





Release flux of heavy metals from river sediments at different flow rates

Jiangang Lu ^{a,b}, Haisheng Cai ^{a,c,*}, Xueling Zhang ^a and Yanmei Fu ^d

^a Key Laboratory of Po-Yang Lake Watershed Agricultural Resources and Ecology of Jiangxi Province/Development Research Center of Selenium-rich Agricultural Industry, Jiangxi Agricultural University, Nanchang 330045, China

^b Jiangxi Water Resources Institute, Nanchang 330013, China

^c Jiangxi Tourism and Commerce Vocational College, Nanchang 330100, China

^d Yuzhang Normal University, Nanchang 330103, China

*Corresponding author. E-mail: caihs1972@163.com

 JL, 0000-0001-7628-8918; HC, 0000-0002-6707-7999; XZ, 0000-0002-8438-187X; YF, 0000-0001-7063-6571

ABSTRACT

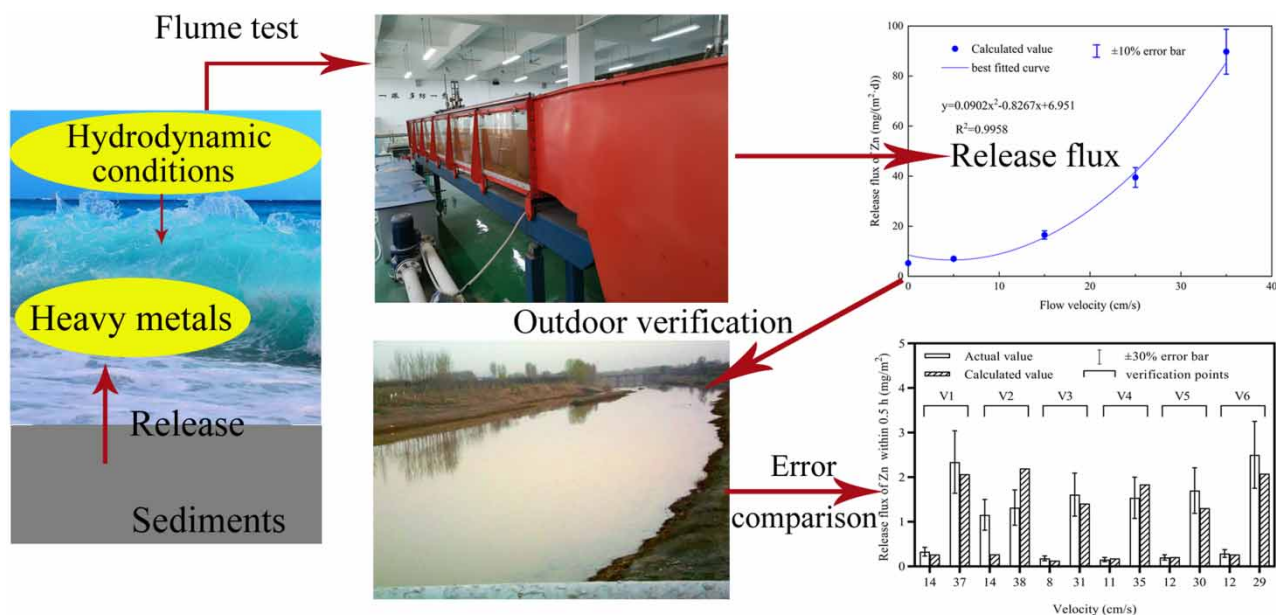
This paper simulates sediment motion under different hydrodynamic conditions, aiming to investigate the release flux of heavy metals in river sediments. During the lab experiments, carried out in a circular rectangular flume device, water velocity in the flume was altered by controlling the gate switch, and the flow rate was controlled from 0 to 1 m/s. Sediment from the Le'an River and chlorine-removed tap water were used as experimental sediment and water, respectively. Through analyses of Cu, Zn, Cd, and Pb concentration in water at different flow rates, the relationship between the release flux (y) of Cu, Zn, Cd, and Pb and the flow rate (x) was established with a fitting error of less than 15%. In order to judge the reliability of the conclusions, experimental results were verified outdoors. The results showed when the sediment particle size is between 0 and 250 μm , within 1 hour, a quadratic polynomial correlation between the release flux of Cu, Cd, and Pb from river sediments and water velocity when the water pH is 5–9 and the flow rate is 0–65 cm/s; when the water pH is 5–9, the flow rate is 0–35 cm/s, the release flux of Zn from river sediments was shown to have a quadratic polynomial relationship with water velocity. The error between the calculated and measured values of heavy metals released from sediment in the Le'an River were within 5–30%. Our results can provide a theoretical reference for the control and treatment of heavy metal pollution in rivers and further improve corresponding water quality models.

Key words: flow rate, heavy metals, Le'an River, release flux, sediment

HIGHLIGHTS

- The heavy metal release flux from sediments in river follows a quadratic polynomial relationship with the flow rate.
- The release of heavy metals in the river sediments at different flow rates within 1 h is greater than the adsorption.
- Flow rate and water pH are two important factors affecting the release of heavy metals from sediments.

GRAPHICAL ABSTRACT



1. INTRODUCTION

Sediments, which are sources and sinks of pollutants, absorb large amounts of nutrient salts and heavy metals (Zhu *et al.* 2017). The heavy metal content of sediments generally reflects the level of heavy metal pollution in water. Changes in environmental conditions trigger the release of heavy metals, which will directly or indirectly influence the water quality through biological enrichment (Byrne *et al.* 2012). Some scholars study the release of heavy metals in constructed wetlands with zeolites as fillers, and found that the upper layer (0–30 cm) of the vertical flow constructed wetlands (VFCWs) was the most effective area for heavy metal removal, showed that significant amounts of Zn and Cu were released from the VFCWs while Pb and Cr rarely escaped, and Zn and Cu existed mainly in the exchangeable state (Zhou *et al.* 2019). Some scholars study the potential impact of indigenous bacterial processes on the release of heavy metals from sediment. It was found that drastic increases in metal (Zn, Cd) release were observed under acidic conditions (Lors *et al.* 2004). Several factors affect the release of heavy metals from sediments; for example, the rise of P (Phosphorus) content in the overlying water will inhibit the release of heavy metals from sediments (Chen *et al.* 2017; Liu *et al.* 2019). Moreover, high salinity and high nitrogen levels can enhance the bioavailability of heavy metals in sediments (Hong *et al.* 2011), or the activity of heavy metals in sediments is relatively high in anoxic environments (Acosta *et al.* 2011). Heavy metal release is also affected by sediment redox potential, sulfide and calcium content (Martin-Torre *et al.* 2015; Zhou *et al.* 2020), or the inherent characteristics of the sediment (Xu *et al.* 2015; Huang *et al.* 2017).

The actions of wind and waves cause an intense exchange of substances between sediments and water bodies (Zhu *et al.* 2013), and the impact on heavy metal release from sediments cannot be ignored (Wang *et al.* 2020). The released amount is related to the resuspension and deposition of sediments (Argese *et al.* 1997). Wind and waves can effectively activate the sediment layer and lead to the adsorption and release of heavy metals in the sediments (Huang *et al.* 2016; Tang *et al.* 2020), thereby resulting in the continuous exchange of heavy metals in the sediments at the water-soil interface (Zhu *et al.* 2017). Many studies have targeted the release of heavy metals from sediments under different hydrodynamic conditions. However, most of them are focused on factors affecting the release of heavy metals from sediments, the different forms and levels of heavy metals in water after release, or the exploration of the migration and transformation rules of the different forms of heavy metals (Zheng *et al.* 2013; Lu *et al.* 2019). Few reports exist on the quantitative analysis of heavy metal release from sediments at different flow rates (Wen *et al.* 2021; Yan *et al.* 2021), and even fewer studies have been conducted on heavy metal release from river sediments (Liu *et al.* 2018a; Cai *et al.* 2021).

