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 #3:

The authors have conducted an interesting study of the chemical evolution of snow as it undergoes metamorphism from snow-firn-ice. The overall goal was to determine the compositions of DOM in snow samples collected from the Dongkemadi glacier and evaluate the changes in DOM composition during transition of snow to firn and then ice. The tool of choice was the use of fluorescence spectrometer and ESI-FTICR-MS. I think the paper is an interesting read. That said, a lot more information, including significant additional qualification of the interpretations, is required to make the manuscript acceptable for publication. The methods sections should be expanded to include the process "blank" analyses for organic carbon. In several places, the authors do not provide adequate references to back their statements (see minor comments).

Thank you very much for your positive comments.

A serious shortcoming is that there is no critical evaluation and discussion of the blanks associated with the DOC measurements. It is well known that snow and ice samples for organic carbon measurements can easily be contaminated (see Preunkert et al., Environ. Sci. Technol. 45, 673-678, 2011). Contaminations can occur due to contact of the samples with sampling materials, during shipping, and storing. Furthermore, a strong source of contamination is the dissolution of organic species during the melting of the samples. The impact of possible contamination during storage, the handling of the samples in the laboratory and during the melting process should have been tested using blanks. The authors provide no information about blank experiments. Did the authors estimate the carbon contamination associated with the filter/ filtration device used for filtering the samples?

We prepared the procedure blanks during our experiment using Milli-Q water (18.2 M Ω). The average DOC concentration of blanks in this study was 5.9 ± 1.2 µmol L⁻¹ and the mass spectrum of FTICR-MS is shown below (Fig. R1). We are so sorry to exclude this analysis in previous manuscript. We add the DOC measurement for blanks in section 2.2 and 2.6 as follows.

"Procedural blanks were prepared using Milli-Q water in 50 mL FalconTM tubes at the beginning of filtration and were used to assess potential contamination that could occur during sampling preparation and filtration. The mean concentration of procedure blanks was $5.9 \pm 1.2 \mu mol L^{-1}$."

"The procedure blank was obtained following the procedure described above to determine if any potential contamination occurred during the sample preprocessing."



Figure R1. Positive ion mode ESI FT-ICR mass spectrum of procedure blank. The mass spectrum was collected by use of procedure blank in 1:1 MeOH: blank.

Ambiguity in biomolecular compound class assignments – I understand that the van Krevelen plot is a useful tool for the visualisation of complex FTICR-MS data. But classifications in this study are based only on the location of formulae in the van Krevelen diagram. It is important to remember that formulae that plot in a particular area of the van Krevelen plot are not strictly representative of all similar molecules, but rather approximate guidelines for identifying compounds of similar composition. Thus, caveats associated with the descriptions of DOM as proteins, lignin, tannin etc., based only on the H/C and O/C without accounting for the N/C or aromaticity index (AImod) should be appropriately explained in the MS. Somewhere in the initial discussion of the van Krevelen diagrams, the authors need to make it very clear that just because a formula falls into a particular category it is not definitely that type of structure, as you only have formula data, not structural data. The fact that FTICR-MS only provides elemental formulae and not structural classes needs to be emphasized so that readers know that when the authors refer to something as lignin, it could represent a molecule with the same formula. It is critical that readers, be made aware of these ambiguities when discussing FTICR-MS data.

Thank you very much for your suggestions. We have added the note for van Krevelen in update manuscripts in section 3.3 as follows.

"Note that the formula in a particular category only according to H:C and O:C ratio in the van Krevelen diagram could include different isomers."

We have calculated the modified aromaticity index (AI_{mod}) for each formula and added the values of AI_{mod} to identify the biomolecular class to which each molecular compound belonged in section

2.7 as follows.

