

1 **Taxon-specific variation in $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ of subfossil invertebrate remains: Insights into**
2 **historical trophodynamics in lake food-webs**

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8 Abstract

9 Carbon and nitrogen stable isotope ratios of sub-fossil invertebrate remains are
10 potentially powerful indicators of nutrient flux, habitat-specific resource utilization, and trophic
11 interactions in lentic food webs, but are rarely estimated for multiple species within lakes. Here
12 we examined historical time series of $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ in remains of individual invertebrate taxa
13 representing pelagic, littoral and benthic habitats during the 20th century in five boreal lakes of
14 central Canada. We applied a novel statistical approach based on Generalized Additive Models
15 (GAMs) to quantify the differences in centennial means and trends (i) between invertebrate
16 remains and sedimentary organic matter (SOM), and (ii) among different taxa within each lake to
17 evaluate the coherence of isotope signals during the 20th century. Differences in mean $\delta^{13}\text{C}$ and
18 $\delta^{15}\text{N}$ were usually significant ($p < 0.05$) between SOM and invertebrate taxa, and among
19 individual taxa, reflecting selective feeding by invertebrates and differences in trophic position
20 within food webs. In contrast, patterns of historical variance in isotope values varied among
21 lakes with few consistent differences between long-term isotopic trends of SOM and invertebrate
22 remains. In particular, SOM and invertebrate isotopic trends were similar in relatively
23 dystrophic lakes, likely due to the importance of terrestrial carbon in both SOM and invertebrate
24 diets. However, significant SOM-invertebrate trend differences were observed for both $\delta^{13}\text{C}$ and
25 $\delta^{15}\text{N}$ in relatively clear-water lakes, possibly reflecting temporal variation in diets or tissue
26 fractionation. Comparisons of historical trends in isotope values among taxa revealed few
27 consistent patterns, likely indicating uncoupled carbon and nitrogen fluxes through invertebrate
28 consumers with different habitat specializations or feeding modes. Together, our findings
29 suggest that taxon-specific $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ can provide valuable insights into historical tropho-
30 dynamics in lake food webs beyond those generated by SOM analysis.

31 1. Introduction

32 Abundance, composition and size of chitinous invertebrate remains preserved in lake
33 sediments are often used as a paleo-environmental indicators of historical changes in food-web
34 composition and trophic structure (Jeppesen et al., 2001). Recently, these analyses have been
35 expanded to include estimates of the elemental composition of sub-fossils, including carbon (C)
36 and nitrogen (N) stable isotope (SI) ratios as proxies for historical trophic dynamics (Frossard et
37 al., 2013b; Perga, 2009; Schilder et al., 2017; van Hardenbroek et al., 2013). The
38 appropriateness of sub-fossil invertebrate SI ratios as paleolimnological proxies of food-web
39 processes is supported by several lines of evidence. First, SI ratios of sub-fossil exoskeletons and
40 resting eggs of invertebrates are predictably related to their whole body values (Perga, 2011;
41 Schilder et al., 2015b), as well as dietary source material (Frossard et al., 2013a; Perga, 2009).
42 Second, SI ratios of invertebrate remains are largely unaffected by acid and base pre-treatments
43 usually used for sediment processing (Perga, 2009; van Hardenbroek et al., 2010). Third,
44 taphonomic and early diagenetic processes appear to have little effect on carbon SI ratios ($\delta^{13}\text{C}$)
45 of invertebrate remains, although corrections for systematic changes in nitrogen SI ratios ($\delta^{15}\text{N}$)
46 with time may be required (Perga, 2011). Finally, the evaluation of SI stratigraphies of subfossil
47 invertebrate remains is facilitated by well-established understanding of the relation between
48 contemporary food-web processes and isotopic composition of living zooplankton and
49 zoobenthos (Grey, 2006). Despite these advances, very little is known of whether SI of sub-fossil
50 remains of individual taxa record whole-lake or habitat-specific processes (changes in C and
51 nutrient fluxes) or historical variation in trophic positions within the food web (Jeppesen et al.
52 2001).

53 Unlike SI composition of invertebrate remains, analysis of $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ of bulk
54 sedimentary organic matter (SOM) has been commonly incorporated into paleolimnological
55 studies. SOM is a heterogeneous mixture of plant-, animal- and microbial-derived organic
56 matter that originated in different habitats of the lake (i.e. littoral, pelagic and benthic) and
57 received from the watershed (Meyers and Teranes, 2001). Consequently, accurate interpretation
58 of the mechanisms underlying historical changes in isotopic values of SOM can be difficult due
59 to the potential for multiple pathways influencing both mass fluxes and isotopic value of source
60 materials within the lake and its watershed (Meyers and Teranes, 2001; van Hardenbroek et al.,
61 2014). In this regard, isotopic trends of sub-fossil invertebrate remains are considered to be
62 better indicators of changes in C and N dynamics within food webs than those of SOM (Griffiths
63 et al., 2010; Simon et al., 2017; van Hardenbroek et al., 2014), given *in situ* primary production
64 likely contribute relatively little to total SOM than terrestrial organic matter (Von Wachenfeldt
65 and Tranvik, 2008), and invertebrate consumers are thought to feed selectively on autochthonous
66 sources (del Giorgio and France 1996; Meili et al. 1996; Doi et al. 2006). However, isotopic
67 trends of sub-fossil invertebrate remains and SOM may not necessarily and universally be
68 different. Not all invertebrate taxa are equally selective in their grazing (Demott, 1982;
69 Tanentzap et al., 2017). Further, in systems where available basal resources are overwhelmingly
70 dominated by allochthonous sources, invertebrates may be forced to feed less-selectively (Cole
71 et al., 2011; Persaud et al., 2009; Tanentzap et al., 2017) and consequently, isotopic trends of
72 invertebrate subfossils may not differ from those of SOM with predominant allochthonous origin
73 (Simon et al., 2017). This urges the need to test the significance of difference in temporal
74 patterns between SOM and invertebrate subfossils in each lake. Yet, in past studies, such

75 differences were inferred only from pair-wise comparisons of raw values without statistically
76 testing their significance (Kattel et al., 2015; Perga et al., 2010; van Hardenbroek et al., 2014).

77 Comparisons of isotopic trends among invertebrate remains from multiple taxa can yield
78 insights into historical changes in energy and nutrient flux through different food-web
79 compartments of lakes. For instance, $\delta^{13}\text{C}$ values within a given ecosystem may differ among
80 invertebrate taxa with different habitat affinities (i.e., pelagic, littoral, benthic) and/or feeding
81 modes owing to differences in dietary sources of C (Matthews and Mazumder, 2003; Persaud et
82 al., 2009), which may converge or further diverge over time in response to changes in basal
83 resources (del Giorgio and France, 1996; Grey et al., 2001; Vadeboncoeur et al., 2003). For
84 $\delta^{15}\text{N}$, diverging or converging temporal trends between two taxa may indicate variations in their
85 relative trophic positions over time (Patoine et al. 2006; Perga et al. 2010) or shifts in selective
86 feeding on alternate N sources (France, 1995a; Mulholland et al., 2000). Fortunately, recent
87 improvements in technology to lower mass required for analysis (Maddi et al., 2006; Perga,
88 2009; van Hardenbroek et al., 2010) have enabled investigators to determine the SI values of
89 individual taxa (Frossard et al., 2013b; van Hardenbroek et al., 2014, 2013; Wooller et al., 2012),
90 although better statistical analysis of the significance of historical trends is still needed.

91 Changes in C and N cycling in Canadian Boreal Shield region during the past century due
92 to environmental perturbations (Anas et al., 2015; Kurz et al., 2014) may have in turn affected
93 trophic dynamics in lacustrine food webs. Several studies have reported not only multi-decadal
94 changes in dissolved organic matter (DOM) inputs to lakes from terrestrial sources, but also
95 altered in-lake C processing in the Boreal Shield region in response to individual or combined
96 effects of changes in temperature and precipitation and/or atmospheric deposition of sulfur and N
97 species from anthropogenic sources, or recovery from it (Dillon and Molot, 2005; Keller et al.,

98 2008; Zhang et al., 2010). In addition, these perturbations have also induced changes in nutrient
99 dynamics and productivity in lakes (Dillon and Molot, 2005; Holtgrieve et al., 2011; Schindler
100 and Lee, 2010). The nature and degree of responses likely vary among individual lakes
101 depending on local characteristics, such as catchment properties, lake morphometry, type and
102 degree of nutrient limitation (Anas et al., 2015; Zhang et al., 2010). These changes in turn may
103 have variably influenced energy and nutrient fluxes through different invertebrate taxa with
104 different habitat or feeding niches within lakes. For instance, changes in allochthonous DOM
105 inputs may strongly influence the diet of non-selective feeders relative to selective feeders of
106 phytoplankton (Persaud et al., 2009; Tanentzap et al., 2017). Further, the diet of littoral
107 consumers can be affected more than that of pelagic consumers by a shift from predominant
108 benthic production to a pelagic regime following eutrophication (Vadeboncoeur et al., 2003).
109 However, such historical food-web consequences of environmental perturbations in Boreal
110 Shield lakes are poorly understood (Persaud et al., 2009) owing to paucity of long-term food-web
111 studies (Schindler and Lee, 2010) .

112 Here, we quantified time series of $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ in SOM and fossils from individual
113 invertebrate taxa during the 20th century in five Boreal Shield lakes of central Canada to evaluate
114 whether taxon-specific SI analyses improve insights concerning historical changes in lacustrine
115 ecosystems. These systems are heterogeneous in terms of local characteristics including lake
116 morphometry, physiochemical conditions, catchment properties and proximity to nearby major
117 source of atmospheric pollutants i.e. Athabasca Oil Sands Region (AOSR). (See below).

118 Specifically, we analyzed chitinous remains of taxa representing different habitats of lakes (and
119 likely different feeding modes) i.e. *Daphnia* and *Bosmina* spp. (pelagic), *Alona* sp. (littoral) and
120 Chironomidae (benthic), and statistically compared the isotopic trajectories (i) between SOM and

121 invertebrate taxa, and (ii) among invertebrate taxa within each lake. We hypothesized that
122 isotopic trends of invertebrate subfossils reflective of food web-specific dynamics would be
123 significantly different from SOM which derives from heterogeneous sources, yet such
124 differences would be system- and/or taxon-specific. Further, we expected that SI trends would
125 be significantly different among invertebrate taxa representing different habitat or feeding niches
126 due to variable food-web related effects of above-mentioned environmental perturbations
127 occurring in the Boreal Shield region. We also hypothesized that among-taxon differences in SI
128 trends would be system-specific as different lakes may have responded differently to
129 environmental perturbations due to differences in local characteristics. To our knowledge, this is
130 the first study where taxon-specific SI analyses of invertebrate remains has been performed on
131 multiple study lakes, and in which robust statistical approaches were applied to compare isotopic
132 trends of invertebrate taxa and SOM.

133 **2. Methods**

134 *2.1. Study lakes*

135 Our five study lakes were a subset of 16 lakes in northwest Saskatchewan (Fig. 1)
136 described in Mushet et al. (2017), that represent 262 basins surveyed by the provincial
137 government during 2007-2009 and 2011 (Scott et al., 2010). Initially, sites were selected to
138 represent type of nutrient limitation i.e. phosphorus-limited vs. nitrogen-limited (based on molar
139 dissolved inorganic nitrogen to total phosphorus [DIN: TP] ratios) and degree of vulnerability to
140 atmospheric emissions from AOSR (distance from the industrial center) (Table 1). Of the 16
141 lakes cored, the five lakes examined here were chosen on the basis of their elevated densities of
142 fossil invertebrate remains (Hesjedal, 2017) which allowed sufficient sample size to conduct

143 taxon-specific SI analyses. Final lake selection included two N-limited lakes (basins 17V, 6E)
144 and three phosphorus-limited lakes (17P, 17F, 13N).