The Le'an River is the secondary tributary of Poyang Lake, the largest freshwater lake in China, and the primary tributary of the Raohe River (Liang *et al.* 2020). The geographical location of Le'an River is shown in Figure 1. The total length of the river is 280 km with a drainage basin of 8,521 km². Studies have shown that the Le'an River is the main source of heavy metals in Poyang Lake (He *et al.* 1998; Niu *et al.* 2017; Zhang *et al.* 2018). The largest open-pit copper mine in Asia (Dexing copper mine) is located near the Dawu River, a tributary of the Le'an River, with serious levels of heavy metal pollution (especially heavy metals such as Cu and Zn, followed by Cd and Pb) (Tao *et al.* 2014). The total length of the Dawu River is 14.3 km. A large amount of acidic and alkaline wastewater from the Dexing Copper Mine is discharged annually into the Dawu River (He *et al.* 1998), which carries it into the Le'an River. This results in serious water pollution and the river is becoming almost a wastewater ditch. The acidic wastewater released from the mine has a pH of 2.5–3.5, and the pH of the alkaline wastewater is about 10.5 (Jiang *et al.* 2018). The acidic space between the 'ditch' is about 2–3.5 and the alkaline space between the 'ditch' is about 10–11 (Zhang *et al.* 2018).

This article simulates the sediment movement dynamics by an indoor sink device and analyzes heavy metal levels in the overlying water at different velocities. Through the laws of mass conservation, we also explored the dynamics of heavy metal release from river sediments at different flow rates. A field test was also conducted to verify the findings of indoor experiments and draw conclusions for the Le'an River in order to provide theoretical reference for further studies on the mechanisms of the internal release of heavy metals from sediment in natural bodies of flowing water.

2. TEST MATERIALS AND METHODS

2.1. Indoor test device

The water tank device (produced by Hohai University, China) is shown in Figure 1. It consists of a water tank (4 m³ volume), water pump, flow meter, water level meter and rectangular glass flume (3.8 m × 0.8 m × 1 m). The pump has a maximum flow rate of 100 m³/h. The gate valve is mainly used to control the flow, and the flow meter is installed behind the pump. In the test, the cascade tailgate and pump gate of the flume outlet are adjusted to ensure a stable water level and flow rate in the flume.

We use chlorine-removed tap water as testing water; 10 cm deep sediment columns from the lower reaches of the Le'an River are collected by a column sediment sampler as the test sediments. The collected sediments are laid on the bottom of a sink, lightly flattened and size-controlled to retain 10 cm thickness. After the samples were left to stand for 24 h, the water tank was filled with water and the device started. The water depth was controlled at 80 cm, and the water was set still for 24 h. The water velocity of the Le'an River is between 0 and 1 m/s all year round (Chen *et al.* 2016), and therefore we set the flow rate to 0, 5, 15, 25, 35, 45, 65, 85 and 100 cm/s by controlling the gate. The velocity was gradually increased from 0 to the highest test value. Each flow rate setting was maintained for 1 h to ensure that water samples in the device were evenly mixed and all water flowed through the flume at least once.

2.2. Outdoor test

In order to enable the indoor test conclusion to be verified outdoors, members of the research team conducted a field test in March, April and May 2019, at the upper reaches of the Le'an River (V1), midstream (V5), downstream (V6), and 1 km, 10 km, 20 km after the tributary of the Dawu River converges (V2, V3, V4). During the test period, all the six verification points (V1, V2, V3, V4, V5, and V6) experienced four different wind speed conditions: strong, moderate, light, and calm (without flow velocity). The specific locations of the verification points are shown in Figure 1. Water samples were collected at different stages of the test at water depths typical for the middle of the water body at the sampling site, with the velocity and depth of water recorded at the time of sampling. During the test, water depths corresponding to the six verification points (V1, V2, V3, V4, V5 and V6) were 1.56 m, 1.65 m, 1.79 m, 1.54 m, 2.13 m, and 0.96 m, respectively.

2.3. Sample collection and analysis

2.3.1. Sample collection

During the indoor test, 50 mL of middle water sample was collected from the middle of the tank every ten minutes at each flow rate. During the outdoor test, 500 ml of water samples were collected under different flow rates. All collected water samples were adjusted to pH < 2 by the addition of HNO₃ (Maoming Xiongda Chemical Co., Ltd, China) and stored at –20 °C in the refrigerator (NR-FC50VT, Panasonic, Japan).

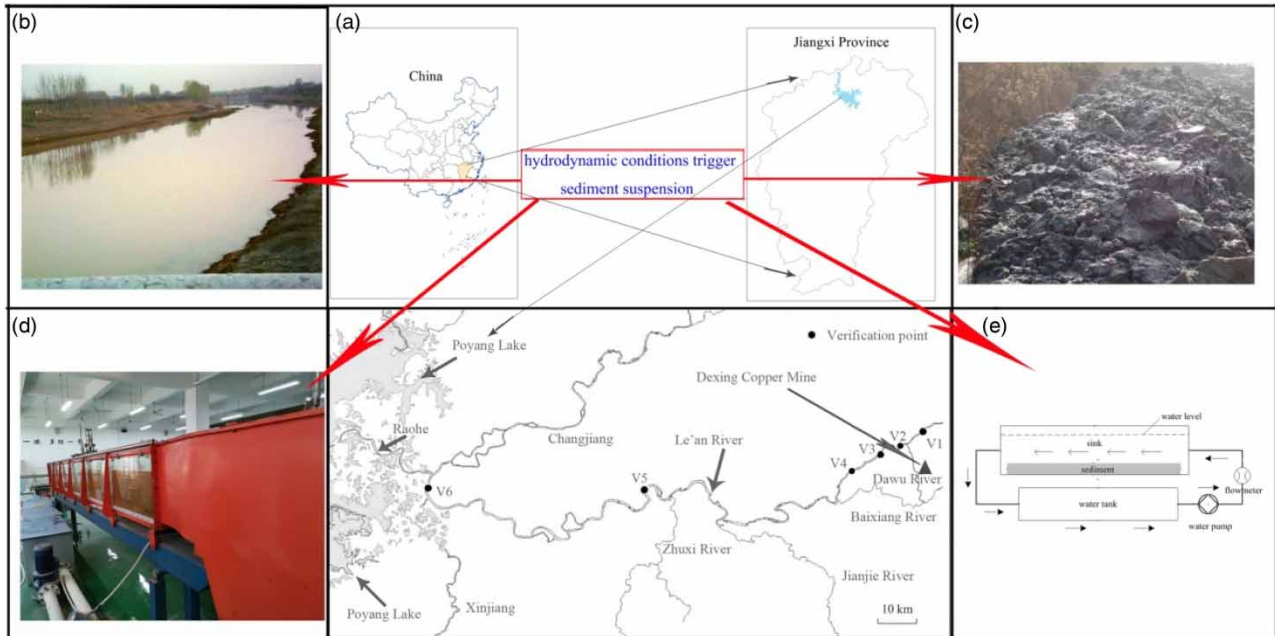


Figure 1 | (a) Location of Le’an River. (b) A picture of Le’an River. (c) A picture exhibiting the sediment of Le’an River. (d) A picture of the dynamic experiment. (e) Schematic diagram of the experimental device and primary equipment are indicated.

2.3.2. Analysis items and methods

The Cu, Zn, Cd and Pb, as representative heavy metals in the Le’an River (Tao *et al.* 2014), were selected as research objects. The applied analytical methods and detection limits of each element are shown in Table 1 (China 2002 (fourth edition) (in Chinese); Yang *et al.* 2017; Liu *et al.* 2018b; Yan *et al.* 2019; Hu *et al.* 2020). The concentration measured in this test is the concentration of heavy metals in raw water. In order to reduce the error, each water sample was measured three times in parallel, and the average value was taken as the final result. The water quality in the lab and at each verification point of the Le’an River is shown in Table 2 (Tao *et al.* 2014; Jiang *et al.* 2018; Zhang *et al.* 2018). The sediment characteristics of Le’an River are presented in Table 3.

