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1 **Mercury and methylmercury concentrations in high altitude lakes**
2 **and fish populations from the French Alps related to watershed**
3 **characteristics**

4
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15
16 **Abstract**

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19 **Keywords** : Mercury, methylmercury, lakes, fish population, French Alps.
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34 1. Introduction

35

36 Mercury is a toxic metal for human and environment, which can be transported on long distant from
37 its emission sources and can contaminate aquatics environment. Emitted by both natural and
38 anthropogenic sources (Pacyna et al., 2006) , elemental gaseous mercury (Hg^0), predominant form in
39 the atmosphere, can join remote areas, like altitude lakes, mainly by atmospheric transport. Hg^0 , can
40 be oxidized to divalent mercury (Hg^{2+}) and deposited onto surfaces and contaminate different
41 reservoirs like catchments and water. Once deposited, one fraction of Hg^{2+} can be methylated by
42 both biotic (Pongratz and Heumann, 1999) and abiotic pathways (Celo et al., 2006) leading to an
43 organo-metallic form: methylmercury (MeHg). This form is very toxic and is able to both accumulate
44 in living organisms and to biomagnify through the food web. Mercury contamination in fish
45 population can be extremely variable, due to the nature of ecosystems (Bjorklund et al., 1984;
46 Drevnick et al., 2007; Lindeberg et al., 2007) . MeHg sources to lakes are numerous and known. The
47 principal sources are atmospheric, via dry and wet deposition (Graydon et al., 2008), the runoff from
48 watershed, - and thus meltwater (Loseto et al., 2004) -, and in-situ production of MeHg by microbial
49 activities in water column and sediment, especially by sulfate-reducing bacteria (SRB) present in
50 anoxic sediments (Compeau, 1985; Warner et al., 2003). Now, It is known, that watershed
51 composition influences the production and accumulation of methylmercury by aquatic organism
52 (Belger and Forsberg, 2006; Bonzongo and Lyons, 2004; Warner et al., 2005) . Therefore, high
53 mercury concentrations in environment not signify that fish population will be highly contaminated.
54 Indeed, watershed composition (Warner et al., 2005) or lakes water with an important organic
55 matter can promote mercury methylation (Ribeiro Guevara et al., 2008) (REF). Therefore, a
56 catchments with abundant vegetation, is susceptible to increase MeHg concentration in water
57 column. It is also known, that mercury and methylmercury concentration increase with size of
58 ecosystem and also with trophic position of fish. More food chain is long, more methylmercury
59 concentration measured in fish will be important. This, can be explained by the fact that
60 methylmercury is bioaccumulable in the organism. However, a study conducted by (Ward et al.,
61 2010), using a large-scale field experiment, examining the relationship between Hg concentrations
62 and growth rate in fish and demonstrates that the variability of mercury concentrations in fish
63 explained by the concentrations of mercury measured in these fish prey, but also by the rate of
64 growth. Indeed, a fast-growing fish are less contaminated than the small fish. These studies show
65 that several factors influence the contamination of fish populations. Lavigne et al (Lavigne et al.,
66 2010, In Press) show again that growth rates influence mercury contamination in fishes . A study
67 conducted in Sweden (Lindqvist et al., 1991), shows that despite the reduction of emissions of Hg in
68 Sweden during the 1970's and 1980's, mercury contaminations in fish populations increase slightly.

69 Rognerud et al. (2005) (Rognerud et al., 2005) studied the contamination of mercury and
70 organochlorine in fish from high elevation lakes in Europe. Results show mercury concentrations in
71 fish populations rather low ($0.02 \mu\text{g}\cdot\text{g}^{-1}$). Several explanations are mentioned by the authors to
72 explain these low mercury levels : i) a trophic level low of fish population studied, ii) a flow of Hg in
73 sediments low, and iii) a low rates of methylation in the cold and clear lakes. However, Blais et al.,
74 (Blais, 2006), shows in a study conducted in the Pyrenees, the existence of a relationship between
75 altitude lakes and mercury contamination. The authors suggest that enhanced deposition and/or
76 retention of mercury is taking place in high-altitude aquatic foodwebs. Despite some studies
77 conducted in Europe, no study was conducted in the French Alps on mercury contamination in fish
78 population.

