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1     **Tracking sources and transfer of contamination according to pollutants variety at the**  
2             **sediment-biota interface using a clam as bioindicator in peri-alpine lakes**

3

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5

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12 **Abstract**

13 Point pollution sources may differently impact lakes littoral, possibly leading to local ecological risks.  
14 The concomitant chemical analysis of littoral-benthic organisms and sediment can provide insights  
15 into the bioavailability and thus the ecological risk of contaminants. In this study, the autochthonous  
16 *Corbicula fluminea* was used to assess the sources and transfer of six trace metals (TMs) and fourteen  
17 Polycyclic Aromatic Hydrocarbons (PAHs) to the littoral-benthic biota of a large lake. The  
18 contaminant concentrations spatially varied with a value scale from 1 to 280 000 times along the lake  
19 littoral in both the sediment and clams. Multiple linear regressions were performed to explain the  
20 spatial variability of *Corbicula fluminea* contamination by considering both watershed and in-lake  
21 sources. The concentration of the sum of PAHs in clams was significantly correlated with sediment  
22 contamination, suggesting that PAHs contamination of the benthic biota mainly occur from the  
23 sediment. Most of the internal TM concentrations of clams were significantly correlated with  
24 stormwater drainage areas in the lake watershed, highlighting the importance of stormwater runoffs in  
25 the littoral biota contamination. The transfer of TMs and PAHs was assessed through the  
26 bioconcentration factor defined as the ratio of internal and sediment concentrations. As, Cd, Cu, Zn  
27 and light molecular weight PAHs were more bioconcentrated in *C. fluminea* than Pb, Sn and heavy  
28 molecular weight PAHs, suggesting differences in their bioavailability. This study underlines the  
29 relevance of using autochthonous organisms as bioindicators of lake littoral biota contamination  
30 concomitantly with sediment matrices, and illustrates the challenge of tracking pollution sources in  
31 lakes.

32 **Keywords**

33 Trace metal, polycyclic aromatic hydrocarbon, bioindicator, bioconcentration, *Corbicula fluminea*,

34 littoral.

## 35 **1. Introduction**

36 The sediment compartment is of great importance when assessing the ecological risk of contaminants  
37 for biological organisms (Atkinson et al., 2007; Bryan and Langston, 1992; Burton, 2018). Chemical  
38 analyses of sediments can be compared to ecotoxicological thresholds from the literature (e.g.  
39 Threshold Effect Concentration (MacDonald et al., 2000)). Sediment can also be used in bioassays as  
40 a contaminated matrix to which study laboratory organisms are exposed to assess their response  
41 (Triffault-Bouchet et al., 2005). In the literature, the sediment is considered as a more stable and  
42 integrative compartment compared to water (Payne et al., 1996; Sutherland, 2000) and can exhibit  
43 significantly higher contaminant concentrations leading to a higher ecological risk (Atkinson et al.,  
44 2007; Bryan and Langston, 1992). However, the binding capacity of pollutants to the sediment greatly  
45 varies according to their molecular structure. As an example, PAHs with low octanol / water  
46 partitioning coefficient ( $K_{ow}$ ) tend to bind to the sediment organic matter in a lesser extent than those  
47 with higher  $K_{ow}$  (Harner et al., 2004; Naffrechoux et al., 2015; Shoeib and Harner, 2002). Secondly,  
48 many abiotic (e.g. temperature, dissolved oxygen, hydrodynamic regime) and biotic (e.g. trophic  
49 interactions, bioturbation) factors can influence the bioavailability of sediment-borne contaminants  
50 (Eggleton and Thomas, 2004). It is therefore essential to perform concomitant measurements in  
51 biological matrices, in addition to sediment, to provide better insights regarding the bioavailability of  
52 sediment-borne contaminants (Guo and Feng, 2018) and their transfer throughout lake food webs.

53 The Asian clam *Corbicula fluminea* is a benthic filter-feeder (Cataldo et al., 2001) commonly used as  
54 a bioindicator of the ecological status of stream and coastal environments over large time (Riva et al.,  
55 2010) and spatial (Goldberg, 1986) scales. *C. fluminea* is a widespread bivalve mollusk living in the  
56 upper part of sediment and filtering water at the sediment surface. These characteristics make *C.*  
57 *fluminea* a great biological model to test for the bioavailability of contamination at the sediment-water  
58 interface.

59 Lake Annecy is a large French peri-alpine lake with an anthropized watershed. Although the physico-  
60 chemical and biological parameters of the water column have been monitored for several decades  
61 (Frossard et al., 2018), only a few studies have focused on anthropogenic contamination (GREBE,

62 2008; Lallée, 2009; Naffrechoux et al., 2015) and mostly concern contamination in the deepest part of  
63 the lake.

64 Using Lake Annecy as case study, we aim at assessing whether the contamination of *C. fluminea*  
65 provides insights (1) into the transfer and bioavailability of sediment-borne pollutants to benthic  
66 organisms in the lake littoral zone, and (2) into the identification of sites and point sources that may  
67 exert ecological risks. In particular, this study attempts to assess the contribution of potential in-lake  
68 versus watershed sources in the contamination of the littoral biota by studying *C. fluminea* multi-  
69 contamination.

70 Two contaminant families were investigated; Trace Metals and Metalloids (TMs) and Polycyclic  
71 Aromatic Hydrocarbons (PAHs). These families are widespread in the lacustrine environment (Järup,  
72 2003; Pacyna et al., 2016; Schwarzenbach et al., 2010) and mainly reflect domestic, industrial and  
73 transport activities (e.g. wood and fossil fuel combustion, fuel leakage, corrosion of roofs and vehicle  
74 bodies, road, brake and tire wear abrasions (Abdel-Shafy and Mansour, 2016; Shen et al., 2013)).  
75 Among these anthropogenic activities, the pollutants emitted from transport activities can both  
76 originate from in-lake (e.g. boats emissions (Hoch, 2001)) and lake watershed. The objective was to  
77 identify a large array of potential pollution sources with these two contaminant families.

78 In addition, TMs and PAHs differ by their mobility, persistence, bioavailability and toxicity (Naimo,  
79 1995; UNEP, 2003), thereby justifying a large-scale assessment of *C. fluminea*'s capacity to reflect  
80 biota contamination in the littoral zone.

