



Temporal and spatial variation and risk assessment of soil heavy metal concentrations for water-level-fluctuating zones of the Three Gorges Reservoir

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Abstract

Purpose This study aimed to reveal the temporal and spatial variation of soil heavy metal concentrations in the Three Gorges Reservoir area (TGR) water-level-fluctuating zone (WLFZ) and evaluated its pollution status and potential ecological risks and provide scientific basis for ecological risk prevention and ecological restoration of the TGR.

Materials and methods This study was based on long-term monitoring of soil heavy metals (Cu, Pb, Cd, and Cr) before water level fluctuation (2008) and after 1 (2009), 4 (2012), or 7 (2015) cycles of water level fluctuation at the altitude of 155–172 m in the Wushan (WS) and Zigui (ZG) sections of the TGR, and pollution status and potential ecological risks of each heavy metal element were evaluated by index of geoaccumulation and potential ecological risk index.

Results and discussion The Cd concentration increased with the increase in the number of reservoir water level fluctuations, whereas the concentrations of Cu, Cr, and Pb varied with the monitoring site. The Cd showed clear horizontal transfer characteristics. Moreover, with the increase of the frequency of water level fluctuations the Cd concentration at ZG (near the dam) were higher than those at WS (away from the dam). After 7 cycles of water level fluctuation, the concentrations of most soil heavy metal were not obvious differences between soil layers (except Pb). Before and after the reservoir water level fluctuation, Cd contamination level changed from pollution-free to strong or extremely polluted, Cu contamination level changed from pollution-free to moderately polluted, and Cr and Pb were pollution-free. Before the fluctuation of the reservoir water level, the potential ecological risk of Cd in the WS reached a classification of strongly polluted, whereas pollution at ZG was considered to be low level. However, after 4 cycles of water level fluctuation the Cd pollution level increased to a very high level, whereas Cu, Cr, and Pb remained consistently low.

Conclusions There is an obvious temporal and spatial variation of heavy metal concentrations for WLFZ of TGR. Cd concentration increased with the increase in the number of reservoir water level fluctuations. Heavy metal concentrations changed from WS > ZG to WS < ZG after the impact of water level fluctuations. After 7 cycles of water level fluctuation, the distribution of heavy metals in different soil layers tends to be uniform (except Pb). Cd pollution is more serious, and there is a strong potential ecological risk.

Keywords Risk assessment · Soil heavy metal concentrations · Temporal and spatial variation · Three Gorges reservoir · Water-level-fluctuating zone

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1 Introduction

The water-level-fluctuating zone (WLFZ) of the Three Gorges reservoir (TGR) area is the most active zone for material transformation and energy flow in aquatic and terrestrial environmental systems. This area can be considered both a pollutant source and a pollutant sink, depending on the environmental conditions. The heavy metal pollution in the soil of water level fluctuation zone is not only harmful to aquatic and terrestrial organisms but can also reach overlying water through

diffusion and dissolution and can thus have an impact on the reservoir water quality and endanger human health. Therefore, study of the temporal and spatial changes of soil heavy metal concentrations and assessment of the potential ecological risk in the WLFZ of the TGR has important theoretical and practical significance for aquatic environment ecological risk control and ecological restoration.

The TGR operation features the characteristic of anti-season water level variation, that is, the high water level emerges in winter (dry season), and the low level occurs in summer (wet season) (Bing et al. 2016), this mode of operation has greatly changed the nature of the soil in the riparian zone. When the environmental conditions change, heavy metals in the soil can be released into the water column, which becomes a secondary source of pollution, leading to contamination of the entire waterbody by pollutants that are considered the most serious pollutants due to their persistence in the environment, bioaccumulation, and high toxicity (Huang et al. 2009). Meanwhile, in flood periods, the heavy metals in the natural environment and inputs from human activities in the river basin can be easily deposited into the riparian zone (Ye et al. 2011), this aggravates the possibility of pollution. Number of factors could directly influence the migration and accumulation of heavy metals in soil, such as differences in soil type, interactions between the elements, soil electrical conductivity, clay content, nutrients, enzyme activity, flooding duration, and other physical and chemical properties (Adams et al. 2004; McCauley et al. 2009; Zhao et al. 2010; Ghosh et al. 2012; Li et al. 2014; Shaheen and Rinklebe 2014).

Although some useful information about the temporal and spatial variation of soil heavy metal concentrations for WLFZ of the TGR was provided in previous studies, further study is necessary for the following reasons: the studies were mainly focused on the change of soil heavy metal concentrations under different soil types and land use patterns before and after reservoir operation, input and output of heavy metals in soil, and potential ecological risk assessment (Muller et al. 2008; Tang et al. 2008a; Ye et al. 2011, 2013; Li et al. 2013; Ao et al. 2014a, b; Tang et al. 2015; Liu et al. 2015; Gao et al. 2016b), since these studies preferred short-term investigations, the conclusions were not consistent. For heavy metals migration, reports were usually limited to sediment analysis, and less soil research (Li and Zhang 2010; Tang et al. 2015; Gao et al. 2016a; Bing et al. 2016). Meanwhile, researchers are more concerned about heavy metals migration in the horizontal direction (Tang et al. 2015), less attention to the vertical direction (Tang et al. 2014), and lack of researches on the migration of heavy metals in the soil profile in WLFZ. Moreover, most of these studies took place in the early stages of the formation of the WLFZ, and there is little information available about heavy metal pollution in reservoirs and their changes after prolonged exploitation (Ye et al. 2011). Because of the instability of the soil environment in the early stage of the WLFZ,

the heavy metal element concentration in the soil fluctuates substantially throughout the year. Therefore, there is likely to be a large uncertainty associated with the estimation of heavy metal concentrations in the WLFZ. Thus, it is thus necessary to increase the regularity of monitoring the change of heavy metal concentrations in the soil by establishing fixed monitoring plots in the WLFZ in different areas of the TGR area and conduct long-term monitoring. However, to the best of our knowledge, such an experiment has not yet been conducted.

