We appreciate the reviewers for their constructive comments and suggestions. The manuscript has been revised accordingly. Our point-by-point responses to the comments are presented below. The comments are in **black** and responses in **blue**. Changes made to the manuscript are in **red**.

Response to the reviewer #1:

The chemical evolution of snow is an interesting subject, and the authors provide some novel results to add to the research field. The paper outlines multiple analyses into the DOM, and find links between DOC and snow evolution, DOC and UV and changes in biologically related DOM components. The lack of statistics is worrying and needs to be addressed as a priority, however, taking the descriptions of values at face value, the story is complex and more effort needs to invested in picking out the likely reasoning behind these changes. I come away with a haze of knowledge about which components increase and decrease in concentration with ageing snow state, but some well rounded sentences in the conclusion, and a good schematic of the overarching findings could make this learning experience all the better.

Thank you very much for your insightful suggestion and positive comments.

Major concerns:

1) STATISTICS. My greatest concern with this manuscript is the severe lack of statistics to show the relationships between different sets of values. From the presentation of standard deviations, to performing appropriate comparative or correlation analyses, this element of the paper is worryingly poor. Without it, there can be no way for either the authors, not the reader, to interpret the results meaningfully. Looking at figures 2 and 3, and assuming that error bars are the SD (please include this in the legend alongside the n values), it's hard to imagine that significant differences would be found between these groups. Statistics would make this more convincing, and until these are included the rest of your arguments / conclusions / hypotheses cannot be considered.

We agree the reviewer's comment. We have added the statistical analysis in update manuscript. The description for figures 2 and 3 were updated as follows which have included the SD alongside the n values. We have included the explanation for SD in the caption figure 2 and 3 and marked the significant differences (letter a, b and c) in the bar graph according to the result of one-way ANOVA analysis.

"Their monthly mean values are presented in Figure 2a and the values decreased in the following order: September (59.6 ± 38.64 μmol L^{-1}), August (35.7 ± 20.2 μmol L^{-1}), October (26.4 ± 15.3 μmol L^{-1}), May (22.8 ± 11.1 μmol L^{-1}), June (19.8 ± 11.4 μmol L^{-1}), and July (18.6 ± 9.5 μmol L^{-1})."

"The average DOC concentration of fresh snow was 26.8 ± 10.8 μmol L^{-1}, which decreased to 15.0 ± 6.0 μmol L^{-1} for fine firn (*t*-test, $p < 0.05$); these concentrations then gradually increased for coarse firn (26.2 ± 16.1 μmol L^{-1}) ($p < 0.05$) and granular ice (34.4 ± 34.9 μmol L^{-1}) ($p > 0.05$) "

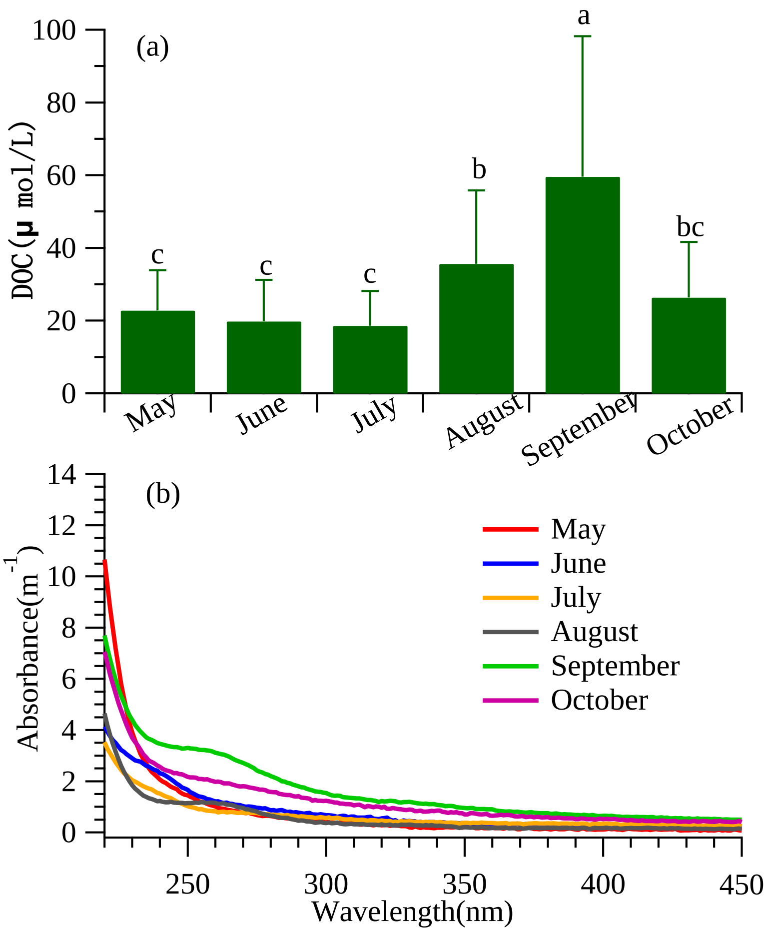


Figure 2. (a) Average mass concentrations and (b) UV-Vis absorbance spectra of dissolved organic carbon (DOC) in snow/ice samples from each month. The error bars indicate the standard deviation. Different letters indicate significant differences in mean values (one-way ANOVA). Mean values with the same superscript letters (a, b and c) were similar and no statistically differences were observed for these samples.

