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

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Abstract

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

Keywords

Trace metal, polycyclic aromatic hydrocarbon, bioindicator, bioconcentration, *Corbicula fluminea,* littoral.

1. Introduction

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

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

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

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

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

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

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

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

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

2. Materials and Methods

2.1. The study lake

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

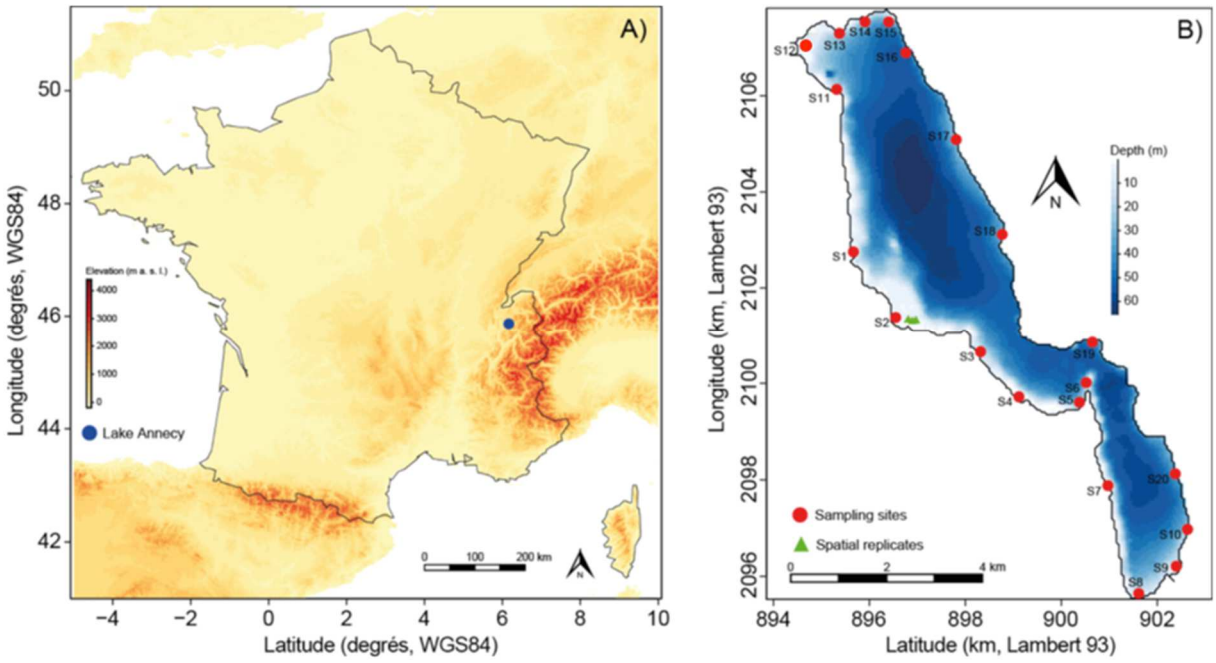


Figure 1. Map of Lake Annecy in the east of France (A) and the locations of the sampling sites (B). Red circles indicate main sampling sites and green triangles replicates for assessment of local contamination variability.