81 We first expected that *C. fluminea* contamination would exhibit significant spatial variability in the  
82 littoral zone due to the influence of local point sources of pollution. In one side, harbors were expected  
83 to exert significant in-lake influence on littoral biota contamination, with a greater concentration of  
84 TMs in organism (Guevara-Riba et al., 2004). In the other side, external sources, such as surface and  
85 stormwater runoffs, were expected to play a role in the contamination by PAHs of the littoral biota  
86 (Brown and Peake, 2006). In addition, two buffer zones around the study sites of respectively 200 and  
87 600 m were assessed in order to account for small and medium-scale sources that may influence *C.*  
88 *fluminea* contamination. We therefore expected that point sources included in the small buffer zone  
89 (i.e. 200 m) would more precisely explain the clams contamination than higher scales. The last

90 assumption was that *C. fluminea* contamination would differ from sediment contamination according  
91 to the bioavailability of the contaminants. Particularly, differences in PAH contaminations in the  
92 sediment and biological matrices should be greater (i.e. by bioconcentration effect) than for TMs due  
93 to their greater reactivity (i.e. lipophilicity, molecular degradation through biological, chemical and  
94 physical processes; Moza et al., 1999). However, some differences should be expected according to  
95 light and heavy molecular weight PAHs that exhibit different molecular properties (e.g. water / octanol  
96 partitioning coefficient  $K_{ow}$ ) (Ha et al., 2019; Noh et al., 2018).

97

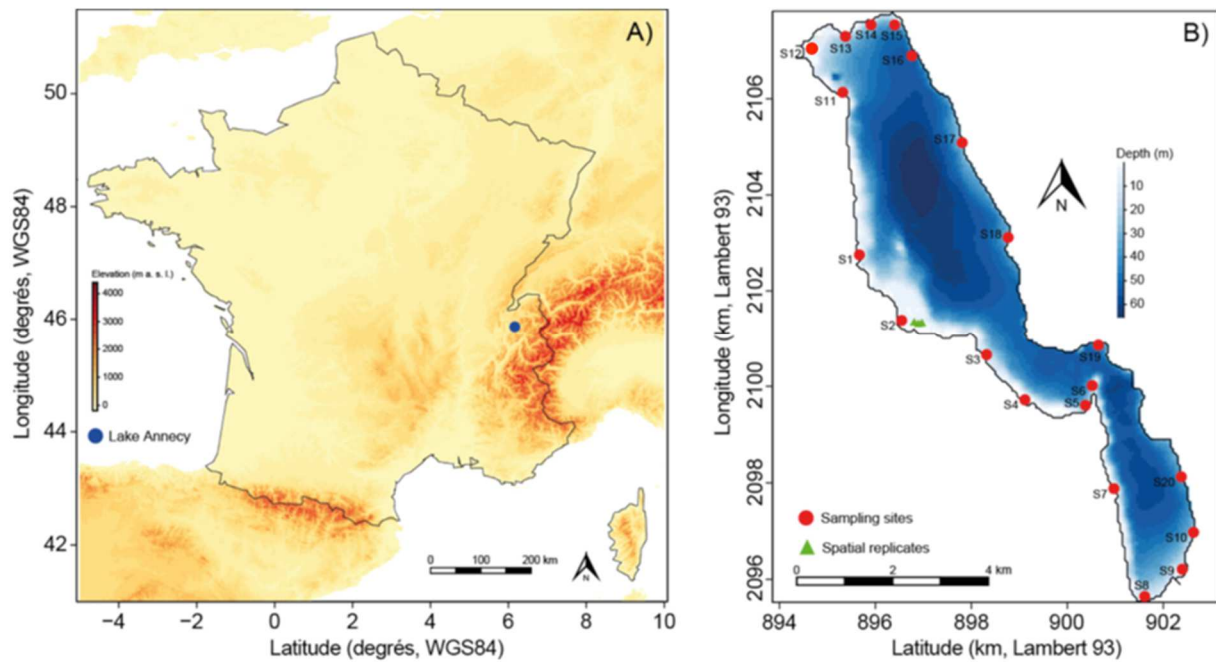
98

## 99 **2. Materials and Methods**

### 100 2.1. The study lake

101 Lake Annecy is a large monomictic and oligotrophic lake located in Savoie (France), on the northwest  
102 edge of the French Alps (45°51'N, 6°10'E, Figure 1A). It has a surface area of 27 km<sup>2</sup> and mean and  
103 maximum depths of 41 and 65 m, respectively. The 250 km<sup>2</sup>-catchment area of Lake Annecy is  
104 covered by forests (56 %), agricultural areas (20 %), urban areas (17 %) and water bodies (6 %)  
105 (Corinne Land Cover, 2012). Its watershed has been subject to rapid anthropization over recent  
106 decades (Rulliere et al., 1972). The total population in the watershed, about 125 000 inhabitants,  
107 doubles in summer during the tourist season.

108



109

110 **Figure 1.** Map of Lake Annecy in the east of France (A) and the locations of the sampling sites (B).

111 Red circles indicate main sampling sites and green triangles replicates for assessment of local  
 112 contamination variability.

113

114 2.2. Sampling sites

115 Twenty littoral sites were selected to cover the overall lake littoral and included different land-use  
 116 contexts (i.e. beaches, reed beds, roadsides, harbors) (Figure 1-B). At all sites, surface sediments (i.e.  
 117 the first 5-cm sediments) and *C. fluminea* were sampled using an Ekman grab at a water depth of 1.5  
 118 to 2.5 m, depending on the topographic constraints. Sediment samples were homogenized, sieved  
 119 through a 2-mm mesh to remove debris and stored at 4°C.

120 The sampled *C. fluminea* were selected according to similar ranges of shell size (i.e. between 1.5 and  
 121 2.5 cm) to limit possible ontogenic differences in contamination. We checked for the  
 122 representativeness of each sample by sampling 3 replicates close to each other (<12m) in the site S2  
 123 (Figure 1). No significant differences in *C. fluminea* contamination for PAHs and TMs were found  
 124 (see details in Figure A1 in the Appendix).

125

126



127 2.3. Contamination quantification

128 The glassware used for sampling and measurements was prior washed in a detergent bath (5%  
129 Gigapur® 05 (Shimitek, France) for 12 h, followed by an acid bath (5% nitric acid) during 6 hours and  
130 rinsed with deionized water.

