

1 Terrestrial food sources matter for mercury
2 bioaccumulation in zooplankton and
3 macroinvertebrates in lakes with differing dissolved
4 organic carbon concentrations

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18

19 **Abstract**

20 Dietary uptake is a key step in conveying both toxic mercury (Hg; particularly as highly
21 bioavailable methylmercury, MeHg) and essential dietary biochemicals, such as polyunsaturated fatty
22 acids (PUFA), across trophic levels within aquatic food webs. Using stable isotopes and fatty acids we
23 evaluated the role of food sources in size-fractionated plankton and littoral macroinvertebrates for the
24 bioaccumulation of total Hg and MeHg in six oligotrophic and one mesotrophic Swedish lakes with
25 differing concentrations of dissolved organic carbon (DOC). We found that the consumption of both
26 algal and terrestrial diets (assessed by PUFA and long-chain saturated fatty acids, respectively)
27 predicted >66% of the Hg concentration variability in meso- (100-500 μm) and macrozooplankton
28 (>500 μm) in oligotrophic lakes. In the mesotrophic lake, total Hg bioaccumulation in higher trophic
29 level biota, carnivorous macroinvertebrates was also significantly related to terrestrial diet sources
30 ($R^2=0.65$, $p<0.01$). However, lake pH and DOC correlated to total Hg bioaccumulation and
31 bioconcentration across all lakes, suggesting the consumption of different diet sources is mediated by
32 the influence of lake characteristics. This field study reveals that using dietary biomarkers (stable
33 isotopes and fatty acids) together with the physico-chemical lake parameters pH and nutrients together
34 improve our ability to predict Hg bioaccumulation in aquatic food webs. Fatty acids used as dietary
35 biomarkers provide correlative evidence of specific diet source retention in consumers and their effect
36 on Hg bioaccumulation, while pH and nutrients are the underlying physico-chemical lake parameters
37 controlling differences in Hg bioaccumulation between lakes.

38

39 **Keywords**

40 Mercury bioaccumulation, methylmercury, fatty acids, stable isotopes, plankton, macroinvertebrates

41

42 **Introduction**

43 High degrees of mercury (Hg) bioaccumulation is common in boreal freshwater lakes
44 (Lindqvist *et al.*, 1991; Munthe *et al.*, 2007), especially in lakes that are small (a few hectares), low in
45 productivity, acidic, and high in dissolved organic carbon (DOC) (e.g., 5-10 mg L^{-1} and even higher)

46 (Sobek *et al.*, 2007; Clayden *et al.*, 2014; Soerensen *et al.*, 2017). High DOC or total organic carbon
47 (TOC) in boreal freshwater lakes usually relates to terrestrial organic matter (OM) (Gadmar *et al.*,
48 2002; Monteith *et al.*, 2007). In small, acidic, and nutrient-poor boreal lakes with high terrestrial OM
49 input (Jansson *et al.*, 2001; Seekell *et al.*, 2015), Hg from soils can be mobilized to lakes due to strong
50 divalent Hg binding to OM (Skylberg *et al.*, 2000; Skylberg *et al.*, 2006). For such lakes with
51 nitrogen limitation, an increase in nitrogen (e.g. after forestry activities, atmospheric nitrogen
52 deposition) will generally increase phytoplankton biomass (Bergström *et al.*, 2005), which can in turn
53 bio-dilute Hg concentration in lake biota (Pickhardt *et al.*, 2002; Karimi *et al.*, 2007). On the other
54 hand, acidic waters with high DOC often have higher concentrations of the highly bioavailable
55 methylmercury (MeHg). Thus, these lakes are generally more prone to facilitate Hg bioaccumulation
56 compared to less acid and/or humic lakes (Edmonds *et al.*, 2012; Clayden *et al.*, 2014). Increasing
57 DOC concentrations derived from terrestrial sources can themselves potentially increase Hg
58 bioaccumulation in estuarine ecosystem food webs (French *et al.*, 2014; Jonsson *et al.*, 2014; Jonsson
59 *et al.*, 2017). This is also relevant in boreal ecosystems as common forest harvest activities can
60 enhance terrestrial OM input to lake (Meunier *et al.*, 2016), resulting in increased DOC concentrations
61 (O'Driscoll *et al.*, 2006; Glaz *et al.*, 2015), and sometimes higher Hg and/or MeHg input from
62 terrestrial to aquatic ecosystems (Eklöf *et al.*, 2013; Kronberg *et al.*, 2016). This can increase Hg
63 bioaccumulation in food webs (Bishop *et al.*, 2009; Hongve *et al.*, 2012; Wu *et al.*, 2018).

64 The effect of dietary terrestrial OM uptake on Hg bioaccumulation in lake food webs,
65 however, is not well understood. The utilization of terrestrial OM by zooplankton for somatic growth
66 and reproduction have been highlighted in several studies (Brett *et al.*, 2009; Cole *et al.*, 2011;
67 McMeans *et al.*, 2015), but the trophic link between terrestrial OM retained in zooplankton and its role
68 for Hg bioaccumulation remains to be resolved.

69 Food sources vary among ecosystems with different environmental conditions and can affect
70 Hg bioaccumulation (Minshall, 1967; Ward *et al.*, 2010). There is evidence from stable isotopes that
71 zooplankton and macroinvertebrates obtain their dietary energy, to various degrees, from
72 allochthonous or autochthonous sources (Hessen *et al.*, 1989; Junger & Planas, 1993; del Giorgio &
73 France, 1996). However, stable isotope analysis has difficulty in identifying source-specific dietary

74 pathways that convey allochthonous and autochthonous C to zooplankton (Carpenter *et al.*, 2005). The
75 same holds true for Hg (Cremona *et al.*, 2009).

76 Fatty acids (FA) are informative dietary source indicators that can help us further understand
77 dietary pathways. Various FA molecules are unique to algae (e.g., polyunsaturated fatty acids, PUFA;
78 Napolitano (1999)), bacteria (e.g., odd and/or branched chain fatty acids derived from bacteria, BFA),
79 or terrestrial-origin organic matter (e.g., long-chain saturated fatty acids, terr. FA) (Kainz *et al.*, 2002;
80 Kainz *et al.*, 2003; Kainz & Fisk, 2009). As such, FA have been used as tracers of dietary sources to
81 examine the bioaccumulation of persistent organic pollutants (POPs) in aquatic food webs
82 (Swackhamer & Skoglund, 1993; Fisk *et al.*, 2001; Lynn *et al.*, 2007). But often the bioaccumulation
83 patterns of toxic MeHg (Lavoie *et al.*, 2013) and FA distribution (Syvaranta & Rautio, 2010) in food
84 webs have been studied separately. Few studies have evaluated underlying links between MeHg and
85 FA compositions simultaneously, particularly at lower trophic levels such as phytoplankton. In
86 particular, the relationship of PUFA (e.g., eicosapentaenoic acid or EPA) with MeHg bioaccumulation
87 in aquatic food webs remains unclear, despite the fact that these particular FA might be helpful as
88 dietary biomarkers to link growth with the cycling of an environmental pollutant such as MeHg (Kainz
89 *et al.*, 2008).

90 The combination of stable isotopes and FA is a valuable tool for better understanding complex
91 resource-consumer relationships (Perga *et al.*, 2006; Lau *et al.*, 2014), as well as elucidating
92 environmental pollutant trajectories from dietary sources (Kainz *et al.*, 2008). Yet, very few studies
93 have utilized the combined dietary biomarkers of stable isotopes and FA analyses in freshwater lakes,
94 and fewer have gone on to relate dietary sources with Hg bioaccumulation in freshwater food webs.
95 This is because diet sources are more challenging to identify in zooplankton due to difficulties in
96 collecting sufficient biomass for quantitative analysis as compared to fishes.

97 In order to fill this gap of knowledge, the aim of this field study was to assess how trophic
98 transfer of Hg and MeHg relates to specific dietary sources in pelagic and littoral food webs of boreal
99 lakes with different DOC concentrations. More specifically, the study investigated whether size-
100 fractionated zooplankton (mesozooplankton, 100-500 μm ; and macrozooplankton, $>500 \mu\text{m}$) and littoral
101 macroinvertebrates selectively retain specific FA compounds while simultaneously accumulating Hg

102 in boreal freshwater lakes. We selected seven boreal lakes along a DOC gradient, with varying land
103 use activities (clear-cut or whole lake nitrogen fertilization), to explore how total Hg and MeHg in
104 these food webs respond to changes in physico-chemical conditions and eventual differences in dietary
105 composition. We hypothesized that increased DOC and inorganic nitrogen, caused by forest clear-cut
106 or nitrogen addition (Deininger *et al.*, 2017a), will result in lower Hg bioaccumulation in zooplankton
107 and macroinvertebrates. This is expected to be caused by Hg bio-dilution linked with increasing
108 phytoplankton biomass growth that supports somatic growth of zooplankton (Karimi *et al.*, 2007) and
109 therefore decreases Hg contents in somatic consumer biomass.

110

111 **Materials and Methods**

112 **1. Sampling**

113 Seven freshwater lakes were sampled, six of which were oligotrophic lakes located close to
114 each other in northern Sweden in the Västerbotten region (64.12 to 64.25° N, 18.76 to 18.80° E)
115 (Table 1). One lake was mesotrophic and located further south, in eastern Sweden within the
116 Stockholm region (59.8458° N, 18.5649° E). The lakes were selected along a DOC concentration
117 gradient to represent the typical variety of mostly unproductive lakes in the Swedish boreal landscape
118 (Meili *et al.*, 1991; Sobek *et al.*, 2007). The selection of Lake Erken, which is the only mesotrophic
119 lake in this study, allowed us to compare the difference in Hg and FA bioaccumulation under different
120 trophic conditions. The catchment areas, particularly those situated in the Västerbotten region, consist
121 mainly of coniferous forest (*Picea abies*, *Pinus sylvestris*) and open *Sphagnum* dominated mires.