145 Study lakes are mostly small (surface area < 65 ha) and shallow (maximum depth < 10
146 m), with the exception of 13N which is both relatively large and deep (Table 1). The pH of each
147 basin was circumneutral (range 6.8-7.3), yet alkalinity varied considerably among lakes (range
148 4.2-16.5 mg L⁻¹ CaCO₃). Study lakes were largely oligotrophic to mesotrophic, as reflected by
149 low water-column concentration of TP (range 5.0-16.5 µg L⁻¹), total nitrogen (TN; range 253-
150 315 µg L⁻¹) and chlorophyll a (range 2.0-6.1 µg L⁻¹). In 13N and 17P, dissolved organic carbon
151 (DOC) concentration (each 7.1 mg L⁻¹) and water color (26.2 and 32.8 mg L⁻¹ Pt units,
152 respectively) were considerably higher than in other study lakes, indicating the greater
153 importance of allochthonous DOM in these lakes (Wetzel, 1983). We refer to Scott *et al.*, (2010)
154 and Anas *et al.*, (2014) for detailed descriptions of survey domain, water chemistry and hydro-
155 morphological attributes of study lakes.

156 2.2. Sediment coring

157 Sediment cores were collected in February and April 2014. A single core was retrieved
158 from the approximate center of each lake (Table 1) using a Glew gravity corer with a 7.6-cm
159 internal diameter tube. The length of the cores varied from 36.5 to 48.5 cm, which were
160 sectioned into 0.5-cm intervals upon return to the base camp. Samples were shipped to Queen's
161 University, Kingston, Ontario where they were stored at 4°C in a cold room until further
162 analyses.

163

164

165 2.3. *Core chronologies*

166 Details on radiometric dating (using ^{210}Pb activities) and age models of the cores are
167 described in Mushet et al. (2017). All cores exhibited robust depth-time chronologies, with the
168 sediment accumulation rates of 17V, 13N and 6E sufficient to achieve a temporal resolution
169 between 2 and 8 years (spaced 0.5 or 1 cm apart, respectively). In contrast, temporal resolutions
170 of cores from lakes 17F and 17P were ~3 - 13 years and 8 - 16 years, respectively, reflecting
171 lower mass accumulation rates (Mushet et al., 2017).

172 2.4. *SI analysis of SOM*

173 Approximately 3 mg of freeze-dried sediment from each interval were packed in tin
174 capsules, and combusted in an Elemental Combustion System (Costech EA) coupled with a
175 Thermo-Finnigan Delta V isotope ratio mass spectrometer (IRMS). Bovine liver and wheat flour
176 were used as internal laboratory standards. Isotopic ratios are reported in the conventional δ
177 notation (‰), relative to Vienna Pee-Dee-Belemnite (C) and atmospheric N_2 (N) standards
178 (Cooper and Wissel, 2012). Agreement between duplicate sub-samples was within 0.2‰ for
179 each element. Acidification of sediment samples indicated that inorganic C was negligible in all
180 cores (Wolfe, 2016).

181 2.5. *SI analysis of subfossil invertebrate remains*

182 To recover sufficient subfossil invertebrate remains for SI analysis, sediment samples
183 were first deflocculated in warm 10% KOH for 2 hrs, then thoroughly rinsed with deionized
184 water and concentrated onto a 106- μm mesh sieve (Perga, 2011; van Hardenbroek et al., 2010).
185 Subsequently, the chitinous remains from each taxon were isolated by hand with fine forceps
186 under a dissecting microscope, placed in pre-weighed tin capsules, and dried at 50°C to constant

187 weight. Subfossil remains included headshields and carapaces of *Bosmina* (average 250 remains
188 sample⁻¹, 175 µg dry weight) and *Alona* (average 115 remains, 110 µg dry weight), ephippia
189 (diapausing eggs) of *Daphnia* (average 88 remains, 105 µg dry weight) and head capsules of
190 chironomids (average 7 remains, 32 µg dry weight). Sample sizes were almost always sufficient
191 to provide accurate $\delta^{13}\text{C}$ measurements, yet in some cases were inadequate to obtain reliable
192 $\delta^{15}\text{N}$ measurements (mostly *Daphnia* ephippia).

193 The protocol for stable isotope analysis of invertebrate subfossils was similar to that of
194 SOM, with additional modification of the combustion system to accommodate the small sample
195 mass (Carman and Fry, 2002; Maddi et al., 2006). Briefly, the high temperature-reactor system
196 was minimized by combining combustion and reduction columns into one column with a smaller
197 inner diameter of 8 mm. The single column was filled halfway with elemental copper with a
198 lower reactor temperature of 800°C to prevent melting of the elemental copper. Flow rate was
199 set to 50 mL min⁻¹. Due to the relatively quick oxidation of the elemental copper and narrow
200 zone of optimum temperature, reactors had to be changed about every 100 samples.

201 2.6. Data analysis

202 Generalized additive models (GAMs) were used to test significant differences of isotopic
203 time series both i) between SOM and invertebrate taxa and ii) among invertebrate taxa, by
204 modifying the approach of Rose et al. (2012). GAMs are a form of semi-parametric regression,
205 which can estimate non-linear relationships between predictors and response in terms of smooth
206 functions of predictors (Wood, 2017). Location scale GAMs where the response is conditionally
207 distributed Gaussian were selected given the considerable differences in variance of $\delta^{13}\text{C}$ and
208 $\delta^{15}\text{N}$ values among sample types (SOM, invertebrates) (Fig.2). Location scale GAMs allow both

209 mean (location) and variance (scale) of the response distribution to be modelled as a function of
210 predictors. For each lake, we fitted an ordered-factor-smooth interaction model of the form:

$$211 \quad y_{ij} \sim N(\mu_{ij}, \sigma_{ij}^2),$$

$$212 \quad \mu_{ij} = \alpha_0 + \alpha_{1j}(\text{sample type}_j) + f_1(\text{time}_i) + f_{2j}(\text{time}_i),$$

$$213 \quad \log(\sigma_{ij}-b) = \gamma_0 + \gamma_{1j}(\text{sample type}_j) + \gamma_2(\text{time interval}_i),$$

214 where sample type is an ordered factor variable representing SOM and invertebrate taxa, while
215 SOM is the reference level. The response y_{ij} (i.e. i th $\delta^{13}\text{C}$ or $\delta^{15}\text{N}$ observation in the j th sample
216 type) was Gaussian-distributed with a mean μ_{ij} and variance σ_{ij}^2 . Here μ_{ij} was modelled as the
217 mean value of the response in the reference sample type (SOM) (model intercept α_0), plus the
218 difference between mean response of the j th sample type and α_0 (α_{1j}), plus the centered smooth
219 function of time (sediment age in years AD) for the reference sample type SOM (f_1), plus the
220 difference between smooth function of time for the j th sample type and f_1 (f_{2j}). In other words,
221 the model tests i) if the trend (change over time) of SOM $\delta^{13}\text{C}/\delta^{15}\text{N}$ was statistically significant
222 and ii) if both means and trends of $\delta^{13}\text{C}/\delta^{15}\text{N}$ of individual invertebrate taxa were significantly
223 different from those of SOM within a given lake. Restricted maximum likelihood (REML)
224 estimation method and thin-plate regression splines were used to parameterize f_1 and f_{2j}
225 (Simpson, 2018; Wood, 2017). We also examined diagnostic plots to check any residual
226 autocorrelation.

227 Given the large differences in variances among sample types within lakes (Fig.2), it was
228 necessary to model σ_{ij} as a part of the GAM. Hence, σ_{ij} -b (log scale) was fitted as a linear
229 function of the sample type γ_{1j} and the time interval represented by each sediment section (γ_2),

230 plus a constant term (model intercept γ_0), while b is a small lower bound on σ_{ij} to avoid any
231 singularities in the model likelihood.

232 Subsequently, we processed the GAM output (following Rose et al., 2012) to both
233 generate smooth SI trends for individual invertebrate taxa and estimate differences in SI trends
234 between pairs of taxa, as well as to generate approximate 95% point-wise confidence intervals
235 (Supplementary methods; Supplementary data). We inferred a significant difference in isotopic
236 trends between a pair of taxa when the confidence interval excluded zero for considerable time
237 duration.

238 All statistical analysis were performed in R version 3.4.3 (R Core Team, 2017) using the
239 packages ‘mgcv’(Wood, 2017; Wood et al., 2016) and ‘ggplot2’(Wickham, 2009).

240

241 3. Results

242 3.1. $\delta^{13}\text{C}$ of SOM and invertebrate taxa

243 Across all study lakes, $\delta^{13}\text{C}$ values of SOM ranged from -29.3 to -22.8‰ (Fig.2a), with
244 lakes 17F and 17V exhibiting more enriched $\delta^{13}\text{C}$ values than the other lakes (mean difference of
245 ~3-4‰). For a given taxon, the $\delta^{13}\text{C}$ of invertebrate remains varied less among lakes (-28 and -
246 24‰) than did SI values of SOM (range), except *Daphnia* which showed relatively high
247 variation in core-wide mean $\delta^{13}\text{C}$ values (~7‰). Within lakes, ranges of $\delta^{13}\text{C}$ often differed
248 among taxa, although there was no consistent pattern among sites as to which taxon was most
249 variable.

250 GAMs, by modeling both mean and variance components, explained a large proportion of
251 the historical changes in $\delta^{13}\text{C}$ in each lake (explained deviance ranging 91-98%). Sample type

252 (i.e., SOM and invertebrate taxa) was a significant predictor ($p < 0.05$) of mean $\delta^{13}\text{C}$ i.e. mean
253 $\delta^{13}\text{C}$ of all invertebrate taxa within a lake was significantly different ($p < 0.05$) from that of SOM
254 in each lake (Fig. 2a). For variance of $\delta^{13}\text{C}$, sample type (i.e., SOM and invertebrate taxa) had a
255 significant effect ($p < 0.05$) for all lakes, while a significant influence ($p < 0.05$) of the time
256 interval (represented by each sediment section) was only detected for lakes 17V and 13N.

257 The $\delta^{13}\text{C}$ of SOM in all lakes exhibited statistically significant ($p < 0.05$) declines during
258 the 20th century (Table 2, Fig. 3), with more prominent trends in lakes 6E and 17F relative to
259 other basins. Trajectories of invertebrate $\delta^{13}\text{C}$ generally followed the declining trend of SOM
260 $\delta^{13}\text{C}$ in each lake (Fig.3), although invertebrate time series exhibited noticeable differences in
261 timing, pattern and degree of change both within and among lakes. In lakes 17V, 17F and 6E,
262 temporal trends of invertebrate taxa varied ($p < 0.05$) from that of SOM, with the exception of
263 *Chironomid* in lake 17V (Table 2 and Fig. 4). On the other hand, no significant differences in
264 trends ($p < 0.05$) between invertebrate taxa and SOM were detected for 13N and 17P.

265 There were few consistent differences in C source for invertebrates both within and
266 among lakes (6E, 17V) with sufficient fossil density of common taxa (Fig. 5). For example, the
267 $\delta^{13}\text{C}$ trajectories of *Daphnia* and *Bosmina* differed significantly ($p < 0.05$) from each other and
268 from other taxa for much of the past 100 years in lake 6E. Similarly, $\delta^{13}\text{C}$ trends in lake 17V
269 were significantly different ($p < 0.05$) between *Bosmina* and *Daphnia*, *Bosmina* and *Chironomid*,
270 *Daphnia* and *Alona*, and *Alona* and *Chironomid*. However, the nature and degree of trend
271 differences for the same pairs of taxa were not similar among lakes.

272 3.2. $\delta^{15}\text{N}$ of SOM and invertebrate taxa

273 The $\delta^{15}\text{N}$ of SOM varied from -1.9 to 4.1‰ across all years in individual study lakes,

274 with lakes 13N and 17P exhibiting more enriched $\delta^{15}\text{N}$ (mean difference ~2.0-3.5‰) with less
275 variability relative to other sites (Fig.2b). Overall, $\delta^{15}\text{N}$ values of invertebrate remains showed a
276 higher variability than that of SOM (-1.9 to 4.1‰), which ranged from -1.4 to 9.7‰ across all
277 study lakes with considerable among-lake differences.

278 Similar to patterns seen with $\delta^{13}\text{C}$, GAM analysis explained most variation in
279 sedimentary $\delta^{15}\text{N}$ for all parameters in each lake (96-99% deviance explained). Within each site,
280 mean $\delta^{15}\text{N}$ values of individual invertebrates were significantly enriched ($p < 0.05$) relative to
281 SOM for most taxa, except *Daphnia* in 17P (Fig .2b). Variance of $\delta^{15}\text{N}$ was significantly ($p <$
282 0.05) influenced by sample type (SOM and invertebrate taxa), but not by the time interval
283 represented by each sediment section ($p > 0.05$).

