Taxon-specific variation in δ13C and δ15N of subfossil invertebrate remains: Insights into historical trophodynamics in lake food-webs

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1 **Taxon-specific variation in δ^{13}C and δ^{15}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 $\delta^{13}C$ and $\delta^{15}N$ 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 $\delta^{13}C$ and

18    $\delta^{15}N$ 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 $\delta^{13}C$ and

25    $\delta^{15}N$ in relatively clear-water lakes, possibly reflecting temporal variation in diets or tissue

26   fractionation. Comparison 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 evaluation of taxon-specific $\delta^{13}C$ and $\delta^{15}N$ can provide valuable insights into

30   historical tropho-dynamics in lake food webs.

1. Introduction

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

Unlike SI composition of invertebrate remains, analysis of δ^{13}C and δ^{15}N of bulk sedimentary organic matter (SOM) has been commonly incorporated into paleolimnological

54   studies. SOM is a heterogeneous mixture of plant-, animal- and microbial-derived organic

55   matter originated in different habitats of the lake (i.e. littoral, pelagic and benthic) and received

56   from the watershed (Meyers and Teranes, 2001). Consequently, accurate interpretation of the

57   mechanisms underlying historical changes in isotopic signals of SOM can be difficult due to the

58   potential for multiple pathways influencing both mass fluxes and isotopic value of source

59   materials within the lake and its watershed (Meyers and Teranes, 2001; van Hardenbroek et al.,

60   2014). In this regard, isotopic trends of sub-fossil invertebrate remains are considered to be

61   better indicators of changes in C and N dynamics within the food webs than are those of SOM

62   (Griffiths et al., 2010; Simon et al., 2017; van Hardenbroek et al., 2014), given *in situ* primary

63   production likely contribute relatively little to total SOM relative to terrestrial organic matter

64   (Von Wachenfeldt and Tranvik, 2008), and invertebrate consumers are thought to feed

65   selectively on autochthonous sources (del Giorgio and France 1996; Meili et al. 1996; Doi et al.

66   2006). However, isotopic trends of sub-fossil invertebrate remains and SOM may not necessarily

67   and universally be different. Not all invertebrate taxa are equally selective in their grazing

68   (Demott, 1982; Tanentzap et al., 2017). Further, in systems where available basal resources are

69   overwhelmingly dominated by allochthonous sources, invertebrates may be forced to feed less-

70   selectively (Cole et al., 2011; Persaud et al., 2009; Tanentzap et al., 2017) and consequently,

71   isotopic trends of invertebrate subfossils may not be different from that of SOM with

72   predominant allochthonous origin (Simon et al., 2017). Therefore, differences in temporal

73   patterns between SOM and invertebrate subfossils within each lake should be tested using

74   powerful statistical methods. Yet, in past studies, differences in temporal patterns between SOM

75   and invertebrates were inferred only from pair-wise comparisons of raw values without

76　statistically testing if temporal trends were significantly different (Kattel et al., 2015; Perga et al.,

77　2010; van Hardenbroek et al., 2014).

78　　　　Comparisons of isotopic trends among taxon-specific invertebrate remains can yield

79　insights into historical changes in energy and nutrient flux through different food-web

80　compartments of lakes. For instance, δ^{13}C values within a given ecosystem may differ among

81　invertebrate taxa with different habitat affinities (i.e., pelagic, littoral, benthic) and/or feeding

82　modes owing to differences in dietary sources of C (Matthews and Mazumder, 2003; Persaud et

83　al., 2009), which may converge or further diverge over time in response to changes in basal

84　resources (del Giorgio and France, 1996; Grey et al., 2001; Vadeboncoeur et al., 2003). For

85　δ^{15}N, diverging or converging temporal trends between two taxa may indicate variations in their

86　relative trophic positions over time (Patoine et al. 2006; Perga et al. 2010) or shifts in selective

87　feeding on alternate N sources (France, 1995a; Mulholland et al., 2000). Fortunately, recent

88　improvements in technology to lower mass required for analysis (Maddi et al., 2006; Perga,

89　2009; van Hardenbroek et al., 2010) have allowed investigators to better determine the SI values

90　of individual taxa (Frossard et al., 2013b; van Hardenbroek et al., 2014, 2013; Wooller et al.,

91　2012), although better statistical analysis of the significance of historical trends is still needed.

92　　　　Changes in C and N cycling in Canadian Boreal Shield region during the past century due

93　to environmental perturbations (Anas et al., 2015; Kurz et al., 2014) may have in turn affected

94　trophic dynamics in lacustrine food webs. Several studies have reported not only multidecadal

95　changes in dissolved organic matter (DOM) inputs to lakes from terrestrial sources, also altered

96　in-lake C processing in Boreal Shield regions in response to individual or combined effects of

97　climate change in temperature and precipitation and/or atmospheric deposition of sulfur and N

98　species from anthropogenic sources or recovery form it (Dillon and Molot, 2005; Keller et al.,

99   2008; Zhang et al., 2010). In addition, these perturbations have also induced changes in nutrient

100   dynamics and consequent changes in productivity in lakes (Dillon and Molot, 2005; Holtgrieve

101   et al., 2011; Schindler and Lee, 2010). The nature and degree of responses likely vary among

102   individual lakes depending on local characteristics including catchment properties, lake

103   morphometry, type and degree of nutrient limitation, etc. (Anas et al., 2015; Zhang et al., 2010).

104   These changes in turn may have variably influenced energy and nutrient fluxes through different

105   invertebrate taxa with different habitat or feeding niches within lakes. For instance, changes in

106   allochthonous DOM inputs may strongly influence the diet of non-selective feeders relative to

107   selective feeders of phytoplankton (Persaud et al., 2009; Tanentzap et al., 2017). Further, the diet

108   of littoral consumers can be affected more than that of pelagic consumers by a shift from

109   predominant benthic production to a pelagic regime following eutrophication (Vadeboncoeur et

110   al., 2003). However, such historical food-web consequences of environmental perturbations in

111   Boreal Shield lakes is poorly understood (Persaud et al., 2009), while SI analysis of subfossil

112   invertebrate remains provides a potential means for evaluating these long-term temporal

113   trophodynamics of individual taxa (Frossard et al., 2014; Perga, 2009; van Hardenbroek et al.,

114   2014).

115   Here, we quantified time series of $\delta^{13}C$ and $\delta^{15}N$ in SOM and fossils from individual

116   invertebrate taxa during the 20th century in five Boreal Shield lakes of central Canada to evaluate

117   whether taxon-specific SI analyses improve insights concerning historical changes in lacustrine

118   ecosystems. These systems are heterogeneous in terms of local characteristics including lake

119   morphometry, physiochemical conditions, catchment properties and proximity to nearby major

120   source of atmospheric pollutants i.e. Athabasca Oil Sands Region (AOSR). (See below).

121   Specifically, we analyzed chitinous remains of taxa representing different habitats of lakes (and

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122   likely with different feeding modes) i.e. *Daphnia* and *Bosmina* spp. (pelagic), *Alona* sp. (littoral)

123   and Chironomidae (benthic) and statistically compared the isotopic trajectories (i) between SOM

124   and invertebrate taxa, and (ii) among invertebrate taxa within each lake. We hypothesized that

125   isotopic trends between invertebrate subfossils reflective of food web-specific dynamics would

126   be significantly different from SOM which derives from heterogeneous sources, yet such

127   differences would be system- and/or taxon-specific. Further, we expected that SI trends would

128   be significantly different among invertebrate taxa representing different habitat or feeding niches

129   due to variable food-web related effects of above-mentioned environmental perturbations

130   occurring in Boreal Shield region. We also expected that among-taxon differences in SI trends

131   would be system-specific as different lakes may have responded differently to environmental

132   perturbations due to differences in local characteristics. To our knowledge, this is the first study

133   where taxon-specific SI analyses of invertebrate remains has been performed on multiple study

134   lakes, and in which robust statistical approaches were applied to compare isotopic trends of

135   invertebrate taxa and SOM.

136   **2. Methods**

137   *2.1. Study lakes*

138   Our five study lakes were a subset of 16 lakes in northwest Saskatchewan (Fig. 1)

139   described in Mushet et al. (2017), that represent 262 basins surveyed by the provincial

140   government during 2007-2009 and 2011 (Scott et al., 2010). Initially, sites were selected to

141   represent type of nutrient limitation i.e. phosphorus-limited vs. nitrogen-limited (based on molar

142   dissolved inorganic nitrogen to total phosphorus [DIN: TP] ratios) and degree of vulnerability to

143   atmospheric emissions from AOSR (based on distance from the industrial center) (Table 1). Of

7

144   the 16 lakes cored, the five lakes examined here were chosen on the basis of their elevated

145   densities of fossil invertebrate remains (Hesjedal, 2017) which allowed sufficient sample size to

146   conduct taxon-specific SI analyses. Final lake selection included two N-limited lakes (basins

147   17V, 6E) and three phosphorus-limited lakes (17P, 17F, 13N).

148   Study lakes are mostly small (surface area < 65 ha) and shallow (maximum depth < 10

149   m), with the exception of 13N which is both relatively large and deep (Table 1). The pH of each

150   basin was circumneutral (range 6.8-7.3), yet alkalinity varied considerably among lakes (range

151   4.2-16.5 mg L^{-1} $CaCO_3$). Study lakes were largely oligotrophic to mesotrophic, as reflected by

152   low water-column concentration of TP (range 5.0-16.5 µg L^{-1}), total nitrogen (TN; range 253-

153   315 µg L^{-1}) and chlorophyll a (range 2.0-6.1 µg L^{-1}). In 13N and 17P, dissolved organic carbon

154   (DOC) concentration (each 7.1 mg L^{-1}) and water color (26.2 and 32.8 mg L^{-1} Pt units,

155   respectively) were considerably higher than in other study lakes, indicating the greater

156   importance of allochthonous DOM in these lakes (Wetzel, 1983). We refer to Scott *et al*., (2010)

157   and Anas *et al*., (2014) for detailed descriptions of survey domain, water chemistry and hydro-

158   morphological attributes of study lakes.

159   *2.2. Sediment coring*

160   Sediment cores were collected in February and April 2014. A single core was retrieved

161   from the approximate center of each lake (Table 1) using a Glew gravity corer with a 7.6-cm

162   internal diameter tube. The length of the cores varied from 36.5 to 48.5 cm, which were

163   sectioned into 0.5-cm intervals upon return to the base camp. Samples were shipped to Queen's

164   University, Kingston, Ontario where they were stored at 4°C in a cold room until further

165   analyses.

