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Des défis du Nord au Sud

Geochemistry of the active layer and permafrost in northwestern Canada: from measurements to Quaternary stratigraphy

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ABSTRACT
The amount and distribution of soluble ions and salts in active layer and permafrost provide a tool to assess present and past changes in active layer thickness, and chemical weathering regimes in a given landscape. This short paper summarizes various methodologies used for the extraction of soluble salts and ions in the active layer and permafrost, and then tests two methods from two sites in northwestern Canada. The results are discussed with respect to the extraction techniques, the timing of permafrost aggradation following sediment deposition (i.e., syngenetic vs epigenetic permafrost) and the history of permafrost (i.e., relict active layer vs residual thaw layer).

RÉSUMÉ
La quantité et la distribution des ions et des sels solubles dans la couche active et le pergélisol constituent un outil pour évaluer les changements passés et présents de l'épaisseur de la couche active et des régimes d'altération chimique d'un paysage. Cet article résume différentes méthodes utilisées pour l'extraction des ions et des sels solubles dans la couche active et le pergélisol, puis teste deux méthodes sur deux sites dans le nord-ouest du Canada. Les résultats sont discutés par rapport aux techniques d'extraction, à l'expansion du pergélisol suivant le dépôt des sédiments (pergélisol syngénétique vs épigénétique) et à l'histoire du pergélisol (couche active relique vs couche de dégel résiduelle).

1 INTRODUCTION

Knowledge on the extent of permafrost degradation to past warm climate intervals (i.e., Medieval Warm Period, early Holocene thermal maximum) is of importance to assess the potential magnitude of changes permafrost environments may experience as a result of climate warming or disturbance. J.R. Mackay was one of the first to recognize the use of Quaternary stratigraphy and identification of unconformities in frozen ground to infer past permafrost conditions, which evolved into the field of cryostratigraphy (e.g., Mackay et al. 1972; Mackay 1975; 1976). Paleo-thaw unconformity results from an increase in active layer thickness followed by the aggradation of permafrost and can be identified by distinct cryostructures in the relict active layer or the former residual thaw layer relative to the underlying permafrost (Murton and French 1994; French 1998; French and Shur 2010). A well-documented paleo-thaw unconformity is found in the western Arctic where the active layer thickened during the early Holocene warm interval (Mackay 1974; Burn 1997).

In the absence of cryostratigraphic observations along natural or anthropogenic exposures (i.e., coastal bluff, headwall of thaw slumps, mining section), alternative approaches must be used to assess the past permafrost conditions. One of these approaches consists of determining the concentration and distribution of soluble ions in permafrost. This stems from the pioneering work on permafrost geochemistry done in the regions of Fairbanks and Barrow, Alaska, USA (e.g., O'Sullivan 1966; Brown 1968; Péwé and Sellman 1973). These studies demonstrated that soluble cations (Ca^{2+}, Mg^{2+}, Na^{+}, K^{+}) and conductivity have a much lower concentration in sediments of interglacial ages (i.e., Holocene, Sangamonian) relative to glacial age loess (i.e., Wisconsin, Illinoian), with a step-like change at the transition between interglacial-glacial age sediments (Figure 1). Variability in the distribution of soluble ions in the Quaternary sediments was attributed to enhanced freshwater leaching during the warmer-wetter interglacial periods. This finding was used to infer that geochemical discontinuities in permafrost, combined with observations on cryostructures, can become a powerful tool to infer past conditions of permafrost (e.g., Mackay and Dallimore, 1992).

Recent studies of permafrost geochemistry focused on the active layer – permafrost boundary. For example, studies on the geochemical composition of the active layer and near-surface permafrost from the Mackenzie Delta (Kokelj and Burn 2005; Lacelle et al. 2014), Herschel Island (Kokelj et al. 2002) and the north slope of Alaska (Ping et al. 1998; Bockheim et al. 1998; Keller et al. 2007) have all shown that soluble ion content is lowest in the active layer but can increase ca. 2 to 7 fold in the top of permafrost. These studies have been conducted in fine-grained material, which can contain significant amounts of unfrozen water at temperatures below 0°C due to their higher specific surface area (Anderson et al. 1973; Williams and Smith 1989). As such, the increase in soluble ions in the top of permafrost has been attributed to leaching in the active layer and the downward transport of soluble ions during summer by water migrating along the negative soil thermal gradient. Therefore, the distribution of soluble ions in the top of permafrost may be used to infer decadal to centennial stability of the active layer and associated growth of the transient layer (e.g., Kokelj and Burn 2003; Lacelle et al. 2014).