122 Water, phytoplankton, zooplankton and macroinvertebrates samples were taken at the same
123 time in the summer of 2015. Based on previous observations that MeHg increases with increasing
124 plankton size (Kainz & Mazumder, 2005), we size-fractionated both phytoplankton (microplankton, 25-
125 100 µm; seston, <25 µm) and zooplankton (macrozooplankton, >500 µm; mesozooplankton, 100-500
126 µm). Different size fractions of plankton samples were taken by vertical hauls from the bottom of the
127 lakes (hypolimnion, i.e., ~1 m above lake sediment) to the lake surface (epilimnion, i.e., top 0.5~1 m)
128 of the lake pelagic zone with a water sampler and filtered through non-metallic plankton nets of

129 different mesh sizes (500 μm , 100 μm , and 25 μm). These collected phyto- and zooplankton samples
130 collected on the nets were transferred to TeflonTM FEP bottles immediately stored on dry ice within 15
131 min and then moved to a -80°C freezer before being freeze-dried at -20°C. Additional plankton
132 samples were collected for plankton community analysis and stored in polypropylene vials at 4°C
133 before being transported for zooplankton and phytoplankton taxonomical analysis.

134 Littoral macroinvertebrates were retained on nets with different mesh size (0.5 mm and 0.25
135 mm) by kick-sampling method (kicking littoral substrate for a few times while placing the mesh net in
136 the direction of flow) in shallow stony and/or sandy habitats (0-1 m depth) (Davies, 2001). Major
137 groups of littoral macroinvertebrates collected for this study included insects (Coleoptera, Diptera, and
138 Odonata) and shellfish (Bivalvia and Gastropoda) (Supporting Information (SI) Table S1). We
139 classified the macroinvertebrates into functional feeding groups (FFG), as collectors: Diptera,
140 Gastropoda and Bivalvia; shredders: Amphipoda and a few Megaloptera and Trichoptera species; and
141 predators: carnivorous Coleoptera, Odonata, Hemiptera, Megaloptera, and Trichoptera (Cummins &
142 Klug, 1979). The sorted samples were stored at -20°C before freeze-drying. These were then
143 homogenized separately by identified taxa using an agate mortar and pestle for further chemical
144 analysis.

145

146 **2. Analysis**

147 **Water chemistry analysis**

148 Total nitrogen (totN), total phosphorus (totP), dissolved organic carbon (DOC), and
149 chlorophyll *a* (Chl *a*) and pH in lake water samples were analyzed at the Geochemical Lab at
150 Department of Aquatic Sciences and Assessment (Swedish University of Agricultural Sciences,
151 Uppsala) according to accredited practices and analytical methods accredited (Swedish Board for
152 Accreditation/Conformity Assessment; SWEDAC). Details of the above-mentioned analytical
153 methods are included in the supporting information (SI Table S2).

154

155 **Plankton and macroinvertebrate taxonomy identification**

156 The phytoplankton samples (plankton size fraction, seston: < 25 μm ; microplankton: 25-100
157 μm) were fixed with Lugol, and variable volume (5 to 50 μL) was settled following the Utermöhl
158 method for their taxonomic composition analysis (Utermöhl, 1958). Samples were counted on an
159 inverted microscope (Leica DMI 3000 B) and at least 400 cells were identified to the genus level.
160 Phytoplankton biovolumes were assigned using reference data (Kremer *et al.*, 2014). The
161 identification of zooplankton and macroinvertebrates was carried out by SWEDAC accredited
162 Biodiversity Lab at Department of Aquatic Sciences and Assessment (Swedish University of
163 Agricultural Sciences, Uppsala). The zooplankton samples (mesoplankton (100-500 μm) and
164 macroplankton (> 500 μm)) were counted (75x magnification) and taxonomically identified to species
165 (150x magnification) using a stereo microscope (Nikon Eclipse Ti-U). Macroinvertebrates were
166 taxonomically identified using a stereo microscope (Nikon SMZ1000; using 6x to 100x magnification)
167 by sorting individuals of abundant taxa to the lowest possible taxonomic unit (i.e. species, genus, or, in
168 some cases, family or order level) upon collection on ice.

169

170 **Total mercury and methylmercury analysis**

171 Total Hg and MeHg concentrations in water were analyzed by the Swedish Environmental
172 Institute (IVL). The MeHg concentration in water was determined by CVAFS, following US EPA
173 1630 method (USEPA, 1998), with a detection limit of 0.02 ng L^{-1} and a quantitation limit of 0.06 ng
174 L^{-1} . The total Hg concentration in water was also identified by CVAFS, but following US EPA 1631
175 method (USEPA, 2002) and performed by IVL. The method has a detection limit of 0.05 ng L^{-1} and a
176 quantitation limit of 0.1 ng L^{-1} .

177 Total Hg and MeHg concentrations of freeze-dried plankton in different size-fractions and
178 macroinvertebrates were measured by Analytical Services at the Biotron Centre for Experimental
179 Climate Change Research at Western University in London, Canada. Both the total Hg concentrations
180 ([THg]) in plankton and macroinvertebrate samples were analyzed with a DMA-80 Total Mercury
181 Analyzer (Milestone Srl) employing US EPA method 7473 (USEPA, 2007), or solid-sampling thermal
182 decomposition amalgamation atomic absorption spectrometry (TDA AAS). The equipment's method
183 detection limit was documented as low as 0.1 ng . The precision of measurement from replicate

184 analyses was greater than 80%. Samples, blanks, CRMs (DORM-2 with [THg] of $4.64 \pm 0.26 \mu\text{g g}^{-1}$
185 d.w., TORT-2 with [THg] of $0.27 \pm 0.06 \mu\text{g g}^{-1}$ d.w., MESS-3 with [THg] of $0.091 \pm 0.009 \mu\text{g g}^{-1}$ d.w.)
186 were run at least once per every 10 samples to assess analysis accuracy. Recoveries for CRMs were
187 within $100 \pm 20\%$, while blanks were $< 10\%$ of the lowest sample [THg].

188 The MeHg concentrations ([MeHg]) in plankton and macroinvertebrate samples were
189 analyzed with a Tekran® 2700 Methyl Mercury Auto-Analysis System (Model 2700; Tekran
190 Instrument Corporation.). The equipment is equipped with atomic fluorescence detection (following
191 EPA Method 1630 (USEPA, 1998)) that has a method detection limit of $0.002 \text{ ng Hg L}^{-1}$. Method
192 blanks were $0.045 \pm 0.01 \text{ ng Hg L}^{-1}$. Sample replicates and CRMs (IAEA-086 with [MeHg] of
193 $0.258 \pm 0.022 \mu\text{g g}^{-1}$ d.w., DORM-3 with [MeHg] of $0.355 \pm 0.056 \mu\text{g g}^{-1}$ d.w., and TORT-3 with
194 [MeHg] of $0.137 \pm 0.012 \mu\text{g g}^{-1}$ d.w.) were run at least once per every 15 samples to assess analysis
195 accuracy (method relative percent difference $< 35\%$).

196 An estimation of total Hg bioconcentration factors (BCF) was calculated using the log ratio of
197 bioaccumulated total Hg concentrations and aqueous total Hg concentrations (Rolffhus *et al.*, 2011; Wu
198 *et al.*, 2019).

199

200 **Fatty acids analysis**

201 Fatty acids analyses were performed at WasserCluster Lunz, Austria. Lipids were extracted
202 and analyzed from freeze-dried, homogenized samples of plankton per size-fraction and
203 macroinvertebrates per species (ca. 1-10 mg dry weight) using chloroform:methanol (2:1 v/v) as
204 described in detail by Heissenberger *et al.* (2010). In brief, total lipids were quantified as mass
205 fractions (mg lipids g^{-1} d.w.) gravimetrically using duplicate measurements. Known volumes of TFA
206 extracts were derivatized to fatty acid methyl esters (FAME) using H_2SO_4 -methanol (incubated at
207 50°C for 16 h). FAME were dried under N_2 before being re-dissolved in hexane and run on a gas
208 chromatograph (Thermo Scientific™ TRACE™ Gas Chromatograph coupled to flame ionization
209 detection) with a Supelco™ SP-2560 column used for separation of FAME. FAME were identified by
210 comparison of their retention times with known standards (37-component FAME mix, Supelco™
211 47885-U; bacterial fatty acids, Supelco™ 47080-U; and the following individual FAME standards:

212 stearidonic acid, O5130 SIGMA™; and n-3 docosapentaenoic acid, Supelco™ 47563-U) and
213 quantified with reference to seven-point calibration curves derived from 2.5, 50, 100, 250, 500, 1000,
214 2000 ng μL⁻¹ solutions of the FAME standard for each identified FA. FAME were expressed as mass
215 fractions (mg FA g dry wt⁻¹) and as individual FA relative proportions (% of total identified FA).