2.2. Sampling sites

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

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

2.3. Contamination quantification

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

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

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

2.4. Bioconcentration factor (BCF)

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

2.5. Explanatory variables of *C. fluminea* contamination

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

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

2.6. Data analysis

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

3. Results

3.1. Sediment and *C. fluminea* contaminations

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

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

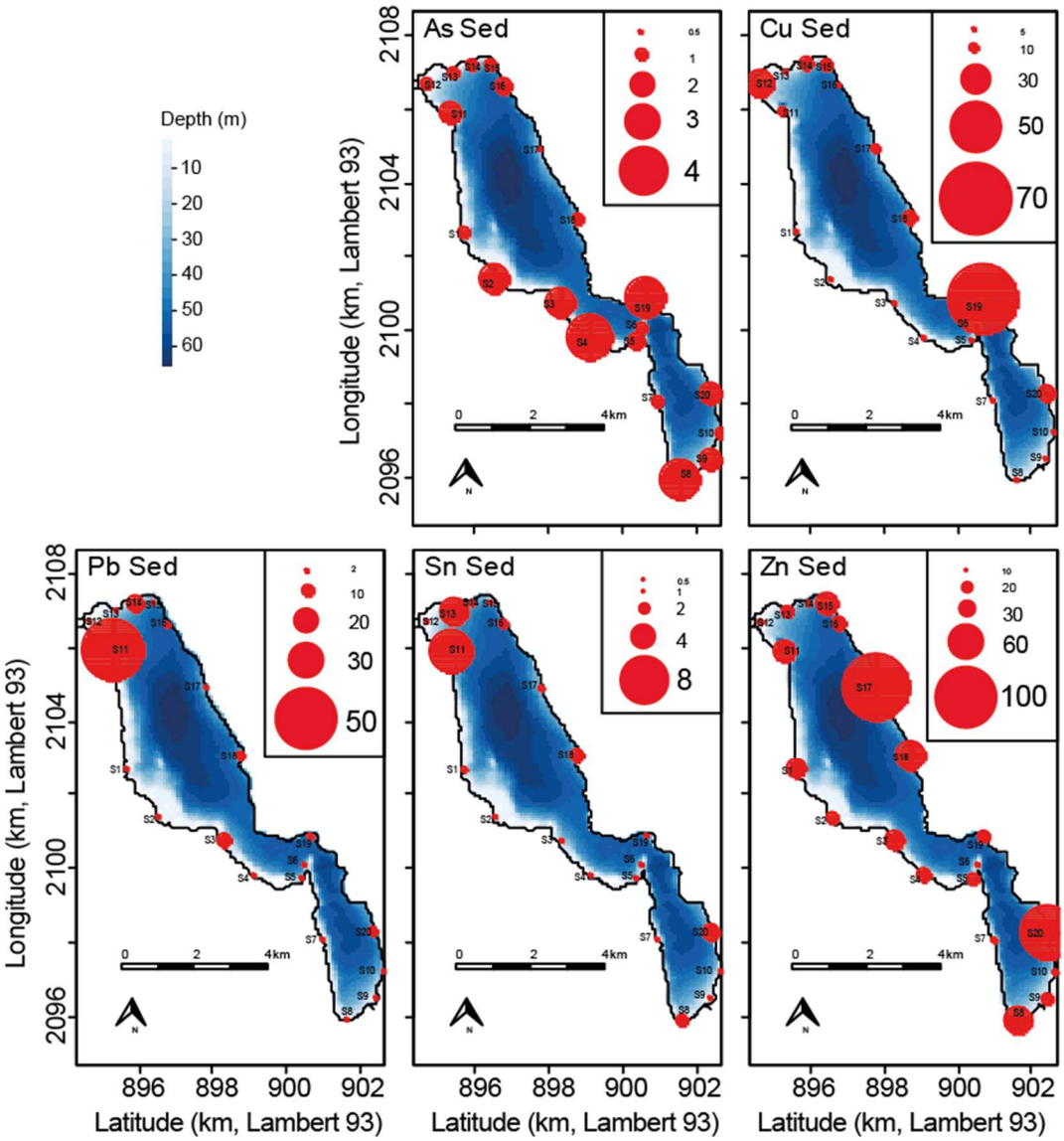


Figure 2. Mapping the TMs sediment contamination (mg kg^{-1} DM) along Lake Annecy littoral.