131 TM concentrations in sediment and organism samples were assessed by inductively coupled plasma  
132 mass spectrometry (ICP-MS) according to the European guideline NF EN ISO17294-2 (AFNOR,  
133 2016). Incertitude measurements were always under 25 % and detection limit for these TMs was about  
134 0.1  $\mu\text{g L}^{-1}$ . PAH concentrations in sediment and organism samples were measured by LCME  
135 Laboratory using high performance liquid chromatography (HPLC) with fluorescence detection. The  
136 employed methodology was similar as the one described in Lécivain et al. (2018). The limit of  
137 detection and quantification corresponded to 6 and 12 times the blank standard deviation, respectively.  
138 Among the 16 PAHs prioritized by the US Environmental Protection Agency (US-EPA), 14 were  
139 quantified; acenaphthene (Ace), fluorene (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene  
140 (Fla), pyrene (Pyr), benzo[a]anthracene (BaA), chrysene (Chr), benzo[b]fluoranthene (BbF),  
141 benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), dibenzo[ah]anthracene (DbahA),  
142 benzo[g,h,i]perylene (BghiP) and indeno[1,2,3-c,d]pyrene (IP). Light molecular weight PAHs, also  
143 called light PAHs are Ace, Flu, Phe, Ant and Pyr, while the others are considered as heavy PAHs. To  
144 limit biases in the analysis of contamination data related to physicochemical properties, PAH  
145 concentrations in sediments and *C. fluminea* were normalized to organic matter (OM) concentration  
146 and lipid content, respectively. The average percentage of lipids for all the *C. fluminea* sampled was  
147 estimated at 12.9% based on C:N ratios (Figure A2 in the Appendix). The organic matter content of  
148 the sediments was estimated by measuring loss on ignition according to Heiri et al. (2001) (Figure  
149 A3). The sum of the 14 PAHs was thus expressed in  $\mu\text{g kg}^{-1}$  OM for the sediments and in  $\mu\text{g kg}^{-1}$   
150 lipids for *C. fluminea*. Grain size distribution of sediment was measured by laser diffractometry  
151 (Malvern Mastersizer 2000G) and presented a homogeneous sandy size group (Figure A3). The grain  
152 size distribution of sampled sediment could not consequently exert a major bias in the contaminant  
153 bioavailability (Eggleton and Thomas, 2004; Reid et al., 2000; Zhang et al., 2014). Concentrations

154 below the detection limits were estimated at a value equal to the detection limit divided by  $\sqrt{2}$   
155 according to Croghan and Egeghy (2003). The raw data of sediment and *C. fluminea* contaminant  
156 concentrations are presented in Table A1 in the Appendix.

157

#### 158 2.4. Bioconcentration factor (BCF)

159 It was assumed that the sampled organisms had already reached an equilibrium regarding the ambient  
160 contamination since they were autochthonous and adult clams (Jacomini et al., 2006; Thorsen et al.,  
161 2007). The transfer from sediment borne contaminants to the clams could so be assessed through the  
162 ratio of the non- normalized contaminant concentrations between *C. fluminea* and the sediments  
163 designated as the bioconcentration factor (BCF) (Jia et al., 2018; Mendoza-Carranza et al., 2016;  
164 Mountouris et al., 2002). A contaminant was considered as bioconcentrated when the BCF was equal  
165 to or greater than 1. The relationship between the BCFs and the sediment contamination was studied to  
166 assess whether potential biological regulation mechanisms may occur in the organisms exposed to an  
167 increasing sediment contamination (McGeer et al., 2003).

168

#### 169 2.5. Explanatory variables of *C. fluminea* contamination

170 We defined explanatory variables which represented in-lake and external contamination sources to  
171 identify which one contributed to contaminant concentration in clams. First, potential in-lake sources  
172 of pollution were considered using the total engine power of boats (hp) in the harbors, to provide a  
173 realistic pollution capacity of the boats (Figure A4 in the Appendix). A total of 1635 boats were  
174 counted throughout the lake littoral, moored at private pontoons and in large public harbors. The  
175 individual power of each engine was read directly on the engine for 70% of the boats and estimated for  
176 the remaining 30%. Second potential watershed sources consisted of the number of stormwater outlets  
177 and their associated drainage areas (Figure A4 in the Appendix). To test the influence of the spatial  
178 scale in *C. fluminea* contamination by these candidate variables, two buffer sizes around the sampling  
179 sites were considered, i.e. 200 and 600 m (Figure A5 in the Appendix). The mean wave energy and  
180 slope and the sediment organic content of the sampling sites were also considered to influence the

181 contamination of *C. fluminea*. The mean wave energy was calculated as a proxy of the hydrodynamic  
182 regime within a 200 m diameter circle around each site, based on the hydrodynamic models described  
183 in Rohweder et al. (2008) and Marchand and Gill (2017), by coupling bathymetric data from the  
184 BRGM / SILA lake (Bisson et al., 2006) and the direction and wind speed from the Météo-France  
185 Meythet station (i.e. hourly data over 4 annual cycles). The average wave energy within a 200 m  
186 diameter circle around the site was calculated over the entire meteorological time range (Figure A6 in  
187 the Appendix). The mean slope (%) within a 50 m radius around the site was calculated from BRGM /  
188 SILA (1990) bathymetric lake data. The sediment contamination was also considered to explain the  
189 contamination of *C. fluminea*.

190

## 191 2.6. Data analysis

192 We assessed the contamination in *C. fluminea* performing multiple linear regressions with backward  
193 selection, using *C. fluminea* contamination (for each TMs separately and for the sum of the PAHs) as  
194 response variable against the potential explanatory variables (i.e. section 2.5). Site S11 was removed  
195 prior to modeling for the PAHs because of its concentration several orders of magnitude higher than  
196 the other sites that can be explained by the vicinity of a main road with huge daily traffic. For the  
197 TMs, sites S7, S8, S10, S13, S16, S17, S18, S20 could not be considered since the biological masses  
198 sampled were not sufficient to carry out the analyses. Results of the models were illustrated in two  
199 heatmaps, one for each study scale (i.e. radii of 200 and 600 m). All the statistical analyses and models  
200 as well as the graphical displays were carried out using R (Team, 2017).

