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## The Cryosphere

### Revised manuscript: tc-2014-177

Dear Editor,

On behalf of my co-authors, I submit the revised version of the manuscript with the title "*Dissolved organic carbon (DOC) in Arctic ground ice*".

Attached you will find:

- response to reviewer #1
- response to reviewer #2
- marked-up manuscript version
- supplement to the manuscript

Thank you very much for your time and efforts in this process and we look forward to seeing this manuscript accepted soon. Please, do not hesitate to contact me if you need further formation.

Best regards,



Michael Fritz

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15 March 2015

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## ***Interactive comment on “Dissolved organic carbon (DOC) in Arctic ground ice” by M. Fritz et al.***

**M. Fritz et al.**

michael.fritz@awi.de

Received and published: 12 March 2015

### **Reply to Anonymous Referee No.1**

We are grateful for the review and acknowledge your comments and suggestions. You will find all replies or changes that have been made below. Reviewer comments are cited in italic font.

Best regards,  
Michael Fritz

(on behalf of the co-authors)

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**Title:** *I recommend a little more info. How about: “The chemical composition and fate of organic carbon (DOC) in Arctic ground ice”*

We would like to keep the original title as it is concise and meaningful to us. We do not present the chemical composition of DOC.

**Abstract:**

**Line 4:** *The “their” is confusing. I think you mean “Permafrost” but the previous sentence ends with info on nutrients so it seems like that is what “their” refers to.*

Replaced by “permafrost”.

**Line 9:** *“using biogeochemical data”*

Changed accordingly.

**Line 17:** *just a curiosity: why refer to snow melt as “pristine” and not just as “snowmelt”?*

In the course of the discussion we come back to this point, because ion-poor (i.e. pristine) snowmelt is able to leach inorganic and organic matter.

**Line 20:** *Perhaps start the “In the yedoma” with “We found that in the Yedoma...”*

Changed accordingly.

**Lines 22-25:** *This is the first time particulate OC is mentioned and it is a main focus of this last sentence. I recommend introducing some of the POC results, data, etc, to set this up. Maybe one sentence to do that?*

The abstract might not be the right chapter to extend the explanation on POC stock/relevance, because our manuscript does not rely on POC data. More information on the size and relevance of permafrost OC pools, also in comparison with DOC data in this study, is given in the following parts of the manuscript: p.79 L. 23ff, p.80 L. 7ff., p.89 L. 5ff., p91 L. 23ff.

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**Page 80: Line 7:** *“several studies have shed light”*

Changed accordingly.

**Page 82: Line 10:** *“It is” is vague. The previous sentence covers a lot of topics so you have to be more specific.*

changed into: “DOC in intrasedimental ice is, however, . . .”

**Page 83:** *The Section title has “component” but line 25 has “components”. Be consistent.*

Component!! Changed accordingly.

**Page 85:**

**Line 10:** *do the “mean concentrations” refer to DIC? If so then state that for clarity.*

DIC added.

**Line 12-13:** *the sentence that starts with “It is obvious” is unnecessary. Move the “Basal glacier ice” sentence to the end of the previous paragraph.*

Changed accordingly.

**Page 86: Line 20:** *“those with more of a continental”*

Changed accordingly.

**Page 89: Line 26:** *“sources that have been”*

Changed accordingly.

**Page 90:**

**Line 4:** *“into frost cracks”?*

There is no need to change anything because the text already reads this way.

**Line 7:** *Unless any of the “leachable components” are close to saturation in other*

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*precipitation/ water courses (which is highly unlikely) the “initial purity” of the snow is irrelevant. I recommend removing this sentence.*

Dou et al. (2008) have shown that the ionic and chemical composition of the solvent plays a role in the ability and strength to leach DOC from permafrost. Therefore, we would like to keep the sentence.

**Line 8-10:** *The “Snow melt feeding” sentence: See previous comment and:*

*1) There are a ton of papers on the age and lability of the DOC in rivers at melt and some suggest this surface flow (ie at snow melt) has bioavailable C. So it is not necessary that the bioavailable C has from lower down in the soil column.*

*2) As the spring snow melt waters trickle downward toward the ice wedges they interact with basal soil material (frozen or not) and this could leaches out carbon.*

*3) Also- since wedges take thousands of years to form and the location of their upper surface likely changes with time there are plenty of spatial and temporal ways that deeper soil pore waters can get integrated into the wedge ice.*

*I recommend some of these ideas be introduced or discussed. The fact that the snow has little ionic strength is not a potential reason for this. And I agree the waters at snow melt start at the surface but they do trickle downward and are not likely frozen until the following winter so there is ample time for subsequent waters, interacting with a deeper active layer, could incorporate deeper carbon.*

Above the discussed paragraph we added the following information: “As the spring snow melt waters trickle downward toward the ice wedges they interact with the basal soil material leaching out carbon. Also, since wedges may take thousands of years to form and the location of their upper surface changes with time, there are plenty of spatial and temporal ways that deeper soil pore waters can get integrated into the wedge ice.”

**Pages 89-90:** *Somewhere in here or elsewhere (?) it is worth noting the potential mineral weathering signature to which their samples provide context. For example- could*

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*the major cations and bicarbonate could be used to decipher silicate versus carbonate weathering. Since carbonate weathering occurs more rapidly with silicate weathering (particularly where “fresh” mineral surfaces are available) their study might be able to use this to identify where in the active layer some compounds are sourced? They could explore a few quick ternary diagrams of Ca, Mg, and Na+K and of SO<sub>4</sub>, HCO<sub>3</sub>, and Cl. I suspect there will be some unique trends and if/where the signatures are more carbonate based they could be able to decipher the location in the soil column? IE the “fresh” mineral surfaces are likely toward the base of the active layer (exposed to weathering the least amount of time on an annual basis). Caveat: the marine localities may have a swamped HCO<sub>3</sub> and Cl signal so it is possible that this will not work. But I recommend they make the plots to explore it. This is in no way a requirement by me for acceptance, etc. Just that they have a unique data set and I am trying to see if there is more info that can be teased from it.*

These are very good points. Unfortunately, we are not aware of any indices, based on the major ion composition in ground ice, to discuss the strength of silicate versus carbonate weathering in permafrost. A detailed study of the solid and liquid mineral composition along depth profiles would be necessary. With such an approach one could determine which layers are prone to leaching and at what depth/age of the deposits does the leaching/weathering happen. For now, we have only dealt with ground ice but not with the whole permafrost systems in a holistic approach.

**Page 92: Line 20:** “while deposition occurred”

Changed into “during deposition” as reviewer 2 recommended.

**Page 93: Line 20:** *I do not like use of the word “overproportionally” I am sorry I cannot provide a better word to use but it has a lot of chemical and physical connotations and I am not sure it is clear enough. Perhaps keep it but then provide what is being “overproportionally” loaded?*

We deleted the word.

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**Table 2 and discussion in the text:** *Are there any data from non-thermokarst ponds? Or perhaps find it in the literature? It would be good for a comparison in this Table and for the study because thermokarst ponds likely have an outsized amount of mineral weathering (ie ions), and carbon (ie leaching from exposed blocks of soil and degraded permafrost). As such, comparing to non-thermokarst ponds could help identify whether there are significant differences?*

The purpose of showing the data on modern surface water was to get an idea about the magnitudes of DOC concentrations we are dealing with. Now, the reader knows that they are comparable. Further reading is recommended to Walter Anthony et al. (2014) where they show more data on DOC in Yedoma and non-Yedoma bottom lake water in eastern Siberia near Cherskii. We do not want to speculate here about the processes why lakes in different geological and catchment settings have different hydrochemical characteristics. On page 91 we also discuss that runoff into lakes and rivers might be already degraded by microbial communities and photochemical reactions in contrast to ground ice. As the focus of this paper is on ground ice we try to keep the presented information as close as possible to our manuscript goals.

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Interactive comment on The Cryosphere Discuss., 9, 77, 2015.

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## ***Interactive comment on “Dissolved organic carbon (DOC) in Arctic ground ice” by M. Fritz et al.***

**M. Fritz et al.**

michael.fritz@awi.de

Received and published: 12 March 2015

### **Reply to Anonymous Referee No.2**

We are grateful for the review and acknowledge your comments and suggestions. You will find all replies or changes that have been made below. Reviewer comments are cited in italic font.

Best regards,  
Michael Fritz

(on behalf of the co-authors)

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**Main comments: 1.** *The first of my main comment regards a combination of terminology and the overlooking of another pool of carbon in ground ice: particulate OC. As the authors describe, the Pleistocene ice wedges are yellowish-brown to grey in colour, clearly visible on the photos in Figure 2. My guess is that the ice also contains particulate matter and carbon, which is currently not assessed and neither mentioned, as the authors have not provided any information on the organic carbon that remained on the filters. I would find it very valuable if this information is included, and if not, at least that this pool is described as a component currently not addressed. Linked to this, the authors use POC to describe the OC pool in permafrost soils. In de "aquatic community", POC is often used to describe the particulate OC fraction in water. The current use of POC in the manuscript is confusing and in most permafrost literature not used like this. I suggest to use soil OC, or just OC, or soil OC (SOC) or something like this.*

We used 0.7  $\mu\text{m}$  GFF filters attached to a syringe. These filters are sealed and cannot be analyzed afterwards. We acknowledge that the particulate fraction is important but it is not the objective of the paper.

The differentiation between dissolved organic carbon (DOC) and particulate organic carbon (POC) is simply a matter of particle size. DOC is defined as the organic matter that is able to pass through a filter of 0.7-0.22  $\mu\text{m}$  pore size. Conversely, particulate organic carbon (POC) is that carbon that is too large and is filtered out of a sample. Unfortunately, there is no global agreement on DOC and POC size differentiation which would guarantee direct comparisons of data from different studies. Mostly, the two pore sizes are used (0.7 and 0.45  $\mu\text{m}$ ). Once more, permafrost seems to a special and understudied case. The terms organic carbon (OC), total organic carbon (TOC) or soil organic carbon (SOC) always pool the dissolved and undissolved carbon fractions. However, studies inside and outside permafrost research have shown that DOC plays a special role in the carbon cycle.

**2.** *My second point concerns the availability of data. Can the data for DOC, water*

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*isotopes, ICP-OES etc. be presented in a supplementary table? Currently no individual sample info is available, and, for example, d18O and dD values are not included either.*

Changed accordingly. We added the table and referenced the supplementary table in the text and figure captions. See **Supplement Table S1**.

**Further comments:**

**p.78, L4:** *It's not clear what "Their" refers to.*

Replaced by "permafrost"

**p. 78, L19:** *Can something be "rapidly stored"? I suggest to rephrase into "rapidly frozen and stored".*

Changed accordingly.

**p. 78, L22:** *"4172 km3" is a number with too many significant numbers given the estimates this has been calculated from. I suggest to replace with "4170" or even better "4200" (please also replace this at a few other occasions in the text).*

Changed accordingly.

**p. 78, L22:** *See first, main comment: replace particulate OC with something else.*

See comment above.

**p. 79, L4:** *"degradation forms as thermokarst", you (also) mean alas deposits?*

Yes, but we do not want a large annotation of landforms and sediment types. The idea is to mention that not only Yedoma is ice-rich but also degraded Yedoma deposits.

**p. 79, L10:** *100 % volume? Isn't ice just ice? This is a bit confusing. Also, this paragraph lists many % numbers, some in weight, some in volume, can these be presented slightly more consistent?*

This relates to your first comment. The ice content of ice wedges is a little less than

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100 %. From an unpublished master's thesis we know that volumetric ice content of ice wedges from Herschel Island in the western Canadian Arctic is >98 %; mostly above 99 %.

Unfortunately, we cannot present the data on ice contents more consistently. In the past many studies have measured ice contents only gravimetrically. For example, leaving out the Schirmer et al. (2011c) review paper with the gravimetric ice content presented here, we would leave out the best data set for east Siberia.

**p. 79, L13:** *I suggest to change "other sediments in Yedoma" with "other ice in Yedoma sediments", because this sentence is about the total ice content, right?*

changed into: "other ice types in Yedoma deposits. . ."

**p. 79, L17-18:** *I think you can remove the definition of massive ice, it just adds to the confusion. p. 80, L24: "particulate OC", see the main comment above.*

We would like to keep the definition, because not everybody is familiar with massive and non-massive ice classification. For now, we mostly deal with massive ice and have not touched non-massive intrasedimental ice.

**p. 80, L2:** *"DOC from permafrost is chemically labile", I think it would be more correct to write "DOC from yedoma sediment and yedoma ice wedges is chemically labile".*

We added the reference of Dou et al. (2008) who showed large quantities of low-molecular-weight DOC in surficial permafrost horizons in northern Alaska, which corresponds to labile DOC. With that we would keep the original statement because labile DOC is not restricted to Yedoma.

**p. 80, L7:** *"that" should be removed.*

Changed accordingly.