Considering the above factors, in this paper, four heavy metals with high ecological risk in the reservoir area (Cu, Pb, Cd, and Cr) were selected as the research object, which was based on the fixed monitoring plots established in the WS and ZG sections of the TGR in 2008. In 2008 (before the formation of the WLFZ), 2009 (1 cycle of water level fluctuations), 2012 (4 cycles of water level fluctuations), and 2015 (7 cycles of water level fluctuations), the heavy metal concentrations in different soil layers at 155–172 m above sea level in the WLFZ were measured. The objectives of our study were (1) to reveal the temporal and spatial variation of heavy metal concentrations in soil of the WLFZ of the canyon area of the TGR, (2) to evaluate the pollution status and potential ecological risk by the index of geoaccumulation and the potential ecological risk index. This research could be helpful for knowing the soil quality changes with the stable operation of reservoir, thus estimating its security to residents and making contributions to strategic decisions for the TGR.

2 Materials and methods

2.1 Study areas

The fixed monitoring plots are located in the typical fluctuation zone of the WS and ZG sections of the TGR. The ZG section of the WLFZ of the fixed monitoring plot is located at the Three Gorges Dam near the town of Maoping. The geographical coordinate is 30°52'36.8"N, 110°55'01"E with a 36° slope, and a slope direction of WN 16°. The soil type is yellow soil developed on granite parent material (Shi et al. 2004). The soil thickness is about 40 cm. The region is characterized by a humid subtropical monsoon climate (Tang et al. 2015), the annual average temperature is 18.0 °C, a ≥ 10 °C annual accumulated temperature of 5723.6 °C, an annual frost-free period of 306 days, an average annual rainfall of 1100.0 mm, and 1631.5 h of annual sunshine. Before submergence, the native vegetation of the plot was a *Pinus massoniana* plantation with a forest age of about 20 years. The canopy closure was 0.5, and the forest litter coverage was 70%. In the spring of 2008, according to the provisions of the reservoir management department, the trees and large shrubs on the plot were cut down and removed.

The WS section of the WLFZ is located in the town of Wuxia, which is located in the upper reaches of ZG, at about 120 km from ZG. The geographical coordinates are 31°03′34.4″N, 109°54′38.6″E with a slope of 41° and a slope direction of WN 25°. The soil type is yellow lime soil developed on limestone parent material (Shi et al. 2004). The soil thickness is about 35 cm. The climate type is the same as that of ZG, with an annual average temperature of 18.4 °C, $a \geq 10$ °C annual accumulated temperature of 5857.3 °C, an annual frost-free period of 305 days, and an average annual rainfall of 1049.3 mm. The native vegetation of the plot is secondary shrubs such as *Hibiscus syriacus*, *Hypericum patulum*, and *Ilex bitorisensis* and the forest litter coverage is about 60%.

All plots were established in August 2008. The vertical projection area is 15 × 16 m. The bottom edge is located at an elevation of 156 m and the top edge is at an elevation of 172 m. As the end of 2008, the highest water level of the reservoir from the elevation of 155 to 172 m, so, when the plot was established, areas above the elevations of 155 m had not yet experienced flooding.

2.2 Research methods

2.2.1 Soil sample collection and treatment

Five soil samples were collected from the plots in August of 2008, 2009, 2012, and 2015; one from the center of the sample plot and the other four from the corners. At each collection point, samples were collected from 0 to 10, 10 to 20, and 20 to 30 cm soil depth. Each soil sample from each sampling position and each depth weighed about 1 kg. Soil samples were collected and brought back to the laboratory. In the laboratory, the soil samples were naturally air-dried and then the gravel, roots, and other debris were removed from the dry soil samples. Finally, soil samples were agate mortar ground and screened by 100 mesh nylon screen, and then loaded into a paper bag and numbered. Each soil sample was subjected to the same collection and processing methods.

2.2.2 Measurement methods

For the analysis of heavy metals, each sample was weighed accurately for 0.10–0.15 g and placed into Teflon digestion tubes, followed by digestion with 5 mL HNO₃, 2 mL HCl, and 2 mL HF in a microwave digestion system (MARS5, CEM, USA). After digestion, the Teflon tubes were cooled to ambient temperature, uncapped, and then placed on a hot plate around 140 °C to allow solutions to evaporate to dryness. After cooling, deionized water was added to bring the final volume to 50 mL. The solution was then filtered through a 0.45- μ m filter membrane and stored in a PVC (polyvinyl chloride) bottle at 4 °C prior to analysis. Four heavy metals (Cu, Pb, Cr, and Cd) were analyzed using plasma emission spectroscopy (IRIS Intrepid II

