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Spatial distribution and solubilization characteristics of metal(loid)s in riparian soils within reservoirs along the middle Jinsha River

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Abstract

Purpose The construction of large dams submerges riparian soils within reservoirs. However, little is known about the influence of water submergence on metal(loid) solubilization from polluted soils. In this study, the spatial distribution and solubilization characteristics of soil-associated metal(loid)s within reservoirs along the middle Jinsha River were evaluated.

Methods Concentrations of soil-associated metal(loid)s were determined and principal component analysis was performed to evaluate the metal(loid) spatial distribution. Then, metal(loid) chemical fractions of highly contaminated soils were analyzed. Finally, ex situ experiments of metal(loid) solubilization were conducted under the influence of changing pH, suspended solid concentration (SSC), and oxidation-reduction potential (ORP) conditions as well as successive drought-wetting cycles.

Results Within the LY, AH, and LKK reservoirs, the soil-associated Ni and Cr originated from specific metal(loid) ores, and Cd and Cu remained from historical industrial pollution posed potential risks to water quality. Water submergence changed pH, SSC, and ORP conditions, which affected metal(loid) solubilization through dissolution-precipitation and sorption-desorption processes. The low SSC variables contributed to over 20% solubilization of total Cd, Cu, and As. Moreover, the neutral-alkaline conditions due to carbonate-dominated lithology limited metal(loid) release from soils. Finally, the decreased ORP promoted solubilization of metal(loid)s bound to Fe-Mn oxyhydroxides. Generally, the solubilization levels of metal(loid)s were dependent on the chemical fractions.

Conclusion Water submergence could induce potential solubilization of metal(loid)s, especially for Cd from historical industrial pollution. Thus, this study highlighted that the investigation on both spatial distribution and solubilization characteristics of metal(loid)s in riparian soils is important for future environmental management of reservoirs.

Keywords Metals and metalloids · Reservoirs · Riparian soils · Spatial distribution · Solubilization characteristics · Water submergence

1 Introduction

Because of rapid urbanization and industrialization, contamination of metals and metalloids has created pervasive environmental problems worldwide (Tshalakatumbay et al. 2015). Metal(loid) pollutants from anthropogenic sources are transported and enriched in soils or sediments via various pathways, such as chemical production, mining and smelting, municipal waste, and agricultural runoff (Hu et al. 2014; Li et al. 2014; Zhao et al. 2015; Yu et al. 2016; Yuan et al. 2019, 2021). Through both terrestrial and aquatic food chains, metal(loid)s can be absorbed by surrounding organisms and transferred to higher trophic levels, which causes potential harmful impacts on ecological and human health (Tshalakatumbay et al. 2015; Lindsay et al. 2017; Li et al. 2018a). It has been reported that chronic exposures to copper (Cu), cadmium (Cd), and mercury (Hg) could induce Wilson, Itai-itai, and Minamata diseases, respectively (Han et al. 2017; Ye et al. 2019). As a result, an improved understanding on the environmental cycling of metal(loid) contaminants is essential for food security, public health, and ecological sustainability.

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In aquatic environments, over 90% of metal(loid)s are adsorbed and fixed onto the particles and eventually deposited in sediments or soils (Bing et al. 2019). However, various natural processes, such as rainfall, flood, snow-melting, and wind disturbance, as well as anthropogenic activities, including hydraulic dredging, water transfer project, shipping, and trawling, could largely change hydrodynamic conditions and influence physicochemical environments (Du Laing et al. 2009; Yuan et al. 2019). Subsequently, the metal(loid) partitioning is significantly altered, which could be greatly affected by solid-liquid physicochemical characteristics, external hydrodynamic conditions, and metal(loid) intrinsic properties (Eggleton and Thomas 2004; Frémion et al. 2016). Therefore, the complex fate of metal(loid)s in disturbed water environments needs to be comprehensively investigated.

Dams, particularly large dams with over 15-m height, have been considered as the strongest hydrodynamic disturbance in many large rivers, which modify the natural regimes of riverine geomorphology and flow velocity, and disrupt the spontaneous transports of nutrients, soils, sediments, and microbes (Ellis and Jones 2013; Chen et al. 2019, 2020). Until 2020, more than 58,700 large dams have been constructed worldwide for various purposes, such as flood control, hydropower production, agricultural irrigation, and water supply (Winemiller et al. 2016; CIGB/ICOLD 2020). After the construction and operation of large dams, the raised water table in reservoirs significantly expands the submerged region of riparian soils (Shu et al. 2017; Song et al. 2017). It has been demonstrated that metal(loid) contaminants could be released from riparian soils after water submergence, which is regarded as a newly formed pollution source threatening the drinking water security and aquatic ecosystem health (Liu et al. 2014; Han et al. 2016, 2017; Pei et al. 2018; Yang et al. 2018). For example, Han et al. (2016, 2017) found that anthropogenic vanadium (V), cobalt (Co), nickel (Ni), zinc (Zn), Cu, Cd, and lead (Pb) in riparian soils originated from coal combustion caused potential ecological impacts on the Miyun Reservoir water. Moreover, the impacts of water level fluctuation within the Three Gorges Reservoir and Manwan Reservoir notably increased the enrichment degree and risk level of Cd in riparian soils after water submergence (Liu et al. 2014; Pei et al. 2018; Yang et al. 2018). However, previous studies mainly focused on the total concentrations, chemical fractions, and ecological risk assessments of metal(loid)s. Little is known about the solubilization characteristics of metal(loid)s influenced by changing aquatic physicochemical conditions after water submergence.

Water submergence in riparian soils within reservoirs largely modifies the micro-environments of the soil-water interface (Sun et al. 2018). Actually, in a dam-reservoir context, increased frequency of high-intensity flood events and water level fluctuation could generate similar alterations in physicochemical conditions, especially during the water release and intake stages in daily regulated reservoirs (Frémion et al. 2017; Chen et al. 2020). The re-suspension of riparian soils, continuous cycles of drought-wetting processes, and rapid changes of water environments (e.g., suspended solid concentration (SSC), acid-alkaline property (pH), and oxidation-reduction potential (ORP)) could notably influence the evolution of metal(loid) mobility (Larner et al. 2008; Pareuil et al. 2008; Du Laing et al. 2009; Lin et al. 2018). Recently, Gao et al. (2017, 2018), Sun et al. (2018), and Xu et al. (2018, 2019) applied various in situ analysis techniques to predict the mobilization characteristics of soil-associated metal(loid)s, including Co, Ni, Cu, Zn, and Cd, within the Miyun Reservoir prior to water submergence. However, the complex field environments brought too much uncertainty to determine the key physicochemical parameters that influenced metal(loid) mobility (Monnin et al. 2018). So, to obtain a comprehensive insight into the impacts of water submergence on metal(loid) solubilization in riparian soils, an indispensable methodological approach should be conducted through independent laboratory sequential extractions and solubilization tests under the different SSC, pH, and ORP conditions as well as drought-wetting cycles. Previous studies have demonstrated that changed hydrodynamic conditions regulated by dam operation can facilitate the solubilization and release of metal(loid)s from sediments to overlying water column, especially for those non-residual metal(loid)s derived from human activities (Frémion et al. 2016, 2017). Thus, in this study, we hypothesized that the disturbed physicochemical factors after water submergence within reservoirs would increase the solubilization of metal(loid)s from riparian soils based on the laboratory solubilization tests.