2.3.3. Calculation method

The present work intends to deduce the release of heavy metals from sediments from the concentration of heavy metals in water according to the law of conservation of mass. The calculation method is as follows (Yiping *et al.* 2004):

$$r = [1,000\bar{V}(C_{\bar{n}} - C_0) + \sum_{j=1}^n 1,000V_j(C_{j-1} - C_n)]/(A.t) \tag{1}$$

In the formula, *r* denotes heavy metal release flux (mg/[m².d]); \bar{V} indicates the total volume of water used in the test (m³); *C_n* is heavy metal concentration in water at the nth sampling (mg/L); *C₀* indicates initial heavy metal concentration in water

Table 1 | Analytical methods and detection limits of heavy metals in water samples

Index	Analytical method	Detection limit (mg/L)	Demanded detection limit (mg/L)
Cu	Extraction-flame atomic absorption spectrometry	0.0002	0.05
Zn	Extraction-flame atomic absorption spectroscopy	0.0005	0.05
Cd	Flameless atomic absorption spectroscopy	0.00001	0.05
Pb	Inductively coupled plasma atomic emission spectrometry	0.0002	0.01

Table 2 | The water quality of lab water and each verification point of Le'an River

Index	Lab water	V1	V2	V3	V4	V5	V6
pH	6.5–7.5	6–8	2.5–4	3–5	5–7	5–9	6–9
Cu ($\mu\text{g/L}$)	7.31	41–85	162–196	157–194	134–172	126–157	62–120
Zn ($\mu\text{g/L}$)	2.57	16–24	68–78	58–62	40–46	33–41	24–30
Cd ($\mu\text{g/L}$)	0.11	0.67–1.3	1.74–1.88	1.52–1.82	1.21–1.38	0.82–1.05	1–1.15
Pb ($\mu\text{g/L}$)	1.86	0.7–1.8	10.1–10.6	8.9–10.6	6.5–8.2	4.3–5.5	1.5–3

Table 3 | Basic parameters of sediment in the Le'an River

Index	Heavy metals content/($\text{mg}\cdot\text{kg}^{-1}$)				
	Cu	Zn	Cd	Pb	Moisture (%)
Le'an River	10.12–764.3	49.24–557.82	0.97–3.85	3.54–15.86	(95 \pm 0.4)%

(mg/L); V_i is sample volume in every sampling (m^3); C_{j-1} denotes heavy metal concentration in water during $j - 1$ sampling (mg/L); C_n is the heavy metal concentration after raw water addition; t indicates heavy metal release time (d); and A is the surface area of sediment in the flume (m^2).

3. RESULTS AND ANALYSIS

3.1. Process of flume test and analysis

3.1.1. Flume test

According to the principles of sediment movement, sediments can be divided into three states: ready to move but still, slightly moving and generally moving (Guo-ren 1999). Heavy metal levels in water vary under different hydrodynamic conditions, and there are some changes in the adsorption and release of the materials in sediments. The characteristics of sediment movement in the flume test in this paper correspond with the sediment movement principles.

Up to a flow rate of about 25 cm/s, the sediment does not move. When the flow velocity of water increases from 0, the shear force on the silt surface gradually rises, and the soil-water interface transforms from the initial relative static state to a thin layer of dilute suspension on the mud surface. At this time, the silt is in a 'ready to move but still' state.

With a further increase in the flow rate, the shear force of the sludge continues to rise. The turbulent action of water flow gradually strengthens, and some small pieces of silt in the flume are washed up and suspended on the mud surface. At this stage, also called the 'slightly moving' state, the water body becomes turbid and the corresponding water velocity is between 25 and 50 cm/s.

When the flow rate increases to about 60–70 cm/s, the turbulent action of water gets more intense, the bottom silt is lifted in pieces, and water becomes turbid in a short time, especially when a certain part of the silt is lifted; the remaining sludge in this area will move rapidly. The stage is called the 'generally moving' state.

Once the flow velocity exceeds 70 cm/s, the state of water flow movement is similar to 'generally moving', and a large amount of sediment suspended in water is observed at this time.

3.1.2. Changes in the concentration of heavy metals after maintaining different flow rates for one hour

After maintaining each flow rates for one hour, the changes in heavy metal levels at different flow rates in the experimental sink are shown in Figure 2. As seen in the figure, with the flow rate rising from 0 to 65 cm/s, the concentration of Cu, Pb and Cd in water gradually increase, and their levels show little change when the flow rate continues to increase further. When the flow rate is less than 25 cm/s, the concentration of each heavy metal elevates slightly, which is attributed to the fact that the sediment has not yet been suspended in large quantities and is only slightly disturbed. Only heavy metals in interstitial water are released at this time. As the flow rate continues to rise, the sediment changes from 'ready to move but still' to 'slightly moving', and the concentration of each substance elevates significantly. This is because heavy metals adsorbed between

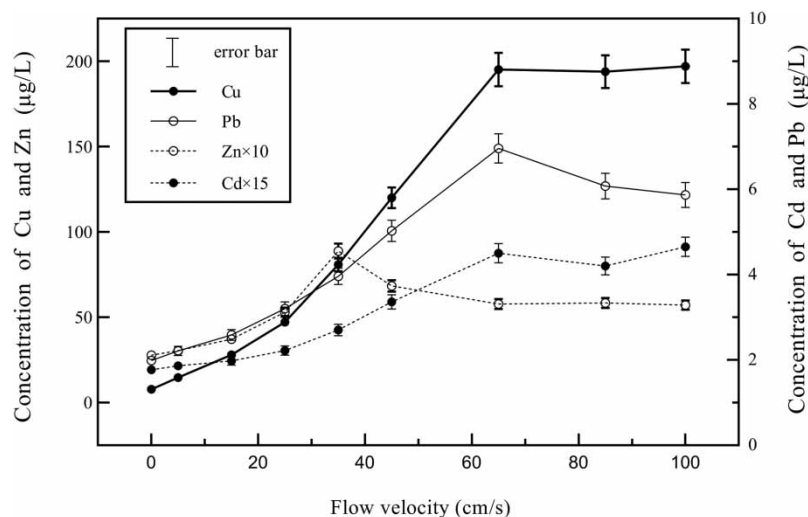


Figure 2 | Changes in heavy metal concentrations at different flow rates. Zn \times 10 represents 10 times the concentration of Zn, Cd \times 15 represents 15 times the concentration of Cd.

the sludge particles are gradually suspended into the overlying water as silt on the surface of the sediment is washed up. Also, heavy metals in the interstitial water are released in large quantities at the same time. When the flow rate reaches 60–70 cm/s, the bottom sludge is lifted in pieces and reaches the ‘generally moving’ state, with the concentration of each substance increased to the maximum of its range. At a flow rate of 65 cm/s, the concentrations of Cu, Cd and Pb in water are observed as 195.29 $\mu\text{g/L}$, 0.3 $\mu\text{g/L}$, and 6.96 $\mu\text{g/L}$, respectively, which are 24.85, 2.54, and 3.5 times, respectively, of those in stagnant water. The experiments conducted in this study demonstrate that the characteristics of silt motion observed at 65 cm/s change little with the continued increase of flow rate, and thereby the level of each substance tends to stabilize.

Interestingly, the dynamics of Zn concentration change are not consistent with that of the other three heavy metals. As the flow rate rises from 0 to 35 cm/s, the level of Zn in water gradually increases, but this trend reverses when the flow rate continues to increase to 65 cm/s; we tested different forms of Zn in the water and found that the decrease is primarily caused by the reduced amount of dissolved Zn. The further increase of flow rate does not have a significant effect on Zn concentration.

3.1.3. Changes in the concentration of heavy metals with time at different flow rates

Changes in the concentration of heavy metals with time at different flow rates are shown in Figure 3. The results show that when the flow rate is 65 cm/s, within 1 h, the concentration of Zn in water gradually decreases with time. In other cases, the concentration of heavy metals in water gradually increased with time. Further linear fitting showed that the fitting effect was good, and the R^2 of the fitting straight line were all above 0.95.

