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Authors: Thompson, M. S., Giesler, R., Karlsson, J., and Klaminder, J.

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Size and characteristics of the DOC pool in near-surface subarctic mire permafrost as a potential source for nearby freshwaters

M. S. Thompson[1,2]
R. Giesler[2]
J. Karlsson[2] *and*
J. Klaminder[2]

[1]Corresponding author: Calgary, Alberta, Canada,
megan.shera@gmail.com
[2]Climate Impacts Research Centre, Department of Ecology and Environmental Science, Umeå University, Box 62, Abisko SE-981 07, Sweden

Abstract

Subarctic peatlands are rich sources of organic carbon for freshwater ecosystems. Where those peatlands are underlain by permafrost, permafrost thaw may cause an initial release of bioavailable dissolved organic carbon (DOC) to surrounding freshwaters. In this study, we measured icebound and potentially leachable (extracted) DOC quantities and indices of DOC quality in active layer and permafrost layers from two subarctic peat mires, Stordalen and Storflaket. Most of the permafrost layers did not contain more organic matter or exportable DOC (as g kg^{-1} dry soil) than the overlying active layer, and there was no difference in aromaticity, molecular weight, or the ratio between labile and recalcitrant DOC extracted from the permafrost and active layer. However, DOC held in segregated ice of the near-surface permafrost had relatively low aromaticity compared to extracted DOC from the same depth. Total icebound and potentially leachable DOC in the Stordalen mire permafrost that is predicted to experience active layer deepening during each of the next 50 years corresponded to about 0.1% of the current annual aquatic export of DOC from the mire. We conclude that the pool of potentially leachable DOC currently stored in permafrost layers is small. We also highlight differences in permafrost organic material between the two studied mire systems, which has an effect on the pool and properties of leachable DOC that is potentially available for export during thaw. Moreover, the geomorphological form of permafrost thaw will influence future hydrological connectedness and DOC production, in turn determining future DOC export from the mires.

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Introduction

Worldwide, the carbon pool contained in permafrost is estimated to be larger than the current atmospheric pool (Schuur et al., 2008), although there is a high degree of uncertainty in current estimates (Hugelius et al., 2013). Arctic permafrost is expected to decrease in area by 20%–90% by the year 2100, while thaw in the form of active layer deepening is expected to increase by 30%–100% in the same time period (Frey and McClelland, 2009). The potential release of permafrost carbon, whether directly to the atmosphere or via hydrological export, represents a major potential impact of climate change and can feed back on the climate system by raising atmospheric CO_2 and CH_4. Furthermore, as permafrost thaws, some of the dissolved carbon will be transported to freshwater systems in runoff or via thermokarst development (Prowse et al., 2006). The export of dissolved organic carbon (DOC) from terrestrial environments is important for the function of Arctic surface waters (Ask et al., 2012), and numerous studies have focused on assessing net changes in the export of DOC following permafrost thaw (Walvoord and Striegl, 2007; Olefeldt and Roulet, 2012; Carey, 2003; Petrone et al., 2007; Frey and Smith, 2005). To what extent potentially leachable DOC originating in permafrost can contribute to these net changes is, however, not well chracterized.

Permafrost has particular hydrological characteristics, generally acting as an aquitard and restricting the flow of infiltrating runoff to the seasonally thawed active layer (Péwé and Sellmann, 1973; MacLean et al., 1999). Because runoff-associated leaching processes do not operate within the permafrost, it is generally held that permafrost layers contain high concentrations of readily soluble compounds compared to the overlying active layer (Keller et al., 2007). Complicating this view is the known transfer of soluble materials from the active layer to the underlying permafrost with successive freeze-thaw cycles (Shur et al., 2005). Thaw water and infiltrating runoff migrates to the cold permafrost layer within the soil profile, forming a thin ice-rich transient layer at the top of the permafrost. Hence, this transient layer can have chemical and physical properties that differ from the underlying permafrost (Shur et al., 2005), including distinct concentrations of DOC. Yet DOC concentrations in deeper permafrost layers have rarely been measured.

Several recent studies have reported thawing of permafrost peat layers in subarctic and arctic mires (O'Donnell et al., 2011; Pokrovsky et al., 2011; Sannel and Kuhry, 2011). Previous studies have found that permafrost thaw may have a significant impact on organic carbon export from thawed soils to aquatic ecosystems (Frey and Smith, 2005; Kawahigashi et al., 2006; O'Donnell et al., 2012). Frey and Smith (2005), for example, used permafrost-free catchments in permafrost zones as a proxy for a post-thaw permafrost catchment. This approach is quite common in studies that are attempting to study permafrost thaw, because it is generally difficult to confirm the nature and rate of permafrost thaw without many years or decades of observational data. However, using this proxy method fails to provide information about how soil conditions might differ in permafrost versus non-frozen soils (i.e., changes in soil development conditions at depth and different DOC production rates related to air and, subsequently, soil temperature differences) (Kalbitz et al., 2000; Kawahigashi et al., 2004). Thus

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a proxy comparison involves confounding factors that influence DOC export, and direct measurement of permafrost DOC can provide a useful supplement to such landscape-scale permafrost carbon export assessments.

The rate of transport of organic compounds to surface waters as a result of permafrost thaw depends on the nature of the permafrost soils. If active layers deepen into poorly developed mineral soils, the potential for adsorption and immobilization of high molecular weight DOC is increased (Carey, 2003; Kawahigashi et al., 2006). If, however, active layers deepen into "unleached" organic soils, DOC export may increase. In addition to active layer deepening, permafrost thaw has caused subsidence of peat palsas (Malmer et al., 2005), submerging previously uplifted peat horizons in pools or adjacent fens and streams. Under such conditions, the export of permafrost DOC to freshwaters would be more direct than via surface runoff. Depending on the specific conditions, exported terrestrial DOC could either be respired in the terrestrial system or be exported and respired in recipient freshwaters (Kawahigashi et al., 2006; Karlsson et al., 2007; Roehm et al., 2009). In order to accurately assess the existing permafrost DOC pool as a potential source for freshwaters, icebound and potentially leachable mobile DOC stocks in permafrost must be assessed both qualitatively and quantitatively.