"To identify the biomolecular class to which each molecular formula belonged, the van Krevelen diagram (Kim et al., 2003) and the modified aromaticity index (AImod) (Koch and Dittmar, 2006, 2016) were used. The van Krevelen diagram is constructed using the molar ratio of hydrogen to carbon (H/C ratio) as the ordinate and the molar oxygen-to-carbon ratio (O/C ratio) as the abscissa. Major biogeochemical classes of compounds (such as lipids compounds, aliphatic/proteins, carbohydrates, etc.) have their own characteristic H/C or O/C ratios. As a result, each class of compounds plots in a specific location on the diagram. It is well recognized that types of compounds can be identified from the location of points in the van Krevelen plot. AImod is a measure of the probable aromaticity for a given molecular formula assuming that half of the oxygen atoms are doubly bound and half are present as σ bonds was calculated as: AImod = (1 + C = 0.5O = S = 0.5[N + P + H])/(C = 0.5O = N = S = P). Formulas with AImod ≥ 0.5 and <0.67 are assigned as aromatics, while formulas with AImod \geq 0.67 are assigned as condensed aromatics (Koch and Dittmar, 2006). The following compound classes were defined based on previous studies (Grannas et al., 2006; Hockaday et al., 2009; Ide et al., 2017): (1) lipids ($0 \leq 1$ O/C < 0.3, 1.5 < H/C < 2.4), (2) aliphatic/proteins ($0.3 \le O/C \le 0.67, 1.5 < H/C < 2.4),$ (3) carbohydrates (0.67 < O/C < 1.2, 1.5 < H/C < 2.4), (4) unsaturated hydrocarbons (0 < O/C < 0.1, 1.5 < H/C < 2.4), (4) unsaturated hydrocarbons (0 < 0/C < 0.1, 1.5 < H/C < 0.4), (4) unsaturated hydrocarbons (0 < 0/C < 0.1), (4) unsaturated hydrocar $0.7 \leq$ H/C \leq 1.5), (5) lignins/carboxylic-rich alicyclic molecules (CRAM) (0.1 \leq O/C \leq $0.67, 0.7 \le H/C \le 1.5, AI < 0.67)$, (6) tannins (0.67 < O/C < 1.2, 0.5 $\le H/C \le 1.5, AI < 0.67)$ 0.67), and (7) condensed aromatics ($0 < O/C \le 0.67, 0.2 \le H/C < 0.7, AI \ge 0.67$)."

Kim, S., And, R. W. K., and Hatcher, P. G.: Graphical Method for Analysis of Ultrahigh-Resolution Broadband Mass Spectra of Natural Organic Matter, the Van Krevelen Diagram, Anal. Chem., 75, 5336–5344, 2003.

Koch, B., and Dittmar, T.: From mass to structure: an aromaticity index for high - resolution mass data of natural organic matter, Rapid Commun. Mass Spectrom., 20, 926 - 932, 2006.

Koch, B., and Dittmar, T.: From mass to structure: an aromaticity index for high - resolution mass data of natural organic matter, Rapid Commun. Mass Spectrom., 30, 250 – 250, 2016.

The authors state that one of the objectives of this study is to examine the compositions and sources of DOM in snow, yet sources are not adequately discussed. The authors identify through fluorescence measurements protein-like and humic like components and through FTICR-MS several lignin, tannin, condensed aromatic, protein formulae among others. What are the sources of these humic, lignin-and tannin-like compounds in snow? These need to be discussed.

We discussed the sources of DOM in section 4.2. As Referee #1 suggested, some contents in this part were over stated. We have reworded this part in the updated manuscript and added a briefly discussion in section 4.2 for the sources of humic, lignin-and tannin-like compounds in snow/ice samples in the update manuscripts.

"The low account of humic-like components (C3) (15%) derived primarily from the glacially

overridden or soils from the surrounding region (Dubnick et al., 2010)."

"A small fraction of terrestrial components, including tannins, lignins/CRAM molecules and unsaturated hydrocarbons were also observed, which derived from ambient vascular plants or soils from the surrounding region. It is interesting to observe that the lignins/CRAM-type compounds contributed 19% – 27% to the total identified formulae in all category samples. A relative high lignins/CRAM content was also reported for Antarctic Ice Sheet, which has a serious lack of plants in the environment similar with the DKMD glacier (Antony et al., 2014). Previous studies have revealed CRAM are ultimately derived from biomolecules with similarities to sterols and hopanoids (Hertkorn et al., 2006), thus suggesting CRAM can be present in the ecosystems rich in algal and/or microbial sources (Xie et al., 2009). The DOM in snow/ice samples also contained small amounts (0.4%–1%) of hydrogen-deficient condensed aromatic constituents (Table 3), i.e., black-carbon-like material, with low O/C ratios (≤ 0.2) and high AImod (≥ 0.67), which could originate from wildfires and charred soil residues (Schmidt et al., 1999), and usually not a pure substance but rather a continuum of condensed aromatic molecules (Jones and Chaloner, 1991)."

