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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

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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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7

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. 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 $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ 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 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 ($\delta^{13}\text{C}$) of invertebrate remains, although corrections for systematic changes in nitrogen SI ratios ($\delta^{15}\text{N}$) 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 $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ of bulk sedimentary organic matter (SOM) has been commonly incorporated into paleolimnological

studies. SOM is a heterogeneous mixture of plant-, animal- and microbial-derived organic matter originated in different habitats of the lake (i.e. littoral, pelagic and benthic) and received from the watershed (Meyers and Teranes, 2001). Consequently, accurate interpretation of the mechanisms underlying historical changes in isotopic signals of SOM can be difficult due to the potential for multiple pathways influencing both mass fluxes and isotopic value of source materials within the lake and its watershed (Meyers and Teranes, 2001; van Hardenbroek et al., 2014). In this regard, isotopic trends of sub-fossil invertebrate remains are considered to be better indicators of changes in C and N dynamics within the food webs than are those of SOM (Griffiths et al., 2010; Simon et al., 2017; van Hardenbroek et al., 2014), given *in situ* primary production likely contribute relatively little to total SOM relative to terrestrial organic matter (Von Wachenfeldt and Tranvik, 2008), and invertebrate consumers are thought to feed selectively on autochthonous sources (del Giorgio and France 1996; Meili et al. 1996; Doi et al. 2006). However, isotopic trends of sub-fossil invertebrate remains and SOM may not necessarily and universally be different. Not all invertebrate taxa are equally selective in their grazing (Demott, 1982; Tanentzap et al., 2017). Further, in systems where available basal resources are overwhelmingly dominated by allochthonous sources, invertebrates may be forced to feed less-selectively (Cole et al., 2011; Persaud et al., 2009; Tanentzap et al., 2017) and consequently, isotopic trends of invertebrate subfossils may not be different from that of SOM with predominant allochthonous origin (Simon et al., 2017). Therefore, differences in temporal patterns between SOM and invertebrate subfossils within each lake should be tested using powerful statistical methods. Yet, in past studies, differences in temporal patterns between SOM and invertebrates were inferred only from pair-wise comparisons of raw values without

statistically testing if temporal trends were significantly different (Kattel et al., 2015; Perga et al., 2010; van Hardenbroek et al., 2014).

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

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

2008; Zhang et al., 2010). In addition, these perturbations have also induced changes in nutrient dynamics and consequent changes in productivity in lakes (Dillon and Molot, 2005; Holtgrieve et al., 2011; Schindler and Lee, 2010). The nature and degree of responses likely vary among individual lakes depending on local characteristics including catchment properties, lake morphometry, type and degree of nutrient limitation, etc. (Anas et al., 2015; Zhang et al., 2010). These changes in turn may have variably influenced energy and nutrient fluxes through different invertebrate taxa with different habitat or feeding niches within lakes. For instance, changes in allochthonous DOM inputs may strongly influence the diet of non-selective feeders relative to selective feeders of phytoplankton (Persaud et al., 2009; Tanentzap et al., 2017). Further, the diet of littoral consumers can be affected more than that of pelagic consumers by a shift from predominant benthic production to a pelagic regime following eutrophication (Vadeboncoeur et al., 2003). However, such historical food-web consequences of environmental perturbations in Boreal Shield lakes is poorly understood (Persaud et al., 2009), while SI analysis of subfossil invertebrate remains provides a potential means for evaluating these long-term temporal trophodynamics of individual taxa (Frossard et al., 2014; Perga, 2009; van Hardenbroek et al., 2014).

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

likely with different feeding modes) i.e. *Daphnia* and *Bosmina* spp. (pelagic), *Alona* sp. (littoral) and Chironomidae (benthic) and statistically compared the isotopic trajectories (i) between SOM and invertebrate taxa, and (ii) among invertebrate taxa within each lake. We hypothesized that isotopic trends between invertebrate subfossils reflective of food web-specific dynamics would be significantly different from SOM which derives from heterogeneous sources, yet such differences would be system- and/or taxon-specific. Further, we expected that SI trends would be significantly different among invertebrate taxa representing different habitat or feeding niches due to variable food-web related effects of above-mentioned environmental perturbations occurring in Boreal Shield region. We also expected that among-taxon differences in SI trends would be system-specific as different lakes may have responded differently to environmental perturbations due to differences in local characteristics. To our knowledge, this is the first study where taxon-specific SI analyses of invertebrate remains has been performed on multiple study lakes, and in which robust statistical approaches were applied to compare isotopic trends of invertebrate taxa and SOM.