The amount and distribution of soluble ions and salts in active layer and permafrost provide a tool to assess present and past changes in active layer thickness, and chemical weathering regimes in a given landscape (e.g., Keller et al. 2007; Lacelle et al. 2014). However, results and associated interpretations of the distribution and concentration of soluble salts and ions in the active layer and permafrost are largely dependent on the methodological approach used to extract the ions, and how the data are portrayed. Further, the extent to which this approach can be used to identify past permafrost conditions is unclear. This study first reviews various methodologies used for the extraction of soluble salts and ions in the active layer and permafrost, and then tests two different methods from two sites in northwestern Canada. The results are discussed with respect to the extraction techniques, the distribution with depth of ions and salts of different solubility, the timing of permafrost aggradation following sediment deposition (i.e., syngenetic vs epigenetic permafrost) and the history of permafrost (i.e., relict active layer vs residual thaw layer). Although the geochemistry of massive ice bodies has been used to infer the source of water and the origin of the ice (e.g., Mackay and Dallimore, 1992; Fritz et al. 2011), analyses of massive ice are not discussed here.

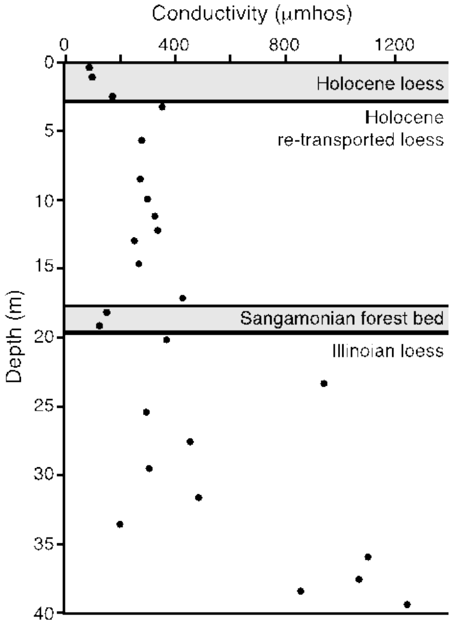

Figure 1. Conductivity, sediment type and age at Eva Creek Gold Placer mining cut, Fairbanks, Alaska (modified from Péwé and Sellman 1973).

2 REVIEW OF SAMPLING AND EXTRACTION TECHNIQUES

A variety of methods are used to sample and analyze the geochemistry of soil water and pore water in the active layer and permafrost, including the soluble salts and ions. Below, we review some of the most commonly used methods.

2.1 Direct sampling of soil water

Soil water from saturated zones in the active layer can be collected using a piezometer or shallow well. The piezometer, consisting of a hollow steel or PVC pipe, is installed in the soil with a screen mesh section placed at the required sampling depth. The groundwater can be hand-pumped to the surface directly into a sample vial through a Teflon tube (i.e., Lacelle et al. 2008). Unsaturated zone porewaters can be sampled using a suction lysometer (i.e., Carey and Quinton 2005). Alternatively, Rhizons samplers may be used in saturated and unsaturated soils to collect the small volume of pore water (i.e., Newman et al. 2015).

Although not commonly used in continuous permafrost regions, direct sampling of soil water from the thawed active layer has the advantage of characterizing the *in situ* geochemical conditions of the water, calculating the saturation of different minerals and assessing the evolution of the carbonate system, including P_{CO2} in the soil water, a key component to the C-cycle in the active layer (e.g., Lacelle et al. 2008).

2.2 Extraction of pore water

The geochemistry of active layer and permafrost is commonly determined by indirect sampling of the water. Soil sampling pits are excavated in the active layer and underlying permafrost samples may be collected with a Cold Region Research and Engineering Laboratory (CRREL) core barrel and the soil samples are brought to the laboratory (e.g., Kokelj et al. 2002; Kokelj and Burn 2005). Extraction of porewaters is best done by decanting, squeezing, or centrifuging the sample. For the samples that are super-saturated, the soil water is readily collected by these methods. For under-saturated samples a known amount of deionized water is added to the sample to reach saturation (i.e. ca. 1:1 soil water ratio; Janzen, 1993), the sample is agitated for a few hours and the excess water is collected. Both the filtered super-saturated and leachate water are then analyzed for geochemical composition and ionic concentrations are corrected to field moisture content or to unit weight of dry soil.

