



**HAL**  
open science

## Assessing the impacts of mining activities on fish health in Northern Québec

Anthony Fontaine, Mackenzie Anne Clifford Martyniuk, Camille Garnier,  
Patrice Couture

► **To cite this version:**

Anthony Fontaine, Mackenzie Anne Clifford Martyniuk, Camille Garnier, Patrice Couture. Assessing the impacts of mining activities on fish health in Northern Québec. *Canadian Journal of Fisheries and Aquatic Sciences*, 2023, 80 (5), pp.771-794. 10.1139/cjfas-2022-0200 . hal-04576828

**HAL Id: hal-04576828**

**<https://univ-pau.hal.science/hal-04576828>**

Submitted on 15 May 2024

**HAL** is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

1    **ASSESSING THE IMPACTS OF MINING ACTIVITIES ON FISH HEALTH**  
2                                    **IN NORTHERN QUÉBEC**

3    Anthony Fontaine<sup>1</sup>, Mackenzie Anne Clifford Martyniuk<sup>1</sup>, Camille Garnier<sup>1</sup>, Patrice Couture<sup>1</sup>

4    <sup>1</sup> *Institut National de la Recherche Scientifique, centre Eau-Terre-Environnement, Québec, Canada*

5

6    **Corresponding author:**

7    Anthony Fontaine

8    anthony.fontaine@univ-pau.fr

9    **ORCID ID:** <https://orcid.org/0000-0001-5779-5789>

10

**ABSTRACT**

11 For several decades, Northern Québec has been exploited by mining companies for its mineral  
12 resources, yet, research documenting the effects of toxic stress on fish health in subarctic  
13 environments remains limited. In this study, one lake directly affected by mining activities in the  
14 Schefferville area, two lakes close to mining facilities in the Fermont and Schefferville areas, and  
15 one reference lake were sampled for water, sediment and fish. Our results suggest that  
16 manganese bioaccumulation induced an oxidative stress in *Catostominae*, as demonstrated by  
17 the positive relationships between manganese concentrations and catalase activity and 8-  
18 hydroxy-2'-deoxyguanosine concentrations, as well as the negative relationship with glutathione  
19 peroxidase activity (both catalase and glutathione peroxidase activities being biomarkers of  
20 antioxidant capacities and 8-hydroxy-2'-deoxyguanosine an indicator of DNA oxidative damage).  
21 Similarly, selenium bioaccumulation was positively correlated with 8-hydroxy-2'-deoxyguanosine  
22 concentrations in *Salmoninae*. These results suggest a prooxidant role of excess selenium and  
23 manganese, and highlight the interspecific variability of fish responses to contaminated areas  
24 around historical and current iron ore mining operations.

25

26 **Keywords:** Northern fish, metal contamination, oxidative stress, Fermont, Schefferville

## 27 **1 INTRODUCTION**

---

28 Fish are ectothermic organisms, meaning that energy allocation is modulated in part by the  
29 environmental temperature (Jobling et al., 1997; Power, 1978). Generally, in cold environments,  
30 the age at maturity is delayed and the growth and reproductive seasons are shorter than in a  
31 warmer climate (Duchesne and Magnan, 1997; Houde, 1989; Stewart et al., 2007). Energy  
32 accumulated is allocated towards the maintenance of homeostasis, growth, reproduction, energy  
33 storage and sometimes migration, though this is species, life stage and habitat dependent  
34 (Jørgensen and Fiksen, 2006). However, when fish are facing a toxic stress such as metal  
35 contamination, detoxification of these stressors and repair of affected biological tissues and  
36 functions represent additional energetic costs. As a consequence, this will reduce allocation of  
37 energy to other vital functions, potentially leading to physiological (Couture and Kumar, 2003;  
38 Maes et al., 2005; McPhee and Janz, 2014) and behavioral (Berntssen et al., 2003; Scott and  
39 Sloman, 2004; Sloman and McNeil, 2012) disruptions. Uptake and internal handling of trace  
40 elements vary according to environmental (e.g. temperature, pH, water hardness) (Karthikeyan  
41 et al., 2007; Köck et al., 1996) and individual variables (e.g. age, length, growth) (Farkas et al.,  
42 2003; Greenfield et al., 2001) that in turn modulate fish health. Additionally, depending on their  
43 trophic position, fish can be exposed to high metal concentrations through their diet (Kuchapski  
44 and Rasmussen, 2015; van der Velden et al., 2013).

45 Metals are naturally occurring at the Earth's surface, but environmental concentrations can  
46 be significantly amplified by recent human activities. Mining operations (from the exploration  
47 phase to after the end of exploitation) increase the input of contaminants into natural  
48 environments which often include aquatic ecosystems. Although some elements are essential for  
49 fish, playing a key role in various metabolic reactions (e.g. barium (Ba), cobalt (Co), copper (Cu),  
50 iron (Fe), manganese (Mn), selenium (Se) and zinc (Zn)), others are classified as non-essential

51 (e.g. aluminum (Al), arsenic (As), cadmium (Cd), mercury (Hg) and lead (Pb)). However, the  
52 essentiality of certain elements, such as chromium (Cr) or nickel (Ni) remains controversial  
53 (Wood, 2011). While essential elements can lead to toxic stress when occurring in elevated  
54 concentrations (Wood et al., 2011a), non-essential elements can result in negative effects on fish  
55 health even at very low concentrations (Wood et al., 2011b). Many physiological disruptions have  
56 been associated with elevated elemental concentrations, including impairment of metabolic  
57 activities (Couture and Pyle, 2012) and hormonal responses (Laflamme et al., 2000; Levesque et  
58 al., 2002), reproduction (Friedmann et al., 1996), reduction of growth (Eastwood and Couture,  
59 2002) and longevity (Couture and Pyle, 2008; Pyle et al., 2008) or impacts on the antioxidant  
60 defence system, as highlighted by the observation of biomarkers of reactive oxygen species  
61 (ROS) (Sevcikova et al., 2011).

62 The ROS are naturally produced by aerobic metabolism (Winston and Di Giulio, 1991) and  
63 are regulated by two defense systems: the activity of antioxidant enzymes, such as superoxide  
64 dismutase (SOD), catalase (CAT) and glutathione peroxidase (GPx), and antioxidant molecules  
65 such as reduced glutathione (GSH) (Di Giulio et al., 1989; Lackner, 1998; Livingstone, 2001).  
66 Generally, metals are associated with the production of ROS in two ways (Sevcikova et al., 2011).  
67 First, some elements such as Fe, Cu, or Cr generate ROS through their redox potential, while Hg,  
68 Pb, Cd, or Ni can directly impact antioxidant mechanisms. Oxidative stress is what occurs when  
69 these antioxidant capacities are exceeded, which then disrupts cellular homeostasis and  
70 culminates in accumulated ROS. This leads to associated oxidative damage, such as lipid  
71 peroxidation and deoxyribonucleic acid (DNA) damage (Halliwell, 1999; Lackner, 1998). Although  
72 fish responses to mining and metal contamination have been well documented in more Southern  
73 regions of Canada (Couture and Pyle, 2012; Sherwood et al., 2000), literature evaluating such  
74 impacts in more Northern locations remains limited to a few scientific studies (Martyniuk et al.,  
75 2020b) and small scale regional environmental monitoring programs (Makivik Coproration, 2021).

76 Even though literature is scarce on the particular impacts of iron ore mining on fish health,  
77 one study case stands out: the Samarco disaster (southeast Brazil). In 2015, roughly 50 million  
78 m<sup>3</sup> of iron mine tailing were discharged in the environment after a dam break. Since then, several  
79 studies have highlighted an increase of metal concentrations (Ba, Co, Cr, Cd, Cu, Fe, Mn, Ni) in  
80 the aquatic ecosystem downstream (da Silva et al., 2022; Passos et al., 2020; Siqueira et al.,  
81 2022). Assessing fish responses to metal contamination, Gabriel et al. (2020) showed that two  
82 demersal fish species captured in the estuary (*Cathorops spixii* and *Genidens genidens*)  
83 expressed high values of As, Cr, Mn, Hg and Se in both liver and muscle tissues. Additionally,  
84 trace metal bioaccumulation was significantly correlated with the expression of metallothioneins  
85 (MT) and GSH, two biomarkers of detoxification processes. Another study from Weber et al.  
86 (2020) demonstrated that two fish species (*Hoplias intermedius* and *Hypostomus affinis*), sampled  
87 along the Doce River in a contaminated area, showed liver damage (vacuolization, necrosis and  
88 hypertrophy of hepatocytes) and a significant response of cytochrome P450 1A (CYP1A), MT and  
89 CAT. *Hoplias intermedius* also expressed significantly higher level of SOD than their counterparts  
90 from the reference site. Together, these results reinforce the conclusion of a long-term chronic  
91 contamination which can be observed in the surrounding ecosystem of iron ore mining operations  
92 (Baeten et al., 2018). Nevertheless, fish response to environmental contamination from iron  
93 mining operations could markedly differ between warm-water species and cold-water species. In  
94 their study, Weber et al. (2020) selected two species that are known to be compliant to strong  
95 environmental changes, yet, they still observed negative effects due to metal contamination.  
96 Northern fish species could be more sensible to such toxic stressors since they experience shorter  
97 periods of high energy accumulation and metabolic activity compared to individuals from Southern  
98 locations. Thus, their tolerance range (i.e. detoxification capacities, behavioural adaptations,  
99 reproductive success) to environmental contamination from mining activities might be more  
100 limited than warm-water species.

101 The Labrador Trough is a region of Mid-Northern Québec recognized for its mining potential,  
102 especially iron (Simard, 2018) in the Fermont and Schefferville areas. The first mining operations  
103 began in Schefferville in the 1950s under the direction of the Iron Ore Company of Canada (IOC)  
104 (Boutet, 2012) while in Fermont, development started in 1970 with the Compagnie minière  
105 Québec Cartier (CMQC) (CLD de Caniapiscau, 2015). Historically, several companies have  
106 exploited claims in the two regions, which has had a major impact on the environment, particularly  
107 the surrounding aquatic ecosystems (Aebischer et al., 2015; Laperrière et al., 2007). More  
108 recently in 2014, further environmental concerns were raised in the region after several incidents.  
109 These included the failure of a dam at the Bloom Lake mine site in Fermont (Gouvernement du  
110 Canada, 2014) and red coloured water identified by Schefferville residents entering aquatic  
111 systems in areas proximate to several of the surrounding mines (Blais, 2019). The objective of  
112 this study was to investigate the effects of past and present mining activities on fish health in both  
113 locations. To assess elemental accumulation in fish with similar feeding strategies and trophic  
114 position, six fish species further grouped in three subfamilies were sampled in three contaminated  
115 lakes from Fermont and Schefferville in addition to a reference lake in the same region, but far  
116 from potential mining influence. Relationships between liver trace element concentrations and  
117 condition indices (Fulton's condition factor (FCF), hepatosomatic index (HSI)) and biomarkers of  
118 antioxidant capacities (CAT, SOD and GPx) as well as oxidative damage (malondialdehyde  
119 (MDA), a lipid peroxide, and 8-hydroxy-2'-deoxyguanosine (8-OHdG), an indicator of DNA  
120 oxidative damage) were investigated to evaluate effects of contamination from mining operations  
121 on fish health.

## 122 **2 MATERIALS AND METHODS**

---

### 123 **2.1 Study area**

124 The towns of Fermont (52°46'59"N, 67°05'00"W) and Schefferville (54°47'59"N,  
125 66°50'00"W) are located in Northern Québec on the provincial boundary between Québec and  
126 Labrador (Figure 1). Geologically, Fermont and Schefferville are located in the Labrador Trough  
127 of the Canadian Shield, which has been globally identified as an exceptionally productive area for  
128 metal mining (Simard, 2018). These two regions are characterized by a subarctic climate with a  
129 short growing season and vegetation transitioning between boreal forest and arctic tundra  
130 (Vowinkel et al., 1975). Recorded average temperatures range from -22.1°C in January to  
131 13.5°C in July for Fermont (Ministère de l'Environnement et de la Lutte contre les changements  
132 climatiques, 2020). In Schefferville, the average temperatures for January and July are -23.3°C  
133 (Gouvernement du Canada, 2020a) and 13.8°C, respectively (Gouvernement du Canada,  
134 2020b). Mining operations, and more specifically iron ore exploitation, have occurred in the region  
135 since the 1950's and have been a dominant force in the local economy of both Fermont and  
136 Schefferville (Boutet, 2012; Simard and Brisson, 2013). Two main iron ore exploitation sites are  
137 currently operational in Fermont: the Mont-Wright Mine (16 Mt/year) owned by ArcelorMittal  
138 Mining Canada G.P. (AMEM), and the Bloom Lake Mine (7.6 Mt/year) by Minerai de fer Québec  
139 (a subsidiary of Champion Iron). Tata Steel Minerals Canada (TSMC) has been the sole mining  
140 operation profiting from iron ore deposit exploitation (1 Mt/year) in Schefferville since 2014.  
141 Historical mining operations in Schefferville involves two companies: IOC which exploited iron ore  
142 from 1954 to 1982 (from 2 to 12 Mt/ year) and Labrador Iron Mine Limited (LIM) from 2011 to  
143 2013 (3.6 Mt produced during the three years of exploitation). Schefferville is also positioned in  
144 the traditional territories of two Indigenous Nations: the Naskapi Nation of Kawawachikamach and  
145 Innu community of Matimekush-Lac-John (Simard and Brisson, 2016). Individuals from both



146 communities still practice traditional hunting and fishing activities on land situated near both  
147 historic and current mining operations.

148 In summer 2019, three lakes reported to have received input from both historical and  
149 present mining operations (Table 1) were sampled to assess the effects of mining activities on  
150 fish health and their environment (Figure 1). In Schefferville, lakes were chosen with the  
151 assistance of an Innu guide, for their proximity to past and present mining activity and identified  
152 as of potential concern from the local community. In addition to be a primary food item, fish  
153 represent important cultural, economical and social resources for Indigenous Nations. As such,  
154 there have been growing concerns since local guides identified mine tailings contaminating the  
155 lakes (Table 1). Lake Perroche ( $54^{\circ}1'40.11''\text{N}$  and  $69^{\circ}37'30.72''\text{W}$ ), located nearly 200 km from  
156 both Fermont and Schefferville and free from any mining influences, was chosen as a reference  
157 site due to its ecological and physicochemical similarity with the aforementioned sampling sites.  
158 In Fermont, Lake Cherny ( $52^{\circ}52'40.02''\text{N}$  and  $67^{\circ}26'0.64''\text{W}$ ) is located about 6 km downstream  
159 from the Bloom Lake mining site. In Schefferville, Lake Denault ( $54^{\circ}50'13.75''\text{N}$  and  
160  $66^{\circ}53'26.79''\text{W}$ ) is directly adjacent to a historical site of IOC exploitation, while Lake Houston  
161 ( $54^{\circ}43'57.31''\text{N}$  and  $66^{\circ}41'5.43''\text{W}$ ) is approximately 10 km from this location, but closer to past  
162 Labrador Iron Mines (LIM) and current TSMC exploitation sites. Sampling occurred between July  
163 15<sup>th</sup> and August 8<sup>th</sup> in Fermont area and in the reference site, and between August 25<sup>th</sup> and  
164 September 3<sup>th</sup> in Schefferville.

## 165 **2.2 Sample collection**

166 Water variables, including pH (Waterproof pHep@5 pH/Temperature Tester – H198128,  
167 Hanna Instruments, Laval, Québec, Canada), temperature ( $^{\circ}\text{C}$ ) and conductivity ( $\mu\text{S}/\text{cm}$ ) (YSI  
168 Model 85, YSI Incorporated, Yellow Springs, Ohio, USA) were measured at each sampling  
169 location. The probe of both instruments was recording approximately 30 cm underwater. Two  
170 surface water and sediment samples (Ekman Bottom Grabs 225  $\text{cm}^2$ , KC Denmark A/S,

171 Silkeborg, Denmark) were also collected at each site to establish trace element concentrations in  
172 addition to water field and transport blanks. Ultrapure water was used to prepare water field and  
173 transport blanks to evaluate potential procedural and material contamination. To sample surface  
174 water, the syringe was rinsed five times consecutively with surface water before setting the filter  
175 (25 mm PES 0.45  $\mu\text{m}$ , Canadian Life Science, Peterborough, ON, Canada). Then, 20 mL of  
176 surface water were sampled and 5 mL were discarded. The remaining 15 mL were stored in trace  
177 metal free tubes at 4°C.

178 To maximize diversity of fish capture, sampling was completed using experimental gill nets  
179 (25 to 150 mm mesh panels, 120 m length, 2 m depth, Fipec Industries, Grande-Rivière, Québec,  
180 Canada), bait traps and angling. A total of ten different species were caught: Northern pike (*Esox*  
181 *lucius*), Lake charr (*Salvelinus namaycush*), Brook charr (*Salvelinus fontinalis*), White sucker  
182 (*Catostomus commersonii*), Longnose sucker (*Catostomus catostomus*), Lake whitefish  
183 (*Coregonus clupeaformis*), Round whitefish (*Prosopium cylindraceum*), Lake chub (*Couesius*  
184 *plumbeus*), Burbot (*Lota lota*) and Slimy sculpin (*Cottus cognatus*). See Table 2 for the details of  
185 capture data. A maximum of 20 individuals from each species were retained from each location.  
186 However, considering the low sample numbers for Burbot ( $n = 3$ ) and Slimy sculpin ( $n = 4$ ), these  
187 species were excluded from any further analysis. Only one Northern pike was captured in a  
188 contaminated site (Lake Cherny) and Lake chub could only be analysed for whole fish metal  
189 concentration, hence, both species were also excluded from this study. Our sampling protocol  
190 was approved by the Comité Institutionnel de Protection des Animaux (CIPA) of the Institut  
191 National de la Recherche Scientifique (INRS) and by the Ministère des Forêts, de la Faune et des  
192 Parcs (MFFP) (SEG permit 18-08-06-061-09-S-P).

193 All fish captured were sacrificed by a sharp blow to the head before fork length ( $\pm 1$  mm)  
194 and mass ( $\pm 1$  g) were measured for each individual. These measurements were then used to  
195 calculate FCF as follows:  $\text{FCF} = (W_T/L^3) \cdot 10^5$  where  $W_T$  represents the total weight of the fish (g)

196 and L the fork length (mm). Prior to further dissection and tissue sample collection, sex and  
197 maturity were determined when possible. Gonad weight ( $\pm 1$  g), as well as liver weight ( $\pm 1$  g)  
198 were also recorded to estimate gonadosomatic index (GSI) and HSI, respectively. The HSI, an  
199 energy status indicator, was estimated as follows:  $HSI = 100 \cdot W_L / W_T$  where  $W_L$  is the liver wet  
200 weight (g). The GSI is used to assess sexual maturity of fish and was determined as follows:  $GSI$   
201  $= 100 \cdot W_G / W_T$  where  $W_G$  is the gonad weight (g). Otoliths, scales, and opercular bones for White  
202 sucker and Longnose sucker were removed and evaluated by MFFP personnel to estimate fish  
203 age. Two independent readings were performed on each structure and if results differed, a third  
204 reading was completed. Otoliths were prioritized compared to other structures.

205 All captured fish were then dissected at the sampling site to collect liver tissue for trace  
206 element analyses, as well as enzyme, lipid peroxidation and DNA damage analyses. Lake chub  
207 were excluded from this procedure, as whole fish tissue was used to perform trace element  
208 analyses for this species. Samples were immediately placed in 15 mL trace metal free tubes and  
209 frozen at  $-20^\circ\text{C}$  until subsequent trace element analyses could be performed. For enzyme, lipid  
210 peroxidation, DNA damage analyses, and protein concentration determination, liver tissue was  
211 sub-sampled and stored in cryogenic tubes at  $-80^\circ\text{C}$ .

## 212 **2.3 Trace Element Analyses**

### 213 **Fish**

214 Trace element analyses for water, sediment and fish liver tissues were all performed at  
215 the Institut National de la Recherche Scientifique – Centre Eau Terre Environnement (INRS-ETE)  
216 in Québec (QC), Canada. Liver samples were lyophilized for a minimum of 48 h (FTS Systems  
217 TMM, Kinetics Thermal Systems, Longueuil, QC, Canada), after which freeze-dried tissues were  
218 divided between total mercury (THg) (approximately  $50\text{-}100$  mg  $\pm$   $0.1$  mg) and additional trace  
219 element analyses (approximately  $10\text{-}15$  mg  $\pm$   $0.1$  mg) (XS205 DualRange, Mettler-Toledo GmbH,

220 Greifensee, Switzerland). In instances where sample weights were insufficient for both analyses,  
221 trace element analyses were prioritized.