XSP, Thermo Fisher Scientific, USA). All glassware and Teflon devices were thoroughly acid-washed. Reagents of analytical grade were used for blanks and calibration curves. The QA/QC procedures were conducted by using the standard reference materials (GBW07401, GBW07405, obtained from Chinese Academy of Measurement Science) with each batch of samples (one blank and one standard for each 20 samples). Rigorous QA/QC protocols include inserting of “blind” standard reference materials for determination of the accuracy of the methods and analytical duplicates to allow estimation of the precision of the method. Satisfactory recoveries are obtained for Cd (93–98%), Cr (98–103%), Pb (94–106%), and Cu (96–103%). Soil pH was measured using the desktop acidity meter (pH 211, Hanna instruments, Italy), with a water to soil ratio of 2.5:1 (State Forestry Administration 1999). All the measurement and analysis work was conducted in the Key Laboratory of Forest Ecology and Environment of the State Forestry Administration, Research Institute of Forest Ecology, Environment and Protection, Chinese Academy of Forestry, Beijing.

2.3 Soil heavy metal pollution and potential ecological risk assessment methods

2.3.1 Index of geoaccumulation

The index of geoaccumulation (I_{geo}) (Muller 1969) is a commonly used quantitative indicator of soil heavy metal pollution evaluation and is calculated as follows:

$$I_{geo} = \log_2(C_i/kB_i),$$

where I_{geo} is the index of geoaccumulation, C_i is the measured concentration of the heavy metal i in soil, B_i is the geochemical background value of the element, and k is the coefficient that takes into account the variation of the background value that may cause the background value to change (taken as 1.5 in this study). The degree of pollution was divided into seven levels according to the size of the I_{geo} value. $I_{geo} < 0$ means pollution-free (PF), $0 \leq I_{geo} < 1$ means pollution-free to moderately polluted (PFM), $1 \leq I_{geo} < 2$ means moderately polluted (MP), $2 \leq I_{geo} < 3$ means moderately to strongly polluted (MS), $3 \leq I_{geo} < 4$ means strongly polluted (S), $4 \leq I_{geo} < 5$ means strongly to extremely polluted (SE), and $I_{geo} \geq 5$ means extremely polluted (E).

2.3.2 Potential ecological risk index

The potential ecological risk index method (Hakanson 1980) takes into account the concentration effects of various heavy metals in sediments, the toxic effects of different heavy metals, and the synergistic effects of multiple heavy metals complex pollution. This method not only reflects the risk of single heavy metal pollution but also reflects the

comprehensive impact of pollution from a variety of heavy metals. The formula is as follows:

$$RI = \sum E_i = \sum [T_i(C_s^i/C_n^i)],$$

where *RI* is the total potential ecological risk index, *E_i* is the potential ecological risk coefficient of heavy metal *i*, and *T_i* is the toxic response coefficient of heavy metal *i*, which reflects the toxicity level of the heavy metal. In this study, the critical heavy metal toxicity coefficient developed by Hakanson (1980) was taken as Cu = 5, Cd = 30, Cr = 2, Pb = 5; *C_sⁱ* is the measured concentration of heavy metal *i* in soil; and *C_nⁱ* is the geochemical background value of heavy metal *i*. Pollution by a single heavy metal was divided into five levels according to the *E_i* value where *E_i* < 40 means low potential ecological risk (L), 40 ≤ *E_i* < 80 means moderate potential ecological risk (M), 80 ≤ *E_i* < 160 means considerable potential ecological risk (C), 160 ≤ *E_i* < 320 means high potential ecological risk (H), and *E_i* ≥ 320 means very high potential ecological risk (VH). The total potential ecological risk index is divided into four levels according to the *RI* value. *RI* < 0 means low ecological risk (LR), 150 ≤ *RI* < 300 means moderate ecological risk (MR), 300 ≤ *RI* < 600 means considerable ecological risk (CR), and *RI* ≥ 600 means very high ecological risk (VHR).

The geochemical background value is one of the key parameters in the *I_{geo}* and potential ecological risk index assessment. In general, soil heavy metal background values in the study area or adjacent area are used as the soil heavy metal background values for the study area, or the soil heavy metal background value in other areas in which the environmental conditionals are similar to those of the study area may be used to replace this value. In this study, the soil heavy metal background value in the TGR area was selected as the geochemical background value (Tang et al. 2008b) (Table 1).

2.4 Statistical analysis

One-way ANOVA (Student–Newman–Keuls test, *p* < 0.05) and least significant difference (LSD) were used to detect the difference between each measurement years. Pearson correlation analysis and two-tailed test methods were applied to establish the relationship between values. All statistical analysis in this study were performed by the software packages SPSS 16.0 (IBM, USA), Excel 2013 (Microsoft, USA), and SigmaPlot 10.0 (Systat Software Inc., USA) for Windows 7 (Microsoft, USA).