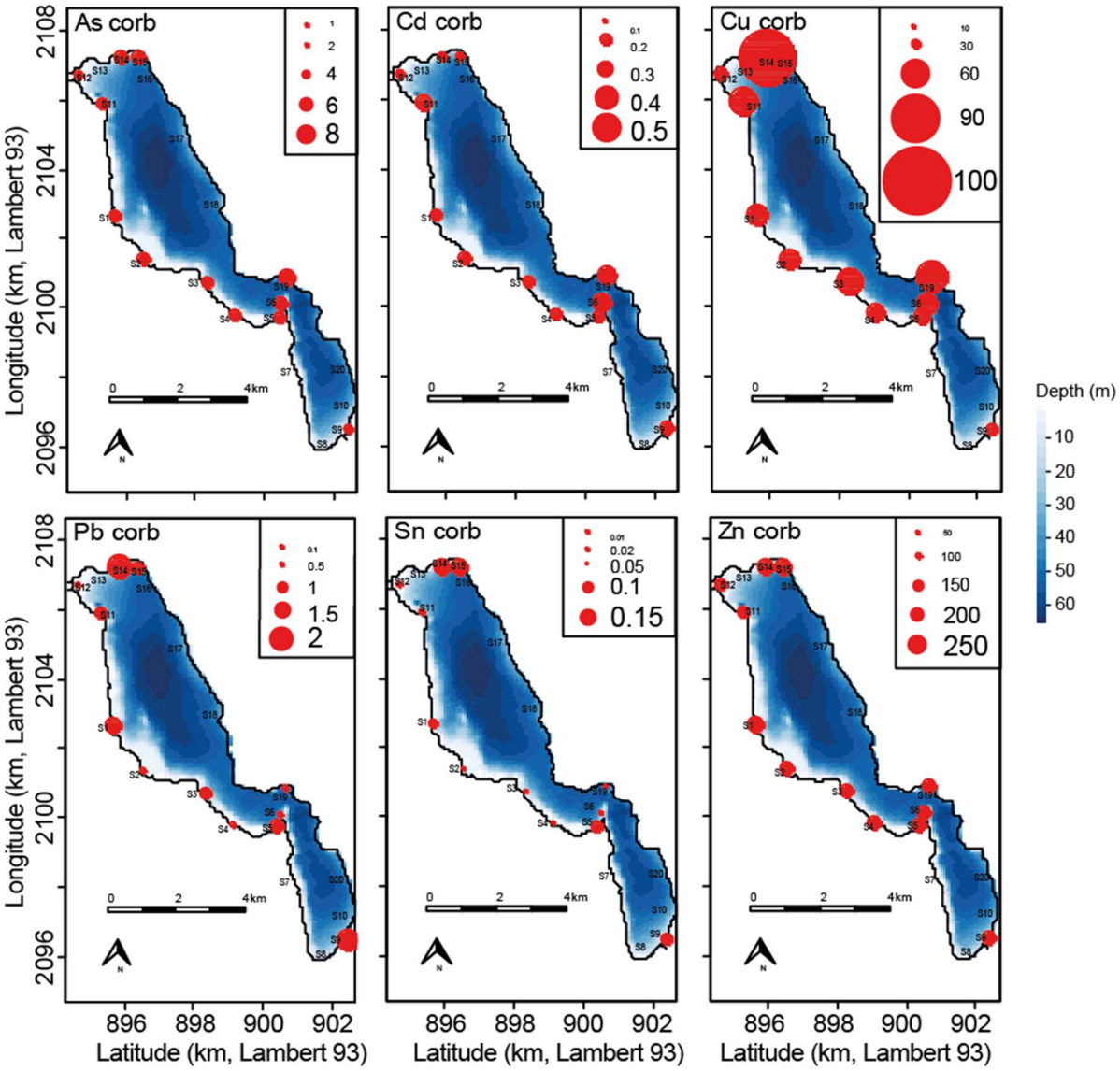


Figure 3. Mapping the TMs *C. fluminea* contamination (mg kg^{-1} DM) along Lake Annecy littoral.

The average sum of PAHs in the lake littoral was equal to $1.10^5 \pm 4.10^5$ µg kg^{-1} OM in the sediment and 165.6 ± 150.2 µg kg^{-1} lipids in *C. fluminea* and also varied considerably according to the location considered in the littoral in both matrices (i.e. sediments and *C. fluminea*; Figure 4).