The specific objectives of this study were (i) to quantify the pool of C that is expected to be potentially leachable following permafrost thaw (including icebound DOC) and (ii) to assess the quality of this C. We conducted analyses of active-layer and near-surface permafrost soil cores with a high vertical resolution from the Stordalen and Storflaket mires in northern Sweden. We assessed concentrations of icebound DOC and water-extractable DOC in permafrost to quantify leachable C pools. In addition, we used spectral absorbance and fluorescence measurements of the dissolved organic matter fractions to estimate the quality of the DOC in the permafrost. Finally, we calculated the size of the pool of leachable DOC found within the upper 0.5 m of permafrost in the Stordalen mire, which is expected to thaw within the coming five decades if the current thawing rate proceeds into the future.

Materials and Methods

STUDY SITES

The two studied mires, Stordalen and Storflaket, are situated about 3 km apart within a large palsa peat complex in the larger (15 km^2) Stordalen catchment (68°20′90″N, 18°58′57″E) about 10 km east of the village of Abisko in northern Sweden. The mires are within the discontinuous permafrost region of northern Scandinavia and permafrost underlies raised plateaus (~1 m above surrounding hollows). Both mires occur in open canopy mountain birch forest (Olefeldt and Roulet, 2012). The active layer on the plateaus is approximately 0.5–1 m thick at maximum thaw (Åkerman and Johansson, 2008; Klaminder et al., 2008). In both mires, a layer of peat is underlain by silty lacustrine sediment with a glacial origin (Klaminder et al., 2008). Plateau vegetation is dominated by dwarf shrubs, lichens, and mosses and indicates ombrotrophic conditions. Vegetation in the adjacent hollows includes graminoids and indicates an increased connection to groundwater and nutrients (Klaminder et al., 2008). The mean annual temperature at Abisko is –0.2 °C and the mean annual precipitation is 329 mm, based on the period 1986–2000 (Malmer et al., 2005). However, a recent analysis of long-term (1913–2006) temperature trends indicated that annual mean temperatures have increased to >0 °C during recent decades (Callaghan et al., 2010).

The two study mires are located along a valley bottom within the same hydrological catchment, with Storflaket (68º20′51″N, 18º15′55″E) located roughly mid-catchment and Stordalen (68º20′90″N, 18º58′57″E) located just above the catchment outlet (Lundin et al., 2013; Olefeldt and Roulet, 2014). The Storflaket mire is connected via bordering streams to catchment flows, while the Stordalen mire complex is bordered by several streams and lakes. There is evidence of past palsa formation from a fen environment at both mires, measured as changes in ^{13}C, and indicating a shift in metabolism from anaerobic to aerobic degradation (Alewell et al., 2011). Carbon dating indicated that these shifts, detected in the palsa active layers, occurred between 155 and 246 years ago at Stordalen and between 212 and 670 years ago at Storflaket (Alewell et al., 2011). The palsa plateaus have undergone permafrost thaw, mainly in the form of active layer deepening at Storflaket (Åkerman and Johansson, 2008) and active layer deepening and significant thermokarst-related subsidence at Stordalen (Malmer et al., 2005; summarized in Åkerman and Johansson, 2008). Alewell et al. (2011) chracterized Storflaket as a relatively undegraded palsa system, compared to the thermokarst-affected Stordalen mire. Both mires were considered in a recent analysis of aquatic C export by Olefeldt and Roulet (2014). That study's permafrost plateau catchment was located on the Stordalen plateau, and the mire containing permafrost in the mixed catchment was Storflaket.

SOIL SAMPLING

The Stordalen and Storflaket mire systems contain intensively studied tracts of peat plateau underlain by permafrost. There is an established record of active layer depth at Storflaket (Åkerman and Johansson, 2008, spanning 29 years) and permafrost extent at Stordalen (Malmer et al., 2005, spanning 30 years). At each mire, three coring sites were identified: one site was identified in the elevated plateau area (P) without evidence of permafrost erosion or collapse, a second site on the elevated plateau at an erosion front (E) directly adjacent to a hollow area, and a third hollow site (H). The hollow sites were generally saturated with water at approximately 10 cm below the surface. At each of these three site types, two cores were taken within 1 m of each other, giving a total of 12 cores from both mires. While the extensive permafrost records at Stordalen and Storflaket were a prerequisite for the current study, other ongoing field research limited the extent of this permafrost sampling program because of the possibility that coring with a relatively large-diameter drill would accelerate permafrost thaw and/or disturb existing study plots, especially at sensitive sites like erosion fronts. For this reason, replicates allowing for statistical comparisons were not collected, and instead sampling sites were distributed across elevation gradients with duplicate cores taken to provide confirmation of the general soil characteristics.

In September 2009, cores of the active layer were collected with a hammer-type corer using a 5.8-cm-diameter inner core tube. Permafrost cores were subsequently collected using a drill with a 7-cm-diameter stainless steel core barrel and a carbide cutting tip powered by an ice auger motor (modified from plans by Calmels et al., 2005). Cores were collected to 1 m below the permafrost table, except at the Stordalen hollow site where the permafrost was <1 m thick. Cores were cut into 5 cm sections in the field, which were then placed into plastic sample bags in coolers and immediately frozen after arrival at the lab. Every section of the three Stordalen (SD) cores were analyzed, while Storflaket (SF) permafrost cores were analyzed continuously from the permafrost surface to 0.3 m below the permafrost table, plus sections at 0.5 cm below and 1 m below the table, where available. A total of 201 core sections were analyzed separately.

EXTRACTION OF DOC

Active layer core sections were thawed and gently homogenized by hand, and 30 g of soil (fresh weight) were placed in 250 mL centrifuge bottles. Extraction of this active layer soil was as follows: a 150 mL portion of deionized MilliQ (MQ) water was added to the soil, equivalent to a soil:solution ratio of 1:5 (weight in g:volume in mL) (Jones and Willett, 2006). This mixture was shaken on an orbital shaker (150 rpm) in darkness at approximately 5 °C for 24 hours. The mixture was thereafter centrifuged (60 min, 14,000 rpm, 5 °C), and the supernatant water removed to separate containers for further analyses.