222 Dry weight (dw) THg results were determined with a Direct Mercury Analyzer (DMA-80,  
223 Milestone Inc., Shelton, USA) according to US EPA method 7473 (EPA, 1998) before conversion  
224 to final wet weight concentrations using percent moisture calculations (van der Velden et al.,  
225 2013). As THg concentrations are generally highly correlated with age and/or length (Gantner et  
226 al., 2010; Muir et al., 2005), obtained data were tested for significant relationships with these  
227 variables using linear regressions ( $\log_{10}\text{THg}$  vs.  $\log_{10}\text{fork length}$ , fork length, or  $\log_{10}\text{age}$ , and  
228 age) (Martyniuk et al., 2020a; Tran et al., 2015) in an effort to determine the necessity of data  
229 length standardization. Certified reference materials TORT-3 (Lobster hepatopancreas, National  
230 Research Council of Canada, NRCC, Halifax, NS, Canada) and DOLT-5 (Dogfish liver, National  
231 Research Council of Canada, NRCC, Halifax, NS, Canada) from the National Research Council  
232 of Canada (NRCC), as well as blanks, were assessed in conjunction with every fifth tissue sample  
233 in the same analytical cycle to establish accuracy and recovery. The average recoveries  
234 (expressed as % of certified values  $\pm$  standard error) were  $97.4 \pm 0.9\%$  and  $90.4 \pm 0.7\%$  for TORT-  
235 3 and DOLT-5 respectively.

236 Liver samples dedicated for additional trace element analyses were digested in 1500  $\mu\text{L}$   
237 of Optima grade nitric acid (70%, v/v, Optima grade, Fisher Scientific, Whitby, ON, Canada) for 3  
238 days at room temperature, before being heated for 2 hours at 60°C. After samples were cooled  
239 to room temperature, 750  $\mu\text{L}$  of hydrogen peroxide (30%, v/v, Optima grade, Fisher Scientific,  
240 Whitby, ON, Canada) and 15  $\mu\text{L}$  of hydrofluoric acid (47 – 51%, v/v, Optima grade, Fischer  
241 Scientific, Whitby, ON, Canada) were added and samples were subsequently digested for an  
242 additional 24 hours. Volume was adjusted to 15 mL with ultrapure water for a final digestion  
243 volume of 10% nitric acid. Regarding trace element analyses of whole fish, a similar protocol was  
244 applied with the following modifications: 5000  $\mu\text{L}$  nitric acid, 2500  $\mu\text{L}$  hydrogen peroxide, 50  $\mu\text{L}$

245 hydrofluoric acid and volume completed to 50 mL with ultrapure water. Concentrations of Al, As,  
246 Ba, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Se, and Zn were then quantified using either inductively  
247 coupled atomic emission spectroscopy (ICP-AES) (Varian Vista XP Axial CCD Simultaneous ICP-  
248 AES, Agilent Technologies, Santa Clara, California, USA) or inductively coupled plasma-mass  
249 spectrometry (ICP-MS) (Model x-7, Thermo Elemental, Winsford, England, UK). The suite of trace  
250 elements analyzed in this study has been chosen considering metals of concerns from iron mining  
251 operations and from other mining industries (i.e. copper, gold, nickel) that took place in the  
252 Canadian Shield. Certified reference materials TORT-3, DOLT-5 and NIST-1515 (Apple leaves,  
253 National Institute of Standards and Technology, NIST, Gaithersburg, MD, USA) from the NRCC  
254 and blanks were digested in triplicate with identical protocols to control for efficacy, accuracy, and  
255 reproducibility. The average recoveries (expressed as % of certified values  $\pm$  standard error) of  
256 TORT-3 were as follows: As ( $99.2 \pm 5.0\%$ ), Cd ( $92.1 \pm 2.8\%$ ), Co ( $101.6 \pm 4.8\%$ ), Cr ( $98.1 \pm$   
257  $6.5\%$ ), Cu ( $88.0 \pm 3.7\%$ ), Fe ( $87.7 \pm 5.6\%$ ), Mn ( $89.4 \pm 5.4\%$ ), Ni ( $84.5 \pm 4.9\%$ ), Pb ( $97.3 \pm 10.4\%$ ),  
258 Se ( $95.3 \pm 5.9\%$ ) and Zn ( $96.3 \pm 8.9\%$ ). The average recoveries of DOLT-5 were as follows: Al  
259 ( $98.1 \pm 16.4\%$ ), As ( $82.4 \pm 3.3\%$ ), Cd ( $91.9 \pm 3.9\%$ ), Co ( $95.0 \pm 5.2\%$ ), Cr ( $93.3 \pm 5.1\%$ ), Cu ( $75.2$   
260  $\pm 5.3\%$ ), Fe ( $89.0 \pm 6.0\%$ ), Mn ( $84.0 \pm 6.4\%$ ), Ni ( $98.3 \pm 6.9\%$ ), Pb ( $84.3 \pm 10.9\%$ ), Se ( $91.1 \pm$   
261  $6.1\%$ ) and Zn ( $100.4 \pm 9.6\%$ ). The average recoveries (expressed as % of certified values  $\pm$   
262 standard error) for NIST-1515 were as follows: Al ( $88.1 \pm 7.1\%$ ), Ba ( $81.6 \pm 4.2\%$ ) and Mn ( $83.5$   
263  $\pm 9.6\%$ ).

## 264 **Water and sediment**

265 Water samples for trace element analyses were acidified to a pH of 2 (Audet and Couture,  
266 2003) using optima grade nitric acid (70%, v/v, Optima grade, Fisher Scientific, Whitby, ON,  
267 Canada) and stored at 4°C until subsequent ICP-AES and ICP-MS analyses were performed.  
268 Field and transport blanks were prepared following identical procedures. Sediment samples for  
269 trace element analyses were digested following a protocol adapted from Tessier et al. (1979)

270 aimed at the extraction of bioavailable elements. Approximately 20 mL of  $\text{NH}_2\text{OH}\cdot\text{HCl}$  0.04 M  
271 prepared in glacial acetic acid (99%, v/v, Optima grade, Fisher Scientific, Whitby, ON, Canada)  
272 was added to 1 g of non-lyophilized sediment before samples were heated at  $96^\circ\text{C}$  ( $\pm 3^\circ\text{C}$ ) for 6  
273 hours. Samples were agitated every 30 min. Sample volume was then adjusted to 20 mL with  
274 ultrapure water before a decantation period of 48 hours. Subsequently, 5 mL of supernatant was  
275 removed and treated to the previously described digestion protocol.

#### 276 **2.4 Enzyme activities, protein concentration, lipid peroxidation and DNA** 277 **damage analyses**

278 Enzyme activities included the quantification of CAT, SOD and GPx activity. Analyses were  
279 performed using a UV/Vis spectrophotometer (Varian Cary 100, Varian Inc., Palo Alto, California,  
280 USA) on 96-well microplates at room temperature ( $20^\circ\text{C}$ ). Liver samples were homogenized in a  
281 buffer solution prepared with 20 mM HEPES, 1 mM EDTA and 0.10% Triton X-100 and activities  
282 were measured as follows:

- 283 • *Catalase (CAT; EC 1.11.1.6)*: Catalase activity was measured using an assay kit (No.  
284 707002 purchased from Cayman Chemical Company Inc. (Ann Arbor, Michigan USA)),  
285 following the manufacturer's protocol at 540 nm.
- 286 • *Superoxide dismutase (SOD; EC 1.15.1.1)*: Superoxide dismutase activity was performed  
287 using an assay kit (No. 706002 purchased from Cayman Chemical Company Inc. (Ann  
288 Arbor, Michigan USA)), following the manufacturer's protocol at 450 nm.
- 289 • *Glutathione peroxidase (GPx; EC 1.11.1.9)*: Glutathione peroxidase activity was prepared  
290 using an assay kit (No. 703102 purchased from Cayman Chemical Company Inc. (Ann  
291 Arbor, Michigan USA)), according to the manufacturer's protocol at 340 nm.

292 Enzyme concentrations are expressed as nmol/min/mg protein for both CAT and GPx activities  
293 and as U/mg protein for SOD activity (one unit of SOD is defined by the manufacturer as the  
294 amount of enzyme needed to exhibit 50% dismutation of the superoxide radical). Protein

295 concentrations, used to normalize the enzyme activities, were determined on liver homogenates  
296 before centrifugation using Coomassie (Bradford) Protein Assay Kit (No. 23200) at a wavelength  
297 of 595 nm with protein concentrations expressed as mg protein per g of liver wet weight (Bradford,  
298 1976).

299 Lipid peroxidation and DNA damage were estimated through the measurement of MDA  
300 and 8-OHdG concentrations, respectively. Concentrations of MDA were estimated using  
301 thiobarbituric acid reactive substances (TBARS) assay kits (No. 10009055) purchased from  
302 Cayman Chemical Company Inc. (Ann Arbor, Michigan USA). Sample preparation and assays  
303 were performed according to manufacturer's instructions with MDA concentrations measured at  
304 540 nm and expressed as  $\mu\text{mol MDA/g protein}$ . The TBARS assay is not specific to MDA but it is  
305 generally acknowledged that MDA represents the majority of TBARS in the liver. For quantification  
306 of 8-OHdG concentrations, DNA was extracted from collected liver samples using Quick-DNA™  
307 Miniprep Plus Kit (Nos. D4068 & D4069) purchased from Zymo Research (Irvine, CA, USA)  
308 following the manufacturer's instructions. Initial DNA concentrations were then measured with a  
309 NanoDrop-2000 spectrophotometer (Thermo Fisher Scientific). Prior to the measurement of 8-  
310 OHdG concentrations, DNA samples were prepared with the following protocol: (1) heat-  
311 denaturation of the sample for 15 min at 100°C followed by 10 min on ice, (2) addition of 1  $\mu\text{L}$   
312 nuclease P1 solution prepared at 0.175  $\mu\text{g}/\mu\text{L}$  in a buffer solution of 200 mM acetic acid and 10  
313 mM  $\text{CaCl}_2$  (pH = 6.5), (3) incubation for 1 hour at 37°C, (4) addition of 11.66  $\mu\text{L}$  of alkaline  
314 phosphatase solution at 3  $\mu\text{g}/\mu\text{L}$  in 1 M Tris buffer solution (pH = 8), (5) incubation for 30 min at  
315 37°C, (6) heating for 10 min at 100°C before final storage on ice. A secondary DNA concentration  
316 measurement was then taken to adjust the sample solution to 20 ng DNA/ $\mu\text{L}$ . The concentrations  
317 of 8-OHdG were then determined using DNA/RNA Oxidative Damage ELISA kit (No. 589320)  
318 purchased from Cayman Chemical Company Inc. (Ann Arbor, Michigan USA) according to

319 manufacturer's protocols. Absorbance was measured at 410 nm and concentrations of 8-OHdG  
320 are expressed as pg of 8-OHdG per  $\mu\text{g}$  of DNA.

## 321 **2.5 Statistical analyses**

322 Statistical analyses were performed using the R (Version 4.0.3, 2020) software package  
323 and RStudio (Version 1.3.1093, © 2009-2020 RStudio, PBC) with a type I error set to  $\alpha = 0.05$ .  
324 To better identify trends of elemental accumulation in ecologically similar fish, species were  
325 grouped by subfamily. Thus, we gathered White sucker and Longnose sucker in the *Catostominae*  
326 subfamily, Lake whitefish and Round whitefish in the *Coregoninae* subfamily and Lake charr and  
327 Brook charr in the *Salmoninae* subfamily. Grouping the species into subfamilies was necessary to  
328 perform statistically valid comparisons of patterns of bioaccumulation and investigate their  
329 relationships with indicators of fish health, due to the low sample size for some species in some  
330 lakes. However, this grouping clearly influences the metal bioaccumulation values for each  
331 subfamily, depending on the relative number of individuals from different species within these  
332 subfamilies and differences in habitat preference and diet of each species. For example, Lake  
333 charr and Brook charr may occupy different ecological niche or forage on different prey types.  
334 Data interpretation took this limitation into consideration.

335 Comparisons between mean values of biometric and biochemical variables and trace  
336 element concentrations were performed using one-way analysis of variance (ANOVA). Data  
337 normality was assessed using residual diagnostic histograms, Q-Q plots and the Shapiro-Wilk W  
338 test (Shapiro and Wilk, 1965). Homoscedasticity was verified using the Bartlett's test (Bartlett,  
339 1937) or the Levene's test (Levene, 1961). Data that did not respect normality or homoscedasticity  
340 conditions were  $\log_{10}$  transformed and potential outliers were evaluated using Cook's Distance  
341 statistic (Zar, 2007). A Tukey-Kramer honestly significant difference post-hoc test was completed  
342 after each significant ANOVA to evaluate significant differences among means of studied groups.  
343 When data were nonparametric, a Kruskal-Wallis test (Kruskal and Wallis, 1952) was performed.



344 Student's t-tests or Wilcoxon/Mann-Whitney test for nonparametric data were used for  
345 *Catostominae*. Principal component analyses (PCA) were computed for each subfamily to  
346 investigate relationships among liver trace element concentrations and biomarkers to reduce the  
347 dimensionality of the dataset. In order not to bias the PCAs with missing values, the *MissMDA*  
348 package was used to impute values that do not have effects in the calculation of the dimensions.  
349 Results of PCAs are represented by biplots that project the dispersion of the variables on two  
350 principal components. The significance of studied parameters is characterized by arrow length  
351 and direction in relation to the two principal components. Major outputs from the PCA were then  
352 examined with Spearman correlations ( $r_s$ ) to identify significant relationships between trace  
353 element concentrations and biomarkers. A Bonferroni correction was applied to account for the  
354 multiple comparisons performed in the correlation matrices.

### 355 **3 RESULTS**

---

#### 356 **3.1 Lake characteristics**

##### 357 **Water parameters**

358 Water chemistry parameters for all sampled lakes are available in Table 1. Each location  
359 expressed very similar values for temperature, while pH varied slightly between the most acidic  
360 lake studied (Perroche, pH 6.6) and Lake Denault (pH 7.7). Values of conductivity showed a  
361 greater variability among lakes but still remained similar to those detailed in a previous study from  
362 Larocque et al. (2006) where the authors reported the conductivity of sixty lakes (mean: 28.7; min:  
363 10.0; max: 170.0  $\mu\text{S}/\text{cm}$ ) distributed from latitude 49°48'N to 59°32'N in northwestern Québec.

##### 364 **Water and sediment trace element concentrations**

365 Considering the high variability and the substantial number of values below the limits of  
366 detection for As, Ba and Ni concentrations in fish tissues, those elements were excluded from  
367 further analyses. Detection limits are listed in Table S1. Trace element concentrations of Al, Cd,  
368 Co, Cr, Cu, Fe, Mn, Pb, Se and Zn for water and sediment are presented in Figure 2. Aqueous  
369 and sediment concentrations of As, Ba and Ni can be found in Table S2. Aqueous concentrations  
370 of Al and Fe expressed a very similar trend among sampled lakes, with mean values being  
371 approximately 5-fold higher for Al and 10-fold higher for Fe in the reference lake than in lakes  
372 Cherny and Houston. Waterborne concentrations of Cd and Co were greater in Lake Cherny  
373 compared to the reference site and Lake Houston. The concentration of Cr was only determined  
374 in Lake Houston samples, since values from the reference lake, lakes Cherny and Denault were  
375 below the detection limits. Mean values of Cu concentrations appeared to be roughly 2-fold higher  
376 in lakes Cherny and Houston compared to the reference site and Lake Denault. Aqueous  
377 concentrations of Mn and Pb were higher in Lake Houston when compared to the reference lake,  
378 lakes Cherny and Denault. Waterborne Se concentration did not differ among sampled lakes. Zinc

379 concentrations were higher in Lake Houston samples, being approximately 4-fold higher than in  
380 the reference lake and Lake Cherny and 15-fold higher than in Lake Denault. In general, aqueous  
381 element concentrations were low in all lakes sampled and never exceeded the Provincial Water  
382 Quality Objectives (PWQO) (Ministère de l'Environnement et de la Lutte contre les changements  
383 climatiques, 2021).

384 In sediment, Lake Houston exhibited generally higher element concentrations compared  
385 to other lakes. The average value of Cd concentration in Lake Houston was roughly 3-fold higher  
386 than in Lake Cherny, 10-fold higher than in the reference site and 45-fold higher than in Lake  
387 Denault. Sediment Cu concentrations were higher in lakes Houston and Denault compared to the  
388 reference lake and Lake Cherny. Concentrations of Co, Se and Zn all exhibited comparable  
389 patterns among study sites, with mean values being the highest in Lake Houston and the lowest  
390 in Lake Denault. Additionally, sediment Fe and Mn concentrations also showed higher elemental  
391 values in Lake Houston when compared to the reference site and Lake Cherny with Fe  
392 concentrations being 6- to 12-fold higher in Lake Houston compared to other lakes. Sediment Mn  
393 concentrations further varied greatly among sampled lakes, with lakes Denault and Houston  
394 demonstrating values 10- to 30-fold higher than in the reference lake and Lake Cherny.  
395 Concentrations of Al and Cr also differed among study sites, with average sediment  
396 concentrations of these elements in Lake Houston being 18-fold higher than in Lake Denault.  
397 Finally, sediment concentrations of Pb were higher in lakes Cherny and Houston than in the  
398 reference site and Lake Denault. Overall, sediment elemental concentrations were consistently  
399 higher than waterborne elemental concentrations.

### 400 **3.2 Biometric variables**

401 Biometric variables and sex ratios of *Salmoninæ*, *Catostominæ* and *Coregoninæ* are  
402 presented in Table 3, except for GSI, presented in Table S3. Observed fish diversity was greatest  
403 in Lake Denault, but success of captures was highly variable among lakes. *Salmoninæ* were

404 collected in the reference lake (Brook charr and Lake charr), Lake Cherny (Lake charr), Lake  
405 Houston (Brook charr) and in Lake Denault (Brook charr and Lake charr). *Catostominæ* were  
406 caught in the reference site (White sucker) and Lake Denault (White sucker and Longnose  
407 sucker). *Coregoninæ* were captured in the reference lake (Lake whitefish), Lake Cherny (Lake  
408 whitefish and Round whitefish) and Lake Denault (Lake whitefish and Round whitefish). Fish  
409 sampled at the reference site were generally longer, heavier and older than fish collected from  
410 contaminated sites. Fulton's condition factor and HSI were also mostly greater in fish from the  
411 reference lake compared to fish from other lakes. *Salmoninæ* from the reference site, especially  
412 Brook charr, exhibited higher values for fork length ( $X^2_{(1,39)} = 36.00, p < 0.0001$ ), whole weight  
413 ( $X^2_{(1,39)} = 32.50, p < 0.0001$ ) and FCF ( $t_{(1,39)} = -4.10, p = 0.0002$ ) when compared to individuals  
414 from Lake Houston. Nevertheless, *Salmoninæ* collected in Lake Houston were older ( $X^2_{(1,39)} =$   
415  $8.42, p = 0.0148$ ) than individuals from the reference site. Similarly, *Catostominæ* demonstrated  
416 higher values for fork length ( $X^2_{(1,20)} = 1.00, p = 0.0004$ ), whole weight ( $X^2_{(1,20)} = 1.00, p = 0.0004$ ),  
417 FCF ( $X^2_{(1,20)} = 8.00, p = 0.0011$ ), HSI ( $X^2_{(1,20)} = 6.00, p = 0.0213$ ) and age ( $X^2_{(1,20)} = 11.50, p =$   
418  $0.0044$ ) in the reference lake compared to individuals from Lake Denault. *Coregoninæ*  
419 (particularly Lake whitefish) collected in Lake Cherny displayed higher values of HSI ( $F_{(2,39)} =$   
420  $13.34, p < 0.0001$ ) compared to HSI values of individuals collected in the reference lake ( $p =$   
421  $0.0006$ ) and Lake Denault ( $p = 0.0006$ ).

### 422 3.3 Trace element bioaccumulation

423 Trace element concentrations of Al, Cd, Co, Cr, Cu, Fe, Hg, Mn, Pb, Se and Zn in the liver  
424 of *Salmoninæ*, *Catostominæ* and *Coregoninæ* are shown in Table 4, while liver concentrations of  
425 As, Ba and Ni are reported in Table S4.