284 All lakes showed significant ($p < 0.05$) historical declines in $\delta^{15}\text{N}$ of SOM and
285 invertebrate taxa (Table 3 and Fig. 6). Specifically, $\delta^{15}\text{N}$ values of SOM and invertebrates
286 declined after ca.1940-1950 in lakes 17V, 17P, 17V and 6E, whereas a continuous, linear decline
287 was detected in 13N over the entire 120 year record. While significant differences ($p < 0.05$) in
288 $\delta^{15}\text{N}$ trends between *Bosmina*, *Alona* and *Chironomid* and SOM in 6E and between *Alona* and
289 SOM in lakes 17V and 17 F were detected (Table 3 and Fig. 7), there was no similarity in the
290 nature or degree of difference in invertebrate-SOM trends among lakes. Similarly, time series of
291 $\delta^{15}\text{N}$ differed significantly ($p < 0.05$) among most pairs of invertebrates in lakes 6E and 17V
292 (Fig.8), with few common difference patterns among pairs of taxa.

293 **4. Discussion**

294 The novel combination of taxon-specific isotope time series and GAMs demonstrated
295 that significant differences in mean $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values exist between SOM and invertebrate

296 taxa in all lakes, yet significant differences in long-term trends were only detected in certain
297 basins. Significant differences in mean $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ between SOM and invertebrate taxa may
298 be reflective of selective feeding by invertebrates (Jones et al., 2008, 1999) and differences in
299 trophic position within food webs (Vander Zanden and Rasmussen, 2001). In contrast,
300 differences in historical trends among sample types (i.e. SOM and invertebrate taxa) within lakes
301 were more pronounced for $\delta^{15}\text{N}$ than $\delta^{13}\text{C}$, suggesting that either the biogeochemistry of N
302 (Bunting et al., 2010) or trophic relationships among taxa (Karlsson et al., 2004) was more
303 variable than the sources or cycling of C in these boreal ecosystems. Overall, few differences in
304 SI trends between SOM and invertebrates and among different taxa were noted in lakes with
305 more elevated DOC content, whereas such differences in SI time series were more pronounced in
306 relatively clear-water lakes, possibly reflecting variation in the importance of allochthonous C to
307 invertebrate consumers (Solomon et al., 2008; Tanentzap et al., 2017), as well as a higher degree
308 of habitat diversity or feeding selectivity in transparent ecosystems (Matthews and Mazumder,
309 2006; Persaud et al., 2009). Together, these findings suggest that the analysis of taxon-specific
310 fossil invertebrate time series provides unique insights into the trophic relationships in lentic
311 ecosystems that cannot be achieved through analysis of bulk sediments alone (See also Wyn et
312 al. 2007; Perga et al. 2010; van Hardenbroek et al. 2014).

313 *4.1. SI variability of SOM and invertebrate remains*

314 Overall, the range of SOM $\delta^{13}\text{C}$ values detected across study lakes (-29.3 to -22.8‰)
315 (Fig.2) is indicative of mixed contributions of C from C3 terrestrial plants (-28 to -27‰) and
316 lacustrine primary producers (-35 to -8‰) (Meyers and Teranes, 2001; Vuorio et al., 2006), with
317 among-lake differences likely related to factors that control origin and fate of accumulated C
318 (Supplementary discussion; Supplementary data). Meanwhile, low SOM $\delta^{15}\text{N}$ values in several

319 study lakes (< 2‰) (Fig.2) indicate that fixed N is the main N source (Meyers and Teranes,
320 2001; Patoine et al., 2006), which is likely derived from terrestrial sources (e.g. plants such as
321 *Alnus* spp. and *Shepherdia canadensis* and microorganisms in deciduous forest) (Bunting et al.,
322 2010; Engstrom and Fritz, 2006), as pigments for N₂-fixing cyanobacteria were not detected in
323 sediments of these lakes (Wolfe, 2016). Among-lake differences in SOM δ¹⁵N on the other hand
324 can arise due to differences in the mass flux or isotopic ratios of fixed DOM-N as well as
325 subsequent modification by biological (terrestrial and in-lake) processes (Supplementary
326 discussion; Supplementary data).

327 The δ¹³C and δ¹⁵N values of *Bosmina* exoskeletons (28.1‰ to -23‰ and 0.9 to 9.7‰,
328 respectively) and *Daphnia ephippia* (-30.6‰ to -23.5‰ and -1.4 to 9‰ respectively) recorded in
329 the present study were similar to contemporary values recorded for these taxa in a synoptic
330 survey of 233 regional boreal lakes (*Bosmina* -31.4 to -22.9‰ and -1.5 to 9‰, respectively;
331 *Daphnia* -35.8 to -24.3‰ and 0.5 to 7.8‰, respectively) (Anas, 2019). This supports the high
332 likelihood that downcore isotopic variations in invertebrate sub-fossils are within the expected
333 range in response to regional environmental variations such as fluctuations in primary
334 productivity, allochthonous organic matter inputs, in-lake biogeochemical transformations and
335 catchment processes (Anas, 2019). Elsewhere highly negative δ¹³C values (~-50 to -40‰) of
336 subfossil chironomid head capsules and *Daphnia ephippia* may reflect contributions of ¹³C-
337 depleted methanogenic C to invertebrate diets (Frossard et al., 2013b; Rinta et al., 2016; Schilder
338 et al., 2017; van Hardenbroek et al., 2013; Wooller et al., 2012), a factor which does not seem to
339 be important in our study (Fig. 2). Instead, invertebrate δ¹³C values herein were usually within
340 the range expected for photoautotrophically-produced C in aquatic and terrestrial ecosystems (-
341 35 to -8‰) (France, 1996, 1995b; Meyers and Teranes, 2001; Vuorio et al., 2006).

342 4.2. *SI trends of SOM*

343 Patterns of sedimentary $\delta^{13}\text{C}$ decline over the 20th century (Fig.3) are similar to those
344 observed of other regional studies of boreal western Canada (Ahad et al., 2011; Curtis et al.,
345 2010). In principle, such depletion of ^{13}C content may arise because of increased abundance and
346 assimilation of ^{13}C -depleted respired DIC (originated from heterotrophic respiration of terrestrial
347 organic matter) by primary producers over the past century (France et al., 1997; Meyers and
348 Teranes, 2001). Alternately, such relatively small declines (1-2‰) may just reflect changes in
349 the isotopic composition of atmospheric CO_2 as a result of accelerated release of ^{13}C -depleted
350 CO_2 from fossil fuels, known as Suess Effect (Schelske and Hodell, 1995).

351 As historical data are not available for much the study region (Wolfe 2016), it is difficult
352 to unambiguously identify the precise mechanism(s) underlying progressive declines in $\delta^{15}\text{N}$ of
353 SOM. However, given that declines occur in all lakes, irrespective of the mode of nutrient
354 limitation (N vs. P) and relationship with respect to aerial deposition of nitrogenous pollutants
355 from AOSR (Mushet et al. 2017; Laird et al. 2017; Also see Supplementary discussion;
356 Supplementary data), we infer that climate-related processes, such as regional brownification
357 (Anas et al., 2015) leading to increased influx of ^{15}N -depleted terrestrial DOM-N (Bunting et al.,
358 2010) may underlie the common trend among lakes. This hypothesis is supported by the
359 observations that terrestrially-derived DOM-N is the major N source to these lakes (see above)
360 and that the abundance of mixotrophic algae has increased in these lakes during the past century
361 (Wolfe 2016; Stevenson et al. 2016).

362

363

364 4.3. Differences in $\delta^{13}\text{C}$ time series between invertebrate remains and SOM

365 Significant within-lake differences in mean $\delta^{13}\text{C}$ of SOM and chitinous remains (Fig.2)
366 are in line with findings of other paleolimnological studies (Kattel et al., 2015; Simon et al.,
367 2017; van Hardenbroek et al., 2014, 2013). In principle, such deviations can arise from
368 preferential ingestion or assimilation of specific fractions of particulate organic matter (POM) or
369 surface sediment by invertebrates (Jones et al., 2008, 1999). Bulk POM is a mixture of several
370 constituents (i.e. algae, detritus, bacteria and small planktonic organisms) each with a different
371 $\delta^{13}\text{C}$ value (del Giorgio and France, 1996; Meili, 1992) and relative contribution to the POM
372 pool. POM is eventually deposited as sediment and, as a result, $\delta^{13}\text{C}$ of surface sediment is
373 generally similar to that of POM in overlying waters (Jones et al., 2008; Meili et al., 1996;
374 Meyers and Teranes, 2001). Contemporary studies which detect differences between $\delta^{13}\text{C}$ of
375 lacustrine POM or surface sediment and invertebrate consumers (cladocerans and chironomids)
376 often attribute these patterns to the effects of preferential grazing and/or assimilation of
377 isotopically-distinct fraction within the diet (del Giorgio and France, 1996; Doi et al., 2006;
378 Jones et al., 2008, 1999; Meili et al., 1996). This isotopic deviation is likely conserved between
379 invertebrate remains and POM detritus even after burial in lake sediments (Schilder et al.,
380 2015a), assuming that diagenetic alterations of $\delta^{13}\text{C}$ is negligible or the same in both materials.
381 Meanwhile, empirical and circumstantial evidence indicates that invertebrate taxa examined here
382 can exhibit feeding selectivity due to physical, sensory and nutritional properties of food items
383 (Supplementary discussion; supplementary data).

384 Significant differences between $\delta^{13}\text{C}$ trends of SOM and invertebrates from different
385 habitats within lakes 17V, 17F and 6E suggest that analysis of taxon-specific SI can provide new
386 insights on the regulation of food-web C dynamics of different lake habitats. For instance,

387 variation in phytoplankton $\delta^{13}\text{C}$ due to changes in primary productivity, DIC content or DIC
388 source (France et al., 1997; Meyers and Teranes, 2001) should be reflected in isotopic values of
389 invertebrate remains if phytoplankton-derived organic matter is grazed or assimilated
390 preferentially by invertebrates. Such fine-scale trophic patterns may be partially or completely
391 masked in analysis of SI in SOM in instances where phytoplankton-derived organic matter is
392 mixed with other sources of POM or DOM (Davidson and Jeppesen, 2013; Meyers and Teranes,
393 2001). For the same reason, SOM $\delta^{13}\text{C}$ may fail to reflect environmentally-induced variations in
394 epiphytic $\delta^{13}\text{C}$ which are faithfully recorded in exoskeletons of littoral herbivore *Alona*, or the
395 incorporation of C from methanogens by benthic chironomids (Schilder et al., 2017; Simon et al.,
396 2017; van Hardenbroek et al., 2014). Finally, historical trends in invertebrate $\delta^{13}\text{C}$ may reflect
397 shifts in feeding selectivity of source population due to temporal variations in relative
398 availability of autochthonous vs. allochthonous resources (Demott, 1982; Tanentzap et al., 2017).
399 Under such conditions, the $\delta^{13}\text{C}$ values of invertebrate remains may be expected to diverge from
400 those of bulk SOM during periods of high feeding selectivity (and converge as feeding
401 selectivity decreases).

402 Trends in $\delta^{13}\text{C}$ of invertebrate remains did not vary significantly from SOM in lakes 17P
403 and 13N (Fig.4), suggesting that existence of SOM-invertebrate trend differences can be system-
404 specific. This may be a result of strong influence of allochthonous inputs on both SOM and
405 invertebrate diets into these dystrophic lakes. In particular, these sites exhibited not only much
406 higher DOC concentrations and water color (Table 1), but also elevated SOM C:N (molar) ratios
407 (ca. 13-15 and 15-17, respectively) compared to other study lakes (Fig. S1; Supplementary data),
408 indicating substantial terrestrial organic matter inputs over the past century (Meyers and Teranes,
409 2001). Although speculative, we suggest that invertebrate consumers in these lakes were relying

410 more on allochthonous C sources (terrestrial detritus and/or heterotrophic bacteria), given they
411 were more abundant than preferred autochthonous foods (Cole et al., 2011; Simon et al., 2017;
412 Solomon et al., 2008; Tanentzap et al., 2017). Allochthonous organic matter may also have
413 largely contributed to SOM in these dystrophic lakes (Dalton et al., 2018; Meyers and Teranes,
414 2001). Such large contribution of allochthonous sources to both invertebrate diets and SOM
415 may have led to temporal coherence between $\delta^{13}\text{C}$ trends of invertebrate remains and SOM.