Table 1 The background value of soil heavy metals in the Three Gorges reservoir area (mg • kg⁻¹)

Elements	Cu	Cd	Cr	Pb
Background value	25.00	0.13	78.03	23.88

3 Results

3.1 Temporal variation of soil heavy metal concentrations

The concentration of each soil heavy metal element in the same soil layer was summarized according to the year of measurement and assessed to determine the significance of the differences between the measurement years (Table 2). The results show that the concentrations of heavy metal were variable (*p* < 0.05) with the increase in the number of reservoir water level fluctuations (except for the concentration of Pb in the 10–20 cm soil layer of WS, which showed no significant difference between the measurement years). The general trend was that the concentration of Cd in each soil layer increased with the increased number of water level fluctuations, and other elements changed irregularly in all plots. The Cu concentration in each soil layer of WS increased first and then decreased, whereas a continual increase was shown at ZG. The Cr concentration decreased in each soil layer of WS, whereas that of ZG increased first and then decreased. The Pb concentration increased in each soil layer at the ZG site, whereas a decrease was found in the 0–10 cm soil layer at WS, and no clear changes were shown at 10–20 cm, and the 20–30 cm soil layer increased first and then decreased.

The average concentrations of heavy metals in the 0–30 cm soil layer in 2008 and 2015 was compared and the results show that the concentration of Cu decreased by 27.15%, Cd increased by 563.8%, Cr decreased 63.47%, and Pb did not change significantly. The concentrations of Cu, Cd, and Pb at ZG increased by 268.3, 2119 and 175.7% respectively, but Cr concentrations did not change significantly.

3.2 Spatial variation of soil heavy metal concentrations

3.2.1 Horizontal variation of soil heavy metal concentrations

The heavy metal concentrations in the same measurement year and the same soil layer were aggregated according to the site (Table 3), and the results show that before the reservoir water level fluctuations, the concentrations of Cd, Cu, and Pb in all layers were higher at WS than at ZG; however, after 4 cycles of fluctuation, the Cu concentrations in all soil layers and the Cd concentrations in the 0–10 cm soil layer had become lower at WS than at ZG.

The correlation analysis of the same heavy metal concentrations in different plots shows that there was a highly significant positive correlation between the Cd concentration within the same soil layer at WS and ZG. The Cu concentration in the 10–20 cm soil layer at WS was significantly positively correlated with the Cu concentration of in the 20–30 cm soil layer at ZG; however, other correlation analyses were not significant.

Table 2 The concentration and ANOVA of heavy metal elements in the same soil layer in different measurement years ($\text{mg} \cdot \text{kg}^{-1}$)

Soil layer/cm	Elements	Plots	Mean \pm Std. error			
			2008	2009	2012	2015
0–10	Cu	WS	22.07 \pm 0.48 b	27.70 \pm 2.30 b	47.09 \pm 2.03 a	13.11 \pm 2.51 c
		ZG	21.56 \pm 10.87 b	21.37 \pm 2.63 b	101.8 \pm 15.79 a	75.64 \pm 5.50 a
	Cd	WS	0.40 \pm 0.02 c	0.38 \pm 0.01 c	2.21 \pm 0.07 b	2.51 \pm 0.06 a
		ZG	0.18 \pm 0.04 b	0.46 \pm 0.06 b	3.24 \pm 0.16 a	3.70 \pm 0.39 a
	Cr	WS	92.11 \pm 0.56 b	101.7 \pm 2.29 a	63.81 \pm 1.15 c	33.69 \pm 1.09 d
		ZG	35.73 \pm 9.03 b	62.27 \pm 9.36 a	82.47 \pm 4.08 a	31.62 \pm 2.65 b
Pb	WS	15.06 \pm 0.47 a	15.25 \pm 1.03 a	15.99 \pm 0.55 a	12.30 \pm 0.41 b	
	ZG	4.95 \pm 0.85 b	3.99 \pm 0.47 b	12.41 \pm 1.60 a	13.71 \pm 2.63 a	
10–20	Cu	WS	20.16 \pm 0.61 bc	27.64 \pm 1.78 b	43.95 \pm 2.54 a	16.50 \pm 5.41 c
		ZG	21.05 \pm 11.04 b	19.79 \pm 4.30 b	106.7 \pm 9.88 a	86.09 \pm 15.13 a
	Cd	WS	0.39 \pm 0.03 c	0.40 \pm 0.01 c	2.29 \pm 0.08 b	2.70 \pm 0.05 a
		ZG	0.17 \pm 0.03 b	0.48 \pm 0.07 b	2.94 \pm 0.29 a	3.52 \pm 0.32 a
	Cr	WS	92.49 \pm 1.83 b	102.0 \pm 2.44 a	64.74 \pm 2.20 c	35.37 \pm 0.60 d
		ZG	36.72 \pm 9.34 b	63.44 \pm 10.96 a	75.38 \pm 5.35 a	31.28 \pm 2.54 b
Pb	WS	14.52 \pm 0.55 a	14.02 \pm 0.59 a	15.18 \pm 0.86 a	13.89 \pm 0.64 a	
	ZG	3.23 \pm 0.69 bc	1.72 \pm 0.32 c	6.52 \pm 1.55 ab	7.84 \pm 2.36 a	
20–30	Cu	WS	20.84 \pm 1.43 bc	27.54 \pm 1.47 b	45.36 \pm 2.26 a	16.36 \pm 4.10 c
		ZG	21.86 \pm 9.84 b	21.87 \pm 5.18 b	102.2 \pm 7.56 a	75.70 \pm 6.97 a
	Cd	WS	0.39 \pm 0.03 c	0.38 \pm 0.00 c	2.34 \pm 0.07 b	2.62 \pm 0.14 a
		ZG	0.14 \pm 0.03 c	0.49 \pm 0.09 c	2.54 \pm 0.21 b	3.42 \pm 0.40 a
	Cr	WS	103.6 \pm 1.30 a	100.8 \pm 1.91 a	66.53 \pm 0.50 b	36.19 \pm 1.58 c
		ZG	42.07 \pm 9.36 bc	52.01 \pm 8.34 ab	66.30 \pm 2.76 a	27.92 \pm 1.23 c
Pb	WS	11.93 \pm 0.29 b	14.84 \pm 0.67 a	15.88 \pm 0.83 a	13.23 \pm 0.61 b	
	ZG	1.68 \pm 0.61 b	1.34 \pm 0.32 b	9.25 \pm 1.48 a	5.57 \pm 2.54 a	