2. Methods

2.1. Study lakes

Our five study lakes were a subset of 16 lakes in northwest Saskatchewan (Fig. 1) described in Mushet et al. (2017), that represent 262 basins surveyed by the provincial government during 2007-2009 and 2011 (Scott et al., 2010). Initially, sites were selected to represent type of nutrient limitation i.e. phosphorus-limited vs. nitrogen-limited (based on molar dissolved inorganic nitrogen to total phosphorus [DIN: TP] ratios) and degree of vulnerability to atmospheric emissions from AOSR (based on distance from the industrial center) (Table 1). Of

the 16 lakes cored, the five lakes examined here were chosen on the basis of their elevated densities of fossil invertebrate remains (Hesjedal, 2017) which allowed sufficient sample size to conduct taxon-specific SI analyses. Final lake selection included two N-limited lakes (basins 17V, 6E) and three phosphorus-limited lakes (17P, 17F, 13N).

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

2.2. Sediment coring

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

2.3. Core chronologies

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

2.4. SI analysis of SOM

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

2.5. SI analysis of subfossil invertebrate remains

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

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

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

2.6. Data analysis

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

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

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

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

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

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

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

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

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

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

3. Results

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

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

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

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

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

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

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

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

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

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

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

4. Discussion

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

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

4.1. SI variability of SOM and invertebrate remains

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

study lakes ($< 2\text{‰}$) (Fig.2) indicate fixed N is the main N source (Meyers and Teranes, 2001; Patoine et al., 2006), which is likely derived from terrestrial sources (e.g. plants such as *Alnus* spp. and *Shepherdia canadensis* and microorganisms in deciduous forest) (Bunting et al., 2010; Engstrom and Fritz, 2006), as pigments for N_2 -fixing cyanobacteria were not detected in sediments of these lakes (Wolfe, 2016). Among-lake differences in SOM $\delta^{15}\text{N}$ on the other hand can arise due to differences in the mass flux or isotopic ratios of fixed DOM-N as well as subsequent modification by biological (terrestrial and in-lake) processes (Supplementary discussion; Supplementary data).

The $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values of *Bosmina* exoskeletons (28.1‰ to -23‰ and 0.9 to 9.7‰, respectively) and *Daphnia ephippia* (-30.6‰ to -23.5‰ and -1.4 to 9‰ respectively) recorded in the present study were similar to contemporary values recorded for these taxa in a synoptic survey of 233 regional boreal lakes (*Bosmina* -31.4 to -22.9‰ and -1.5 to 9‰, respectively; *Daphnia* -35.8 to -24.3‰ and 0.5 to 7.8‰, respectively) (Anas, 2019). This supports the high likelihood that downcore isotopic variations in invertebrate sub-fossils are within the expected range in response to regional environmental variations such as fluctuations in primary productivity, allochthonous organic matter inputs, in-lake biogeochemical transformations and catchment processes (Anas, 2019). Elsewhere highly negative $\delta^{13}\text{C}$ values (~ -50 to -40‰) of subfossil chironomid head capsules and *Daphnia ephippia* may reflect contributions of ^{13}C -depleted methanogenic carbon to invertebrate diets (Frossard et al., 2013b; Rinta et al., 2016; Schilder et al., 2017; van Hardenbroek et al., 2013; Wooller et al., 2012), a factor which does not seem to be important in our study (Fig. 2). Instead, invertebrate $\delta^{13}\text{C}$ values herein were usually within the range expected for photoautotrophically-produced C in aquatic and terrestrial ecosystems (-35 to -8‰) (France, 1996, 1995b; Meyers and Teranes, 2001; Vuorio et al., 2006).