426 ***Salmoninæ***

427 In *Salmoninæ*, liver concentrations of Al ( $X^2 = 28.20$ ,  $p < 0.0001$ ), Cd ( $F_{(3,44)} = 21.3$ ,  $p$   
428  $< 0.0001$ ), Co ( $F_{(3,44)} = 4.07$ ,  $p = 0.0124$ ), Cr ( $F_{(3,44)} = 29.88$ ,  $p < 0.0001$ ), Cu ( $F_{(3,44)} = 7.16$ ,  $p =$   
429  $0.0005$ ), THg ( $X^2 = 13.33$ ,  $p = 0.0040$ ), Pb ( $F_{(3,44)} = 6.46$ ,  $p = 0.0010$ ), Se ( $F_{(3,44)} = 66.64$ ,  $p <$   
430  $0.0001$ ) and Zn ( $F_{(3,40)} = 12.98$ ,  $p < 0.0001$ ) varied by lake. However, trace element concentrations  
431 were generally higher in *Salmoninæ* from Lake Houston compared to other lakes. For example,  
432 concentrations of Al, Cd and Cr were higher in fish collected from Lake Houston compared to  
433 *Salmoninæ* captured in the reference lake ( $p = 0.0001$ ,  $p < 0.0001$  and  $p < 0.0001$  respectively)  
434 and lakes Cherny ( $p = 0.0011$ ,  $p = 0.0007$  and  $p = 0.0004$  respectively) and Denault ( $p < 0.0001$ ,  
435  $p = 0.0122$  and  $p < 0.0001$  respectively). Liver Co concentrations were higher in *Salmoninæ*  
436 collected from Lake Houston compared to *Salmoninæ* sampled from Lake Denault ( $p = 0.0242$ ).  
437 The concentrations of Cu were greater in liver tissues sampled from Lake Houston than in those  
438 from the reference site ( $p = 0.0008$ ). Individuals captured from Lake Cherny exhibited higher  
439 values of THg than fish from the reference lake ( $p = 0.0076$ ) and Lake Houston ( $p = 0.0058$ ), while  
440 those collected in Lake Denault displayed greater THg concentrations than fish from the reference  
441 lake ( $p = 0.0042$ ) and Lake Houston ( $p = 0.0050$ ). Liver Pb concentrations were greater in fish  
442 collected from the reference lake ( $p = 0.0026$ ) and Lake Houston ( $p = 0.0017$ ) than those from  
443 Lake Denault. However, individuals from Lake Denault showed higher Se concentrations than  
444 *Salmoninæ* collected in the reference site ( $p < 0.0001$ ) and lakes Cherny ( $p = 0.0042$ ) and  
445 Houston ( $p < 0.0001$ ). The concentrations of Se were also greater in fish sampled in lakes  
446 Houston ( $p < 0.0001$ ) and Cherny ( $p = 0.0012$ ) compared to the reference lake. Finally, the  
447 concentrations of Zn were higher in fish captured in Lake Houston compared to the reference lake  
448 ( $p = 0.0013$ ), lakes Cherny ( $p = 0.0216$ ) and Denault ( $p < 0.0001$ ), while Zn concentrations of  
449 *Salmoninæ* from the reference site were greater than in fish from Lake Denault ( $p = 0.0056$ ).

**450 *Catostominæ***

451 In *Catostominæ*, concentrations of Cr, Pb and Zn did not vary significantly by site. Concentrations  
452 of Al ( $t_{(1,37)} = 6.01$ ,  $p < 0.0001$ ), Mn ( $X^2_{(1,37)} = 217.00$ ,  $p < 0.0001$ ) and Se ( $t_{(1,37)} = 2.48$ ,  $p = 0.0299$ )  
453 differed by sampling location, with fish from Lake Denault exhibiting significantly greater  
454 concentrations than *Catostominæ* from the reference site. In contrast, concentrations of Cd  
455 ( $X^2_{(1,37)} = 7.00$ ,  $p < 0.0001$ ), Co ( $t_{(1,37)} = -4.82$ ,  $p = 0.0011$ ), Cu ( $t_{(1,37)} = -2.83$ ,  $p = 0.0154$ ), Fe ( $t_{(1,37)}$   
456  $= -4.59$ ,  $p = 0.0005$ ) and THg ( $X^2_{(1,33)} = 0.00$ ,  $p < 0.0001$ ) were higher in *Catostominæ* collected  
457 in the reference lake than in fish from Lake Denault.

**458 *Coregoninæ***

459 In *Coregoninæ*, liver Al ( $F_{(2,42)} = 7.84$ ,  $p = 0.0013$ ), Cd ( $F_{(2,42)} = 19.84$ ,  $p < 0.0001$ ), Co ( $F_{(2,43)}$   
460  $= 13.01$ ,  $p < 0.0001$ ), Cr ( $F_{(2,43)} = 21.12$ ,  $p < 0.0001$ ), Cu ( $F_{(2,42)} = 14.90$ ,  $p < 0.0001$ ), Fe ( $F_{(2,43)} =$   
461  $4.45$ ,  $p = 0.0176$ ), THg ( $F_{(2,41)} = 11.41$ ,  $p = 0.0001$ ), Mn ( $F_{(2,43)} = 14.36$ ,  $p < 0.0001$ ), Pb ( $F_{(2,36)} =$   
462  $5.31$ ,  $p = 0.0096$ ), Se ( $F_{(2,43)} = 29.98$ ,  $p < 0.0001$ ) and Zn ( $F_{(2,40)} = 25.4$ ,  $p < 0.0001$ ) concentrations  
463 significantly differed by sampling locations. The concentrations of Al, Cd and Co were higher in  
464 samples from lakes Denault ( $p = 0.0038$ ,  $p = 0.0018$  and  $p < 0.0001$  respectively) and the  
465 reference site ( $p = 0.0133$ ,  $p < 0.0001$  and  $p = 0.0059$  respectively) than in liver tissue sampled  
466 from Lake Cherny. Liver Cr and Zn concentrations in sampled *Coregoninæ* from the reference  
467 lake were greater than in fish from lakes Cherny ( $p < 0.0001$  and  $p < 0.0001$  respectively) and  
468 Denault ( $p = 0.0096$  and  $p < 0.0001$  respectively). The concentrations of Cu and THg were higher  
469 in individuals captured in the reference lake ( $p < 0.0001$  and  $p = 0.0001$  respectively) and Lake  
470 Cherny ( $p = 0.0004$  and  $p = 0.0002$  respectively) than *Coregoninæ* captured in Lake Denault. The  
471 concentrations of Fe were greater in *Coregoninæ* from Lake Denault ( $p = 0.0278$ ) compared to  
472 *Coregoninæ* from Lake Cherny and *Coregoninæ* collected in the reference lake ( $p = 0.0017$ ) and  
473 Lake Denault ( $p < 0.0001$ ) expressed higher Mn concentrations than those from Lake Cherny.  
474 Individuals caught in the reference site exhibited higher Pb concentrations than individuals from

475 Lake Cherny ( $p = 0.0069$ ). The concentrations of Se in *Coregoninæ* from Lake Denault were  
476 greater than in *Coregoninæ* captured in the reference lake ( $p < 0.0001$ ) and Lake Cherny ( $p <$   
477  $0.0001$ ), while Se concentrations in liver tissue sampled from Lake Cherny were higher than in  
478 samples from the reference site ( $p = 0.0040$ ).

### 479 **3.4 Relationships between trace element concentrations and biomarkers**

480 The PCAs and Spearman correlations performed to assess the relationships between  
481 trace element concentrations and biomarkers are presented in Figure 3 and Table 5 for  
482 *Salmoninæ*, Figure 4 and Table 6 for *Catostominæ* and Figure 5 and Table 7 for *Coregoninæ*,  
483 respectively. See Table S5 for the results of enzyme activities (CAT, SOD and GPx activities) and  
484 biomarkers of oxidative stress (MDA and 8-OHdG concentrations).

#### 485 ***Salmoninæ***

486 The PCA computed for *Salmoninæ* led to two principal components explaining 60.6% of  
487 the total variability. However, an ellipse could not be drawn for *Salmoninæ* collected in Lake  
488 Cherny despite individuals having been collected in all four lakes as a result of a low sample size  
489 ( $n = 2$ ) in this location. The first dimension (Dim1) explained 33.3% of the overall variance and  
490 distinctly separated *Salmoninæ* captured in Lake Denault (on the left side of the axis) from  
491 *Salmoninæ* sampled in Lake Houston (on the right side of the axis). Individuals collected in the  
492 reference site (Lake Perroche) represent an overlap of other two populations. Fork length, total  
493 weight, THg and Se concentrations, age, FCF, HSI, and GPx activity correlated negatively with  
494 Dim1. On the other hand, the activity of CAT, MDA and 8-OHdG concentrations as well as  
495 concentrations of Fe, Cu, Co, Cd, Cr, Mn, Al and Zn correlated positively with Dim1. This first axis  
496 appeared to detail relationships dependent on fish growth, as Se and THg concentrations were  
497 highly associated to biometric variables (fork length, total weight, age and HSI) and GPx activity.

498 Results obtained from Spearman correlations further supported this trend, as positive  
499 correlations were observed between concentrations of THg and length, weight and age.  
500 Additionally, Se concentrations were positively correlated to age and 8-OHdG concentrations.  
501 The concentrations of MDA and 8-OHdG appeared to be linked with concentrations of Fe, Cu,  
502 Co, Cd and Cr, while CAT activity was associated with concentrations of Mn, Al and Zn. Spearman  
503 correlations also revealed that Cd concentrations displayed a negative correlation with FCF, while  
504 the MDA and 8-OHdG concentrations were positively correlated.

505 The second principal component (Dim2) explained 27.3% of the total variability observed  
506 for the examined variables. Fulton's condition factor and concentrations of Pb and Zn correlated  
507 negatively with Dim2 while all the other variables (except Al concentrations) correlated positively  
508 with Dim2. A particularly high loading of SOD activity was additionally observed. This second  
509 principal component separated *Salmoninæ* sampled in contaminated sites (lakes Denault and  
510 Houston) on the top of the axis from *Salmoninæ* collected at the reference site on the bottom of  
511 the axis.

#### 512 ***Catostominæ***

513 The PCA performed to evaluate relationships between trace element concentrations and  
514 biomarkers for *Catostominæ* led to two principal components explaining 54.6% of the total  
515 variability. Since *Catostominæ* were caught in two sampling sites only (the reference lake and  
516 Lake Denault), two ellipses are displayed on the biplot. The first principal component (Dim1)  
517 explained 36.8% of the total variability and strongly separated both populations of *Catostominæ*.  
518 The positive (right) side of Dim1 is defined by the high loading of FCF, fork length, total weight,  
519 age and concentrations of Cd and THg. With the exemption of Cd concentrations, these results  
520 mirror the similar pattern determined for *Salmoninæ*.

521 Spearman correlations support the strong positive relationship between THg and Cd  
522 concentrations and all biometric variables (total weight, fork length, FCF and age). Hepatosomatic



523 index, GPx and SOD activities, MDA concentrations and Co concentrations were also associated  
524 positively with Dim1. In contrast to our observations for *Salmoninæ*, Se concentrations did not  
525 express any significant correlations with biometric or biochemical biomarkers in *Catostominæ*.  
526 Concentrations of Mn, CAT activity and 8-OHdG concentrations loaded negatively on Dim1. The  
527 concentrations of Mn were negatively correlated to fork length and total weight. Additionally, Mn  
528 concentrations were strongly and negatively correlated with Cd and THg concentrations. Finally,  
529 the activity of GPx was positively correlated to MDA concentrations.

530 The second principal component (Dim2) explained 17.8% of the total variability and was  
531 characterized by the high positive loading of concentrations of Fe, Cr, Al and Pb. The  
532 concentrations of these metals were higher in *Catostominæ* from Lake Denault than in fish from  
533 the reference site, however, they did not appear to covary with biometric or biochemical  
534 biomarkers.

### 535 ***Coregoninæ***

536 The PCA performed for *Coregoninæ* led to two principal components explaining 52.2% of  
537 the overall variability. Three ellipses were determined representing each sampling site where  
538 *Coregoninæ* were captured (the reference site, lakes Cherny and Denault). The first principal  
539 component (Dim1) explained 29.9% of the total variance and patterns in the data appeared to be  
540 dictated by parameters indicative of fish growth, as total weight, fork length, age, FCF and THg,  
541 Cu, Cr and Cd concentrations were all positively correlated. Spearman correlations further  
542 support these observations as positive correlations were determined for both Cu and THg  
543 concentrations with fork length, total weight and age, in addition to FCF for THg concentrations.  
544 Concentrations of Se did not tend to generally covary with biometric variables. However, Mn  
545 concentrations were negatively correlated to fork length, total weight and FCF, as mostly identified  
546 for *Salmoninæ* and *Catostominæ*.

547           The activities of CAT, GPx and SOD, 8-OHdG and MDA concentrations, as well as  
548 concentrations of Al, Co, Fe, Mn, Pb and Se were negatively associated to Dim1. Additionally,  
549 CAT activity was positively correlated with MDA concentrations. Although Mn concentrations and  
550 CAT activity loaded similarly on the negative (left) side of Dim1, no significant correlation could  
551 be determined between these two variables.

552           The second principal component (Dim2) explained 22.3% of the observed total variability.  
553 Similar to observations for *Salmoninæ*, the majority of examined variables were positioned on the  
554 positive side (top) of the axis. Only HSI correlated negatively. In addition, Spearman values  
555 detailed negative correlation between HSI and concentrations of Cd. Finally, *Coregoninæ*  
556 collected in Lake Cherny appeared to be distinctly separated from fish collected in Lake Denault,  
557 though overlap with *Coregoninæ* from the reference lake was observed.

## 558 **4 DISCUSSION**

---

559 While a small sub-set of studies have described the environmental contamination resulting  
560 from iron mining operations in the Fermont (Jacques et al., 2020) and Schefferville (Aebischer et  
561 al., 2015; Laperrière et al., 2007) areas, as well as the effects of trace metal contamination from  
562 mining activities on fish elsewhere in the province of Québec (Couture et al., 2008a; Gauthier et  
563 al., 2009; Martyniuk et al., 2020b; Ponton et al., 2016; Pyle et al., 2008), scientific research  
564 detailing these subjects still remains limited in our region of study. Furthermore, with the  
565 exemption of a few studies that documented trace metal accumulation and the associated impacts  
566 (Amundsen et al., 2011; Moiseenko and Kudryavtseva, 2001) in other Northern regions, to the  
567 best of our knowledge there are no reports in the scientific literature reporting on the effects of  
568 trace element contamination on fish health and condition, especially antioxidant capacities and  
569 oxidative damage, near past and present iron ore exploitation sites in subarctic regions. Our study  
570 addresses this knowledge gap.

### 571 **4.1 Element concentrations in water and sediment among sampling sites**

572 Aqueous element concentrations varied among our study sites. The highest  
573 concentrations of Al and Fe were observed in Lake Perroche, our reference location. However,  
574 this can potentially be explained by the lower pH value measured in that lake, leading to greater  
575 trace metal solubility (Lofts et al., 2008). The relationship between pH and aqueous trace metal  
576 solubility may also explain the higher concentrations of Mn, Pb and Zn in Lake Houston in  
577 comparison to those recorded at other locations. Nevertheless, all aqueous element  
578 concentrations reported in any of our study lakes remained below the PWQO guidelines for  
579 Québec, which has been designed to protect aquatic organisms (Ministère de l'Environnement et  
580 de la Lutte contre les changements climatiques, 2021).

581 In sediment, concentrations of Cd, Cu, Se and Zn were higher in Lake Houston compared  
582 to the reference lake, where values for these elements were comparable with sediment  
583 concentrations determined by Couture et al. (2008b) at their reference sites. However, they  
584 reported element concentrations 20-fold higher for Cd, Se and Zn and over 600-fold higher for Cu  
585 in contaminated lakes from the Rouyn-Noranda area compared to Lake Houston. Mean values of  
586 Fe in Lake Houston were 10-fold higher than in the reference site and 5-fold greater than the  
587 highest values determined in twelve lakes by Pyle et al. (2005) in the Sudbury area. Accumulation  
588 of Fe in the sediment of Lake Houston clearly reflects the impact of iron mining activities in the  
589 Schefferville area, whereas Rouyn-Noranda and Sudbury areas were affected by copper and  
590 nickel mining operations. In addition to Fe contamination, lakes Houston and Denault displayed  
591 significantly higher concentrations of Mn in sediment compared to the reference site. A past study  
592 has indicated that iron smelting in Schefferville is associated with Mn, as Leslie and Bourque  
593 (1984) indicated that iron ore extracted from this location contained from trace amounts up to 10%  
594 Mn. Undoubtedly, Mn concentrations determined in sediment in both lakes can be attributed to  
595 iron ore exploitations in Schefferville. The comparison between element concentrations in  
596 sediment and water samples among our study sites provides insight into the long-term deposition  
597 processes of elements in sediment while aqueous element concentrations indicate short-term  
598 inputs, which is also supported by Pyle et al. (2005).

599 Overall, lakes from Schefferville (Denault and Houston) appeared to be more negatively  
600 affected by mining operations than Lake Cherny in Fermont and the reference location. Our data  
601 also clearly indicates the effect of distance on aqueous habitat contamination by trace elements  
602 from mining operations, as Lake Cherny is located farther from past and current mining operation  
603 sites than lakes Denault and Houston. In Lake Denault, while other elemental concentrations were  
604 similar to the reference lake, the high sediment concentrations of Mn reflect the legacy of past  
605 iron ore exploitations (IOC, closed in 1982). However, as Lake Houston is affected by current

606 (TSMC) and more recent (LIM, closed in 2014) mining activities, we observed higher  
607 concentrations of Cd, Cu, Fe, Mn, Se and Zn than in sediment from the reference lake. With this  
608 location, apparent short-term inputs, manifested by surface water from watersheds upstream and  
609 atmospheric transport are less pronounced.

## 610 **4.2 Element bioaccumulation among fish subfamilies and relationships with** 611 **biometric and biochemical biomarkers**

### 612 ***Salmoninæ***

613 In *Salmoninæ*, higher liver concentrations of THg were measured in fish from lakes  
614 Denault (Schefferville) and Cherny (Fermont) compared to the reference location. Low THg  
615 concentrations in the liver of *Salmoninæ* from Lake Houston were expected given the associations  
616 between THg and species dependent trophic position. Total mercury, and more specifically MeHg,  
617 is known to bioaccumulate and reach elevated concentrations in top predatory fish, such as Lake  
618 charr (Chételat et al., 2015; Lockhart et al., 2005). However, only Brook charr, a smaller salmonid,  
619 was present in Lake Houston. A shift in dietary preferences toward larger prey may explain the  
620 elevated hepatic THg concentrations observed in *Salmoninæ* from Lake Cherny. In our study,  
621 THg accumulation itself was not associated with any significant effect.

622 In Lake Houston, metal concentrations of Al, Cd, Co, Cr, Cu and Pb were more elevated  
623 in comparison to concentrations measured in *Salmoninæ* from the reference site. Furthermore,  
624 observed concentrations were greater than those detailed in previous reports of trace metal  
625 concentrations in the liver of another *Salmoninæ* species, Arctic charr (*Salvelinus alpinus*),  
626 collected in the Deception Bay region of Northern Québec (Martyniuk et al., 2020b) and other  
627 Northern regions of the circumpolar Arctic (Allen-Gil et al., 2003). These elevated concentrations  
628 may be explained by dietary metal contamination or by a slower growth in Lake Houston  
629 *Salmoninæ* due to a decrease in food availability. Both alternatives would be consistent with the  
630 heavy contamination of the sediment in this lake. In a study presented by Besser et al. (2001),

631 the authors demonstrated that different trophic levels (periphyton, benthic invertebrates and Brook  
632 charr) exhibited different responses to metal contamination, which they associated with foraging  
633 habits and food intake within each trophic level. The authors suggested that Cu concentrations  
634 measured in benthic invertebrates and Brook charr were linked to the larger scale negative  
635 impacts observed on fish populations and benthic invertebrate communities. Considering metal  
636 concentrations measured in sediment from Lake Houston, our results could indicate a comparable  
637 scenario. Furthermore, although Brook charr were longer, heavier and in a better condition in the  
638 reference lake than in Lake Houston, they were older in Lake Houston, indicating faster growth in  
639 fish from the reference site. As Lake Houston and the reference lake are experiencing a very  
640 similar seasonality given their geographical proximity, the overall poorer condition of Lake  
641 Houston *Salmoninæ* may be linked to indirect effects associated with reductions in food  
642 availability. Nevertheless, our sampling only involves a limited number of fish (n = 20), all captured  
643 in the same year. Therefore, inter-individual and inter-annual variability is expected given the  
644 environment under study.

