon season, the mixing ratios of ISOC

and Fe ranged from 80-10100 ng $g^{-1}$ and 20-340 ng $g^{-1}$, respectively, and the mixing ratios of ILAPs in most of the ice samples increased remarkably from the top to the bottom. This result is highly consistent with a previous study by Doherty et al. (2013). Fig. S5 shows that the vertical profiles of the mass mixing ratios of BC, ISOC, and Fe for the ice samples in the Xiaodongkemadi glacier were more complicated than those for the other regions. With the exception of the surface layer at sites 53 and 54, most values of $C_{BC}^{est}$ ranged from 10 to 280 ng $g^{-1}$ in the Xiaodongkemadi glacier; therefore, this glacier was the cleanest region of all the studied glaciers. 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. Doherty et al. (2013) also found that the ILAPs could be scavenged with the snow meltwater, therefore leading to a much higher concentration of BC in the surface snow. At sites 56-58, $f_{non\text{-}BC}^{est}$ was lower than 38%, and $\mathring{A}_{tot}$ ranged from 1-2.5. These results are consistent with the fossil fuel combustion source due to industrial activities. Because single layer samples are not shown, the vertical profiles of $C_{BC}^{est}$ are plotted in Fig. S6 for all ice samples, which were collected in the Qiumianleiketage, Hariqin, and Gurenhekou glaciers. Except for the sites in Fig. S6d, S6e, S6i, and S6h, the other sites reveal the trapping and scavenging effects of a higher mass concentration of BC in the surface layer due to the melting process. Previous studies have also illustrated that the ILAPs could become trapped and integrated at the surface of the snowpack due to melting and sublimation to enrich the surface concentrations (Conway et al., 1996; Painter et al., 2012; Doherty et al., 2013).

### 3.2 Water-soluble organic carbon

It is well known that the WSOC component has a large variation ranging from 20% to 99% of total carbonaceous particles (Saxena and Hildemann, 1996; Mayol-Bracero et al., 2002). There is a very large fraction representing the average WSOC (>80%) to TOC, which can absorb solar light and enhance cloud formation through their direct and indirect climate effects (Ram et al., 2010). These results are highly consistent with

previous studies showing that fossil fuel combustion plays a key role in leading to the higher fraction of WSOC to total organic carbon (TOC) (Zhang et al., 2012). The concentration of WSOC during the field campaign varied from 400 to 43600 ng $g^{-1}$, with a median of 1400 ng $g^{-1}$ (Table 1). The median values of the WSOC/TOC ratio were 0.85 for the monsoon season and 0.9 for the non-monsoon season during this field campaign (Fig. 6). No remarkable correlation was observed between WSOC and ISOC in both monsoon and non-monsoon seasons. As WSOC is considered as a stable indicator of the primary biomass burning, with a small contribution of fossil fuel combustion, we indicate that a mixture emission sources from the biomass burning and fossil fuel combustion might be contributing significantly to the TOC concentrations in these TP glacier regions.

### 3.3 Contributions to particulate light absorption by ILAPs

BC and OC were mainly emitted from biomass burning and biofuel and fossil fuel combustion, while mineral dust was emitted from the local soil or desert regions (Streets et al., 2001; Painter et al., 2007; Chen and Bond, 2010; Bond et al., 2013; Kaspari et al., 2014; Pu et al., 2017). The contributions to particulate light absorption by BC, OC, and Fe have been investigated using the ISSW measurements across northern China and North America (Wang et al., 2013; Doherty et al., 2014). The fractional contributions to absorption by BC, ISOC, and Fe at 450 nm in surface glaciers are shown in Fig. 7, and the concentrations of BC, ISOC, and Fe are given in Table S1. BC plays a dominant role in particulate light absorption with values ranging from 20-87% in all glacier regions. ISOC is the second highest absorber in glacier regions, and there are large variations of light absorption of ISOC during the field campaign with values ranging from 0.5-58%. The light absorption due to ILAPs in the TP glacier regions is not only from industrial and biomass burning but also with a small contribution from local soil dust. Note that the median fraction of light absorption due to Fe is ~13%, with the highest light absorption of iron being higher than 40% in the Gurenhekou glacier. This result is an indication that mineral dust plays a key role in affecting the spectral absorption properties due to the soil and mineral dust at site 67. Although the total light absorption was dominated by BC and ISOC in all selected glaciers, we note that the relative spatial distribution of the total light

absorption due to Fe (>17%) also play key roles to affected the snowmelt in the southern glaciers than that of northern glaciers across the TP regions (Fig. S7).

### 3.4 Enrichment factor (EF)

Briefly, EF values ranging from 0.1 to 10 indicate significant input from crustal sources. Conversely, EF values that larger than 10 exhibit a major contribution from anthropogenic activities. Referring to the EF analysis (Fig. 8), the mean EF of Fe < 5 in each glacier can be assumed to customarily originate from crustal sources. Recent studies have also indicated that light-absorbing particles in snow are dominated by local soil dust in some typical regions over northern China (Wang et al., 2013), and northern America (Doherty et al., 2014). Comparable with Fe, the other trace metals with the mean EF of ⩾5.0 were moderately to highly enriched predominantly from anthropogenic emissions (Hsu et al., 2010). For example, Pacyna (2001) reported that fossil fuel combustion is a major source of Cr. Cu primarily originates from emissions from fossil fuel combustion and industrial processes, while Pb and Zn are known to be drawn from the traffic-related activities and coal burning (Christian et al., 2010; Contini et al., 2014). Together with high EF values observed for Cu, Zn, and Cd in our ice samples clearly suggested that the TP glaciers have already been polluted by human activities, such as biomass burning, fossil fuel burning, and the coal burning. For instance, high level of Pb and Cr have demonstrated a link to coal combustion (Zhang et al., 2013a, 2013b; Mokhtar et al., 2014), abundant Cu and Cd were associated with traffic-related dusts (Cheng et al., 2010). Therefore, we concluded that the natural dust source and anthropogenic emission source are both non-negligible to the ILAPs in the TP glaciers.

### 3.5 Source apportionment

Given the importance of the climate effect in our understanding of the ILAPs in TP glaciers, we present a PMF receptor model to analyze the source attribution of ILAPs in these glaciers. In this study, two datasets including the mass concentrations of the chemical components and the ILAPs in ice and the associated uncertainty datasets were used to run the PMF 5.0 model. The details of the techniques have already been

illustrated by Hegg et al. (2009, 2010) and Pu et al. (2017). The factor loadings (apportionment of species mass to individual factors) for the 3-factor solution of the source profiles based on the PMF 5.0 model are given in Fig. 9 (in both measured mass concentration and the % total mass allocated to each factor). It is evident that the first factor (top panel) was obviously characterized by high loadings of $Cl^-$, $CL_{salt}$, $SO_4^{2-}$, and $NO_3^-$, which are well known markers for the urban or local industrial pollutions (Alexander et al., 2015). Although $Cl^-$ to $Na^+$ are usually considered as a potential product of emission source of sea salt, but also a high loading of $Cl^-$ to $CL_{salt}$ indicated another source in addition to sea salt such as industrial emission and coal combustion (Hailin et al., 2008; Kulkarni, 2009). Additionally, the highest loading of $NH_4^+$ is also suggested as an indicator of coal combustion (Pang et al., 2007). Compared with the first factor, the highest loading of Al (90.3%) and Fe (87.3%) are well-known markers for the urban or regional mineral dust (Pu et al., 2017). Therefore, the second factor or source profile is easily interpretable as a natural mineral dust source. 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). It is well known that $K^+$ and $K_{Biosmoke}$ are the major indicators of biomass burning source (Zhang et al., 2013a). Therefore, it is easily interpretable that the highest loadings of $K^+$ and $K_{Biosmoke}$ are well representative the biomass burning source (Fig. 9c). However, it is also important to note that the lowest mass loading of $C_{BC}^{max}$ in this factor is a bit unexpected. 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). Therefore, we interpreted the third factor normally considered a predominantly biomass burning product. However, the major emission of BC in TP glaciers originated from the local mineral dust source instead of the biomass burning and industrial pollution than previous studies (Zhang et al., 2013a; Pu et al., 2017).

Finally, the chemical composition and mean source apportionment of the ILAPs to the three sources in the TP glaciers were given in Fig. 10. Note that the apportionment is of the light absorption by insoluble particles in the surface glaciers. On average, the source appointment of the ILAPs in all TP glaciers by mineral dust is close to 37.5%, while the

industrial emission and biomass burning contributes 33% and 29.4%. Specifically, the largest biomass burning contribution of the light-absorption of ILAPs was found in the Qiyi glacier, which is close to the human activity regions (Guan et al., 2009; Li et al., 2016). In the Meikuang, Qiumianleiketage, Gurenhekou, and Xiaodongkemadi glaciers, the mineral dust contribution of light absorption is much larger (>47.9%) than that of industrial pollution and biomass burning, especially in the Meikuang glacier. In these regions, the percent of the MD light absorption is ranging from 20.4-31.1%, while the light absorption by biomass burning is in the range of 18.5-35.8%. Industrial pollution constitutes a major fraction in the Yuzhufeng glacier. Chemical analysis shows that the percentages of the chemical species in the Yuzhufeng and Meikuang glaciers are much similar. The attribution of the total anions by chloride, nitrate, and sulphate is higher 52% and 48% than the other chemical species in the Yuzhufeng and Meikuang glaicers. In the Hariqin glacier, the largest attribution of the sulphate is up to 45.4%. As shown in Fig. 10, the source apportionment of the light absorption by insoluble particles in the surface glaciers is dominated by mineral dust and the industrial pollution in most glaciers, only with a large fraction of the light absorption due to biomass burning in the Yuzhufeng glacier. These results are highly consistent with the previous studies (Andersson et al., 2015). They found that the contributions of coal-combustion-sourced BC are the most significant for the TP glaciers. Based on the model simulations, Zhang et al. (2015) revealed that the largest contribution to annual mean BC burden and surface deposition in the entire TP regions is from biofuel and biomass (BB) emissions in South Asia, followed by fossil fuel (FF) emissions from South Asia.

## 4 Conclusions

In this study, the ILAPs observations in 7 glacier regions across the Tibetan Plateau are presented using the ISSW technique along with chemical analysis. 67 vertical profiles of ice samples are analyzed during the monsoon and non-monsoon seasons from 2013-2015. There are no apparent differences in the mixing ratios of ILAPs in the ice samples during seasonal transitions. The results indicate that the variations of $Å_{tot}$ and $Å_{non\text{-}BC}$ for all ice samples range from 1.4-3.7 and 1.9-5.8, respectively, excluding site 16. The lower

absorption Ångström exponent ($\mathring{A}_{tot}$ <2) suggested that the sites 30, 51, and 56-58, 65 were primarily influenced by fossil fuel emission, whereas the rest of the sites were heavily influenced by mineral dust and biomass burning. Another notable feature is the large variation of the ILAPs in the ice samples. By excluding some of the highest ILAPs values in the ice samples, the values of $C_{BC}^{est}$, $C_{ISOC}$, and $C_{Fe}$ range from 100-1000 ng $g^{-1}$, 10-2700 ng $g^{-1}$, and 10-1000 ng $g^{-1}$, respectively. Among the samples, the lower concentrations of BC were found in the Xiaodongkemadi, Hariqin and Gurenhekou glaciers, with the median concentrations of 33 ng $g^{-1}$, 24 ng $g^{-1}$, and 28 ng $g^{-1}$, respectively.

We also present a PMF receptor model to analyze the source attributions of ILAPs in these glaciers. We found that the anthropogenic air pollution is much heavy across the northern glaciers due to human activities, but the major emissions of the light absorption by insoluble particles in TP glaciers originated from the local mineral dust and industrial pollution sources, followed by the biomass burning source. We note that 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. Therefore, the natural mineral dust source and anthropogenic emission source are both non-negligible to the ILAPs in the TP glaciers.

## 5 Data availability

All datasets and codes used to produce this study can be obtained by contacting Xin Wang (wxin@lzu.edu.cn).

*Competing interests*. The authors declare that they have no conflicts of interest.

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**Table 1**. Statistics of the ice variables measured using an ISSW for each glacier.

| Region | Latitude | Longitude | | $C_{BC}^{equiv}$ | $C_{BC}^{max}$ | $C_{BC}^{est}$ | $f_{non\text{-}BC}^{est}$ | $\mathring{A}_{tot}$ | ISOC | WSOC | Fe |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | (N) | (E) | | (ng g$^{-1}$) | (ng g$^{-1}$) | (ng g$^{-1}$) | (%) | | (ppm) | (ppm) | (ppb) |
| Qiyi glacier | 39°14’28” | 97°45’27” | average | 414 | 299 | 238（116, 313） | 42（15, 66） | 2.59 | 1.21 | 5.43 | 181.3 |
| | | | median | 176 | 128 | 94（29, 124） | 41（17, 70） | 2.62 | 0.66 | 2.25 | 93.94 |
| | | | minimum | 26 | 29 | 25（13, 35） | 21（—, 53） | 0.8 | 0.08 | 0.62 | 19.8 |
| | | | maximum | 2651 | 2230 | 1877（1182, 2109） | 73（41, —） | 3.73 | 11.59 | 43.55 | 2414 |
| Qiumianleiketage | 36°41’47” | 90°43’44” | average | 421 | 296 | 238（139, 402） | 44（24, 81） | 2.80 | 1.43 | 1.99 | 231.7 |
| | | | median | 307 | 215 | 172（64, 218） | 44（24, 81） | 2.76 | 1.06 | 2.23 | 184 |
| | | | minimum | 139 | 93 | 62（19, 93） | 37（12, 64） | 2.45 | 0.54 | 1.17 | 105.9 |
| | | | maximum | 995 | 662 | 558（143, 678） | 56（27, 86） | 3.08 | 3.97 | 2.56 | 625.2 |
| Meikuang glacier | 35°40’24” | 94°11’10” | average | 493 | 328 | 260（119, 331） | 42（15, 37） | 2.65 | 2.14 | 2.80 | 218.8 |
| | | | median | 197 | 156 | 133（76, 153） | 44（16, 69） | 2.64 | 0.61 | 3.05 | 125.5 |
| | | | minimum | 24 | 23 | 19（17, 24） | 16（—, 17） | 1.37 | 0.13 | 0.62 | 32.24 |
| | | | maximum | 4696 | 2817 | 2292（109, 2938） | 62（23, 85） | 3.56 | 16.89 | 8.06 | 1224 |
| Yuzhufeng glacier | 35°38’43” | 94°13’36” | average | 457 | 312 | 233（94, 295） | 51（—, 37） | 2.84 | 1.51 | 2.85 | 438.4 |
| | | | median | 317 | 201 | 160（116, 204） | 48（26, 87） | 2.95 | 1.02 | 2.76 | 212.3 |
| | | | minimum | 52 | 35 | 24（8, 35） | 15（—, 37） | 1.82 | 0.07 | 0.8 | 45.1 |
| | | | maximum | 2630 | 1608 | 1169（72, 1603） | 110（6, 49） | 3.7 | 9.16 | 5.92 | 3513 |
| Hariqin glacier | 33°08’23” | 92°05’34” | average | 476 | 327 | 256（100, 385） | 48（26, 82） | 2.79 | 1.59 | 1.67 | 171.2 |
| | | | median | 54 | 37 | 23（9, 30） | 48（26, 82） | 2.87 | 0.22 | 1.08 | 52.15 |
| | | | minimum | 36 | 24 | 13（4, 22） | 19（—, 41） | 1.96 | 0.08 | 0.72 | 26.69 |
| | | | maximum | 3990 | 2702 | 2131（682, 2784） | 64（32, 84） | 3.52 | 9.64 | 4.92 | 1049 |
| Xiaodongkemadi | 33°04’08” | 92°04’24” | average | 253 | 171 | 152（76, 177） | 37（15, 63） | 2.28 | 0.95 | 1.82 | 173.0 |

| Region | Latitude | Longitude | | $C_{BC}^{equiv}$ | $C_{BC}^{max}$ | $C_{BC}^{est}$ | $f_{non\text{-}BC}^{est}$ | $\mathring{A}_{tot}$ | ISOC | WSOC | Fe |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | (N) | (E) | | (ng g$^{-1}$) | (ng g$^{-1}$) | (ng g$^{-1}$) | (%) | | (ppm) | (ppm) | (ppb) |
| | | | median | 62 | 47 | 53（37, 65） | 36（13, 59） | 2.18 | 0.19 | 1.41 | 62.08 |
| | | | minimum | 13 | 12 | 9（6, 18） | 8（—, 19） | 1.08 | 0.01 | 0.45 | 10.22 |
| | | | maximum | 2770 | 1849 | 1637（596, 2031） | 86（25, 90） | 3.63 | 6.97 | 5.91 | 2129 |
| Gurenhekou glacier | 30°11’17” | 90°27’23” | average | 382 | 292 | 247（212, 591） | 46（16, 71） | 2.42 | 0.62 | 1.21 | 182.1 |
| | | | median | 61 | 46 | 30（19, 44） | 48（18, 75） | 2.46 | 0.13 | 0.85 | 97.99 |
| | | | minimum | 28 | 23 | 15（10, 24） | 27（7, 52） | 1.34 | 0.02 | 0.41 | 31.51 |
| | | | maximum | 4674 | 3634 | 3080（1876, 3884） | 61（26, 85） | 2.92 | 5.22 | 4.65 | 911.2 |