416 4.4. Differences in $\delta^{13}\text{C}$ trends among invertebrate taxa

417 The significant among-taxon differences in $\delta^{13}\text{C}$ trends of lakes 17V and 6E indicate that
418 flux of C through invertebrates can differ among taxa, habitats and lakes, and that all pathways
419 may have decoupled through time (Fig.5). Dietary sources of C for invertebrates, as well as the
420 associated $\delta^{13}\text{C}$ values of C source can be impacted by habitat-specific or whole-lake
421 environmental changes (Persaud et al., 2009; Vadeboncoeur et al., 2003; van Hardenbroek et al.,
422 2014). In particular, changes in relative availability of different C sources (i.e. phytoplankton,
423 epiphyton, terrestrial, methane-derived C) can be induced by alteration in water transparency by
424 primary producers (Solomon et al., 2008; Vadeboncoeur et al., 2003), physical turbidity
425 (Scheffer et al., 1993) or DOC influx (Carpenter et al., 1988), leading to shifts in the diets of
426 primary consumers (Solomon et al., 2008; Vadeboncoeur et al., 2003). For instance, a switch
427 from predominantly benthic production to a pelagic regime following eutrophication can shift the
428 diet of littoral consumers (e.g. *Alona*) from ^{13}C -enriched epiphyton ((Hecky and Hesslein, 1995)
429 to ^{13}C -depleted phytoplankton and suspended detritus, while diet of pelagic consumers (e.g.,
430 *Bosmina*, *Daphnia*) remain unaffected (Vadeboncoeur et al., 2003). Further, $\delta^{13}\text{C}$ of littoral
431 epiphyton may respond differently compared to pelagic phytoplankton to temporal changes in
432 DIC content and dominant chemical form of inorganic C (CO_2 , HCO_3^- , CO_3^{2-}) due to boundary

433 layer effects (Hecky and Hesslein, 1995; McPherson et al., 2015; Woodland et al., 2012), thereby
434 altering historical trends in $\delta^{13}\text{C}$ of littoral consumers relative to pelagic taxa. Differences in
435 degree of feeding selectivity may have contributed to differences in $\delta^{13}\text{C}$ through time between
436 pelagic taxa *Bosmina* and *Daphnia*. For example, $\delta^{13}\text{C}$ trajectory of less-selective *Daphnia* may
437 be more affected by temporal variations in relative availability of autochthonous vs.
438 allochthonous sources compared to more selective *Bosmina* (Demott, 1982; DeMott and Kerfoot,
439 1982; Tanentzap et al., 2017). Meanwhile, lack of among-lake similarity in trend difference
440 patterns for pairs of common taxa suggests that individual lakes may have been variably
441 influenced by regional environmental changes (e.g. climatic forcing) owing to differences in
442 local characteristics, leading to differential temporal changes in C fluxes through different
443 habitats or food web compartments in individual lakes.

444 Time series of $\delta^{13}\text{C}$ did not differ significantly among invertebrates through time in three
445 lakes (13N, 17P, 17F; Fig.5), suggesting the path or process of C assimilation was relatively
446 consistent in food webs of these lakes. Such consistency in the two dystrophic lakes (13N, 17P)
447 may reflect the paramount importance of terrestrial DOM as a food-web C source in all habitats
448 and feeding niches (Solomon et al., 2011), either because of the importance of ^{13}C -depleted
449 respired DIC to primary producers in all habitats, or, the consistent availability of terrestrial
450 detritus and/or heterotrophic bacteria as a C source to primary consumers (see above). In lake
451 17F, both pelagic and benthic habitats were likely autotrophic due to light penetration to the
452 bottom, as indicated by dominance of benthic over planktonic diatoms over the past century
453 (Laird et al., 2017). The unchanged diatom composition in this lake (Laird et al., 2017) suggests
454 that is limnological conditions have remained largely unaltered through time, with few changes

455 in water transparency and the ratio of benthic to pelagic autotrophy. Therefore, the dietary
456 sources of C to invertebrates may have remained consistent within 17F's habitats through time.

457 *4.5. Differences in $\delta^{15}\text{N}$ time series between invertebrate remains and SOM*

458 The significantly higher mean $\delta^{15}\text{N}$ of invertebrate remains relative to SOM in our study
459 lakes (Fig.7) is characteristic of a 2-4‰ trophic fractionation between diet and consumer
460 (Vander Zanden and Rasmussen, 2001), assuming that SOM $\delta^{15}\text{N}$ represents available dietary
461 sources (Perga et al., 2010). The $\delta^{15}\text{N}$ enrichment relative to SOM varied among invertebrate
462 taxa within each lake (Figs.2, 7), likely as a result of differences in trophic position of individual
463 taxa within the local aquatic food web, as well as variability in metabolic pathways of protein
464 synthesis (Kling et al., 1992; Vander Zanden and Rasmussen, 2001). Meanwhile, among-lake
465 differences in invertebrate $\delta^{15}\text{N}$ enrichment relative to SOM may be associated with lake-specific
466 factors that can influence the magnitude of ^{15}N fractionation by consumers. For instance,
467 reduced availability of N in some food resources (i.e. high C: N ratios) may lead to elevated diet-
468 tissue ^{15}N fractionation in consumers (Adams and Sterner, 2000). Enriched $\delta^{15}\text{N}$ values of
469 consumers can also arise under conditions of low-food quantity or quality as a result of increased
470 turnover of internal nitrogen and preferential excretion of ^{14}N (Adams and Sterner, 2000;
471 Karlsson et al., 2004)

472 Significant differences between $\delta^{15}\text{N}$ trends of SOM and invertebrate subfossils in lakes
473 17V, 17F and 6E (Fig. 7) may have arisen from shifts in dietary sources of N and/or changes in
474 magnitude of isotopic fractionation in invertebrate consumers. For example, $\delta^{15}\text{N}$ of invertebrate
475 consumers can covary with bacterial contribution to the diet (Grey et al., 2004, 2001; Karlsson et
476 al., 2004). Empirical evidence suggests that bacterial $\delta^{15}\text{N}$ may differ from other basal resources

477 either due to elevated isotopic fractionation depending on biochemical composition and
478 availability of nitrogen substrate (McGoldrick et al., 2008; Steffan et al., 2015) or assimilation of
479 isotopically light nitrogen substrate (e.g. excreted ammonia) (Grey et al., 2004). In addition, few
480 SI studies (e.g. France, 1995a; Mulholland et al., 2000) suggest that, although not definitive,
481 $\delta^{15}\text{N}$ variation of invertebrate consumers can be reflective of shifts in relative dietary importance
482 of autochthonous vs. allochthonous sources. Alternately, changes in $\delta^{15}\text{N}$ offset between SOM
483 and invertebrates can reflect variation in trophic position of taxa over decades, assuming that
484 SOM represents an isotopic baseline (Griffiths et al., 2010; Perga et al., 2010). In this case,
485 climatically-induced changes in DOC flux could favour shifts from an algae-dominated to a
486 bacterivorous protozoa-dominated diet (i.e. microflagellates and ciliates) (DeMott and Kerfoot,
487 1982; Ngochera and Bootsma, 2011), which is in turn reflected by temporal changes in $\delta^{15}\text{N}$ of
488 invertebrate consumers. Finally, shifts N-rich algae to N-poor allochthonous food sources may
489 lead to elevated $\delta^{15}\text{N}$ in aquatic invertebrates (Maguire and Grey, 2006) as a result of increased
490 diet-tissue ^{15}N fractionation (Adams and Sterner, 2000; Karlsson et al., 2004). **Similar to findings**
491 **for $\delta^{13}\text{C}$ trends, $\delta^{15}\text{N}$ trends of invertebrate subfossils did not differ significantly from SOM in**
492 **two dystrophic lakes (13N, 17P) likely due to paramount and consistent importance of**
493 **allochthonous sources to both SOM (Dalton et al., 2018; Meyers and Teranes, 2001) and**
494 **invertebrate diets (Cole et al., 2011; Simon et al., 2017; Solomon et al., 2008; Tanentzap et al.,**
495 **2017), again indicating existence of SOM-invertebrate trend differences can be system-specific.**

496 *4.6. Differences in $\delta^{15}\text{N}$ trends among invertebrate taxa*

497 The presence of significant among-taxon differences in $\delta^{15}\text{N}$ trends in lakes 17V and 6E
498 suggests that trophic relationships among taxa may vary in lakes at centennial scales (Fig. 8). As
499 noted above, variable offsets in $\delta^{15}\text{N}$ among pairs of species may result from habitat-specific

500 changes in resource materials with different $\delta^{15}\text{N}$ values, resource limitation, or changes in
501 metabolic processes of individual taxa. In addition, lake-specific changes in the influx of
502 nitrogenous materials may also influence the degree of agreement in $\delta^{15}\text{N}$ trends between pairs
503 of species, particularly from different habitats. In particular, the $\delta^{15}\text{N}$ of individual taxa may be
504 selectively influenced by dietary shifts or changes in isotopic fractionation while the $\delta^{15}\text{N}$ of the
505 other taxon remains unaffected (e.g. *Alona* vs. *Bosmina* in 17V). Alternately, $\delta^{15}\text{N}$ variations of
506 both taxa can be influenced by environmental and physiological mechanisms simultaneously,
507 asynchronously or differentially (e.g. *Alona* vs. *Bosmina* in 6E), as has been seen in modern lake
508 time series (Patoine et al. 2006). Regardless of the underlying mechanism, the observation that
509 historical differences between pairs of common taxa (e.g., *Bosmina* v. *Alona*) vary among lakes
510 (17V, 6E) suggests the possibility of lake-specific food web responses to environmental changes
511 during the past century.

512 4.7. Caveats

513 Due to methodological constraints, our SI analyses were conducted at a relatively coarse
514 taxonomic resolution relative to modern limnological studies (e.g., *Daphnia* spp., chironomids,
515 etc.). Consequently, interpretations of taxon-specific isotopic variability may be confounded in
516 part by variation in $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ values among con-specific taxa. For example, although
517 chironomid larvae are generally considered as detritivores, feeding mode may vary among species
518 (e.g. deposit feeding, filter feeding and engulfing), which may have consequences for $\delta^{13}\text{C}$ and
519 $\delta^{15}\text{N}$ variability (Griffiths et al., 2010; Solomon et al., 2008; van Hardenbroek et al., 2014).
520 Similarly, differences in daphniid body size can influence the accessibility to both very small and
521 large food particles (Kasprzak and Lathrop, 1997). Furthermore, *Daphnia* ephippia may
522 integrate time differently than invertebrate exoskeletons, and may provide a different metric of

523 resource use or trophic position compared to remains deposited as a result of adult death or
524 molting. Ehippia are produced within a very short time frame in response to specific
525 environmental cues such as food limitation, reduced photoperiod and crowding (Kleiven et al.,
526 1992). Hence, they may only provide a temporal snapshot of $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ of the source
527 population during their brief period of formation (Griffiths et al., 2010; Perga, 2009). In
528 contrast, invertebrate exoskeletons may provide a relatively long-term representation isotopic
529 composition of the source population.

530 **5. Conclusions**

531 Our study demonstrates that $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ analysis of taxon-specific invertebrate
532 remains can provide unique insights into historical trophodynamics in lake food webs, different
533 from those derived from an analysis of bulk sediments. Statistically-significant differences in
534 mean values of $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ between SOM and invertebrate remains likely reflected selective
535 feeding or trophic fractionation, and appear to be ubiquitous in study lakes despite limnological
536 differences among lakes. Further, the absence of significant differences in historical trends
537 among trophic levels or taxa in relatively dystrophic lakes suggests a stabilizing role of terrestrial
538 C contributions in food-web processes (cf. Wetzel 1989). On the other hand, the higher
539 variability in SOM-invertebrate SI time series in relatively clear lakes may reflect decadal-scale
540 variation in dietary sources of elements or their metabolic processing by invertebrates. That said,
541 it is clear that further refinement of causal mechanisms requires substantial additional work,
542 including additional paleoenvironmental analyses and comparisons with long-term monitoring
543 data. For instance, simultaneous evaluation of $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ of chlorins (i.e. chlorophylls and
544 their phaeopigment derivatives) may help to better understand if and how isotopic trends of
545 invertebrate remains are related to changes in aquatic primary productivity (Hayes et al., 1987).

546 Based on our results, we suggest that evaluation of isotopic trends of invertebrate remains
547 relative to SOM, will be a particularly informative means of unraveling past food-web carbon,
548 nutrient and trophic dynamics.