2.3 Soil : water extractions

A similar approach to the extraction of pore water is the soil-water extraction technique. The active layer and permafrost samples are first brought to the laboratory and are oven dried. To determine the most efficient and complete extraction method (total soluble ions and salts), various soil-water ratios can be tested (1:1, 1:5, 1:10, 1:25, etc). The soil-water mixture is shaken for 1 hour, centrifuged, and the leachate water decanted, filtered and analyzed for its ionic content. Figure 2 shows the results for a salt-rich permafrost sample collected near Navy Road (Inuvik, NT, Canada) using different soil-water ratios. Based on the results, soluble ions at this site are completely leached with a soil-water ratio > 1:5. Soils from other regions may require different soil-water ratios due to differing salt concentrations. It should be noted that the pore water and soil:water extraction methods may misrepresent cation concentrations due to cationexchange capacity, which is enhanced with the addition of low-salinity deionized water.

2.4 Acid extractions

For minerals that are insoluble at room temperature and atmospheric P_{CO2} (i.e., carbonates), soil samples may be leached and digested with various acids. Some of the most common procedures (e.g., Blum et al. 2002; Keller et al. 2007; Conklin 2014) include initial leaching with 1M HNO_3 at room temperature for 20 hours to dissolve easily soluble minerals (i.e., most carbonates, and minor amount of phosphates and sulfides). Subsequently, concentrated HNO_3 at 150°C for 3 hours is used to partially digest less soluble but still potentially weatherable minerals such as biotite, plagioclase, potassium feldspar, sulfides, and some oxides and clays. However, considering that a variety of acid extraction techniques are employed, the reader should consult the literature on this topic to determine which one is best for their needs.

2.5 Representing soluble salt and ion content in soils

To calculate the soluble salt and ion content in soils, the soil bulk density and ice content are required. Soluble salt contents (SSC) in the soil (expressed as equivalents m^{-2}) are calculated by summing over the required soil column the product of the salt concentration (C, meq kg^{-1}), the horizon thickness (T, m), the soil bulk density (BD, g cm^{-3}), and the weight fraction of the >2 mm soil fraction and/or ice (CF):

[1] $SSC = C \times BD \times T \times (1-CF)$

This equation assumes that all of the soluble salts and ions in the soil are held in the <2 mm soil fraction. Using this equation, the soluble salt and ion content in soils can be calculated for various soil depths.

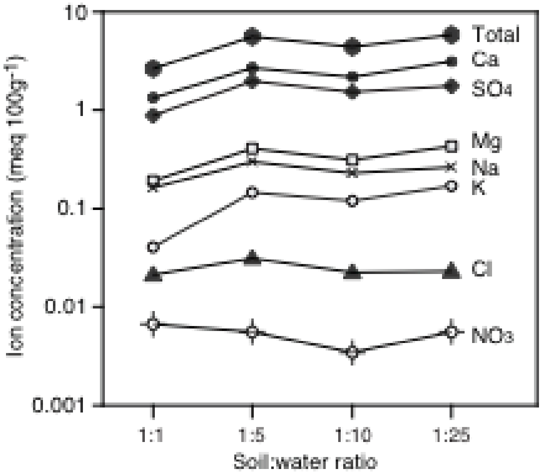

Figure 2. Graph of soluble ion concentrations measured at different soil-water ratios. The permafrost sample (150 cm depth) is from a hummock near Navy Road, Inuvik, NT, Canada.

3 GEOCHEMISTRY OF ACTIVE LAYER AND PERMAFROST IN NORTHWESTERN CANADA

The Peel Plateau is an ice-rich hummocky moraine landscape that extends along the eastern margins of the Richardson Mountains (Catto 1996). The surface sediments consist of glacial sediments deposited during the Late Pleistocene advance and retreat of the Laurentide Ice Sheet (Duk-Rodkin and Hughes 1992; Lacelle et al. 2013). Climate data obtained from the Inuvik and Fort McPherson meteorological stations indicates a mean annual air temperature of –7.3°C, with the region receiving <250 mm in annual precipitation. With mean annual temperatures > –2.5°C, permafrost is relatively warm (O'Neill et al. 2015).