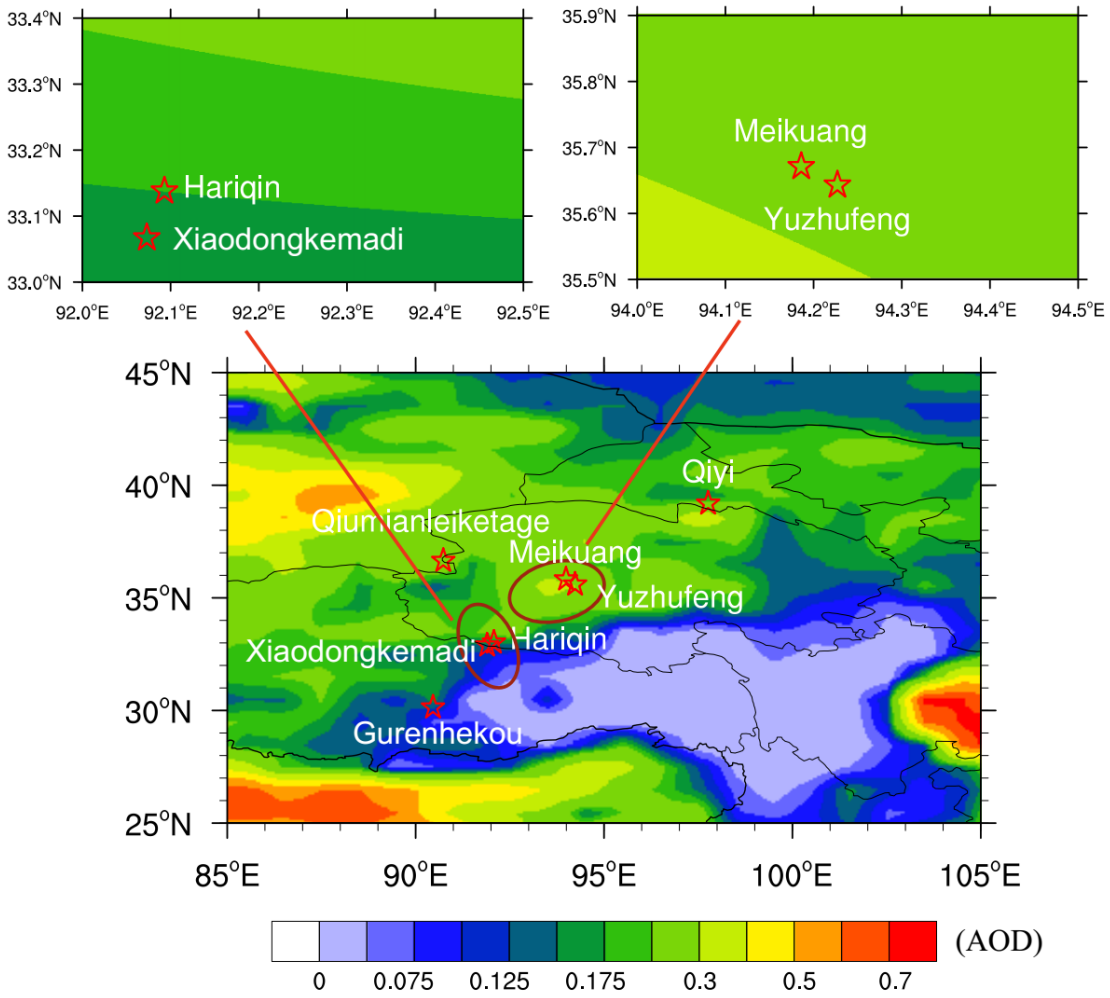


**Figure 1.** Spatial distribution of the averaged AOD retrieved from Aqua-MODIS over Tibetan Plateau from 2013 to 2015, and larger values represent higher optical depth and smaller values represent smaller optical depth. The red stars are the sampling locations (see also Table 1): 1, Qiyi glacier (97.76° E, 39.24° N, 4850 m a.s.l); 2, Qiumianleiketage glacier (90.73° E, 36.70° N, 5240 m a.s.l); 3, Meikuang glacier (94.19° E, 35.67° N, 4983 m a.s.l); 4, Yuzhufeng glacier (94.23° E, 35.65° N, 5200 m a.s.l); 5, Hariqin glacier (92.09° E, 33.14° N, 5100 m a.s.l); 6, Xiaodongkemadi glacier (92.07° E, 33.07° N, 5600 m a.s.l); 7, Gurenhekou glacier (90.46° E, 30.19° N, 5655 m a.s.l).

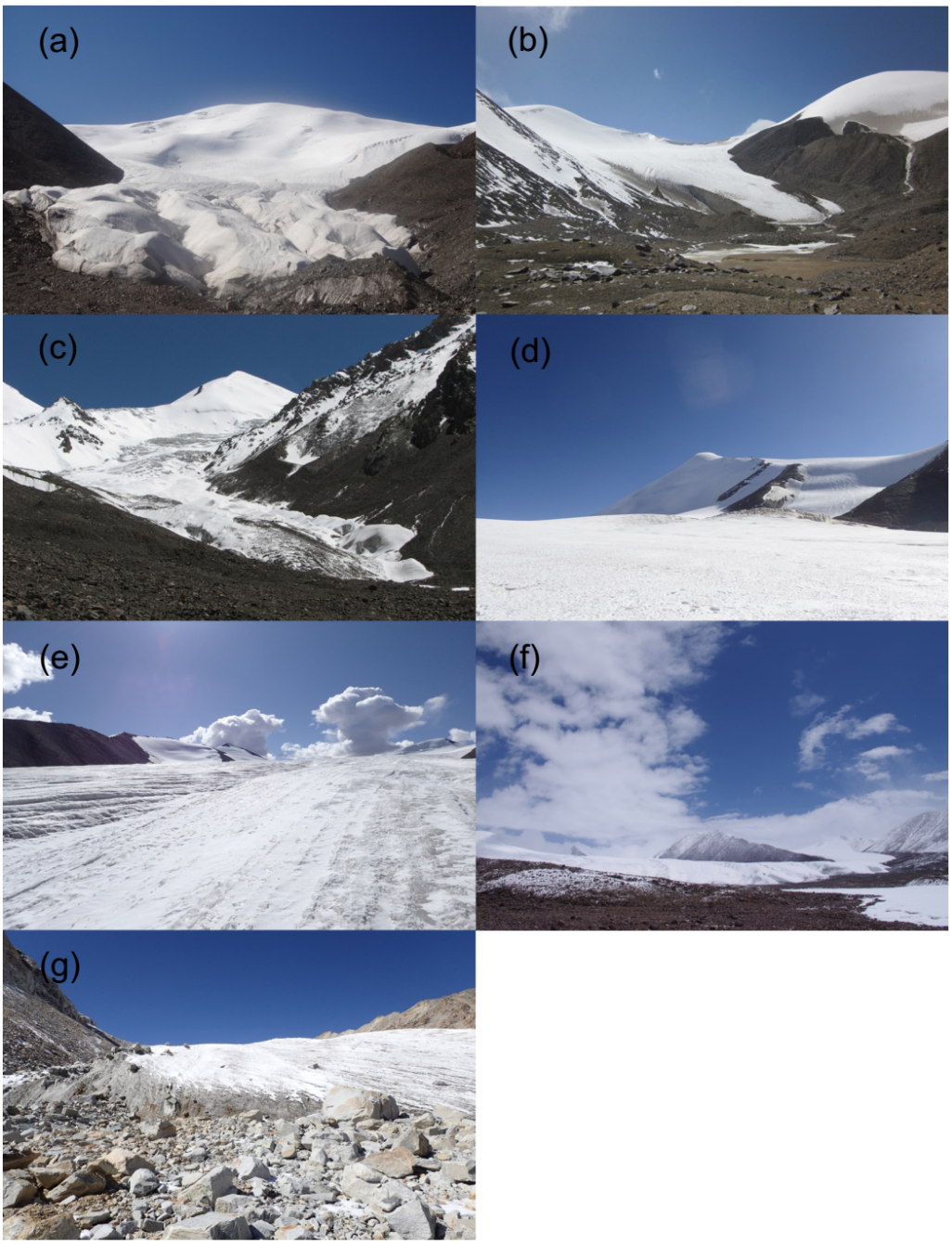


**Figure 2.** Pictures of ice sampling location in the (a) Qiyi, (b) Qiumianleiketage, (c) Meikuang, (d) Yuzhufeng, (e) Hariqin, (f) Xiaodongkemadi, (g) Gurenhekou glaciers, respectively.

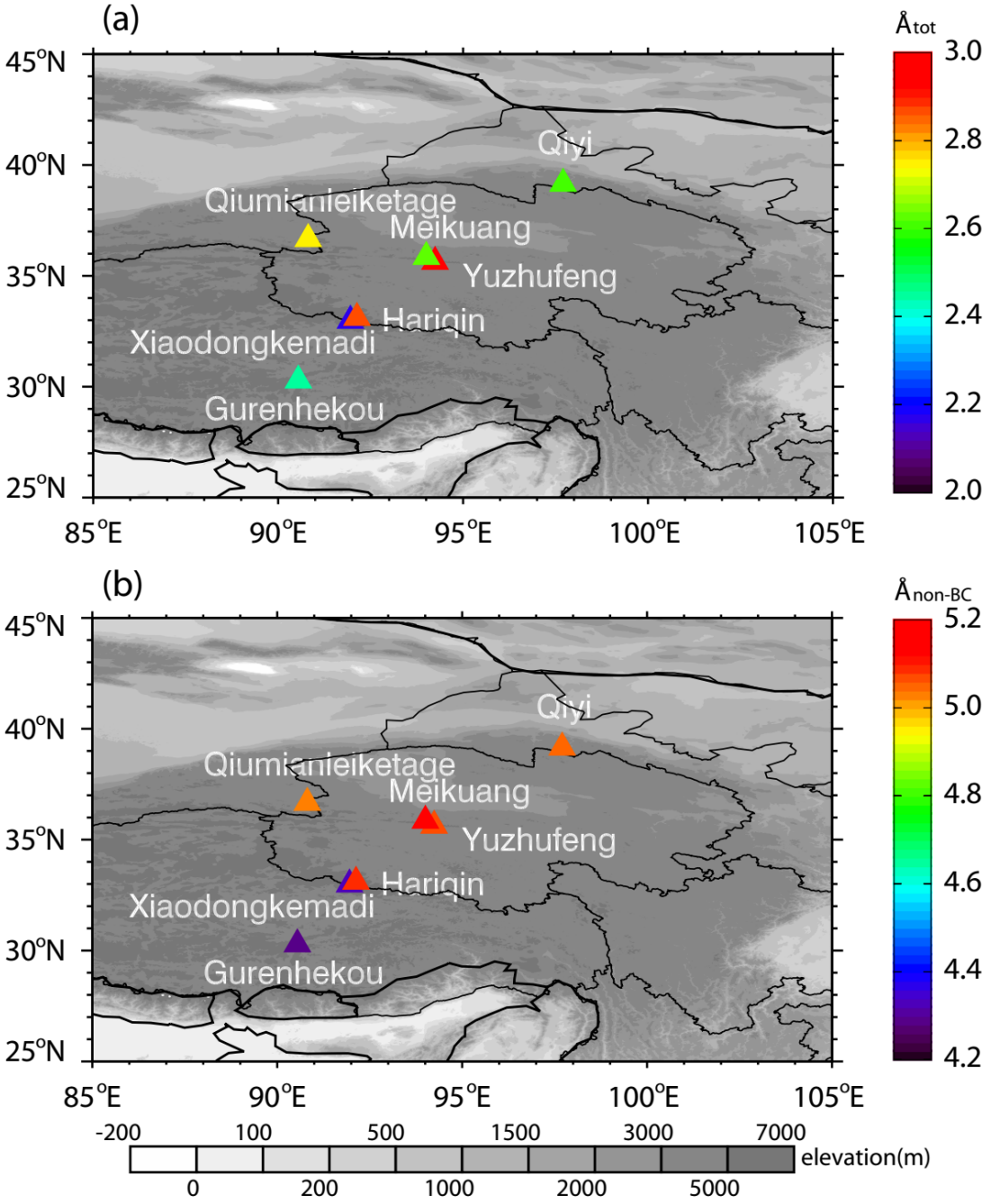


**Figure 3.** The spatial distribution of the median absorption Ångström exponent for (a) total particulate constituents ($Å_{tot}$), and (b) non-BC particulate constituents ($Å_{non\text{-}BC}$) in each glacier (see Fig. 1).

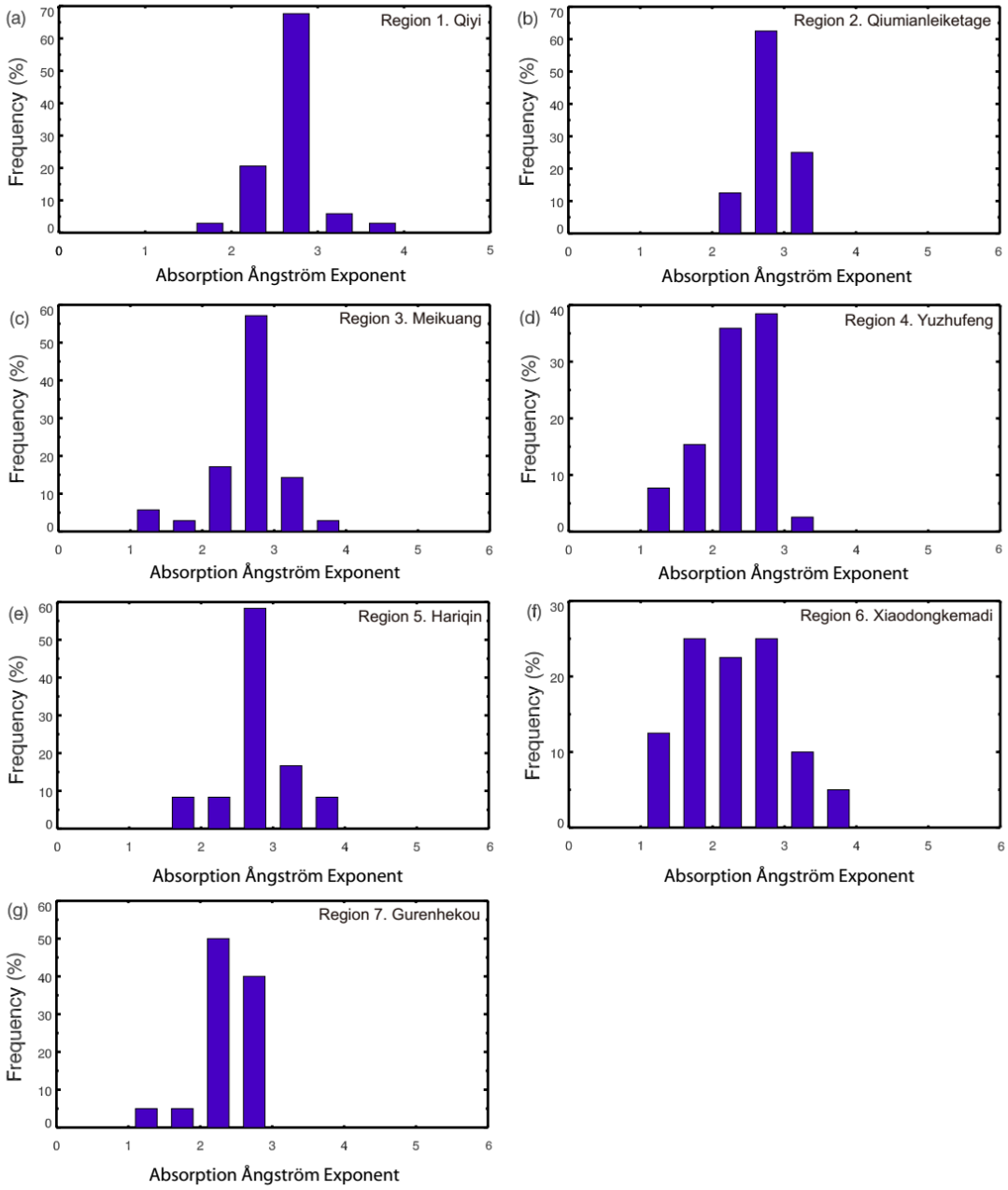


**Figure 4.** Histograms of the frequency of $\mathring{A}_{tot}$ (450-600 nm) for ice samples in each of the glacier region. Samples from all vertical profiles are included.