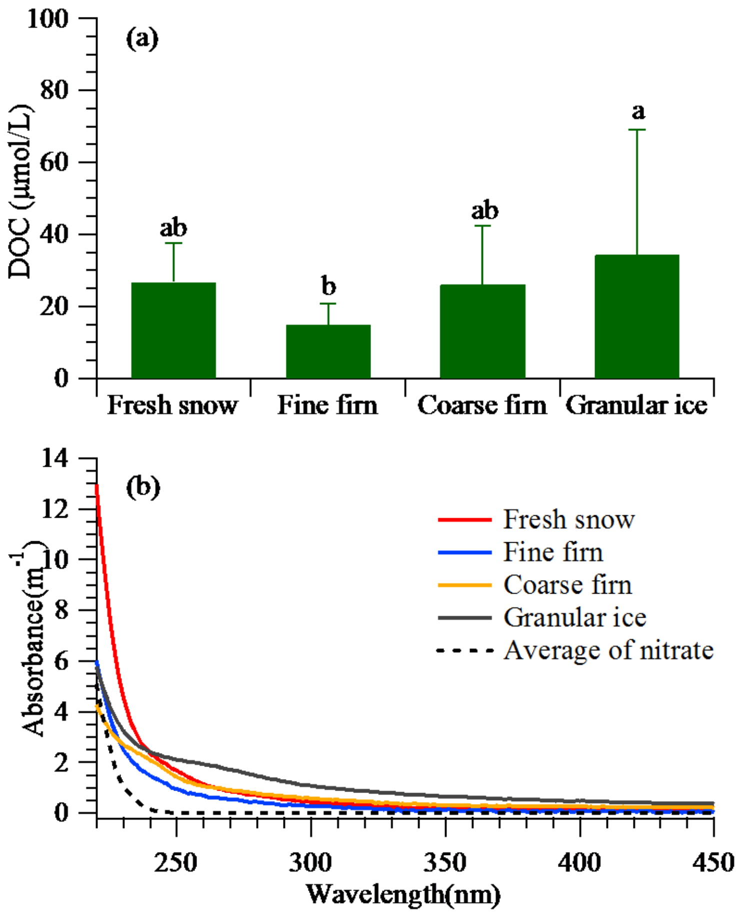


Figure 3. (a) Average mass concentrations and (b) UV-Vis absorbance spectra of dissolved organic carbon (DOC) in each category of snow/ice sample. The error bars indicate the standard deviation. Different letters indicate significant differences in mean values (one-way ANOVA). Mean values with the same superscript letters (a and b) were similar and no statistically differences were observed for these samples.

2) The INTRODUCTION needs to be sharpened up to priorities the background information that is relevant to this subject area (see below for specific details)

Thank you for your very valuable suggestion. We have revised and reorganized first two paragraphs as follows:

"Mountain glaciers are a critical source of fresh water, and their response to climate change has an impact on the regional water supply. In addition, glacier melt water also exerts an important influence on the biogeochemical properties of downstream ecosystems (Hood et al., 2015; Robinson et al., 2016; Saros et al., 2010). Hood et al. (2009) observed that the high bioavailability of dissolved organic matter (DOM) in glacial streams had a high impact on marine microorganisms in the coastal zone. Due to the high abundances and large degree of chemical diversity of DOM in global glacial streams (e.g., C, H, O, N, P, and S) (Fegel et al., 2016; Hood et al., 2015; Singer et al., 2012), the biogeochemical effects of glacial melt water have recently attracted much attention (Bhatia et al., 2013; Milner et al., 2017).

As the "Water Tower of Asia", the Tibetan Plateau (TP) glaciers have exhibited dramatic decline in recent decades, with models predicting that glacier area shrinkage could be as high as 26.7% by 2050, and with glacier runoff increasing until its maximum in about 2030 (Li et al., 2008; Yao et al., 2012). Consequently, the dissolved substances stored in glacial environments would be liberated to the downstream ecosystems and enter contemporary biogeochemical cycles. For example, Liu et al. (2016) estimated the release rates of dissolved organic carbon (DOC) from the glaciers in Western China at 15.4 ±6.1 Gg yr^{-1} which was more efficient than those observed in the Polar Region relative to their glacier storage. Xu et al. (2013) found that contributions of DOM could account for more than 50% of the dissolved substances in snow samples collected from glaciers in the Himalayas. This high content of DOM has also been shown to be highly biolabile, with past estimates from TP glaciers showing biolability (Spencer et al. (2014). Our previous work revealed a predominant biological component of DOM in cryoconite (which is a dark-colored, dust-like material found on the surfaces of glaciers) from the TP (Feng et al., 2016). However, the dynamic evolution of chemical components of DOM during the snow/ice melting process remains poorly characterized."