Different lowercase letters indicate significant differences between the different measurement years, at the level of 0.05

Table 3 Correlation analysis of soil heavy metal concentration in WS and ZG

Plots	ZG	Elements	Soil layers/cm	Cu			Cd			Cr			Pb		
				0–10	10–20	20–30	0–10	10–20	20–30	0–10	10–20	20–30	0–10	10–20	20–30
				WS	Cu	0–10	0.380	0.341	0.465	–	–	–	–	–	–
			10–20	0.361	0.267	0.499*	–	–	–	–	–	–	–	–	–
			20–30	0.327	0.469	0.481	–	–	–	–	–	–	–	–	–
	Cd	0–10				0.966**	0.951**	0.932**	–	–	–	–	–	–	–
		10–20				0.970**	0.967**	0.949**	–	–	–	–	–	–	–
		20–30				0.929**	0.928**	0.915**	–	–	–	–	–	–	–
	Cr	0–10							0.280	0.323	0.299	–	–	–	–
		10–20							0.256	0.298	0.257	–	–	–	–
		20–30							0.186	0.249	0.223	–	–	–	–
	Pb	0–10										–0.276	–0.265	0.014	–
		10–20										0.185	0.033	0.203	–
		20–30										0.128	0.061	0.288	–

* Correlation is significant at the 0.05 level

** Correlation is significant at the 0.01 level. Did not do correlation analysis. The same below

3.2.2 Variation of heavy metal concentrations in soil profile

The concentrations of Cu, Pb, Cd, and Cr in different soil layers of the same year were summarized (Fig. 1) and the results show that the concentration of some elements were obvious differences between soil layers in certain measurement years, but after 7 cycles of the reservoir water level fluctuations, only the Pb concentration in the soil at ZG showed a obvious difference, and there were no obvious differences in the concentrations of other elements between soil layers. The correlation analysis of the heavy metal concentrations between soil layers shows that there was a positive correlation between the same element concentration in different soil layers, and most correlations were significant (Table 4 and Table 5).

3.2.3 Soil heavy metals pollution degree and potential ecological risk assessment

The mean values of the heavy metal elements in different monitoring plots and different measurement years were used to calculate the geoaccumulation index (Table 6) and the potential ecological risk index (Table 7) of soil heavy metals. Before water level fluctuation, the soil Cu concentration at WS and ZG was classified as pollution-free. After 4 cycles of water level fluctuations, WS became pollution-free to moderately polluted and ZG became moderately polluted; however, after 7 cycles of water level fluctuations, WS became pollution-free once more. Before water level fluctuation, the Cd at WS was considered pollution-free to moderately polluted, and that at ZG was pollution-free. However, after 4 cycles of water level