166   *2.3. Core chronologies*

167   Details on radiometric dating (using ^{210}Pb activities) and age models of the cores are

168   described in Mushet et al. (2017). All cores exhibited robust depth-time chronologies, with the

169   sediment accumulation rates of 17V, 13N and 6E sufficient to achieve a temporal resolution

170   between 2 and 8 years (spaced 0.5 or 1 cm apart, respectively). In contrast, temporal resolutions

171   of cores from lakes 17F and 17P were ~3 - 13 years and 8 - 16 years, respectively, reflecting

172   lower mass accumulation rates (Mushet et al., 2017).

173   *2.4. SI analysis of SOM*

174   Approximately 3 mg of freeze-dried sediment from each interval were packed in tin

175   capsules, and combusted in an Elemental Combustion System (Costech EA) coupled with a

176   Thermo-Finnigan Delta V isotope ratio mass spectrometer (IRMS). Bovine liver and wheat flour

177   were used as internal laboratory standards. Isotopic ratios are reported in the conventional δ

178   notation (‰), relative to Vienna Pee-Dee-Belemnite (C) and atmospheric N_2 (N) standards

179   (Cooper and Wissel, 2012). Agreement between duplicate sub-samples was within 0.2‰ for

180   each element. Acidification of sediment samples indicated that inorganic C was negligible in all

181   cores (Wolfe, 2016).

182   *2.5. SI analysis of subfossil invertebrate remains*

183   To recover sufficient subfossil invertebrate remains for SI analysis, sediment samples

184   were first deflocculated in warm 10% KOH for 2 hrs, then thoroughly rinsed with deionized

185   water and concentrated onto a 106-μm mesh sieve (Perga, 2011; van Hardenbroek et al., 2010).

186   Subsequently, the chitinous remains from each taxon were isolated by hand with fine forceps

187   under a dissecting microscope, placed in pre-weighed tin capsules, and dried at 50°C to constant

188   weight. Subfossil remains included headshields and carapaces of *Bosmina* (average 250 remains

189   sample[-1], 175 µg dry weight) and *Alona* (average 115 remains, 110 µg dry weight), ephippia

190   (diapausing eggs) of *Daphnia* (average 88 remains, 105 µg dry weight) and head capsules of

191   chironomids (average 7 remains, 32 µg dry weight). Sample sizes were almost always sufficient

192   to provide accurate δ^{13}C measurements, yet in some cases were inadequate to obtain reliable

193   δ^{15}N measurements (mostly *Daphnia* ephippia).

194   The protocol for stable isotope analysis of invertebrate subfossils was similar to that of

195   SOM, with additional modification of the combustion system to accommodate the small sample

196   mass (Carman and Fry, 2002; Maddi et al., 2006). Briefly, the high temperature-reactor system

197   was minimized by combining combustion and reduction columns into one column with a smaller

198   inner diameter of 8 mm. The single column was filled halfway with elemental copper with a

199   lower reactor temperature of 800°C to prevent melting of the elemental copper. Flow rate was

200   set to 50 mL min[-1]. Due to the relatively quick oxidation of the elemental copper and narrow

201   zone of optimum temperature, reactors had to be changed about every 100 samples.

202   *2.6. Data analysis*

203   Generalized additive models (GAMs) were used to test significant differences of isotopic

204   time series both i) between SOM and invertebrate taxa and ii) among invertebrate taxa, by

205   modifying the approach of Rose et al. (2012). GAMs are a form of semi-parametric regression,

206   which can estimate non-linear relationships between predictors and response in terms of smooth

207   functions of predictors (Wood, 2017). Location scale GAMs where the response is conditionally

208   distributed Gaussian were selected given the considerable differences in variance of δ^{13}C and

209   δ^{15}N values among sample types (SOM, invertebrates) (Fig.2),Location scale GAMs allow both

210   mean (location) and variance (scale) of the response distribution to be modelled as a function of

211   predictors. For each lake, we fitted an ordered-factor-smooth interaction model of the form:

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

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

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

215   where sample type is an ordered factor variable representing SOM and invertebrate taxa, while

216   SOM is the reference level. The response y_{ij} (i.e. ith δ^{13}C or δ^{15}N observation in the jth sample

217   type) was Gaussian-distributed with a mean μ_{ij} and variance σ_{ij}^{2}. Here μ_{ij} was modelled as the

218   mean value of the response in the reference sample type (SOM) (model intercept α_0), plus the

219   difference between mean response of the jth sample type and α_0 (α_{1j}), plus the centered smooth

220   function of time (sediment age in years AD) for the reference sample type SOM (f_1), plus the

221   difference between smooth function of time for the jth sample type and f_1 (f_{2j}). In other words,

222   the model tests i) if the trend (change over time) of SOM δ^{13}C/δ^{15}N was statistically significant

223   and ii) if both means and trends of δ^{13}C/ δ^{15}N of individual invertebrate taxa were significantly

224   different from those of SOM within a given lake. Restricted maximum likelihood (REML)

225   estimation method and thin-plate regression splines were used to parameterize f_1 and f_{2j}

226   (Simpson, 2018; Wood, 2017). We also examined diagnostic plots to check any residual

227   autocorrelation.

228   Given the large differences in variances among sample types within lakes (Fig.2), it was

229   necessary to model σ_{ij} as a part of the GAM. Hence, σ_{ij}-b (log scale) was fitted as a linear

230   function of the sample type γ_{1j} and the time interval represented by each sediment section (γ_2),

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

232   singularities in the model likelihood.

233   Subsequently, we processed the GAM output (following Rose et al., 2012) to both

234   generate smooth SI trends for individual invertebrate taxa and estimate differences in SI trends

235   between pairs of taxa, as well as to generate approximate 95% point-wise confidence intervals

236   (Supplementary methods; Supplementary data). We inferred a significant difference in isotopic

237   trends between a pair of taxa when the confidence interval excluded zero for considerable time

238   duration.

239   All statistical analysis were performed in R version 3.4.3 (R Core Team, 2017) using the

240   packages 'mgcv'(Wood, 2017; Wood et al., 2016) and 'ggplot2'(Wickham, 2009).

241

242   **3. Results**

243   *3.1. $\delta^{13}C$ of SOM and invertebrate taxa*

244   Across all study lakes, $\delta^{13}C$ values of SOM ranged from -29.3 to -22.8‰ (Fig.2a), with

245   lakes 17F and 17V exhibiting more enriched $\delta^{13}C$ values than the other lakes (mean difference of

246   ~3-4‰). For a given taxon, the $\delta^{13}C$ of invertebrate remains varied less among lakes (-28 and -

247   24‰) than did SI values of SOM (range), except *Daphnia* which showed relatively high

248   variation in core-wide mean $\delta^{13}C$ values (~7‰). Within lakes, ranges of $\delta^{13}C$ often differed

249   among taxa, although there was no consistent pattern among sites as to which taxon was most

250   variable.

251   GAMs, by modeling both mean and variance components, explained a large proportion of

252   the historical changes in $\delta^{13}C$ in each lake (explained deviance ranging 91-98%). Sample type

253   (i.e., SOM and invertebrate taxa) was a significant predictor ($p < 0.05$) of mean $\delta^{13}C$ i.e. mean

254    $\delta^{13}C$ of all invertebrate taxa within a lake was significantly different ($p < 0.05$) from that of SOM

255   in each lake (Fig. 2a). For variance of $\delta^{13}C$, sample type (i.e., SOM and invertebrate taxa) had a

256   significant effect ($p < 0.05$) for all lakes, while a significant influence ($p < 0.05$) of the time

257   interval (represented by each sediment section) was only detected for lakes 17V and 13N.

258   The $\delta^{13}C$ of SOM in all lakes exhibited statistically significant ($p < 0.05$) declines during

259   the 20th century (Table 2, Fig. 3), with more prominent trends in lakes 6E and 17F relative to

260   other basins. Trajectories of invertebrate $\delta^{13}C$ generally followed the declining trend of SOM

261    $\delta^{13}C$ in each lake (Fig.3), although invertebrate time series exhibited noticeable differences in

262   timing, pattern and degree of change both within and among lakes. In lakes 17V, 17F and 6E,

263   temporal trends of invertebrate taxa varied ($p < 0.05$) from that of SOM, with the exception of

264   *Chironomid* in lake 17V (Table 2 and Fig. 4). On the other hand, no significant differences in

265   trends ($p < 0.05$) between invertebrate taxa and SOM were detected for 13N and 17P.

266   There were few consistent differences in C source for invertebrates both within and

267   among lakes (6E, 17V) with sufficient fossil density of common taxa (Fig. 5). For example, the

268    $\delta^{13}C$ trajectories of *Daphnia* and *Bosmina* differed significantly ($p < 0.05$) from each other and

269   from other taxa for much of the past 100 years in lake 6E. Similarly, $\delta^{13}C$ trends in lake 17V

270   were significantly different ($p < 0.05$) between *Bosmina* and *Daphnia*, *Bosmina* and *Chironomid*,

271   *Daphnia* and *Alona,* and *Alona* and *Chironomid*. However, the nature and degree of trend

272   differences for same pairs of taxa were not similar among lakes.

273   *3.2. $\delta^{15}N$ of SOM and invertebrate taxa*

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

275 with lakes 13N and 17P exhibiting more enriched $\delta^{15}N$ (mean difference ~2.0-3.5‰) with less

276 variability relative to other sites (Fig.2b). Overall, $\delta^{15}N$ values of invertebrate remains showed a

277 higher variability than that of SOM (-1.9 to 4.1‰), which ranged from -1.4 to 9.7‰ across all

278 study lakes with considerable among-lake differences.

279 Similar to patterns seen with $\delta^{13}C$, GAM analysis explained most variation in

280 sedimentary $\delta^{15}N$ for all parameters in each lake (96-99% deviance explained). Within each site,

281 mean $\delta^{15}N$ values of individual invertebrates were significantly enriched ($p < 0.05$) relative to

282 SOM for most taxa, except *Daphnia* in 17P (Fig .2b). Variance of $\delta^{15}N$ was significantly ($p <$

283 0.05) influenced by sample type (SOM and invertebrate taxa), but not by the time interval

284 represented by each sediment section ($p > 0.05$).