**p. 80, L25-26-27:** *Yes, this study measures DOC at the source, but only in ice wedges, not in the (total) permafrost. Maybe specify?*

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Changed into “(i.e. ground ice in permafrost). . .”

**p. 81, L25:** *Only here you briefly define Yedoma. I think this should come a bit earlier in the manuscript.*

We added information on Yedoma origin in the introduction.

**p. 81, L16:** *and in many other places in the manuscript: please have a thorough look at your use of hyphens. In this sentence you correctly write "ground-ice conditions" with a hyphen between "ground" and "ice" because they together are an adjective to "conditions". At some other place (e.g. p.79, L5, 8, 14, 15, 25 and 28) you do not do this.*

Changed accordingly.

**p. 82, L2:** *You use both "late glacial" but also "late Pleistocene" throughout the text. If they mean the same thing, I suggest to just stick to one of these.*

The “late Pleistocene” (MIS 2-5) spans a longer time than the “late glacial” (Bölling to Younger Dryas). In some occasions we have the possibility to narrow the age of the studied ice bodies.

**p. 82, L14-15:** *You only included three surface water samples from thermokarst lakes. Compared to the rest of the pretty extensive dataset, I find this number a bit poor. There must be more data available in the literature.*

The purpose of showing the data on modern surface water was to get an idea about the magnitudes of DOC concentrations we are dealing with. Now, we can see that they are comparable. Further reading is recommended to Walter Anthony et al. (2014) where they show more data on DOC concentrations in bottom water from thermokarst lakes in Yedoma and non-Yedoma landscapes in eastern Siberia near Cherskii. Unfortunately, the hydrochemical data sets were not comparable so that external data could not be included into the statistical analyses.

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**p. 82, L18:** *I suggest to move "(purified water)" to directly behind "pre-cleaned".*

Changed accordingly.

**p. 83, L10:** *What other standards besides VSMOW did you use for your d18O and dD analysis? A standard that is sufficiently depleted such as SLAP should be included to be able to calibrate the ice wedge stable isotope composition that could reach values of e.g. -260 for dD.*

For all stable isotope measurements, a three-point calibration is used with an internal standard set selected from the list below related to the expected values of the samples. All standards have been calibrated against V-SMOW and SLAP. Since the isotope laboratory in Potsdam works since more than 15 years with depleted ground ice samples (also Antarctic ice) it is especially well-equipped for analyses with depleted isotope signature. Standards are generally selected from the pool of standards below covering the complete range of natural water isotope composition. This pool is routinely renewed and several new standards were calibrated to replace the older ones.

Standard	Target value sample vs. SMOW [‰]	Tolerance [ $\pm$ ]
NGT	-34.40	0.1
NWH	-48.25	0.1
HDW2	-12.70	0.1
dc1	-54.05	0.1
KARA	-0.10	0.1
Sez	-27.00	0.1

**p. 85, L13-14-15:** *This sentence is superfluous I think because it overlaps mostly with L3-4. Maybe expand L3-4 instead?*

Also according to the comment of reviewer 1 we removed one of the two sentences and moved the last sentence to end of the previous paragraph.

**p. 86, L22:** *Maybe write "climate conditions of formation"?*

Changed accordingly.

**p. 86, L22-25:** *This is interesting I think. Can you elaborate a bit on the reasons why you see this pattern?*

More interpretation and discussion of these points can be found in section 5.2 (page 90). In this chapter we added the following sentences that discuss the reviewer point towards stable water isotope characteristic: "A similar differentiation of ice wedges (and all ground ice types) is done along the second PCA axis (Fig. 5). Differences in stable water isotopes indicate the predominant climate variations between the late Pleistocene and the Holocene which are also reflected in the landscape (i.e. distance to sea; maritime vs. continental)."

**p. 87, L4-5:** *Is there a simple way to explain what these percentages mean?*

The percentage value here is based on the cation spectrum only. This means that all measured cations sum up to 100 %. This is statistically more robust than using individual sample concentrations which can differ in magnitudes. We added this information to the text.

**p. 87, L16:** *I think you should here instead of the McGuire paper refer to Holmes et al. 2012, which gives an updated estimate of pan-arctic DOC of 34 Tg/year (Holmes, R. M. et al. Seasonal and Annual Fluxes of Nutrients and Organic Matter from Large Rivers to the Arctic Ocean and Surrounding Seas. Estuaries and Coasts 35, 369-382, doi:10.1007/s12237-011-9386-6 (2012))*

We added the number of Holmes et al. (2012) and the reference.

**p. 88, L13:** *You've used WIV before, so I suggest to replace "wedge ice volumes" here with "WIV" to avoid confusion with volume of ice in square kilometers.*

Changed accordingly.

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**p. 88, L27-28:** *I suppose it makes sense that non-massive ice in soils is more DOC-rich, but it would not harm giving a brief explanation here why you think this is the case.*

We added: “Higher DOC concentrations in intrasedimental ice than in massive ice are certainly due to the long-term contact of soil moisture with soil organic matter prior to freezing.”

**p. 89, L5-9:** *What is this calculation based upon? I suggest to clarify this or leave it out. Also, I do not understand the last sentence here.*

This is a back-of-the-envelope calculation which is based on an overall average for sedimentary TOC values from the literature and our overall DOC average on massive ground ice. We slightly changed the used values and added some references of TOC values in permafrost deposits.

We added some information towards a better understanding of the last sentence. “. . .because TOC comprises both POC and DOC.” The message is that we need to differentiate permafrost TOC into POC and DOC because they react differently in time and space (e.g. transport, deposition, residence time, degradability, bioavailability).

**p.89, L12:** *replace "on" with "in"*

Changed accordingly.

**p. 89, L15:** *the term "mineralization" is a bit confusing as this word is also used as "degradation" sometimes. Maybe use "ion content" or "conductivity" or so instead?*

Changed accordingly.

**p. 89, L26:** *Only here you explain how ice wedges are formed. Would it be more appropriate to explain this earlier in the text? You only mention they are formed syn-genetically but don't say anything more.*

We added the following information to the introduction:

Ice wedges are one of the most common types of ground ice in permafrost. They form

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when thermal contraction cracks open in winter, which are periodically filled with snow melt water in spring that quickly (re)freezes at negative ground temperatures to form ice veins and finally vertically foliated ice wedges.

**p. 90, L21-23:** *Is the sentence "Marine ion .... into coastal ones" really needed? I feel it mostly holds information that has already been stated elsewhere in this paragraph.*

This is the only occasion in the manuscript where we present arguments why present-day coastal sites can host ice wedges with a completely different hydrochemical signature. A difference in age is accompanied by a difference in distance from the coast, which seems to be relevant for ion transport mechanisms.

**p. 91, L19-20 and before:** *I agree with the last sentence of this paragraph but I do not follow how this statement follows from the above sentences. I find it a bit confusing, and the point you are trying to make unclear. First of all, you say both ice wedge DOC and DOC in runoff of lakes and rivers are both bioavailable (right?) but in between these two things you use "In contrast" (L17). Also, the sentence "One destination of the fresh, young and therefore most bioavailable ...". You mean the destination of vegetation debris before ending up in the ice wedge DOC? And (L13): I think concentrations of DOC are also lower because all the vegetation debris and surface soils have already been actively flushed out by the spring flood when discharge and therefore, then, also DOC is high. All in all, I propose to reconsider the arguments that you use to arrive at the final sentence of this paragraph "The highlights ... of bioavailable DOC".*

The reviewer has a point here and we agree that DOC concentrations are also lower because all the vegetation debris and surface soils have already been actively flushed out by the spring flood. This is exactly what we write. To improve the structure, we changed the order of arguments so that the “in contrast”-sentence is not a contrast anymore but an addition “In addition, dissolved organic matter compounds in runoff into lakes and rivers can become rapidly degraded by microbial communities and photochemical reactions (Striegl et al., 2005; Olefeldt and Roulet, 2012; Cory et al., 2014).“

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Now the structure of arguments is the following:

1. Guo et al. (2007) have shown in experiments and from natural river samples that intensive leaching of DOC from young and fresh plant litter and upper soil horizons occurs during the snowmelt period (i.e. spring).
2. Later in the season (i.e. summer/autumn), they found that DOC yields decreased in rivers draining permafrost areas. This indicates that deepening of the active layer and leaching of deeper seasonally frozen soil horizons released lower DOC concentrations due to the refractory and insoluble character of the remaining organic matter compounds. In addition, dissolved organic matter compounds in runoff into lakes and rivers can become rapidly degraded by microbial communities and photochemical reactions.
3. Our conclusion based on sentences 1 and 2 is that the fresh, young and therefore most bioavailable DOC components will become incorporated in ice wedges which are basically fed by spring meltwater with a short transport pathway and small residence time.

We end this paragraph with the sentence: “This highlights the importance of ground ice and especially ice wedges as a vital source of bioavailable DOC.”

**p. 91, L22:** *replace "particulate fraction" by something else.*

This sentence is based on the dichotomy between dissolved and particulate. We replaced “particulate fraction” by “POC”.

**p. 92, L21:** *"while" sounds strange, maybe use "with" or "during"?*

Changed accordingly.

**p. 92, L22:** *"On the other hand" sounds incorrect here as this sentence adds up to the argument made by the previous sentence. Suggest to replace with "Also".*

Changed accordingly.

**p. 92, L24:** *What are "forbs"?*

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forbs = non-graminoid herbaceous vascular plants (see Willerslev et al., 2014 Nature)

**p. 94, L1:** Please replace "mineralization" with something else.

We replaced "sparsely mineralized ice wedge meltwater" by "ice wedge meltwater with a low ion content".

**p. 94, bulletpoints:** *Just an idea, is it possible to arrange these conclusions parallel to the bulletpoints you list in the introduction? A parallel construction of objectives conclusions would benefit the reader.*

Changed accordingly.

**p.94, L16:** round up "4172 km<sup>3</sup>" to "4170" or "4200".

Changed accordingly. 4200 throughout.

**p. 95, L1-2:** *Maybe rephrase into " we propose that future studies shall strive to".*

Changed accordingly.

**p. 95, bulletpoints:** *I think some of these points do not follow from the analyses/ conclusions in this paper, and/or I find the points that are made a bit unclear. For example, why should DOC from coastal erosion be better quantified?*

We re-arranged some of the conclusions and clarified some points in the outlook section to better match the open questions.

**p. 95, L8:** *"what remains POC and what is going to become DOC", maybe just write "what fraction of soil OC will be leached as DOC".*

Changed accordingly.

**Figure 3:** *capitalize holocene.*

Changed accordingly.

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Please also note the supplement to this comment:

<http://www.the-cryosphere-discuss.net/9/C208/2015/tcd-9-C208-2015-supplement.pdf>

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Interactive comment on The Cryosphere Discuss., 9, 77, 2015.

**TCD**

9, C208–C219, 2015

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1 **Dissolved organic carbon (DOC) in Arctic ground ice**

2

3 **M. Fritz<sup>1</sup>, T. Opel<sup>1</sup>, G. Tanski<sup>1</sup>, U. Herzschuh<sup>1,2</sup>, H. Meyer<sup>1</sup>, A. Eulenburg<sup>1</sup> and H.**  
4 **Lantuit<sup>1,2</sup>**

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10

11 Correspondence to: M. Fritz (Michael.Fritz@awi.de)

12

1 **Abstract**

2 Thermal permafrost degradation and coastal erosion in the Arctic remobilize  
3 substantial amounts of organic carbon (OC) and nutrients which have been accumulated in  
4 late Pleistocene and Holocene unconsolidated deposits. ~~Permafrost~~~~their~~ vulnerability to thaw  
5 subsidence, collapsing coastlines and irreversible landscape change is largely due to the  
6 presence of large amounts of massive ground ice such as ice wedges. However, ground ice  
7 has not, until now, been considered to be a source of dissolved organic carbon (DOC),  
8 dissolved inorganic carbon (DIC) and other elements, which are important for ecosystems and  
9 carbon cycling. Here we show, using **biogeochemical** data from a large number of different  
10 ice bodies throughout the Arctic, that ice wedges have the greatest potential for DOC storage  
11 with a maximum of 28.6 mg L<sup>-1</sup> (mean: 9.6 mg L<sup>-1</sup>). Variation in DOC concentration is  
12 positively correlated with and explained by the concentrations and relative amounts of  
13 typically terrestrial cations such as Mg<sup>2+</sup> and K<sup>+</sup>. DOC sequestration into ground ice was more  
14 effective during the late Pleistocene than during the Holocene, which can be explained by  
15 rapid sediment and OC accumulation, the prevalence of more easily degradable vegetation  
16 and immediate incorporation into permafrost. We assume that pristine snowmelt is able to  
17 leach considerable amounts of well-preserved and highly bioavailable DOC as well as other  
18 elements from surface sediments, which are rapidly **frozen and** stored in ground ice,  
19 especially in ice wedges, even before further degradation. **We found that ice wedges** ~~in~~  
20 Yedoma region ~~ice wedges~~ represent a significant DOC (45.2 Tg) and DIC (33.6 Tg) pool in  
21 permafrost areas and a fresh-water reservoir of 4, ~~200472~~ km<sup>3</sup>. This study underlines the need  
22 to discriminate between particulate OC and DOC to assess the availability and vulnerability  
23 of the permafrost carbon pool for ecosystems and climate feedback upon mobilization.