201

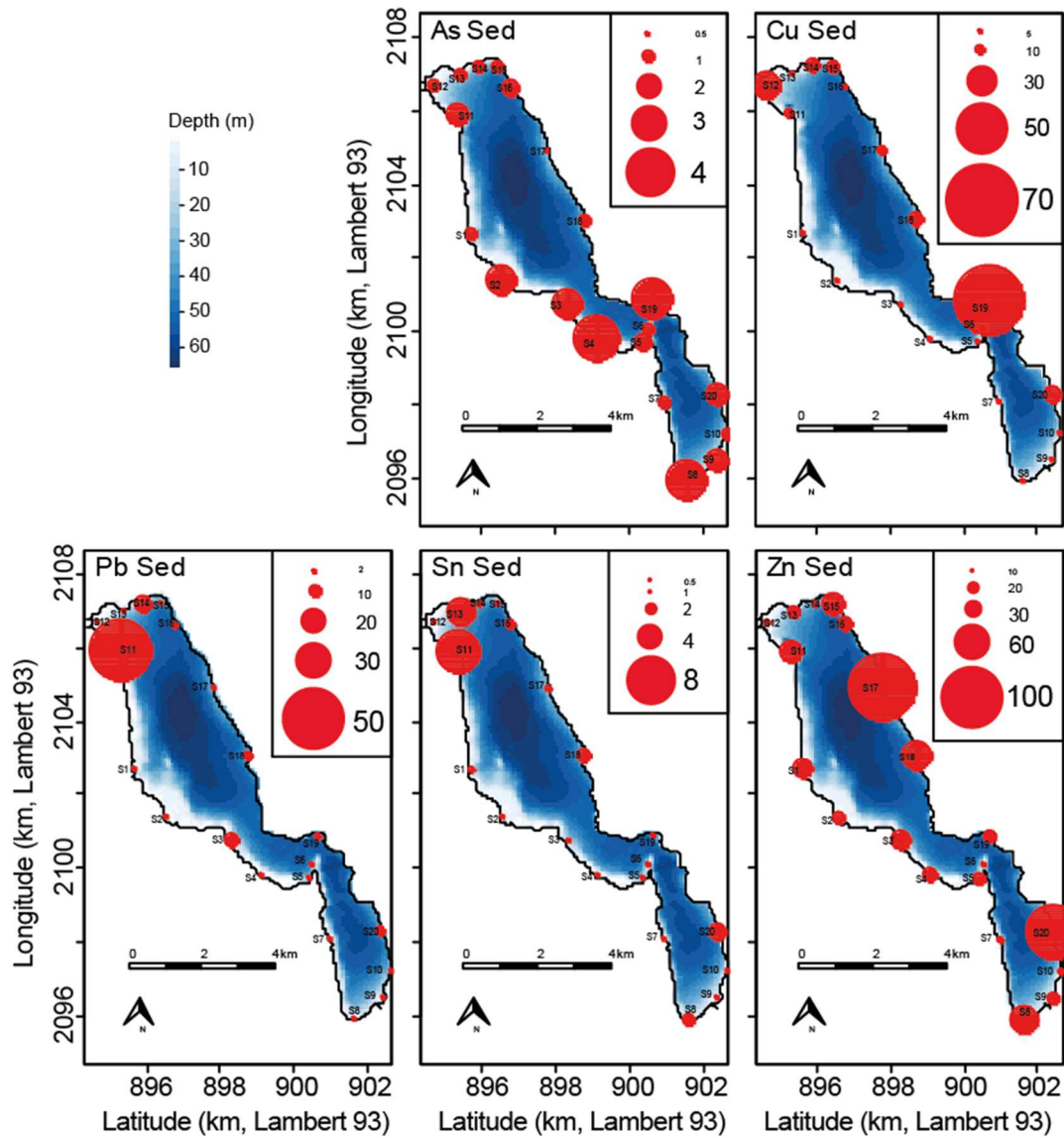
202

## 203 3. Results

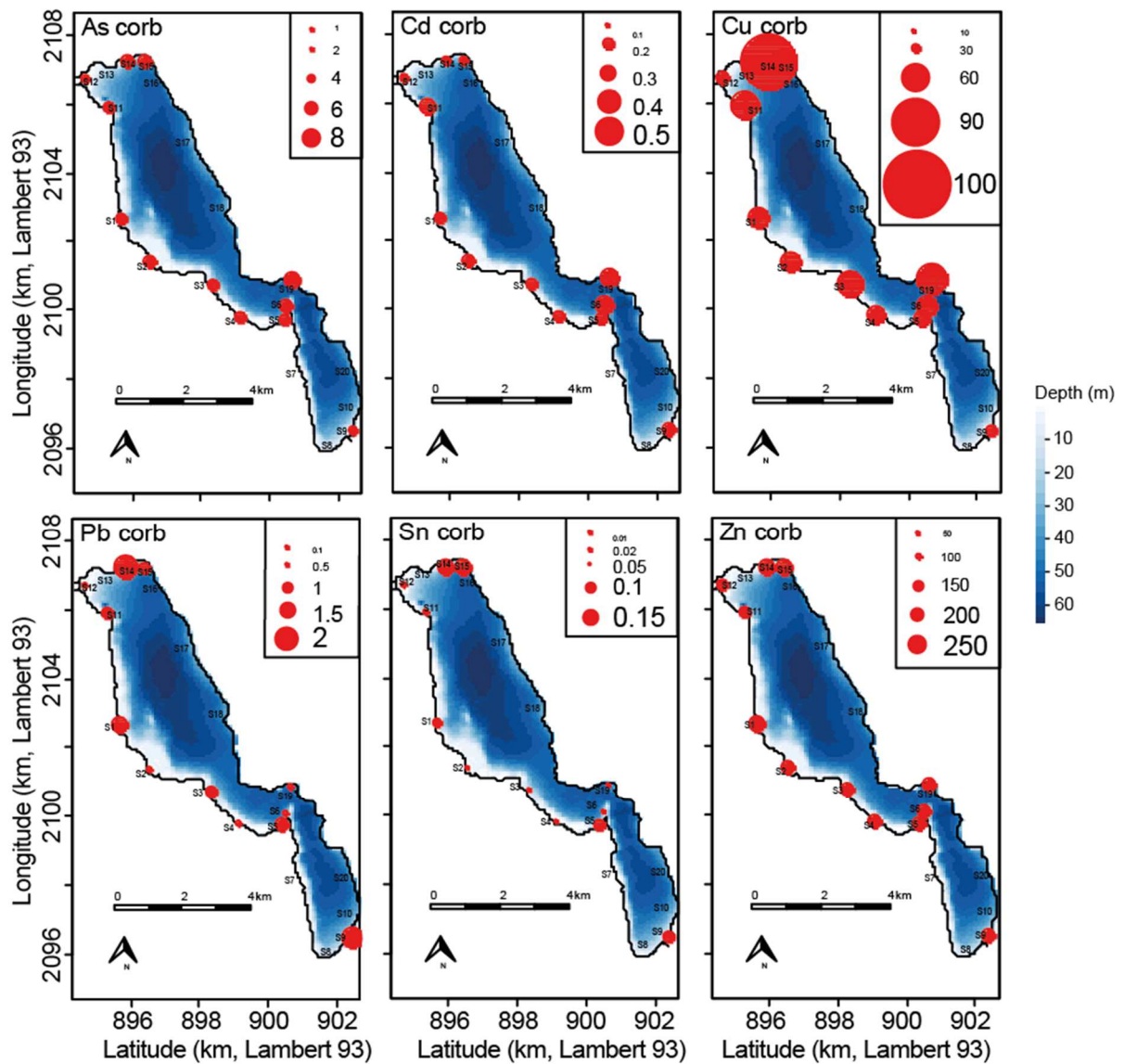
### 204 3.1. Sediment and *C. fluminea* contaminations

