



Changes in mobility of trace metals at the sediment-water-biota interfaces following laboratory drying and reimmersion of a lacustrine sediment

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Abstract

Alterations in the timing, frequency, and magnitude of water level fluctuations (WLF) in lakes may result in important changes in abiotic parameters that can affect sediment-borne contaminant mobility at the sediment-water-biota interfaces in littoral zones. This study aims to assess the mobility of trace metals (TMs)—Cd, Cr, Cu, Ni, Pb, and Zn—under laboratory-simulated WLF (i.e., drying and reimmersion of sediments) through a three-pronged approach. One surficial sediment was sampled from the shoreline of a large French lake exhibiting an artificially limited WLF. A sample was enriched with a solution of TMs to ensure significant measurements of mobility. The spiked and naturally contaminated sediments were dried and reimmersed. The first approach consisted in measuring the mobility of TMs from the sediment to the water column under resuspensions of particles through leaching tests. The second approach assessed the partitioning of TMs between the different binding forms within the sediments through sequential extraction tests. The last approach tested the changes in TM bioconcentration in organisms exposed to sediment through microcosm assays. The hypothesis was that WLF may increase mobility from the sediment to the water column relative to mobility from the residual to easily mobilizable fractions within the sediments and consequently increase the bioconcentration of less inert trace metals, mostly Cd and Zn. This hypothesis was partly rejected as TM binding forms mainly increased toward the residual fractions within the sediment, especially for Cd and Zn, and bioconcentration mainly decreased following WLF. However, TM concentration increased in the water column when WLF included great resuspension of particles. The study also provides insights into the complex relationships among contaminant mobility to the water column, bioavailability, and bioconcentration, especially in the context of large abiotic disturbances such as WLF. These findings may be useful for further management strategies for WLF-regulated lakes and reservoirs.

Keywords Heavy metal · Laboratory microcosm · Sequential extractions · Leaching test · Lake littoral · Bioconcentration · Mobility · Bioavailability

Introduction

Alterations of the timing, frequency, and magnitude of water level regimes may result in important changes in the

ecological structure and functioning of littoral habitats (Adams 2002; Strayer and Findlay 2010; Evtimova and Donohue 2014, 2016). The impacts of water level regulations on the abiotic characteristics of littoral zones and the negative consequences on biological productivity and diversity have been widely studied (Leira and Cantonati 2008; Hirsch et al. 2017). Restoring natural water level fluctuations (WLF) in artificially regulated lakes is hence increasingly considered as a tool for ecological restoration (Coops and Hesper 2002; Coops et al. 2003; Geest et al. 2005). However, little attention has been paid to the influence of these changes on abiotic parameters, e.g., sediment-borne contaminants and their mobility at the sediment-water-biota interfaces (Frémion et al. 2016, 2017). WLF induce the drying and reflooding of shoreline surficial sediments, which may affect biogeochemical

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cycling in sediments through changes in temperature, pH, redox potential, particle resuspension, and microbial activity. These alterations can affect the lability, solubility, and speciation of contaminants, potentially resulting in toxicity of an originally non-toxic littoral site. The mobility of sediment-borne contaminants under sediment drying has been often studied in the context of management of dredged sediments (Guevara-Riba et al. 2004; Charrasse et al. 2018; Kim et al. 2018), but little is known about the mobility and bioavailability of contaminants from shoreline surface sediments that undergo WLF after immersion by standing waters for a long period (Skoulikidis et al. 2008).

Lake Bourget (Savoie, France) is a large deep lake for which WLF (i.e. the vertical difference between the summer and winter water level) were artificially restricted to 30 cm year⁻¹ in the early 1980s for hydroelectricity production (Miquet 1997). The restoration of larger WLF is currently being argued for ecological rehabilitation purpose. Among other contaminants, trace metals (TMs) have been measured in littoral surficial sediments at low to moderately toxic levels (Lécrivain et al. 2018).

The aim of this study is to assess the mobility and bioavailability of six TMs (Cd, Cr, Cu, Ni, Pb, and Zn) from a littoral surficial sediment of Lake Bourget through laboratory WLF simulations. The simulations consisted in drying and reimmersing these sediments and assessing the mobility of sediment TMs following three approaches. The first approach consisted in assessing the mobility of TMs between the sediment and the water column under reimmersion, including vigorous resuspension of particles simulated through leaching tests. The second approach was based on sequential extraction tests to appraise the potential changes in the binding forms of TMs within the sediment under reimmersion without mechanical resuspension of particles. The third approach used microcosm bioassays in which laboratory aquatic organisms were exposed to these sediments to determine the changes in TM bioconcentration after WLF. Half of the naturally contaminated sediment was spiked with TMs to ensure the detection of contaminant mobility via these three approaches since littoral sediments are coarse sediments and are likely to adsorb less metal on particle surfaces than fine offshore sediments (Nedrich and Burton 2017a; Lécrivain et al. 2018).

