- 1 We are very grateful for the editor and reviewers and appreciate your comments and suggestions. All
- 2 responses or changes have been made below. The responses are marked blue.
- 3 Thank you very much
- 4 Kind regards,
- 5 Fangping Yan
- 6 (on behalf of the co-authors)

7 Reply to Anonymous Referee No.1

8 1. It is assumed that the strong relationship between Ca²⁺ and DOC (Fig. S3) reflects a primary source
9 for Ca²⁺ and DOC from the same allochthonous source. Could not the DOC however be produced by
10 later autochthonous or heterotrophic biological activity within the snowpack/ice surface, catalysed by
11 nutrients associated with the dust? This should be at least discussed, and abstract and concluding
12 statements adjusted accordingly.

13 Response: Thanks for the meaningful suggestion. Yes, DOC may also be produced by these activities.

We discussed these potential sources in the section 3.2. The abstract and concluding statements wereadjusted accordingly.

16 2. How was discharge measured? You give the discharge data in the supplementary info, but you need

to either a) give details of methods used to discharge at the gauging station or b) cite a reference forthis data.

- 19 Response: Thank you for your advice. The discharge measurement of LHG glacier has been discussed 20 (Gao et al., 2014). In detail: The hydrological gauging site was setup at about 0.8 km downstream of 21 the glacier terminus. It meets the requirements for a hydrological gauging site. Horizon walls were built 22 on the both sides of the river, and an automatic barometric sensor (HOBO Water Level Logger, Onset, 23 America) was installed in the wall to record water pressure every 10 minutes to calculate the water 24 levels. There was a bridge across the river to facilitate the flow velocity measurement using propeller 25 blade current meter (Model LS25-1, Huazheng Hydrometric Instrument Ltd). The river channel was 26 divided into nine segments in which flow velocity and water depth were measured. Coupled with mean 27 flow velocity, width of each segment and water depth, discharge at specific water level was obtained. 28 By including maximum and minimum water level in a year, a discharge relationship with water levels 29 was developed. Therefore, using the HOBO water lever record, discharge of all seasons was calculated. 30 This part was added in the supplementary information. Below is the picture of the gauging station of
- 31 the glacier.





- 34 samples
- 35 Response: The sum of total samples has been changed to numbers of different individual samples.
- 36 4. Line 82-how was ice sampled-using an ice axe? Shallow drill? To what depth? Were they also
- 37 collected in the same plastic bottles after crushing?
- **38** Response: The 0-3 cm and 3-5 cm ice was sampled using a pre-cleaned ice axe and collected in the 125
- 39 mL pre-cleaned polycarbonate bottles after crushing similar with other samples, this information was
- 40 added in section 2.2.
- 41 5. Line 115–how were the plastic bottles cleaned?
- 42 Response: The plastic polycarbonate bottles were firstly cleaned by ultrapure water for three times,
- 43 then soaked into 1 M HCl for 24 h (Spencer et al., 2009), after that washed for three times using
- ultrapure water, finally soaked into ultrapure water for over 24 h. This information was added into themethod part 2.2.
- 46 6. Line 162–I'd use pre-combusted or pre-baked rather than pre-burned.
- 47 Response "Pre-burned" has been changed to "pre-combusted" through the whole text.

48 7. Line 164-I am unclear as to the methodology here. You state that the experimental samples are first filtered through 0.7um nominal filters, then incubated. But won't the filtration remove much of the 49 biological activity? Plus isn't refrigerating the already filtered samples to act as controls effectively the 50 51 same as the non control samples? Do you mean instead that samples were actually incubated prior to 52 filtration, and then filtered at each time point then refrigerated? If so, please rewrite. And if so, please 53 state the values of controls and place them on Fig 3. Note that you are unlikely to remove all microorganisms when filtering through the nominal 0.7um filters, so they shouldn't be expected to be 54 55 sterile i.e. the initial samples may also have had some biological activity, hence the BDOC values 56 should be seen as minimum values.

- 57 Response: Thanks for the suggestions. Although it might remove some part of the biological activity,
- this method was used in previous researches (e.g. Spencer et al., 2014). We adopted this method for the purpose of easy comparison with previous results. In detail, we filtered the samples, then started the experiment: firstly, refrigerated the two filtered original samples, other samples were put in the outside natural environment, and every 3 days 2 samples were put into the refrigerator to keep frozen till analysis. The control value was added on Figure 3. According to the other reviewer, Figure 3 was changed to Figure 4 as below.





Figure 4 Exponential deceases in DOC concentrations during the biodegradation experiment. Note: The blue point
 is calculated using equations derived from the experimental data (black point). Mean values ± standard deviations

- 67 of duplicate treated samples are presented.
- 68 8. Line 177–DOC could also be influenced by microbial activity–see point 1 above.
- 69 Response: We agree that DOC was influenced not only by the mineral dust but also the microbial
- 70 activities. "Microbial activity" has been added in the sentences.
- 71 9. Line 189–again, should mention potential biological activity here
- 72 Response: We added the potential biological activity in the sentence.
- 73 10. Line 198–you estimate from extrapolation that 43.2% DOC could be re-mineralized within 28 days.

How does 28 days compare with the likely residence time of supraglacial runoff and river runoff,

75 where will the water be in this time-still in a river, or lake, could the DOC survive long enough to 76 impact additional downstream ecosystems?

- 77 Response: Supraglacial runoff of LHG glacier No. 12 is the headwater of Xiaochangma River, which
- 78 disappears as underground water at the mouth of LHG valley and appears as the spring downstream,
- 79 flowing into Shule River in the place of Changma. Therefore, DOC can survive long enough to impact
- the downstream ecosystem.
- 81 11. Line 205–I don't think Anesio et al 2009 looked at viruses. A good additional reference for viruses
- 82 would be Bellas et al 2012 'Viral impacts on bacterial communi-ties in Arctic cryoconite' Env Res Lett
- 83 8 http://iopscience.iop.org/article/10.1088/1748-9326/8/4/045021.
- Response: Reference "Bellas et al., 2013" was added in the text, and the section of DOC sources wasrewritten according to your point 1.
- 12. I found that the number of acronyms made it harder to read. For those used only a couple of times
- 87 (e.g. BrC for brown carbon, WSOC for water soluble organic matter). I'd write them out in full each
- time simply to aid readability.
- 89 Response: These acronyms were rewritten in full name in the text.
- 90 13. There are too many decimal points e.g. in abstract 6,949.4 kg (line 27) should be rewritten as 6,950
- 91 kg; 425.8 (line 26) should be rewritten as 426. And the same throughout the main text and
- 92 supplementary information.
- 93 Response: Adjusted accordingly throughout the main text and supplementary information.
- 94 14. Figures:

- 1) Fig. 2. Add in error bars both ways, plus put n = x under each bar for sample numbers.
- 96 Response: Adjusted accordingly.



- 98 Figure 2 Average DOC concentrations of ice, snow and proglacial streamwater for LHG glacier.
- 99 2) Fig. 3. I would have thought that the relationship here could also be adequately described by a linear
- 100 regression. Also, need to put control (refrigerated) values on here.

- 101 Response: The control values were added on the Figure 3. Yes, based on the data only, the linear
- regression could be adequate. However, according to the DOC bioavailablity, the exponential one can
- be more authentic (Spencer et al., 2015), because some DOC are bio-refractory, so that DOC cannot
- reach zero with long enough resident time. The modified figures are shown on Figure 3, and this Figure
- 105 is now Figure 4 based on the other reviewer's comment as below.





Figure 4 Exponential deceases in DOC concentrations during the biodegradation experiment. Note: The blue point
 is calculated using equations derived from the experimental data (black point). Mean values ± standard deviations
 of duplicate treated samples are presented.

- 110 3) Fig. 4. Would help to put some lines as dotted/dashed (when printing in black+white)
- 111 Response: Yes, the line types have been changed in Figure 4, and the spectrum of desert sand was
- deleted according to the other reviewer's comment. Figure 4 is now changed to Figure 5 in the main
- text as below.



- 115 Figure 5 Absorption spectra for the DOC in snow and ice of LHG glacier and the dust and desert sand from
- surrounding areas.

- 4) Fig. 5. Again, be better to have one line dotted or dashed plus have different symbols to aid
- 118 interpretation when printing in black + white.
- 119 Response: The line and symbol types have been changed, and this Figure is now changed to Figure 6 in
- 120 the main text based on the other reviewer's comment.



122 Figure 6 Comparison of DOC concentrations (A) and MAC365 (B) between surface and subsurface ice.

123 5) Fig. 6. How was discharge calculated and smoothed from raw data?

