

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 #2:

This manuscript presents some very interesting results on the evolution of organic matter in the snowpack from DKMD glacier on the Tibetan Plateau. It is becoming apparent that organic matter may behave differently from atmospheric aerosol species in the snowpack as it ages, and the authors present a detailed description of some of these changes and the possible mechanisms involved.

Thank you very much for your positive comments.

Despite these interesting results, I find that the manuscript as written is very confusing and the authors omit key descriptions of how and where the snow was sampled. This leaves the reader without a context in which to frame the results that are presented. Specifically, the authors have sampled snow from a large number of sites above and below the equilibrium line (5600 m) on the surface of DKMD glacier, yet refer to changes in the snowpack as it "melts" over the summer season. Surely, melting, and the generation of meltwater and subsequent snowpack flushing and mobilization of DOM within the snowpack, will differ greatly between snow found below the ELA (in the ablation zone) and snow found above the ELA (in the accumulation zone). I would even question the existence of a firm layer within the snowpack in the lower ablation zone, yet the authors seem group all of their samples together and discuss them as being equal in structural composition, and by extension, the generation of meltwater. Further, I question the use of the term "melting" throughout the manuscript. The first line of the abstract mentions snowpack "metamorphism", which implies processes that occur as the snowpack ages, and does not necessarily include the production of meltwater. The use of the term "melting" throughout the remainder of the manuscript implies the transformation of the snowpack from the solid to liquid phase, or at the very least, the generation of some meltwater which would be expected to mobilize a significant fraction of the DOM due to flushing. I believe that "metamorphism" would be the more correct term to use here, at least in the accumulation zone, unless melting truly is happening, in which case the authors would need to specify this and integrate it into their interpretation. Additionally, do you have a multi-year snowpack in the accumulation zone? It would be interesting to hear about the patterns that you see over several years.

Thank you very much for your wonderful comments. We have added more information to describe the state of this glacier during melting period as follows. Actually, this glacier is a small glacier with a length of 2.8 km and an average width of 0.5 – 0.6 km which is more easily influenced by the weather conditions, and then most of the part is located below ELA. Yao et al. (2012) have shown that this glacier is undergone fast retreat during last 20 years with annual mass loss rate of about 300 mm w.e. and usually looked as typical glacier in the TP to display the melting state of glacier in this region. The data of ELA is cited from the literature 2002 years which are not exactly correct for the glacier now. The ELA in the DKMD glacier has varied in the

range 5450–5850 m a.s.l. during the past 14 years (1989–2002). During the sample collecting, we found the surface melting covering everywhere. The snow pit dugged at the upper section of the glacier during the end of the melting period was only 120 cm due to the strong melting. Under this strong melting situation, the feature of the same type snow sample is similar and can be combined together.

“Due to the rapid increase in air temperature that has occurred in recent decades, this glacier has experienced severe ablation (Pu et al., 2008), with an annual mass loss rate of about 300 mm w.e. during the last 20 years (Yao et al., 2012). With a gentle surface and without any avalanche or surface moraines, this glacier is typical of glaciers on the TP and displays the melting state of numerous glaciers in this region. The equilibrium-line altitude (ELA) for the DKMD glacier has varied in the range 5450–5850 m a.s.l. during recent years (1989–2002). The DKMD glacier is an extremely continental (polar) glacier, with an annual precipitation of approximately 300 mm and it is dominated by summer precipitation from the Asian monsoon. Only 21 days showed a daily average air temperature above 0 °C, and the maximum air temperature was 6.5 °C. Temperatures at the ELA are estimated at –10 °C. In such extremely cold and dry conditions, ablation of surface snow represents a slow and continuous process that provides a good model for studying the chemical evolution of DOM across the different physical snow types.”

Specific comments are as follows:

Line 7: what are the "stages of snowmelt"?

This means different physical descriptions of snow representing the different stages of snowmelt, i.e., fresh snow for new snow, fine firn for a slight thaw of fresh snow, coarse firn for further freeze-thaw processes of fine snow, and granular ice for the final state of snow. We have added a figure (Figure S1) to show the evolution of snow type as follows.