3.1.4. Relationship between release flux and flow rate

The release flux of each substance is calculated according to Formula (1). Results show that a rising flow rate results in an increasingly greater release flux rate of Cu, Zn, Cd, and Pb from sediment. At a flow rate of 35 cm/s, the release flux of Zn is 17.1 times that in stagnant water. When the flow rate is 65 cm/s, the release flux of Cu, Cd and Pb are 136.9, 14.6, and 14.8 times, respectively, those in stagnant water (Table 4). Among these heavy metals, the release flux of Cu shows the most significant hike, which is attributed to the lower background concentration of Cu in the water used in the experiment (Table 2), and the higher Cu pollution level in sediments (Table 3).

According to the relationship established between the released flux and flow velocity above, quadratic fitting was performed, with results shown in Figure 4. It is concluded that, within a specific flow rate range, the release fluxes of the four heavy metals in sediment all rise with increased flow rate (The flow rate range corresponding to the release flux of Cu, Cd, and Pb is 0–65 cm/s, and the flow rate range corresponding to the release flux of Zn is 0–35 cm/s.). When the hydrodynamic strength declines, the suspended sediment sinks again, and will not continue to float or complete the release of heavy metals until the next hydrodynamic strength surge occurs.

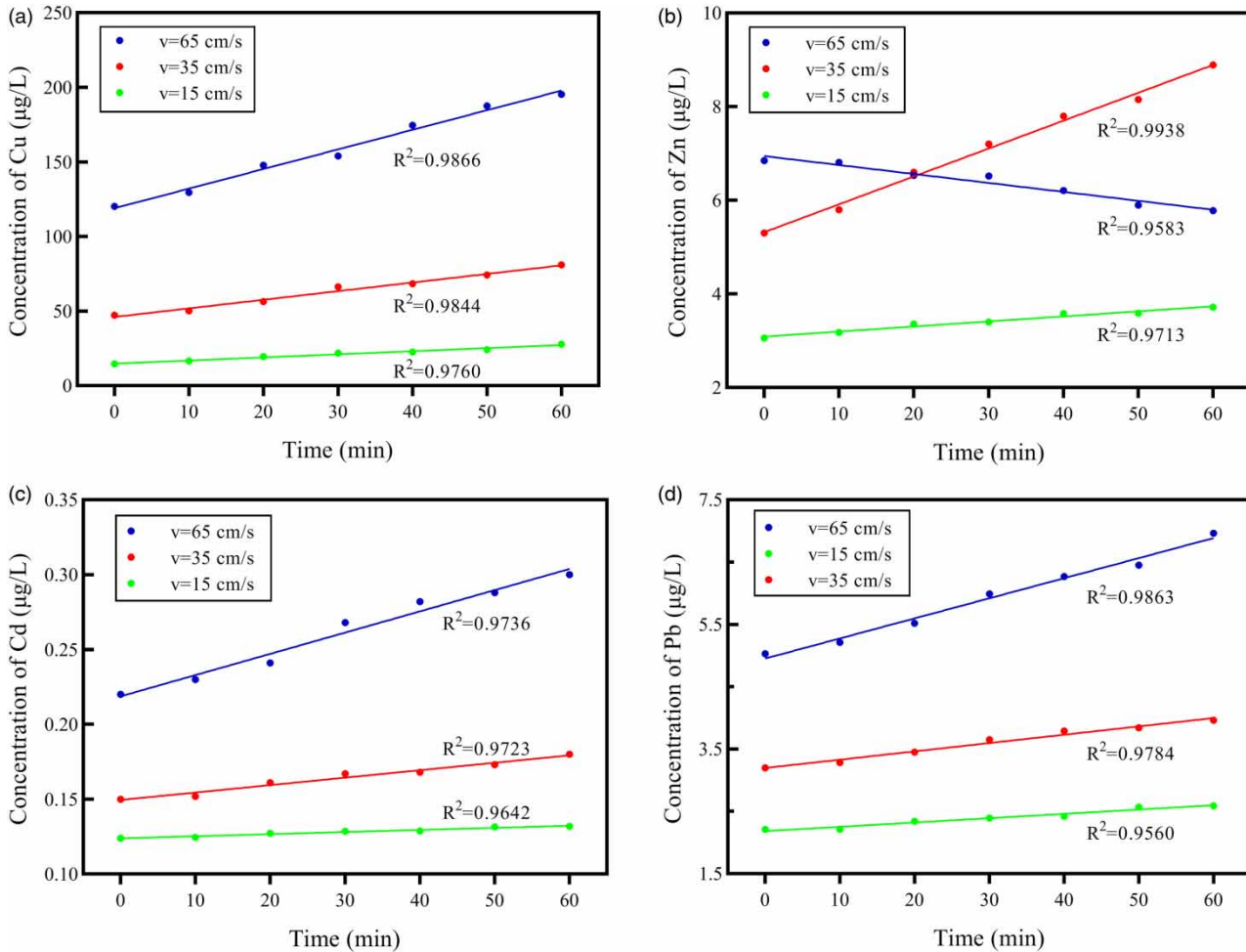


Figure 3 | Changes in the concentration of heavy metals with time at different flow rates.

Table 4 | Content and release rate of heavy metals at different water flow rates

Index	Flow velocity (cm/s)	Concentration (µg/L)	Ratio	Release flux (mg/(m ² .d))	Ratio
Cu	0	7.86	21.37	13.75	136.9
	65	195.26		1,878.08	
Cd	0	0.12	2.76	0.13	14.6
	55	0.3		1.9	
Pb	0	1.99	3.82	3.25	14.8
	65	6.96		48.5	
Zn	0	2.78	1.93	5.25	17.1
	35	8.89		89.75	

3.2. Results of outdoor test

A comparison of the measured and calculated values of heavy metal release flux from sediments within a period of 0.5 h at different water flow rates is shown in Figure 4 (the corresponding flow velocities at each verification point in the outdoor test are selected according to the characteristics of sediment movement. The velocities at each point are 0–25, 25–50, 60–70 cm/s.). In order to determine the heavy metal content in water within this period, calculated values are based on the fitting formula, and the measured values (actual value) are calculated from experimental data, not from fitting formulae.

