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

(Article begins on next page)

10 November 2024

1 **Review: Impacts of permafrost degradation on inorganic chemistry of surface**

2 **fresh water**

3

4 Nicola Colombo[1,2,*], Franco Salerno[3], Stephan Gruber[2], Michele Freppaz[4], Mark Williams[5], Simona

5 Fratianni[1], Marco Giardino[1]

6

7 [1] University of Turin, Department of Earth Sciences, Via Valperga Caluso, 35, 10125, Turin, Italy

8 [2] Carleton University, Department of Geography and Environmental Studies, Loeb Building, 1125 Colonel

9 By Drive, Ottawa, ON, K1S 5B6, Canada

10 [3] CNR-IRSA (National Research Council - Water Research Institute), Via del Mulino, 19, 20047,

11 Brugherio, Italy

12 [4] University of Turin, Department of Agricultural, Forest and Food Sciences, Largo Paolo Braccini, 2,

13 10095, Grugliasco, Italy

14 [5] University of Colorado, Department of Geography, Boulder CO 80309, USA

15 *Correspondence to*: nicola.colombo@unito.it

16

17 **Abstract**

18 Recent studies have shown that climate change is impacting the inorganic chemical characteristics of

19 surface fresh water in permafrost areas and affecting aquatic ecosystems. Concentrations of major ions

20 (e.g., Ca^{2+}, Mg^{2+}, SO_4^{2-}, NO_3^-) can increase following permafrost degradation with associated deepening

21 of flow pathways and increased contributions of deep groundwater. In addition, thickening of the active

22 layer and melting of near-surface ground ice can influence inorganic chemical fluxes from permafrost into

23 surface water. Permafrost degradation has also the capability to modify trace element (e.g., Ni, Mn, Al,

24 Hg, Pb) contents in surface water. Although several local and regional modifications of inorganic

25 chemistry of surface fresh water have been attributed to permafrost degradation, a comprehensive review

26 of the observed changes is lacking. The goal of this paper is to distil insight gained across differing

27 permafrost settings through the identification of common patterns in previous studies, at global scale. In

28 this review we focus on three typical permafrost configurations (pervasive permafrost degradation,

29 thermokarst, and thawing rock glaciers) as examples and distinguish impacts on (i) major ions and (ii)

30 trace elements. Consequences of warming climate have caused spatially-distributed progressive

31 increases of major ion and trace element delivery to surface fresh water in both polar and mountain areas

32 following pervasive permafrost degradation. Moreover, localised releases of major ions and trace

33 elements to surface water due to the liberation of soluble materials sequestered in permafrost and ground

34 ice have been found in ice-rich terrains both at high latitude (thermokarst features) and high elevation

35 (rock glaciers). Further release of solutes and related transport to surface fresh water can be expected

36 under warming climatic conditions. However, complex interactions among several factors able to influence

37 the timing and magnitude of the impacts of permafrost degradation on inorganic chemistry of surface fresh

38 water (e.g., permafrost sensitivity to thawing, modes of permafrost degradation, characteristics of

39 watersheds) require further conceptual and mechanistic understanding together with quantitative

40 diagnosis of the involved mechanisms in order to predict future changes with confidence.

41

1. Introduction

43 Average atmospheric temperature has increased globally over the last decades and, as a response,

44 the cryosphere is changing (Fountain et al., 2012). Permafrost, a component of the cryosphere, is

45 widespread in the Arctic and Antarctica, and in cold mountains, including densely populated areas of the

46 European Alps and Asian mountain ranges (Gruber, 2012). Permafrost interacts with climate (Walter

47 Anthony et al., 2006; Schuur et al., 2015), hydrology (e.g., Liljedahl et al., 2016; Yang et al., 2017),

48 ecosystems (Jorgenson et al., 2001; Vonk et al., 2015), geophysical processes (e.g., Gruber et al., 2004;

49 Gruber and Haeberli, 2009), and human systems (Nelson et al., 2002; Harris et al., 2009).

50 Recent reviews have focused on the impacts of permafrost warming and degradation on river

51 biogeochemistry (Frey and McClelland, 2009) and aquatic ecosystems (Vonk et al., 2015) in the Arctic.

52 Notwithstanding, local and regional modifications of water hydrochemistry due to permafrost degradation

53 have been reported from many locations, globally. Given the sparsity of data available, understanding and

54 analysing permafrost degradation impacts on inorganic chemistry of surface fresh water will benefit from

55 identifying common patterns in existing studies. The present review thus aims to distil insight gained

56 across differing permafrost environments and configurations globally.

57   Following a brief background section, we distinguish three typical example configurations of permafrost

58 thaw. For those, we review impacts of permafrost degradation on major ions (e.g., Ca^{2+}, Mg^{2+}, SO_4^{2-}, NO_3^-

59) and on trace elements (e.g., Ni, Mn, Al, Hg, Pb).

60

61 **2.   Permafrost: definition and main characteristics**

62   Permafrost is defined as ground (soil and/or rock, including ice and organic material, plus air/gas in

63 unsaturated ground) that remains at or below 0 °C for at least two consecutive years (Muller, 1943). Thus,

64 permafrost is a thermal phenomenon and it can, but does not need to, contain water or ice. Most

65 permafrost areas experience seasonal thaw, during which ground surface temperature rises above 0 °C.

66 The layer of ground that thaws on a seasonal basis is called "active layer" (ACGR, 1988). The depth of

67 permafrost (permafrost base) is determined by the geothermal heat flux, subsurface properties, and the

68 long-term surface temperature. The upper limit of permafrost (permafrost table) is largely controlled by

69 surface temperatures during summer and near-surface ground properties (Williams and Smith, 1989;

70 French, 2007). The difference in depth between the permafrost table and the permafrost base is the

71 permafrost thickness (see Figure 2 in ACGR, 1988, source: https://ipa.arcticportal.org). Permafrost

72 regions are often divided into a continuous permafrost zone, where more than ~90% of the surface area is

73 underlain by permafrost, a discontinuous zone, of which ~10% to ~90% is underlain by permafrost, and a

74 zone of isolated patches of permafrost, where permafrost occurs under less than ~10% of the ground. The

75 discontinuous zone is often divided into extensive discontinuous permafrost (~90% to ~50%) and sporadic

76 discontinuous permafrost (~50% to ~10%, Heginbottom et al., 1995).

77   The global permafrost region (exposed land surface below which some permafrost can be expected) is

78 estimated to be $22\pm3 \times 10^6$ km^2, approximately 17% of the global land area (Gruber, 2012). Zhang et al.

79 (1999; 2003) estimated that permafrost could underlie nearly 25% of the exposed land area of the

80 Northern Hemisphere. Despite occupying only 0.36% (49,800 km^2) of the Antarctic region, permafrost is

81 estimated to be present beneath all ice-free terrain, except at the lowest elevations of the maritime

82 Antarctic and sub-Antarctic islands (Vieira et al., 2010). Although with increased spatial heterogeneity

83 imposed by topography, permafrost is also widespread in many mountain ranges such as the European

84 Alps (e.g., Boeckli et al., 2012), the Hindu Kush Himalaya (e.g., Gruber et al., 2017), and the Andes (e.g.,

85 Brenning, 2005).

86   The aggradation and degradation of permafrost usually occur in association with a change of mean

87 ground temperature due to microclimatic (ground cover) and climatic changes (Williams and Smith, 1989).

88 *Permafrost degradation* refers to the decrease of volume of a body of permafrost, whereas *permafrost*

89 *thaw* or *thawing* is used to refer to increasing liquid water content and other changes of physical

90 characteristics in response to energy input within a body of rock or soil while still satisfying the definition of

91 permafrost. The term *thawed permafrost* indicates ground that no longer is permafrost. Global warming

92 over the past several decades is now large enough that regional climate change is emerging above the

93 noise of natural variability (Hansen and Sato, 2016). Despite the small magnitude of warming relative to

94 weather fluctuations, effects of the warming already have notable environmental, social and economic

95 impacts. In this context, the degradation of permafrost is seen as a major challenge in the current

96 discussion of globally rising air temperatures (IPCC, 2013), because of the possible effects on climate

97 (e.g., Harden et al., 2012; Schuur et al., 2015), infrastructure stability (e.g., Nelson et al., 2001; Streletskiy

98 et al., 2012), hydrology (e.g., Liljedahl et al., 2016; Walvoord and Kurylyk, 2016), natural hazards in

99 mountainous areas (e.g., Gruber et al., 2004; Harris et al., 2009), and aquatic biogeochemistry and

100 ecosystems (e.g., Frey and McClelland, 2009; Vonk et al., 2015).

101

102 **3. Typical configurations of permafrost thaw**

103   We focus on three typical configurations in which permafrost degradation can impact the inorganic

104 chemistry of water bodies. While not exhaustive, these examples currently reflect major interests in this

105 research field:

106 i) Pervasive permafrost degradation, leading to spatially-distributed modifications in groundwater-

107   surface water connectivity and causing volumes of permafrost to thaw and become subject to

108   weathering and leaching;

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

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

111   (i) <u>Pervasive permafrost degradation</u>. Permafrost thaw affects hydrology through increasing hydrologic

112 permeability with ice loss in pores (Bense et al., 2009; Kurylyk et al., 2014) and through the release of

113 water stored in frozen material. Permafrost is commonly an aquitard (Woo, 2012; Kane et al., 2013),

114 supporting perched water tables and increasing near-surface soil moisture (Gorbunov, 1978; Ishikawa et

115 al., 2005). Permafrost thaw, through talik growth and higher permeability of partially-frozen material,

116 changes the hydrologic connectivity in a catchment and affects the rates and amounts of flow along

117 differing paths (Hinzman et al., 2005; Rogger et al., 2017), which often produce water with distinct

118 geochemical signatures (Clark et al., 2001).

119   Due to thaw and degradation, new hydrological pathways in permafrost are expected to enhance the

120 interaction between mineralised groundwater and surface water, driving transitional permafrost

121 environments from surface-water dominated systems to groundwater-dominated systems (e.g., Frey and

122 McClelland, 2009). Indeed, recent studies have attributed increases in groundwater contributions to base

123 flow of several rivers in permafrost watersheds to permafrost degradation (Walvoord and Striegl, 2007; St.

124 Jacques and Sauchyn, 2009; Kolosov et al., 2016).

125   Most near-surface effects of permafrost thaw stem from increasing Active Layer Thickness (ALT),

126 which has been shown to occur at large scale, across entire landscapes (e.g., Åkerman and Johansson,

127 2008). Recently, Luo et al. (2016) investigated the spatiotemporal characteristics of ALT across the

128 Northern Hemisphere from 1990 to 2015. Significant trends of increasing ALT have been observed at

129 approximately 43 % of the investigated 169 sites analysed. Regionally, increasing ALT have been found in

130 North America (e.g., Oelke et al., 2004; Burn and Zhang, 2009), Asia (e.g., Frauenfeld et al., 2004; Oelke

131 et al., 2004; Wu et al., 2012), Antarctica (e.g., Guglielmin and Cannone, 2012; Guglielmin et al., 2014),

132 and Europe (e.g., Åkerman and Johansson, 2008; Harris et al., 2009), although variation exists due to

133 local conditions.

134   Increasing ALT can expose newly-thawed soils to near-surface water and increase chemical fluxes

135 from freshly-exposed strata (e.g., Keller et al., 2007; Keller et al., 2010). Furthermore, geochemical

136 constituents previously sequestered within the near-surface permafrost (e.g., Kokelj and Burn, 2005) can

137 be released.

138   Although spatially-distributed modifications in groundwater-surface water connectivity and increasing

139   soil volumes subject to weathering and leaching are distinct processes associated with permafrost

140   degradation, the impacts of these processes on the inorganic chemistry of surface water are not easily

141   discernible. Indeed, both result in diffuse increases of geochemical fluxes into the receiving water bodies

142   and only few studies distinguish the main sources of such fluxes (e.g., Keller et al., 2007, 2010;

143   Lamhonwah et al., 2017).

144   (ii) <u>Thermokarst</u>. Permafrost warming and increasing ALT can cause self-reinforcing thaw of ice-rich

145   ground leading to the development of thermokarst (Jorgenson and Osterkamp, 2005; Rowland et al.,

146   2010). The term thermokarst includes a suite of processes and landforms that involve lowering, and often

147   collapsing, of the land surface because of melting excess ground ice. Kokelj and Jorgenson (2013)

148   classified the principal thermokarst types as: a) hillslope processes, including retrogressive thaw slumps,

149   active layer detachment slides, and thermal erosion gullies; b) thaw lake processes, including lake

150   expansion/reduction, drainage, and lake basin evolution; c) wetland processes, including peatland

151   collapse and the development of bogs and fens. Thermokarst can also occur on active rock glaciers

152   (details in (iii) Rock glacier thawing), where increased melting of permafrost ice can cause the formation of

153   small thermokarst lakes (Kääb and Haeberli, 2001). Thermokarst is commonly observed throughout most

154   permafrost regions including Canada (e.g., Watanabe et al., 2011), Alaska (e.g., Andresen and Lougheed,

155   2015), Russia (e.g., Manasypov et al., 2014), Mongolia (e.g., Sharkhuu, 1998), China (e.g., Wu et al.,

156   2010), Antarctica (e.g., Gooseff et al., 2016) and the European Alps (e.g., Kääb and Haeberli, 2001).

157   Thermokarst development has the potential to alter the geochemical signature of surface water

158   because it facilitates mobilisation of sediments and solutes previously sequestered in ground ice (Gooseff

159   et al., 2009), causing localised and strong influences on hydrochemical systems (Malone et al., 2013).