285 All lakes showed significant ($p < 0.05$) historical declines in $\delta^{15}N$ of SOM and

286 invertebrate taxa (Table 3 and Fig. 6). Specifically, $\delta^{15}N$ values of SOM and invertebrates

287 declined after ca.1940-1950 in lakes 17V, 17P, 17V and 6E, whereas a continuous, linear decline

288 was detected in 13N over the entire 120 year record. While significant differences ($p < 0.05$) in

289 $\delta^{15}N$ trends between *Bosmina, Alona* and *Chironomid* and SOM in 6E and between *Alona* and

290 SOM in lakes 17V and 17 F were detected (Table 3 and Fig. 7), there was no similarity in the

291 nature or degree of difference in invertebrate-SOM trends among lakes. Similarly, time series

292 of $\delta^{15}N$ differed significantly ($p < 0.05$) among most pairs of invertebrates in lakes 6E and 17V

293 (Fig.8), with few common difference patterns among pairs of taxa.

294 **4. Discussion**

295 The novel combination of taxon-specific isotope time series and GAMs demonstrated

296 that significant differences in mean $\delta^{13}C$ and $\delta^{15}N$ values exist between SOM and invertebrate

14

297   taxa in all lakes, yet significant differences in long-term trends were only detected in certain

298   basins. Significant differences in mean $\delta^{13}C$ and $\delta^{15}N$ between SOM and invertebrate taxa may

299   be reflective of selective feeding by invertebrates (Jones et al., 2008, 1999) and differences in

300   trophic position within food webs (Vander Zanden and Rasmussen, 2001). In contrast,

301   differences in historical trends among sample types (i.e. SOM and invertebrate taxa) within lakes

302   were more pronounced for $\delta^{15}N$ than $\delta^{13}C$, suggesting that either the biogeochemistry of N

303   (Bunting et al., 2010) or trophic relationships among taxa (Karlsson et al., 2004) was more

304   variable than the sources or cycling of C in these boreal ecosystems. Overall, few differences in

305   SI trends between SOM and invertebrates and among different taxa were noted in lakes with

306   more elevated DOC content, whereas such differences in SI time series were more pronounced in

307   relatively clearwater lakes, possibly reflecting variation in the importance of allochthonous C to

308   invertebrate consumers (Solomon et al., 2008; Tanentzap et al., 2017), as well as a higher degree

309   of habitat diversity or feeding selectivity in transparent ecosystems (Matthews and Mazumder,

310   2006; Persaud et al., 2009). Together, these findings suggest that the analysis of taxon-specific

311   fossil invertebrate time series provides unique insights into the trophic relationships in lentic

312   ecosystems that cannot be achieved through analysis of bulk sediments alone (See also Wyn et

313   al. 2007; Perga et al. 2010; van Hardenbroek et al. 2014).

314   *4.1. SI variability of SOM and invertebrate remains*

315   Overall, the range of SOM $\delta^{13}C$ values detected across study lakes (-29.3 to -22.8‰)

316   (Fig.2) is indicative of mixed contributions of C from C3 terrestrial plants (-28 to -27‰) and

317   lacustrine primary producers (-35 to -8‰) (Meyers and Teranes, 2001; Vuorio et al., 2006), with

318   among-lake differences likely related to factors that control origin and fate of accumulated C

319   (Supplementary discussion; Supplementary data). Meanwhile, low SOM $\delta^{15}N$ values in several

320   study lakes (< 2‰) (Fig.2) indicate fixed N is the main N source (Meyers and Teranes, 2001;

321   Patoine et al., 2006), which is likely derived from terrestrial sources (e.g. plants such as *Alnus*

322   spp. and *Shepherdia canadensis* and microorganisms in deciduous forest) (Bunting et al., 2010;

323   Engstrom and Fritz, 2006), as pigments for N_2-fixing cyanobacteria were not detected in

324   sediments of these lakes (Wolfe, 2016). Among-lake differences in SOM $\delta^{15}N$ on the other hand

325   can arise due to differences in the mass flux or isotopic ratios of fixed DOM-N as well as

326   subsequent modification by biological (terrestrial and in-lake) processes (Supplementary

327   discussion; Supplementary data).

328   The $\delta^{13}C$ and $\delta^{15}N$ values of *Bosmina* exoskeletons (28.1‰ to -23‰ and 0.9 to 9.7‰,

329   respectively) and *Daphnia* ephippia (-30.6‰ to -23.5‰ and -1.4 to 9‰ respectively) recorded in

330   the present study were similar to contemporary values recorded for these taxa in a synoptic

331   survey of 233 regional boreal lakes (*Bosmina* -31.4 to -22.9‰ and -1.5 to 9‰, respectively;

332   *Daphnia* -35.8 to -24.3‰ and 0.5 to 7.8‰, respectively) (Anas, 2019). This supports the high

333   likelihood that downcore isotopic variations in invertebrate sub-fossils are within the expected

334   range in response to regional environmental variations such as fluctuations in primary

335   productivity, allochthonous organic matter inputs, in-lake biogeochemical transformations and

336   catchment processes (Anas, 2019). Elsewhere highly negative $\delta^{13}C$ values (~-50 to -40‰) of

337   subfossil chironomid head capsules and *Daphnia* ephippia may reflect contributions of ^{13}C-

338   depleted methonogenic carbon to invertebrate diets (Frossard et al., 2013b; Rinta et al., 2016;

339   Schilder et al., 2017; van Hardenbroek et al., 2013; Wooller et al., 2012), a factor which does not

340   seem to be important in our study (Fig. 2). Instead, invertebrate $\delta^{13}C$ values herein were usually

341   within the range expected for photoautotrophically-produced C in aquatic and terrestrial

342   ecosystems (-35 to -8‰) (France, 1996, 1995b; Meyers and Teranes, 2001; Vuorio et al., 2006).

343

4.2. SI trends of SOM

345   Patterns of sedimentary δ^{13}C decline over the 20th century (Fig.3) are similar to those

346   observed of other regional studies of boreal western Canada (Ahad et al., 2011; Curtis et al.,

347   2010). In principle, such depletion of ^{13}C content may arise because of increased abundance and

348   assimilation of ^{13}C-depleted respired DIC (originated from heterotrophic respiration of terrestrial

349   organic matter) by primary producers over the past century (France et al., 1997; Meyers and

350   Teranes, 2001). Alternately, such relatively small declines (1-2‰) may reflect changes in the

351   isotopic composition of atmospheric CO_2 as a result of accelerated release of ^{13}C-depleted CO_2

352   from fossil fuels, known as Suess Effect (Schelske and Hodell, 1995).

353   As historical data are not available for much the study region (Wolfe 2016), it is difficult

354   to unambiguously identify the precise mechanism(s) underlying progressive declines in δ^{15}N of

355   SOM. However, given that declines occur in all lakes, irrespective of the mode of nutrient

356   limitation (N vs. P) and relationship with respect to aerial deposition of nitrogenous pollutants

357   from AOSR (Mushet et al. 2017; Laird et al. 2017), we infer that climate-related processes, such

358   as regional brownification (Anas et al., 2015) leading to increased influx of ^{15}N-depleted

359   terrestrial DOM-N (Bunting et al., 2010) may underlie the common trend among lakes. This

360   hypothesis is supported by the observations that terrestrially-derived DOM-N is the major N

361   source to these lakes (see above) and that the abundance of mixotrophic algae has increased in

362   these lakes during the past century (Wolfe 2016; Stevenson et al. 2016).

4.3. Differences in δ^{13}C time series between invertebrate remains and SOM

364   Significant within-lake differences in mean $\delta^{13}C$ of SOM and chitinous remains (Fig.2)

365   are in line with findings of other paleolimnological studies (Kattel et al., 2015; Simon et al.,

366   2017; van Hardenbroek et al., 2014, 2013). In principle, such deviations can arise from

367   preferential ingestion or assimilation of specific fractions of particulate organic matter (POM) or

368   surface sediment by invertebrates (Jones et al., 2008, 1999). Bulk POM is a mixture of several

369   constituents (i.e. algae, detritus, bacteria and small planktonic organisms) each with a different

370    $\delta^{13}C$ value (del Giorgio and France, 1996; Meili, 1992) and relative contribution to the POM

371   pool. POM is eventually deposited as sediment and, as a result, $\delta^{13}C$ of surface sediment is

372   generally similar to that of POM in overlying waters (Jones et al., 2008; Meili et al., 1996;

373   Meyers and Teranes, 2001). Contemporary studies which detect differences between $\delta^{13}C$ of

374   lacustrine POM or surface sediment and invertebrate consumers (cladocerans and chironomids)

375   often attribute these patterns to the effects of preferential grazing and/or assimilation of

376   isotopically-distinct fraction within the diet (del Giorgio and France, 1996; Doi et al., 2006;

377   Jones et al., 2008, 1999; Meili et al., 1996). This isotopic deviation is likely conserved between

378   invertebrate remains and POM detritus even after burial in lake sediments (Schilder et al.,

379   2015a), assuming that diagenetic alterations of $\delta^{13}C$ is negligible or the same in both materials.

380   Meanwhile, empirical and circumstantial evidence indicates that invertebrate taxa examined here

381   can exhibit feeding selectivity due to physical, sensory and nutritional properties of food items

382   (supplementary discussion: Supplementary data).

383   Significant differences between $\delta^{13}C$ trends of SOM and invertebrates from different

384   habitats within lakes 17V, 17F and 6E suggest that analysis of taxon-specific SI can provide new

385   insights on the regulation of food-web carbon dynamics of different lake habitats. For instance,

386   variation in phytoplankton $\delta^{13}C$ due to changes in primary productivity, DIC content or DIC

387 source (France et al., 1997; Meyers and Teranes, 2001) should be reflected in isotopic values of

388 invertebrate remains if phytoplankton-derived organic matter is grazed or assimilated

389 preferentially by invertebrates. Such fine-scale trophic patterns may be partially or completely

390 masked in analysis of SI in SOM in instances where phytoplankton-derived organic matter is

391 mixed with other sources of POM or DOM (Davidson and Jeppesen, 2013; Meyers and Teranes,

392 2001). For the same reason, SOM δ^{13}C may fail to reflect environmentally-induced variations in

393 epiphytic δ^{13}C which are faithfully recorded in exoskeletons of littoral herbivore *Alona*, or the

394 incorporation of C from methanogens by benthic chironomids (Schilder et al., 2017; Simon et al.,

395 2017; van Hardenbroek et al., 2014). Finally, historical trends in invertebrate δ^{13}C may reflect

396 shifts in feeding selectivity of source population due to temporal variations in relative

397 availability of autochthonous vs. allochthonous resources (Demott, 1982; Tanentzap et al., 2017).