Past studies have shown a redistribution of most TMs from the residual to mobile fractions following drought of anoxic sediments (Stephens et al. 2001). Other studies have demonstrated that the speciation of metals depends on their concentrations in the sediment (Fan et al. 2002), on the organic content in the sediment (Lin et al. 2018; Yuan et al. 2018), or even on the carbonate and sulfide content (Nedrich and Burton 2017b). Others have only shown slight changes in oxid to suboxic sediments (Frémion et al. 2017).

Following these past results, our study hypotheses were that sediment drying and reflooding may increase TM

concentration in the water column following sediment resuspension (approach 1), may increase TM abundance from the residual to easily mobilizable fractions within the sediment (approach 2), and consequently increase the TM internal concentrations of the organisms (approach 3). These changes in TM mobility at the sediment-water-biota interfaces are expected to be greater for Cd and Zn, since they are considered as less inert metals than Cr, Cu, Ni, and Pb (Wu et al. 2011; Tiquio et al. 2017).

This study is expected to increase knowledge of the behavior of sediment-borne contaminants under controlled WLF and provide broader perspectives for management tools related to WLF in lake ecological restoration.

Materials and methods

Sampling sites and method

Surface (0–5 cm) sediment was sampled in the southeastern littoral of Lake Bourget (45° 40' 3.3" N, 5° 53' 36.3" E) at a water depth of 1 m using an Ekman grab in an area with a gentle slope. The sediment was then homogenized in a plastic tray and placed in pre-washed 2-L polyethylene bottles. The sample was transported to the laboratory in coolers; it was then wet sieved through 2-mm mesh to remove debris and macrofauna and stored at 4 °C.

Sampled sediment characterization

The grain size distribution, loss on ignition, and total content of trace metals of the sediment were assessed according to the method described in Lécrivain et al. (2018). Briefly, three sub-samples of the sieved sediment were dried at 105 °C for 24 h. One sub-sample was used for grain size measurements by laser diffractometry (Malvern Mastersizer 2000G). The second sub-sample was used for water content determination and further ignited at 550 °C for 4 h to determine the loss on ignition (LOI). The percentage of organic carbon (OC) was calculated from the LOI using the relationship $OC (\%) = LOI (\%) / 2.13$ (Dean Jr 1974). The last sediment sample was dried and digested in a microwave oven (CEM/MARS Xpress, 2 mL of ultra-pure HNO₃ + 6 mL of ultra-pure HCl, 180 °C for 30 min, filtration on a 0.45-µm Whatman™ filter, and dilution of the residue in 25 mL of ultra-pure water). The resulting sample was analyzed by atomic absorption spectrometry (AAS). The total sediment contents of Cd, Cr, Cu, Ni, and Pb were measured in a graphite furnace at 1500 °C, and the total sediment content of Zn was measured with an air-acetylene flame (PerkinElmer, model PinAAcle 900T, detection limits: Cd = 0.1 µg L⁻¹, Cr = 0.040 µg L⁻¹, Cu = 0.014 µg L⁻¹, Ni = 0.07 µg L⁻¹, Pb = 0.05 µg L⁻¹, Zn = 0.02 µg L⁻¹). Certified reference material (River sediment

LGC6 187) was used for standardization, and triplicate measurements were done to ensure that analytical errors were under 2%.

Sediment spiking

Half of the sediment was spiked with Cd (added as Cd(NO₃)₂, 4H₂O; Merck), Cr (added as CrO₃, Merck), Cu (added as CuCl₂, 2H₂O, Sigma-Aldrich), Ni (added as NiSO₄, 7H₂O, Acros Organics), and Zn (added as Zn(CH₃COO)₂, Prolabo) to achieve the following respective final concentrations: 2, 75, 90, 30, and 200 mg kg⁻¹ DM. These nominal concentrations were chosen as the means of the respective threshold effect concentration (TEC) and probable effect concentration (PEC) (MacDonald et al. 2000) to increase the probability of observing biological effects of sediment contamination on the exposed organisms. The final sediment concentrations of Cd, Cr, Cu, Ni, and Zn were 1.30 ± 0.22, 62.39 ± 14.53, 60.20 ± 11.19, 13.18 ± 2.47, and 150 ± 30 mg kg⁻¹, respectively. Throughout this study, the non-spiked sediment will be called sediment A, and the spiked one will be called sediment B.