343

344 4.2. *SI trends of SOM*

345 Patterns of sedimentary $\delta^{13}\text{C}$ 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 $\delta^{15}\text{N}$ 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).

363 4.3. *Differences in $\delta^{13}\text{C}$ time series between invertebrate remains and SOM*

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

Significant differences between $\delta^{13}\text{C}$ trends of SOM and invertebrates from different habitats within lakes 17V, 17F and 6E suggest that analysis of taxon-specific SI can provide new insights on the regulation of food-web carbon dynamics of different lake habitats. For instance, variation in phytoplankton $\delta^{13}\text{C}$ due to changes in primary productivity, DIC content or DIC

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

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

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

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

The significant among-taxon differences in $\delta^{13}\text{C}$ trends of lakes 17V and 6E indicate that flux of carbon through invertebrates can differ among taxa, habitats and lakes, and that all pathways may have decoupled through time (Fig.5). Dietary sources of C for invertebrates, as well as the associated $\delta^{13}\text{C}$ values of C source can be impacted by habitat-specific or whole-lake environmental changes (Persaud et al., 2009; Vadeboncoeur et al., 2003; van Hardenbroek et al., 2014). In particular, changes in relative availability of different carbon sources (i.e. phytoplankton, epiphyton, terrestrial, methane-derived carbon) can be induced by alteration in water transparency by primary producers (Solomon et al., 2008; Vadeboncoeur et al., 2003), physical turbidity (Scheffer et al., 1993) or DOC influx (Carpenter et al., 1988), leading to shifts in the diets of primary consumers (Solomon et al., 2008; Vadeboncoeur et al., 2003). For instance, a switch from predominantly benthic production to a pelagic regime following eutrophication can shift the diet of littoral consumers (e.g. *Alona*) from ^{13}C -enriched epiphyton ((Hecky and Hesslein, 1995) to ^{13}C -depleted phytoplankton and suspended detritus, while diet of pelagic consumers (e.g., *Bosmina*, *Daphnia*) remain unaffected (Vadeboncoeur et al., 2003). Further, $\delta^{13}\text{C}$ of littoral epiphyton may respond differently compared to pelagic phytoplankton to temporal changes in DIC content and dominant chemical form of inorganic C (CO_2 , HCO_3^- , CO_3^{2-}) due to boundary layer effects (Hecky and Hesslein, 1995; McPherson et al., 2015;

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

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

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

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

Significant differences between $\delta^{15}\text{N}$ trends of SOM and invertebrate subfossils in lakes 17V, 17F and 6E (Fig. 7) may have arisen from shifts in dietary sources of N and/or changes in magnitude of isotopic fractionation in invertebrate consumers. For example, $\delta^{15}\text{N}$ of invertebrate consumers can covary with bacterial contribution to the diet (Grey et al., 2004, 2001; Karlsson et al., 2004). Empirical evidence suggests that bacterial $\delta^{15}\text{N}$ may differ from other basal resources either due to elevated isotopic fractionation depending on biochemical composition and availability of nitrogen substrate (McGoldrick et al., 2008; Steffan et al., 2015) or assimilation of isotopically light nitrogen substrate (e.g. excreted ammonia) (Grey et al., 2004). In addition, few

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

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

The presence of significant among-taxon differences in $\delta^{15}\text{N}$ trends in lakes 17V and 6E suggests that trophic relationships among taxa may vary in lakes at centennial scales (Fig. 8). As noted above, variable offsets in $\delta^{15}\text{N}$ among pairs of species may result from habitat-specific changes in resource materials with different $\delta^{15}\text{N}$ values, resource limitation, or changes in metabolic processes of individual taxa. In addition, lake-specific changes in the influx of nitrogenous materials may also influence the degree of agreement in $\delta^{15}\text{N}$ trends between pairs

of species, particularly from different habitats. In particular, the $\delta^{15}\text{N}$ of individual taxa may be selectively influenced by dietary shifts or changes in isotopic fractionation while the $\delta^{15}\text{N}$ of the other taxon remains unaffected (e.g. *Alona* vs. *Bosmina* in 17V). Alternately, $\delta^{15}\text{N}$ variations of both taxa can be influenced by environmental and physiological mechanisms simultaneously, asynchronously or differentially (e.g. *Alona* vs. *Bosmina* in 6E), as has been seen in modern lake time series (Patoine et al. 2006). Regardless of the underlying mechanism, the observation that historical differences between pairs of common taxa (e.g., *Bosmina* v. *Alona*) vary among lakes (17V, 6E) suggests the possibility of lake-specific food web responses to environmental changes during the past century.