24

## 1 Introduction

Vast parts of the coastal lowlands of Siberia, Alaska and Canada consist of unconsolidated organic-rich fine-grained deposits. These sediments, that occur as glacial and Yedoma-type sediments (including their degradation forms as thermokarst), are characterized by high ground-ice contents, both on a volumetric (vol%) and gravimetric (weight %) basis (Brown et al., 1997; Zhang et al., 1999; Grosse et al., 2013; Schirrmeister et al., 2013). Yedoma deposits, which formed during the late Pleistocene cold stages in unglaciated Beringia (Schirrmeister et al., 2013), for instance, are characterized by absolute ground-ice contents, excluding ice wedges, of 40-60 weight % (Schirrmeister et al., 2011c). Ice wedges are one of the most common types of ground ice in permafrost. They form when thermal contraction cracks open in winter, which are periodically filled with snow meltwater in spring that quickly (re)freezes at negative ground temperatures to form ice veins and finally vertically foliated ice wedges. The ice wedges are themselves characterized by volumetric ice contents closing 100 vol% and make much of the subsurface in these Yedoma deposits. Recent calculations of ice-wedge volumes in East Siberian Pleistocene Yedoma and Holocene thermokarst deposits show contents of 48 vol% and 7 vol%, respectively (Strauss et al., 2013). Combining ice wedges and other ice types sediments-in Yedoma deposits gives a mean volumetric ground-ice content for those regions between 60 and 82 vol% (Zimov et al. 2006a, b; Schirrmeister et al., 2011b, c; Strauss et al., 2013). High ground-ice contents are also typical for coastal Alaska (43-89 vol%; Kanevskiy et al., 2011, 2013) and the western Canadian Arctic (50-60 vol%; French, 1998). The presence of massive ice (i.e. gravimetric ice content >250% on dry soil weight basis; cf. van Everdingen, 1998) and excess ice, which is visible ice that exceeds the pore space, is the key factor for the vulnerability of permafrost to warmer temperatures and mechanical disturbance, as ice melt will initiate surface subsidence and thermal collapse, also known as thermokarst (Czudek and Demek, 1970). Permafrost soils hold approximately 50% of the global soil carbon pool (Tarnocai et al., 2009; Hugelius et al., 2014), mostly as particulate organic carbon (POC). These calculations of permafrost OC stocks, however, subtract the ground-ice content (Zimov et al. 2006a, b; Tarnocai et al. 2009; Strauss et al., 2013; Hugelius et al., 2013, 2014) and therefore disregard the OC, especially the amount of dissolved organic carbon (DOC), contained in large ground-ice bodies such as ice wedges and other types of massive ice. Although these numbers might be small compared to the POC stocks in peat and mineral soils, DOC from permafrost is chemically labile (Dou et al., 2008; Vonk et al., 2013a, b) and may directly enter local food

1 webs. Due to its lability, DOC can become quickly mineralized by microbial communities  
2 and photochemical reactions (Battin et al., 2008; Vonk et al., 2013a, b; Cory et al., 2014) and  
3 returned to the atmosphere when released due to permafrost degradation (Schuur et al., 2009;  
4 Schuur and Abbot, 2011).

5 | As mentioned above, several studies ~~that~~ have shed light on the POC stocks contained in  
6 permafrost (e.g. Zimov et al., 2006a; Tarnocai et al., 2009; Schirrmeister et al., 2011b; Strauss  
7 et al., 2013; Hugelius et al., 2013, 2014; Walter Anthony et al., 2014) and how much of these  
8 stocks is potentially mobilized due to thermal permafrost degradation and coastal erosion  
9 (Rachold et al., 2004; Jorgenson and Brown, 2005; Lantuit et al., 2009; McGuire et al., 2009;  
10 Ping et al., 2011; Schneider von Deimling et al., 2012; Vonk et al., 2012; Günther et al., 2013;  
11 | 2015; Wegner et al., under review). DOC fluxes have also been quantified in western Siberian  
12 catchments (Frey and Smith, 2005) and monitoring efforts of the large rivers draining  
13 permafrost areas and entering into the Arctic Ocean have provided robust estimations of the  
14 riverine DOC export (Raymond et al., 2007; McGuire et al., 2009). However, DOC stocks in  
15 permafrost ground ice and the resulting potential DOC fluxes in response to coastal erosion  
16 and thermal degradation are still unknown (Guo et al., 2007; Duo et al., 2008). At this  
17 moment, any inference about DOC stocks in permafrost and fluxes from permafrost is derived  
18 from measurements in secondary systems such as lake (e.g. Kling et al., 1991; Walter  
19 Anthony et al., 2014), river (e.g. Benner et al., 2004; Finlay et al., 2006; Guo et al., 2007;  
20 Raymond et al., 2007; Holmes et al., 2012) and ocean waters (e.g. Opsahl and Benner, 1997;  
21 Dittmar and Kattner, 2003; Cooper et al., 2005) or from laboratory experiments (Dou et al.,  
22 2008). In contrast, the purpose of this study was to sample and measure DOC at the source  
23 | (i.e. permafrost ground ice in permafrost) directly, before it gets altered by natural processes  
24 such as exposition to the atmosphere, lithosphere and hydrosphere.

25

26 Here, we present an Arctic-wide study on DOC stocks in ground ice, aiming at incorporating  
27 massive ground ice into the Arctic permafrost carbon budget. The specific objectives of our  
28 study are:

- 29 • to quantify DOC contents in different massive ground ice types,
- 30 • to calculate DOC stocks in massive ground ice at the Arctic level,
- 31 • to put ground-ice-related DOC stocks into the context of the terrestrial Arctic OC  
32 pools and fluxes.

- 1 | • and to introduce relationships between organic and inorganic geochemical parameters,  
2 | stable water isotopes, stratigraphy, and genetic and spatial characteristics to shed light  
3 | on the origin of DOC and the processes of carbon sequestration in ground ice;  
4 | ~~to calculate DOC stocks in massive ground ice at the Arctic level;~~  
5 | • ~~and to put ground ice related DOC stocks into the context of the terrestrial Arctic OC~~  
6 | ~~pools and fluxes.~~  
7 |



## 2 Study area and study sites

This study was carried out along the coastal lowlands of east Siberia, Alaska and northwest Canada (Fig. 1). All study sites, except for the Fairbanks area, are located within the zone of continuous permafrost. The sites cover a wide and representative range of geomorphological settings, terrain units and ground-ice conditions (Table 1). All studied ground-ice bodies were found in ice-rich unconsolidated Holocene and late Pleistocene deposits. Outcrops in permafrost were either accessible due to strong rates of coastal erosion along the ice-rich coasts forming steep exposures (Forbes, 2011) or were technically constructed for research purposes such as the CRREL Permafrost Tunnel in Barrow or for mining such as the Vault Creek Tunnel near Fairbanks, Alaska.

Coastal outcrops in Siberia were dominated by large late Pleistocene ice wedges reaching up to 20 m in depth and up to 6 m in width (Schirrmeyer et al., 2011c). They formed syngenetically during periods of rapid sedimentation of Ice Complex deposits, also known as Yedoma (Schirrmeyer et al., 2013). Holocene epigenetic and syngenetic ice wedges of 1 – 6 m in depth and <1.0 – 3.5 m in width were encountered in exposed thermokarst depressions of late glacial to Holocene origin and within the Holocene peaty cover deposits. Besides ice wedges, other types of massive ground ice were sampled, such as buried glacier ice, buried lake ice and a fossil snow patch (Fig. 2). In some cases, massive ground ice occupied as much as 90 vol% of 40 m coastal exposures eroding up to 10 m a<sup>-1</sup> (Lantuit et al., 2012). The focus of this paper is on massive ground ice; non-massive ice (in particular pore ice and intrasedimental ice such as ice lenses) was excluded from this first attempt to calculate DOC stocks in ground ice, because of the complex genetic processes associated with the interaction with enclosing sediment and the relatively small amount of ice relative to massive ice bodies. DOC in intrasedimental ice is, however, not considered to be insignificant.

## 1 3 Material and methods

### 2 3.1 Laboratory analyses

3 A total number of 101 ice samples from 29 ice bodies and 3 surface water samples  
4 from 3 thermokarst lakes were studied. Ice blocks were cut with a chain saw in the field and  
5 kept frozen until further processing with a band saw in a cold lab at  $-15^{\circ}\text{C}$  for removal of  
6 partially melted margins and cleaning of the edges. Samples  $\geq 50$  ml were thawed at  $4^{\circ}\text{C}$  in  
7 pre-cleaned (purified water) glass beakers (~~purified water~~) covered with pre-combusted  
8 aluminium foil ( $550^{\circ}\text{C}$ ). Meltwater was filtered with gum-free syringes equipped with glass  
9 fibre filters (Whatman<sup>TM</sup> GF/F; pore size:  $0.7\ \mu\text{m}$ ) and acidified with  $20\ \mu\text{l}$   $\text{HCl}_{\text{suprapur}}$  (30%)  
10 to  $\text{pH} < 2$  in order to prevent microbial conversion. DOC concentrations ( $\text{mg L}^{-1}$ ) were  
11 measured with a high-temperature ( $680^{\circ}\text{C}$ ) combustion total organic carbon analyzer  
12 (Shimadzu TOC-V<sub>CPH</sub>). Internal acidification is used to convert inorganic carbon into  $\text{CO}_2$ ,  
13 which is stripped out of solution. Non-purgeable organic carbon compounds are combusted  
14 and converted to  $\text{CO}_2$  and measured by a non-dispersive infrared detector (NDIR). The  
15 device-specific detection limit is  $0.4\ \mu\text{g L}^{-1}$ . For each sample, one measurement with three to  
16 five repetitions was performed and results were averaged.

17 Further analyses for hydrochemical characterization included pH, electrical conductivity,  
18 major anions and cations, and stable water isotopes ( $\delta^{18}\text{O}$ ,  $\delta\text{D}$ ). Stratigraphic investigations  
19 and stable water isotopes ~~and~~ were used to differentiate between genetic ice types and to  
20 assess their approximate age (i.e. Holocene and late Pleistocene). Analyses of  $\delta^{18}\text{O}$  and  $\delta\text{D}$   
21 ~~analyses~~ were carried out with a mass spectrometer (Finnigan MAT Delta-S) using the water-  
22 gas equilibration technique (for further information see [Horita et al., 1989](#); [Meyer et al.,](#)  
23 [2000](#)). The isotopic composition is expressed in delta per mil notation ( $\delta$ , ‰) relative to the  
24 Vienna Standard Mean Ocean Water (VSMOW) standard. The reproducibility derived from  
25 long-term standard measurements is established with  $1\sigma$  better than  $\pm 0.1\ \text{‰}$  for  $\delta^{18}\text{O}$  and  
26  $\pm 0.8\ \text{‰}$  for  $\delta\text{D}$  ([Meyer et al., 2000](#)). Samples for ion analysis were passed through cellulose-  
27 acetate filters (Whatman<sup>TM</sup> CA; pore size  $0.45\ \mu\text{m}$ ). Afterwards, samples for the cation  
28 analyses were acidified with  $\text{HNO}_3_{\text{suprapur}}$  (65%) to prevent microbial conversion processes  
29 and adsorptive accretion, whereas samples for anion analyses were kept cool. The cation  
30 content was analysed by Inductively Coupled Plasma-Optical Emission Spectrometry (ICP-  
31 OES, Perkin-Elmer Optima 3000 XL), while the anion content was determined by Ion  
32 Chromatography (IC, Dionex DX-320). Hydrogen carbonate concentrations were measured

1 by titration with 0.01 M HCl using an automatic titrator (Metrohm 794 Basic Titrino). Based  
2 on  $\text{HCO}_3^-$  concentrations we approximated the dissolved inorganic carbon (DIC)  
3 concentrations using the molecular weights.  
4

## 5 **3.2 Statistical methods**

### 6 **3.2.1 Principal Component Analysis (PCA)**

7 | Principal components analysis (PCA) was used to summarize the variation in a biplot  
8 | by reducing dimensionality of the data while retaining most of the variation in the data set  
9 | (Jolliffe, 2002). Ordinally scaled variables (i.e. chemical data set) were log-transformed,  
10 | centered and standardized except for pH,  $\delta^{18}\text{O}$ ,  $\delta\text{D}$ , latitude, and longitude not being log-  
11 | transformed due the inter-sample invariance. Ice types (ice wedge, buried lake ice, basal  
12 | glacier ice, snow pack ice, surface water), relative age (Pleistocene, Holocene, recent) were  
13 | coded with dummy variables and were superimposed as inactive supplementary variables on  
14 | the ordination plot to enable rough assumptions about the relationship between chemical  
15 | composition, ground-ice formation and age. The whole data set was reduced to 92 samples  
16 | and 23 variables by removing those containing missing values. PCA was performed with  
17 | focusing on inter-species correlation and was implemented using CANOCO 4.5 software for  
18 | Windows (ter Braak and Šmilauer, 2002).  
19

### 20 **3.2.2 Univariate Tree Models (UTM)**

21 | A powerful tool to explore the relationship between a single continuous response  
22 | variable (DOC concentration) and multiple explanatory variables is a regression tree (Zuur et  
23 | al., 2007). Tree models perform well with non-linearity and interaction between explanatory  
24 | variables. UTM is used to find interactions missed by other methods and also indicate the  
25 | relative importance of different explanatory variables. UTM was performed using the  
26 | computing environment R and Brodgar 2.6.5 software for Windows (ter Braak and Šmilauer,  
27 | 2002).  
28

## 1 4 Results

### 2 4.1 DOC and DIC concentrations

3 Table 2 provides an overview of mean DOC and DIC concentrations and range for  
4 each ground-ice types. We found strong variations of DOC concentrations within and across  
5 individual ground-ice types. The highest DOC concentrations were found in ice wedges with  
6 a mean of 9.6 mg L<sup>-1</sup> and a maximum of 28.6 mg L<sup>-1</sup>. Late Pleistocene ice wedges were  
7 characterized by higher mean DOC concentrations than Holocene ones with 11.1 and  
8 7.3 mg L<sup>-1</sup>, respectively. Other ice types had average DOC concentrations between 1.8 and  
9 3.0 mg L<sup>-1</sup> and their range was narrower than in ice wedges (Table 2, Fig. 3). Modern surface  
10 water gave DOC values between 5.5 and 5.876 mg L<sup>-1</sup>.