645 *Salmoninæ* sampled in the Schefferville area, particularly those captured in Lake Denault,  
646 demonstrated high liver concentrations of Se compared to fish from the reference site, as well as  
647 previous studies on *Salmoninæ* species (Arribère et al., 2008; Herrmann et al., 2016). Similar to  
648 observations for liver THg, Se concentrations displayed a positive correlation with fish age,  
649 indicating that *Salmoninæ* bioaccumulate this element, most likely through dietary sources, since  
650 aqueous Se does not induce elemental bioaccumulation even in the most contaminated areas  
651 (Stewart et al., 2010; Wood et al., 2011a). Liver Se concentration was strongly and positively  
652 correlated to 8-OHdG concentrations, suggesting that Se is contributing to oxidative stress and  
653 resulting DNA damage in our sampled individuals (Spallholz, 1994; Spallholz et al., 2001).  
654 Although non-significant, the positive correlation of liver Se with SOD and GPx activities also  
655 suggests that this oxidative stress induced an antioxidant response. A review published by Lemly

656 (1993a) recommended that 12  $\mu\text{g/g}$  should be considered as a toxic threshold for freshwater  
657 fishes based upon the evaluation of numerous studies demonstrating negative impacts linked to  
658 liver Se accumulation between 12 and 86  $\mu\text{g/g}$ . *Salmoninæ* populations from Lake Denault, but  
659 also those from lakes Houston and Cherny, exceeded that threshold (liver Se concentrations  
660 ranging from 14 to 90  $\mu\text{g/g}$  for these lakes) indicating that fish populations at these locations may  
661 be subjected to elemental Se-induced toxicity. The very fine boundary between essentiality and  
662 toxicity of Se (Hamilton, 2004) represents a substantial environmental threat in our region of  
663 study, especially for *Salmoninæ*, which are particularly sensitive (Lemly, 1993a). However, given  
664 that the liver Se concentrations in our *Salmoninæ* were strongly and positively correlated with  
665 liver Cd, Cu and Hg concentrations, this field study cannot exclude that the increase in oxidative  
666 damage and antioxidant capacity that we observed in our system may be a response to the  
667 contaminant mixture.

668 Bioaccumulation of Se through the aquatic food web can lead to deleterious impacts in  
669 early-life stages of fish such as teratogenicity and ultimately lead to population level effects  
670 through recruitment impairment (Janz et al., 2010; Lemly, 1993b, 2004). Palace et al. (2004)  
671 suggested that oxidative stress in fish embryos could be a trigger for these effects. We could not  
672 evaluate the toxicity of Se on early life-stages of *Salmoninæ* in our study. However, Chapman  
673 (2007) demonstrated that Northern fish species could tolerate higher concentrations of Se than  
674 more temperately located species and recommends the adoption of a “site and species-specific  
675 studies” approach before drawing conclusions. Additionally, Se is known to have a protective  
676 effect against MeHg (Ralston et al., 2008) when the molar ratio between Se and Hg approaches  
677 or exceeds a 1:1 stoichiometry (Peterson et al., 2009; Ralston et al., 2007). Our results could  
678 suggest a preventive action of Se against Hg toxicity (Se/THg molar ratios  $> 1$ ). Since  
679 demethylation takes place in the liver of fish (Wood et al., 2011b) and we only measured THg and  
680 not MeHg, we do not know the proportion of THg that was in the methylated form in our samples.

681 Thus, it remains difficult to establish whether Se may be a protective or a toxic element given the  
682 elevated Se concentrations measured in this study, even for Northern species. In other  
683 circumstances (Se/Hg ratio < 1:1 stoichiometry), the very high affinity between Se and Hg induces  
684 Se-Hg bonds, potentially leading to a decrease in Se availability for vital functions (Ralston and  
685 Raymond, 2010).

### 686 ***Catostominæ***

687 *Catostominæ* in Lake Denault displayed significantly elevated concentrations of Mn  
688 compared to those from the reference site. Manganese is an essential trace metal for fish and is  
689 required for many biological functions (Goldhaber, 2003; Olmedo et al., 2013). However, more  
690 recently it has become an emerging contaminant of interest in ecotoxicological studies (Pinsino  
691 et al., 2012). The negative correlations determined between liver Mn concentration and biometric  
692 variables in *Catostominæ* (as well as in *Coregoninæ*) indicate that Mn does not bioaccumulate  
693 with age in fish liver. This is consistent with previous findings presented in Nussey (2000), that  
694 suggested that Mn is well regulated through homeostatic mechanisms. As described in Bendell-  
695 Young and Harvey (1986), at the water-sediment interface, Mn can be characterized as an  
696 intermediate state between dissolved and particulate forms. White sucker and Longnose sucker  
697 are both groundfish, feeding on benthic invertebrates. Their feeding behaviour induces a  
698 resuspension of particles in the upper sedimentary layer where their prey is located, leading to  
699 Mn contamination through both waterborne and dietary sources. Furthermore, several studies  
700 have reported that Mn has the potential to disturb the abundance and the diversity of benthic  
701 invertebrates (Dittman and Buchwalter, 2010; Wellnitz et al., 1994; Wilson et al., 2019). Therefore,  
702 in our study, a decrease in prey availability due to Mn contamination could potentially explain the  
703 lower condition observed in *Catostominæ* captured in Lake Denault, as these fish are directly  
704 exposed to metal contamination from both contact with sediment and food consumption (Bendell-  
705 Young and Harvey, 1986; Hamilton et al., 1990; Munkittrick and Dixon, 1988).



706 Similar to other essential and non-essential metals, excessive Mn accumulation can lead  
707 to immune response disruptions (Aliko et al., 2018) and induce oxidative stress (Copat et al.,  
708 2020; Hedayati et al., 2014) through the production of ROS or interference with antioxidant  
709 capacities. Under laboratory conditions, Vieira et al. (2012) exposed goldfish to aqueous Mn at  
710 0.1 and 1 mM (5.5 and 55 mg/L, respectively) for 96 h. They observed an increase of GPx activity  
711 and an inhibition of CAT activity in liver tissue. The authors hypothesized that GPx represented  
712 the first defense against waterborne Mn exposition. Their observation is supported by another  
713 study from Gabriel et al. (2013) where the authors exposed juvenile tambaqui (*Colossoma*  
714 *macropomum*) to aqueous Mn (3.9 mg/L Mn<sup>2+</sup>) for 96 hours. They reported a significant increase  
715 in liver glutathione-S-transferase (GST) activity and GSH content along with unchanged CAT  
716 activity. Their studies contrast strongly with the results of our study, in which wild fish were  
717 chronically exposed to water Mn concentrations ranging from 0.49 to 3.18 µg/L (3 orders of  
718 magnitude lower than the laboratory studies cited above), but also exposed to an unknown level  
719 of dietary Mn, and where CAT activity was stimulated, but GPx activity inhibited, by Mn. Although  
720 the comparison of results obtained from these studies with ours highlights the sharp contrast  
721 between short-term responses of antioxidant mechanisms observed in acute laboratory  
722 exposures and the long-term adaptations of animals chronically exposed to contaminants in the  
723 field, both support that exposure to high Mn concentrations leads to a response of antioxidant  
724 capacities. Our data allow to hypothesize that long-term (life-long) exposures to Mn like in our  
725 study induce catalase, inhibit GPx and lead to oxidative damage to DNA (as indicated by 8-OHdG)  
726 and metabolic costs. The negative correlations between liver Mn and Cd and THg concentrations  
727 strongly support that in the case of the *Catostominae* from our study area, Mn, and not other  
728 metals, is the metal of concern for the health of these fish.

729 Although liver Se concentration was higher in *Catostominae* from Lake Denault compared  
730 to individuals from the reference location, our study did not reveal any significant relationships

731 with the biomarkers of effects examined. This suggests that Se accumulation did not reach toxic  
732 levels in *Catostominae*. Additionally, liver Se concentrations in individuals from Lake Denault were  
733 below the toxic threshold of 12 µg/g proposed by Lemly (1993a), which supports our observations.  
734 Liver THg and Cd concentrations were higher in fish from the reference lake compared to Lake  
735 Denault and were positively correlated to biometric variables, clearly demonstrating the  
736 bioaccumulation potential of these two metals (Croteau et al., 2005; Kidd et al., 2012). As  
737 observed in *Salmoninae*, an increase of GPx activity in *Catostominae* appeared to efficiently  
738 prevent oxidative damage that may be induced by these elements, since the liver concentration  
739 of both Cd and Hg was negatively correlated with 8-OHdg concentration, an indicator of DNA  
740 oxidative damage.

#### 741 ***Coregoninae***

742 In our study, the *Coregoninae* subfamily was represented by Lake whitefish and Round  
743 whitefish, with both species being part of the *Salmonidae* family (Phillips and Rab, 2001).  
744 *Coregoninae* also share similar ecological characteristics with *Catostominae*, in terms of habitat  
745 and food preferences, even if *Coregoninae* can forage in deeper waters. These genetic and  
746 ecological similarities with both *Salmoninae* and *Catostominae*, respectively, were reflected in the  
747 observed patterns of liver element concentrations and their associated effects. As observed with  
748 *Catostominae*, *Coregoninae* from Lake Denault exhibited higher liver concentrations of Mn when  
749 compared to the reference site. Bioaccumulation of Mn also appeared to be regulated through  
750 cellular homeostasis, as suggested by the negative correlations between Mn concentrations and  
751 biometric biomarkers.

752 With *Coregoninae*, we reported negative correlations between HSI and liver concentrations  
753 of Cd. Past laboratory studies have demonstrated that fish exposed to both dietary and  
754 waterborne Cd displayed a decrease of HSI and disruptions to the antioxidant system (Drag-  
755 Kozak et al., 2019; Jamwal et al., 2018; Pereira et al., 1993; Pretto et al., 2011). Lake Cherny

756 presented higher aqueous and sediment Cd concentrations compared to the reference lake and  
757 Lake Denault. However, these elevated habitat concentrations appeared to not be reflected by  
758 liver Cd accumulation, since *Coregoninæ* from Lake Cherny exhibited lower concentrations of  
759 hepatic Cd compared to fish from the reference site and Lake Denault. Currently, our data does  
760 not provide an explanation for this trend. We propose two alternative hypotheses to explain this  
761 observation. An increased detoxification of intracellular Cd through GSH and metallothionein (MT)  
762 mediation in *Coregoninæ* from Lake Cherny, induced by chronic exposure, could decrease Cd  
763 accumulation (Chowdhury et al., 2005; Wood et al., 2011b). Alternately, an antagonistic action  
764 with calcium could lead to a decrease in Cd bioavailability (Brzóska and Moniuszko-Jakoniuk,  
765 1997), in agreement with our observation that Lake Cherny exhibited the highest conductivity  
766 value among the lakes studied.

767       As observed in *Salmoninæ*, *Coregoninæ* from lakes Cherny and Denault exhibited higher  
768 liver Se concentration than fish sampled in the reference site. Nevertheless, no correlation could  
769 be established between Se concentrations and biomarkers of oxidative stress or oxidative  
770 damage measured in this research. The absence of these associations provides further support  
771 for the hypothesis that cold-water species can tolerate higher Se bioaccumulation, as liver Se  
772 concentration in *Coregoninæ* from Lake Denault were above the potential toxic threshold of 12  
773 µg/g. Additionally, compared to fish collected in the reference site and Lake Cherny, *Coregoninæ*  
774 captured in Lake Denault displayed lower liver THg concentrations. In a study from Amundsen et  
775 al. (2011), Whitefish (*Coregonus lavaretus*) feeding on plankton rather than on benthic  
776 invertebrates exhibited higher Hg concentrations, suggesting that a pelagic feeding behaviour  
777 leads to an increase of Hg bioaccumulation. Two additional studies (Kahilainen et al., 2017; Keva  
778 et al., 2017) further supported this observation. This suggests that *Coregoninæ* from Lake Denault  
779 may be primarily feeding on benthic invertebrates, since both *Catostominæ* and *Coregoninæ* from

780 this lake expressed low liver THg and high Mn concentrations compared to their counterparts  
781 from the reference site.

## 782 **5 CONCLUSION**

---

783 Our study highlighted the interspecific variation of fish responses to contaminated  
784 environments near past and current mining areas in Northern Québec. Fish sampled in Lake  
785 Cherny (Fermont area) exhibited a generally better condition and appeared to be less impacted  
786 by trace element contamination than fish captured in lakes Houston and Denault (Schefferville  
787 area). Sediment Mn concentrations in Lake Denault and sediment concentrations of Cd, Cu, Fe,  
788 Mn, Se and Zn, in addition to water concentrations of Mn and Pb in Lake Houston were higher  
789 than those determined in the reference site. These observations can be attributed to the impacts  
790 of both historical and current mining operations, as iron ore exploitation in the Schefferville area  
791 has been more intense and precedes exploitation in the Fermont area. Trace element  
792 accumulation in the liver of the fish studied differed among subfamilies, particularly for Mn, Se  
793 and THg concentrations, and can most likely be related to trophic position and feeding behaviours.  
794 In Lake Denault, benthic fish from the *Catostominæ* subfamily appeared to be negatively affected  
795 by environmental Mn contamination, a legacy of past mining operations (IOC). This was evident  
796 through the evaluation of relationships between Mn concentrations and biomarkers of oxidative  
797 stress. *Salmoninæ* also exhibited indication of potential negative effects associated with  
798 contamination from mining activity, as high values of Se measured in fish from contaminated lakes  
799 compared to individuals from the reference lake were associated with an increase of 8-OHdG  
800 concentrations. In Lake Houston, *Salmoninæ* also demonstrated higher liver concentrations of Al,  
801 Cd, Co, Cr, Cu, Pb and lower condition than fish from the reference location. We could  
802 hypothesize that at sites impacted by more recent iron exploitation as in Lake Houston (LIM closed  
803 in 2014 and current TSMC activities), negative effects from sediment contamination are indirectly  
804 contributing to deteriorate fish health. Yet, as we did not collect information on prey abundance,  
805 food webs and since we have only one period of sampling, further research should be carried out  
806 to examine the magnitude of contamination upon benthic fauna. Lastly, fish collected in Lake

807 Cherny appeared to be in a similar condition and health status as those captured from Lake  
808 Perroche, the reference site. Only liver THg concentrations in fish from Lake Cherny were  
809 concerning and could be possibly explained by a shift in foraging strategies to larger prey items.  
810 It is important to consider that although strong correlations can be identified between liver trace  
811 element concentrations and biochemical biomarkers, field studies do not allow for clear attribution  
812 of an element's effect on the antioxidant system. The biomarkers under study are not entirely  
813 element-specific and in a natural ecosystem where environmental variables cannot be controlled  
814 as in laboratory studies, additive or antagonistic effects may occur. Further studies are required  
815 to have a better understanding of the impacts associated with mining activities on Northern fish  
816 species. Specifically, our study points to the relative importance of feeding behaviour on trace  
817 element concentrations in fish and their associated effects on fish health. Therefore, stable  
818 isotope analyses would be a valuable tool to estimate relationships between the parameters  
819 evaluated here and ecological variables such as trophic level and feeding behaviour.

820

## ACKNOWLEDGEMENTS

821 The authors wish to acknowledge Environment and Climate Change Canada's Environmental  
822 Damages Fund for funding this project and the Ministère des Forêts, de la Faune et des Parcs for  
823 determining fish ages. We thank Conrad André, our Innu guide in Schefferville, François  
824 Jinchereau, our bush pilot and adviser for the Fermont and Lake Perroche sampling, and the  
825 intern students (Christophe Dumoulin and Philippe Lagueux) who participated in field work. Lastly,  
826 we would like to express our gratitude to J. Perreault, J.F. Dutil and S. Prémont for their advice  
827 and direction in laboratory analyses.

828

829 **Competing interests:** The authors declare there are no competing interests.

830 **CRedit authorship contribution statement: Anthony Fontaine:** Methodology, Validation,  
831 Formal analysis, Investigation, Writing – Original Draft, Visualization; **Mackenzie Anne Clifford**  
832 **Martyniuk:** Methodology, Validation, Formal analysis, Investigation, Writing – Review & Editing;  
833 **Camille Garnier:** Investigation, Writing – Review & Editing; **Patrice Couture:** Conceptualization,  
834 Methodology, Validation, Investigation, Resources, Writing – Review & Editing, Supervision,  
835 Project administration, Funding acquisition

836 **Funding statement:** This project was funded by Environment and Climate Change Canada's  
837 Environmental Damages Fund (EDF-PQ-2017 J002).

### 838 **Data availability statement**

839 All untreated data for this study is available here:  
840 [https://borealisdata.ca/privateurl.xhtml?token=13b5b017-30f5-4cf3-8de4-](https://borealisdata.ca/privateurl.xhtml?token=13b5b017-30f5-4cf3-8de4-44ef8ae00c69)  
841 [44ef8ae00c69](https://borealisdata.ca/privateurl.xhtml?token=13b5b017-30f5-4cf3-8de4-44ef8ae00c69)

842 **REFERENCES**

- 
- 843 Aebischer, S., Cloquet, C., Carignan, J., Maurice, C., and Pienitz, R. 2015. Disruption of the  
844 geochemical metal cycle during mining: Multiple isotope studies of lake sediments from  
845 Schefferville, subarctic Québec. *Chemical Geology* **412**: 167-178.  
846 doi:10.1016/j.chemgeo.2015.07.028.
- 847 Aliko, V., Qirjo, M., Sula, E., Morina, V., and Faggio, C. 2018. Antioxidant defense system,  
848 immune response and erythron profile modulation in gold fish, *Carassius auratus*, after  
849 acute manganese treatment. *Fish & shellfish immunology* **76**: 101-109.
- 850 Allen-Gil, S.M., Ford, J., Lasorsa, B.K., Monetti, M., Vlasova, T., and Landers, D.H. 2003. Heavy  
851 metal contamination in the Taimyr Peninsula, Siberian Arctic. *Science of the Total*  
852 *Environment* **301**(1-3): 119-138.
- 853 Amundsen, P.-A., Kashulin, N.A., Terentjev, P., Gjelland, K.Ø., Koroleva, I.M., Dauvalter, V.A.,  
854 Sandimirov, S., Kashulin, A., and Knudsen, R. 2011. Heavy metal contents in whitefish  
855 (*Coregonus lavaretus*) along a pollution gradient in a subarctic watercourse.  
856 *Environmental Monitoring and Assessment* **182**(1): 301-316.
- 857 Arribére, M., Guevara, S.R., Bubach, D., Arcagni, M., and Vigliano, P. 2008. Selenium and  
858 mercury in native and introduced fish species of patagonian lakes, Argentina. *Biological*  
859 *Trace Element Research* **122**(1): 42-63.
- 860 Audet, D., and Couture, P. 2003. Seasonal variations in tissue metabolic capacities of yellow  
861 perch (*Perca flavescens*) from clean and metal-contaminated environments. *Canadian*  
862 *Journal of Fisheries and Aquatic Sciences* **60**(3): 269-278. doi:10.1139/f03-020.
- 863 Baeten, J., Langston, N., and Lafreniere, D. 2018. A spatial evaluation of historic iron mining  
864 impacts on current impaired waters in Lake Superior's Mesabi Range. *Ambio* **47**(2): 231-  
865 244.
- 866 Bartlett, M.S. 1937. Properties of sufficiency and statistical tests. *Proceedings of the Royal*  
867 *Society of London. Series A-Mathematical and Physical Sciences* **160**(901): 268-282.
- 868 Bendell-Young, L.I., and Harvey, H.H. 1986. Uptake and tissue distribution of manganese in the  
869 white sucker (*Catostomus commersoni*) under conditions of low pH. *Hydrobiologia* **133**(2):  
870 117-125.
- 871 Berntssen, M.H.G., Aatland, A., and Handy, R.D. 2003. Chronic dietary mercury exposure causes  
872 oxidative stress, brain lesions, and altered behaviour in Atlantic salmon (*Salmo salar*) parr.  
873 *Aquatic Toxicology* **65**(1): 55-72. doi:10.1016/s0166-445x(03)00104-8.
- 874 Besser, J.M., Brumbaugh, W.G., May, T.W., Church, S.E., and Kimball, B.A. 2001. Bioavailability  
875 of metals in stream food webs and hazards to brook trout (*Salvelinus fontinalis*) in the  
876 upper Animas River watershed, Colorado. *Archives of Environmental Contamination and*  
877 *Toxicology* **40**(1): 48-59.
- 878 Blais, A. 2019. Minière Tata Steel : plusieurs déversements suspects d'eaux rouges sèment  
879 l'inquiétude [Online article]. Available from  
880 [https://www.journaldequebec.com/2019/05/29/plusieurs-deversements-suspects-deaux-](https://www.journaldequebec.com/2019/05/29/plusieurs-deversements-suspects-deaux-rouges-denonces)  
881 [rouges-denonces](https://www.journaldequebec.com/2019/05/29/plusieurs-deversements-suspects-deaux-rouges-denonces) [accessed October 29, 2020].