79 Here, we present the results of a study conducted in four lakes situated in the French Alps (Bramant
80 lakes, Crop lake, la Sagne lake and Poursollet lake). The main objectives of this paper are to
81 understand and explain mercury contamination in fish population and understand the influence of
82 watershed composition in altitude.

83

84 **2. Methods**

85 ***2.1. Site description:***

86 The four lakes studied are situated in the French Alps, in particularly in “Belledonne Massif” and
87 “Grandes Rousses Massif”. For this study we have selected, the Bramant Lake ($45^{\circ}12'00''$ N,
88 $E6^{\circ}10'34''$ E), the Crop lake ($45^{\circ}12'28''$ N, $5^{\circ}59'16''$ E), the La Sagne lake ($45^{\circ}13'15''$ N, $E6^{\circ}04'33''$ E),
89 and Poursollet Lake ($45^{\circ}03'08''$ N, $E5^{\circ}54'00''$ E). The altitude of lakes is respectively 2448m, 1906m,
90 2067m and 1648m above sea level (a.s.l.) and their distance from Grenoble is ~ 35 km, ~ 20 km, ~ 27
91 km, ~ 21 km, respectively. The different types of land cover were assessed using matricial Landsat 7
92 satellite images. The snapshots were interpolated to cover the whole region using GRASS 6.0, and
93 then analyzed with Quantum GIS for vegetation coverage. Six classes of coverage were considered,
94 including tree population, wild grass, land, rock, ice/snow and lakes. The resulting is presented in
95 figure 1. Table 1 resume different information of these 4 lakes, and figure 1 show watershed
96 composition of each lake and the study area.

97

98 ***2.2. Water samples and analysis:***

99 Water column have been sampled in September 2008 for analyses of Total mercury and
100 Methylmercury. Water column is sampled using a Niskin™ bottle, and each sample are collected in
101 acid-washed 250 mL Teflon bottles for THg and in acid-washed 125 mL Teflon bottle for MeHg using
102 clean sampling techniques (Ferrari et al., 2000). All water samples were maintained frozen at -20°C
103 and in the dark until analysis.

104 *2.2.1. Total Mercury analysis*

105 Each sample of water was oxidised with 0,5 ‰ (v/v) of BrCl to dissociate all the mercury complexes
106 and to oxidize THg to divalent form, Hg²⁺. Excess BrCl was neutralized with hydroxylamine
107 hydrochloride (0,5 ‰ v/v), and THg was determined using cold vapor atomic fluorescence
108 spectrometry after reduction of Hg²⁺ to Hg⁰ by stannous chloride (Bloom and Crecelius, 1983), using a
109 Tekran 2600 analyser (Tekran Inc.) according to EPA method 1631 revision E, with a detection limit of
110 0.1 ng.L⁻¹. Each sample was analyzed in triplicate. THg concentrations are presented as mean ± 1
111 standard deviation.

112

113 *2.2.2. Methylmercury analysis*

114 MeHg concentrations were determined by capillary gas chromatography coupled with atomic
115 fluorescence spectrometry (GC-AFS) as described by *Cai et al., (Cai et al., 1996)* and is briefly described
116 here. The determination of organomercury in water samples involves an adsorbent pre-
117 concentration of the organomercurials onto sulfydryl-cotton fibers followed by elution with acidic KBr
118 and CuSO₄ and extraction in methylene chloride.

119

120 **2.3. Fish sample and analysis:**

121 All lakes have been fished using three different experimental nets of 20 m length with mesh size of
122 15, 20 or 27 mm were used for fish collection. Fish species, total length (mm), weight (g), sex, were
123 determined for all specimens whenever possible. The anatomic structures needed for age
124 determination were also collected. A piece of fish muscle was taken from the caudal region for
125 mercury analysis. Pieces of fish flesh were conserved frozen at -20°C and in the dark until analysis.