WLF simulation

Sub-samples of sediments A and B were dried in an oven at 40 °C for 6 days to ensure slow, total drying. The other sub-samples remained wet and were stored at 4 °C for use as controls in the simulation.

Approach 1

Leaching tests were performed on the dried and control (always wet) sediments according to the European standard (EN 12457-2 2002) to assess the leachate concentrations of the trace metals that could be transferred from the sediment to the water column after reflooding with mechanic particle resuspension. The leachate dissolved TM concentrations were measured using AAS as previously described.

Approach 2

Another sub-sample of the dried sediments was slowly rewetted until water saturation with a synthetic medium (pH 7.7, hardness 60 mg CaCO₃ L⁻¹, alkalinity 120 mg CaCO₃ L⁻¹, conductivity 290 μS cm⁻¹, phosphorous 100 μg L⁻¹, nitrogen 1308 μg L⁻¹, FeEDTANa₂, 6H₂O 109.4 μg L⁻¹, oligo-elements and vitamins of M4 medium (Elendt and Bias 1990)). Then, 250 g of fresh sediment with a width of approximately 2 cm was introduced in acid-rinsed beakers (four replicates for each sediment (spiked and non-spiked)) and submerged in 2 L of the same synthetic medium.

The beakers were aerated and left to stand for 7 days for stabilization and maturation in the dark at 20 °C.

Sequential extraction tests were performed according to the method of Pueyo et al. (2001). Tests were performed on wet (sequential extraction test “SE1”), dried (sequential extraction test “SE2”), and rewetted (sequential extraction test “SE3”) sediments to assess the mobility of the trace metals along the WLF simulation between the four fractions: (F1) exchangeable metal, (F2) Fe-Mn oxides and hydroxide-bound metal, (F3) organic matter and sulfide-associated metal, and (F4) residual metal. For each sediment, the extractions were performed in triplicate, and a cleaning step with ultra-pure water was introduced between each extraction. The supernatants were separated by centrifugation (20 min at 8000×g), filtered with a 0.2-μm nylon mesh, and acidified with concentrated nitric acid. The TM concentrations in the sediment fractions were measured by AAS as previously described.

Approach 3

Microcosm assays were performed using the previously cited beakers containing sediments that always remained wet throughout the whole study as controls.

The laboratory organisms used for this experiment originated from a laboratory room exempt from possible contamination at constant temperature (20 ± 2 °C) and under a light regime of 16 h of light and 8 h of darkness. *Chironomus riparius* midges were bred in plastic aquaria (34 × 18 × 19 cm) containing a layer of artificial sediment comprising 2 kg of dry Fontainebleau sand (particle size 150–300 μm) (VWR Prolabo) mixed with 600 mg of finely ground fish food flakes (TetraMin®) submerged in 6 L of clean and aerated groundwater. Approximately 75% of the water column was renewed weekly, and the midges were fed finely crushed TetraMin®. Forty *C. riparius* first-instar larvae were introduced into the microcosms. The larvae were homogeneous in individual size (i.e., coefficient of variation of less than 10%). During exposure, the larvae were fed 0.33 mL per individual per day of a finely crushed suspension of 10 g L⁻¹ TetraMin®. Half of the water column was gently renewed every other day to prevent abnormally high contaminant concentrations, and the water column quality was monitored to prevent bias in the fitness response of the organisms. The dissolved oxygen concentration (mg L⁻¹), temperature (°C), conductivity (μS cm⁻¹), pH, and Na⁺, K⁺, Ca²⁺, NH₄⁺, Mg²⁺, Cl⁻, NO₂⁻, NO₃⁻, PO₄³⁻, and SO₄²⁻ dissolved concentrations (mg L⁻¹) were measured in the beakers every other day following the methods described by Perrodin et al. (2016).

The larvae were exposed for 10 days to prevent emergence. At the final time (tf) of exposure, the larvae were collected, counted, and placed for 4 h in beakers filled with fresh synthetic media to exclude gut contents from the internal bioconcentrated TME concentrations.