3) The DISCUSSION should attempt to draw out and summarize the findings of the study. This is a complex story with concentrations going up and down between different sample groups at different times. A schematic could be the ticket here to imprint the hypothesized story in the head of the reader; pulling together a narrative of snow melt, freeze-thaw enrichment, exogenous sources and microbial activity.

We have deleted the discussion on cryoconite and added the explanation for the DOM enrichment during the melting process:

"Previous study has observed that supraglacial communities during the melt season are photosynthetically active, with production rates often exceeding respiration rates (Boetius et al., 2015), therefore, DOM is produced and enriched (i.e., net autotrophy) at this time (Bagshaw et al., 2016), which includes bacterial production (Xie et al., 2009), chlorophyll-a derived from algal phototrophs and the rainbow of pigment colors (Foreman et al., 2013). In addition, wet and dry deposition from the atmosphere also make an important contribution to the accumulation of DOM on the surface of the glacier during the ablation season (Clement et al., 2012; Spencer et al., 2014)."

We have deleted line of 394-400 in discussion part.

At the end of discussion, in order to illustrate the linkage between the changes in DOM and changes in biomolecular composition, we have added the discussion for the results of redundancy analysis (RDA).

"In addition, from fresh snow to granular ice, the changes in lignins/CRAM, lipids and aliphatic/proteins-like components showed a positive correlation with the changes in DOC concentration (Figure 9), suggesting that biochemical evolution plays an important role in composition of DOM during the process of snow melting."

Minor concerns:

Title: I think this should be reconsidered. The snow is physically evolving, the resulting effect is changes to the chemistry. Perhaps this is a good chance to be bold and pull in the concept of biology into this title. In general the title would be more appealing as a statement of the findings, rather than a description of the study.

Thank you very much for your suggestion.

The title is revised to "Physical and biochemical evolution of dissolved organic matter across the ablation season for a glacier on the central Tibetan Plateau"

Introduction:

The introduction seems a little off-track and unhelpful for getting into the subject matter. You start your ms about water supply, however this is very tenuously linked to the purpose of the project. The same is true for lines 43 - 48. Please remove these sections / replace with another concept of why this subject is important to study / make clearer the links between water supply and your study. Lines 29 - 31 also seem a distraction, as do lines 36-38

We have removed the sentences that are note close related with the topic of this manuscript. The introduction is focused on the biogeochemical effect of DOM and more easily to follow now as follows.

"Mountain glaciers are a critical source of fresh water, and their response to climate change has an impact on the regional water supply. In addition, glacier melt water also exerts an important influence on the biogeochemical properties of downstream ecosystems (Hood et al., 2015; Robinson et al., 2016; Saros et al., 2010). Hood et al. (2009) observed that the high bioavailability of dissolved organic matter (DOM) in glacial streams had a high impact on marine microorganisms in the coastal zone. Due to the high abundances and large degree of chemical diversity of DOM in global glacial streams (e.g., C, H, O, N, P, and S) (Fegel et al., 2016; Hood et al., 2015; Singer et al., 2012), the biogeochemical effects of glacial melt water have recently attracted much attention (Bhatia et al., 2013; Milner et al., 2017).

As the "Water Tower of Asia", the Tibetan Plateau (TP) glaciers have exhibited dramatic decline in recent decades, with models predicting that glacier area shrinkage could be as high as 26.7% by 2050, and with glacier runoff increasing until its maximum in about 2030 (Li et al., 2008; Yao et al., 2012). Consequently, the dissolved substances stored in glacial environments would be liberated to the downstream ecosystems and enter contemporary biogeochemical cycles. For example, Liu et al. (2016) estimated the release rates of dissolved organic carbon (DOC) from the glaciers in Western China at 15.4 ±6.1 Gg yr-1 which was more efficient than those observed in the Polar Region relative to their glacier storage. Xu et al. (2013) found that contributions of DOM could account for more than 50% of the dissolved substances in snow samples collected from glaciers in the Himalayas. This high content of DOM has also been shown to be highly biolabile, with past estimates from TP glaciers showing biolability (Spencer et al. (2014). Our previous work revealed a predominant biological component of DOM in cryoconite (which is a dark-colored, dust-like material found on the surfaces of glaciers) from the TP (Feng et al., 2016). However, the dynamic evolution of chemical components of DOM during the snow/ice melting process remains poorly characterized."

Line 42 - please include appropriate references to this statement

We have added the appropriate references here as follows.

