Review: Impacts of permafrost degradation on inorganic chemistry of surface fresh water

Published version:

DOI:10.1016/j.gloplacha.2017.11.017

Review: Impacts of permafrost degradation on inorganic chemistry of surface fresh water

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Abstract

Recent studies have shown that climate change is impacting the inorganic chemical characteristics of surface fresh water in permafrost areas and affecting aquatic ecosystems. Concentrations of major ions (e.g., Ca^{2+}, Mg^{2+}, SO_4^{2-}, NO_3^-) can increase following permafrost degradation with associated deepening of flow pathways and increased contributions of deep groundwater. In addition, thickening of the active layer and melting of near-surface ground ice can influence inorganic chemical fluxes from permafrost into surface water. Permafrost degradation has also the capability to modify trace element (e.g., Ni, Mn, Al, Hg, Pb) contents in surface water. Although several local and regional modifications of inorganic chemistry of surface fresh water have been attributed to permafrost degradation, a comprehensive review of the observed changes is lacking. The goal of this paper is to distil insight gained across differing permafrost settings through the identification of common patterns in previous studies, at global scale. In this review we focus on three typical permafrost configurations (pervasive permafrost degradation, thermokarst, and thawing rock glaciers) as examples and distinguish impacts on (i) major ions and (ii) trace elements. Consequences of warming climate have caused spatially-distributed progressive increases of major ion and trace element delivery to surface fresh water in both polar and mountain areas following pervasive permafrost degradation. Moreover, localised releases of major ions and trace elements to surface water due to the liberation of soluble materials sequestered in permafrost and ground ice have been found in ice-rich terrains both at high latitude (thermokarst features) and high elevation (rock glaciers). Further release of solutes and related transport to surface fresh water can be expected under warming climatic conditions. However, complex interactions among several factors able to influence the timing and magnitude of the impacts of permafrost degradation on inorganic chemistry of surface fresh water (e.g., permafrost sensitivity to thawing, modes of permafrost degradation, characteristics of watersheds) require further conceptual and mechanistic understanding together with quantitative diagnosis of the involved mechanisms in order to predict future changes with confidence.

1. Introduction

Average atmospheric temperature has increased globally over the last decades and, as a response, the cryosphere is changing (Fountain et al., 2012). Permafrost, a component of the cryosphere, is widespread in the Arctic and Antarctica, and in cold mountains, including densely populated areas of the European Alps and Asian mountain ranges (Gruber, 2012). Permafrost interacts with climate (Walter Anthony et al., 2006; Schuur et al., 2015), hydrology (e.g., Liljedahl et al., 2016; Yang et al., 2017), ecosystems (Jorgenson et al., 2001; Vonk et al., 2015), geophysical processes (e.g., Gruber et al., 2004; Gruber and Haeberli, 2009), and human systems (Nelson et al., 2002; Harris et al., 2009).

Recent reviews have focused on the impacts of permafrost warming and degradation on river biogeochemistry (Frey and McClelland, 2009) and aquatic ecosystems (Vonk et al., 2015) in the Arctic. Notwithstanding, local and regional modifications of water hydrochemistry due to permafrost degradation have been reported from many locations, globally. Given the sparsity of data available, understanding and analysing permafrost degradation impacts on inorganic chemistry of surface fresh water will benefit from identifying common patterns in existing studies. The present review thus aims to distil insight gained across differing permafrost environments and configurations globally.

Following a brief background section, we distinguish three typical example configurations of permafrost thaw. For those, we review impacts of permafrost degradation on major ions (e.g., Ca^{2+}, Mg^{2+}, SO_4^{2-}, NO_3^-) and on trace elements (e.g., Ni, Mn, Al, Hg, Pb).

2. Permafrost: definition and main characteristics

Permafrost is defined as ground (soil and/or rock, including ice and organic material, plus air/gas in unsaturated ground) that remains at or below 0 °C for at least two consecutive years (Muller, 1943). Thus, permafrost is a thermal phenomenon and it can, but does not need to, contain water or ice. Most permafrost areas experience seasonal thaw, during which ground surface temperature rises above 0 °C. The layer of ground that thaws on a seasonal basis is called “active layer” (ACGR, 1988). The depth of permafrost (permafrost base) is determined by the geothermal heat flux, subsurface properties, and the long-term surface temperature. The upper limit of permafrost (permafrost table) is largely controlled by surface temperatures during summer and near-surface ground properties (Williams and Smith, 1989; French, 2007). The difference in depth between the permafrost table and the permafrost base is the permafrost thickness (see Figure 2 in ACGR, 1988, source: https://ipa.arcticportal.org). Permafrost regions are often divided into a continuous permafrost zone, where more than ~90% of the surface area is underlain by permafrost, a discontinuous zone, of which ~10% to ~90% is underlain by permafrost, and a zone of isolated patches of permafrost, where permafrost occurs under less than ~10% of the ground. The discontinuous zone is often divided into extensive discontinuous permafrost (~90% to ~50%) and sporadic discontinuous permafrost (~50% to ~10%, Heginbottom et al., 1995).

The global permafrost region (exposed land surface below which some permafrost can be expected) is estimated to be $22 \pm 3 \times 10^6$ km^2, approximately 17% of the global land area (Gruber, 2012). Zhang et al. (1999; 2003) estimated that permafrost could underlie nearly 25% of the exposed land area of the Northern Hemisphere. Despite occupying only 0.36% (49,800 km^2) of the Antarctic region, permafrost is estimated to be present beneath all ice-free terrain, except at the lowest elevations of the maritime Antarctic and sub-Antarctic islands (Vieira et al., 2010). Although with increased spatial heterogeneity imposed by topography, permafrost is also widespread in many mountain ranges such as the European Alps (e.g., Boeckli et al., 2012), the Hindu Kush Himalaya (e.g., Gruber et al., 2017), and the Andes (e.g., Brenning, 2005).

The aggradation and degradation of permafrost usually occur in association with a change of mean ground temperature due to microclimatic (ground cover) and climatic changes (Williams and Smith, 1989). *Permafrost degradation* refers to the decrease of volume of a body of permafrost, whereas *permafrost thaw* or *thawing* is used to refer to increasing liquid water content and other changes of physical characteristics in response to energy input within a body of rock or soil while still satisfying the definition of permafrost. The term *thawed permafrost* indicates ground that no longer is permafrost. Global warming over the past several decades is now large enough that regional climate change is emerging above the noise of natural variability (Hansen and Sato, 2016). Despite the small magnitude of warming relative to weather fluctuations, effects of the warming already have notable environmental, social and economic impacts. In this context, the degradation of permafrost is seen as a major challenge in the current discussion of globally rising air temperatures (IPCC, 2013), because of the possible effects on climate (e.g., Harden et al., 2012; Schuur et al., 2015), infrastructure stability (e.g., Nelson et al., 2001; Streletskiy et al., 2012), hydrology (e.g., Liljedahl et al., 2016; Walvoord and Kurylyk, 2016), natural hazards in mountainous areas (e.g., Gruber et al., 2004; Harris et al., 2009), and aquatic biogeochemistry and ecosystems (e.g., Frey and McClelland, 2009; Vonk et al., 2015).

3. Typical configurations of permafrost thaw

We focus on three typical configurations in which permafrost degradation can impact the inorganic chemistry of water bodies. While not exhaustive, these examples currently reflect major interests in this research field:

i) Pervasive permafrost degradation, leading to spatially-distributed modifications in groundwater-surface water connectivity and causing volumes of permafrost to thaw and become subject to weathering and leaching;

ii) thermokarst and associated localised mobilisations of sediments and solutes;

iii) rock-glacier thawing and related localised export of solute-rich water.

(i) Pervasive permafrost degradation. Permafrost thaw affects hydrology through increasing hydrologic permeability with ice loss in pores (Bense et al., 2009; Kurylyk et al., 2014) and through the release of water stored in frozen material. Permafrost is commonly an aquitard (Woo, 2012; Kane et al., 2013), supporting perched water tables and increasing near-surface soil moisture (Gorbunov, 1978; Ishikawa et al., 2005). Permafrost thaw, through talik growth and higher permeability of partially-frozen material, changes the hydrologic connectivity in a catchment and affects the rates and amounts of flow along differing paths (Hinzman et al., 2005; Rogger et al., 2017), which often produce water with distinct geochemical signatures (Clark et al., 2001).

Due to thaw and degradation, new hydrological pathways in permafrost are expected to enhance the interaction between mineralised groundwater and surface water, driving transitional permafrost environments from surface-water dominated systems to groundwater-dominated systems (e.g., Frey and McClelland, 2009). Indeed, recent studies have attributed increases in groundwater contributions to base flow of several rivers in permafrost watersheds to permafrost degradation (Walvoord and Striegl, 2007; St. Jacques and Sauchyn, 2009; Kolosov et al., 2016).

Most near-surface effects of permafrost thaw stem from increasing Active Layer Thickness (ALT), which has been shown to occur at large scale, across entire landscapes (e.g., Åkerman and Johansson, 2008). Recently, Luo et al. (2016) investigated the spatiotemporal characteristics of ALT across the Northern Hemisphere from 1990 to 2015. Significant trends of increasing ALT have been observed at approximately 43 % of the investigated 169 sites analysed. Regionally, increasing ALT have been found in North America (e.g., Oelke et al., 2004; Burn and Zhang, 2009), Asia (e.g., Frauenfeld et al., 2004; Oelke et al., 2004; Wu et al., 2012), Antarctica (e.g., Guglielmin and Cannone, 2012; Guglielmin et al., 2014), and Europe (e.g., Åkerman and Johansson, 2008; Harris et al., 2009), although variation exists due to local conditions.