The survival rate was expressed as the percentage of living individuals at the final time of the assays over the initial number of introduced individuals. The individual growth size (day^{-1}) was calculated according to Eq. (1) (AFNOR 2006), where l_f and l_0 are the final and initial individual body lengths (mm), respectively, and t is the duration of the assays (days).

$$gr = \frac{\ln(l_f) - \ln(l_0)}{t} \quad (1)$$

The cephalic capsules were also measured to check that all larvae reached the fourth instar (Bour et al. 2015).

Internal TME concentrations, which corresponded to the bioconcentrated fractions of TMs, were measured on dried and pooled individuals from the same beaker (CEM/MARS Xpress, 3 mL of ultra-pure HNO_3 + 1 mL of ultra-pure HCl + 1 mL of H_2O_2 , 180 °C, 30 min, dilution of the residue in 10 mL of ultra-pure water). The samples were then analyzed by AAS using the same method as for the sediment samples.

Data analysis

The sediments that remained wet during the whole study were used as a control for these tests, called “control,” and the tested sediments that had undergone WLF simulations following the three approaches were called “WLF sediments.”

For sequential extraction tests, the sum of the first three fractions (F1 + F2 + F3) is often considered the most mobilizable fraction (Kartal et al. 2006; Horváth et al. 2013; Liu et al. 2016). This fraction will be called the “easily mobilizable fraction” in the study.

The influence of WLF simulations on TM mobility in the three approaches was statistically assessed using type III sum of squares analysis of variance (ANOVA) (on TM concentrations (in leachate for approach 1, in sediment fractions in approach 2, and in organisms in approach 3)), with treatment (control and WLF) and sediments (A and B) as fixed factors. For all variables, the normality and the homoscedasticity of the residues were tested using the Shapiro-Wilk’s test and the Levene’s test, respectively.

Following the ANOVA results in approach 2, the repartition within the fractions of each TM was also statistically compared between the sequential extraction tests SE1–SE2 and SE1–SE3 using the function `prop.test` from the *pgirmess* package (Giraudoux and Giraudoux 2018) for which null hypothesis expected similar proportions among sequential extraction results (Newcombe 1998).

For all approaches, the ratios of the TM concentrations (in leachate for approach 1, in the easily mobilizable fraction in approach 2, and in organisms in approach 3) measured in the WLF sediments to those in the controls were calculated to illustrate the evolution (increase or decrease) of the mobility of the TMs following WLF simulations and compare the

evolution among the three approaches; these ratios were called $\text{Ratio}_{\text{leaching}}$, $\text{Ratio}_{\text{extraction}}$, and $\text{Ratio}_{\text{organism}}$, respectively.

All statistical analyses as well as graphical displays were performed using R (Team 2017). Throughout the manuscript, values following “±” correspond to the standard deviation of the data.

Results

Main sediment characterization

The sampled sediment was mainly sandy ($94.5 \pm 1.0\%$) and poorly organic ($1.8 \pm 0.2\%$) (Table 1), which is characteristic of a surficial sediment exposed to hydrodynamic regimes in the littoral zone (Lécrivain et al. 2018).

Sediment A was always under the TEC, whereas sediment B was always above, except for Ni and Pb. Both sediments were under the PEC.

Approach 1

An increase in Cu, Ni, and Pb leachate concentrations was observed for WLF sediments compared to the control and was significant for Ni in sediment A and Cu in sediment B (Fig. 1).

The TM concentrations in the leachates from both the control and WLF sediments were significantly higher in sediment B than in sediment A, even when the total sediment concentrations were similar (e.g., Ni), except for Pb. This may be due to the higher TM mobility linked to the spiking of sediment B, which did not undergo the same aging process of TM contamination as sediment A.

Table 1 Grain size distribution (%), organic carbon content (%), and mean TM contents (\pm standard deviation (SD)), and TEC and PEC in mg kg^{-1} DM (dry mass) of the two sediments ($n = 3$)

	Sediment A	Sediment B	TEC*	PEC*
Sand (%)	94.5 ± 1.0		–	–
Silt (%)	5.3 ± 1.0		–	–
Clay (%)	0.2 ± 0.0		–	–
D50 (µm)	551.9 ± 11.4			
OC (%)	1.8 ± 0.2		–	–
Cd (mg kg^{-1})	0.09 ± 0.01	1.30 ± 0.22	0.99	4.98
Cr (mg kg^{-1})	25.35 ± 4.08	62.39 ± 14.53	43.4	111.00
Cu (mg kg^{-1})	6.33 ± 0.17	60.20 ± 11.19	31.6	149.00
Ni (mg kg^{-1})	14.23 ± 7.24	13.18 ± 2.47	22.7	49.00
Pb (mg kg^{-1})	5.81 ± 0.85	5.64 ± 0.67	35.8	128.00
Zn (mg kg^{-1})	46.86 ± 0.24	149.12 ± 25.60	121.00	459.00