In summer 2012 and 2013, active layer and near-surface permafrost samples were collected at four sites on the Peel Plateau and six sites in Mackenzie Delta region. Here we present the results from two sites on the Peel Plateau: i) CB slump: 67°10'N, 135°44'W, 595m; ii) Wilson slump: 67°13'N, 135°47'W, 610m. Active layer samples were collected at every 4 cm and placed in a sealed plastic bag. Permafrost samples were collected with a hand-held Hilti power head with 3.5'' diameter diamond bit barrel by coring horizontally into the headwall of the slumps (Figure 3B). All samples were weighed at the Aurora Research Institute laboratory (Inuvik, NT). When supernatant water was present in the active layer sample, the conductivity of the pore water was measured using a SympHony 3P90MS multimeter and the values were then corrected to account for the amount of water in each sample. The melt water from ice-bearing permafrost was extracted, filtered, and conductivity was determined using the SympHony meter and $\delta^{18}O$ and δD was determined using a LGR liquid isotope analyzer. Gravimetric water content (GWC) was calculated for all samples after drying the wet sediment at 105^oC for a minimum of 24 hours. The total soluble ions in the active layer and permafrost were subsequently extracted using a 1:10 soil-water ratio. Samples were shaken for an hour, centrifuged, measured for conductivity and filtered prior to determination of cations and anions concentrations. The leachate conductivity and ionic values were corrected to account for the soil-water ratio.

3.1 CB slump

In late summer 2012, the depth of the active layer was 48 cm. Based on changes in cryostructures observed in the headwall of the slump (from reticulate to suspended), the depth of the paleo-thaw unconformity was inferred at 115 cm (Fig. 3). The GWC ranged from 35 to 170%. With the exception of the O horizon, GWC is lowest in the active layer and relict active layer (<60%) and highest at the top of permafrost and below the paleo-thaw unconformity. $\delta^{18}O$ measurements were only made on the permafrost below the paleo-thaw unconformity where sufficient water was recoverable for analysis. The $\delta^{18}O$ values of the ground ice are highest immediately below the paleo-thaw unconformity (ca. –26‰) and vary between –28.7 and –27.3‰ below it. These values are similar to other sites in northwestern Canada where it has been suggested that the ground ice formed during Late Pleistocene and has not experienced thaw since (e.g., Mackay and Dallimore, 1992; Lacelle et al. 2004; Murton et al. 2004; Fritz et al. 2011).

Like $\delta^{18}O$, pore water conductivity could only be measured in the permafrost below the paleo-thaw unconformity. The pore water conductivity values follow a similar trend to the GWC. Pore water conductivity is highest immediately below the paleo-thaw unconformity (2.0 dS cm^{-1}) and then varies from 1.1 to 1.9 dS cm^{-1}. The conductivity values determined from leachate (1:10 soil-water ratio) is 3 to 6 times higher than the pore water values. Leachate conductivity values are lowest in the active layer (1.2-3.4 dS cm^{-1}) and increases to 3.3 dS cm^{-1} at the top of permafrost. Leachate conductivity values progressively increase from the top of permafrost to the top of the paleo-thaw unconformity (115 cm depth). Below this depth, leachate conductivity values range from 5.3 to 7.2 dS cm^{-1}. The dominant soluble ions in the active layer and permafrost are SO_4^{2-}, Ca^{2+} and Mg^{2+}. Their concentration is lowest in the active layer (<1000 meq 100 g^{-1} soils) and highest in the permafrost below the paleo-thaw unconformity (>1000 meq 100 g^{-1} soils). Cl^- and NO_3^-, which are very soluble, have the lowest concentration in the active layer and permafrost.