The Jinsha River originates from the Qinghai-Tibet Plateau with a length of 2290 km, which is an important water source for the Yangtze River, the mother river in China (Yuan et al. 2019). Over the past decade, a cascade of six large hydropower dams has been constructed and operated along the middle reaches of the Jinsha River, which modified hydrodynamic processes, increased river water table, and impacted riparian ecosystems within reservoirs (Li et al. 2018b; Xia and Xu 2018; Xiong et al. 2019). Previous studies have analyzed the variations of metal(loid)s in surface water, suspended particles, and riverbed sediments as well as the biological accumulation of fishes (e.g., *Silurus asotus*, *Cyprinus carpio*, *Carassius auratus*) in the Jinsha River (Teng et al. 2011; Wu et al. 2013; Li et al. 2018c; Yuan et al. 2019, 2021). It is clear that an investigation on the spatial distribution and solubilization characteristics of metal(loid)s in riparian soils within reservoirs is crucial to drinking water security along the middle Jinsha River.

2 Materials and methods

2.1 Study area and field sampling

The Jinsha River is one of the largest rivers in southwestern China, flowing through the Qinghai Province, Tibetan autonomous region, Sichuan Province, and Yunnan Province (Yuan et al. 2019). The six cascading large dams constructed on the midstream of the Jinsha River include the Liyuan (LY), Ahai (AH), Jin'anqiao (JAQ), Longkaikou (LKK), Ludila (LDL), and Guanyinyan (GYY) dams (Fig. 1). Detailed information about these six dams, such as reservoir area, annually river flux, normal and dead water levels, regulation frequency, storage capacities, and operation time, is presented in Supplementary Table S1.

The field sampling was conducted in May 2017, the early rainy season, with no extreme weather condition during the sampling periods. A total of 21 riparian topsoil samples (0–20 cm) periodically affected by floods (< 5 m from the high water-level line) were collected with a plastic shovel (Chen et al. 2019). There were seven sites in the upper reaches labeled Up1-7; 12 sites in the middle reservoir-regulated reach labeled LY1/2, AH1/2, JAQ1/2, LKK1/2, LDL1/2, and GYY1/2; and two sites in the downstream area labeled Down1-2 (Fig. 1). Geographic coordinates (longitude and latitude) of sampling sites were recorded by a handheld device

equipped with a global positioning system (GPSmap 62s, Garmin, USA) (Table S2). At each sampling site, three replicate sampling plots of 1×1 m^2 were selected randomly along a ~10-m longitudinal river transect.

2.2 Metal(loid) sequential extraction and soil property analysis

All soil samples were sealed in 100-mL sterile plastic centrifuge tubes and stored in the ice boxes at 4 °C, then freeze-dried to a stable weight. These freeze-dried soil samples were powdered, homogenized, and sieved through a 1-mm mesh. One gram of soil sample was used for the measurement of four different metal(loid) fractions including exchangeable, reducible, oxidizable, and residual parts based on a four-stage sequential extraction procedure proposed by Tokalioğlu et al. (2000). Briefly, the exchangeable metal(loid)s (M_{exc}) were extracted with 0.11 M acetic acid; the reducible metal(loid)s (M_{red}) bound to iron-manganese-aluminum (Fe-Mn-Al) oxides and hydroxides were extracted with 0.50 M hydroxylamine hydrochloride (pH 1.5 adjusted with HNO_3); and the oxidizable metal(loid)s (M_{oxid}) bound to organic matter and sulphide were extracted with 30% (w/w) hydrogen peroxide (H_2O_2) and 1 M ammonium acetate (pH 1.5). The last fraction, residual metal(loid)s (M_{res}) underwent the same procedure than for the determination of total metal(loid)s, i.e.,
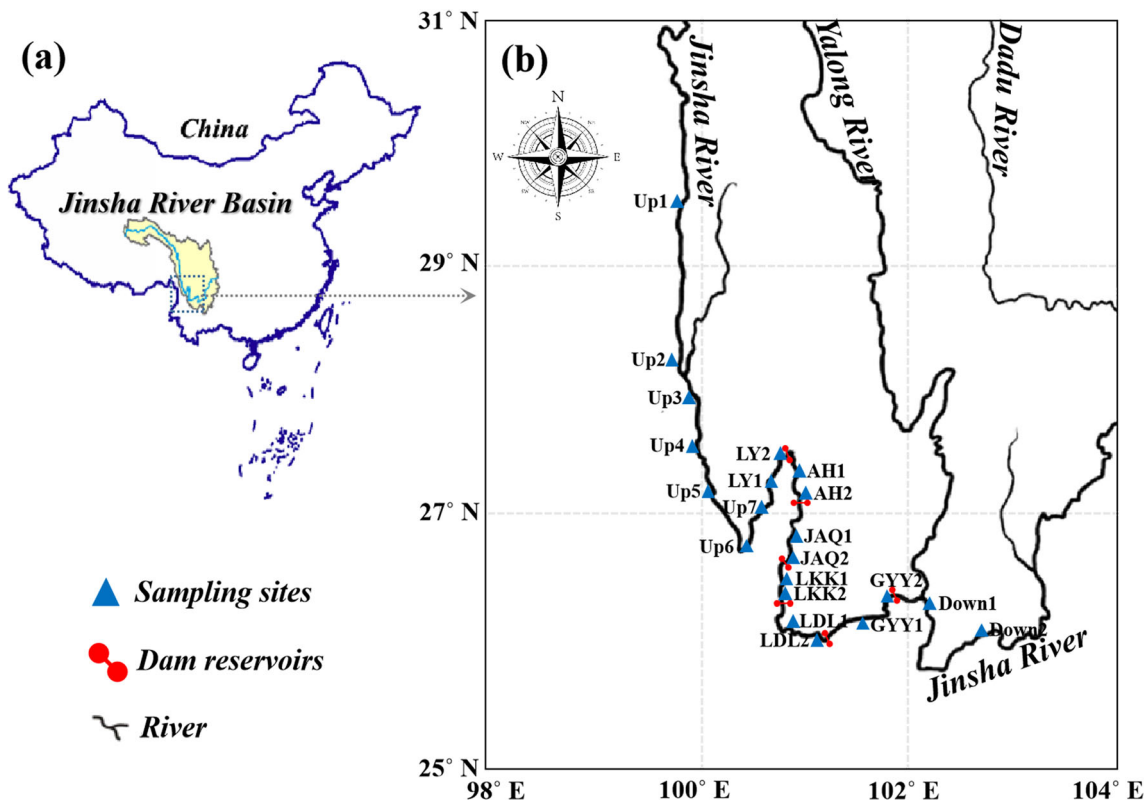
Fig. 1 Sampling sites of riparian soils along the Jinsha River. (a) Location of the Jinsha River Basin. (b) Distribution of sampling sites and dams along the middle Jinsha River

digestion in a microwave oven (ETHOS-A, Milestone, Italy) with hydrochloric/nitric/hydrofluoric (HCl/HNO$_3$/HF = 2/5/1) mixed acids, according to the digestion protocol of the USEPA 3052 (Strady et al. 2017).