398 Under such conditions, the δ^{13}C values of invertebrate remains may be expected to diverge from

399 those of bulk SOM during periods of high feeding selectivity (and converge as feeding

400 selectivity decreases).

401 Trends in δ^{13}C of invertebrate remains did not vary significantly from SOM in lakes 17P

402 and 13N (Fig.4), suggesting existence of SOM-invertebrate trend differences can be system-

403 specific. This may be a result of strong influence of allochthonous inputs on both SOM and

404 invertebrate diets in these dystrophic lakes. In particular, these sites exhibited not only much

405 higher DOC concentrations and water color (Table 1), but also elevated SOM C:N (molar) ratios

406 (ca. 13-15 and 15-17, respectively) compared to other study lakes (Fig. S1; Supplementary data),

407 indicating substantial terrestrial organic matter inputs over the past century (Meyers and Teranes,

408 2001). Although speculative, we suggest that invertebrate consumers in these lakes were relying

409 more on allochthonous C sources (terrestrial detritus and/or heterotrophic bacteria), given they

410   were more abundant than preferred autochthonous foods (Cole et al., 2011; Simon et al., 2017;

411   Solomon et al., 2008; Tanentzap et al., 2017). Allochthonous organic matter may also have

412   largely contributed to SOM in these dystrophic lakes (Dalton et al., 2018; Meyers and Teranes,

413   2001). Such large contribution of allochthonous sources to both invertebrate diets and SOM

414   may have led to temporal coherence between δ^{13}C trends of invertebrate remains and SOM.

415   *4.4. Differences in δ^{13}C trends among invertebrate taxa*

416   The significant among-taxon differences in δ^{13}C trends of lakes 17V and 6E indicate that

417   flux of carbon through invertebrates can differ among taxa, habitats and lakes, and that all

418   pathways may have decoupled through time (Fig.5). Dietary sources of C for invertebrates, as

419   well as the associated δ^{13}C values of C source can be impacted by habitat-specific or whole-lake

420   environmental changes (Persaud et al., 2009; Vadeboncoeur et al., 2003; van Hardenbroek et al.,

421   2014). In particular, changes in relative availability of different carbon sources (i.e.

422   phytoplankton, epiphyton, terrestrial, methane-derived carbon) can be induced by alteration in

423   water transparency by primary producers (Solomon et al., 2008; Vadeboncoeur et al., 2003),

424   physical turbidity (Scheffer et al., 1993) or DOC influx (Carpenter et al., 1988), leading to shifts

425   in the diets of primary consumers (Solomon et al., 2008; Vadeboncoeur et al., 2003). For

426   instance, a switch from predominantly benthic production to a pelagic regime following

427   eutrophication can shift the diet of littoral consumers (e.g. *Alona*) from ^{13}C-enriched epiphyton

428   ((Hecky and Hesslein, 1995) to ^{13}C-depleted phytoplankton and suspended detritus, while diet of

429   pelagic consumers (e.g., *Bosmina*, *Daphnia*) remain unaffected (Vadeboncoeur et al., 2003).

430   Further, δ^{13}C of littoral epiphyton may respond differently compared to pelagic phytoplankton to

431   temporal changes in DIC content and dominant chemical form of inorganic C (CO_2, HCO_3^-,

432   CO_3^{2-}) due to boundary layer effects (Hecky and Hesslein, 1995; McPherson et al., 2015;

433   Woodland et al., 2012), thereby altering historical trends in δ^{13}C of littoral consumers relative to

434   pelagic taxa. Differences in degree of feeding selectivity may have contributed to differences in

435   δ^{13}C through time between pelagic taxa *Bosmina* and *Daphnia*. For instance, δ^{13}C trajectory of

436   less-selective *Daphnia* may be more affected by temporal variations in relative availability of

437   autochthonous vs. allochthonous sources compared to more selective *Bosmina* (Demott, 1982;

438   DeMott and Kerfoot, 1982; Tanentzap et al., 2017). Meanwhile, lack of among-lake similarity in

439   trend difference patterns for pairs of common taxa suggests that individual lakes may have been

440   variably influenced by regional environmental changes (e.g. climatic forcing) owing to

441   differences in local characteristics, leading to differential temporal changes in C fluxes through

442   different habitats or food web compartments in individual lakes.

443   Time series of δ^{13}C did not differ significantly among invertebrates through time in three

444   lakes (13N, 17P, 17F; Fig.5), suggesting the path or process of C assimilation was relatively

445   consistent in food webs of these lakes. Such consistency in the two dystrophic lakes (13N, 17P)

446   may reflect the paramount importance of terrestrial DOM as a food-web C source in all habitats

447   and feeding niches (Solomon et al., 2011), either because of the importance of ^{13}C-depleted

448   respired DIC to primary producers in all habitats, or, the consistent availability of terrestrial

449   detritus and/or heterotrophic bacteria as a C source to primary consumers (see above). In lake

450   17F, both pelagic and benthic habitats were likely autotrophic due to light penetration to the

451   bottom, as indicated by dominance of benthic over planktonic diatoms over the past century

452   (Laird et al., 2017). The unchanged diatom composition in this lake (Laird et al., 2017) suggests

453   that is limnological conditions have remained largely unaltered through time, with few changes

454   in water transparency and the ratio of benthic to pelagic autotrophy. Therefore, that the dietary

455   sources of C to invertebrates may have remained consistent within 17F's habitats through time.

4.5. Differences in δ^{15}N time series between invertebrate remains and SOM

457   The significantly higher mean δ^{15}N of invertebrate remains relative to SOM in our study

458   lakes (Fig.7) is characteristic of a 2-4‰ trophic fractionation between diet and consumer

459   (Vander Zanden and Rasmussen, 2001), assuming that SOM δ^{15}N represents available dietary

460   sources (Perga et al., 2010). The δ^{15}N enrichment relative to SOM varied among invertebrate

461   taxa within each lake (Figs.2, 7), likely as a result of differences in trophic position of individual

462   taxa within the local aquatic food web, as well as variability in metabolic pathways of protein

463   synthesis (Kling et al., 1992; Vander Zanden and Rasmussen, 2001). Meanwhile, among-lake

464   differences in invertebrate δ^{15}N enrichment relative to SOM may be associated with lake-specific

465   factors that can influence the magnitude of ^{15}N fractionation by consumers. For instance,

466   reduced availability of N in some food resources (i.e. high C: N ratios) may lead to elevated diet-

467   tissue ^{15}N fractionation in consumers (Adams and Sterner, 2000). Enriched δ^{15}N values of

468   consumers can also arise under conditions of low-food quantity or quality as a result of increased

469   turnover of internal nitrogen and preferential excretion of ^{14}N (Adams and Sterner, 2000;

470   Karlsson et al., 2004)

471   Significant differences between δ^{15}N trends of SOM and invertebrate subfossils in lakes

472   17V, 17F and 6E (Fig. 7) may have arisen from shifts in dietary sources of N and/or changes in

473   magnitude of isotopic fractionation in invertebrate consumers. For example, δ^{15}N of invertebrate

474   consumers can covary with bacterial contribution to the diet (Grey et al., 2004, 2001; Karlsson et

475   al., 2004). Empirical evidence suggests that bacterial δ^{15}N may differ from other basal resources

476   either due to elevated isotopic fractionation depending on biochemical composition and

477   availability of nitrogen substrate (McGoldrick et al., 2008; Steffan et al., 2015) or assimilation of

478   isotopically light nitrogen substrate (e.g. excreted ammonia) (Grey et al., 2004). In addition, few

479   SI studies (e.g. France, 1995a; Mulholland et al., 2000) suggest that, although not definitive,

480    $\delta^{15}N$ variation of invertebrate consumers can be reflective of shifts in relative dietary importance

481   of autochthonous vs. allochthonous sources. Alternately, changes in $\delta^{15}N$ offset between SOM

482   and invertebrates can reflect variation in trophic position of taxa over decades, assuming that

483   SOM represents an isotopic baseline (Griffiths et al., 2010; Perga et al., 2010). In this case,

484   climatically-induced changes in DOC flux could favour shifts from an algae-dominated to a

485   bacterivorous protozoa-dominated diet (i.e. microflagellates and ciliates) (DeMott and Kerfoot,

486   1982; Ngochera and Bootsma, 2011), which is in turn reflected by temporal changes in $\delta^{15}N$ of

487   invertebrate consumers. Finally, shifts N-rich algae to N-poor allochthonous food sources may

488   lead to elevated $\delta^{15}N$ in aquatic invertebrates (Maguire and Grey, 2006) as a result of increased

489   diet-tissue ^{15}N fractionation (Adams and Sterner, 2000; Karlsson et al., 2004). Similar to findings

490   for $\delta^{13}C$ trends, $\delta^{15}N$ trends of invertebrate subfossils did not differ significantly from SOM in

491   two dystrophic lakes (13N, 17P) likely due to paramount and consistent importance of

492   allochthonous sources to both SOM (Dalton et al., 2018; Meyers and Teranes, 2001) and

493   invertebrate diets (Cole et al., 2011; Simon et al., 2017; Solomon et al., 2008; Tanentzap et al.,

494   2017), again indicating existence of SOM-invertebrate trend differences can be system-specific.

495   *4.6. Differences in $\delta^{15}N$ trends among invertebrate taxa*

496   The presence of significant among-taxon differences in $\delta^{15}N$ trends in lakes 17V and 6E

497   suggests that trophic relationships among taxa may vary in lakes at centennial scales (Fig. 8). As

498   noted above, variable offsets in $\delta^{15}N$ among pairs of species may result from habitat-specific

499   changes in resource materials with different $\delta^{15}N$ values, resource limitation, or changes in

500   metabolic processes of individual taxa. In addition, lake-specific changes in the influx of

501   nitrogenous materials may also influence the degree of agreement in $\delta^{15}N$ trends between pairs

502   of species, particularly from different habitats. In particular, the δ^{15}N of individual taxa may be

503   selectively influenced by dietary shifts or changes in isotopic fractionation while the δ^{15}N of the

504   other taxon remains unaffected (e.g. *Alona* vs. *Bosmina* in 17V). Alternately, δ^{15}N variations of

505   both taxa can be influenced by environmental and physiological mechanisms simultaneously,

506   asynchronously or differentially (e.g. *Alona* vs. *Bosmina* in 6E), as has been seen in modern lake

507   time series (Patoine et al. 2006). Regardless of the underlying mechanism, the observation that

508   historical differences between pairs of common taxa (e.g., *Bosmina* v. *Alona*) vary among lakes

509   (17V, 6E) suggests the possibility of lake-specific food web responses to environmental changes

510   during the past century.