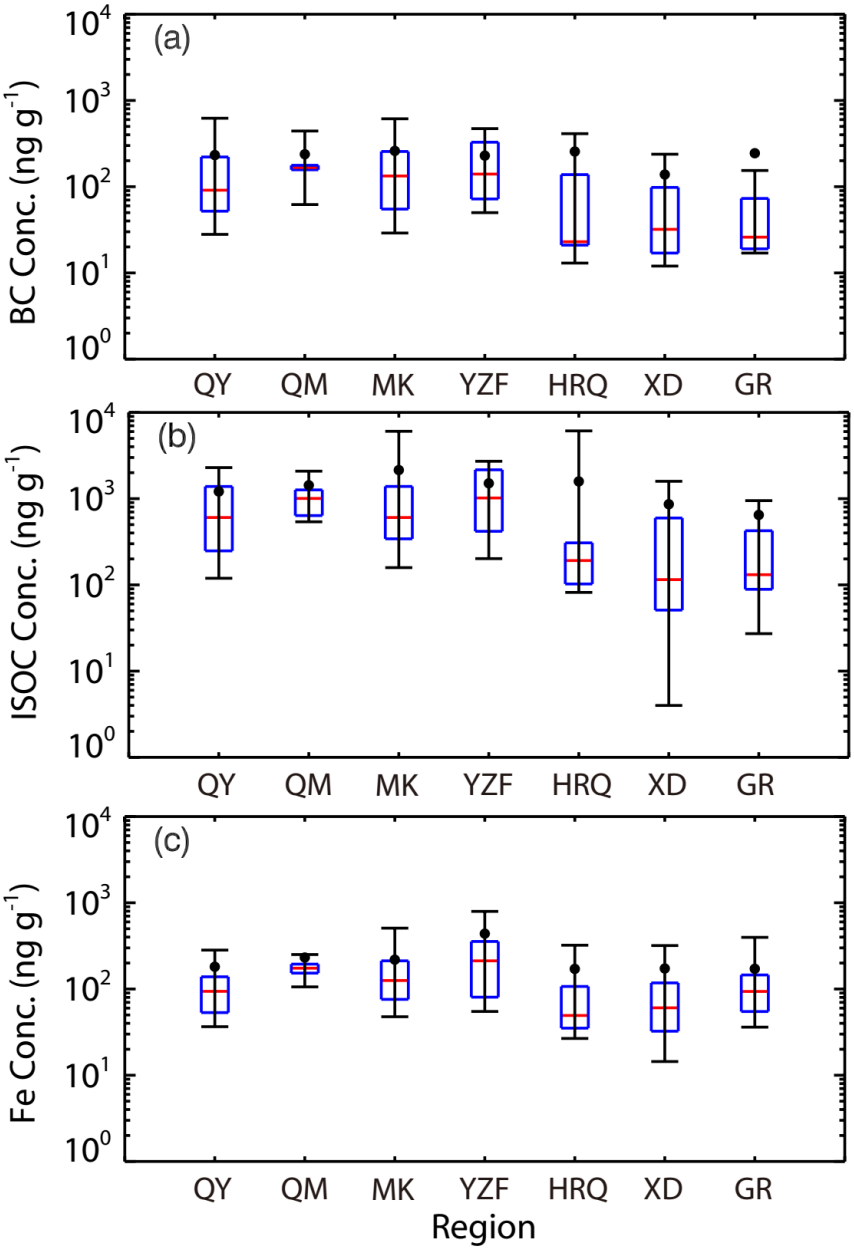


**Figure 5.** Box plots of the region variations in (a) BC concentration, (b) ISOC concentration, and (c) Fe concentration of the seven glaciers. QY, QM, MK, YZF, HRQ, XD, and GR represent the following glaciers: Qiyi, Qiumianleiketage, Meikuang, Yuzhufeng, Hariqin, Xiaodongkemadi, Gurenhekou glaciers, respectively. Error bars are 10$^{th}$, 25$^{th}$, median, 75$^{th}$, and 90$^{th}$ percentiles of the data. The dot symbol represents the average concentrations of the ILAPs in ice samples in each glacier.

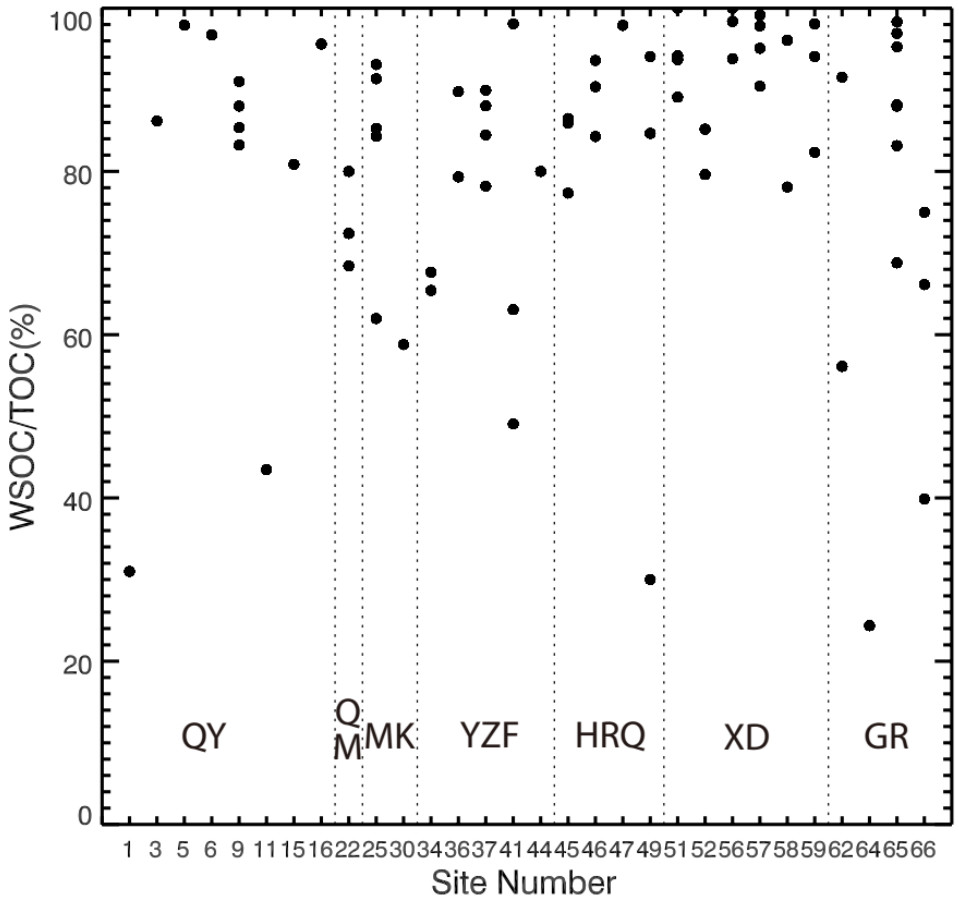


**Figure 6.** The mass fraction of WSOC to TOC of each ice sample.

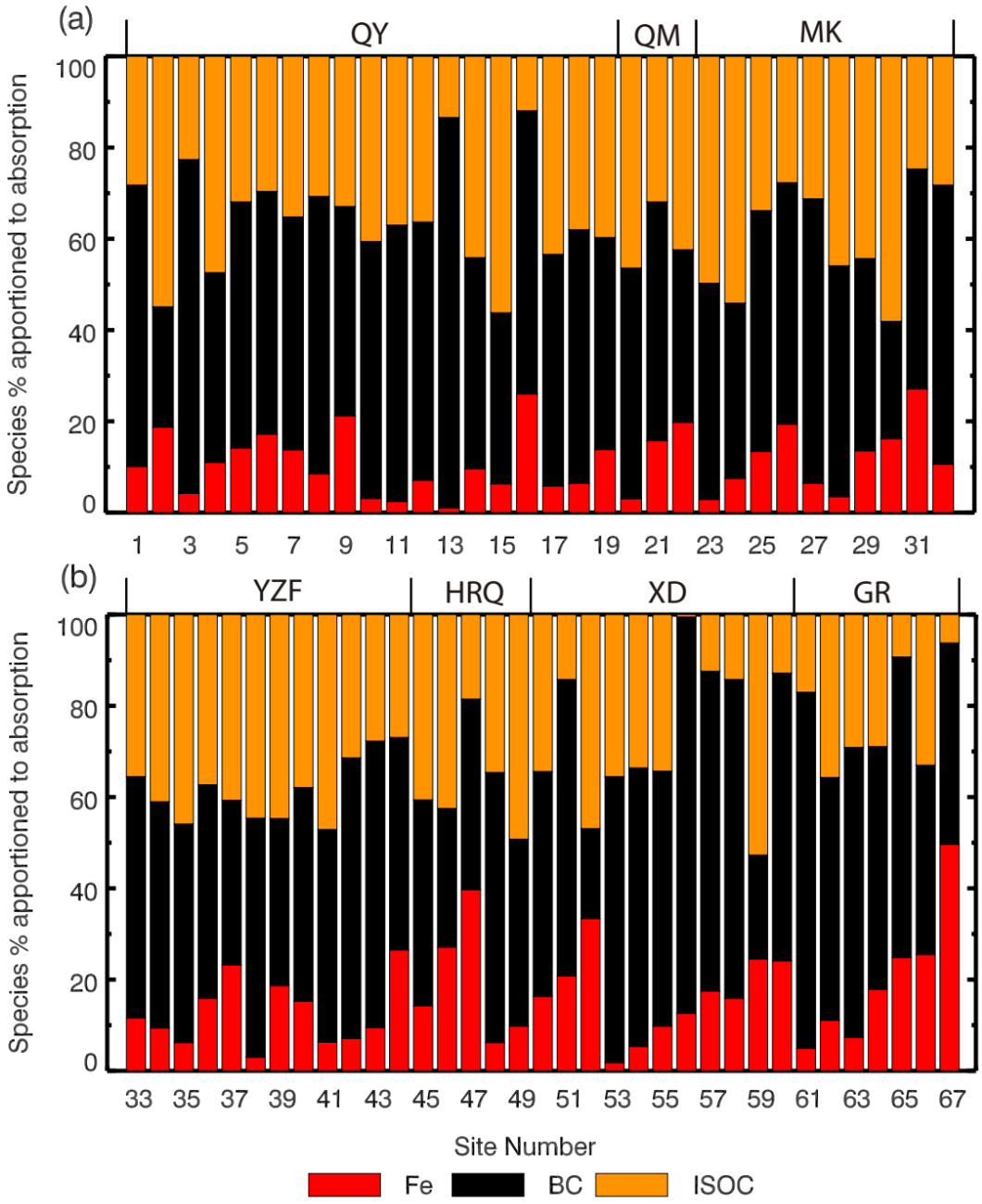


**Figure 7.** Relative contributions to total light absorption by BC, ISOC, and Fe oxide (assumed to be in the form of goethite) for surface ice samples in each sampling site (See Table 1).