160   The effects of thermokarst are mostly local, but the transport of solutes into downstream aquatic

161   ecosystems can sometimes impact large areas (e.g., Kokelj et al., 2013). Moreover, the formation of new

162   features such as thaw ponds and lakes can modify the hydrochemical characteristics of large areas (e.g.,

163   Manasypov et al., 2014).

164   (iii) <u>Rock-glacier thawing</u>. Rock glaciers are distinctive and sometimes abundant geomorphological

165   features of cold mountain regions (Barsch, 1996). They are slowly-flowing mixtures of rock debris and ice

166 and formed by a continuum of processes from glacial to periglacial (Haeberli et al., 2006). The thawing of

167 ice-rich talus slopes (cf., Gruber and Haeberli, 2009), although not studied in detail, is likely part of the

168 same continuum. Rock glaciers are considered as the visible expression of long term creep of ice-rock

169 mixtures under permafrost conditions (Humlum, 2000; Ikeda and Matsuoka, 2006; Berthling, 2011), and

170 for this reason, they are often used for first-order mapping of permafrost distribution at coarse scales (e.g.,

171 Brenning, 2005; Cremonese et al., 2011; Schmid et al., 2015). Compared to glaciers, rock glaciers are

172 believed to react more slowly to rising temperature because of the retardation of heat transfer through

173 thick debris mantles (Barsch, 1996). During the cold season, ice is formed within rock glaciers, their "mass

174 balance", however, is difficult to determine. Emerging observations suggest the thermal and kinematic

175 state of many rock glaciers is changing in concert with climate change (e.g., Roer et al., 2005; Haeberli et

176 al., 2006; Scapozza et al., 2014). Many active (moving) rock glaciers loose some water due to melt during

177 summer. This is because frequently, their tongues extent into microclimatic situations in which the moving

178 ice core undergoes slow degradation. However, at decadal and longer time scales, due to global warming,

179 accelerated melting of the ice within rock glaciers may represent an increasing hydrological contribution to

180 downstream areas (Brenning, 2005). For this reason, active rock glaciers are generally considered to be

181 sources of fresh water (Burger et al., 1999), especially in arid areas, small catchments, and during low

182 flow conditions (e.g., Corte, 1976; Brenning, 2005; Azócar and Brenning, 2010; Millar et al., 2013;

183 Rangecroft et al., 2015). Finally, melting of permafrost ice in active rock glaciers in particular, and also in

184 ice-rich talus (cf., Gruber and Haeberli 2009), is able to increase the porosity of the sediments. In turn, this

185 can increase the storage capacity, causing changes in the discharge pattern (Rogger et al., 2017).

186 The thaw of rock glaciers due to climate change has the potential to export enriched-solute fluxes and

187 thereby modify the inorganic chemistry of downstream impounded surface waters (e.g., Ilyashuk et al.,

188 2014) and streams (e.g., Thies et al., 2013).

189 Local and regional modifications of inorganic chemistry of surface fresh water following different

190 configurations of permafrost thaw and degradation have been reported globally and are listed in Tab. 1.

191 The impacts on major ions and trace elements are discussed in the following chapters.

192

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)

221   in the period 1982-2014 (Fig. 2), attributing this evidence to altered hydrological flowpaths and increased

222   weathering due to widespread permafrost degradation. Several other studies of Northern-Hemisphere

223   high-latitude streams reported increasing concentrations of ions such as Ca^{2+} and SO_4^{2-} (Noatak National

224   Preserve - Alaska, Stottlemyer, 2001), Ca^{2+} and Mg^{2+} (sub-Arctic - Sweden, Giesler et al., 2014), Ca^{2+},

225   Mg^{2+}, K^+, and Na^+ (Caribou-Poker Creeks Research Watershed - Alaska, Petrone et al., 2006, 2007),

226   SO_4^{2-}, Ca^{2+}, Mg^{2+}, and alkalinity (Mackenzie basin - Canada, Tank et al., 2016), HCO_3^-, Cl^-, SO_4^{2-}, Ca^{2+},

227   and Mg^{2+} (Central Siberian Plateau - Russia, Kolosov et al., 2016), following permafrost degradation.

228   Similar findings have been reported for streams and lakes of the Colorado Rocky Mountains (USA) over

229   the last two-three decades, with increasing concentrations of Ca^{2+} and SO_4^{2-} (Baron et al., 2009; Caine,

230   2010; Todd et al., 2012), and Ca^{2+}, Mg^{2+}, and SO_4^{2-} (Mast et al., 2011).

231   Although increases in major ions concentrations of surface water have been found in several

232   transitional permafrost areas, the causal link with permafrost degradation is not well established. Keller et

233   al. (2007, 2010) and Douglas et al. (2013) proposed the use of Ca/Sr, Ca/Na, Ca/Ba, and $^{87}Sr/^{86}Sr$ trends

234   in soils and streams for quantitatively assessing the effects of increasing ALT on mineral fluxes to surface

235   water, where the parent material and soil geochemistry are variable with depth. For instance, geochemical

236   trends point to carbonate concentrations in the ground increasing with depth (higher concentrations in the

237   permafrost than in the active layer), due to decreasing frequency of thaw and hence weathering intensity.

238   Correspondingly, a progressive increase of carbonate weathering signals during the summer season

239   (especially in late summer) was found, associated with increasing thaw depth. New insights on

240   mechanisms and sources of inorganic solutes in surface waters draining permafrost-affected soils can

241   also be gained by using stable isotopes of inorganic solutes such as Ca (Bagard et al., 2013), Mg

242   (Mavromatis et al., 2014, 2016), and Si (Pokrovsky et al., 2013). For example, Pokrovsky et al. (2013)

243   analysed Si isotopic composition of large and small rivers, surface flow, interstitial soil solutions, plant litter

244   and soils in Central Siberia, finding significant variations of the dissolved Si isotopic composition. These

245   variations were linked to 1) the high concentrations of suspended silicate matter during the spring flood, 2)

246   the progressive increase of ALT during summer accompanied by secondary mineral precipitation in

247   surface layers and plant uptake of Si, and 3) the impact of the permafrost-hosted deep underground

248 waters during winter. Under climate change, the resulting permafrost degradation may be the most

249 important factor controlling the Si isotopic composition in the rivers.

250 Short-time increases in the depth of thaw are also capable to enhance mineral fluxes from permafrost

251 terrain. Increases in major ion concentrations (mainly Ca^{2+}, Mg^{2+}, Na^+, SO_4^{2-}, and Cl^-) in surface water

252 have been linked to active-layer thickening following strong thermal perturbations (warm summer

253 conditions) and summer rainfall (Melville Island - Canada, Dugan et al., 2012, Lewis et al., 2012;

254 Lafrenière and Lamoureux, 2013; Roberts et al., 2017) with flushing of solutes stored in near-surface

255 permafrost (Melville Island - Canada, Lamhonwah et al., 2017) as well as to fire disturbance (Central

256 Siberia - Russia, Parham et al., 2013).

257 In the McMurdo Dry Valleys (Antarctica) the presence of unusual surface-flow-seep features has been

258 attributed to the potential melting of complex-origin sub-surface ice (e.g., excess ground ice, snow

259 patches, refrozen precipitation that has accumulated in the subsurface, or buried glacier ice) in permafrost

260 (Harris et al., 2007). Seeps showed a different geochemical signature in comparison to glacial meltwater,

261 strongly enriched in solutes (Na^+, Ca^{2+}, Mg^{2+}, Cl^-, NO_3^-, SO_4^{2-} and HCO_3^-). These features associated with

262 water tracks (Levy et al., 2011) might contribute to the desert hydrological cycle exporting solute-

263 concentrated fluxes from sub-surface ice to surface water, enhancing the connectivity between the

264 cryosphere and the hydrosphere in the Antarctic polar desert (Levy et al., 2011).

265 Future increases in air temperature, and rain, may enhance the release of major ions from solute-rich

266 layers in near-surface permafrost. Indeed, several studies reported a solute enrichment at the base of the

267 active layer and at the top of permafrost in different permafrost environments due to ion redistribution

268 during thermally-induced moisture migration (Herschel Island - Canada, Kokelj et al., 2002; Northwest

269 Territories - Canada, Kokelj and Burn, 2003, 2005; Lacelle et al., 2014), greater availability of water for

270 chemical aqueous reactions where drainage is inhibited by subjacent permafrost (Yukon Territory -

271 Canada, Lacelle et al., 2007; Devon Island - Canada, Lacelle et al., 2008), mineral dissolution reactions

272 taking place in loamy layers slowly flushed by advective flow, with the reaction products transported

273 upward by diffusion to surficial peat layers frequently flushed during summer (Ilulissat - Greenland, Jessen

274 et al., 2014), physical modifications (landslides) of active layer geochemistry (Yamal Peninsula - Russia,

275 Leibman and Streletskaya, 1997), and cryogenic concentration of salts during freezing and humidity-
276 driven salt separation at the soil surface (McMurdo Dry Valleys - Antarctica, Levy et al. 2012).

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

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

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

303 (Frey and McClelland, 2009, Pokrovsky et al., 2015). For instance, Hobbie et al. (1999) demonstrated that

304 permafrost thawing along the MilkyWay stream in northern Alaska contributed 30% of inflowing phosphate

305 into Toolik Lake, causing bottom-up trophic effects. Similarly, increasing concentrations of dissolved

306 silicate with a decrease in permafrost coverage were found in Alaska (MacLean et al., 1999) and in

307 western Siberia (Frey et al., 2007a). However, assuming future climate warming and increasing vegetation

308 density in permafrost-bearing zones, transient uptake of nutrients during summer period may attenuate

309 the expected increase in surface water concentrations linked to progressive involvement of thawed

310 mineral horizons, as hypothesised by Pokrovsky et al. (2015) for Si dynamics in western Siberian rivers

311 (Russia).

312

313 **4.2   Thermokarst**

314 Thermokarst disturbances in permafrost terrain such as retrogressive thaw slumps, thermal erosion

315 gullies, active-layer detachment slides, and catastrophic thaw of frozen palsa peat bogs have been found

316 to strongly modify major ion concentrations of surface water (mainly SO_4^{2-}, Ca^{2+}, Mg^{2+}, Na^+, and Cl^-). The

317 concentrations of solutes released from thermokarst can be several orders of magnitude higher than in

318 background water, due to the liberation of soluble materials sequestered in ground ice. Such instances

319 have been reported globally, but mainly in Arctic areas: Ellesmere Island, Canada (Kokelj and Lewkowicz,

320 1999); Melville Island, Canada (Dugan et al., 2012); Banks Island, Canada (Rudy et al., 2017); Northwest

321 Territories, Canada (Kokelj et al., 2005; Malone et al., 2013); Mackenzie Delta, Canada (Kokelj et al.,

322 2009); Peel Plateau and Mackenzie Mountains, Canada (Kokelj et al., 2013); Brooks Range, Alaska

323 (Abbott et al., 2015); western Siberia, Russia (Loiko et al., 2017); and McMurdo Dry Valleys, Antarctica

324 (Gooseff et al., 2016).

325 Thermokarst can affect surface water chemistry for several decades with the magnitude of effects

326 depending on the proportion of a catchment area influenced by the disturbance (Kokelj et al., 2005) (Fig.

327 3), although generalisations are difficult since hydrochemical modifications can depend on several factors

328 such as the type of disturbance, its stabilisation rate, the downstream connectivity, and the characteristics

329 of the impacted area (e.g., physical and geochemical characteristics of near-surface permafrost,

330 mineralogical composition of bedrock/sediments).

331   Thermokarst processes can also release Si, and N and P forms into surface water. For example, Si

332   and P were found to be higher in small depressions and permafrost subsidences in actively thawing

333   frozen palsa peat bog areas than in stable palsa peat bog zones in western Siberia, Russia (Loiko et al.

334   2017). Moreover, thermokarst gullies and thaw slumps have been shown to increase N and P form

335   concentrations in water tracks, streams and lakes (Brooks Range - Alaska, Bowden et al., 2008; Harms et

336   al., 2014; McMurdo Dry Valleys - Antarctica, Gooseff et al., 2016). Louiseize et al. (2014) (Melville Island -

337   Canada) demonstrated that even small thermokarst occurrences (active-layer detachments) are capable

338   to promote the export of microbially-derived nitrate as result of limited nitrate retention and enhanced

339   nitrification in mineral soils exposed in the scar zones. Abbott et al. (2015) (Brooks Range - Alaska) also

340   suggested that aquatic disturbance of inorganic nitrogen by thermokarst can be long-lived. Conversely,

341   Thompson et al. (2012) (Mackenzie Delta uplands - Canada) found that lakes unaffected by thaw slumps

342   had higher concentrations of the nutrients than slump-affected lakes, despite the high concentrations of

343   inorganic N and P in runoff pools within the slump sites. Within-lake biological uptake, sedimentation and

344   degradation of nutrients were assumed to be the main drivers of this effect.

345

346   **4.3   Rock-glacier thawing**

347   Thies et al. (2007) found strong increases in electrical conductivity (up to 19-fold), SO_4^{2-} (up to 26-fold),

348   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

349   glaciers in the Central Eastern Alps in Europe (Austria and Italy) (Fig. 4). These trends have been

350   attributed to the melting of ice in the rock glaciers and an associated release of solutes, although no direct

351   evidence of the driving processes was provided. A similar evidence was reported in 20 remote high-

352   elevation lakes located in Himalaya (Mt. Everest Region, Nepal) over the past two decades. Here,

353   significant enrichments in solutes have been attributed to the retreat of debris-covered glaciers (Salerno et

354   al., 2016) due to the sulphide oxidation processes that occur in subglacial environments, often microbially

355   mediated.

356   Increased geochemical fluxes from thawing permafrost and ice melt in rock glaciers have been also

357   found to influence the seasonal hydrochemistry of small streams in the Austrian Alps (Krainer and Mostler,

358   2002; Berger et al., 2004; Krainer et al., 2007), although no specific analyses on major ions were

359   conducted and instead, conductivity measurements were used as proxy. Williams et al. (2006) in 2003

360   filled this gap, reporting strong seasonal increases for SO_4^{2-} (60-fold), Mg^{2+} (30-fold) and Ca^{2+} (20-fold) in

361   the outflow of rock glacier RG5 in the Colorado Front Range (Rocky Mountains - USA) compared to

362   nearby streams and lakes during the low-flow season. Longer-term measurements from 1998 to 2002

363   showed the same pattern of elevated concentrations of these solutes in the fall. The elevated

364   concentrations of major ions in rock glacier outflows and downstream lakes are consistent with the results

365   of a precursor study performed by Giardino et al. (1992), who noted that water exiting rock glaciers in the

366   San Juan Mountains of Colorado (USA) acquired a significant dissolved load during movement through

367   the rock glaciers. Specific hydrochemical characteristics in rock glacier meltwaters were found also in the

368   hyperarid Agua Negra river basin - Andes of Argentina (meltwaters enriched in SO_4^{2-}, HCO_3^- and Ca^{2+},

369   Lecomte et al., 2008) and in Central Eastern Alps - Austria (meltwaters enriched in SO_4^{2-}, Ca^{2+} and Mg^{2+},

370   Thies et al., 2013).

371   During the last decades, the increasing solute concentrations in lakes with rock glaciers in their

372   catchments were attributed to the enhanced melting of ice due to atmospheric warming (Thies et al.,

373   2007). Williams et al. (2006) further supported this hypothesis, assuming this process may occur

374   seasonally, with a flushing of mineralised water at the end of summer when the 0 ºC isotherm proceeds

375   towards the ice under conditions of degrading permafrost. Williams et al. (2006) reported the highest ion

376   concentrations in rock glacier outflows during particularly dry years. In this case, there would be more

377   internal melt than in wetter years, with a higher proportion of mineralised groundwater. The authors also

378   hypothesised that long-lasting snow cover might be able to reduce solute export by delaying subsurface

379   melt and by dilution of solute-rich water with snowmelt. Not only snowmelt, but also rain has been

380   reported to lower solute contents in rock glacier outflows (Krainer et al., 2007) through dilution effects.

381   Conversely, Thies et al. (2007) hypothesised that solute fluxes from rock glaciers intensify after periods of

382   intense rainfall during summer and fall, as these might enhance water percolation through rock glaciers

383   and thus cause the flushing of geochemical products. In summary, three hypotheses exist: (a) Warm

384   periods cause increased subsurface melt, which releases solutes. (b) Dry periods reduce the input of

385   meteoric water and thus reduce the dilution of rock-glacier outflow. (c) The percolation of meteoric water

386   through the rock glacier exports solutes. Considerable uncertainty exists about the processes, and their

387   interactions, driving the export of solutes from internal ice in rock glaciers.

388   Recent research attributed increased NO_3^- concentrations in surface water melt in permafrost and rock

389   glaciers (Rocky Mountains - USA, Baron et al., 2009). Williams et al. (2007) found that nitrate

390   concentrations in the outflow of a rock glacier were higher than all other analysed surface waters of the

391   same area in the Colorado Front Range, Rocky Mountains (USA), from 1998 to 2004 (Fig. 5). Fegel et al.

392   (2016) provides addition support that elevated biogeochemical and microbial characteristics of rock

393   glaciers in the Cascade Mountains, Rocky Mountains, and Sierra Nevada (USA) result in elevated nitrate

394   content in surface water. Further, microbial communities adapted to the extreme environment of the

395   interior of rock glaciers have been indicated as potential sources of the elevated nitrate in the rock glacier

396   outflows (Williams et al. 2007). The observed increase in surface-water nitrate concentration may also be

397   a result of melt water flushing microbially-active sediments (Barnes et al., 2014) and/or active microbial

398   populations in sediment pockets within talus fields in the Rocky Mountains (USA) (Ley et al., 2004).

399

5.   Impacts on trace elements

5.1   Pervasive permafrost degradation

402   Although several studies dealt with the analysis of trace element concentrations in surface fresh water

403   in cold areas, especially in the boreal and sub-Arctic regions (e.g., Rember and Trefry, 2004; Voss et al.,

404   2015), researches devoted to the investigation of the links with permafrost dynamics are rare and mainly

405   based on short time series. Typically, mobilisation of trace elements to surface water during early spring is

406   attributed to snowmelt when precipitation that has accumulated during winter is released (McNamara et

407   al., 1997; Rember and Trefry, 2004; Pokrovsky et al., 2010), with trace elements also potentially

408   incorporated from upper-soil horizons and thawing ponds (Rember and Trefry, 2004). Differently, Barker

409   et al. (2014) reported increasing concentrations of trace elements (Al, Ba, Fe and Mn) in stream water

410   during late fall in Alaska (Brooks Range), correlated with increased ALT and weathering fluxes from

411   freshly-exposed mineral surface (metal sources). Bagard et al. (2011) discriminated two distinct seasonal

412   peaks of trace element fluxes in Central Siberia (Russia): the first one in the beginning of spring snowmelt

413   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

442   affected by acid drainage. Here, acid drainage was a natural phenomenon which effects were amplified by

443   the construction of a road. The removal of the surficial sediments, and consequent deepening the active

444   layer, allowed seasonal thawing and oxidation of sulphide-rich minerals in the excavated area. Increase of

445   ALT under warmer climate scenarios might enhance acid drainage at sites with sulphide-rich minerals by

446   allowing access to atmospheric oxygen to penetrate deeper in the ground.

447

448   **5.2   Thermokarst**

449   Trace element dynamics in thermokarst ponds and lakes have been found to be primarily related to the

450   evolutionary stage of these features. For example, Manasypov et al. (2014) investigated the

451   hydrochemical characteristics of thermokarst ponds and lakes with different degrees of permafrost

452   influence located in the northern part of western Siberia, across a latitudinal gradient of ~900 km. The

453   authors evidenced a geographical gradient in trace element concentrations related to thermokarst lake

454   evolution (from small permafrost depressions to larger lakes), in relation to different degrees of carbon

455   and element leaching from frozen peat soils. They estimated that the acceleration of permafrost thaw in

456   Siberia might increase the amount of small thermokarst lakes and, in turn, increase the concentrations of

457   most trace elements by 200-400%. Shirokova et al. (2013) also reported a clear decrease in some trace

458   element concentrations (e.g., Fe, Al, Ti, Zr and rare earth elements) from thermokarst depressions, to

459   ponds, and finally, to lakes in the central part of western Siberia (Russia) (Fig. 7). They attributed this

460   pattern to two main factors: i) increase in water residence time in large lakes, which leads to more efficient

461   mineralisation of organo-mineral colloids by aerobic heterotrophic bacterioplankton; ii) decrease in peat

462   abrasion at the lake borders as the perimeter-volume ratio decreases with size. Furthermore, trace

463   element concentrations (e.g., Fe, Al, Ni, Pb) have been found to increase in hollows, depressions and

464   permafrost subsidences in an area characterised by thawing frozen peat in western Siberia, Russia (Loiko

465   et al., 2017). Here, the authors reported decreasing concentrations with increasing surface area of the

466   water body, along the hydrological continuum: soil water → hollows → depressions and permafrost

467   subsidences → thaw ponds → thermokarst lakes. Thermokarst lakes can also have an important role in

468   the storage of some trace elements (Al, Cr, Cd, Pb) comparable to, or even higher than, the transport of

469   these elements by rivers (western Siberia - Russia, Polishchuk et al., 2017). Hot summer periods, and

470 cycles of ice formation and melting have been reported to drive in-lake biological uptake and release of

471 some elements (Zn, Mn, Co, Mo, Cu, Si, Pb, Cd) (western Siberia - Russia, Pokrovsky et al., 2013), and

472 seasonal differences in trace element concentrations (western Siberia - Russia, Manasypov et al., 2015),

473 respectively. In addition, Audry et al. (2011) observed that in differently-developed thermokarst lakes in

474 western Siberia, sediments at the early stage of ecosystem development are a sink for water column Cu,

475 Zn, Cd, Pb and Sb due to authigenic sulphides precipitation, confirming the progressive depletion trends

476 of trace element concentrations in the water column during the chronosequence of lake maturation (from

477 permafrost subsidence to mature and drained lakes) reported by Pokrovsky et al. (2011), who analysed

478 20 lakes and small ponds of various sizes and ages in the same area. Conversely, the sediments are a

479 source of dissolved Co, Ni and As to the water column due to upward diffusive fluxes, regardless the

480 ecosystem development stage. However, the concentrations of these elements were found to be low due

481 to sorption processes on Fe-bounding particles and/or large-size organo-mineral colloids.

482 Several studies have been conducted focusing on Hg and Pb dynamics in thermokarst-affected areas.

483 For instance, Klaminder et al. (2008) assessed the impact of permafrost degradation on the increase in

484 Hg mobility from soils to lake surface water in a palsa peat complex in sub-Arctic Sweden. Main driving

485 factors were accelerated soil/peat erosion, increasing hydrological connectivity, and increased runoff

486 which led to the exposure of soluble elements. Gordon et al. (2016) (Northwest Territories - Canada)

487 further demonstrated that new hydrological pathways due to permafrost thaw can increase peatland

488 hydrological connectivity, creating areas of high Hg concentrations at critical points (fens) before water

489 discharged to a wider drainage system. Small permafrost thaw ponds also contribute in controlling the

490 local and regional fluxes of Hg, as demonstrated by MacMillan et al. (2015) in sub-Arctic and high-Arctic

491 Canada. Elevated concentrations of Hg and methylmercury have been found to be strongly related to

492 inputs of organic matter and nutrients into surface water. Moreover, Rydberg et al. (2010) reported that

493 long-term changes in climate, permafrost and mire dynamics had large effects on the release of Hg from

494 palsa mires to lakes in sub-arctic Sweden. Particularly, the recent thawing of permafrost has increased

495 thermokarst erosion, resulting in an enhanced Hg transport from the mire to lake water. In their study of

496 sub-Arctic Sweden, Klaminder et al. (2010) reported a similar process for Pb, attributing its increase to the

497 thermokarst erosion of peat. Conversely, Deison et al. (2012) found that lakes affected by the

498   development of retrogressive thaw slumps in the Mackenzie Delta (Canada) had lower Hg levels in

499   surface sediments when compared to not-affected lakes. The authors attributed these results to the

500   effects of thaw slumping of clay-rich permafrost tills, which resulted in high inorganic sedimentation rates

501   that caused a dilution of organic material and associated Hg in the sediments.

502

503   **5.3   Rock-glacier thawing**

504   An increase in trace element concentrations in high-elevation Alpine lakes (Central Eastern Alps,

505   Austria and Italy) has been observed, and it has been attributed to the influence of ice melting within rock

506   glaciers (Thies et al., 2007). For instance, unexpectedly high Ni, Mn and Al concentrations, exceeding the

507   European Union limits for drinking water by more than an order of magnitude, have been reported for a

508   remote high-mountain lake situated at the toe of an active rock glacier. In contrast, negligible

509   concentrations of these elements (and other solutes) have been recorded in an adjacent pond without a

510   rock glacier in the catchment (Central Eastern Alps - Italy, Ilyashuk et al., 2014). Surprisingly, the lithology

511   did not contain high Ni concentrations, thus the source of Ni remains unknown. In addition, other springs

512   derived from rock glaciers in the Central Eastern Alps exhibited high concentrations of trace elements

513   such as Ni, Co, Cu, Fe, Mn and Zn (Krainer, 2014), pointing to rock-glacier ice as a potential origin.

514   Increasing acidity of rock-glacier affected surface waters accompanied by high heavy metal

515   concentrations have been also reported to cause significant changes in aquatic ecosystems (Central

516   Eastern Alps - Austria, Thies et al., 2013) and severe ecological damages (Central Eastern Alps - Italy,

517   Ilyashuk et al., 2017).

518   Results from geochemical analyses of ice cores sampled from a medium-sized active rock glacier

519   (Lazaun) in Central Eastern Alps (Italy) are reported by Krainer et al. (2015). The ice near the base is

520   approximately 10,300 years old and trace elements (Ni, Co, Cu, Zn) are enriched in three horizons within

521   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

522   BP), with Ni concentrations up to 0.49 mg L^{-1}. High Ni concentrations were also measured at the rock

523   glacier spring, indicating that Ni in spring water was derived from distinct depth intervals within the

524   permafrost ice (Krainer et al., 2011). Again, the lithology was reported to not contain high concentrations

525   of trace elements. However, only few rock glaciers in Central Eastern Alps display meltwater with

526 abnormally high trace element concentrations. Considerable uncertainty still exists about the origin of the

527 high concentrations of trace elements in rock-glacier ice.

528

529 **6. Research perspectives**

530   Several studies report major ion distribution in near-surface permafrost, finding increasing

531 concentrations at the top of permafrost, generally corresponding with ice-rich layers. However, relatively

532 few researches have combined geochemical analyses on changing near-surface permafrost and resulting

533 effects on surface-water characteristics. Performing combined investigations may provide insight into the

534 relationship between concentrations of major ions in near-surface permafrost and modifications of

535 surface-water geochemistry in case of permafrost degradation, both on intra- and inter-annual bases. Few

536 studies in the Arctic also demonstrated that rainfall events can flush solutes from near-surface permafrost

537 to the surface and increase ion concentrations in surface water. Projected warming and rainfall

538 augmentation in Arctic areas (IPCC, 2013) might cause increases in major ion concentrations in surface

539 water in permafrost areas; in turn, increasing runoff might reduce ion concentrations since discharge is

540 generally negatively correlated with concentrations of major ions because of dilution effects (Petrone et

541 al., 2006, 2007), although the "chemostatic" behaviour in stream water (solute concentrations in stream

542 water are not determined by simple dilution, Godsey et al., 2009) should be also taken into account. The

543 ability to adequately incorporate these processes into models will be relevant for predicting the

544 hydrochemical impacts of climatic changes in transitional permafrost areas. Future studies should also

545 focus on the bedrock/sediment composition and particularly on the geochemical composition of

546 permafrost ice that might be locally enriched in ions (due to lithological characteristics), which can be

547 released when permafrost ice melts. Finally, research in highly sensitive areas such as watersheds where

548 acid drainage is possible due to lithological properties (e.g., pyritiferous shale presence) will help to

549 understand how permafrost degradation may cause a deterioration of surface water quality.

550   Major ion concentrations downstream of rock glaciers have mainly been investigated in the European

551 Alps and in mountain chains of North America. Extending these studies to areas such as in the Andes

552 (e.g., Brenning, 2005) and in the Hindu Kush Himalaya (e.g., Schmid et al., 2015; Gruber et al., 2017) will

553 allow better understanding the dynamics and potential effects of rock-glacier thaw on downstream water

554   quality. The establishment of baselines for future monitoring and the better understanding of processes of

555   solute export from rock glaciers will thereby be important elements of such studies. In this framework,

556   investigating the hydrological connections between rock glaciers and adjacent surface waters through the

557   application on non-invasive techniques (Colombo et al., 2017) could provide new insights on the rock-

558   glacier role, and dynamics, in exporting concentrated chemical fluxes into high-elevation hydrological

559   systems. Moreover, it has been shown that melting of ice in active rock glaciers can increase their storage

560   capacity. May chemical composition of outflowing water be affected by this process? Fresh water stored in

561   rock glaciers can be important for local water management only during the melt season (summer) since

562   during the cold season (winter) discharge is extremely low or absent. Future studies on water quality

563   originating from rock glaciers should also focus on relict rock glaciers due to their higher storage capacity

564   and residence time for fresh water which can also be released during winter. Finally, extending

565   investigations to other common debris features with permafrost such as talus slopes (Sass, 2006; Lambiel

566   and Pieracci, 2008; Scapozza et al., 2011) will allow understanding the effects of thawing permafrost on

567   water quality in cold mountains more broadly. Do talus slopes with significant amount of ground ice (cf.,

568   Gruber and Haeberli, 2009) behave like rock glaciers in their export of solutes during permafrost

569   degradation? Trends of major ions and ion-isotope ratios in surface water may be considered as important

570   additions to the permafrost monitoring conducted in mountains (c.f., PERMOS, 2016).

571   Given that most studies on geochemistry in near-surface permafrost and ground ice considered only

572   major ions, future research on trace element concentrations and their variations within the active layer,

573   permafrost and massive ground ice occurrences is necessary for evaluating the effect they might have on

574   ecosystems when thawing.

575   High trace element concentrations have been reported downstream of rock glaciers and in three

576   horizons within the ice core of a rock glacier in the Central Eastern Alps. Since lithology was not found to

577   contain high trace element concentrations, alternative hypotheses (e.g., atmospheric fallout) to chemical

578   weathering should be tested in order to understand the origin and the potential for future release of trace

579   elements.

580   Beside specific research needs, several broader questions emerge for the understanding of permafrost

581   degradation and its effects on inorganic chemistry of surface water. These are especially important in arid

582 areas, such as some areas in the Andes and Himalaya, where fresh water stored in permafrost can be

583 important for local water management (Brenning, 2005; Rangecroft et al., 2013; Rangecroft et al., 2015;

584 Gruber et al., 2017) in small catchments. What is the role of permafrost in controlling water quality in these

585 areas and what duration will the transient effects of permafrost thaw have? What will be the long-term

586 implications of permafrost degradation on water quality of local freshwater reservoirs? What are the

587 possible socioeconomic consequences of water quality deterioration due to permafrost degradation? What

588 are suitable adaptation strategies to mitigate the possible impacts of permafrost degradation on water

589 supply quality of local populations?

590

591 **7. Conclusion**

592 This review of permafrost-thaw impacts on inorganic chemistry of surface fresh water has revealed

593 several common patterns.

594 Spatially-distributed progressive increases of major ion delivery to surface fresh water have been

595 reported in both polar and mountain areas following permafrost degradation. This is the result of

596 increasing contributions of highly mineralised groundwater and enhanced interactions of soil water with

597 deep mineral strata. Localised releases of major ions to surface water due to the liberation of soluble

598 materials sequestered in permafrost and ground ice have been found in ice-rich terrains both at high

599 latitude (thermokarst features) and high elevation (rock glaciers). Generalisations must be considered

600 carefully though, because the intensity of hydrochemical modifications depends on several factors such as

601 the characteristics of the watersheds (e.g., physical and geochemical characteristics of permafrost), their

602 hydrological connectivity, the magnitude and type of disturbance in case of thermokarst, and the extent of

603 permafrost or rock glaciers in the catchment. Permafrost degradation can also increase the export of

604 inorganic N and P forms, and Si, from permafrost to surface water due to deeper flow paths that bypass

605 shallow organic layers where most retention and removal of nutrients occur (N), and because of enhanced

606 mineral weathering (P and Si). However, significant uncertainty has been shown on DIN dynamics, mainly

607 dependent on regional and landscape-scale differences in Arctic areas (e.g., Alaska and Siberia). In

608 addition, thermokarst can enhance the export of inorganic nutrients from permafrost to surface water,

609 although biological uptake, sedimentation and degradation of nutrients might mitigate these effects.

610   Increases in nitrate concentrations have been also attributed to rock glacier melt water flushing microbially

611   active sediments.

612   Trace element dynamics in permafrost areas have been shown to exhibit two seasonal peaks in

613   geochemical fluxes: one at the beginning of the snowmelt (early spring), related to the flushing of surficial

614   soil horizons and previously mobilised species stored in them; the second occurring in late summer and

615   early fall, associated with deeper mineral-soil horizons. The formation of new thermokarst features (e.g.,

616   ponds, lakes, palsa mires) and leaching from frozen peat can be relevant for increasing trace element

617   concentrations in permafrost catchments and for affecting the locations of high concentrations due to

618   newly-developing hydrological pathways. Alternatively, increasing sediment erosion due to thermokarst

619   processes has been reported to potentially mitigate these effects through increasing sedimentation rates

620   and dilution of concentrations. Moreover, stabilisation and increasing size of older thermokarst features

621   might lead to decreasing trends because of stabilising shorelines, reduced peat abrasion, and more

622   efficient mineralisation. Increasing trends in trace element concentrations have been also reported in

623   streams and lakes in mountain areas, where enhanced chemical weathering on fresh mineral surfaces

624   exposed by permafrost thaw and contribution of metal-enriched groundwater have been hypothesised to

625   be the main drivers. Nevertheless, in the Central Eastern Alps, high concentrations of trace elements

626   (e.g., Ni) in surface water have been found to be unrelated to the lithological setting, hence rejecting the

627   hypothesis of mineral weathering as the only process involved.

628   In conclusion, several factors influence the timing and magnitude of the impacts of permafrost

629   degradation on inorganic chemistry of surface fresh water. These include permafrost sensitivity to thawing

630   (e.g., amount of ground ice), modes of permafrost degradation (e.g., increasing ALT, thermokarst) and

631   environmental characteristics of watersheds (e.g., soil properties, lithology, land cover, topography,

632   hydrological connectivity). Further mobilisation of solutes and related transport to surface fresh water

633   appear reasonable under warming climatic conditions. This might develop through gradual concentration

634   increase in case of progressive and diffused thawing of permafrost, and/or through distinct temporal peaks

635   in case of localised thermokarst and rock glacier thawing. As a result, modifications of inorganic water

636   chemistry might have significant implications for ecological and human systems.

637

Reference list

Abbott, B.W., Jones, J.B., Godsey, S.E., Larouche, J.R., Bowden, W.B., 2015. Patterns and persistence of hydrologic carbon and nutrient export from collapsing upland permafrost. Biogeosciences 12, 3725-3740.

ACGR (Associate Committee On Geotechnical Research), 1988. Glossary of Permafrost and Related Ground Ice Terms. Permafrost Subcommittee, National Research Council of Canada, Technical Memorandum 142.

Åkerman, H.J.A., Johansson, M., 2008. Thawing permafrost and thicker active layers in subarctic Sweden. Permafrost and Periglacial Processes 19, 279-92.

Andresen, C.G., Lougheed, V.L., 2015. Disappearing Arctic tundra ponds: Fine-scale analysis of surface hydrology in drained thaw lake basins over a 65 year period (1948-2013). Journal of Geophysical Researches 120, 466-479.

Audry, S., Pokrovsky, O.S., Shirokova, L.S., Kirpotin, S.N., Dupré, B., 2011. Organic matter mineralization and trace element post-depositional redistribution in Western Siberia thermokarst lake sediments. Biogeosciences 8: 3341-3358.

Azócar, G.F., Brenning, A., 2010. Hydrological and geomorphological significance of rock glaciers in the dry Andes, Chile (27°-33° S). Permafrost and Periglacial Processes 21, 42-53.

Bagard, M.-L., Chabaux, F., Pokrovsky, O.S., Viers, J., Prokushkin, A.S., Stille, P., Rihs, S., Schmit, A.D., Dupre, B., 2011. Seasonal variability of element fluxes in two Central Siberian rivers draining high latitude permafrost dominated areas. Geochimica et Cosmochimica Acta 75, 3335-3357.

664   Bagard, M.-L., Schmitt, A.-D., Chabaux, F., Pokrovsky, O.S., Viers, J., Stille, P., Labolle, F., Prokushkin,

665   A.S., 2013. Biogeochemistry of stable Ca and radiogenic Sr isotopes in a larch-covered permafrost-

666   dominated watershed of Central Siberia. Geochimica et Cosmochimica Acta 114, 169-187.

667   Barker, A.J., Douglas, T.A., Jacobson, A.D., McClelland, J.W., Ilgen, A.G., Khosh, M.S., Lehn, G.O.,

668   Trainor, T.P., 2014. Late season mobilization of trace metals in two small Alaskan arctic watersheds as

669   a proxy for landscape scale permafrost active layer dynamics. Chemical Geology 381, 180-193.

670   Barnes, R.T., Williams, M.W., Parman, J.N., Hill, K., Caine, N., 2014. Thawing glacial and permafrost

671   features contribute to nitrogen export from Green Lakes Valley, Colorado Front Range, USA.

672   Biogeochemistry 117, 413-430.

673   Baron, J.S., Schmidt, T.M., Harman, M.D., 2009. Climate-induced changes in high-elevation stream

674   nitrate dynamics. Global Change Biology 15 (7), 1777-1789.

675   Barsch, D., 1996. Rockglaciers: Indicators for the Present and Former Geoecology in High Mountain

676   Environments. Springer-Verlag: Berlin-Heidelberg.

677   Bense, V.F., Ferguson, G., Kooi, H., 2009. Evolution of shallow groundwater flow systems in areas of

678   degrading permafrost. Geophysical Research Letters 36, L22401 (doi:10.1029/2009GL039225).

679   Berger, J., Krainer, K., Mostler, W., 2004. Dynamics of an active rockglacier (Ötztal Alps, Austria).

680   Quaternary Research 62, 233-242.

681   Berthling, I., 2011. Beyond confusion: Rock glaciers as cryo-conditioned landforms. Geomorphology 131,

682   98-106.

683   Boeckli, L., Brenning, A., Gruber, S., Noetzli, J., 2012. Permafrost distribution in the European Alps:

684   calculation and evaluation of an index map and summary statistics, The Cryosphere 6, 807-820.

685   Bowden, W.B., Gooseff, M.N., Balser, A., Green, A., Peterson, B.J., Bradford, J., 2008. Sediment and

686   nutrient delivery from thermokarst features in the foothills of the North Slope, Alaska: Potential impacts

687   on headwater stream ecosystems. Journal of Geophysical Research 113, G02026

688   (doi:10.1029/2007JG000470).

689   Brenning, A., 2005. Geomorphological, hydrological, and climatic significance of rock glaciers in the

690   Andes of Central Chile (33-35 S). Permafrost and Periglacial Processes 16, 231-240.

691   Burger, K.C., Degenhardt, J.J., Giardino, J.R., 1999. Engineering geomorphology of rock glaciers.

692   Geomorphology 31, 93-132.

693   Burn, C.R., Zhang, Y., 2009. Permafrost and climate change at Herschel Island (Qikiqtaruk), Yukon

694   Territory, Canada. Journal of Geophysical Research 114, F02001 (doi: 10.1029/2008JF001087).

695   Caine, N., 2010. Recent hydrologic change in a Colorado alpine basin: an indicator of permafrost thaw?

696   Annals of Glaciology 51 (56), 130-134.

697   Clark, I.D., Lauriol, B., Harwood, L., Marschner, M., 2001. Groundwater contributions to discharge in a

698   permafrost setting, Big Fish River, N.W.T., Canada. Arctic Antarctic and Alpine Research 33, 62-69.

699   Colombo, N., Sambuelli, L., Comina, C., Colombero, C., Giardino, M., Gruber, S., Viviano, G., Vittori

700   Antisari, L., Salerno, F., 2017. Mechanisms linking active rock glaciers and impounded surface water

701   formation in high-mountain areas. Earth Surface Processes and Landforms, doi:10.1002/esp.4257.

702   Corte, A., 1976. The hydrological significance of rock glaciers. Journal of Glaciology 17, 157-158.

703   Cremonese, E., Gruber, S., Phillips, M., Pogliotti, P., Boeckli, L., Noetzli, J., Suter, C., Bodin, X., Crepaz,

704   A., Kellerer-Pirklbauer, A., Lang, K., Letey, S., Mair, V., Morra di Cella, U., Ravanel, L., Scapozza, C.,

705   Seppi, R., Zischg, A., 2011. Brief Communication: "An inventory of permafrost evidence for the

706   European Alps". The Cryosphere 5, 651-657.

707   Deison, R., Smol, J.P., Kokelj, S.V., Pisaric, M.F.J., Kimpe, L.E., Poulain, A.J., Sanei, H., Thienpont, J.R.,

708   Blais, J.M., 2012. Spatial and temporal assessment of mercury and organic matter in thermokarst

709   affected lakes of the Mackenzie Delta Uplands, NT, Canada. Environmental Science & Technology 46,

710   8748-8755.

711   Douglas, T.A., Blum, J.D., Guo, L., Keller, K., Gleason, J.D., 2013. Hydrogeochemistry of seasonal flow

712   regimes in the Chena River, a subarctic watershed draining discontinuous permafrost in interior Alaska

713   (USA). Chemical Geology 335, 48-62.

714   Dugan, H.A., Lamoureux, S.F., Lewis, T., Lafrenière, M.J., 2012. The Impact of permafrost disturbances

715   and sediment loading on the Limnological characteristics of two High Arctic lakes. Permafrost and

716   Periglacial Processes 23 (2), 119-126.

717   Fegel, T.S., Baron, J.S., Fountain, A.G., Johnson, G.F., Hall, E.K., 2016. The differing biogeochemical

718   and microbial signatures of glaciers and rock glaciers. Journal of Geophysical Research 121, 919-932.

719   Fountain, A.G., Campbell, J.L., Schuur, E.A.G., Stammerjohn, S.E., Williams, M.W., Ducklow, H.W., 2012.

720   The Disappearing Cryosphere: Impacts and Ecosystem Responses to Rapid Cryosphere Loss.

721   BioScience 62, 405-415.

722   Frauenfeld, O.W., Zhang, T.J., Barry, R.G., Gilichinsky, D., 2004. Interdecadal changes in seasonal freeze

723   and thaw depths in Russia. Journal of Geophysical Research 109, D05101

724   (doi:10.1029/2003JD004245).

725   French, H.M., 2007. The Periglacial Environment. Third edition, John Wiley & Sons, Chichester.

726   Frey, K.E., McClelland, J.W., 2009. Impacts of permafrost degradation on arctic river biogeochemistry.

727   Hydrological Processes 23, 169-182.

728   Frey, K.E., McClelland, J.W., Holmes, R.M., Smith, L.C., 2007b. Impacts of climate warming and

729   permafrost thaw on the riverine transport of nitrogen and phosphorus to the Kara Sea. Journal of

730   Geophysical Research 112, G04S58 (doi:10.1029/2006JG000369).

731   Frey, K.E., Siegel, D.I., Smith, L.C., 2007a. Geochemistry of west Siberian streams and their potential

732   response to permafrost degradation. Water Resources Research 43, W03406

733   (doi:10.1029/2006WR004902).

734   Giardino, J.R., Vitek, J.D., DeMorett, J.L., 1992. A model of water movement in rock glaciers and

735   associated water characteristics. Proceedings of the 22nd Annual Binghamton Symposium in

736   Geomorphology, 159-184.

737   Giesler, R., Lyon, S.W., Mörth, C.-M., Karlsson, J., Karlsson, E.M., Jantze, E.J., Destouni, G., Humborg,

738   C., 2014. Catchmentscale dissolved carbon concentrations and export estimates across six subarctic

739   streams in northern Sweden, Biogeosciences 11, 525-537.

740   Godsey, S.E., Kirchner, J.W., Clow, D.W., 2009. Concentration-discharge relationships reflect chemostatic

741   characteristics of US catchments. Hydrological Processes 23 (13), 1844-1864.

742   Gooseff, M.N., Balser, A., Bowden, W.B., Jones, J.B., 2009. Effects of hillslope thermokarst in Northern

743   Alaska. Eos, Transactions American Geophysical Union 90 (4), 29-30.

744   Gooseff, M.N., Van Horn, D., Sudman, Z., McKnight, D.M., Welch, K.A., Lyons, W.B., 2016. Stream

745   biogeochemical and suspended sediment responses to permafrost degradation in stream banks in

746   Taylor Valley, Antarctica. Biogeosciences 13, 1723-1732.

747   Gorbunov, A.P., 1978. Permafrost investigations in high-mountain regions. Arctic and Alpine Research 10,

748   283-294.

749   Gordon, J., Quinton, W., Branfireun, B.A., Olefeldt, D., 2016. Mercury and methylmercury biogeochemistry

750   in a thawing permafrost wetland complex, Northwest Territories, Canada. Hydrological Processes 30,

751   3627-3638.

752   Gruber, S., 2012. Derivation and analysis of a high-resolution estimate of global permafrost zonation. The

753   Cryosphere 6, 221-233.

754   Gruber, S., Fleiner, R., Guegan, E., Panday, P., Schmid, M.-O., Stumm, D., Wester, P., Zhang, Y., Zhao,

755   L., 2017. Review article: Inferring permafrost and permafrost thaw in the mountains of the Hindu Kush

756   Himalaya region. The Cryosphere 11, 81-99.

757   Gruber, S., Haeberli, W., 2009. Mountain permafrost. In R. Margesin (Ed.), *Permafrost Soils.* Berlin,

758   Heidelberg: Springer Berlin Heidelberg, 33–44. (doi: 10.1007/978-3-540-69371-0).

759   Gruber, S., Hoelzle, M., Haeberli, W., 2004. Permafrost thaw and destabilization of Alpine rock walls in the

760   hot summer of 2003. Geophysical Research Letters 31, L13504 (doi:10.1029/2004GL020051).

761   Guglielmin, M., Cannone, N., 2012. A permafrost warming in a cooling Antarctica? Climatic Change 111,

762   177-195.

763   Guglielmin, M., Dalle Fratte, M., Cannone, N., 2014. Permafrost warming and vegetation changes in

764   continental Antarctica. Environmental Research Letters 9, 045001 (doi:10.1088/1748-

765   9326/9/4/045001).

766   Haeberli, W., Hallet, B., Arenson, L., Elconin, R., Humlum, O., Kääb, A., Kaufmann, V., Ladanyi, B.,

767   Matsuoka, N., Springman, S., and Vonder Mühll, D., 2006. Permafrost creep and rock glacier

768   dynamics. Permafrost and Periglacial Processes 17, 189-214.

769   Hansen, J., Sato M., 2016. Regional climate change and national responsibilities. Environmental

770   Research Letters 11: 034009 (doi:10.1088/1748-9326/11/3/034009).

771   Harden, J.W., Koven, C.D., Ping, C., Hugelius, G., McGuire, A.D., Camill, P., Jorgenson, T., Kuhry, P.,

772   Michaelson, G.J., O'Donnell, J.A., Schuur, E.A.G., Tarnocai, C., Johnson, K., Grosse, G., 2012. Field

773   information links permafrost carbon to physical vulnerabilities of thawing. Geophysical Research

774   Letters 39, L15704 (doi:10.1029/2012GL051958).

775   Harms, T.K., Abbott, B.W., Jones, J.B., 2014. Thermo-erosion gullies increase nitrogen available for

776   hydrologic export. Biogeochemistry 117 (2-3), 299-311.

777   Harms, T.K., Jones Jr., J.B., 2012. Thaw depth determines reaction and transport of inorganic nitrogen in

778   valley bottom permafrost soils. Global Change Biology 18, 2958-2968.

779   Harris, H., Arenson, L.U., Christiansen, H.H., Etzelmüller, B., Frauenfelder, R., Gruber, S., Haeberli, W.,

780   Hauck, C., Hoelzle, M., Humlum, O., Isaksen, K., Kääb, A., Kern-Lütschg, M.A., Lehning, M.,

781   Matsuoka, N., Murton, J., Noetzli, J., Phillips, M., Ross, N., Seppälä, M., Springman, S.M., Vonder

782   Mühll, D., 2009. Permafrost and climate in Europe: Monitoring and modelling thermal,

783   geomorphological and geotechnical responses. Earth-Science Reviews 92, 117-171.

784   Harris, K.J., Carey, A.E., Lyons, W.B., Welch, K.A., Fountain, A.G., 2007. Solute and isotope

785   geochemistry of subsurface ice melt seeps in Taylor Valley, Antarctica. Geological Society of America

786   Bulletin 119 (5-6), 548-555.

787   He, M., Wang, Z., Tang, H., 1997. Spatial and temporal patterns of acidity and heavy metals in predicting

788   the potential for ecological impact on the Le An river polluted by acid mine drainage. Science of the

789   Total Environment 206, 67-77.

790   Heginbottom, J.A., Dubreuil, M.A., Harker, P.A., 1995. Canada-Permafrost. National Atlas of Canada, 5th

791   Edition. National Atlas Information Service, Natural Resources Canada, Ottawa, Plate 2.1 MCR 4177.

792   Hinzman, L.D., Bettez, N.D., Bolton, W.R., Chapin, F.S., Dyurgerov, M.B., Fastie, C. L., Griffith, B.,

793   Hollister, R.D., Hope, A., Huntington, H.P., Jensen, A.M., Jia, G.J., Jorgenson, T., Kane, D.L., Klein,

794   D.R., Kofinas, G., Lynch, A.H., Lloyd, A.H., McGuire, A.D., Nelson, F.E., Oechel, W.C., Osterkamp,

795   T.E., Racine, C.H., Romanovsky, V.E., Stone, R.S., Stow, D.A., Sturm, M., Tweedie, C.E., Vourlitis,

796   G.L., Walker, M. D., Walker, D.A., Webber, P.J., Welker, J.M., Winker, K.S., Yoshikawa, K., 2005.

797   Evidence and implications of recent climate change in northern Alaska and other Arctic regions.

798   Climatic Change 72, 251-298.

799   Hirst, C., Andersson, P.S., Shaw, S., Burke, I.T., Kutscher, L., Murphy, M.J., Maximov, T., Pokrovsky,

800   O.S., Mörth, C-M., Porcelli, D., 2017. Characterisation of Fe-bearing particles and colloids in the Lena

801   River basin, NE Russia. Geochimica et Cosmochimica Acta 213, 553-573.

802   Hobbie, J.E., Peterson, B.J., Bettez, N., Deegan, L., O'Brien, W.J., Kling, G.W., Kipphut, G.W., Bowden,

803   W.B., Hershey, A.E., 1999. Impact of global change on the biogeochemistry and ecology of an Arctic

804   freshwater system. Polar Research 18 (2), 207-214.

805   Humlum, O. (2000). The geomorphic significance of rock glaciers: estimates of rock glacier debris

806   volumes and headwall recession rates in West Greenland. Geomorphology 35, 41-67.

807   Ikeda, A., Matsuoka, N., 2006. Pebbly versus bouldery rock glaciers: Morphology, structure and

808   processes. Geomorphology 73, 279-296.

809   Ilyashuk, B.P., Ilyashuk, E.A., Psenner, R., Tessadri, R., Koinig, K.A., 2014. Rock glacier outflows may

810   adversely affect lakes: lessons from the past and present of two neighboring water bodies in a

811   crystalline-rock watershed. Environmental Science & Technology 48, 6192-6200.

812   Ilyashuk, B.P., Ilyashuk, E.A., Psenner, R., Tessadri, R., Koinig, K.A., 2017. Rock glaciers in crystalline

813   catchments: hidden permafrost-related threats to alpine headwater lakes. Global Change Biology,

814   doi:10.1111/gcb.13985.

815   Ishikawa, M., Sharkhuu, N., Zhang, Y., Kadota, T., and Ohata, T., 2005. Ground thermal and moisture

816   conditions at the southern boundary of discontinuous permafrost, Mongolia. Permafrost and Periglacial

817   Processes 16: 209-216.

818   IPCC, 2013. Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the

819   Fifth Assessment Report of the Intergovernmental Panel on Climate Change. Cambridge University

820   Press, Cambridge, United Kingdom and New York, NY, USA.

821   Jessen, S., Holmslykke, H.D., Rasmussen, K., Richardt, N., Holm, P.E., 2014. Hydrology and pore water

822   chemistry in a permafrost wetland, Ilulissat, Greenland. Water Resources Research 50 (6), 4760-4774.

823   Jones Jr., J.B., Petrone, K.C., Finlay, J.C., Hinzman, L.D., Bolton, W.R., 2005. Nitrogen loss from

824   watersheds of interior Alaska underlain with discontinuous permafrost. Geophysical Research Letters

825   32, L02401 (doi:10.1029/2004GL021734).

826   Jorgenson, M.T., Osterkamp, T.E., 2005. Response of boreal ecosystems to varying modes of permafrost

827   degradation. Canadian Journal of Forest Research 35, 2100-2111.

828   Jorgenson, M.T., Racine, C.H., Walters, J.C., Osterkamp, T.E., 2001. Permafrost degradation and

829   ecological changes associated with a warming climate in Central Alaska. Climatic Change 48, 551-579.

830   Kääb, A., Haeberli, W., 2001. Evolution of a high mountain thermokarst lake in the Swiss Alps. Arctic,

831   Antarctic and Alpine Research 33(4), 385-390.

832   Kane, D.L., Yoshikawa, K., McNamara, J.P., 2013. Regional groundwater flow in an area mapped as

833   continuous permafrost, NE Alaska (USA). Hydrogeology Journal 21, 41-52.

834   Keller, K., Blum, J.D., Kling, G.W., 2007. Geochemistry of soils and streams on surfaces of varying ages

835   in Arctic Alaska. Arctic, Antarctic, and Alpine Research 39, 84-98.

836   Keller, K., Blum, J.D., Kling, G.W., 2010. Stream geochemistry as an indicator of increasing permafrost

837   thaw depth in an arctic watershed. Chemical Geology 273 (1–2), 76-81.

838   Klaminder, J., Hammarlund, D., Kokfelt, U., Vonk, J.E., Bigler, C., 2010. Lead contamination of subarctic

839   lakes and its response to reduced atmospheric fallout: can the recovery process be counteracted by

840   the ongoing climate change? Environmental Science & Technology 44, 2335-2340.

841   Klaminder, J., Yoo, K., Rydberg, J., Giesler, R., 2008. An explorative study of mercury export from a

842   thawing palsa mire. Journal of Geophysical research 113, G04034 (doi:10.1029/2008JG000776).

843   Koch, J.C., Runkel, R.L., Striegl, R., McKnight, D.M., 2013. Hydrologic controls on the transport and

844   cycling of carbon and nitrogen in a boreal catchment underlain by continuous permafrost. Journal of

845   Geophysical Research 118, 698-712.

846   Kokelj, S.V., Burn, C.R., 2003. Ground ice and soluble cations in near-surface permafrost, Inuvik,

847   Northwest Territories, Canada. Permafrost and Periglacial Processes 143, 275-289.

848   Kokelj, S.V., Burn, C.R., 2005. Geochemistry of the active layer and near-surface permafrost, Mackenzie

849   delta region, Northwest Territories, Canada. Canadian Journal of Earth Sciences 42, 37-48.

850   Kokelj, S.V., Jenkins, R.E., Milburn, D., Burn, C.R., Snow, N., 2005. The influence of thermokarst

851   disturbance on the water quality of small upland lakes, Mackenzie Delta region, Northwest Territories,

852   Canada. Permafrost and Periglacial Processes 16, 343-353.

853   Kokelj, S., Jorgenson, M.T., 2013. Advances in thermokarst research. Permafrost and Periglacial

854   Processes 24, 108-119.

855   Kokelj, S.V., Lacelle, D., Lantz, T.C., Tunnicliffe, J., Malone, L., Clark, I.D., Chin, K.S., 2013. Thawing of

856   massive ground ice in mega slumps drives increases in stream sediment and solute flux across a

857   range of watershed scales. Journal of Geophysical Research 118, 681-692.

858   Kokelj, S.V., Lewkowicz, A.G., 1999. Salinization of permafrost terrain due to natural geomorphic
859   disturbance, Fosheim Peninsula, Ellesmere Island. Arctic 52, 372-385.

860   Kokelj, S.V., Smith, C.A.S., Burn, C.R., 2002. Physical and chemical characteristics of the active layer and
861   permafrost, Herschel Island, western Arctic coast, Canada. Permafrost and Periglacial Processes 13,
862   171-185.

863   Kokelj, S.V., Zajdlik, B., Thompson, M.S., 2009. The impacts of thawing permafrost on the chemistry of
864   lakes across the subarctic boreal tundra transition, Mackenzie Delta region, Canada. Permafrost and
865   Periglacial Processes 20, 185-200.

866   Kolosov, R.R., Prokushkin, A.S., Pokrovsky, O.S., 2016. Major anion and cation fluxes from the Central
867   Siberian Plateau watersheds with underlying permafrost. IOP Conference Series: Earth and
868   Environmental Science 48, 012018, doi:10.1088/1755-1315/48/1/012018.

869   Krainer, K., 2014. Permafrost and Climate Change in North and South Tyrol. In: permAfrost, Austrian
870   Permafrost Research Initiative, Final Report, 51-67.

871   Krainer, K., Bressan, D., Dietre, B., Haas, J.N., Hajdas, I., Lang, K., Mair, V., Nickus, U., Reidl, D., Thies,
872   H., Tonidandel, D., 2015. A 10,300-year-old permafrost core from the active rock glacier Lazaun,
873   southern Ötztal Alps (South Tyrol, northern Italy). Quaternary Research 83(2), 324-335.

874   Krainer, K., Mostler, W. 2002. Hydrology of active rock glaciers: Examples from the Austrian Alps. Arctic,
875   Antarctic, and Alpine Research 34, 142-149.

876   Krainer, K., Mostler, W., Spötl, C., 2007. Discharge from active rock glaciers, Austrian Alps: a stable
877   isotope approach. Austrian Journal of Earth Sciences 100, 102-112.

878   Krainer, K., Nickus, U., Thies, H., Tessadri, R., 2011. Geochemical analyses of permafrost ice and of
879   permafrost water springs. PermaNET Project report, WP7.

880   Kurylyk, B.L., MacQuarrie, K.T.B., McKenzie, J.M., 2014. Climate change impacts on groundwater and
881   soil temperatures in cold and temperate regions: Implications, mathematical theory, and emerging
882   simulation tools. Earth-Science Reviews 138, 313-334.

883   Lacelle, D., Doucet, A., Clark, I.D., Lauriol, B., 2007. Acid drainage generation and seasonal recycling in
884   disturbed permafrost near Eagle Plains, northern Yukon Territory, Canada. Chemical Geology 243,
885   157-177.

886 Lacelle, D., Fontaine, M., Forest, A.P., Kokelj, S.V., 2014. High-resolution stable water isotopes as tracers

887 of thaw unconformities in permafrost: A case study from western Arctic Canada. Chemical Geology

888 368, 85-96.

889 Lacelle, D., Juneau, V., Pellerin, A., Lauriol, B., Clark, I.D., 2008. Weathering regime and geochemical

890 conditions in a polar desert environment, Haughton impact structure region, Devon Island, Canada.

891 Canadian Journal of Earth Sciences 45, 1139-1157.

892 Lafrenière, M.J., Lamoureux, S.F., 2013. Thermal perturbation and rainfall runoff have greater impact on

893 seasonal solute. Permafrost and Periglacial Processes 24, 241-251.

894 Lambiel, C., Pieracci, K., 2008. Permafrost distribution in talus slopes located within the alpine periglacial

895 belt, Swiss Alps. Permafrost and Periglacial Processes 19, 293-304.

896 Lamhonwah, D., Lafrenière, M.J., Lamoureux, S.F., Wolfe, B.B., 2017. Evaluating the hydrological and

897 hydrochemical responses of a High Arctic catchment during an exceptionally warm summer.

898 Hydrological Processes 31(12), 2296-2313.

899 Lecomte, K.L., Milana, J.P., Formica, S.M., Depetris, P.J., 2008. Hydrochemical appraisal of ice- and rock-

900 glacier meltwater in the hyperarid Agua Negra drainage basin, Andes of Argentina. Hydrological

901 Processes 22, 2180-2195.

902 Leibman, M.O., Streletskaya, I.D., 1997. Landslide induced changes in the chemical composition of

903 active-layer soils and surface-water runoff, Yamal Peninsula, Russia. Proceedings of the International

904 Symposium on Physics, Chemistry and Ecology of Seasonally Frozen Soils, 120-126.

905 Levy, J.S., Fountain, A.G., Gooseff, M.N., Welch, K.A., Lyons, W.B., 2011. Water tracks and permafrost in

906 Taylor Valley, Antarctica: Extensive and shallow groundwater connectivity in a cold desert ecosystem.

907 Geological Society of America Bulletin 123 (11-12), 2295-2311.

908 Levy, J.S., Fountain, A.G., Welch, K.A., Berry Lyons, W., 2012. Hypersaline "wet patches" in Taylor

909 Valley, Antarctica. Geophysical Research Letters 39, L05402 (doi:10.1029/2012GL050898).

910 Ley, R.E., Williams, M.W., Schmidt, S.K., 2004. Microbial population dynamics in an extreme environment:

911 controlling factors in talus soils at 3,750 m in the Colorado Rocky Mountains. Biogeochemistry 68 (3),

912 313-335.

913   Lewis, T., Lafrenière, M.J., and Lamoureux, S.F., 2012. Hydrochemical and sedimentary responses of

914   paired High Arctic watersheds to unusual climate and permafrost disturbance, Cape Bounty, Melville

915   Island, Canada. Hydrological Processes 26 (13), 2003-2018.

916   Liljedahl, A.K. Boike, J., Daanen, R.P., Fedorov, A.N., Frost, G.V., Grosse, G., Hinzman, L.D., Iijma, Y.,

917   Jorgenson, J.C., Matveyeva, N., Necsoiu, M., Raynolds, M.K., Romanovsky, V.E. Schulla, J., Tape,

918   K.D., Walker, D.A., Wilson, C.J., Yabuki, H., Zona, D., 2016. Pan-Arctic ice-wedge degradation in

919   warming permafrost and its influence on tundra hydrology. Nature Geoscience 9,

920   (doi :10.1038/NGEO2674).

921   Loiko, S.V., Pokrovsky, O.S., Raudina, T.V., Lim, A., Kolesnichenko, L.G., Shirokova, L.S., Vorobyev,

922   S.N., Kirpotin, S.N., 2017. Abrupt permafrost collapse enhances organic carbon, CO_2, nutrient and

923   metal release into surface waters. Chemical Geology 471, 153-165.

924   Louiseize, N.L., Lafrenière, M.J., Hastings, M.G., 2014. Stable isotopic evidence of enhanced export of

925   microbially derived NO3- following active layer slope disturbance in the Canadian High Arctic.

926   Biogeochemistry 121, 565-580.

927   Luo, D., Wu, Q., Jin, H., Marchenko, S.S., Lü, L., Gao, S., 2016. Recent changes in the active layer

928   thickness across the northern hemisphere. Environmental Earth Sciences 75:555 (doi:10.1007/s12665-

929   015-5229-2).

930   MacLean, R., Oswood, M.W., Irons, J.S., McDowell, W.H., 1999. The effect of permafrost on stream

931   biogeochemistry: A case study of two streams in the Alaskan (U.S.A.) taiga. Biogeochemistry 47, 239-

932   267.

933   MacMillan, G.A., Girard, C., Chételat, J., Laurion, I., Amyot, M., 2015. High methylmercury in Arctic and

934   subarctic ponds is related to nutrient levels in the warming eastern Canadian Arctic. Environmental

935   Science & Technology 49, 7743-7753.

936   Malone, L., Lacelle, D., Kokelj, S., Clark, I.D., 2013. Impacts of hillslope thaw slumps on the geochemistry

937   of permafrost catchments (Stony Creek watershed, NWT, Canada). Chemical Geology 356, 38-49.

938   Manasypov, R.M., Pokrovsky, O.S., Kirpotin, S.N., Shirokova, L.S., 2014. Thermokarst lake water across

939   the permafrost zones of western Siberia. The Cryosphere 8, 1177-1193.

940   Manasypov, R.M., Vorobyev, S.N., Loiko, S.V., Kritzkov, I.V., Shirokova, L.S., Shevchenko, V.P., Kirpotin,

941   S.N., Kulizhsky, S.P., Kolesnichenko, L.G., Zemtzov, V.A., Sinkinov, V.V., Pokrovsky, O.S., 2015.

942   Seasonal dynamics of organic carbon and metals in thermokarst lakes from the discontinuous

943   permafrost zone of western Siberia. Biogeosciences 12, 3009-3028.

944   Mast, M.A., Turk, J.T., Clow, D.W., Campbell, D.H., 2011. Response of lake chemistry to changes in

945   atmospheric deposition and climate in three high-elevation wilderness areas of Colorado.

946   Biogeochemistry 103 (1-3), 27-43.

947   Mavromatis, V. Prokushkin, A.S., Pokrovsky, O.S., Viers, J., Korets, M.A., 2014. Magnesium isotopes in

948   permafrost-dominated Central Siberian larch forest watersheds. Geochimica et Cosmochimica Acta

949   147, 76-89.

950   Mavromatis, V., Rinder, T., Prokushkin, A.S., Pokrovsky, O.S., Korets, M.A., Chmeleff, J., Oelkers, E.H.,

951   2016. The effect of permafrost, vegetation, and lithology on Mg and Si isotope composition of the

952   Yenisey River and its tributaries at the end of the spring flood. Geochimica et Cosmochimica Acta 191,

953   32-46.

954   McClelland, J.W., Stieglitz, M., Pan, F., Holmes, R.M., Peterson, B.J., 2007. Recent changes in nitrate

955   and dissolved organic carbon export from the upper Kuparuk River, North Slope, Alaska. Journal of

956   Geophysical Research 112: G04S60 (doi:10.1029/2006JG000371).

957   McNamara, J.P., Kane, D.L., Hinzman, L.D., 1997. Hydrograph separations in an Arctic watershed using

958   mixing model and graphical techniques. Water Resources Research 33 (7), 1707-1719.

959   Millar, C.I., Westfall, R.D., Delany, D.L., 2013. Thermal and hydrologic attributes of rock glaciers and

960   periglacial talus landforms: Sierra Nevada, California, USA. Quaternary International 310, 169-180.

961   Muller, S.W., 1943. Permafrost or permanently frozen ground and related engineering problems. U.S.

962   Engineers Office, Strategic Engineering Study, Special Report No. 62.

963   Nelson, F.E., Anisimov, O.A., Shiklomanov, N.I., 2001. Brief communication: Subsidence risk from

964   thawing permafrost. Nature 410, 889-890.

965   Nelson, F.E., Anisimov, O.A., Shiklomanov, N.I., 2002. Climate change and hazard zonation in the circum-

966   Arctic permafrost regions. Natural Hazards 26, 203-225.

967 Nordstrom, D.K., 2011. Hydrogeochemical processes governing the origin, transport and fate of major and

968   trace elements from mine wastes and mineralized rock to surface waters. Applied Geochemistry 26,

969   1777-1791.

970 Oelke, C., Zhang, T.J., Serreze, M.C., 2004. Modeling evidence for recent warming of the Arctic soil

971   thermal regime. Geophysical Research Letters 31, L07208 (doi: 10.1029/2003GL019300).

972 Parham, L.M., Prokushkin, A.S., Pokrovsky, O.S., Titov, S.V., Grekova, E., Shirokova, L.S., McDowell,

973   W.H., 2013. Permafrost and fire as regulators of stream chemistry in basins of the Central Siberian

974   Plateau. Biogeochemistry 116, 55-68.

975 PERMOS, 2016. Permafrost in Switzerland 2010/2011 to 2013/2014. Noetzli, J., Luethi, R., and Staub, B.

976   (Eds.), Glaciological Report (Permafrost) No. 12-15 of the Cryospheric Commission of the Swiss

977   Academy of Sciences.

978 Petrone, K.C., Hinzman, L.D., Shibata, H., Jones, J.B., Boone, R.D., 2007. The influence of fire and

979   permafrost on sub-arctic stream chemistry during storms. Hydrological Processes 21 (4), 423-434.

980 Petrone, K.C., Jones, J.B., Hinzman, L.D., Boone, R.D., 2006. Seasonal export of carbon, nitrogen, and

981   major solutes from Alaskan catchments with discontinuous permafrost. Journal of Geophysical

982   Resources 111, G02020 (doi:10.1029/2005JG000055).

983 Pokrovsky, O.S., Manasypov, R.M., Loiko, S.V., Krickov, I.A., Kopysov, S.G., Kolesnichenko, L.G.,

984   Vorobyev, S.N., Kirpotin, S.N., 2016. Trace element transport in western Siberian rivers across a

985   permafrost gradient. Biogeosciences 13, 1877-1900.

986 Pokrovsky, O.S., Manasypov, R.M., Loiko, S.V., Shirokova, L.S., Krickov, I.A., Pokrovsky, B.G.,

987   Kolesnichenko, L.G., Kopysov, S.G., Zemtzov, V.A., Kulizhsky, S.P., Vorobyev, S.N., Kirpotin, S.N.,

988   2015. Permafrost coverage, watershed area and season control of dissolved carbon and major

989   elements in western Siberian rivers. Biogeosciences 12 (21), 6301-6320.

990 Pokrovsky, O.S., Reynolds, B.C., Prokushkin, A.S, Schott, J., Viers, J., 2013. Silicon isotope variations in

991   Central Siberian rivers during basalt weathering in permafrost-dominated larch forests. Chemical

992   Geology 355, 103-116.

993   Pokrovsky, O.S., Shirokova, L.S., Kirpotin, S.N., Audry, S., Viers, J., Dupré, B., 2011. Effect of permafrost

994   thawing on organic carbon and trace element colloidal speciation in the thermokarst lakes of western

995   Siberia. Biogeosciences 8, 565-583.

996   Pokrovsky, O.S., Shirokova, L.S., Kirpotin, S.N., Kulizhsky, S.P., Vorobiev, S.N., 2013. Impact of western

997   Siberia heatwave 2012 on greenhouse gases and trace metal concentration in thaw lakes of

998   discontinuous permafrost zone. Biogeosciences 10, 5349-5365.

999   Pokrovsky, O.S., Viers, J., Shirokova, L.S., Shevchenko, V.P., Filipov, A.S., Dupré, B., 2010. Dissolved,

1000   suspended, and colloidal fluxes of organic carbon, major and trace elements in the Severnaya Dvina

1001   River and its tributary. Chemical Geology 273, 136-149.

1002   Polishchuk, Y.M., Bogdanov, A.N., Polishchuk, V.Y., Manasypov, R.M., Shirokova, L.S., Kirpotin, S.N.,

1003   Pokrovsky, O.S., 2017. Size Distribution, Surface Coverage, Water, Carbon, and Metal Storage of

1004   Thermokarst Lakes in the Permafrost Zone of the Western Siberia Lowland. Water 9, 228,

1005   doi:10.3390/w9030228.

1006   Rangecroft, S., Harrison, S., Anderson, K., 2015. Rock glaciers as water stores in the Bolivian Andes: an

1007   assessment of their hydrological importance. Arctic, Antarctic, and Alpine Research 47(1), 89-98.

1008   Rangecroft, S., Harrison, S., Anderson, K., Magrath, J., Castel, A.P., Pacheco, P., 2013. Climate change

1009   and water resources in arid mountains: An Example from the Bolivian Andes. Ambio 42, 852-863.

1010   Raudina, T.V., Loiko, S.V., Lim, A.G., Krickov, I.V., Shirokova, L.S., Istigechev, G.I., Kuzmina, D.M.,

1011   Kulizhsky, S.P., Vorobyev, S.N., Pokrovsky, O.S., 2017. Dissolved organic carbon and major and trace

1012   elements in peat porewater of sporadic, discontinuous, and continuous permafrost zones of western

1013   Siberia. Biogeosciences 14, 3561-3584.

1014   Rember, R., Trefry, J., 2004. Increased concentrations of dissolved trace metals and organic carbon

1015   during snowmelt in rivers of the Alaskan arctic. Geochimica et Cosmochimica Acta 68, 477-489.

1016   Roberts, K.E., Lamoureux, S.F., Kyser, T.K., Muir, D.C.G., Lafrenière, M.J., Iqaluk, D., Pieńkowski, A.J.,

1017   Normandeau, A., 2017. Climate and permafrost effects on the chemistry and ecosystems of High Arctic

1018   Lakes. Scientific Reports 7, 13292 (doi:10.1038/s41598-017-13658-9).

1019   Roer, I., Kääb, A., Dikau, R., 2005. Rock glacier acceleration in the Turtmann valley (Swiss Alps):

1020   Probable controls. Norsk Geografisk Tidsskrift 59, 157-163.

1021   Rogger, M., Chirico, G.B., Hausmann, H., Krainer, K., Brückl, E., Stadler, P., Blöschl, G., 2017. Impact of

1022   mountain permafrost on flow path and runoff response in a high alpine catchment. Water Resources

1023   Research 53(2), 1288-1308.

1024   Rowland, J.C. Jones, C.E., Altmann, G., Bryan, R., Crosby, B.T., Hinzman, L.D., Kane, D.L., Lawrence,

1025   D.M., Mancino, A., Marsh, P., McNamara, J.P., Romanovsky, V.E., Toniolo, H., Travis, B.J., Trochim,

1026   E., Wilson, C.J., Geernaert, G.L., 2010. Arctic landscapes in transition: responses to thawing

1027   permafrost. Eos 91 (26), 229-230.

1028   Rudy, A.C.A., Lamoureux, S.F., Kokelj, S.V., Smith, I.R., England, J.H., 2017. Accelerating thermokarst

1029   transforms ice-cored terrain triggering a downstream cascade to the ocean. Geophysical Research

1030   Letters, doi:10.1002/2017GL074912.

1031   Rühland, K.M., Smol, J.P., Wang, X., Muir, D.C.G., 2003. Limnological characteristics of 56 lakes in the

1032   Central Canadian Arctic Treeline Region. Journal of Limnology 62 (1), 9-27.

1033   Rydberg, J., Klaminder, J., Rosén, P., Bindler, R., 2010. Climate driven release of carbon and mercury

1034   from permafrost mires increases mercury loading to sub-arctic lakes. Science of the Total Environment

1035   408, 4778-4783.

1036   Salerno, F., Rogora, M., Balestrini, R., Lami, A., Tartari, G.A., Thakuri, S., Tartari, G., 2016. Glacier

1037   melting increases the solute concentrations of Himalayan glacial lakes. Environmental Science &

1038   Technology 50(17): 9150-9160.

1039   Sass, O., 2006. Determination of the internal structure of alpine talus deposits using different geophysical

1040   methods (Lechtaler Alps, Austria). Geomorphology 80, 45-58.

1041   Scapozza, C., Lambiel, C., Baron, L, Marescot, L., Reynard, E., 2011. Internal structure and permafrost

1042   distribution in two alpine periglacial talus slopes, Valais, Swiss Alps. Geomorphology 132(34), 208-221.

1043   Scapozza, C., Lambiel, C., Bozzini, C., Mari, S., Conedera, M., 2014. Assessing the rock glacier

1044   kinematics on three different timescales: a case study from the southern Swiss Alps. Earth Surface

1045   Processes & Landforms 39, 2056-2069.

1046   Schmid, M.O., Baral, P., Gruber, S., Shahi, S., Shrestha, T., Stumm, D., Wester, P., 2015. Assessment of

1047   permafrost distribution maps in the Hindu Kush Himalayan region using rock glaciers mapped in

1048   Google Earth. The Cryosphere 9, 2089-2099.

1049   Schuur, E.A.G., McGuire, A.D., Schädel, C., Grosse, G., Harden, J.W., Hayes, D.J., Hugelius, G., Koven,

1050   C.D., Kuhry, P., Lawrence, D.M., Natali, S.M., Olefeldt, D., Romanovsky, V.E., Schaefer, K., Turetsky,

1051   M.R., Treat, C.C., Vonk, J.E., 2015. Climate change and the permafrost carbon feedback. Nature 520,

1052   171-179.

1053   Sharkhuu, N., 1998. Trends of permafrost development in the Selenge River Basin, Mongolia.

1054   Proceedings of the 7th International Conference on Permafrost, 979-985.

1055   Shirokova, L.S., Pokrovsky, O.S., Kirpotin, S.N., Desmukh, C., Pokrovsky, B.G., Audry, S., Viers, J., 2013.

1056   Biogeochemistry of organic carbon, CO2, CH4, and trace elements in thermokarst water bodies in

1057   discontinuous permafrost zones of Western Siberia. Biogeochemistry 113, 573-593.

1058   St-Jacques, J.M., Sauchyn, D.J., 2009. Increasing winter baseflow and mean annual streamflow from

1059   possible permafrost thawing in the Northwest Territories, Canada. Geophysical Research Letters 36,

1060   L01401 (doi:10.1029/2008GL035822).

1061   Stottlemyer, R., 2001. Biogeochemistry of a treeline watershed, northwestern Alaska. Journal of

1062   Environmental Quality 30 (6), 1990-1998.

1063   Streletskiy, D.A., Shiklomanov, N.I., Nelson, F.I., 2012. Permafrost, infrastructure and climate change: A

1064   GIS based landscape approach to geotechnical modeling. Arctic, Antarctic and Alpine Research 44(3),

1065   368-380.

1066   Striegl, R.G., Aiken, G.R., Dornblaser, M.M., Raymond, P.A., Wickland, K.P., 2005. A decrease in

1067   discharge-normalized DOC export by the Yukon River during summer through autumn. Geophysical

1068   Research Letters 32, L21413 (doi:10.1029/2005GL024413).

1069   Szopińska, M., Szumińska, D., Polkowska, Z., Machowiak, K., Lehmann, S., Chmiel, S., 2016. The

1070   chemistry of river-lake systems in the context of permafrost occurrence (Mongolia, Valley of the

1071   Lakes). Part I. Analysis of ion and trace metal concentrations. Sedimentary Geology 340, 74-83.

1072   Tank, S.E., Frey, K.E., Striegl, R.G., Raymond, P.A., Holmes, R.M., McClelland, J.W., Peterson, B.J.,

1073   2012. Landscape-level controls on dissolved carbon flux from diverse catchments of the circumboreal.

1074   Global Biogeochemical Cycles 26, GB0E02 (doi:10.1029/2012GB004299).

1075   Tank, S.E., Striegl, R.G., Frey, K.E., McClelland, J.W., Kokelj, S.V., 2016. Multi-decadal increases in

1076   dissolved organic carbon and alkalinity flux from the Mackenzie drainage basin to the Arctic Ocean.

1077   Environmental Research Letters 11 (5), 1-10.

1078   Thies, H., Nickus, U., Tessadri, R., Psenner, R., 2007. Unexpected response of high Alpine Lake water to

1079   climate warming. Environmental Science & Technology 41, 7424-7429.

1080   Thies, H., Nickus, U., Tolotti, M., Tessadri, R., Krainer, K. 2013. Evidence of rock glacier melt impacts on

1081   water chemistry and diatoms in high mountain streams. Cold Regions Science and Technology 96, 77-

1082   85.

1083   Thompson, M.S., Wrona, F.J., Prowse, T.D., 2012. Shifts in plankton, nutrient and light relationships in

1084   small tundra lakes caused by localized permafrost thaw. Arctic 65, 367-376.

1085   Todd, A.S., Manning, A-H., Verplanck, P.L., Crouch, C., McKnight, D.M., Dunham, R., 2012. Climate-

1086   change-driven deterioration of water quality in a mineralized watershed. Environmental Science &

1087   Technology 46 (17), 9324-9332.

1088   Toohey, R.C., Herman-Mercer, N.M., Schuster, P.F., Mutter, E., Koch, J.C., 2016. Multi-decadal increases

1089   in the Yukon River Basin of chemical fluxes as indicators of changing flowpaths, groundwater, and

1090   permafrost. Geophysical Research Letters, (doi:10.1002/2016GL070817).

1091   Vieira, G., Bockheim, J., Guglielmin, M., Balks, M., Abramov, A.A., Boelhouwers, J., Cannone, N.,

1092   Ganzert, L., Gilichinsky, D., Goryachkin, S., López-Martínez, J., Meiklejohn, I., Raffi, R., Ramos, M.,

1093   Schaefer, C., Serrano, E., Simas, F., Sletten, R., Wagner, D., 2010. Thermal state of permafrost and

1094   active-layer monitoring in the Antarctic: advances during the International Polar Year 2007-2008.

1095   Permafrost and Periglacial Processes 21, 182-197.

1096   Vonk, J.E., Tank, S.E., Bowden, W.B., Laurion, I., Vincent, W.F., Alekseychik, P., Amyot, M., Billet, M.F.,

1097   Canário, J., Cory, R.M., Deshpande, B.N., Helbig, M., Jammet, M., Karlsson, J., Larouche, J.,

1098   MacMillan, G., Rautio, M., Walter Anthony, K.M., Wickland, K.P., 2015. Reviews and syntheses:

1099   Effects of permafrost thaw on Arctic aquatic ecosystems. Biogeosciences 12, 7129-7167.

1100   Voss, B.M., Peucker-Ehrenbrink, B., Eglinton, T.I., Spencer, R.G.M., Bulygina, E., Galy, V., Lamborg,

1101   C.H., Ganguli, P. M., Montluçon, D.B., Marsh, S., Gillies, S.L., Fanslau, J., Epp, A., Luymes, R., 2015.

1102   Seasonal hydrology drives rapid shifts in the flux and composition of dissolved and particulate organic

1103   carbon and major and trace ions in the Fraser River, Canada. Biogeosciences 12, 5597-5618.

1104   Walter Anthony, K.M., Zimov, S.A., Chanton, J.P., Verbyla, D., Chapin, F.S., 2006. Methane bubbling from

1105   Siberian thaw lakes as a positive feedback to climate warming. Nature 443, 71-75.

1106   Walvoord, M.A., Kurylyk, B.L., 2016. Hydrologic impacts of thawing permafrost-A review. Vadose Zone

1107   Journal 15(6) (doi:10.2136/vzj2016.01.0010).

1108   Walvoord, M., Striegl, R., 2007. Increased groundwater to stream discharge from permafrost thawing in

1109   the Yukon river basin: Potential impacts on lateral export of carbon and nitrogen. Geophysical

1110   Research Letters 34, L12402 (doi:10.1029/2007GL030216).

1111   Watanabe, S., Laurion, I., Chokmani, K., Pienitz, R., Vincent, W.F., 2011. Optical diversity of thaw ponds

1112   in discontinuous permafrost: A model system for water color analysis. Journal of Geophysical

1113   Research 116, G02003 (doi:10.1029/2010JG001380).

1114   Williams, M.W., Knauf, M., Caine, M., Liu, F., Verplanck, P.L., 2006. Geochemistry and source water of

1115   rock glacier outflow, Colorado Front Range. Permafrost and Periglacial Processes 17, 13-33.

1116   Williams, M.W., Knauf, M., Cory, R., Caine, N., Liu, N., 2007. Nitrate content and potential microbial

1117   signature of rock glacier outflow, Colorado Front Range. Earth Surface Processes and Landforms 32,

1118   1032-1047.

1119   Williams, P.J., Smith, M.W., 1989. The frozen earth. Fundamentals of geocryology. Cambridge University

1120   Press, Cambridge.

1121   Woo, M., (2012). Permafrost hydrology. Springer-Verlag Berlin Heidelberg.

1122   Wu, J.C., Sheng, Y., Wu, Q.B., Wen, Z., 2010. Processes and modes of permafrost degradation on the

1123   Qinghai-Tibet Plateau. Science in China 53 (1), 150-158.

1124   Wu, Q., Zhang, T., Liu, Y., 2012. Thermal state of the active layer and permafrost along the Qinghai-

1125   Xizang (Tibet) railway from 2006 to 2010. Cryosphere 6 (3), 607-612.

1126   Yang, Y., Wu, Q., Hou, Y., Zhang, Z., Zhan, J., Gao, S., Jin, H., 2017. Unravelling of permafrost

1127   hydrological variabilities on Central Qinghai-Tibet Plateau using stable isotopic technique. Science of

1128   the Total Environment 605-606: 199-210.

1129　Zhang, T., Barry, R.G., Knowles, K., Heginbottom, J.A., Brown, J., 1999. Statistics and characteristics of

1130　　　permafrost and ground-ice distribution in the Northern Hemisphere. Polar Geography 23, 132-154.

1131　Zhang, T., Barry, R.G., Knowles, K., Ling, F., Armstrong, R.L., 2003. Distribution of seasonally and

1132　　　perennially frozen ground in the Northern Hemisphere. Proceedings of the 8th International Conference
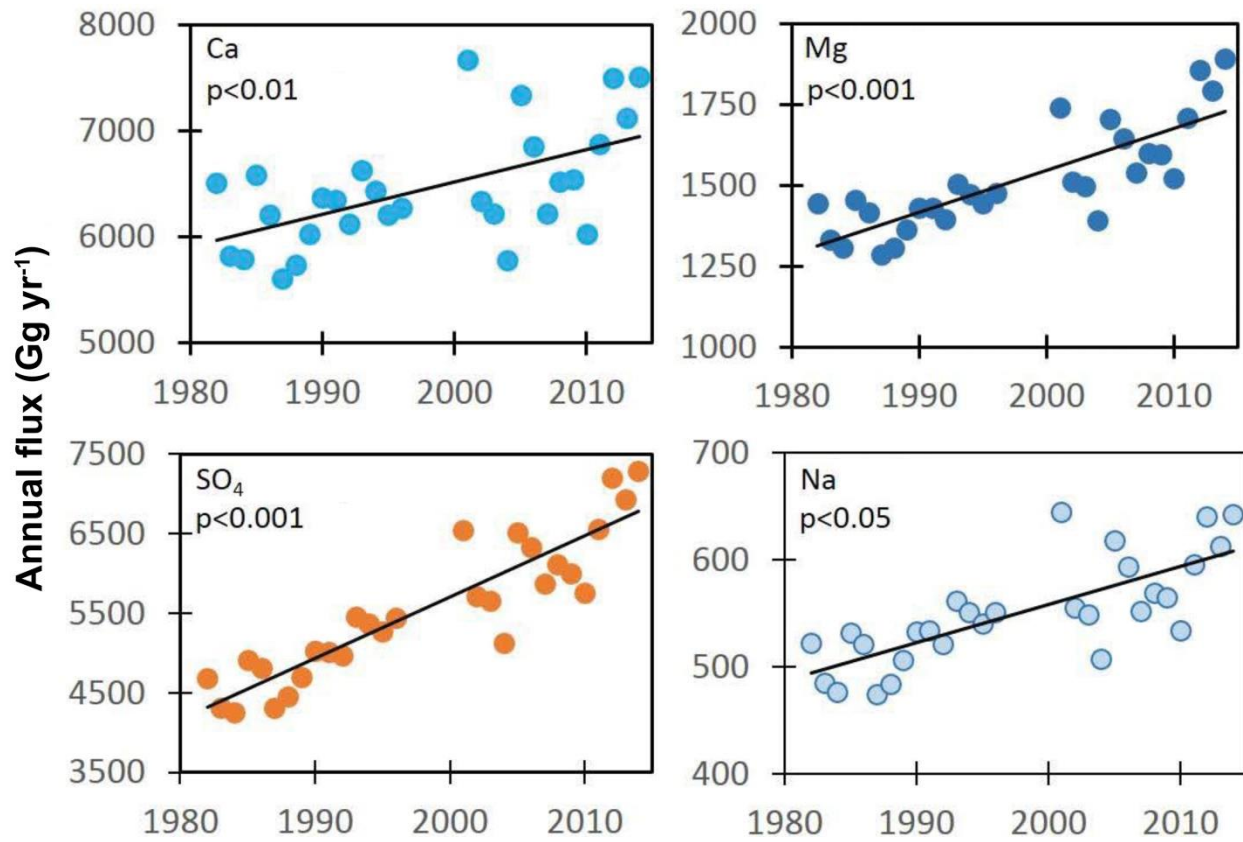
1133　　　on Permafrost, 1289-1294.

1134 **Figures and Tables**
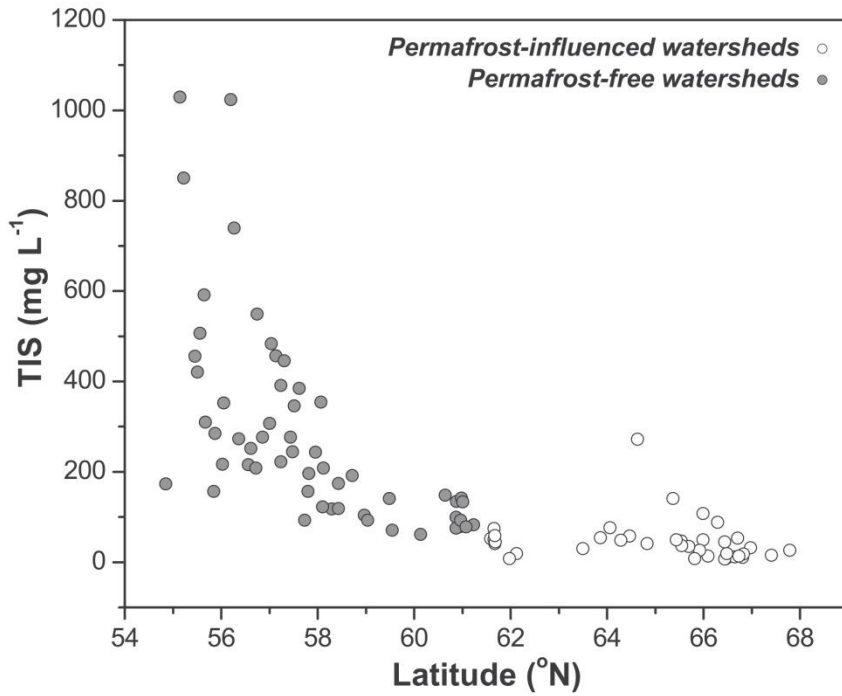
1135

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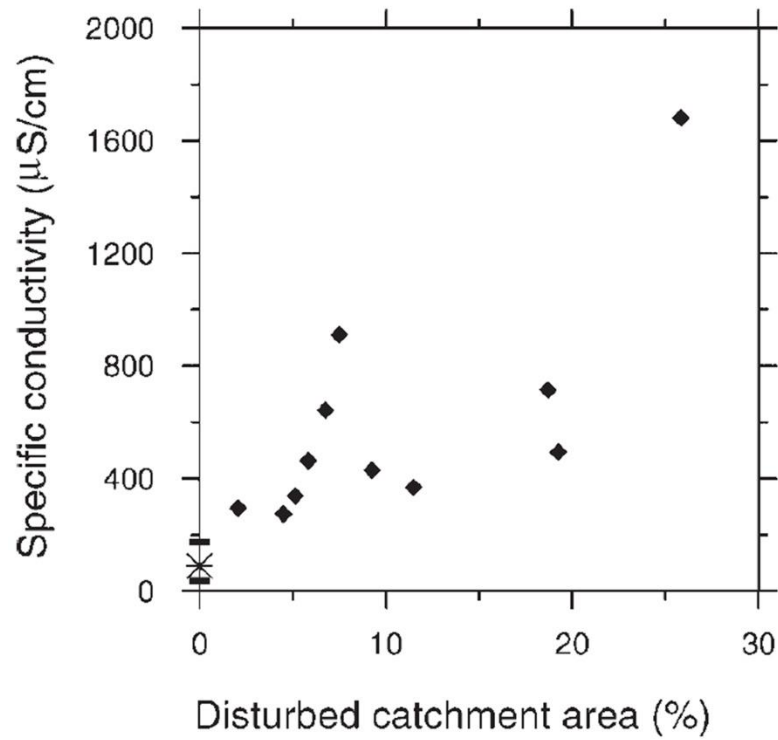
1137 a function of latitude. Permafrost limit is approximately located at 61°N (from Frey et al., 2007a).

1138

1139   Figure 2 - Annual flux increases in the Yukon River with Thiel-Sen trend lines (adapted from Toohey et al.,

1140   2016).

1141

1142   Figure 3 - Percentage of catchment area influenced by thermokarst disturbance (slumping) and specific

1143   conductivity (as index of total solute concentrations, mainly represented by Ca^{2+}, Mg^{2+}, and SO_4^{2-}) of lake

1144   water, upland lakes, Mackenzie Delta region. The (*) indicates the mean specific conductivity of the 11

1145   pristine lakes measured by Kokelj et al. (2005). The (–) indicates maximum and minimum values for

1146   pristine lakes (from Kokelj et al., 2005).
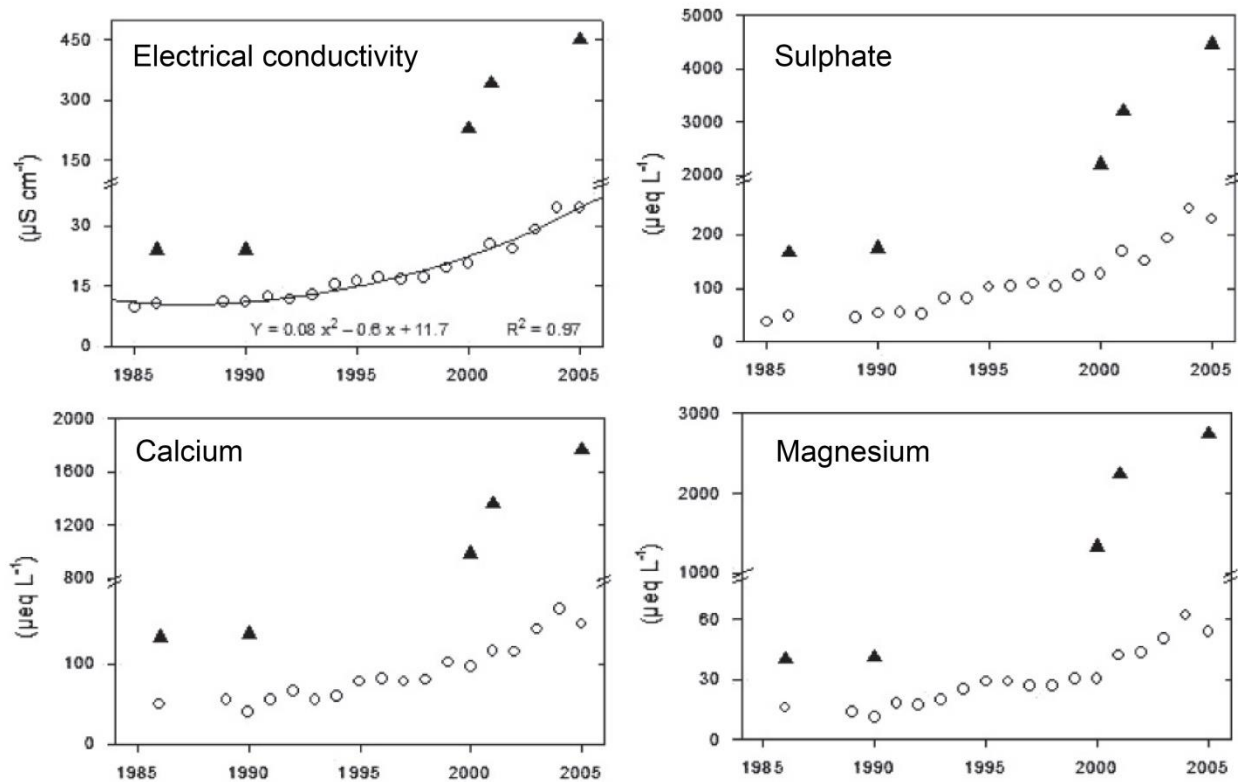
1147

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).
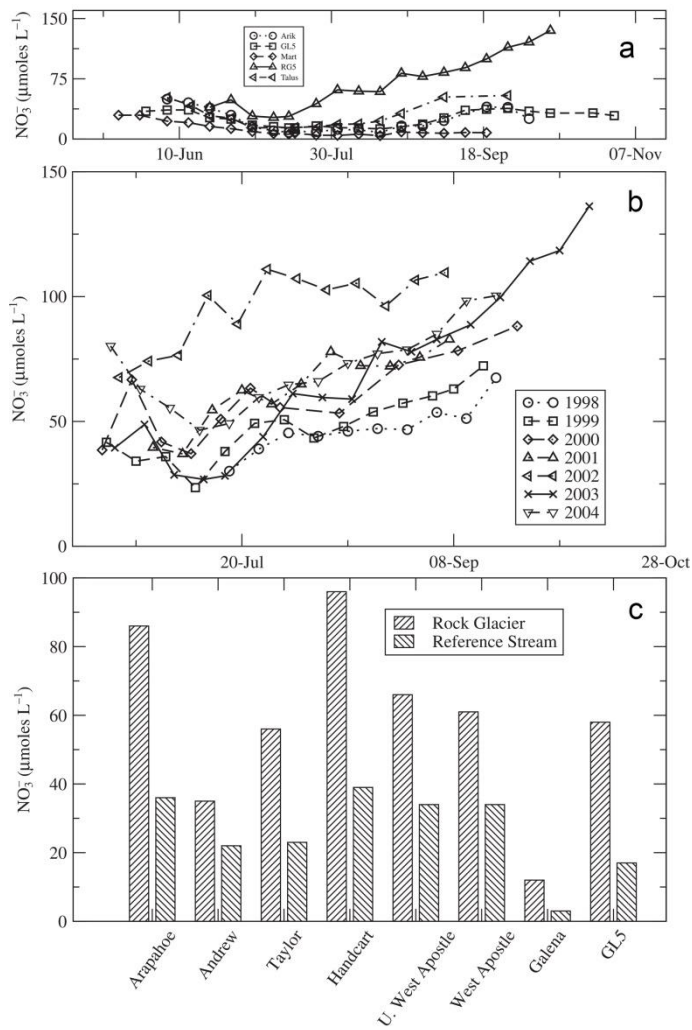
1153

1154   Figure 5 - a) Time series of nitrate in surface water of the Green Lakes Valley, for 2003. Arik is Arikaree

1155   Glacier outflow; GL5 is the outlet of Green Lakes 5; Mart is the outlet of the Martinelli catchment; RG5 is

1156   the Green Lake 5 rock glacier outflow; talus is the blockfield site (for site details refer to Williams et al.,

1157   2007). b) Time series of nitrate concentrations in the outflow of RG5 during the snow-free season from

1158   1998 to 2004. c) Comparison of nitrate concentrations from the outflow of 7 rock glaciers and co-located

1159   surface waters, August 2003 (adapted from Williams et al., 2007).

1160
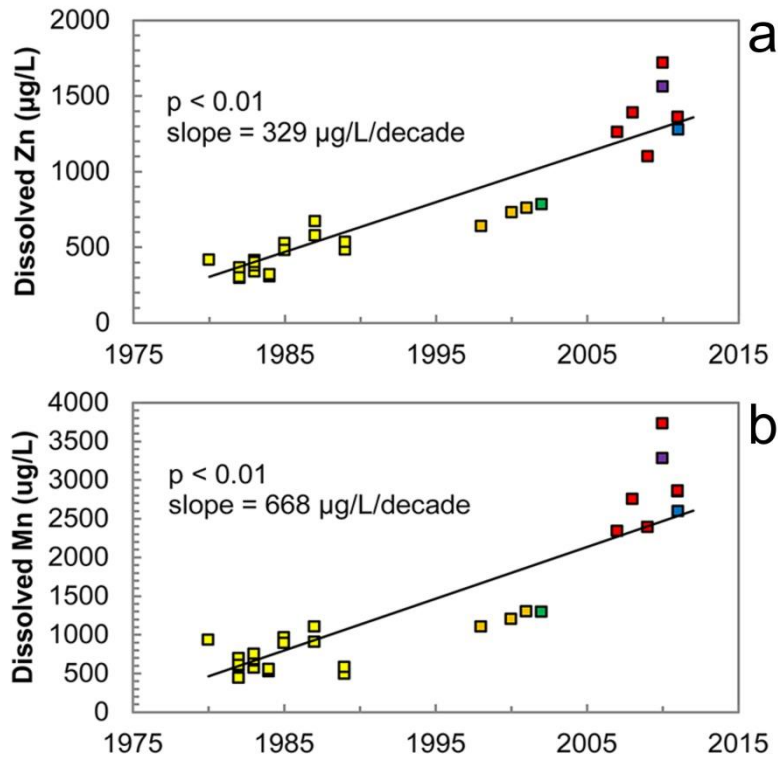
1161

1162   Figure 6 - Trends in September dissolved Zn (a) and Mn (b) concentrations (µg/L) in the Upper Snake

1163   River. Different colours represent different data sources (further details in Todd et al., 2012). p-values

1164   were computed using the Mann-Kendall Test, and lines shown are Kendall-Theil lines (adapted from Todd

1165   et al., 2012).
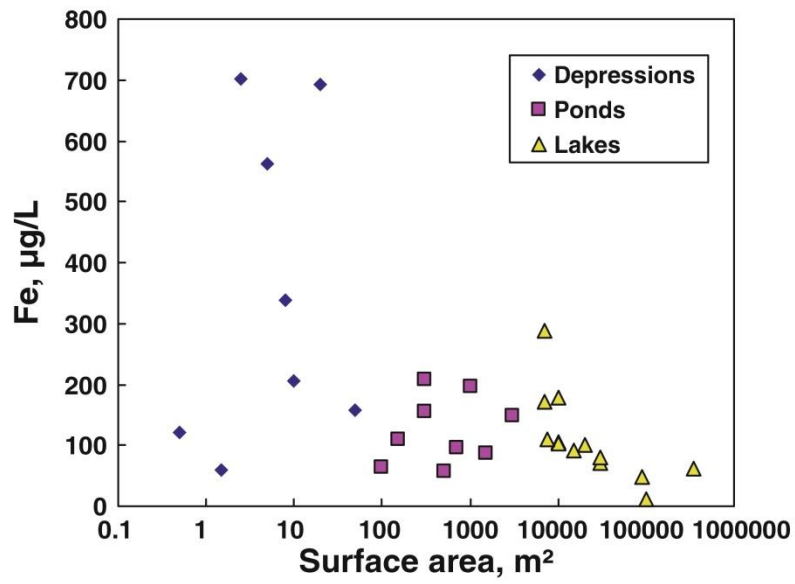
1166

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	Colorado Front Range - USA	RG	M
	2007			
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	Sub-Arctic - Sweden	TK	T
	2010			
MacMillan et al.	2015	Sub-arctic, high-Arctic - Canada	TK	T
Manasypov et al.	2014	Western Siberia - Russia	TK	T
	2015			
Pokrovsky et al.	2011	Western Siberia - Russia	TK	T
	2013			
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	Central Eastern Alps - Italy	RG	T
	2017			
Krainer	2014	Central Eastern Alps - Austria, Italy	RG	T
Krainer et al.	2011	Central Eastern Alps - Italy	RG	T

1169

1170 Table 1 - Selected papers reporting information on the impacts of permafrost degradation on inorganic

1171 chemistry of surface fresh water globally. PD: Pervasive permafrost degradation, TK: Thermokarst, RG:

1172 Rock-glacier thawing, M: Major ions, T: Trace elements.