Concentrations of Cu, Zn, Ni, chromium (Cr), Cd, and Pb were measured by an inductively coupled plasma-mass spectrometer (ICP-MS) (Thermo XII series, MA, USA) and arsenic (As) and Hg were measured by an atomic fluorescence spectrometer (AFS) (Reilly AF-630A, Beijing, China) with external calibrations. To quickly and accurately analyze different chemicals, Fe, Mn, and Al were measured with an inductively coupled plasma-atomic emission spectrometer (ICP-AES) (PE 2100DV, MA, USA) (Yuan et al. 2019). Analytical quality assurance of metal(loid) measurements was assured by standard operating procedures with reference materials and duplicate samples (< 5% of relative standard deviation). Moreover, by comparing the sum of four extracted fractions with total metal(loid) concentrations, the accuracy of measurement was evaluated to be satisfactory (± 5%).

Concentrations of soil organic carbon (SOC) and total nitrogen (TN) were analyzed using an elemental analyzer (Flash, EA-1112, Italy). Total phosphorus (TP) was measured according to the frame of the Standard Measurements and Testing (SMT) Programme of the European Commission (Ruban et al. 2001). Soil pH and electrical conductivity (EC) values were measured at a ratio of 1:5 solid-liquid by a calibrated multi-parameter probe (HQ40d, HACH, CO, USA). The average particle diameter (D_{50}) of riparian soils was obtained using a laser diffraction particle size analyzer (Malvern, Mastersizer 2000, UK).

2.3 Solubilization characteristics of metal(loid)s

2.3.1 Influence of changing physicochemical conditions

To evaluate the influence of SSC, pH, and ORP on the solubilization of soil-associated metal(loid)s, three of the most polluted samples within reservoirs were selected to conduct the metal(loid) solubilization studies. The effect of SSC on metal(loid) solubilization was analyzed using 0.05, 0.1, 0.5, 1, and 2 g L^{-1} of SSC with neutral pH values (7.0 ± 0.5) adjusted by HNO$_3$ and NaOH. In the experiments of pH and ORP impacts, 50-mg soils with duplicates was exactly weighted and introduced into 100-mL sealed plastic centrifuge tubes with 50 mL distilled water (SSC = 1 g L^{-1}). The pH ranging from 4 to 10 (4, 5, 6, 7, 8, 9, and 10) was used to study the metal(loid) solubilization. Moreover, to investigate the influence of ORP conditions, different concentrations of sodium ascorbate were applied in 0, 0.005, 0.01, 0.05, and 0.1 M with the ORP variations from +393 to +14 mV (Pareuil et al. 2008). The pH values were adjusted to neutral condition (7.0 ± 0.5). All of the experimentally sealed glass bottles (100 mL) were intensively shaken at 180 r min^{-1} for 14 days (Frémion et al.

2017). At the end of the experiments, the mixed samples were centrifuged at 8000 g for 15 min and filtered by 0.2-μm Nylon filters (Shanghai Xin Ya Purification Equipment CO., LTD, China). Considering the potential interference of sodium ascorbate, the filtered water samples were further digested with HNO$_3$ and H$_2$O$_2$ before metal(loid) concentration analysis (Pareuil et al. 2008).

2.3.2 Impact of continuous drought-wetting cycles

To estimate the impact of frequent water drawdowns on the solubilization of metal(loid) within reservoirs, successive drought-wetting cycles were simulated to test the metal(loid) release from these most polluted samples. Each drought-wetting cycle lasted 24 h with 18-h wetting and 6-h drought processes according to the actual operation conditions of daily regulated reservoirs along the Jinsha River (i.e., LY, AH, and LKK reservoirs) and referring to the previous studies on the Vaussaire Reservoir (Liu et al. 2014; Frémion et al. 2017; Xiong et al. 2019). The experiment was conducted for 7 days with constant aquatic conditions (50 g L^{-1} of SSC, 7.0±0.5 of pH, and +381~+322 mV of ORP). During the wetting period, the 100-mL sealed glass bottles were shaken at 80 r min^{-1} with 50mL distilled water. And then, the supernatant was collected by centrifugation at 8000g. The remaining soils were oven-dried at 35 °C and then rewetted with new distilled water. All of the water samples after each day process were acidified by concentrated HNO$_3$ for metal(loid) concentration analysis.

2.3.3 Calculation of metal(loid) solubilization percentages

The percentages of metal(loid)s dissolved from riparian soils were calculated using the following formula (Frémion et al. 2017):

$$P_{\text{Solubilization}}(\%) = \frac{M_{\text{Solubilization}}}{M_{\text{Total}}} \times 100\% = \frac{C_{\text{Solubilization}}}{SSC \times C_{\text{Total}}} \times 100\% \tag{1}$$

where, the $M_{\text{Solubilization}}$ and M_{Total} represent the weights of metal(loid)s in the solutions and initial soils, respectively; the $C_{\text{Solubilization}}$ and C_{Total} represent the concentrations of metal(loid)s in the solutions (μg L^{-1}) and initial soils (mg kg^{-1}), respectively; and the SSC is expressed in g L^{-1}.

2.4 Data analysis

Statistical analyses were performed by SPSS application for Windows (version 19.0, IBM, USA). Pearson correlation analysis was applied to test the relationships between metal(loid) variables. To reveal the composition difference of

metal(loid) concentrations between soil samples, principal component analysis (PCA) was performed based on the Pearson correlation matrix. Moreover, a varimax rotation was used for a better representation of metal(loid) variables on the principal component factors. Significant differences of metal(loid) solubilization percentages between soil samples were compared by paired t-tests ($P < 0.05$).

3 Results

## 3.1 Characteristics of metal(loid)s in riparian soils along the Jinsha River

3.1.1 Metal(loid) concentrations and physicochemical properties

The total concentrations of metal(loid)s in riparian soils along the Jinsha River are shown in Fig. 2. The concentration ranges of each metal(loid) were as follows: 21.34–120.21 mg kg^{-1} for Cu, 23.07–87.14 mg kg^{-1} for Zn, 24.06–478.35 mg kg^{-1} for Ni, 57.42–649.58 mg kg^{-1} for Cr, 0.093–13.245 mg kg^{-1} for Cd, 10.49–70.39 mg kg^{-1} for Pb, 0.013–0.098 mg kg^{-1} for Hg, and 4.43–28.99 mg kg^{-1} for As (Fig. 2). Because of significant high concentrations of Cd, Ni, and Cr in the LY and AH reservoirs (i.e., LY2 and AH1 sites), the skewness coefficients of these metal(loid)s were over 2.0 with high positively-ly skewed distributions (Table S3). Moreover, the coefficients of variation (CV) of Cd, Ni, and Cr were notably higher than those of other metal(loid)s, especially the CV value for Cd (311.63%). Compared with the Chinese background values of metal(loid)s in soils, the concentrations of Zn and Hg in the whole Jinsha River riparian soils were basically low, whereas the maximum contents of Cd, Ni, Cr, and Cu were 61-fold, 11-fold, 10-fold, and 2.6-fold higher than the background values of corresponding regions (Yunnan Province), respectively (Table S3; Chen et al. 1991).