216 PUFA were investigated as algal-derived FA compounds (i.e. linoleic acid (LIN; 18:2*n*-6), α-
217 linoleic acid (ALA; 18:3*n*-3), arachidonic acid (ARA; 20:4*n*-6), eicosapentaenoic acid (EPA; 20:5*n*-3),
218 stearidonic acid (SDA; 18:4*n*-3), and docosahexaenoic acid (DHA; 22:6*n*-3)) that are required for
219 somatic growth of various consumers, as they are integral parts of cell membranes (Kainz &
220 Mazumder, 2005). The odd-saturated and branched-chained FA (i.e. the sum of C15:0 and C17:0 and
221 their iso- and anteiso-series, also known as mono-unsaturated FA) were used as bacterial-derived FA
222 (BFA), while long-chain saturated FAs were considered as terrestrial plant derived FA (terr. FA) (Sun
223 *et al.*, 2000).

224

225 **Stable isotopes analysis and trophic level estimation**

226 A subset sample of plankton per size-fraction and macroinvertebrate per species were selected
227 for stable isotope analysis to identify the natural abundance of ¹³C and ¹⁵N. The dry weight biomass of
228 biological tissue used for stable isotope analysis was approximately 1 mg. The analysis was carried
229 out by the Stable Isotope Facility at UC Davis, United States. Stable isotope values are expressed in δ
230 notation as parts per mille deviation from a standard reference using equation 1:

$$231 \quad \delta^{13}C \text{ or } \delta^{15}N = \left(\frac{R_{\text{sample}}}{R_{\text{standard}}} - 1 \right) \times 1000 \quad [1]$$

232 where *R* is ¹³C/¹²C, or ¹⁵N/¹⁴N

233 The δ¹³C values can be used to assess the consumption of aquatic vs. terrestrial carbon in an
234 individual's diet. The δ¹³C values become heavier with increasing utilization of terrestrial diet (e.g.,
235 vascular plants), but tends to decrease with increasing lipid content (McCutchan *et al.*, 2003). The
236 δ¹⁵N values increase along with trophic level, so it can be used to compare Hg biomagnification across
237 systems as the slope of the regression of log-transformed [Hg] vs. δ¹⁵N (Lavoie *et al.*, 2013).

238 Estimation of trophic levels (TL) in littoral macroinvertebrates using $\delta^{15}\text{N}$ was performed
239 using equation 2:

$$240 \quad TL_{consumer} = \frac{\delta^{15}\text{N}_{consumer} - \delta^{15}\text{N}_{baseline}}{\Delta N} + TL_{baseline} \quad [2]$$

241 where $TL_{consumer}$ is the trophic level of an individual macroinvertebrate species with a measured
242 $\delta^{15}\text{N}_{consumer}$. $\delta^{15}\text{N}_{baseline}$ is a commonly used isotope ratio for a primary consumer occupying a trophic
243 level of $TL_{baseline}$. Isotope enrichment factors (ΔN) of 3.4 per mille was applied (Borga *et al.*, 2012;
244 Jardine *et al.*, 2013).

245

246 **Statistical analysis**

247 The statistical analysis of the data was performed using the software JMP 10 (© SAS Institute
248 Inc.). We used Tukey-Kramer HSD for comparing total Hg and MeHg concentrations by different
249 biological sample types, plankton size fractions, and macroinvertebrates FFG. Regression analyses
250 were used to test whether FA predicted total Hg and MeHg concentrations in plankton and
251 macroinvertebrates. We log-transformed total Hg and MeHg concentrations in water, plankton, and
252 macroinvertebrates to achieve data normality (Shapiro & Wilk, 1965). These values are presented in
253 brackets to represent the log-transformation (i.e. $\log[\text{THg}]_{\text{plankton}}$, $\log[\text{MeHg}]_{\text{water}}$). FA contents
254 were also log-transformed to achieve data normality and expressed in brackets (i.e. $\log[\text{PUFA}]$). The
255 level of significance for all tests was set at $p < 0.05$.

256 We used Partial Least Square (PLS) analysis to assess the relative importance of different
257 environmental factors and FA in explaining the variation of total Hg bioaccumulation in plankton and
258 macroinvertebrates from oligotrophic lakes. PLS allows for the combination of relevant multi-
259 collinear predictor variables and their use in a linear regression equation to predict the response
260 variables. The predictive PLS model plot also yields a variable influence on the projection (VIP) that
261 ranks the relative importance of predictor variables for explaining the response variables (Eriksson *et*
262 *al.*, 2006). VIP scores are calculated for each variable based on the PLS loading weights. A variable
263 with a VIP score close to or greater than 1 (one) can be considered important in the model (Eriksson *et*
264 *al.*, 2006). Thus, we applied the criteria of VIP scores > 1 to identify highly influential predictor

265 variables; variables with VIP scores >0.8 but <1 were identified as moderately influential; while those
266 with VIP scores >0.5 but <0.8 were considered as making relatively small contributions to the
267 prediction of response variables in the PLS model. We used regression coefficients and VIP to
268 evaluate the contribution of each predictor for an individual response. The greater the coefficient size,
269 the stronger the response variable is correlated to the predictor variable. For model output, we
270 displayed the relation between the predictor variables and the response variables in loading scatter
271 plots, whereas the score scatter plots showed how the samples were situated with respect to each other
272 based on predictor variables.

273 Total Hg bioaccumulation, or $\log[\text{THg}]$, were used as response variable in our PLS model.
274 Abiotic, environmental factors (i.e., pH, DOC, and nutrient concentrations) of the aquatic ecosystems
275 and biotic, dietary FA biomarkers (PUFA, BFA, and terr. FA), as well as trophic level (TL), were
276 considered as the predictor variables.

277

278 **Results**

279 **Lake characteristics**

280 The ranges of pH, DOC, and Chl *a*, lake surface water temperature, and concentration of
281 aqueous total Hg and MeHg, totN, and totP in the investigated lakes are listed in Table 1. The
282 Västerbotten lakes were all low in the nutrients i.e. totN ($< 500 \mu\text{g L}^{-1}$) and totP ($<25 \mu\text{g L}^{-1}$), as well
283 as low in pH (4.89-6.51). DOC concentrations were high in most of the Västerbotten lakes ($>20 \text{mg L}^{-1}$),
284 except for lakes Mångstenstjärn and Lillsjöleden (10.3 and 13.5mg L^{-1} , respectively). The
285 mesotrophic Lake Erken had the highest nutrient concentrations (totN: $712 \mu\text{g L}^{-1}$; totP: $60.3 \mu\text{g L}^{-1}$)
286 and pH levels (7.87), but lower DOC (11.2mg L^{-1}). Increasing DOC concentrations in lakes were
287 significantly associated with low pH (correlation $r=-0.89$, $p<0.0001$).

288 Total Hg concentrations in lake water ranged from 1.2 to 11ng L^{-1} in the oligotrophic lakes,
289 and the lowest concentration was found in the mesotrophic Lake Erken (0.31ng L^{-1}). MeHg
290 concentrations in all lakes were low ($<0.45 \text{ng L}^{-1}$), and even below the method detection limit in Lake

291 Erken ($<0.06 \text{ ng L}^{-1}$). Aqueous total Hg concentrations in lakes were negatively correlated with pH
292 ($r=-0.78, p=0.04$) and positively correlated with DOC ($r=0.90, p<0.01$).

293

294 **Plankton and macroinvertebrates**

295 The phytoplankton community composition did not vary among the oligotrophic lakes. The
296 phytoplankton communities in most of the lakes were dominated by small Euglenophyta (mostly
297 *Trachelomonas sp.* 10-11 μm). In the mesotrophic Lake Erken, phytoplankton largely dominated by
298 diatoms, particularly *Fragilaria sp.* $<100 \mu\text{m}$ (Figure 1a). The zooplankton communities in the
299 epilimnion of the investigated lakes was dominated by rotifers 100-500 μm , except in lake Lillsjöliden
300 and Nedre Björntjärn, where there was a higher cladoceran abundance (Figure 1b). Rotifers dominated
301 in the hypolimnion across all lakes (Figure 1c).

302 The cluster analysis based on phytoplankton and zooplankton community compositions
303 (Figure 1d, 1e, and 1f), showed that Lake Erken was significantly different from the other lakes in its
304 phytoplankton community composition. However, Lake Erken did not seem to be more different
305 compared to other lakes in terms of the zooplankton composition. There was no difference in either
306 $\delta^{13}\text{C}$ or $\delta^{15}\text{N}$ from plankton across all size fractions ($p>0.05$) in all lakes.

307 The macroinvertebrate community differed in its taxonomic composition and abundance
308 greatly among lakes, with Lake Erken having both the highest diversity and abundance of
309 macroinvertebrates from six different classes: Clitellata, Gastropoda, Bivalvia, Insecta, Malacostraca,
310 Arachnida. The oligotrophic lakes were low in species biodiversity (SI Table S1). The differences in
311 insect group diversity at the level of order (*Ephemeroptera*, *Diptera*, *Hemiptera*, *Coleoptera*,
312 *Lepidoptera*, *Trichoptera*, *Megaloptera*, *Neuroptera*, and *Odonata*) was not marked among the lakes.
313 The stable carbon isotope ratios ($\delta^{13}\text{C}$) in zooplankton and macroinvertebrates were significantly
314 different between the oligotrophic lakes and the mesotrophic Lake Erken ($p<0.001$), but there was no
315 significant difference in terms of $\delta^{15}\text{N}$ values of macroinvertebrates ($p>0.05$) (Table 2, Figure 3).