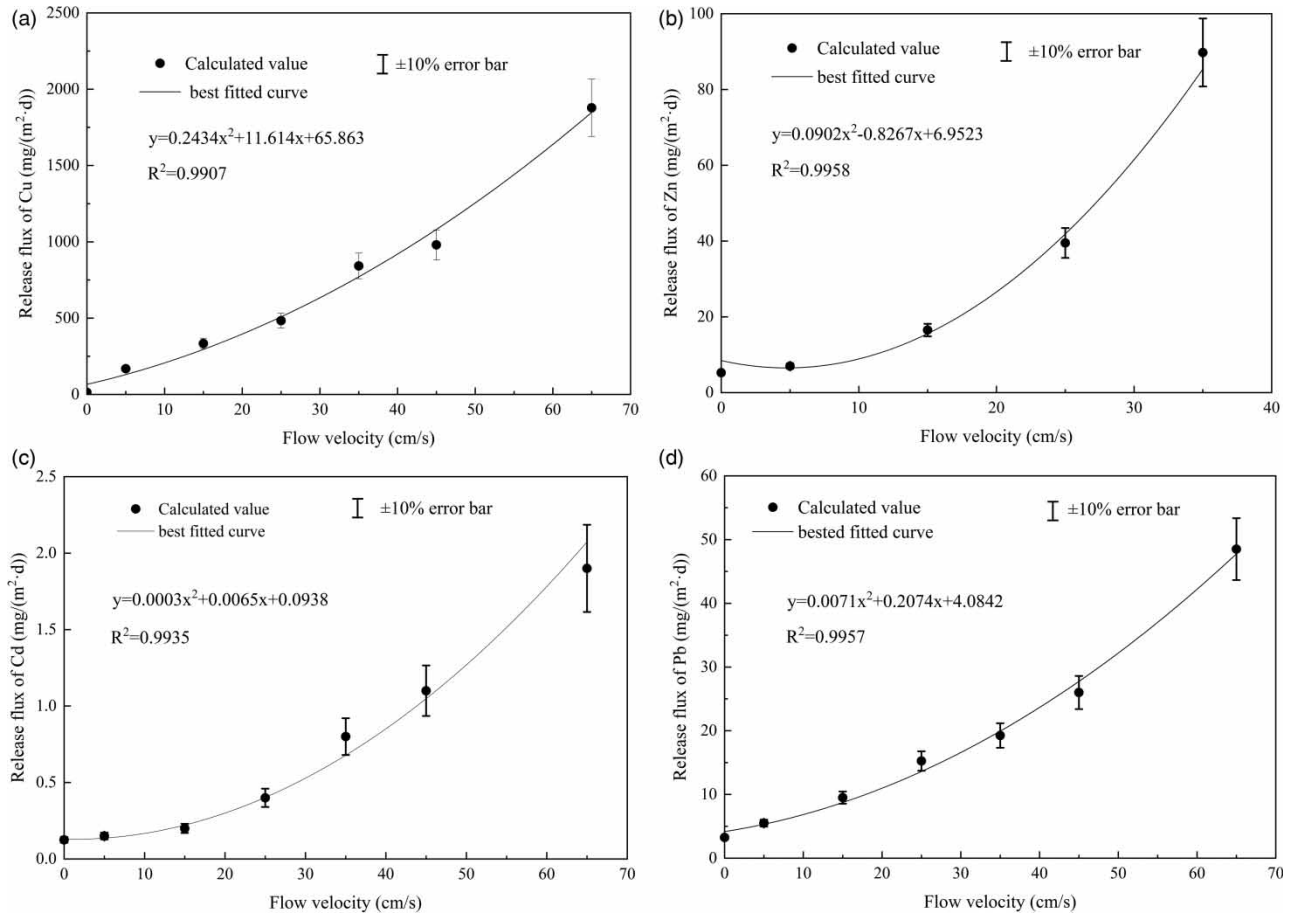


Figure 4 | Response curve of relationship between release flux of heavy metals and flow velocity.

According to Figure 5, the error between the calculated value and the measured value of each verification point (except V2 and V3) is less than 30%, which indicates that the relationship between the release flux and the flow rate of each heavy metal, as determined in our indoor experiments, can be considered essentially adequate for the prediction of actual heavy metal flux events.

4. DISCUSSION

The absorption and release of heavy metals in the water-sediment interface depend on multiple factors, and there is a close relationship between the absorption and release (Huang *et al.* 2015). The factors affecting the absorption of heavy metals by sediments include temperature, pH, dissolved oxygen, hydrodynamic conditions, ion concentration, content of organic matter, etc (Wen & Allen 1999; Soltan *et al.* 2001; Li *et al.* 2009). Among them, pH and ion concentration have a greater impact on the absorption capacity (Soltan *et al.* 2001; He *et al.* 2009). However, the absorption kinetics of sediments generally reach absorption equilibrium within one or two hours (He *et al.* 2015). This paper did not carry out a test on the absorption effect of heavy metals in sediments under different flow rates, because the release amount discussed in this paper is the amount released under the combined action of absorption-release. It also shows that the total amount of heavy metals released from sediment under different flow rates is larger than that of absorption in 1 hour.

In this experiment, each hydrodynamic condition was only maintained for one hours. In the field verification experiment, the content of heavy metal in water was measured for only 0.5 hour. In a natural state, there is a dynamic absorption-release equilibrium at the water-sediment interface, and it is reported that it takes at least two days to reach equilibrium under hydrodynamic conditions (He *et al.* 2015). However, we did not test the time to reach equilibrium in our experiment settings. As heavy metals in water include dissolved state and suspended state (Coynel *et al.* 2007; Yu *et al.* 2019), studies have shown that

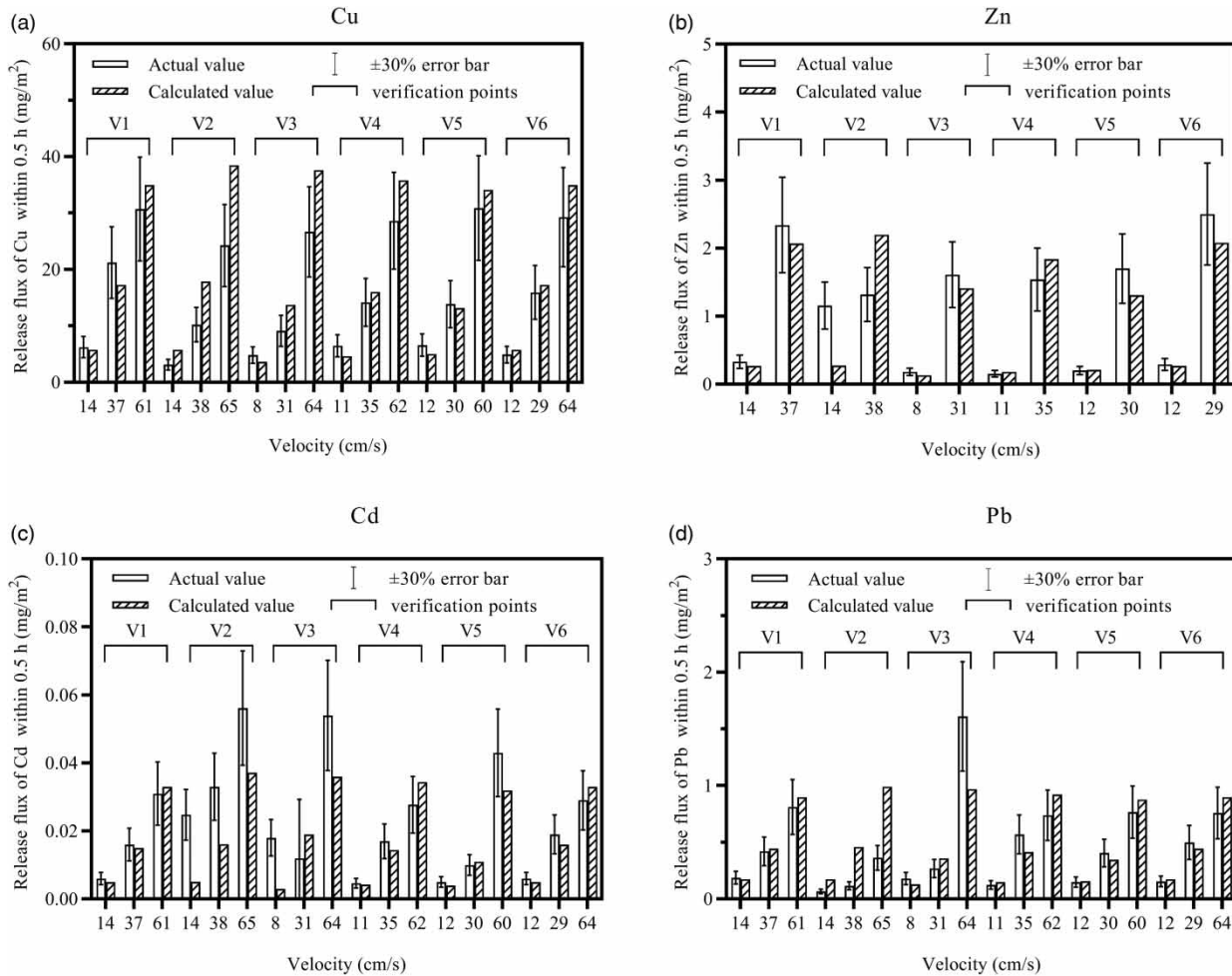


Figure 5 | Comparison of calculation results and measured values of heavy metal release flux within 0.5 h under different flow rates.

Cu, Zn, and Pb are primarily released in dissolved state and Cd is release in suspended state under hydrodynamic conditions (Argese *et al.* 1997). In this paper, since we measured the total heavy metal concentration in raw water, the migration and transformation characteristics of different forms of heavy metals in water were not analyzed. Therefore, our next step was to study the time required to reach release equilibrium and the morphology migration and transformation of heavy metals.