The physicochemical properties of riparian soils along the Jinsha River are presented in Table S4. The pH values of riparian soils varied from 6.98 to 9.68 with an average of

Fig. 2 Spatial distribution of metal(loid)s in riparian soils along the Jinsha River. I-VI represent the LY, AH, JAQ, LKK, LDL, and GYY dams
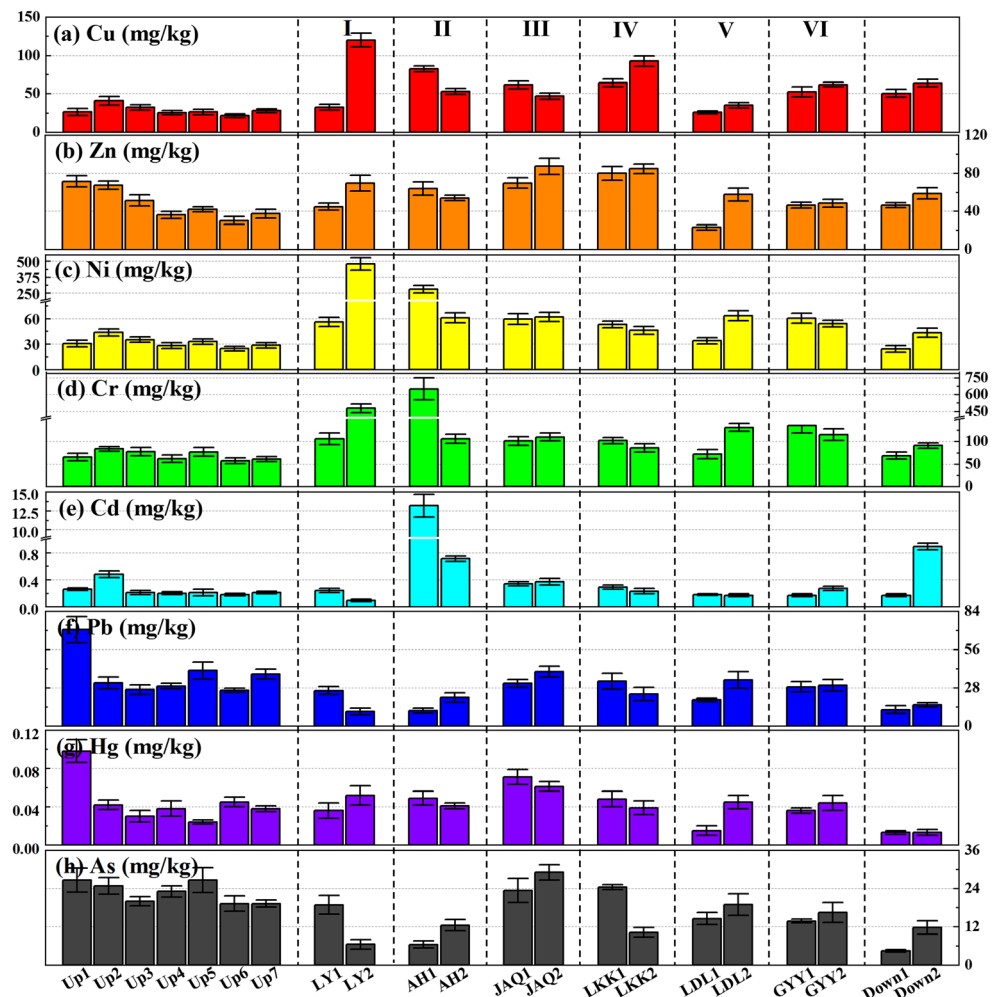
8.55, indicating that the lithology of the Jinsha River Basin is mildly alkaline. The EC, SOC, TN, and TP were 43.3–216.5 µs cm^{-1}, 5.79–92.43 g kg^{-1}, 203.87–1058.76 µg kg^{-1}, and 138.07–958.12 µg kg^{-1}, respectively. The D_{50} values of riparian soils varied from 25.48 to 192.73 µm, which showed a decreasing trend from upstream to downstream areas. The average concentrations of Mn, Fe, and Al were 1.61 g kg^{-1}, 43.93 g kg^{-1}, and 58.33 g kg^{-1}, respectively (Table S4).

3.1.2 Pearson correlation and principal component analysis of metal(loid)s

To interpret the potential sources and coexisting relationships of metal(loid)s in riparian soils, the Pearson correlation and principal component analysis (PCA) were applied. The coefficient of Kaiser-Meyer-Olkin was 0.568 (> 0.50), indicating that the proportion of variance in variables caused by underlying factors was acceptable (moderately adequate) (Patil et al. 2020). And Bartlett's test was significant ($P < 0.001$), justifying the use of PCA to analyze the variables (Yuan et al. 2019). The factor scores of metal(loid)s and sampling sites, as well as the interpretive percentages of the first two principal components, are shown in Fig. 3. Both principal components accumulatively explained 75.78% of the total variances, whose eigenvalues were > 1. The metal(loid) elements were made up of two main groups. The first group of PCA was characterized by Cr, Ni, Cu, and Cd, and the correlations between these four metals were significant, especially between Cr and Ni ($r = 0.882$, $P < 0.001$) (Table S5), indicating their

Fig. 3 Principal component analysis of riparian soil samples based on the metal(loid) loadings with varimax rotation solution

similar sources or transport fates. The second group of PCA included Hg, Pb, Zn, and As, and the Pb and As showed a high correlation ($r = 0.791$, $P < 0.001$). Because of high concentrations of Cd, Ni, Cr, and Cu, the factor scores of sampling sites at LY2, AH1, and LKK2 were notably different from others, which were regarded as the most polluted riparian soils in the Jinsha River (Fig. 3).

The relationships between soil physicochemical factors and metal(loid)s were also analyzed (Table S6). The pH and D_{50} values showed negative correlations with most metal(loid)s, particularly with Cu ($r_{(pH)} = -0.763$, $P < 0.001$; $r_{(D50)} = -0.613$, $P < 0.01$). On the other hand, the SOC, TP, Fe, and Al were positively associated with metal(loid)s, for example, Cu and Fe ($r = 0.623$, $P < 0.01$), suggesting their potential coexisting patterns in terms of adsorption, complexation, and precipitation.

3.2 Metal(loid) chemical fractions of most polluted riparian soils

Generally, the chemical speciation of metal(loid)s was an important intrinsic property that governs metal(loid) mobility, bio-availability, and toxicity (Frémion et al. 2017). In this study, three most polluted riparian soil samples (i.e., LY2, AH1, and LKK2) were selected for further chemical fraction analysis (Fig. 4) and metal(loid) solubilization experiments.

For almost all metal(loid)s, the exchangeable fraction was lower than 5% of the total concentration, except for Cd and As, both of which were more mobile than other metal(loid)s. The reducible fraction bound with Fe-Mn-Al oxides and hydroxides ranged from 1 to 10% of the total Cr, Zn, and Ni to 10–35% for the Cu, Cd, Pb, and As elements. The oxidizable metal(loid) fraction bound to organic matter and sulfide is significantly sensitive to redox conditions. This part of the most metal(loid)s was limited with < 10% of the total concentration in riparian soils. The last remaining part of metal(loid)s was the residual fraction, which accounted for a significant percentage, varying from 37.67 to 92.25%. Among all metal(loid)s, the proportions of residual fraction for Cd and As were relatively low, whereas those for Cr, Ni, and Zn were higher than 80% of the total concentration.