- 124 Response: By measuring water levels and flow velocity in different parts of the river channel in
- 125 different seasons, a relationship between discharge and water levels was developed using the minimum,
- 126 maximum and usual water levels. Coupled with HOBO water lever record, discharge of whole ablation
- 127 period was calculated. Detained method was added in the supplementary information.
- 128 6) Fig. S2. Typo–should be elevation, not evelation
- 129 Response: Has been corrected.
- 130 7) Fig. S4–use different symbols for ice and snow to aid readability.
- 131 Response: The symbols are changed to recognize easily in Figure S4, and now it is Figure S3 based on
- the other reviewer's comment as below.





- 135 15. Tables:
- 136 1) Table 1. Use 3 sig figures throughout (e.g. 332.4 should be 332)
- 137 Response: Adjusted accordingly.
- 138 2) Table 2. Footnote unclear
- 139 Response: Very sorry for the mistake. The footnote was rewritten.
- 140 3) Table S1. Please clarify resolution–e.g. for snowpack I presume it is vertical resolution, for ice I
- 141 presume horizontal distance on glacier, or is it calculated vertical distance?
- 142 Response: Yes, for snowpack it is vertical resolution, for surface ice and snow it is elevation interval143 (horizontal distance).
- 4) Table S2 BK2 (top line) is out of line. Plus would be better to replace BK numbers with date to
- show how they encompass the time of study. Could also include the mean+STDEV at bottom.

146	Response: BK numbers have been adjusted accordingly and the mean+STDEV was added at bottom.
147	
148	References
149	Gao, X., Xie, X., Qin, X.: Analysis on a floor happened at the No. 12 glacier in Laohugou Valley, Qilian Mountain
150	in June 2013, Journal of Northwest Normal University: Natural Science, 50, 88-91, 2014 (in Chinese with
151	English abstract).
152	Spencer, R. G., Stubbins, A., Hernes, P. J., Baker, A., Mopper, K., Aufdenkampe, A. K., Dyda, R. Y., Mwamba, V.
153	L., Mangangu, A. M., and Wabakanghanzi, J. N.: Photochemical degradation of dissolved organic matter and
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155	2009.
156	Spencer, R. G. M., Guo, W., Raymond, P. A., Dittmar, T., Hood, E., Fellman, J., and Stubbins, A.: Source and
157	biolability of ancient dissolved organic matter in glacier and lake ecosystems on the Tibetan Plateau, Geochim.
158	Cosmochim. Acta, 142, 64-74, 2014.
159	Spencer, R. G., Mann, P. J., Dittmar, T., Eglinton, T. I., McIntyre, C., Holmes, R. M., Zimov, N., and Stubbins, A.:
160	Detecting the signature of permafrost thaw in Arctic rivers, Geophys. Res. Lett., 42, 2830-2835, 2015.
161	
162	
163	Reply to Anonymous Referee No.2
164	This study examines radiative forcing of dissolved organic carbon in snow and ice and its contribution

to carbon flux returned to the atmosphere using samples collected form the Laohugou glacier No. 12 165 (LHG glacier)) in the north-eastern Tibetan Plateau. Radiative forcing is very small (0.1_0.1%) in 166 167 comparison with black carbon in ice but constitutes about 10% of the black carbon forcing in snow 168 although the uncertainty on this estimation is close to the estimation itself. I suggest that the authors 169 comment on the importance of this forcing carefully given that these are not large figures. Figure 6S and text given in lines 256-258 are confusing as they suggest much higher radiative forcing of 170 171 dissolved organic carbon. This should be explained more clearly. I also suggest that the two themes -172 radiative forcing and release of carbon from the glacier into the atmosphere should be given more 173 distinct separation in the text and the latter given more prominence that it has now (a very short section 174 3.4).

175 Response: Thanks a lot for the suggestions. Sorry for the confusing statement about Figure 6S and lines 176 256-258. Figure 6S was intended to explain the relatively higher radiative forcing of snowpit compared 177 to glacier ice. Lines 256-258 were deleted from the text. For section 3.4, we combined DOC 178 concentration of proglacial streamwater collected previously across the TP (Table S2) to estimate the 179 total output of DOC from glacier region of the TP. The result showed that about 12.7-13.2 Gg DOC 180 $(Gg=10^9 \text{ g})$ was exported from glaciers of the TP, which was higher than that of DOC deposition in the glacier region (Li et al., 2016), indicating glacier of the TP is a carbon source at present environment 181 182 condition. Therefore, we extend this part of our MS to glaciers of the entire TP.

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Table S2 Information of the studied glaciers and DOC concentrations (µg L⁻¹) of proglacial streamwater samples

across the TP.						
Classier ID	Classical and	Mountain rongo	DOC (monsoon)	DOC		
	Glacier hame	Wouldani Talige	DOC (monsoon)	(non-monsoon)		
LHG	Laohugou glacier No. 12	Qilian	325	394		
TGL	Xiaodongkemadi glacier	Tanggula	150	212		
EV	East Rongbu glacier	Middle Himalaya	139	171		
ZD	Zhadang glacier	Nyainq êntanglha	169	222		
DM	Demula glacier	Eastern	102	124		
DIVIL		Himalaya	105	134		
QY	Qiangyong glacier	Central Himalaya	124	167		

191 Lines 82-83 and lines 102-107: I suggest that Supplement Table 1 should be given together with Fig. 1

in the main text. Both snow pits should be shown on Fig. 1. Or were they in the same place? If yes,clarify in the text (line 103).

194 Response: Yes, the two snowpits are almost in the same site and marked on Figure 1. Supplement

195Table 1 was moved into the main text.

196 Lines 82-83 and Section 2.2: How did you measure discharge? Explain

197 Response: Thanks a lot for this question, which was also asked by the other reviewer. In detail: The 198 hydrological gauging site was setup at about 0.8 km downstream of the glacier terminus. It meets the 199 requirements for a hydrological gauging site. Horizon walls were built on the both sides of the river, 200 and an automatic barometric sensor (HOBO Water Level Logger, Onset, America) was installed in the 201 wall to record water pressure every 10 minutes to calculate the water levels. There was a bridge across 202 the river to facilitate the flow velocity measurement using propeller blade current meter (Model LS25-1, 203 Huazheng Hydrometric Instrument Ltd). The river channel was divided into nine segments in which 204 flow velocity and water depth were measured. Coupled with mean flow velocity, width of each 205 segment and water depth, discharge at specific water level was obtained. By including maximum and 206 minimum water level in a year, a discharge relationship with water levels was developed. Therefore, 207 using the HOBO water lever record, discharge of all seasons was calculated. This part was added in the 208 supplementary information.

Line 105 and Fig. 1: What is the 'eastern tributary'? Is it a tributary of the glacier or of the stream? It isnot clear from Fig. 1.

Response: Sorry for the missing of the description of this glacier. "Eastern tributary" is one of the
branches of LHG glacier No. 12 (Figure 1). "It is divided into two parts of western and eastern branch
at the elevation of 4560 m a.s.l (Dong et al., 2014)" has been added in section 2.1.

Line 105: How did you collect ice samples? How did you store them? Crushed or melted before placing in a bottle?

216 Response: Surface ice (0-3 and 3-5cm) samples were collected using an ice axe directly into 125 mL

pre-cleaned polycarbonate bottles after crushing. This method of sample collection was added insection 2.2.

Line 105: Were your samples collected from the surface?

220 Response: Yes, they were collected from the surface. "71 snow/ice samples" was changed to "29

surface snow and 42 surface ice samples".

Line 105: Please clarify how many samples of snow or ice collected. You currently give one number

223 for all.

224 Response: "71 snow/ice samples" was changed to "29 surface snow and 42 surface ice samples".

Lines 110-111: 'Clean Hands –Dirty Hands' procedure: Please explain in plain English, avoiding
 jargon, what it is.

Response: "Clean Hands-Dirty Hands" is the sample collection protocol in which the person who takes
charge of the sample collection should not touch any other things except the samples to avoid
contamination, the hands are "clean hands", while the other people can take charge of other processes
and these hands called "dirty hands".

Lines 115-119: Your numbers of samples from the deserts are low and sand may not make a useful
 comparison as it is not the material to undergo long-range transport (too large particles). Please
 comment on the spatial homogeneity / heterogeneity of mineral and elemental composition of desert
 material

Response: Data of desert sand were deleted from Figure 5. In this study, we focused on the dust in thedesert sand, and the desert sands are well mixed, for instance, the 1870s/1880s ratios study showed

that for Taklimakan Desert sands are close to the average of Kunlun moraines, river sediments around

the Taklimakan Desert and the Tibetan soils. Therefore, the Taklimakan Desert sands are derived from

- 239 moraines and river sediments around the desert or from Tibetan soils and are homogenized by aeolian
- activity in the desert (Hattori et al., 2003). Furthermore, because dusts loaded on the glaciers are well
- 241 mixed during long distant transport from the desert region, mineral and elemental compositions of dust
- 242 deposited on glacier are also homogenized (Wu et al., 2009). Therefore, "Mineral and elemental
- composition of desert sands of west China are homogenized by aeolian activity (Hattori et al., 2003), so
- that the dust samples collected in this study are representative of desert sourced dust in west China"was added into the MS.
- Line 117: Is Dunhuang a desert location? Please clarify.
- 247 Response: Yes, it is a desert location. This information was added in section 2.2.
- 248 Line 147: Provide references supporting your first sentence.
- Response: References "Kaspari et al., 2014", "Qu et al., 2014" and "Ming et al., 2013" were added inthe sentence.
- Line 161: Use 'pre-combusted' instead of 'pre-burned'.
- 252 Response: "pre-burned" in line 161 and 162 was changed to "pre-combusted".