In general, the release flux of heavy metals from river sediments is closely related to flow turbulence (Zhu *et al.* 2014), flow turbulence affects the movement of pollutants in the vertical direction (Cheng *et al.* 2020). In the laboratory test, we collected the surface, middle and bottom water for detection, the results showing that the concentration of heavy metals in water is different in the vertical direction, the concentration of heavy metals in the bottom water is slightly higher, followed by the middle layer, and the lowest in the surface layer, but the difference is not significant; further analysis showed that the concentration of heavy metals in the middle water was roughly the average of the concentration of heavy metals in the surface and bottom layers. Some scholars take Taihu Lake as the research object (the water depth is 4.8 m), found that the concentration of heavy metals in the vertical direction changes exponentially (Zheng *et al.* 2013). The reason for the difference may be that both the laboratory test water depth and the outdoor test water depth of this research are relatively shallow (0.7 m and 2.1 m, respectively), and the heavy metals in water are quickly mixed completely in the vertical direction. Therefore, both the laboratory and outdoor experiments of this research collect intermediate water bodies for testing.

According to formula 1, the release flux is proportional to the increased concentration of heavy metals before and after the test (The impact of the small amount of water collected during testing is almost negligible). Figure 3 shows that the concentration of heavy metals in water at different flow rates within 1 hour increases linearly with time (When the flow rate is 65 cm/s, the concentration of Zn in water is excluded), this is consistent with the conclusions of Kyung-Yup Hwang

(Hwang *et al.* 2011). We did not analyze the concentration of heavy metals in water after 1 h, because the variation concentration of heavy metals in water after 1 h at different flow rates fluctuates greatly, and R^2 is small after linear fitting, indicating that the conclusion of the release flux of heavy metals in this paper is applicable within 1 h.

At present, similar tank experiments in academia are dominated by annular tanks (Huang *et al.* 2012a, 2012b; Mahdavi *et al.* 2013; Zheng *et al.* 2013; Sun *et al.* 2015; Xu *et al.* 2015), and pollutant release flux is calculated mainly by calculating the shear stress of the soil and water interface (Wang *et al.* 2011, 2016). In this paper, a rectangular flume was selected for the experiment, most scholars perform an annular flume simulation of lakes or reservoirs. While the research object in this paper is a river, the hydraulic characteristics of the rectangular flume are closer to reality, and the use of flow velocity to characterize the release flux in this paper is easier to understand and measure. In general, the indoor test should have a similar hydrodynamic condition to the outdoor test. In this paper, the outdoor test mainly affects the water velocity through the wind speed, while it is difficult to simulate different wind speed indoors, so different flow rates are directly set to achieve the purpose of the test. Wind speed determines the degree of wind impact, not only affecting horizontal pollutants transport, but also regulating vertical dynamics (Wang *et al.* 2020). However, the results of the indoor test in this paper are less error when applied to the outdoor test. We analyze that this is because the outdoor verification test select the river as research object and the water depth is relatively shallow, so the influence brought by turbulence is less than the flow velocity when the river water depth of is less than 2.1 m (The water depth of the six points in the field verification test in this paper is almost all less than 2.1 m.). Therefore, for the deep water reservoirs and lakes with obvious turbulence phenomenon, the applicability of this conclusion remains to be further studied.

In this paper, indoor rectangular flume experiments were carried out to reveal the dynamics of heavy metal release from river sediments under different flow rates. The relationship between heavy metal release flux and water velocity proved quadratic polynomial, indicating that the flow rates play an essential role in the circulation of heavy metals in rivers. It also shows that attempting to reduce heavy metal pollution in a river, the internal release of heavy metals from sediments should also be controlled in addition to addressing external pollution. These findings are similar to the conclusions of Reddy (Reddy *et al.* 1996) on Lake Apopka or those of Sondergaard (Sondergaard *et al.* 1992) on Lake Arreso. In our experiment, when the flow rate increases in the range of 35–65 cm/s, the concentration of Zn in water decreases. We speculate that certain hydrodynamic conditions (such as at different flow velocities (Huang *et al.* 2015) and wind speeds (Yuan *et al.* 2020)) can promote the adsorption of dissolved Zn by the sediment. Apart from the quadratic fitting error of 15% for Cu, the fitting errors of the release fluxes of the other three elements are less than 10%. However, as we verified the above patterns in outdoor tests, it was found that the heavy metal release fluxes of two points (V2 and V3) do not meet those patterns, and the errors between the measured and theoretical values at the other four verification points are within 30%. We assume that the mismatch at V2 and V3 is due to the large fluctuation of water quality and strong acidity (Table 2). It indicates that pH (2.5–5, Table 2) has a significant influence on the release of heavy metals from sediments, which is consistent with previous research conclusions (Atkinson *et al.* 2007; Wen *et al.* 2021). The verification errors for both points V1 and V6 are within 20%. This is because the pollution levels of heavy metals at these two points are low, especially at point V1. V1 is near the source of the Le'an River and it is not polluted by any heavy metals. The verification error at point V4 is within 30%, as this point is in the buffer zone after where the wastewater is discharged from the Dexing Copper Mine into the Le'an River. Since the impact of water pH remains at this point, the conclusions of this article are applicable only when the pH is between 5 and 9. The accuracy of the outdoor verification of the equation, however, is not as high as that of the indoor test (the water used for indoor testing was chlorine-removed tap water), indicating that there are further factors affecting the release of heavy metals from sediment.

We could not determine the acceptable margin of error; however, we can refer to the overview of the water quality model that currently considers simulation results acceptable within a 30% margin of error from the water quality model (Li *et al.* 2011; Wu *et al.* 2019; Chen *et al.* 2020). Though we can identify various factors (such as pH, dissolved oxygen (Atkinson *et al.* 2007), temperature (Huang *et al.* 2017), the inherent characteristics of the sediment (Xu *et al.* 2015), high salinity and high nitrogen levels (Hong *et al.* 2011), the content of P in the overlying water (Chen *et al.* 2017), sediment redox potential (Martin-Torre *et al.* 2015), sulfide and calcium content (Zhou *et al.* 2020)) that affect the heavy metal release from sediments, many issues are yet to be studied: the extent that each factor (especially hydrodynamic conditions and pH) influences heavy metal release from sediments, the release characteristics of dissolved and suspended heavy metals in sediments under different hydrodynamic conditions, the release time to reach a relative equilibrium, the total amount of heavy metals released from sediments under different environmental conditions. Here we analyzed the particle size of sediments in the Le'an River particularly (0–250 μm), since particle size also has a certain impact on nutrient release from sediments (Zhu

et al. 2014). Since both indoor and outdoor test sediment samples are obtained from the Le'an River, the effect of sediment particle size on the results of this paper can be neglected. Whether the principles established herein are applicable to other flowing water bodies with different particle size needs further evaluation.

In this paper, the mechanism of heavy metal release from sediments under different fluid flow conditions is discussed and validated through a field test, thereby providing a theoretical reference for further understanding of the endogenous release mechanism of sediments.

5. CONCLUSIONS

In the present work, eight groups of hydrodynamic experiments with velocities ranging 0–1 m/s are set up in a rectangular flume to simulate the principle of river sediment movement dynamics under different flow rates, and the concentration of heavy metals in water is measured at different flow rates. The relationship between the release flux of heavy metals from sediments and the flow velocity are subsequently determined. Results demonstrated that: (1) when the sediment particle size is between 0 and 250 μm , the water depth is below 2.1 m, within 1 hour, the heavy metal release flux from sediments in river follows a quadratic polynomial relationship with the flow rate; (2) the release of heavy metals in the river sediments at different flow rates within 1 h is greater than the adsorption; (3) for rivers with a water depth of less than 2.1 m, the flow velocity has a greater impact on the release flux of heavy metals from sediments than flow turbulence; (4) flow rate and water pH are two important factors affecting the release of heavy metals from sediments; (5) within a certain flow rate and pH range, the error between the measured and calculated values of heavy metal concentrations in a field test on the Le'an River was within 30%, indicating that the relationship established by the present work between the release flux of heavy metals from sediment and the water velocity is basically adequate, and the experimental device can be operated in a feasible manner.