Increasing ALT can expose newly-thawed soils to near-surface water and increase chemical fluxes from freshly-exposed strata (e.g., Keller et al., 2007; Keller et al., 2010). Furthermore, geochemical constituents previously sequestered within the near-surface permafrost (e.g., Kokelj and Burn, 2005) can be released.

Although spatially-distributed modifications in groundwater-surface water connectivity and increasing soil volumes subject to weathering and leaching are distinct processes associated with permafrost degradation, the impacts of these processes on the inorganic chemistry of surface water are not easily discernible. Indeed, both result in diffuse increases of geochemical fluxes into the receiving water bodies and only few studies distinguish the main sources of such fluxes (e.g., Keller et al., 2007, 2010; Lamhonwah et al., 2017).

(ii) Thermokarst. Permafrost warming and increasing ALT can cause self-reinforcing thaw of ice-rich ground leading to the development of thermokarst (Jorgenson and Osterkamp, 2005; Rowland et al., 2010). The term thermokarst includes a suite of processes and landforms that involve lowering, and often collapsing, of the land surface because of melting excess ground ice. Kokelj and Jorgenson (2013) classified the principal thermokarst types as: a) hillslope processes, including retrogressive thaw slumps, active layer detachment slides, and thermal erosion gullies; b) thaw lake processes, including lake expansion/reduction, drainage, and lake basin evolution; c) wetland processes, including peatland collapse and the development of bogs and fens. Thermokarst can also occur on active rock glaciers (details in (iii) Rock glacier thawing), where increased melting of permafrost ice can cause the formation of small thermokarst lakes (Kääb and Haeberli, 2001). Thermokarst is commonly observed throughout most permafrost regions including Canada (e.g., Watanabe et al., 2011), Alaska (e.g., Andresen and Lougheed, 2015), Russia (e.g., Manasypov et al., 2014), Mongolia (e.g., Sharkhuu, 1998), China (e.g., Wu et al., 2010), Antarctica (e.g., Gooseff et al., 2016) and the European Alps (e.g., Kääb and Haeberli, 2001).

Thermokarst development has the potential to alter the geochemical signature of surface water because it facilitates mobilisation of sediments and solutes previously sequestered in ground ice (Gooseff et al., 2009), causing localised and strong influences on hydrochemical systems (Malone et al., 2013). The effects of thermokarst are mostly local, but the transport of solutes into downstream aquatic ecosystems can sometimes impact large areas (e.g., Kokelj et al., 2013). Moreover, the formation of new features such as thaw ponds and lakes can modify the hydrochemical characteristics of large areas (e.g., Manasypov et al., 2014).

(iii) Rock-glacier thawing. Rock glaciers are distinctive and sometimes abundant geomorphological features of cold mountain regions (Barsch, 1996). They are slowly-flowing mixtures of rock debris and ice and formed by a continuum of processes from glacial to periglacial (Haeberli et al., 2006). The thawing of ice-rich talus slopes (cf., Gruber and Haeberli, 2009), although not studied in detail, is likely part of the same continuum. Rock glaciers are considered as the visible expression of long term creep of ice-rock mixtures under permafrost conditions (Humlum, 2000; Ikeda and Matsuoka, 2006; Berthling, 2011), and for this reason, they are often used for first-order mapping of permafrost distribution at coarse scales (e.g., Brenning, 2005; Cremonese et al., 2011; Schmid et al., 2015). Compared to glaciers, rock glaciers are believed to react more slowly to rising temperature because of the retardation of heat transfer through thick debris mantles (Barsch, 1996). During the cold season, ice is formed within rock glaciers, their “mass balance“, however, is difficult to determine. Emerging observations suggest the thermal and kinematic state of many rock glaciers is changing in concert with climate change (e.g., Roer et al., 2005; Haeberli et al., 2006; Scapozza et al., 2014). Many active (moving) rock glaciers loose some water due to melt during summer. This is because frequently, their tongues extent into microclimatic situations in which the moving ice core undergoes slow degradation. However, at decadal and longer time scales, due to global warming, accelerated melting of the ice within rock glaciers may represent an increasing hydrological contribution to downstream areas (Brenning, 2005). For this reason, active rock glaciers are generally considered to be sources of fresh water (Burger et al., 1999), especially in arid areas, small catchments, and during low flow conditions (e.g., Corte, 1976; Brenning, 2005; Azócar and Brenning, 2010; Millar et al., 2013; Rangecroft et al., 2015). Finally, melting of permafrost ice in active rock glaciers in particular, and also in ice-rich talus (cf., Gruber and Haeberli 2009), is able to increase the porosity of the sediments. In turn, this can increase the storage capacity, causing changes in the discharge pattern (Rogger et al., 2017).

The thaw of rock glaciers due to climate change has the potential to export enriched-solute fluxes and thereby modify the inorganic chemistry of downstream impounded surface waters (e.g., Ilyashuk et al., 2014) and streams (e.g., Thies et al., 2013).

Local and regional modifications of inorganic chemistry of surface fresh water following different configurations of permafrost thaw and degradation have been reported globally and are listed in Tab. 1. The impacts on major ions and trace elements are discussed in the following chapters.

4. Impacts on major ion content

4.1 Pervasive permafrost degradation

A strong divergence of inorganic solute concentrations between catchments with differing permafrost extents has been reported, whereby lower concentrations are associated with higher permafrost extent. This has been attributed to permafrost (a) inhibiting the infiltration of surface water into deep mineral soil strata, and (b) confining mineral-rich groundwater in the subpermafrost zone without hydrological connection to surface water (Frey and McClelland, 2009; Woo, 2012; Kane et al., 2013). For example, Frey et al. (2007a) analysed Total Inorganic Solutes (TIS, defined as sum of Ca^{2+}, K^{+}, Mg^{2+}, Na^{+}, Si, Cl^{-}, SO_4^{2-} and HCO_3^{-}) as a function of latitude in West Siberian streams and rivers in Russia, showing that TIS concentrations were 289 mg L^{-1} in permafrost-free watersheds compared to 48 mg L^{-1} on average in watersheds with permafrost (Fig. 1). Other studies highlighted differences in concentrations of major ions, such as HCO_3^{-} (Yukon River basin - Alaska, Striegl et al., 2005; Siberia - Russia, Yukon - Alaska, and Mackenzie - Canada, Tank et al., 2012), Ca^{2+} and Mg^{2+} (Caribou-Poker Creeks Research Watershed - Alaska, MacLean et al., 1999), Cl^{-}, SO_4^{2-}, Ca^{2+}, K^{+}, Mg^{2+}, and Na^{+} (Central Canadian sub-arctic - Canada, Rühland et al., 2003), Na^{+}, Ca^{2+}, Cl^{-}, and SO_4^{2-} (Central Mongolia, Szopińska et al., 2016), SO_4^{2-}, Ca^{2+}, Mg^{2+}, Na^{+}, K^{+}, and DIC (Dissolved Inorganic Carbon) (western Siberia - Russia, Pokrovsky et al., 2015), among areas with different permafrost distribution. In western Siberia, Pokrovsky et al. (2015) also provided evidence of the importance of plant litter and ground vegetation leaching as element sources comparing K^{+} concentrations as a function of latitude. Indeed, lower solute concentrations were measured in permafrost-bearing zones in spring, during intense plant litter leaching, due to lower biomass and primary productivity of forest-tundra and tundra biomes compared to the productive taiga zone of the western Siberia. Based on their results, Pokrovsky et al. (2015) hypothesised that, assuming a short-term climate warming scenario of hundreds of years, an increase in ALT will be capable to increase the export of major elements from transitional permafrost watersheds.

Increasing contributions of highly-mineralised groundwater, and increased contact (time and volume) between soil water and mineral surfaces have been proposed as the main drivers of ion enrichment in surface water of several areas subject to permafrost degradation. A recent investigation performed by Toohey et al. (2016) showed an increase in Ca^{2+}, Mg^{2+}, SO_4^{2-}, and Na^{+} in the Yukon River Basin (Alaska) in the period 1982-2014 (Fig. 2), attributing this evidence to altered hydrological flowpaths and increased weathering due to widespread permafrost degradation. Several other studies of Northern-Hemisphere high-latitude streams reported increasing concentrations of ions such as Ca^{2+} and SO_4^{2-} (Noatak National Preserve - Alaska, Stottlemyer, 2001), Ca^{2+} and Mg^{2+} (sub-Arctic - Sweden, Giesler et al., 2014), Ca^{2+}, Mg^{2+}, K^+, and Na^+ (Caribou-Poker Creeks Research Watershed - Alaska, Petrone et al., 2006, 2007), SO_4^{2-}, Ca^{2+}, Mg^{2+}, and alkalinity (Mackenzie basin - Canada, Tank et al., 2016), HCO_3^-, Cl^-, SO_4^{2-}, Ca^{2+}, and Mg^{2+} (Central Siberian Plateau - Russia, Kolosov et al., 2016), following permafrost degradation. Similar findings have been reported for streams and lakes of the Colorado Rocky Mountains (USA) over the last two-three decades, with increasing concentrations of Ca^{2+} and SO_4^{2-} (Baron et al., 2009; Caine, 2010; Todd et al., 2012), and Ca^{2+}, Mg^{2+}, and SO_4^{2-} (Mast et al., 2011).