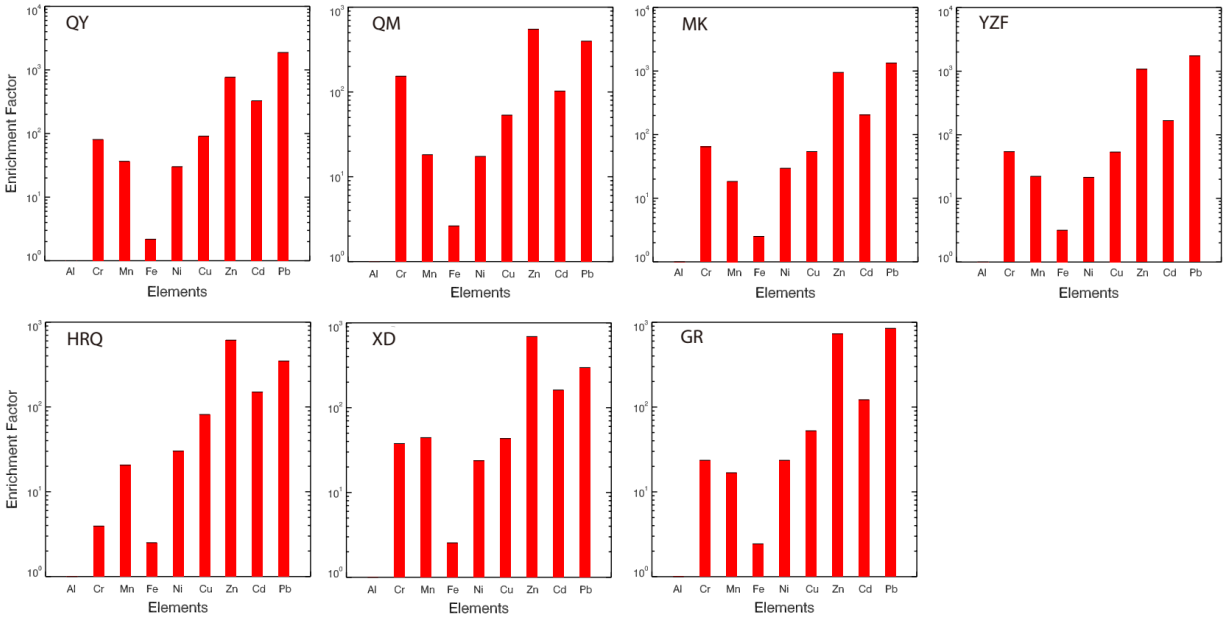


**Figure 8.** Average enrichment factors of trace metals in surface ice samples at each region.

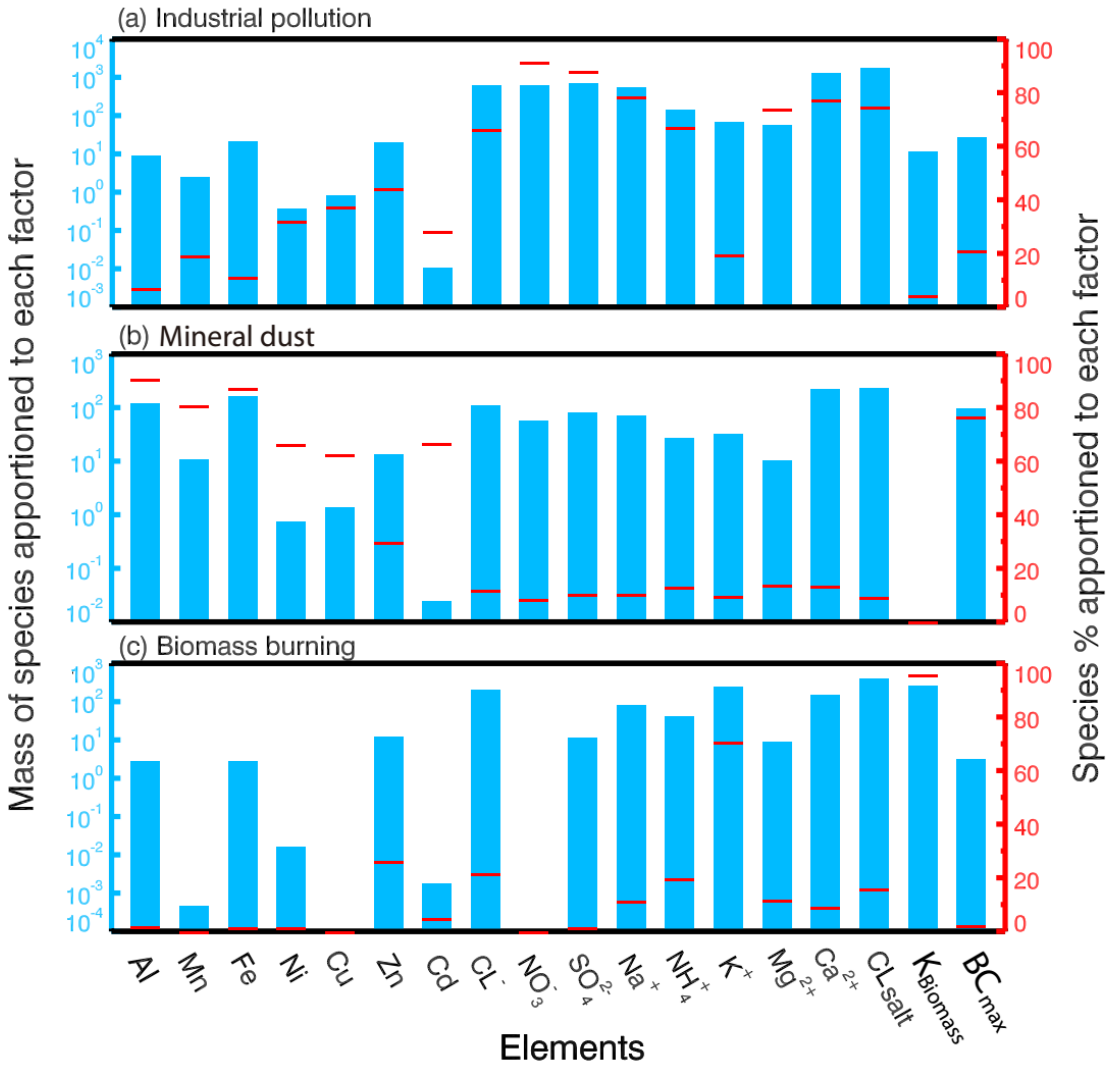


**Figure 9.** Source profiles for the three factors/sources that were resolved by the PMF 5.0 model.

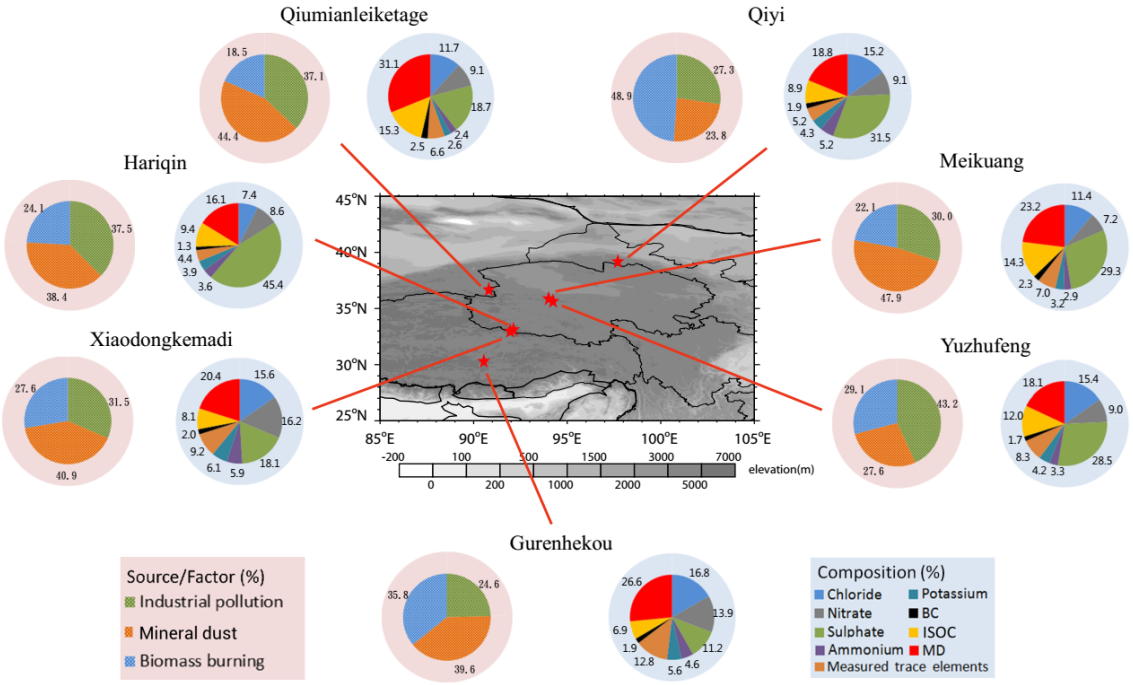


**Figure 10.** Chemical composition and source apportionment for the seven glaciers in the TP regions. Note that the apportionment is of the light absorption by insoluble particles in the surface glaciers.

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# Quantifying light absorption and its source attribution of insoluble light-absorbing particles in Tibetan Plateau glaciers from 2013-2015

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**Abstract.** Amounts of insoluble light-absorbing particles (ILAPs) deposited on the surface of snow and ice can significantly reduce the snow albedo and accelerate the snow melting process. In this study, ~~–~~67 ~~snow/~~ice samples were collected in 7 high mountain glaciers over the Tibetan Plateau (TP) regions from May 2013 to October 2015. The mixing ratio of black carbon (BC), organic carbon (OC), and mineral dust (MD) was measured using an integrating sphere/integrating sandwich spectrophotometer (ISSW) system associated with the chemical analysis by assuming the light absorption of mineral dust due to iron oxide. The results indicate that mass mixing ratios of BC, ISOC, and MD show a large variation of 10-3100 ng $g^{-1}$, 10-17000 ng $g^{-1}$, 10-3500 ng $g^{-1}$, with a mean value of 218±397 ng $g^{-1}$, 1357±2417 ng $g^{-1}$, 241±452 ng $g^{-1}$ on TP glaciers during the entire ~~snow~~ ice field campaign, respectively. The chemical elements and the selected carbonaceous particles were also analyzed of the attributions of the particulate light absorption based on a positive matrix factorization (PMF) receptor model. On average, the industrial pollution (33.1%), biomass/biofuel burning (29.4%), and ~~soil~~ mineral dust (37.5%) were the major sources of the ILAPs in TP glaciers. Although the ~~soil~~ mineral dust assumed to be the highest contributor to the mass loading of ILAPs, we noted that the averaged light absorption of BC (50.7%) and ISOC (33.2%) was largely responsible for the measured light absorption in the high mountain glaciers at the wavelengths of 450-600 nm.

## 1 Introduction

The Tibetan Plateau (TP), known as the highest plateau in the world, and its surrounding areas contain the largest snow and ice mass outside the polar regions (Qin et al., 2006). 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).~~Ample evidence has indicated that the greatest decrease in length and area and the most negative mass balance of high glaciers in the TP regions is associated with the deposition of black carbon (BC) over the past decade (Yao et al., 2012; Xu et al., 2009a; Xu et al., 2006).~~ The unusual increase in temperature over the TP is now considered one of the major contributors to glacial shrinkage (Ding et al., 2006). Climate models indicated that black carbon (BC)~~BC~~ heats the troposphere by absorbing solar radiation (Jacobson, 2001~~; Jacobi et al., 2015~~; Ji et al., 2016; Yang et al., 2017), and BC reduces snow and ice albedos when it is deposited on their surface, thus leading to the acceleration of snowmelt (~~Hadley and Kirchstetter, 2012;~~ Hansen and Nazarenko, 2004; Flanner et al., 2007, 2009; Hadley and Kirchstetter, 2012; Yasunari et al., 2015; Schmale et al., 2017; Zhang et al., 2017, 2018~~; Flanner et al., 2009; Flanner et al., 2007~~). For example, a mixing ratio of 10 ng $g^{-1}$ of BC in snow can reduce snow albedo by 1%, which has a similar effect to that of 500 ng $g^{-1}$ of mineral dust on the albedo of snow and ice at 500 nm wavelength (Warren and Wiscombe, 1980; Warren, 1982; Wang et al., 2017).

Recently, Li et al. (2016) exhibited that similar contributions from fossil fuel (46±11%) and biomass (54±11%) combustion of the BC sources based on the dual-carbon isotopes technique from aerosol and snowpit samples in the TP regions. Bond et al. (~~2014~~2013) indicated that the best estimate of climate forcing from BC deposition on snow and sea ice in the industrial era is +0.13 W $m^{-2}$ with 90% uncertainty bounds of +0.04 to +0.33 W $m^{-2}$. In addition to BC, organic carbon (OC) and mineral dust (MD) also substantially contribute to springtime snowmelt and surface warming through snow darkening effects (Painter et al., 2010; Huang et al., 2011; ~~Yasunari et al., 2015;~~ Painter et al., 2012; Wang et al., 2013;~~Painter et al., 2010;~~ Kaspari et al., 2014; Wang et al., 2014; Yasunari et al.,

2015~~Wang et al., 2013; Huang et al., 2011~~). Water-soluble organic carbon (WSOC) and insoluble organic carbon (ISOC) are the major components of organic carbon in the atmosphere, snow and sea ice. In addition to the strong light absorption of ISOC and WSOC may also influence the regional and global climate through the heating and evaporating of clouds and by acting as cloud condensation nuclei (CCN) (Chen and Bond, 2010; ~~Witkowska and Lewandowska, 2016;~~ Alexander et al., 2012; Witkowska and Lewandowska, 2016). Moreover, WSOC also plays a key role in affecting human health due to its toxic effects (McConnell and Edwards, 2008; Wang et al., 2015). Although the mass mixing ratio of insoluble organic carbon in the snow and ice has been widely investigated in previous studies (Xu et al., 2006, 2009; Flanner et al., 2009; Wang et al., 2013), there are still limited studies that measure the mass mixing ratios of both WSOC and ISOC in ice samples, especially across the TP regions (Li et al., 2016; Yan et al., 2016).~~Although the mass mixing ratio of insoluble organic carbon in the snow and ice has been widely investigated in previous studies, there are still limited studies that measure the mass mixing ratios of both WSOC and ISOC in snow/ice samples, especially across the TP regions.~~ 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).~~–~~

Due to the importance of the climate effects by insoluble light-absorbing particles (ILAPs), numerous snow surveys have been conducted to investigate the light absorption of ILAPs and their potential source attribution in snow (Xu et al., 2009a, b; Doherty et al., 2010, 2014; Hegg et al., 2010; Wang et al., 2015; Jenkins et al., 2016; Niu et al., 2017; Wang et al., 2017). For instance, Hegg et al. (2009) found out that the light absorption in Arctic snow by ILAPs is mainly originated from two distinct biomass burning sources, a pollution source, and a marine source based on the EPA PMF receptor model. Huang et al. (2011) conducted the first snow survey over northern China, and the sources of ILAPs in seasonal snow in the region were explored based on a positive matrix factorization (PMF) with backward trajectory cluster analysis (Zhang et al., 2013a). Wang et al. (2013) indicated that soil dust was found to be the major contributor to snow particulate

absorption in Inner Mongolia regions and Qilian mountains over northern China. Recently, vertical profiles of ILAPs in seasonal snow were performed from 67 North American sites, and biomass/biofuel burning, soil and fossil fuel pollution are explored as the major sources of particulate light absorption based on the chemical and optical data, and these were (Doherty et al., 2014). However, the assessments of the light absorption and its emission sources of ILAPs on the TP glaciers are sparse due to limited observations. Here, we present a snow survey on collecting the ~~snow/ice~~ice samples on 7 high mountain glaciers on the TP regions from 2013-2015. By using an integrating sphere/integrating sandwich spectrophotometer (ISSW) system associated with the chemical analysis, the particulate light absorption of BC, ISOC, and MD in TP glaciers was evaluated. Finally, the relative attribution of emission sources of the ILAPs in these regions was explored based on a positive matrix factorization (PMF) receptor model.

## 2 Site description and methods

### 2.1 Site description and sample collection

As shown in Fig. 1, the spatial distribution of aerosol optical depth (AOD) retrieved from Moderate-resolution Imaging Spectrometer (MODIS) sensors are ranging from 0.1 to 0.4 from south to north near the high mountain glacier regions over TP regions from 2013-2015. The Qiyi glacier (39°14' N, 97°45' E) is located in the eastern part of the TP, with an elevation of 6178 m. It is classified bucket-valley glaciers according to its shape, and is classified subcontinental glacier according to the physical characteristics of glacier. ~~above the equilibrium line altitude (ELA) at 4130 m a.s.l.~~ The ~~Tanggula~~Xiaodongkemadi glacier (33°04' N, 92°04' E) is located ~~at 5743 m a.s.l.~~ in the central Qinghai-Tibetan Plateau. It is 2.8 km in length ~~,~~ and the average snowline ~~in the Tanggula glacier~~ is 5560 m a.s.l. The annual mean air temperature at the equilibrium line altitude is in the range -5~-7℃. The surrounding region is mainly tundra. The Yuzhufeng glacier (35°38' N, 94°13' E) is the highest peak across the eastern Kunlun Mountains, with an elevation of 6178 m. The Meikuang and Qiumianleiketage glaciers are also located over the Kunlun Mountains, and these glaciers have an average altitude of 5100 m and 5500 m a.s.l, respectively. The Meikuang glacier is located in the eastern Kunlun Mountains, where is

characterized by alluvial deposits and sand dunes, the glacier covers an area of 1.1 $km^2$, is 1.8 km in length (Xiao et al., 2002). The Qiumianleiketage glacier is located in the Heyuan District of the Nagora River, the largest river in the Qaidam Basin, which originated in the Kunlun Mountains of the Qinghai-Tibet Plateau. The length of the glacier is 2.6 km, the area is 1.73 $km^2$. The glaciers of Hariqin and Meikuang have similar altitudes but are from different mountains ~~(Li et al., 2016)~~. The ~~Yangbajing~~Gurenhekou glacier is located on the south-eastern margin of the Nyenchen Tanglha Mountains, and seated about 90 km northwest of Lhasa, the capital city of Tibet (Liang et al., 1995). To investigate the enrichment of ILAPs via wet and dry deposition on ~~high~~ glaciers, we collected 67 ice samples in seven glaciers on the Tibetan Plateau ~~67 vertical profiles of snow/ice samples with seasonal transitions were obtained at 7 high mountain sites~~ from May 2013 to October 2015 (Fig. ~~1~~2). 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. 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. In order to analyze light absorption by ILAPs in the ice samples 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. The ice samples were filtered at laboratories in Lanzhou University. The samples were quickly melted and then immediately filtered through a 0.2-μm Nuclepore filter. Additional details on the ice filtration processes have been previously reported (Doherty et al., 2010, 2014; Wang et al., 2013).

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### 2.2 Optical analysis

An updated integrating sphere/integrating sandwich spectrophotometer (ISSW) is used to calculate the mass mixing ratio of BC in snow, which is similar with the instrument

developed by Grenfell et al. (2011). Compared with the ISSW spectrophotometer developed by Grenfell et al. (2011), the major difference is that we used two integrating sphere to calculate the relative attenuation instead of the integrating sandwich diffuser to reduce the diffuse radiation during the measuring process. 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). Briefly, 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 (Atn) is expressed as:

$$Atn=\ln[S_0(\lambda)/S(\lambda)] \quad (1)$$

The following measured parameters included equivalent BC ($C_{BC}^{equiv}$), maximum BC ($C_{BC}^{max}$), estimated BC ($C_{BC}^{est}$), fraction of light absorption by non-BC ILAPs ($f_{non-BC}^{est}$), the non-BC absorption Ångström exponent ($Å_{non\text{-}BC}$) and the absorption Ångström exponent of all ILAPs ($Å_{tot}$), were calculated by using the wavelength dependence of the measured spectral light absorption and by assuming that the MACs of the BC, OC, and Fe are 6.3, 0.3, and 0.9 $m^2\ g^{-1}$, respectively, at 550 nm and that the absorption Ångström exponents (Å or AAE) for BC, OC, and Fe are 1.1, 6, and 3, respectively (Doherty et al., 2010, 2014; Grenfell et al., 2011; Wang et al., 2013). These parameters are defined as follows:

1. $C_{BC}^{max}$ (ng $g^{-1}$): *maximum BC* is the maximum possible BC mixing ratio in snow by assuming all light absorption is due to BC at the wavelengths of 650-700 nm.

2. $C_{BC}^{est}$ (ng $g^{-1}$): *estimated BC* is the estimated snow BC mixing ratio derived by separating the spectrally resolved total light absorption.

3. $C_{BC}^{equiv}$ (ng $g^{-1}$): *equivalent BC* is the amount of BC that would be needed to produce absorption of solar energy by all insoluble particles in snow for the wavelength-integrated from 300-750 nm.

4. $Å_{tot}$: *absorption Ångström exponent* is calculated for all insoluble particles deposited on the filter between 450 and 600 nm.

5. $Å_{non\text{-}BC}$: *non-BC absorption Ångström exponent* is defined as the light absorption by non-BC components of the insoluble particles in snow between 450-600 nm.

6. $f^{est}_{non-BC}$ (%): *fraction of light absorption by non-BC light absorbing particles* is the integrated absorption due to non-BC light absorbing particles, which is weighted by the down-welling solar flux from snow at the wavelengths of 300-750 nm.