549

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557

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833 Table 1. Limnological characteristics of five study lakes

Variable	Lake				
	17V	17P	17F	13N	6E
Elevation (m)	533.0	558.0	525.0	471.0	434.0
Surface area (ha)	62.0	63.0	21.0	150.0	18.3
Maximum depth (m)	7.4	7.4	8.8	15.4	9.6
Coring depth (m)	5.4	7.4	7.9	12.2	6.0
Distance from AOSR (km)	287.2	291.4	268.7	177.6	105.7
Molar DIN:TP	2.8	7.8	7.8	13.4	3.3
TN ($\mu\text{g L}^{-1}$)	290.0	253.0	299.0	253.0	314.0
TP ($\mu\text{g L}^{-1}$)	14.0	7.7	6.5	5.0	16.5
Chlorophyll a ($\mu\text{g L}^{-1}$)	5.4	2.0	3.2	3.1	6.1
pH	7.1	6.8	7.0	7.1	7.3
Alkalinity ($\text{mg L}^{-1} \text{CaCO}_3$)	7.5	4.2	7.8	11.6	16.5
DOC (mg L^{-1})	3.4	7.1	4.4	7.1	3.4
Color ($\text{mg L}^{-1} \text{Pt}$)	4.8	32.8	6.8	26.6	8.3

834 Note: The values are averages of lake surveys conducted in 2007- 2009 and 2011 by

835 Saskatchewan Ministry of Environment (Scott et al., 2010). See text for descriptions of variables.

836 Table 2. Model summaries for SOM and invertebrate $\delta^{13}\text{C}$ time series of study lakes

Lake	Covariate	EDF	Ref. DF	χ^2	<i>p</i> -value
17V	Trend _{SOM}	2.41	9.00	36.43	1.16×10^{-6}
	Trend _{Daphnia-SOM}	1.21	5.00	3.74	0.04
	Trend _{Bosmina-SOM}	1.88	5.00	15.85	4.91×10^{-5}
	Trend _{Alona-SOM}	1.90	5.00	18.60	1.32×10^{-5}
	Trend _{Chironomid-SOM}	9.07×10^{-6}	5.00	0.00	0.51
17P	Trend _{SOM}	1.76	2.00	30.54	4.05×10^{-8}
	Trend _{Daphnia-SOM}	6.44×10^{-6}	2.00	0.00	0.66
	Trend _{Bosmina-SOM}	2.60×10^{-5}	2.00	0.00	0.37
	Trend _{Alona-SOM}	0.13	2.00	0.30	0.13
	Trend _{Chironomid-SOM}	4.92×10^{-4}	2.00	0.00	0.17
17F	Trend _{SOM}	3.34	9.00	730.81	$< 2.00 \times 10^{-16}$
	Trend _{Bosmina-SOM}	0.96	4.00	27.42	9.49×10^{-8}
	Trend _{Alona-SOM}	2.21	4.00	87.59	$< 2.00 \times 10^{-16}$

	Trend _{Chironomid-SOM}	0.97	4.00	31.41	1.21×10^{-8}
13N	Trend _{SOM}	5.06	9.00	479.50	$< 2.00 \times 10^{-16}$
	Trend _{Bosmina-SOM}	2.47×10^{-5}	9.00	0.00	0.62
	Trend _{Alona-SOM}	6.57×10^{-6}	9.00	0.00	0.62
	Trend _{Chironomid-SOM}	5.99×10^{-5}	9.00	0.00	0.37
6E	Trend _{SOM}	7.23	9.00	917.35	$< 2.00 \times 10^{-16}$
	Trend _{Daphnia-SOM}	4.04	5.00	23.47	5.14×10^{-5}
	Trend _{Bosmina-SOM}	2.52	5.00	61.91	2.50×10^{-15}
	Trend _{Alona-SOM}	3.27	5.00	31.83	1.75×10^{-7}
	Trend _{Chironomid-SOM}	2.11	5.00	10.95	2.77×10^{-3}

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844 Table 3. Model summaries for SOM and invertebrate $\delta^{15}\text{N}$ time series of study lakes

Lake	Covariate	EDF	Ref. DF	χ^2	<i>p</i> -value
17V	Trend _{SOM}	3.97	9.00	293.02	$< 2.00 \times 10^{-16}$
	Trend _{Bosmina-SOM}	4.61×10^{-5}	5.00	0.00	0.60
	Trend _{Alona-SOM}	1.84	5.00	9.96	2.89×10^{-3}
	Trend _{Chironomid-SOM}	3.76×10^{-5}	5.00	0.00	0.56
17P	Trend _{SOM}	1.84	2.00	24.71	1.47×10^{-6}
	Trend _{Daphnia-SOM}	5.30×10^{-7}	2.00	0.00	1.00
	Trend _{Bosmina-SOM}	8.81×10^{-7}	2.00	0.00	0.68
	Trend _{Alona-SOM}	0.44	2.00	1.41	0.07
	Trend _{Chironomid-SOM}	6.96×10^{-5}	2.00	0.00	0.38
17F	Trend _{SOM}	5.23	9.00	2013.11	$< 2.00 \times 10^{-16}$
	Trend _{Bosmina-SOM}	3.79×10^{-5}	5.00	0.00	0.54
	Trend _{Alona-SOM}	1.52	5.00	4.77	0.04
	Trend _{Chironomid-SOM}	6.95×10^{-5}	5.00	0.00	0.93

13N	Trend _{SOM}	9.58	9.00	23.06	8.96×10^{-7}
	Trend _{Bosmina-SOM}	4.97	5.00	1.13	0.13
	Trend _{Alona-SOM}	5.58×10^{-5}	5.00	0.00	0.33
	Trend _{Chironomid-SOM}	0.08	5.00	0.10	0.26
6E	Trend _{SOM}	2.55	9.00	54.96	6.86×10^{-14}
	Trend _{Daphnia-SOM}	0.00	5.00	0.00	0.32
	Trend _{Bosmina-SOM}	1.76	5.00	11.14	1.00×10^{-3}
	Trend _{Alona-SOM}	3.52	5.00	40.75	3.00×10^{-9}
	Trend _{Chironomid-SOM}	0.84	5.00	5.61	9.87×10^{-3}

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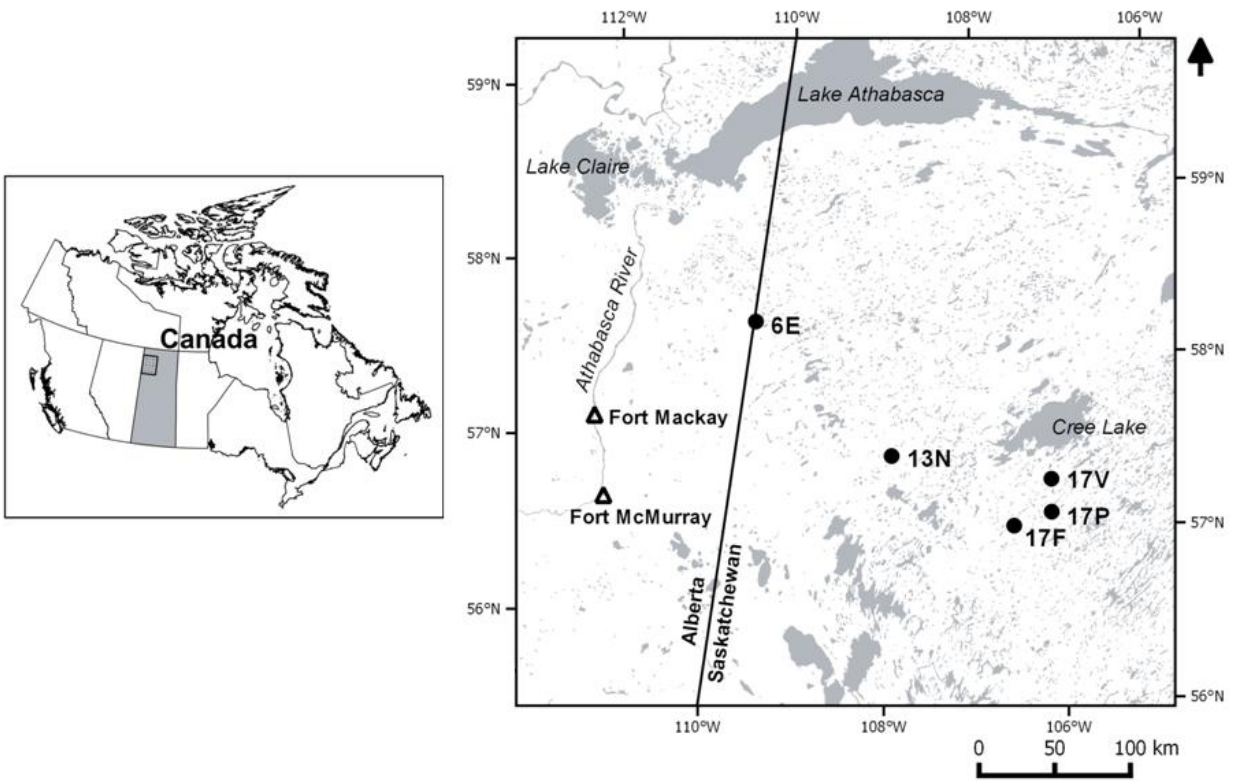
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855 Fig.1. Sampling domain and locations of five study lakes in northwest Saskatchewan.