Permafrost core sections were thawed at room temperature (20 °C) overnight in darkness. If the thawed permafrost had supernatant water overlying the saturated soil after thawing (i.e., excess ice in frozen permafrost), the entire core section was homogenized, placed in a centrifuge bottle, and left to settle for a further 24 hours in darkness at room temperature. The settling of the supernatant water overnight was necessary to provide an accurate measure of the excess ice in the permafrost core sections, but was not ideal for the maintenance of the DOC in the ice before measuring. The samples were not refrigerated because the associated vibration would inhibit the fine clays and silt from settling. We acknowledge that some of the DOC in the excess ice would be degraded during the settling period, so that the DOC concentrations reported below are likely underestimates. Additionally, Roehm et al. (2009) showed that permafrost soil extractions inoculated with lake bacteria showed an approximately 25% increase in DOC aromaticity after 48 hours of incubation at 3.5 °C. Therefore, DOC aromaticity in the permafrost ice might be overestimated in our results, although none of these samples was inoculated with bacteria.

After settling, supernatant volumes were recorded in the centrifuge bottles by measuring equivalent volumes of water in centrifuge bottles filled to the marked level of the settled soil and supernatant water. The samples were thereafter centrifuged (60 min, 14,000 rpm, 5 °C) and the supernatant thaw water removed to a separate bottle for further analyses. A subsample of 30 g of this centrifuged permafrost core section was then transferred to a 250 mL centrifuge bottle and extracted as for the active layer soils. Permafrost soil sections without overlying thaw water were treated in the same manner as active layer samples.

For selected sections from three Stordalen cores (P, E, and H sites), the extraction procedure was repeated sequentially three times and the solution separated after each consecutive extraction. This step was meant to roughly estimate the potential labile DOC pool after initial permafrost thaw. This sequential extraction was performed only on core sections located 20 cm above and below the permafrost table. The interval between extractions was variable between 1 and 4 days, with one interval at 7 days (for three samples from the SD-H active layer). Samples were refrigerated between sequential extractions.

For all core sections, after the 30 g fresh weight soil portion was removed for extraction, the remaining unextracted soil was retained to determine loss on ignition (LOI).

ANALYSES

All thawed ice water and extraction water was filtered through 0.45 μm syringe filters (Filtropur S, Sarstedt AG & Co.) before analyzing for DOC and UV-visible light absorbance. DOC was obtained after acidification of the water samples on a Shimadzu TOC analyzer by the catalytic combustion technique (Shimadzu TOC-VcPH total organic carbon analyzer). Absorbance was measured on a Jasco V-650 spectrophotometer with continuous scans at each nm from 200 to 900 nm (scan speed: 400 nm min^{-1}, slit width 1 nm). The spectral absorbance was used to characterize the DOM extracted from the cores. Specific UV absorbance (SUVA) was calculated as UV absorbance at 254 nm (in m^{-1}) divided by DOC concentration (mg L^{-1}) (Weishaar et al., 2003). SUVA is an indicator of the aromaticity of the DOM (higher SUVA means more aromatic DOM) (Weishaar et al., 2003). In soil extracts from the Stordalen mire, SUVA was also the strongest predictor of aquatic bacterial DOC consumption rates in soil extracts sampled from the Stordalen mire (higher SUVA means slower bacterial consumption) (Roehm et al., 2009). The slope of the log-transformed spectral absorbance between 275 nm and 295 nm provides an estimate of DOC molecular weight ($S_{275–295}$, Helms et al., 2008). DOC molecular weight is higher in samples that have lower $S_{275–295}$. Fluorescence index (FI) was obtained from fluorescence scans (emλ: 450nm:500nm, exλ:370 nm) measured on a Perkin Elmer LS-55 spectrofluorometer (McKnight et al., 2001). FI provides a ratio of the labile:recalcitrant nature of DOM. LOI was determined for samples dried at 50 °C for 3 days as the mass loss after heating at 550 °C for 5 hours.

CALCULATIONS

Excess ice content (I_c, reported as %) in the permafrost core sections was calculated using saturated soil and supernatant thawwater volumes using the following equation:

$$I_C = \left\{ \left(W_V \times 1.09 \right) \div \left[S_V + \left(W_V \times 1.09 \right) \right] \right\} \times 100 \qquad (1)$$

where W_v is supernatant volume (cm^3), 1.09 = water to ice volume conversion, and S_v is saturated soils volume (cm^3) (Kokelj and Burn, 2005).

To estimate the areal ice and extractable DOC content in the Stordalen cores only (because sufficient ice volume and subsequently ice DOC data were not available for the relatively ice-poor Storflaket cores, and detailed measurements at 0.5 m below the permafrost table were not made), the dry bulk density of the soil was multiplied by each section volume to obtain the soil weight (g). This was then multiplied by the mean concentration of DOC in permafrost soils (g g dry $weight^{-1}$) to obtain the areal ice DOC content (in g m^{-2}) (after Kokelj and Burn, 2003). To estimate the pool of exportable C in segregated ice and soils at the Stordalen mire accumulated in permafrost layers that are expected to thaw during the coming decades, the cumulative DOC areal content at 0.5 m below the permafrost table was multiplied by the area of the Stordalen mire (Malmer et al., 2005). The P and E sites in this study were averaged to obtain an areal DOC estimate coinciding with "hummock" sites (7.28 ha), and H sites coincided with "wet" and "semiwet" sites (6.19 ha) described by Malmer et al. (2005).

STATISTICS

In addition to depth profiles of organic matter quantity and quality, the changes in DOC yield and SUVA across the sequential extractions were compared using paired Wilcoxon signed-rank tests (data were not normally distributed). We used linear regression to test for the relationship between DOC concentrations (mg L^{-1}, dependent variable) and organic matter as LOI (independent variable) as well as DOC and ice volume (mL, independent variable) following log-transformation of the data. This analysis was meant to provide more information about the location of DOC pools in the soil profile relative to soil organic matter and patterns in ice content. Because DOC is capable of migrating between the active layer and the permafrost, particularly during the formation of an ice-rich transient layer (Shur et al., 2005), shallow permafrost organic matter may not correlate with the amount of available DOC at a given depth.

Results

PHYSICAL PROPERTIES AND ICE CONTENT

Approximately 1 m of permafrost soil was collected at each sampling site. The active layer varied in depth from 0.44 to 0.65 m, and the total core depth ranged from 1 to 1.8 m. At the hollow sites (SD-H and SF-H), the coring drill broke through the bottom of the permafrost layer into unconsolidated water-logged sediments between 1 and 1.5 m below the ground surface. At these sites, the active layer was also waterlogged approximately 0.1 m below the surface.