511   again indicating the consistent importance of allochthonous DOM as food web N source

512   in all habitats and feeding niches.

513

514   *4.7. Caveats*

515   Due to methodological constraints, our SI analyses were conducted at a relatively coarse

516   taxonomic resolution relative to modern limnological studies (e.g., *Daphnia* spp., chironomids,

517   etc.). Consequently, interpretations of taxon-specific isotopic variability may be confounded in

518   part by variation in δ^{15}N and δ^{13}C values among con-specific taxa. For example, although

519   chironomid larvae are generally considered as detrivores, feeding mode may vary among species

520   (e.g. deposit feeding, filter feeding and engulfing), which may have consequences for δ^{13}C and

521   δ^{15}N variability (Griffiths et al., 2010; Solomon et al., 2008; van Hardenbroek et al., 2014).

522   Similarly, differences in daphniid body size can influence the accessibility to both very small and

523   large food particles (Kasprzak and Lathrop, 1997). Furthermore, *Daphnia* ephippia may

524 integrate time differently than invertebrate exoskeletons, and may provide a different metric of

525 resource use or trophic position compared to remains deposited as a result of adult death or

526 molting. Ephippia are produced within a very short time frame in response to specific

527 environmental cues such as food limitation, reduced photoperiod and crowding (Kleiven et al.,

528 1992). Hence, they may only provide a temporal snapshot of $\delta^{13}C$ and $\delta^{15}N$ of the source

529 population during their brief period of formation (Griffiths et al., 2010; Perga, 2009). In

530 contrast, invertebrate exoskeletons may provide a relatively long-term representation isotopic

531 composition of the source population.

5. Conclusions

533   Our study demonstrates that $\delta^{13}C$ and $\delta^{15}N$ analysis of taxon-specific invertebrate

534 remains can provide unique insights into historical trophodynamics in lake food webs, different

535 from those derived from an analysis of bulk sediments. Statistically-significant differences in

536 mean values of $\delta^{13}C$ and $\delta^{15}N$ between SOM and invertebrate remains likely reflected selective

537 feeding or trophic fractionation, and appear to be ubiquitous in study lakes despite limnological

538 differences among lakes. Further, the absence of significant differences in historical trends

539 among trophic levels or taxa in relatively dystrophic lakes suggests a stabilizing role of terrestrial

540 C contributions in food-web processes (cf. Wetzel 1989). On the other hand, the higher

541 variability in SOM-invertebrate SI time series in relatively clear lakes may reflect decadal-scale

542 variation in dietary sources of elements or their metabolic processing by invertebrates. That said,

543 it is clear that further refinement of causal mechanisms requires substantial additional work,

544 including additional paleoenvironmental analyses and comparisons with long-term monitoring

545 data. For instance, simultaneous evaluation of $\delta^{13}C$ and $\delta^{15}N$ of chlorins (i.e. chlorophylls and

546 their phaeopigment derivatives) may help to better understand if and how isotopic trends of

547 invertebrate remains are related to changes in aquatic primary productivity (Hayes et al., 1987).

548 Based on our results, we suggest that evaluation of isotopic trends of invertebrate remains

549 relative to SOM, will be a particularly informative means of unraveling past food-web carbon,

550 nutrient and trophic dynamics.

551

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559

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843   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 ($\mu g\ L^{-1}$)	290.0	253.0	299.0	253.0	314.0
TP ($\mu g\ L^{-1}$)	14.0	7.7	6.5	5.0	16.5
Chlorophyll a ($\mu g\ L^{-1}$)	5.4	2.0	3.2	3.1	6.1
pH	7.1	6.8	7.0	7.1	7.3
Alkalinity (mg L^{-1} 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 (mg L^{-1} Pt)	4.8	32.8	6.8	26.6	8.34

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

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

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848 Table 2. Model summaries for SOM and invertebrate δ^{13}C time series of study lakes

Lake	Covariate	EDF	Ref. DF	χ^2	*p*-value
17V	Trend$_{SOM}$	2.41	9.00	36.43	1.16×10^{-6}
	Trend$_{Daphnia\text{-}SOM}$	1.21	5.00	3.74	0.04
	Trend$_{Bosmina\text{-}SOM}$	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	$\text{Trend}_{\text{SOM}}$	7.23	9.00	917.35	$< 2.00 \times 10^{-16}$
	$\text{Trend}_{\textit{Daphnia}\text{-SOM}}$	4.04	5.00	23.47	5.14×10^{-5}
	$\text{Trend}_{\textit{Bosmina}\text{-SOM}}$	2.52	5.00	61.91	2.50×10^{-15}
	$\text{Trend}_{\textit{Alona}\text{-SOM}}$	3.27	5.00	31.83	1.75×10^{-7}
	$\text{Trend}_{\textit{Chironomid}\text{-SOM}}$	2.11	5.00	10.95	2.77×10^{-3}

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

Lake	Covariate	EDF	Ref. DF	χ^2	*p*-value
17V	$\text{Trend}_{\text{SOM}}$	3.97	9.00	293.02	$< 2.00 \times 10^{-16}$
	$\text{Trend}_{\textit{Bosmina}\text{-SOM}}$	4.61×10^{-5}	5.00	0.00	0.60

	Trend$_{Alona\text{-}SOM}$	1.84	5.00	9.96	2.89×10^{-3}
	Trend$_{Chironomid\text{-}SOM}$	3.76×10^{-5}	5.00	0.00	0.56
17P	Trend$_{SOM}$	1.84	2.00	24.71	1.47×10^{-6}
	Trend$_{Daphnia\text{-}SOM}$	5.30×10^{-7}	2.00	0.00	1.00
	Trend$_{Bosmina\text{-}SOM}$	8.81×10^{-7}	2.00	0.00	0.68
	Trend$_{Alona\text{-}SOM}$	0.44	2.00	1.41	0.07
	Trend$_{Chironomid\text{-}SOM}$	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$_{Bosmina\text{-}SOM}$	3.79×10^{-5}	5.00	0.00	0.54
	Trend$_{Alona\text{-}SOM}$	1.52	5.00	4.77	0.04
	Trend$_{Chironomid\text{-}SOM}$	6.95×10^{-5}	5.00	0.00	0.93
13N	Trend$_{SOM}$	9.58	9.00	23.06	8.96×10^{-7}
	Trend$_{Bosmina\text{-}SOM}$	4.97	5.00	1.13	0.13
	Trend$_{Alona\text{-}SOM}$	5.58×10^{-5}	5.00	0.00	0.33
	Trend$_{Chironomid\text{-}SOM}$	0.08	5.00	0.10	0.26

6E	$Trend_{SOM}$	2.55	9.00	54.96	6.86×10^{-14}
	$Trend_{Daphnia\text{-}SOM}$	0.00	5.00	0.00	0.32
	$Trend_{Bosmina\text{-}SOM}$	1.76	5.00	11.14	1.00×10^{-3}
	$Trend_{Alona\text{-}SOM}$	3.52	5.00	40.75	3.00×10^{-9}
	$Trend_{Chironomid\text{-}SOM}$	0.84	5.00	5.61	9.87×10^{-3}

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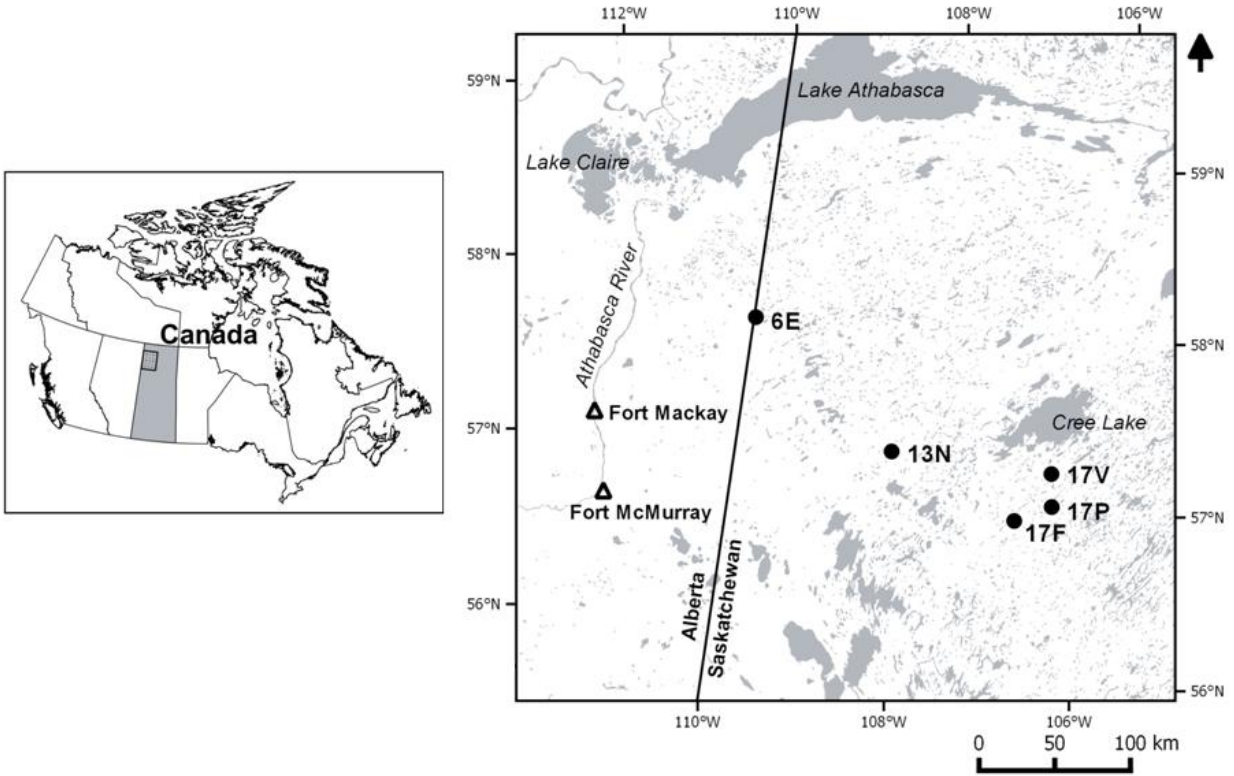
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867   Fig.1. Sampling domain and locations of five study lakes in northwest Saskatchewan.