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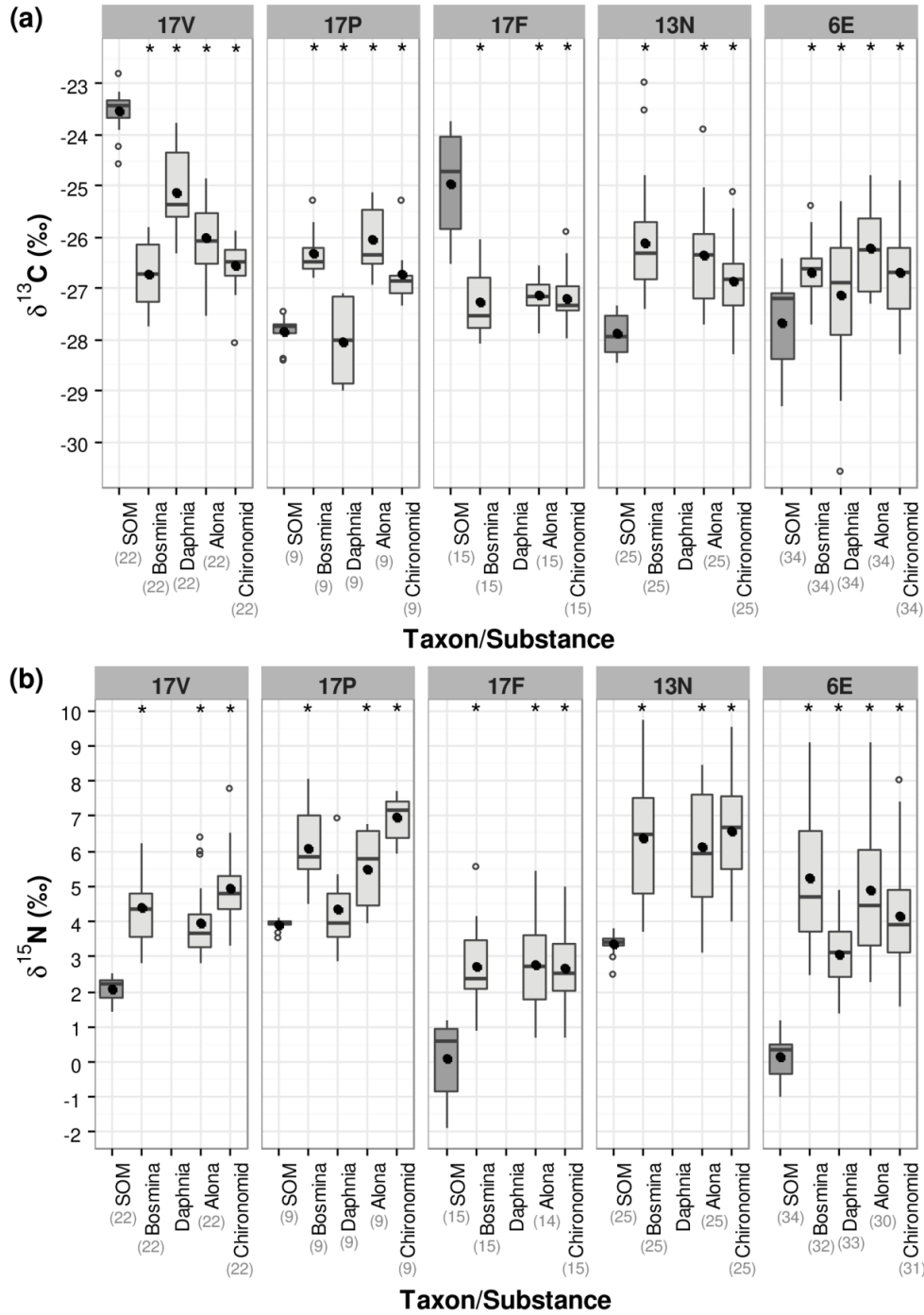
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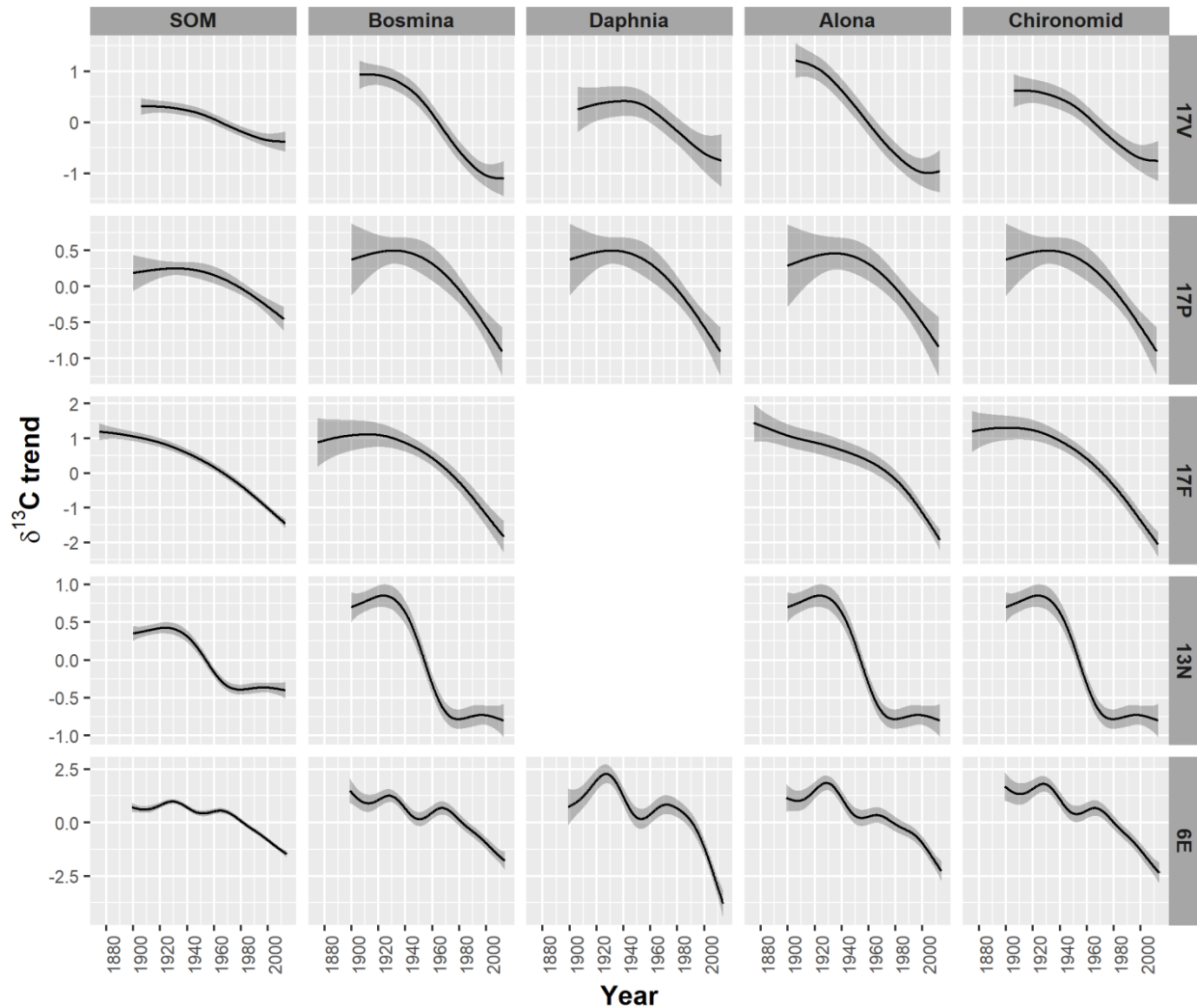
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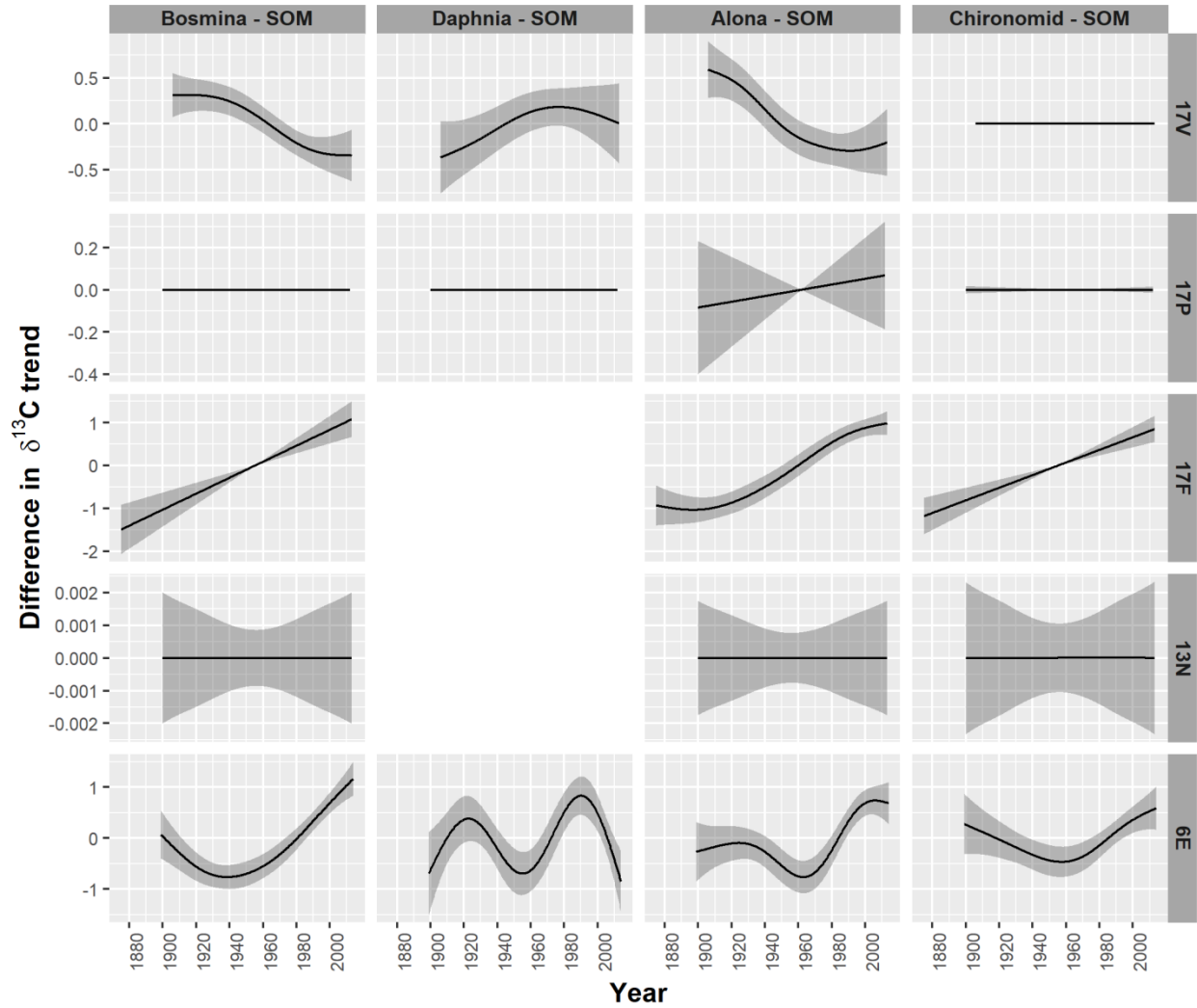
864 Fig.2. Comparison of observed isotopic values among sample types i.e. SOM and invertebrate
 865 taxa within study lakes. (a) Boxplots of $\delta^{13}\text{C}$ values of sample types. (b) Boxplots of $\delta^{15}\text{N}$ values
 866 of sample types. The grey box represents quartiles (25-75%), the horizontal bar in the box is the
 867 median, the whiskers extend to the furthest data point that is within 1.5 times the interquartile

868 range and open circles (○) show the outliers. Solid circles (●) indicate the mean values and
 869 asterisks (*) denote the invertebrate taxa with significantly different ($p < 0.05$) mean values from
 870 that of SOM.



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 872 Fig.3. $\delta^{13}\text{C}$ trends of SOM and invertebrate taxa of five study lakes. Solid lines denote the fitted
 873 trends (smooth functions) and shaded regions represent the point-wise approximate 95%
 874 confidence intervals. The columns represent sample types and the rows represent study lakes.

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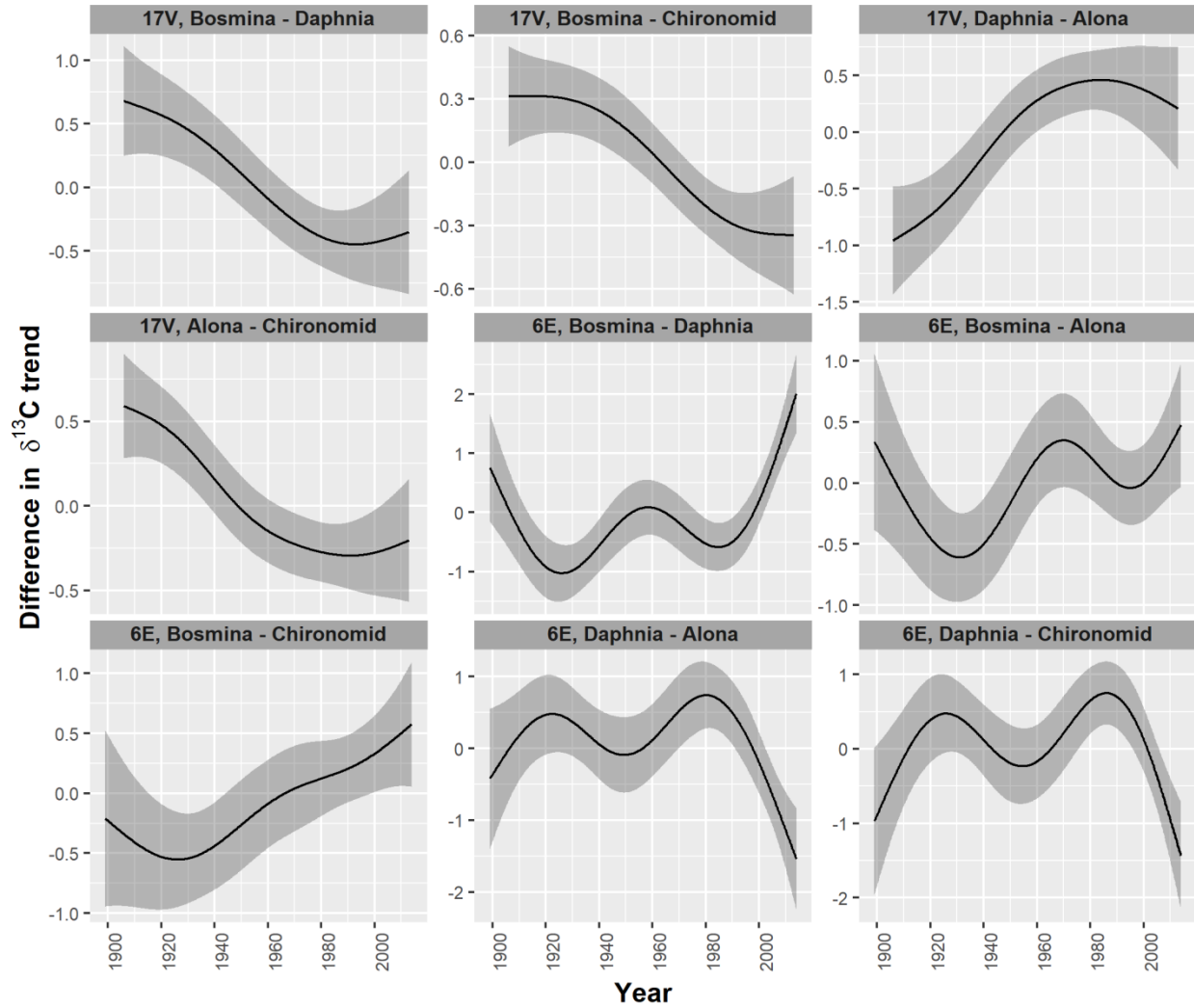
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877 Fig.4. Within-lake differences in $\delta^{13}\text{C}$ trends between invertebrate taxa and SOM. The columns
 878 represent pair-wise comparisons and the rows represent study lakes. Solid lines denote the
 879 estimated differences in trends (difference smooths) and shaded regions represent the point-wise
 880 approximate 95% confidence intervals.