126

127 *2.3.1. Age determination*

128 Age determinations were determined using operculum method (*Campbell and Babaluk 1979; Pépin*
129 *and Lévesque 1985; Babaluk and Campbell 1987; Babaluk et al. 1993*). All age estimations were
130 performed at least twice - usually by two different and independent readers - or until agreement was
131 reached on an age value. If disagreement persisted on the age value after the third reading, the
132 structure was discarded and the age data rejected.

133 *2.3.2. Total mercury analysis*

134 Total Hg concentrations in the fish muscles were determined by flameless atomic absorption
135 spectrometry. Analyses were carried out automatically after drying by thermal decomposition at
136 750°C, under an oxygen flow (AMA 254; Leco-France). The validity of the analytical method was
137 checked during each series of measurements against one standard biological reference materials

138 (TORT-2, lobster hepatopancreas from NRCC-CNRC, Ottawa, Canada). Hg values were consistently
139 within the certified ranges.

140

141 2.3.3. Fish standardized length

142 The average fish lengths were calculated for each lake, and the mean value of all lakes was calculated
143 according to the distribution of averages for all lakes. The three mean values obtained were then
144 used for modeling at standardized length (L_{std}) rounded at the nearest 210 mm. In this study, L_{std}
145 values act as modeling constants for each lake under study.

146

147 3. Results

148 3.1. Total mercury and methylmercury concentrations in water column.

149 Figure 2 shows THg and MeHg concentrations in water column on the four lakes studied: Bramant
150 Lake , Crop Lake, La Sagne Lake and Poursollet Lake in September 2008. THg and MeHg
151 concentrations measured are low compared to others studies. For Bramant Lake (figure2a), THg
152 concentrations profiles are obtained on depth of 19 meters. THg concentrations in water surface is
153 inferior to detection limit ($<0.1 \text{ ng.L}^{-1}$). In the water column, THg concentrations range between 1.5
154 and detection limit ($<0.1 \text{ ng.L}^{-1}$). For MeHg, levels are between 2.1 pg.L^{-1} and 3.7 pg.L^{-1} for the first
155 twelve meters. After we observe an increase up to 16 meters where methylmercury levels increase
156 from 3.6 pg.L^{-1} to 9.3 pg.L^{-1} . Then we have a significant decrease until to 18 meters, with a
157 methylmercury concentration of 3.7 pg.L^{-1} .

158 Regarding Crop lake (figure 2b), profiles is obtained with a depth of 29 meters. With our detection
159 limit, we measure only THg on surface and at 2 meters deep, corresponding to a concentration of 0.1
160 ng.L^{-1} and 0.9 ng.L^{-1} respectively. Others depth, THg concentrations is not detected by our analyze
161 method. The methylmercury concentrations in water column from Crop lake range between 1.90 to
162 9.06 pg.L^{-1} . In detail, until ten meters, we observe an increase of methylmercury levels until 9.06 pg.L^{-1}
163 1 , and then methylmercury concentrations decrease to 3.02 pg.L^{-1} until 20 meters. After we observe
164 an increase until to 29 meters deep to reach a methylmercury concentration of 8.97 pg.L^{-1} .

165 Figure 2c, shows THg concentration profiles to La Sagne lake until to 17 meters of depth. THg
166 concentrations measured are relatively high compared to others lakes. In surface we measured a
167 concentration of 3.12 ng.L^{-1} then, THg levels decrease until 8 meters to reach 1.69 ng.L^{-1} . From 8
168 meters concentrations increase to reach a maximum of 4.34 ng.L^{-1} at 10 meters deep. Then, THg
169 levels decrease until 17 meters to reach 2.97 ng.L^{-1} . Regarding MeHg concentrations in water
170 column, we observe a MeHg concentration not homogeneous, concentrations range between 2.19 to
171 6.61 pg.L^{-1} .