11 The highest DIC concentrations were found in modern surface water with on average  
12 22.6 mg L<sup>-1</sup> and a maximum of 40.2 mg L<sup>-1</sup> (Table 2, Fig. 3). DIC concentrations were lower  
13 in ground ice but varied strongly across ice types. With 8.5 mg L<sup>-1</sup> late Pleistocene ice wedges  
14 were characterized by almost four times higher mean DIC concentrations than Holocene ones  
15 (2.2 mg L<sup>-1</sup>; Fig. 3). Buried glacier and lake ice had similar mean DIC concentrations (around  
16 9 mg L<sup>-1</sup>) but showed large ranges; from values around zero up to 25 mg L<sup>-1</sup>. Basal glacier  
17 ice, buried lake ice, and snow pack ice show mean DOC concentrations between 1.8 and  
18 3.0 mg L<sup>-1</sup>. For individual sample values see Supplement Table S1.

19 ~~It is obvious that ice wedges were characterized by highest mean DOC concentrations but~~  
20 ~~rather low DIC concentrations compared to other ice types. Basal glacier ice, buried lake ice,~~  
21 ~~and snow pack ice show mean DOC concentrations between 1.8 and 3.0 mg L<sup>-1</sup>.~~

### 23 4.2 Correlation matrix

24 With the help of a correlation matrix environmental processes and chemical  
25 relationships can be visualized that may help to explain the sequestration of DOC into ground  
26 ice. Pearson's correlation coefficients were calculated and plotted in a correlation matrix in  
27 order to assess the degree of association between DOC, chemical properties, stable water  
28 isotopes and spatial variables (Fig. 4). A strong positive correlation suggests a mutual driving  
29 mechanism whereas negative values imply an inverse association. Most importantly, DOC is  
30 positively related to the relative proportion of Mg<sup>2+</sup> in the cation spectrum (R=0.65). Further

1 positive relations between DOC and other parameters, although less pronounced, involve  $K^+$   
2 ( $R=0.36$ ),  $HCO_3^-$  ( $R=0.36$ ) and latitude ( $R=0.38$ ). The only significantly negative relationship  
3 with regard to DOC exists together with  $Na^+$  ( $R=-0.44$ ) (Fig. 4). Climate-driven parameters  
4 such as  $\delta^{18}O$ ,  $\delta D$ , and D-excess do not explain DOC concentrations.  
5

### 6 4.3 Principal components

7 The first two axes of the PCA explain 43.9% of the variation in the data (Fig. 5).  $Cl^-$   
8 and  $Na^+$  ions are positively correlated with the first axis in descending order of correlation,  
9 whereas  $Ca^{2+}$ ,  $Mg^{2+}$ , and  $HCO_3^-$  ions and pH are negatively correlated. Parameters positively  
10 correlated with PCA axis 2 include information on the ice origin as Pleistocene and basal  
11 glacier ice. In contrast,  $\delta D$ ,  $\delta^{18}O$ , DOC concentration, and information on the ice origin as ice  
12 wedge and Holocene ground ice are negatively correlated with PCA axis 2. Variations in  
13  $SO_4^{2-}$  and  $NO_3^-$  concentration as well as information on latitude and longitude are not  
14 correlated with the first two PCA axes. The separation of ice samples in the PCA ordination  
15 plot leads to three distinct groups: (1) Holocene ice wedges and recent surface water samples  
16 are entirely negatively related to the second axis, whereas (2) Pleistocene ice wedges are  
17 entirely negatively related to the first axis. (3) Pleistocene basal glacier ice and buried lake ice  
18 is positively related to the second axis. This separation might be related to the different  
19 processes of ice formation and climate variation.

20  $Na^+$  and  $Cl^-$ -dominated samples represent Holocene ice wedges from coastal cliffs in east  
21 Siberia (Muostakh Island and Oyogos Yar). The majority of ice wedges with a terrestrial ion  
22 composition ( $Mg^{2+}$ ,  $Ca^{2+}$ ,  $HCO_3^-$ ) are of late Pleistocene age in areas such as Mamontov Klyk,  
23 Bol'shoy Lyakhovsky Island, Yukon Coast and the Fairbanks area. The first axis probably  
24 separates samples with a strong marine impact at its upper end from those with more of a  
25 rather—continental background. The second axis might represent climate conditions of  
26 formation. The majority of Pleistocene ice samples with a depleted stable water isotope  
27 composition show positive sample scores whereas Holocene ground ice being enriched in  
28 heavy stable water isotopes mostly shows negative sample scores and therefore plots in the  
29 lower part of the PCA (Fig. 5).  
30

#### 1 4.4 Univariate Tree Model (UTM)

2 UTM (Fig. 6a) shows that differences in DOC concentrations can be explained according to  
3 inorganic geochemical properties. The first two nodes split on  $Mg^{2+}$  with a threshold value of  
4 16% of the cation spectrum. The next nodes split according to thresholds in  $K^+$  of 2.30 and  
5 2.65%, respectively (Fig. 6a). Threshold percentages presented here are based on the cation  
6 spectrum only. This means that all measured cations sum up to 100 %. This is statistically  
7 more robust than using individual sample concentrations which can have different  
8 magnitudes. We end up with four statistically significant groups (i.e. nodes) with different  
9 mean DOC concentrations ( $mg\ L^{-1}$ ) of each group, also showing the number of observations  
10 in each group (n). With the UTM information – that inorganic geochemistry explains the  
11 variability in DOC concentration – we can make assumptions about relations between carbon  
12 sequestration in different water types. DOC concentration is not independent from inorganic  
13 geochemical composition. Cross validation (Fig. 6b) confirms statistical significance of the  
14 model result.

15

16

## 1 5 Discussion

### 2 5.1 DOC stocks in ground ice and relevance to carbon cycling

3 While the riverine DOC export to the Arctic Ocean has been estimated to ~~as much as~~  
4 ~~33-34~~ Tg a<sup>-1</sup> (McGuire et al. 2009; Holmes et al., 2012), comparable numbers for the DOC  
5 input by coastal erosion and thermal permafrost degradation (also known as thermokarst) are  
6 not available yet. This knowledge gap includes the DOC contribution derived from melting  
7 ground ice from ice-rich permafrost. Table 2 provides an overview of DOC contents in  
8 different massive ground-ice types from the North American and Siberian Arctic. Because of  
9 their wide spatial distribution in Arctic lowlands and the measured DOC concentrations, we  
10 conclude that from massive ground-ice types ice wedges hold the greatest potential for DOC  
11 storage with a maximum of 28.6 mg L<sup>-1</sup>. This is in good agreement with DOC measurements  
12 in a so far limited number of ice wedges by Douglas et al. (2011) in Alaska and Vonk et al.  
13 (2013b) in east Siberia who showed DOC concentrations of 18.4 – 68.5 mg L<sup>-1</sup> (n=5) and  
14 8.8 – 15 mg L<sup>-1</sup> (n=3), respectively.

15 Ulrich et al., (2014) have calculated maximum wedge ice volumes (WIV), which range from  
16 31.4 to 63.2 vol% for late Pleistocene Yedoma deposits and from 6.6 to 13.2 vol% for  
17 Holocene thermokarst deposits in east Siberia and Alaska. Strauss et al. (2013) have shown  
18 similar averages for WIV of 48 vol% in late Pleistocene Yedoma and 7.0 vol% for Holocene  
19 thermokarst deposits. Together with average DOC concentrations of 11.1 mg L<sup>-1</sup> (max. 28.6)  
20 this would lead to 5.3 g DOC per m<sup>3</sup> (max. 18.1) for late Pleistocene ice wedges in the upper  
21 late Pleistocene permafrost column (Table 3) and a DOC pool of 43.0 Tg DOC based on  
22 416,000 km<sup>2</sup> of undisturbed Yedoma in Beringia and a mean thickness of 19.4 m (Strauss et  
23 al., 2013). DOC stocks in ice wedges in Holocene thermokarst deposits are much lower with  
24 on average 0.51 g m<sup>-3</sup> and a maximum of 2.6 g m<sup>-3</sup> due to much lower ~~wedge-ice~~  
25 ~~volumes~~ WIV (cf. Ulrich et al., 2014) and slightly lower DOC concentrations (Table 3). With  
26 on average 2.2 Tg the Holocene ice wedge DOC pool is much lower than the late Pleistocene  
27 pool, mainly because of lower WIV, an average thickness of 5.5 m for thermokarst deposits  
28 and despite their greater extent (775,000 km<sup>2</sup>) than undegraded Yedoma deposits (Strauss et  
29 al., 2013). Even stronger differences are characteristic for DIC pools in late Pleistocene ice  
30 wedges (32.9 Tg) compared to Holocene ice wedges (0.66 Tg) in the same areas. Based on  
31 the above-mentioned spatial coverage of Yedoma and thermokarst deposits including  
32 sediment thickness and WIV, we conclude that in the study area ice wedges represent a

1 significant DOC (45.2 Tg) and DIC (33.6 Tg) pool in permafrost areas and a fresh-water  
2 reservoir of 4,172–200 km<sup>3</sup> (see Table 3).  
3 However, all types of non-massive intrasedimental ice, raising the total ground-ice volume to  
4 ~80% (Schirmer et al., 2011b; Strauss et al., 2013), are still excluded. Unpublished DOC  
5 concentrations in non-massive intrasedimental ice from Muostakh Island (Siberia) and the  
6 Yukon Coast (Canada) show overall averages of 327 and 100 mg L<sup>-1</sup>, respectively (Fritz,  
7 unpublished data). Higher DOC concentrations in intrasedimental ice than in massive ice are  
8 certainly due to the long-term contact of soil moisture with soil organic matter prior to  
9 freezing. We therefore suggest that incorporating DOC from non-massive ground-ice types  
10 would lead to a significant rise in DOC stocks in permafrost of at least one order of  
11 magnitude. However, a differentiation between particulate and dissolved OC in permafrost is  
12 not done yet, although the technical means via rhizon soil moisture sampling is already  
13 available on a cost- and time-efficient basis. Nevertheless, we are aware of the fact that DOC  
14 makes up a limited proportion of the whole permafrost carbon stocks. A cautious estimation  
15 of the ratio of DOC and POC is in the order of ~1/24000 if we consider about 24 wt% total  
16 organic carbon (TOC) in sediments (e.g. Schirmer et al., 2011b,c; Strauss et al., 2013) and  
17 about 10 mg L<sup>-1</sup> DOC in massive ground ice. This ratio will become much smaller if POC and  
18 DOC in the whole permafrost column would be differentiated, because TOC comprises both  
19 POC and DOC.  
20

## 21 5.2 Carbon sequestration and origin in relation to inorganic geochemistry

22 The origin and sequestration process into ground ice seems to play an important role  
23 in the magnitude and bioavailability of DOC. Sequestration of OC into ground ice is a  
24 complex process that is dependent on water source, freezing process, organic matter origin  
25 and inorganic geochemical signature of the ambient water to form ground ice.  
26 Figures 4 and 6a show that the ~~total mineralization~~ electrical conductivity (i.e. total ion  
27 content) of ground ice is unrelated to DOC but that the ion composition and therefore the ion  
28 source seems to be relevant. Mg<sup>2+</sup> and K<sup>+</sup> are the most significant parameters for explaining  
29 variations in DOC concentrations (Fig. 6a). Higher Mg<sup>2+</sup> and K<sup>+</sup> fractions of the cations  
30 spectrum are positively related to higher DOC concentrations (Fig. 4). We recognize that in  
31 the node (group) with the highest average DOC concentrations ( $\bar{C}$  = 11.9 mg L<sup>-1</sup>, n=40) we  
32 find most of the Pleistocene ice wedges and to a lesser extent Holocene ice wedges (Fig. 6a).