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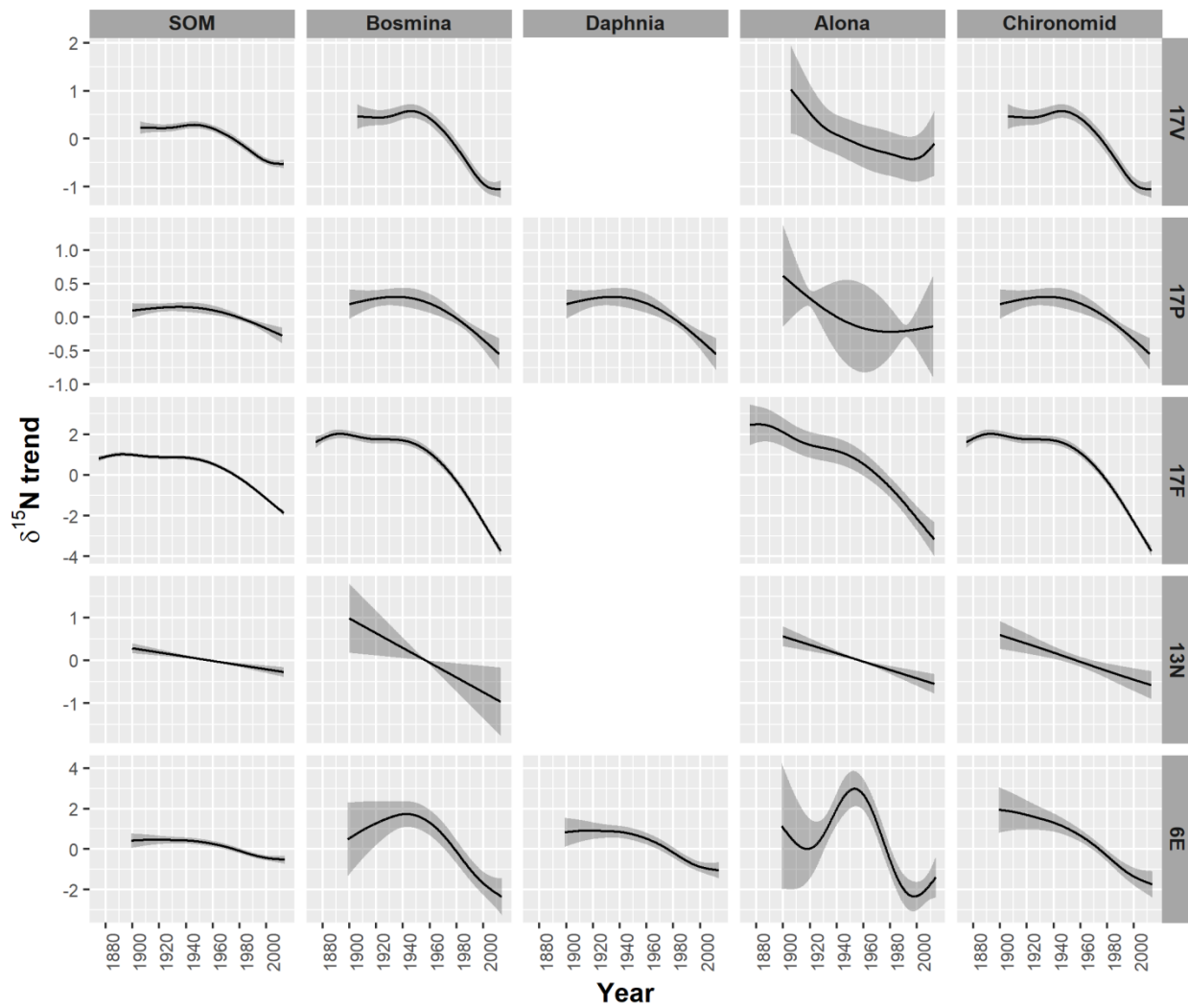
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885 Fig.5. Within-lake differences in $\delta^{13}\text{C}$ trends among invertebrate taxa. The columns represent
 886 pair-wise comparisons and the rows represent study lakes. Solid lines denote the estimated
 887 differences in trends and shaded regions represent the point-wise approximate 95% confidence
 888 intervals.

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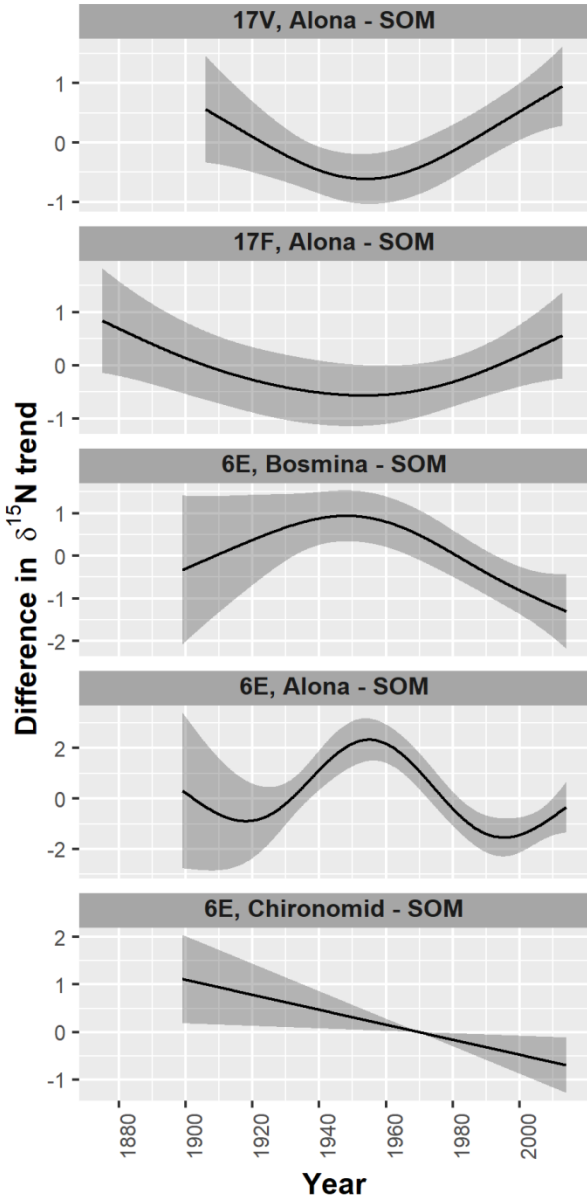
893 Fig.6. $\delta^{15}\text{N}$ trends of SOM and invertebrate taxa of five study lakes. Solid lines denote the fitted
 894 trends (smooth functions) and shaded regions represent the point-wise approximate 95%
 895 confidence intervals. The columns represent sample types and the rows represent study lakes.

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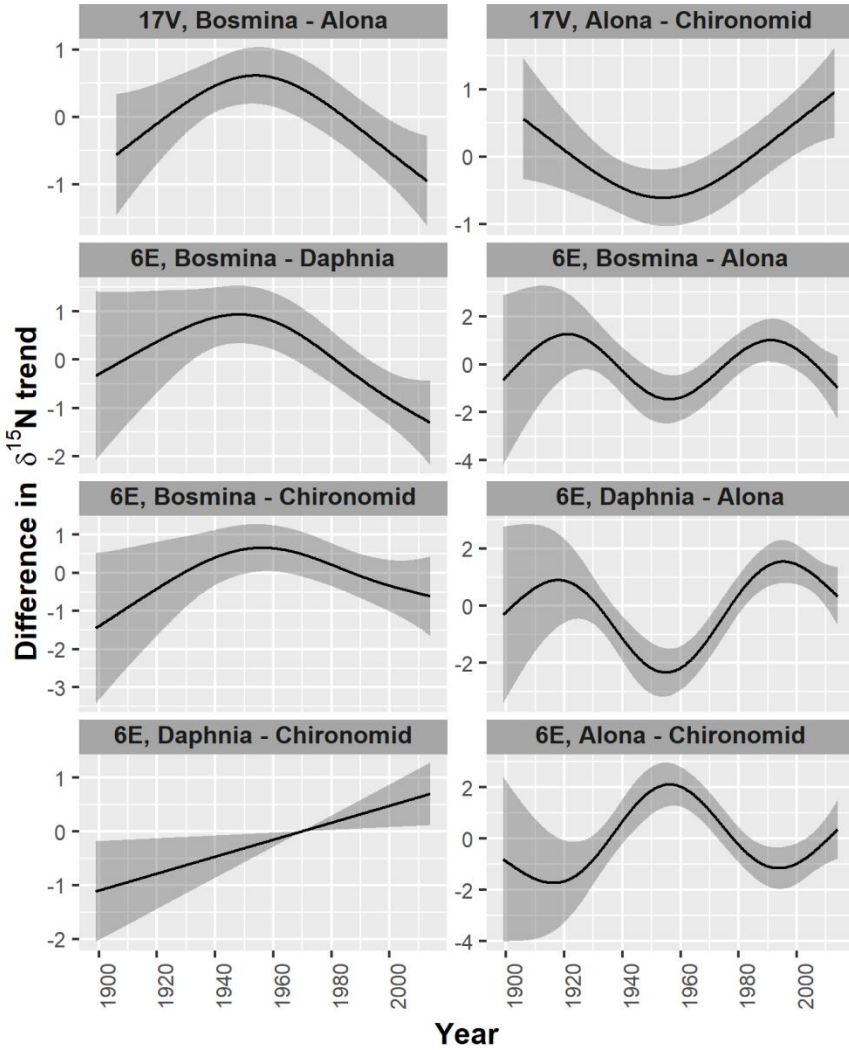
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901 Fig.7. Within-lake differences in $\delta^{15}\text{N}$ trends between invertebrate taxa and SOM. The columns
 902 represent pair-wise comparisons and the rows represent study lakes. Solid lines denote the
 903 estimated differences in trends (difference smooths) and shaded regions represent the point-wise
 904 approximate 95% confidence intervals.

905



906

907 Fig.8. Within-lake differences in $\delta^{15}\text{N}$ trends among invertebrate taxa. The columns represent
 908 pair-wise comparisons and the rows represent study lakes. Solid lines denote the estimated
 909 differences in trends and shaded regions represent the point-wise approximate 95% confidence
 910 intervals.