At almost all coring sites, organic matter (OM, measured as LOI) decreased to 25% or lower between 0.5 and 1 m from the ground surface, generally 0.5 m below the permafrost table (Fig. 1). OM at SD-P decreased within the active layer, about 20 cm below the ground surface, and continued to decline to a sustained low of <10% at 0.5 m below the permafrost table. However, there was virtually no decrease in OM with depth at SF-P. In contrast to the other sample sites, OM at SF-P remained consistently above 80% as deep as 0.5 m below the permafrost table, and above 50% at the bottom of the sample core. In general, the change from surface organic soils to deeper mineral soils was gradual at the Stordalen plateau (SD-P) but more abrupt with depth at the erosion front sites (SD-E and SF-E). At SD-P and SD-E, there were localized peaks in OM at 0.75 below the ground surface. At SF-E, OM "rebounded," increasing to approximately 45% toward the bottom of the core. At the hollow sites (SD-H and SF-H), silt and sand were present in high quantities in the deep active layer (>0.3 m below surface) and throughout the permafrost layer, making OM patterns with depth somewhat erratic despite the general trend of decreasing OM with depth.

Ice content was low at both P sites (<20%), but ranged from 35.3% to 48.8% at the remaining core sites (Fig. 1). The three Stordalen sites contained more excess ice, and the pattern with depth is more discernible than in the Storflaket cores. The core at SD-E contained high ice content below the permafrost table, extending from 0.6 to 0.9 m below the ground surface. Below that point, excess ice content steadily decreased with depth to approximately 20%. At SD-P, in contrast, there were peaks in excess ice content (to ~20%) just below the permafrost table and at 1–1.3 m below the ground surface, below which ice content decreased. Ice content at SD-H was moderate, with no apparent depth trend. The cores collected at the Storflaket sites SF-P and SF-E had several sections with no recoverable excess ice. Excess ice content was high and variable at SF-H, reaching a maximum of approximately 48% 0.3 m below the permafrost table.

The subsurface peaks in the excess ice content indicate that a transient layer may exist at SD-P, and both E and H sites, located in the top 0.4 to 0.5 m of frozen ground (Fig. 1). At SD-P a second potential ice-rich transient layer is located approximately 0.75 m below the permafrost layer. The E sites near the subsided hollow sites had high ice content, which may reflect an intermediate state between the subsided H sites and "stable" P sites and thus represent at-risk areas for future subsidence (Mackay, 1992). The consistently low ice content at the P sites indicate that these sites are at the lowest risk for thermokarst development.

Visual examination of the duplicate core OM data indicated no difference between duplicates for all sites. Ice content in the SD-E duplicate was higher than in the principal core (peaked at ~57%) and was lower in the SF-E duplicate (>10%), indicating variability but supporting the pattern of higher ice content at Stordalen versus Storflaket sites.

DOC QUANTITY

In general, the amount of DOC extracted from the soil cores mirrored the relative patterns in OM at both Stordalen and Storflaket (Fig. 1). At both SD-E and SD-P, localized peaks in extracted DOC occurred at 0.75 m below the ground surface before decreasing gradually with depth. However, only SD-E had a similar peak in icebound DOC (Fig. 1). The remaining Stordalen cores (P and H) had low icebound DOC concentrations. At the Storflaket E and H sites, the DOC concentration in ice was equivalent to that of the DOC extracted from the surrounding permafrost soil (Fig. 1). This high concentration is noteworthy because ice DOC was generally contained in a smaller volume of thaw water (5–90 mL) than the DOC in the 150 mL of extraction water, and was not subjected to the shaking procedure of the extractions. In contrast to SF-E and SF-H, peak extracted DOC in the permafrost at SF-P was >1 g kg dry $soil^{-1}$ and differed from peak icebound DOC concentrations, which were <1 g kg^{-1} in the three samples that were collected.

The cumulative areal icebound and extracted DOC content (Fig. 2), which was calculated for Stordalen peaked at SD-E just below the permafrost table before leveling off, but increased gradually with depth at SD-P. Total DOC held in the uppermost 0.5 m of permafrost at the Stordalen mire (area delineated by Malmer et al., 2005) was 11.3 kg for icebound DOC and 26.4 kg for extracted DOC. Over the 1.3×10^5 m^2 of plateau and hollow in the Stordalen mire, this 37.7 kg of DOC converts to an average of 0.28 g C m^{-2}.

At the SD-P and SD-E sites, OM (as LOI, % composition) significantly predicted DOC (mg kg dry $soil^{-1}$) in regressions for both the excess ice and extraction samples ($p < 0.001$). However, at SD-H, OM did not significantly predict DOC in the excess ice or extractions ($p > 0.05$). Ice volume was not a significant predictor of ice DOC at any of the Stordalen sites ($p > 0.05$). This suggests that the ice-rich transient layers do not contain particularly high concentrations of DOC compared to the surrounding permafrost.

For extracted DOC, the core duplicate at SD-E did not contain the near-surface peak in permafrost evident in the principal core. At SF-E there was a peak (0.75 g kg^{-1}) in the permafrost at 0.75 m below the surface, and active layer DOC was relatively lower in the duplicate core (resembling extracted DOC at SF-P). The duplicate ice DOC at SF-P was relatively high ~1–1.5 m below the ground surface (0.5 g kg^{-1}), indicating potential for high ice DOC concentrations at the Storflaket plateau. Much of the variability in duplicate core DOC was evident at erosion front sites and indicates the need for more spatially extensive sampling at these sites to increase accuracy in the DOC pool estimate. All remaining duplicate core data for extracted and icebound DOC was not appreciably different from the principal core data.

DOC PROPERTIES

In general, the three measures of DOC quality indicate no trend with depth in extracted DOC aromaticity (SUVA), molecular weight (MW, $S_{275–295}$), or labile:recalcitrant nature (FI) at most of the Storflaket and Stordalen sites (Fig. 1). The exception was site SD-P, where aromaticity, molecular weight, and relative recalcitrant content of extracted DOC decreased with depth, particularly at 0.75 m below the ground surface, where $S_{275–295}$ peaked (Fig. 1). It is interesting to note that this is the same depth at which a localized peak in OM, extracted DOC, and ice content occurred at SD-P. At Stordalen, extracted permafrost DOC was least aromatic, with the lowest MW and the highest labile:recalcitrant ratio at SD-P, and was the most aromatic with the highest MW and the lowest labile:recalcitrant ratio at SD-H (Fig. 1). Within the active layer, extracted DOC properties were variable, with all three indicators overlapping in value between the three Stordalen sites.