ACKNOWLEDGEMENTS

The research is supported by National Natural Science Foundation of China (31660140), National Science and Technology Support Program Project of China (2012BAC11B00), Land Development and Consolidation Project in Jiangxi Province (9131207800), Science and Technology Project of Jiangxi Provincial Water Resources Department (202022YBKT15). We would like to thank Ningning Shi and Guanghui Feng for their assistance with field sampling and major ions analysis. We also would like to thank the anonymous reviewers for their helpful comments and suggestions.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

REFERENCES

- Acosta, J. A., Jansen, B., Kalbitz, K., Faz, A. & Martinez-Martinez, S. 2011 Salinity increases mobility of heavy metals in soils. *Chemosphere* **85** (8), 1318–1324.
- Argese, E., Ramieri, E., Bettiol, C., Pavoni, B., Chiozzotto, E. & Sfriso, A. 1997 Pollutant exchange at the water/sediment interface in the Venice canals. *Water Air and Soil Pollution* **99** (1–4), 255–263.
- Atkinson, C. A., Jolley, D. F. & Simpson, S. L. 2007 Effect of overlying water pH, dissolved oxygen, salinity and sediment disturbances on metal release and sequestration from metal contaminated marine sediments. *Chemosphere* **69** (9), 1428–1437.
- Byrne, P., Wood, P. J. & Reid, I. 2012 The impairment of river systems by metal mine contamination: a review including remediation options. *Critical Reviews in Environmental Science and Technology* **42** (19), 2017–2077.
- Cai, Y. B., Xu, N., Meng, F. D., Li, F. Y., Xie, Y. & Zhang, H. 2021 Speciation and release kinetics simulation of Zn and Cd from river sediment contaminated by gold mining. *Water Air and Soil Pollution* **232** (1) 749, 21.
- Chen, H. Y., Chen, R. H., Teng, Y. G. & Wu, J. 2016 Contamination characteristics, ecological risk and source identification of trace metals in sediments of the Le'an River (China). *Ecotoxicology and Environmental Safety* **125**, 85–92.
- Chen, M., Ding, S., Zhang, L., Li, Y., Sun, Q. & Zhang, C. 2017 An investigation of the effects of elevated phosphorus in water on the release of heavy metals in sediments at a high resolution. *Science of the Total Environment* **575**, 330–337.
- Chen, F., Zhang, C., Brett, M. T. & Nielsen, J. M. 2020 The importance of the wind-drag coefficient parameterization for hydrodynamic modeling of a large shallow lake. *Ecological Informatics* **59**, 101006.
- Cheng, P., Wang, X. & Feng, C. 2020 Numerical simulation of phosphorus release from resuspended sediment. *Acta Mechanica Sinica* **36** (6), 1191–1201.
- China Ministry of Ecology and Environment of the People's Republic of 2002 (fourth edition)(in Chinese) *Water and Waste Water Monitoring and Analysis Method*. China Environmental Publishing Group, Beijing.

- Coyne, A., Schäfer, J., Blanc, G. & Bossy, C. 2007 Scenario of particulate trace metal and metalloid transport during a major flood event inferred from transient geochemical signals. *Applied Geochemistry* **22** (4), 821–836.
- Guo-ren, D. 1999 Incipient motion of coarse and fine sediment. *Journal of Sediment Research* **6**, 1–9 (in Chinese).
- He, M. C., Wang, Z. J. & Tang, H. X. 1998 The chemical, toxicological and ecological studies in assessing the heavy metal pollution in Le An River, China. *Water Research* **32** (2), 510–518.
- He, J., Xue, H.-X., Lü, C.-W., Fan, Q.-Y., Liang, Y., Sun, Y., Shen, L.-L. & Bai, S. 2009 The impacts of common ions on the adsorption of heavy metal. *Environmental Geology* **58** (7), 1499–1508.
- He, J., Deng, W., Chen, C., Xu, X., Wang, S., Liu, W. & Wu, X. 2015 Ammonia nitrogen adsorption and release characteristics of surface sediments in Dianchi Lake, China. *Environmental Earth Sciences* **74** (5), 3917–3927.
- Hong, Y. S., Kinney, K. A. & Reible, D. D. 2011 Effects of cyclic changes in pH and salinity on metals release from sediments. *Environmental Toxicology and Chemistry* **30** (8), 1775–1784.
- Hu, S., Wang, Y., Chen, L., Wang, Z., Tong, Z., Deng, Q. & Luo, S. 2020 Determination of Cr in water and sediment of Haidian Creek by cloud point extraction-flame atomic absorption spectrometry. *Desalination and Water Treatment* **188**, 333–341.
- Huang, J. Z., Ge, X. P. & Wang, D. S. 2012a Distribution of heavy metals in the water column, suspended particulate matters and the sediment under hydrodynamic conditions using an annular flume. *Journal of Environmental Sciences* **24** (12), 2051–2059.
- Huang, J. Z., Ge, X. P., Yang, X. F., Zheng, B. & Wang, D. S. 2012b Remobilization of heavy metals during the resuspension of Liangshui River sediments using an annular flume. *Chinese Science Bulletin* **57** (27), 3567–3572.
- Huang, Y. X., Zhang, D. F., Li, Y. H., Xu, Z. H., Yuan, S. J. & Wang, L. 2015 Heavy metals sorption on river sediments: kinetics, equilibria and relative selectivities. *Fresenius Environmental Bulletin* **24** (9), 2792–2799.
- Huang, L., Fang, H., He, G., Jiang, H. & Wang, C. 2016 Effects of internal loading on phosphorus distribution in the Taihu Lake driven by wind waves and lake currents. *Environmental Pollut* **219**, 760–773.
- Huang, Y., Zhang, D., Xu, Z., Yuan, S., Li, Y. & Wang, L. 2017 Effect of overlying water pH, dissolved oxygen and temperature on heavy metal release from river sediments under laboratory conditions. *Archives of Environmental Protection* **43** (2), 28–36.
- Hwang, K.-Y., Kim, H.-S. & Hwang, I. 2011 Effect of resuspension on the release of heavy metals and water chemistry in anoxic and oxic sediments. *CLEAN – Soil, Air, Water* **39** (10), 908–915.
- Jiang, Y. H., Xie, H. Q., Zhang, H., Xie, Z. L. & Cao, Y. 2018 Dissolved heavy metals distribution and risk assessment in the Le'an River subjected to violent mining activities. *Polish Journal of Environmental Studies* **27** (4), 1559–1572.
- Li, Y., Wang, X. L., Huang, G. H., Zhang, B. Y. & Guo, S. H. 2009 Adsorption of Cu and Zn onto Mn/Fe oxides and organic materials in the extractable fractions of river surficial sediments. *Soil & Sediment Contamination* **18** (1), 87–101.
- Li, Y. P., Acharya, K. & Yu, Z. B. 2011 Modeling impacts of Yangtze River water transfer on water ages in Lake Taihu, China. *Ecological Engineering* **37** (2), 325–334.
- Liang, Y., Xiao, H., Liu, X. & Shi, H. 2020 The risk and phytotoxicity of metal(loid)s in the sediment, floodplain soil, and hygrophilous grasses along Le'an River. *International Journal of Environmental Science and Technology* **17** (4), 1963–1974.
- Liu, G. M., An, Q., Wang, L. J., Jia, X., Feng, S. H., Xia, S. M., Wu, Y. N. & Qi, H. 2018a Release and kinetics of arsenic and plumbum in the Songhua River surficial sediments. *Environmental Science and Pollution Research* **25** (1), 541–551.
- Liu, J., Liu, B.-b., Han, M., Jia, N. & Zhang, C.-l. 2018b Methodology research for determination of total phosphorus in water by inductively coupled plasma-atomic emission spectrometry. *Spectroscopy and Spectral Analysis* **38** (6), 1880–1883.
- Liu, J. J., Diao, Z. H., Xu, X. R. & Xie, Q. 2019 Effects of dissolved oxygen, salinity, nitrogen and phosphorus on the release of heavy metals from coastal sediments. *Science of the Total Environment* **666**, 894–901.
- Lors, C., Tiffreau, C. & Laboudigue, A. 2004 Effects of bacterial activities on the release of heavy metals from contaminated dredged sediments. *Chemosphere* **56** (6), 619–630.
- Lu, F., Zhang, H. Q., Jia, Y. G., Liu, W. Q. & Wang, H. 2019 Migration and diffusion of heavy metal Cu from the interior of sediment during wave-induced sediment liquefaction process. *Journal of Marine Science and Engineering* **7** (12), 449.
- Mahdavi, A., Kashfeipour, S. M. & Omid, M. H. 2013 Effect of sorption process on cadmium transport. *Proceedings of the Institution of Civil Engineers-Water Management* **166** (3), 152–162.
- Martin-Torre, M. C., Ruiz, G., Galan, B. & Viguri, J. R. 2015 Generalised mathematical model to estimate Zn, Pb, Cd, Ni, Cu, Cr and As release from contaminated estuarine sediment using pH-static leaching tests. *Chemical Engineering Science* **138**, 780–790.
- Niu, Y., Niu, Y., Yu, H., Jiang, X., Guo, X., Pang, Y. & Xu, X. 2017 Concentration distribution and toxicity of heavy metals in surface sediment of Poyang Lake, China. *Wetlands* **39** (S1), 55–62.
- Reddy, K. R., Fisher, M. M. & Ivanoff, D. 1996 Resuspension and diffusive flux of nitrogen and phosphorus in a hypereutrophic lake. *Journal of Environmental Quality* **25** (2), 363–371.
- Soltan, M. E., Rashed, M. N. & Taha, G. M. 2001 Heavy metal levels and adsorption capacity of Nile River sediments. *International Journal of Environmental Analytical Chemistry* **80** (3), 167–186.
- Sondergaard, M., Kristensen, P. & Jeppesen, E. 1992 Phosphorus release from resuspended sediment in the shallow and wind-exposed Lake Arresø, Denmark. *Hydrobiologia* **228** (1), 91–99.
- Sun, Z. H., Xu, G. H., Hao, T., Huang, Z., Fang, H. R. & Wang, G. 2015 Release of heavy metals from sediment bed under wave-induced liquefaction. *Marine Pollution Bulletin* **97** (1–2), 209–216.