Although increases in major ions concentrations of surface water have been found in several transitional permafrost areas, the causal link with permafrost degradation is not well established. Keller et al. (2007, 2010) and Douglas et al. (2013) proposed the use of Ca/Sr, Ca/Na, Ca/Ba, and $^{87}Sr/^{86}Sr$ trends in soils and streams for quantitatively assessing the effects of increasing ALT on mineral fluxes to surface water, where the parent material and soil geochemistry are variable with depth. For instance, geochemical trends point to carbonate concentrations in the ground increasing with depth (higher concentrations in the permafrost than in the active layer), due to decreasing frequency of thaw and hence weathering intensity. Correspondingly, a progressive increase of carbonate weathering signals during the summer season (especially in late summer) was found, associated with increasing thaw depth. New insights on mechanisms and sources of inorganic solutes in surface waters draining permafrost-affected soils can also be gained by using stable isotopes of inorganic solutes such as Ca (Bagard et al., 2013), Mg (Mavromatis et al., 2014, 2016), and Si (Pokrovsky et al., 2013). For example, Pokrovsky et al. (2013) analysed Si isotopic composition of large and small rivers, surface flow, interstitial soil solutions, plant litter and soils in Central Siberia, finding significant variations of the dissolved Si isotopic composition. These variations were linked to 1) the high concentrations of suspended silicate matter during the spring flood, 2) the progressive increase of ALT during summer accompanied by secondary mineral precipitation in surface layers and plant uptake of Si, and 3) the impact of the permafrost-hosted deep underground waters during winter. Under climate change, the resulting permafrost degradation may be the most important factor controlling the Si isotopic composition in the rivers.

Short-time increases in the depth of thaw are also capable to enhance mineral fluxes from permafrost terrain. Increases in major ion concentrations (mainly Ca^{2+}, Mg^{2+}, Na^{+}, SO_4^{2-}, and Cl^{-}) in surface water have been linked to active-layer thickening following strong thermal perturbations (warm summer conditions) and summer rainfall (Melville Island - Canada, Dugan et al., 2012, Lewis et al., 2012; Lafrenière and Lamoureux, 2013; Roberts et al., 2017) with flushing of solutes stored in near-surface permafrost (Melville Island - Canada, Lamhonwah et al., 2017) as well as to fire disturbance (Central Siberia - Russia, Parham et al., 2013).

In the McMurdo Dry Valleys (Antarctica) the presence of unusual surface-flow-seep features has been attributed to the potential melting of complex-origin sub-surface ice (e.g., excess ground ice, snow patches, refrozen precipitation that has accumulated in the subsurface, or buried glacier ice) in permafrost (Harris et al., 2007). Seeps showed a different geochemical signature in comparison to glacial meltwater, strongly enriched in solutes (Na^{+}, Ca^{2+}, Mg^{2+}, Cl^{-}, NO_3^{-}, SO_4^{2-} and HCO_3^{-}). These features associated with water tracks (Levy et al., 2011) might contribute to the desert hydrological cycle exporting solute-concentrated fluxes from sub-surface ice to surface water, enhancing the connectivity between the cryosphere and the hydrosphere in the Antarctic polar desert (Levy et al., 2011).

Future increases in air temperature, and rain, may enhance the release of major ions from solute-rich layers in near-surface permafrost. Indeed, several studies reported a solute enrichment at the base of the active layer and at the top of permafrost in different permafrost environments due to ion redistribution during thermally-induced moisture migration (Herschel Island - Canada, Kokelj et al., 2002; Northwest Territories - Canada, Kokelj and Burn, 2003, 2005; Lacelle et al., 2014), greater availability of water for chemical aqueous reactions where drainage is inhibited by subjacent permafrost (Yukon Territory - Canada, Lacelle et al., 2007; Devon Island - Canada, Lacelle et al., 2008), mineral dissolution reactions taking place in loamy layers slowly flushed by advective flow, with the reaction products transported upward by diffusion to surficial peat layers frequently flushed during summer (Ilulissat - Greenland, Jessen et al., 2014), physical modifications (landslides) of active layer geochemistry (Yamal Peninsula - Russia, Leibman and Streletskaya, 1997), and cryogenic concentration of salts during freezing and humidity-driven salt separation at the soil surface (McMurdo Dry Valleys - Antarctica, Levy et al. 2012).

Permafrost degradation can also increase the export of NO_3^- from soils to surface water due to deeper pathways that bypass the shallow organic soil horizons where most retention and removal of inorganic nitrogen occurs (Caribou Poker Creeks Research Watershed and Upper Kuparuk River Basin - Alaska, Harms and Jones, 2012). For instance, increased nitrification and decreased nitrate uptake due to enhanced water-inorganic soil and limited water-organic soil contacts have been demonstrated to lead to higher nitrate concentrations in surface water in permafrost environments. Jones et al. (2005) investigated three watersheds underlain by discontinuous permafrost in interior Alaska (Caribou Poker Creeks Research Watershed) over a four-year period. The results showed that deeper flow paths in areas of less extensive permafrost facilitated nitrate export, with the loss of permafrost that could also have released stored nitrogen inaccessible to terrestrial plant uptake, resulting in high nitrification and leaching loss rates. Petrone et al. (2006) followed up on this work and highlighted the sensitivity of nitrate fluxes to changes in active layer thickness, with higher concentrations associated with deeper movements of water into soils. The authors further demonstrated the role of the active layer as source of nitrate during the summer season, especially during summer storms, as also reported by Koch et al. (2013) in the Richardson Catchment (Alaska). Furthermore, McClelland et al. (2007) found a substantial increase in nitrate export from the Upper Kuparuk River (Alaska) between the late 1970s and early 2000s, attributing it to mechanisms potentially linked to warming and permafrost thaw.

Conversely, comparison of 96 rivers and streams in western Siberia found no differences in Dissolved Inorganic Nitrogen (DIN) concentrations linked to variations in permafrost extent (Frey et al., 2007b). Frey and McClelland (2009) attributed the difference in findings for western Siberia and Interior Alaska to the greater remineralisation (and subsequent nitrification) of organic nitrogen in the Alaskan watersheds (intermediate permafrost extent), in contrast with a limited remineralisation of organic nitrogen in western Siberia due to limited soil water saturation in peat-dominated terrains and enhanced denitrification due to wet conditions.

Deepening of flow paths can also lead to increasing concentrations of inorganic phosphate and silicate in soil water due to enhanced mineral weathering, which is the primary source of these inorganic nutrients

(Frey and McClelland, 2009, Pokrovsky et al., 2015). For instance, Hobbie et al. (1999) demonstrated that permafrost thawing along the MilkyWay stream in northern Alaska contributed 30% of inflowing phosphate into Toolik Lake, causing bottom-up trophic effects. Similarly, increasing concentrations of dissolved silicate with a decrease in permafrost coverage were found in Alaska (MacLean et al., 1999) and in western Siberia (Frey et al., 2007a). However, assuming future climate warming and increasing vegetation density in permafrost-bearing zones, transient uptake of nutrients during summer period may attenuate the expected increase in surface water concentrations linked to progressive involvement of thawed mineral horizons, as hypothesised by Pokrovsky et al. (2015) for Si dynamics in western Siberian rivers (Russia).

4.2 Thermokarst

Thermokarst disturbances in permafrost terrain such as retrogressive thaw slumps, thermal erosion gullies, active-layer detachment slides, and catastrophic thaw of frozen palsa peat bogs have been found to strongly modify major ion concentrations of surface water (mainly SO_4^{2-}, Ca^{2+}, Mg^{2+}, Na^+, and Cl^-). The concentrations of solutes released from thermokarst can be several orders of magnitude higher than in background water, due to the liberation of soluble materials sequestered in ground ice. Such instances have been reported globally, but mainly in Arctic areas: Ellesmere Island, Canada (Kokelj and Lewkowicz, 1999); Melville Island, Canada (Dugan et al., 2012); Banks Island, Canada (Rudy et al., 2017); Northwest Territories, Canada (Kokelj et al., 2005; Malone et al., 2013); Mackenzie Delta, Canada (Kokelj et al., 2009); Peel Plateau and Mackenzie Mountains, Canada (Kokelj et al., 2013); Brooks Range, Alaska (Abbott et al., 2015); western Siberia, Russia (Loiko et al., 2017); and McMurdo Dry Valleys, Antarctica (Gooseff et al., 2016).

Thermokarst can affect surface water chemistry for several decades with the magnitude of effects depending on the proportion of a catchment area influenced by the disturbance (Kokelj et al., 2005) (Fig. 3), although generalisations are difficult since hydrochemical modifications can depend on several factors such as the type of disturbance, its stabilisation rate, the downstream connectivity, and the characteristics of the impacted area (e.g., physical and geochemical characteristics of near-surface permafrost, mineralogical composition of bedrock/sediments).

Thermokarst processes can also release Si, and N and P forms into surface water. For example, Si and P were found to be higher in small depressions and permafrost subsidences in actively thawing frozen palsa peat bog areas than in stable palsa peat bog zones in western Siberia, Russia (Loiko et al. 2017). Moreover, thermokarst gullies and thaw slumps have been shown to increase N and P form concentrations in water tracks, streams and lakes (Brooks Range - Alaska, Bowden et al., 2008; Harms et al., 2014; McMurdo Dry Valleys - Antarctica, Gooseff et al., 2016). Louiseize et al. (2014) (Melville Island - Canada) demonstrated that even small thermokarst occurrences (active-layer detachments) are capable to promote the export of microbially-derived nitrate as result of limited nitrate retention and enhanced nitrification in mineral soils exposed in the scar zones. Abbott et al. (2015) (Brooks Range - Alaska) also suggested that aquatic disturbance of inorganic nitrogen by thermokarst can be long-lived. Conversely, Thompson et al. (2012) (Mackenzie Delta uplands - Canada) found that lakes unaffected by thaw slumps had higher concentrations of the nutrients than slump-affected lakes, despite the high concentrations of inorganic N and P in runoff pools within the slump sites. Within-lake biological uptake, sedimentation and degradation of nutrients were assumed to be the main drivers of this effect.