Lines 188-189: In the text you state "Therefore, the distributions of DOC concentrations in the glacier surface snow and ice were influenced by complicated factors, such as the terrain, surface moraine and atmosphere circulation". (i) In which way does the 'terrain' (whatever it means here) influence DOC? (ii) What is the impact of atmospheric circulation? I suppose you can't make quantitative conclusions

257 in the absence of continuous measurements but you should at least comment and refer to literature. (iii)

- In Section 2.2 you refer to the collection of samples from deserts so I assume that 'mineral dust'
 implies 'desert dust'. If this is the case, make it clear. How can you tell input of desert dust in DOC
 concentrations from input of material from local moraines?
- 200 concentrations from input of material from local moranies?
- 261 Response: For question (i), 'Terrain' means different slopes and faces of the glacier, which will cause
- different enrichment of particles on the glacier surface, finally cause variations of DOC concentration.
- 263 (ii) 'Atmospheric circulation' is deleted because it hardly influences the contribution of DOC in glacier
- surface ice. (iii) "Mineral dust" is "desert dust" Therefore, "mineral dust" was replaced by "desert
- sourced mineral dust". Research on Sr-Nd isotopic compositions (Xu et al., 2012) has shown that dust
- 266 loaded on LHG glacier was mainly derived from long range transported dust rather than local 267 moraines.

- 268 Lines 210-211: Concentrations of Ca^{2+} in desert dust. Have you compared your desert dust samples 269 with the samples from the local moraines? What other tracers can you use? Do you have absorption 270 spectra for material from local moraines and how is it different from those for your desert material 271 samples?
- Response: Sorry for not doing this comparison because according to previous research on Sr-Nd
 isotopic compositions (Xu et al., 2012), it was well constrained that dust loaded on LHG glacier was
 mainly transported from deserts rather than local moraines. Therefore we only check the desert sourced
 dust. Furthermore, according to our observation during sampling collecting process, we found local
 moraines belong to coarse crusted sand and stones, which should contain little organic matter and hard
- to be transported to the glacier surface.
- Section 3.2: You should bring Supplement Figure 1 into this section and add comments on the profiles
 of DOC in your snow pits highlighting differences between the layers containing dust and the relatively
 clean layers.
- 281 Response: Supplement Figure 1 was brought into the main text as Figure 3. "Moreover, the profiles of
- 282 DOC in two snowpits varied with the dust content, DOC concentration of dust layer was much higher283 than that of clean layers." was added in section 3.2.
- Lines 256-258 and Fig. 6S: Please explain the elevation dependence of DOC relative to BOS moreclearly
- 286 Response: Lines 256-258 were deleted for the misunderstanding.
- Add more detailed comments on Fig. 6S.
- 288 Response: Paragraph 2 in section 3.3.3 was rewritten and changed to "The high radiative forcing ratio
 289 of snowpit samples was caused by its higher DOC/BC (0.65) than that of surface ice (0.012) (Fig. S5),
- and the low ratio of DOC/BC in surface ice was caused by enrichment of BC in surface glacier ice
- during the intensive ablation period (Xu et al., 2009)".
- Equation 3 show that radiative forcing depends on concentration and Fig. 6S shows the ratios between
 black carbon and dissolved carbon but a couple of sentences would be required to clarify and
 strengthen you message.
- Response: Two sentences were added into the MS. "It is obvious that the value of is closely connectedwith relative concentrations between DOC and BC." was added into the method part 2.3.2. Meanwhile,
- the expression in part 3.3.3 was modified to "The high radiative forcing ratio of snowpit samples was
- 298 caused by its higher DOC/BC (0.65) than that of surface ice (0.012) (Fig. S5), and the low ratio of
- 299 DOC/BC in surface ice was caused by enrichment of BC in surface glacier ice during the intensive
- ablation period (Xu et al., 2009)."
- 301 Section 3.3.3: Too many abbreviations (BrC, WSOC) make reading this section difficult.
- 302 Response: These abbreviations were rewritten in full name in the text.
- 303 Line 287: Provide references after ": : :the European Alps and Alaska"
- Response: References "Singer et al., 2012" and "Fellman et al., 2015" were added in the sentence.
- 305 Section 3.4 is very brief. Can you expand it and give it more prominence?
- 306 Response: We measured DOC concentrations of proglacial streamwater samples at other 5 glaciers
- during last three years. Although only two data were achieved for each glacier, the total flux of DOC
- 308 for all the glaciers of the TP was estimated and the following information was added in the section 3.4.
- 309 "When it comes to the entire TP, it is obvious that proglacial streamwater DOC concentrations
 310 (Table S2) showed similar spatial variation to that of snowpit DOC (Li et al., 2016), with high and low
 311 value appeared at north and south TP, respectively, reflecting good succession of proglacial

- 312 streamwater DOC concentration to that of snowpit samples. Therefore, it was calculated that DOC flux
- in proglacial streamwater of the entire TP glacier was around 12.7-13.2 Gg C (Gg = 10^9 g) based on
- average proglacial streamwater DOC concentration of 193 μ g L⁻¹ (Table S2) and annual glacial
- meltwater runoff in China of 66-68.2 km³ (Xie et al., 2006), which is higher than that of DOC

deposition (5.6 Gg C) at glacier region of the TP, agree well with the negative water balance of the

317 glaciers of the TP. Therefore, the TP glaciers can be considered as a carbon source under present

- 318 environment condition."
- Section Conclusions (Lines 291-293) and Abstract: radiative forcing of DOC is 0.1±0.1% of BC in ice
- and 9.5 ± 8.4 % for snow. So this in effect is an almost zero addition in case of ice and might be close to zero addition for snow. I suggest that you should convert this into W m⁻² using data from literature to
- be more convincing.
- 323 Response: Thanks a lot for the suggestion. Since the radiative forcing ratio of surface ice is almost zero,
- 324 we converted the ratio of snowpit into W m^{-2} based on the previous published data of black carbon in
- snowpit of LHG glacier (Ming et al., 2013). "Based on the previous published radiative forcing data of
- 326 black carbon of snowpit of LHG (Ming et al., 2013), for the first time, it is estimated that the radiative
- forcing caused by snowpit DOC was 0.43 W m^{-2} , accounting for around 10 % of the radiative forcing
- 328 caused by BC" was added into conclusions section and section abstract was adjusted accordingly.
- 329 Figures and Tables Tables 1 and 2: Why are your references in parentheses?
- 330 Response: These references were changed according to the writing standards.
- Combine Fig. 1 with Table 1 in the Supplement and show both snow pits.
- Response: Table 1 in supplement information was moved into the main text, the other snowit wasmarked on Figure 1.
- Fig. 2: Show error bars both ways and add the number of samples in each category.
- **335** Response: Adjusted accordingly on Figure 2.



336

337 Figure 2 Average DOC concentrations of ice, snow and proglacial streamwater for LHG glacier.

Fig. 3: Why do you need exponential fit here? Linear regression describes this relationship well. Yourstandard deviation bars are impossible to see.

Response: Yes, based on the data only, the linear regression could be adequate. However, according to

- the DOC bioavailability, the exponential one can be more authentic (Spencer et al., 2015), because
- some DOC are bio-refractory, so that DOC cannot reach zero with long enough resident time. The

343 differences between the parallel samples were very small, so the standard deviation bars were very

- 344 short.
- Fig. 4: What is 'desert' and what is 'dust'?

346 Response: "Desert" was desert sand collected from the desert at Dunhuang; "Dust" was the dust fall

347 collected during dust storm events at Dunhuang. The spectrum of desert sand was deleted because of

the low numbers of the samples. Figure 4 was changed to Figure 5 based on the previous comments asbelow.



350

357

Figure 5 Absorption spectra for the DOC in snow and ice of LHG glacier and the dust and desert sand from

352 surrounding areas.

Fig.S1: Use the same scales on X and Y axes for both profiles for an easier comparison. Move this

354 figure to the main text.

Response: This Figure was moved to the main text as Figure 3 and the scales were adjusted accordingly.



Figure 3 Variation in DOC concentrations in profiles of studied snowpits. The gray rectangles are dirty layers.

359 Fig. 6S: Are these average ratios for all samples?

360 Response: They were average DOC and BC ratios of snowpits and surface ice, respectively, "snow"

was changed to "snowpit", "ice" was changed to "surface ice" on this figure, and "6S" was changed to"5S".