205 The TMs contamination of the sediment exhibited a high spatial heterogeneity along lake littoral with  
206 a value scale from 1 to 8 (As), 9 (Pb), 12 (Cu), 31 (Zn) and 42 (Sn) times (Figure 2). Similarly,

207 internal concentrations in clams were also highly variable with a value scale from 1 to 2 (As, Cd), 3  
 208 (Sn) and 4 (Cu, Pb) times (Figure 3). However, the pattern of TMs contamination was significantly  
 209 different between the sediment and the organisms along the lake littoral (Chi<sup>2</sup> tests, p-value < 0.05).  
 210



211  
 212 **Figure 2.** Mapping the TMs sediment contamination (mg kg<sup>-1</sup> DM) along Lake Annecy littoral.  
 213



214

215 **Figure 3.** Mapping the TMs *C. fluminea* contamination (mg kg<sup>-1</sup> DM) along Lake Annecy littoral.

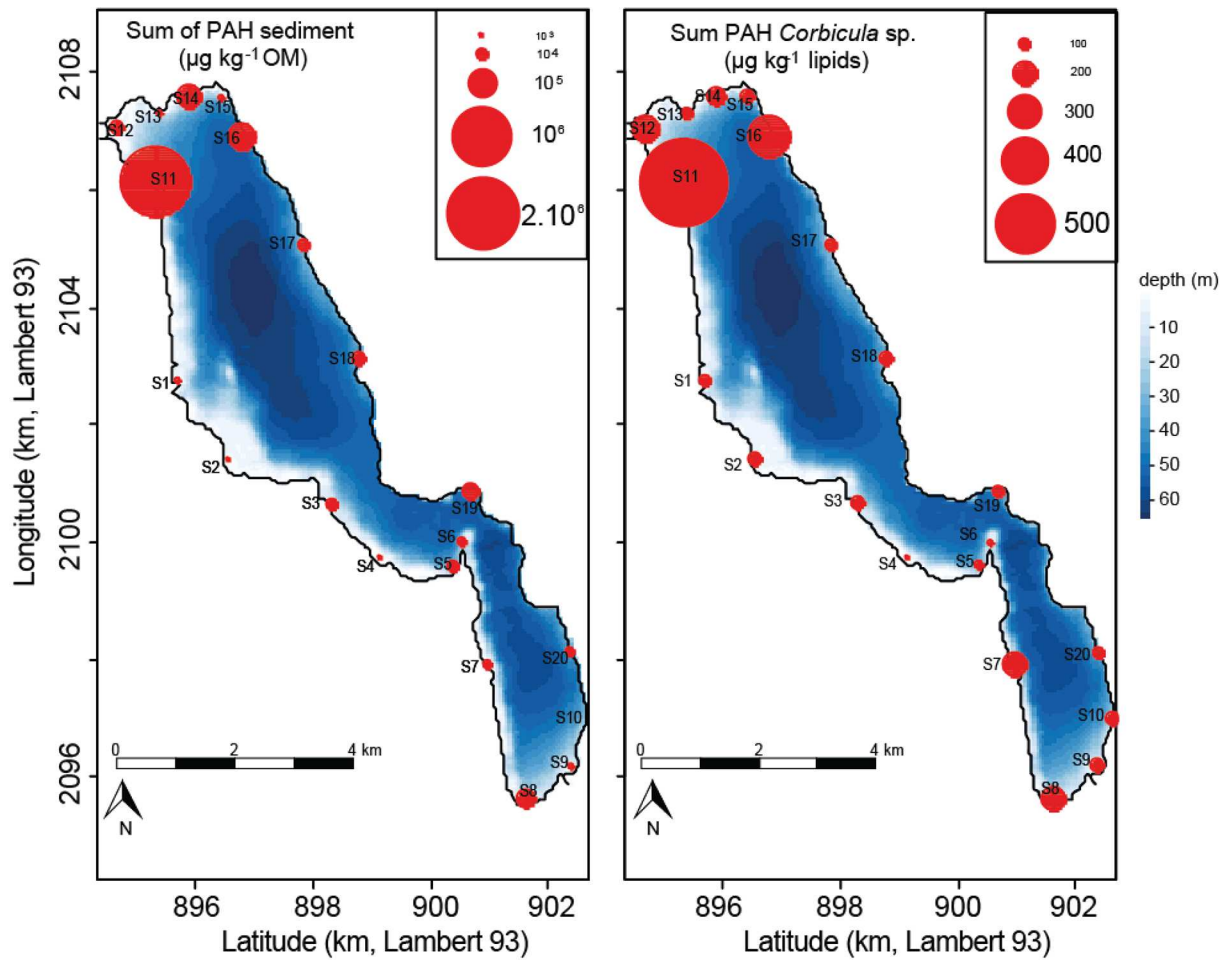
216

217 The average sum of PAHs in the lake littoral was equal to  $1.10^5 \pm 4.10^5 \mu\text{g kg}^{-1}$  OM in the sediment

218 and  $165.6 \pm 150.2 \mu\text{g kg}^{-1}$  lipids in *C. fluminea* and also varied considerably according to the location

219 considered in the littoral in both matrices (i.e. sediments and *C. fluminea*; Figure 4).

220



221

222 **Figure 4.** Map of the sum of PAHs in the sediment ( $\mu\text{g kg}^{-1}$  OM) on the left side and in *C. fluminea*

223 ( $\mu\text{g kg}^{-1}$  lipids) on the right side.