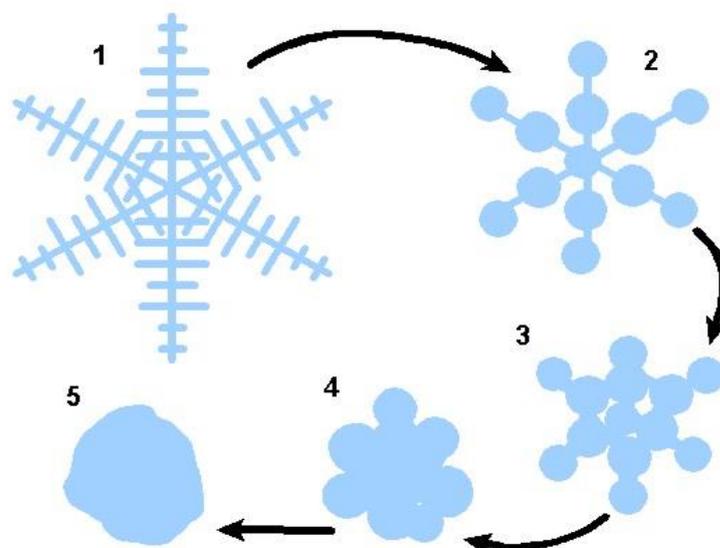


Figure S1. The physical evolution (metamorphism) of snow: fresh snow (stage 1), fine firn (stage

2), coarse firm (stage 3), granular ice (stage 4) and glacier ice (stage 5).

Line 51-52: not sure what "more efficient to be released" means

This means that the release rate of DOC for Chinese mountain glacier is higher than the polar regions based on the relative value of the glacier storage. The amount of DOC released annually from Chinese glaciers is ~7 – 20% of the amount released annually from the Greenland ice sheet, while Chinese glaciers account for ~0.2% of the volume of the Greenland ice sheet (Liu et al., 2016).

Line 52: "Polar Regions" doesn't need to be capitalized.

We have corrected the "Polar Regions" to "polar regions" in the revised version.

Lines 62-64: unclear what the difference between ablation and melting is here.

Sorry for confusing for you. We have revised this sentence and used the uniform word to express the same meaning as follows

“During ablation, glaciers can undergo leaching effects, in which certain amounts of dissolved matter, such as water-soluble inorganic ions, are percolated downwards and exported into the glacier runoff (Eichler et al., 2001; Hou and Qin, 1999). Sun et al. (1998) also found the chemical composition could also be changed during the melting process.”

Line 74: again, is this really snowmelt or metamorphism, firnification?

Yes. This is snowmelt here because we found the surface melting covering everywhere during the sample collecting.

Line 170: "widely used", yet you only provide a single reference. Can you list a few more?

We have revised this sentence and added more reference as follows to represent more exactly here.

“The combination of EEMs and PARAFAC has been a useful method applied to characterize DOM in aquatic environments and in glacier research (Barker et al., 2013; Barker et al., 2009; D'Andrilli et al., 2017; Dubnick et al., 2010).”

Barker, J. D., Dubnick, A., Lyons, W. B., and Chin, Y. P.: Changes in Dissolved Organic Matter (DOM) Fluorescence in Proglacial Antarctic Streams, *Arctic Antarctic & Alpine Research*, 45, 305–317, 2013.

D'Andrilli, J., Foreman, C. M., Sigl, M., Prisco, J. C., and McConnell, J. R.: A 21 000-year record of fluorescent organic matter markers in the WAIS Divide ice core, *Climate of the Past*, 13, 1–15, 2017.

Dubnick, A., Barker, J., Sharp, M., Wadham, J., Lis, G., Telling, J., Fitzsimons, S., and Jackson, M.: Characterization of dissolved organic matter (DOM) from glacial environments using total fluorescence spectroscopy and parallel factor analysis, *Annals of Glaciology*, 51, 111–122, 2010.

Line 256: why "gradually increased for coarse firn"? This suggests that [DOC] increased with depth in the coarse firn layer. Is this what you mean?