4.3 Rock-glacier thawing

Thies et al. (2007) found strong increases in electrical conductivity (up to 19-fold), SO_4^{2-} (up to 26-fold), Ca^{2+} (up to 13-fold), and Mg^{2+} (up to 68-fold) from 1985 to 2005 in two lakes at the toe of active rock glaciers in the Central Eastern Alps in Europe (Austria and Italy) (Fig. 4). These trends have been attributed to the melting of ice in the rock glaciers and an associated release of solutes, although no direct evidence of the driving processes was provided. A similar evidence was reported in 20 remote high-elevation lakes located in Himalaya (Mt. Everest Region, Nepal) over the past two decades. Here, significant enrichments in solutes have been attributed to the retreat of debris-covered glaciers (Salerno et al., 2016) due to the sulphide oxidation processes that occur in subglacial environments, often microbially mediated.

Increased geochemical fluxes from thawing permafrost and ice melt in rock glaciers have been also found to influence the seasonal hydrochemistry of small streams in the Austrian Alps (Krainer and Mostler, 2002; Berger et al., 2004; Krainer et al., 2007), although no specific analyses on major ions were conducted and instead, conductivity measurements were used as proxy. Williams et al. (2006) in 2003 filled this gap, reporting strong seasonal increases for SO_4^{2-} (60-fold), Mg^{2+} (30-fold) and Ca^{2+} (20-fold) in the outflow of rock glacier RG5 in the Colorado Front Range (Rocky Mountains - USA) compared to nearby streams and lakes during the low-flow season. Longer-term measurements from 1998 to 2002 showed the same pattern of elevated concentrations of these solutes in the fall. The elevated concentrations of major ions in rock glacier outflows and downstream lakes are consistent with the results of a precursor study performed by Giardino et al. (1992), who noted that water exiting rock glaciers in the San Juan Mountains of Colorado (USA) acquired a significant dissolved load during movement through the rock glaciers. Specific hydrochemical characteristics in rock glacier meltwaters were found also in the hyperarid Agua Negra river basin - Andes of Argentina (meltwaters enriched in SO_4^{2-}, HCO_3^- and Ca^{2+}, Lecomte et al., 2008) and in Central Eastern Alps - Austria (meltwaters enriched in SO_4^{2-}, Ca^{2+} and Mg^{2+}, Thies et al., 2013).

During the last decades, the increasing solute concentrations in lakes with rock glaciers in their catchments were attributed to the enhanced melting of ice due to atmospheric warming (Thies et al., 2007). Williams et al. (2006) further supported this hypothesis, assuming this process may occur seasonally, with a flushing of mineralised water at the end of summer when the 0 ºC isotherm proceeds towards the ice under conditions of degrading permafrost. Williams et al. (2006) reported the highest ion concentrations in rock glacier outflows during particularly dry years. In this case, there would be more internal melt than in wetter years, with a higher proportion of mineralised groundwater. The authors also hypothesised that long-lasting snow cover might be able to reduce solute export by delaying subsurface melt and by dilution of solute-rich water with snowmelt. Not only snowmelt, but also rain has been reported to lower solute contents in rock glacier outflows (Krainer et al., 2007) through dilution effects. Conversely, Thies et al. (2007) hypothesised that solute fluxes from rock glaciers intensify after periods of intense rainfall during summer and fall, as these might enhance water percolation through rock glaciers and thus cause the flushing of geochemical products. In summary, three hypotheses exist: (a) Warm periods cause increased subsurface melt, which releases solutes. (b) Dry periods reduce the input of meteoric water and thus reduce the dilution of rock-glacier outflow. (c) The percolation of meteoric water through the rock glacier exports solutes. Considerable uncertainty exists about the processes, and their interactions, driving the export of solutes from internal ice in rock glaciers.

Recent research attributed increased NO_3^- concentrations in surface water melt in permafrost and rock glaciers (Rocky Mountains - USA, Baron et al., 2009). Williams et al. (2007) found that nitrate concentrations in the outflow of a rock glacier were higher than all other analysed surface waters of the same area in the Colorado Front Range, Rocky Mountains (USA), from 1998 to 2004 (Fig. 5). Fegel et al. (2016) provides addition support that elevated biogeochemical and microbial characteristics of rock glaciers in the Cascade Mountains, Rocky Mountains, and Sierra Nevada (USA) result in elevated nitrate content in surface water. Further, microbial communities adapted to the extreme environment of the interior of rock glaciers have been indicated as potential sources of the elevated nitrate in the rock glacier outflows (Williams et al. 2007). The observed increase in surface-water nitrate concentration may also be a result of melt water flushing microbially-active sediments (Barnes et al., 2014) and/or active microbial populations in sediment pockets within talus fields in the Rocky Mountains (USA) (Ley et al., 2004).

5. Impacts on trace elements

5.1 Pervasive permafrost degradation

Although several studies dealt with the analysis of trace element concentrations in surface fresh water in cold areas, especially in the boreal and sub-Arctic regions (e.g., Rember and Trefry, 2004; Voss et al., 2015), researches devoted to the investigation of the links with permafrost dynamics are rare and mainly based on short time series. Typically, mobilisation of trace elements to surface water during early spring is attributed to snowmelt when precipitation that has accumulated during winter is released (McNamara et al., 1997; Rember and Trefry, 2004; Pokrovsky et al., 2010), with trace elements also potentially incorporated from upper-soil horizons and thawing ponds (Rember and Trefry, 2004). Differently, Barker et al. (2014) reported increasing concentrations of trace elements (Al, Ba, Fe and Mn) in stream water during late fall in Alaska (Brooks Range), correlated with increased ALT and weathering fluxes from freshly-exposed mineral surface (metal sources). Bagard et al. (2011) discriminated two distinct seasonal peaks of trace element fluxes in Central Siberia (Russia): the first one in the beginning of spring snowmelt and related to the uppermost soil horizon rich in organics, and the second one occurring late in summer and related to deeper mineral soil horizons. Permafrost gradient was found to be the most important environmental parameter in controlling trace element concentrations in western Siberia (Russia), where Pokrovsky et al. (2016) analysed approximately 60 large and small rivers (<100 to ≤150,000 km^2 watershed area) during spring flood and summer, and winter baseflow, across a 1500-km latitudinal gradient. The main factors controlling the shift of river feeding from surface and subsurface flow to deep underground flow in the permafrost-bearing zone were the depth of the active layer and its position in organic or mineral horizons of the soil profile. The authors also suggested that under climate warming scenarios the change in the relative degree of the peat and mineral soil leaching to the river might cause a decrease in divalent metal organic complexes and an increasing delivery of trivalent and tetravalent hydrolysates in the form of organo-ferric colloids to the river via supra-permafrost flow and hyporheic influx. Differently, a recent study performed by Raudina et al. (2017) in western Siberia (Russia) reported that the concentration of most trace elements in peat porewater did not show any statistically significant trend with latitude. Applying a space-for-time substitution approach for the climate warming scenario in western Siberia, the authors also predicted a decrease in the feeding of small rivers and lakes by peat soil leachates in transitional permafrost areas with frozen peatlands. Finally, similar to major elements, the use of stable isotopes of inorganic solutes such as Fe (Hirst et al., 2017) will allow to improve the knowledge about the impacts of future permafrost evolution on surface waters.

In a multi-decadal study (from 1980s to 2000s) performed in a mineralised alpine watershed with natural acidic in the Colorado's Rocky Mountains (USA), Todd et al. (2012) provided evidence of increasingly high concentrations (increase ranging from 100% to 400%) of Zn and other metals of ecological concern (e.g., Mn) in the Upper Snake River during low-flow months (late summer-early fall) (Fig. 6). Increased rock weathering on freshly-exposed mineral surfaces and sulphide oxidation due to the degradation of permafrost and the opening of new subsurface pathways, causing the migration of groundwater and oxygen into the subsurface, were indicated as driving factors. Acid drainage is indeed a potential mechanism capable to strongly influence water quality, through the release of metals (e.g., Fe, Zn, Pb, Cu) from sulphide-rich minerals due to the exposure to oxygen and water (e.g., He et al., 1997; Nordstrom, 2011), in areas where permafrost is degrading. In northern Yukon Territory (Canada), Lacelle et al. (2007) reported water quality deterioration due to acidity and high metal concentrations in a stream affected by acid drainage. Here, acid drainage was a natural phenomenon which effects were amplified by the construction of a road. The removal of the surficial sediments, and consequent deepening the active layer, allowed seasonal thawing and oxidation of sulphide-rich minerals in the excavated area. Increase of ALT under warmer climate scenarios might enhance acid drainage at sites with sulphide-rich minerals by allowing access to atmospheric oxygen to penetrate deeper in the ground.