1 All study areas are represented here. Both,  $Mg^{2+}$  and  $K^+$  have typically high shares in  
2 terrestrial water types because Mg and K are major elements in clay minerals and feldspars. In  
3 combination with terrestrial  $HCO_3^-$  and  $Ca^{2+}$  the mobility of  $Mg^{2+}$  is high in  $Mg/Ca(HCO_3)_2$   
4 solutions (Granse and Führs, 2013).

5  
6 Ice wedges are fed by meltwater from atmospheric sources that have been in contact with  
7 vegetation and sediments of the tundra surface before meltwater infiltrated the frost cracks in  
8 spring. By contrast, glacier ice, buried snow bank ice, and lake ice is primarily fed by  
9 atmospheric waters having less interaction with carbon and ion sources. Yet, the yellowish  
10 brown to gray late Pleistocene and the milky-white Holocene ice wedges have incorporated  
11 sediments and organic matter that originates from surface soils and vegetation debris that was  
12 carried along with the meltwater into the frost crack (e.g. Opel et al., 2011). Spring snow melt  
13 water interacts with the soil material leaching out carbon as it trickles downward toward the  
14 ice wedges. Also, since wedges may take thousands of years to form and the location of their  
15 upper surface changes with time, there are numerous spatial and temporal ways that deeper  
16 soil pore waters can get incorporated into the wedge ice. Leaching of DOC from relatively  
17 young surface organic matter takes place (Guo et al., 2007; Lachniet et al., 2012) as well as  
18 dissolution of ions from sediment particles. Snow melt feeding ice wedges strongly attracts  
19 leachable components because of its initial purity. This might be the reason why especially  
20 ice wedges contain relatively high amounts of bioavailable DOC with low-molecular weight  
21 compounds that can be old but remained fresh over millennia (Vonk et al., 2013b).

22  
23 Principal component analysis clusters ice wedges into two main groups along the first axis  
24 based on  $Na^+$  and  $Cl^-$  dominating Holocene ice wedges in modern coastal settings and  $Mg^{2+}$ ,  
25  $Ca^{2+}$  and  $HCO_3^-$  for Pleistocene ice wedges and Holocene ones being far from coasts (Fig. 5).  
26 This pattern depicts the competing influence of maritime and terrestrial/continental  
27 conditions. A similar differentiation of ice wedges (and all ground ice types) is done along the  
28 second PCA axis (Fig. 5). Differences in stable water isotopes indicate the predominant  
29 climate variations between the late Pleistocene and the Holocene which are also reflected in  
30 the landscape (i.e. distance to sea; maritime vs. continental). Distance from the coastline is  
31 crucial for the incorporation of marine-derived ions through aerosols such as NaCl via sea  
32 spray. While the Fairbanks area is the only site far inland, all other study sites except for  
33 Samoylov Island in the central Lena River Delta are coastal areas today. However, during the

1 late Pleistocene global sea level was lower and large parts of the shallow circum-arctic  
2 shelves were subaerially exposed. Present-day coastal sites were located up to hundreds of  
3 kilometers inland. Marine ion transport via sea spray is not expected to have played a role on  
4 inland sites but indeed since the rapid marine transgression during the Holocene that changed  
5 far inland sites into coastal ones. Input of sea spray is only relevant during the open-water  
6 season so that a prolonged ice cover during the late Pleistocene (Nørgaard-Pedersen et al.,  
7 2003; Bradley and England, 2008) should have further reduced the influx of sea salt.  
8 Additionally, sustained dry conditions (Carter, 1981; Alfimov and Berman, 2001; Murton,  
9 2009) probably increased eolian input of terrestrial material into ice wedges which is then  
10 directly mirrored in the hydrochemical signature.

11  
12 So far we have shown that coastal/maritime and terrestrial environmental conditions can be  
13 differentiated based on inorganic hydrochemistry and that terrestrial surface OC sources feed  
14 the DOC signal in ice wedges. DOC sequestration into ground ice is also dependent on active  
15 layer properties, vegetation cover, vegetation communities, and deposition rates. Long-term  
16 stable surfaces and relatively constant active layer depths will lead to substantially leached  
17 soil layers in terms of DOC (Guo and Macdonald, 2006) and inorganic solutes (Kokelj et al.,  
18 2002). Based on  $\Delta^{14}\text{C}$  values and  $\delta^{13}\text{C}$  ratios on DOC from soil leaching experiments and  
19 natural river water samples, Guo et al. (2007) have shown that intensive leaching of DOC  
20 from young and fresh plant litter and upper soil horizons occurs during the snowmelt period.  
21 Later in the season, DOC yields decreased in rivers draining permafrost areas, indicating that  
22 deepening of the active layer and leaching of deeper seasonally frozen soil horizons were  
23 accompanied by much lower concentrations of DOC due to the refractory and insoluble  
24 character of the remaining organic matter compounds. ~~In addition~~In contrast, dissolved  
25 organic matter compounds in runoff into lakes and rivers are can become rapidly degraded by  
26 microbial communities and photochemical reactions (Striegl et al., 2005; Olefeldt and Roulet,  
27 2012; Cory et al., 2014). One destination of the fresh, young and therefore most bioavailable  
28 DOC components will be ice wedges (Vonk et al., 2013b), where the chemical character is  
29 preserved because of immediate freezing. ~~In contrast, dissolved organic matter compounds in~~  
30 ~~runoff into lakes and rivers are rapidly degraded by microbial communities and~~  
31 ~~photochemical reactions (Striegl et al., 2005; Olefeldt and Roulet, 2012; Cory et al., 2014).~~  
32 This highlights the importance of ground ice and especially ice wedges as a vital source of  
33 bioavailable DOC.

1

### 2 **5.3 DOC mobility and quality upon permafrost degradation**

3 The absolute numbers of DOC in permafrost might be still small compared to the  
4 ~~particulate fraction~~POC. However, POC from both peat and mineral soil has a relatively slow  
5 decomposition rate after thaw compared to DOC (Schuur et al., 2008). Organic matter from  
6 melting ground ice was shown to be highly bioavailable and can even enhance organic matter  
7 degradation of the host material by increased enzyme activity in ice wedge meltwater (Vonk  
8 et al., 2013b). Bioavailability experiments with Yedoma DOC from thaw streams fed by ice  
9 wedge meltwater in NE Siberia illustrated the rapid decomposability of Yedoma OC, with OC  
10 losses of up to 33% in 14 days (Vonk et al 2013a). Incubations with increasing amounts of ice  
11 wedge water in the Yedoma-water suspension enhanced DOC loss over time. Vonk et al.  
12 (2013b) concluded that ice wedges contain a DOM pool of reduced aromaticity and can be  
13 therefore regarded as an old but readily available carbon source with a high content of low-  
14 molecular weight compounds. Additionally, a co-metabolizing effect through high potential  
15 enzyme activity in ice wedges upon thaw leads to enhanced degradation rates of organic  
16 matter of the host material. When studying organic matter cycling in permafrost areas we  
17 have to abandon the paradigm, which holds true for temperate regions and Arctic  
18 oceanography, that old OC is refractory and that only young OC is fresh, bioavailable, and  
19 therefore relevant for foods webs and greenhouse gas considerations.

20 We suggest that reduced organic matter degradation during cold periods is the main reason  
21 why late Pleistocene syngenetic ice wedges have incorporated more DOC on average than  
22 Holocene ice wedges. Incorporation of soluble organic matter into ground ice might have  
23 been more effective than today due to various reasons. Ice Complex deposits in the coastal  
24 lowlands formed during the late Pleistocene cold period, when high accumulation rates of  
25 fine-grained sediments and organic matter were accompanied by rapidly aggrading permafrost  
26 (Hubberten et al., 2004). This means that ~~on the one hand~~, organic matter is less decomposed  
27 because it was rapidly incorporated into perennially frozen ground and into the surrounding  
28 syngenetic ice wedges as the permafrost table rose together with the rising surface during  
29 ~~while~~ deposition (Schirrmeister et al., 2011b). ~~One the other hand~~Also, colder annual air  
30 temperatures led to reduced decomposition rates of organic matter which originated from  
31 vegetation communities dominated by easily decomposable forbs (Willerslev et al., 2014) in  
32 contrast to resistant sedge-moss-shrub tundra vegetation since postglacial times (Andreev et

1 al., 2011). Additionally, low precipitation and reduced runoff presumably retained more DOC  
2 in the landscape, ready to be transported into frost cracks.

3

4 Guo et al. (2007) concluded that most of the DOC in Arctic rivers is derived from young and  
5 fresh plant litter and upper soil horizons. Leaching of deeper seasonally frozen soil horizons  
6 were accompanied by much lower DOC concentrations due to the refractory and insoluble  
7 character of the remaining organic matter compounds (Guo et al., 2007). DOC  
8 impoverishment in the active layer is logical as it is leached each season over a long time  
9 under modern climate conditions, where permafrost aggradation is much slower than during  
10 cold stages; if it happens at all. The quantity and quality of DOC pools in deeper permafrost is  
11 probably much higher because of – so far – suppressed remobilization. Dou et al. (2008)  
12 studied the production of DOC as water-extractable organic carbon (WEOC) yields from  
13 organic-rich soil horizons in the active layer and permafrost from a coastal bluff near Barrow  
14 (Alaska) facing the Beaufort Sea. Besides high DOC yields in the uppermost horizon (0-5 cm  
15 below surface) the second highest DOC yields derived from permafrost although the sampled  
16 horizon showed lower soil OC contents than others (Dou et al., 2008). Interestingly, higher  
17 fractions of low-molecular-weight DOC, which is regarded to be more bioavailable, were  
18 generally found at greater depths. This supports the view that permafrost deposits hold a great  
19 potential for mobilizing large quantities of highly bioavailable organic matter upon  
20 degradation. Coastal erosion and thermokarst often expose old and deep permafrost strata.  
21 Contained organic matter is directly exposed to the atmosphere and transferred into coastal  
22 and fresh-water ecosystems without degradation because of short travel and residence times.  
23 Therefore, Arctic coastal zones are supposed to receive ~~overproportionally~~ high loads of  
24 bioavailable dissolved and particulate organic matter. Dou et al. (2008) used pure water  
25 (presumably MilliQ) and natural sea water as solvent for studying the production of DOC. It  
26 turned out that seawater extraction significantly reduced DOC yields which were attributed  
27 mainly to reduced solubility of humic substances due to the presence of polyvalent cations  
28 such as  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  in seawater (Aiken and Malcolm, 1987). On the one hand Dou et al.  
29 (2008) invoked that a laboratory setup using pure water and dried/rewetted soil samples  
30 would lead to an overestimation of DOC input to the Arctic Ocean during coastal erosion. On  
31 the other hand and based on the large ground-ice volumes in coastal cliffs (Lantuit et al.,  
32 2012), we suggest that ~~sparsely mineralized~~ ice wedge meltwater with a low ion content is

1 probably able to leach greater amounts of DOC from permafrost upon thaw than other natural  
2 surface water.

3

4 An open question remains how much DOC can be found in intrasedimental ice and how much  
5 DOC is produced upon degradation of old permafrost (e.g. late Pleistocene Yedoma type) for  
6 example as a result of coastal erosion. The answer to this question is crucial to follow the fate  
7 of permafrost organic matter upon re-mobilization. Additionally, robust estimations of carbon  
8 release are crucial for predicting the strength and timing of carbon-cycle feedback effects, and  
9 thus how important permafrost thaw will be for climate change this century and beyond.

10

## 1 6 Conclusions and outlook

2 Ground ice in ice-rich permafrost deposits contains DOC, DIC and other nutrients,  
3 which are relevant to the global carbon cycle, Arctic fresh-water habitats and marine food  
4 webs upon release.

5 The following conclusions can be drawn from this study:

- 6 • Ice wedges represent a significant DOC (45.2 Tg) and DIC (33.6 Tg) pool in the studied  
7 permafrost areas and a considerable fresh-water reservoir of 4, ~~172-200~~ km<sup>3</sup>.
- 8 • Syngenetic late Pleistocene ice wedges have the greatest potential to host a large pool of  
9 presumably bioavailable DOC because of i) highest measured average DOC  
10 concentrations in combination with ii) their wide spatial (lateral, vertical) distribution in  
11 ice-rich permafrost areas and iii) the sequestration of fresh and easily leachable OC  
12 compounds.
- 13 • Increased incorporation of DOC into ground ice is linked to relatively high proportions of  
14 terrestrial cations, especially Mg<sup>2+</sup> and K<sup>+</sup>. This indicates that leaching of terrestrial  
15 organic matter is the most relevant process of DOC sequestration into ground ice.