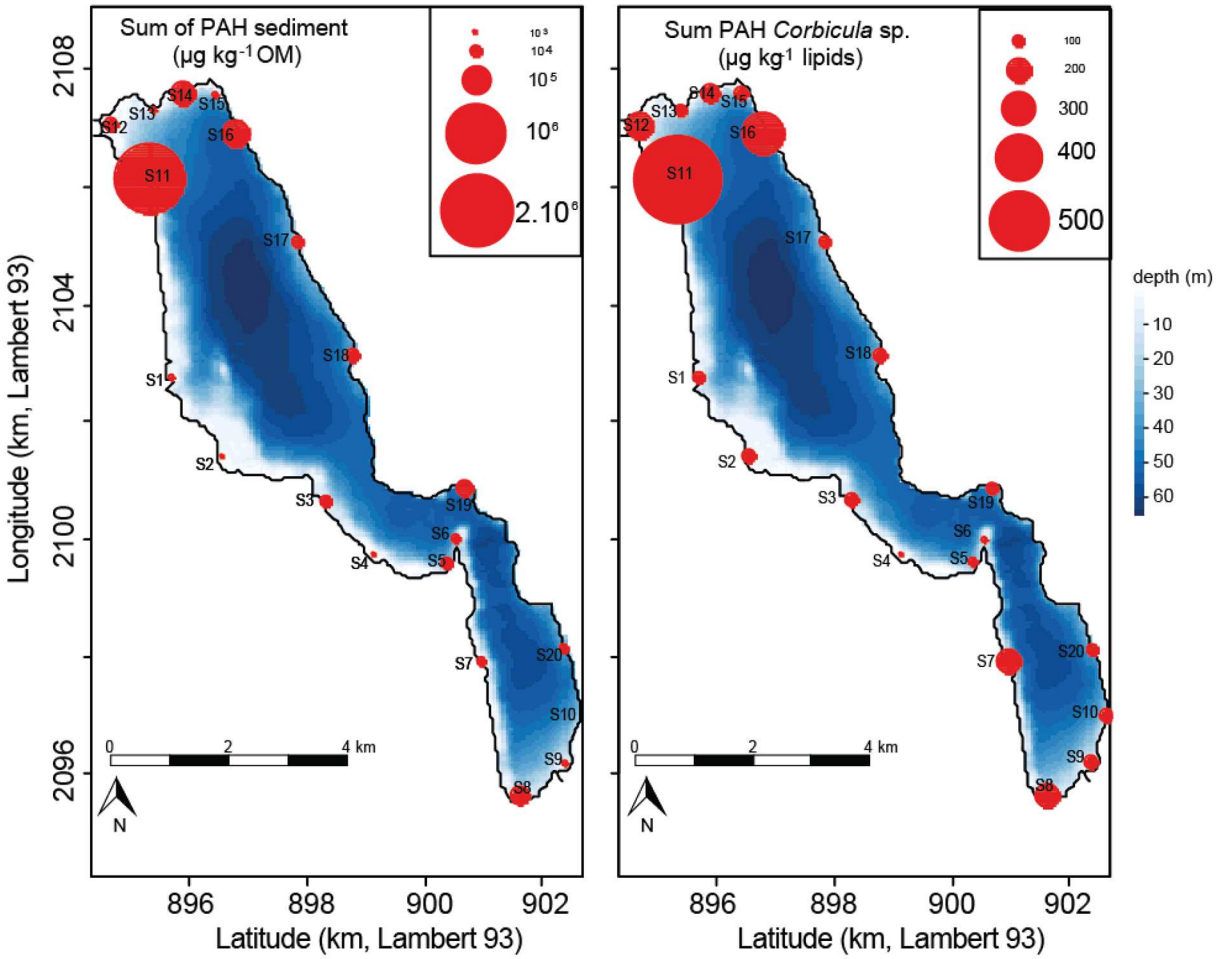


Figure 4. Map of the sum of PAHs in the sediment (µg kg^{-1} OM) on the left side and in *C. fluminea* (µg kg^{-1} lipids) on the right side.

S11 exhibited the highest contamination with concentrations of 1.7 10^6 µg kg^{-1} OM in the sediment and 734.1 µg kg^{-1} lipids in *C. fluminea.* In addition, *C. fluminea* exhibited lower proportions of heavy PAHs (23 ± 10 %) than the sediment (62 ± 10 %) (Figure 5).