- Xu, B., Cao, J., Hansen, J., Yao, T., Joswia, D. R., Wang, N., Wu, G., Wang, M., Zhao, H., and Yang, W.: Black
 soot and the survival of Tibetan glaciers, PNAS, 106, 22114-22118, 2009.
- Xu, J., Yu, G., Kang, S., Hou, S., Zhang, Q., Ren, J., and Qin, D.: Sr–Nd isotope evidence for modern aeolian dust
 sources in mountain glaciers of western China, Journal of Glaciology, 58, 859-865, 2012.
- 396
- **397** Other relevant changes are made as follow:
- 398 Line 29: "43.2%" was changed to "46.3%".

- Line 33-36: "The radiative forcing of DOC relative to black carbon (BC) was calculated to be 9.5 \pm
- 400 8.4 % in snow and 0.1±0.1 % in ice, respectively, implying the necessity of accounting for DOC in
- 401 future radiative forcing investigations in the glacierized region on the TP, especially when these areas
- are covered by fresh snow" was changed to "Meanwhile, autotrophic or heterotrophic biological
- 403 activities and autochthonous carbon could also contribute to glacier DOC. The radiative forcing of
- 404 snowpit DOC was considered to be 0.43 W m^{-2} , implying the necessity of accounting DOC of snow for
- 405 accelerating melt of glaciers on the TP".
- Line 197: "glacier" was changed to "Xiaodongkemadi glacier"; "Dong Rongbuk" was changed to "EastRongbu".
- 408 Line 223: "43.2%" was changed to "46.3%".
- 409 Line 360-361: "the number of glacial rivers across the entire TP and surrounding areas may be large
- 410 due to the glacial area of approximately $100,000 \text{ km}^2$ (Yao et al. 2012). These factors need to be
- 411 comprehensively studied in the future" was changed to "when it comes to the entire TP DOC flux from
- 412 glaciers of the entire TP was around 12.7-13.2 Gg C".
- 413 References: All the references were changed according to the manuscript preparation guidelines.

414 **Concentration, sources and light absorption**

415 characteristics of dissolved organic carbon on a typical

416 glacier, the northern Tibetan Plateau

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- Abstract. Light-absorbing dissolved organic carbon (DOC) constitutes a major part of the organic 431 432 carbon in glacierized regions. It has important influences on the carbon cycle and radiative forcing of 433 glaciers. However, currently, few data are available in the glacierized regions of the Tibetan Plateau (TP). In this study, DOC characteristics of a typical glacier (Laohugou glacier No. 12 (LHG glacier)) in 434 435 the northern TP were investigated. Generally, DOC concentrations on LHG glacier were comparable to 436 those in other regions around the world. DOC concentrations in snowpits and surface snow were 332-4 ± 132.3 µg L⁻¹ and 229.3 ± 104.4 µg L⁻¹, respectively, which were slightly higher than those of the 437 438 Greenland ice sheet. DOC concentration of surface ice (superimposed ice) was $425.86 \pm 269.970 \ \mu g \ L^{-1}$, comparable to that of the Antarctic ice sheet. The average discharge-weighted DOC of proglacial 439 streamwater was $237.58 \pm 95.66 \ \mu g \ L^{-1}$, which is lower than that of Mendenhall glacier, Alaska. The 440 annual DOC flux released from this glacier was estimated to be 6,949.4 kg C yr⁻¹, of which 463.2% of 441

442	DOC was bioavailable and could be decomposed into CO_2 within one month of its release. The mass
443	absorption cross section (MAC) of DOC at 365 nm was 1.4 \pm 0.4 m ² g ⁻¹ in snow and 1.3 \pm 0.7 m ² g ⁻¹ in
444	ice, similar to the values of dust transported from adjacent deserts. Based on this finding and the
445	significant relationship between DOC and Ca ²⁺ , the main source of DOC was-might be desert mineral
446	dust. Meanwhile, autochthonous, autotrophic or heterotrophic biological activities and autochthonous
447	carbon could also contribute to glacier DOC. The radiative forcing of snowpit_DOC relative to black
448	carbon (BC) was considered to be 0.43 W m^{-2} 9.5 $\pm 8.4 \text{ \% in snow and } 0.1 \pm 0.1 \text{ \% in ice}$, implying the
449	necessity of accounting-for DOC of snow for accelerating melt of glaciers in future radiative forcing
450	investigations in the glacierized regions on the TP, especially when these areas are covered by fresh
451	snow .
452	Key words: dissolved organic carbon, concentration, light absorption, LHG glacier, the Tibetan
453	Plateau

455 1 Introduction

Ice sheets and mountain glaciers cover 11 % of the land surface of the Earth and store 456 approximately 6 Pg (1 Pg = 10^{15} g) of organic carbon, the majority of which (77 %) is in the form of 457 458 dissolved organic carbon (DOC) (Hood et al., 2015). The annual global DOC release through glacial runoff is approximately 1.04 \pm 0.18 Tg C (1 Tg = 10¹² g) (Hood et al., 2015). Therefore, glaciers not 459 460 only play an important role in the hydrological cycle by contributing to sea-level rise (Rignot et al., 461 2003; Jacob et al., 2012) but also potentially influence the global carbon cycle (Anesio and 462 Laybourn-Parry, 2012; Hood et al., 2015) in the context of accelerated glacial ice loss rates. In addition, 463 a large portion of glacier-released DOC has proven to be highly bioavailable, influencing the balance of 464 downstream ecosystems (Hood et al., 2009; Singer et al., 2012; Spencer et al., 2014).

465 Although DOC storage in ice sheets is much larger than that of mountain glaciers, the annual 466 mountain glacier-derived DOC dominates the global DOC release (Hood et al., 2015). Currently, many 467 studies on the concentration, age, composition, storage and release of DOC have been conducted 468 around the world (Hood et al., 2009; Stubbins et al., 2012; May et al., 2013; Bhatia et al., 2013; 469 Lawson et al., 2014; Fellman et al., 2015; Hood et al., 2015). The sources of glacier-derived DOC were 470 found to be diverse (Bhatia et al., 2010; Stubbins et al., 2012; Singer et al., 2012; Spencer et al., 2014), 471 with large variations in concentrations and ages (Hood et al., 2009; Singer et al., 2012; Hood et al., 472 2015). For example, a study on the Greenland ice sheet showed that the concentration of exported DOC 473 exhibited slight temporal variations during the melting period, with subtly higher values in early May 474 than in late May and July (Bhatia et al., 2013). Additionally, the concentration of total organic carbon 475 in snow across the East Antarctic ice sheets exhibited remarkable spatial variations due to the marine 476 source of organic carbon (Antony et al., 2011). Studies on both radiocarbon isotopic compositions and 477 biodegradable DOC (BDOC) have proposed that ancient organic carbon from glaciers was much easier 478 for microbes to utilize in glacier-fed rivers and oceans, implying that large amounts of this DOC will 479 return to the atmosphere quickly as CO_2 and participate in the global carbon cycle, thereby producing a 480 positive feedback in the global warming process (Hood et al., 2009; Singer et al., 2012; Spencer et al., 481 2014). In addition to black carbon (BC), another DOC fraction known as water-soluble brown carbon 482 (WS BrC) has also been considered a warming component in the climate system (Andreae and 483 Gelencs é, 2006; Chen and Bond, 2010). This type of DOC exhibits strong light-absorbing properties in 484 the ultraviolet wavelengths (Andreae and Gelencs ér, 2006; Chen and Bond, 2010; Cheng et al., 2011).

The relative radiative forcing caused by water-soluble organic carbon (WSOC, the same as DOC) relative to BC in aerosols was estimated to account for 2-10 % and approximately 1 % in a typical pollution area of North China (Kirillova et al., 2013) and a remote island in the Indian Ocean (Bosch et al., 2014), respectively. Unfortunately, so far, few direct evaluations have been conducted in the glacierized regions around the world, including the Tibetan Plateau (TP), where DOC accounts for a large part of the carbonaceous matter (Legrand et al., 2013; May et al., 2013) and potentially contributes to the radiative forcing in the glacierized region.