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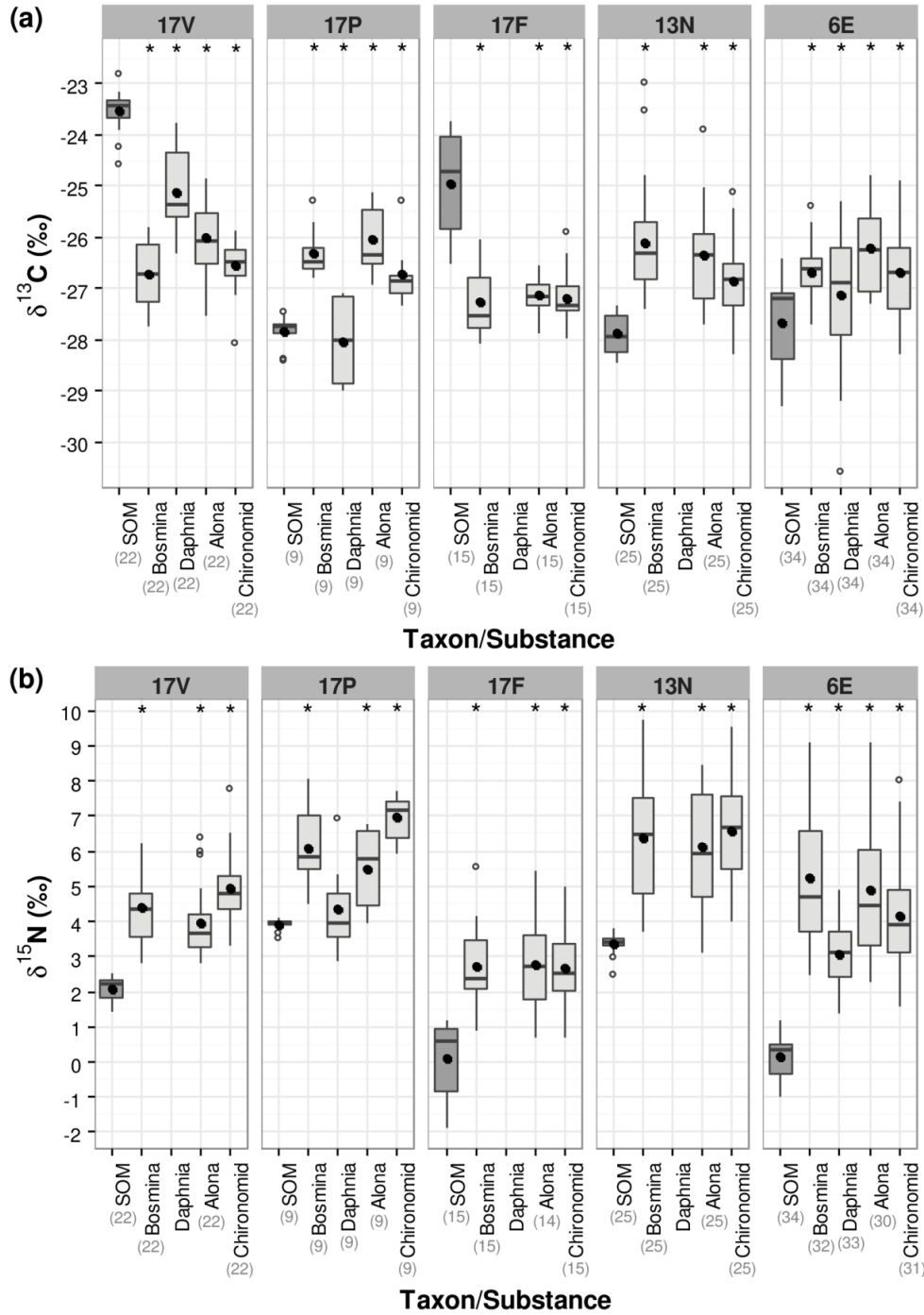
Fig.2. Comparison of isotopic values among sample types i.e. SOM and invertebrate taxa within study lakes. (a) Boxplots of $\delta^{13}C$ values of sample types. (b) Boxplots of $\delta^{15}N$ values of sample types. Solid circles (●) indicate the mean values and asterisks (*) denote the invertebrate taxa with significantly different ($p < 0.05$) mean values from that of SOM.
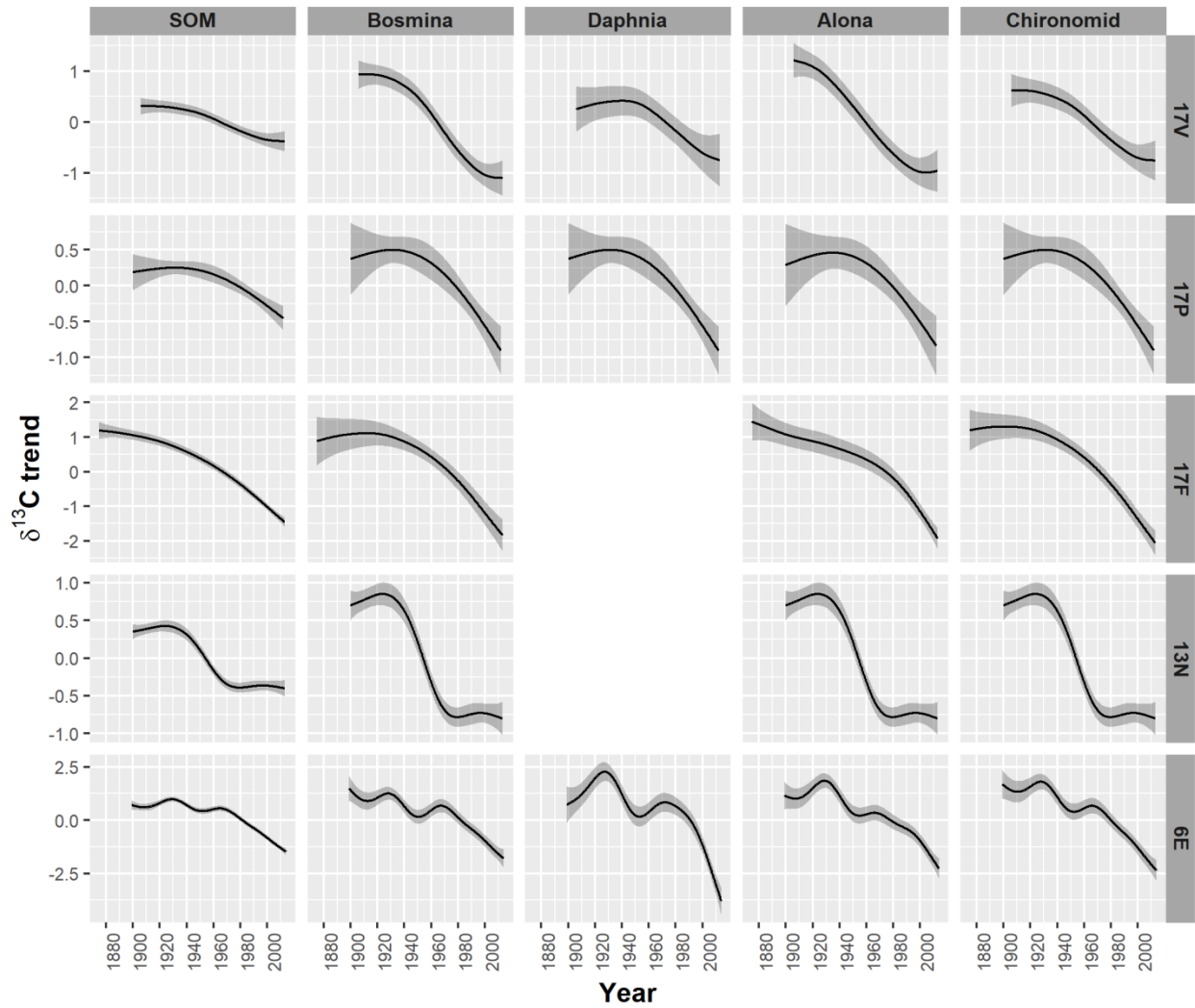
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Fig.3. δ^{13}C trends of SOM and invertebrate taxa of five study lakes. Solid lines denote the fitted

trends (smooth functions) and shaded regions represent the point-wise approximate 95%

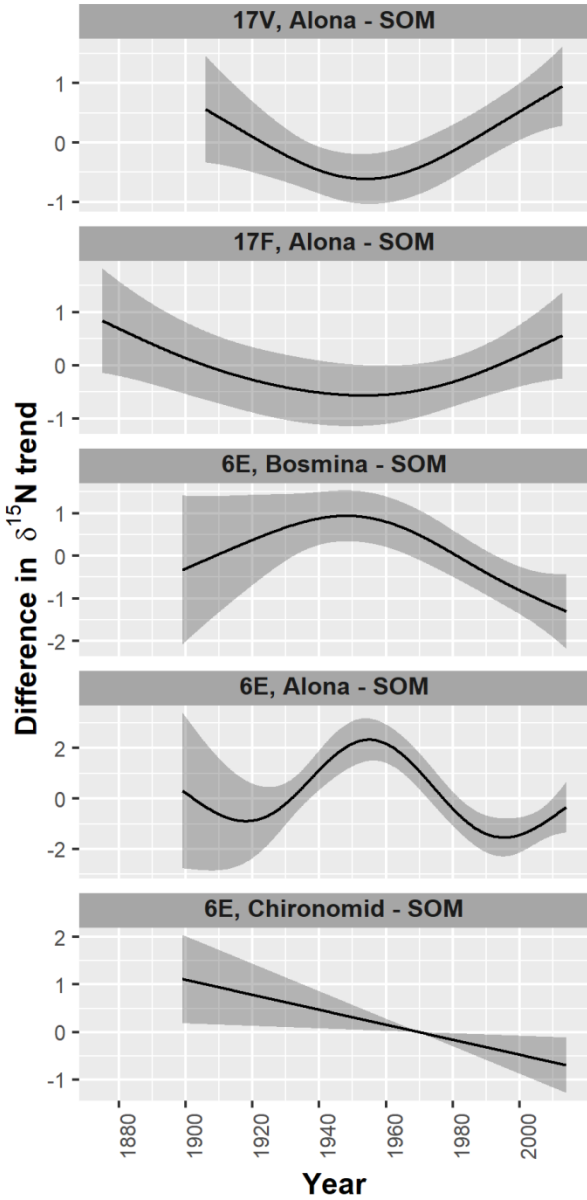confidence intervals. The columns represent sample types and the rows represent study lakes.