Antony, R., Grannas, A. M., Willoughby, A. S., Sleighter, R. L., Thamban, M., and Hatcher, P. G.: Origin and Sources of Dissolved Organic Matter in Snow on the East Antarctic Ice Sheet, Environ. Sci. Technol., 48, 6151–6159, 2014.

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.

Hertkorn, N., Benner, R., Frommberger, M., Schmitt-Kopplin, P., Witt, M., Kaiser, K., Kettrup, A., and Hedges, J. I.: Characterization of a major refractory component of marine dissolved organic matter, Geochim. Cosmochim. Acta, 70, 2990–3010, 2006.

Jones, T. P., and Chaloner, W. G.: Fossil charcoal, its recognition and palaeoatmospheric significance, Global & Planetary Change, 97, 39–50, 1991.

Schmidt, M. W. I., Skjemstad, J. O., Gehrt, E., and Kögel - Knabner, I.: Charred organic carbon in German chernozemic soils, Eur. J. Soil Sci., 50, 351 - 365, 1999.

Xie, J., Wang, N., Pu, J., and Chen, L.: Study of bacterial diversity recoved from glacier snow of the northern tibetan plateau, Journal of Glaciology and Geocryology, 31, 342–349, 2009.

DOM that shares the same regions of the van Krevelen diagram with other microbial studies can only suggest similar chemical character, and not microbial production (Line 381). And since microbial production was not directly tested with the current experimental design, this statement is inappropriate. Did the authors measure microbial abundances in these samples? If data for microbial numbers is available, these should be included. If not, are there other papers that have described microbial numbers/diversity in snow or ice from this region?

We did not analyze the microbial abundances in these samples. We have revised this sentence as follows, and to support our results, we have added the related reference which has given the fingerprint of DOM definitely produced by the microbial in superglacial snow samples.

"In addition, the distribution of DOM in Van Krevelen diagrams for our samples are similar with

those derived from in-situ microbial compounds with high H/C and low O/C ratios (Antony et al., 2017), which is characteristic of lipids and aliphatic/proteins-like material (Figure 6)."

Several samples were collected from each elevation. Were all of the replicate samples also analysed by FTICR-MS? If yes, then the authors should report some value of reproducibility. For example see Sleighter et al., Analytical Chemistry, 2012, 84, 9184-9191.

We did not do reproducibility analysis due to the limited sample volume for FTICR-MS measurement. For each category, we only had one sample. We will pay attention on this reproducibility analysis in the future. For clarify this issue, we mentioned this point in the updated manuscript in section 2.6 as follows.

"Note that we only had one sample for each category due to the limited sample volume for FT-ICR MS measurement."

Minor comments: Line 46 – one of the

We have reorganized the introduction part. This sentence has been deleted in the updated manuscript.

Line 51 – 'they were more efficient to be released'. It is not clear what you mean by that.

This means the mountain glacier released more DOC than the polar regions relative to their glacier volume. This sentence has been revised as follows:

"Liu et al. (2016) estimated the release rates of dissolved organic carbon (DOC) from the glaciers in Western China at 15.4 \pm 6.1 Gg yr⁻¹ which was more efficient than those observed in the Polar Region relative to their glacier storage."

Line 55 – How is a discussion on cryoconites relevant to this study? You either need to establish a link, or delete this section.

We have deleted this sentence.

Line 63 – lost how? to where?

We have revised this sentence 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 79 - 1.767 km². Is this correct?

Yes. This is a correct area.

Line 115 - What is the detection limit of the instrument?

We have added the detection limit of the instrument in the update manuscript in section 2.2 as follows.

"The detection limit for DOC was ~50 μ gC L⁻¹ defined as three times the standard deviation of low concentration samples."

Line 200 - edit 'methanoland 3' to 'methanol and 3'

We have corrected "methanoland 3" to "methanol and 3".

Line 215 - FI-ICR or FT-ICR?

We have corrected "FI-ICR" to "FT-ICR'.

Line 226 – what is DBE? Expand the abbreviation and provide an explanation as to what it signifies

We have added the full name of DBE in the update manuscripts in section 2.7 as follows.

"For the chemical formula $C_c H_h O_o N_n S_s$, the double bond equivalence (DBE) was calculated using the following equation: DBE = (2c+2-h+n)/2."