No. This means the DOC concentration in coarse firn sample was higher than fine firn sample. We just sampled the surface snow sample from DKMD (Figure 1) not the snow pit sample, so the fresh snow, fine firn, coarse firn and the granular ice were all sampled from the surface of sampling points on Figure 1. At the beginning of ablation season, most of the snow samples maybe attributed to the fresh snow and fine firn, but at the middle or late of the ablation season, most of the samples belonged to the coarse firn and granular ice. We use the average value of each category from more than 8 samples distributed widely on the surface of the glacier to represent the average state of DOC content and deduce the evolution of DOM in surface snow.

Line 272: "component"?

This was a mistake. We have changed “component” to “category”.

Line 275: again, "melting stage"? Is it melting?

Yes. From the fresh snow to granular ice, it is a freeze-thaw processes as shown in figure S1.

Line 282: you can delete "as described in the Supporting Information section".

We have deleted this in the updated manuscript.

Line 285: "important microbial origin". I think that what you mean is that C1&C2 have been associated with microbially-produced DOM, and it constitutes a large % of the total DOM fluorescence.

Yes. We revised this sentence as follows to make it more clearly.

“The relative contributions of C1 and C2 account for an average value of approximately 80% of the total DOM fluorescence in each category of samples (Figure 5), suggesting that a considerable amount of DOM in snow/ice samples in this region similar with DOM derived from the microbial sources (Barker et al., 2010; Yamashita and Tanoue, 2003).”

Lines 325-327: Is this true of all layers, or an integrated average for the snowpack? "snowmelt could have lost DOC" is misleading.

Sorry for making you confuse here. We have reworded this sentence as follows.

“It is worth noting that all snow/ice samples in this study were collected from the surface of the glacier, not from snowpack or snow pit samples. In addition, the values presented here are the integrated average values for each month, which include samples in different categories. The different mass concentrations of DOC in each month represent the average state of surface snow/ice. The lowest mass concentrations of DOC were observed in the summer period (June and July), therefore we conclude that the melting process of surface snow/ice could selectively lose DOC.”

Line 328: I remain confused by the word "melted". Are you claiming that in May, all that you were sampling was unaltered snow, and that over the course of an ablation season this snow was metamorphosed into firn? I don't think that this is what you mean...

I mean the samples from the May were mainly composed by the fresh snow, also including some fine firn samples, because May is the beginning of the ablation season, the melting in ablation zone is not very intense, but at the June, July and August most of the samples were coarse firn and granular ice which induced by the intense melting process.

Line 334: are you suggesting that the proglacial stream has received the nutrients flushed from the snowpack during summer? Surely this would be from the snow in the ablation zone only?

Yes, the proglacial stream will receive the nutrients flushed from the snow/ice during summer. The melting of the glacier could occur in the surface of the ablation zone and also in the bottom of the glacier (Williams et al., 2006), which the contribution of runoff from surface stream of the glacier could up to more than 30%. In addition, the water from the bottom of glacier could also from surface stream which went down the bottom through ice crevasse. Therefore, the mass concentration of DOC in proglacial stream could be closely related with the mass concentration of DOC in the surface snow/ice.

Line 366: all studies using fluorescence to characterize glacier melt have found that protein-like fluorescence dominates supraglacial DOM.

Agree, we have revised this sentence as follows:

“This finding is consistent with that of other studies, which recorded protein-like fluorescence dominating supraglacial DOM (Barker et al., 2009; Dubnick et al., 2010; Hood et al., 2009).”

Line 369: why use the Lake Taihu comparison? Why would it be relevant to glacier snow?

Accept. We have deleted this comparison with Lake Taihu here.

Line 422-424 loss of DOM from the snowpack? I don't get it. Where's the DOM going? Is it being flushed from the snowpack, volatilized, ...? Likewise "enrichment" in lower levels? Is DOM being produced?

As we discussed in section 4.1, the average DOC concentration in fresh snow samples was $26.8 \mu\text{mol L}^{-1}$ and decreased to $15 \mu\text{mol L}^{-1}$ in fine firn samples which mainly induced by percolation. The percolated DOM could be transported to the proglacial stream as illustrated by increased DOC concentration in June and July from measurement in proglacial stream.