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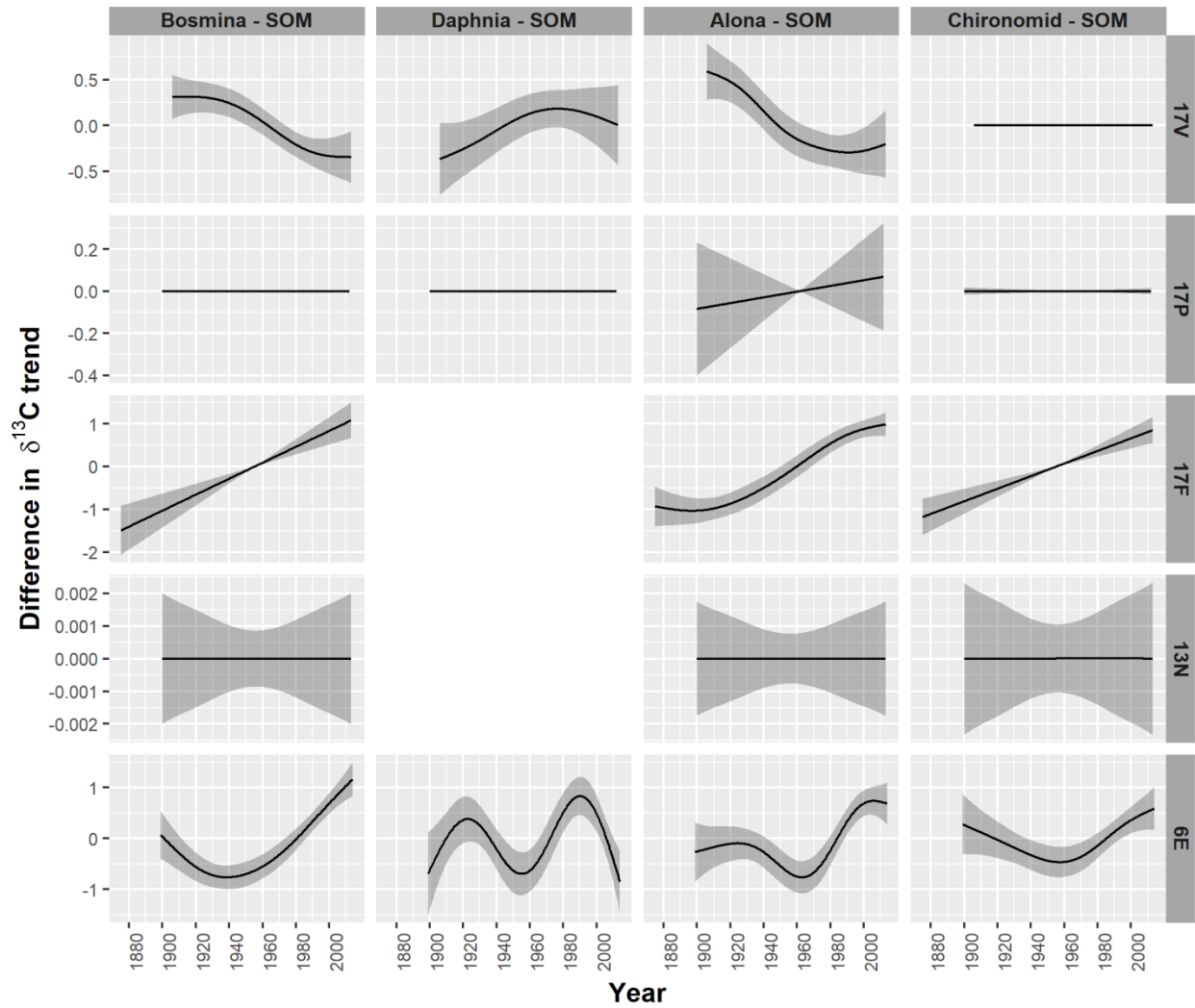
Fig.4. Within-lake differences in δ¹³C trends between invertebrate taxa and SOM. The columns

represent pair-wise comparisons and the rows represent study lakes. Solid lines denote the

estimated differences in trends (difference smooths) and shaded regions represent the point-wise

approximate 95% confidence intervals.

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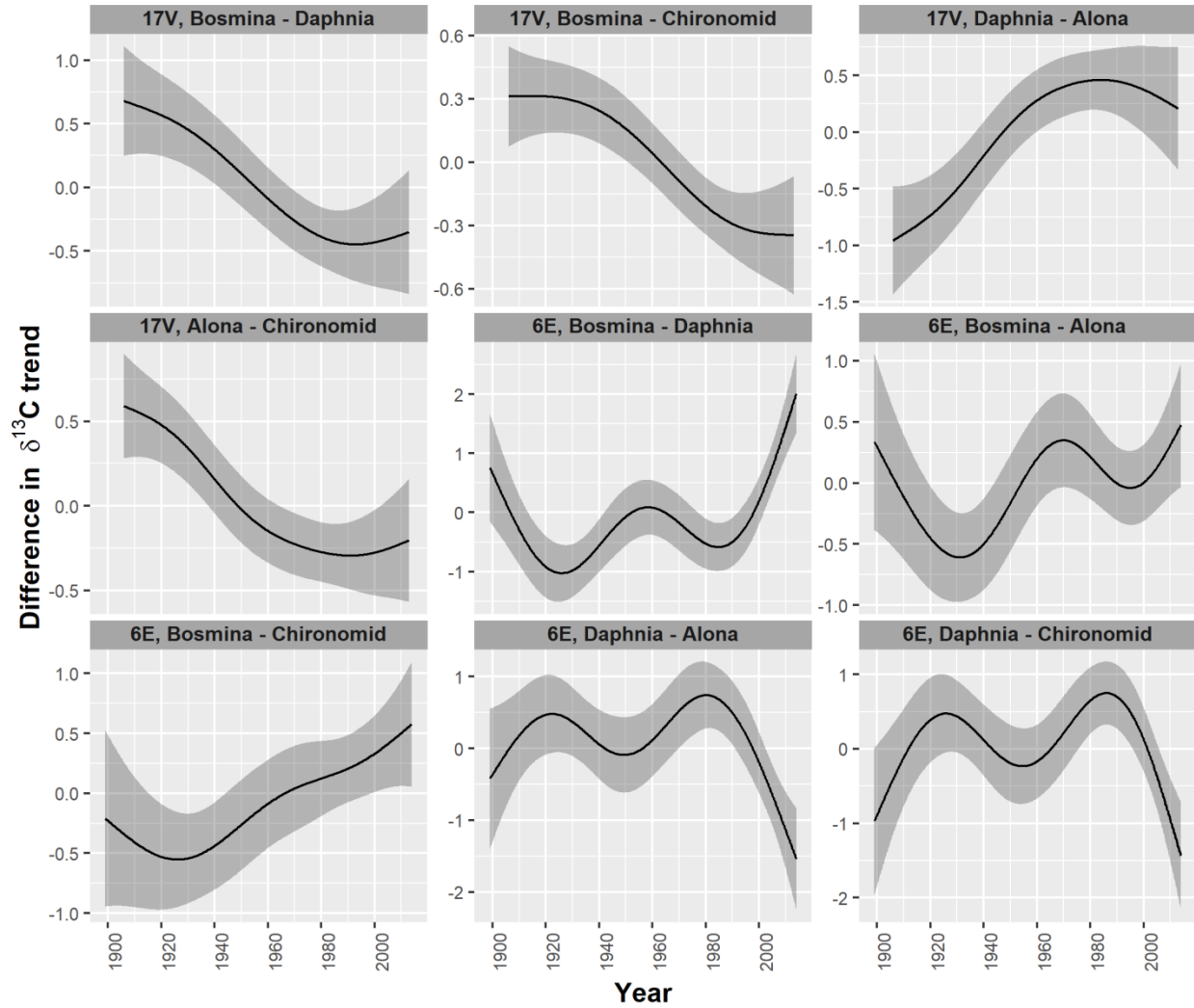
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Fig.5. Within-lake differences in δ^{13}C trends among invertebrate taxa. The columns represent

pair-wise comparisons and the rows represent study lakes. Solid lines denote the estimated

differences in trends and shaded regions represent the point-wise approximate 95% confidence

intervals.