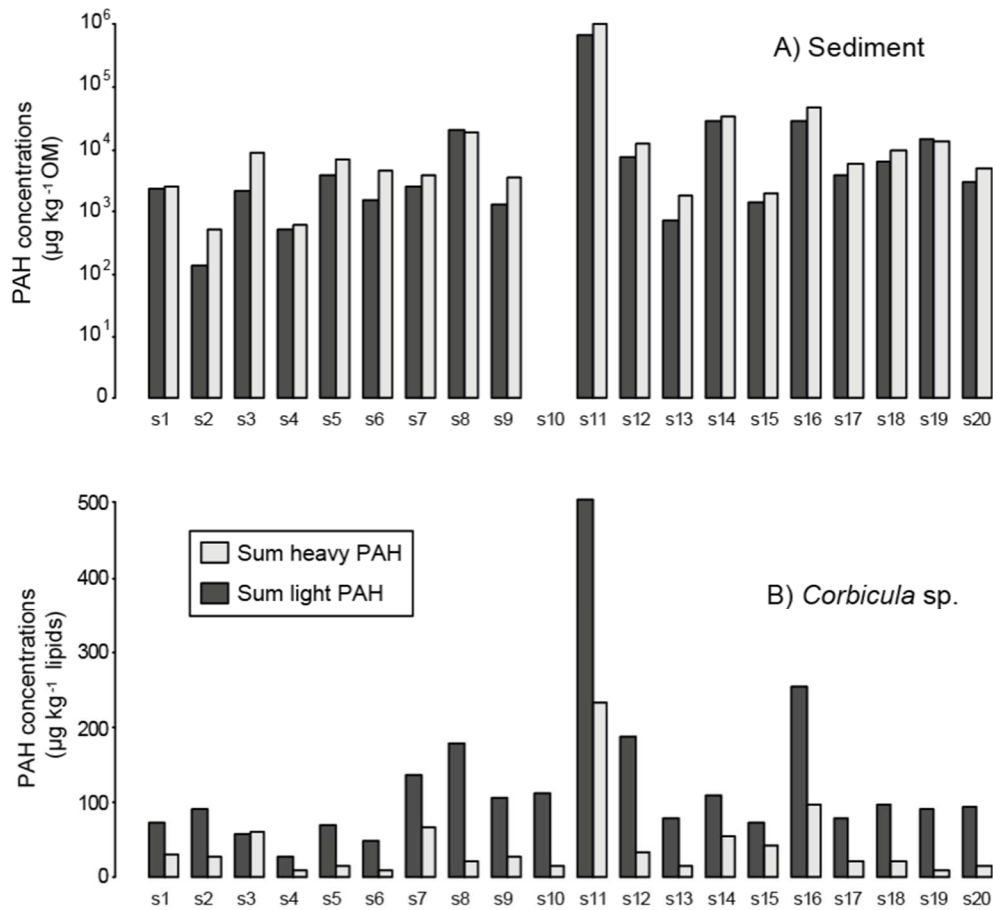
224

225 S11 exhibited the highest contamination with concentrations of  $1.7 \cdot 10^6 \mu\text{g kg}^{-1}$  OM in the sediment

226 and  $734.1 \mu\text{g kg}^{-1}$  lipids in *C. fluminea*. In addition, *C. fluminea* exhibited lower proportions of heavy

227 PAHs ( $23 \pm 10 \%$ ) than the sediment ( $62 \pm 10 \%$ ) (Figure 5).

228



229

230 **Figure 5.** Barplots representing the composition in light and heavy PAHs in A) the sediments ( $\mu\text{g kg}^{-1}$   
 231 OM) and B) *C. fluminea* ( $\mu\text{g kg}^{-1}$  lipids).

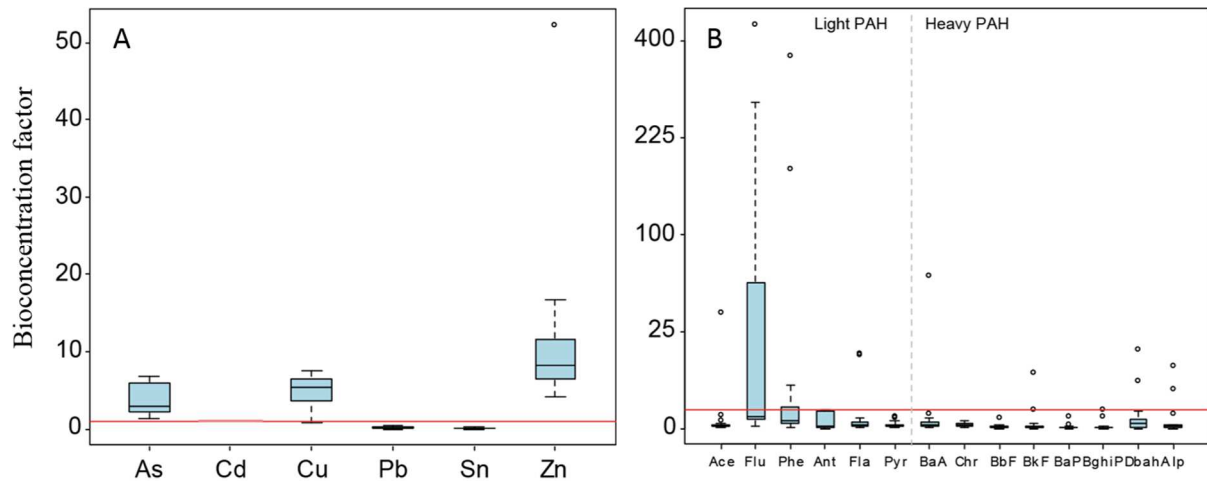
232

233 3.2. Contamination transfer

234 BCFs differed significantly among contaminants for both PAHs and TMs and among sites for PAHs  
 235 (ANOVA,  $p\text{-value} < 0.01$ ; Figure 6). For TMs, the BCFs for As, Cu and Zn were  $> 1$  while they were  
 236  $< 1$  for Pb and Sn (Figure 6A). The BCF for Cd could not be calculated, as it was not detected in the  
 237 sediment.

238 BCFs were higher for light PAHs compared to heavy ones, especially Flu for which values exceeded  
 239 400 (Figure 6B). Regarding the spatial distribution of BCFs along the lake littoral, S2 and S3 had the  
 240 highest BCFs (Table A2 in the Appendix).

241



242

243 **Figure 6.** Boxplots of the bioconcentration factors (BCF) for A) TMs and B) PAHs. The horizontal  
 244 line represents a BCF equal to 1.

245

246 3.3. Contamination sources

247 All the candidate variables were significantly related to *C. fluminea* contamination for at least one  
 248 contaminant for a given buffer radius, and the R-squared were relatively high and varied from 0.49 to  
 249 0.9 (except 0.28 for Pb at 200 m radius, Table 1).