172 Figure 2c shows THg and MeHg profiles for Poursollet lake. THg and MeHg profile is obtained on
173 depth of 5 meters with an increase of THg concentration with depth. THg concentrations increase
174 with depth. In surface water, we measure 0.4 ng.L⁻¹ until 1.5 ng.L⁻¹ to 5 meters of depth. Concerning
175 MeHg levels, we measured concentrations range 4.73 to 9.21 pg.L⁻¹. Maximum is measured to 4
176 meters deep (9.21 pg.L⁻¹).

177

178 **3.2. THg muscle concentration in fish**

179 Different species of fishes are caught during this fishing campaign. We caught a total of 109 fishes,
180 including *Salvelinus alpinus* (n=66), *Salvelinus namaycush* (n=32), *Oncorhynchus mykiss* (n=10) and
181 *Salmo trutta fario* (n=1). We have sample a portion of caudal muscle, for determination of THg and
182 MeHg. For a standard length of 210mm, statistical analysis shows that no difference exists between
183 species in each lakes regarding THg concentration in muscle. That's why, we can compared each lake,
184 considering the fish community. Figure 3 shows different concentrations obtained for each lake, for a
185 standard length of 210mm. We can observe that there is no difference for La Sagne, Crop and
186 Poursollet lake, with THg concentration of 0.19, 0.21, 0.14 mg.kg⁻¹, respectively. Regarding fishes of
187 Bramant Lake, we note no difference with fishes of Poursollet Lake, but a significant difference with
188 La Sagne and Crop Lake. Regarding

189

190 **4. Discussion**

191 **4.1. Total mercury and methylmercury concentrations in lakes.**

192 Results presented in this study show mercury and methylmercury concentrations in water column
193 relatively low, but similar compared to small lakes situated in U.S.A (Barbiaz and Andren, 1995;
194 Monson and Brezonik, 1998). In surface water, mercury comes mainly from atmospheric deposition
195 (dry or wet), runoff and groundwater (Driscoll et al., 2007). In altitude, lakes are covered by snow
196 and ice during 6-7 months a year (December to June), and the main water supply for the lake are
197 runoff and melt water. These waters are drained by watershed and we know that the transport of
198 mercury is primarily mediated by dissolved organic carbon (DOC), a leading carrier of mercury
199 through the watershed (Grigal, 2002). Also, a forest or vegetation coverage on watershed, promote
200 mercury absorption and may be leached in throughfall (Lindberg et al 2005). Three lakes studied
201 have few vegetation and forest coverage. The watershed composition is mainly composed of rocks
202 (Figure X). Mercury can be eluted more easily (not retained by vegetation and forest that could be
203 the watershed, and therefore the organic matter) in the direction of Lake. In addition, the renewal of
204 the lake in summer is very important, In particular with via runoff resulting mainly meltwater. When
205 the ice cover melts, and that the runoff joins the lake, lake volume increases rapidly and a brewing of
206 water occurs, allowing for a substantial renewal of the lake waters. As a result, mercury arriving

207 mainly via runoff in lake is evacuated quickly by effluent. Therefore, mercury present in the lake has
208 too little time to reside in the water column or to deposit in the lake bottom. Thus, possibility of
209 mercury methylation is also low. In addition, these lakes are dimictic, that is to say, that we observe
210 two stratifications by year: an inverse stratification in winter when lake is frozen, and a thermal
211 stratification in warm season. We know that when a lake is stratified, the hypolimnion (water layer
212 situated at the bottom of lake and above the sediment) becomes anoxic, which may promote
213 mercury méthylation (REF). Now, on the profiles observe (figure x), we see that methylmercury
214 concentration not excess 2 % of total mercury. We know that an important brewing and renewal of
215 the lake water occurs in summer, that why, the hypolimnion is anoxic too little time, which reduce
216 the possibility of mercury methylation.