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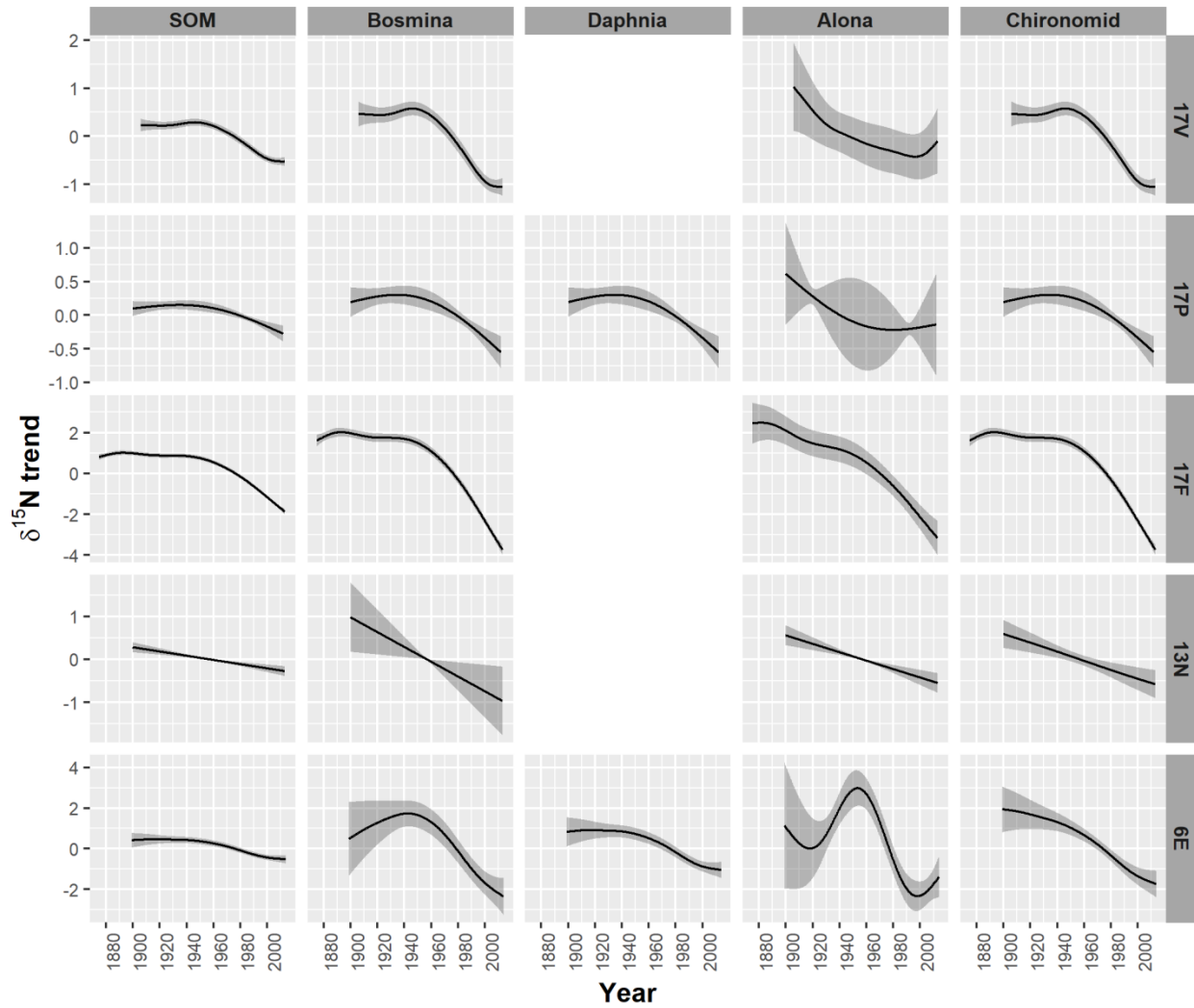
Fig.6. δ^{15}N trends of SOM and invertebrate taxa of five study lakes. Solid lines denote the fitted

trends (smooth functions) and shaded regions represent the point-wise approximate 95%

confidence intervals. The columns represent sample types and the rows represent study lakes.
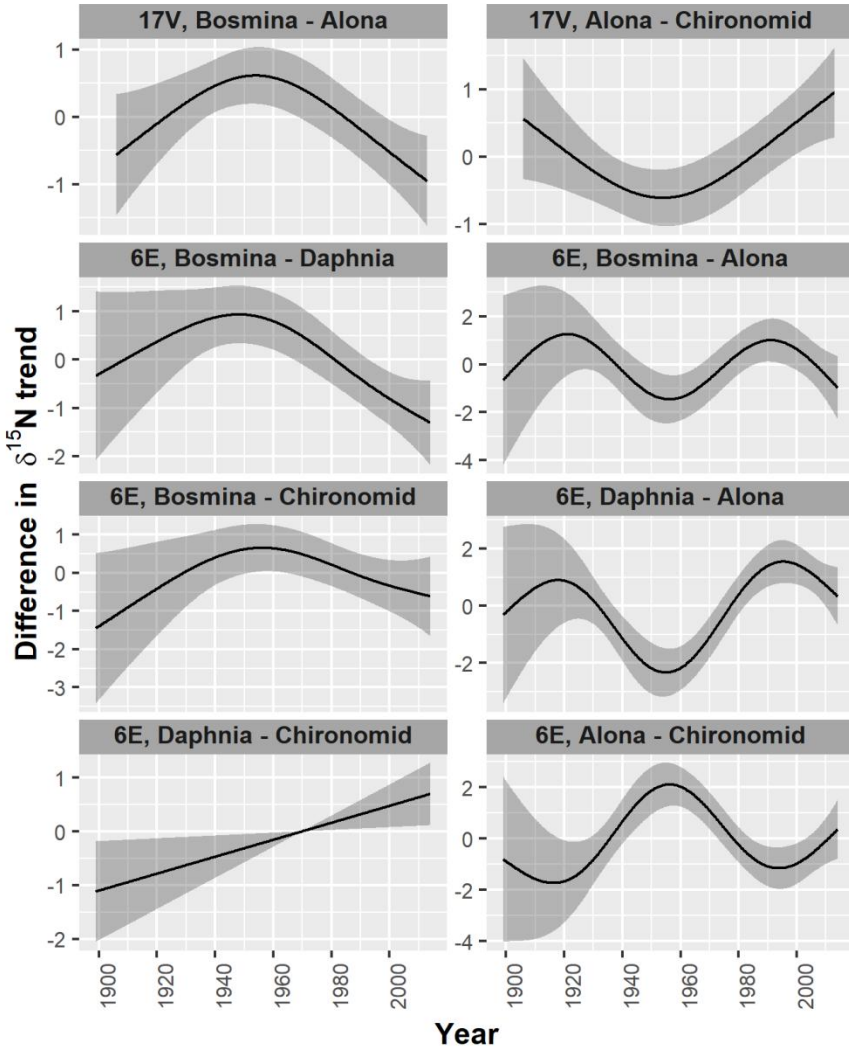
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Fig.7. Within-lake differences in δ^{15}N trends between invertebrate taxa and SOM. The columns

represent pair-wise comparisons and the rows represent study lakes. Solid lines denote the

estimated differences in trends (difference smooths) and shaded regions represent the point-wise

approximate 95% confidence intervals.

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Fig.8. Within-lake differences in δ^{15}N trends among invertebrate taxa. The columns represent

920   pair-wise comparisons and the rows represent study lakes. Solid lines denote the estimated

921   differences in trends and shaded regions represent the point-wise approximate 95% confidence

922   intervals.

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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. α_0, α_{1j}, γ_0, γ_{1j}, γ_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 $\hat{V}_\beta$. The standard errors were provided by the diagonal elements of;

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

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 $\delta^{13}C/\delta^{15}N$ 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 $\delta^{13}C$ of SOM among study lakes

Differences in mean $\delta^{13}C$ 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 $\delta^{13}C$ 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 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 $\delta^{13}C$ values can influence $\delta^{13}C$ 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 $\delta^{13}C$ values (Davidson and Jeppesen, 2013; Jones and Grey, 2011). Finally, post-depositional diagenetic effects may alter SOM $\delta^{13}C$ differently in lakes with varying SOM composition due to differential diagenesis of their SOM constituents (Meyers and Teranes, 2001).

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

Among-lake variability in mean $\delta^{15}N$ 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 $\delta^{15}N$ 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 δ^{15}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 δ^{15}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 δ^{15}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 δ^{15}N values in autochthonous organic matter relative to the nitrogen source. In lakes with limited DIN supply on the other hand, δ^{15}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 δ^{15}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* (Bleiwas 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 detrivorous chironomid larvae, preferential utilization of phyotoplankton 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 $\delta^{13}C$ and $\delta^{15}N$ 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).
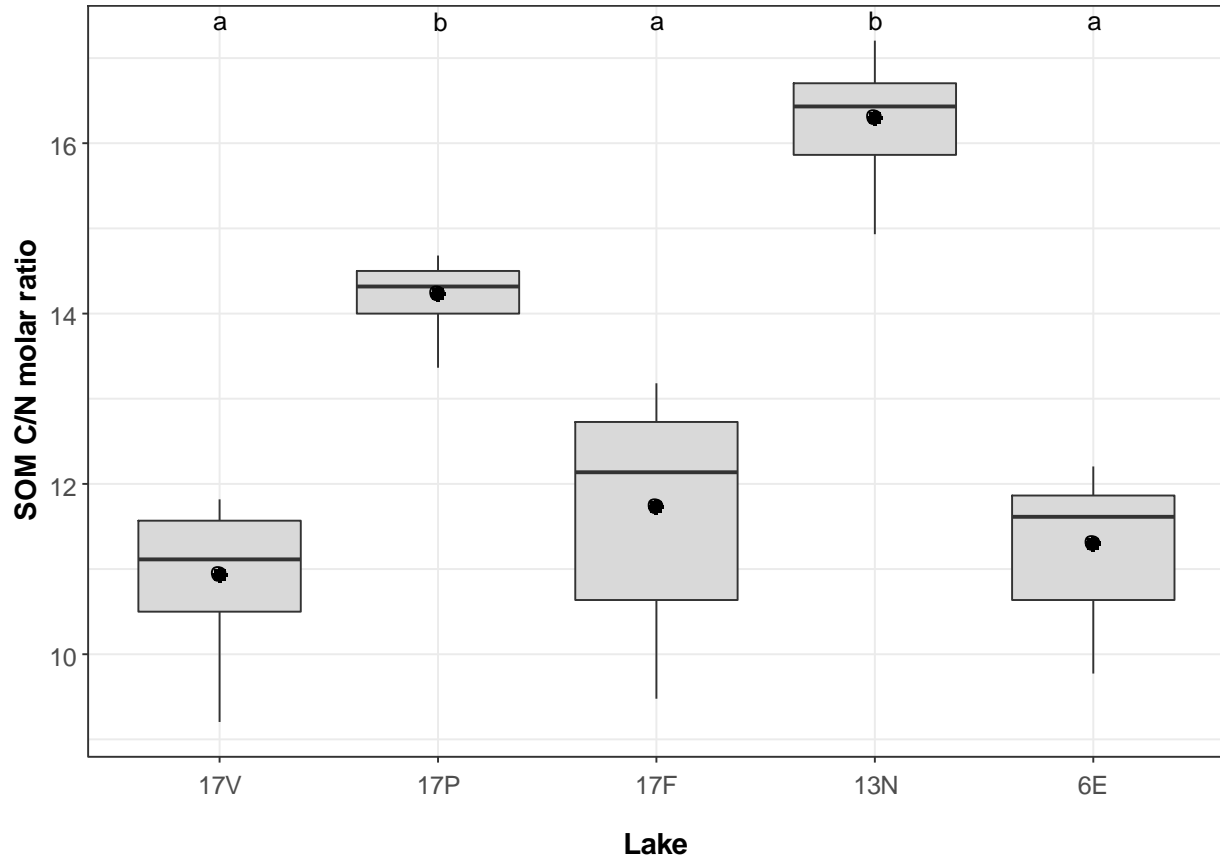
Fig.S1. Comparison of SOM C/N ratios among study lakes. Solid circles (●) indicate the mean

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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