16

17 Based on our results about the stocks and chemical behavior of DOC in massive ground-ice  
18 bodies we propose that further studies shall strive to:

- 19 • quantify DOC fluxes in the Arctic from thawing permafrost, melting ground ice and ice  
20 wedges due to coastal erosion,
- 21 • differentiate between DOC and POC in permafrost including non-massive intrasedimental  
22 ice,
- 23 • quantify DOC production from permafrost in different stratigraphic settings and with  
24 different natural solvents to answer the question, what fraction of soil OC will be leached  
25 as remains POC and what is going to become DOC due to leaching,
- 26 • assess the age and lability of DOC versus POC in permafrost and the potential impact on  
27 coastal food webs and fresh-water ecosystems.

28

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12

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8 **Tables**

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10 **Table 1.** Summary of study areas, study sites, stratigraphy of the host sediments, ground-ice inventory and the studied ice types.

Region	Location	Longitude	Latitude	Stratigraphy and host sediments	Ground-ice conditions (inventory, ground-ice types, sampled ice types marked in <i>italicbold</i> )	Reference
Western Laptev Sea	Cape Mamontov Klyk	117.2	73.6	<ul style="list-style-type: none"> <li>Fluvial bottom sands – Late Weichselian Ice Complex – late glacial to Holocene thermokarst deposits – Holocene valley deposits – Holocene cover deposits</li> <li>Yedoma hills (20-40 m a.s.l.) of ice-rich permafrost sequences with wide and deep syngenetic ice wedges separated by thermoerosional valleys and thermokarst depressions</li> </ul>	Ice-rich permafrost sequences with wide and deep syngenetic <i>late Pleistocene ice wedges</i>	Schirmeister et al., 2008; Schirmeister et al., 2011b; Boereboom et al., 2013
Lena Delta	Samoylov Island	126.4	72.4	<ul style="list-style-type: none"> <li>First terrace (0-10 m a.s.l.): early to late Holocene delta floodplain, along the main river channels in the central and eastern parts of the delta; fluvial facies from organic-rich sands to silty-sandy peats towards bottom-up</li> <li>Modern to late Holocene floodplain; alluvial facies from peaty sands to silty-sandy peats bottom-up</li> </ul>	Ice-rich permafrost with active and buried syngenetic <i>Holocene ice wedges</i>  Ice-rich permafrost with epigenetic <i>Holocene ice wedges</i>	Schwamborn et al., 2002; Schirmeister et al., 2011a; Meyer et al., 2015
Eastern Laptev Sea	Muostakh Island	129.9	71.6	<ul style="list-style-type: none"> <li>Late glacial and Holocene cover deposits on top of Ice Complex</li> <li>Middle to Late Weichselian Ice Complex</li> </ul>	Very ice-rich permafrost, <i>late Pleistocene ice wedges</i> , <i>Holocene ice wedges</i>	Schirmeister et al., 2011b, c (and references therein), Günther et al., 2015

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Dmitry Laptev Strait	Oyogos Yar coast	143.5	72.7	<ul style="list-style-type: none"> <li>• Alternation of wide thermokarst depressions (alases) and hills representing remnants of Ice-Complex deposits (Yedoma)</li> <li>• Late glacial to Holocene thermokarst deposits and on top of Ice Complex</li> <li>• Taberite formed during Weichselian to Holocene transition</li> <li>• Late Weichselian Ice Complex</li> <li>• Middle Weichselian Ice Complex</li> </ul>	<p>Late Pleistocene and <i>Holocene ice wedges</i>.</p> <p>All ice wedges were sampled at a coastal bluff at an elevation of about 10 m a.s.l. in a central alas depression</p>	<p>Wetterich et al., 2009; Opel et al., 2011; Schirrmeister et al., 2011b</p>	<p><b>Formatiert:</b> Schriftart: Nicht Fett, Kursiv</p>
New Siberian Islands	Bol'shoy Lyakhovsky Island	143.9	73.2	<ul style="list-style-type: none"> <li>• Late Holocene cover deposits and Holocene valley deposits</li> <li>• Late glacial to Holocene thermokarst deposits</li> <li>• Taberite formed during Weichselian to Holocene transition</li> <li>• Middle Weichselian Ice Complex</li> </ul>	<p><i>Late Pleistocene ice wedges</i></p>	<p>Meyer et al., 2002; Andreev et al., 2004, 2009; Schirrmeister et al., 2011b; Wetterich et al., 2011, 2014</p>	<p><b>Formatiert:</b> Schriftart: Nicht Fett, Kursiv</p> <p><b>Formatiert:</b> Schriftart: Nicht Fett, Kursiv</p>
Northern Alaska	Barrow (CRREL Permafrost Tunnel)	-156.7	71.3	Buried ice-wedge system under about three meters of late glacial to early Holocene ice-rich sediments	<p><i>Late glacial ice wedges</i>, Holocene ice wedges</p>	<p>Sellman and Brown, 1973; Meyer et al., 2010a, b</p>	<p><b>Formatiert:</b> Schriftart: Nicht Fett, Kursiv</p>
Interior Alaska	Fairbanks (Vault Creek Tunnel)	-147.7	65.0	Discontinuous permafrost. Late Pleistocene ice-rich silty, loess-like organic-rich sediments between 12-15 m thick with large intersecting ice wedges	<p><i>Late Pleistocene ice wedges</i>, Holocene ice wedges</p>	<p>Shur, et al. 2004; Meyer et al., 2008</p>	<p><b>Formatiert:</b> Schriftart: Nicht Fett, Kursiv</p>
<u>Yukon Coast</u>	<u>Komakuk Beach</u>	<u>-140.5</u>	<u>69.6</u>	<ul style="list-style-type: none"> <li>• <u>Middle and late Holocene ice-rich peat, polygonal tundra</u></li> <li>• <u>Early Holocene thaw-lake sediments, peat, ice wedge casts</u></li> <li>• <u>Late Wisconsin (i.e. Late Weichselian) proluvial, alluvial, eolian deposits</u></li> </ul>	<p><u>Holocene ice wedges</u>, <u>Holocene snow pack ice (fossil snow bank)</u></p>	<p><u>Rampton, 1982; Fritz et al., 2012</u></p>	<p><b>Formatiert:</b> Schriftart: Kursiv</p> <p><b>Kommentar [MF1]:</b> This site was added because we had missed it before.</p>

Yukon Coast	Herschel Island	-139.1	69.6	<ul style="list-style-type: none"> <li>• Retrogressive thaw slumps along the coast exposing massive ground ice and ice-rich sediments</li> <li>• Holocene cover deposits and slope material along steep coastal bluffs</li> <li>• Mixed origin of marine, near-shore and terrestrial deposits</li> <li>• Push end-moraine of Late Wisconsin age</li> </ul>	<p><i>Buried glacier ice of ≥ 20 m thickness within Late Wisconsin diamicton</i></p> <p><i>Late Wisconsin ice wedges truncated by mass movement and early Holocene thaw unconformity</i></p> <p>Epigenetic and anti-syngenetic <i>Holocene ice wedges</i></p> <p><i>Buried lake ice, fossil snow bank ice</i></p>	<p>Mackay, 1959; Rampton, 1982; Fritz et al., 2011, 2012</p>	<p>Formatiert: Schriftart: Nicht Fett, Kursiv</p> <p>Formatiert: Schriftart: Nicht Fett, Kursiv</p> <p>Formatiert: Schriftart: Nicht Fett, Kursiv</p> <p>Formatiert: Schriftart: Nicht Fett, Kursiv</p>
Yukon Coast	Roland Bay	-139.0	69.4	<ul style="list-style-type: none"> <li>• Retrogressive thaw slumps along the coast exposing massive ground ice and ice-rich sediments</li> <li>• Holocene cover deposits and slope material along steep coastal bluffs</li> <li>• Late Wisconsin diamicton</li> </ul>	<p><i>Late Wisconsin and Holocene ice wedges</i></p>	<p>Rampton, 1982</p>	<p>Formatiert: Schriftart: Nicht Fett, Kursiv</p> <p>Formatiert: Schriftart: Nicht Fett, Kursiv</p> <p>Formatiert: Schriftart: Nicht Fett, Kursiv</p>
Yukon Coast	Kay Point	-138.2	69.2	<ul style="list-style-type: none"> <li>• Retrogressive thaw slumps along the coast exposing massive ground ice and ice-rich sediments</li> <li>• Holocene cover deposits and slope material along steep coastal bluffs</li> <li>• Moraine (ridge) of Late Wisconsin age</li> </ul>	<p>Presumably <i>Late Wisconsin buried glacier ice, Holocene ice wedges</i></p>	<p>Rampton, 1982; Harry et al., 1985</p>	<p>Formatiert: Schriftart: Nicht Fett, Kursiv</p>

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1 | [Table 2.](#) Summarized DOC and DIC concentrations of different massive ground-ice types.  
 2 | [For individual sample values see Supplement Table S1.](#)

Ice type	DOC		No. of ice bodies	No. of samples	DIC		No. of ice bodies	No. of samples	Stratigraphic affiliation
	DOC Mean [mg L <sup>-1</sup> ]	DOC concentration range [mg L <sup>-1</sup> ]			DIC Mean [mg L <sup>-1</sup> ]	DIC concentration range [mg L <sup>-1</sup> ]			
Ice wedge ice	9.6	1.6–28.6	22	72	4.7	0.3–19.8	21	66	Holocene, Late Pleistocene
Basal glacier ice	1.8	0.7–3.8	5	22	9.3	0.1–25.4	4	19	Late Pleistocene
Buried lake ice	2.0	0.3–5.2	1	6	8.8	0.3–22.9	1	6	Late Pleistocene
Snow pack ice	3.0	n.a.	1	1	n.a.	n.a.	0	0	Holocene
<i>Modern surface water</i>	5.6	5.5–5.7	3	3	22.6	5.0–40.2	3	3	<i>recent</i>

3 | Three modern surface water samples are from three different water bodies representing thermokarst  
 4 | ponds along the Yukon Coast.

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1 **Table 3.** DOC stocks and pools in late Pleistocene and Holocene permafrost containing ice  
 2 wedges (IW) based on calculated wedge-ice volumes (WIV) in Yedoma and thermokarst  
 3 basin deposits. All other ground-ice types, especially non-massive intrasedimental ice, are not  
 4 included.

	DOC concentrati on in Pleistocen e IW mg L <sup>-1</sup>	DOC concentrati on in Holocene IW mg L <sup>-1</sup>	WIV in Pleistoce ne Yedoma deposits vol%	WIV in Holocene thermoka rst deposits vol%	DOC stocks in Pleistoce ne permafro st <sup>c</sup> g m <sup>-3</sup>	DOC stocks in Holocen e permafro st <sup>c</sup> g m <sup>-3</sup>	DOC pools in Pleistoce ne permafro st <sup>c, d</sup> Tg	DOC pools in Holocen e permafro st <sup>c, d</sup> Tg
Min	2.4	1.6	16.7 <sup>a</sup>	1.0 <sup>a</sup>	0.4	0.02	3.2	0.07
Mea n	11.1	7.3 <del>3</del>	48.0 <sup>b</sup>	7.0 <sup>b</sup>	5.3	0.51	43.0	2.2
Max	28.6	19.5	63.2 <sup>a</sup>	13.2 <sup>a</sup>	18.1	2.6	145.9	11.0

5 <sup>a</sup> WIV data by [Ulrich et al., 2014](#). <sup>b</sup> Mean WIV data by [Strauss et al., 2013](#). <sup>c</sup> This includes ice wedges  
 6 only. <sup>d</sup> According to [Strauss et al. \(2013\)](#) undisturbed Pleistocene Yedoma covers 416,000 km<sup>2</sup> with a  
 7 mean thickness of 19.4 m, whereas Holocene thermokarst deposits cover 775,000 km<sup>2</sup> with a mean  
 8 thickness of 5.5 m.