5.2 Thermokarst

Trace element dynamics in thermokarst ponds and lakes have been found to be primarily related to the evolutionary stage of these features. For example, Manasypov et al. (2014) investigated the hydrochemical characteristics of thermokarst ponds and lakes with different degrees of permafrost influence located in the northern part of western Siberia, across a latitudinal gradient of ~900 km. The authors evidenced a geographical gradient in trace element concentrations related to thermokarst lake evolution (from small permafrost depressions to larger lakes), in relation to different degrees of carbon and element leaching from frozen peat soils. They estimated that the acceleration of permafrost thaw in Siberia might increase the amount of small thermokarst lakes and, in turn, increase the concentrations of most trace elements by 200-400%. Shirokova et al. (2013) also reported a clear decrease in some trace element concentrations (e.g., Fe, Al, Ti, Zr and rare earth elements) from thermokarst depressions, to ponds, and finally, to lakes in the central part of western Siberia (Russia) (Fig. 7). They attributed this pattern to two main factors: i) increase in water residence time in large lakes, which leads to more efficient mineralisation of organo-mineral colloids by aerobic heterotrophic bacterioplankton; ii) decrease in peat abrasion at the lake borders as the perimeter-volume ratio decreases with size. Furthermore, trace element concentrations (e.g., Fe, Al, Ni, Pb) have been found to increase in hollows, depressions and permafrost subsidences in an area characterised by thawing frozen peat in western Siberia, Russia (Loiko et al., 2017). Here, the authors reported decreasing concentrations with increasing surface area of the water body, along the hydrological continuum: soil water → hollows → depressions and permafrost subsidences → thaw ponds → thermokarst lakes. Thermokarst lakes can also have an important role in the storage of some trace elements (Al, Cr, Cd, Pb) comparable to, or even higher than, the transport of these elements by rivers (western Siberia - Russia, Polishchuk et al., 2017). Hot summer periods, and cycles of ice formation and melting have been reported to drive in-lake biological uptake and release of some elements (Zn, Mn, Co, Mo, Cu, Si, Pb, Cd) (western Siberia - Russia, Pokrovsky et al., 2013), and seasonal differences in trace element concentrations (western Siberia - Russia, Manasypov et al., 2015), respectively. In addition, Audry et al. (2011) observed that in differently-developed thermokarst lakes in western Siberia, sediments at the early stage of ecosystem development are a sink for water column Cu, Zn, Cd, Pb and Sb due to authigenic sulphides precipitation, confirming the progressive depletion trends of trace element concentrations in the water column during the chronosequence of lake maturation (from permafrost subsidence to mature and drained lakes) reported by Pokrovsky et al. (2011), who analysed 20 lakes and small ponds of various sizes and ages in the same area. Conversely, the sediments are a source of dissolved Co, Ni and As to the water column due to upward diffusive fluxes, regardless the ecosystem development stage. However, the concentrations of these elements were found to be low due to sorption processes on Fe-bounding particles and/or large-size organo-mineral colloids.

Several studies have been conducted focusing on Hg and Pb dynamics in thermokarst-affected areas. For instance, Klaminder et al. (2008) assessed the impact of permafrost degradation on the increase in Hg mobility from soils to lake surface water in a palsa peat complex in sub-Arctic Sweden. Main driving factors were accelerated soil/peat erosion, increasing hydrological connectivity, and increased runoff which led to the exposure of soluble elements. Gordon et al. (2016) (Northwest Territories - Canada) further demonstrated that new hydrological pathways due to permafrost thaw can increase peatland hydrological connectivity, creating areas of high Hg concentrations at critical points (fens) before water discharged to a wider drainage system. Small permafrost thaw ponds also contribute in controlling the local and regional fluxes of Hg, as demonstrated by MacMillan et al. (2015) in sub-Arctic and high-Arctic Canada. Elevated concentrations of Hg and methylmercury have been found to be strongly related to inputs of organic matter and nutrients into surface water. Moreover, Rydberg et al. (2010) reported that long-term changes in climate, permafrost and mire dynamics had large effects on the release of Hg from palsa mires to lakes in sub-arctic Sweden. Particularly, the recent thawing of permafrost has increased thermokarst erosion, resulting in an enhanced Hg transport from the mire to lake water. In their study of sub-Arctic Sweden, Klaminder et al. (2010) reported a similar process for Pb, attributing its increase to the thermokarst erosion of peat. Conversely, Deison et al. (2012) found that lakes affected by the development of retrogressive thaw slumps in the Mackenzie Delta (Canada) had lower Hg levels in surface sediments when compared to not-affected lakes. The authors attributed these results to the effects of thaw slumping of clay-rich permafrost tills, which resulted in high inorganic sedimentation rates that caused a dilution of organic material and associated Hg in the sediments.

5.3 Rock-glacier thawing

An increase in trace element concentrations in high-elevation Alpine lakes (Central Eastern Alps, Austria and Italy) has been observed, and it has been attributed to the influence of ice melting within rock glaciers (Thies et al., 2007). For instance, unexpectedly high Ni, Mn and Al concentrations, exceeding the European Union limits for drinking water by more than an order of magnitude, have been reported for a remote high-mountain lake situated at the toe of an active rock glacier. In contrast, negligible concentrations of these elements (and other solutes) have been recorded in an adjacent pond without a rock glacier in the catchment (Central Eastern Alps - Italy, Ilyashuk et al., 2014). Surprisingly, the lithology did not contain high Ni concentrations, thus the source of Ni remains unknown. In addition, other springs derived from rock glaciers in the Central Eastern Alps exhibited high concentrations of trace elements such as Ni, Co, Cu, Fe, Mn and Zn (Krainer, 2014), pointing to rock-glacier ice as a potential origin. Increasing acidity of rock-glacier affected surface waters accompanied by high heavy metal concentrations have been also reported to cause significant changes in aquatic ecosystems (Central Eastern Alps - Austria, Thies et al., 2013) and severe ecological damages (Central Eastern Alps - Italy, Ilyashuk et al., 2017).

Results from geochemical analyses of ice cores sampled from a medium-sized active rock glacier (Lazaun) in Central Eastern Alps (Italy) are reported by Krainer et al. (2015). The ice near the base is approximately 10,300 years old and trace elements (Ni, Co, Cu, Zn) are enriched in three horizons within the ice at depths of 4.2 m (approx. 2600 yr BP), 9.9 m (approx. 3200y BP) and 12.4 m (approx. 3500 yr BP), with Ni concentrations up to 0.49 mg L^{-1}. High Ni concentrations were also measured at the rock glacier spring, indicating that Ni in spring water was derived from distinct depth intervals within the permafrost ice (Krainer et al., 2011). Again, the lithology was reported to not contain high concentrations of trace elements. However, only few rock glaciers in Central Eastern Alps display meltwater with abnormally high trace element concentrations. Considerable uncertainty still exists about the origin of the high concentrations of trace elements in rock-glacier ice.

6. Research perspectives

Several studies report major ion distribution in near-surface permafrost, finding increasing concentrations at the top of permafrost, generally corresponding with ice-rich layers. However, relatively few researches have combined geochemical analyses on changing near-surface permafrost and resulting effects on surface-water characteristics. Performing combined investigations may provide insight into the relationship between concentrations of major ions in near-surface permafrost and modifications of surface-water geochemistry in case of permafrost degradation, both on intra- and inter-annual bases. Few studies in the Arctic also demonstrated that rainfall events can flush solutes from near-surface permafrost to the surface and increase ion concentrations in surface water. Projected warming and rainfall augmentation in Arctic areas (IPCC, 2013) might cause increases in major ion concentrations in surface water in permafrost areas; in turn, increasing runoff might reduce ion concentrations since discharge is generally negatively correlated with concentrations of major ions because of dilution effects (Petrone et al., 2006, 2007), although the “chemostatic” behaviour in stream water (solute concentrations in stream water are not determined by simple dilution, Godsey et al., 2009) should be also taken into account. The ability to adequately incorporate these processes into models will be relevant for predicting the hydrochemical impacts of climatic changes in transitional permafrost areas. Future studies should also focus on the bedrock/sediment composition and particularly on the geochemical composition of permafrost ice that might be locally enriched in ions (due to lithological characteristics), which can be released when permafrost ice melts. Finally, research in highly sensitive areas such as watersheds where acid drainage is possible due to lithological properties (e.g., pyritiferous shale presence) will help to understand how permafrost degradation may cause a deterioration of surface water quality.

Major ion concentrations downstream of rock glaciers have mainly been investigated in the European Alps and in mountain chains of North America. Extending these studies to areas such as in the Andes (e.g., Brenning, 2005) and in the Hindu Kush Himalaya (e.g., Schmid et al., 2015; Gruber et al., 2017) will allow better understanding the dynamics and potential effects of rock-glacier thaw on downstream water quality. The establishment of baselines for future monitoring and the better understanding of processes of solute export from rock glaciers will thereby be important elements of such studies. In this framework, investigating the hydrological connections between rock glaciers and adjacent surface waters through the application on non-invasive techniques (Colombo et al., 2017) could provide new insights on the rock-glacier role, and dynamics, in exporting concentrated chemical fluxes into high-elevation hydrological systems. Moreover, it has been shown that melting of ice in active rock glaciers can increase their storage capacity. May chemical composition of outflowing water be affected by this process? Fresh water stored in rock glaciers can be important for local water management only during the melt season (summer) since during the cold season (winter) discharge is extremely low or absent. Future studies on water quality originating from rock glaciers should also focus on relict rock glaciers due to their higher storage capacity and residence time for fresh water which can also be released during winter. Finally, extending investigations to other common debris features with permafrost such as talus slopes (Sass, 2006; Lambiel and Pieracci, 2008; Scapozza et al., 2011) will allow understanding the effects of thawing permafrost on water quality in cold mountains more broadly. Do talus slopes with significant amount of ground ice (cf., Gruber and Haeberli, 2009) behave like rock glaciers in their export of solutes during permafrost degradation? Trends of major ions and ion-isotope ratios in surface water may be considered as important additions to the permafrost monitoring conducted in mountains (c.f., PERMOS, 2016).

Given that most studies on geochemistry in near-surface permafrost and ground ice considered only major ions, future research on trace element concentrations and their variations within the active layer, permafrost and massive ground ice occurrences is necessary for evaluating the effect they might have on ecosystems when thawing.

High trace element concentrations have been reported downstream of rock glaciers and in three horizons within the ice core of a rock glacier in the Central Eastern Alps. Since lithology was not found to contain high trace element concentrations, alternative hypotheses (e.g., atmospheric fallout) to chemical weathering should be tested in order to understand the origin and the potential for future release of trace elements.