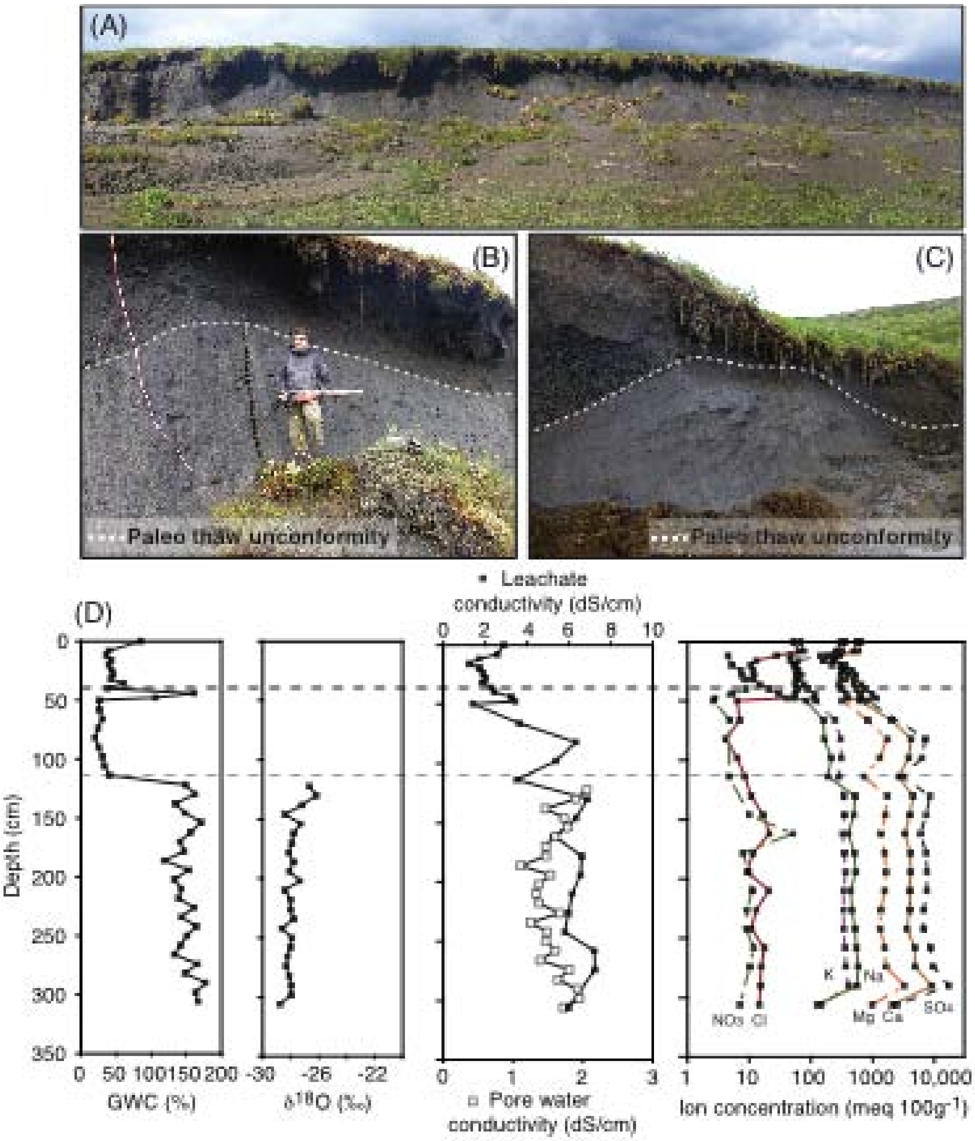

Figure 3. A) Field photograph (July 2012) showing headwall of the CB slump on the Peel Plateau, NT, Canada. B-C) Photographs of two sections of the headwall showing a change in cryostructures, from reticulate to suspended. The contact between the two is interpreted as representing the early Holocene paleo-thaw unconformity. D) Gravimetric water content (GWC), $\delta^{18}O$, pore water and leachate (1:10 soil-water ratio) conductivity and leachate ion concentration in the active layer and shallow permafrost. The thick dashed line represents the active layer and the thin dashed line represents the paleo-active layer.

3.2 Wilson slump

In summer 2012, the active layer thickness in the undisturbed terrain upslope of the Wilson slump was ca. 45 cm. Based on the observed change from reticulate to suspended cryostructures, the depth of the paleo-thaw unconformity was inferred to be at 175 cm (Fig. 4). At this site, the GWC ranged from 30 to 295% with peaks occurring at 80 and 192 cm depths (295% and 200%, respectively). The GWC is lowest within the active layer (<50%). Unlike at the CB slump site, there is little difference in GWC above and below the paleo-thaw unconformity. The $\delta^{18}O$ values of the ground ice show a progressive decrease from the bottom of the active layer (–22‰) to below the paleo-thaw unconformity where values remain near –29‰.

Pore water conductivity could only be measured in the permafrost. Pore water conductivity fluctuates throughout the uppermost 3 m of permafrost, from 0.2 to 2.5 dS cm^{-1}, the latter occurring as peaks at 80 and 192 cm depth, which corresponds to GWC maximum values. Leachate conductivity values are 2 to 9 times higher then those from the pore water. Leachate conductivity is lowest in the active layer (<3.5 dS cm^{-1}) and highest in the permafrost below the paleo-thaw unconformity (>5.1 dS cm^{-1}). Between the top of permafrost and the top of the paleo-thaw unconformity, leachate conductivity values have intermediate values, fluctuating between 3.8 and 6.5 dS cm^{-1}, which contrast with the CB slump site. The dominant soluble ions in the active layer and permafrost are SO_4^{2-}, Ca^{2+} and Mg^{2+}. Their concentration is lowest in the active layer (<2500 meq 100 g^{-1} soils) and highest below the paleo-thaw unconformity (>5000 meq 100 g^{-1} soils). Cl^- and $NO3^-$ have the lowest concentrations and shows little variation with depth.