It is well known that the aerosol composition and the size distribution are key parameters that affect the absorption Ångström exponent. Doherty et al. (2010) reported that the value of the absorption Ångström exponent of OC was close to 5, which is consistent with previous studies with values ranging from 4-6 (Kirchstetter et al., 2004). Several studies indicated that the absorption Ångström exponent of mineral dust ranged from 2 to 5 (Fialho et al., 2005; Lafon et al., 2006). The variation in the absorption Ångström exponents for urban and industrial fossil fuel emissions is typically in the range of 1.0-1.5 (Millikan, 1961; Bergstrom et al., 2007), which is slightly lower than that of biomass burning, which primarily falls in the range of 1.5-2.5 (Kirchstetter et al., 2004; Bergstrom et al., 2007). Although the source attribution of the insoluble light-absorbing particles in the samples is not a dominant determinant of the value of the absorption Ångström exponent, fossil fuel burning may have a lower absorption Ångström exponent (<2) than 2-5 (Millikan, 1961; Fialho et al., 2005). In this study, we noted that the absorption Ångström exponent ($Å_{tot}$) is due to a mix state of BC and non-BC impurities on our filters, and the calculations of $Å_{tot}$ and $Å_{non\text{-}BC}$ could be found in the study of Doherty et al. (2014). The OC mixing ratio was also determined according to Eq. (2) in Wang et al. (2013), and the Fe concentration was determined according to the inductively coupled plasma-mass spectrometry (ICP-MS) measurements.

### 2.3 Chemical analysis

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). Meanwhile, to quantify WSOC, about 10 ml of the filter liquor was injected into a total carbon analyzer (TOC-V, Shimadzu). The method detection limit (MDL) used was 4 μg $l^{-1}$~~μg/l~~ with a precision of ±5% (Cong et al., 2015). The definition of TOC (Total

Organic Carbon) in this study is calculated as the total WSOC measured by carbon analyzer and the ISOC calculated by ISSW instrument.

The major metallic elements (Al, Cr, Mn, Fe, Ni, Cu, Zn, Cd, Pb) were analyzed by an inductively coupled plasma-mass spectrometry (ICP-MS, X-7 Thermo Elemental) at the Institute of Tibetan Plateau Research in Beijing. The detection limits are Al, 0.238 ng $ml^{-1}$~~ng/ml~~; Cr, 0.075 ng $ml^{-1}$~~ng/ml~~; Mn, 0.006 ng $ml^{-1}$~~ng/ml~~; Fe, 4.146 ng $ml^{-1}$~~ng/ml~~; Ni, 0.049 ng $ml^{-1}$~~ng/ml~~; Cu, 0.054 ng $ml^{-1}$~~ng/ml~~; Zn, 0.049 ng $ml^{-1}$~~ng/ml~~; Cd, 0.002 ng $ml^{-1}$~~ng/ml~~; Pb, 0.002 ng $ml^{-1}$~~ng/ml~~. Generally speaking, we acidified all ~~snow/ice~~ice samples to pH<2 with ultra-pure $HNO_3$, then let settle for 48h. The instrument adopted national standard materials and standard materials of the United States as the quality monitoring samples. The relative deviation between most of the measured values of the elements and the standard reference values is within 10%. ~~We note that the measurement precision is ranging from 2-10%.~~ Details on these procedures are given in Li et al. (2009) and Cong et al. (2010)~~Gao et al. (2003)~~.

Meanwhile, for the filtrated ~~snow/ice~~ice samples, we measured the major anions ($Cl^-$, $NO_2^-$, $NO_3^-$, $SO_4^{2-}$) and cations ($Na^+$, $NH_4^+$, $K^+$, $Mg^{2+}$, $Ca^{2+}$) with an ion chromatograph using a CS12 column for cations and an AS11 column for anions at the Institute of Tibetan Plateau Research in Beijing. All the detection limit of the ions was 1 μg $l^{-1}$~~μg/l~~. In addition, except for the anions and cations and trace elements, $CL_{salt}$, MD and biosmoke K ($K_{Biosmoke}$) were determined to assess the mass contributions of the major components in the ~~snow/ice~~ice samples. $CL_{salt}$ was estimated as follows in accordance with Pio et al. (2007), by adding to sodium, chloride, and sea-salt contributions of magnesium, calcium, potassium, and sulfate, as follows:

$$CL_{salt}=Na^+_{Ss}+Cl^-+Mg^{2+}_{S_s}+Ca^{2+}_{S_s}+K^+_{S_s}+SO^{2-}_{4S_s}$$

$$=Na^+_{Ss}+Cl^-+0.12Na^+_{Ss}+0.038Na^+_{Ss}+0.038Na^+_{Ss}+0.25Na^+_{Ss} \quad (~~1~~2)$$

$$Na_{Ss}= Na_{Total}\text{-}Al\cdot(Na/Al)_{Crust} \quad (~~3~~2)$$

Where $(Na/Al)_{Crust}$=0.33 , and represents the Na/Al ratio in the dust materials (Wedepohl, 1995). With 0.12, 0.038, 0.038, and 0.25 being the mass rations in seawater of magnesium to sodium, calcium to sodium, as well as potassium to sodium and sulfate to sodium, respectively.

The MD content was calculated by a straightforward method, and the Al concentration in dust was estimated at 7% (Zhang et al., 2013b):

$$MD=Al/0.07 \quad (3\text{~~4~~})$$

We determined $K_{Biosmoke}$ as follows (Pu et al., 2017):

$$K_{Biosmoke}=K_{Total}-K_{Dust}-K_{Ss} \quad (4\text{~~5~~})$$

$$K_{Dust}=Al \cdot (K/Al)_{Crust} \quad (5\text{~~6~~})$$

$$K_{Ss}=Na_{Ss} \cdot 0.038 \quad (6\text{~~7~~})$$

Where $(K/Al)_{Crust}$ is 0.37 and represents the K/Al ratio in the dust materials (Wedepohl, 1995) and $Na_{Ss}$ is estimated by Eq. (2~~3~~).

## 2.4 Enrichment factor (EF)

To evaluate the relative contributions of trace elements from natural (e.g., mineral and soil dust) versus anthropogenic sources, an inter-annual comparison of $EF_c$ values, which represent the enrichment of a given element relative to its concentration in the crust of the earth. The primary uncertainty in these calculations is attributed to the differences between chemical compositions in the snow and the reference crustal composition. The ~~$EF_c$~~EF is defined as the concentration ratio of a given metal to that of Al, which is a reliable measure of crustal dust, normalized to the same concentration ratio characteristic of the upper continental crust (Wedepohl, 1995), calculated with the following equation:

$$\text{~~}EF_c\text{~~EF} = \frac{(X/Al)_{snow}}{(X/Al)_{crust}} \quad (8)$$

## 2.5 Source apportionment

The Positive Matrix Factorization (PMF 5.0) is considered as a generally accepted receptor model to determine source apportionment of the ILAPs when source emission profiles are unknown (Paatero and Tapper, 1994). Details of the PMF procedure used in this study are also similar to the previous work as discussed in Hegg et al. (2009, 2010). Generally, the mass concentration of the chemical species and the uncertainty were used

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as the input. The final data set used for the PMF analysis contained 189 samples with 18 elements whereby only elements that have high recovery were used. The uncertainty value of each variable in each sample estimated from an empirical equation. The PMF model was run for 3 to 6 factors with 6 random seeds, but only a three-factor solution of the ILAPs in TP glaciers could provide the most meaningful results. $Q$ values (modified values) for the 3-factor solution (both robust and true) were closest to the theoretical $Q$ value of any of the factor numbers for which the model was run, suggesting that the 3-factor solution was optimal.

## 3. Results and Discussion

### 3.1 Regional averages

~~Over~~ 67 ~~snow/ice~~ice samples were collected at 7 sites from 2013 to 2015 across the Tibetan plateau field campaign. ~~Each vertical snow/ice sample was cut into several pieces.~~ The general information of $C_{BC}^{est}$, $C_{BC}^{max}$, $C_{BC}^{equiv}$, $f_{non\text{-}BC}^{est}$, $\mathring{A}_{tot}$, and $\mathring{A}_{non\text{-}BC}$ of the ~~snow/ice~~ice samples are given in Table 1 for each glacier. The lower median values of $C_{BC}^{est}$ could be found in the ~~Tanggula~~Xiaodongkemadi, Hariqin, and ~~Yangbajing~~Gurenhekou glaciers on the south of the TP regions, while the other glaciers show~~s~~ a relative higher range (94-172 ng $g^{-1}$) on the north edge of the TP regions. Details of the vertical profiles of all ice samples collected in each site could also be found in Table S1. Based on the parameters listed in Table 1, we can analyze the major characteristics of the BC and non-BC components in the samples during these field campaigns from 2013-2015 in the TP glaciers. ~~Details of the vertical profiles of all snow/ice~~ice ~~samples collected in each site could also be found in Table S1. During the field campaign from 2013-2015,~~We found that the lowest concentration of BC in the ~~snow/ice~~ ice samples is found in the ~~Tanggula~~Xiaodongkemadi glacier, with a value of $C_{BC}^{est}$ ~10 ng $g^{-1}$. In contrast, the highest values of $C_{BC}^{est}$, $C_{BC}^{max}$, and $C_{BC}^{equiv}$ are 3100 ng $g^{-1}$, 3600 ng $g^{-1}$, and 4700 ng $g^{-1}$, respectively, taken in the ~~Yangbajing~~Gurenhekou region. ~~We note that there are no apparent differences in ILAPs between the cold and warm seasons in high glacier regions.~~

~~It is well-known that the aerosol composition and the size distribution are key parameters that affect the absorption Ångström exponent. Doherty et al. (2010) reported that the value of the absorption Ångström exponent of OC was close to 5, which is consistent with previous studies with values ranging from 4-6 (Kirchstetter et al., 2004). Several studies indicated that the absorption Ångström exponent of mineral dust ranged from 2 to 5 (Fialho et al., 2005; Lafon et al., 2006). The variation in the absorption Ångström exponents for urban and industrial fossil fuel emissions is typically in the range of 1.0-1.5 (Millikan, 1961; Bergstrom et al., 2007), which is slightly lower than that of biomass burning, which primarily falls in the range of 1.5-2.5 (Kirchstetter et al., 2004; Bergstrom et al., 2007). Although the source attribution of the insoluble light-absorbing particles in the samples is not a dominant determinant of the value of the absorption Ångström exponent, fossil fuel burning may have a lower absorption Ångström exponent (<2) than 2.5 (Fialho et al., 2005; Millikan, 1961).~~ Generally, the median of the absorption Ångström exponent for total particulate constituents ($Å_{tot}$) exceeds 1.0 at all locations (Fig. ~~2a~~3a). As shown in Fig. ~~2a~~3a, the lowest median value of $Å_{tot}$ (~2.1) is found in the ~~Tanggula~~Xiaodongkemadi glacier, while the other glaciers exhibit much higher values (2.5-2.9). The results indicated that the emission of the ILAPs in the ~~Tanggula~~Xiaodongkemadi glacier likely originated from the combustion sources, which is also consistent with the previous studies (Bond et al., 1999, 2001; ~~Bergstrom et al., 2007;~~ Schnaiter et al., 2003, 2005; Bergstrom et al., 2007; Clarke et al., 2007). Except the Hariqin glacier, the other glaciers show an increased trend of the absorption Ångström exponent for non-BC particulate constituents ($Å_{non\text{-}BC}$) from the south to north regions in the TP regions (Fig. ~~2b~~3b). $Å_{tot}$ and $Å_{non\text{-}BC}$ for all ~~snow/ice~~ ice samples were in the range of 1.4-3.7 and 1.9-5.8, respectively (Table S1). ~~The lower absorption Ångström exponent ($Å_{tot}$ <2) found in the Meikuang (site 30), Tanggula~~Xiaodongkemadi ~~(sites 51, 56, 57, 58) and Yangbajing~~Gurenhekou ~~glaciers (site 65) suggested that the sites were primarily influenced by fossil fuel emission, whereas the other sites were heavily influenced by soil mineral dust and biomass burning. Another notable feature is that a lower $Å_{non\text{-}BC}$ suggests a higher percentage of mineral dust in all snow/ice ice samples, while a higher $Å_{non\text{-}BC}$ reflects that the non-BC ILAPs in snow and ice were mainly dominated by OC (Wang et~~

al., 2013; see Eq. 3). Histograms of the absorption Ångström exponent by region are shown in Fig. 34. In the Yuzhufeng and TanggulaXiaodongkemadi glaciers, there is a large variation of the absorption Ångström exponent (~1-4), reflecting that the ILAPs are not only dominated by BC in these regions but also influenced by non-BC absorbers such as OC and mineral dust. In contrast, a common feature in the other regions is that they show a less variable of the absorption Ångström exponent, ranging from 2.5-3.

BC and other ILAPs are integrated into the snowpack and ice surface by dry and wet deposition, such as gravity, turbulence, and precipitation. For instance, Flanner et al. (2012) indicated that BC nucleates ice very poorly via direct deposition of vapor, so most relevant mechanisms involve liquid water. Qi et al. (2017) exhibited that the major process of wet scavenging is in-cloud scavenging, which occurs in two stages: aerosol activation to form cloud droplets, and removal of droplets by precipitation. Therefore, the investigation of the mixing ratios of ILAPs in each glacier could be useful to analyze the emission sources of the air pollutants. As shown in Fig. 45, a notable feather is that there are large biases between the median and the average values of the concentration of ILAPs in snow/ice ice in each glacier. Due to all of the snow/ice ice samples were collected in the individual period from 2013-2015, there are large variations of the concentrations of the ILAPs in snow/ice ice samples. Therefore, we note that median values are more representative in each glacier. Based on our sampling locations shown in Fig. 1, the Qiyi, Qiumianleiketage, Meikuang, Yuzhufeng glaciers were located in the northern part of Tibetan Plateau, while Xiaodongkemadi, Hariqin, Gurenhekou glaciers were located in the southern part of Tibetan Plateau. As shown in Fig. 5, the median values of the $C_{BC}^{est}$ and $C_{ISOC}$ obviously showed a significant decreasing trend from the northern TP to the southern TP. The mass concentration of BC in northern TP glaciers was higher than that in southern TP glaciers, which shows a good agreement with Ming et al. (2013). Comparing with the Hariqin, Xiaodongkemadi and Gurenhekou glaciers, the relative higher values of the $C_{BC}^{est}$ andand $C_{ISOC}$ in Qiyi, Qiumianleiketage, Yuzhufeng, and Meikuang glaciers are possibly influenced by human activities. Doherty et al. (2010, 2014) and Wang et al. (2013) indicated that the light absorption of Fe across the Northern

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Hemisphere is mainly originated from the local ~~soil~~ mineral dust. Therefore, the concentrations of Fe in ~~snow/ice~~ice samples in each glacier are exhibited in Fig. ~~4c~~5c.

The Yuzhufeng glacier is located in the northern part of the Loess Plateau, close to the Meikuang glacier. Twelve ~~snow/ice~~ice samples were collected in the Yuzhufeng glacier. The depth of these ice samples collected in Yuzhufeng glacier are ranging from 15 to 45 cm as Table S1 shown. As shown in Fig. S2, most values of $C_{BC}^{est}$ in this region range from ~100-1000 ng g$^{-1}$, with a few values lower than 100 ng g$^{-1}$. One notable feature is that the highest concentrations of $C_{BC}^{equiv}$ and $C_{BC}^{max}$ for the surface layer are 2600 ng g$^{-1}$ and 1600 ng g$^{-1}$, respectively, at site 41. While the concentration of ISOC and $\mathring{A}_{tot}$ are ~~8600~~ 9160 ng g$^{-1}$ and 3.41. ~~Based on~~Due to the non-BC fraction of the light absorption ($f_{non\text{-}BC}^{est}$) is 0.56, we pointed out that the light absorption in the surface glacier at site 41 wasn't only influenced by BC, but also possibly related to the ISOC, and MD. The large variation of the absorption Ångström exponent distribution is an indication of the complicated emission sources of ILAPs in the ~~snow/ice~~ ice samples. In the Yuzhufeng glacier, $\mathring{A}_{tot}$ generally varied between ~2 and 3.7, and the average value of $f_{non\text{-}BC}^{est}$ is close to 50%, so these results also reveal that the ILAPs in ~~snow/ice~~ ice samples are heavily influenced by anthropogenic air pollutants. Large variations of ISOC (measured by ISSW) and WSOC are also observed, with values ranging from ~10-17000 ng g$^{-1}$ and ~410-43000 ng g$^{-1}$, respectively. Except for site 23, the values of $C_{BC}^{est}$ in the Meikuang glacier are much lower than those in the Yuzhufeng glacier, with values ranging from ~20-670 ng g$^{-1}$ and with a median value of 130 ng g$^{-1}$. The median value of the mass concentration of ISOC is ~600 ng g$^{-1}$ in the Meikuang glacier. The fraction of total particulate light absorption due to non-BC constituents is typically ~16-62%, and $\mathring{A}_{non\text{-}BC}$ (5.12) in this region is highly similar to that found in the Yuzhufeng glacier (5.06). ~~This suggests that the ILAPs have similar emission sources in the Yuzhufeng and Meikuang glaciers.~~ In addition, there appears to be no significant difference in the mixing ratios of ILAPs in the ~~snow/ice~~ ice samples via dry and wet deposition in these glaciers between the ~~cold and warm~~monsoon and non-monsoon seasons (Fig. S2, S3, S4, S5, S6). We noted that several reasons could lead this discrepancy. First of all, all of the ice samples were collected in an individual time. Another major issue is that except the long-range

transport of ILAPs, local air pollutants can also affect the ILAPs in the glacier via wet and dry deposition. For instance, Li et al. (2016) indicated that the Fossil fuel contributions of BC is much higher in the Laohugou No. 12 glacier (close to Qiyi glacier) due to human activities. Finally, based the study by Xu et al. (2009), the ILAPs in the TP glacier also can be affected by the European air to reach that location on the eastern plateau, and thus lead the greater proportion of the heavy leading of the BC in the glacier. For instance, the mass concentration of BC in the Muztagh Ata and Xiaodongkemadi glaciers in northern part of Tibetan Plateau is much higher than that in the East Rongbuk, Noijin Kangsang and Zuoqiupo glacier, which are located in the southern part of Tibetan Plateau.