"Bhatia, M. P., Kujawinski, E. B., Das, S. B., Breier, C. F., Henderson, P. B., and Charette, M. A.: Greenland meltwater as a significant and potentially bioavailable source of iron to the ocean, Nat. Geosci., 6, 274–278, 2013.
Milner, A. M., Khamis, K., Battin, T. J., Brittain, J. E., Barrand, N. E., Füreder, L., Cauvy-Fraunié, S., Gíslason, G. M., Jacobsen, D., and Hannah, D. M.: Glacier shrinkage driving global changes in downstream systems, Proc. Natl. Acad. Sci. U. S. A., 114, 9770, 2017."

line 47 Just in this area? What about other glaciers world-wide?

We have deleted this sentence in the update manuscript.

Lines 50 - 52: state why this is of importance to know

We have rephrased this section (see response above) and this sentence is used to emphases on the rapid melting of glacier in the TP which will release plenty of DOM.

Lines 55 - 61: Again this moves away from the specificity of the paper. Either remove, or make links as to why this information is important to assimilate in the introduction.

We have rephrased this section (see response above).

Experimental Details:
line 86 - I don't see any sample lines with five in them.

We made a mistake here and have revised it as follows.

"At each elevation, two to four parallel samples were collected along the transect of the glacier (Figure 1)".

Line 87 - better to describe this sampling strategy as being done along transects of similar elevation?

We have revised this sentence as follows.

"At each elevation, two to four parallel samples were collected along the transect of the glacier

(Figure 1)”.

Line 91 - what was the filtrate collected into?

We have revised this sentence as follows.

“The filtered samples were collected in 50 ml FalconTM tubes and frozen at −20 °C in the dark before transportation, during which the samples were kept frozen in a cooler box filled with blue ice.”

Line 92 - Did you freeze after filtration?

Yes and please refer to the response above.

Line 96 - include a figure to show the different ice types so that others can replicate these methods with more confidence. Is there a reference to use for the evolution of snow types?

We have added a figure (Figure R1) in the supplementary information of the updated manuscript (Figure S1) to show the different ice types and included a reference for evolution of snow type as follows.

“Hock, R.: Glacier melt: a review of processes and their modelling, Progress in Physical Geograph y, 27, 362–391, 2005.”

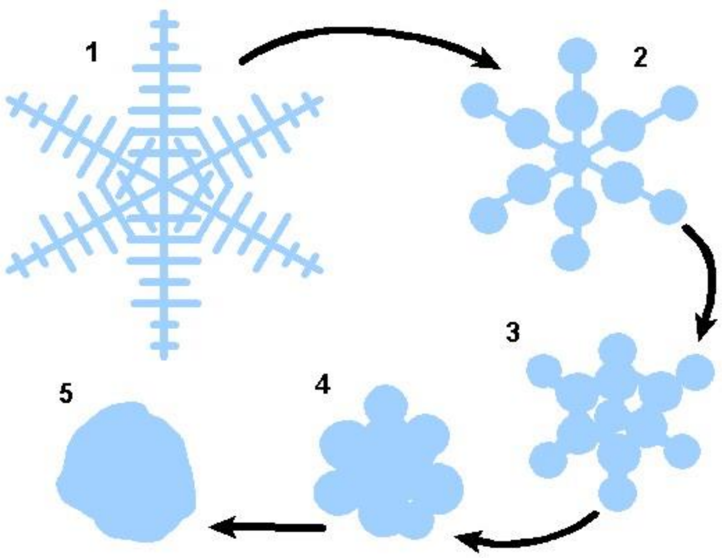


Figure R1. The physical evolution (metamorphism) of snow: fresh snow (stage 1), fine firn (stage 2), coarse firn (stage 3), granular ice (stage 4) and glacier ice (stage 5).

Line 105 - I think these mixed samples would be of interest. Surely they help to add to the story. Please include them.

Thank you for your kindly suggestion. As shown in figure R2, the mass concentration of mix snow samples were similar with those of the fresh snow and coarse ice which suggested a mixed information. The UV absorption of mix snow samples showed the same trend with fresh snow between 220-250 nm, and then showed a same variation with coarse firn after 250 nm. In order to more clarify the trend of the DOM evolution during snow ablation, we think it is better to remove these mixed samples.