- Tang, C., Li, Y., He, C. & Acharya, K. 2020 Dynamic behavior of sediment resuspension and nutrients release in the shallow and wind-exposed Meiliang Bay of Lake Taihu. *Science of the Total Environment* **708**, 135131.
- Tao, L., Liu, G. N., Liu, X. H., Zhang, C., Cheng, D. M., Wang, A. J. & Li, R. P. 2014 Trace metal pollution in a Le'an River tributary affected by non-ferrous metal mining activities in Jiangxi Province, China. *Chemistry and Ecology* **30** (3), 233–244.
- Wang, P. F., Wang, C., Ouyang, P., Qian, J. & Shi, R. J. 2011 Physiological responses of *Vallisneria spiralis* L. induced by different hydraulic conditions when exposed to copper and nitrogen. *African Journal of Biotechnology* **10** (38), 7441–7452.
- Wang, H., Zhou, Y. & Wang, X. 2016 Transport dynamics of Cr and Zn between deposited sediment and overlying water. *CLEAN – Soil, Air, Water* **44** (11), 1453–1460.
- Wang, H., Kaisam, J. P., Liang, D., Deng, Y. & Shen, Y. 2020 Wind impacts on suspended sediment transport in the largest freshwater lake of China. *Hydrology Research* **51** (4), 815–832.
- Wen, X. H. & Allen, H. E. 1999 Mobilization of heavy metals from Le An River sediment. *Science of the Total Environment* **227** (2–3), 101–108.
- Wen, T., Zhong, H., Fu, G. & Zhong, Y. 2021 Effects of environmental factors on release amount of heavy metal and structure of microbial community in sediments. *International Journal of Environmental Science and Technology* 1–12.
- Wu, Z., Wang, S. & Ji, N. 2019 Phosphorus (P) release risk in lake sediment evaluated by DIFS model and sediment properties: a new sediment P release risk index (SPRRI). *Environmental Pollution* **255**, 113279.
- Xu, W. H., Li, X. D., Wai, O., Huang, W. L. & Yan, W. 2015 Remobilization of trace metals from contaminated marine sediment in a simulated dynamic environment. *Environmental Science and Pollution Research* **22** (24), 19905–19911.
- Yan, Z.-X., Qu, J.-K., Yu, Z.-H., Song, J. & Wei, G.-Y. 2019 Multi-spectral fitting-determination of trace cobalt in high purity nickel by inductively coupled plasma atomic emission spectrometry. *Chinese Journal of Analytical Chemistry* **47** (3), 423–428.
- Yan, H. J., Zhang, H. Y., Shi, Y. J., Zhou, P., Li, H., Wu, D. L. & Liu, L. 2021 Simulation on release of heavy metals Cd and Pb in sediments. *Transactions of Nonferrous Metals Society of China* **31** (1), 277–287.
- Yang, X., Jia, Z., Yang, X., Li, G. & Liao, X. 2017 Cloud point extraction-flame atomic absorption spectrometry for pre-concentration and determination of trace amounts of silver ions in water samples. *Saudi Journal of Biological Sciences* **24** (3), 589–594.
- Yiping, L., Yong, P., Jun, L., Gang, Z., Ling, D., Junpeng, P. & Chao, W. 2004 On the relation between the release rate of TN, TP from sediment and water velocity. *Journal of Lake Science* **16** (04), 318–324 (in Chinese).
- Yu, J., Chen, Q., Zhang, J., Zhong, J., Fan, C., Hu, L., Shi, W., Yu, W. & Zhang, Y. 2019 In situ simulation of thin-layer dredging effects on sediment metal release across the sediment-water interface. *Science of the Total Environment* **658**, 501–509.
- Yuan, W., Hua, W., Fan, L., Deng, Y. & Zeng, Y. 2020 Impact of wind on copper footprints in a large river-connected lake. *Water and Environment Journal* **34**, 586–596.
- Zhang, J., Han, L., Ji, Y., Wei, J., Cai, G., Gao, G., Wu, J. & Yao, Z. 2018 Heavy meta investigation and risk assessment along the Le'An River from non-ferrous metal mining and smelting activities in Poyang, China. *Journal of Environmental Biology* **39** (4), 536–545.
- Zheng, S. S., Wang, P. F., Wang, C., Hou, J. & Qian, J. 2013 Distribution of metals in water and suspended particulate matter during the resuspension processes in Taihu Lake sediment, China. *Quaternary International* **286**, 94–102.
- Zhou, Y. C., Gu, T. F., Yi, W. T., Zhang, T. Q. & Zhang, Y. P. 2019 The release mechanism of heavy metals from lab-scale vertical flow constructed wetlands treating road runoff. *Environmental Science and Pollution Research* **26** (16), 16588–16595.
- Zhou, J., Zhang, M., Ji, M., Wang, Z., Hou, H., Zhang, J., Huang, X., Hursthouse, A. & Qian, G. 2020 Evaluation of heavy metals stability and phosphate mobility in the remediation of sediment by calcium nitrate. *Water Environment Research* **92** (7), 1017–1026.
- Zhu, H.-w., Cheng, P.-d., Zhong, B.-c. & Wang, D.-z. 2013 Hydrodynamic effects on contaminants release due to rususpension and diffusion from sediments. *Journal of Hydrodynamics* **25** (5), 731–736.
- Zhu, H. W., Wang, D. Z. & Cheng, P. D. 2014 Dynamic release process of pollutants during suspended sediment transport in aquatic system. *Applied Mathematics and Mechanics-English Edition* **35** (11), 1411–1420.
- Zhu, L., Li, X., Zhang, C. & Duan, Z. 2017 Pollutants' release, redistribution and remediation of black smelly river sediment based on re-suspension and deep aeration of sediment. *International Journal of Environmental Research and Public Health* **14** (4), 374.

First received 6 May 2021; accepted in revised form 24 July 2021. Available online 6 August 2021