Beside specific research needs, several broader questions emerge for the understanding of permafrost degradation and its effects on inorganic chemistry of surface water. These are especially important in arid areas, such as some areas in the Andes and Himalaya, where fresh water stored in permafrost can be important for local water management (Brenning, 2005; Rangecroft et al., 2013; Rangecroft et al., 2015; Gruber et al., 2017) in small catchments. What is the role of permafrost in controlling water quality in these areas and what duration will the transient effects of permafrost thaw have? What will be the long-term implications of permafrost degradation on water quality of local freshwater reservoirs? What are the possible socioeconomic consequences of water quality deterioration due to permafrost degradation? What are suitable adaptation strategies to mitigate the possible impacts of permafrost degradation on water supply quality of local populations?

7. Conclusion

This review of permafrost-thaw impacts on inorganic chemistry of surface fresh water has revealed several common patterns.

Spatially-distributed progressive increases of major ion delivery to surface fresh water have been reported in both polar and mountain areas following permafrost degradation. This is the result of increasing contributions of highly mineralised groundwater and enhanced interactions of soil water with deep mineral strata. Localised releases of major ions to surface water due to the liberation of soluble materials sequestered in permafrost and ground ice have been found in ice-rich terrains both at high latitude (thermokarst features) and high elevation (rock glaciers). Generalisations must be considered carefully though, because the intensity of hydrochemical modifications depends on several factors such as the characteristics of the watersheds (e.g., physical and geochemical characteristics of permafrost), their hydrological connectivity, the magnitude and type of disturbance in case of thermokarst, and the extent of permafrost or rock glaciers in the catchment. Permafrost degradation can also increase the export of inorganic N and P forms, and Si, from permafrost to surface water due to deeper flow paths that bypass shallow organic layers where most retention and removal of nutrients occur (N), and because of enhanced mineral weathering (P and Si). However, significant uncertainty has been shown on DIN dynamics, mainly dependent on regional and landscape-scale differences in Arctic areas (e.g., Alaska and Siberia). In addition, thermokarst can enhance the export of inorganic nutrients from permafrost to surface water, although biological uptake, sedimentation and degradation of nutrients might mitigate these effects.

Increases in nitrate concentrations have been also attributed to rock glacier melt water flushing microbially active sediments.

Trace element dynamics in permafrost areas have been shown to exhibit two seasonal peaks in geochemical fluxes: one at the beginning of the snowmelt (early spring), related to the flushing of surficial soil horizons and previously mobilised species stored in them; the second occurring in late summer and early fall, associated with deeper mineral-soil horizons. The formation of new thermokarst features (e.g., ponds, lakes, palsa mires) and leaching from frozen peat can be relevant for increasing trace element concentrations in permafrost catchments and for affecting the locations of high concentrations due to newly-developing hydrological pathways. Alternatively, increasing sediment erosion due to thermokarst processes has been reported to potentially mitigate these effects through increasing sedimentation rates and dilution of concentrations. Moreover, stabilisation and increasing size of older thermokarst features might lead to decreasing trends because of stabilising shorelines, reduced peat abrasion, and more efficient mineralisation. Increasing trends in trace element concentrations have been also reported in streams and lakes in mountain areas, where enhanced chemical weathering on fresh mineral surfaces exposed by permafrost thaw and contribution of metal-enriched groundwater have been hypothesised to be the main drivers. Nevertheless, in the Central Eastern Alps, high concentrations of trace elements (e.g., Ni) in surface water have been found to be unrelated to the lithological setting, hence rejecting the hypothesis of mineral weathering as the only process involved.

In conclusion, several factors influence the timing and magnitude of the impacts of permafrost degradation on inorganic chemistry of surface fresh water. These include permafrost sensitivity to thawing (e.g., amount of ground ice), modes of permafrost degradation (e.g., increasing ALT, thermokarst) and environmental characteristics of watersheds (e.g., soil properties, lithology, land cover, topography, hydrological connectivity). Further mobilisation of solutes and related transport to surface fresh water appear reasonable under warming climatic conditions. This might develop through gradual concentration increase in case of progressive and diffused thawing of permafrost, and/or through distinct temporal peaks in case of localised thermokarst and rock glacier thawing. As a result, modifications of inorganic water chemistry might have significant implications for ecological and human systems.

Acknowledgments

Nicola Colombo and Franco Salerno equally contributed to this paper. We greatly acknowledge the critical proof reading by Renée Leduc and Carina Schuh, and the valuable suggestions provided by Christopher Burn (Carleton University, Ottawa, Canada). Two anonymous reviewers provided valuable feedback and input during the review of this manuscript.

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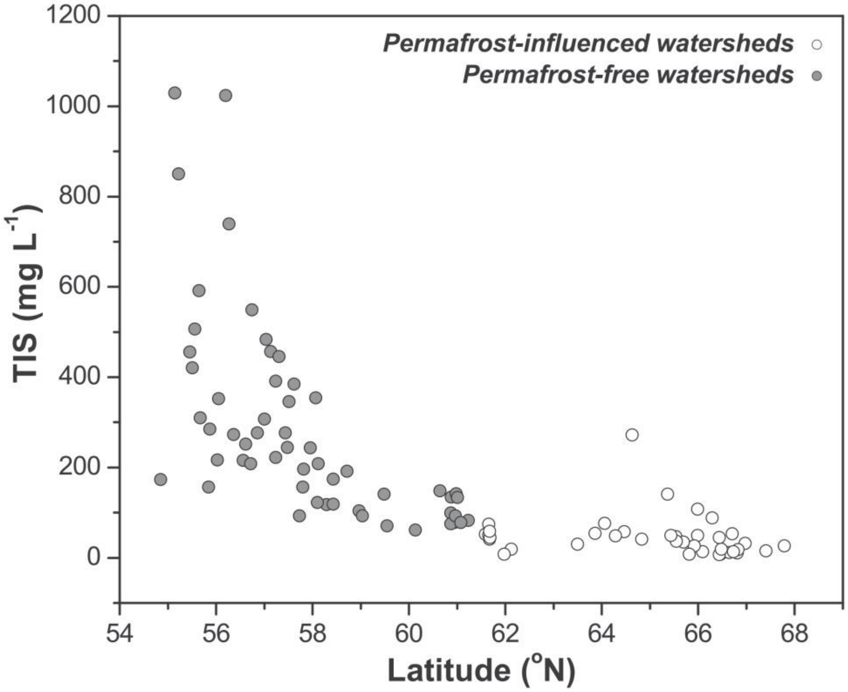
1134 **Figures and Tables**



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1136 Figure 1 - TIS (total inorganic solutes, defined as sum of Ca^{2+}, K^+, Mg^{2+}, Na^+, Si, Cl^-, SO_4^{2-} and HCO_3^-) as

1137 a function of latitude. Permafrost limit is approximately located at 61°N (from Frey et al., 2007a).

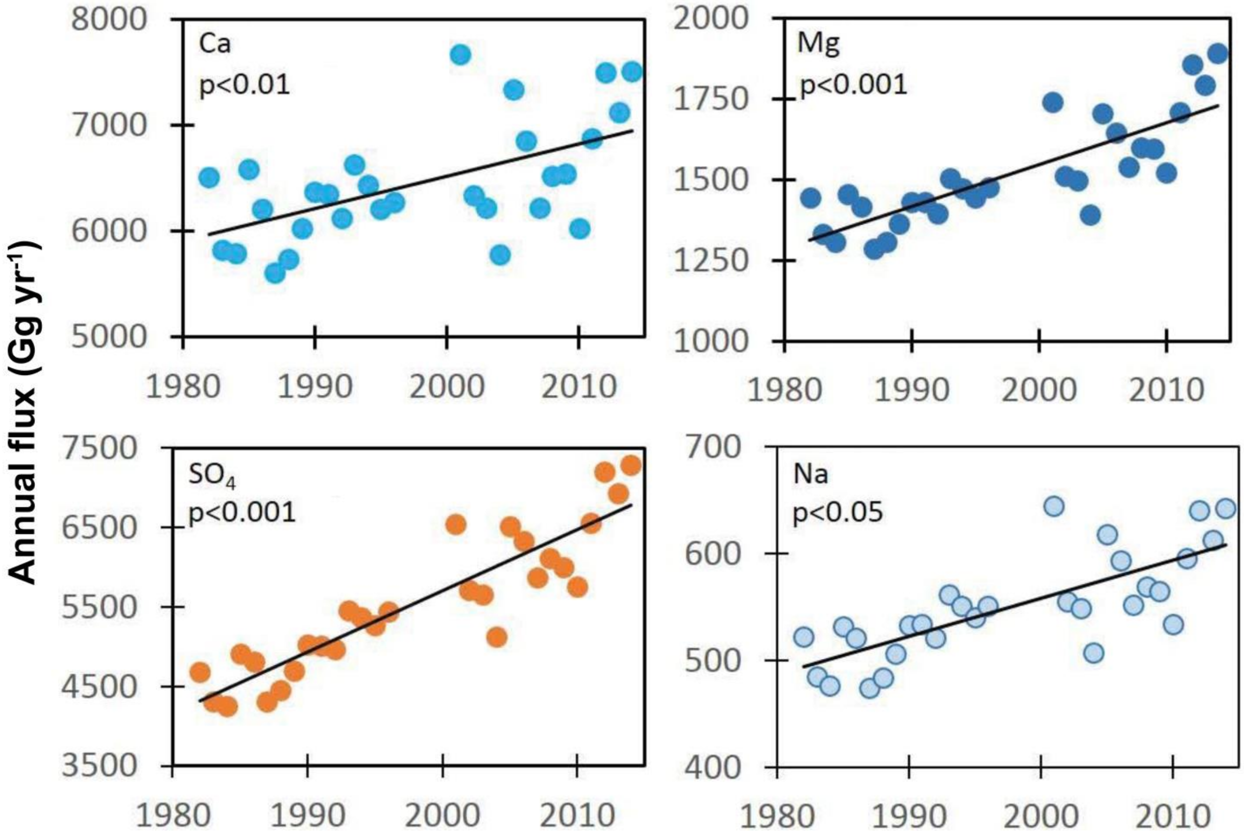

Figure 2 - Annual flux increases in the Yukon River with Thiel-Sen trend lines (adapted from Toohey et al., 2016).