9  
 10

1 **Figures**

2

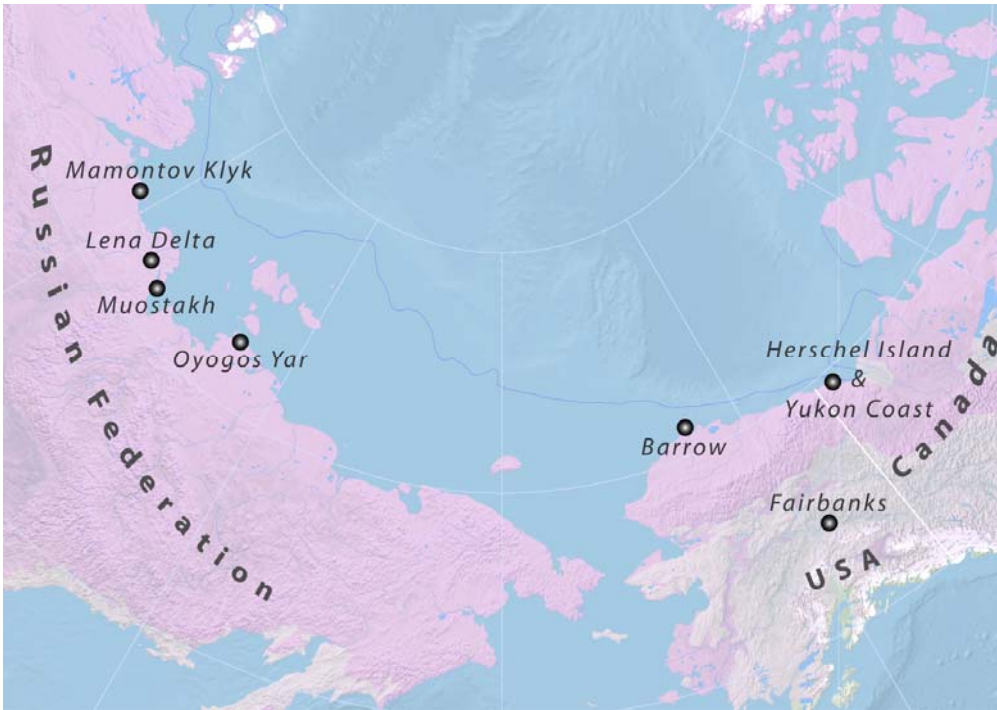


Figure 1. Study area and study sites (dots) for massive ground ice sampling in the Arctic lowlands of Siberia and North America. All study sites are located within the zone of continuous permafrost (dark purple), except for the Fairbanks area, which is the zone of discontinuous permafrost (light purple). Blue line in the Arctic Ocean marks the northerly extent of submarine permafrost according to Brown et al. (1997).

3

4



Ice-rich late Pleistocene permafrost, Oyogos Yar, Russia



Ice-rich permafrost coast, Muostakh, Russia



Epigenetic Holocene ice wedge, Lena Delta (Samoylov Island), Russia



Syngenetic late Pleistocene ice wedge, Mamontov Klyk, Russia



Buried glacier ice, Herschel Island, Canada



Snow pack ice, Herschel Island, Canada



Buried lake ice, Herschel Island, Canada

Figure 2. Ground-ice conditions and examples of studied ground-ice types in the Siberian and North American Arctic. Place names are plotted on Fig. 1.

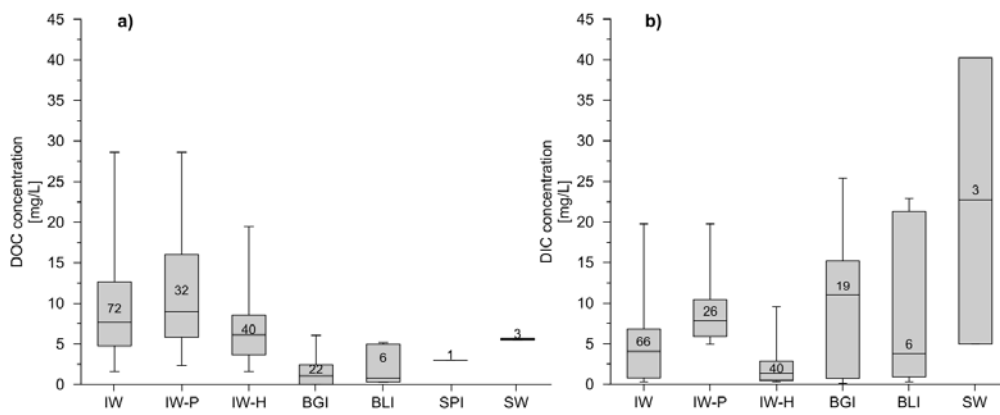


Figure 3. Boxplots of (a) DOC and (b) DIC concentrations in different massive ground-ice types. Plots show the number of samples in each category, minimum, maximum, median, 25 per cent-quartile and 75 per cent quartile as edge of boxes. IW: Ice wedges (all), IW-P: Pleistocene ice wedges, IW-H: Holocene ice wedges, BGI: Buried glacier ice, BLI: Buried lake ice, SPI: Snow pack ice, SW: Surface water. [For individual sample values see Supplement Table S1.](#)

1  
2



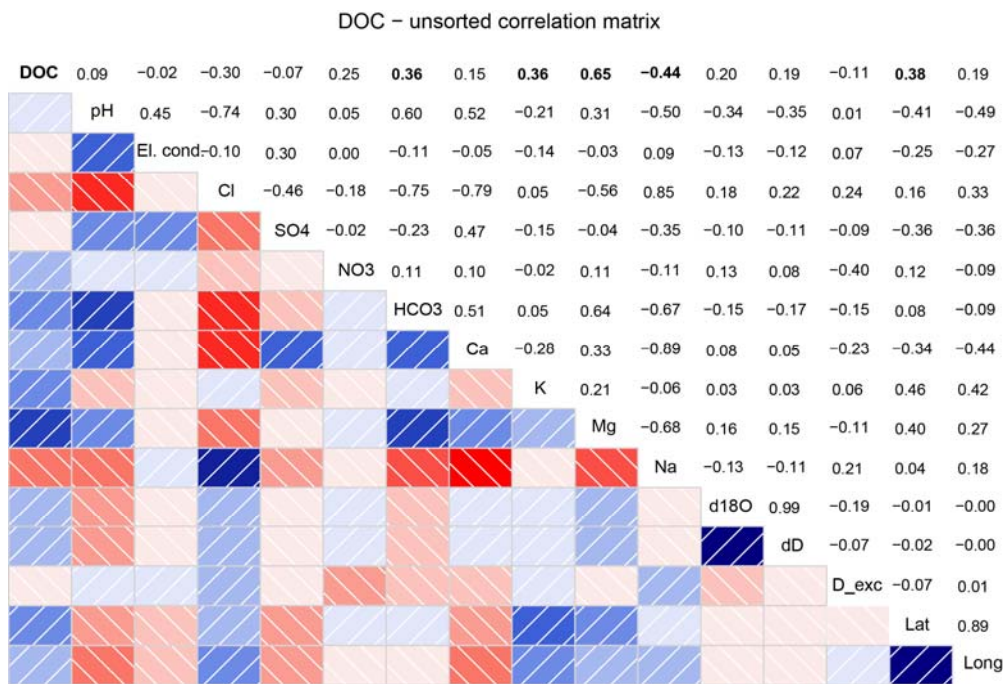


Figure 4. Correlation matrix. Correlations mentioned in the text are printed in bold. Strong positive correlations of paired variables are indicated by dark bluish colors, while strong anti-correlations are depicted in red. Hatching from the upper right to the lower left depict positive correlations, whereas negative correlations are reversely hatched for better perceptibility in a black-and-white print. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

1

2

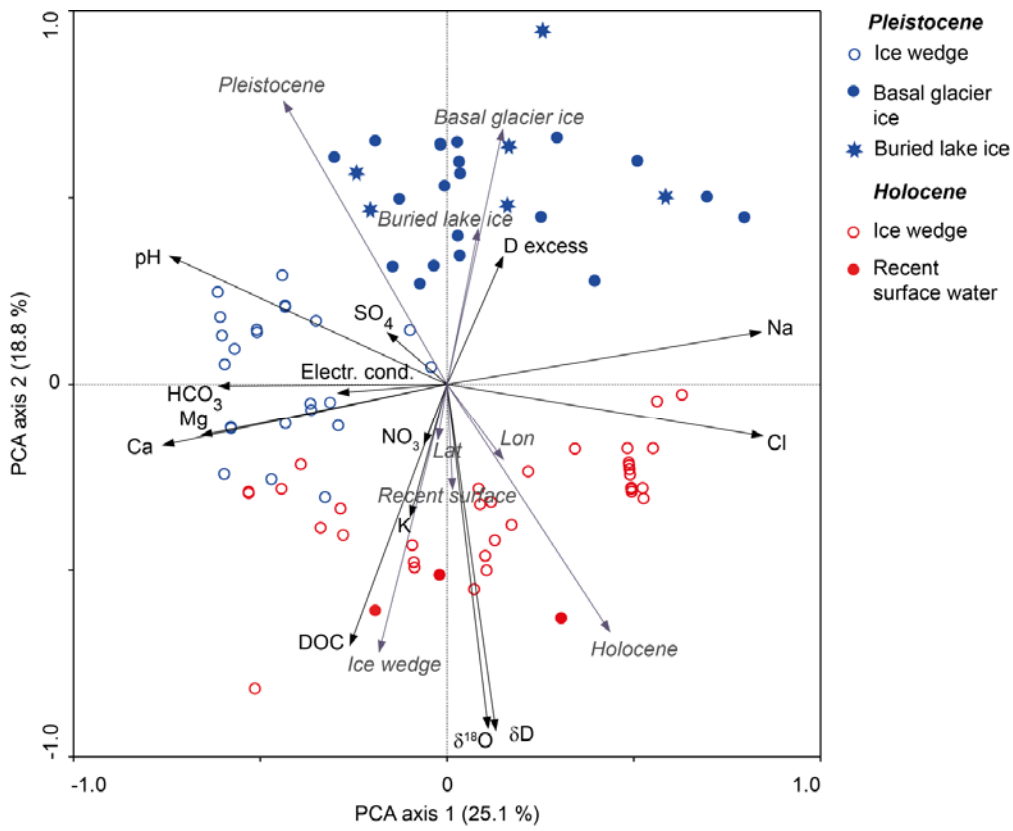


Figure 5. PCA biplot for ground-ice data. Inactive supplementary parameters (i.e. ice wedge, buried lake ice, basal glacier ice, snow pack ice, surface water, Pleistocene, Holocene, recent) are shown in grey italic. [For individual sample values see Supplement Table S1.](#)

1  
2

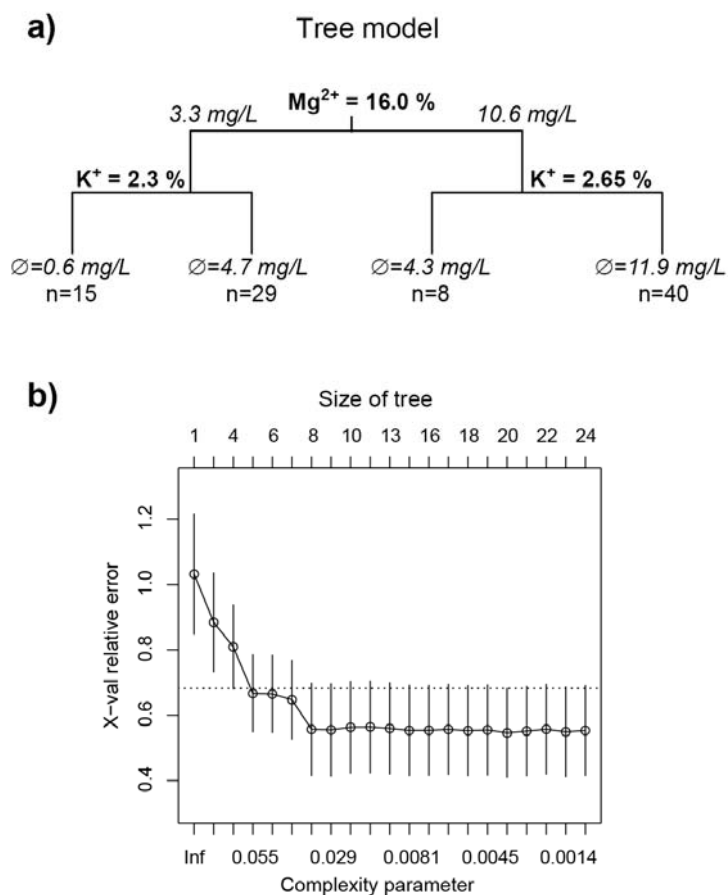


Figure 6. Univariate Tree Model (UTM) explains variability pattern in DOC concentration. a) Tree model focuses on DOC concentration as response variable. UTM uses 92 observations and a set of 22 explanatory variables.  $Mg^{2+}$  and  $K^+$  ions are most important to explain differences in DOC concentrations. Mean DOC concentrations of each group in  $mg L^{-1}$ . Number of observations in each group (n). b) Cross validation determines the statistically significant size of the tree model. The dotted line is obtained by the mean value of the errors (x-error) of the cross validations plus the standard deviation of the cross validations upon convergence. [For individual sample values see Supplement Table S1.](#)