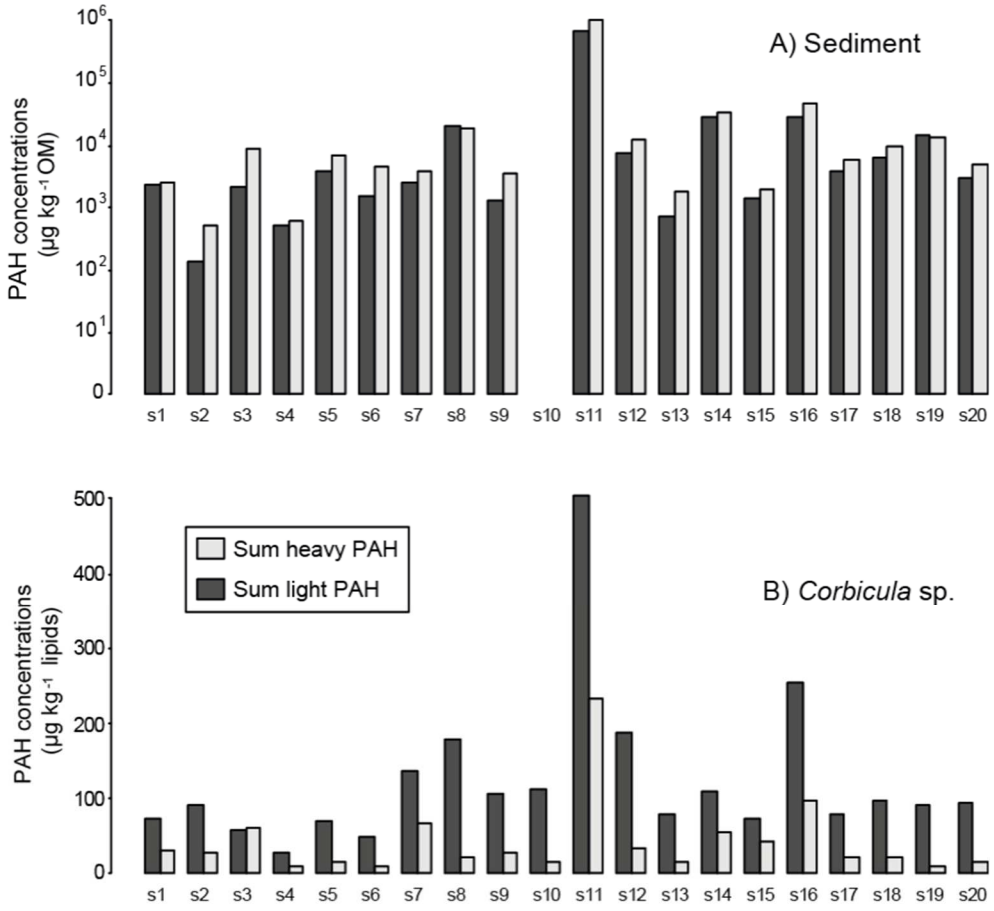


Figure 5. Barplots representing the composition in light and heavy PAHs in A) the sediments (µg kg^{-1} OM) and B) *C. fluminea* (µg kg^{-1} lipids).

3.2. Contamination transfer

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

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

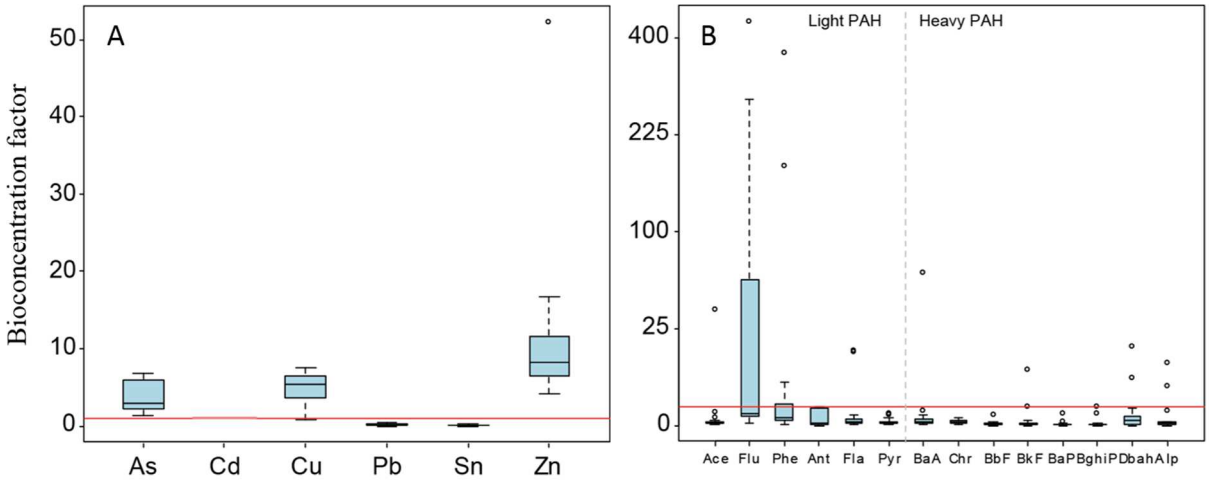


Figure 6. Boxplots of the bioconcentration factors (BCF) for A) TMs and B) PAHs. The horizontal line represents a BCF equal to 1.

3.3. Contamination sources

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

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

Contaminants	As		Cd		Cu		Pb		Sn		Zn		Sum of PAHs	
Radius	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^2	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

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

contamination. However, at a large scale (i.e. radius = 600 m), some significant negative relationships indicate that the influence of the explanatory variables may differ from one TM to another (Figure 7-B). For example, the number of stormwater outlets exerted a significant positive influence on the contamination for As and Cd but a negative one for Cu, Pb and Sn.