Although the four-stage sequential extraction procedure exists certain artificial limitations, the proportion of metal(loid)s bound to the non-residual fractions, i.e., the summation of exchangeable, reducible, and oxidizable fractions, could still abstractly reflect the mobility of metal(loid)s (Frémion et al. 2017). Therefore, the theoretical metal(loid) mobility in this study decreased as follows: Cd (47.80%) > As (43.98%) > Cu (23.44%) ≈ Pb (23.14%) > Zn (16.71%) > Ni (12.96%) > Cr (8.14%) (Fig. 4). Furthermore, the non-residual fractions of
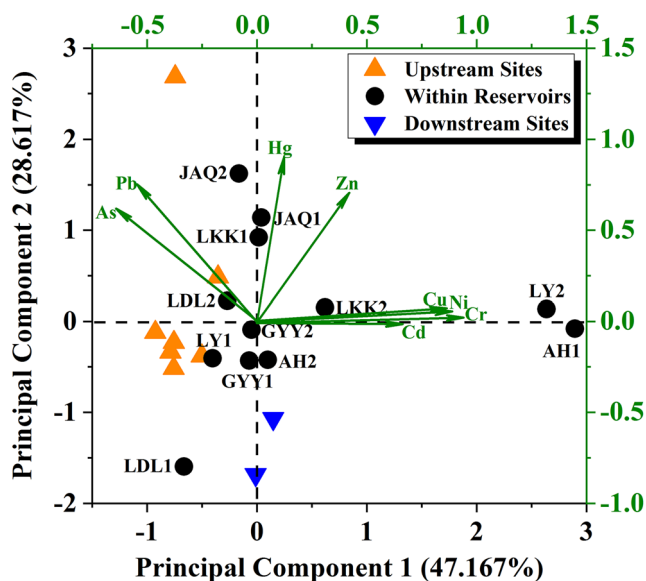
Cu, Zn, Cr, and As showed no significant difference between three polluted riparian soils, whereas the mobility of Ni at LKK2, Cd at AH1, and Pb at LY2 and LKK2 was notably higher than other sampling sites ($P < 0.05$).

3.3 Influence of physicochemical conditions on metal(loid) solubilization

3.3.1 Influence of suspended solid concentration

After water submergence within reservoirs, the SSC values in overlying water were increased by the suspension of riparian soils. The percentages of metal(loid) solubilization ($P_{Solubilization}$) were negatively correlated with the SSC values ($-0.905 < r < -0.574$, $P < 0.05$) (Table S7). When SSC < 0.1 g L^{-1}, more than 10% of the total Cd, Cu, and As was dissolved from the soil matrix, while for SSC > 1 g L^{-1}, these metal(loid) solubilization only exceeded 1% of the total concentration (Fig. 5a). The $P_{Solubilization}$ of each metal(loid) was different from one to another, which decreased in the order of Cu > Cd > As > Zn > Ni > Mn > Cr > Fe > Pb > Al. Because of low total contents in riparian soils, the dissolved concentrations of Hg were lower than the detection limit. The paired t-tests of

$P_{Solubilization}$ between different soil samples suggested that the solubilization of Cu at LY2, Cd at AH1, and As at LKK2 was significantly higher than other sampling sites ($P < 0.05$) under the changing SSC values (Table S8a).

3.3.2 Influence of acidic-alkaline property (pH values)

Variation of acid-alkaline property (i.e., pH values) after water submergence was another key controlling factor that affected soil-associated metal(loid) mobility. The decrease of water pH values led to a strong enhancement of cationic metal(loid) discharge from riparian soils, such as Zn, Ni, Cd, and Pb. The correlations between pH values and $P_{Solubilization}$ of these cationic metal(loid)s were notably negative ($-0.773 < r < -0.552$, $P < 0.01$) (Fig. 5b; Table S7). Similar behaviors were also observed for major elements, like Fe and Mn, suggesting that the acid conditions (pH < 5) could induce the increased solubilization of Fe-Mn oxides and hydroxides. On the contrary, the $P_{Solubilization}$ of Cr and As showed positive associations with the pH values ($0.603 < r < 0.818$, $P < 0.01$), indicating that alkaline conditions (pH > 9) favored the release of oxyanion species, such as CrO_4^{2-} and AsO_4^{3-}. It was interesting that the maximum solubilization of Cu occurred in
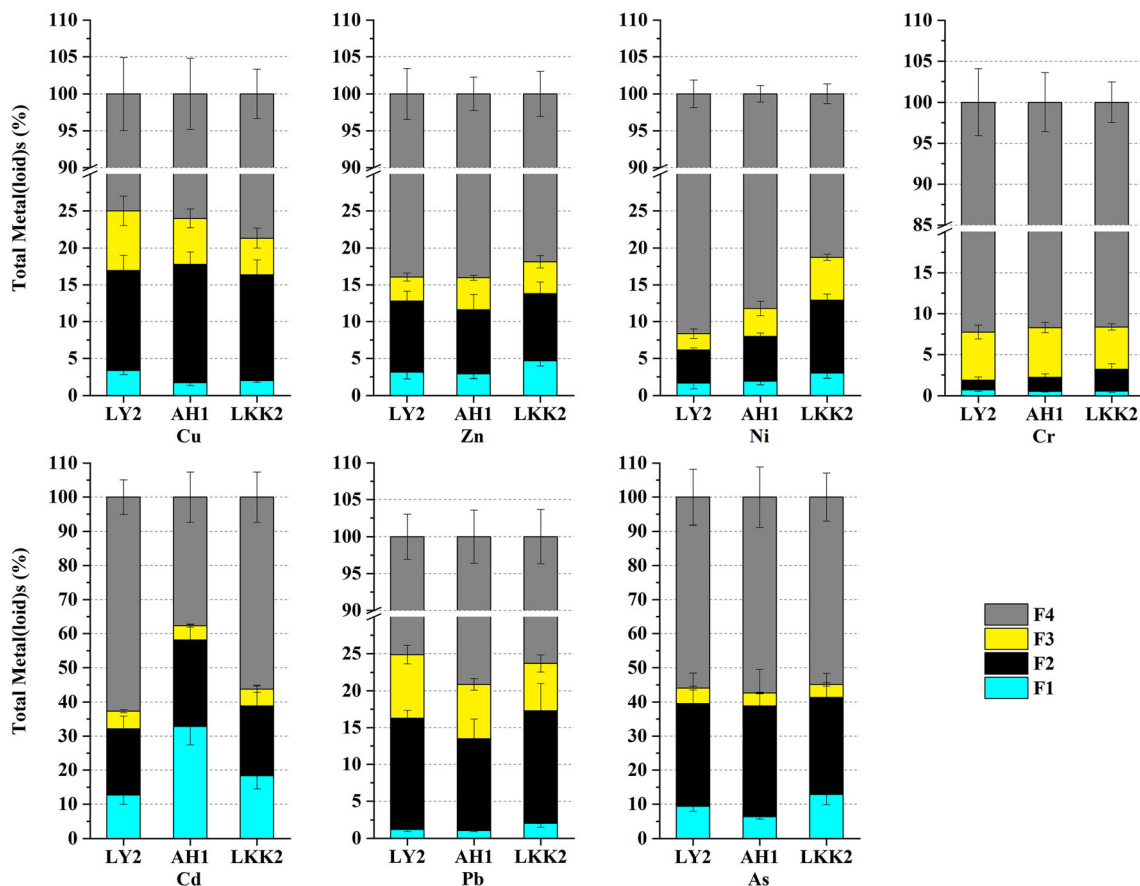

Fig. 4 Chemical fractions of metal(loid)s in the most polluted riparian soils according to the four-stage sequential extraction procedure

particularly acidic and alkaline media, which ranged from 5.46 to 10.11% of the total contents when pH units were 4 and 10. Regarding all metal(loid)s, there was no significant difference of $P_{\text{Solubilization}}$ between three soil samples under the changing pH values, except $P_{\text{Solubilization-Pb}}$ between LY2 and AH1 ($P < 0.05$) (Table S8b).