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Anas, M. U. M., Simpson, G. L., Leavitt, P. R., Cumming, B.F., Laird, K. R., Scott, K. A., Das, B., Wolfe, J. D., Hesjedal, B., Mushet, G. R., Walker, A., Meegahage, B. J. and Wissel, B. 2019. Taxon-specific variation in $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ of subfossil invertebrate remains: Insights into historical trophodynamics in lake food-webs. *Ecol. Indic.*

Supplementary data

Supplementary methods: Statistical testing of among-taxon trend differences

In order to test significant differences of isotopic trends among different invertebrate taxa within each lake, we post-processed the results of the above analysis as described in Rose et al. (2012). This involved using the prediction matrix X_p of the fitted GAM which yields the fitted response values (\hat{y}_p) for a new set of high resolution data points over the time-scale of each core (p) when multiplied by the vector of fitted coefficients i.e. $\alpha_0, \alpha_{1j}, \gamma_0, \gamma_{1j}, \gamma_2$ and penalized coefficient sets of basis functions representing the fitted smooths f_1 and f_{2j} ($\hat{\beta}$) i.e.;

$$\hat{y}_p = X_p \hat{\beta}$$

For a given pair of invertebrate taxa to be compared (a and b), the elements of X_p for first the taxon (a) were subtracted from those corresponding to second taxon (b) and then, the columns of the resultant differenced matrix $X_{p(a,b)}$ that were not involved in the comparison were set to zero. Subsequently, the difference between trends for the pair of taxa $D_{p(a,b)}$ was obtained by;

$$D_{p(a,b)} = X_{p(a,b)} \hat{\beta}$$

To determine the uncertainty of estimated difference ($D_{p(a,b)}$), we computed the standard errors for $D_{p(a,b)}$ by using the variance-covariance matrix of the estimated model coefficients $\hat{V}_{\hat{\beta}}$. The standard errors were provided by the diagonal elements of;

$$X_{p(a,b)} \hat{V}_{\hat{\beta}} X_{p(a,b)}^T$$

where $X_{p(a,b)}^T$ is a matrix transpose of $X_{p(a,b)}$. Using these standard errors, point-wise approximate 95% confidence intervals for $D_{p(a,b)}$ were generated. We inferred a significant difference in isotopic trends between a pair of taxa when the confidence interval excluded zero for considerable time duration.

A similar approach was used to generate $\delta^{13}\text{C}/\delta^{15}\text{N}$ smooth trends for individual invertebrate taxa. The only difference being that instead of subtracting the elements of X_p related to two taxa from one another, the elements of X_p corresponding to the taxon of interest were added to those corresponding to SOM. The estimated isotopic trends for taxa were inferred as statistically significant when the point-wise approximate 95% confidence intervals did not overlap zero for considerable time duration.

Supplementary discussion

Differences in mean $\delta^{13}\text{C}$ of SOM among study lakes

Differences in mean $\delta^{13}\text{C}$ of SOM among study lakes can be related to number of factors that control origin and fate of accumulated carbon. First, they can be associated with among-lake differences in organic matter sources to bulk sediment (e.g. terrestrial plants, phytoplankton, epiphyton and macrophytes) that likely vary in their $\delta^{13}\text{C}$ values (Aichner et al., 2010; Brenner et al., 2006; Davidson and Jeppesen, 2013; Meyers and Teranes, 2001). Second, among-lake variation in primary productivity may have contributed to these differences, given the greater algal discrimination against ^{13}C under more productive conditions (Meyers and Teranes, 2001; Schelske and Hodell, 1995). Third, varying importance of dissolved inorganic carbon (DIC) sources (atmospheric, geogenic and respired) with different $\delta^{13}\text{C}$ values can influence $\delta^{13}\text{C}$ values of autochthonously-derived organic material (France et al., 1997; Meyers and Teranes, 2001; Rau, 1978). Fourth, among-lake differences in methane production in sediments is another possible factor, as strongly ^{13}C -depleted methane can lead to a large reduction in SOM $\delta^{13}\text{C}$ values (Davidson and Jeppesen, 2013; Jones and Grey, 2011). Finally, post-depositional diagenetic effects may alter SOM $\delta^{13}\text{C}$ differently in lakes with varying SOM composition due to differential diagenesis of their SOM constituents (Meyers and Teranes, 2001).

Differences in mean $\delta^{15}\text{N}$ of SOM among study lakes

Among-lake variability in mean $\delta^{15}\text{N}$ of SOM is likely a function of anthropogenic and natural impacts on origin and transformation of nitrogen (inorganic and organic) in lakes. First, among-lake variability in $\delta^{15}\text{N}$ of terrestrially-derived dissolved organic nitrogen (DON; dominant form of nitrogen in our study lakes; Anas et al. 2014) can be related to differences in terrestrial vegetation characteristics. For instance, peatlands may act as denitrification hotspots where ^{14}N is preferentially out-gassed, resulting in higher $\delta^{15}\text{N}$ values of residual DON entering the lakes (Lindau et al., 1997; Wray and Bayley, 2007). In contrast, nitrogen fixation by plants such as *Alnus* spp. and *Shepherdia canadensis* and microorganisms in deciduous forest may result in DON inputs with low $\delta^{15}\text{N}$ into lakes (Bunting et al., 2010; Engstrom and Fritz, 2006; Wolfe, 2016). Second, atmospheric deposition of (likely) ^{15}N -depleted reactive nitrogen from nearby (i.e., Athabasca Oil Sands Region) and long-range industrial sources and subsequent assimilation by lacustrine primary producers may vary among study lakes (Holtgrieve et al., 2011; Wolfe et al., 2006; Wolfe, 2016). However, the amount of deposited reactive nitrogen transferred from the catchment to the lake is determined by soil nitrogen retention and terrestrial uptake, which may again vary among lakes (Anas et al., 2015; Hobbs et al., 2016; Wolfe, 2016). Third, type and degree of nutrient limitation in lakes may contribute to differences in $\delta^{15}\text{N}$ of autochthonous organic matter. In phosphorus-limited lakes with sufficient dissolved inorganic nitrogen (DIN) supply to sustain primary production, preferential uptake of ^{14}N may result in lower $\delta^{15}\text{N}$ values in autochthonous organic matter relative to the nitrogen source. In lakes with limited DIN supply on the other hand, $\delta^{15}\text{N}$ of autochthonous organic matter can be similar to the DIN source due to minimal fractionation against ^{15}N by algae, according to Rayleigh distillation kinetics (Meyers and Teranes, 2001; Talbot, 2001). Finally, isotopic fractionation during other biogeochemical processes occurring in lakes and catchments (e.g. ammonification, nitrification, denitrification) may also influence $\delta^{15}\text{N}$ of the DIN pool used by primary producers (Kendall 1998; Robinson 2001, Anas et al., unpublished).

Feeding selectivity of invertebrate taxa

Empirical and circumstantial evidence indicates that invertebrate taxa examined here can exhibit feeding selectivity due to physical, sensory and nutritional properties of food items. Properties which affect ingestion include size, shape, flavour, surface characteristics (sheathes, cell projections, flagella, etc.) and nutrient content (Bogdan and Gilbert, 1982; Brett et al., 2009; Butler et al., 1989; Cyr and Curtis, 1999; DeMott, 1986; Knisely and Geller, 1986), while differential digestion in the gut can affect assimilation (Porter, 1973). *Bosmina*, a pelagic suspension-feeder, is known to graze selectively on certain phytoplankton taxa, such as the chlorophytes *Chlamydomonas* and *Cosmarium* (Bleiwass and Stokes, 1985; Bogdan and Gilbert, 1982; Demott, 1982; DeMott, 1986). The other pelagic filter-feeder, *Daphnia* is also known to preferentially utilize phytoplankton and bacteria (Brett et al., 2009; Grey et al., 2000; Grey and Jones, 1999; Gu et al., 1994; Knisely and Geller, 1986), yet likely less selective relative to *Bosmina* (Demott, 1982; DeMott and Kerfoot, 1982). The feeding ecology of the littoral cladoceran *Alona* is poorly established, although circumstantial evidence suggests they may prefer epiphytic over planktonic algae (Downing, 1981; Sakuma et al., 2004). For detritivorous chironomid larvae, preferential utilization of phytoplankton detritus and methane-oxidizing bacteria in surface sediments has been reported (Doi et al., 2006; Johnson, 1987; Jones et al., 2008; Solomon et al., 2008). However, the degree of selective feeding by a given taxon may vary depending on the temporal and spatial differences in availability of preferred food items and presence of different species, tribes, subfamilies within the broader taxonomic group (Cole et al., 2011; Solomon et al., 2008; Tanentzap et al., 2017; van Hardenbroek et al., 2014).

Impacts of industrial deposition

The study lakes are located downwind of and in near proximity to the AOSR, a major source of atmospheric sulphur and nitrogen oxides, as well as base cations (Fenn et al., 2015; Percy, 2013). Even though the study lakes are less sensitive to acidification due to their high geological buffering capacity (Laird et al., 2017), N-limited (or N-P co-limited) lakes are still vulnerable to increases in primary productivity due to deposition of reactive N (Curtis et al., 2010; Fenn et al., 2015). Consequently, it is feasible that some food-web related changes induced by enhanced lake productivity could be reflected by $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ trajectories of invertebrate remains. However, as we did not detect any changes in trajectories corresponding to intensified industrial development ca. post 1980 in any of the lakes, we believe that atmospheric deposition of industrial pollutants played a negligible role in the patterns recorded in this study. Although speculative, such conclusion is in agreement with other paleolimnological proxies from the same sediment cores (i.e., molar C:N ratios of SOM, diatom assemblages, scaled-chrysophytes and cladoceran composition), which indicated only limited industrial impacts (Hesjedal 2017; Laird et al. 2017; Mushet et al. 2017).

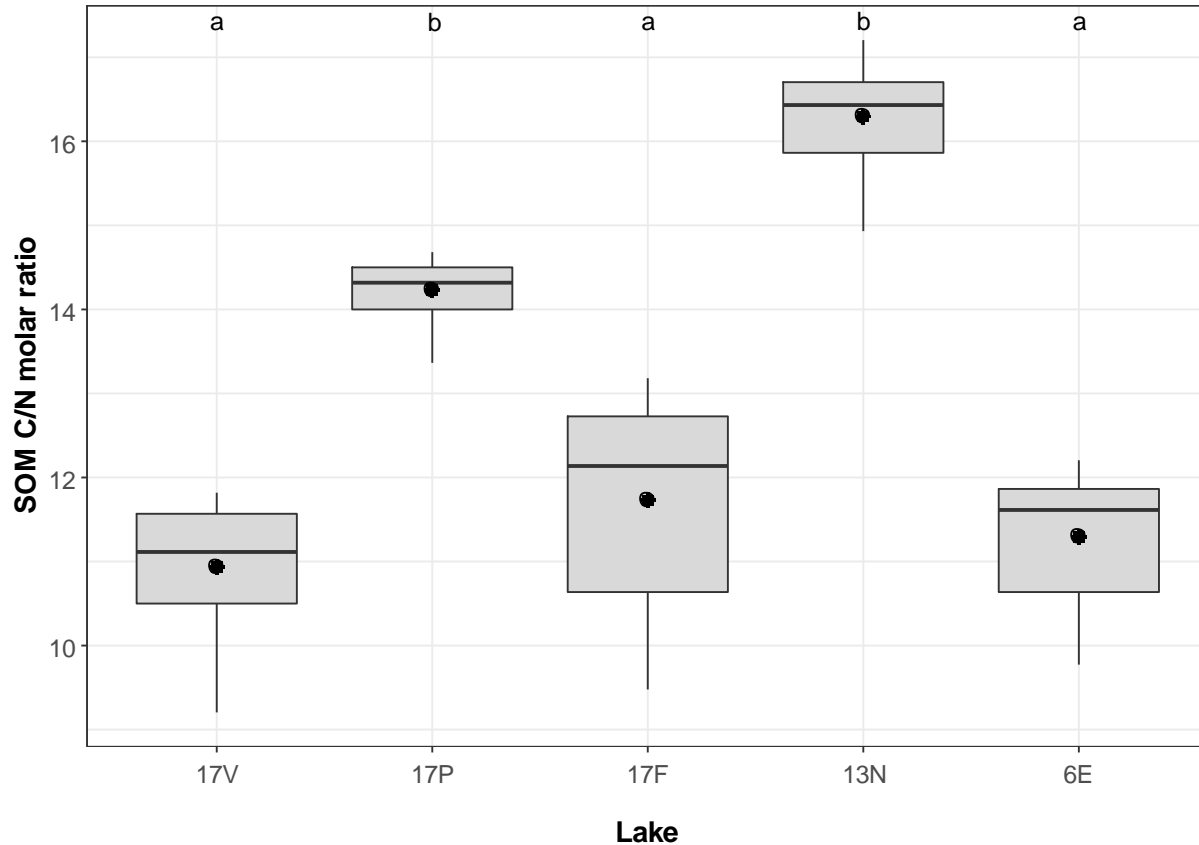


Fig.S1. Comparison of SOM C/N ratios among study lakes. The grey box represents quartiles (25-75%), the whiskers extend to the furthest data point that is within 1.5 times the interquartile range, the horizontal bar in the box is the median and solid circles (●) indicate the mean values. Median values of lakes indicated by different letters are significantly different (adjusted $p < 0.05$) according to Kruskal-Wallis test followed by Dunn's test for multiple comparisons (Dunn, 1964).

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