Line 229-230 - A description of van Krevelen analysis should be included here

Agree. We have included the description of van Krevelen analysis in the updated manuscript (section 4.1).

"The van Krevelen diagram is constructed using the molar ratio of hydrogen to carbon (H/C ratio) as the ordinate and the molar oxygen-to-carbon ratio (O/C ratio) as the abscissa. Major biogeochemical classes of compounds (such as lipids compounds, aliphatic/proteins, carbohydrates, etc.) have their own characteristic H/C or O/C ratios. As a result, each class of compounds plots in a specific location on the diagram. It is well recognized that types of compounds can be identified from the location of points in the van Krevelen plot."

Line 236- what is CRAM? Please provide the definition and expand the abbreviation.

We have provided the full name of CRAM in the updated manuscript.

"(5) lignins/carboxylic-rich alicyclic molecules (CRAM) $(0.1 \le O/C \le 0.67, 0.7 \le H/C \le 1.5)$."

Lines 234-238 – please back these classifications with the appropriate reference(s)

We have added the appropriate references here.

"Grannas, A. M., Hockaday, W. C., Hatcher, P. G., Thompson, L. G., and Ellen, M. T.: New revelations on the nature of organic matter in ice cores, J. Geophys. Res.: Atmos., 111, 2006. Hockaday, W. C., Purcell, J. M., Marshall, A. G., Baldock, J. A., and Hatcher, P. G.: Electrospray and photoionization mass spectrometry for the characterization of organic matter in natural waters: a qualitative assessment, Limnol. Oceanogr.: Methods, 7, 81–95, 2009. Ide, J., Ohashi, M., Takahashi, K., Sugiyama, Y., Piirainen, S., Kortelainen, P., Fujitake, N., Yamase, K., Ohte, N., and Moritani, M.: Spatial variations in the molecular diversity of dissolved organic matter in water moving through a boreal forest in eastern Finland, Sci. Rep., 7, 2017."

Lines 242-245 – How do the values obtained in your study compare with that of other studies that report DOC values for snow from the TP or other mountain glaciers?

We have compared the DOC concentration of this study with the Yulong glacier located in southeastern TP.

"The DOC concentrations of these samples ranged from $1.1 - 189 \ \mu mol \ L^{-1}$, exhibiting a similar DOC concentration to snow/ice samples ($32.5 - 169 \ \mu mol \ L^{-1}$) from the Yulong mountain glacier located in southeastern TP (Niu et al., 2016)."

Line 251, 311 - Please do not use the word significant if no statistical tests were performed.

Agree. We have replaced significant with other words.

"The average absorbance spectra observed during each month exhibited obvious absorbance between 220–450 nm (Figure 2b)."

We have included *t* test here.

"Therefore, the ratio between CHON and CHO increased significantly, from 0.287 to 0.393 (p<0.05), from fine firm to granular ice."

Line 264 – what are these obvious differences? State them.

We have stated the differences as follows:

"The total absorption coefficient of each category showed obvious differences in its characteristics between 240–450 nm, between which the granular ice showed most obvious absorption peak, then

followed by coarse firn, fresh snow, and fine firn showed the lowest absorption peak."

Line 269-272 – this needs to be supported with a reference. Consider changing 'aromatic components' to 'aromatic constituents'

Agree. We have changed 'aromatic components' to 'aromatic constituents', and included the next reference to support this sentence.

Weishaar, J. L., Aiken, G. R., Bergamaschi, B. A., Fram, M. S., Fujii, R., and Mopper, K.: Evaluation of specific ultraviolet absorbance as an indicator of the chemical composition and reactivity of dissolved organic carbon, Environ. Sci. Technol., 37, 4702, 2003.

Line 281 - 'as is described in the Supporting Information section' is redundant

Agree. We have deleted this description.

Line 293 – what do these percentages indicate? Do they refer to the contribution of formulae of a particular biomolecular compound class to the total identified formulae? Please specify.

Yes. We restated this sentence as follows:

"The assigned molecular compounds are mainly concentrated in three biomolecular classes (Figure 6, Table 3), i.e., lipids (29.2% - 42%), aliphatic/proteins (33% - 40.3%), and lignins/CRAM (19% - 27%), the percentages refer to the contribution of each particular biomolecular compound class to the total identified formulae."