3.3.3 Influence of oxidation-reduction potential

After dam construction, the water submergence in riparian soils decreased the ORP values of soil-water micro-interface, which significantly increased metal(loid) solubilization, except Cu ($P > 0.05$), as evidenced by laboratory tests (Fig. 5c; Table S7). The solubilization percentages of all metal(loid)s were lower than 6% under the completely oxidized conditions (ORP > +350 mV). However, the reduced conditions (ORP < +50 mV) enhanced the $P_{\text{Solubilization}}$ of As, Cd, and Pb to 31.96–35.72%, 28.06–40.68%, and 8.99–11.58%, respectively. Moreover, the $P_{\text{Solubilization}}$ of two major elements, Fe and Mn, both of which are well-known for their high oxidation-reduction sensitivity, increased from < 0.5% under oxidized conditions to > 15% when ORP was less than +100 mV. On the contrary, the Zn, Ni, and Cr showed limited solubilization from riparian soils even under reduction conditions. The

paired t-tests revealed that, under the changing ORP values, the $P_{\text{Solubilization}}$ of Ni at LKK2, Cd at AH1, and Mn at LKK2 was significantly higher than other sampling sites ($P < 0.05$) (Table S8c).

3.4 Effect of drought-wetting cycles on metal(loid) solubilization

The successive drought-wetting cycles were applied to simulate the effect of frequent water drawdowns on metal(loid) solubilization from riparian soils after water submergence (Fig. 6). Within the 7 rounds of drought-wetting cycles, less than 0.5% of the total soil-associated metal(loid)s were released into overlying water, and, the $P_{\text{Solubilization}}$ of each metal(loid) decreased in the order of Cu > Ni > Cd ≈ As > Cr > Fe ≈ Mn ≈ Al. The dissolved concentrations of Hg, Zn, and Pb at all samples as well as Cd at LY2 were too low to be determined.

After the first round of drought-wetting cycle, the $P_{\text{Solubilization}}$ of Cu was slightly decreased, while those of Ni and Cd were notably increased almost an order of magnitude (Fig. 6). Furthermore, the $P_{\text{Solubilization}}$ of Ni, Cd, and Mn posed a slight decrease at the last few drought-wetting cycles. But, the $P_{\text{Solubilization}}$ of Cr, As, Fe, and Al showed no

Fig. 5 Evolution of metal(loid) solubilization percentages ($P_{\text{Solubilization}}$) as functions of (a) suspended solid concentration (SSC), (b) acid-alkaline property (pH values), and (c) oxidation-reduction potential (ORP)
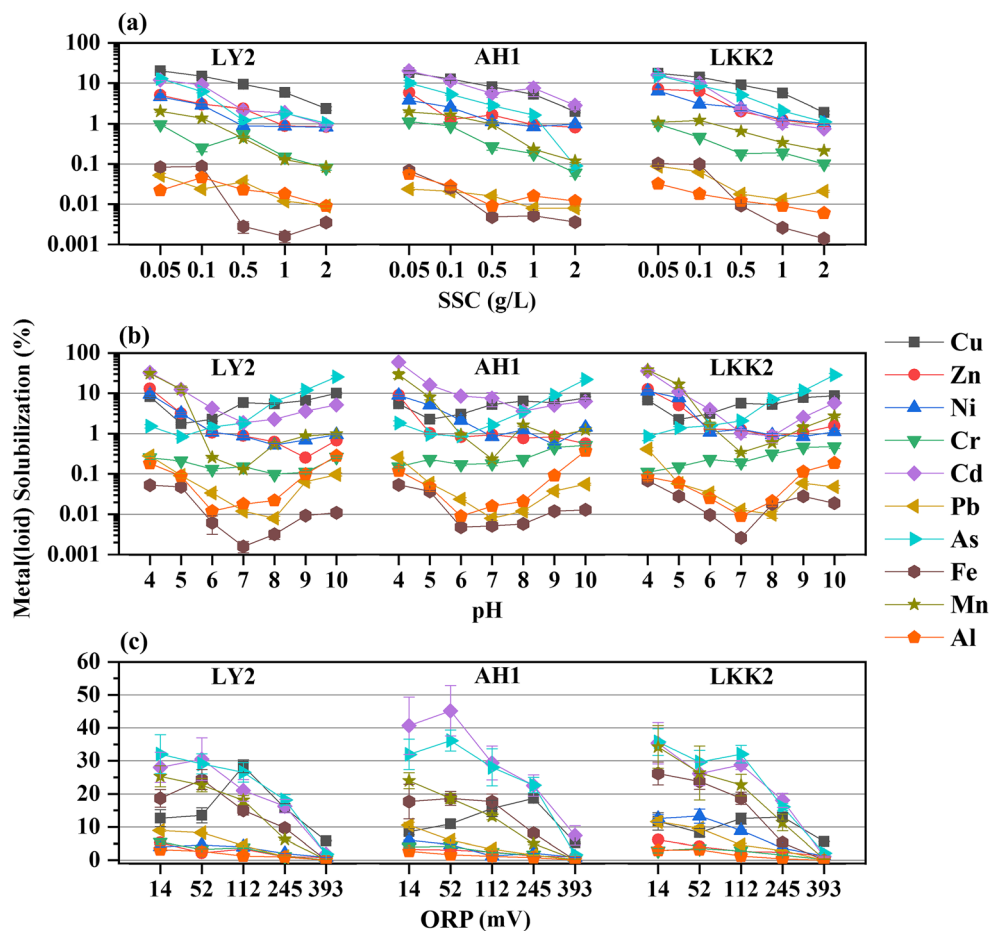
significant variations within the whole drought-wetting cycles. The comparison of $P_{Solubilization}$ between different soil samples was also analyzed by paired t-tests (Table S9). The Ni and As at LY2 and LKK2 as well as the Cd and Cr at AH1 had significant higher $P_{Solubilization}$ than other sampling sites ($P < 0.01$).