492 The TP has the largest number of glaciers at moderate elevations. Most of the glaciers on the TP 493 are experiencing intensive retreat because of increases in temperature (Kang et al., 2010; Yao et al., 494 2012; Yao, 2004a; Kang et al., 2015; Zhang et al., 2015) and anthropogenic carbonaceous particle 495 deposition (Ming et al., 2008; Xu et al., 2009; Qu et al., 2014; Kaspari et al., 2014). However, to date, 496 no study has quantitatively evaluated the light absorption characteristics of DOC in the glacierized 497 regions on the TP, despite some investigations of concentrations and sources of DOC (Spencer et al., 498 2014; Yan et al., 2015). The primary results of these studies have shown that DOC concentrations in 499 snowpits in the northern TP are higher than those in the southern TP (Yan et al., 2015). In addition, a 500 large fraction of the ancient DOC in the glaciers in the southern TP has high bioavailability 501 characteristics (Spencer et al., 2014). However, knowledge of DOC in TP glaciers remains lacking due 502 to the large area and diverse environments of the TP and the relatively limited samples and studies. 503 Therefore, numerous snow (n=67), ice (n=42) and proglacial streamwater samples (n=201)(n=310)504 were collected from a typical glacier in the northeastern TP (Laohugou glacier No. 12) based on the 505 preliminary research of snowpit samples (Yan et al., 2015) (Table §1, Fig. 1). The concentrations of DOC and major ions $(Ca^{2+}, Mg^{2+}, Na^+, K^+, NH_4^+, Cl^-, NO_3^-)$ and SO₄²⁻) and DOC light absorbance were 506 507 measured to comprehensively investigate the sources, light absorption properties and carbon dynamics 508 in this glacierized region to provide a basis for the study of DOC across the TP and other regions in the 509 future.

510 2 Methodology

511 **2.1 Study area and sampling site**

Laohugou glacier No. 12 (LHG glacier) (39 05'-40'N, 96 07'-97 04'E 4260-5481 m) is the largest
mountain glacier (9.85 km, 20.4 km²) in the Qilian Mountains located on the northeastern edge of the
TP (Du et al., 2008; Dong et al., 2014a). It is divided into two part of western and eastern branch at the

elevation of 4560 m a.s.l (Dong et al., 2014a). This glacier is surrounded by large arid and semi-arid
regions (sandy deserts and the Gobi desert) (Fig. 1). The area of the glacier covers approximately 53.6 %
of the entire LHG glacier basin (Du et al., 2008; Li et al., 2012).

LHG glacier features typical continental and arid climate characteristics (Li et al., 2012; Zhang et al., 2012b). Precipitation occurs mainly from May to September, accounting for over 70 % of the total annual precipitation (Zhang et al., 2012b). The monthly mean air temperatures in the ablation zone of the glacier range from -18.4 $\$ in December to 3.4 $\$ in July (Li et al., 2012). Like other glaciers on the TP, LHG glacier has been experiencing significant thinning and shrinkage at an accelerated rate since the mid-1990s (Du et al., 2008; Zhang et al., 2012b).

524 2.2 Sample collection

525 Two snowpits were dug in 2014 and 2015 almost in the same site in the accumulation zone of 526 LHG glacier. In total, 15 and 23 snow samples were collected in 2014 and 2015, respectively, at a 527 vertical resolution of 5 cm for each snowpit. Moreover, 29 surface snow and 42 surface ice 71 snow/ice 528 samples were collected along the eastern tributary at an approximate elevation interval of 50 or 100 m 529 from the terminus to the accumulation zone, and 201 proglacial streamwater samples were collected at 530 the gauge station during the melting period (Fig. 1, Table $\frac{1}{5}$). The concentrations of glacier DOC have 531 been observed to be very low and prone to contamination, causing an overestimation of DOC 532 concentrations (Legrand et al., 2013). Therefore, before sample collection, polycarbonate bottles were 533 firstly cleaned by ultrapure water for three times, then soaked into 1 M HCl for 24 h (Spencer et al., 534 2009), and rinsed three times using ultrapure water, finally soaked into ultrapure water for over 24 h; 535 during the whole sampling procedure, snow samples were collected directly into 125-mL pre-cleaned 536 bottles, surface ice (0-3 cm and 3-5 cm) samples were collected using an ice axe directly into 537 polycarbonate bottles after crushing, while proglacial streamwater samples were filtered immediately after collection before putting into bottles. All ice and snow sample were filtered as soon as possible 538 539 after they were melted samples were collected according to the "Clean hands Dirty Hands" principle to 540 prevent any contamination. During all the processes, the person who takes charge of sample collection 541 should not touch any other things to avoid contamination. Meanwhile, at least one blank was made for 542 every sampling process to confirm that the contamination was low (Table S12). Meanwhile, another 543 batch of samples were also collected for BC concentration measurement following the protocol 544 discussed in detail in our earlier work (Qu et al., 2014); these results will be presented in another article. 545 In order to evaluate DOC release on of the totalentire TP, DOC concentrations of proglacial streamwater
546 samples of other five glaciers in monsoon and non-monsoon seasons were measured, respectively (Fig.
547 1, Table S2).

548 All the collected samples were stored in 125 mL pre cleaned polycarbonate bottles and were 549 kept frozen and in the dark in the field, during transportation and in the laboratory until analysis. In 550 addition, two sand samples from the desert and four dust fall samples from Dunhuang, a desert location (39 53'-41 35'N, 92 13'-93 30'E) - a-potential source region of the dust deposited on LHG glacier -551 552 were collected to compare the light absorption characteristics of dust-sourced DOC to those of the snowpit and ice samples. Mineral and elemental composition of desert sands of west China are 553 homogenized by aeolian activity (Hattori et al., 2003), so that the dust samples collected in this study 554 555 are representative of desert sourced dust in west China.

556 2.3 Laboratory analyses

557 2.3.1 Concentration measurements of DOC and major ions

558 DOC concentrations were determined using a TOC-5000A analyzer (Shimadzu Corp, Kyoto, 559 Japan) after the collected samples were filtered through a PTFE membrane filter with 0.45-um pore size (Macherey-Nagel) (Yan et al., 2015). The detection limit of the analyzer was 15 µg L⁻¹, and the 560 average DOC concentration of the blanks was $31.92 \pm 7.4 \ \mu g \ L^{-1}$, demonstrating that contamination can 561 562 be ignored during the pre-treatment and analysis processing of these samples (Table S12). The major cations (Ca²⁺, Mg²⁺, Na⁺, K⁺ and NH₄⁺) and major anions (Cl⁻, NO₃⁻ and SO₄²⁻) were measured using a 563 Dionex-6000 Ion Chromatograph and a Dionex-3000 Ion Chromatograph (Dionex, USA), respectively. 564 The detection limit was 1 μ g L⁻¹, and the standard deviation was less than 5 % (Li et al., 2007; Li et al., 565 2010). The average ion concentrations of the blank were very low and could be ignored (Na⁺, K⁺, Mg²⁺, 566 F^+ , SO_4^{2-} , CI^- , $NO_3^- < 1 \ \mu g \ L^{-1}$; $NH_4^+ = 1.42 \ \mu g \ L^{-1}$; $Ca^{2+} = 1.24 \ \mu g \ L^{-1}$). 567

568 2.3.2 Light absorption measurements

The light absorption spectra of DOC <u>was-were</u> measured using an ultraviolet-visible absorption spectrophotometer (SpectraMax M5, USA), scanning wavelengths from 200-800 nm at a precision of 5 nm. The mass absorption cross section (MAC) was calculated based on the Lambert-Beer Law (Bosch et al., 2014; Kirillova et al., 2014a; Kirillova et al., 2014b):

573
$$MAC_{DOC} = \frac{-\ln|\frac{1}{I_0}|}{C.L} = \frac{A}{C.L} \times \ln(10)$$
(1)

where I_0 and I are the light intensities of the transmitted light and incident light, respectively; A is the absorbance derived directly from the spectrophotometer; C is the concentration of DOC; and L is the absorbing path length (1 cm).

577 In order to investigate the wavelength dependence of DOC light absorption characteristics, the
578 Absorption Ångström Exponent (AAE) was fitted by the following equation (Kirillova et al.,
579 2014b;Kirillova et al., 2014a):

580
$$\frac{A(\lambda_1)}{A(\lambda_2)} = \left(\frac{\lambda_2}{\lambda_1}\right)^{AAE}$$
(2)

AAE values were fitted from the wavelengths of 330 to 400 nm; within this wavelength range, light absorption by other inorganic compounds can be avoided (such as nitrate) (Cheng et al., 2011).The radiative forcing caused by BC has been widely studied (Kaspari et al., 2014; Qu et al., 2014; Ming et al., 2013). Therefore, in this study, using a simplistic model (the following algorithm), the amount of solar radiation absorbed by DOC compared to BC was estimated:

586
$$f = \frac{\int_{300}^{2500} I_{0}(\lambda) \cdot \left\{1 - e^{-(MAC_{365}\left(\frac{365}{\lambda}\right)^{AAE_{DOC}} \cdot C_{DOC} \cdot h_{ABL})}\right\} d\lambda}{\int_{300}^{2500} I_{0}(\lambda) \cdot \left\{1 - e^{-(MAC_{550}\left(\frac{550}{\lambda}\right)^{AAE_{BC}} \cdot C_{EC} \cdot h_{ABL})}\right\} d\lambda}$$
(3)

587 where λ is the wavelength; I₀ (λ) is the clear sky solar emission spectrum determined using the Air 588 Mass 1 Global Horizontal (AM1GH) irradiance model (Levinson et al., 2010); MAC₃₆₅ and MAC₅₅₀ are 589 the mass absorption cross section of DOC at 365 nm and mass absorption cross section of BC at 550 590 nm, respectively; h_{ABL} is the vertical height of the atmospheric boundary layer; and AAE_{DOC} and AAE_{BC} are the Absorption Ångström Exponents (AAEs) of DOC and BC. In this simplistic model, 591 following a previous study, we used MAC₅₅₀ = 7.5 $\pm 1.2 \text{ m}^2 \text{g}^{-1}$ (Bond and Bergstrom, 2006), and AAE 592 593 for BC was set as 1, while h_{ABL} was set to 1000 m, which has little influence on the integration from 594 the wavelengths of 300 - 2500 nm (Kirillova et al., 2013; Bosch et al., 2014; Kirillova et al., 2014a; 595 Kirillova et al., 2014b). It is obvious that the value of "f" is closely connected with relative 596 concentrations between DOC and BC.