Sample ID	Region	Location	Latitude	Longitude	Ice type	Stratigraphy [Holocene, Pleistocene]	DOC [mg/L]	DIC [mg/L]	d18O [‰]	δD [‰]	D excess [‰]	pH	Electrical conductivity [μS/cm]	Cl [mg/L]	SO4 [mg/L]	NO3 [mg/L]	HCO3 [mg/L]	Ca [mg/L]	K [mg/L]	Mg [mg/L]	Na [mg/L]
Oy7-11-IW7 11-12	Dmitry Laptev Strait	Oyogos Yar coast	72.70	143.50	Ice wedge	Holocene	6.13	0.42	-23.56	-183.1	5.4	5.61	39.5	7.96	0.31	BDL	2.14	0.34	0.85	0.46	4.25
Oy7-11-IW7 1-2	Dmitry Laptev Strait	Oyogos Yar coast	72.70	143.50	Ice wedge	Holocene	5.40	0.59	-25.69	-199.6	5.9	6.03	35.5	5.39	0.29	BDL	2.97	0.31	0.75	0.32	2.37
Oy7-11-IW7 13-14	Dmitry Laptev Strait	Oyogos Yar coast	72.70	143.50	Ice wedge	Holocene	6.15	0.48	-24.02	-184.9	7.3	5.83	48.0	8.42	0.32	BDL	2.44	0.37	1.12	0.51	4.33
Oy7-11-IW7 15-16	Dmitry Laptev Strait	Oyogos Yar coast	72.70	143.50	Ice wedge	Holocene	6.31	0.43	-24.88	-192.1	6.9	na	na	6.36	0.45	BDL	2.19	0.31	0.67	0.30	3.27
Oy7-11-IW7 17-18	Dmitry Laptev Strait	Oyogos Yar coast	72.70	143.50	Ice wedge	Holocene	7.75	0.53	-25.47	-196.9	6.9	5.53	36.5	5.52	0.39	0.22	2.67	0.36	0.87	0.41	2.62
Oy7-11-IW7 19-20	Dmitry Laptev Strait	Oyogos Yar coast	72.70	143.50	Ice wedge	Holocene	8.22	0.55	-25.10	-194.6	6.2	5.58	37.2	5.42	0.57	BDL	2.78	0.34	0.77	0.30	2.66
Oy7-11-IW7 21-22	Dmitry Laptev Strait	Oyogos Yar coast	72.70	143.50	Ice wedge	Holocene	8.12	0.99	-25.90	-199.7	7.5	6.29	48.9	5.67	1.25	0.22	5.03	0.30	2.09	0.25	2.27
Oy7-11-IW7 23-24	Dmitry Laptev Strait	Oyogos Yar coast	72.70	143.50	Ice wedge	Holocene	9.59	1.41	-25.87	-198.9	8.1	6.66	77.7	7.82	0.77	BDL	7.17	0.31	3.33	0.25	2.00
Oy7-11-IW7 3-4	Dmitry Laptev Strait	Oyogos Yar coast	72.70	143.50	Ice wedge	Holocene	5.87	0.36	-24.74	-191.9	6.0	5.56	35.3	7.05	0.31	BDL	1.83	0.32	0.50	0.37	3.60
Oy7-11-IW7 5-6	Dmitry Laptev Strait	Oyogos Yar coast	72.70	143.50	Ice wedge	Holocene	5.59	0.44	-25.54	-197.6	6.8	5.72	26.8	5.10	0.24	BDL	2.21	0.26	0.62	0.28	2.39
Oy7-11-IW7 7-8	Dmitry Laptev Strait	Oyogos Yar coast	72.70	143.50	Ice wedge	Holocene	6.63	0.36	-24.95	-192.3	7.3	5.75	47.8	10.20	0.37	BDL	1.83	0.38	0.61	0.55	5.25
Oy7-11-IW7 9-10	Dmitry Laptev Strait	Oyogos Yar coast	72.70	143.50	Ice wedge	Holocene	16.03	0.60	-24.34	-189.3	5.5	5.73	59.1	9.14	0.31	BDL	3.05	0.43	0.34	0.54	4.59
RB12-IW1	Yukon Coast	Roland Bay	69.43	-139.00	Ice wedge	Pleistocene	2.43	8.46	-27.58	-217.1	3.5	7.62	444.0	6.08	163.14	1.39	43.02	57.30	3.59	12.40	10.80
RB12-IW2	Yukon Coast	Roland Bay	69.43	-139.00	Ice wedge	Holocene	12.66	2.79	-23.87	-181.9	9.0	7.12	73.4	12.64	1.43	BDL	14.19	4.06	1.18	1.32	7.28
RB12-IW3	Yukon Coast	Roland Bay	69.43	-139.00	Ice wedge	Holocene	10.87	2.61	-23.23	-177.7	8.2	7.18	75.5	11.77	3.39	BDL	13.27	4.65	1.23	1.57	6.94
RB12-IW4	Yukon Coast	Roland Bay	69.43	-139.00	Ice wedge	Pleistocene	6.38	6.42	-26.02	-204.0	4.1	7.53	366.0	5.98	130.29	0.23	32.64	45.90	3.45	9.16	9.33
RB12-IW5	Yukon Coast	Roland Bay	69.43	-139.00	Ice wedge	Holocene	15.77	3.75	-23.19	-175.3	10.2	7.21	56.1	4.84	1.87	BDL	19.07	5.52	1.60	1.54	3.03
RB12-IW6	Yukon Coast	Roland Bay	69.43	-139.00	Ice wedge	Holocene	16.29	1.83	-22.10	-167.0	9.8	6.94	39.1	3.56	0.95	0.24	9.30	3.27	1.22	1.02	2.26
RB12-IW7	Yukon Coast	Roland Bay	69.43	-139.00	Ice wedge	Holocene	15.54	1.83	-21.45	-162.0	9.6	6.95	39.3	3.80	0.63	BDL	9.30	3.35	1.26	1.01	2.19
RB12-IW8	Yukon Coast	Roland Bay	69.43	-139.00	Ice wedge	Holocene	8.57	3.00	-21.55	-162.7	9.6	7.37	64.4	5.80	6.58	0.34	15.25	5.76	1.30	1.60	2.96
RB12-IW9	Yukon Coast	Roland Bay	69.43	-139.00	Ice wedge	Holocene	6.62	6.75	-23.99	-183.3	8.6	7.76	98.6	3.44	11.61	0.26	34.32	10.90	2.16	2.66	2.78
SP H2O 1-1m	Yukon Coast	Roland Bay	69.33	-139.03	Surface water	Recent	5.71	22.72	-15.67	-131.3	-5.9	7.57	384.8	56.45	9.35	BDL	115.51	33.03	2.41	8.61	30.85
TSA12-IW1-01	Yukon Coast	Herschel Island	69.58	-138.94	Ice wedge	Holocene	8.08	1.26	-21.20	-158.3	11.3	6.88	43.4	6.29	0.87	0.52	6.41	2.11	0.92	0.83	3.41
TSA12-IW1-02	Yukon Coast	Herschel Island	69.58	-138.94	Ice wedge	Holocene	8.28	1.98	-20.74	-154.9	11.0	7.25	62.3	10.45	1.05	0.22	10.07	3.45	1.05	1.43	5.11
TSA12-IW1-03	Yukon Coast	Herschel Island	69.58	-138.94	Ice wedge	Holocene	6.26	4.41	-20.91	-157.5	9.8	7.45	68.3	7.37	1.53	BDL	22.42	6.05	0.75	1.78	3.85
TSC12-IW1-01	Yukon Coast	Herschel Island	69.58	-138.95	Ice wedge	Holocene	13.24	2.85	-20.41	-152.5	10.8	7.20	92.7	16.10	3.39	0.23	14.49	5.99	0.92	1.92	8.33
TSC12-IW1-02	Yukon Coast	Herschel Island	69.58	-138.95	Ice wedge	Holocene	19.48	9.54	-20.50	-154.7	9.3	7.65	230.2	28.71	20.27	BDL	48.51	11.80	3.88	4.38	24.60
TSD09-6-1	Yukon Coast	Herschel Island	69.60	-139.23	Basal glacier ice	Pleistocene	0.74	2.61	-32.70	-253.2	8.4	8.00	68.2	6.90	3.02	0.17	13.27	4.19	0.37	1.14	4.39
TSD09-6-2	Yukon Coast	Herschel Island	69.60	-139.23	Basal glacier ice	Pleistocene	3.80	14.01	na	na	na	8.00	346.0	21.58	54.49	BDL	71.24	15.05	6.66	8.79	36.35
TSD09-IW-5-1	Yukon Coast	Herschel Island	69.60	-139.23	Ice wedge	Pleistocene	8.90	7.26	-28.36	-225.2	1.7	8.19	175.0	24.09	5.92	BDL	36.91	9.65	5.87	3.58	15.00
TSD12-IW1-71	Yukon Coast	Herschel Island	69.57	-139.02	Ice wedge	Pleistocene	5.85	6.72	-25.23	-194.6	7.2	7.85	173.1	10.09	33.97	BDL	34.17	20.10	1.60	2.87	5.89
TSD12-IW1-72	Yukon Coast	Herschel Island	69.57	-139.02	Ice wedge	Pleistocene	9.02	6.21	-25.77	-198.6	7.6	7.76	359.0	13.88	111.43	BDL	31.58	40.40	7.40	8.73	11.20
TSD12-IW1-73	Yukon Coast	Herschel Island	69.57	-139.02	Ice wedge	Pleistocene	7.89	4.95	-25.30	-193.3	9.1	7.68	161.8	8.06	36.96	BDL	25.17	14.70	3.96	4.29	5.13
TSD12-IW2-01	Yukon Coast	Herschel Island	69.57	-139.01	Ice wedge	Holocene	2.94	1.92	-24.17	-183.0	10.3	7.30	44.3	7.14	0.26	BDL	9.76	3.00	0.32	0.51	3.85
TSD12-IW2-02	Yukon Coast	Herschel Island	69.57	-139.01	Ice wedge	Holocene	4.25	2.07	-22.41	-169.6	9.7	7.25	47.2	7.09	0.31	0.15	10.53	3.77	0.39	0.60	3.71
TSD12-IW2-03	Yukon Coast	Herschel Island	69.57	-139.01	Ice wedge	Holocene	6.03	2.04	-23.97	-182.6	9.2	6.80	50.2	8.24	0.42	BDL	10.37	3.10	0.73	0.86	3.92
TSD12-MI-01	Yukon Coast	Herschel Island	69.57	-139.02	Basal glacier ice	Pleistocene	1.06	3.27	-31.47	-245.1	6.6	7.70	77.7	13.85	0.91	0.17	16.63	4.87	0.27	0.69	8.14
TSD12-MI-02	Yukon Coast	Herschel Island	69.57	-139.02	Basal glacier ice	Pleistocene	1.03	4.17	-31.67	-246.0	7.3	7.65	87.9	13.91	1.42	0.23	21.20	6.27	0.44	1.33	7.53
TSD12-MI-03	Yukon Coast	Herschel Island	69.57	-139.02	Basal glacier ice	Pleistocene	6.07	22.60	-31.62	-245.8	7.1	8.09	386.0	27.74	51.88	0.40	114.86	10.60	5.90	6.45	60.20
TSD12-MI-04	Yukon Coast	Herschel Island	69.57	-139.02	Basal glacier ice	Pleistocene	5.72	25.39	-32.98	-258.4	5.4	8.10	504.0	68.41	32.15	BDL	129.05	9.98	6.20	5.85	79.30
TSD12-MI-05	Yukon Coast	Herschel Island	69.57	-139.02	Basal glacier ice	Pleistocene	0.00	1.65	-33.40	-259.8	7.4	7.24	47.7	9.50	0.53	BDL	8.39	2.35	BDL	0.30	6.10
TSD12-MI-06	Yukon Coast	Herschel Island	69.57	-139.02	Basal glacier ice	Pleistocene	1.80	15.78	-33.47	-260.5	7.2	8.01	236.5	22.43	10.73	BDL	80.23	9.44	3.98	5.22	20.50
TSD12-MI-07	Yukon Coast	Herschel Island	69.57	-139.02	Basal glacier ice	Pleistocene	2.17	13.92	-33.24	-259.0	7.0	7.92	206.5	19.09	15.23	BDL	70.78	6.58	4.80	3.67	26.40
TSD12-MI-08	Yukon Coast	Herschel Island	69.57	-139.02	Basal glacier ice	Pleistocene	0.00	0.72	-32.00	-248.5	7.6	7.18	26.5	5.11	0.78	BDL	3.66	1.08	BDL	0.24	3.00
TSD12-MI-09	Yukon Coast	Herschel Island	69.57	-139.02	Basal glacier ice	Pleistocene	1.85	13.89	-34.44	-267.5	8.0	7.79	175.6	5.41	18.73	BDL	70.62	10.50	3.96	4.10	15.90
TSD12-MI-10	Yukon Coast	Herschel Island	69.57	-139.02	Basal glacier ice	Pleistocene	3.12	17.88	-34.14	-265.2	7.9	8.01	379.0	55.72	22.68	0.27	90.91	8.39	5.76	4.59	58.20
TSD-MI-6	Yukon Coast	Herschel Island	69.60	-139.23	Basal glacier ice	Pleistocene	0.77	na	-31.74	-249.7	4.2	7.10	28.0	na	na	na	na	na	na	na	na

na not available (no measurement)

BDL Below Detection Limit (For statistical analyses, this value was set to zero)