316 There were differences in stable isotope ratios and trophic levels among the
317 macroinvertebrates FFG (Table 2). For example, $\delta^{13}\text{C}$ values were higher in collectors ($F(2,54)=12$,

318 $p < 0.0001$) than in predators and shredders across all lakes, while $\delta^{15}\text{N}$ values were significantly
319 enriched in predators ($F(2,54)=25, p < 0.0001$). In short for the oligotrophic lakes, $\delta^{13}\text{C}$: Collectors \geq
320 Shredders \geq Predators, $\delta^{15}\text{N}$ and TL: Predators \geq Collectors \geq Shredders. While for the mesotrophic
321 Lake Erken: $\delta^{13}\text{C}$: Collectors \geq Predators \geq Shredders, $\delta^{15}\text{N}$ and TL: Predators $>$ Shredders \geq Collectors.

322

323 **Hg bioaccumulation and fatty acids**

324 Phytoplankton and zooplankton [THg], [MeHg], and %MeHg across all size-fractions did not
325 differ significantly (Table 2). [MeHg] in littoral macroinvertebrates across oligotrophic lakes
326 ($151.3 \pm 105.5 \text{ ng g}^{-1} \text{ d.w.}$) were significantly higher than in phytoplankton ($6.1 \pm 6.2 \text{ ng g}^{-1} \text{ d.w.}$) and
327 zooplankton ($6.6 \pm 3.2 \text{ ng g}^{-1} \text{ d.w.}$) ($F(2,18)=11, p < 0.001$), although there was a great variation for
328 macroinvertebrates [MeHg]. MeHg(%) across oligotrophic lakes was also higher in
329 macroinvertebrates ($51.8 \pm 30.7\%$) than in phytoplankton ($3.6 \pm 5.7\%$) and zooplankton ($2.3 \pm 2.2\%$)
330 ($F(2,18)=15, p < 0.001$). Mass fractions of terrestrial FA [terr. FA] were significantly higher in
331 phytoplankton than in zooplankton and macroinvertebrates ($F(2, 56)=7, p < 0.01$) in the oligotrophic
332 lakes, while [BFA] were significantly higher in macroinvertebrates from the oligotrophic lakes than in
333 the mesotrophic Lake Erken ($F(1,58)=6, p=0.02$) (Table 2). The [PUFA], [BFA], and [terr.FA] were
334 not generally different between oligotrophic lakes and the mesotrophic Lake Erken for phytoplankton,
335 zooplankton, and macroinvertebrates ($p > 0.05$), respectively.

336 Lake trophic status and DOC were more influential on pelagic food web Hg bioaccumulation
337 compared to forest clear-cut and nitrogen addition. Both phytoplankton and zooplankton [THg] and
338 [MeHg] were significantly higher in oligotrophic lakes than in the mesotrophic Lake Erken ($p < 0.01$),
339 regardless of lake treatment effects (Figure 4a and 4b). The lakes with clear-cut (only Lillsjöliden and
340 Stortjärn) had the highest [MeHg] in both phytoplankton and zooplankton (Figure 4b). The highest
341 phytoplankton and zooplankton [THg] were found in Lake Nedre Björntjärn ($p < 0.01$), which had both
342 nitrogen addition and organic matter input from forest clear-cut. Phytoplankton and zooplankton
343 [PUFA], [BFA], and [terr. FA] from Lake Nedre Björntjärn was also the highest compared to other
344 lakes ($p < 0.01$, Figure 4c, 4d, and 4e). In lakes with high DOC ($\geq 20 \text{ mg L}^{-1}$), [THg], [MeHg] and all

345 FA mass fractions in both phytoplankton and zooplankton were significantly higher ($p<0.05$)
346 compared with lakes with less DOC (10-15 mg L⁻¹) (Figure 4f, 4g, 4h, 4i, 4j).

347 Neither forest clear-cut nor nitrogen addition lead to significant shifts in macroinvertebrate FA
348 concentrations ($p>0.05$). Both macroinvertebrate [THg] and [MeHg] in the clear-cut lakes (only
349 Lillsjöleden and Stortjärn) were the highest as compared to other oligotrophic lakes or Lake Erken
350 ($p<0.01$). Macroinvertebrate Hg accumulation differed significantly across FFG among all lakes: The
351 predators [THg] (255.8 ± 141.9 ng g⁻¹ d.w.) and [MeHg] (192.6 ± 76.6 ng g⁻¹ d.w.) were the highest
352 among all FFG (for [THg]: $F(2,44)=9$, $p<0.001$; for [MeHg]: $F(2,11)=8$, $p<0.01$) (Figure 5a and 5b).
353 Both [THg] and [MeHg] of macroinvertebrates were higher in lakes with high DOC than in low DOC
354 lakes (Figure 5c and 5d). Neither FA concentrations in macroinvertebrates in high DOC lakes did not
355 differ with low DOC ones ($p>0.05$). Macroinvertebrates [THg] increased significantly with increasing
356 $\delta^{15}\text{N}$ and TL in both oligotrophic lakes and the mesotrophic Lake Erken ($p<0.05$). However, none of
357 FA types correlated with $\delta^{15}\text{N}$ or TL in macroinvertebrates from oligotrophic lakes ($p>0.05$).

358 Regardless of lake treatment effects, both phytoplankton and zooplankton [THg] in
359 oligotrophic lakes were closely related to its FA content (Figure 6a, 6b, 6c); [PUFA] explained 68%
360 ($R^2=0.68$, $F(1, 19)=40$, $p<0.01$) and [terr.FA] 57% ($R^2=0.57$, $F(1, 19)=28$, $p<0.01$) of the [THg]
361 variability in plankton. More specifically, zooplankton (meso- and macrozooplankton) [THg] in high
362 DOC lakes significantly correlated with [PUFA] ($R^2=0.55$, $F(1, 6)=7$, $p=0.03$) and [terr.FA] ($R^2=0.72$,
363 $F(1, 6)=15$, $p<0.01$). In the oligotrophic lakes, the THg bioconcentration factor (BCF_{THg}), calculated as
364 the ratio of [THg] in phytoplankton (seston and microplankton) and aqueous [THg], correlated
365 positively with [terr.FA] ($R^2=0.41$, $F(1, 9)=6$, $p=0.03$). The [PUFA] ($R^2=0.37$, $F(1, 45)=26$, $p<0.01$)
366 and [BFA] ($R^2=0.38$, $F(1, 44)=27$, $p<0.01$) were more closely related to macroinvertebrates [THg] for
367 all lakes (Figure 6d, 6e). Particularly, carnivorous macroinvertebrates [terr.FA] was closely related to
368 [THg] ($R^2=0.65$, $p<0.01$) in the mesotrophic Lake Erken, along with PUFA ($R^2=0.71$, $p<0.01$) and
369 bacterial fatty acids ($R^2=0.59$, $p<0.02$). Neither FA type predicted [THg] or [MeHg] in
370 macroinvertebrates in high DOC lakes.

371 Both phytoplankton and zooplankton [MeHg] in oligotrophic lakes decreased with increasing
372 [terr.FA] ($R^2=0.40$, $F(1, 10)=7$, $p=0.03$) (Figure 7). Zooplankton [MeHg] in high DOC lakes decreased
373 with increasing [terr.FA] ($R^2=0.93$, $F(1, 3)=28$, $p=0.03$).

374

375 **PLS analysis of Hg bioaccumulation**

376 The total Hg bioaccumulation in the zooplankton and macroinvertebrates was predicted in a
377 PLS model with predictor variables included water chemistry (i.e., pH, DOC, nutrients) and
378 biomarkers (stable isotope values and fatty acids). The PLS analysis predicted 77% of response
379 variation for log[THg] (Table S3), a detailed list of the model loading scores are listed in Table S4.
380 pH, [terr.FA] and totP were the top three influential predictor variable in predicting total Hg
381 bioaccumulation in zooplankton and macroinvertebrates, followed by DOC, aqueous Hg, and total
382 nitrogen concentrations (Table 3). While [PUFA], [BFA], chlorophyll a and trophic levels, were the
383 least influential VIPs for THg variation in zooplankton and macroinvertebrates. The first component
384 of PLS predicted 67% of total Hg variation (Table S3), and the two most influential predictor variables
385 were pH and DOC, according to the loadings score of the first component (Table S4).

386

387 **Discussion**

388 This field study provides empirical evidence that terrestrial food sources are closely associated
389 with THg bioaccumulation in boreal freshwater food webs. We found that the contribution from
390 terrestrial and algal-derived OM to Hg bioaccumulation were equally important for meso- and
391 macrozooplankton in oligotrophic lakes, i.e., THg variation in zooplankton were equally explained by
392 algal-derived diet ($R^2=0.67$, $p<0.01$) and terrestrial OM diet ($R^2=0.66$, $p<0.01$) (Figure 6a and 6c).
393 Either direct uptake of terrestrial OM or ingestion of bacteria that sequester OM likely resulted in this
394 elevated Hg in zooplankton and macroinvertebrates. This suggests that non-essential Hg in these
395 aquatic consumers is associated with the retention of equally non-essential terrestrial OM, from a
396 nutritional point of view (Grey *et al.*, 2001; Jansson *et al.*, 2007; Cole *et al.*, 2011; Karlsson *et al.*,
397 2012; Lau *et al.*, 2012). The OM-Hg interactions have been intensively studied with a general
398 consensus that OM mobilizes total Hg and MeHg from soil to waters (Skjellberg *et al.*, 2000; Shanley

399 & Bishop, 2012; Kronberg *et al.*, 2016). The high abundance of cladocerans across oligotrophic lakes
400 is strongly exposed to terrestrial food sources (Berggren *et al.*, 2014) and, once ingested, Hg may be
401 equally transferred via such terrestrial as well as algal food sources, as seen in this study. However, the
402 extent of zooplankton or macroinvertebrates uptake of terrestrial OM was restrained in high DOC
403 lakes, as was the case for MeHg bioaccumulation (Wu *et al.*, 2019). This likely suggests that
404 zooplankton depends on essential nutrients from algal food sources to sustain their somatic growth and
405 reproduction (Karlsson *et al.*, 2012; Masclaux *et al.*, 2012; Wenzel *et al.*, 2012). Retaining algae and
406 bacteria in zooplankton can be closely related to MeHg bioaccumulation (Kainz & Mazumder, 2005)
407 and sizes (Kainz *et al.*, 2008). Previous studies by Jansson *et al.* (2007) and Lau *et al.* (2012) have
408 shown that macroinvertebrates can utilize terrestrial OM. Going a step further, we were able to find
409 that both pelagic and littoral consumers were exposed to Hg via terrestrial OM uptake. This could be
410 seen in the mesotrophic Lake Erken where total Hg bioaccumulation in macroinvertebrates correlated
411 with terrestrial fatty acids (Figure 6f), suggesting that terrestrial OM may contribute to Hg
412 bioaccumulation even in a mesotrophic lake. At the same time, the feeding behavior of
413 macroinvertebrates was still critical to their Hg bioaccumulation, which is in accordance with previous
414 studies (Parkman & Meili, 1993).