597 2.3.3 In situ DOC bioavailability experiment

598 The bioavailability experiment was conducted from August 17th to 31st, 2015, at the glacier 599 terminus during fieldwork. In brief, surface ice samples were collected in-pre-burned pre-combusted 600 (550 °C, 6 h) aluminum basins and melted in the field. The melted samples were filtered through 601 pre-burned pre-combusted glass fiber filters (GF/F 0.7 µm) into 12 pre-cleaned 125-mL polycarbonate 602 bottles and wrapped with three layers of aluminum foil to avoid solar irradiation. Two samples were 603 refrigerated immediately after filtering to obtain initial DOC concentrations; the others were placed 604 outside at the terminus of the glacier, and 2 samples were refrigerated every 3 days to get 605 corresponding DOC values. The BDOC was calculated based on the discrepancies between the initial 606 and treated samples.

607 3 Results and discussion

608 3.1 DOC concentrations and bioavailability

609 **3.1.1 Snowpits**

610 LHG glacier is surrounded by arid and semi-arid regions and frequently influenced by strong dust 611 storms (Dong et al., 2014b) (Fig. 1). Therefore, heavy desert sourced mineral dust deposition 612 contributes to high DOC concentrations on LHG glacier. The average DOC concentration of the snowpit samples was 332.4 \pm 132.3 μ g L⁻¹ (Fig. 2), with values ranging from 124.4 μ g L⁻¹ to 581.9 μ g 613 L^{-1} (Fig. S13). The highest values appeared in the dirty layers (Fig. S13), similar to the pattern 614 615 observed in the Greenland summit (Hagler et al., 2007) and glaciers in the southern TP (Xu et al., 616 2013), indicating that DOC concentrations were mainly influenced by dust deposition in this region in 617 addition to the potential microbial activities (Anesio et al., 2009). Spatially, our results were higher than those of Xiaodongkemadi glacier on Mountain Tanggula-glacier (TGL) in the middle TP and 618 619 DongEast Rongbuk glacier on Mount Everest (EV) in the southern TP (Fig. 1) (Yan et al., 2015) but 620 similar to the mercury distribution on the TP (Zhang et al., 2012a). Moreover, the DOC concentrations 621 on LHG glacier were also higher than those of Alaskan glaciers (Stubbins et al., 2012) and the 622 Greenland summit (Hagler et al., 2007) (Table 24).

623 3.1.2 Surface snow and ice

The average DOC concentration in LHG glacier surface snow was significantly lower than that in surface ice because more impurities are present in the latter (Fig. 2). Like those of the snowpits, DOC concentrations in the glacier surface ice (Fig. 2) were higher than those in the southern TP (Nyainqentanglha glacier) (Spencer et al., 2014) and subsurface ice (0.5 m beneath the glacier surface) 628 in a European Alpine glacier (Singer et al., 2012) (Table 2) but comparable to that in the surface ice of 629 the Antarctic ice sheet (Hood et al., 2015). However, the DOC concentrations in surface snow (Fig. 630 2Table 2) were higher than those in the Greenland ice sheet (Hagler et al., 2007), mainly due to the 631 heavy dust load of LHG glacier-(Table 2). No significant relationship was found between DOC 632 concentration and elevation for either the surface snow or ice (Fig. S_{12}), suggesting no "altitude effect" 633 on DOC in this glacier. This finding is similar to the mercury distribution pattern in surface snow of 634 this glacier (Huang et al., 2014). Therefore, the distributions of DOC concentrations in the glacier 635 surface snow and ice were influenced by complicated factors, such as the terraindifferent slopes (Hood and Scott, 2008) and surface-cryoconite holes moraineand atmosphere circulation. Furthermore, DOC 636 637 concentrations of snow and ice of this glacier were within the range of previously reported values for 638 glacieried regions outside the TP.

639 **3.1.3 DOC bioavailability**

640 Previous studies conducted under controlled conditions (stable temperature) have shown that 641 glacier-derived DOC is more bioavailable than terrestrial-derived DOC (Hood et al., 2009; Fellman et 642 al., 2010; Spencer et al., 2014). Our results showed that the amount of DOC being consumed decreased exponentially over time ($R^2 = 0.98$) (Fig. <u>4</u>³), with approximately 26.7 % (from 41<u>6.9-7</u>µg L⁻¹ to 30<u>5.66</u> 643 μ g L⁻¹) degraded within 15 days during the experiment (average temperature: 3.77-8 ± 3.65-7 °C; range: 644 645 -4.778-11.364 °C). The BDOC reached 43.26.32 % if the experiment duration was extended to 28 days, 646 according to the equation derived from the 15-day experiment (Fig. 43). Despite different incubation 647 conditions, this finding agrees well with the reports of BDOC from a glacier in the southern TP (28-day 648 dark incubation at 20 °C, 46-69 % BDOC) (Spencer et al., 2014) and European Alpine glaciers (50-day 649 dark incubation at 4 °C, 59±20 % BDOC) (Singer et al., 2012). Therefore, the previous results obtained 650 in the laboratory closely reflect the real situation and can be used to estimate the bioavailability of 651 glacier-derived DOC.

652 **3.2 Sources of snowpit DOC**

2 5.2 Sources of showpit DOC

The sources of glacier DOC are diverse and include microbial_autochthonous or *in situ* biological
activit<u>iesy (viruses, bacteria and algae) in subglacial systems</u> (Anesio et al., 2009; Bellas et al., 2013),
allochthonous carbon derived from overridden soils and vegetation in subglacial systems (Bhatia et al.,
2010); terrestrial inputs (DOC deposition from vascular plants and dust) (Singer et al., 2012) and

657 anthropogenic sources (fossil fuel and biomass combustion) (Stubbins et al., 2012; Spencer et al., 2014)(Anesio et al., 2009; Hood et al., 2009; Bhatia et al., 2010; Stubbins et al., 2012; Singer et al., 658 659 2012; Spencer et al., 2014; Antony et al., 2014). Research on glacier microbial activity suggests that 660 globally only cryoconite holes can potentially fix about 64 Gg carbonC per year (Anesio et al., 2009). Moreover, viral induced mortality at the cost of heterotrophic bacterial community plays a dominant 661 662 role in carbon cycle and other nutrients transformation in supraglacier ecosystems (Bellas et al., 2013). In this study, major ions were adopted as indicators to investigate the potential sources of snowpit DOC, 663 664 because the sources of major ions in snowpit samples from Tibetan glaciers have been investigated in 665 detail (Kang et al., 2002; Kang et al., 2008; Wu et al., 2011; Yan et al., 2015). Moreover, the profiles of DOC in two snowpits varied with the dust content, DOC concentration of dust layer was much higher 666 than that of clean layers. Furthermore, <u>I</u>it was found that DOC and Ca²⁺ (a typical indicator of mineral 667 dust (Yao, 2004b)) were significantly related ($R^2 = 0.84$, Fig. S23), suggesting that the major source of 668 669 DOC was desert sourced mineral dust, which is consistent with the previous DOC source investigations 670 of snowpits on this glacier (Yan et al., 2015). In addition, the combined study of geochemistry and 671 backward trajectories for LHG glacier showed that the dust particles on the glacier were mainly derived 672 from the deserts to the west and north of the study area (Dong et al., 2014a; Dong et al., 2014b).