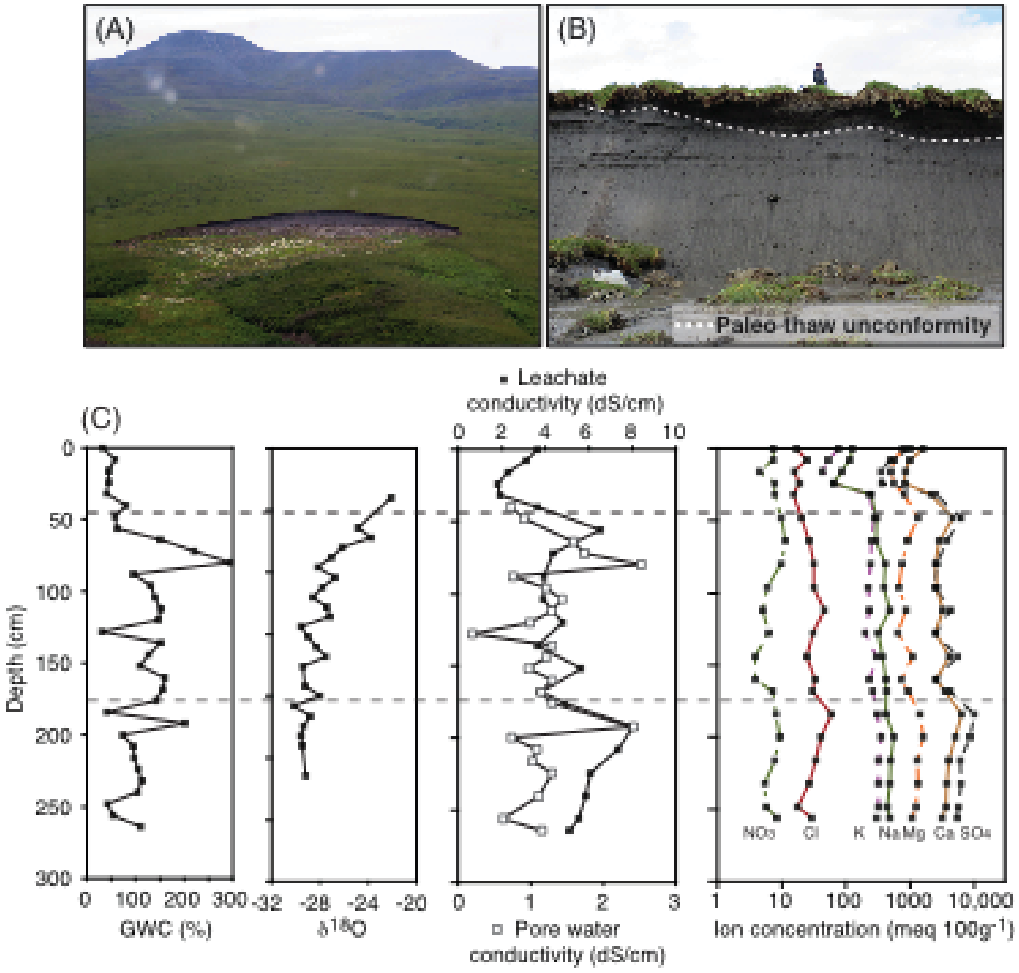

Figure 4. A) Field photograph (July 2012) showing headwall of the Wilson slump on the Peel Plateau, NT, Canada. B-C) Two sections of the headwall showing a change in cryostructures, from reticulate to suspended. The contact between the two is interpreted as representing the early Holocene paleo-thaw unconformity. D) Gravimetric water content (GWC), $\delta^{18}O$, pore water and leachate (1:10 soil-water ratio) conductivity and leachate ion concentration in the active layer and shallow permafrost. The thick dashed line represents the active layer and the thin dashed line represents the paleo-active layer.