415

416 **Effects of nitrogen addition and clear-cut**

417 We had expected to see lower zooplankton Hg bioaccumulation in lakes with nitrogen
418 addition and forest clear-cut, due to previous findings that increased nutrient in lakes will promote
419 phytoplankton biomass production (Deininger *et al.*, 2017a) which can lead to biodilution of
420 zooplankton Hg (Karimi *et al.*, 2007). By contrast, there was much higher THg in zooplankton from
421 the lake with nitrogen addition. The MeHg was also higher in zooplankton and macroinvertebrates
422 from clear-cut lakes. This suggests both pelagic and littoral food webs in these lakes were more
423 sensitive towards elevated Hg fluxes, but less sensitive to nutrient inputs. Indeed there was an increase
424 of phytoplankton biomass in the nitrogen addition lake, but the additional nutrients and resulting
425 dietary energy were not efficiently transferred up to zooplankton (Deininger *et al.*, 2017b). Light
426 attenuation in brown water lakes (DOC concentrations > 15 mg L⁻¹) could have overridden the effects

427 of phytoplankton production and biomass increase after nitrogen addition (Deininger *et al.*, 2017a). A
428 similar situation occurred in forest clear-cut lakes as the phytoplankton community changed little
429 while lake nutrient and DOC remained stable (Deininger *et al.*, 2019). The elevation of zooplankton
430 Hg bioaccumulation was likely a consequence of decreased or stable phytoplankton biomass as lake
431 DOC increased and led to increased zooplankton utilization of terrestrial OM-Hg complexes.

432 According to the first component loadings of our PLS model (Table S4), the lake
433 characteristics pH and DOC most important for Hg bioaccumulation. In boreal ecosystems, low pH is
434 usually coupled with high DOC (in our case, $r=-0.89$, $p<0.0001$), which contributed to elevated Hg
435 bioavailability, resulting in increased Hg uptake by aquatic consumers (Kelly *et al.*, 2003). This likely
436 has to do with the fact that MeHg in its neutral form methylmercury-chloride has higher lipid
437 solubility under higher DOC (or lower pH), allowing MeHg to be efficiently adsorbed and transfused
438 across cell membranes (Pickhardt & Fisher, 2007). Thus, if MeHg bioavailability is greatly promoted
439 under low pH in high DOC freshwaters, i.e., in our investigated humic lakes, this can significantly
440 contribute to elevated Hg content in aquatic consumers, regardless of consumer selectivity towards
441 different diet sources. There is also a likelihood of spurious correlations between total Hg
442 bioaccumulation and DOC, due to a strong correlation of aqueous total Hg concentrations with DOC
443 (in our case, $r=0.90$, $p<0.01$) (Braaten *et al.*, 2019). Increased lake DOC also has large impacts on lake
444 conditions, i.e., light attenuation (Ask *et al.*, 2009) and oxygen depletion (Couture *et al.*, 2015), which
445 possibly affect the growth and feeding of zooplankton and macroinvertebrates on various dietary
446 sources, and eventually Hg bioaccumulation.

447

448 **Carbon isotopic values and role of methanotrophic bacteria**

449 The oligotrophic lake food webs were significantly more depleted in ^{13}C compared to the
450 mesotrophic Lake Erken (Figure 3). One possible explanation is the zooplankton uptake of
451 methanotrophic bacteria supported by terrestrial OM (Kankaala *et al.*, 2006; Deines *et al.*, 2009; Cole
452 *et al.*, 2011). It was clear that THg exposure closely related to bacterial diet in both zooplankton and
453 macroinvertebrates (Figure 6), although we do not have direct proof of zooplankton or
454 macroinvertebrate from these lakes feeding on methanotrophic bacteria. It is likely that methanotrophs

455 have contributed to Hg bioaccumulation via terrestrial OM in these oligotrophic lakes we have
456 investigated, due to high prevalence of methane emission in lakes (Summons *et al.*, 1994; Bastviken *et*
457 *al.*, 2004).

458

459 **Role of dietary sources in MeHg bioaccumulation**

460 We did not find a positive correlation of MeHg bioaccumulation with either algal-derived or
461 bacterial fatty acids as Kainz and Mazumder (2005) have shown in monomictic coastal lakes. Instead,
462 MeHg bioaccumulation declined with increasing terrestrial diet in pelagic food webs, or bacterial diet
463 in littoral food webs (Figure 7). It was likely that both terrestrial and bacterial diets were largely
464 respired and little integrated to consumer metabolism for growth as suggested by Brett *et al.* (2009);
465 Karlsson *et al.* (2012). The decreased MeHg bioaccumulation associated with increasing bacterial FA
466 was also similar to what has been described in de Wit *et al.* (2012) about macroinvertebrates sampled
467 in Norwegian streams during the summer. This reflects both zooplankton and macroinvertebrates
468 selectively assimilating high quality algal-derived diet for growth and reproduction, even under
469 circumstances with abundant terrestrial OM availability. The limited number of samples analyzed for
470 both MeHg and PUFA for extrapolation of statistical correlations may, however, have contributed to
471 the failure to find a relationship. It was challenging to obtain all MeHg results for zooplankton due to
472 limited sample biomass, especially in the clear-cut lake Struptjärn where the bloom of flagellated
473 microalga *G. semen* occurred, which was also reported by Deininger *et al.* (2019).

474

475 **Conclusion**

476 Our study provides evidence on pelagic and littoral food web Hg bioaccumulation responses
477 to terrestrial carbon and nitrogen inputs. This is important given the wide extent of timber harvest in
478 forested landscapes (Furukawa *et al.*, 2015) and increasing human activities in agricultural and
479 industrial fields that promote nitrogen loading to surface waters (Vitousek *et al.*, 1997). With
480 evidences from fatty acids as dietary biomarkers, we have shown that zooplankton and
481 macroinvertebrates accumulate more Hg after clear-cut and nitrogen addition in high DOC lakes due

482 to light limitation and oxygen depletion. Thus, we suggest that high DOC lakes are more vulnerable
483 towards additional terrestrial nutrient input regarding Hg bioaccumulation. This study also highlights
484 the importance of macroinvertebrates feeding behavior and the potentially critical role of
485 methanotrophs for MeHg bioaccumulation in oligotrophic lakes. Further investigations are still needed
486 as less is known about the physiological requirements of zooplankton and littoral macroinvertebrates
487 for terrestrial OM and their interaction with Hg.

488

489 **Associated content**

490 **Supporting information**

491 We included a table summarized the macroinvertebrate community info in Table S1, and the analytical
492 methods and instruments for water chemistry analysis in Table S2. Table S3 and S4 are PLS model
493 scores for both predictor variables and response variables.

494

495 **Acknowledgements**

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500 acids analysis in their lab, as well as the analytical facility and staff at the Biotron Centre for
501 Experimental Climate Change Research at Western University for their great support in analyzing total
502 and methylmercury.