673 3.3 Light absorption characteristics of DOC

674 **3.3.1 AAE**

675 The Absorption Ångström Exponent (AAE) is generally used to characterize the spectral 676 dependence of the light absorption of DOC, which is important input data for radiative forcing 677 calculations (supporting information). The fitted AAE₃₃₀₋₄₀₀ values (supporting information) ranged 678 from 1.2 to 15.2 (5.0 \pm 5.9) for snow samples and from 0.3 to 8.4 (3.4 \pm 2.7) for ice samples (Fig. S45). 679 The relatively low $AAE_{330-400}$ values of the ice indicated that the DOC experienced strong 680 photobleaching due to long-duration exposure to solar irradiation. Previous studies have found that the 681 AAE values of brown carbon (BrC)in aged aerosols (Zhao et al., 2015) and secondary organic aerosols 682 (SOAs) (Lambe et al., 2013) were much lower compared to that of the primary values. Therefore, the 683 large divergence in AAE values might suggest different chemical compositions of DOC due to multiple 684 possibilities, such as different sources and photobleaching processes. Regardless, the average AAE 685 value of the snow samples was comparable to that of atmospheric aerosols in urban areas in South Asia 686 (New Delhi, India) (Kirillova et al., 2014b) (Table 2). In general, the AAE₃₃₀₋₄₀₀ values had a negative relationship with MAC₃₆₅, especially in the ice samples (Fig. S45), suggesting that stronger absorbing DOC might contribute to lower AAE values, which was also found in other aerosol studies (Chen and Bond, 2010; Bosch et al., 2014; Kirillova et al., 2014b).

690 3.3.2 MAC₃₆₅

691 The mass absorption cross section at 365 nm (MAC₃₆₅) for DOC is another input data point for the 692 radiative forcing calculation. The light absorption ability at 365 nm is selected to avoid interferences of 693 non-organic compounds (such as nitrate) and to be consistent with previous investigations (Hecobian et al., 2010; Cheng et al., 2011). The MAC₃₆₅ was 1.4 \pm 0.4 m² g⁻¹ in snow and 1.3 \pm 0.7 m² g⁻¹ in glacier 694 695 ice (Fig. S45), both of which were higher than those of water soluble organic carbon WSOC in outflow 696 in northern China (Kirillova et al., 2014a) and a receptor island in the Indian Ocean (Bosch et al., 2014). 697 Meanwhile, the values were comparable to DOC concentrations in typical urban aerosols associated 698 with biomass combustion in winter in Beijing, China (Cheng et al., 2011) and in New Delhi, India 699 (Kirillova et al., 2014b) (Table <u>32</u>). The MAC values for DOC from different sources vary widely. Normally, the MAC_{365} of DOC derived from biomass combustion can be as high as 5 $m^2\ g^{\text{-}1}$ 700 701 (Kirchstetter, 2004) (Table 32). Correspondingly, the values for SOAs can be as low as 0.001-0.088 m² 702 g^{-1} (Lambe et al., 2013). Due to the remote location of LHG glacier, it was considered that the snowpit 703 DOC should be SOAs with low MAC₃₆₅ values; however, the high MAC₃₆₅ value of the snowpit DOC 704 indicated that DOC may not be entirely derived from SOAs. Here, it was proposed that mineral 705 dust-sourced DOC caused the high MAC₃₆₅ values in the snowpit samples. For instance, the light 706 absorption characteristics of DOC from both snowpit and ice showed similar patterns to those of water 707 soluble organic carbon WSOC in dust from the adjacent deserts, further indicating that LHG glacier 708 DOC was transported via desert sourced mineral dust and shared similar light absorption characteristics 709 (Fig. 54). Moreover, the difference in light absorption characteristics (especially for wavelengths larger 710 than 400 nm) between snow/ice samples and aerosols in Beijing, China, also indicated different sources 711 (Fig. <u>54</u>). Light absorbance was significantly correlated with DOC concentrations in both snow and ice 712 samples (Fig. S_{24}), indicating that DOC was one of the absorption factors. Nevertheless, the MAC₃₆₅ 713 values of surface ice (0-3 cm) were lower than those of subsurface layers (3-5 cm), despite its higher 714 DOC concentrations (Fig. 65), reflecting stronger DOC photobleaching in the surface ice due to the 715 direct exposure to solar irradiation.

716 3.3.3 Radiative forcing of DOC relative to BC

717	The radiative forcing contributed by water soluble organic carbon WSOC relative to BC in
718	aerosols has been proposed to be as high as 2-10 % (Kirillova et al., 2013; Kirillova et al., 2014a).
719	Furthermore, it was estimated that brown carbon BrC-accounts for a higher ratio of 20 % of the direct
720	radiative forcing of aerosols at the top of the atmosphere because BC concentrations decrease faster
721	than brown carbon BrC-in the high-altitude atmosphere (Liu et al., 2014). Because the studied glacier is
722	located at high elevations near the top of the troposphere and features relatively high DOC/BC ratios in
723	the snowpit samples (Fig. S6), the radiative forcing caused by DOC relative to BC should also be high.
724	Our results showed that the relative radiative forcing caused by DOC relative to BC ranged from
725	2.1 % to 30.4 % (9.5 \pm 8.4 %) for snowpit samples and from 0.01 % to 0.5 % (0.1 \pm 0.1%) for surface
726	ice samples (Fig. S45)., The high radiative forcing ratio of snowpit samples was caused by its higher
727	DOC/BC (0.65) than that of surface ice (0.012) (Fig. S5), and the low ratio of DOC/BC in surface ice
728	was caused by enrichment of BC in surface glacier ice during the intensive ablation period (Xu et al.,
729	2009)mainly because of the higher DOC/BC ratio (0.65) in the snowpit samples than in the ice samples
730	(0.012) (Fig. S6). The value in ice was much lower due to the enrichment of BC in surface glacier ice
731	during the intensive ablation period (Xu et al., 2009). Therefore, the relative radiative forcing
732	contribution of DOC relative to BC in snowpit samples on LHG glacier was comparable with those of
733	aerosols in urban areas of New Delhi, India (Kirillova et al., 2014b). The value in ice was much lower
734	due to the enrichment of BC in surface glacier ice during the intensive ablation period (Xu et al., 2009).
735	Snowpit samples can be approximately considered to be fresh snow; thus, it is concluded that radiative
736	forcing caused by DOC is a non-ignorable contributor in addition to BC in reducing the albedo of a
737	glacier when the glacier is covered by fresh snow.

I

3.4 DOC export during the melt season

739The two-year average discharge-weighted DOC concentration was 237.58 ± 95.66 µg L⁻¹ during740the melting period, comparable with the proglacial streamwater of Mount Nyainqentanglha glacier in741the southern TP (Spencer et al., 2014). Seasonally, high DOC concentrations appeared during the low742discharge periods (May to July and September to October) (Fig. 76), suggesting that DOC743concentrations were slightly enriched to some extent. However, there were no clear diurnal variations744in the DOC concentrations with the discharge, suggesting that the discharge from different parts of the745glacier was well mixed at the glacier terminus (Fig. S67).

that discharge (rather than DOC concentrations) played a dominant role in the DOC mass flux. Hence, the majority of the glacier DOC export occurred during the summer melting season. Over the whole melting season, the annual flux of DOC from LHG glacier was 192.0 kg km⁻² yr⁻¹, with peak DOC fluxes from mid-late July to late August (70 % of the annual flux). Combined with the value of BDOC determined above, at least 3,001.53211 kg C yr⁻¹ was ready to be decomposed and returned to the atmosphere as CO₂ within one month of its release, producing positive feedback in the global warming process.

754 When it comes to the entire TP, it is obvious that proglacial streamwater DOC concentrations 755 (Table S2) showed similar spatial variation to that of snowpit DOC (Li et al., 2016), with high and low 756 value appeared at north and south TP, respectively, reflecting good succession of proglacial 757 streamwater DOC concentration to that of snowpit samples. Therefore, it was calculated that DOC flux 758 in proglacial streamwater of the total TP glacier was around 12.7-13.2 Gg C (Gg = 10^9 g) based on average proglacial streamwater DOC concentration of 193 μ g L⁻¹ (Table S2) and annual glacial 759 meltwater runoff in China of 66-68.2 km³ (Xie et al., 2006), which is higher than that of DOC 760 761 deposition (5.6 Gg C) at glacial region of the TP (Li et al., 2016), agree well with the negative water 762 balance of the glaciers of the TP. Therefore, the TP glaciers can be considered as a carbon source under present environment condition. 763