4 Discussion

The development of six cascade hydropower dams along the middle Jinsha River has created significant social and economic benefits, which saved over 40 million raw coal every year (He et al. 2015; Xia and Xu 2018). However, the dam construction also caused various environmental pollution and ecological damage (Yuan et al. 2019; Chen et al. 2020). Because of the changed hydrodynamic condition and flow regime, a total of 182.4 km^2 of water area has been formed within these reservoirs under normal water level conditions, which contained 134.7 km^2 of newly submerged areas (He et al. 2015). Furthermore, after dam operations, the frequency of high-intensity flood events was increased within reservoirs, so that the micro-interface environments between river water and riparian soil were notably altered, especially during water release/intake stages in daily regulated reservoirs, such as the LY, AH, and LKK reservoirs (Xia and Xu 2018; Chen et al. 2019, 2020). Therefore, the metal(loid) contaminants had a complex fate in changing aquatic environments when influenced by dam-reservoir disturbance. The study on spatial distribution and solubilization characteristics of metal(loid)s in riparian soils within cascade reservoirs is crucial to drinking water security and aquatic ecosystem health along the middle Jinsha River after water submergence.

4.1 Spatial distribution and ecological risks of metal(loid)s in riparian soils

The metal(loid)s in riparian soils presented different spatial distribution patterns along the Jinsha River (Figs. 2 and 3).

The significantly high skewness and variation coefficients of Cr, Ni, Cd, and Cu revealed great heterogeneous distribution of metal(loid)s (Table S3). The concentrations of Cr, Ni, Cu, and Cd in riparian soils within the LY and AH reservoirs were notably higher than those in other reaches of the Jinsha River and many other large rivers in China, such as the Yangtze River (Pei et al. 2018; Bing et al. 2019), Lancang-Mekong River (Liu et al. 2014; Strady et al. 2017), and Yellow River (Bai et al. 2012; Sun et al. 2015).

The significant enrichment of Cr and Ni in the upper midstream was probably from soil parent materials, whose distribution trend was strongly consistent with the spatial variation of metal(loid)s in riverbed sediments along the Jinsha River, as reported in our previous study (Yuan et al. 2019). The list of national key monitoring enterprises in 2016 revealed that there was almost no metal(loid) contamination associated company located in the middle Jinsha River, owing to the project requirements of the Sanjiangyuan National Natural Reserve, the source areas of Yangtze, Yellow, and Lancang rivers. Moreover, both the Cr and Ni had relatively high residual fraction percentages, suggesting that these two metal(loid)s posed less mobility in riparian soils (Fig. 4). According to the conditions of the Sanjiang Tethyan Metallogenesis in southwestern China, large amounts of base metal and rare metal ores are deposited in the middle catchments of the Jinsha River (Hou et al. 2007). Thus, the elevated concentrations of soil-associated Cr and Ni within the LY and AH reservoirs were mainly originated from the natural sources, for example, the chromite ore (Wu et al. 2013; Yuan et al. 2019).

However, the past anthropogenic activities have also left a legacy of metal(loid) pollution, for example, the copper metallurgy previously recorded in the Yunnan Lake sediments (Zhao 2013; Hillman et al. 2015). In this study, the Cu posed elevated concentrations within the LY reservoir, which was co-occurred with the major element Fe in riparian soils (Fig. 2; Table S6). Over the past decades, numerous copper-ferrous mining activities were conducted around the Jinsha River bank, and thus large amounts of Cu-rich wastewater and
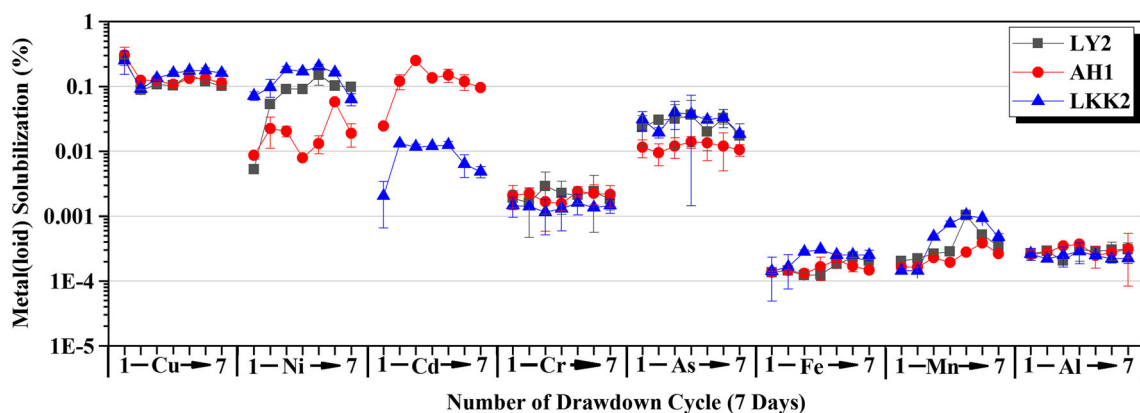

Fig. 6 Changes of metal(loid) solubilization percentages ($P_{Solubilization}$) as a function of the number of drawdown (drying/wetting) cycles

residues were abandoned irresponsibly without effective treatments (Yuan et al. 2021). Accompanied with the surface run-off and weathering erosion, the Cu pollutant was eventually transported into the riparian soils and riverbed sediments of the catchment, which subsequently caused severe threats to surrounding terrestrial and aquatic ecosystems (Eggleton and Thomas 2004; Yuan et al. 2021). In addition, the significant Cd contamination left over by history with high percentages of non-residual fraction happened in riparian soil within the AH reservoir (Figs. 2 and 4). It has been reported that the long-term addition of excessive amounts of fertilizers and fungicides could cause the enrichment of Cd as well as Cu and Zn (Yang et al. 2013). However, the concentrations of these three metal(loid)s did not increase together at the AH1 site, and the agriculture-related environmental parameters, such as SOC, TN, and TP, showed no significant correlation with Cd (Tables S5, S6). Therefore, the Cd contamination was mainly derived from the industrial sources, such as metallurgy, mining, and smelting (Strady et al. 2017). Although most industrial activities have been forbidden since the ecological protection, the historical Cd pollution still posed great hazards in riparian soils along the middle Jinsha River.

To determine whether the metal(loid)s in riparian soils posed ecological threats to aquatic biota after water submergence, the sediment quality guidelines (SQGs) were firstly applied to assess the potential risks of each metal(loid) (Macdonald et al. 2000). The adverse biological effect cannot happen when the metal(loid) contents are lower than the threshold effect concentrations (TECs), whereas it may occur as the metal(loid) contents reach the probable effect concentrations (PECs) (Bing et al. 2019). In this study, the concentrations of Zn and Hg in riparian soils were entirely below the TECs, and the Pb and As posed limited adverse ecological effects, whose concentrations were basically lower than the half of PECs (Table S3). However, the concentrations of Ni and Cr at the LY2 site and Cd, Ni, and Cr at the AH1 site were notably higher than the thresholds of PECs. In addition, the contents of Cu at LY2 and LKK2 sites reached the half of PECs, indicating that potential ecological risks may exist in these riparian soils (Yuan et al. 2019). Therefore, we concluded that, according to the SQGs, the Zn, Hg, Pb, and As in riparian soils along the whole Jinsha River posed low ecological risks, while the Ni, Cr, Cd, and Cu could threaten the aquatic biota within the LY, AH, and LKK reservoirs, once riparian soils were submerged.

4.2 Solubilization characteristics of metal(loid)s affected by water submergence

The water submergence in riparian soils within reservoirs could greatly change various physicochemical conditions, such as pH, SSC, and ORP, in soil-water micro-interface. These environmental factors played an important role in the influences on metal(loid) partitioning between particulate and dissolved phases, which subsequently alter metal(loid) bio-availability through the dissolution-precipitation and sorption-desorption processes (Eggleton and Thomas 2004). Therefore, the three most polluted riparian soils within the AH, LY, and LKK reservoirs were selected for a series of ex situ analysis of metal(loid) solubilization under the influence of changing pH, SSC, and ORP conditions as well as successive drought-wetting cycles.