*From MacDonald et al. (2000)

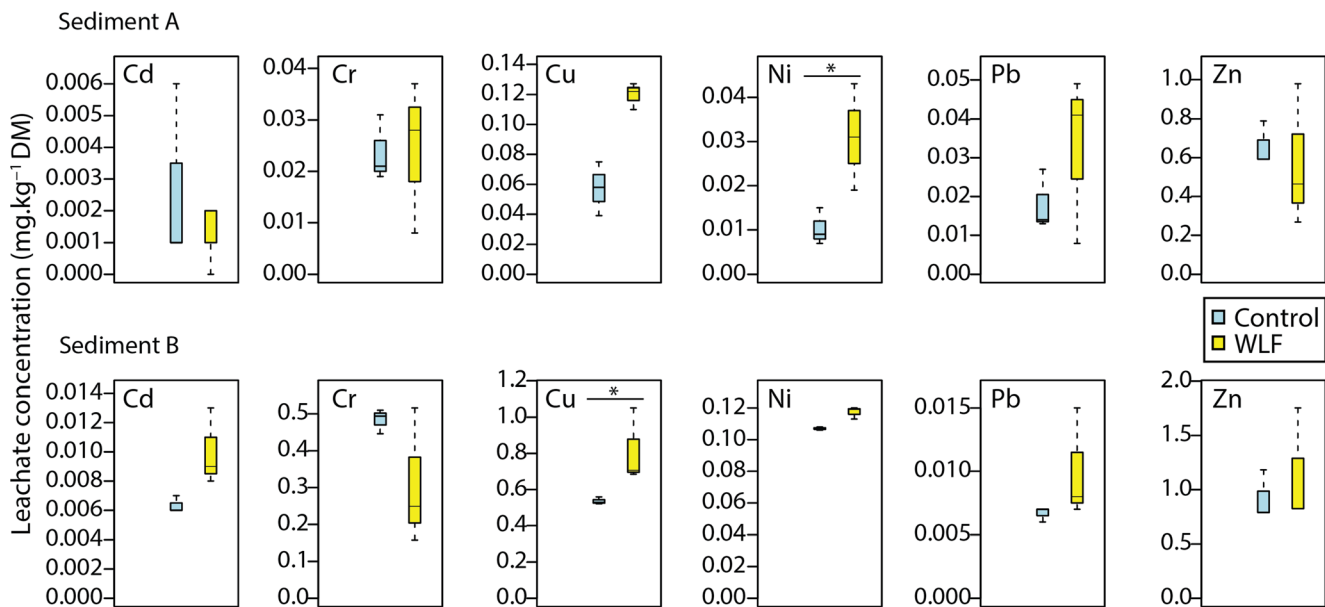


Fig. 1 Leaching test results for the control sediments (blue) and WLF sediments (yellow) for the natural (a) and spiked (b) sediments ($n = 3$). The stars indicate significant differences between the control and WLF sediments

Approach 2

Type III sum of squares ANOVA revealed significant differences in the distributions between fractions according to both the sequential extraction test (SE1, SE2, and SE3) and the TM. Following these results, the differences between the results of the sequential extraction tests regarding the distributions between the fractions were statistically analyzed. The WLF simulation was assessed by comparing the results of the sequential extraction tests SE1 and SE3 and was characterized by an upward trend of the residual fraction (F4) in both sediments (Fig. 2), which was more or less marked according

to the TM and significant for Zn in both sediments and for Cr in sediment B (Table 2).

More precisely, drying (comparison between the sequential extraction tests SE1 and SE2) was also characterized by an upward trend of F4 in both sediments and was significant for Zn in sediment A (Table 2).

Exploring the fraction analysis further, Cd and Zn exhibited the greatest abundance in F2, Cu and Pb were highly bound to organic matter as they were abundant in F3, and Cr and Ni exhibited the greatest abundance in F4.

Overall, the WLF simulation was characterized by a reduction of the easily mobilizable fraction (F1 + F2 + F3).