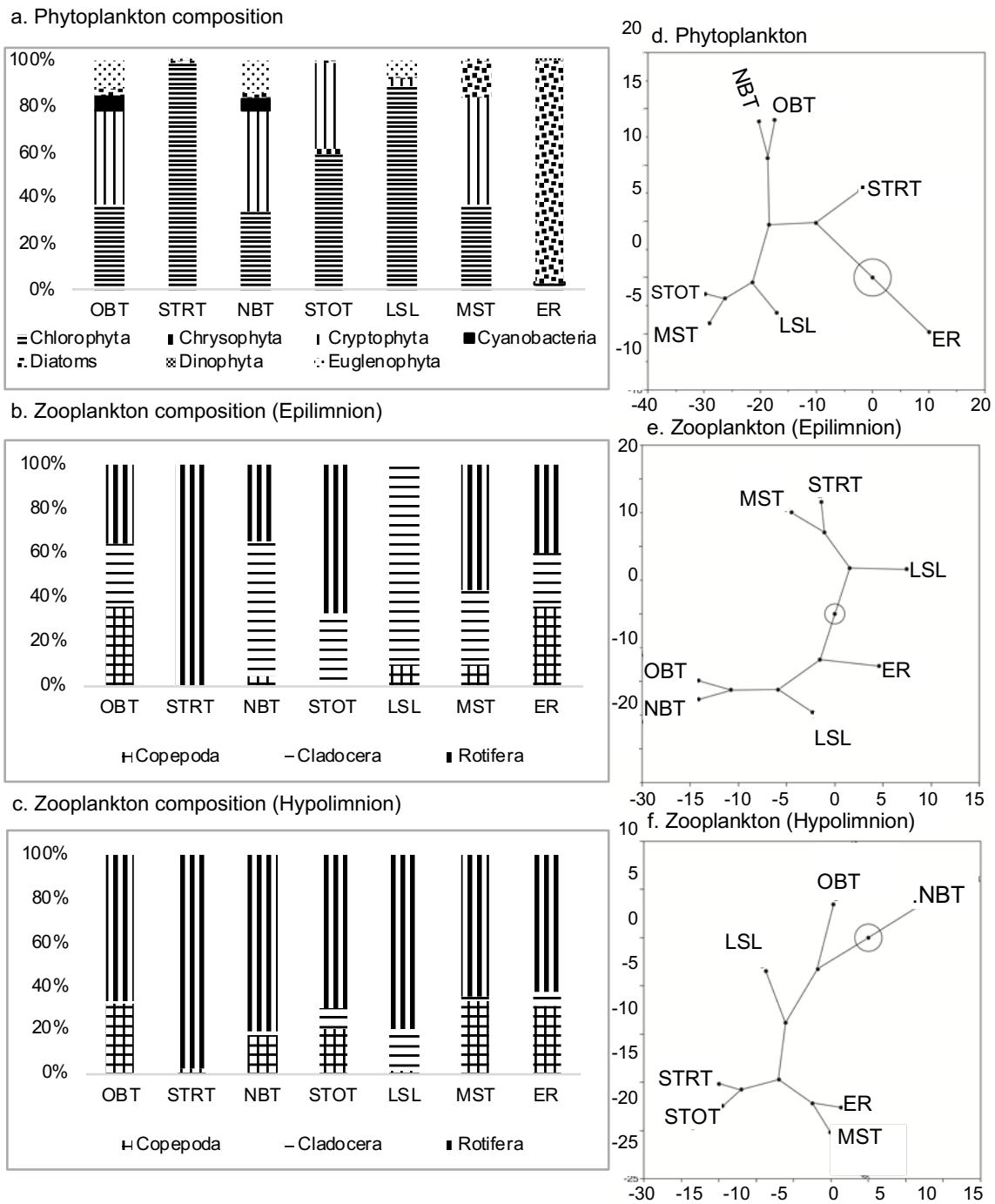
503 Pianpian Wu holds a postgraduate stipend from the China Scholarship Council (CSC) (2013-
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506 Framework (Contract 2013-6978).

Table 1. Summary lake characteristics, extent of forest clear-cut, and water chemistry analysis results from the sampling campaign in 2015. DOC: dissolved organic carbon, Chl *a*: Chlorophyll *a*.

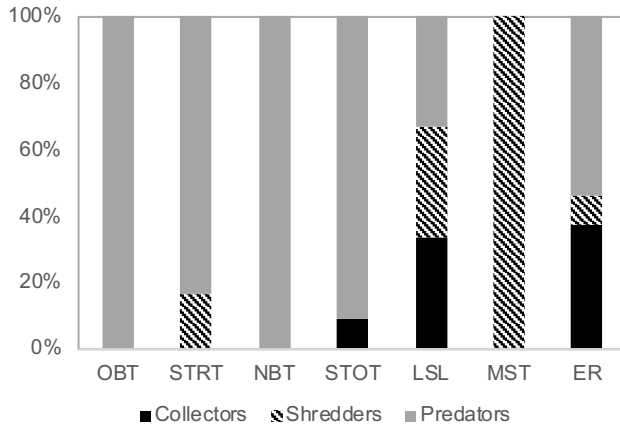
Lake	Abbreviation	Lake type	Lake treatment	Mean depth (m)	Secchi depth (m)	Surface area (ha)	Catchment area (ha)	%Clear-cut of forest area in watershed	DOC (mg L ⁻¹)	pH	Chl <i>a</i> (µg L ⁻¹)	Total Hg (ng L ⁻¹)	MeHg (ng L ⁻¹)	totP (µg L ⁻¹)	totN (µg L ⁻¹)
Övre Björntjärn	OBT	Humic	Clear-cut reference	4	0.5	4.8	284	11.6	27.2	4.89	1.5	7.1	0.29	15.6	510
Struptjärn	STRT	Humic*	Clear-cut	3.8	0.5	3.1	1183	37.4	25.1	5.24	34	11	0.45	24	547
Nedre Björntjärn	NBT	Humic	N addition & Clear-cut	6	0.4	3.2	324.9	10.7	23	4.99	1.5	6.4	0.28	17.4	432
Stortjärn	STOT	Humic	Clear-cut Reference	2.7	0.5	3.9	86.6	/	19.6	5.28	2.5	3.8	0.16	11.7	469
Lillsjöleden	LSL	Clear	Clear-cut	3.8	1.2	0.8	29.2	57.6	13.5	6.24	2.4	2.1	0.33	11.4	318
Mångstenstjärn	MST	Clear	N addition Reference	5.3	1.1	1.8	14.1	/	10.3	6.51	2.3	1.2	0.14	11	298
Erken	ER	Clear	/	9	2.9	2400	14100	/	11.2	7.87	8	0.31	<0.06	60.3	712

*Lake Struptjärn was undergoing an algal bloom of *G. semen* from an invasive species during the sampling campaign in 2015 (Deininger *et al.*, 2019).

Note: Whole lake fertilization of nitrogen (N) addition was implemented in Nedre Björntjärn in 2012 and 2013, while forest clear-cut activity took place in the November 2012 in the catchments of Lillsjöleden and Struptjärn. The catchment of lake Övre Björntjärn was clear-cut in November 2013, and Nedre Björntjärn was clear-cut in June 2014. Later in 2014, the harvested areas around lake Lillsjöleden and Struptjärn were subject to site preparation.



512 *Figure 1. Upper left (a): phytoplankton community classified in phylum group based on percentage density across lakes. Middle left (b):*
 513 *epilimnion (surface water) zooplankton community of different zooplankton classified in subclass group by percentage across lakes. Lower*
 514 *left (c): hypolimnion (close to the water bottom) zooplankton community classified in subclass group by percentage across lakes. Upper right*
 515 *(d): Constellation plot of phytoplankton community clustered in lakes. Middle right (e): Constellation plot of epilimnion zooplankton*
 516 *community clustered in lakes. Lower right (f): Constellation plot of hypolimnion zooplankton community clustered in lakes. OBT: Övre*
 517 *Björntjärn; STRT: Struptjärn; NBT: Nedre Björntjärn; STOT: Stortjärn; LSL: Lillsjöleden; MST: Mångstenstjärn; ER: Erken.*



518

519 *Figure 2. Relative abundance of macroinvertebrate functional feeding groups based on number of taxa collected from investigated lakes.*

520 OBT: Övre Björntjärn; STRT: Struptjärn; NBT: Nedre Björntjärn; STOT: Stortjärn; LSL: Lillsjöleden; MST: Mångstenstjärn; ER: Erken.

521

522 *Table 2. Summary of total Hg (THg), MeHg, the proportion of MeHg in total Hg (%MeHg), stable isotope ratios $\delta^{13}C$ and $\delta^{15}N$,*

523 *trophic level (TL), total lipids, bacteria FA (BFA), terrestrial FA (terr. FA), and algae derived PUFA in biological samples. THg,*

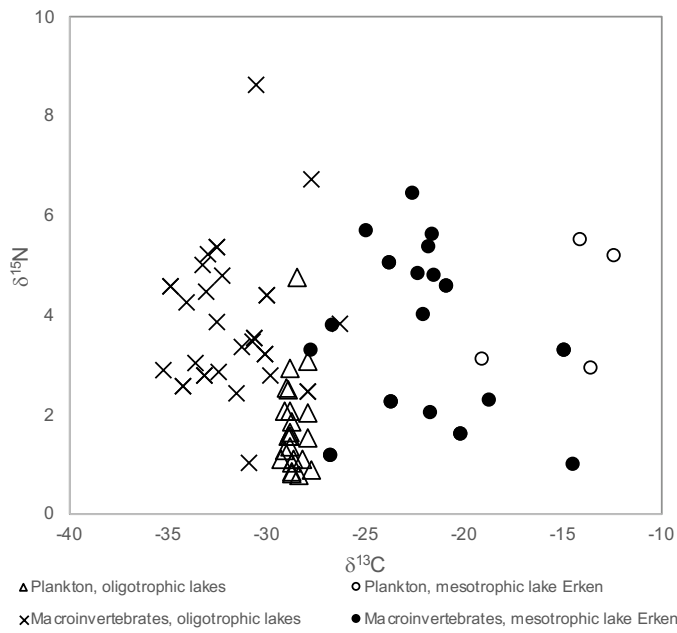
524 *MeHg, total lipids, BFA, PUFA, and terr. FA concentrations in dry weight (d.w.). All values stated as Mean \pm Standard deviation*