764 4 Conclusions and implications

765 The concentrations and light absorption characteristics of DOC on a typical glacier in the northern 766 TP were reported in this study. The mean DOC concentrations of snowpit samples, fresh snow, surface 767 ice and proglacial streamwater were $332.4 \pm 132.3 \ \mu g \ L^{-1}$, $229.3 \pm 104.4 \ \mu g \ L^{-1}$, $425.86 \pm 269.970 \ \mu g$ L^{-1} and 237.58 ±95.66 µg L^{-1} , respectively. These values were slightly higher or comparable to those of 768 other regions, such as the European Alps and Alaska (Singer et al., 2012; Fellman et al., 2015). DOC in 769 the snowpit samples was significantly correlated with Ca²⁺, a typical cation in mineral dust, indicating 770 771 that mineral dust transported from adjacent arid regions made important contributions to DOC of the 772 studied glacierized regions except autochthonous or *in situ* biological activities. In addition, the light 773 absorption profile of the snowpit DOC was similar to that of dust from potential source deserts, 774 providing further evidence of the influence of desert sourced mineral dust on snowpit DOC. Based on 775 the previous published radiative forcing data of black carbon in snowpit of LHG (Ming et al., 2013), 776 For for the first time, it is estimated that the radiative forcing caused by snowpit_DOC was 0.43 W 777 m^{-2} accounts for 9.5 ± 8.4 % and 0.1 ± 0.1 % relative to that of BC in the snowpit samples and surface 778 ice, respectively, accounting for around 10 % of the radiative forcing caused by BC. Therefore, in 779 addition to BC, DOC is also an important agent in terms of absorbing solar radiation in glacierized 780 regions, especially when the glacier is covered by fresh snow., which contains high DOC/BC ratios. It 781 has also been proven that water-insoluble organic carbon has-a stronger light absorption ability (Chen 782 and Bond, 2010). Therefore, the total contribution of OC to light absorption in glacierized regions 783 should be higher, which requires further study in the future. Wet deposition is the most effective way of 784 removing carbonaceous matter from the atmosphere (Vignati et al., 2010), and the removal ratio of OC 785 in remote areas is almost the same as that of BC after long-range transport from source regions (Garrett 786 et al., 2011). Because snowpit samples directly reflect the wet and dry deposition of carbonaceous 787 matter, it is assumed that the contribution of radiative forcing for water soluble organic carbon WSOC 788 relative to BC in the atmosphere in glacierized regions should be close to that of the snowpit samples in 789 this study.

790 Because proglacial streamwater from different parts of the glacier is well mixed, no clear diurnal 791 variations in DOC concentrations have been found. Combined with discharge and the corresponding DOC concentration, it was calculated that approximate 192.0 kg km⁻² yr⁻¹ of DOC was released from 792 793 LHG glacier. It was also calculated that approximately 463.32 % of the DOC could be decomposed within 28 days; thus, $3,\frac{001.5211}{1000}$ kg C yr⁻¹ would return to the atmosphere as CO₂, producing positive 794 795 feedback in the warming process. Although the flux of DOC from the studied glacier is small, when it 796 comes to the entire TP he number of glacial rivers across the entire TP and surrounding areas may be large due to the glacial area of approximately 100,000 km² (Yao et al. 2012). These factors need to be 797 798 comprehensively studied in the future.DOC flux from glaciers of the total TP was around 12.7-13.2 Gg 799 <u>C</u>.

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2957-2996, 2015.

985	Table 1 . Sampling information for snow, ice and proglacial streamwater in this study.						
	Sample type	Sampling site	Number (n)	Index			
	<u>Snowpit</u>	<u>30th July, 2014</u>	<u>5 cm</u>	<u>4989 m</u>	<u>15</u>	DOC, absorbance, ions	
	<u>Snowpit</u>	25th August, 2015	<u>5 cm</u>	<u>5050 m</u>	<u>23</u>	DOC, absorbance, ions	
<u>Sur</u>	face fresh snow	<u>4th August, 2014</u>	<u>100 m</u>	<u>4450-4900 m</u>	<u>18</u>	DOC	
<u>{</u>	Surface ice	<u>6th August, 2014</u>	<u>100 m</u>	<u>4350-4900 m</u>	<u>20</u>	DOC	
	Surface snow	<u>16th July, 2015</u>	<u>50 m</u>	<u>4350-4850 m</u>	<u>11</u>	DOC	
	Surface ice	15th August, 2015	<u>50 m</u>	<u>4350-4850 m</u>	<u>11</u>	DOC	
	Surface ice	25th August, 2015	<u>50 m</u>	<u>4350-4600 m</u>	<u>6</u>	DOC, absorbance	
<u>s</u>	ubsurface ice	25th August, 2015	<u>50 m</u>	<u>4350-4600 m</u>	<u>5</u>	DOC, absorbance	
Prog	lacial streamwater	<u>29th-30th July, 2014</u>	<u>2h (day),4h (night)</u>	<u>4210 m</u>	<u>17</u>	DOC	
Prog	acial streamwater	<u>20th May-9th</u> October, 2015	Every day	<u>4210 m</u>	<u>184</u>	DOC	

^{*} for snowpit it is vertical resolution, for surface ice and snow it is horizontal distance.

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Table 24. Comparison of DOC concentrations in snow and, ice and proglacial streamwater from the glacier in this

study and glaciers in other regions.					
Sites	DOC concentration (µg L ⁻¹) Sample types		References		
Laohugou glacier (LHG)	332 .4 ±132 .3	Snowpit	This study		
Tanggula glacier (TGL)	21 6.9 7 ±14 2.8 3	Snowpit	(Van et al. (2015)		
Mount Everest (EV)	15 2.5<u>3</u> ±56.11	Snowpit	(Tail et al. <u>(</u> ; 2013)		
Mendenhall Glacier, Alaska	190	snowpit	<mark>(</mark> Stubbins et al. , (2012)		
Greenland ice sheet	40 .3<u>1</u> - 5<u>6.97</u>	Snowpit	(Hagler et al. , <u>(</u> 2007)		
Laohugou glacier (LHG)	229 <mark>.3</mark> ±104.4	Surface snow	This study		
Greenland ice sheet	111 .2	Surface snow	(Hagler et al. , (2007)		
Juneau Icefield, Southeast Alaska	100 - 300	Fresh snow/snowpits	(Fellman et al. ,(2015)		
Laohugou glacier (LHG)	42 5.8<u>6</u> ±269.9<u>70</u>	Surface ice	This study		
Mount Nyainqentanglha Glacier	212 <mark>.4</mark>	Glacier ice	(Spencer et al., (2014)		
Antarctic ice sheet	460 <u>±</u> 120	Surface ice	(Hood et al. , <u>(</u> 2015)		
Alpine glacier	138 ±96	Subsurface ice	(Singer et al. , <u>(</u> 2012)		
Laohugou glacier (LHG)	23 7.5<u>8</u> ±95.66	Proglacial streamwater	This study		
Mount Nyainqentanglha Glacier	26 1.6 2	Proglacial streamwater	(Spencer et al. ;(2014)		
Mendenhall Glacier, Alaska	380 ±20	Proglacial streamwater	(Stubbins et al., (2012)		

Table 32. Mass absorption cross section (MAC) and Absorption Ångström Exponent (AAE₃₃₀₋₄₀₀) of ice and snow from LHG glacier and aerosols from other regions.

<u> </u>							
-	Site/Source	MAC $(m^2 g^{-1})$	AAE ₃₃₀₋₄₀₀	λ <u>(MAC)</u>	References		
_	LHC algoian	1.4 ± 0.4 (snow)	5.0 ± 5.9 (snow)	265	This study		
	LIG glacier	1.3 ±0.7 (ice)	3.4 ±2.7 (ice)	303			
	Biomass smoke	5.0	4.8	350	(Kirchstetter, <u>et al.</u> (2004)		
	Secondary organic aerosols	0.001 - 0.088	5.2 - 8.8	405	(Lambe et al. , <u>(</u> 2013)		
	Wood smoke	0.13 - 1.1	8.6 - 17.8	400	(Chen and Bond, (2010)		
	HULIS, Arctic snow	2.6 ± 1.1	6.1 ^{<u>×</u>a}	250	(Voisin et al. , 2012)		
	Beijing, China (winter)	1.79 ± 0.24	7.5	365	(Cheng et al. , (2011)		
_	Beijing, China (summer)	0.71 ± 0.20	7.1	365	(Cheng et al., (2011)		

^a[∞] the wavelength range for AAE of this study is 的波长范围为 300 - 550 nm

998	Figure 1.	Location	map of	LHG	glacier	No.	12.
	<u> </u>				~		

- 999 Figure 2. Average DOC concentrations of ice, snow and proglacial streamwater for LHG glacier.
- 1000 Figure 3. Variation in DOC concentrations in profiles of studied snowpits. The gray rectangles are dirty layers.
- 1001 Figure <u>34</u>. Exponential deceases in DOC concentrations during the biodegradation experiment. Note: The blue
- 1002 point is calculated using equations derived from the experimental data (black point).
- Figure 4<u>5</u>. Absorption spectra for DOC in snow and ice of LHG glacier and the dust and desert sand-from
 surrounding areas.
- 1005 Figure <u>65</u>. Comparison of DOC concentrations (A) and MAC₃₆₅ (B) between surface and subsurface ice.
- 1006 Figure <u>76</u>. The discharge, DOC concentrations and fluxes exported from LHG glacier. Note: The concentrations
- 1007 with error bars include more than one sample on that day.























Figure <u>76</u>. The discharge, DOC concentrations and fluxes exported from LHG glacier. Note: The concentrations with error bars include more than one sample on that day.