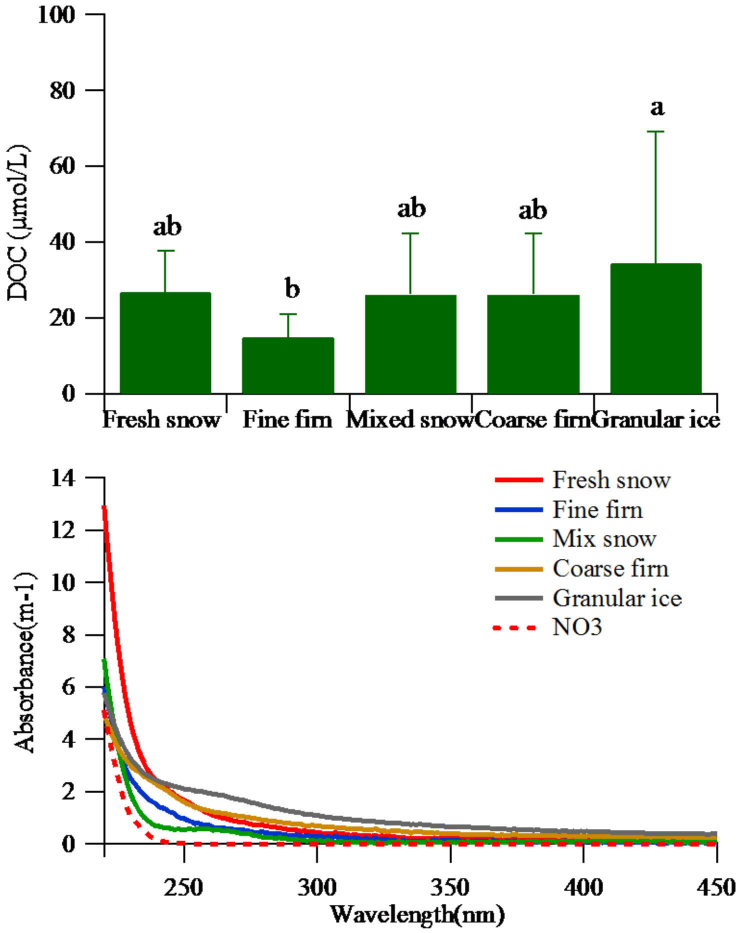


Figure R2. Average mass concentrations of dissolved organic carbon (DOC) in snow/ice samples from different snow/ice type. The error bars indicate the standard errors. Different letters indicate significant differences in mean values (one-way ANOVA). Mean values with the same superscript letters (a, b and c) were similar and no statistically differences were observed for these samples.

Results:

Line 242 - "from" instead of "in"

Revised as suggested by the Referee.

Lines 244 - 245 and 249 - 250 - why show the maximum values, why not mean with SD? Surely this is more representative of your samples.

These values for DOC concentration in each month are mean values not the maximum. We have revised this sentence to make more clear as follows. In addition, we added the SD for each value.

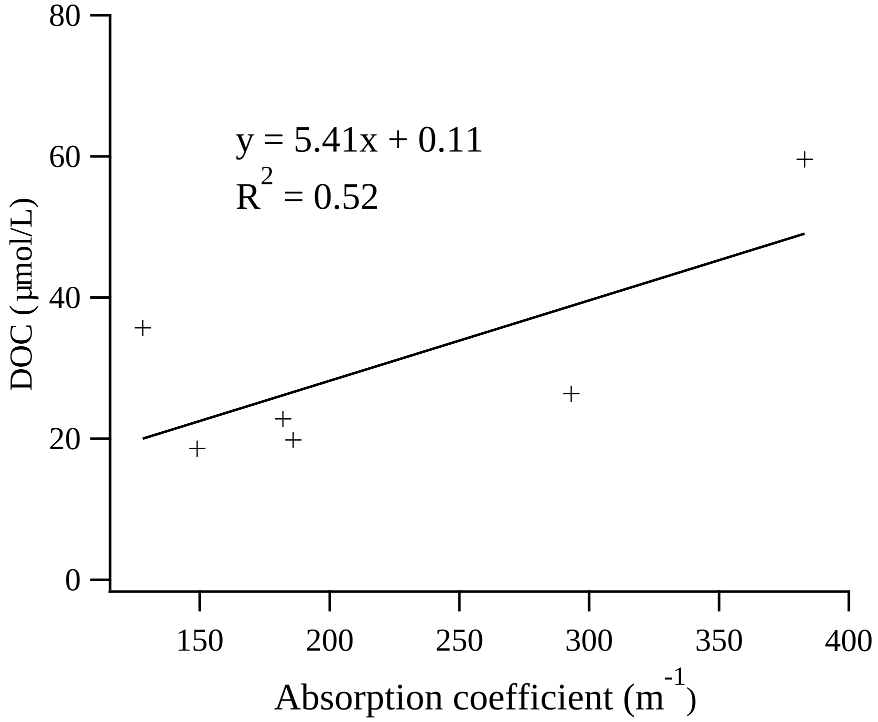
"Their monthly mean values are presented in Figure 2a and the values decreased in the following order: September (59.6 ± 38.64 μmol L^{-1}), August (35.7 ± 20.2 μmol L^{-1}), October (26.4 ± 15.3 μmol L^{-1}), May (22.8 ± 11.1 μmol L^{-1}), June (19.8 ± 11.4 μmol L^{-1}), and July (18.6 ± 9.5 μmol L^{-1})."

"The sum of the absorbance between 220-450 nm in each monthly average spectrum varied evidently; in accordance with the order from high to low was September (383 ± 241), October (293 ± 133), June (186 ± 83), May (182 ± 93), July (149 ± 96) and August (128 ± 75)."