525 *(Number of samples).*

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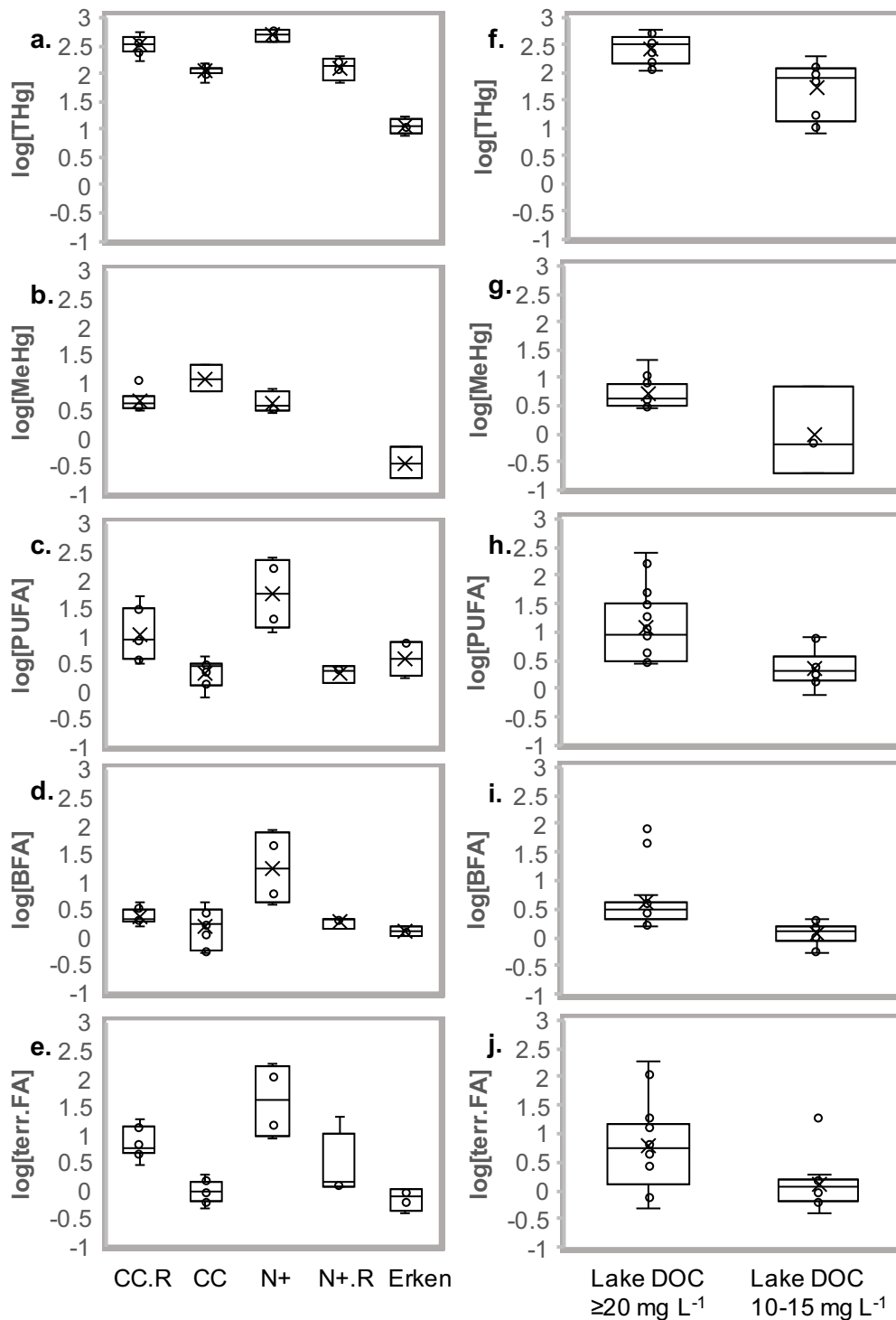
	Phytoplankton seston (<25 μm), microplankton (25-100 μm)	Zooplankton mesozooplankton (100-500 μm), macrozooplankton (> 500 μm)	Macroinvertebrates					
			Oligotrophic lakes			Mesotrophic lake		
			Collectors	Shredders	Predators	Collectors	Shredders	Predators
THg (ng g^{-1} d.w.)	244.7 \pm 216.4(14)	198.1 \pm 132.6(14)	/	91.7 \pm 27.5(3)	234.0 \pm 132.7(24)	18.6 \pm 8.4(9)	57.9(1)	80.5 \pm 41.6(9)
MeHg (ng g^{-1} d.w.)	5.4 \pm 6.1(8)	5.5 \pm 3.9(6)	/	6.74(1)	192.6 \pm 76.6(7)	8.0 \pm 4.7(4)	/	5.2(1)
%MeHg	3.6 \pm 5.7(8)	2.3 \pm 2.2(6)	/	8.9(1)	62.4 \pm 19.1(7)	66.0 \pm 28.3(4)	/	3.9(1)
$\delta^{13}C$	-26.3 \pm 5.6(14)	-26.8 \pm 4.6(14)	-27.9(1)	-31.9 \pm 1.5(5)	-31.9 \pm 2.2(26)	-19.8 \pm 4.7(9)	-23.6(1)	-23.1 \pm 2.2(13)
$\delta^{15}N$	2.1 \pm 1.5(13)	2.1 \pm 1.1(14)	2.5(1)	2.5 \pm 0.8(5)	4.2 \pm 1.4(26)	1.9 \pm 0.9(9)	2.3(1)	4.9 \pm 0.8(13)
TL	1.0 \pm 0.1(14)	2.0 \pm 0.1(14)	2.3(1)	2.2 \pm 0.3(4)	2.8 \pm 0.5(14)	2.3 \pm 0.2(6)	2.4(1)	3.1 \pm 0.2(13)
Total lipids (mg g^{-1} d.w.)	161.4 \pm 245.8(14)	77.1 \pm 37.7(14)	51.9 \pm 17.2(3)	116.3 \pm 23.1(6)	112.3 \pm 50.4(27)	44.1 \pm 24.2(9)	82.2 \pm 0.9(2)	116.0 \pm 52.6(13)
PUFA (mg g^{-1} d.w.)	43.2 \pm 82.5(11)	8.6 \pm 13.0(14)	19 \pm 10.3(3)	42.5 \pm 10.4(6)	39.2 \pm 19(27)	19 \pm 8.4(9)	27.5 \pm 0.4(2)	48.0 \pm 25.6(13)
BFA (mg g^{-1} d.w.)	13.5 \pm 26.8(11)	2.3 \pm 1.4(14)	8.3 \pm 2.1(3)	14.5 \pm 2.7(6)	14.0 \pm 7.1(27)	4.4 \pm 0.4(9)	10.8 \pm 0.4(2)	12.8 \pm 5.9(13)
terr. FA (mg g^{-1} d.w.)	28.1 \pm 56.3(13)	4.2 \pm 4.7(14)	0.6 \pm 0.4(3)	1.7 \pm 1.4(6)	1.3 \pm 0.7(27)	2.7 \pm 6.2(9)	0.9 \pm 0.0(2)	1.2 \pm 0.9(13)

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Figure 3. Stable isotopes values of $\delta^{13}\text{C}$ (x-axis) and $\delta^{15}\text{N}$ (y-axis) plot by sample type and lake trophic status. Patterns represent samples from different trophic status, triangles: plankton from oligotrophic lakes; crosses: macroinvertebrates from oligotrophic lakes; circles: plankton from the mesotrophic Lake Erken; dots: macroinvertebrates from the mesotrophic Lake Erken.