In Storflaket, extracted DOC aromaticity at SF-P also dipped at 0.75 m below the ground surface, and also coincided with a localized peak in extracted DOC concentrations. Despite the similar pattern with depth, actual SUVA values in SF-P permafrost were higher, indicating more aromaticity, than at SD-P. Where SUVA declined below the permafrost table at SF-P, there was also a slight decrease in MW and increase in the labile:recalcitrant ratio, but the pattern was not as apparent as at the Stordalen plateau. At SF-E and SF-H there was no apparent pattern with depth, and, in contrast to Stordalen, the relative values for aromaticity were lower and the labile:recalcitrant ratio of extracted DOC at SF-H was higher than at SF-P and SF-E. MW indicator values were overlapping across the three Storflaket sites through the active layer and permafrost sections.

The mean SUVA of ice DOC was lower than for extracted DOC at most sites (Fig. 1), indicating that icebound DOC has lower aromaticity compared to the DOC extracted from permafrost soils. Only at SD-H did ice SUVA resemble extracted SUVA values. At Stordalen, icebound DOC MW decreased with depth at all sites, particularly at 0.75 m below the ground surface for SD-P and SD-E, and the labile:recalcitrant ratio was higher in icebound than extracted DOC (Fig. 1) . The general picture is that icebound DOC at Stordalen was less aromatic, with lower MW and higher labile:recalcitrant ratio than in extracted DOC. There was a noticeable difference between icebound and extracted DOC SUVA in Storflaket, where icebound DOC had relatively low aromaticity (Fig. 1), but there was little discernible difference in the other two DOC quality indices at the Storflaket sites.

Visual examination of the duplicate core SUVA data indicated variability at SF-E, where there was low aromaticity measured 0.75 m below the ground surface (min SUVA approximately 0.75 mg C L^{-1} m^{-1}, similar to SUVA patterns at SF-P). At SF-H, aromaticity in the duplicate core peaked at the permafrost table (max SUVA approximately 3.75 mg C L^{-1} m^{-1}). There was also variability in MW data between duplicate cores at SD-E, where there was a localized peak above and trough below the permafrost table. This pattern was not apparent in the principal core. The duplicate cores at both SF-E and SF-P had relatively low MW (high $S_{275–295}$) in the active layers (≥0.010 nm^{-1}), but showed little variability in permafrost. In contrast, the duplicate core at SF-H showed relatively high MW (<0.005 nm^{-1}) in the permafrost layers. For the labile:recalcitrant ratio, the duplicates had similar vertical patterns as the principal cores but were relatively high at SD-H and low at SF-H. Differences between remaining duplicate core DOC quality indices were not apparent.

SEQUENTIAL EXTRACTION—STORDALEN

Second and third sequential extractions combined yielded 64% of the DOC in the first extraction at SD-E, 75% at SD-H, and 81% at SD-P (Table 1). The aromaticity of the DOC increased with sequential extractions. The stepwise decrease in DOC and increase in SUVA with each extraction were significant (paired Wilcoxon signed-rank tests, $p < 0.05$).

Discussion

DOC QUANTITIES IN ACTIVE LAYER AND PERMAFROST SOILS

According to our estimate, the Stordalen mire stores an average of 0.28 g C m^{-2} as icebound and extractable DOC within the upper 0.5 m of the permafrost table. Currently, the active layer deepens at a rate of approximately 1 cm yr^{-1} (Åkerman and Johansson, 2008), indicating that about 2% of the total pool of icebound and potentially readily leachable DOC stored in the upper 0.5 m of the permafrost is released annually. This corresponds to a rate of about 0.0056 g C m^{-2} yr^{-1}, which in turn represents only 0.1% of the total DOC export from the Stordalen peatland complex, estimated at 5.3 g C m^{-2} yr^{-1} from 2008 data (Olefeldt and Roulet, 2012). This potential annual release of permafrost DOC is also low when compared to estimates of current DOC export for fen areas (5.2–8.1 g C m^{-2} yr^{-1}) or bog and palsa areas (2.5–3.5 g C m^{-2} yr^{-1}) located in the Stordalen mire (Olefeldt and Roulet, 2012). In the absence of subsidence, the results indicate that continued active layer deepening at Stordalen mire is not likely to significantly increase DOC export from palsas. In fact, active layer deepening has slowed recently in the Abisko region (attributed to dry summer periods) (Christiansen et al., 2010). In contrast, if palsa subsidence continues to occur at Stordalen, the rate of DOC export to freshwaters will be closely related to increased hydrological connectivity and subsequent fen development. Therefore, the dominant geomorphic form of thaw (active layer deepening vs. subsidence) is an important factor to consider in predicting the net effect of permafrost thaw on the future C export from the Stordalen mire.

Although we did not have the required data to estimate the C pool within 0.5 m of the permafrost table for the Storflaket mire, it is reasonable to assume from the extracted DOC results that the Storflaket plateau permafrost may act as a more significant pool of potentially leachable DOC during permafrost thaw than the Stordalen mire permafrost. Lundin et al. (2013) found that some of the largest annual aquatic C emissions (mostly as CO_2) in the Stordalen catchment occur in the streams immediately below the Storflaket mire (>5000 g C m^{-2} yr^{-1}). In comparison, annual C emissions in the streams (500–2000 or 2000–5000 g C m^{-2} yr^{-1}) surrounding the Stordalen mire were lower. Our results highlight important differences in permafrost OM content between the two mires, which plays an important role in determining the amount of potentially leachable DOC at each location.