Lines 247 - 248 - use correlation analysis to prove this

The scatter plot between DOC concentration and sum of absorption coefficient (between 220-450 nm) in each month is shown below which presents a good correlation between them. We have added this information in the updated manuscript as follows.

"The variability in the chemical contents of the snow/ice samples showed a correlation (R^2 = 0.52, p > 0.5) with the variability in their UV-Visible absorbance spectra (Figure S3)."



Line 251 - what does "exhibited significant absorbance" mean? How significant?

We have replaced significant with obvious.

Lines 255 - 275 - Every time a comparison or correlation is made, you need to state the statistical significance of these. There are numerous cases of this in this section. All need to be dealt with.

We have added the statistical analysis here, and revised the similar issue in the updated manuscript.

"The average DOC concentration of fresh snow was 26.8 ± 10.8 μmol L^{-1}, which decreased to 15.0 ± 6.0 μmol L^{-1} for fine firn (*t*-test, p<0.05); these concentrations then gradually increased for coarse firn (26.2 ± 16.1 μmol L^{-1}) (p<0.05) and granular ice (34.4 ± 34.9 μmol L^{-1}) (p>0.05).

The sum values of the average absorbance between 220–450 nm in four categories were fresh snow (212 ± 90), fine firn (116 ± 58), coarse firn (170 ± 92) and granular ice (267 ± 181), which showed a significant correlation (R2 = 0.93, p < 0.05) with DOC concentration (Figure S4). The average $SUVA_{254}$ values obtained for each category sample were 1.96 ± 0.40 for fresh snow, 1.85 ± 0.49 for fine firn, 2.39 ± 1.63 for coarse firn, and then increased to 3.05 ± 1.93 (p < 0.05) for granular ice, which revealed a significantly high abundance of aromatic constituents in granular ice samples (Weishaar et al., 2003). The values of the S_R for each category were 1.56 ± 0.05 for fresh snow, 1.76 ± 0.68 for fine firn, decreased to 1.6 ± 0.04 for coarse firn (p > 0.05), and 1.57 ± 0.03 for granular ice (p > 0.05), which suggests that the molecular weight of the DOM changed during melting stage (Helms et al., 2008)."

Line 281 - can you include the 4 components from this study into the table as C1- 4.

We have included the 4 component into the Table S1.

Line 294 - it's not a clear trend unless you use statistics to analyze the trends

Agree, we have added the *t* tests analyses for these results in the update manuscript as follows.

"The relative contributions of aliphatic/proteins and lignins/CRAM increased from 33% to 40.3% (t-test, p<0.05) and from 19% to 27% (p<0.05), respectively, from fine firn to granular ice. The relative contributions of lipids (from 42% to 29.2%, p<0.05), unsaturated hydrocarbon content (from 3% to 1.7%, p<0.05), and condensed aromatics (from 1% to 0.5%, p<0.05) all exhibited significant decreasing trends, yet carbohydrates showed a decrease (from 2% to 0.8%, p<0.05) from fine firn to coarse firn, and then increased up to 1.4% (p<0.05) in granular ice. Notably, the fractions of aliphatic/proteins and lignins/CRAM classes in fresh snow were higher than those in fine firn (p<0.05)."

Line 295 - what does "generally" mean?

We have deleted it.

Line 302, 303, 309 - 313 – stats

Agree, we have added the *t* tests analyses for these results in the update manuscript as follows.

“The average molecular mass of the assigned DOM gradually increased from fine firn (376) to granular ice (396) ($p<0.05$) (Table 3), and the assigned molecular number increased from 2469 to 3933 (Table 4) ($p>0.05$).

We found that the contributions of CHO molecules decreased from 77.3% to 71.7% ($p < 0.05$) from fine firn to granular ice, while those of CHON molecules increased from 22.2% to 28.2% ($p < 0.05$). Therefore, the ratio between CHON and CHO increased significantly, from 0.287 to 0.393 ($p < 0.05$), from fine firn to granular ice. We also calculated the percentages of molecules with low C/N ratios (≤ 20) in each category of samples, the value increased from 10 to 17.7% ($p < 0.05$) from fresh snow to granular ice.”

Line 305 - unsure what this means

We have restated this sentence as follows.

“The relative mass content of carbon (Cm) in each average formula of the molecular classes showed different values in four category samples, thus reflecting the chemical composition of DOM which could be altered by the biochemical dynamics during the melting process of snow ice (Table S2).”

Line 316 - state as a percentage. When looking at these unique components does it say anything about the flow of molecules?

We have added the percentage number of each overlapped component in four categories. In addition, for the unique components in each category, we analyzed the percentage contribution of CHO and CHON molecular (pie chart) to reveal the changes of the DOM component in melting process.