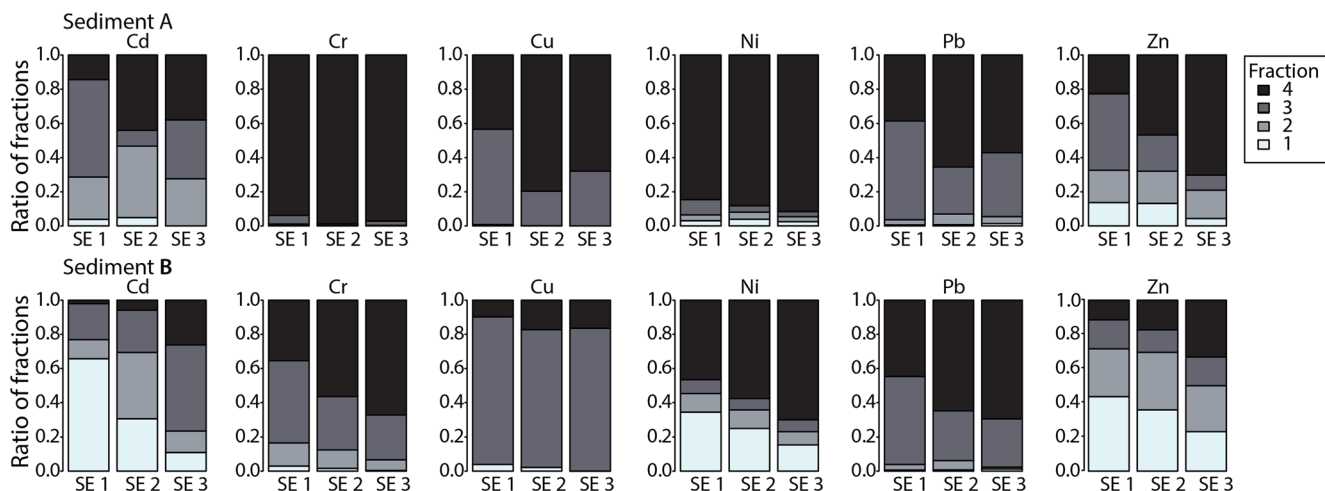


Fig. 2 Fractionation of Cd, Cr, Cu, Ni, Pb, and Zn according to the BCR protocol. The results are expressed as the mean percentage of the contribution of each fraction (concentrations in mg kg^{-1} DM converted

in ratios over the total concentration, $n = 3$). SE1, SE2, and SE3 correspond to extraction tests performed on the control (wet), dried, and WLF sediments, respectively

Table 2 Prop.test results to assess significant differences between the sequential extraction tests (SE1, SE2, and SE3) in the distribution of each TM between the four fractions

Element		Cd	Cr	Cu	Ni	Pb	Zn
Sediment A	Chi SE1–SE2	0.13	1.24	1.95	0.27	1.42	12.41
	<i>p</i> value SE1–SE2	0.99	0.74	0.58	0.97	0.70	0.01
	Chi SE1–SE3	0.06	0.37	0.92	0.35	0.53	31.44
	<i>p</i> value SE1–SE3	1.00	0.95	0.82	0.95	0.91	0.00
Sediment B	Chi SE1–SE2	0.41	5.10	1.64	0.38	0.63	4.50
	<i>p</i> value SE1–SE2	0.94	0.16	0.65	0.95	0.89	0.21
	Chi SE1–SE3	0.53	12.38	2.18	1.78	0.86	23.33
	<i>p</i> value SE1–SE3	0.91	0.01	0.54	0.62	0.84	0.00

Approach 3

There was no significant difference between the WLF and control sediments or between sediments A and B in terms of the survival (mean = 78 ± 9%) and growth rates (mean = 0.30 ± 0.005 day⁻¹) of *C. riparius*.

A downward trend was observed in internal concentrations between the organisms exposed to the control and WLF sediments, with a significant decrease in Pb in sediment A (Fig. 3). By contrast, Cd concentrations significantly increased in organisms exposed to WLF sediment B.

Comparison of TM mobility among the three approaches

Ratio_{leaching} was greater than 1 (Table 3) except for Cd in sediment A and Cr in sediment B, suggesting that hydrodynamic flows related to WLF may increase TM mobility from the sediment to the water column.

By contrast, ratio_{extraction} was less than 1, suggesting a decrease in the easily mobilizable fraction in the sediment following WLF simulation.