The active layer at Storflaket has deepened by about 0.2 m between 1978 and 2006 (Åkerman and Johansson, 2008). This means that the lower 0.2 m of the active layer at Storflaket is recently thawed permafrost, and we can consider the DOC extracted from the active layer just above the permafrost table as representative of recently thawed layers. There was no apparent peak in extracted DOC above the permafrost table at the Storflaket hollow or erosion front site, but there was a small peak at the Storflaket plateau site. This indicates that recently thawed permafrost layers on the plateau are a source of DOC that is greater than is derived from the shallow active layer, indicating a potential increase in post-thaw

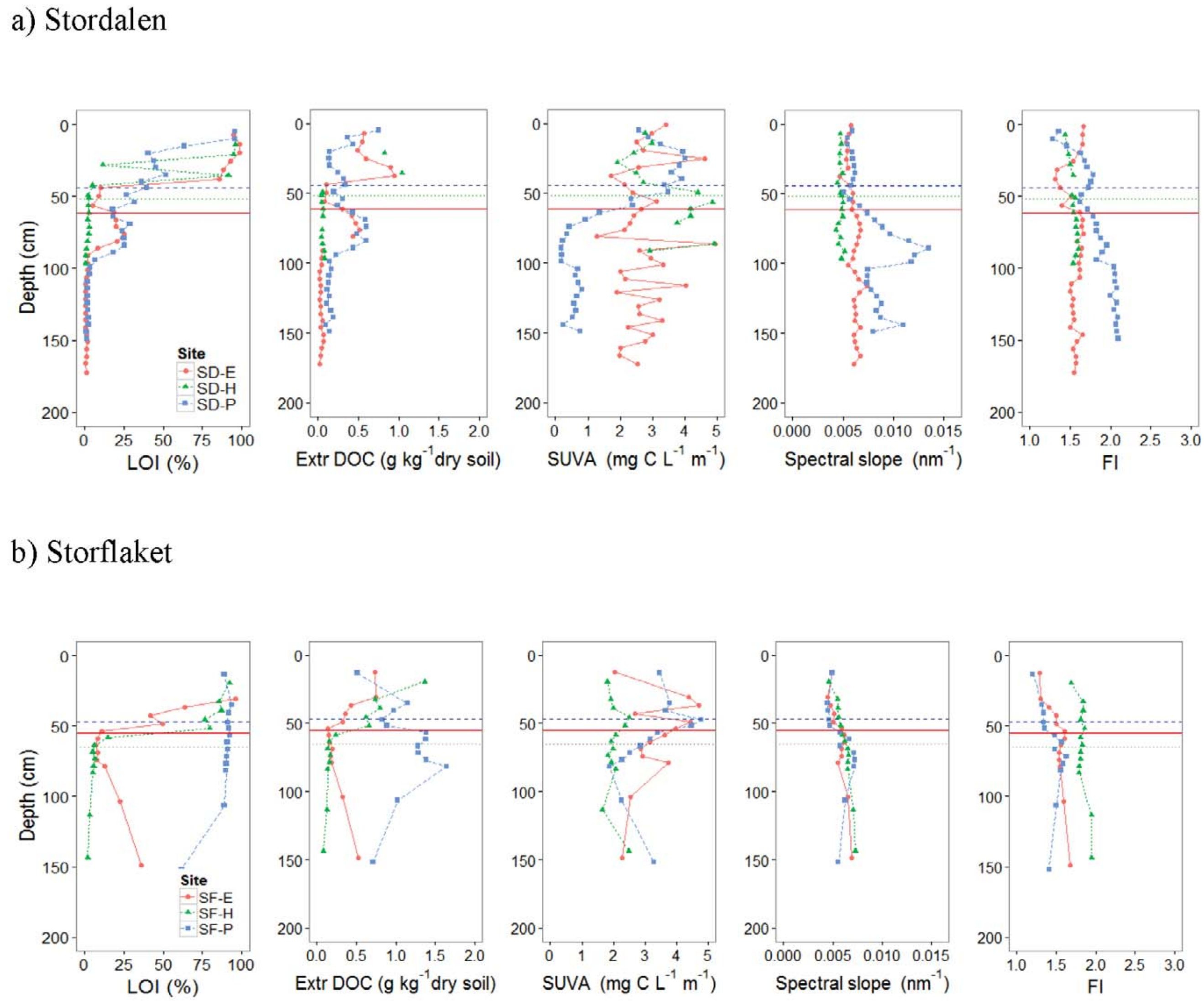

FIGURE 1. Soil organic matter (LOI), ice content, extracted and icebound DOC (in g per kg dry soil), specific UV absorbance (SUVA), spectral slope ($S_{275-295}$), and fluorescence index (FI) for cores from the Stordalen (SD) and Storflaket (SF) mires. Coring sites are plateau (-P), erosion front (-E), and hollow (-H). Depth is from the ground surface at 0 m. The permafrost table at each site is indicated by a horizontal line that corresponds to each plotted line.

soil DOC production, and that a shift in DOC export may have already occurred at Storflaket.

QUALITATIVE PROPERTIES OF C WITHIN THE ACTIVE LAYER AND PERMAFROST LAYER

Indices of DOC quality showed that extracted DOC at most of the Stordalen and Storflaket core sites was not consistently qualitatively different between active layer and permafrost layers. Comparison of measured DOC quality at Stordalen with a previous study shows that the permafrost DOC sampled in this study was generally less aromatic, with a higher labile:recalcitrant ratio than in extracted permafrost DOC sampled by Roehm et al. (2009) (from Stordalen; mean SUVA, 6.56 mg C L^{-1} m^{-1}; mean FI, 1.35), although it should be noted that the extraction method differed from the one used here (shaken for 12 h, centrifuged at 10,000 rpm for 10 min, filtered at 0.22 µm, Roehm et al., 2009). Olefeldt and Roulet (2012) measured runoff from a Stordalen palsa catchment, which had a range in SUVA of 1.59–4.41 mg C L^{-1} m^{-1} and a range in FI of 1.36–1.52. These ranges are comparable to extracted DOC in Stordalen permafrost, especially near the top of the permafrost table, while deeper permafrost SUVA and FI at the Stordalen plateau had lower aromaticity and was less labile (SUVA generally < 1 mg C L^{-1} m^{-1}; FI > 1.5). The results further indicate that the DOC that may be released with active layer deepening at the Stordalen mire is likely to resemble the quality of current runoff with initial thaw, and may become less aromatic and more labile as thaw continues into deeper permafrost layers. At the same time, the sequential extraction results indicate that long-term leaching of thawed permafrost layers will increase the aromaticity of exported DOC. Roehm et al. (2009) have shown that permafrost DOC with high SUVA was relatively biodegradable by lake bacteria, and we have refrained from discussing SUVA as an indicator of biodegradability in freshwater systems because of the variety of soils analyzed,