again indicating the consistent importance of allochthonous DOM as food web N source in all habitats and feeding niches.

4.7. Caveats

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

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

5. Conclusions

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

invertebrate remains are related to changes in aquatic primary productivity (Hayes et al., 1987).
Based on our results, we suggest that evaluation of isotopic trends of invertebrate remains
relative to SOM, will be a particularly informative means of unraveling past food-web carbon,
nutrient and trophic dynamics.

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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 ($\mu\text{g L}^{-1}$)	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	8.34

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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 $\delta^{13}\text{C}$ 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 _{<i>Alona</i>-SOM}	1.90	5.00	18.60	1.32×10^{-5}
	Trend _{<i>Chironomid</i>-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 _{<i>Daphnia</i>-SOM}	6.44×10^{-6}	2.00	0.00	0.66
	Trend _{<i>Bosmina</i>-SOM}	2.60×10^{-5}	2.00	0.00	0.37
	Trend _{<i>Alona</i>-SOM}	0.13	2.00	0.30	0.13
	Trend _{<i>Chironomid</i>-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 _{<i>Bosmina</i>-SOM}	0.96	4.00	27.42	9.49×10^{-8}
	Trend _{<i>Alona</i>-SOM}	2.21	4.00	87.59	$< 2.00 \times 10^{-16}$
	Trend _{<i>Chironomid</i>-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 _{<i>Bosmina</i>-SOM}	2.47×10^{-5}	9.00	0.00	0.62
	Trend _{<i>Alona</i>-SOM}	6.57×10^{-6}	9.00	0.00	0.62
	Trend _{<i>Chironomid</i>-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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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	<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 _{<i>Alona</i>-SOM}	1.84	5.00	9.96	2.89×10^{-3}
	Trend _{<i>Chironomid</i>-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 _{<i>Daphnia</i>-SOM}	5.30×10^{-7}	2.00	0.00	1.00
	Trend _{<i>Bosmina</i>-SOM}	8.81×10^{-7}	2.00	0.00	0.68
	Trend _{<i>Alona</i>-SOM}	0.44	2.00	1.41	0.07
	Trend _{<i>Chironomid</i>-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 _{<i>Bosmina</i>-SOM}	3.79×10^{-5}	5.00	0.00	0.54
	Trend _{<i>Alona</i>-SOM}	1.52	5.00	4.77	0.04
	Trend _{<i>Chironomid</i>-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 _{<i>Bosmina</i>-SOM}	4.97	5.00	1.13	0.13
	Trend _{<i>Alona</i>-SOM}	5.58×10^{-5}	5.00	0.00	0.33
	Trend _{<i>Chironomid</i>-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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Fig.1. Sampling domain and locations of five study lakes in northwest Saskatchewan.

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

Fig.3. $\delta^{13}\text{C}$ 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.

Fig.4. Within-lake differences in $\delta^{13}\text{C}$ 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.

Fig.5. Within-lake differences in $\delta^{13}\text{C}$ 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.

Fig.6. $\delta^{15}\text{N}$ 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.

Fig.7. Within-lake differences in $\delta^{15}\text{N}$ 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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919 Fig.8. Within-lake differences in $\delta^{15}\text{N}$ 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. $\alpha_0, \alpha_{1j}, \gamma_0, \gamma_{1j}, \gamma_2$ 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 $\hat{V}_{\beta}$. 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 $\delta^{13}\text{C}/\delta^{15}\text{N}$ 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 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* (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 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. 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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