It has been demonstrated that, after dam constructions, the suspended sediment concentrations and loads were significantly decreased within cascade reservoirs of the Jinsha River (Li et al. 2018b). However, the "starving flow" in turn promoted the erosion of river channels and riparian soils, particularly during flood seasons, with frequent high-intensity flood events (Chen et al. 2019). According to the previous studies on the responses of the Jinsha River sediment loads to cascade reservoirs, the SSC ranged from 2.70 mg L^{-1} during the low flow periods to 1.96 g L^{-1} during the reservoir discharge periods (Li et al. 2018b; Chen et al. 2020). In this study, the metal(loid) solubilization analysis highlighted that the lower the content of suspended riparian soils was, the higher the solubilization percentages of metal(loid)s were (Fig. 5a). The variation of $P_{Solubilization}$ was owing to the changes of both soil-water micro-interface area and adsorption equilibria, as described by the previous studies on sediment-associated metal(loid) partitioning (Frémion et al. 2016, 2017). Moreover, the intrinsic propriety of chemical fractions was another factor that governs metal(loid) solubilization (Eggleton and Thomas 2004). The metal(loid)s, like Cu, Cd, and As, with high mobility posed relatively significant solubilization characteristics, whereas the Cr and Ni strongly bound to solid phases presented low solubility (Fig. 5a). In addition, regarding the individual metal(loid), the difference of chemical fractions caused notable spatial variability of metal(loid) solubilization, especially for the Cd between AH1 and other two sites (Fig. 4, Table S8).

The metal(loid) solubilization was also largely dependent on the water pH values. The acidic conditions notably increased the release of cationic metal(loid)s from riparian soils (Fig. 5b), which is because the elevated proton (H$^+$) replaced various cations that originally adsorbed onto the surface sites and binding ligands of soil particles (Frémion et al. 2017). Moreover, the increased solubilization of major elements, such as Fe, Mn, and Al, with the decreasing pH values, could also enhance the release of metal(loid)s bound to Fe-Mn-Al oxides and hydroxides (Martin 2005). On the other hand, the solubilization of Cr and As was positively correlated with the pH values owing to the competition between hydroxide (OH$^-$) and oxyanion species, for example, CrO$_4^{2-}$ and AsO$_4^{3-}$, for the adsorption on soil particles (Cappuyns and Swennen 2008). It was unexpected that the Cu solubilization showed increasing trends under both extremely acidic and alkaline

conditions (Fig. 5b). The enhanced release of Cu from riparian soils under low pH values was undoubtedly because of the elevated H^+ contents, whereas the increased Cu solubilization under high pH values was attributed to the simultaneous release of dissolved organic matters, consistent with the findings in the previous study (Frémion et al. 2017). Because of the carbonate-dominated lithology of the Jinsha River catchment, the pH values of river water, riverbed sediments, and riparian soils all presented slightly alkaline property (Table S4; Wu et al. 2013; Yuan et al. 2019). Therefore, under natural conditions, the specific neutral-alkaline conditions may limit the metal(loid) release from riparian soils after water submergence, even for the most mobile elements.

Within cascade reservoirs, the ORP conditions of soil-water micro-interface were also influenced by water dissolved oxygen and soil organic matters through the daily regulated water release/intake processes (Frémion et al. 2017). The reduced conditions could promote the solubilization and release of metal(loid)s from riparian soils (Fig. 5c). Because of the relatively high percentages of reducible metal(loid) fractions, the $P_{Solubilization}$ of Cd, As and Pb were significantly affected by the ORP conditions. However, the Zn, Ni, and Cr mostly bound to the residual fractions presented limited variations of $P_{Solubilization}$ in response to ORP variations (Figs. 4 and 5c). The Fe and Mn, as oxidation-reduction sensitive major elements, play an important role in regulating the solubilization of most metal(loid)s (Simpson et al. 1998). Low ORP conditions led to the significant dissolution of Fe and Mn, which subsequently stimulated the synchronous release of metal(loid)s associated with Fe-Mn oxides and hydroxides (Fig. 5c). For the individual metal(loid), the higher reducible fractions of Cd at AH1 and Ni at LKK2 existed, the higher solubilization percentages of relevant metal(loid)s were (Fig. 4, Table S87). Moreover, the enhanced solubilization of As under the reduced conditions was also owing to its own reducing reaction, which may vary from AsO_4^{3-} (V) to AsO_3^{3-} (III) and become more soluble than that under oxidizing conditions (Pareuil et al. 2008; Frémion et al. 2017).

In addition to the changing SSC, pH, and ORP conditions, the frequent water drawdowns, accompanied with the significant water level variations regulated by dam operations, also affect the metal(loid) solubilization (Larner et al. 2008; Du Laing et al. 2009; Lin et al. 2018). In fact, before dam constructions, the riparian soils were completely under the aerobic conditions with low contents of reductive substances, for example, sulphide (S^{2-}). Furthermore, the carbonate-dominated lithology acted as an effective buffer, which inhibited the changes of acidic-alkaline conditions. Thus, the metal(loid) solubilization presented overall low sensitivity to the drought-wetting cycles, except Cu, Ni, and Cd (Fig. 6). At the initial phases of the cycles, the slightly decreased Cu and notably increased Ni and Cd solubilization were attributed to the simultaneous release of organic matters from riparian

soils. After that, the metal(loid) solubilization remained stable until the last few stages. Finally, the solubilization was reduced because the most mobile metal(loid) fractions were about to be released completely. The solubilization of metal(loid)s during drought-wetting cycles was mainly dependent on the chemical fractions. The higher mobility of Ni at LY2 and LKK2 and the Cd at AH1 posed, the more metal(loid) solubilization was (Table S9).

5 Conclusions

Because of the specific metal(loid) ores and historical industrial contamination along the middle Jinsha River, significant Cr, Ni, Cd, and Cu were enriched in riparian soils within the LY, AH, and LKK reservoirs. According to the SQGs, the Zn, Hg, Pb, and As had low ecological risks, while the Ni, Cr, Cd, and Cu posed potential threats to aquatic biota after water submergence. The physicochemical conditions, such as pH, SSC, and ORP, played an important role in the influences on metal(loid) solubilization through dissolution-precipitation and sorption-desorption processes. The low SSC variables could contribute to over 20% solubilization of total Cd, Cu, and As contents. Moreover, the neutral-alkaline pH conditions may limit the metal(loid) release from riparian soils after water submergence because of the carbonate-dominated lithology. Finally, the decreased ORP values were favorable for more solubilization of metal(loid)s bound to Fe-Mn oxyhydroxides or influenced by the reducing reaction. Under changing physicochemical conditions, the solubilization levels of metal(loid)s between different sampling soils were mainly dependent on the chemical fractions. Thus, this study highlighted that the investigation on both spatial distribution and solubilization characteristics of metal(loid)s in riparian soils is important for future environmental management in reservoirs.

Declarations

Conflict of interest The authors declare that they have no competing interests.

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