- 882 Boutet, J.-S. 2012. Développement ferrifère et mondes autochtones au Québec subarctique,  
883 1954-1983. *Recherches amérindiennes au Québec* **40**(3): 35-52. doi:10.7202/1009367ar.
- 884 Bradford, M.M. 1976. A rapid and sensitive method for the quantitation of microgram quantities of  
885 protein utilizing the principle of protein-dye binding. *Analytical Biochemistry* **72**(1-2): 248-  
886 254.
- 887 Brzóska, M.M., and Moniuszko-Jakoniuk, J. 1997. The influence of calcium content in diet on  
888 cumulation and toxicity of cadmium in the organism Review. *Archives of toxicology* **72**(2):  
889 63-73.
- 890 Chapman, P.M. 2007. Selenium thresholds for fish from cold freshwaters. *Human and Ecological*  
891 *Risk Assessment* **13**(1): 20-24.
- 892 Chételat, J., Amyot, M., Arp, P., Blais, J.M., Depew, D., Emmerton, C.A., Evans, M., Gamberg,  
893 M., Gantner, N., Girard, C., Graydon, J., Kirk, J., Lean, D., Lehnherr, I., Muir, D., Nasr, M.,  
894 Poulain, A.J., Power, M., Roach, P., Stern, G., Swanson, H., and van der Velden, S. 2015.  
895 Mercury in freshwater ecosystems of the Canadian Arctic: recent advances on its cycling  
896 and fate [Research Support, Non-U.S. Gov't, Review]. *The Science of the Total*  
897 *Environment* **509-510**: 41-66. doi:10.1016/j.scitotenv.2014.05.151.
- 898 Chowdhury, M., Baldisserotto, B., and Wood, C. 2005. Tissue-specific cadmium and  
899 metallothionein levels in rainbow trout chronically acclimated to waterborne or dietary  
900 cadmium. *Archives of Environmental Contamination and Toxicology* **48**(3): 381-390.
- 901 CLD de Caniapiscou. 2015. Profil démographique et socioéconomique Fermont 2015. Available  
902 from <http://caniapiscou.net/files/view/f7f5fc8cdc8094f> [accessed November 01, 2020].
- 903 Copat, C., Rizzo, M., Zuccaro, A., Grasso, A., Zuccarello, P., Fiore, M., Mancini, G., and Ferrante,  
904 M. 2020. Metals/metalloids and oxidative status markers in saltwater fish from the Ionic  
905 Coast of Sicily, Mediterranean Sea. *International Journal of Environmental Research*  
906 **14**(1): 15-27.
- 907 Couture, P., and Kumar, P.R. 2003. Impairment of metabolic capacities in copper and cadmium  
908 contaminated wild yellow perch (*Perca flavescens*). *Aquatic Toxicology* **64**(1): 107-120.
- 909 Couture, P., and Pyle, G. 2008. Live fast and die young: metal effects on condition and physiology  
910 of wild Yellow perch from along two metal contamination gradients. *Human and Ecological*  
911 *Risk Assessment: An International Journal* **14**(1): 73-96.  
912 doi:10.1080/10807030701790322.
- 913 Couture, P., and Pyle, G. 2012. Field studies on metal accumulation and effects in fish. pp. 417-  
914 473.
- 915 Couture, P., Rajotte, J.W., and Pyle, G.G. 2008a. Seasonal and regional variations in metal  
916 contamination and condition indicators in Yellow perch (*Perca flavescens*) along two  
917 polymetallic gradients. III. Energetic and physiological indicators. *Human and Ecological*  
918 *Risk Assessment: An International Journal* **14**(1): 146-165.  
919 doi:10.1080/10807030701790314.
- 920 Couture, P., Busby, P., Gauthier, C., Rajotte, J.W., and Pyle, G.G. 2008b. Seasonal and regional  
921 variations of metal contamination and condition indicators in Yellow perch (*Perca*  
922 *flavescens*) along two polymetallic gradients. I. Factors influencing tissue metal  
923 concentrations. *Human and Ecological Risk Assessment: An International Journal* **14**(1):  
924 97-125. doi:10.1080/10807030701790330.

- 925 Croteau, M.N., Luoma, S.N., and Stewart, A.R. 2005. Trophic transfer of metals along freshwater  
926 food webs: evidence of cadmium biomagnification in nature. *Limnology and*  
927 *Oceanography* **50**(5): 1511-1519.
- 928 da Silva, A.P.V., Silva, A.O., de Lima, F.R.D., Benedet, L., de Jesus Franco, A., de Souza, J.K.,  
929 Júnior, A.C.R., Batista, É.R., Inda, A.V., and Curi, N. 2022. Potentially toxic elements in  
930 iron mine tailings: Effects of reducing soil pH on available concentrations of toxic elements.  
931 *Environmental Research* **215**: 114321.
- 932 Di Giulio, R.T., Washburn, P.C., Wenning, R.J., Winston, G.W., and Jewell, C.S. 1989.  
933 Biochemical responses in aquatic animals: a review of determinants of oxidative stress.  
934 *Environmental Toxicology and Chemistry: An International Journal* **8**(12): 1103-1123.
- 935 Dittman, E.K., and Buchwalter, D.B. 2010. Manganese bioconcentration in aquatic insects: Mn  
936 oxide coatings, molting loss, and Mn (II) thiol scavenging. *Environmental Science &*  
937 *Technology* **44**(23): 9182-9188.
- 938 Drag-Kozak, E., Pawlica-Gosiewska, D., Gawlik, K., Socha, M., Gosiewski, G., Łuszczek-Trojnar,  
939 E., Solnica, B., and Popek, W. 2019. Cadmium-induced oxidative stress in Prussian carp  
940 (*Carassius gibelio* Bloch) hepatopancreas: ameliorating effect of melatonin.  
941 *Environmental Science and Pollution Research* **26**(12): 12264-12279.
- 942 Duchesne, J.-F., and Magnan, P. 1997. The use of climate classification parameters to investigate  
943 geographical variations in the life history traits of ectotherms, with special reference to the  
944 white sucker (*Catostomus commersoni*). *Ecoscience* **4**(2): 140-150.
- 945 Eastwood, S., and Couture, P. 2002. Seasonal variations in condition and liver metal  
946 concentrations of yellow perch (*Perca flavescens*) from a metal-contaminated  
947 environment. *Aquatic Toxicology* **58**(1-2): 43-56. doi:10.1016/s0166-445x(01)00218-1.
- 948 EPA, U. 1998. Method 7473 (SW-846): mercury in solids and solutions by thermal decomposition,  
949 amalgamation, and atomic absorption spectrophotometry, Revision 0. Washington, DC  
950 **17**.
- 951 Farkas, A., Salánki, J., and Specziár, A. 2003. Age- and size-specific patterns of heavy metals in  
952 the organs of freshwater fish *Abramis brama* L. populating a low-contaminated site. *Water*  
953 *research* **37**(5): 959-964.
- 954 Friedmann, A.S., Watzin, M.C., Brinck-Johnsen, T., and Leiter, J.C. 1996. Low levels of dietary  
955 methylmercury inhibit growth and gonadal development in juvenile walleye (*Stizostedion*  
956 *vitreum*). *Aquatic Toxicology* **35**(3-4): 265-278.
- 957 Gabriel, D., Riffel, A.P.K., Finamor, I.A., Saccol, E.M., Ourique, G.M., Goulart, L.O., Kochhann,  
958 D., Cunha, M.A., Garcia, L.O., and Pavanato, M.A. 2013. Effects of subchronic  
959 manganese chloride exposure on tambaqui (*Colossoma macropomum*) tissues: oxidative  
960 stress and antioxidant defenses. *Archives of Environmental Contamination and*  
961 *Toxicology* **64**(4): 659-667.
- 962 Gabriel, F.Â., Hauser-Davis, R.A., Soares, L., Mazzuco, A.C.A., Rocha, R.C.C., Saint Pierre, T.D.,  
963 Saggiaro, E., Correia, F.V., Ferreira, T.O., and Bernardino, A.F. 2020. Contamination and  
964 oxidative stress biomarkers in estuarine fish following a mine tailing disaster. *PeerJ* **8**:  
965 e10266.
- 966 Gantner, N., Muir, D.C., Power, M., Iqaluk, D., Reist, J.D., Babaluk, J.A., Meili, M., Borg, H.,  
967 Hammar, J., and Michaud, W. 2010. Mercury concentrations in landlocked Arctic char  
968 (*Salvelinus alpinus*) from the Canadian Arctic. Part II: influence of lake biotic and abiotic

- 969 characteristics on geographic trends in 27 populations. *Environmental Toxicology and*  
970 *Chemistry: An International Journal* **29**(3): 633-643.
- 971 Gauthier, C., Campbell, P.G., and Couture, P. 2009. Condition and pyloric caeca as indicators of  
972 food web effects in fish living in metal-contaminated lakes [Research Support, Non-U.S.  
973 Gov't]. *Ecotoxicology and Environmental Safety* **72**(8): 2066-2074.  
974 doi:10.1016/j.ecoenv.2009.08.005.
- 975 Goldhaber, S.B. 2003. Trace element risk assessment: essentiality vs. toxicity. *Regulatory*  
976 *toxicology and pharmacology* **38**(2): 232-242.
- 977 Gouvernement du Canada. 2014. Amende de 7,5 millions de dollars à Bloom Lake General  
978 Partner pour infractions environnementales. Available from  
979 [https://www.canada.ca/fr/environnement-changement-climatique/services/application-](https://www.canada.ca/fr/environnement-changement-climatique/services/application-lois-environnementales/notifications/bloom-lake-general-partner-infractions.html)  
980 [lois-environnementales/notifications/bloom-lake-general-partner-infractions.html](https://www.canada.ca/fr/environnement-changement-climatique/services/application-lois-environnementales/notifications/bloom-lake-general-partner-infractions.html)  
981 [accessed October 20, 2020].
- 982 Gouvernement du Canada. 2020a. Rapport de données quotidiennes pour juillet 2019,  
983 Schefferville, Québec. Available from  
984 [https://climat.meteo.gc.ca/climate\\_data/daily\\_data\\_f.html?StationID=49649&timeframe=](https://climat.meteo.gc.ca/climate_data/daily_data_f.html?StationID=49649&timeframe=2&StartYear=1840&EndYear=2020&Day=16&Year=2019&Month=1)  
985 [2&StartYear=1840&EndYear=2020&Day=16&Year=2019&Month=1](https://climat.meteo.gc.ca/climate_data/daily_data_f.html?StationID=49649&timeframe=2&StartYear=1840&EndYear=2020&Day=16&Year=2019&Month=1) [accessed  
986 November 01, 2020].
- 987 Gouvernement du Canada. 2020b. Rapport de données quotidiennes pour janvier 2019,  
988 Schefferville, Québec. Available from  
989 [https://climat.meteo.gc.ca/climate\\_data/daily\\_data\\_f.html?StationID=49649&timeframe=](https://climat.meteo.gc.ca/climate_data/daily_data_f.html?StationID=49649&timeframe=2&StartYear=1840&EndYear=2020&Day=16&Year=2019&Month=1)  
990 [2&StartYear=1840&EndYear=2020&Day=16&Year=2019&Month=1](https://climat.meteo.gc.ca/climate_data/daily_data_f.html?StationID=49649&timeframe=2&StartYear=1840&EndYear=2020&Day=16&Year=2019&Month=1) [accessed  
991 November 01, 2020].
- 992 Greenfield, B.K., Hrabik, T.R., Harvey, C.J., and Carpenter, S.R. 2001. Predicting mercury levels  
993 in yellow perch: use of water chemistry, trophic ecology, and spatial traits. *Canadian*  
994 *Journal of Fisheries and Aquatic Sciences* **58**(7): 1419-1429.
- 995 Halliwell, B. 1999. Antioxidant defence mechanisms: from the beginning to the end (of the  
996 beginning). *Free Radical Research* **31**(4): 261-272.
- 997 Hamilton, S.J. 2004. Review of selenium toxicity in the aquatic food chain. *Science of the Total*  
998 *Environment* **326**(1-3): 1-31.
- 999 Hamilton, S.J., Buhl, K.J., Faerber, N.L., Bullard, F.A., and Wiedmeyer, R.H. 1990. Toxicity of  
1000 organic selenium in the diet to chinook salmon. *Environmental Toxicology and Chemistry: An International Journal* **9**(3): 347-358.
- 1002 Hedayati, A., Hoseini, S.M., and Ghelichpour, M. 2014. Acute toxicity of waterborne manganese  
1003 to *Rutilus caspicus* (Yakovlev, 1870)—gill histopathology, immune indices, oxidative  
1004 condition, and saltwater resistance. *Toxicological & Environmental Chemistry* **96**(10):  
1005 1535-1545.
- 1006 Herrmann, S., Nimmo, D., Carsella, J., Herrmann-Hoesing, L., Turner, J., Gregorich, J., Heuvel,  
1007 B.V., Nehring, R., and Foutz, H. 2016. Differential accumulation of mercury and selenium  
1008 in brown trout tissues of a high-gradient urbanized stream in Colorado, USA. *Archives of*  
1009 *Environmental Contamination and Toxicology* **70**(2): 204-218.
- 1010 Houde, E.D. 1989. Comparative growth, mortality, and energetics of marine fish larvae:  
1011 temperature and implied latitudinal effects. *Fishery Bulletin* **87**.

- 1012 Jacques, O., Pienitz, R., and Ibrahim, G. 2020. Paleolimnological assessment of long-term  
1013 changes in a boreal recreational lake of the Fermont mining region (subarctic Quebec,  
1014 Canada). *Lake and Reservoir Management*: 1-21.
- 1015 Jamwal, A., Lemire, D., Driessnack, M., Naderi, M., and Niyogi, S. 2018. Interactive effects of  
1016 chronic dietary selenomethionine and cadmium exposure in rainbow trout (*Oncorhynchus*  
1017 *mykiss*): A preliminary study. *Chemosphere* **197**: 550-559.
- 1018 Janz, D.M., DeForest, D.K., Brooks, M.L., Chapman, P.M., Gilron, G., Hoff, D., Hopkins, W.A.,  
1019 McIntyre, D.O., Mebane, C.A., and Palace, V.P. 2010. Selenium toxicity to aquatic  
1020 organisms. *Ecological assessment of selenium in the aquatic environment*: 141-231.
- 1021 Jobling, M., Wood, C.M., and McDonald, D.G. 1997. Temperature and growth: modulation of  
1022 growth rate via temperature change. *In* *Global Warming*. pp. 225-254.
- 1023 Jørgensen, C., and Fiksen, Ø. 2006. State-dependent energy allocation in cod (*Gadus morhua*).  
1024 *Canadian Journal of Fisheries and Aquatic Sciences* **63**(1): 186-199. doi:10.1139/f05-209.
- 1025 Kahilainen, K.K., Thomas, S.M., Nystedt, E.K., Keva, O., Malinen, T., and Hayden, B. 2017.  
1026 Ecomorphological divergence drives differential mercury bioaccumulation in polymorphic  
1027 European whitefish (*Coregonus lavaretus*) populations of subarctic lakes. *Science of the*  
1028 *Total Environment* **599**: 1768-1778.
- 1029 Karthikeyan, S., Palaniappan, P., and Sabhanayakam, S. 2007. Influence of pH and water  
1030 hardness upon nickel accumulation in edible fish *Cirrhinus mrigala*. *Journal of*  
1031 *Environmental Biology* **28**(2): 489.
- 1032 Keva, O., Hayden, B., Harrod, C., and Kahilainen, K.K. 2017. Total mercury concentrations in  
1033 liver and muscle of European whitefish (*Coregonus lavaretus* (L.)) in a subarctic lake-  
1034 Assessing the factors driving year-round variation. *Environmental Pollution* **231**: 1518-  
1035 1528.
- 1036 Kidd, K., Clayden, M., and Jardine, T. 2012. Bioaccumulation and biomagnification of mercury  
1037 through food webs. *Environmental chemistry and toxicology of mercury*. Wiley, Hoboken:  
1038 455-499.
- 1039 Köck, G., Triendl, M., and Hofer, R. 1996. Seasonal patterns of metal accumulation in Arctic char  
1040 (*Salvelinus alpinus*) from an oligotrophic Alpine lake related to temperature. *Canadian*  
1041 *Journal of Fisheries and Aquatic Sciences* **53**(4): 780-786.
- 1042 Kruskal, W.H., and Wallis, W.A. 1952. Use of ranks in one-criterion variance analysis. *Journal of*  
1043 *the American statistical Association* **47**(260): 583-621.
- 1044 Kuchapski, K.A., and Rasmussen, J.B. 2015. Food chain transfer and exposure effects of  
1045 selenium in salmonid fish communities in two watersheds in the Canadian Rocky  
1046 Mountains. *Canadian Journal of Fisheries and Aquatic Sciences* **72**(7): 955-967.
- 1047 Lackner, R. 1998. "Oxidative stress" in fish by environmental pollutants. *In* *Fish Ecotoxicology*.  
1048 Springer. pp. 203-224.
- 1049 Laflamme, J.-S., Couillard, Y., Campbell, P.G., and Hontela, A. 2000. Interrenal metallothionein  
1050 and cortisol secretion in relation to Cd, Cu, and Zn exposure in yellow perch, *Perca*  
1051 *flavescens*, from Abitibi lakes. *Canadian Journal of Fisheries and Aquatic Sciences* **57**(8):  
1052 1692-1700.
- 1053 Laperrière, L., Fallu, M.-A., Hausmann, S., Pienitz, R., and Muir, D. 2007. Paleolimnological  
1054 evidence of mining and demographic impacts on Lac Dauriat, Schefferville (subarctic

- 1055 Québec, Canada). *Journal of Paleolimnology* **40**(1): 309-324. doi:10.1007/s10933-007-  
1056 9162-6.
- 1057 Larocque, I., Pienitz, R., and Rolland, N. 2006. Factors influencing the distribution of chironomids  
1058 in lakes distributed along a latitudinal gradient in northwestern Quebec, Canada. *Canadian*  
1059 *Journal of Fisheries and Aquatic Sciences* **63**(6): 1286-1297.
- 1060 Lemly, A.D. 1993a. Guidelines for evaluating selenium data from aquatic monitoring and  
1061 assessment studies. *Environmental Monitoring and Assessment* **28**(1): 83-100.
- 1062 Lemly, A.D. 1993b. Teratogenic effects of selenium in natural populations of fresh water fish.  
1063 *Ecotoxicology and Environmental Safety* **26**(2): 181-204.
- 1064 Lemly, A.D. 2004. Aquatic selenium pollution is a global environmental safety issue.  
1065 *Ecotoxicology and Environmental Safety* **59**(1): 44-56.
- 1066 Leslie, K., and Bourque, Y. 1984. Géologie et métallogénie de la Gaspésie et de la région de la  
1067 fosse du Labrador. Le Manganèse dans la région de Schefferville. Ministère de l'énergie  
1068 et des ressources: 121-125.
- 1069 Levene, H. 1961. Robust tests for equality of variances. *Contributions to probability and statistics.*  
1070 *Essays in honor of Harold Hotelling*: 279-292.
- 1071 Levesque, H., Moon, T., Campbell, P., and Hontela, A. 2002. Seasonal variation in carbohydrate  
1072 and lipid metabolism of yellow perch (*Perca flavescens*) chronically exposed to metals in  
1073 the field. *Aquatic Toxicology* **60**(3-4): 257-267.
- 1074 Livingstone, D. 2001. Contaminant-stimulated reactive oxygen species production and oxidative  
1075 damage in aquatic organisms. *Marine Pollution Bulletin* **42**(8): 656-666.
- 1076 Lockhart, W., Stern, G., Low, G., Hendzel, M., Boila, G., Roach, P., Evans, M., Billeck, B.,  
1077 DeLaronde, J., and Friesen, S. 2005. A history of total mercury in edible muscle of fish  
1078 from lakes in northern Canada. *Science of the Total Environment* **351**: 427-463.
- 1079 Lofts, S., Tipping, E., and Hamilton-Taylor, J. 2008. The chemical speciation of Fe (III) in  
1080 freshwaters. *Aquatic Geochemistry* **14**(4): 337-358.
- 1081 Maes, G.E., Raeymaekers, J.A., Pampoulie, C., Seynaeve, A., Goemans, G., Belpaire, C., and  
1082 Volckaert, F. 2005. The catadromous European eel *Anguilla anguilla* (L.) as a model for  
1083 freshwater evolutionary ecotoxicology: relationship between heavy metal  
1084 bioaccumulation, condition and genetic variability. *Aquatic Toxicology* **73**(1): 99-114.
- 1085 Makivik Coporation. 2021. Nunavik Research Centre. Available from  
1086 <https://www.makivik.org/nunavik-research-centre/> [accessed July 20, 2022].
- 1087 Martyniuk, M.A.C., Couture, P., Tran, L., Beaupre, L., and Power, M. 2020a. Seasonal variation  
1088 of total mercury and condition indices of Arctic charr (*Salvelinus alpinus*) in Northern  
1089 Quebec, Canada. *Science of the Total Environment* **738**: 139450.  
1090 doi:10.1016/j.scitotenv.2020.139450.
- 1091 Martyniuk, M.A.C., Couture, P., Tran, L., Beaupre, L., Urien, N., and Power, M. 2020b. A seasonal  
1092 comparison of trace metal concentrations in the tissues of Arctic charr (*Salvelinus alpinus*)  
1093 in Northern Quebec, Canada. *Ecotoxicology*. doi:10.1007/s10646-020-02248-7.
- 1094 McPhee, D.L., and Janz, D.M. 2014. Dietary selenomethionine exposure alters swimming  
1095 performance, metabolic capacity and energy homeostasis in juvenile fathead minnow.  
1096 *Aquatic Toxicology* **155**: 91-100.