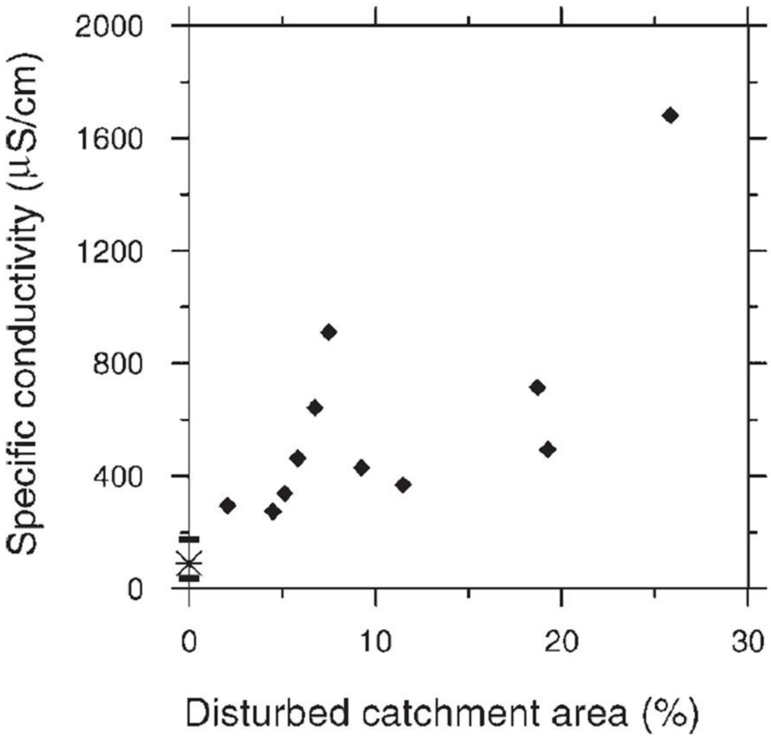

Figure 3 - Percentage of catchment area influenced by thermokarst disturbance (slumping) and specific conductivity (as index of total solute concentrations, mainly represented by Ca^{2+}, Mg^{2+}, and SO_4^{2-}) of lake water, upland lakes, Mackenzie Delta region. The (*) indicates the mean specific conductivity of the 11 pristine lakes measured by Kokelj et al. (2005). The (–) indicates maximum and minimum values for pristine lakes (from Kokelj et al., 2005).

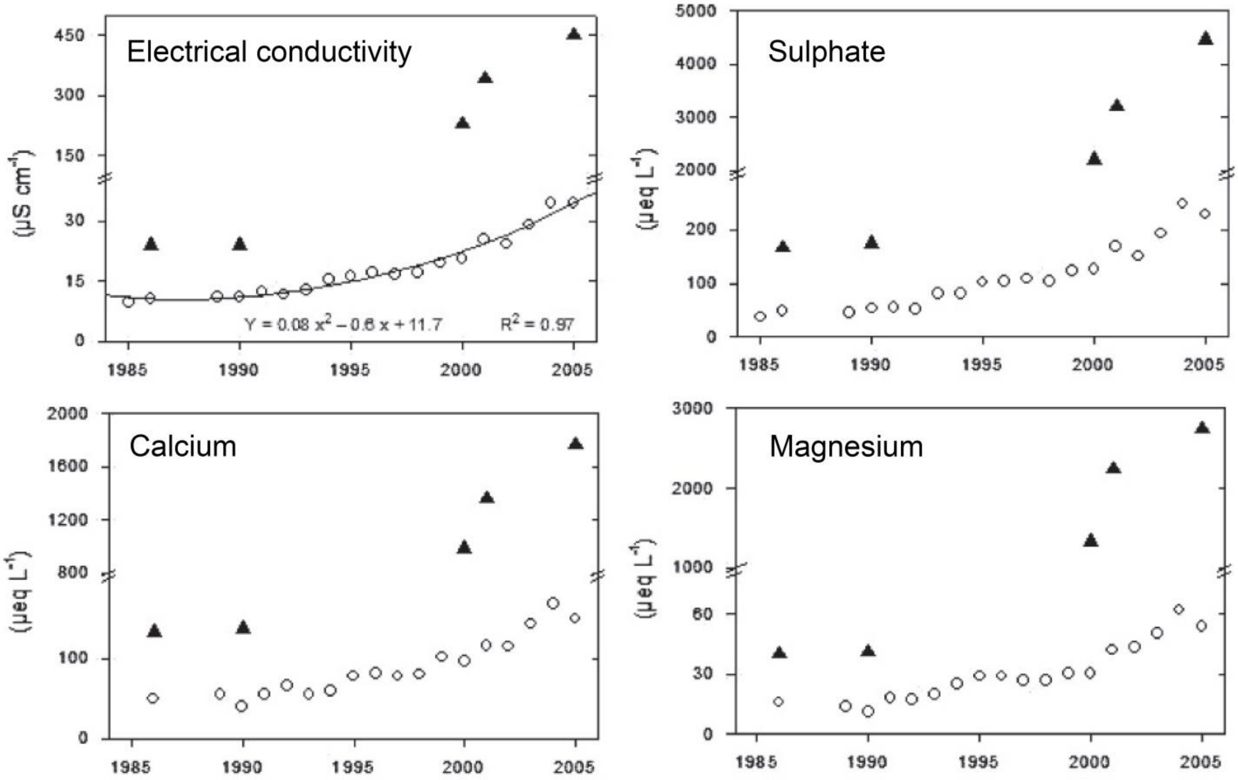

Figure 4 - Electrical conductivity, sulphate, calcium and magnesium concentrations in lake water of Rasass See (black triangles) and Schwarzsee ob Sölden (open circles) lakes (1985-2005). Values for each lake represent mean values of four to seven discrete samples along the lake vertical profile taken during holomixis. Variability among single values is <5 %. Horizontal lines show break in vertical scale (adapted from Thies et al., 2007).

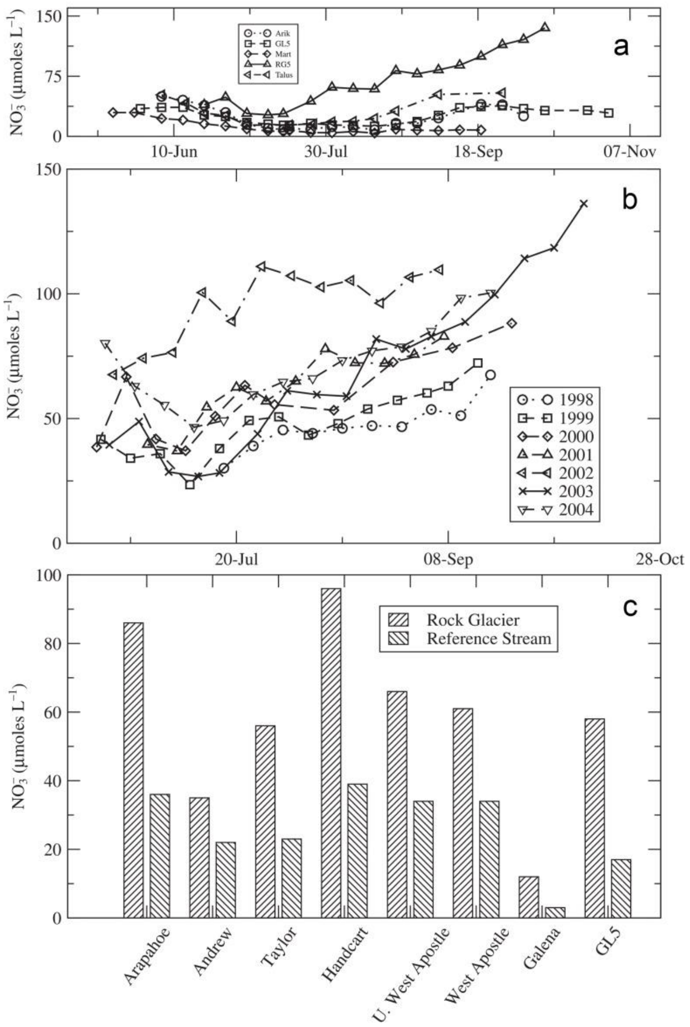

Figure 5 - a) Time series of nitrate in surface water of the Green Lakes Valley, for 2003. Arik is Arikaree Glacier outflow; GL5 is the outlet of Green Lakes 5; Mart is the outlet of the Martinelli catchment; RG5 is the Green Lake 5 rock glacier outflow; talus is the blockfield site (for site details refer to Williams et al., 2007). b) Time series of nitrate concentrations in the outflow of RG5 during the snow-free season from 1998 to 2004. c) Comparison of nitrate concentrations from the outflow of 7 rock glaciers and co-located surface waters, August 2003 (adapted from Williams et al., 2007).