Fig. 1 Analysis of soil heavy metals in different soil layer of WS and ZG

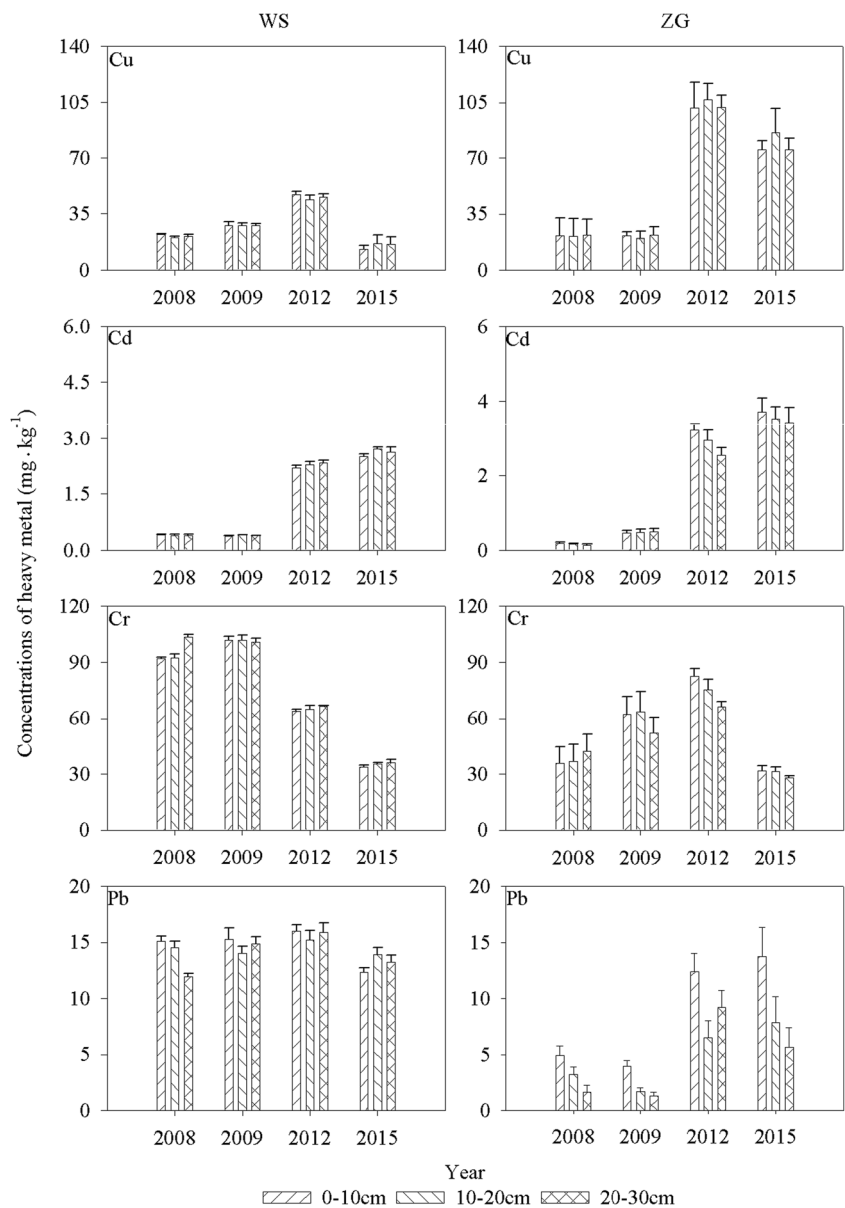


Table 4 Correlation analysis of heavy metal elements in different soil layers of WS

Elements	Soil layers/cm	Cu			Cd			Cr			Pb		
		0–10	10–20	20–30	0–10	10–20	20–30	0–10	10–20	20–30	0–10	10–20	20–30
Cu	0–10	1	0.825**	0.912**	–	–	–	–	–	–	–	–	–
	10–20		1	0.746**	–	–	–	–	–	–	–	–	–
	20–30			1	–	–	–	–	–	–	–	–	–
Cd	0–10				1	0.992**	0.983**	–	–	–	–	–	–
	10–20					1	0.978**	–	–	–	–	–	–
	20–30						1	–	–	–	–	–	–
Cr	0–10							1	0.991**	0.974**	–	–	–
	10–20								1	0.974**	–	–	–
	20–30									1	–	–	–
Pb	0–10										1	0.051	0.050
	10–20											1	0.311
	20–30												1

fluctuations Cd concentration at WS increased to strongly polluted and ZG increased to strongly and strongly to extremely polluted. In contrast, Cr and Pb remained at pollution-free levels before and after the impact of reservoir water level fluctuations. The results of the classification of soil heavy metals pollution degree were as follows: Cd > Cu > Cr > Pb.

Before water level fluctuation, the potential ecological risk of Cd in soil of WS was considerable, and that at ZG was low; however, after 4 cycles of water level fluctuations these all increased to very high. In contrast, Cu, Cr, and Pb remained at consistently low levels of potential ecological risk. The RI calculation for both sites indicated that before the formation of the WLFZ and after 1 cycle of water level fluctuation, the *RI* was low; however, after 4 cycles of water level fluctuations, the *RI* increased to high or very high.

4 Discussion

4.1 Effects of reservoir water level fluctuation on heavy metal concentrations in soil of the WLFZ

A growing body of literature has shown that the concentration of heavy metal elements in the WLFZ will change substantially with the increase of the number of fluctuation cycles of reservoir water level. In September 2008 and June 2009, soil samples were collected in 12 sites (from Chongqing to Zigui) in the WLFZ and heavy metals (Hg, As, Cr, Pb, Cu, Zn, Fe, and Mn) were determined, and the results show that the Hg, Cr, Pb, Cu, and Zn concentrations in soil after submergence were higher than those before submergence. Yang et al. (2011) studied the changes of

Table 5 Correlation analysis of heavy metal elements in different soil layers of ZG

Elements	Soil layers/cm	Cu			Cd			Cr			Pb		
		0–10	10–20	20–30	0–10	10–20	20–30	0–10	10–20	20–30	0–10	10–20	20–30
Cu	0–10	1	0.844**	0.906**	–	–	–	–	–	–	–	–	–
	10–20		1	0.937**	–	–	–	–	–	–	–	–	–
	20–30			1	–	–	–	–	–	–	–	–	–
Cd	0–10				1	0.979**	0.974**	–	–	–	–	–	–
	10–20					1	0.987**	–	–	–	–	–	–
	20–30						1	–	–	–	–	–	–
Cr	0–10							1	0.960**	0.910**	–	–	–
	10–20								1	0.948**	–	–	–
	20–30									1	–	–	–
Pb	0–10										1	0.571**	0.664**
	10–20											1	0.774**
	20–30												1

Table 6 Index of geoaccumulation and evaluation of pollution degree of heavy metal in different measurement years