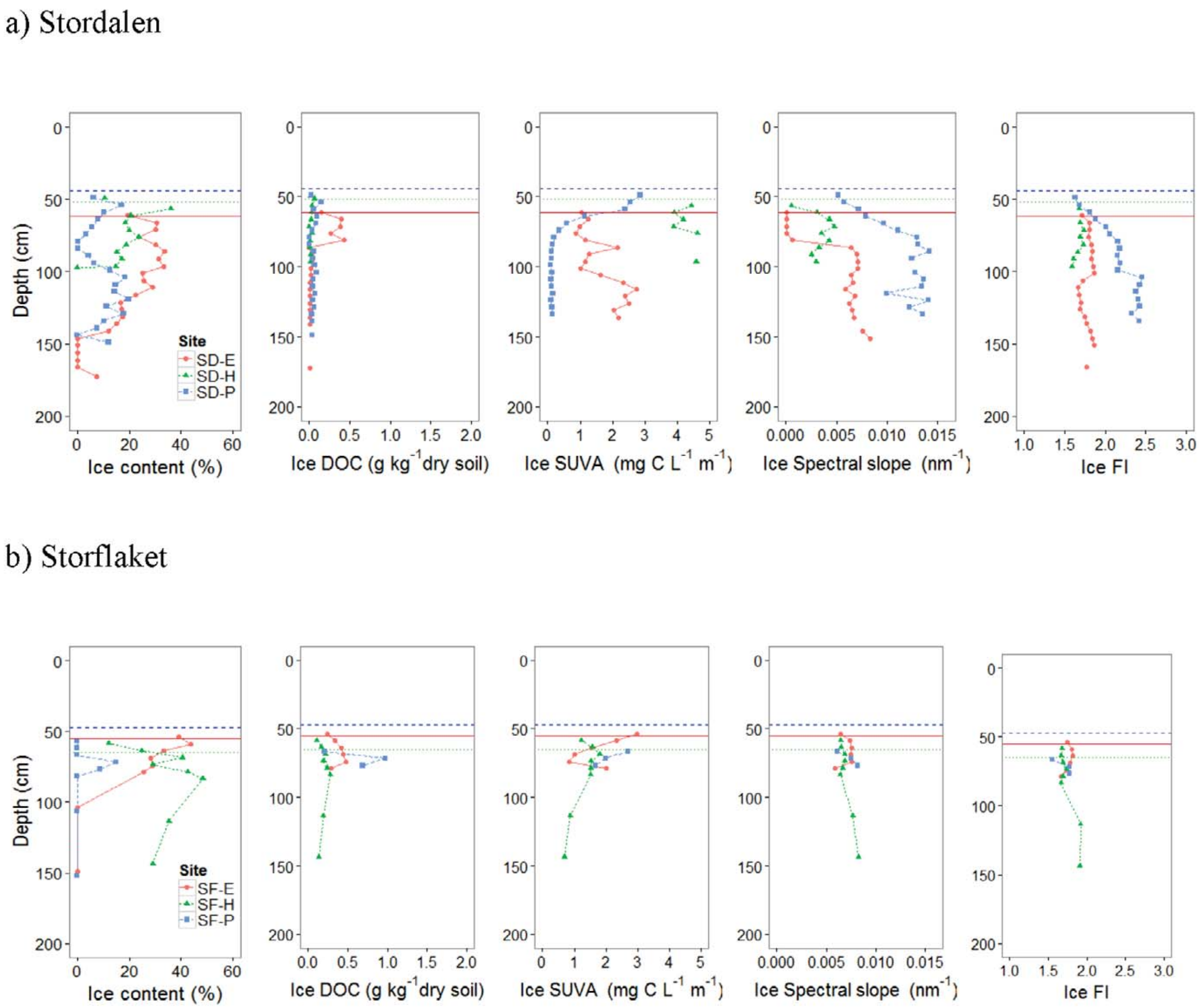

FIGURE 1 (continued).

particularly organic versus mineral, and the weakened link between SUVA and biodegradability where DOC is derived from mineral soils (summarized in Roehm et al., 2009).

IMPACTS OF PERMAFROST THAW

The results of this study indicate that a difference in potential geomorphic change between the study mires exists, with implications for DOC export. Ice content in the Storflaket cores are high near the permafrost table, but there is no excess ice in most of the measured permafrost profile, especially at SF-P and SF-E. In contrast, ice content at SD-E was high and at SD-P increased 0.5 m below the permafrost table. These characteristics indicate that with thaw the Storflaket plateau, having a more vertically extensive peat layer and lower ice content, may be less susceptible to thermokarst-related subsidence than the Stordalen plateau, where sands and silts occur alongside larger volumes of segregated ice (Mackay, 1992). In effect, permafrost thaw in each of the study mires seems likely to continue in its currently dominant form—active layer deepening at Storflaket and active layer deepening with significant subsidence at Stordalen.

In the case of active layer deepening, the DOC released directly from thawing permafrost will most likely vary in nature as different soil components thaw. When excess ice thaws, relatively low molecular weight and labile DOC will be released, especially at Stordalen. As the new active layer strata are infiltrated by runoff, DOC will be mobilized and transported to nearby freshwaters. With further infiltration and continuing losses, less labile and higher molecular weight DOC will be leached from the former permafrost layers. At the Storflaket plateau, the direct export of DOC to freshwaters may increase as the near-surface permafrost layer thaws, leading to export of DOC with relatively low aromaticity. In contrast, the export of DOC from the Stordalen plateau is likely to resemble current DOC export from the active layer, and if thaw increases exposure to infiltrating flows along the hydrological flow path, the exposure will reach mineral soil layers in approximately 50 years. This increases the likelihood of DOC adsorption and removal from infiltrating flows (Kawahigashi et al., 2006; O'Donnell