217 Runoff may come from others lakes situated in watershed. In the four lakes studied (Bramant lake, La
218 Sagne lake, Poursollet Lake and Crop lake), mercury concentrations are never greater than 0.5 ng.L^{-1}
219 for 3 lakes, except La Sagne lake, where mercury concentration in surface water is equal to 3.12 ng.L^{-1} .
220 This difference can be explained by the fact that La Sagne lake is situated in the watershed composed
221 of numerous lakes connected together (figure 1). Here, La Sagne Lake is a lake of order 5. It is known,
222 that the lake order can be affect total mercury contamination. That's why we can consider that the
223 difference between mercury concentrations in water column of each lake, may be due to an
224 accumulation of mercury from water of different runoff situated upstream of La Sagne Lake. To
225 explore possible inputs of total mercury to La Sagne Lake by runoff, we have sampled runoff water.
226 Results confirmed that we have a runoff contribution of total mercury in water of La Sagne Lake.
227 Indeed, mercury concentrations measured in runoff are similar to mercury concentration in surface
228 water and water column.

229 Finally, we cannot exclude the fact that the low mercury and methylmercury concentration in water,
230 can be due to a reduction of ions Hg to Hg^0 by photochemical process (Amyot et al., 1997) or by a
231 microbial activities in water column.

232

233 *4.2. Total mercury concentration in fish population*

234 Total mercury concentrations in fish population, for a standard length of 210 mm, showed in this
235 study are relatively low (inferior to 0.22 mg.kg^{-1}) and not exceeded 0.5 mg.Kg^{-1} , fish consumption
236 advisory limit established for mercury by the World Health Organization. In addition, there are
237 similar to others studies in Europe for all lakes (Blais, 2006; Rognerud et al., 2005). In these studies,
238 Blais et al., show that in The French Pyrénées, the altitude can be a factor explaining mercury
239 concentration in fish population. In fact, authors explain that in altitude, deposition and/or retention
240 of Hg is more important, that why mercury concentration in fish are higher in altitude. However, in
241 our study, with a range elevation of 800 meters, we observe no difference with mercury

242 concentration in fish population between three lakes (Crop Lake, La Sagne Lake and Poursollet Lake).
243 Instead, Bramant Lake located at 2448m a.s.l (lake more elevated), is lake where mercury
244 concentration is lowest. In addition, we knew that watershed characteristics may influence mercury
245 concentration in water column, by a renewal water in summer, that why we can think, that the little
246 time of residence of mercury in water column, can explain the low mercury concentration in fish
247 population.

248 Regarding Study of Rognerud et al., authors explain the low mercury concentrations by : i) a trophic
249 level low of fish population studied, ii) a flow of Hg in sediments low, and iii) a low rates of
250 methylation in the cold and clear lakes. The two first hypotheses can explain the low level of mercury
251 in fish population. Indeed, in our lakes, and mountain lakes generally, food web is brief. Fish
252 population feeds mainly of insects or, we can observe a phenomenon of cannibalism between
253 species. But, it knew that trophic position influe on mercury contamination and that a fish with a
254 trophic level high will be more contaminated. Consequently, fish population studied here has a
255 trophic level low, combined with a short food web, may explain the low mercury level in fish
256 population. In addition, the watershed composition is mainly composed of rock, consequently, when
257 runoffs join lakes, few little particles and few DOC join water, essential elements for a methylation
258 rates elevated.

259 Finally, the low mercury concentration in fish population studied here, can be explain by the fact that
260 these lakes are situated in altitude, and that in this region, mercury contamination is mainly due to
261 by atmospheric mercury transport and deposition, and that these lakes are located far away of
262 anthropogenic sources of pollution. This can explain the difference with study conducted in Canada
263 or Amazonia.

264

265 **5. Conclusions**

266 The present article provides the first study on mercury contamination in water column and fish
267 population in the French Alps. We show that Hg and MeHg concentration in water lake and fish
268 muscles are very low in comparison to others studies in North-America, but similar with studies
269 conducted in Europe. We explain these results by watersheds compositions and by the fact that in
270 summer, an important water renewal conducted Hg in direction of effluents, consequently, Hg
271 cannot reside in water column or to deposit in sediment. A significant effort must be however
272 conducted to identify the source of Hg and to know mercury distribution in fishes.