Plots	Year	Cu		Cd		Cr		Pb	
		I_{geo}	Degree	I_{geo}	Degree	I_{geo}	Degree	I_{geo}	Degree
WS	2008	-0.83	PF	0.96	PFM	-0.29	PF	-1.37	PF
	2009	-0.44	PF	0.96	PFM	-0.21	PF	-1.28	PF
	2012	0.28	PFM	3.51	S	-0.85	PF	-1.19	PF
	2015	-1.29	PF	3.71	S	-1.74	PF	-1.45	PF
ZG	2008	-0.80	PF	-0.32	PF	-1.62	PF	-3.44	PF
	2009	-0.84	PF	1.26	MP	-0.97	PF	-3.93	PF
	2012	1.46	MP	3.86	S	-0.66	PF	-1.93	PF
	2015	1.08	MP	4.15	SE	-1.94	PF	-1.98	PF

soil heavy metal concentrations in the WLFZ of Xiaojiang River and found that the concentrations of Cu and Pb in 2010 were higher than that in 2009. Heavy metal concentrations tend to increase under a periodic alternate drying–wetting environment for a long time (Wang et al. 2013). This trend is consistent with the findings for before 2012 in the current study; however, this trend was not sustained. In our study, comparison of 2015 and 2012 monitoring results showed that most of the soil heavy metal concentrations showed a decreasing trend excluding Cr (Table 2). This change may be related to the difference in heavy metal element input and output because the fluctuation zone was subjected to a different frequency of water level fluctuation. At the beginning of the WLFZ formation, the vegetation of the WLFZ was seriously damaged by the alternation of the dry and wet soil, and the new vegetation was not fully developed (Zhu et al. 2015). Subsequently, the WLFZ was strongly affected by surge scour and erosion, and its soil environment was in a very unstable state. Moreover, the TGR upstream and reservoir area pollution prevention and control was still in the initial stage. As a result, short-term and long-term heavy metal monitoring results are different.

4.2 Effects of geographical location on heavy metal concentrations in soil of the WLFZ

There was a significant change in the geographical space of heavy metal concentrations in the soil of the WLFZ. The adsorption of heavy metal in soils is affected by soil pH, organic matter, and soil mineral composition; the adsorption of heavy metals in soils increases with the increase of pH, soil organic matter, and iron-manganese oxides (McBride et al. 1997; Naidu et al. 1997; Fontes and Gomes 2003; Covelo et al. 2004). In addition, changes in water level play an important role in the movement and fixation of heavy metals, with the change from aerobic to reductive conditions, the reductive dissolution of Fe and Mn hydroxides takes place, and it is also controlled by pH, salinity, organic matter content, and temperature (Rinklebe and Du Laing 2011). In reducing conditions, metals such as Fe and Mn and commonly associated pollutants such as, Cd, Cr, Mo, Ni, and Zn can be released from the solid phase to pore waters (Shaheen et al. 2014; Hindersmann and Mansfeldt 2014). Flood recession followed by the drying and aeration of floodplain soils reverses the processes of metal dissolution. In an oxic environment, Fe and Mn re-precipitate as oxides and scavenge heavy metals back to the solid state

Table 7 Potential ecological risk assessment of heavy metal in different measurement years

Plots	Year	Cu		Cd		Cr		Pb		RI	Degree
		E_i	Degree	E_i	Degree	E_i	Degree	E_i	Degree		
WS	2008	4.21	L	90.00	C	2.46	L	2.90	L	99.57	LR
	2009	5.53	L	90.00	C	2.60	L	3.08	L	101.21	LR
	2012	9.09	L	526.15	VH	1.67	L	3.29	L	540.20	CR
	2015	3.06	L	602.31	VH	0.90	L	2.75	L	609.02	VHR
ZG	2008	4.30	L	36.92	L	0.98	L	0.69	L	42.89	LR
	2009	4.20	L	110.77	C	1.53	L	0.49	L	116.99	LR
	2012	20.69	L	671.54	VH	1.90	L	1.97	L	696.10	VHR
	2015	15.83	L	819.23	VH	0.78	L	1.90	L	837.74	VHR

(Du Liang et al. 2009). The current study showed heavy metal concentrations changed from $WS > ZG$ to $WS < ZG$ after the impact of water level fluctuations. The study of soil pH, which is a key factor affecting the adsorption of heavy metal ions in soil, during the period from 2008 to 2015, indicated that the soil pH of the WS did not change significantly, but the soil pH in ZG increased from 5.03 (2008) to 7.59 (2015). Soil heavy metals immobilization appeared to be governed by the soil pH increase, which induced a greater retention of metals on soil particles (Rees et al. 2014), it is one of the reasons for the heavy metal concentrations of the soil in ZG. In addition, there are lateral transfer characteristics of heavy metals (Cr, Ni, Cu, Zn, As, Cd, and Pb) in the WLFZ of the TGR (Tang et al. 2015). Bing et al. (2016) found that the Cd concentrations in the riparian sediments increased towards the dam, and the high accumulation of heavy metals in the riparian sediments emerged between Fuling and Fengjie, Chongqing, China, and those in the submerged sediments occurred in areas close to the dam. Saha et al. (2002) argued that the first-order hydrolysis equilibrium constants (pK_1) of ions can reflect the size of competitive adsorption capacity for heavy metal ions by soil colloids. Hydroxyl metal ions that are formed by the hydrolysis of metal ions are more susceptible to soil adsorption than metal ions themselves; the size of the adsorption affinity decreases with the increase of the negative logarithm of pK_1 ($Cr (3.9) > Pb (7.8) > Cu (8.0) > Cd (10.1)$). Therefore, relative to other heavy metals, the stability of Cd is poor, and its migration is stronger. Meanwhile, Cd is typical anthropogenic metal affected by human activities (Zhang and Shan 2008; Sayadi and Sayyed 2011; Sayadi et al. 2015a, b). Cd has been associated to a greater extent with colloidal materials in surface runoff that can easily be transported in river flow (Wakida et al. 2008). In this study, the Cd showed clear horizontal transfer characteristics. Moreover, with the increase of the frequency of water level fluctuations the Cd concentrations at ZG (near the dam) were higher than those at WS (located in the upper reaches of Zigui). The closer to the dam, the wider the water surface, the slower the water flow, and thus the more conducive the conditions to the deposition of Cd carried in solution in the WLFZ (Friedl and Wüest 2002; Fremion et al. 2016).