Line 295 - 298 - why no mention of carbohydrates?

We have included the description for changes in carbohydrates in the update manuscripts.

"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."

Line 302-303 – Please be advised that the number of molecular formulae can be greatly dependent on sampling method, SPE procedure, ionization mechanism, FTICR-MS parameters, data acquisitions etc. Therefore, when tracking changes in the number of molecular formulae amongst samples, appropriate disclaimers should be provided.

Thank you for your kindly advise. We have provided the disclaimers in the updated manuscripts in section 2.6.

"It is important to note that the mass spectra of different characteristic molecules may be affected by different solid-phase extraction, ionization mechanism, parameters and data acquisitions." line 305 – not sure what this line means

We have revised 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 307 - did you detect any S containing formulae? If yes, why were these not discussed?

We have not detected any S containing formulae in our samples.

Line 327 – what do you mean by lost DOC?

We have revised this sentence as follows:

"The monthly average DOC concentrations during summer (June, July, and August) were all at the low range of their values, suggesting that the snowmelt could release DOC to glacial runoff through leaching effects."

Lines 331 -332 – consider rephrasing this sentence. A decrease in DOC values do not indicate which compounds/compound categories were reduced in concentration.

Accept. We have replaced 'compounds' by 'DOC' in this sentence.

"This indicates that approximately 44% of the DOC was lost during the first stage of snowmelt."

Line 337 - edit 'Water-' to 'water-'

We have revised.

Line 342 – what is meant by enrichment of DOM? What contributed to this enrichment?

We have discussion the enrichment as follows.

"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)."

Line 350-358- It is not clear what point the authors are trying to make by comparing with cryoconite samples.

We have deleted this sentence.

Line 369 – why is a comparison made to lake Taihu? How is it relevant to your study?

We have deleted this sentence.

Line 379 – what is meant by more intense microbial signals? I do not understand why a comparison with cryoconite samples is important?

We have deleted this sentence.

Line 400 – do you mean 'which is the dominant mass fraction'?

We have revised this part as follows.

"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 (Despr s 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 401 – How was the chemical composition different? State the differences.

We have included the comparison of chemical composition between fresh snow and granular ice samples as follows:

"Comparing with fresh snow, granular ice samples contained more aliphatic/proteins and lignins/CRAM compounds, less lipids, carbohydrates, unsaturated hydrocarbons and condensed aromatics components."

Line 402-404 – Cite a reference to back this claim.

We have revised this sentence as follows:

"The fact that the absorbance observed at 240 - 450 nm was higher in granular ice than it was in fresh snow reflects the more chromophores DOM in granular ice (Beine et al., 2011; Liu et al., 2009; Malik and Joens, 2000)."

Line 404-406 – it is not clear how you conclude that there is a variation in DOM molecular weight based on the variation in S240-400 values. Please explain and provide appropriate references. Also, please explain how these results suggest the presence of microbially transformed and newly produced DOM.

We have recalculated the spectral slope coefficient between 275-295 nm and 350-400 nm and gave the results and discussion of slope ratio (S_R) (the ratio of $S_{275-295}$ to $S_{350-400}$) of each category in the 3.1 section and 4.2 section respectively as follows.

"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 suggested molecular weight of the DOM changed during melting stage (Helms et al., 2008)."

"The S_R values of DOM in each category exceed 1.0, and according to Helms et al. (2008), S_R values higher than 1.0 are typical for endogenous sources. Meanwhile, previous work has suggested an inverse relationship of DOM molecular weight with spectral slope coefficient (Miller et al., 2009), therefore, molecular weight of the DOM increased during the snowmelt process."

Helms, J. R., Stubbins, A., Ritchie, J. D., Minor, E. C., Kieber, D. J., and Mopper, K.: Absorption spectral slopes and slope ratios as indicators of molecular weight, source, and photobleaching of chromophoric dissolved organic matter, Limnol. Oceanogr., 53, 955–969, 2008.

Line 407 – what other chemical processes?

This is a wrong description. We have deleted this sentence.

Line 408 -411 – Perhaps the authors intend to say 'snowpack microbes exhibit diverse enzyme activities and contribute to the degradation of DOM in these environments'? But in the previous line, the authors talk about newly produced DOM. This claim needs to be supported with appropriate reference(s) (for example see Smith et al., 2017, Nat. Geosci; Antony et al., 2017, Environ. Sci. Technol.)