Ratio_{organism} was less than or close to 1, except for Cd for both sediments. These results are in agreement with ratio_{extraction} as the internal TM concentrations decreased with a decrease in the more mobile fractions in the sediment following the WLF simulation. The variations in the Ratio_{organism} values were less important compared to ratio_{extraction} and ratio_{leaching}.

Discussion

WLF in lake littoral zones are a complex phenomenon that can induce the mobility of sediment-borne pollutants following drastic changes in abiotic parameters. In this study, the mobility of TMs following WLF simulations was observed within the sediment fractions, between the sediment and the water column, and between the sediment and the biota. However, the mobility pattern varied greatly according to the TM.

The sequential extraction tests showed that Cd and Zn were most present in the easily mobilizable fraction (F1 + F2 + F3). In particular, F1 and F2 had greater abundances of these elements compared to the other TMs in both the controls and WLF sediments. By contrast, Cr, Cu, Ni, and Pb are more inert TMs (Calmano et al. 1993; Wu et al. 2011; Liu et al. 2016; Tiquio et al. 2017; Frémion et al. 2017), consistent with their higher abundance in the residual fraction for both the control and WLF sediments in the sequential extraction tests. F3 was the fraction with the second greatest abundance of these elements, which are known to naturally bind to organic matter and sulfides, especially Cu (Frémion 2016; Tiquio et al. 2017).

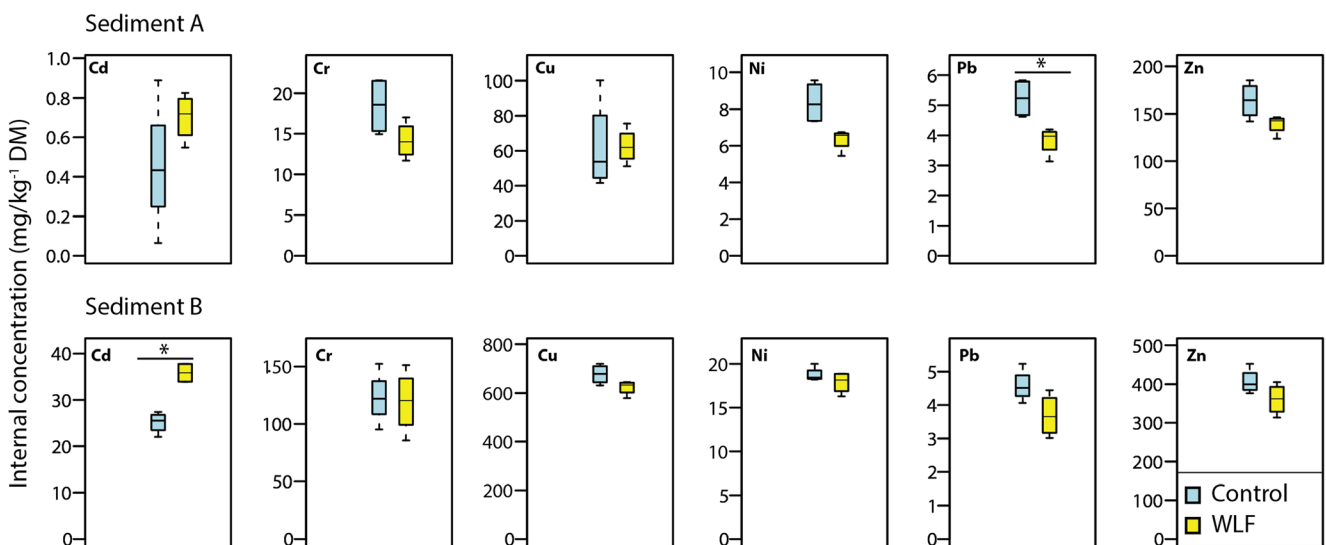


Fig. 3 Internal concentrations (mg kg⁻¹ DM) in *C. riparius* (n = 4) exposed to the control (blue) and WLF sediments (yellow) from sediments A and B

Table 3 Ratio_{extraction} ($n = 3$), Ratio_{leaching} ($n = 3$), and Ratio_{organism} ($n = 4$) for each TM and sediment. (+) and (–) symbolize an increase or decrease in the concentration, respectively, between the control and WLF sediments