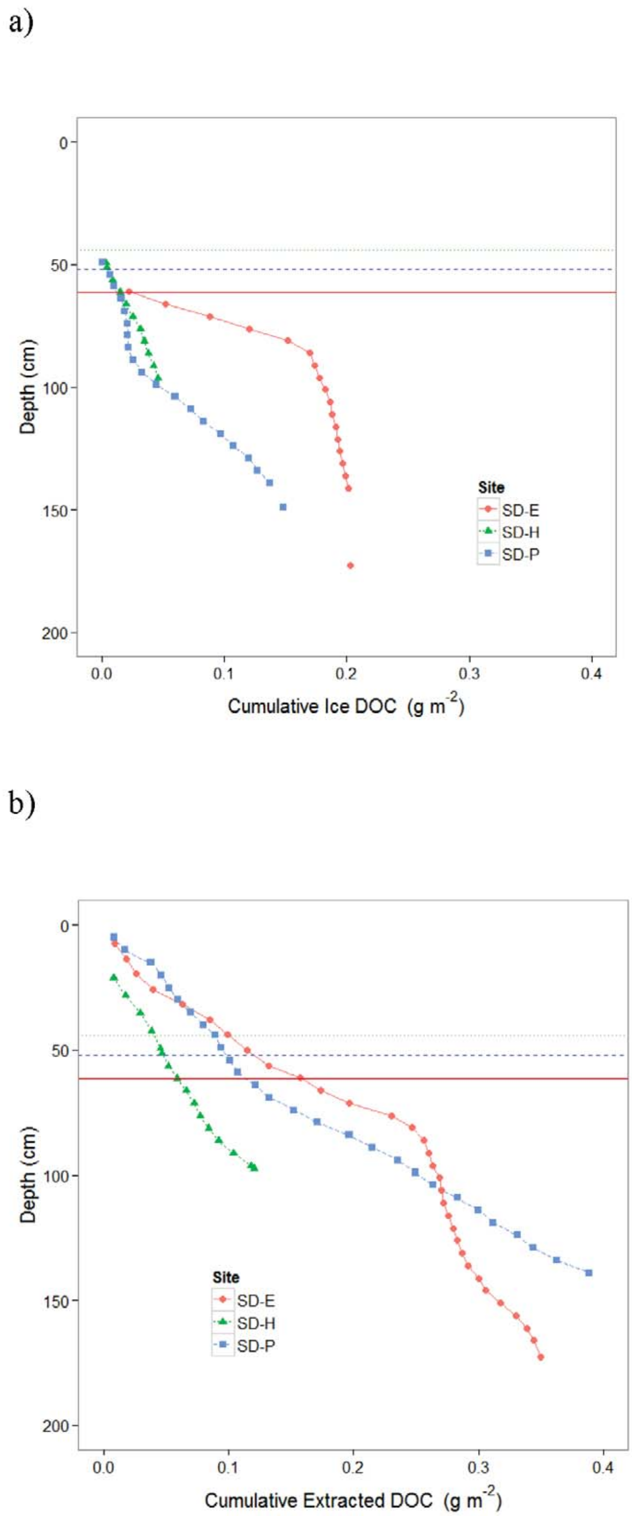

FIGURE 2. (a) Areal DOC content in permafrost ice and (b) full-core MilliQ water extraction (soil weight:solution volume ratio of 1:5). Data are presented as cumulative DOC with depth. Depth is from the ground surface at 0 m. The permafrost table at each site is indicated by a horizontal line that corresponds to each plotted line.

et al., 2012). Under this scenario, the release and export of DOC from Stordalen might actually be even less than before thaw, but further research is required to determine whether this will occur in the future. It is also important to note potential future changes in precipitation patterns, as a recent study of climate trends in the area indicates wetter summer and fall conditions, drier winter conditions and an increase in extreme precipitations events (Callaghan et al., 2010), all of which could impact infiltrating runoff.

TABLE 1

Mean extracted (a) dissolved organic carbon (DOC) concentration and (b) SUVA of Stordalen core sections subjected to three sequential extractions (± SD). For all extractions, core section *n* = 8 except for SD-H where *n* = 7, and span the 20 cm above and below the permafrost table.

Core	1st extraction	2nd extraction	3rd extraction
	(a) DOC (g kg^{-1} dry soil)		
SD-E	0.366 ± 0.291	0.141 ± 0.130	0.092 ± 0.079
SD-H	0.049 ± 0.007	0.018 ± 0.002	0.018 ± 0.006
SD-P	0.362 ± 0.147	0.177 ± 0.051	0.118 ± 0.032
	(b) SUVA (mg C L^{-1} m^{-1})		
SD-E	2.36 ± 0.42	3.79 ± 0.52	4.37 ± 0.32
SD-H	4.47 ± 0.89	5.65 ± 0.93	4.71 ± 1.16
SD-P	2.43 ± 1.28	3.52 ± 1.13	4.12 ± 0.58

At both the Stordalen and Storflaket mires, peat plateaus are surrounded by lower hollows with relatively high water tables, or that are connected to aquatic ecosystems, such as fens or small streams. Where subsidence of the peat plateau has occurred in Stordalen, palsa plateau soils have been inundated, with increased hydrological connectivity to aquatic systems, particularly fens (Malmer et al., 2005; Klaminder et al., 2008). Hence the influence of permafrost thaw on DOC export to freshwaters will be greatly influenced by the geomorphic changes that result, whether active layer deepening alone or including surface subsidence. By comparing catchments with and without permafrost peatlands, as well as investigating permafrost plateau runoff and changes in DOC at fen sites, Olefeldt and Roulet (2014) found that as "collapse fens" continue to expand with permafrost thaw at Stordalen, catchment DOC and its aromaticity will increase. However, the study also found it was likely that an increase in DOC export would result from increased hydrological conductivity and high DOC production from recent fen vegetation, as opposed to resulting strictly from the release or degradation of permafrost DOC stores (Olefeldt and Roulet, 2014). Our results support this conclusion, especially in the Stordalen mire where extractable DOC in the permafrost was equivalent to or less than DOC extracted from the overlying active layer.

CONCLUSIONS AND IMPLICATIONS

Our estimate of potential DOC export from Stordalen permafrost over the next 50 years was 0.28 g C m^{-2}, with a corresponding annual export estimate of 0.0056 g C m^{-2} yr^{-1} or 0.1% of the total annual DOC export from the Stordalen peatland complex; hence, the release of potentially readily leachable DOC from thawing permafrost layers is small in magnitude. Potentially leachable DOC from permafrost layers was generally not aromatic or recalcitrant compared with DOC from overlying active layers.

There were important differences in permafrost conditions between the Stordalen and Storflaket mires, specifically in OM and ice content. The relatively high ice content and low organic matter in Stordalen permafrost makes the palsa plateau in that mire relatively susceptible to surface subsidence as permafrost thaws. Conversion of palsa areas to wet or fen areas within Stordalen has been well documented, and our results indicate that this process is likely to continue. In contrast, the Storflaket permafrost had relatively low ice content, and thaw at that mire may continue to be dominated by active layer deepening. Because of the relatively high concentration of extractable DOC in near-surface permafrost at Storflaket, that palsa mire could become a more significant source of DOC to freshwaters as the active layer deepens.

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