From fine firn to granular ice, DOC concentration increased from $15 \mu\text{mol L}^{-1}$ to $34.4 \mu\text{mol L}^{-1}$, enriched by microbial production in situ.

Line 431 Lignins increasing with depth. Isn't this just an accumulation of recalcitrant DOM with downward flushing through the snowpack?

No. The increasing of lignin and proteins compounds were mainly due to the humidation and microbiology.

Line 436-438 This last sentence is very general to the point of being inconsequential.

Agree. We have reworded the conclusion part and revised this sentence as follows:

“Against the background of global warming, the melting of glaciers and release of DOM to downstream ecosystems is changing, leading to a pressing need to further understand both the sources of DOM and seasonality of DOM in glacier environments as it has relevance for local carbon cycles and ecosystem processes.”

Table 1: you need to provide references for your fluorescence peak assignments

Agree. We have added references for each fluorescence peak.

Table 1. Description of the four-component PARAFAC model. Wavelengths in parentheses represent secondary peaks.

Component number	Excitation maxima nm	Emission maxima nm	Description
C1	<230 285	322	Protein-like (tryosine-like or tyrosine-like), Peak T or Peak B (Dubnick et al., 2010)
C2	270	308	Protein-like (tyrosine-like), Peak B (Coble et al., 1998; Fellman et al., 2010)
C3	<230	424	Humic-like, Peak M (Fellman et al., 2010; Fellman et al., 2014)
C4	240 290	340	Protein-like (tryptophan-like), Peak T (Fellman et al., 2010)

Fig 1: the lines and labels for the mountain ranges are distracting and don't add any useful information

We have revised this figure as follows.

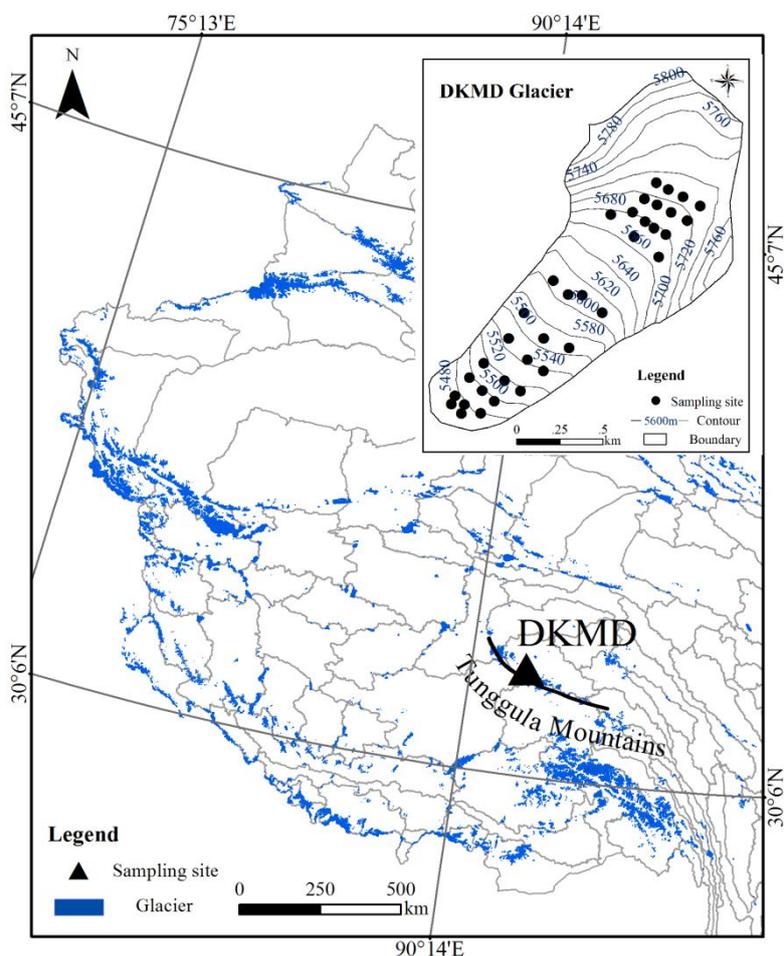


Figure 1. Map showing the locations of glaciers in the Tibetan Plateau and the distribution of snow/ice samples collected on the Dongkemadi (DKMD) glacier.