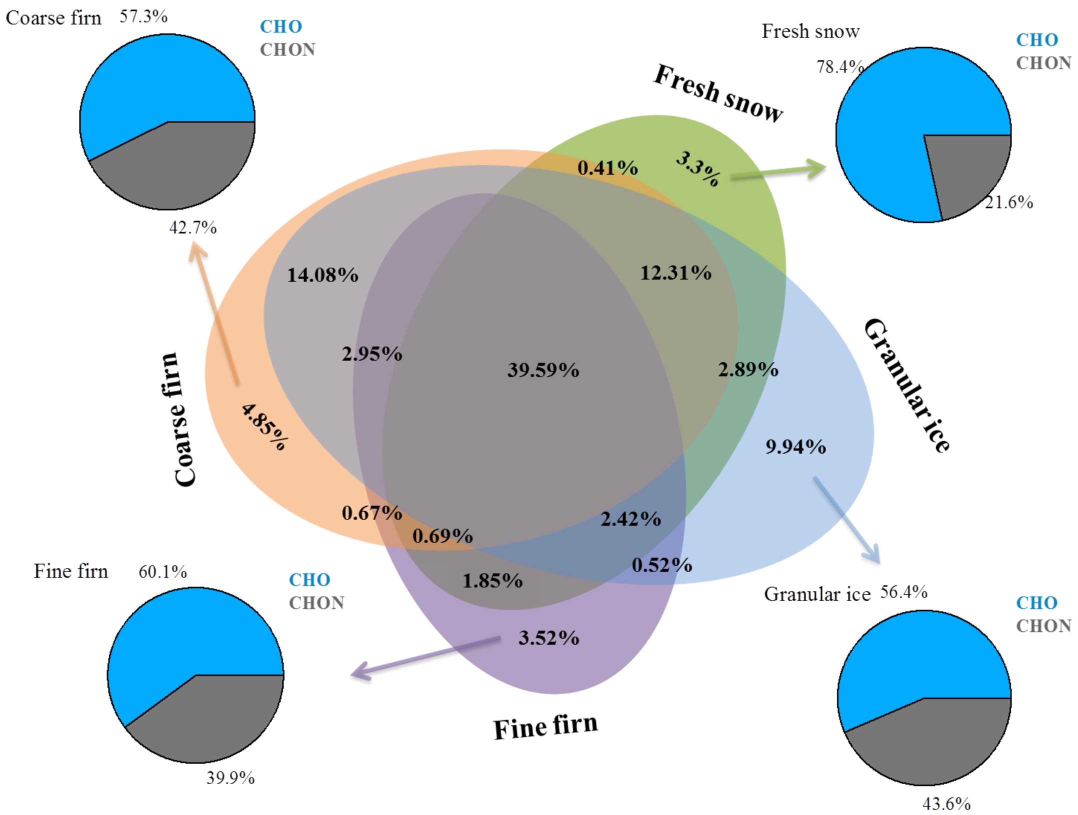


Figure 7. Four-way Venn diagram shows the overlap in molecular formulas between the fresh snow, fine firn, coarse firn and granular ice. The numbers within the Venn diagram are the percentage of overlapped component in four categories and unique component in each category. The relative contributions of CHO and CHON molecular classes are shown in unique components in the four pie charts i.e. Fresh snow, Fine firn, Coarse firn and Granular ice.

DISCUSSION:

Line 349 - I don't see the need for the discussion on cryoconite - consider removing

We have deleted the discussion on cryoconite.

Line 373 - find a better term than "fresh nature"

We have changed the "fresh nature" to "fresh feature".

Line 379 - I'm unsure what you mean by "intense" but given how these cryoconite samples were prepared, I don't think that this is a fair comparison. I'd suggest removing it and finding a more appropriate comparison if necessary.

We have removed this sentence in the update manuscript.

Line 385 - please be more clear about what you are discussing here - which analysis / result does this relate to? In general, condense this paragraph and stay focused on the discussions that matter.

Thank you for your suggestion. We have revised this part as follows.

"Mixed chemical compositions were found not only in granular ice samples but also in fresh snow samples, which were mainly composed of atmospheric aerosols from within and outside of clouds. These mixed chemical characteristics were also seen in other studies of fog water (Zhao et al., 2013) and ambient aerosols (Mazzoleni et al., 2012) in other remote areas. This suggests that the aerosols in this remote area of the TP mostly originated from natural sources. Previous study has suggested that primary biological aerosol particles (PBAPs), including bacteria, spores and pollen, which could be more abundant during summer due to more favorable air temperature and humidity conditions (Toprak and Schnaiter, 2013), may be important for formation of clouds and precipitation (Després et al., 2012). Therefore, biological aerosols could be an important source of the microbially derived components (including lipids and aliphatic/proteins) in fresh snow."

Line 389 - 394 seem redundant, Lines 394 - 400 seems extensive given this was not found in the sample.

Thank you for your suggestion. We have deleted the Line 394-400, and revised Line 389-394 as follows to illustrate the source of DOM in fresh snow.