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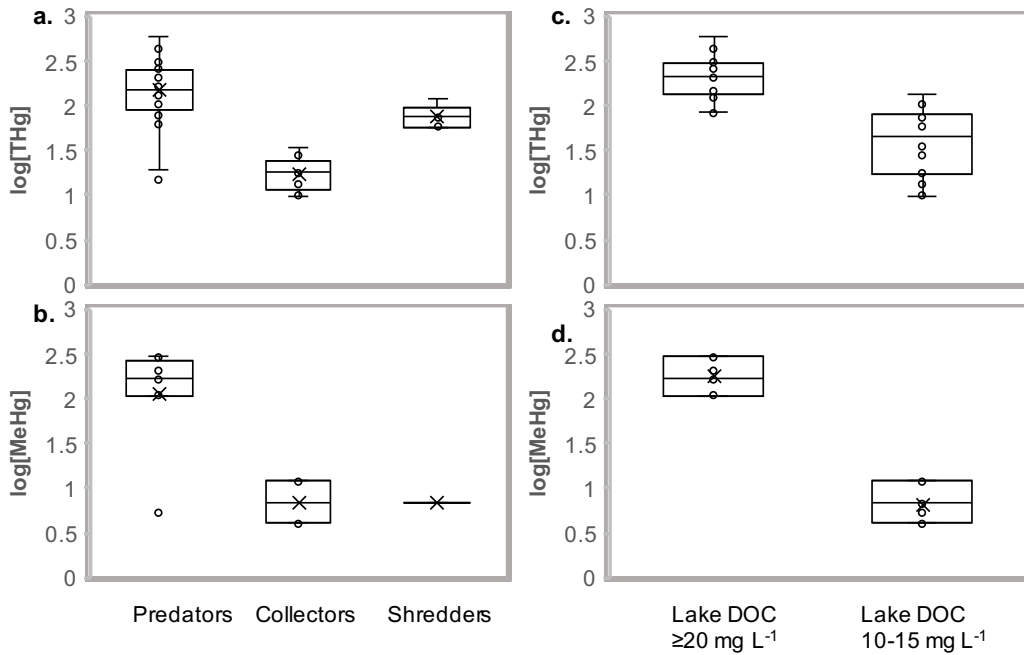
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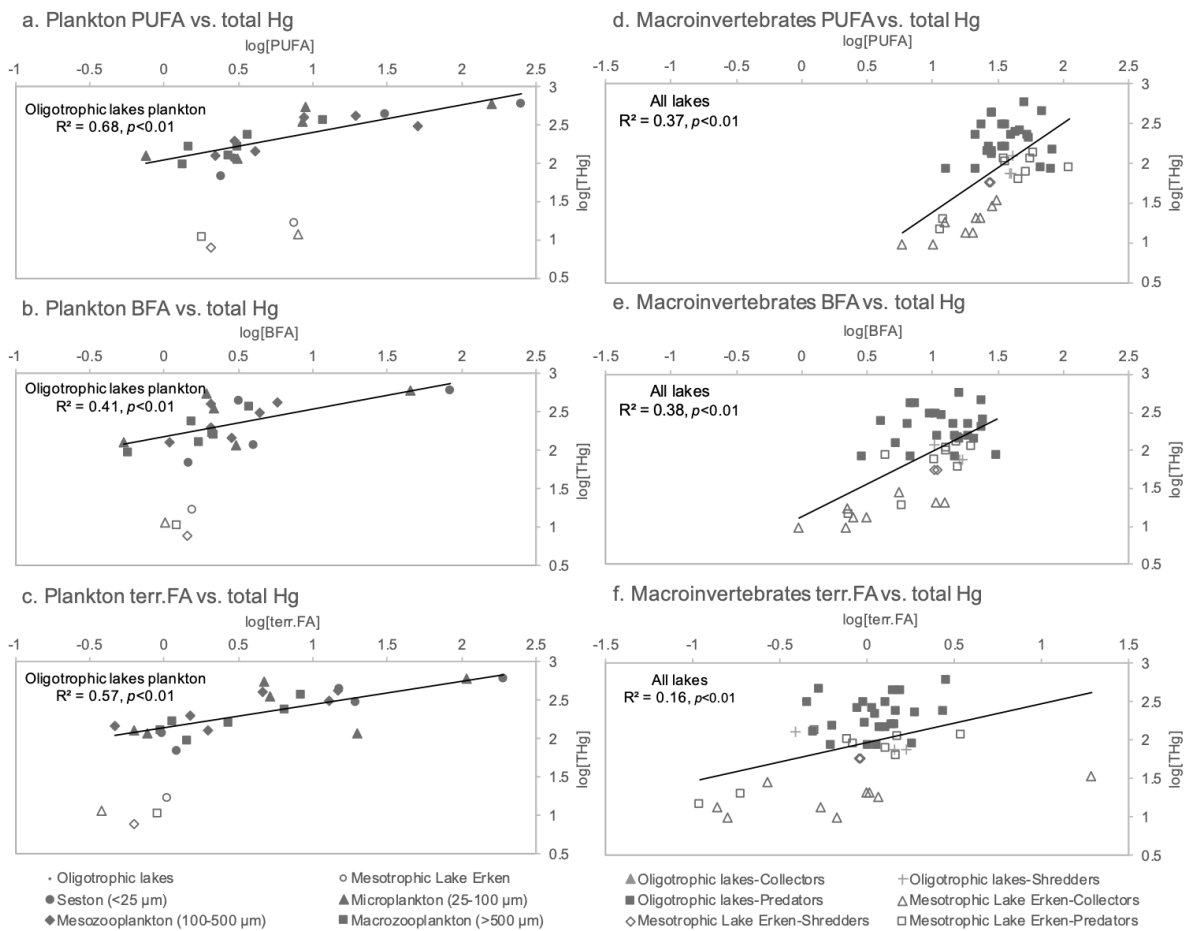
Figure 4. Left top to left bottom (a, b, c, d, and e): Box-whisker plots of plankton (phytoplankton and zooplankton) concentrations of (a) total mercury, (b) methylmercury, (c) algal-derived polyunsaturated fatty acids (PUFA), (d) bacterial fatty acids (BFA), (e) terrestrial fatty acids (terr.FA) in terms of lake treatments or location. CC.R: Clear-cut reference lakes (Övre Björntjärn and Stortjärn); CC: Clear-cut lakes (Lillsjöleden and Struptjärn); N+: whole lake nitrogen fertilization lake (Nedre Björntjärn); N+.R: Reference lake to the nitrogen fertilization one (Mångstenstjärn); Erken: Mesotrophic Lake Erken. Right top to right bottom (f, g, h, i, and j): Box-whisker plots of plankton concentrations of (a) total mercury, (b) methylmercury, (c) PUFA, (d) BFA, (e) terr.FA in lakes with different DOC levels (High DOC

541 concentration $\geq 20 \text{ mg L}^{-1}$ (Övre Björntjärn, Stortjärn, Struptjärn, and Nedre Björntjärn) and median DOC concentration 10-15 mg L^{-1}
542 (Lillsjöleden, Mångstenstjärn, and Erken). All concentrations log-transformed.



543
544 Figure 5. Box-whisker plots of macroinvertebrates concentrations of (a) total mercury (b) methylmercury in three different functional
545 feeding groups: predators, collectors, and shredders; macroinvertebrate concentrations of (c) total mercury, (d) methylmercury in lakes with
546 different DOC levels (High DOC concentration $\geq 20 \text{ mg L}^{-1}$ (Övre Björntjärn, Stortjärn, Struptjärn, and Nedre Björntjärn) and median DOC
547 concentration 10-15 mg L^{-1} (Lillsjöleden, Mångstenstjärn, and Erken)). All concentrations log-transformed.

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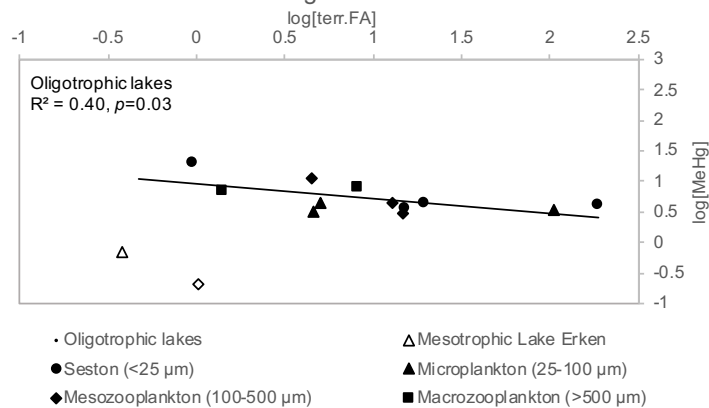
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Figure 6. Left top to left bottom (a, b, and c): Total mercury concentrations related to (a) algal-derived polyunsaturated fatty acids (PUFA) concentrations, and (b) total bacterial fatty acids (BFA) concentrations, as well as (c) total terrestrial fatty acids (terr.FA) concentrations of size-fractionated seston (<25 μm), microplankton (25-100 μm), mesozooplankton (100-500 μm), and macrozooplankton (>500 μm) in both oligotrophic lakes (dark filled symbols) and the mesotrophic Lake Erken (light-colored outlined symbols). Right top to right bottom (d, e, and f): Total mercury concentrations related to (d) PUFA, and (e) BFA, as well as (f) terr.FA concentrations of macroinvertebrates functional feeding groups (FFG) across all lakes (oligotrophic lakes in filled and crossed symbols, the mesotrophic Lake Erken in light-colored outlined symbols). All concentrations log-transformed.

Plankton terr. FA vs. MeHg



558

559 Figure 7. Methylmercury concentrations related to terrestrial fatty acids (*terr.FA*) concentrations of different size-fractionated seston (<25
 560 μm), microplankton (25-100 μm), mesozooplankton (100-500 μm), and macrozooplankton (>500 μm) in both oligotrophic lakes (dark filled
 561 symbols) and the mesotrophic Lake Erken (light-colored outlined symbols). All concentrations log-transformed.

562

563 Table 3. Variable Influence on the Projection (VIP) scores from our PLS analysis explained with regression models for
 564 predicting total mercury bioaccumulation in zooplankton and macroinvertebrates in all lakes. DOC: dissolved organic
 565 carbon concentration in mg L^{-1} . *totP*: aqueous total phosphorous concentration in $\mu\text{g L}^{-1}$. *totN*: aqueous total nitrogen
 566 concentration in $\mu\text{g L}^{-1}$. *Chl a*: aqueous chlorophyll a concentration in $\mu\text{g L}^{-1}$. $[\text{THg}]_{\text{water}}$: aqueous total Hg concentration in
 567 ng L^{-1} . $[\text{MeHg}]_{\text{water}}$: aqueous MeHg concentration in ng L^{-1} . $\log[\text{PUFA}]$: log-transformed polyunsaturated fatty acids
 568 concentration in mg g^{-1} on dry weight basis(d.w.). $\log[\text{BFA}]$: log-transformed bacterial fatty acids concentration in mg g^{-1}
 569 d.w. $\log[\text{terr.FA}]$: log-transformed bacterial fatty acids concentrations in mg g^{-1} d.w. TL: trophic level. Influential predictor
 570 variables with VIP scores >1 are marked in bold.

Predictor Variables	VIP	Coefficient
pH	1.34	-0.12
<i>log[terr. FA]</i>	1.25	0.42
<i>totP</i>	1.25	-0.17
DOC	1.19	0.04
<i>[THg]water</i>	1.12	0.08
<i>[MeHg]water</i>	1.10	0.09
<i>totN</i>	1.04	-0.19
<i>log[BFA]</i>	0.66	0.10
<i>log10[PUFA]</i>	0.55	0.08
<i>Chl a</i>	0.53	0.04
TL	0.32	0.10

571

Predictor Variables	VIP	Coefficient
<i>log[terr.FA]</i>	1.42	0.75
pH	1.04	-0.15
Trophic level	0.9	0.42
DOC	0.88	0.15
<i>log[PUFA]</i>	0.86	0.06

572

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