Fig 3: was fresh snow sampled in the summer? Did you have any summer snowfall?

Most of the fresh snow were sampled in May. There was snowfall during summer, but we did not collect snowfall samples. The snowfall in the surface of the galcire could be melting within several days due to the high air temperature during summer (6.5 °C).

Fig 3: the x-axis for abs in figs 2-3 don't match which is distracting when comparing the spectra

We have revised this two figures as follows:

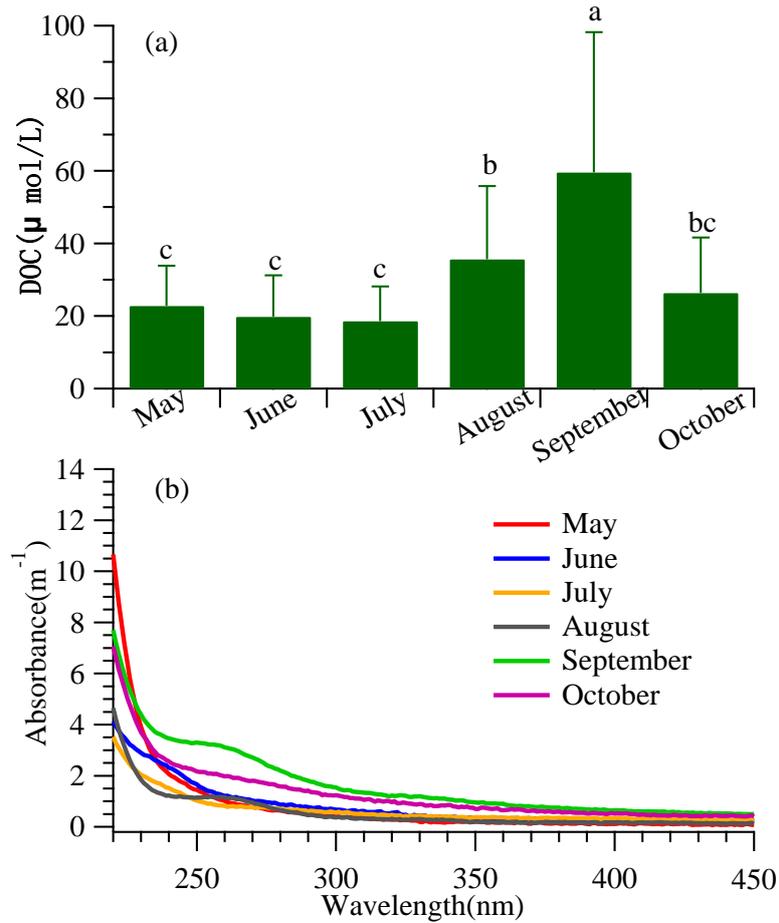


Figure 2. (a) Average mass concentrations and (b) UV-Vis absorbance spectra of dissolved organic carbon (DOC) in snow/ice samples from each month. The error bars indicate the standard deviation. Different letters indicate significant differences in mean values (one-way ANOVA). Mean values with the same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