	Element	Cd	Cr	Cu	Ni	Pb	Zn
Sediment A	Ratio _{extraction}	0.10 (–)	0.66 (–)	0.31 (–)	0.64 (–)	0.40 (–)	0.19 (–)
	Ratio _{leaching}	0.50 (–)	1.03 (+)	2.09 (+)	3.00 (+)	1.81 (+)	0.87 (–)
	Ratio _{organism}	1.55 (+)	0.77 (–)	1.01 (+)	0.76 (–)	0.73 (–)	0.85 (–)
Sediment B	Ratio _{extraction}	0.16 (–)	0.60 (–)	0.50 (–)	0.58 (–)	0.38 (–)	0.46 (–)
	Ratio _{leaching}	1.58 (+)	0.64 (–)	1.52 (+)	1.10 (+)	1.50 (+)	1.23 (+)
	Ratio _{organism}	1.43 (+)	0.97 (–)	0.92 (–)	0.95 (–)	0.81 (–)	0.89 (–)

A general decrease in the easily mobilizable fraction (F1 + F2 + F3) was observed within the sediments for all TMs following WLF (ratio_{extraction} < 1). However, the highest decreases were observed for Cd and Zn, which may be due to the fact that these two TMs are naturally reactive elements (Liu et al. 2016; Frémion et al. 2017). This general decrease in the easily mobilizable fraction was consistent with a general decrease in the internal concentrations observed in the exposed *C. riparius* through the bioassays (ratio_{organism} < 1) along with a decrease in the Cd and Zn concentrations in the leaching tests following WLF observed in the naturally contaminated sediment (sediment A, ratio_{leaching} < 1). However, the increase in the leachates observed for Cd and Zn in the spiked sediment (sediment B, ratio_{leaching} > 1) underlines the high reactivity of these elements especially when it comes to spiked sediments, for which the aging time (i.e., the time during which the speciation and availability of metals change within the sediment (Ghoveisi Hossein et al. 2018)) may not be comparable with that of naturally contaminated sediments (Ardestani and van Gestel 2016; Costello et al. 2016). Overall, the tested hypothesis whereby the TMs would move toward the most mobilizable fractions within the sediment and consequently induce an increase in their bioconcentration by benthic organisms following FLF was rejected in this study.

However, strong increases in the dissolved concentration of the more inert TMs (i.e., Cr, Cu, Ni, and Pb) in the leachate following WLF were observed in the leaching tests. By resuspending particles into oxygen-rich water, the leaching tests may have increased the oxidation of the organic matter and sulfides that the sediment drying had probably initiated (Simpson et al. 2000; Wu et al. 2011; Nedrich and Burton 2017a). This reaction may have led to the release of dissolved Cr, Cu, Ni, and Pb in the water column. Thus, drying and reimmersing lacustrine littoral sediment under high hydrodynamic flow may induce a transport of TMs from the sediment to the water column and exert a risk for the exposed organisms. This result highlights the importance of including hydrodynamic considerations while predicting contaminant mobility under water level fluctuations.

In addition, opposite results (increase vs decrease) were obtained for some TMs in terms of ratio_{extraction}, ratio_{leaching}, and ratio_{organism}, further indicating the complex relationships among contaminant mobility, bioavailability, and

bioconcentration, especially in the context of specific abiotic disturbances such as WLF. As an example, the differences obtained among the TMs in ratio_{extraction} and in ratio_{leaching} were higher than the differences in ratio_{organism}, suggesting that *C. riparius* larvae might regulate TM bioconcentration by reducing assimilation, enhancing excretion, or both (Adams et al. 2000; McGeer et al. 2003).

Conclusion

The study hypotheses were that sediment drying and reflooding could (i) increase TM concentration in the water column following sediment resuspension (approach 1), (ii) increase TM abundance from the residual to the easily mobilizable fractions within the sediment (approach 2), and (iii) consequently increase the bioconcentration of the TMs in the organisms (approach 3). Laboratory experiments partially confirmed these hypotheses since WLF including sediment resuspension under their reflooding induced an increase in TM dissolved concentrations in the water column. In this case, the oxidation of the organic matter and sulfides may have played a role in the release of the TMs. However, both sequential extraction tests and bioassays concluded that TM mobility within the sediment and bioconcentration decreased following WLF without sediment resuspension. These results underline the importance of including hydrodynamic regime when assessing the impacts of WLF on contaminant mobility at the sediment-water-biota interfaces. Moreover, the results provided insights on the complex relationships among contaminant mobility, bioavailability, and bioconcentration, especially in the context of large abiotic disturbances such as WLF, illustrated by the different trends obtained through this three-pronged approach.

These findings may provide useful preliminary conclusions for further management strategies of dam reservoirs that undergo large WLF with sediment resuspensions as well as standing waters for which WLF restorations are planned for ecological considerations.

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