4.3 Changes of heavy metal concentrations in different soil layers

It is generally believed that heavy metal can be enriched in the surface soil. Vertical metal displacement is favored by the frequent fluctuation of water-table levels and, in addition to the visible accumulation of the secondary Fe and Mn oxyhydroxides, may manifest in mineral breakdown and pseudomorphing or high levels of exchangeable and specifically adsorbed metals (Hudson-Edwards et al. 1998). Tang et al. (2015) showed that the heavy metal concentrations in the sediments of the WLFZ in the TGR was higher than that in

the 0–5 cm soil layer, and the 0–5 cm was higher than 5–10 cm soil layer. Wang et al. (2015) also found that heavy metal (As, Cu, Pb, and Zn) in sediments were significantly higher than those in soil. The results of the current study are not exactly the same, and most of the soil heavy metal elements do not indicate this phenomenon. The results of our study show that the content of Pb in 0–10 cm soil was higher than that of other soil layers after 7 cycles of water level fluctuation, and the other heavy metal concentrations were not obvious differences between soil layers. This may be related to soil properties in different study areas. The research of Tang et al. (2015) and Wang et al. (2015) focused on the backwater area of the reservoir, before the formation of the WLFZ. The areas assessed in the aforementioned studies were more densely farmed and thus the soil had a loose cultivation layer atop a hardened layer; however, in this study areas, the native vegetation were *Pinus massoniana* plantation and secondary shrub, the characteristics of these plots were rich in soil organic matter and showed clear forest soil characteristics. The exchange resistance of soil heavy metal in soil was relatively small, causing the distribution of heavy metals in soil layers to be more even.

4.4 Risk assessment of heavy metals in the WLFZ

In the present study, the I_{geo} showed that the Cd pollution was the most serious, with the rule of $Cd > Cu > Cr > Pb$. The RI showed that there was a very high potential ecological risk of Cd, whereas the potential ecological risk of Cu, Cr, and Pb was low. Ye et al. (2013) and Gao et al. (2014) evaluated the heavy metal pollution in soil and sediments of the WLFZ, which were located in the main road and tributaries of the Yangtze River, and the results showed that Cd was moderately polluted (soil samples were collected in 2009), similar to the evaluation of this study in 2009. But, in this study, we found that Cd contamination level change from pollution-free (all plots, 2008) to strong (WS, 2015) or extremely polluted (ZG, 2015) and Cu contamination level change from pollution-free (all plots, 2008) to moderately polluted (ZG, 2015). Previous results of the potential ecological risk index show that Cd had a very high potential ecological risk, whereas the potential ecological risk of Cu, Cr, and Pb was low (Fu et al. 2009; Lin et al. 2012; Gao et al., 2014, 2016b); in current study, we found that the potential ecological risk level of Cd change from considerable (WS, 2008) or low (ZG, 2008) to very high (all plots, 2015). Total heavy metal concentrations in soils are also directly related to soil parent material. However, mobility of human-originated metals is higher than pedogenic and lithospheric sources (Kaasalainen and Yli-Halla 2003; Singani and Ahmadi 2012). Fertilizers, pesticides and mulch are important agricultural inputs for agricultural production (Satarug et al. 2003; Zhang and Zhang 2007; Wu et al. 2009; Zhang et al. 2011). Nevertheless, the long-term

excessive application has resulted in the heavy metal contamination of soils (Su et al. 2014). The vast majority of pesticides are organic compounds, and a few are organic-inorganic compound or pure mineral, and some pesticides contain Hg, As, Cu, Zn, and other heavy metals (Arao et al. 2010). Heavy metals are brought to the soil by irrigative sewage and are fixed in the soil in different ways. It causes heavy metals (Cd, Pb, Cr, Cu, etc.) to continually accumulate in the soil year by year (Su et al. 2014). Soil Cd pollution affects the growth and development of plants, leading to the reduction of plant growth quality. Moreover, it may also pass through the food chain into the human body, thus posing a serious threat to food safety and human health. In the TGR aquatic environment, ecological risk prevention and control of Cd pollution of the soil in the WLFZ should be considered in detail in future research.

5 Conclusions

There is an obvious temporal and spatial variation of heavy metal concentrations for WLFZ of TGR. Cd concentration increased with the increase in the number of reservoir water level fluctuations, whereas the concentrations of Cu, Cr, and Pb varied with the monitoring site. Heavy metal concentrations changed from $WS > ZG$ to $WS < ZG$ after the impact of water level fluctuations. After 7 cycles of water level fluctuation, the distribution of heavy metals in different soil layers tends to be uniform (except Pb). Cd pollution is more serious, and there is a strong potential ecological risk. The hydrological characteristics of the reservoir, the geographical location of the water-level-fluctuating zone, and the human disturbance are the main driving factors of the temporal and spatial changes of the heavy metal elements in the soil.

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