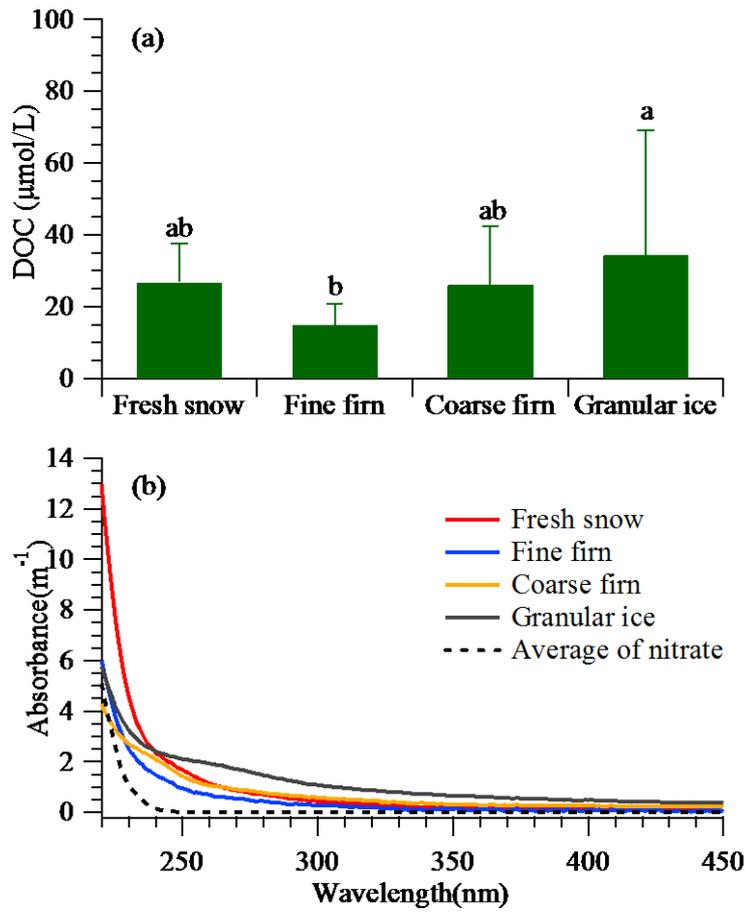


Figure 3. (a) Average mass concentrations and (b) UV-Vis absorbance spectra of dissolved organic carbon (DOC) in each category of snow/ice sample. The error bars indicate the standard deviation. Different letters indicate significant differences in mean values (one-way ANOVA). Mean values with the same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

Fig 7 I don't think that this figure adds anything to the manuscript. What are we supposed to get from it, that there's overlap?

This figure is used to display the unique components which was newly produced during melting in each snow/ice categories. Based on the composition of the unique components, we can know some information of their sources. We have revised the figure 7 which we add the percentage of CHO and CHON molecular class in unique component in each category sample (pie chart).

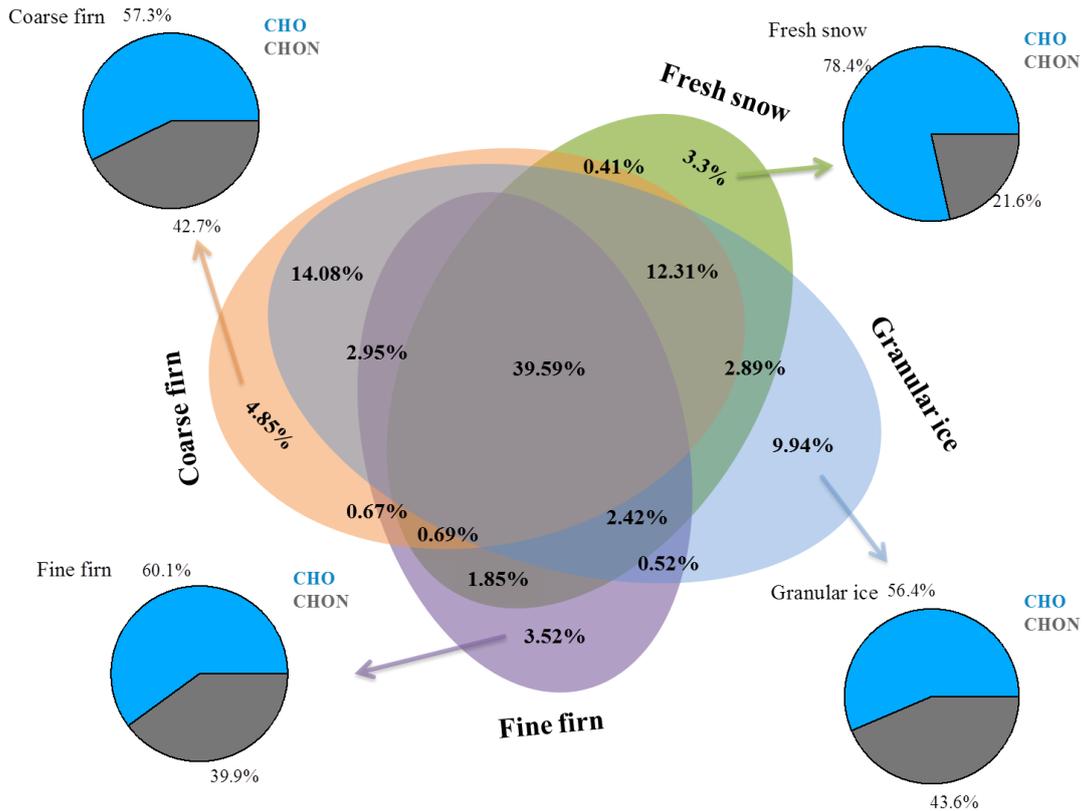
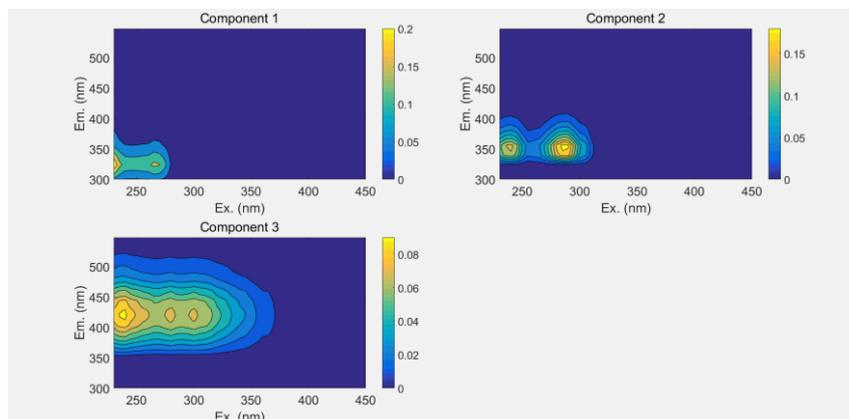