273

274

275

276

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283

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285

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355

356

357 **Table 1 : Lakes and watershed characteristics.**

Lakes	Location	Altitude (meter)	Lake area (km ²)	Catchment area (km ²)	% of tree	% of wild grass	% of land	% of rock	% of snow/ice
Poursollet	45°03'08" N, E5°54'00" E	1649	0.017	0,71	57,16	33,01	2,43	0,97	0,12
Crop	45°12'28" N, 5°59'16" E	1906	0.051	1,71	4,11	48,4	10,49	15,51	1,01
La Sagne	45°13'15" N, E6°04'33" E	2067	0.065	11,19	0,31	13,47	8,8	62,48	3,64
Bramant	45°12'00" N, E6°10'34" E	2448	0.144	5,81	0,78	9,33	7,59	40,74	36,85

358

359 Table 2: Characteristics for fish population and mercury concentrations in fish muscles. THg
 360 predicted, for a standard length of 210mm is shown.

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Variable	Unit	n	Mean	SE	range
Poursollet					
Length	mm	7	277	28.9	230-300
Weight	g	7	322	197	273-374
Age	yr	7	6.42	0.66	4-8
THg	mg.kg ⁻¹	7	0.10	0.039	0.07-0.16
THg predicted (st. length of 210 mm)	mg.kg ⁻¹	7	0.14	/	/
Crop					
Length	mm	35	192	12.9	130-620
Weight	g	35	285	88	24-4000
Age	yr	35	4.97	0.29	3-12
THg	mg.kg ⁻¹	35	0.19	0.017	0.009-0.65
THg predicted (st. length of 210 mm)	mg.kg ⁻¹	35	0.21	/	/
La Sagne					
Length	mm	21	213	16.7	135-520
Weight	g	21	191	113	38-2300
Age	yr	21	5.33	0.38	4-8
THg	mg.kg ⁻¹	21	0.20	0.022	0.06-0.75
THg predicted (st. length of 210 mm)	mg.kg ⁻¹	21	0.19	/	/
Bramant					
Length	mm	46	208	11.2	135-275
Weight	g	46	124	77	27-272
Age	yr	46	4.58	0.25	2-10
THg	mg.kg ⁻¹	46	0.097	0.015	0.03-0.25
THg predicted (st. length of 210 mm)	mg.kg ⁻¹	46	0.083	/	/