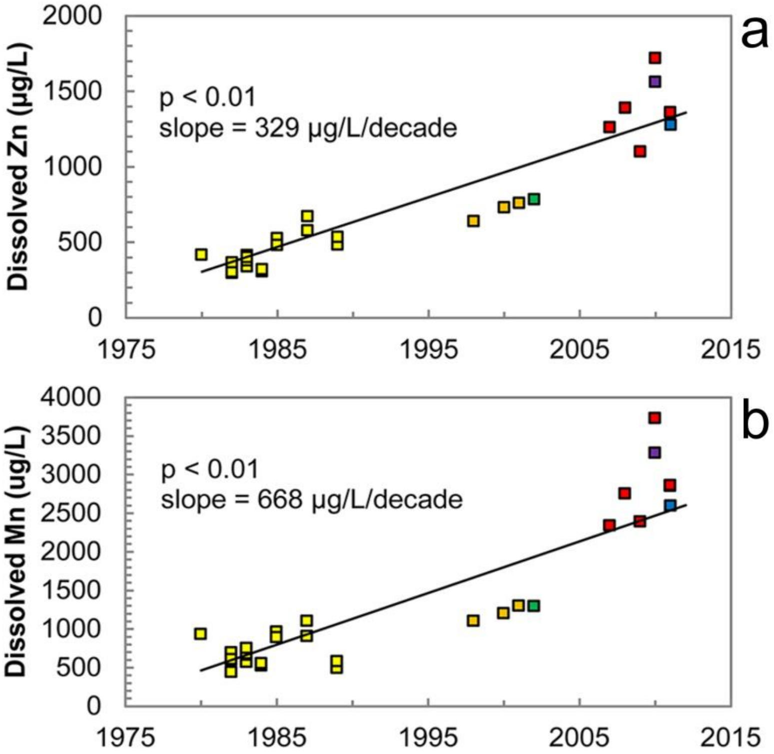

Figure 6 - Trends in September dissolved Zn (a) and Mn (b) concentrations (µg/L) in the Upper Snake River. Different colours represent different data sources (further details in Todd et al., 2012). p-values were computed using the Mann-Kendall Test, and lines shown are Kendall-Theil lines (adapted from Todd et al., 2012).

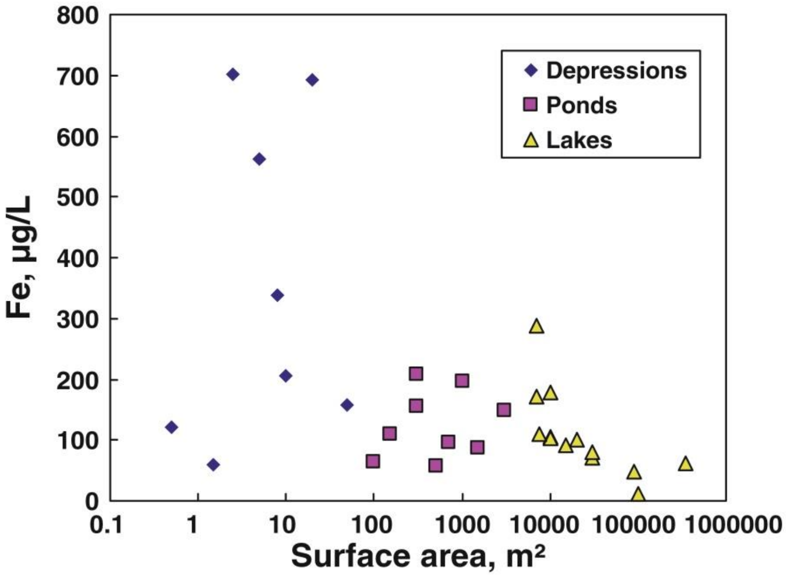

Figure 7 - Iron (<0.45 µm) concentration as a function of water body surface area in depressions, ponds and lakes. Uncertainties (±2σ) are the same or smaller than symbol sizes (from Shirokova et al., 2013).

Author	Year	Study area	Permafrost configuration	Analytes
Bagard et al.	2013	Central Siberia - Russia	PD	M
Caine	2010	Colorado Rocky Mountains - USA	PD	M
Douglas et al.	2013	Chena River - Alaska	PD	M
Frey et al.	2007a 2007b	Western Siberia - Russia	PD	M
Giesler et al.	2014	Sub-Arctic - Sweden	PD	M
Harms and Jones	2012	Caribou Poker Creeks Research Watershed, Upper Kuparuk River Basin - Alaska	PD	M
Harris et al.	2007	McMurdo Dry Valleys - Antarctica	PD	M
Hobbie et al.	1999	MilkyWay stream - Alaska	PD	M
Jones et al.	2005	Caribou Poker Creeks Research Watershed - Alaska	PD	M
Keller et al.	2007 2010	Brooks Range - Alaska	PD	M
Koch et al.	2013	Richardson Catchment - Alaska	PD	M
Kolosov et al.	2016	Central Siberian Plateau - Russia	PD	M
Lafrenière and Lamoureux	2013	Melville Island - Canada	PD	M
Lamhonwah et al.	2017	Melville Island - Canada	PD	M
Levy et al.	2011	McMurdo Dry Valleys - Antarctica	PD	M
Lewis et al.	2012	Melville Island - Canada	PD	M
MacLean et al.	1999	Caribou-Poker Creeks Research Watershed - Alaska	PD	M
Mast et al.	2011	Colorado Rocky Mountains - USA	PD	M
Mavromatis et al.	2014 2016	Central Siberia - Russia Yenisey River - Russia	PD	M
McClelland et al.	2007	Upper Kuparuk River - Alaska	PD	M
Parham et al.	2013	Central Siberia - Russia	PD	M
Petrone et al.	2006 2007	Caribou-Poker Creeks Research Watershed - Alaska	PD	M
Pokrovsky et al.	2013 2015	Central Siberia - Russia Western Siberia - Russia	PD	M
Roberts et al.	2017	Melville Island - Canada	PD	M
Rühland et al.	2003	Central Canadian sub-Arctic	PD	M
Stottlemyer	2001	Noatak National Preserve - Alaska	PD	M
Striegl et al.	2005	Yukon River basin - Alaska	PD	M
Szopińska et al.	2016	Central Mongolia	PD	M
Tank et al.	2012 2016	Siberia - Russia, Yukon - Alaska, Mackenzie - Canada	PD	M
Todd et al.	2012	Colorado Rocky Mountains - USA	PD	M / T
Dugan et al.	2012	Melville Island - Canada	PD / TK	M
Baron et al.	2009	Colorado Rocky Mountains - USA	PD / RG	M
Abbott et al.	2015	Brooks Range - Alaska	TK	M
Bowden et al.	2008	Brooks Range - Alaska	TK	M
Gooseff et al.	2016	McMurdo Dry Valleys -Antarctica	TK	M
Harms et al.	2014	Brooks Range - Alaska	TK	M
Kokelj and Lewkowicz	1999	Ellesmere Island - Canada	TK	M
Kokelj et al.	2005	Northwest Territories - Canada	TK	M
Kokelj et al.	2009	Mackenzie Delta - Canada		

Kokelj et al.	2013	Peel Plateau, Mackenzie Mountains - Canada		
Loiko et al.	2017	Western Siberia - Russia	TK	M / T
Louiseize et al.	2014	Melville Island - Canada	TK	M
Malone et al.	2013	Northwest Territories - Canada	TK	M
Rudy et al.	2017	Banks Island - Canada	TK	M
Thompson et al.	2012	Mackenzie Delta uplands - Canada	TK	M
Barnes et al.	2014	Colorado Rocky Mountains - USA	RG	M
Berger et al.	2004	Central Eastern Alps - Austria	RG	M
Fegel et al.	2016	Cascade Mountains, Rocky Mountains, Sierra Nevada - USA	RG	M
Giardino et al.	1992	San Juan Mountains of Colorado - USA	RG	M
Krainer and Mostler	2002	Central Eastern Alps - Austria	RG	M
Krainer et al	2007	Central Eastern Alps - Austria	RG	M
Lecomte et al.	2008	Agua Negra river basin - Andes of Argentina	RG	M
Thies et al.	2007	Central Eastern Alps - Austria, Italy	RG	M / T
	2013	Central Eastern Alps - Austria		
Williams et al.	2006 2007	Colorado Front Range - USA	RG	M
Bagard et al.	2011	Central Siberia - Russia	PD	T
Barker et al.	2014	Brooks Range - Alaska	PD	T
Hirst et al.	2017	Lena River Basin - Russia	PD	T
Lacelle et al.	2007	Northern Yukon Territory - Canada	PD	T
Pokrovsky et al.	2016	Western Siberia - Russia	PD	T
Raudina et al.	2017	Western Siberia - Russia	PD	T
Audry et al.	2011	Western Siberia - Russia	TK	T
Deison et al.	2012	Mackenzie Delta - Canada	TK	T
Gordon et al.	2016	Northwest Territories - Canada	TK	T
Klaminder et al.	2008 2010	Sub-Arctic - Sweden	TK	T
MacMillan et al.	2015	Sub-arctic, high-Arctic - Canada	TK	T
Manasypov et al.	2014 2015	Western Siberia - Russia	TK	T
Pokrovsky et al.	2011 2013	Western Siberia - Russia	TK	T
Polishchuk et al.	2017	Western Siberia - Russia	TK	T
Rydberg et al.	2010	Sub-Arctic - Sweden	TK	T
Shirokova et al.	2013	Western Siberia - Russia	TK	T
Ilyashuk et al.	2014 2017	Central Eastern Alps - Italy	RG	T
Krainer	2014	Central Eastern Alps - Austria, Italy	RG	T
Krainer et al.	2011	Central Eastern Alps - Italy	RG	T

Table 1 - Selected papers reporting information on the impacts of permafrost degradation on inorganic chemistry of surface fresh water globally. PD: Pervasive permafrost degradation, TK: Thermokarst, RG: Rock-glacier thawing, M: Major ions, T: Trace elements.