- 1097 Ministère de l'Environnement et de la Lutte contre les changements climatiques. 2020. Normales  
1098 climatiques du Québec 1981-2010. Available from  
1099 <http://www.environnement.gouv.qc.ca/climat/normales/sommaire.asp> [accessed  
1100 November 01, 2020].
- 1101 Ministère de l'Environnement et de la Lutte contre les changements climatiques. 2021. Critères  
1102 de qualité de l'eau de surface. Available from  
1103 [https://www.environnement.gouv.qc.ca/eau/criteres\\_eau/index.asp](https://www.environnement.gouv.qc.ca/eau/criteres_eau/index.asp) [accessed February  
1104 16, 2021].
- 1105 Moiseenko, T., and Kudryavtseva, L. 2001. Trace metal accumulation and fish pathologies in  
1106 areas affected by mining and metallurgical enterprises in the Kola Region, Russia.  
1107 *Environmental Pollution* **114**(2): 285-297.
- 1108 Muir, D., Wang, X., Bright, D., Lockhart, L., and Köck, G. 2005. Spatial and temporal trends of  
1109 mercury and other metals in landlocked char from lakes in the Canadian Arctic  
1110 archipelago. *Science of the Total Environment* **351**: 464-478.
- 1111 Munkittrick, K.R., and Dixon, D.G. 1988. Growth, fecundity, and energy stores of White Sucker  
1112 (*Catostomus commersoni*) from lakes containing elevated levels of copper and zinc.  
1113 *Canadian Journal of Fisheries and Aquatic Sciences* **45**(8): 1355-1365. doi:10.1139/f88-  
1114 159.
- 1115 Nussey, H. 2000. Bioaccumulation of chromium, manganese, nickel and lead in the tissues of the  
1116 moggel, *Labeo umbratus* (Cyprinidae), from Witbank Dam, Mpumalanga. *Water SA* **26**(2):  
1117 269-284.
- 1118 Olmedo, P., Hernández, A., Pla, A., Femia, P., Navas-Acien, A., and Gil, F. 2013. Determination  
1119 of essential elements (copper, manganese, selenium and zinc) in fish and shellfish  
1120 samples. Risk and nutritional assessment and mercury–selenium balance. *Food and*  
1121 *Chemical Toxicology* **62**: 299-307.
- 1122 Palace, V.P., Spallholz, J.E., Holm, J., Wautier, K., Evans, R.E., and Baron, C.L. 2004.  
1123 Metabolism of selenomethionine by rainbow trout (*Oncorhynchus mykiss*) embryos can  
1124 generate oxidative stress. *Ecotoxicology and Environmental Safety* **58**(1): 17-21.
- 1125 Passos, L.S., Gnocchi, K.G., Pereira, T.M., Coppo, G.C., Cabral, D.S., and Gomes, L.C. 2020. Is  
1126 the Doce River elutriate or its water toxic to *Astyanax lacustris* (Teleostei: Characidae)  
1127 three years after the Samarco mining dam collapse? *Science of the Total Environment*  
1128 **736**: 139644.
- 1129 Pereira, J., Mercaldo-Allen, R., Kuropat, C., Luedke, D., and Sennefelder, G. 1993. Effect of  
1130 cadmium accumulation on serum vitellogenin levels and hepatosomatic and  
1131 gonadosomatic indices of winter flounder (*Pleuronectes americanus*). *Archives of*  
1132 *Environmental Contamination and Toxicology* **24**(4): 427-431.
- 1133 Peterson, S.A., Ralston, N.V., Whanger, P.D., Oldfield, J.E., and Mosher, W.D. 2009. Selenium  
1134 and mercury interactions with emphasis on fish tissue. *Environmental Bioindicators* **4**(4):  
1135 318-334.
- 1136 Phillips, R., and Rab, P. 2001. Chromosome evolution in the Salmonidae (Pisces): an update.  
1137 *Biological Reviews* **76**(1): 1-25.
- 1138 Pinsino, A., Matranga, V., and Roccheri, M.C. 2012. Manganese: a new emerging contaminant in  
1139 the environment. *Environmental Contamination*: 17-36.

- 1140 Ponton, D.E., Caron, A., Hare, L., and Campbell, P.G.C. 2016. Hepatic oxidative stress and metal  
1141 subcellular partitioning are affected by selenium exposure in wild yellow perch (*Perca*  
1142 *flavescens*). *Environmental Pollution* **214**: 608-617. doi:10.1016/j.envpol.2016.04.051.
- 1143 Power, G. 1978. Fish population structure in Arctic lakes. *Journal of the Fisheries Board of*  
1144 *Canada* **35**(1): 53-59. doi:DOI 10.1139/f78-008.
- 1145 Pretto, A., Loro, V.L., Baldisserotto, B., Pavanato, M.A., Moraes, B.S., Menezes, C., Cattaneo,  
1146 R., Clasen, B., Finamor, I.A., and Dressler, V. 2011. Effects of water cadmium  
1147 concentrations on bioaccumulation and various oxidative stress parameters in *Rhamdia*  
1148 *quelen*. *Archives of Environmental Contamination and Toxicology* **60**(2): 309-318.
- 1149 Pyle, G., Rajotte, J., and Couture, P. 2005. Effects of industrial metals on wild fish populations  
1150 along a metal contamination gradient [Research Support, Non-U.S. Gov't]. *Ecotoxicology*  
1151 *and Environmental Safety* **61**(3): 287-312. doi:10.1016/j.ecoenv.2004.09.003.
- 1152 Pyle, G., Busby, P., Gauthier, C., Rajotte, J., and Couture, P. 2008. Seasonal and regional  
1153 variations in metal contamination and condition indicators in Yellow Perch (*Perca*  
1154 *flavescens*) along two polymetallic gradients. II. Growth patterns, longevity, and condition.  
1155 *Human and Ecological Risk Assessment: An International Journal* **14**(1): 126-145.  
1156 doi:10.1080/10807030701790504.
- 1157 Ralston, N.V., and Raymond, L.J. 2010. Dietary selenium's protective effects against  
1158 methylmercury toxicity. *Toxicology* **278**(1): 112-123.
- 1159 Ralston, N.V., Blackwell, J.L., and Raymond, L.J. 2007. Importance of molar ratios in selenium-  
1160 dependent protection against methylmercury toxicity. *Biological Trace Element Research*  
1161 **119**(3): 255-268.
- 1162 Ralston, N.V., Ralston, C.R., Blackwell III, J.L., and Raymond, L.J. 2008. Dietary and tissue  
1163 selenium in relation to methylmercury toxicity. *Neurotoxicology* **29**(5): 802-811.
- 1164 Scott, G.R., and Sloman, K.A. 2004. The effects of environmental pollutants on complex fish  
1165 behaviour: integrating behavioural and physiological indicators of toxicity. *Aquatic*  
1166 *Toxicology* **68**(4): 369-392.
- 1167 Sevcikova, M., Modra, H., Slaninova, A., and Svobodova, Z. 2011. Metals as a cause of oxidative  
1168 stress in fish: a review. *Veterinarni Medicina* **56**(11): 537-546.
- 1169 Shapiro, S.S., and Wilk, M.B. 1965. An analysis of variance test for normality (complete  
1170 samples)†. *Biometrika* **52**(3-4): 591-611. doi:10.1093/biomet/52.3-4.591.
- 1171 Sherwood, G.D., Rasmussen, J.B., Rowan, D.J., Brodeur, J., and Hontela, A. 2000. Bioenergetic  
1172 costs of heavy metal exposure in yellow perch (*Perca flavescens*): in situ estimates with  
1173 a radiotracer (<sup>137</sup>Cs) technique. *Canadian Journal of Fisheries and Aquatic Sciences*  
1174 **57**(2): 441-450.
- 1175 Simard, M. 2018. L'industrie minière au Québec : situation, tendances et enjeux. *Études*  
1176 *canadiennes / Canadian Studies*(85): 193-217. doi:10.4000/eccs.1579.
- 1177 Simard, M., and Brisson, C. 2013. L'industrie minière et le développement urbain en milieu  
1178 nordique : l'exemple de Fermont au Québec. *Cybergeo*. doi:10.4000/cybergeo.25817.
- 1179 Simard, M., and Brisson, C. 2016. Les vulnérabilités des villes minières nordiques : le cas de  
1180 Schefferville au Québec. *Revue Organisations & Territoires* **25**(1).  
1181 doi:10.1522/revueot.v25i1.67.
- 1182 Siqueira, D., Cesar, R., Lourenço, R., Salomão, A., Marques, M., Polivanov, H., Teixeira, M.,  
1183 Vezzoni, M., Santos, D., and Koifman, G. 2022. Terrestrial and aquatic ecotoxicity of iron

- 1184 ore tailings after the failure of VALE SA mining dam in Brumadinho (Brazil). *Journal of*  
1185 *Geochemical Exploration* **235**: 106954.
- 1186 Sloman, K., and McNeil, P. 2012. Using physiology and behaviour to understand the responses  
1187 of fish early life stages to toxicants. *Journal of Fish Biology* **81**(7): 2175-2198.
- 1188 Spallholz, J.E. 1994. On the nature of selenium toxicity and carcinostatic activity. *Free Radical*  
1189 *Biology and Medicine* **17**(1): 45-64.
- 1190 Spallholz, J.E., Shriver, B.J., and Reid, T.W. 2001. Dimethyldiselenide and methylseleninic acid  
1191 generate superoxide in an in vitro chemiluminescence assay in the presence of  
1192 glutathione: implications for the anticarcinogenic activity of L-selenomethionine and L-Se-  
1193 methylselenocysteine. *Nutrition and cancer* **40**(1): 34-41.
- 1194 Stewart, D., Carmichael, T., Sawatzky, C., Mochnacz, N., and Reist, J. 2007. Fish life history and  
1195 habitat use in the Northwest Territories: round whitefish (*Prosopium cylindraceum*).  
1196 *Canadian Manuscript Report of Fisheries and Aquatic Sciences* **2795**.
- 1197 Stewart, R., Grosell, M., Buchwalter, D.B., Fisher, N.S., Luoma, S.N., Mathews, T., Orr, P., and  
1198 Wang, W. 2010. Bioaccumulation and trophic transfer of selenium. *In* *Ecological*  
1199 *assessment of selenium in the aquatic environment*.
- 1200 Tessier, A., Campbell, P.G., and Bisson, M. 1979. Sequential extraction procedure for the  
1201 speciation of particulate trace metals. *Analytical chemistry* **51**(7): 844-851.
- 1202 Tran, L., Reist, J., and Power, M. 2015. Total mercury concentrations in anadromous Northern  
1203 Dolly Varden from the northwestern Canadian Arctic: a historical baseline study. *Science*  
1204 *of the Total Environment* **509**: 154-164.
- 1205 van der Velden, S., Dempson, J.B., Evans, M.S., Muir, D.C., and Power, M. 2013. Basal mercury  
1206 concentrations and biomagnification rates in freshwater and marine food webs: effects on  
1207 Arctic charr (*Salvelinus alpinus*) from eastern Canada [Research Support, Non-U.S.  
1208 Gov't]. *Science of the Total Environment* **444**: 531-542.  
1209 doi:10.1016/j.scitotenv.2012.11.099.
- 1210 Vieira, M.C., Torronteras, R., Córdoba, F., and Canalejo, A. 2012. Acute toxicity of manganese in  
1211 goldfish *Carassius auratus* is associated with oxidative stress and organ specific  
1212 antioxidant responses. *Ecotoxicology and Environmental Safety* **78**: 212-217.
- 1213 Vowinckel, T., Oechel, W., and Boll, W. 1975. The effect of climate on the photosynthesis of *Picea*  
1214 *mariana* at the subarctic tree line. 1. Field measurements. *Canadian Journal of Botany*  
1215 **53**(7): 604-620.
- 1216 Weber, A.A., Sales, C.F., de Souza Faria, F., Melo, R.M.C., Bazzoli, N., and Rizzo, E. 2020.  
1217 Effects of metal contamination on liver in two fish species from a highly impacted  
1218 neotropical river: a case study of the Fundão dam, Brazil. *Ecotoxicology and*  
1219 *Environmental Safety* **190**: 110165.
- 1220 Wellnitz, T.A., Grief, K.A., and Sheldon, S.P. 1994. Response of macroinvertebrates to blooms of  
1221 iron-depositing bacteria. *Hydrobiologia* **281**(1): 1-17.
- 1222 Wilson, M.J., McTammany, M.E., and Bohr, H.R. 2019. Manganese oxides as localized drivers of  
1223 benthic invertebrate density and community structure. *Hydrobiologia* **838**(1): 85-98.
- 1224 Winston, G.W., and Di Giulio, R.T. 1991. Prooxidant and antioxidant mechanisms in aquatic  
1225 organisms. *Aquatic Toxicology* **19**(2): 137-161.
- 1226 Wood, C.M. 2011. An introduction to metals in fish physiology and toxicology: basic principles. *In*  
1227 *Fish physiology*. Elsevier. pp. 1-51.



- 1228 Wood, C.M., Farrell, A.P., and Brauner, C. 2011a. Homeostasis and Toxicology of Essential  
1229 Metals. *In* Homeostasis and Toxicology of Essential Metals.
- 1230 Wood, C.M., Farrell, A.P., and Brauner, C. 2011b. Homeostasis and Toxicology of Non-Essential  
1231 Metals. *In* Homeostasis and Toxicology of Non-Essential Metals.
- 1232 Zar, J.H. 2007. Biostatistical Analysis. Prentice-Hall, Inc., Upper Saddle River, NJ, USA.

## TABLES

1234

1235 Table 1: Water (temperature, pH and conductivity) and lake (maximal length, maximal width and mining inputs) parameters and fishing usage of  
 1236 sampled sites in the summer 2019.

Lake	Water parameters			Lake parameters			Fishing usage
	Temperature (°C)	pH	Conductivity (µS/cm)	Maximal length (m)	Maximal width (m)	Mining inputs	
Perroche	15.3	6.6	5.7	2090	364	None	Recreational fishing outfitter
Cherny	15.7	7.0	52.0	3300	866	Upstream sources (6 km distance) of metal contamination (from 2011 to 2014) including: (i) 200 000 m <sup>3</sup> of iron mine tailing released in the environment after a dam break, (ii) 14 500 L of iron sulfate discharged and (iii) several incidents of mine tailings discharged after heavy rainfall	None known
Houston	15.1	6.8	19.0	2690	388	Atmospheric contamination from current mining exploitation (TSMC) in addition to historical atmospheric contamination (LIM, from 2011 to 2013)	Subsistence fishing
Denault	15.8	7.7	34.3	2035	505	Immediate contamination from mine tailings (IOC, from 1954 to 1982) with residuals being in direct contact with the lake	Subsistence fishing

1237

1238

Abbreviations: TSMC (Tata Steel Minerals Canada), LIM (Labrador Iron Mine Limited) and IOC (Iron Ore Company of Canada)

1239 Table 2: Capture data for fish species collected in summer 2019 at the reference site (Lake  
 1240 Perroche), as well as in the Fermont (Lake Cherny) and Schefferville (lakes Houston, Denault  
 1241 and Star) areas.

1242

Lake	Perroche	Cherny	Houston	Denault	Star	TOTAL
<b>Lake charr</b> ( <i>Salvelinus namaycush</i> )	3	2		7		12
<b>Brook charr</b> ( <i>Salvelinus fontinalis</i> )	20		20	1		41
<b>White sucker</b> ( <i>Catostomus commersonii</i> )	7			14		21
<b>Longnose sucker</b> ( <i>Catostomus catostomus</i> )				20		20
<b>Round whitefish</b> ( <i>Prosopium cylindraceum</i> )		2		6		8
<b>Lake whitefish</b> ( <i>Coregonus clupeaformis</i> )	20	20		3		43
<b>Northern pike</b> ( <i>Esox lucius</i> )	11	1				12
<b>Lake chub</b> ( <i>Couesius plumbeus</i> )	11	20		20		51
<b>Slimy sculpin</b> ( <i>Cottus cognatus</i> )	1			2	1	4
<b>Burbot</b> ( <i>Lota lota</i> )				3		3
<b>TOTAL</b>	<b>73</b>	<b>45</b>	<b>20</b>	<b>76</b>	<b>1</b>	<b>215</b>

1244 Table 3: Fork length, total weight, FCF, HSI, sex ratio and age of *Salmoninæ*, *Catostominae* and *Coregoninae* captured in the summer of 2019.  
 1245 Values are expressed as mean  $\pm$  standard deviation with ranges (minimum; maximum); n.a., not available. Different letters indicate significant  
 1246 differences for the given species between sampled lakes and sample number is provided for HSI and age in brackets when different from total  
 1247 sample number.

Lake	Subfamily	Species	N	Fork length (mm)	Whole weight (g)	FCF	HSI	Sex	Age (years)
Perroche	<i>Salmoninæ</i>	Brook charr	20	319 $\pm$ 100 <sup>a</sup> 127; 493	500 $\pm$ 486 <sup>a</sup> 21; 1997	1.14 $\pm$ 0.17 <sup>a</sup> 0.79; 1.67	0.94 $\pm$ 0.36 0.57; 2.25	55.0% Male 40.0% Female 5.0% n.d.	3 $\pm$ 1 <sup>b</sup> 1; 6 (18)
		Lake charr	3	568 $\pm$ 43 518; 598	2133 $\pm$ 497 1562; 2467	1.15 $\pm$ 0.02 1.12; 1.17	1.28 $\pm$ 0.46 0.79; 1.70	66.7% Male 33.3% Female	9 $\pm$ 2 8; 11
	<i>Catostominae</i>	White sucker	7	502 $\pm$ 37 <sup>a</sup> 434; 548	1883 $\pm$ 386 <sup>a</sup> 1198; 2326	1.47 $\pm$ 0.06 <sup>a</sup> 1.41; 1.59	1.99 $\pm$ 2.37 <sup>a</sup> 0.97; 7.35	57.1% Male 42.9% Female	11 $\pm$ 3 <sup>a</sup> 8; 18
	<i>Coregoninae</i>	Lake whitefish	20	280 $\pm$ 118 108; 487	440 $\pm$ 530 14; 1692	1.19 $\pm$ 0.13 1.01; 1.47	0.67 $\pm$ 0.16 <sup>b</sup> 0.43; 0.94	65.0% Male 25.0% Female 10.0% n.d.	4 $\pm$ 2 1; 8
Cherny	<i>Salmoninæ</i>	Lake charr	2	570 $\pm$ 50 534; 605	1958 $\pm$ 663 1489; 2426	1.04 $\pm$ 0.08 0.98; 1.10	1.02 $\pm$ 0.51 0.66; 1.38	50.0% Male 50.0% Female	9 (1)
		Lake whitefish	20	281 $\pm$ 74 152; 454	355 $\pm$ 319 39; 1450	1.26 $\pm$ 0.13 1.05; 1.55	0.89 $\pm$ 0.18 <sup>a</sup> 0.57; 1.12 (19)	45.0% Female 45.0% Male 10.0% n.d.	5 $\pm$ 2 2; 11
	<i>Coregoninae</i>	Round whitefish	2	222 $\pm$ 23 205; 238	109 $\pm$ 42 79; 138	0.97 $\pm$ 0.08 0.92; 1.02	1.17 $\pm$ 0.30 0.96; 1.38	100.0% Male	4 $\pm$ 1 3; 4

Houston	<i>Salmoninae</i>	Brook charr	20	186 ± 30 <sup>b</sup> 153; 270	66 ± 38 <sup>b</sup> 36; 190	0.97 ± 0.10 <sup>b</sup> 0.74; 1.25	0.97 ± 0.42 0.36; 1.89 (18)	70.0% Male 30.0% Female	4 ± 1 <sup>a</sup> 3; 5
Denault	<i>Salmoninae</i>	Brook charr	1	133	23	0.98	n.a.	100.0% Female	2
		Lake charr	7	534 ± 104 398; 645	1809 ± 935 634; 3165	1.09 ± 0.14 0.90; 1.26	1.13 ± 0.27 0.87; 1.60	57.1% Male 42.9% Female	8 ± 2 5; 11
	<i>Catostominae</i>	White sucker	14	230 ± 94 <sup>b</sup> 171; 456	257 ± 426 <sup>b</sup> 53; 1362	1.17 ± 0.14 <sup>b</sup> 1.01; 1.49	0.64 ± 0.47 <sup>b</sup> 0.28; 1.33 (7)	28.6% Female 14.3% Male 57.1% n.d.	6 ± 3 <sup>b</sup> 4; 13
		Longnose sucker	20	220 ± 52 165; 384	145 ± 147 50; 720	1.12 ± 0.10 0.99; 1.28	0.79 ± 0.25 0.42; 1.08 (8)	15.8% Male 5.3% Female 78.9% n.d.	5 ± 2 3; 12
	<i>Coregoninae</i>	Lake whitefish	3	184 ± 44 155; 234	82 ± 66 42; 159	1.15 ± 0.08 1.08; 1.24	0.50 ± 0.14 <sup>b</sup> 0.38; 0.65	66.7% Female 33.3% n.d.	2 ± 0 2; 2
		Round whitefish	6	231 ± 23 213; 273	117 ± 35 84; 179	0.93 ± 0.09 0.83; 1.06	0.59 ± 0.21 0.44; 0.74 (2)	83.3% Female 16.7% Male	4 ± 1 3; 5

1248  
1249

Abbreviations: Fulton's condition factor (FCF), hepatosomatic index (HSI)

1250 Table 4: Trace element concentrations of Al, Cd, Co, Cr, Cu, Fe, Hg, Mn, Pb, Se, Zn in the liver of *Salmoninae*, *Catostominae* and *Coregoninae*  
 1251 captured in the summer 2019. Values are expressed in  $\mu\text{g/g dw}$  as mean  $\pm$  standard deviation with ranges (minimum; maximum) and sample  
 1252 number in brackets. THg concentrations are expressed in wet weight ( $\mu\text{g/g}$ ). Different letters indicate significant differences of the given subfamily  
 1253 among lakes.  
 1254

Lake	Perroche			Cherry			Houston			Denault		
	Subfamily	<i>Salmoninae</i>	<i>Catostominae</i>	<i>Coregoninae</i>	<i>Salmoninae</i>	<i>Coregoninae</i>	<i>Salmoninae</i>	<i>Coregoninae</i>	<i>Salmoninae</i>	<i>Salmoninae</i>	<i>Catostominae</i>	<i>Coregoninae</i>
Al		13 $\pm$ 14 <sup>b</sup> 1; 72 (23)	13 $\pm$ 12 <sup>b</sup> 4; 36 (7)	23 $\pm$ 38 <sup>a</sup> 4; 172 (18)	3 $\pm$ 2 <sup>b</sup> 2; 5 (2)	7 $\pm$ 5 <sup>b</sup> 1; 17 (21)	37 $\pm$ 33 <sup>a</sup> 8; 150 (16)	5 $\pm$ 5 <sup>b</sup> 1; 16 (7)	183 $\pm$ 332 <sup>a</sup> 8; 1853 (31)	27 $\pm$ 17 <sup>a</sup> 7; 56 (6)		
Cd		0.8 $\pm$ 0.5 <sup>b</sup> 0.3; 2.2 (23)	3.6 $\pm$ 1.7 <sup>a</sup> 1.2; 5.7 (7)	1.2 $\pm$ 0.6 <sup>a</sup> 0.3; 2.6 (18)	0.5 $\pm$ 0.1 <sup>b</sup> 0.4; 0.5 (2)	0.5 $\pm$ 0.3 <sup>b</sup> 0.1; 1.4 (22)	3.2 $\pm$ 2.1 <sup>a</sup> 0.6; 8.7 (16)	1.3 $\pm$ 0.6 <sup>b</sup> 0.5; 2.4 (7)	0.7 $\pm$ 0.6 <sup>b</sup> 0.1; 3.1 (31)	1.1 $\pm$ 0.3 <sup>a</sup> 0.6; 1.6 (6)		
Co		0.54 $\pm$ 0.29 <sup>ab</sup> 0.25; 1.48 (23)	0.72 $\pm$ 0.34 <sup>a</sup> 0.40; 1.39 (7)	0.31 $\pm$ 0.11 <sup>a</sup> 0.15; 0.63 (18)	0.32 $\pm$ 0.11 <sup>ab</sup> 0.25; 0.40 (2)	0.21 $\pm$ 0.07 <sup>b</sup> 0.10; 0.38 (22)	1.09 $\pm$ 0.99 <sup>a</sup> 0.29; 3.43 (16)	0.39 $\pm$ 0.21 <sup>b</sup> 0.16; 0.82 (7)	0.30 $\pm$ 0.13 <sup>b</sup> 0.12; 0.73 (31)	0.48 $\pm$ 0.24 <sup>a</sup> 0.22; 0.90 (6)		
Cr		0.56 $\pm$ 0.16 <sup>b</sup> 0.35; 0.94 (23)	0.57 $\pm$ 0.14 0.39; 0.77 (7)	0.95 $\pm$ 0.41 <sup>a</sup> 0.46; 2.17 (18)	0.44 $\pm$ 0.10 <sup>b</sup> 0.37; 0.51 (2)	0.40 $\pm$ 0.26 <sup>b</sup> 0.19; 1.40 (22)	1.78 $\pm$ 0.99 <sup>a</sup> 0.66; 4.09 (16)	0.47 $\pm$ 0.20 <sup>b</sup> 0.30; 0.82 (7)	0.68 $\pm$ 0.84 0.05; 4.69 (30)	0.50 $\pm$ 0.20 <sup>b</sup> 0.21; 0.71 (6)		
Cu		48 $\pm$ 20 <sup>b</sup> 7; 93 (23)	54 $\pm$ 26 <sup>a</sup> 28; 103 (7)	21 $\pm$ 11 <sup>a</sup> 9; 57 (18)	107 $\pm$ 47 <sup>ab</sup> 74; 140 (2)	15 $\pm$ 5 <sup>a</sup> 9; 25 (22)	94 $\pm$ 47 <sup>a</sup> 21; 205 (16)	83 $\pm$ 30 <sup>ab</sup> 25; 115 (7)	35 $\pm$ 33 <sup>b</sup> 7; 192 (31)	7 $\pm$ 1 <sup>b</sup> 5; 8 (5)		
Fe		624 $\pm$ 368 158; 1661 (23)	969 $\pm$ 525 <sup>a</sup> 346; 1606 (7)	277 $\pm$ 116 <sup>ab</sup> 136; 603 (18)	642 $\pm$ 327 411; 873 (2)	217 $\pm$ 92 <sup>b</sup> 131; 511 (22)	971 $\pm$ 555 52; 1978 (17)	632 $\pm$ 301 308; 1044 (7)	397 $\pm$ 661 <sup>b</sup> 56; 3702 (31)	340 $\pm$ 122 <sup>a</sup> 183; 480 (6)		

Hg	0.30 ± 0.25 <sup>b</sup> 0.11; 1.29 (22)	0.26 ± 0.12 <sup>a</sup> 0.12; 0.45 (7)	0.28 ± 0.16 <sup>a</sup> 0.12; 0.66 (17)	1.18 ± 0.32 <sup>a</sup> 0.95; 1.40 (2)	0.22 ± 0.10 <sup>a</sup> 0.11; 0.54 (22)	0.15 ± 0.04 <sup>b</sup> 0.10; 0.21 (7)	0.37 ± 0.27 <sup>a</sup> 0.14; 0.86 (7)	0.04 ± 0.01 <sup>b</sup> 0.01; 0.07 (27)	0.05 ± 0.01 <sup>b</sup> 0.04; 0.07 (5)
Mn	3.9 ± 1.3 1.5; 6.3 (23)	4.9 ± 1.1 <sup>b</sup> 3.8; 7.0 (7)	5.2 ± 1.9 <sup>a</sup> 3.0; 9.2 (18)	3.3 ± 0.5 2.9; 3.7 (2)	3.6 ± 0.8 <sup>b</sup> 2.2; 5.4 (22)	4.3 ± 1.8 2.7; 8.9 (17)	3.3 ± 1.4 1.6; 5.8 (7)	39.1 ± 30.2 <sup>a</sup> 7.1; 109.9 (31)	7.0 ± 2.5 <sup>a</sup> 4.4; 10.1 (6)
Pb	0.22 ± 0.49 <sup>a</sup> 0.00; 2.22 (23)	0.14 ± 0.10 0.04; 0.29 (7)	0.25 ± 0.28 <sup>a</sup> 0.01; 0.98 (16)	0.02 ± 0.01 <sup>ab</sup> 0.01; 0.02 (2)	0.05 ± 0.06 <sup>b</sup> 0.00; 0.25 (17)	0.13 ± 0.14 <sup>a</sup> 0.02; 0.61 (16)	0.01 ± 0.01 <sup>b</sup> 0.00; 0.03 (7)	0.29 ± 0.40 0.01; 1.56 (31)	0.64 ± 1.47 <sup>ab</sup> 0.00; 3.63 (6)
Se	5.4 ± 2.4 <sup>c</sup> 3.4; 15.0 (23)	7.6 ± 2.5 <sup>b</sup> 4.8; 11.6 (7)	8.8 ± 2.0 <sup>c</sup> 6.1; 13.1 (18)	20.1 ± 2.5 <sup>b</sup> 18.3; 21.9 (2)	11.3 ± 2.7 <sup>b</sup> 6.4; 17.3 (22)	14.0 ± 7.7 <sup>b</sup> 2.8; 27.5 (16)	90.0 ± 46.2 <sup>a</sup> 52.0; 174.2 (7)	11.4 ± 5.9 <sup>a</sup> 4.2; 30.4 (31)	20.1 ± 4.3 <sup>a</sup> 17.2; 28.6 (6)
Zn	124 ± 35 <sup>b</sup> 58; 178 (23)	64 ± 17 40; 89 (7)	158 ± 44 <sup>a</sup> 104; 266 (15)	85 ± 17 <sup>bc</sup> 73; 97 (2)	88 ± 24 <sup>b</sup> 50; 132 (22)	206 ± 96 <sup>a</sup> 87; 481 (17)	47 ± 5 <sup>c</sup> 43; 50 (7)	63 ± 14 45; 92 (31)	76 ± 27 <sup>b</sup> 50; 126 (6)

1255

1256

Abbreviations: Aluminum (Al), cadmium (Cd), cobalt (Co), chromium (Cr), copper (Cu), iron (Fe), mercury (Hg), manganese (Mn), lead (Pb), selenium (Se), zinc (Zn)

1257 Table 5: Spearman correlations (rs) among biometric biomarkers (fork length, whole weight, FCF, HSI and age), biochemical biomarkers (8-  
 1258 OHdG, MDA, CAT, SOD and GPx) and trace element concentrations (Cd, Cu, Hg, Mn and Se) in the liver of *Salmoninæ* (n = 53) from sampled  
 1259 lakes in the summer 2019. Bolded values indicate significant correlations and the significance level of the correlation is denoted as follows: p <  
 1260 0.05 \*; p < 0.001 \*\*; p < 0.0001\*\*\*.

	Fork length	Whole weight	FCF	HSI	Age	8-OHdG	MDA	CAT	SOD	GPx	Cd	Cu	Hg	Mn
Whole weight	<b>0.99***</b>	1												
FCF	0.42	<b>0.49*</b>	1											
HSI	0.17	0.19	0.12	1										
Age	<b>0.57*</b>	<b>0.57*</b>	0.03	0.16	1									
8-OHdG	-0.12	-0.24	0.24	-0.13	-0.44	1								
MDA	-0.09	-0.09	0.03	-0.03	-0.23	<b>0.69*</b>	1							
CAT	-0.25	-0.34	-0.22	-0.50	-0.41	0.31	0.38	1						
SOD	0.41	0.17	-0.01	-0.43	0.21	0.56	0.38	0.07	1					
GPx	0.46	0.54	0.07	-0.14	0.33	0.18	0.00	-0.25	0.43	1				
Cd	-0.27	-0.29	<b>-0.51*</b>	-0.03	0.36	0.15	0.41	0.24	0.30	-0.50	1			
Cu	-0.22	-0.25	-0.43	-0.01	0.18	0.42	-0.10	0.08	0.33	0.46	0.40	1		
Hg	<b>0.89***</b>	<b>0.88***</b>	0.21	0.15	<b>0.91***</b>	-0.47	-0.33	-0.49	-0.03	0.46	0.24	0.07	1	
Mn	-0.27	-0.25	-0.09	-0.02	-0.33	-0.36	0.10	0.20	-0.34	-0.43	0.22	-0.09	-0.34	1
Se	0.18	0.15	-0.26	0.03	<b>0.59*</b>	<b>0.74*</b>	0.53	0.11	0.69	0.46	<b>0.61**</b>	<b>0.56*</b>	<b>0.60*</b>	-0.02

1261

1262 Abbreviations: Fulton's condition factor (FCF), hepatosomatic index (HSI), 8-hydroxy-2-deoxyguanosine (8-OHdG), malondialdehyde (MDA), catalase (CAT), superoxide  
 1263 dismutase (SOD), glutathione peroxidase (GPx), cadmium (Cd), copper (Cu), mercury (Hg), manganese (Mn), selenium (Se)



1264 Table 6: Spearman correlations (rs) among biometric biomarkers (fork length, whole weight, FCF, HSI and age), biochemical biomarkers (8-  
 1265 OHdG, MDA, CAT, SOD and GPx) and trace element concentrations (Cd, Cu, Hg, Mn and Se) in the liver of *Catostomineæ* (n = 41) from sampled  
 1266 lakes in the summer 2019. Bolded values indicate significant correlations and the significance level of the correlation is denoted as follows: p <  
 1267 0.05 \*; p < 0.001 \*\*; p < 0.0001\*\*\*.

	Fork length	Whole weight	FCF	HSI	Age	8-OHdG	MDA	CAT	SOD	GPx	Cd	Cu	Hg	Mn
Whole weight	<b>0.98***</b>	1												
FCF	<b>0.64*</b>	<b>0.74***</b>	1											
HSI	0.60	0.59	<b>0.58</b>	1										
Age	<b>0.82***</b>	<b>0.78***</b>	<b>0.58*</b>	0.44	1									
8-OHdG	-0.43	-0.45	-0.49	-0.23	-0.48	1								
MDA	0.24	0.25	0.42	0.28	0.16	0.07	1							
CAT	-0.36	-0.33	-0.11	-0.64	-0.36	0.25	0.12	1						
SOD	0.28	0.26	0.27	0.20	0.24	-0.06	0.49	-0.07	1					
GPx	0.40	0.39	0.46	0.51	0.29	-0.22	<b>0.55*</b>	0.11	0.33	1				
Cd	<b>0.80***</b>	<b>0.78***</b>	<b>0.66*</b>	0.68	<b>0.83***</b>	-0.53	0.27	-0.35	0.31	0.47	1			
Cu	0.19	0.14	-0.02	0.12	0.19	-0.10	-0.20	-0.36	0.05	-0.12	0.28	1		
Hg	<b>0.95***</b>	<b>0.94***</b>	<b>0.74**</b>	0.58	<b>0.77***</b>	-0.56	0.22	-0.34	0.22	0.56	<b>0.76***</b>	0.16	1	
Mn	<b>-0.71**</b>	<b>-0.70**</b>	-0.50	-0.65	-0.56	0.53	-0.17	0.49	-0.10	-0.39	<b>-0.60*</b>	-0.34	<b>-0.66*</b>	1
Se	0.13	0.12	0.09	0.01	0.30	0.14	-0.09	-0.03	-0.08	-0.15	0.27	0.06	0.00	0.24

1268

1269 Abbreviations: Fulton's condition factor (FCF), hepatosomatic index (HSI), 8-hydroxy-2-deoxyguanosine (8-OHdG), malondialdehyde (MDA), catalase (CAT), superoxide  
 1270 dismutase (SOD), glutathione peroxidase (GPx), cadmium (Cd), copper (Cu), mercury (Hg), manganese (Mn), selenium (Se)

1271 Table 7: Spearman correlations (rs) among biometric biomarkers (fork length, whole weight, FCF, HSI and age), biochemical biomarkers (8-  
 1272 OHdG, MDA, CAT, SOD and GPx) and trace element concentrations (Cd, Cu, Hg, Mn and Se) in the liver of *Coregoninæ* (n = 51) from sampled  
 1273 lakes in the summer 2019. Bolded values indicate significant correlations and the significance level of the correlation is denoted as follows: p <  
 1274 0.05 \*; p < 0.001 \*\*; p < 0.0001\*\*\*.

	Fork length	Whole weight	FCF	HSI	Age	8-OHdG	MDA	CAT	SOD	GPx	Cd	Cu	Hg	Mn
Whole weight	<b>0.99***</b>	1												
FCF	<b>0.69***</b>	<b>0.75***</b>	1											
HSI	-0.05	-0.03	0.14	1										
Age	<b>0.94***</b>	<b>0.92***</b>	<b>0.61**</b>	0.01	1									
8-OHdG	0.00	0.00	-0.15	-0.42	0.00	1								
MDA	-0.13	-0.12	-0.16	-0.16	-0.24	0.45	1							
CAT	-0.38	-0.38	-0.40	-0.34	-0.44	0.42	<b>0.62*</b>	1						
SOD	-0.30	-0.28	-0.26	-0.44	-0.33	0.38	0.23	0.36	1					
GPx	0.28	0.28	0.13	-0.26	0.22	0.14	0.08	0.21	0.26	1				
Cd	0.40	0.39	0.05	<b>-0.54*</b>	0.44	0.46	-0.05	0.01	0.31	0.27	1			
Cu	<b>0.56*</b>	<b>0.54*</b>	0.41	-0.32	<b>0.52*</b>	0.27	-0.09	-0.29	-0.01	0.07	0.40	1		
Hg	<b>0.90***</b>	<b>0.90***</b>	<b>0.75***</b>	-0.16	<b>0.82***</b>	0.28	-0.18	-0.36	-0.19	0.23	0.38	<b>0.61*</b>	1	
Mn	<b>-0.55*</b>	<b>-0.57*</b>	<b>-0.64**</b>	-0.20	-0.48	0.20	-0.15	0.21	0.44	-0.02	0.24	-0.38	-0.52	1
Se	0.24	0.21	0.06	0.14	0.40	0.10	-0.09	-0.28	-0.08	0.15	0.19	-0.05	0.01	-0.08

1275

1276 Abbreviations: Fulton's condition factor (FCF), hepatosomatic index (HSI), 8-hydroxy-2-deoxyguanosine (8-OHdG), malondialdehyde (MDA), catalase (CAT), superoxide  
 1277 dismutase (SOD), glutathione peroxidase (GPx), cadmium (Cd), copper (Cu), mercury (Hg), manganese (Mn), selenium (Se)

1           **ASSESSING THE IMPACTS OF MINING ACTIVITIES ON FISH**  
2                           **HEALTH IN NORTHERN QUÉBEC**

3 Anthony Fontaine<sup>1</sup>; Mackenzie Anne Clifford Martyniuk<sup>1</sup>; Camille Garnier<sup>1</sup>; Patrice Couture<sup>1</sup>

4 <sup>1</sup> *Institut National de la Recherche Scientifique, centre Eau-Terre-Environnement, Québec, Canada*

5

6 **Corresponding author:**

7 Anthony Fontaine

8 anthony.fontaine@univ-pau.fr

9 **ORCID ID:** <https://orcid.org/0000-0001-5779-5789>

10

## FIGURE CAPTIONS

11 Figure 1: Map of the sampled lakes in the region of Schefferville (left) and Fermont (right)  
12 during summer 2019. The orange triangle on the bottom map indicates the relative distance of  
13 Lake Perroche (the reference site) from Fermont and Schefferville.

14

15 Figure 2: Trace element concentrations of aluminum (Al), cadmium (Cd), cobalt (Co),  
16 chromium (Cr), copper (Cu), iron (Fe), manganese (Mn), lead (Pb), selenium (Se) and zinc  
17 (Zn) for water (panel A) and sediment (panel B) from lakes sampled in the summer 2019. Water  
18 (n = 2) and sediment (n = 2) trace element concentrations are expressed in  $\mu\text{g/L}$  and  $\mu\text{g/g dw}$   
19 respectively. Values below the detection limits were excluded from the figure. A threshold value  
20 corresponding to the Provincial Water Quality Objectives from Québec (Ministère de  
21 l'Environnement et de la Lutte contre les changements climatiques, 2021) is also displayed on  
22 panel A.