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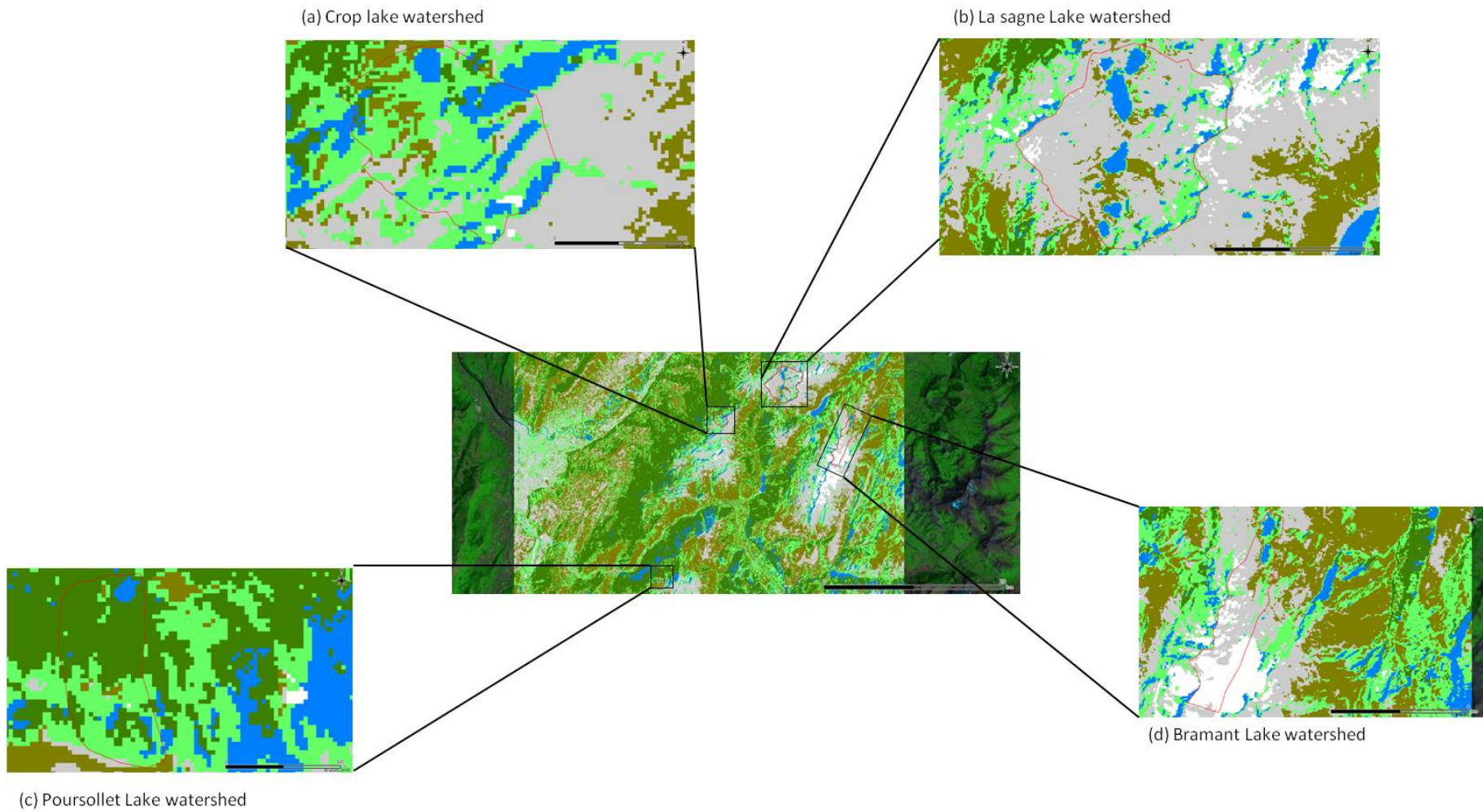
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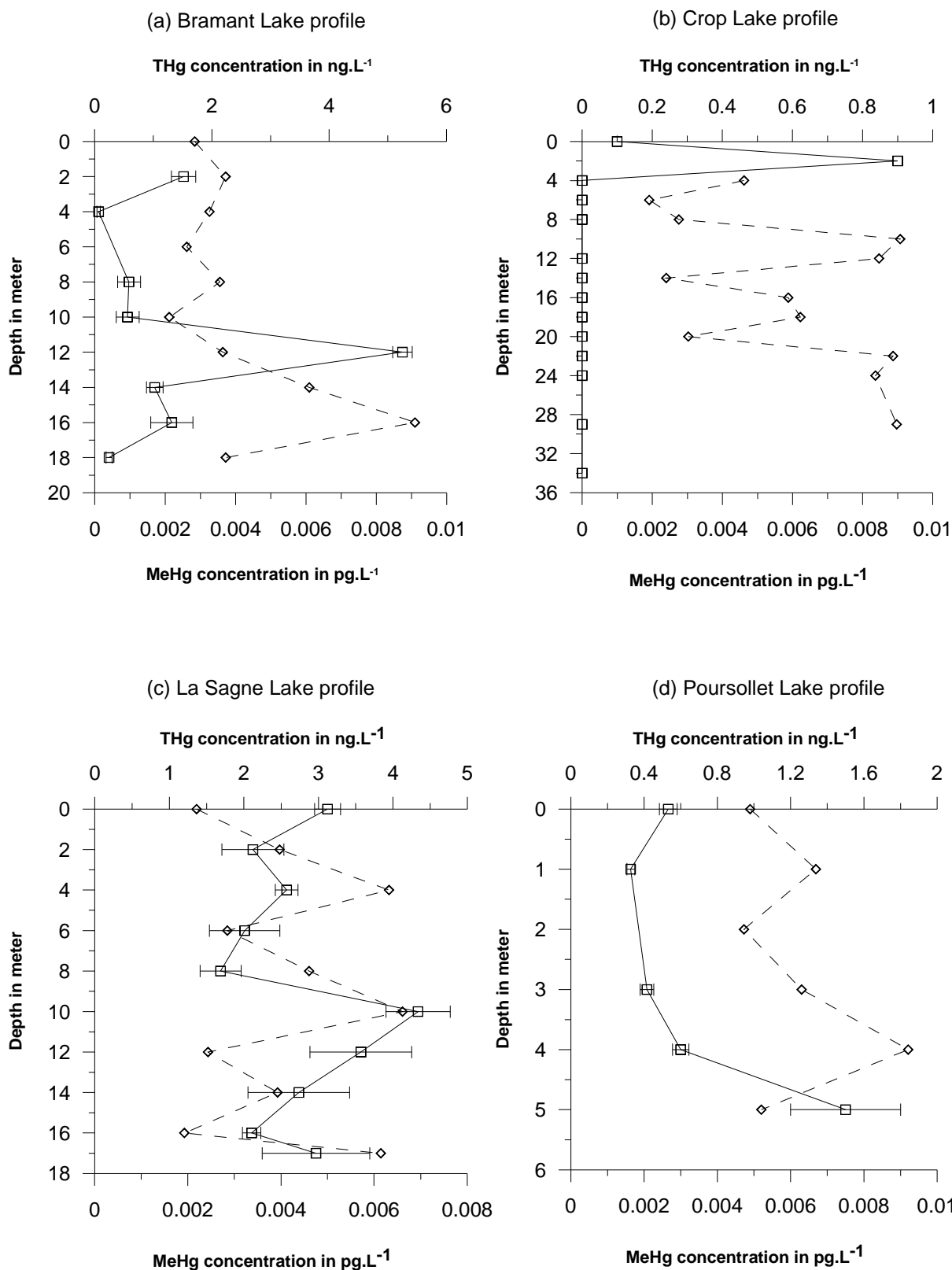
366 **Figure 1 : Map of watershed composition of Crop lake (a), La Sagne lake (b), Poursollet Lake (c) and Bramant lake (d).**