In the Qiyi glacier (Fig. S4), the $C_{BC}^{est}$ are much similar than those in the Meikuang glacier, with values ranging from ~20-720 ng $g^{-1}$, which does not include the highest value of 1900 ng $g^{-1}$ at site 13. The fraction of total particulate light absorption due to non-BC constituents~~s~~ $f_{non\text{-}BC}^{est}$ is typically ~20-70%, with a median value of 41%. This information along with the lower $\mathring{A}_{tot}$ (2.6) indicates that BC plays the dominant role in influencing the light absorption in this region. 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).~~The vertical profiles of ILAPs in the snow/ice samples were collected in the warm season from 2014 to 2015.~~ In the ~~warm~~ monsoon season, the mixing ratios of ISOC and Fe ranged from 80-10100 ng $g^{-1}$ and 20-340 ng $g^{-1}$, respectively, and the mixing ratios of ILAPs in most of the ~~snow/ice~~ ice samples increased remarkably from the top to the bottom. This result is highly consistent with a previous study by Doherty et al. (2013). Fig. S5 shows that the vertical profiles of the mass mixing ratios of BC, ISOC, and Fe for the ~~snow/ice~~ ice samples in the ~~Tanggula~~Xiaodongkemadi glacier were more complicated than those for the other regions. With the exception of the surface layer at sites 53 and 54, most values of $C_{BC}^{est}$ ranged from 10 to 280 ng $g^{-1}$ in the ~~Tanggula~~Xiaodongkemadi glacier; therefore, this glacier was the cleanest region of all the studied glaciers. 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

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samples.~~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 deposition on the snow/ice surface.~~ Doherty et al. (2013) also found that the ILAPs could be scavenged with the snow meltwater, therefore leading to a much higher concentration of BC in the surface snow. At sites 56-58, $f_{non\text{-}BC}^{est}$ was lower than 38%, and $\mathring{A}_{tot}$ ranged from 1-2.5. These results are consistent with the fossil fuel combustion source due to industrial activities. Because single layer samples are not shown, the vertical profiles of $C_{BC}^{est}$ are plotted in Fig. S6 for all ~~snow/ice~~ ice samples, which were collected in the Qiumianleiketage, Hariqin, and ~~Yangbajing~~Gurenhekou glaciers. Except for the sites in Fig. S6d, S6e, S6i, and S6h, the other sites reveal the trapping and scavenging effects of a higher mass concentration of BC in the surface layer due to the melting process. Previous studies have also illustrated that the ILAPs could become trapped and integrated at the surface of the snowpack due to melting and sublimation to enrich the surface concentrations (Conway et al., ~~2002~~1996; Painter et al., 2012; Doherty et al., 2013~~; Painter et al., 2012~~).

### ~~3.2 Relationship between BC and ISOC~~

~~The largest contribution to both BC and OC emissions is from biomass burning, but biofuel and fossil fuel burning play key roles in influencing the mass concentration of BC and OC (Bond et al., 2004, 2006; Chen et al., 2010; Akagi et al., 2011). The OC/BC ratio could also represent the relative abilities of aerosol light scattering and absorption; therefore, this ratio is an indication that the origins of the carbonaceous particles are due to different emission sources during the transport and deposition processes (Chow et al., 1996; Turpin and Huntzicher, 1995). For instance, Bond et al. (2004) revealed that the higher ratio of OC/BC (7-8) was mainly due to open biomass burning and biofuel combustion rather than fossil fuel combustion. Koch (2009) revealed that an OC/BC ratio of ~4.0 was assumed to indicate emission sources due to fossil fuel combustion, while an OC/BC ratio higher than 12 was due to residential coal combustion (Cao et al., 2005). Further studies have also been performed to analyze the variations in the abundance of OC and BC among sources in the atmosphere such as coal, diesel, gasoline, and wood~~

~~combustion (Watson et al., 1994, 2001). However, only few studies have revealed the mass mixing ratios of OC and BC in the Loess Plateau, especially in the high glacier regions via dry and wet deposition. As illustrated in Fig. 5, the ISOC/BC ratios range from 1 to 18 during the warm season and from 0.05 to 18.4 during the cold season, with median values of 6 and 5, respectively. High ISOC/BC ratios (>3) in all TP glaciers mainly revealed the primary emissions of carbonaceous aerosols due to fossil fuel and biofuel burning. The highest ISOC/BC ratio (18.4) found at site 52 resulted that the emission sources is mainly due to residential coal combustion. Only slight differences were found in the ISOC/BC ratios (6 and 5) between warm and cold seasons, respectively. The lowest values for the ISOC/BC ratios (<2) varied substantially at several sampling sites, suggesting that the natural combustion sources were the dominant factor. In order to better illustrate the spatial distribution of the mixing ratio of the ISOC/BC in snow/ice, the median values of the mixing ratio of ISOC/BC in each glacier is given in Fig. S7. The result indicates that the relative lower mixing ratios of ISOC/BC are found in the southern glaciers, while much higher in the northern glaciers. Furthermore, the ISOC concentration exhibited a strong correlation with BC ($y=4.350x+343.80$, $R^2=0.54$) (Fig. 6), which suggested influences from similar emission sources (e.g., biomass burning and residential and commercial coal combustion).~~

**3.~~3~~ 2 Water-soluble organic carbon**

It is well known that the WSOC component has a large variation ranging from 20% to 99% of total carbonaceous particles (~~Mayol-Bracero et al., 2002;~~ Saxena and Hildemann, 1996; Mayol-Bracero et al., 2002). There is a very large fraction representing the average WSOC (>80%) to TOC, which can absorb solar light and enhance cloud formation through their direct and indirect climate effects (Ram et al., 2010). These results are highly consistent with previous studies showing that fossil fuel combustion plays a key role in leading to the higher fraction of WSOC to total organic carbon (TOC) (~~Andreae and Rosenfeld~~Zhang et al., 2012~~08~~). The concentration of WSOC during the field campaign varied from 400 to 43600 ng $g^{-1}$, with a median of 1400 ng $g^{-1}$ (Table 1). The median values of the WSOC/TOC ratio were 0.85 for the ~~warm~~ monsoon season and 0.9

for the ~~cold~~ non-monsoon season during this field campaign (Fig. ~~7~~6). No remarkable correlation was observed between WSOC and ISOC in both ~~cold and warm~~monsoon and non-monsoon seasons. As WSOC is considered as a stable indicator of the primary biomass burning, with a small contribution of fossil fuel combustion, we indicate that a mixture emission sources from the biomass burning and fossil fuel combustion might be contributing significantly to the TOC concentrations in these TP glacier regions.

### 3.~~4~~ 3 Contributions to particulate light absorption by ILAPs

BC and OC were mainly emitted from biomass burning and biofuel and fossil fuel combustion, while mineral dust was emitted from the local soil or desert regions (Streets et al., 2001; Painter et al., 2007; Chen and Bond, 2010; Bond et al., 2013; Kaspari et al., 2014; ~~Painter et al., 2007; Chen and Bond, 2010; Streets et al., 2001;~~ Pu et al., 2017). The contributions to particulate light absorption by BC, OC, and Fe have been investigated using the ISSW measurements across northern China and North America (Wang et al., 2013; Doherty et al., 2014). The fractional contributions to absorption by BC, ISOC, and Fe at 450 nm in surface ~~high~~ glaciers are shown in Fig. ~~8~~7, and the concentrations of BC, ISOC, and Fe are given in Table S1. BC plays a dominant role in particulate light absorption with values ranging from 20-87% in all ~~high~~ glacier regions. ISOC is the second highest absorber in ~~high~~ glacier regions, and there are large variations of light absorption of ISOC during the field campaign with values ranging from 0.5-58%. The light absorption due to ILAPs in the TP glacier regions is not only from industrial and biomass burning but also with a small contribution from local soil dust. Note that the median fraction of light absorption due to Fe is ~13%, with the highest light absorption of iron being higher than 40% in the ~~Yangbajing~~Gurenhekou glacier. This result is an indication that mineral dust plays a key role in affecting the spectral absorption properties due to the soil and mineral dust at site 67. Although the total light absorption was dominated by BC and ISOC in all selected glaciers, we note that the relative spatial distribution of the total light absorption due to Fe (>17%) also play key roles to affected the snowmelt in the southern glaciers than that of northern glaciers across the TP regions (Fig. ~~S8~~S7).

### 3.~~5~~ 4 Enrichment factor (EF)

Briefly, EF values ranging from 0.1 to 10 indicate significant input from crustal sources. Conversely, EF values that larger than 10 exhibit a major contribution from anthropogenic activities. Referring to the EF analysis (Fig. ~~9~~8), the mean EF of Fe < 5 in each glacier can be assumed to customarily originate from crustal sources. Recent studies have also indicated that light-absorbing particles in snow are dominated by local soil dust in some typical regions over northern China (Wang et al., 2013), and northern America (Doherty et al., 2014). Comparable with Fe, the other trace metals with the mean EF of ⩾5.0 were moderately to highly enriched predominantly from anthropogenic emissions (Hsu et al., 2010). For example, Pacyna (2001) reported that fossil fuel combustion is a major source of Cr. Cu primarily originates from emissions from fossil fuel combustion and industrial processes, while Pb and Zn are known to be drawn from the traffic-related activities and coal burning (Christian et al., 2010; Contini et al., 2014~~; Christian et al., 2010~~). Together with high EF values observed for Cu, Zn, and Cd in our ~~snow/ice~~ ice samples clearly suggested that the TP glaciers have already been polluted by human activities, such as biomass burning, fossil fuel burning, and the coal burning. For instance, high level of Pb and Cr have demonstrated a link to coal combustion (Zhang et al., 2013a, 2013b; Mokhtar et al., 2014~~; Zhang et al., 2013a, 2013b~~), abundant Cu and Cd were associated with traffic-related dusts (Cheng et al., 2010). Therefore, we concluded that the natural dust source and anthropogenic emission source are both non-negligible to the ILAPs in the TP glaciers.

### 3.~~6~~ 5 Source apportionment

Given the importance of the climate effect in our understanding of the ILAPs in TP glaciers, we present a PMF receptor model to analyze the source attribution of ILAPs in these glaciers. In this study, two datasets including the mass concentrations of the chemical components and the ILAPs in ~~snow/ice~~ ice and the associated uncertainty datasets were used to run the PMF 5.0 model. The details of the techniques have already been illustrated by Hegg et al. (2009, 2010) and Pu et al. (2017). The factor loadings

(apportionment of species mass to individual factors) for the 3-factor solution of the source profiles based on the PMF 5.0 model are given in Fig. ~~10~~ 9 (in both measured mass concentration and the % total mass allocated to each factor). It is evident that the first factor (top panel) was obviously characterized by high loadings of $Cl^-$, $CL_{salt}$, $SO_4^{2-}$, and $NO_3^-$, which are well known markers for the urban or local industrial pollutions (Alexander et al., 2015). Although $Cl^-$ to $Na^+$ are usually considered as a potential product of emission source of sea salt, but also a high loading of $Cl^-$ to $CL_{salt}$ indicated another source in addition to sea salt such as industrial emission and coal combustion (Hailin et al., 2008; Kulkarni, 2009). Additionally, the highest loading of $NH_4^+$ is also suggested as an indicator of coal combustion (Pang et al., 2007). Compared with the first factor, the highest loading of Al (90.3%) and Fe (87.3%) are well-known markers for the urban or regional ~~soil~~ mineral dust (Pu et al., 2017). Therefore, the second factor or source profile is easily interpretable as a natural ~~soil~~ mineral dust source. But a notable feature is that the relative high mass loading of $C_{BC}^{max}$ (76.4%) to that of the previously identified ~~soil~~ mineral dust source as reported by Pu et al. (2017). It is well known that $K^+$ and $K_{Biosmoke}$ are the major indicators of biomass burning source (Zhang et al., 2013a). Therefore, it is easily interpretable that the highest loadings of $K^+$ and $K_{Biosmoke}$ are well representative the biomass burning source (Fig. ~~10c~~9c). However, it is also important to note that the lowest mass loading of $C_{BC}^{max}$ in this factor is a bit unexpected. 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 ~~soil~~ mineral dust (Bond et al., 2006). Therefore, we interpreted the third factor normally considered a predominantly biomass burning product. However, the major emission of BC in TP glaciers originated from the local ~~soil~~ mineral dust source instead of the biomass burning and industrial pollution than previous studies (~~Pu et al., 2017;~~ Zhang et al., 2013a; Pu et al., 2017).

Finally, the chemical composition and mean source apportionment of the ILAPs to the three sources in the TP glaciers were given in Fig. ~~11~~10. Note that the apportionment is of the light absorption by insoluble particles in the surface glaciers. On average, the source appointment of the ILAPs in all TP glaciers by ~~soil~~ mineral dust is close to 37.5%, while

the industrial emission and biomass burning contributes 33% and 29.4%. Specifically, the largest biomass burning contribution of the light-absorption of ILAPs was found in the Qiyi glacier, which is close to the human activity regions (Guan et al., 2009; Li et al., 2016). In the Meikuang, Qiumianleiketage, ~~Yangbajing~~Gurenhekou, and ~~Tanggula~~Xiaodongkemadi glaciers, the ~~soil~~ mineral dust contribution of light absorption is much larger (>47.9%) than that of industrial pollution and biomass burning, especially in the Meikuang glacier. In these regions, the percent of the MD light absorption is ranging from 20.4-31.1%, while the light absorption by biomass burning is in the range of 18.5-35.8%. Industrial pollution constitutes a major fraction in the Yuzhufeng glacier. Chemical analysis shows that the percentages of the chemical species in the Yuzhufeng and Meikuang glaciers are much similar. The attribution of the total anions by chloride, nitrate, and sulphate is higher 52% and 48% than the other chemical species in the Yuzhufeng and Meikuang glaicers. In the Hariqin glacier, the largest attribution of the sulphate is up to 45.4%. As shown in Fig. ~~11~~10, the source apportionment of the light absorption by insoluble particles in the surface glaciers is dominated by ~~soil~~ mineral dust and the industrial pollution in most glaciers, only with a large fraction of the light absorption due to biomass burning in the Yuzhufeng glacier. These results are highly consistent with the previous studies (Andersson et al., 2015~~; Li et al., 2016~~). They found that the contributions of coal-combustion-sourced BC are the most significant for the TP glaciers. Based on the model simulations, Zhang et al. (2015) revealed that the largest contribution to annual mean BC burden and surface deposition in the entire TP regions is from biofuel and biomass (BB) emissions in South Asia, followed by fossil fuel (FF) emissions from South Asia.