We also noted that many more variable were detected to have an effect on clams contamination in TMs at the larger scale (600 m radius, Figure 7-B) than at the local scale (200 m radius, Figure 7-A), except for the proportion of organic matter in sediment. R-square were also similar or higher for the 600 m radius confirming the better prediction power at this scale (Table 1).

Considering the sum of PAHs, the main explanatory variable was the sediment contamination while the slope of the site had a marginally negative influence on internal PAH concentrations in *C. fluminea* whatever the scale of the study (Figure 7). This result was similar for both radii.

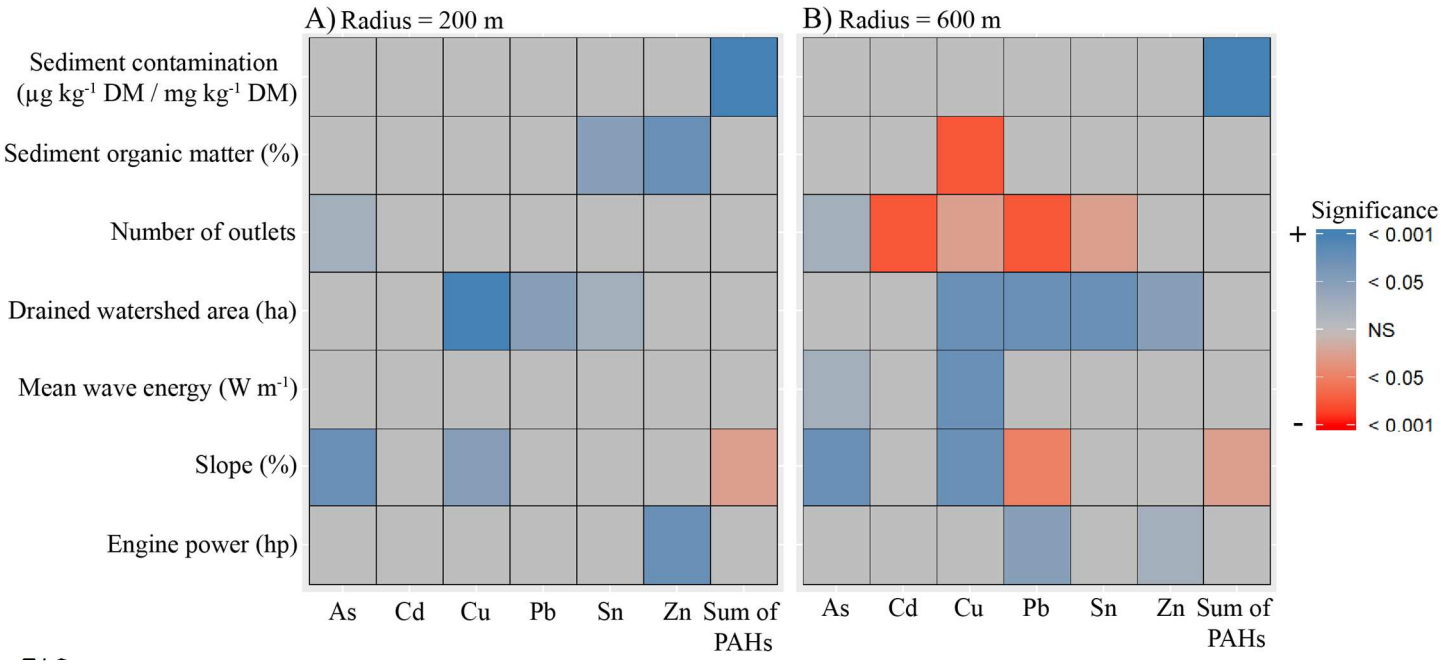


Figure 7. Heatmaps illustrating the results from the multiple linear regressions relating *C. fluminea* contamination to candidate variables according to A) radius = 200 m and B) radius = 600 m. Blue and red colors symbolize a positive (+) and a negative (-) relationship with *C. fluminea* contamination respectively. The significance rates were depicted by color intensity according to the legend intensity 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 ± 19.4 µg kg^{-1} DM)
284 compared to polluted sites such as Lake Kentucky (171.5 µg kg^{-1} 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

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

5. Conclusion

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