Agree. We have included these two references in the update manuscript.

Line 414 – please include appropriate references and explain how a change in DBE values suggests the occurrence of oxidation process.

We have revised this sentence as follows and added a reference here.

"The relatively higher abundances of the molecules gradually changing from high DBE- to zero DBE-containing from fresh snow to granular ice, accompanied by increased oxygen content (Figure 8), previous study has proved approximately two oxygen atoms contributed to one unity

change in DBE (Bae et al., 2011), this suggests there was oxidation process during the snowmelt."

Line 418 - edit 'ranged in' to 'ranged from'

Revised as the Referee suggested.

Line 427 - rephrase 'composition of DOM' to 'composition of identified DOM formulae'

Revised as the Referee suggested.

Line 431 – edit 'relative contributions of proteins and lignins' to 'relative contributions of proteins and lignins to the total identified formulas'

Revised as the Referee suggested.

Line 436-438 – I understand that the authors want to provide a broader perspective to their study, but written as is, this is too general a statement. Consider revising.

Revised as the Referee suggested.

Table 1 – Description of component number C1. Do you mean 'tryptophan-like or tyrosine- like'? You need to provide appropriate references to support these classifications.

Agree. We have included the reference in the Table.

Line 619 - DOM

We have revised in the update manuscripts.

Table 2 -You need to indicate in the table, the number of samples analysed for each corresponding SD value.

We have added the number of each category samples in the Table 2.

Table 3 - The authors should not assume that the readers are aware that m/z corresponds to mean mass. Please include m/z in parenthesis next to mean mass. You need to write clearly in the table heading that the numbers in the table correspond to the total number of molecular formulae identified for each sample category.

Agree. We have revised the table heading in the update manuscripts.

"Table 4. Total number and number and percentage of elemental formulas assigned to the CHO, CHON, and CHN groups in fresh snow, fine firn, coarse firn and granular ice ^a ^aNote the numbers in the table correspond to the total number of molecular formulae

identified for each elemental formulas group."

Table 4 – same comment as above. Please indicate what the numbers reported in the table correspond to. What does total mean?

Agree. We have revised the table heading same as above in the update manuscripts.

Figure 2 – please provide error bar/statistical calculation details in the caption

Agree. We have provided error bar details in the caption.

"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 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."

Figure 3 – same comment as above

Agree. We have provided error bar details in the caption.

"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 errors. 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."

Figure 6 – why is the relative intensity of each peak important to show in the figure? Especially since you do not discuss relative intensities of molecular formulae in the main text.

Agree, we have revised this figure as follows:



Figure 6. Van Krevelen diagrams for the mass spectra of samples of (a) fresh snow, (b) fine firn, (c) coarse firn, and (d) granular ice. The regions in the plots indicate the different classes of biomolecular compounds.

Figure 7 - The overlap of molecular formulae in the venn diagram will be better represented as percentages

Agree, the overlap of molecular formulae in the venn diagram has been represented as percentages and we also added the relative contributions of CHO and CHON molecular classes in each unique components.



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 numbers of m/z molecular formulas and 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 (a) Fresh snow, (b) Fine firn, (c) Coarse firn and (d) Granular ice.

Figure 8 – This figure seems unnecessary, given that you only mention the results pertaining to this figure in the main text and do not have a detailed discussion of the same.

We have included the results of this figure in section 3.4 and add the discussion for this figure in section 4.2 as follows.

"The changes in DBE-containing and oxygen-containing in molecules in each category samples were shown in Figure 8, from fresh snow to granular ice, the relatively higher abundances of the molecules transferred from high DBE– containing to zero –DBE containing meanwhile the oxygen content was increasing, this reveals the changes in molecular structure during the snowmelt."

"The relatively higher abundances of the molecules gradually changing from high DBE- to zero DBE-containing molecules from fresh snow to granular ice, accompanied by increased oxygen content (Figure 8), previous study has shown that approximately two oxygen atoms contributed to a unity change in DBE (Bae et al., 2011), which suggests the occurrence of oxidation process

during the snowmelt."

Supplementary figure S2. What is Kendrick mass defect analysis? Or Kendrick mass? No description of this has been provided in the MS or in the supporting information. Neither is there a reference to Figure S2 in the main text.

We have deleted this figure.

Reference: