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4 **Lichen, moss and peat control of C, nutrient and trace metal regime**
5 **in lakes of permafrost peatlands**

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33 **Abstract**

34 Permafrost thaw in continental lowlands produces large number of thermokarst
35 (thaw) lakes, which act as a major regulator of carbon (C) storage in sediments and C
36 emission in the atmosphere. Here we studied thaw lakes of the NE European permafrost
37 peatlands - shallow water bodies located within frozen peat bogs and receiving the
38 majority of their water input from lateral (surface) runoff. We also conducted mesocosm
39 experiments via interacting lake waters with frozen peat and dominant ground vegetation
40 - lichen and moss. There was a systematic decrease in concentrations of dissolved C,
41 CO₂, nutrients and metals with an increase in lake size, corresponding to temporal
42 evolution of the water body and thermokarst development. We hypothesized that ground
43 vegetation and frozen peat provide the majority of C, nutrients and inorganic solutes in
44 the water column of these lakes, and that microbial processing of terrestrial organic
45 matter controls the pattern of CO₂ and nutrient concentrations in thermokarst lakes.
46 Substrate mass-normalized C, nutrient (N, P, K), major and trace metal release was
47 maximal in moss mesocosms. After first 16 h of reaction, the pCO₂ increased ten-fold in
48 mesocosms with moss and lichen; this increase was much less pronounced in experiments
49 with permafrost peat. Overall, moss and lichen were the dominant factors controlling the
50 enrichment of the lake water in organic C, nutrients, and trace metals and rising the CO₂
51 concentration. The global significance of obtained results is that the changes in ground
52 vegetation, rather than mere frozen peat thawing, may exert the primary control on C,
53 major and trace element balance in aquatic ecosystems of tundra peatlands under climate
54 warming scenario.

55

56

57 **1. Introduction**

58 The surface waters of permafrost peatlands are known to exhibit strong CO₂
59 oversaturation with respect to atmosphere and thus, yield sizeable CO₂ emissions

60 (Laurion et al., 2010; Lundin et al., 2013; Roiha et al., 2015; Sepulveda-Jauregui et al.,
61 2015; Vonk et al., 2015; Elder et al., 2018; Karlsson et al., 2021). These waters also
62 contain high Dissolved Organic Carbon (DOC) concentrations, rendering them important
63 reservoirs of C and related elements in Northern Hemisphere. Today, C biogeochemical
64 cycles in C-rich peatland waters from the permafrost zone are characterized in Canadian
65 Plane lakes (Negandhi et al., 2013; Bouchard et al., 2014; Matveev et al., 2016, 2019;
66 Arsenault et al., 2018; Wauthy et al., 2018), small bogs of northern Sweden (Roehm et al.,
67 2009; Hodgkins et al., 2014, 2016), and most extensively, in western Siberian lowland
68 (WSL, Frey and Smith, 2005; Frey et al., 2007; Pokrovsky et al., 2011, 2013; Shirokova
69 et al., 2013, 2020; Manasypov et al., 2014, 2015; Loiko et al., 2017; Serikova et al.,
70 2019).

71 Among various surface waters of permafrost landscapes, recently formed
72 thermokarst lakes are most interesting because they emit sizeable amount of CO₂ and
73 CH₄ to the atmosphere (Walter et al., 2008; Walter-Anthony et al., 2012, 2014; Hogdkins
74 et al., 2014; Ewing et al., 2015; Mann et al., 2015; Natali et al., 2015; Roiha et al., 2015;
75 Schuur et al., 2015; Wik et al., 2016; Serikova et al., 2019). Further, in case of flooding
76 or drainage, these lakes may deliver substantial amount of dissolved nutrients and metals
77 to adjacent hydrological network (Kokelj et al., 2009; Pokrovsky et al., 2011, 2016a, b).
78 The main interest to these shallow surface waters is in their location at the thawing
79 permafrost front, typically in the discontinuous and isolated permafrost zone, where the
80 soil temperature often fluctuates within the highly 'fragile' temperature range (0 to -2°C)
81 of the permafrost (Romanovsky et al., 2010). As a result, under on-going climate
82 warming and permafrost thaw, these waters may *i*) spread their surfaces much faster than
83 those in the region with mineral substrates and *ii*) become highly vulnerable to water
84 heating and heterotrophic respiration of dissolved and particulate organic matter (OM)
85 into CO₂.

86 The area coverage of thaw ponds and small depressions in permafrost-affected
87 peatlands of Northern Eurasia is not fully quantified but may range from 5 to 20% of the
88 territory which includes Western Siberia Lowland (WSL), Northern Siberian Lowland,
89 Kolyma and Indigirka Lowland, of overall area > 2 million km² (Polishchuk et al., 2020;
90 Zabelina et al., 2021). In Europe, the permafrost peatlands widely occur in the NE part of
91 it, within the Bolshezemelskaya Tundra (200,000 km²), although biogeochemical
92 parameters of related thermokarst lakes are quite poorly known.

93 The main source of DOM in thaw ponds of frozen peatlands is leaching of C from
94 allochthonous reservoirs of solid OM, such as peat and surrounding vegetation (mosses,
95 lichens). The magnitude of this process is not yet quantified, and the relative importance
96 of three main organic ‘solid’ substrates - moss, lichen and peat - in providing DOC, CO₂
97 and nutrients to these thaw waters is not known. This, in turn, does not allow predicting
98 the response of these aquatic ecosystems to the change of environmental conditions such
99 as vegetation regime and increasing the active layer thickness (ALT) and the
100 precipitation. The latter will likely bring about a surface drainage through litter layer
101 rather than through underlying mineral soils. To assess the capacity of ground vegetation
102 and peat soil to modify the chemical and gaseous regime of thaw ponds, mere
103 observations of natural environments are not sufficient and ecosystem (mesocosm-level)
104 manipulations are needed.

105 Indeed, the ecosystem manipulation or mesocosm experiments allow
106 understanding the system behavior under various external impacts (i.e., Hering et al.,
107 2015; de Rozari et al., 2016; Conroy et al., 2016). Although the mesocosm experiments
108 were extensively used for boreal, permafrost- and peat-bearing aquatic landscapes
109 (Bridgham et al., 1999; Judd and Kling, 2002; Blodau and Moore, 2003; Blodau et al.,
110 2004; Basiliko et al., 2006; Balcarczyk et al., 2009; Green et al., 2014; Richardson et al.,
111 2015; Nydahl et al., 2019), only one study (Manasypov et al., 2017) dealt with
112 thermokarst lakes of permafrost peatlands. However, in their study, the pCO₂ and

113 dissolved nutrient patterns were not assessed, and two important substrates contributing
114 to the lake water hydrochemical composition – moss and frozen peat – were not tested.

115 In this work, we performed mesocosm experiments on isolated water volumes of
116 thermokarst thaw ponds and we characterized the change of chemical and gaseous (CO₂)
117 parameters of pond water in response to the addition of peat, moss and lichen. We
118 hypothesized that peat will be an important source of chromophoric DOM (CDOM) and
119 nutrients, whereas a non-aromatic, bioavailable DOM and nutrients may be released from
120 moss and lichen. To test this hypothesis, we quantified the relative capacity of various
121 organic substrates to change the chemical composition (DOC, nutrients, metals, dissolved
122 CO₂) of pond water over the time scale (10 to 20 days) comparable to the water residence
123 time in small thaw ponds. Further, we characterized the hydrochemical regime of small
124 thermokarst water bodies of poorly studied Bolshezemelskaya Tundra of NE Europe, and
125 we quantified the C, nutrient, and trace element regime concentration change in natural
126 ponds and lakes depending on their surface area (degree of maturity). We aimed at
127 comparing the pattern of dissolved CO₂, DOC, nutrients and metals in natural sequence
128 of thermokarst lake evolution to that in the mesocosm manipulation experiments in order
129 to obtain quantitative constraints on future changes of thermokarst lake water chemical
130 composition in response to vegetation change and frozen peat thaw. Based on obtained
131 pattern of dissolved carbon, pCO₂ and elements, we provided first-order predictions of
132 thermokarst lake water hydrochemistry change under various climate warming scenario.

133

134 **2. Study site, experiments and analyses**

135 *2.1. Physio-geographical context of thaw ponds and thermokarst lakes of the*
136 *Bolshezemelskaya Tundra*

137 The BolsheZemelskaya Tundra (BZT) peatland is underlayed by permafrost,
138 discontinuous on the eastern part and sporadic to isolated on the western part. The BZT is
139 a hilly moraine lowland located between rivers Pechora and Usa (from the west and

140 south) and the Polar Ural and Pai-Khoi ridge from the east. The dominant altitudes are
141 between 100 and 150 m, created by hills and moraine ridges, composed of sands and silt
142 with boulders. Between the moraines and ridges there are many lakes, mostly of
143 thermokarst origin. The dominant soils are Histosol of peat bogs and podzol-gleys in the
144 south. The mean annual temperature is -3.1°C and the mean annual precipitation is 503
145 mm. The dominant vegetation of the tundra zone is mosses, lichens and dwarf shrubs.
146 The peat (1.0-1.5 m thick) overlays Quaternary glacial deposits over Cretaceous sands,
147 clays and silts. The dominant soil catenae are flat-mound bogs along the watershed
148 divides, which comprise the Hemic Cryic Histosols on mounds and Fibric Gelic Histosols
149 on troughs (the soil nomenclature is according to IUSS, 2014). The average thickness of
150 the active layer ranges from 145 ± 20 cm within the depressions to 41 ± 5 cm at the peat
151 mounds.

152 The thermokarst lakes are shallow (0.5 to 1.5 m) closed basins without distinct
153 inlet and outlets, with watershed surface area smaller or comparable to the lake water
154 area. They receive the surface inflow via lateral drainage of surrounding soil litter and
155 ground vegetation and shallow subsoil water flow over the permafrost table, which is so-
156 called suprapermafrost flow (Manasypov et al., 2015, 2017; Raudina et al., 2018). The
157 early stage of lake formation includes soil subsidences, lichen and moss submergence,
158 and peat abrasion (Audry et al., 2011; Kirpotin et al., 2011; Shirokova et al., 2013). It
159 follows by lateral spreading of shallow water bodies, when the ratio of the lake
160 circumference to the lake volume diminishes which leads to progressive decrease of
161 allochthonous (terrestrial) organic matter input to the lake.

162 The leaching of surrounding organic substrates occurs via (1) rain and melt snow
163 that provide a lateral surface input occurring over moss and lichen cover (Manasypov et
164 al., 2015), (2) direct interaction of the lake water with wave- and thermokarst-abraded
165 peat at the lake shore (Pokrovsky et al., 2011; Shirokova et al., 2013), and finally, (3)
166 suprapermafrost water interaction with frozen peat layer during water travel from

167 elevated points of relief to the lake, over the permafrost table (Raudina et al., 2018).
168 Taking into account these three main processes supplying the C and inorganic solutes to
169 the thaw lakes, in the present study, we modelled the effect of three main organic
170 substrates – moss, lichen and frozen peat – interacting with thermokarst lake waters at
171 controlled mesocosm conditions.

172 To assess natural variability of lake solutes as a function of lake size and degree of
173 maturity, between July 14 and July 26 of 2016 and 2017, we sampled 29 stagnant isolated
174 surface waters located within upland frozen peat bog, including depressions (permafrost
175 subsidence), thaw ponds and thermokarst lakes (**Fig. 1 A**). Depressions in moss and
176 lichen cover are typically filled by water from thawing of ground ice and ranged in size
177 from 0.5 to 2 m² with an average depth between 0.1 and 0.3 m. A series of thaw ponds
178 (10 to 1000 m² size, 0.3 to 0.5 m depth), and thermokarst lakes (10³ to 10⁶ m², 0.5 to 1.5
179 m depth) represented several stages of thermokarst water body maturation, from youngest
180 small size ponds to large mature thermokarst lakes (Zabelina et al., 2021). These water
181 bodies are similar to the sequence of thermokarst lakes and ponds described in Northern
182 Europe (Matthews et al., 1997), Alaska (Hinkel et al., 2003) and western Siberia (Audry
183 et al., 2011; Kirpotin et al., 2011).

184 The surface waters were collected from the shore (depressions) or the PVC boat
185 (thaw ponds and thermokarst lakes). The water samples were filtered on-site using sterile
186 single use Sartorius filters (acetate cellulose, 0.45 µm) for analyses of cations and anions.
187 Unfiltered water samples for nutrient analyses were placed into 250-mL Milli-Q pre-
188 cleaned PVC bottles and kept refrigerated until arrival to the laboratory, within 2-3 h after
189 collection. Conceptual approach of field observations combined with mesocosm
190 experiments in this study is illustrated in **Fig. 1 B**.

191

192 *2.2. Mesocosm experiments*

193 Closed-system mesocosms experiments were performed with thermokarst lake
194 water (pH ~ 5.5) using three organic substrates that are likely to control the
195 hydrochemistry of thermokarst lakes during ecosystem maturation. We selected a typical
196 (6760 m², depth of 1.0±0.5 m) thermokarst lake with moderate DOC (15 mg/L)
197 concentration. The lake was located in the palsa peat bog of Bolshezemelskaya Tundra,
198 40 km east of the Narian-Mar town (63°46'59" N, 75°39'08" E).

199 Peat from frozen layer of permafrost (50-70 cm from the surface which is 10 cm
200 below the maximal active layer depth), moss and lichen constituting ca. 30% and 70% of
201 peatland surface vegetation, were collected in the 100 m vicinity of thermokarst lake and
202 dried at 40°C using Osmofilm® bags during 5 days. The oligotrophic peat was 35%
203 decomposed and contained *Sphagnum angustifolium* (20%), cotton-grass *Eriophorum*
204 (25%), herbs *Scheuchzeria palustris* (15%), bog-sedge (5%), lichens (5%), dwarf shrubs
205 (5%) and some amount of wood debris (pine) and green mosses. The water was collected
206 on 15 July 2017 from the surface (0-20 cm) of the lake and immediately pre-filtered
207 through 100 µm nylon net to remove zooplankton, insects and large organic debris into
208 50-L PVC tanks. Duplicate tanks with lichen *Cladonia stellaris* (5.9 g_{dry}/L), moss
209 *Sphagnum palustris* (9.2 g_{dry}/L) and peat (20 g_{dry}/L) addition were closed with a 100 µm
210 nylon net and placed directly in the lake ensuring their vertical orientation and no
211 overflow of the lake water (**Fig. 1 A**). Additional aeration of incubated water was
212 achieved through periodic opening of the tank and gentle stirring of the content with pre-
213 cleaned plastic shovel. Control experiments included the same tanks filled with 100 µm-
214 filtered lake water without addition of organic or substrate. The wind –induced wave
215 movement in the lake provided sufficient agitation of the interior of the tank. The
216 experiment lasted 16 days. The CO₂ analyses were run each 2-3 h during first day. The
217 sampling for dissolved major and trace elements was performed after 1, 2, 4, 6, 9, 12 and
218 16 day of exposure in order to assess both short-term and long-term effect of added
219 substrate on lake water hydrochemistry.

220

221 2.3. *Chemical analyses*

222 The water temperature, pH, dissolved oxygen, and electric conductivity in natural
223 waters and mesocosm experiments were measured directly in the lake water column or
224 inside the tank using Hanna and WTW portable instruments. Dissolved CO₂
225 concentration was measured using portable submersible Vaissala Carbocap® GM70
226 Hand-held carbon dioxide meter with GMP222 probes (accuracy 1.5%). Filtered waters
227 of natural thaw ponds and lakes and of mesocosm samplings were processed using the
228 analytical approaches employed by the GET Laboratory (Toulouse) to analyze low total
229 dissolved solid (TDS), DOM-rich waters from boreal and permafrost-bearing settings
230 (Pokrovsky *et al.*, 2011, 2016 a, b; Shirokova *et al.*, 2013). The major anion
231 concentrations (Cl⁻ and SO₄²⁻) were analyzed by ion chromatography (Dionex 2000i),
232 with an uncertainty of 2%. The DOC and Dissolved Inorganic Carbon (DIC) were
233 determined by a Shimadzu TOC-VSCN Analyzer with an uncertainty of 3% and a
234 detection limit of 0.1 mg/L. Specific UV absorbance (SUVA) was measured as
235 absorbance at 254 nm normalized for DOC concentration in l mg⁻¹ m⁻¹). The major and
236 trace elements were measured by quadrupole ICP-MS (7500ce, Agilent Technologies).
237 Indium and rhenium were used as internal standards. The international geostandard
238 SLRS-5 (Riverine Water Reference Material for Trace Metals, certified by the National
239 Research Council of Canada) was used to check the validity and reproducibility of
240 analyses. Good agreement existed between our replicated measurements of SLRS-5 and
241 the certified values (relative difference < 15%).

242 The nutrient analyses were based on colorimetric assays (Koroleff, 1983a, b). Total
243 dissolved organic nitrogen (DON) was measured from the difference between the total
244 dissolved nitrogen (persulfate oxidation) and the total dissolved inorganic nitrogen (DIN,
245 or the sum of NH₄⁺, NO₂⁻ and NO₃⁻). Uncertainties of DON and DIN analyses were
246 between 10 and 20 % and detections limits were between 10 and 50 µg/l. Si concentration

247 was measured by spectrophotometry with molybdate blue with an uncertainty of $\pm 5\%$
248 and a detection limit of $2 \mu\text{g/L}$.

249 The total bacterial number and dominant cell size morphology were quantified
250 using DAPI fluorescence method (Porter and Feig, 1980). In addition, we performed
251 oligotrophic and eutrophic bacteria (OB and EB, respectively) count in the course of
252 mesocosm experiments, following the standard methodology used in biodegradation
253 experiments of peat waters (i.e., Stutter et al., 2013) as also described previously
254 (Shirokova et al., 2013, 2019; Chupakova et al., 2018).

255 *2.4. Quantification of element release from organic substrates*

256 In order to calculate the dynamics of element concentration and the amount of
257 element released in the mesocosms, we normalized the difference between element
258 concentration in the experiment and the control (lake water without added substrate) to
259 the concentration of dry substrate in the mesocosm. This allowed tracing the net amount
260 of element (i , mol L^{-1}) released from the substrate (peat, moss or lichen) to the lake water
261 in the course of mesocosm experiment (Δi , $\text{mmol kg}_{\text{dry substrate}}^{-1}$). We estimated the
262 maximal rate (V_{max} , $\text{mmol kg}^{-1} \text{h}^{-1}$) of element release from substrate as a derivative of
263 average maximal change of concentration over the first 24 h. We also quantified the mean
264 rate of element release (V_{mean}) over the full duration of experiments, and the maximal
265 effect of concentration increase determined as the average of 2-3 neighboring
266 concentrations, typically after 100 h of reaction. The initial concentration used to trace
267 these dependences was the one measured 5-10 minutes following the addition of organic
268 substrate. The reason for this choice is that some elements could be present as loosely
269 bound on the surface of organic matter or within the remaining water of humid substrates,
270 notably moss and peat, and quickly released to the aqueous solution, immediately after
271 substrate addition to the reactors. The negative value of V_{max} rate was indicative of
272 element concentration decrease in the course of experiments; in such case, the rates were
273 not quantified.

274

275 3. Results

276 *3.1. Evolution of element concentration in the natural sequence of lake growth and*
277 *maturation reflected in the thermokarst lake size*

278 The evolution of major hydrochemical parameters in the course of lake maturation
279 (increase in size) was characterized via a Mann Kendall test of probability at 90 and 95%
280 level of significance (**Fig. S1** of the Supplement). It is illustrated as a plot of
281 concentration versus thaw pond and lake area for pH, pCO₂, DOC and DIC (**Fig. 2 A-D**).
282 There was a systematic increase in pH (ca, 3 units) with lake size increase from 0.1 to 10⁵
283 m², whereas pCO₂ and DOC concentration decreased about ten times with lake growth
284 and maturation, when the lake size increased over 6 orders of magnitude. The lowest pH
285 values and the highest concentrations of DOC and pCO₂ were observed at the very
286 beginning of thermokarst activity - in small soil depressions, subsidences, water puddles
287 and thaw ponds (< 1-10 m²). The evolution of nutrient (N, P, Si), major cation (Ca) and
288 metal (Fe, Al) concentrations over the lake maturation was less pronounced (**Fig. 3**). The
289 change of pH value and element (*C_i*) concentration with lake area (*S_{lake}*) can be described
290 by a linear function:

$$291 \quad [C_i] = A \times \log S_{\text{lake}} + B \quad (1)$$

292 with parameters A and B of the regression listed in **Table S1**. Considering the trend of
293 element concentration with lake size increase, two main families of solutes were
294 distinguished (**Fig. S1**). The value of Specific Conductivity (S.C.) and concentrations of
295 eutrophic and oligotrophic bacteria, CO₂, DOC, nutrients (NO₂, NO₃, Si, Fe), and some
296 trace elements (Al, Co, Cd, Pb, Sb, Cs, Tl) decreased with lake size; these components
297 formed the first group of elements. The second group of components demonstrated
298 significant (*p* < 0.05) increase in concentration with lake size: O₂, Li, Ca, K, Y, REE, As,
299 Hf, U and the pH value. The other elements did not show any significant (*p* > 0.05, *r* <

300 0.3) correlation with lake size and the probability of their concentration increase or
301 decrease was below 90% (**Fig. S1**).

302

303 *3.2. Mesocosmes of thermokarst lakes from peat palsa plateau*

304 The addition of organic substrates to the lake water caused progressive change of
305 concentration of most dissolved components. The maximal element yield, mean and
306 maximal rates of DOC, nutrient, bacteria, major component (**Table 1**) and trace element
307 (**Table S2**) release were normalized to dry weight of organic substrate and averaged
308 across replicates. These allowed to assess the degree of substrate effect on lake water
309 enrichment (or impoverishment) by each element. For convenience, the element
310 concentration evolution in the mesocosms and in the control enclosures are depicted for
311 several representative major and trace components in **Fig. S2** of the Supplement and
312 specified by groups below.

313

314 3.2.1. Major parameters of lake water (pH, pCO₂, carbon and bacteria).

315 The pH of the lake water strongly decreased by ca. 1.5 units, immediately after
316 addition of all three organic substrates (**Fig. 4 A**). The peat addition produced the lowest
317 pH (ca. 3.75) whilst the moss and lichen addition yielded similar pH drop at the
318 beginning of experiments; the pH then remained stable for the full duration of exposure.
319 Specific conductivity steadily increased during experiments with mosses but did not
320 evolve in other treatments (**Fig. 4 B**). The DIC was an order of magnitude higher in
321 experiments with moss and lichen compared to peat and lake water control incubation
322 (**Table 1**). The oxygen regime was similar (80-100% saturation) between lake water
323 control and peat addition, but the O₂ level strongly decreased (ca., by a factor of 5 to 10)
324 after first 50-100 h of experiments with moss and lichen thus producing net negative ΔO_2
325 values (**Fig. 4 D**).

326 The pCO₂ pattern in experimental mesocosms demonstrated a 10-fold increase
327 over the first 200 hours of lake water reaction with moss and lichen (**Fig. 4 C; Fig. S2 B**).
328 The degree of pCO₂ increase relative to the control followed the order moss > lichen >>
329 peat. However, when normalized to the amount of biomass used in mesocosm incubation,
330 the moss, lichen, and peat released 19, 34 and 4.7 mmol CO₂ kg⁻¹, respectively. The
331 DOC concentration was the highest in experiments with moss addition, rising 8 times,
332 from 20 mg/L to 160 mg/L over 2 weeks of reaction (**Fig. 4 E; S2 C**). Lichen and peat
333 addition produced similar, 2-3 fold increase in DOC over first 284 h of reaction. The
334 mass-normalized DOC yield at the end of incubation was equal to 1500, 580, and 89
335 mmol DOC kg⁻¹ for moss, lichen and peat, respectively (**Table 1**).

336 The UV absorbency and SUVA₂₅₄ were drastically different among three
337 substrates and evolved different to that of DOC (**Fig. 4 F; S2 D**). Peat and moss addition
338 produced a 2-fold increase in absorbency whereas lichens did not affect the release of
339 CDOM. The SUVA decreased after 5 days of reaction with moss and lichen but remained
340 essentially constant in experiments with peat addition, when slightly positive rate
341 increase was observed.

342 The concentration of eutrophic culturable bacteria steadily increased in moss and
343 lichen mesocosms, but decreased after initial rise at 80-100 h in experiments with peat
344 additions (**Fig. S3 A**). The number of oligotrophic bacteria significantly increased with
345 time for all substrates (**Fig. S3 B**) and was ca. 1 to 2 orders of magnitude higher than that
346 in lake water control run. The mesocosms with moss and lichen addition exhibited the
347 highest concentration of cultivable bacteria. Consistent with cultivable (OB and EB)
348 bacterial numbers, the total bacterial (TB) number measured by DAPI was the highest in
349 moss and lichen treatments ((20-40)×10⁶ cell mL⁻¹ without systematic evolution), which
350 was sizably lower in peat treatment ((1-2)×10⁶ cell mL⁻¹). The latter was comparable with
351 TB in the lake water control (5-6)×10⁶ cell mL⁻¹.

352

353 3.2.3. Nutrients (Si, P, N)

354 Dissolved Si exhibited the minimal changes in the course of experiments with
355 stable concentrations achieved over the first day of reaction (**Fig. 5 A, S2 H**). The
356 biomass-normalized Si release was equal to 1.4, 0.97 and 0.26 mmol kg⁻¹ for moss, lichen
357 and peat treatments (**Table 1**). Total dissolved P was dominated (> 90%) by PO₄ whose
358 yield was orders of magnitude higher in moss and lichen mesocosms compared to peat
359 addition (**Fig. 5 B**). Total dissolved N was dominated by NO₃ which was generally stable
360 from the very beginning of experiment and did not systematically evolved over 380 h of
361 exposure (**Fig. 5 C**); mosses and lichens released significant amount of nitrate, unlike
362 peat. Addition of moss yielded strong increase in NH₄⁺ after 4 days of reaction however
363 the agreement between replicates was quite low (**Fig. 5 D**).

364

365 3.2.3. Major and trace elements

366 The final metal yield achieved after ~300 h of reaction for each substrate listed in
367 **Table S2** allowed distinguishing two main group of elements, depending on the moss or
368 peat control on their release in the mesocosms. Examples of major and trace elements are
369 shown in **Fig. S4**. Strong enrichment of the lake water in Li, Na, Mg, K, Fe, Cu, As, Rb,
370 Cs, Ga, Nb and Tl upon the moss addition persisted over full duration of experiments and
371 was significant (p < 0.0001) compared to peat and lichen treatments. Therefore, these
372 elements may serve as indicators of the dominance of moss leaching on thermokarst lake
373 water's chemistry. In contrast, high concentrations of lithogenic Be, Al, V, Ni, Y, heavy
374 REEs, Zr and Hf were observed in mesocosms with peat addition; these elements may be
375 indicative of the influence of peat on thermokarst lake water composition. Addition of
376 lichens enriched aqueous solution in Pb but removed Fe, Al, Ni, Ba, Y, Ga and Zr, as
377 supported by mean and maximal rates of element concentration increase after addition of
378 substrates (see **Table 1** and **Table S2**).

379

380 4. Discussion

381 4.1. Lake size control on C, nutrient and metal concentration in natural settings

382 We observed gradual decrease of S.C., pCO₂, DOC, some nutrients (Si, NO₃), and
383 trace metals (Fe, Al Zn, Co, Cd, Pb, Cs) from small thaw depressions to thaw ponds and
384 lakes. This is consistent with evolution of water hydrochemistry during thaw pond and
385 thermokarst lake growth and maturation in other frozen peatlands of the world, such as
386 WSL (Pokrovsky et al., 2011; Shirokova et al., 2013; Manasypov et al., 2014) and palsa
387 peat bog of Canada (Peura et al., 2020). A decrease in concentrations of C, nutrients and
388 metals with lake size may be caused by several parallel mechanisms operating at the lake
389 shore/lake watershed and within the lake water column as depicted in **Fig. 6**. The
390 leaching of DOC and inorganic solutes occurs from ground vegetation (mainly moss and
391 lichen) and during peat abrasion at the lake shore (Shirokova et al., 2017; Payandi-
392 Rolland et al., 2020a). The delivery of these substrates decreases with an increase in the
393 ratio of lake circumference and lake watershed area to the lake volume (Manasypov et al.,
394 2015). Non-negligible amount of C can be delivered via diffusion from lake sediments
395 into the water column (Audry et al., 2011; Shirokova et al., 2020). An increase in the lake
396 volume and associated decrease of solute flux lead to mere dilution of dissolved C,
397 nutrient and metal concentration upon growth of thermokarst lake depth and area. This is
398 reflected by a linear response of DOC concentration to the logarithm of lake water areas
399 (**Fig. 2 C**). In addition to surface vegetation and upper (unfrozen) peat horizons, thawing
400 of ground ice during summer period (Connolly et al., 2020; Lim et al., 2021) may provide
401 sizeable input of DOC and related elements. It can be expected that the role of moss and
402 lichen cover is especially pronounced for small water bodies whereas the suprapermafrost
403 flow-delivered peat leaching products are more important for large lakes. The mineral
404 horizons can be leached by subsoil (shallow underground) waters, especially in large
405 lakes which exhibit deeper drainage basins and higher hydrological connectivity with
406 subsurface reservoirs (Ala-aho et al., 2018).

407 The mechanisms that are capable removing the solutes via DOM and nutrient
408 processing within the lake water column include heterotrophic bacterial activity (Comte
409 et al., 2015; Deshpande et al., 2016; Roiha et al., 2015, 2016), photodegradation
410 (although of limited importance, Laurion et al., 2021), and photosynthesis (Tank et al.,
411 2009; Rautio et al., 2011; Przytulska et al., 2016; Bogard et al., 2019; Wauthy and
412 Rautio, 2020). Heterotrophic bacterial and photodegradation of DOM lead to coagulation
413 of Fe, Mn and trace metals and their transfer to lake sediments in the form of particulate
414 OC and metal hydroxides as it is known from laboratory (Oleinikova et al., 2017a, b) and
415 field experiments (Shirokova et al., 2019; Payandi-Rolland et al., 2020b). The second
416 mechanism is nutrient uptake by macrophytes, periphyton, and plankton, whose
417 photosynthesis brings about a pH rise and a pCO₂ decrease in large thermokarst lakes
418 compared to small thaw ponds and depressions as it is known from field observations in
419 thermokarst lakes of the Mackenzie Delta (Squires et al., 2009; Tank et al., 2009). Note
420 here that the pH increase in the course of lake maturation (**Fig. 2 A**) reflects, from the one
421 hand, the phytoplankton and periphyton photosynthetic activity (Shirokova et al., 2015).
422 From the other hand, lake size increase leads to increasing connectivity with shallow
423 ground water and subsurface waters (Manasypov et al., 2015; 2020). These waters have
424 higher pH due to their reaction with soil mineral horizons and thus can decrease the
425 overall acidity of large thermokarst lakes.

426

427 *4.2. Behavior of C, nutrient and trace metal in thermokarst lakes after moss, lichen and*
428 *peat addition in mesocosm experiments.*

429 The pattern of aqueous CO₂, DOC, nutrient and metal concentration in the presence
430 of moss, lichen and peat substrates in the mesocosm experiments can serve as an
431 experimental model of thermokarst water body evolution in the natural sequence of peat
432 thawing and lake lateral spreading. The highest pCO₂ values observed at the very
433 beginning of moss and lichen reaction with lake water (**Fig. 4 C, S2 B, Table 1**) are

434 consistent with elevated CO₂ measured in permafrost thaw subsidence with submerged
435 mosses in this study (i.e., several thousand ppm, x 10 oversaturated with atmosphere, **Fig.**
436 **2 B**). High pCO₂ in small waterbodies and depressions was also reported in western
437 Siberian (Polishchuk et al., 2018) and NE European (Zabelina et al., 2021) permafrost
438 peatlands. Sizable CO₂ production by submerged moss and lichen may occur due to both
439 respiration of live bryophytes, fungi and algae and heterotrophic (bacterial) degradation
440 of DOC that was leached from moss and lichen biomass. The response of CO₂ increase to
441 moss and lichen addition (1-2 days, **Fig. 4 C**), was much faster than the DOC increase (≥
442 5 days, **Fig. 4 E**). This is also reflected in higher values of V_{mean} for CO₂ compared to
443 DOC in the presence of both moss and lichen (**Table 1**). Such a difference suggests that
444 the respiration of live lichen and moss rather than degradation of biomass is the main
445 driving factor of rapid CO₂ rise. The O₂ concentration strongly decreased after first day of
446 reaction with moss and lichen (**Fig. 4 D; S2 A**). This indicates that the heterotrophic
447 degradation consuming O₂ from lake water was the primary cause of partially anoxic
448 conditions in these experiments. It is worth noting that for moss and lichen, the rates of
449 initial (0-24 h) O₂ decrease (7±1; 2.7±0.8 mmol kg⁻¹) and CO₂ increase (6±2; 4±1 mmol
450 kg⁻¹) were comparable (**Fig. 4C**, insert). Furthermore, the heterotrophic degradation
451 control on O₂ level in mesocosms is consistent with higher bacterial number measured in
452 moss and lichen experiment compared to peat experiment (section 3.2.1). Therefore,
453 results of the present study suggest that the plant-derived “fresh” DOM can be a driven
454 factor of CO₂ rise, unlike soil refractory DOM. This is consistent with recent mesocosm
455 experiments in a meso-eutrophic lake of the boreal zone (Nydahl et al., 2019). In these
456 experiments, with an addition of colored allochthonous (soil) DOC, there was no
457 relationship between bacterial activities and pCO₂.

458 The sphagnum moss was the most efficient substrate in terms of DOC and CO₂
459 production. The reason of higher reactivity of moss compared to lichen is not clear; the
460 higher degradability of moss may be caused by its essentially wet status in natural

461 conditions thus allowing to initiate fast and active heterotrophic bacterial activity,
462 necessary for an enhanced CO₂ and DOC production. In contrast, in dry lichens and
463 frozen peat, the bacteria were rather dormant and thus not capable to efficiently degrade
464 particulate organic matter at the time scale of mesocosm experiments. Interestingly, that
465 at the end of experiment, corresponding to stable (steady) concentration of aqueous
466 components, the mass-normalized effects of moss exceeded that of lichen and peat by a
467 factor of 2 to 5 (see **Table 1** and **Table S2**). In particular, the moss mesocosm yielded the
468 highest level of nutrients, notably PO₄³⁻, NO₃⁻, NH₄⁺, K, Fe, Mn, Zn. These nutrients may
469 be potentially limiting for aquatic plankton and periphyton of surface waters from frozen
470 peatlands. Thus, high ammonium concentrations in surface waters derived from
471 decomposition of vegetation are well known for wetland ecosystems (Camargo and
472 Alvaro, 2006). We therefore suggest that, at the beginning of thermokarst development,
473 corresponding to early stages of soil subsidences, the degradation and leaching of mosses
474 rather than that of lichens and peat determine the evolution of C, nutrient and metal
475 concentration in thaw waters of peatlands. In particular, high DOC and pCO₂ values at
476 the beginning of thermokarst development, observed in the present study of NE European
477 peatlands (**Fig. 2 B, C; S2 B, C**) and supported by other observations (Zabelina et al.,
478 2021 and references therein) are most likely linked to moss, and in a lesser degree, to
479 lichen degradation. Note that the presence of dense *Sphagnum* moss was reported to be
480 the main driver of variation in DOM composition in thermokarst water bodies of the
481 North European permafrost peatlands (Hodgkins et al., 2016).

482 The finding that frozen peat releases quite low amount of nutrients compared to
483 dominant terrestrial vegetation may have a global significance. It means that in case of
484 permafrost thaw and tundra landscape modification, the primary factor of DOC
485 concentration and CO₂ emission flux change will be the ground vegetation regime
486 (colonization, flooding, drying), rather than thaw of frozen peat layer and reacting of
487 thawed peat with surface waters. Moreover, given that peat addition is not capable

488 increasing pCO₂ much higher above that of the control, it is possible that progressive
489 thawing of frozen peat and its reaction with surface fluids might not produce drastic
490 change in CO₂ emission pattern from inland waters of frozen peatlands. In contrast, the
491 regime of ground vegetation flooding and decay in these wetlands might significantly
492 increase CO₂ concentration and relevant emission fluxes at the time scale of several days
493 of reaction. The reason for this is that the response of aquatic ecosystem chemical
494 composition and CO₂ emission to the submerging of mosses is on the order of first hours
495 to first days, which is comparable with the water residence time in local depressions and
496 ephemeral puddles and ponds. This conclusion is at odds with current paradigm that
497 thawing of permafrost will greatly enhance C fluxes in aquatic system, microbial activity,
498 and CO₂ emission rate (Voigt et al., 2019, Hayes et al., 2014). In case of permafrost
499 peatlands of continental lowlands, the reactivity of frozen peat in aquatic systems will
500 have a subordinate impact on C, nutrient and metal regime in surface waters, compared to
501 lateral expansion or retreat of moss cover and flooding of moss and lichens by surface
502 waters. At the same time, the relative importance of ground vegetation in element
503 delivery to the thermokarst lakes is likely to vary depending on season. Thus, in a recent
504 study of selenium (Se) biogeochemistry in thermokarst lakes of western Siberia
505 (Pokrovsky et al., 2018), we demonstrated that, in addition to direct atmospheric fallout,
506 Se input to the lakes occurs via surface flux over moss and lichen in June (spring) and via
507 supra-permafrost flow in summer and autumn. These mechanisms are fully consistent
508 with those revealed in the present study using mesocosm approach. In particularly in
509 spring, Se, together with our trace metals of atmospheric origin, can be mobilized directly
510 from melted snow and indirectly from snow water interacting with plant litter and
511 moss/lichen biomass because the underlying peat is frozen.

512 The two groups of trace element revealed during addition of various substances to
513 the lake water (section 3.2.3) can be indicative either of moss (alkalis and alkaline-earth
514 metals, Fe, As, Cu, Ga, Nb, Tl) or peat (Be, V; Ni, Al and trivalent and tetravalent

515 hydrolysates) control on thermokarst lake water chemistry. It is not possible to ascertain
516 specific processes (leaching, adsorption, heterotrophic degradation) controlling these
517 element behavior in the mesocosm experiments without assessing the role of organic and
518 organo-mineral colloids that are likely to control element speciation in surface waters of
519 permafrost peatlands. These mechanisms may include the destabilization of Fe-organic
520 complexes, liberation of ionic Fe (III), its coagulation in the form of Fe oxyhydroxides,
521 and coprecipitation of TE with Fe hydroxides, as it happens in WSL rivers and lakes
522 (Pokrovsky et al., 2016b) and observed in experimental studies of organo-ferric colloids
523 degradation by microbial activity (Oleinikova et al., 2017b). A strong link of trivalent
524 and tetravalent hydrolysates to Fe oxy(hydr)oxides in the form of organo-ferric colloids
525 was already evidenced in the water column and porewater sediments of thermokarst lakes
526 from permafrost peatlands (Pokrovsky et al., 2011; Audry et al., 2011; Shirokova et al.,
527 2013). The elements exhibiting a concomitant release with DOC are likely to be present
528 as strongly-bound organic species. Typically, these are divalent transition metals (Me^{2+}).
529 Their organo-colloidal status is maintained in a wide range of pH and DOC
530 concentrations in thaw ponds and thermokarst lakes of western Siberia (Pokrovsky et al.,
531 2011, 2013, 2016a; Shirokova et al., 2013).

532

533 *4.3. The role of permafrost thaw and vegetation shift in hydrochemistry of thermokarst* 534 *lakes subjected to climate warming*

535 Thermokarst development in permafrost peatlands involves lateral spreading of
536 the lake without significant deepening of its basin (Polishchuk et al., 2017; Manasypov et
537 al., 2020). This leads to a decrease of the ratio water volume : border line (perimeter).
538 This was first reported for Arctic tundra by Britton (1957) and later evidenced in western
539 Siberia (Kirpotin et al., 2009, 2011; Audry et al., 2011; Manasypov et al., 2020). Such a
540 decrease yields a concomitant decrease in DOC, conductivity (major cations), Fe, Al, and
541 trace element of the lake water with an increase in the lake size (Pokrovsky et al., 2011,

542 2013; Shirokova et al., 2013; Manasypov et al., 2014, 2015). Based on results of natural
543 observation in NE Europe and mesocosm experiments, we interpret this trend as due to
544 the dominance of moss and lichen degradation at the beginning of thermokarst lake
545 formation, when small depressions in the moss/lichen cover are filled by water either
546 from thawing of soil ice or of atmospheric origin (stage 1 in **Fig. 6**). The accumulation of
547 organic and inorganic solutes in the newly-formed depressions triggers DOM, nutrient
548 and metal processing in the water column. This occurs via heterotrophic plankton
549 mineralization and sedimentation to the bottom under the dominance of allochthonous
550 input from vegetation leaching (stage 2 in **Fig. 6**). At later stages of lake development, the
551 peat becomes the main supplier of C, nutrient and metal to the water column via
552 suprapermafrost inflow as well as wave abrasion of peat at the lake shore. However,
553 relatively low reactivity of peat (i.e., Payandi-Rolland et al., 2020a and this study) does
554 not allow sufficient accumulation of organic and inorganic components in the lake water
555 column. Instead, mere dilution by atmospheric precipitation and enhanced processing of
556 autochthonous DOM and nutrients (such as photo- and biodegradation and uptake by
557 plankton and periphyton) lead to overall decrease of C, nutrient and metal concentration
558 in large lakes compared to small thaw ponds (stage 3, **Fig. 6**).

559 In this study, we hypothesized that peat is an important source of chromophoric
560 DOM (CDOM) and nutrients, whereas a non-aromatic DOM easily available by bacteria
561 may be released from mosses and lichens. As a result, the pattern of CO₂ concentration is
562 determined by a combination of nutrients and DOC delivery from solid source to aqueous
563 phase. This pattern was found to be drastically different between bryophytes (mosses),
564 symbiotic organisms (lichens) and frozen peat. This finding therefore confirms our
565 hypothesis. We further anticipated that progressive change of the ratio of lake water to
566 surrounding organic substrates in the course of lake evolution will produce a systematic
567 change in lake water chemical composition: from moss and lichen degradation control at
568 the beginning of thaw pond formation, at the stage of permafrost subsidence/depression,

569 to coastal peat abrasion and dissolution control at the later stages of lake growth and
570 maturation. At the current state of knowledge, this hypothesis cannot be unambiguously
571 verified, as it requires an estimation of vegetation cover at the small thaw ponds and
572 thermokarst lakes watershed, which is possible only with high-resolution GIS
573 approaches.

574 According to results of this study, short-term (1-10 years) consequences of
575 climate warming in permafrost peatlands will be mostly pronounced through the change
576 of ground vegetation regime. On a longer perspective (50-100 years), the increase in the
577 active layer depth (currently 1 cm y⁻¹, Johansson et al., 2011) will involve much larger
578 amount of peat in water transport within the lake watersheds. This will inevitably increase
579 the relative role of peat in element delivery from the land to the lentic waters of
580 permafrost peatlands. Furthermore, a sizable amount of highly labile C and related
581 elements can be mobilized from currently frozen peat ice into the hydrological network
582 via the suprapermafrost flow (Lim et al., 2021).

583 Overall, an increase in the active layer thickness, lake water temperature and the
584 duration of unfrozen period due to climate warming should enhance carbon and element
585 leaching from moss, lichen and peat to the thermokarst lakes. However, compared to
586 wave-induced peat abrasion at the lake border, the moss and lichen colonization,
587 submerging and degradation during permafrost thaw and subsidence formation are more
588 important drivers of thermokarst lake water's chemical composition evolution under the
589 on-going environmental changes. Note that this conclusion supports recent findings on
590 very young age of C emitted from permafrost waters which reveals that inland water
591 carbon emissions from permafrost landscapes may be more sensitive to changes in
592 contemporary carbon turnover than the release of pre-aged carbon from thawing
593 permafrost (Dean et al., 2020).

594

595 **5. Concluding remarks**

596 Thawing of frozen peatlands produces depressions, thaw ponds and thermokarst
597 lakes encountered in the NE European discontinuous permafrost zone (Bolshezemelskaya
598 Tundra). There was a systematic decrease in concentrations of dissolved OC, CO₂,
599 nutrient and metal with an increase in the water body size. In the course of lake
600 maturation (over six order of magnitude increase in the surface area), the pH increased by
601 ca. 3 units while the pCO₂ and DOC concentrations decreased ten-fold. Total P, Ca, Mg
602 increased their concentrations by a factor of 2 to 3, whereas Si, Fe, Al, NO₃ decreased
603 concentrations with lake size increase. We interpret these changes as due to a
604 combination of external factors (delivery of solutes from lake watershed) and DOM,
605 nutrient and metal processing in the water column.

606 Mesocosm experiments on thermokarst lake water in the presence of frozen peat,
607 moss and lichen demonstrated that moss and lichen leaching and degradation, rather than
608 peat abrasion, act as a main driver of thermokarst lake water's evolution. The mass-
609 normalized release of DOC, macro nutrients (Si, N, P), major elements (K, Na, Ca, Mg),
610 micronutrients and trace metals (Fe, Mn, Cu, As, Rb, Cs, Tl) was controlled essentially
611 by moss and, in a lesser degree, lichen with almost negligible impact of peat. When
612 comparing to natural sequence of lakes and ponds chemical composition, these mesocosm
613 experiments allowed to characterize the water chemistry during the first step of thaw
614 pond formation, when the moss leaching determines the hydrochemical features of
615 surface water. Upon progressive increase in the lake size, the lichen and moss leaching at
616 the watershed surface become less important than the delivery of peat leaching products
617 by the suprapermafrost flow. This happens due to an increase in the ratio of lake
618 circumference to the lake volume which decreased the delivery of external terrestrial
619 (allochthonous) material. In contrast, when lake water volume increases, the relative role
620 of autochthonous processes of DOM, nutrient and metal transformation, such as photo-
621 and bio-degradation, and phytoplankton/periphyton activity also increase. This leads to

622 gradual removal of DOC, nutrients and metals due to C emission to atmosphere and
623 element deposition to the lake bottom.

624 Because moss leaching is likely to be most important process controlling
625 hydrochemistry of dissolved elements and CO₂ emission/burial pattern in thermokarst
626 water bodies, the response of the moss coverage regime to permafrost thaw and climate
627 warming in NE European frozen peatlands will chiefly define the DOC and nutrient
628 cycling in lentic waters and CO₂ emission fluxes. Given that the vegetation biomass
629 leaching reactions are very fast, on the order of first hours to days, the hydrological
630 pattern of moss and lichen flooding rather than biogeochemical reactions within the peat
631 soil profile will be controlling short-term response of peat palsa landscapes to permafrost
632 thaw and temperature rise. This contrasts a dominant view that thawing of frozen peat
633 provides the main contribution to the negative feedback of environmental response to the
634 climate change. At least on a short-term scale, the peat thaw may be a subordinate factor
635 of biogeochemical C fluxes compared to the change of wetting and flooding regime of the
636 lake shoreline and moss persistence/growth on the tundra surface.

637

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645 **References**

646 Ala-Aho, P., Soulsby, C., Pokrovsky, O.S., Kirpotin, S.N., Karlsson, J., et al., 2018.
647 Using stable isotopes to assess surface water source dynamics and hydrological
648 connectivity in a high-latitude wetland and permafrost influenced landscape. *J.*
649 *Hydrology* 556, 279–293.

650 Arsenault, J., Talbot, J., Moore, T.R., 2018. Environmental controls of C, N and P
651 biogeochemistry in peatland ponds. *Sci. Total Environ.* 631-632, 714-722.

652 Audry, S., Pokrovsky, O.S., Shirokova, L.S., Kirpotin, S.N., Dupré, B., 2011. Organic
653 matter mineralization and trace element post-depositional redistribution in
654 Western Siberia thermokarst lake sediments. *Biogeosciences* 8, 3341–3358.
655 <http://dx.doi.org/10.5194/bg-8-3341-2011>.

656 Balcarczyk, K.L., Jones, J.B., Jaffé, R., Maie, N., 2009. Stream dissolved organic matter
657 bioavailability and composition in watersheds underlain with discontinuous

658 permafrost. *Biogeochemistry* 94, 255–270. [http://dx.doi.org/10.1007/s10533-](http://dx.doi.org/10.1007/s10533-009-9324-x)
659 009-9324-x.

660 Basiliko, N., Moore, T.R., Jeannotte, R., Bubier, J.L., 2006. Nutrient input and carbon
661 and microbial dynamics in an ombrotrophic bog. *Geomicrobiol. J.* 23, 531–543.
662 DOI: 10.1080/01490450600897278.

663 Blodau, C., Moore, T., 2003. Experimental response of peatland carbon dynamics to a
664 water table fluctuation. *Aquat. Sci.* 65, 47–62.
665 <http://dx.doi.org/10.1007/s000270300004>.

666 Blodau, C., Basiliko, N., Moore, T.R., 2004. Carbon turnover in peatland mesocosms
667 exposed to different water table levels. *Biogeochemistry* 67, 331–351.
668 <http://dx.doi.org/10.1023/B:BIOG.0000015788.30164.e2>.

669 Bogard, M.J., Kuhn, C.D., Johnston, S.E., Striegl, R.C., Holtgrieve, G.W. et al., 2019.
670 Negligible cycling of terrestrial carbon in many lakes of the arid circumpolar
671 landscape. *Nature Geoscience* 12, 180-185.

672 Bouchard, F., Francus, P., Pienitz, R., Laurion, I., Feyte, S., 2014. Subarctic thermokarst
673 ponds: Investigating recent landscape evolution and sediment dynamics in
674 thawed permafrost of Northern Québec (Canada). *Arct. Antarct. Alp. Res.* 46
675 (1) 251-271., DOI : 10.1657/1938-4246-46.1.251

676 Bridgham, D.S., Pastor, J., Updegraff, K., Malterer, J.T., Johnson, K., Harth, C., Chen, J.,
677 1999. Ecosystem control over temperature and energy flux in northern peatlands.
678 *Ecol. Appl.* 9, 1345–1358.

679 Britton, M.E., 1957. Vegetation of the Arctic Tundra, in: Hanson, H.P. (Ed.), *Arctic*
680 *biology* (18th Annual Biology Colloquium). Corvallis, Ore., Oregon State
681 College, pp. 26–61.

682 Camargo, J., Alvaro A., 2006. Ecological and toxicological effects of inorganic nitrogen
683 pollution in aquatic systems: A global assessment. *Environment Internat.* 32, 831-
684 849.

685 Chupakova, A. A., Chupakov, A. V., Neverova, N. V., Shirokova, L. S., and Pokrovsky,
686 O. S., 2018. Photodegradation of river dissolved organic matter and trace metals in
687 the largest European Arctic estuary. *Sci. Total Environ.* 622–623, 1343–1352.

688 Comte J., Monier A., Crevecoeur S., Lovejoy C., Vincent W.F., 2015. Microbial
689 biogeography of permafrost thaw ponds across the changing northern landscape.
690 *Ecography* 28, 1-10. doi: 10.1111/ecog.01667.

691 Connolly, C.T., Cardenas, M.B., Burkart, G.A., Spencer, R.G.M., McClelland, J.W.,
692 2020. Groundwater as a major source of dissolved organic matter to Arctic
693 coastal waters. *Nat. Commun.* 11, 1–8. [https://doi.org/10.1038/s41467-020-](https://doi.org/10.1038/s41467-020-15250-8)
694 15250-8

695 Conroy, E., Turner, J.N., Rymaszewicz, A., Bruen, M., O'Sullivan, J.J., Lawler, D.M.,
696 Lally, H., Kelly-Quinn, M., 2016. Evaluating the relationship between biotic and
697 sediment metrics using mesocosms and field studies. *Sci. Total. Environ.* 568,
698 1092–1101. <http://dx.doi.org/10.1016/j.scitotenv.2016.06.168>.

699 de Rozari, P., Greenway, M., El Hanandeh, A., 2016. Phosphorus removal from
700 secondary sewage and septage using sand media amended with biochar in
701 constructed wetland mesocosms. *Sci. Total Environ.* 569–570, 123–133.
702 <http://dx.doi.org/10.1016/j.scitotenv.2016.06.096>.

703 Dean, J. F., Meisel, O. H., Martyn Rosco, M., Marchesini, L.B., Garnett, M.H.,
704 Lenderink, H., ... Dolman, A.J., 2020. East Siberian Arctic inland waters emit
705 mostly contemporary carbon. *Nature Comm.* 11(1), doi:10.1038/s41467-020-
706 15511-6.

707 Deshpande, B. N., Crevecoeur, S., Matveev, A., and Vincent, W. F., 2016. Bacterial
708 production in subarctic peatland lakes enriched by thawing permafrost,
709 *Biogeosciences* 13, 4411-4427.

710 Elder, C.D., Xu, X., Walker, J., Schnell, J.L. et al., 2018. Greenhouse gas emissions from
711 diverse Arctic Alaskan lakes are dominated by young carbon. *Nat. Clim. Chang.* 8,

- 166-171.
- Ewing, S.A., O'Donnell, J.A., Aiken, G.R., Butler, K., Butman, D., Windham-Myers, L., Kanevsiy, M.Z., 2015. Long-term anoxia and release of ancient, labile carbon upon thaw of Pleistocene permafrost. *Geophys. Research. Lett.* 42, 10730–10738. <http://dx.doi.org/10.1002/2015GL066296>.
- Frey, K.E., Siegel, D.I., Smith, L.C., 2007. Geochemistry of west Siberian streams and their potential response to permafrost degradation. *Water Res. Research* 43, W03406. <http://dx.doi.org/10.1029/2006WR004902>.
- Frey, K.E., Smith, L.C., 2005. Amplified carbon release from vast West Siberian peatlands by 2100. *Geophys. Res. Lett.* 32, L09401. <http://dx.doi.org/10.1029/2004GL022025>.
- Green, S.M., Baird, A.J., Boardman, C.P., Gauci, V., 2014. A mesocosm study of the effect of restoration on methane (CH₄) emissions from blanket peat. *Wetl. Ecol. Manag.* 22, 523–537. <http://dx.doi.org/10.1007/s11273-014-9349-3>.
- Hayes, D.J., Kicklighter, D.W., McGuire A.D., Chen, M., Zhuang, Q., Yuan, F., Melillo, J.M., Wullschleger, S.D., 2014. The impacts of recent permafrost thaw on land-atmosphere greenhouse gas exchange. *Environ. Res. Lett.* 9, 045005 doi:10.1088/1748-9326/9/4/045005.
- Hering, D., Carvalho, L., Argillier, C., Beklioglu, M., Borja, A., Cardoso, A.C., Duelt, H., Ferreirah, T., Globevnik, L., Hanganuj, J., Hellsten, S., Jeppesen, E., Kodeš, V., Solheim, A.L., Nögeso, T., Ormerod, S., Panagopoulos, Y., Schmutz, S., Venohrs, M., Birka, S., 2015. Managing aquatic ecosystems and water resources under multiple stress — An introduction to the MARS project. *Sci. Total Environ.* 503–504, 10–21.
- Hinkel, K.M., Eisner, W.R., Bockheim, J.G., Nelson, F.E., Peterson, K.M., Dai, X., 2003. Spatial extent, age, and carbon stocks in drained thaw lake basins on the Barrow Peninsula, Alaska. *Arct. Antarct. Alp. Res.* 35: 291–300
- Hodgkins, S.B., Tfaily, M.M., McCalley, C.K., Logan, T.A., Crill, P.M., Saleska, S.R., Rich, V.I., Chanton, J.P., 2014. Changes in peat chemistry associated with permafrost thaw increase greenhouse gas production. *PNAS* 111, 5819–5824. <http://dx.doi.org/10.1073/pnas.1314641111>.
- Hodgkins, S.B., Tfaily, M.M., Podgorski, D.C., McCalley, C.K., Saleska, S.R., Cooper, W.T., 2016. Elemental composition and optical properties reveal changes in dissolved organic matter along a permafrost thaw chronosequence in a subarctic peatland. *Geochim. Cosmochim. Acta* 187, 123–140.
- IUSS Working Group WRB, 2014. World Reference Base for Soil Resources 2014. International soil classification system for naming soils and creating legends for soil maps. World Soil Resources Reports No. 106. FAO, Rome
- Johansson, M., Åkerman, J., Keuper, F., Christensen, T.R., Lantuit, H., Callaghan, T. V., 2011. Past and present permafrost temperatures in the Abisko area: Redrilling of boreholes. *Ambio* 40, 558–565. <https://doi.org/10.1007/s13280-011-0163-3>
- Judd, K.E., Kling, G.W., 2002. Production and export of dissolved C in arctic tundra mesocosms: the roles of vegetation and water flow. *Biogeochemistry* 60, 213–234. <http://dx.doi.org/10.1023/A:1020371412061>.
- Karlsson, J., Serikova, S., Rocher-Ros G., Denfeld B., Vorobyev S.N., Pokrovsky O.S., 2021. Carbon emission from Western Siberian inland waters. *Nature Communication* 12, 825. <https://doi.org/10.1038/s41467-021-21054-1>.
- Kirpotin, S., Berezin, A., Bazanov, V., Polishchuk, Yu., Vorobiov, S., Mironycheva-Tokoreva, N., Kosykh, N., Volkova, I., Dupre, B., Pokrovsky, O., Kouraev, A., Zakharova, E., Shirokova, L., Mognard, N., Biancamaria, S., Viers, J., Kolmakova, M., 2009. Western Siberia wetlands as indicator and regulator of climate change on the global scale. *Int. J. Environ. Stud.* 66, 409–421. <http://dx.doi.org/10.1080/00207230902753056>.

- 765 Kirpotin, S., Polishchuk, Y., Bryksina, N., Sugaipova, A., Kouraev, A., Zakharova, E.,
766 Pokrovsky, O.S., Shirokova, L.S., Kolmakova, M., Manassypov, R., Dupre, B.,
767 2011. West Siberian palsa peatlands: Distribution, typology, cyclic development,
768 present day climate-driven changes, seasonal hydrology and impact on CO₂
769 cycle. *Int. J. Environ. Stud.* 68, 603–623.
770 <http://dx.doi.org/10.1080/00207233.2011.593901>.
- 771 Kokelj, S.V., Zajdlík, B., Thompson, M.S., 2009. The impacts of thawing permafrost on
772 the chemistry of lakes across the subarctic boreal-tundra transition, Mackenzie
773 Delta region, Canada. *Permafrost Periglac. Proc.* 20, 185–199.
774 <http://dx.doi.org/10.1002/ppp.641>.
- 775 Koroleff, F., 1983a. Total and organic nitrogen. In: Grasshoff K., Ehrhardt M. &
776 Kremling K. (eds.), *Methods for Seawater Analysis*, Verlag Chemie Weinheim,
777 pp. 162–168.
- 778 Koroleff, F., 1983b. Determination of phosphorus. In: Grasshoff, K., Ehrhardt, M.,
779 Kremling, K. (eds.), *Methods for Seawater Analysis*. Verlag Chemie Weinheim,
780 pp. 125–136.
- 781 Laurion, I., Vincent, W.F., MacIntyre, S., Retamal, L., Dupont, C., Francus, P., Pienitz,
782 R., 2010. Variability in greenhouse gas emissions from permafrost thaw ponds.
783 *Limnol. Oceanogr.* 55, 115–133. <http://dx.doi.org/10.4319/lo.2010.55.1.0115>.
- 784 Laurion, I., Massicotte, P., Mazoyer, F., Negandhi, K., Mladenov, N., 2021. Weak
785 mineralization despite strong processing of dissolved organic matter in Eastern
786 Arctic tundra ponds. *Limnol. Oceanogr.* 66 (S1), S47-S63. doi:
787 10.1002/lno.11634.
- 788 Lim, A.G., Loiko, S.V., Kuzmina, D., Krickov, I.V., Shirokova, L.S., Kulizhsky, S.P.,
789 Vorobyev, S.N., Pokrovsky, O.S., 2021. Dispersed ground ice of permafrost
790 peatlands: a non-accounted for source of C, nutrients and metals. *Chemosphere*
791 226, Art No 128953. <https://doi.org/10.1016/j.chemosphere.2020.128953>.
- 792 Loiko, S.V., Pokrovsky, O.S., Raudina, T., Lim, A., Kolesnichenko, L.G., Shirokova,
793 L.S., Vorobyev, S.N., Kirpotin, S.N., 2017. Abrupt permafrost collapse enhances
794 organic carbon, CO₂, nutrient, and metal release into surface waters. *Chemical*
795 *Geol.* 471, 153–165.
- 796 Lundin, E. J., Giesler, R., Persson, A., Thompson, M. S. & Karlsson, J. Integrating
797 carbon emissions from lakes and streams in a subarctic catchment. *J. Geophys.*
798 *Res. Biogeosciences* 118, 1200–1207 (2013).
- 799 Manasypov, R.M., Pokrovsky, O.S., Kirpotin, S.N., Shirokova, L.S., 2014. Thermokarst
800 lake waters across the permafrost zones of western Siberia. *The Cryosphere* 8,
801 1177–1193. <http://dx.doi.org/10.5194/tc-8-1177-2014>.
- 802 Manasypov, R.M., Vorobyev, S.N., Loiko, S.V., Kritzkov, I.V., Shirokova, L.S.,
803 Shevchenko, V.P., Kirpotin, S.N., Kulizhsky, S.P., Kolesnichenko, L.G.,
804 Zemtsov, V.A., Sinkinov, V.V., Pokrovsky, O.S., 2015. Seasonal dynamics of
805 organic carbon and metals in thermokarst lakes from the discontinuous
806 permafrost zone of western Siberia. *Biogeosciences* 12, 3009–3028.
807 <http://dx.doi.org/10.5194/bg-12-3009-2015>.
- 808 Manasypov, R.M., Shirokova, L.S., Pokrovsky, O.S., 2017. Experimental modeling of
809 thaw lake water evolution in discontinuous permafrost zone: role of peat and
810 lichen leaching and ground fire. *Science Total Environ.* 580, 245-257.
- 811 Manasypov, R.M., Lim, A.G., Krickov, I.V., Shirokova, L.S., Vorobyev, S.N.,
812 Pokrovsky, O.S., 2020. Spatial and seasonal variability of C, nutrient and trace
813 metal concentration in thermokarst lakes of western Siberia across a permafrost
814 gradient. *Water*, 12, Art No 1830; doi:10.3390/w12061830.
- 815 Mann, P.J., Eglinton, T.I., McIntyre, C.P., Zimov, N., Davydova, A., Vonk, J.E., Holmes,
816 R.M., Spencer, R.G.M., 2015. Utilization of ancient permafrost carbon in
817 headwaters of Arctic fluvial networks. *Nat. Commun.* 6, 7856.
818 <http://dx.doi.org/10.1038/ncomms8856>.

- 819 Matthews, J. A., Dahl, S.-O., Berrisford, M.S., Nesje, A., 1997. Cyclic development and
820 thermokarstic degradation of palsas in the mid-alpine zone at Leirpullan,
821 Dovrefjell, Southern Norway. *Permafr. Periglac. Process.* 8, 107–122.
- 822 Matveev, A., Laurion, I., Deshpande, B.N., Bhiry, N., Vincent, W.F., 2016. High
823 methane emissions from thermokarst lakes in subarctic peatlands. *Limnol.*
824 *Oceanogr.* 61 (S1), S150-S164.
- 825 Matveev, A., Laurion, I., Warwick, V.F., 2019. Winter accumulation of methane and its
826 variable timing of release from thermokarst lakes in subarctic peatlands. *J.*
827 *Geophys. Res. - Biogeosciences* 124(11), 3521-3535.
- 828 Natali, S.M., Schuur, E.A.G., Mauritz, M., Schade, J.D., Celis, G., Crummer, K.G.,
829 Johnston, C., Krapek, J., Pegoraro, E., Salmon, V.G., Webb, E.E., 2015.
830 Permafrost thaw and soil moisture driving CO₂ and CH₄ release from upland
831 tundra. *J. Geophys. Res.-Bioge.* 120, 525–537.
832 <http://dx.doi.org/10.1002/2014JG002872>.
- 833 Negandhi, K., Laurion, I., Whitticar, M. J., Galand, P. E., Xu, X. Lovejoy, C., 2013. Small
834 thaw ponds: An unaccounted source of methane in the Canadian high Arctic.
835 *PLoS ONE*, 8 (11) : e78204.
- 836 Nydahl, A.C., Wallin M.B., Tranvik, L.J., Hiller, C., Attermeyer, K., Julie A. Garrison,
837 J.A., Chaguaceda, F., Scharnweber, K., Weyhenmeyer, G.A., 2019. Colored
838 organic matter increases CO₂ in meso-eutrophic lake water through altered light
839 climate and acidity. *Limnol. Oceanogr.* 64(2), 744-756, DOI:
840 [10.1002/lno.11072](http://dx.doi.org/10.1002/lno.11072).
- 841 Oleinikova, O.V., Drozdova, O.Y., Lapitskiy, S.A., Demin, V.V., Bychkov, A.Y.,
842 Pokrovsky, O.S., 2017a. Dissolved organic matter degradation by sunlight
843 coagulates organo-mineral colloids and produces low-molecular weight fraction
844 of metals in boreal humic waters. *Geochim. Cosmochim. Acta* 211, 97–114.
- 845 Oleinikova, O., Shirokova, L.S., Gerard, E., Drozdova, O.Yu., Lapitsky, S.A., Bychkov,
846 A.Y., Pokrovsky; O.S., 2017b. Transformation of organo-ferric peat colloids by
847 a heterotrophic bacterium. *Geochim. Cosmochim. Acta* 205, 313–330.
- 848 Payandi-Rolland, D., Shirokova, L.S., Nakhle, P., Tesfa, M., Abdou, A., Causserand, C.,
849 Lartiges, B., Rols, J.-L., Guérin, F., Bénézeth, P., Pokrovsky, O.S., 2020a.
850 Aerobic release and biodegradation of dissolved organic matter from frozen
851 peat: Effects of temperature and heterotrophic bacteria. *Chem. Geol.* 536,
852 119448. <https://doi.org/10.1016/j.chemgeo.2019.119448>.
- 853 Payandi-Rolland, D., Shirokova, L.S., Tesfa, M., Lim, A.G., Kuzmina, D., Benezeth, P.,
854 Karlsson, J., Giesler, R., Pokrovsky, O.S., 2020b. Dissolved organic matter
855 biodegradation along a hydrological continuum in a discontinuous permafrost
856 area: case study of Northern Siberia and Sweden. *Science Total Environ.* 749,
857 Art No 141463.
- 858 Peura, S., Wauthy, M., Simone, D., Eiler, A., Einarsdottir, K., Rautio, M., Bertilsson, S.,
859 2020. Ontogenic succession of thermokarst thaw ponds is linked to dissolved
860 organic matter quality and microbial degradation potential. *Limnol. Oceanogr.*
861 65, S248-S263.
- 862 Pokrovsky, O.S., Shirokova, L.S., Kirpotin, S.N., Audry, S., Viers, J., Dupré, B., 2011.
863 Effect of permafrost thawing on organic carbon and trace element colloidal
864 speciation in the thermokarst lakes of western Siberia. *Biogeosciences.* 8, 565–
865 583. <http://dx.doi.org/10.5194/bg-8-565-2011>.
- 866 Pokrovsky, O.S., Shirokova, L.S., Kirpotin, S.N., Kulizhsky, S.P., Vorobiev, S.N., 2013.
867 Impact of Western Siberia heat wave 2012 on greenhouse gases and trace metal
868 concentration in thaw lakes of discontinuous permafrost zone. *Biogeosciences*
869 10, 5349–5365. <http://dx.doi.org/10.5194/bg-10-5349-2013>.
- 870 Pokrovsky, O.S., Manasypov, R.M., Loiko, S., Krickov, I.A., Kopysov, S.G.,
871 Kolesnichenko, L.G., Vorobyev, S.N., Kirpotin, S.N., 2016a. Trace element

872 transport in western Siberia rivers across a permafrost gradient. *Biogeosciences*
873 13, 1877–1900. <http://dx.doi.org/10.5194/bg-13-1877-2016>.

874 Pokrovsky, O.S., Manasypov, R.M., Loiko, S.V., Shirokova, L.S., 2016b. Organic and
875 organo-mineral colloids of discontinuous permafrost zone. *Geochim.*
876 *Cosmochim. Acta* 188, 1–20. <http://dx.doi.org/10.1016/j.gca.2016.05.035>.

877 Polishchuk, Y. M., I. N. Muratov, and V. Y. Polishchuk. 2020. Remote research of
878 spatiotemporal dynamics of thermokarst lakes fields in Siberian permafrost. In:
879 O.S. Pokrovsky, S. N. Kirpotin, A. I. Malov [eds], (2020) *The Arctic: Current*
880 *Issues and Challenges*. Nova Publ., Inc. New York, 425 pp, ISBN: 978-1-53617-
881 306-2.

882 Polishchuk, Y.M., Bogdanov, A.N., Muratov, I.N., Polishchuk, V.Y., Lim, A.,
883 Manasypov, R.M., Shirokova, L.S., Pokrovsky, O.S., 2018. Minor contribution of
884 small thaw ponds to the pools of carbon and methane in the inland waters of the
885 permafrost-affected part of the Western Siberian Lowland. *Environ. Res. Lett.* 13,
886 04500. <https://doi.org/10.1088/1748-9326/aab046>.

887 Polishchuk, Y.M., Bogdanov, A.N., Polishchuk, V.Y., Manasypov, R.M., Shirokova, L.S.,
888 Kirpotin, S.N., Pokrovsky, O.S., 2017. Size-distribution, surface coverage, water,
889 carbon and metal storage of thermokarst lakes (> 0.5 ha) in permafrost zone of the
890 Western Siberia Lowland. *Water* 9, 228, doi:10.3390/w9030228.

891 Raudina, T.V., Loiko, S.V., Lim, A., Manasypov, R.M., Shirokova, L.S., Istigechev, G.I.,
892 Kuzmina, D.M., Kulizhsky, S.P., Vorobyev, S.N., Pokrovsky, O.S., 2018.
893 Permafrost thaw and climate warming may decrease the CO₂, carbon, and metal
894 concentration in peat soil waters of the Western Siberia Lowland. *Sci. Total*
895 *Environ.* 634, 1004–1023. <https://doi.org/10.1016/j.scitotenv.2018.04.059>

896 Porter, K. G., and Feig, Y. S., 1980. The use of DAPI for identifying and counting
897 aquatic microflora, *Limnol. Oceanogr.* 25, 943–948.

898 Przytulska A., Comte J., Crevecoeur S., Lovejoy C., Laution I., Vincent W.F. (2016)
899 Phototrophic pigment diversity and picophytoplankton in permafrost thaw lakes.
900 *Biogeosciences* 13, 13-26.

901 Rautio, M., Dufresne, F., Laurion, I., Bonilla, S., Vincent, W.F., Christoffersen, K.S.,
902 2011. Shallow freshwater ecosystems of the circumpolar Arctic. *Écoscience* 18,
903 204–222. <http://dx.doi.org/10.2980/18-3-3463>.

904 Richardson, H., Waldron, S., Whitaker, J., Ostle, N., 2015. Measures of peatland carbon
905 cycling from peat mesocosm incubation experiments. NERC Environmental
906 Information Data Centre. [http://dx.doi.org/10.5285/e15fbbab-1cdd-4509-81a3-
907 aa050e927dd0](http://dx.doi.org/10.5285/e15fbbab-1cdd-4509-81a3-aa050e927dd0).

908 Roehm, C.L., Giesler, R., Karlsson, J., 2009. Bioavailability of terrestrial organic carbon
909 to lake bacteria: the case of a degrading high-latitude boreal and Arctic
910 permafrost mire complex. *J Geophys Res* 114, Art No G03006.

911 Roiha, T., Rautio, M., Laurion, I., 2015. Carbon dynamics in highly net heterotrophic
912 subarctic thaw ponds. *Biogeosciences* 12, 7223–7237.
913 <http://dx.doi.org/10.5194/bg-12-7223-2015>.

914 Roiha, T., Peura, S., Cusson, M., Rautio, M., 2016. Allochthonous carbon is a major
915 regulator of bacterial growth and community composition in subarctic freshwaters.
916 *Sci. Rep.* 6: 34456, doi: 10.1038/srep34456.

917 Romanovsky, V.E., Smith, S.L., Christiansen, H.H., 2010. Permafrost thermal state in the
918 polar Northern Hemisphere during the international polar year 2007–2009: a
919 synthesis. *Permafr. Periglac. Process.* 21, 106–116.
920 <https://doi.org/10.1002/ppp.689>

921 Schuur, E.A.G., McGuire, A.D., Schädel, C., Grosse, G., Harden, J.W., Hayes, D.J.,
922 Hugelius, G., Koven, C.D., Kuhry, P., Lawrence, D.M., Natali, S.M., Olefeldt, C.,
923 Romanovsky, V.E., Schaefer, K., Turetsky, M.R., Treat, C.C., Vonk, J.E., 2015.
924 Climate change and the permafrost carbon feedback. *Nature* 520, 171–179.
925 <http://dx.doi.org/10.1038/nature14338>.

- 926 Sepulveda-Jauregui, A., Walter Anthony, K. M., Martinez-Cruz, K., Greene, S. &
 927 Thalasso, F., 2015. Methane and carbon dioxide emissions from 40 lakes along a
 928 north–south latitudinal transect in Alaska. *Biogeosciences* 12, 3197–3223.
- 929 Serikova, S., Pokrovsky, O.S., Laudon, H., Krickov, I.V., Lim, A.G., Manasypov, R.M.,
 930 Karlsson, J., 2019. High carbon emissions from thermokarst lakes of Western
 931 Siberia. *Nature Communications*, 10, Art No 1552.
 932 <https://doi.org/10.1038/s41467-019-09592-1>.
- 933 Shirokova, L.S., Pokrovsky, O.S., Kirpotin, S.N., Desmukh, C., Pokrovsky, B.G., Audry,
 934 S. Viers, J., 2013. Biogeochemistry of organic carbon, CO₂, CH₄, and trace
 935 elements in thermokarst water bodies in discontinuous permafrost zones of
 936 Western Siberia. *Biogeochemistry* 113, 573–593.
 937 <http://dx.doi.org/10.1007/s10533-012-9790-4>.
- 938 Shirokova, L.S., Kunhel, L., Rols, J.-L., Pokrovsky, O.S., 2015. Experimental modeling
 939 of cyanobacterial bloom in a thermokarst lake. *Aquat. Geochem.* 21, 487–511.
 940 <http://dx.doi.org/10.1007/s10498-015-9269-8>.
- 941 Shirokova, L.S., Bredoire, R., Rols, J.-L., Pokrovsky, O.S., 2017. Moss and peat leachate
 942 degradability by heterotrophic bacteria: The fate of organic carbon and trace
 943 metals. *Geomicrobiol. J.* 34(8), 641–655.
 944 <http://dx.doi.org/10.1080/01490451.2015.1111470>.
- 945 Shirokova, L.S., Chupakov, A.V., Zabelina, S.A., Neverova, N.V., Payandi-Rolland, D.,
 946 Causserand, C., Karlsson, J., Pokrovsky, O.S., 2019. Humic surface waters of frozen
 947 peat bogs (permafrost zone) are highly resistant to bio- and photodegradation.
 948 *Biogeosciences* 16, 2511–2526. <https://doi.org/10.5194/bg-16-2511-2019>.
- 949 Shirokova, L.S., Payandi-Rolland, D., Lim, A., Manasypov, R.M., Allen, J., Bénézet P.,
 950 Rols, J.-L., Karlsson, J., Pokrovsky, O.S., 2020. Diurnal cycle of C concentration
 951 and emission fluxes in humic thaw ponds of frozen peatlands: application for C
 952 balance constraints. *Sci. Total Environ.* 737, Art No 139671,
 953 <https://doi.org/10.1016/j.scitotenv.2020.139671>
- 954 Squires, M.M., Lesack, L.F.W., Hecky, R.E., Guilford, S.J., Ramlal, P., Higgins, S.N.,
 955 2009. Primary production and carbon dioxide metabolic balance of a lake-rich Arctic
 956 river floodplain: Partitioning of phytoplankton, epipelon, macrophyte, and epiphyton
 957 production among lakes on the Mackenzie Delta. *Ecosystems* 12, 853–872, doi:
 958 10.1007/s10021-009-9263-3.
- 959 Stutter, M.I., Richards, S., Dawson, J.J.C., 2013. Biodegradability of natural dissolved
 960 organic matter collected from a UK moorland stream. *Water Res.* 47(3), 1169–
 961 1180.
- 962 Tank, S.E., Lesack, L.F.W., Hesslein, R.H., 2009. Northern delta lakes as summertime
 963 CO₂ absorbers within the Arctic landscape. *Ecosystems* 12, 144–157, doi:
 964 10.1007/s10021-008-9213-5.
- 965 Voigt, C., Marushchak, M.E., Mastepanov, M., Lamprecht, R.E., Christensen, T.R....
 966 Biasi C., 2019. Ecosystem carbon response of an Arctic peatland to simulated
 967 permafrost thaw. *Global Change Biology* 25, 1746–1764, doi:
 968 10.1111/gcb.14574.
- 969 Vonk, J.E., Tank, S.E., Bowden, W.B. Laurion, I., Vincent, W.F., Alekseychik, P.,
 970 Amyot, M., Billet, M.F., Canário, J., Cory, R.M., Deshpande, B.N., Helbig, M.,
 971 Jammet, M., Karlsson, J., Larouche, J., MacMillan, G., Rautio, M., Walter
 972 Anthony, K.M., Wickland, K.P., 2015. Reviews and syntheses: Effects of
 973 permafrost thaw on Arctic aquatic ecosystems. *Biogeosciences* 12, 7129–7167.
 974 <http://dx.doi.org/10.5194/bg-12-7129-2015>.
- 975 Walter, K.M., Chanton, J.P., Chapin III, F.S., Schuur, E.A.G., Zimov, S.A., 2008.
 976 Methane production and bubble emissions from arctic lakes: Isotopic
 977 implications for source pathways and ages. *J. Geophys. Res.* 113, G00A08.
 978 <http://dx.doi.org/10.1029/2007JG000569>.
- 979 Walter Anthony, K.M., Anthony, P., Grosse, G., Chanton, J., 2012. Geologic methane

980 seeps along boundaries of Arctic permafrost thaw and melting glaciers. *Nature*
981 *Geosci.* 5, 419–426. <http://dx.doi.org/10.1038/ngeo1480>.

982 Walter Anthony, K.M., Zimov, S.A., Grosse, G., Jones, M.C., Anthony, P.M., Chapin III,
983 F.S., Finlay, J.C., Mack, M.C., Davydov, S., Frenzel, P., Frolking, S., 2014. A
984 shift of thermokarst lakes from carbon sources to sinks during the Holocene
985 epoch. *Nature* 511, 452–456. <http://dx.doi.org/10.1038/nature13560>.

986 Wauthy, M., Rautio, M., Christoffersen, K.S., Forsström, L., Laurion, I., Mariash, H.,
987 Peura, S., Warwick, V.F., 2018. Increasing dominance of terrigenous organic
988 matter in circumpolar freshwaters due to permafrost thaw. *Limnol. Oceanogr.*
989 *Letters* 3 (3), 186-198, DOI : 10.1002/lol2.10063

990 Wauthy M., Rautio M., 2020. Permafrost thaw stimulates primary producers but has a
991 moderate effect on primary consumers in subarctic ponds. *Ecosphere* 11(6), Article
992 e03099.

993 Wik, M., Varner, R.K., Anthony, K.W., MacIntyre, S., Bastviken, D., 2016. Climate-
994 sensitive northern lakes and ponds are critical components of methane release.
995 *Nature Geosci.* 9, 99–105. <http://dx.doi.org/10.1038/ngeo2578>.

996 Zabelina, S.V., Shirokova, L.S., Klimov, S.I., Chupakov, A.V., Lim, A.G., et al., 2021.
997 Seasonal and spatial variations of CO₂ and CH₄ concentrations and fluxes in
998 surface waters of frozen peatlands (NE Europe): morphological and
999 hydrochemical control. *Limnology Oceanography* 66 (S1), S216-S230. doi:
1000 10.1002/lno.11560.

1001

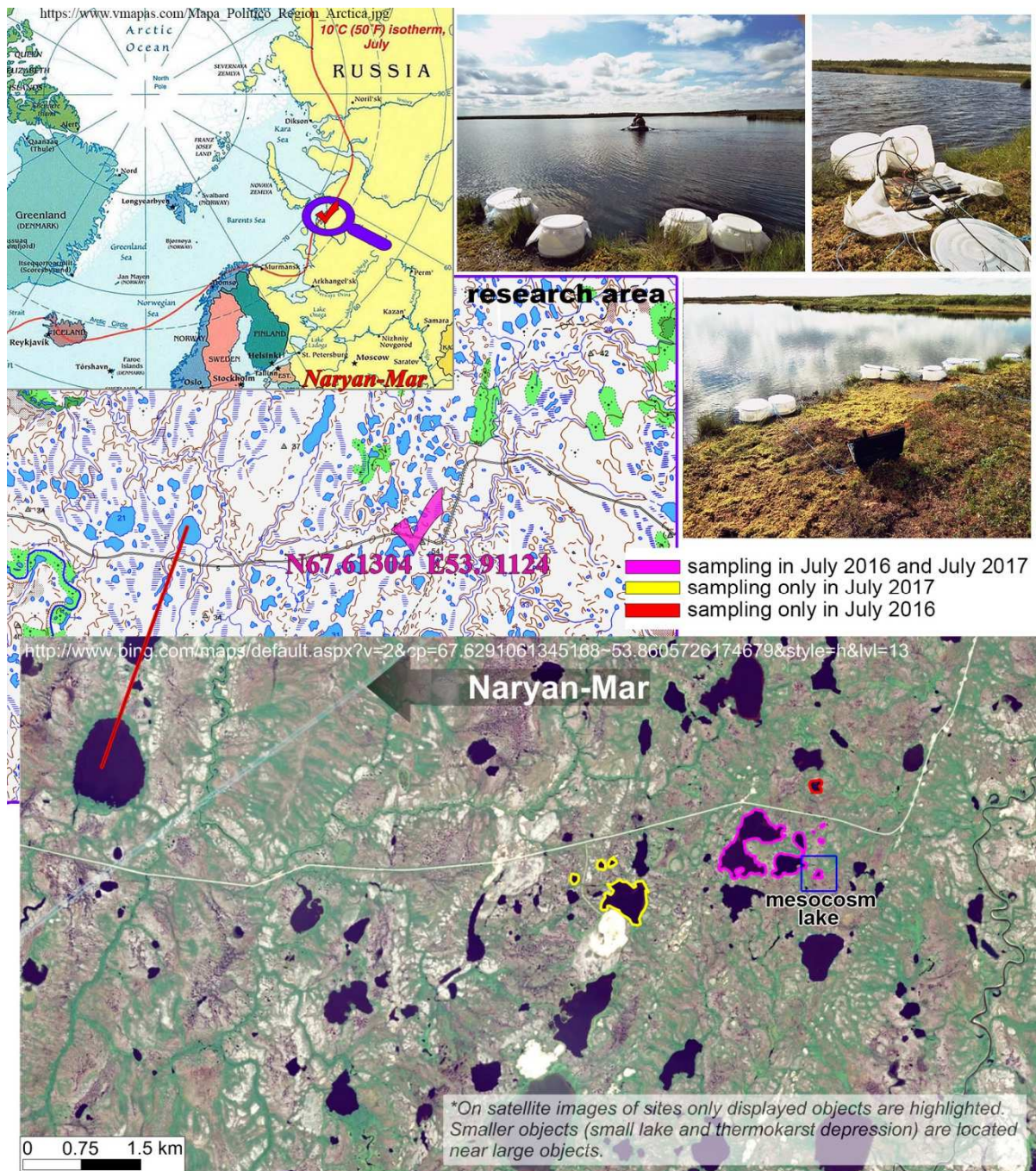
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1010 **Figure 1 A:** The map of the study site with position of sampled lakes and photos of
1011 mesocosm run at the lake shore.

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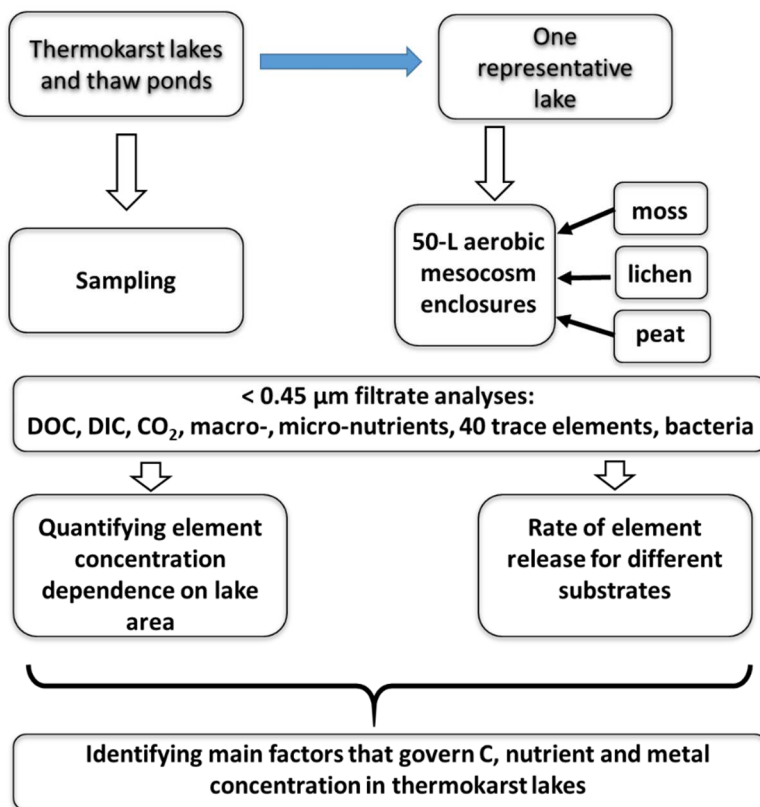
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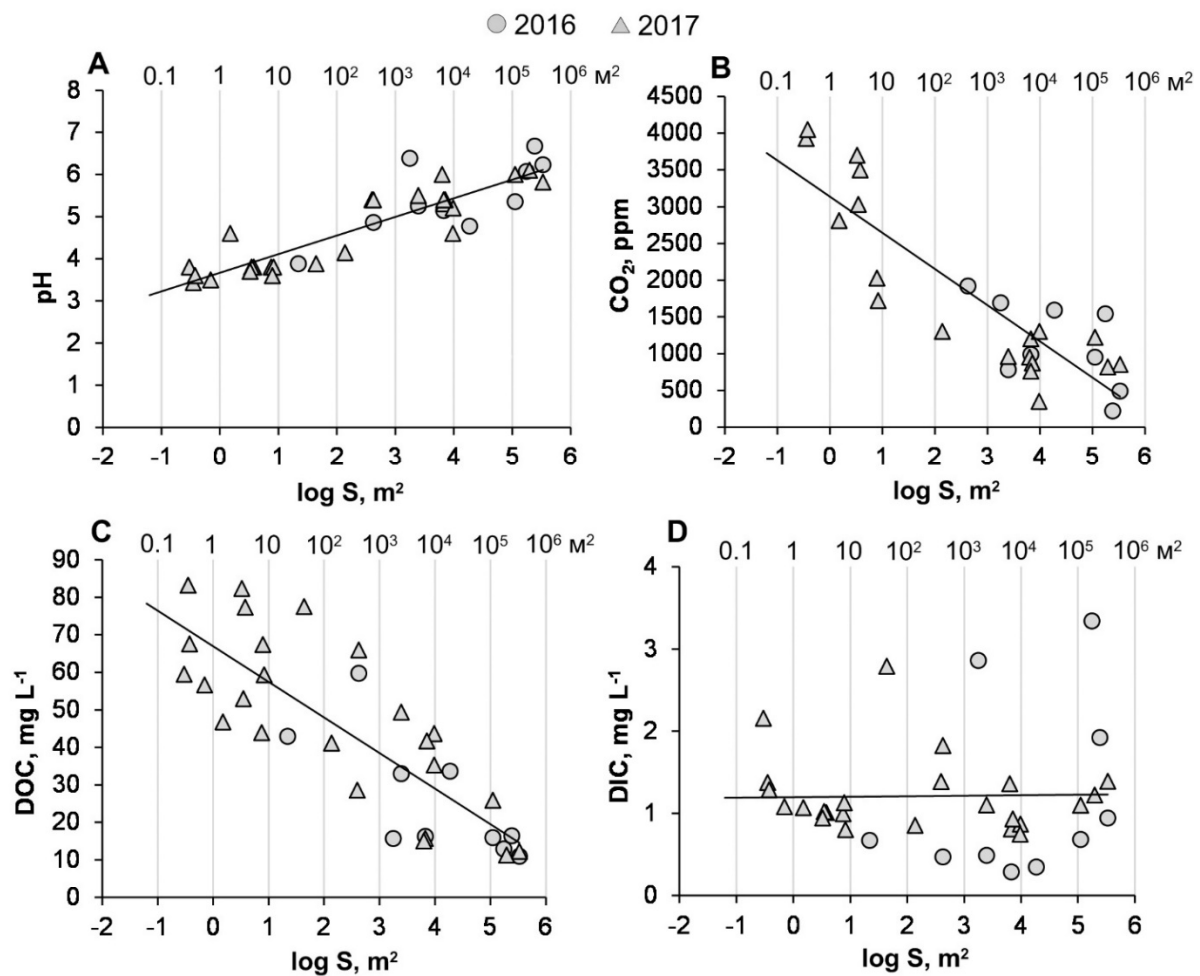
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1021 **Figure 1 B:** The work flow chart of the field observations, mesocosm experiments and
1022 analyses performed in this study.



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1025 **Figure 2.** pH (A), pCO₂ (B), DOC (C) and DIC (D) concentration in depressions, thaw

1026 ponds and thermokarst lakes as a function of logarithm of lake surface area (log S).

1027 Parameters of linear relationships (Eqn. 1) are listed in Table S1 of the Supplement.

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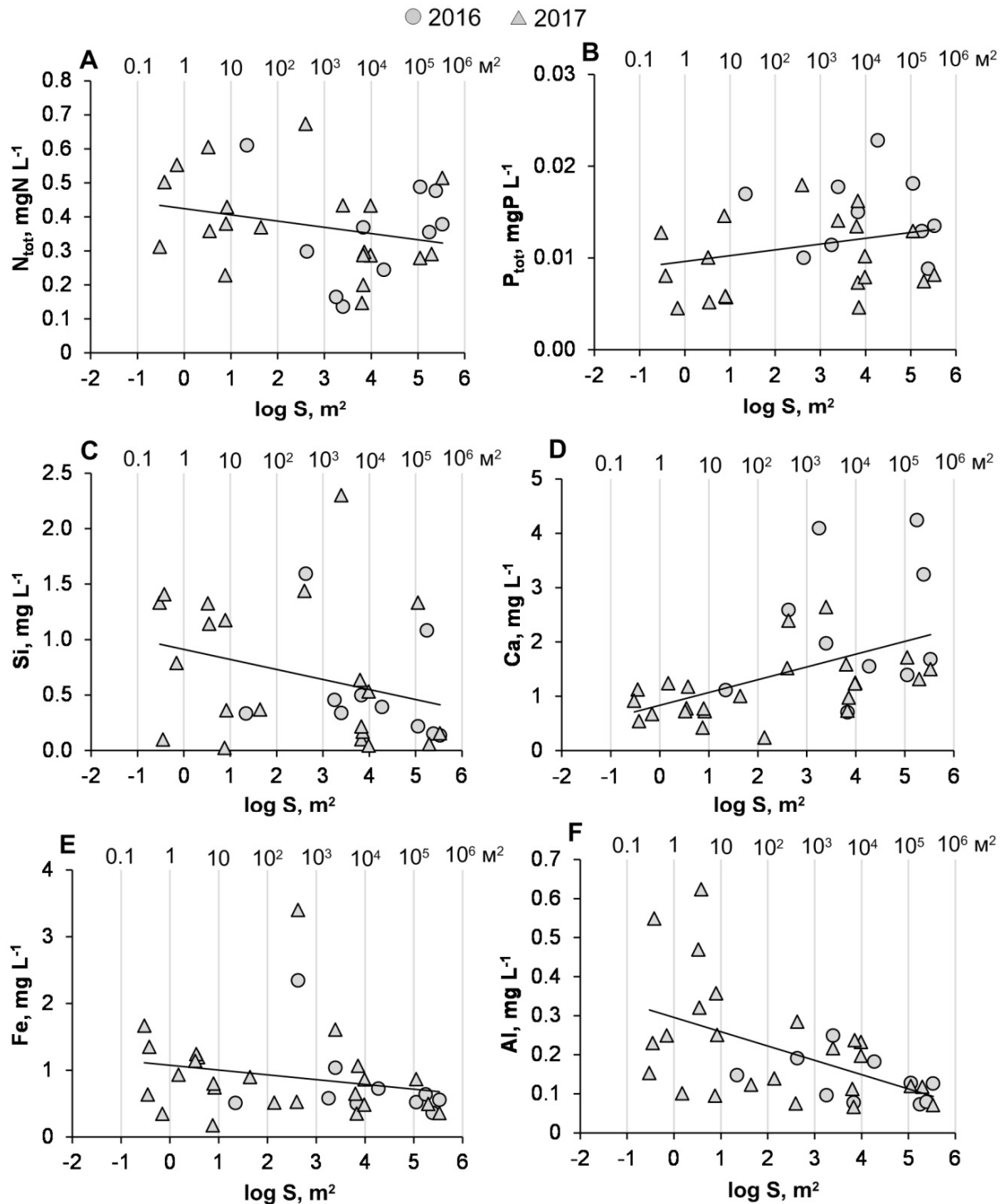
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1040 **Figure 3.** Total dissolved N (A), P (B), Si (C), Ca (D), Fe (E) and Al (F) concentrations
 1041 in depressions, thaw ponds and thermokarst lakes as a function of logarithm of
 1042 lake surface area (log S). Parameters of linear relationships are listed in Table S1.

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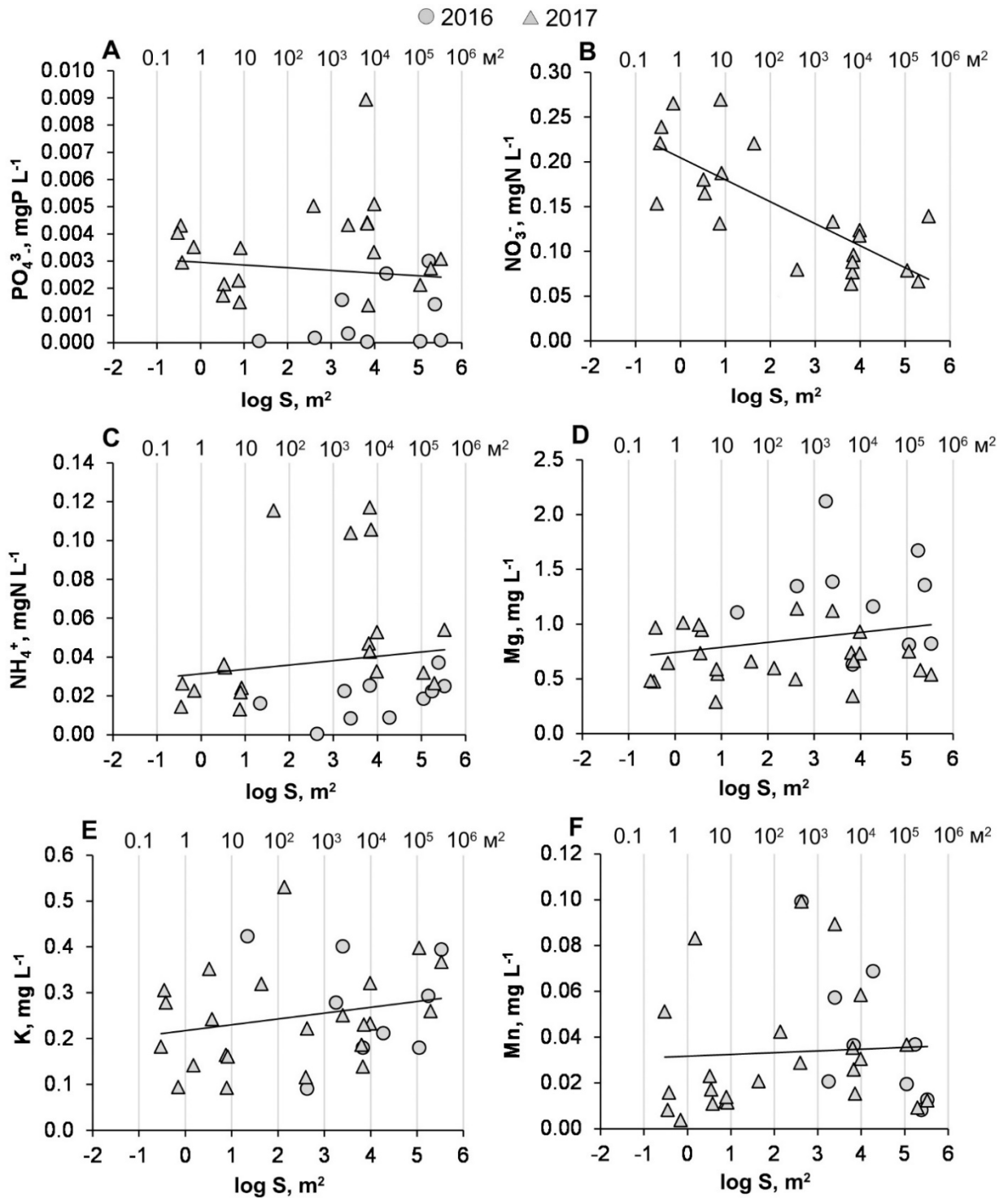
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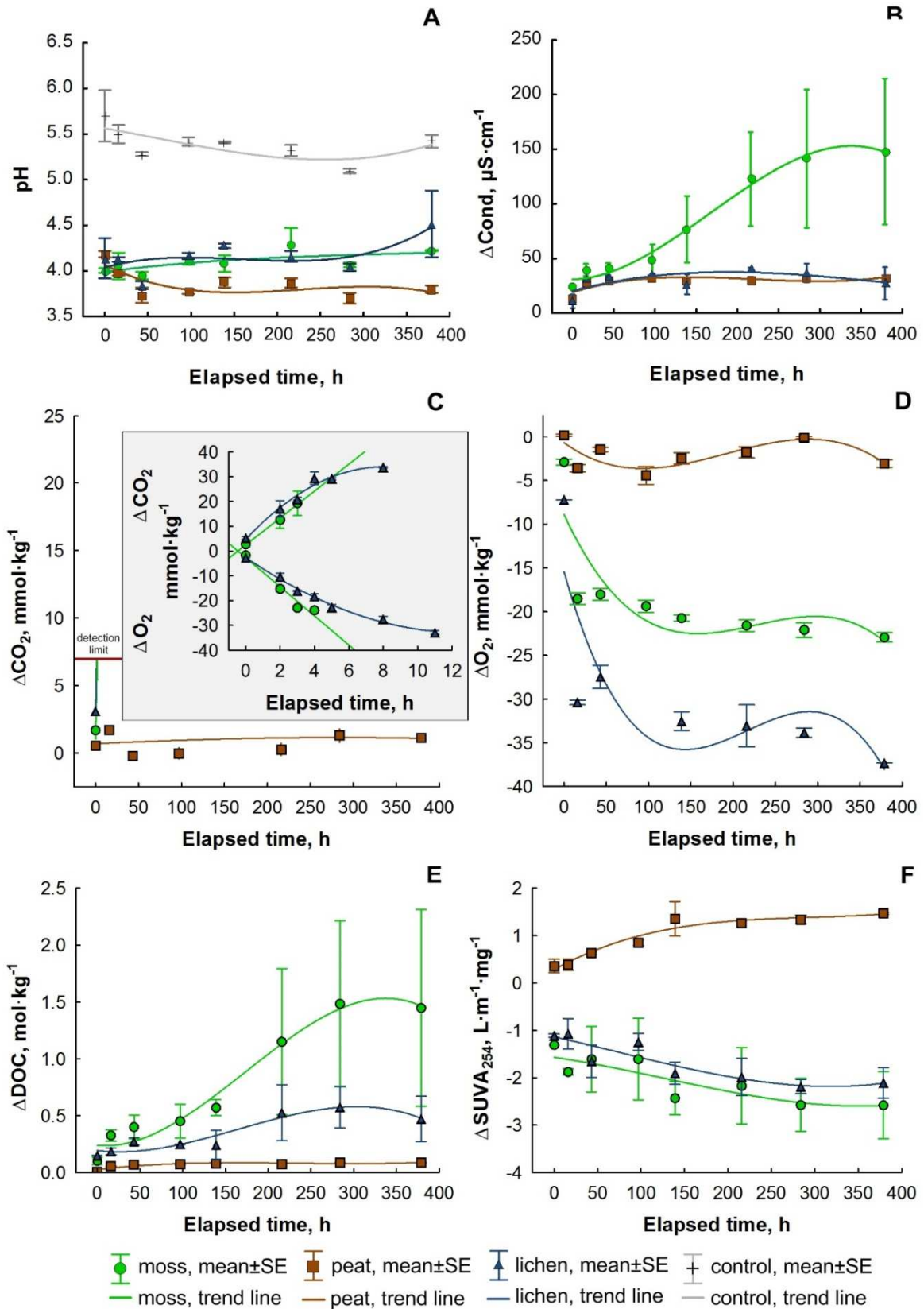
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1052 **Figure 3**, continued. Phosphate, nitrate, ammonia, Mg, K and Mn concentrations in
1053 thermokarst water bodies as a function of logarithm of lake surface area ($\log S$).

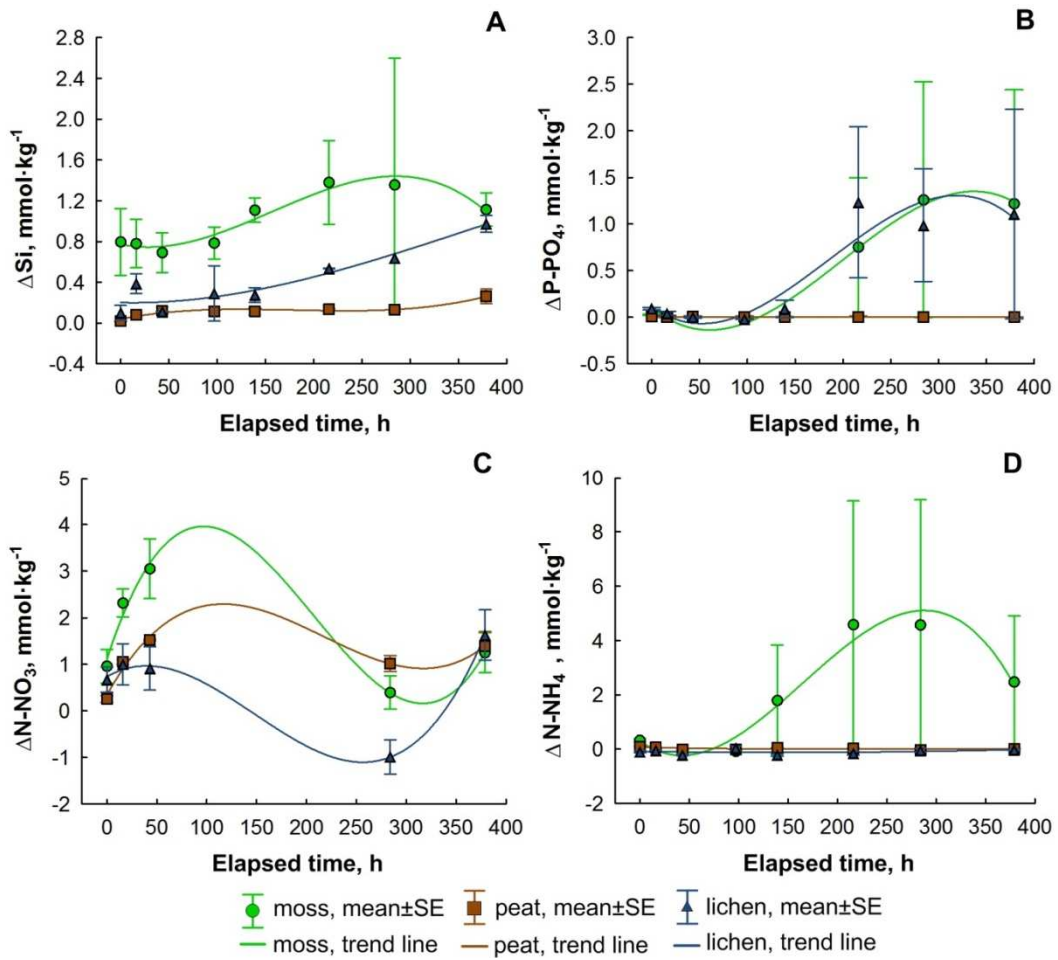
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1063 **Figure 4.** The pH value (A), and the difference between experiment (moss, lichen or
1064 peat) and control (lake water without substrate) in Specific Conductivity (B), O $_2$ (C),
1065 pCO $_2$ (D), DOC (E) concentrations and SUVA $_{254}$ (F) in mesocosms as a function of
1066 exposure time. Here and in other figures below, symbols represent the mean; the error
1067 bars correspond to the S.E. of two independent duplicate experiments. Solid lines
1068 represent a polynomial fit to the data that was used to calculate the maximal rates and
1069 concentration yields.