250

251 **Table 1.** Adjusted R-squared (Adjusted-R<sup>2</sup>) values for each variable and radius that indicate the  
 252 proportion of variations in *C. fluminea* contamination explained in the final models.

Contaminants	As		Cd		Cu		Pb		Sn		Zn		Sum of PAHs	
	200 m	600 m	200 m	600 m	200 m	600 m	200 m	600 m	200 m	600 m	200 m	600 m	200 m	600 m
Adjusted-R <sup>2</sup>	0.54	0.63	NA	0.53	0.9	0.83	0.28	0.58	0.49	0.57	0.78	0.6	0.59	0.59

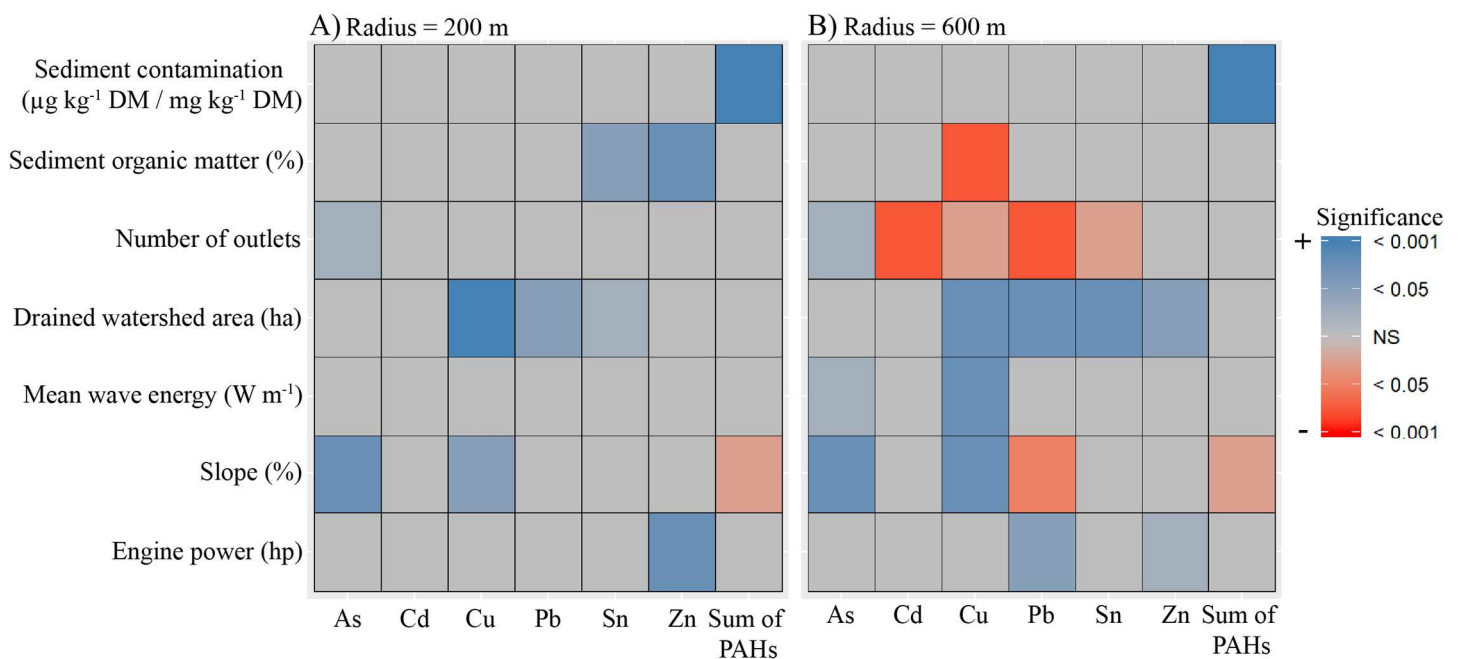
253

254

255 For the TMs, the sediment contamination was not correlated with that of *C. fluminea*, meaning that the  
 256 concentrations in the clams were likely not directly impacted by the sediment contamination (Figure  
 257 7). All other variables were at least significant for one TM and, most of them were positively  
 258 correlated with *C. fluminea* contamination, suggesting that these variables may favor their



259 contamination. However, at a large scale (i.e. radius = 600 m), some significant negative relationships  
 260 indicate that the influence of the explanatory variables may differ from one TM to another (Figure 7-  
 261 B). For example, the number of stormwater outlets exerted a significant positive influence on the  
 262 contamination for As and Cd but a negative one for Cu, Pb and Sn.  
 263 We also noted that many more variable were detected to have an effect on clams contamination in  
 264 TMs at the larger scale (600 m radius, Figure 7-B) than at the local scale (200 m radius, Figure 7-A),  
 265 except for the proportion of organic matter in sediment. R-square were also similar or higher for the  
 266 600 m radius confirming the better prediction power at this scale (Table 1).  
 267 Considering the sum of PAHs, the main explanatory variable was the sediment contamination while  
 268 the slope of the site had a marginally negative influence on internal PAH concentrations in *C. fluminea*  
 269 whatever the scale of the study (Figure 7). This result was similar for both radii.  
 270



272 **Figure 7.** Heatmaps illustrating the results from the multiple linear regressions relating *C. fluminea*  
 273 contamination to candidate variables according to A) radius = 200 m and B) radius = 600 m. Blue and  
 274 red colors symbolize a positive (+) and a negative (-) relationship with *C. fluminea* contamination  
 275 respectively. The significance rates were depicted by color intensity according to the legend intensity  
 276 on the right side of the figure. “NS” means “Non significant”.

277

278

## 279 4. Discussion

### 280 4.1. *C. fluminea* as a bioindicator of littoral contamination

281 As an invasive species and thus a highly widespread organism in freshwater ecosystems,  
282 autochthonous *C. fluminea* may provide useful information on the contamination patterns of lake  
283 littorals. In this study, mean PAH contamination of *C. fluminea* was low ( $21.5 \pm 19.4 \mu\text{g kg}^{-1} \text{DM}$ )  
284 compared to polluted sites such as Lake Kentucky ( $171.5 \mu\text{g kg}^{-1} \text{DM}$ ; (Lapviboonsuk and  
285 Loganathan, 2007)). In contrast, mean TM contaminations of *C. fluminea* were very similar to those  
286 measured in the *C. fluminea* from the littoral zone of Lake Bourget, located 20 kilometers from Lake  
287 Annecy (Table A3 in the Appendix). Similar concentrations were also reported by Shoults and Wilson  
288 et al. (2010) from the Altamaha River (Georgia, USA), except for Cd for which the contamination in  
289 Lake Annecy was on average 10 times lower than in the Altamaha River and even lower than the  
290 detection threshold in the sediment. In this study, the magnitude of both TM and PAH contaminations  
291 of *C. fluminea* varied from 5 to 10 times among sites and the patterns of contamination were related to  
292 different contamination sources. These benthic organisms may therefore efficiently reflect the spatial  
293 differences in the magnitude of contamination that the littoral faces.