4 DISCUSSION

4.1 Effect of extraction technique

Soluble salts and ions in the active layer and shallow permafrost were determined using two different extraction techniques: i) pore water analysis, a method derived from studies on soil salinity developed in the context of agricultural soil studies (e.g., Conklin 2014); and ii) leachate analysis using a 1:10 soil-water ratio, the latter reflecting total soluble ions (Figure 2). Using the pore water analysis technique, the soluble ion concentration is highly dependent on the amount of water present in the sample, with more soluble ions being dissolved under higher water content (Figure 5). As such, the adaptation of the pore water analysis method to permafrost studies may underestimate the total soluble ions associated with the soil.

Based on the active layer and permafrost samples used in this study, the pore water analysis technique results in conductivity values being 2 to 9 times lower than the 1:10 soil-water extraction technique. As such, this method-dependent difference in soluble salt and ion concentration may lead to different interpretations of the results, especially on the abundance and distribution of soluble ions in active layer and permafrost profiles. For example, at the CB and Wilson slump sites, both soluble ion extraction methods show increased conductivity in the top of permafrost and just below the paleo-thaw unconformity; however the depth and magnitude of solute enrichments varies (Figures 3 and 4). At the Wilson slump, leachate conductivity shows solute enrichment at 56 cm depth (6.5 dS cm^{-1}), whereas it is found at 80 cm depth (2.5 dS cm^{-1}) based on pore water conductivity.

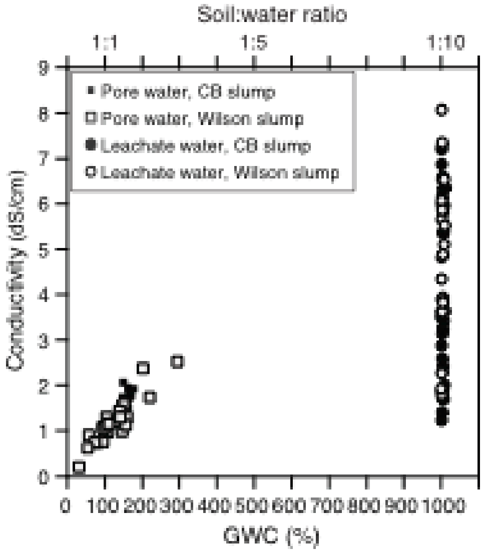

Figure 5. Graph of conductivity measured in the pore water and leachate water (1:10 soil-water ratio). Pore water conductivity is related positivity to field water content. Variation in conductivity of the leachate water provides a standardization that allows for comparison between samples.

4.2 Source of soluble salts and ions

Relations between the various ion species suggest that the source of SO_4, Ca and Mg originates from the dissolution of gypsum or Ca+Mg sulfates (i.e., Malone et al. 2013). Conversely, the Na/Cl ratio in the active layer at both sites approaches the sea-salt ratio (R_{sea}=0.86; Kleene et al. 1986), however the Na/Cl ratio is much higher in the permafrost suggesting an alternate source of Na. Finally, NO_3 shows little variations between the active layer and permafrost due to its high solubility. This topic will be explored in greater details in a follow-up manuscript.

4.3 Effect of history of permafrost on geochemical conditions

On the Peel Plateau, radiocarbon dating of horse mandible from a site nearby both slumps (Lacelle et al. 2013) and *Bison priscus* remains nearby Tsiigehtchic (Zazula et al. 2009) constrains the presence of the Laurentide Ice Sheet between 18,500 and 13,500 ^{14}C cal BP, which deposited ca. 10-50 m of glacial sediments (Duk-Rodkin and Hughes, 1992). Permafrost in the newly deposited glacial sediments is likely syngenetic and formed during the Allerød-Bøllering and Younger Dryas periods. During the early Holocene warm interval, near-surface permafrost experienced deep thaw, with active layer thickness approaching 2 to 2.5 times that of today, as evidenced from the change in cryostructures in the headwall of slumps (e.g., Burn 1997). Following this warm interval, permafrost subsequently aggraded to present day conditions. Based on this history of permafrost and the ground ice content at both sites (Figures 3 and 4), Type F permafrost is found on the Peel Plateau (e.g., Jorgenson et al. 2010), which corresponds to ice-rich syngenetic permafrost overlain by a layer of thawed and refrozen soils containing lower ice content. At the CB and Wilson slump sites, total soluble ions concentration is highest below the paleo-thaw unconformity as a result of the syngenetic permafrost conditions that prevented leaching in the newly deposited glacial sediments and preserved soluble ions. We suggest that soluble salt and ions in type F permafrost may be 1-2 orders of magnitude higher below the maximum historical depth of thaw (i.e., below the paleo-thaw unconformity) relative to the active layer. In the case of epigenetic permafrost, one is not to expect similar soluble ion gradients across the paleo-thaw unconformity as substantial leaching could have occurred in the soils prior to the aggradation of permafrost. Future studies should investigate the distribution and concentration of soluble ions in Type A-E permafrost of the Jorgenson et al. (2010) classification.