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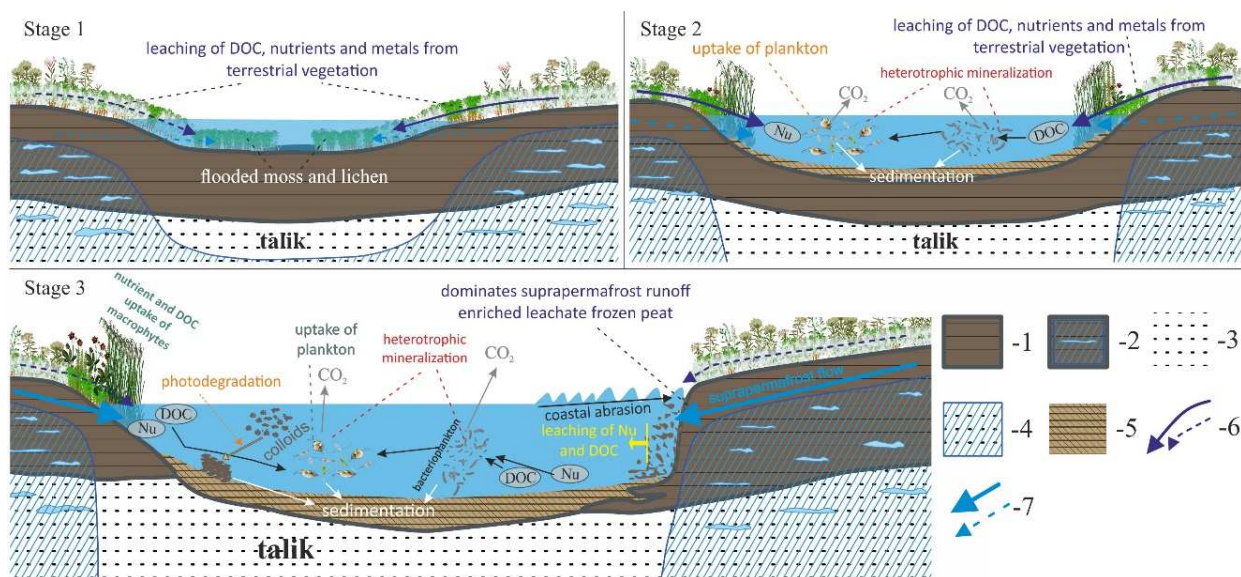


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Figure 5. Evolution of the difference in Si (A), PO_4 (B), NO_3 (C), and NH_4 (D) concentrations between the mesocosm (moss, lichen or peat) and control in the course of experiments. See Fig. 4 caption for detailed legend.

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Legend:

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1 - Peat of active (unfrozen) layer

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2 - Frozen peat with ice lenses

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3 - Unfrozen mineral horizons

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4 - Frozen mineral horizons

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5 - Lake sediments

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6 - Flux of DOC and nutrients that are leached from plant litter

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7 - Flux of DOC and nutrients that are leached from peat (both active and permafrost layer)

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Figure 6. A cartoon of thaw pond and thermokarst lake evolution and the change of

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sources of DOC and nutrients in the course of evolution, from surface moss and

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lichen to frozen peat. A three main stage sequence of thermokarst lake formation

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and development includes:

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(1) soil depression filled by water which interacted with submerged moss and

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lichens thus leaching DOM and solutes to the lake;

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(2) beginning of in-lake processing of DOM via heterotrophic plankton

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mineralization and sedimentation to the bottom, occurring under the dominance of allochthonous input from vegetation leaching;

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(3) stage of large mature lake, influenced by nutrients and DOM input from thawed and frozen peat leaching and nutrients and CO₂ uptake by phytoplankton,

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under on-going DOM, nutrient and metal processing via photo- and biodegradation in the water column.

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1119 **Table 1.** The mean and maximal release rates and maximal concentration yield of major
 1120 components normalized to dry weight of substrate in mesocosm experiments. Significant
 1121 ($p < 0.05$) results are labelled with asterisk (*).

Index	Substr.	R ²	F-test ($p=0.05$)	$\mu_{\text{mean}} \pm \text{SE}, \text{X} \cdot \text{hr}^{-1}$	$\mu_{\text{max}} \pm \text{SE}, \text{X} \cdot \text{hr}^{-1}$	$\Delta C_{\text{max}} \pm \text{SE}$	$T_{\Delta C_{\text{max}}}$
EB, CFU · 10 ⁶ · kg ⁻¹	moss	0.64	7.2*	0.05 ± 0.02	0.1 ± 0.4	21 ± 7	379
	peat	0.74	11.3*	0.022 ± 0.001	0.036 ± 0.004	2.1 ± 0.1	97
	lichen	0.88	29.2*	0.00074 ± 0.00001	0.33 ± 0.03	58 ± 3	379
OB, CFU · 10 ⁶ · kg ⁻¹	moss	0.92	48.3*	0.044 ± 0.002	0.08 ± 0.02	17.5 ± 0.8	379
	peat	0.49	3.8*	0.012 ± 0.005	0.05 ± 0.02	5 ± 1	216
	lichen	0.92	45.0*	0.11 ± 0.02	0.23 ± 0.09	42 ± 6	379
O ₂ , mmol · kg ⁻¹ (long)	moss	0.68	8.62*	-0.128 ± 0.007	-0.98 ± 0.09	-22.9 ± 0.7	379
	peat	0.52	4.3*	-0.05 ± 0.01	-0.24 ± 0.04	-4 ± 1	97
	lichen	0.68	8.62*	-0.182 ± 0.008	-1.45 ± 0.02	-37	97
CO ₂ , mmol · kg ⁻¹ (short, for peat long)	moss	0.78	14.4*	6 ± 2	6 ± 2	19 ± 5	3
	peat	0.10	0.44	0.0026 ± 0.0007	-0.112 ± 0.0064	4.7 ± 0.7	139
	lichen	0.95	80.5*	4.23 ± 0.04	6.0 ± 0.8	33.8 ± 0.4	8
DIC, mmol · kg ⁻¹	moss	0.31	1.78	0.14 ± 0.03	0.7 ± 0.2	14 ± 3	97
	peat	0.38	2.42	0.006 ± 0.006	0.01 ± 0.02	1 ± 1	216
	lichen	0.90	37.7*	0.323 ± 0.007	0.54 ± 0.06	31.4 ± 0.7	97
DOC, mmol · kg ⁻¹	moss	0.52	4.39*	5 ± 4	8 ± 13	1484 ± 1034	284
	peat	0.78	14.0*	0.52 ± 0.05	1.5 ± 0.1	89 ± 2	284
	lichen	0.50	4.01*	2 ± 1	4 ± 5	577 ± 184	284
NH ₄ ⁺ , mmol(N) · kg ⁻¹	moss	0.34	2.08	0.02 ± 0.03	0.04 ± 0.12	5 ± 6	216
	peat	0.36	2.25	-0.0002 ± 0.0001	0.0006 ± 0.0005	-0.04 ± 0.02	284
	lichen	0.11	0.49	0.0002 ± 0.0002	0.0002 ± 0.0010	0.03 ± 0.08	97
NO ₃ ⁻ , mmol(N) · kg ⁻¹	moss	0.81	8.58*	0.05 ± 0.03	0.08 ± 0.06	3.1 ± 0.9	43
	peat	0.85	11.1*	0.030 ± 0.002	0.050 ± 0.008	1.53 ± 0.02	43
	lichen	0.80	8.18*	0.01 ± 0.02	0.02 ± 0.04	1.6 ± 0.5	379
NO ₂ ⁻ , μmol(N) · kg ⁻¹	moss	0.70	9.44*	0.4 ± 0.6	1 ± 2	15 ± 10	43
	peat	0.81	17.5*	0.05 ± 0.02	0.10 ± 0.06	9.8 ± 0.5	379
	lichen	0.09	0.4	0.04 ± 0.06	0.1 ± 0.2	-21 ± 2	16
N _{tot} , mmol(N) · kg ⁻¹	moss	0.76	6.3*	0.014 ± 0.009	0.04 ± 0.03	8 ± 2	284
	peat	0.88	14.3*	0.006 ± 0.003	0.04 ± 0.01	3.4 ± 0.5	379
	lichen	0.75	6.09*	0.03 ± 0.01	0.09 ± 0.08	12 ± 4	379
PO ₄ ³⁻ , μmol(P) · kg ⁻¹	moss	0.38	2.46	4 ± 6	7 ± 42	1262 ± 1790	284
	peat	0.04	0.16	-0.016 ± 0.007	0.01 ± 0.14	0.7 ± 0.4	139
	lichen	0.43	3.03	5 ± 4	11 ± 7	1234 ± 812	216
P _{tot} , μmol(P) · kg ⁻¹	moss	0.47	1.79	0.4 ± 0.2	0.7 ± 0.3	1647 ± 2146	379
	peat	0.19	0.45	-0.02 ± 0.20	0.27 ± 0.05	54 ± 32	0
	lichen	0.54	2.32	5 ± 4	7 ± 5	1695 ± 1068	284
Si, mmol · kg ⁻¹	moss	0.20	0.99	0.003 ± 0.005	0.005 ± 0.007	1.4 ± 0.6	216
	peat	0.75	11.8*	0.0007 ± 0.0003	0.0024 ± 0.0009	0.26 ± 0.07	379

	lichen	0.76	12.76*	0.0023±0.0004	0.0035±0.0009	0.97±0.08	379
Na, mmol·kg ⁻¹	moss	0.80	15.8*	0.08±0.04	0.11±0.1	32±16	379
	peat	0.69	8.98*	0.0005±0.0002	0.0036±0.0005	-0.035±0.005	379
	lichen	0.63	6.67*	0.03±0.02	0.06±0.02	9±1	284
K, mmol·kg ⁻¹	moss	0.62	6.53*	0.2±0.2	0.3±0.4	70±49	379
	peat	0.13	0.6	0.00002±0.00020	-0.0014±0.0004	-0.03±0.05	379
	lichen	0.34	2.05	0.185±0.005	0.37±0.02	22.5±0.1	97
Ca, mmol·kg ⁻¹	moss	0.24	1.27	0.01±0.01	0.01±0.03	3±3	284
	peat	0.68	8.55*	0.008±0.002	0.019±0.008	0.7±0.2	379
	lichen	0.85	22.5*	-0.015±0.001	-0.03±0.01	-2.21±0.01	139
Mg, mmol·kg ⁻¹	moss	0.34	2.04	0.02±0.02	0.02±0.05	5±7	379
	peat	0.81	17.2*	0.0005±0.0001	0.002±0.001	-0.45±0.02	379
	lichen	0.82	18.2*	-0.025±0.003	-0.10±0.02	-1.6±0.1	97
Al, mmol·kg ⁻¹	moss	0.48	3.69*	0.0014±0.0005	0.003±0.003	0.14±0.03	97
	peat	0.91	39.3*	0.00041±0.00003	0.0018±0.0003	0.149±0.007	379
	lichen	0.99	263*	-0.0014±0.0002	-0.0028±0.0007	-0.223±0.006	379
Fe, mmol·kg ⁻¹	moss	0.54	4.63*	0.004±0.002	0.005±0.006	1.4±0.4	284
	peat	0.91	42.3*	0.00040±0.00004	0.0007±0.0004	0.086±0.009	379
	lichen	0.85	22.2*	-0.0054±0.0001	-0.0167±0.0005	-0.959±0.007	97

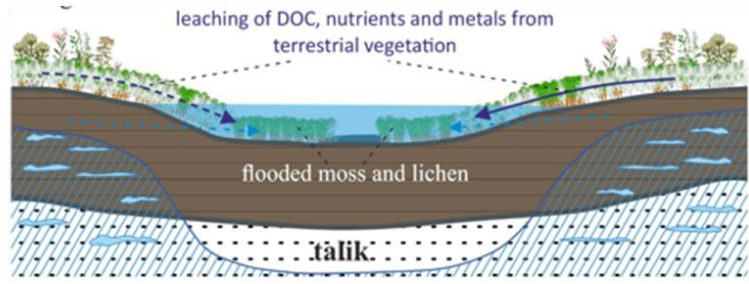
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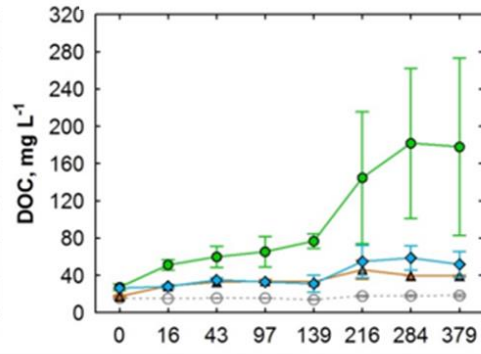
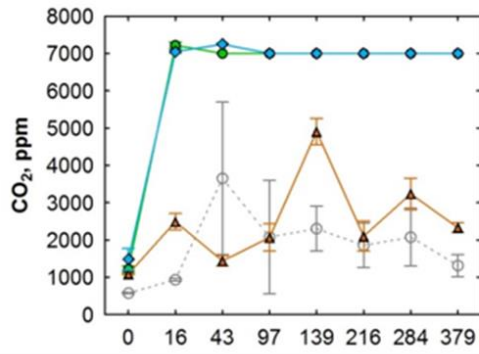
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○ control ● moss ▲ peat ◆ liches



Elapsed time in mesocosm, hr