294

### 295 4.2. In-lake versus watershed sources of contamination

296 In this study, both the 200 m and 600 m buffers were expected to account for small and medium-scale  
297 constraints that may influence *C. fluminea* contaminations. The explanatory powers of the multiple  
298 linear regression models performed with the 600 m buffer radii were higher in term of value and in  
299 number of significant variables, emphasizing the importance of selecting an appropriate buffer zone in  
300 such studies. The explanatory powers of the multiple linear regression models used to explain *C.*  
301 *fluminea* contamination were rather high although variable, suggesting that other non-considered  
302 parameters could also contribute to (i.e. sources) or influence (i.e. environmental variables) the  
303 contamination of organisms, such as bioturbation (Remaili et al., 2017; Schaffner et al., 1997).

304 The significant positive correlations of *C. fluminea* contamination by Cu, Pb, Sn and Zn with the  
305 drainage area in the lake watershed highlighted the potential role of stormwater as a major source of  
306 littoral biota contamination by TMs. This result is consistent with other studies that highlighted the  
307 influence of stormwater in lake contamination (McLellan et al., 2007; Zhang et al., 2016). Stormwater  
308 was also expected to explain the PAH contamination of *C. fluminea* (Brown and Peake 2005),  
309 although no significant correlation have been identified in this study. The influence of the total  
310 number of stormwater outlets was not consistent between the different TMs (i.e. positive and negative  
311 correlations). This result could have been influenced by the drainage area and watershed land cover,  
312 which could vary considerably between the outlets.

313 As expected, *C. fluminea* contamination was also positively correlated with the total engine power of  
314 boats in harbors for Pb and Zn, elements generally measured in high abundance in harbors (Guevara-  
315 Riba et al., 2004). Clam As and Cd contaminations exhibited less significant correlations with the  
316 candidate variables. This result may be partly due to the fact that these elements are highly mobile  
317 between the different compartments (i.e. sediment, water and biota) (Frémion et al., 2017) and  
318 therefore possibly less directly related to point sources.

319

#### 320 4.3. *C. fluminea* as an indicator of contaminant bioavailability

321 The lack of significant correlation between the TM concentrations in *C. fluminea* and those in the  
322 sediment, as well as the differences in BCF between the different contaminants, suggest substantial  
323 variability in contaminant bioavailability. These results demonstrated that chemical analyses  
324 performed on solely abiotic matrices do not provide robust evidence of the bioavailability of the  
325 contaminants and consequently their effective ecological risk (Guo and Feng, 2018). The molecular  
326 characteristics of contaminants can induce significant differences in their uptake (e.g. passive or active  
327 transfers), storage and excretion by the organism, as illustrated in our study by the higher  
328 bioconcentration of light PAHs compared to heavy ones. Such differential bioconcentration was  
329 already highlighted by previous studies (Meador et al., 1995; Noh et al., 2018). An explanation of such  
330 variability is that environmental variables, such as slope and wave energy in this study, influence the

331 benthic organism exposure to littoral contamination. Indeed, these two variables could reflect the  
332 disturbance level at the sampling location leading to a transport and re-suspension of the sediment and  
333 so an increase in the exposure of benthic organisms to littoral contamination (Eggleton and Thomas,  
334 2004; Frémion et al., 2016; Phelps, 2016). In addition, Cu contamination in *C. fluminea* was  
335 negatively correlated with the abundance of organic matter in the sediment, which was in agreement  
336 with the fact that Cu is highly bound to the organic matter fraction of the sediment (Frémion, 2016;  
337 Tiquio et al., 2017), which may reduce its bioavailability. These environmental variables can therefore  
338 be considered in the ecological risk assessment.

339

340

## 341 **5. Conclusion**

342 *C. fluminea* appeared to appropriately reflect the spatial heterogeneity of lake littoral contamination  
343 (both in magnitude and profile) and the target contaminated “hot-spots” locally. The identification of  
344 stormwater runoffs as potentially important sources of TM contamination for the littoral biota  
345 provided insights into the assessment of the ecological risk of the pollution in the littoral zone of Lake  
346 Annecy and useful information for identifying and suggesting remedial actions (e.g. stormwater  
347 management). However, regarding persistent organic pollutants such as PAHs, linking organisms and  
348 potential watershed sources was not possible in this study. This result underlines possible changes in  
349 the contamination pattern from the source to the biota for persistent organic pollutants. This study  
350 supports the need to consider littoral contamination as complementary to that at a single site in the  
351 deepest zone (i.e. management monitoring) to fully appreciate the pressure of contamination on the  
352 lake. In view to continuing this work, it would be interesting to couple the variability of BCF between  
353 the contaminants with foodweb models to predict the preferential transfer of contaminants to  
354 patrimonial fishes such as whitefish and arctic char in Lake Annecy. In light of the bioconcentrated  
355 TMs and PAHs in *C. fluminea* in the current study, enzymatic biomarkers in these autochthonous  
356 organisms could be performed to obtain bioindicators of the detrimental effects caused by their  
357 contamination and thus go further in the ecological risk <sup>[11]</sup><sub>SEP</sub>assessment. In addition, performing risk  
358 assessment at higher trophic levels would be relevant since *C. fluminea* is an important source of

359 protein for dominant macrobenthic invertebrates in many temperate lakes, such as crayfish (Antón et  
360 al., 2000; Simon and Garnier-Laplace, 2005).

361

## 362 **Appendix**

363 In addition to the details of TMs and PAHs contents in clams and sediments in study sites, the  
364 Appendix file provides information about intra-site variability of *C. fluminea* contamination, the  
365 method used for estimating lipid contents in the organisms, sediment characterization, spatial  
366 distribution of potential in-lake and watershed sources of contamination, relationships between the  
367 results of linear regressions and different buffer sizes for the buffer sizes selection and hydrodynamics  
368 details on Lake Annecy shoreline.

369

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374

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384

385

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387 **Literature**

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