According to French (1998) and French and Shur (2010), permafrost above the historical maximum depth of thaw is classified either as: i) a *relict active layer*, if during its development the depth of seasonal freezing and thawing reaches the top of permafrost, or ii) a *residual thaw layer* if the depth of seasonal freezing remained above the permafrost table. Assuming all other conditions remain the same, the frequency and magnitude of freshwater leaching is likely different in a relict active layer compared to in a residual thaw layer. For example, Lacelle et al. (2008) demonstrated that chemical weathering is highest at the base of the active layer, where water and P_{CO2} availability is higher, which leads to increased leaching and weathering; this could be enhanced within a residual thaw layer that remains perennially unfrozen for extended periods of time.

Figure 6 compares the distribution and concentration of total soluble ions at the CB and Wilson slumps. Although similar abundance and vertical trends are observed within the active layer, differences are observable between the top of permafrost and the paleo-thaw unconformity and also below this unconformity. The low total soluble ions throughout the active layer is attributed to cumulative seasonal freshwater leaching since the time of deposition of the glacial sediments (i.e., > 15,000 years). A first peak in total soluble ions is observed just below the top of permafrost, corresponding to a peak in ice content, and these likely developed by downward migration of water and leached soluble ions during the summer along a negative thermal gradient over decadal to centennial time-scale (e.g., Cheng et al. 1983; Mackay 1983; Kokelj et al. 2005). Between the top of permafrost and the paleo-thaw unconformity, the total soluble ions at the CB slump site increases from the base of the active layer to the top of the paleo-thaw unconformity; whereas at the Wilson slump, total soluble ions have a slightly lower abundance and vary little with depth within that zone. Below the paleo-thaw unconformity, the total soluble ions at both sites is greater than 400 mg 100 g^{-1} soils with a peak in abundance just below the thaw unconformity. However, the peak in abundance is ca. 1.2 times higher at the Wilson site (650 mg 100 g^{-1} soils). The distinct patterns at both sites are tentatively interpreted as the CB slump site developing a relict active layer, whereas at the Wilson slump, where the unconformity occurs at much greater depth, resulted in the development of a residual thaw layer; although the timing and duration of this layer is unknown. The demonstrate the effect of a residual thaw layer on soluble salts and ions in permafrost, future studies should investigate modern sites where the depth of seasonal freezing remained above the permafrost table. Given the increase in winter air temperature and snow cover, the development of residual thaw layers in continuous permafrost may become more extensive.

5 CONCLUSIONS

The following conclusions can be reached:

1. Different extraction techniques of soluble salts and ions in active layer and permafrost can yield different patterns of variation with depth. The use of a common extraction technique of soluble salts and ions in the active layer and permafrost is likely required to make results from different studies comparable.
2. Combined with observations of cryostructures, analyses of pore water and leachate soluble ions can be effective tools to infer past thaw depths in syngenetic permafrost. However, their application to epigenetic permafrost remains unclear.

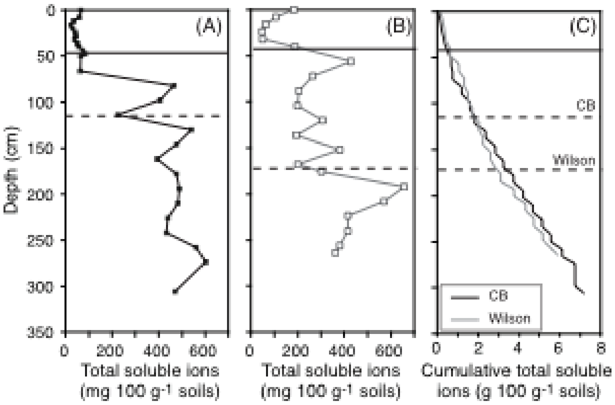

Figure 6. Total soluble ions at the (A) CB slump, (B) Wilson slump. (C) Cumulative total soluble ions at both sites.

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