"Previous study has suggested that primary biological aerosol particles (PBAPs), including bacteria, spores and pollen, which could be more abundant during summer due to more favorable air temperature and humidity conditions (Toprak and Schnaiter, 2013), may be important for formation of clouds and precipitation (Després et al., 2012). Therefore, biological aerosols proved to be the important source of the microbially derived components (including lipids and aliphatic/proteins) in fresh snow."

Line 407 - can you link changes in DOM with changes in the compositions of the biomolecules? Perhaps using a redundancy analysis. What can you tell from this? This ms discussion needs a deeper look at the biological component of this process if you deem it to be an important aspect.

Agree, we have done the redundancy analysis and showed the results as follows and added the discussion for this in the updated manuscripts.

"A redundancy analysis (RDA) was used to evaluate relationships between the changes in DOC concentration and changes in each chemical composition of DOM (Figure 9). The results showed a positive correlation between DOC concentration and the compositions of lignins/CRAM, aliphatic/proteins and lipids, indicating that concentration of DOC changes in each category samples was depended, to a large extent, upon the changes in the compositions of lignins/CRAM, aliphatic/proteins and lipids."

"In addition, from fresh snow to granular ice, the changes in lignins/CRAM, lipids and aliphatic/proteins-like components showed a positive correlation with the changes in DOC concentration (Figure 9), suggesting that biochemical evolution plays an important role in composition of DOM during the process of snow melting.”

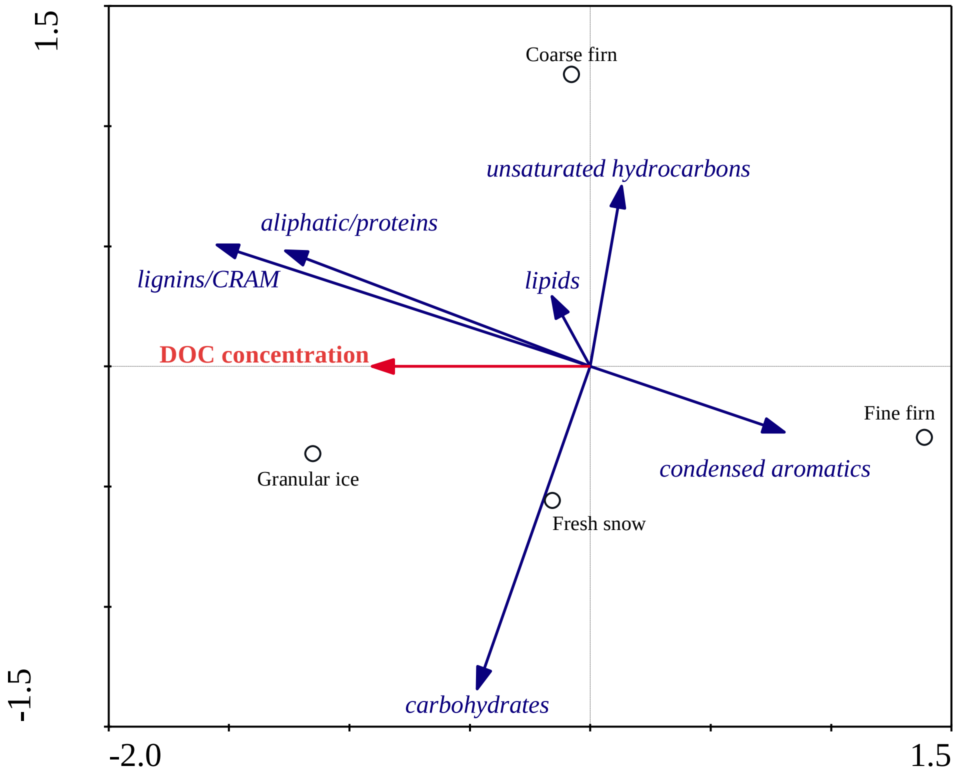


Figure 9. Redundancy analysis (RDA) biplot visualizing the link changes in DOC concentration with changes in the compositions of each chemical component assigned by FT-ICR-MS. Angles between vectors represent correlation; i.e., smaller angles indicate higher correlation. Red arrows denote DOC concentration for each category samples, and blue arrows denote the numbers of each chemical component in each category samples, and blank circles denote the four category samples.

Provide a discussion on the correlation between DOC and UV.

Agree, we have added a discussion on the correlation between DOC and UV as follows.

“The significant correlation (R^2 = 0.93, $p < 0.05$) between DOC concentration and the sum of absorption also proved the increase of CDOM in the pool of glacial DOM during the melting process.”

CONCLUSION: rather than a repeat of your results, summarize it in an amenable manor.

We have rewritten the part of conclusion.

Line 436 - this is a discussion not a conclusion of your work. Either bulk it up and place it in the discussion or remove altogether. It otherwise it is redundant.

We have rewritten the part of conclusion.