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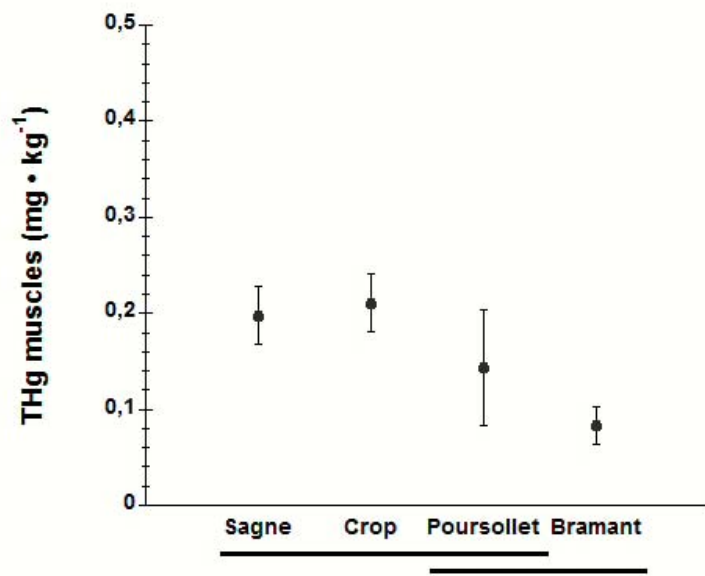
390 **Figure 2 : THg and MeHg profiles in ng.L⁻¹ in water column (—) represent THg and (---)represent MeHg**
 391 **profiles.**

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428 Figure 3 : THg concentration in fish population for a standard length of 210mm

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