## 4 Conclusions

In this study, the ILAPs observations in 7 ~~high~~ glacier regions across the Tibetan Plateau are presented using the ISSW technique along with chemical analysis. ~~Approximately~~ 67 vertical profiles of ~~snow/ice~~ ice samples are analyzed during the ~~warm and cold~~monsoon and non-monsoon seasons from 2013-2015. There are no apparent differences in the mixing ratios of ILAPs in the ~~snow/ice~~ ice samples during seasonal transitions. The

results indicate that the variations of $Å_{tot}$ and $Å_{non\text{-}BC}$ for all ~~snow/ice~~ ice samples range from 1.4-3.7 and 1.9-5.8, respectively, excluding site 16. The lower absorption Ångström exponent ($Å_{tot}$<2) suggested that the sites 30, 51, and 56-58, 65 were primarily influenced by fossil fuel emission, whereas the rest of the sites were heavily influenced by mineral dust and biomass burning. Another notable feature is the large variation of the ILAPs in the ~~snow/ice~~ ice samples. By excluding some of the highest ILAPs values in the ~~snow/ice~~ ice samples, the values of $C_{BC}^{est}$, $C_{ISOC}$, and $C_{Fe}$ range from 100-1000 ng $g^{-1}$, 10-2700 ng $g^{-1}$, and 10-1000 ng $g^{-1}$, respectively. Among the samples, the lower concentrations of BC were found in the ~~Tanggula~~Xiaodongkemadi, Hariqin and ~~Yangbajing~~Gurenhekou glaciers, with the median concentrations of 33 ng $g^{-1}$, 24 ng $g^{-1}$, and 28 ng $g^{-1}$, respectively. ~~The large contribution of high ISOC/BC ratios (>3) at all high glacier sites mainly indicated that the primary emissions of carbonaceous aerosols were due to fossil fuel and biofuel burning.~~

~~BC and ISOC play dominant roles in particulate light absorption, with values ranging from 20-87% and 0.5-58%, respectively, in all high glacier regions. However, the relative spatial distribution of the total light absorption due to Mineral dust also plays key roles to affect the snowmelt in the TP glaciers. In this study, w~~We also present a PMF receptor model to analyze the source attributions of ILAPs in these glaciers. We found that the anthropogenic air pollution is much heavy across the northern glaciers due to human activities, but the major emissions of the light absorption by insoluble particles in TP glaciers originated from the local ~~soil~~ mineral dust and industrial pollution sources, followed by the biomass burning source. We note that the $C_{BC}^{max}$ is not only attributed to the biomass burning emission, but also associated with the industrial activities associated with the local ~~soil~~ mineral dust. Therefore, the natural mineral dust source and anthropogenic emission source are both non-negligible to the ILAPs in the TP glaciers.

## 5 Data availability

All datasets and codes used to produce this study can be obtained by contacting Xin Wang (wxin@lzu.edu.cn).

*Competing interests.* The authors declare that they have no conflicts of interest.

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**Table 1**. Statistics of the ~~ice core~~ice variables measured using an ISSW for each glacier.

| Region | Latitude | Longitude | | $C_{BC}^{equiv}$ | $C_{BC}^{max}$ | $C_{BC}^{est}$ | $f_{non\text{-}BC}^{est}$ | $\mathring{A}_{tot}$ | ISOC | WSOC | Fe |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | (N) | (E) | | (ng g$^{-1}$) | (ng g$^{-1}$) | (ng g$^{-1}$) | (%) | | (ppm) | (ppm) | (ppb) |
| Qiyi glacier | 39°14'28" | 97°45'27" | average | 414 | 299 | 238（116, 313） | 42（15, 66） | 2.59 | 1.21 | 5.43 | 181.3 |
| | | | median | 176 | 128 | 94（29, 124） | 41（17, 70） | 2.62 | 0.66 | 2.25 | 93.94 |
| | | | minimum | 26 | 29 | 25（13, 35） | 21（—, 53） | 0.8 | 0.08 | 0.62 | 19.8 |
| | | | maximum | 2651 | 2230 | 1877（1182, 2109） | 73（41, —） | 3.73 | 11.59 | 43.55 | 2414 |
| Qiumianleiketage | 36°41'47" | 90°43'44" | average | 421 | 296 | 238（139, 402） | 44（24, 81） | 2.80 | 1.43 | 1.99 | 231.7 |
| | | | median | 307 | 215 | 172（64, 218） | 44（24, 81） | 2.76 | 1.06 | 2.23 | 184 |
| | | | minimum | 139 | 93 | 62（19, 93） | 37（12, 64） | 2.45 | 0.54 | 1.17 | 105.9 |
| | | | maximum | 995 | 662 | 558（143, 678） | 56（27, 86） | 3.08 | 3.97 | 2.56 | 625.2 |
| Meikuang glacier | 35°40'24" | 94°11'10" | average | 493 | 328 | 260（119, 331） | 42（15, 37） | 2.65 | 2.14 | 2.80 | 218.8 |
| | | | median | 197 | 156 | 133（76, 153） | 44（16, 69） | 2.64 | 0.61 | 3.05 | 125.5 |
| | | | minimum | 24 | 23 | 19（17, 24） | 16（—, 17） | 1.37 | 0.13 | 0.62 | 32.24 |
| | | | maximum | 4696 | 2817 | 2292（109, 2938） | 62（23, 85） | 3.56 | 16.89 | 8.06 | 1224 |
| Yuzhufeng glacier | 35°38'43" | 94°13'36" | average | 457 | 312 | 233（94, 295） | 51（—, 37） | 2.84 | 1.51 | 2.85 | 438.4 |
| | | | median | 317 | 201 | 160（116, 204） | 48（26, 87） | 2.95 | 1.02 | 2.76 | 212.3 |
| | | | minimum | 52 | 35 | 24（8, 35） | 15（—, 37） | 1.82 | 0.07 | 0.8 | 45.1 |
| | | | maximum | 2630 | 1608 | 1169（72, 1603） | 110（6, 49） | 3.7 | 9.16 | 5.92 | 3513 |
| Hariqin glacier | 33°08'23" | 92°05'34" | average | 476 | 327 | 256（100, 385） | 48（26, 82） | 2.79 | 1.59 | 1.67 | 171.2 |
| | | | median | 54 | 37 | 23（9, 30） | 48（26, 82） | 2.87 | 0.22 | 1.08 | 52.15 |
| | | | minimum | 36 | 24 | 13（4, 22） | 19（—, 41） | 1.96 | 0.08 | 0.72 | 26.69 |
| | | | maximum | 3990 | 2702 | 2131（682, 2784） | 64（32, 84） | 3.52 | 9.64 | 4.92 | 1049 |
| Xiaodongkemadi | 33°04'08" | 92°04'24" | average | 253 | 171 | 152（76, 177） | 37（15, 63） | 2.28 | 0.95 | 1.82 | 173.0 |

| Region | Latitude | Longitude | | $C_{BC}^{equiv}$ | $C_{BC}^{max}$ | $C_{BC}^{est}$ | $f_{non\text{-}BC}^{est}$ | $\mathring{A}_{tot}$ | ISOC | WSOC | Fe |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | (N) | (E) | | (ng g$^{-1}$) | (ng g$^{-1}$) | (ng g$^{-1}$) | (%) | | (ppm) | (ppm) | (ppb) |
| | | | median | 62 | 47 | 53（37, 65） | 36（13, 59） | 2.18 | 0.19 | 1.41 | 62.08 |
| | | | minimum | 13 | 12 | 9（6, 18） | 8（—, 19） | 1.08 | 0.01 | 0.45 | 10.22 |
| | | | maximum | 2770 | 1849 | 1637（596, 2031） | 86（25, 90） | 3.63 | 6.97 | 5.91 | 2129 |
| Gurenhekou glacier | 30°11’17” | 90°27’23” | average | 382 | 292 | 247（212, 591） | 46（16, 71） | 2.42 | 0.62 | 1.21 | 182.1 |
| | | | median | 61 | 46 | 30（19, 44） | 48（18, 75） | 2.46 | 0.13 | 0.85 | 97.99 |
| | | | minimum | 28 | 23 | 15（10, 24） | 27（7, 52） | 1.34 | 0.02 | 0.41 | 31.51 |
| | | | maximum | 4674 | 3634 | 3080（1876, 3884） | 61（26, 85） | 2.92 | 5.22 | 4.65 | 911.2 |

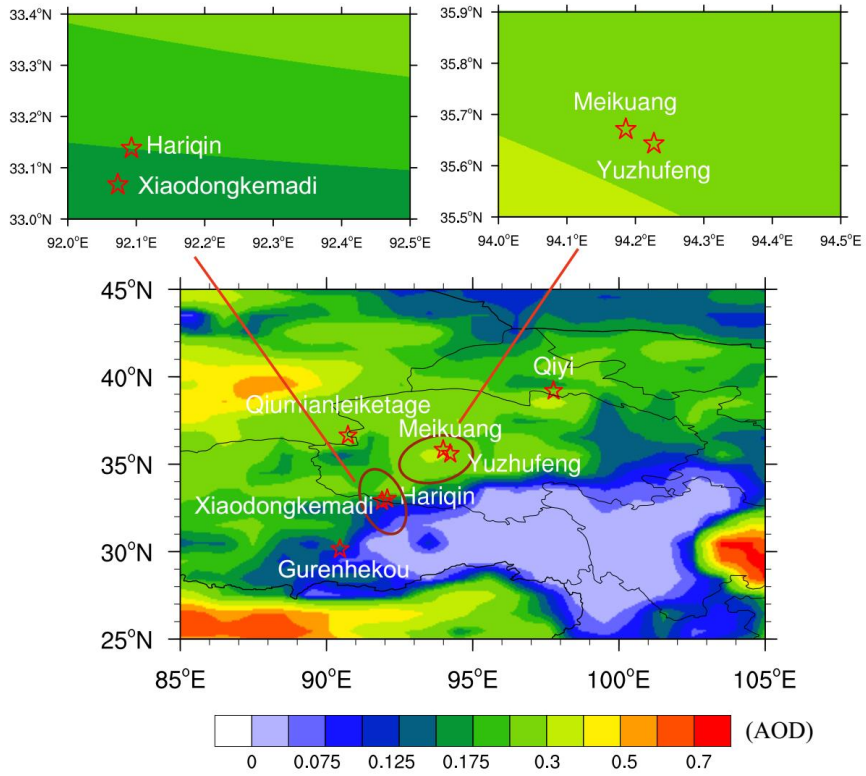


**Figure 1.** Spatial distribution of the averaged AOD retrieved from Aqua-MODIS over Tibetan Plateau from 2013 to 2015, and larger values represent higher optical depth and smaller values represent smaller optical depth. The red stars are the sampling locations (see also Table 1): 1, Qiyi glacier (97.76° E, 39.24° N, 4850 m a.s.l); 2, Qiumianleiketage glacier (90.73° E, 36.70° N, 5240 m a.s.l); 3, Meikuang glacier (94.19° E, 35.67° N, 4983 m a.s.l); 4, Yuzhufeng glacier (94.23° E, 35.65° N, 5200 m a.s.l); 5, Hariqin glacier (92.09° E, 33.14° N, 5100 m a.s.l); 6, Xiaodongkemadi glacier (92.07° E, 33.07° N, 5600 m a.s.l); 7, Gurenhekou glacier (90.46° E, 30.19° N, 5655 m a.s.l).

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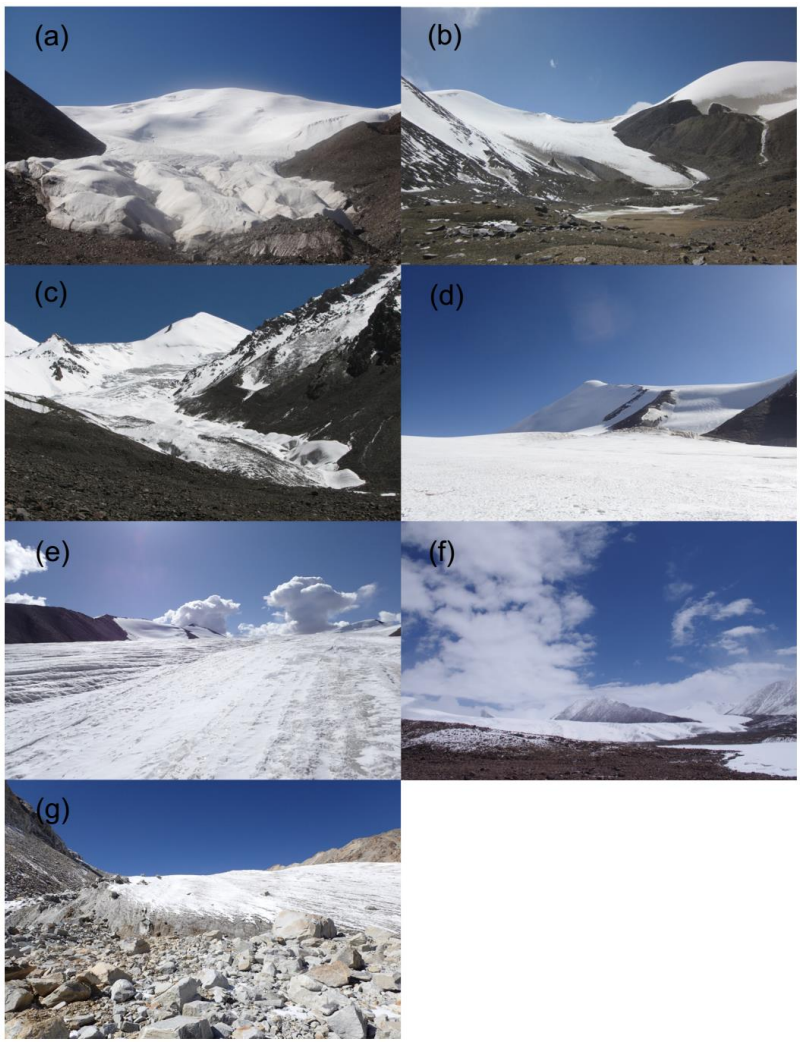


**Figure 2.** Pictures of ice sampling location in the (a) Qiyi, (b) Qiumianleiketage, (c) Meikuang, (d) Yuzhufeng, (e) Hariqin, (f) Xiaodongkemadi, (g) Gurenhekou glaciers, respectively.

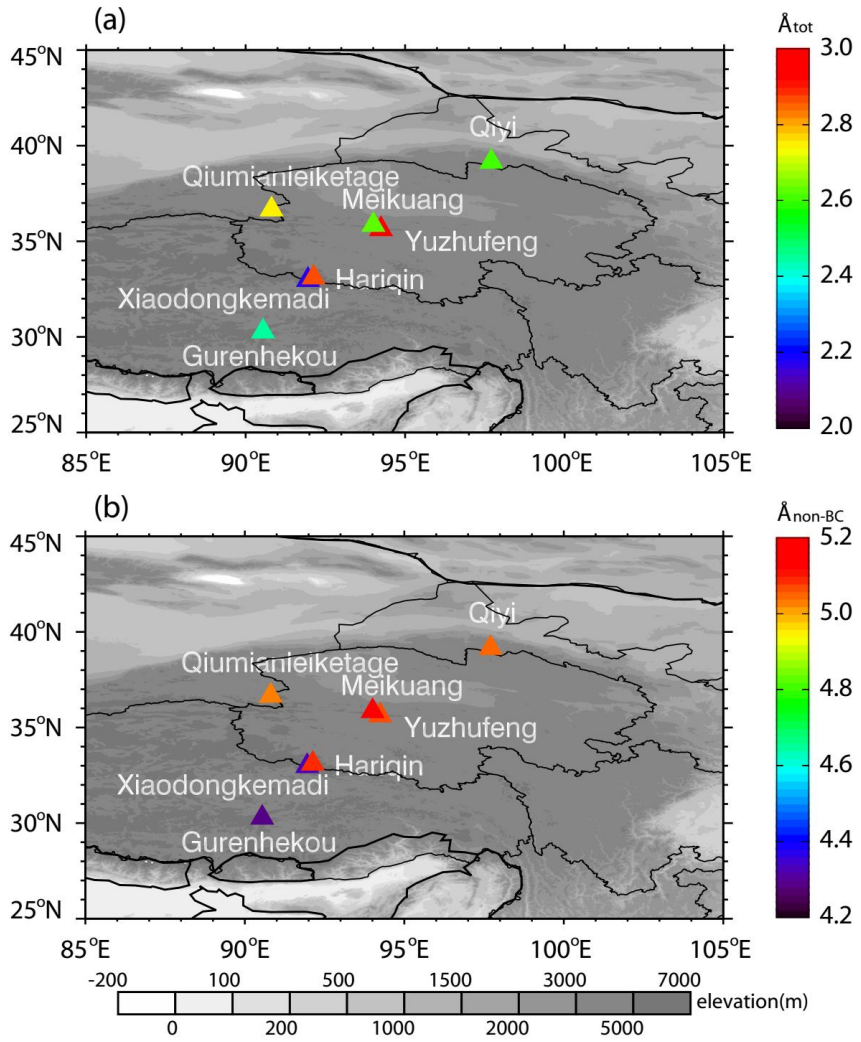


**Figure ~~23~~23.** The spatial distribution of the median absorption Ångström exponent for (a) total particulate constituents ($Å_{tot}$), and (b) non-BC particulate constituents ($Å_{non\text{-}BC}$) in each glacier (see Fig. 1).