23

24 Figure 3: Principal component analysis (PCA) of biometric biomarkers (fork length, whole  
25 weight, age, Fulton's condition factor (FCF) and hepatosomatic index (HSI)), biochemical  
26 biomarkers (8-hydroxy-2'-deoxyguanosine (8-OHdG), malondialdehyde (MDA), catalase  
27 (CAT), superoxide dismutase (SOD) and glutathione peroxidase (GPx)) and trace element  
28 concentrations (aluminum (Al), cadmium (Cd), cobalt (Co), chromium (Cr), copper (Cu), iron  
29 (Fe), manganese (Mn), mercury (Hg), lead (Pb), selenium (Se) and zinc (Zn)) of *Salmoninae*  
30 (n = 53) among the sampled lakes. Each point indicates an individual fish and ellipses  
31 represent individuals from the same site.

32

33 Figure 4: Principal component analysis (PCA) of biometric biomarkers (fork length, whole  
34 weight, age, Fulton's condition factor (FCF) and hepatosomatic index (HSI)), biochemical

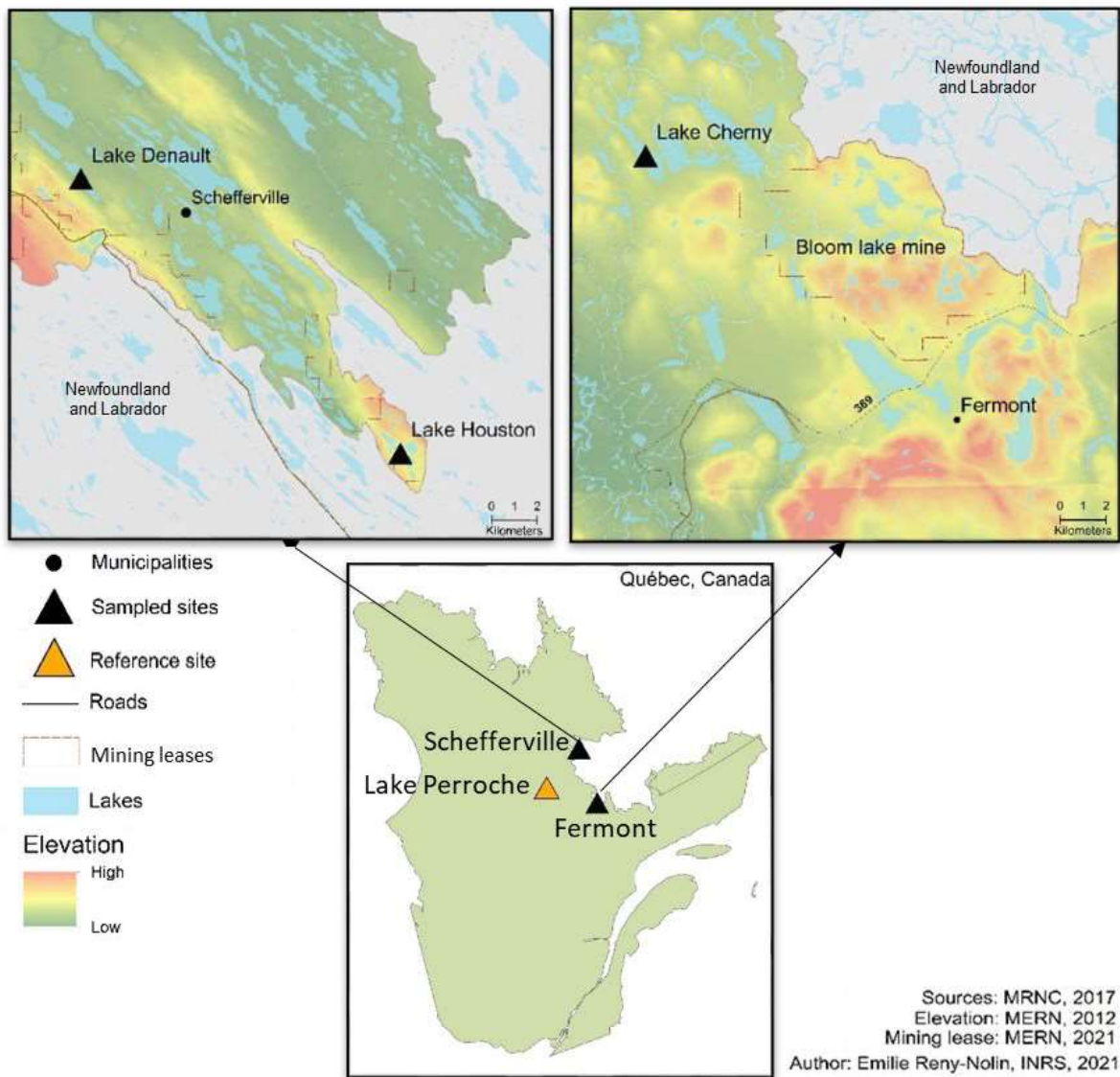
35 biomarkers (8-hydroxy-2'deoxyguanosine (8-OHdG), malondialdehyde (MDA), catalase  
36 (CAT), superoxide dismutase (SOD) and glutathione peroxidase (GPx)) and trace element  
37 concentrations (aluminum (Al), cadmium (Cd), cobalt (Co), chromium (Cr), copper (Cu), iron  
38 (Fe), manganese (Mn), mercury (Hg), lead (Pb), selenium (Se) and zinc (Zn)) of *Catostominae*  
39 (n = 41) among the sampled sites. Each point indicates an individual fish and ellipses represent  
40 individuals from the same site.

41

42 Figure 5: Principal component analysis (PCA) of biometric biomarkers (fork length, whole  
43 weight, age, Fulton's condition factor (FCF) and hepatosomatic index (HSI)), biochemical  
44 biomarkers (8-hydroxy-2'deoxyguanosine (8-OHdG), malondialdehyde (MDA), catalase  
45 (CAT), superoxide dismutase (SOD) and glutathione peroxidase (GPx)) and trace element  
46 concentrations (aluminum (Al), cadmium (Cd), cobalt (Co), chromium (Cr), copper (Cu), iron  
47 (Fe), manganese (Mn), mercury (Hg), lead (Pb), selenium (Se) and zinc (Zn)) of *Coregoninae*  
48 (n = 51) among the sampled sites. Each point indicates an individual fish and ellipses represent  
49 individuals from the same site.

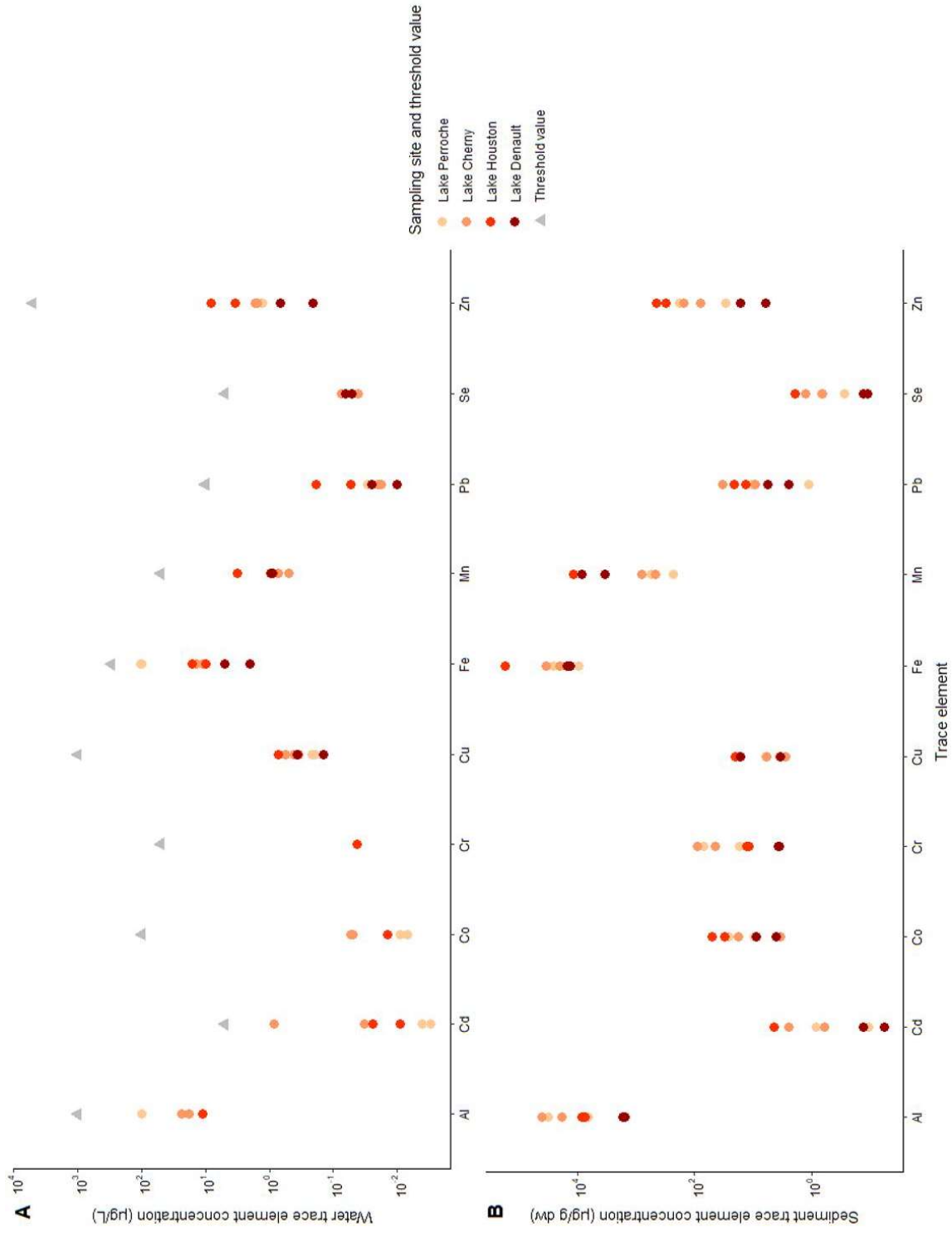
50

# FIGURES



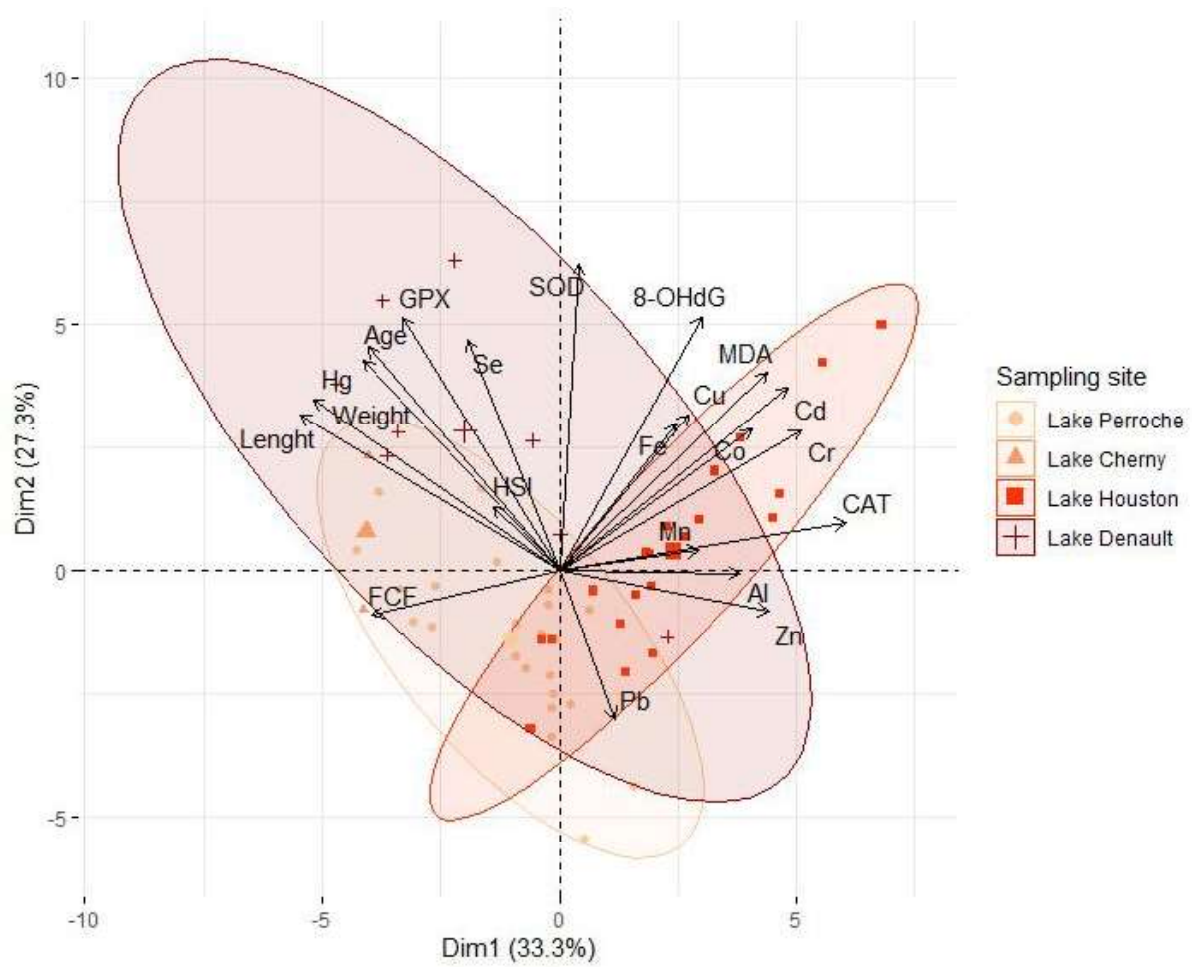
51

52 Figure 1



53

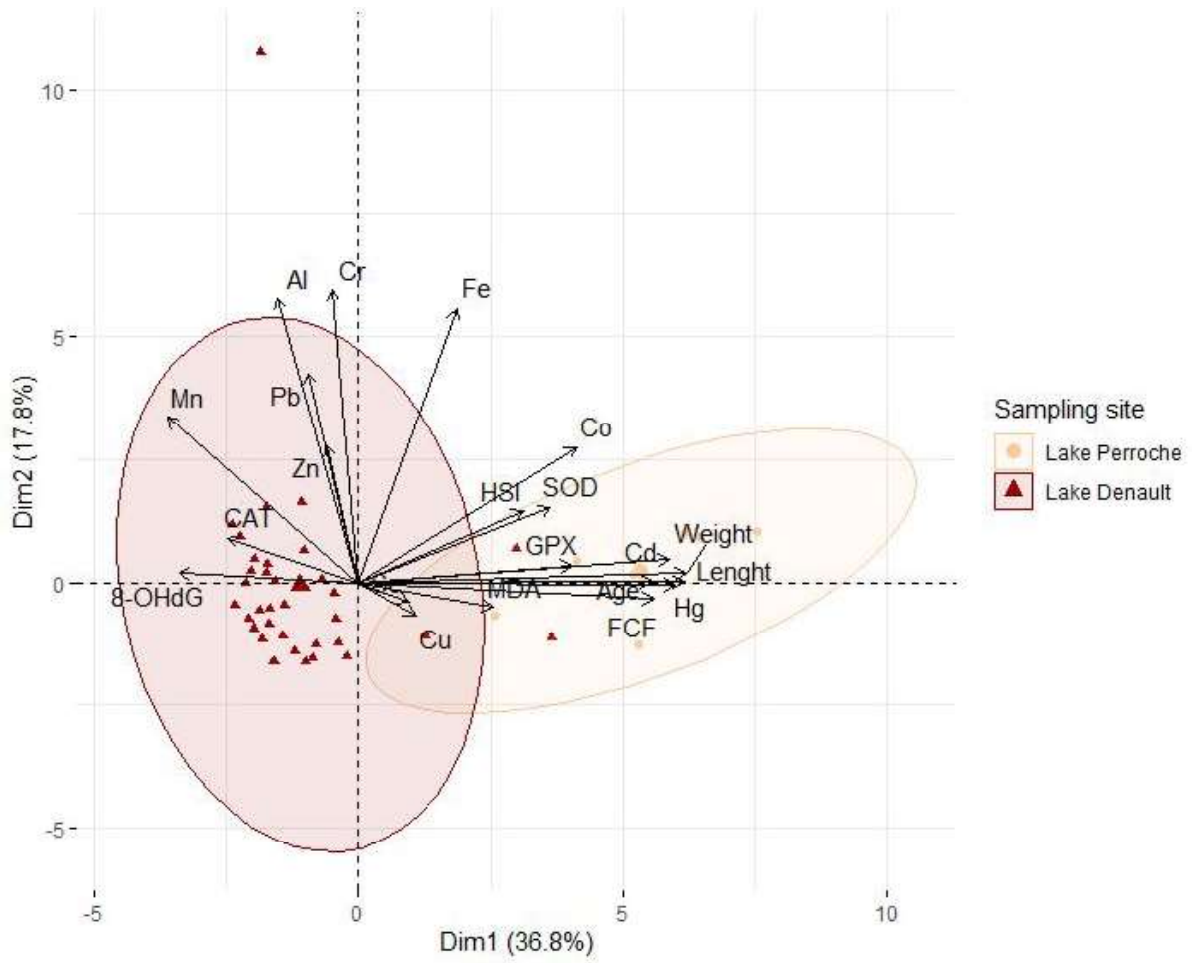
54 Figure 2



55

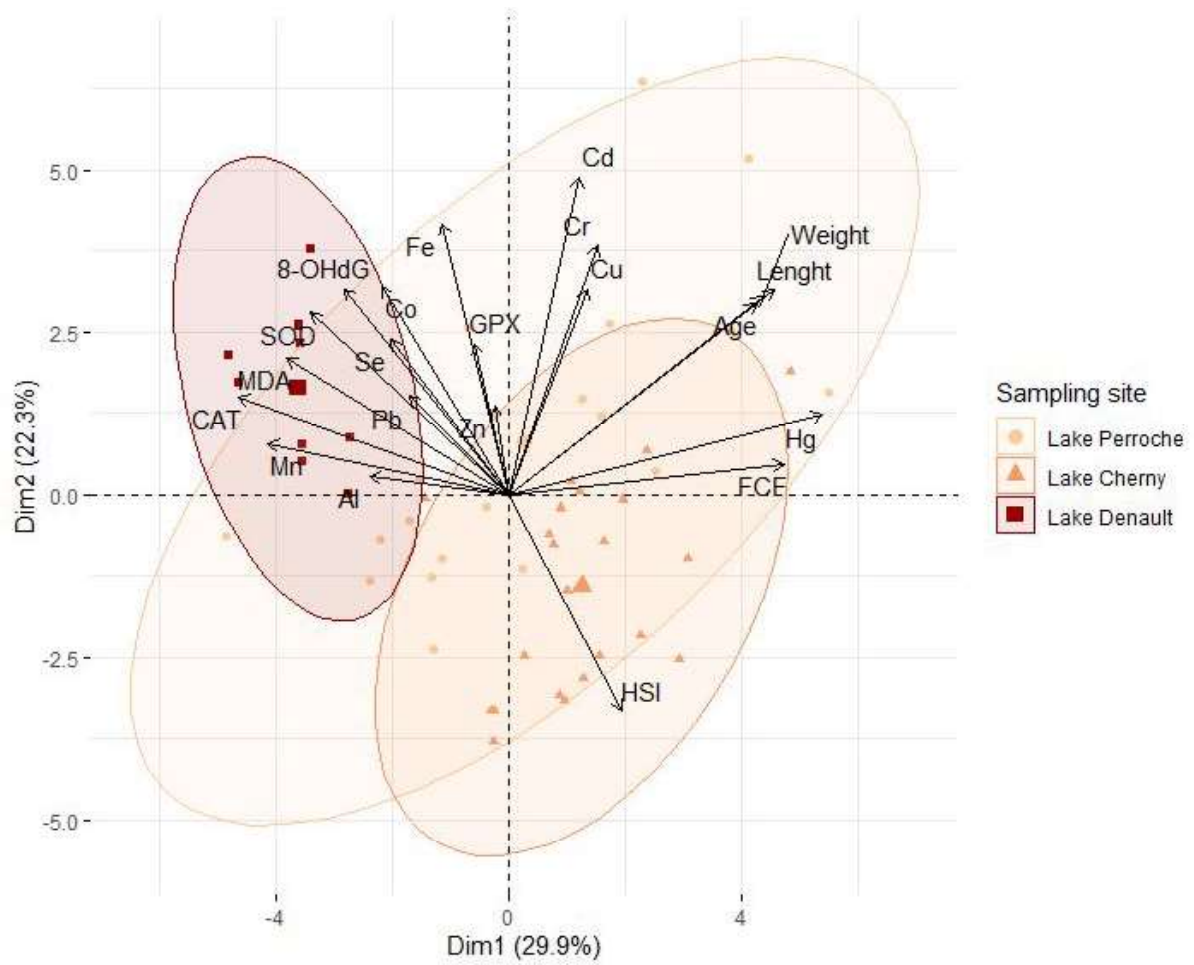
56 Figure 3





57

58 Figure 4



59

60 Figure 5