Figure 7. Four-way Venn diagram shows the overlap in molecular formulas between the fresh snow, fine firn, coarse firn and granular ice. The numbers within the Venn diagram are the percentage of overlapped component in four categories and unique component in each category. The relative contributions 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.

Generally- I think that C1 and C4 are probably the same. Have you run a 3 component PARAFAC model?

Yes, we have run the 3 component PARAFAC and shown the result figure as follows, we found the C1 and C2 in this figure corresponding to the C1 and C4 in Figure 4, this suggested C1 and C4 are different components in our samples, on the contrary, C2 in Figure 4 has been overlapped with other component in 3 component PARAFAC model.



Reference:

Coble, P. G., Del Castillo, C. E., and Avril, B.: Distribution and optical properties of CDOM in the Arabian Sea during the 1995 Southwest Monsoon, *Deep Sea Res., Part II*, 45, 2195–2223, 1998.

Dubnick, A., Barker, J., Sharp, M., Wadham, J., Lis, G., Telling, J., Fitzsimons, S., and Jackson, M.: Characterization of dissolved organic matter (DOM) from glacial environments using total fluorescence spectroscopy and parallel factor analysis, *Ann. Glaciol.*, 51, 111–122, 2010.

Fellman, J. B., Hood, E., and Spencer, R. G. M.: Fluorescence spectroscopy opens new windows into dissolved organic matter dynamics in freshwater ecosystems: a review, *Limnol. Oceanogr.*, 55, 2452–2462, 2010.

Fellman, J. B., Hood, E., Spencer, R. G. M., Stubbins, A., and Raymond, P. A.: Watershed Glacier Coverage Influences Dissolved Organic Matter Biogeochemistry in Coastal Watersheds of Southeast Alaska, *Ecosystems*, 17, 1014–1025, 2014.

Williams, M., Knauf, M., Caine, N., Liu, F., and Verplanck, P.: Geochemistry and source waters of rock glacier outflow, Colorado Front Range, *Permafrost and Periglacial Processes*, 17, 13–33, 2006.

Yao, T., Thompson, L., Yang, W., Yu, W., Yang, G., Guo, X., Yang, X., Duan, K., Zhao, H., and Xu, B.: Different glacier status with atmospheric circulations in Tibetan Plateau and surroundings, *Nat. Clim. Change*, 2, 663–667, 2012.