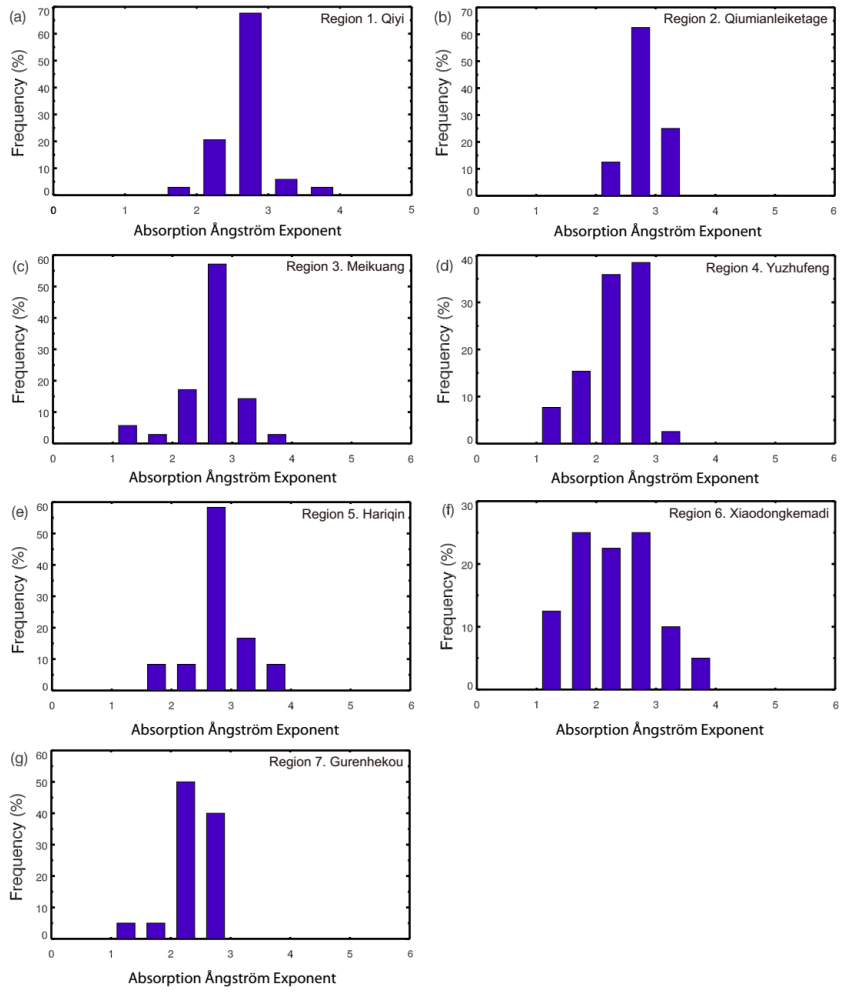


**Figure ~~3~~4.** Histograms of the frequency of $\mathring{A}_{tot}$ (450-600 nm) for ~~snow/ice~~ ice samples in each of the glacier region. Samples from all vertical profiles are included.

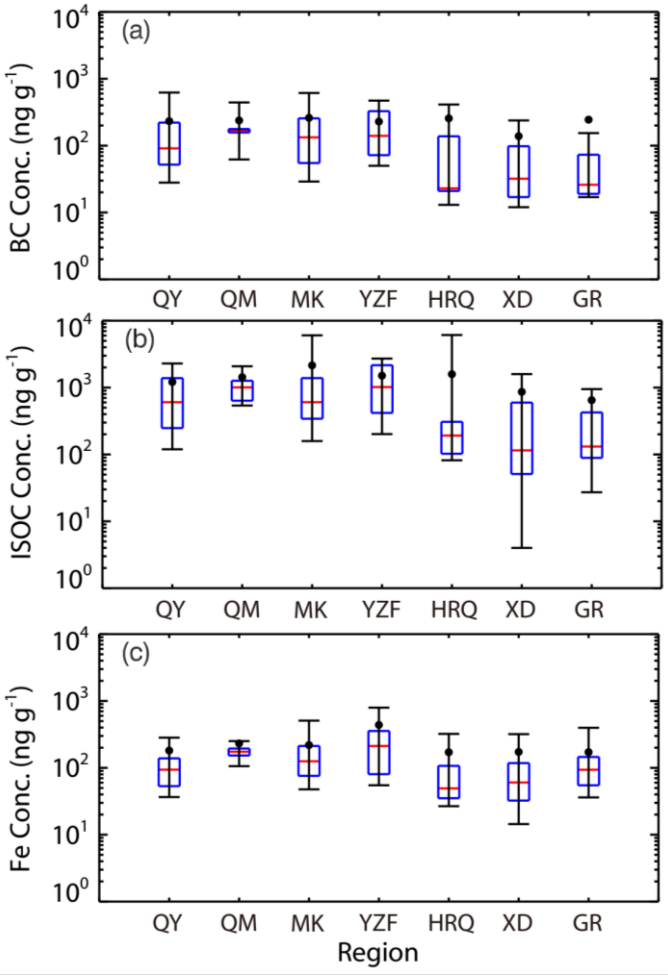


**Figure 45.** Box plots of the region variations in (a) BC concentration, (b) ISOC concentration, and (c) Fe concentration of the seven glaciers. QY, QM, MK, YZF, HRQ, XD, and GR represent the following glaciers: Qiyi, Qiumianleiketage, Meikuang, Yuzhufeng, Hariqin, Xiaodongkemadi, Gurenhekou glaciers, respectively. Error bars are 10$^{th}$, 25$^{th}$, median, 75$^{th}$, and 90$^{th}$ percentiles of the data. The dot symbol represents the average concentrations of the ILAPs in ~~ice core~~ice samples in each glacier.

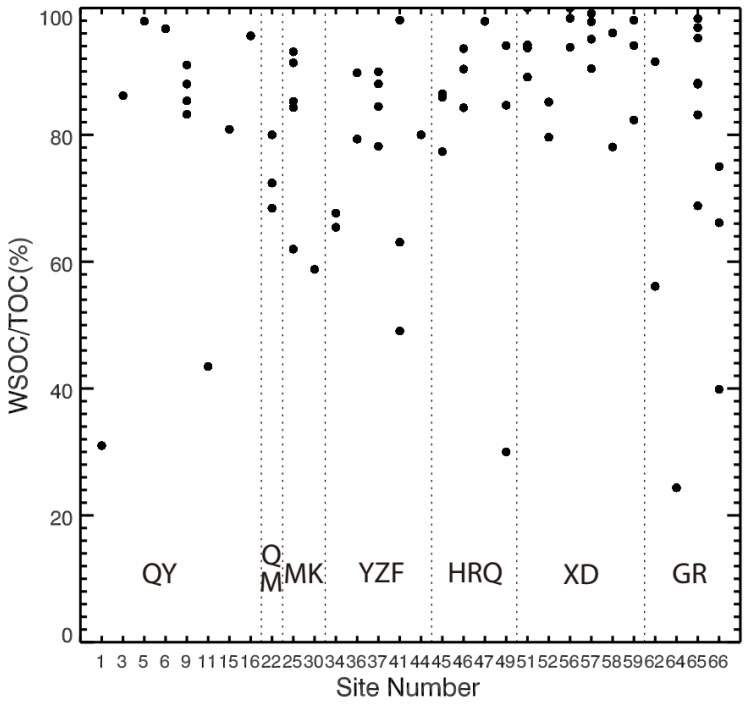


**Figure ~~5~~6.** The mass fraction of WSOC to TOC of each ~~snow/ice~~ ice sample.

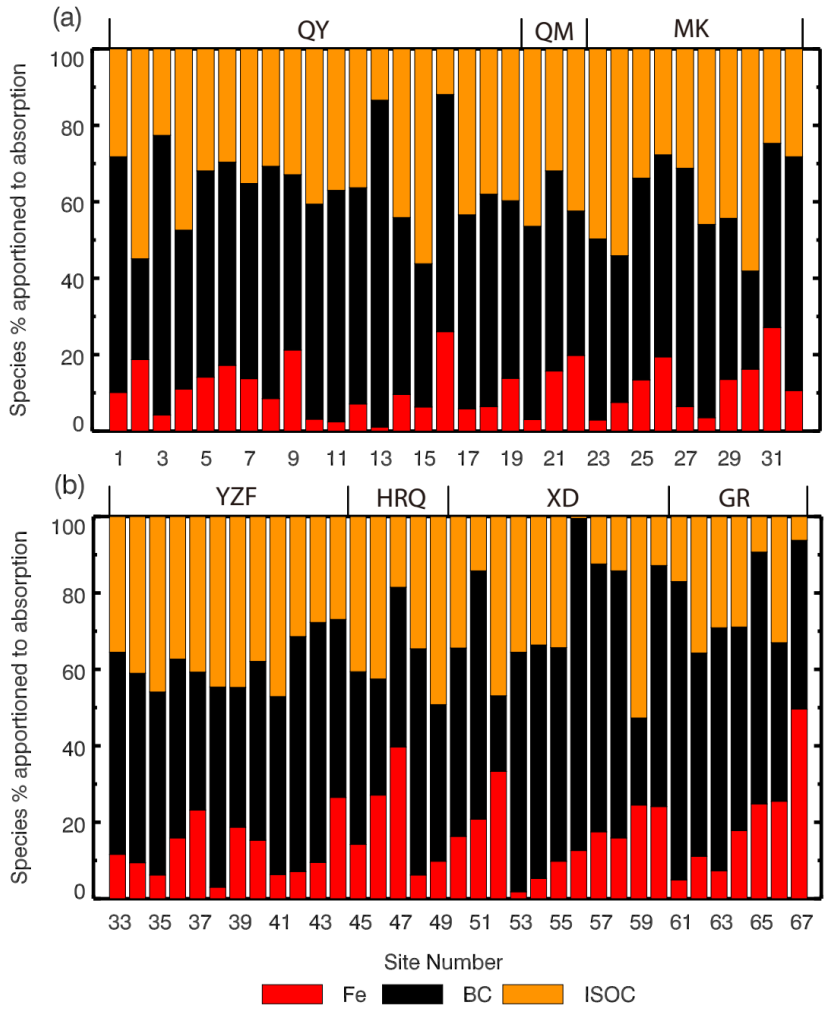


**Figure 67.** Relative contributions to total light absorption by BC, ISOC, and Fe oxide (assumed to be in the form of goethite) for surface ~~snow/ice~~ ice samples in each sampling site (See Table 1).

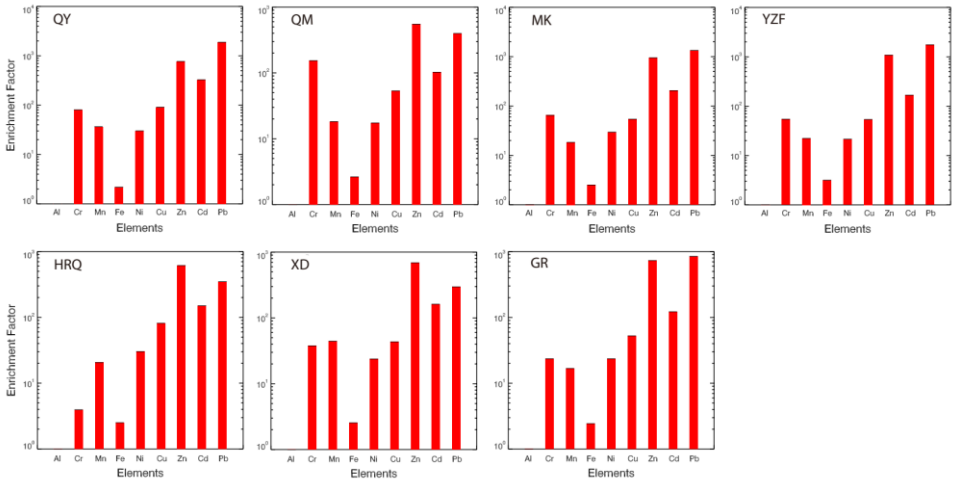


**Figure ~~7~~8.** Average enrichment factors of trace metals in surface ~~snow/ice~~ ice samples at each region.

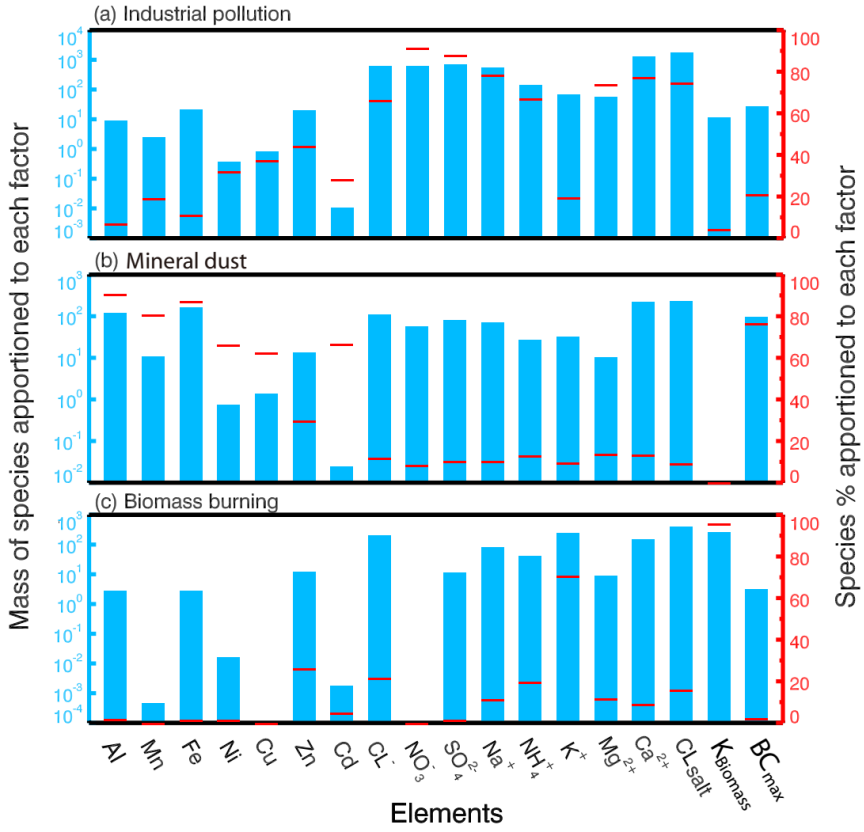


**Figure ~~8~~ 9.** Source profiles for the three factors/sources that were resolved by the PMF 5.0 model.

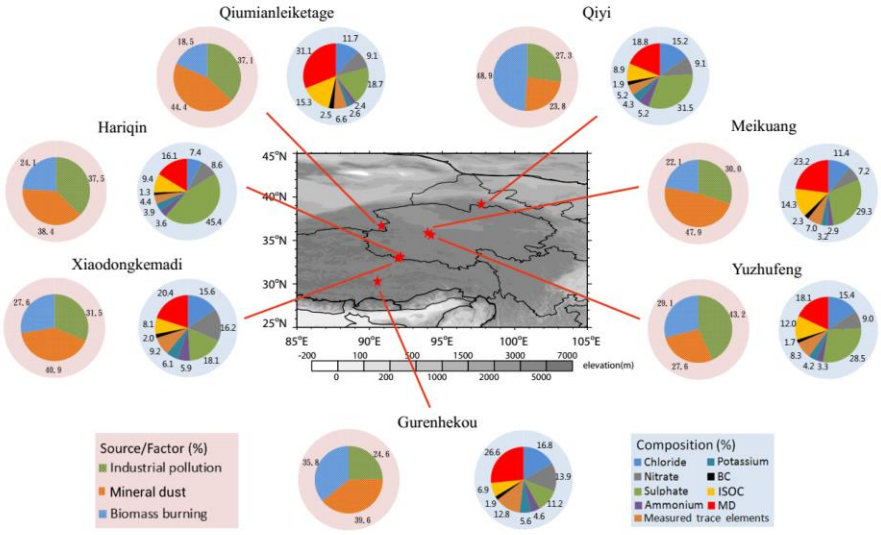


**Figure ~~9~~10.** Chemical composition and source apportionment for the seven glaciers in the TP regions. Note that the apportionment is of the light absorption by insoluble particles in the surface glaciers.

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