

## Remobilization of heavy metals during the resuspension of Liangshui River sediments using an annular flume

HUANG JianZhi, GE XiaoPeng\*, YANG XiaoFang, ZHENG Bei & WANG DongSheng

State Key Laboratory of Environmental Aquatic Chemistry, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China

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The release and transport of heavy metals (Ni, Cr, Cu and Pb) from Liangshui River sediments into the overlying water column during the resuspension event were determined using an annular flume with a velocity ranging from 0.15 to 0.35 m/s. It is shown that the suspended particulate matters (SPM) increased as much as nearly 25 times from 165 to 4220 mg/L as the velocity increased. Heavy metals showed an increase in dissolved phase as the velocity increased due to their desorption from the SPM. Acid-soluble heavy metals increased as the velocity increased, indicating that part of the heavy metals transformed from stable phase to labile phase during resuspension. Heavy metal concentrations in the SPM on volume normalization increased by approximately 2–6 times. However, on the mass weighted basis they decreased, approaching the bulk-sediment contents at high velocity, due to the “particle concentration effect”. The distribution coefficients ( $K_D$ ) of heavy metals were higher at slower velocity during the sediment resuspension, which could be attributed to the decrease of fine particles (silt/clay fraction) during resuspension.

**heavy metals, sediment resuspension, suspended particulate matters, acid-soluble heavy metals, annular flume**

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Contaminated sediments of heavy metals are currently a widespread problem with the potential to threaten water quality and aquatic ecosystems [1,2]. In most cases, heavy metals entering into rivers can be stored in the river sediments in various forms. However, the contaminants may not be permanently sequestered in sediments. When events such as storms, ship traffic or remedial dredging occur, the metals can potentially be released into the water column due to sediment resuspension, which would cause second river pollution.

Sediment resuspension is a ubiquitous natural process in river, marine and estuarine areas, and usually occurs when bottom shear stress is sufficient enough to disrupt the cohesion of the particles. The main factors that determine the magnitude and duration of resuspension are the shear stress exerted on sediments, and the characteristics of sediments,

e.g. grain size, organic content, etc. All these variables influence the resettling velocities of the particles, and, ultimately, their residence time in the water column [3]. However, few studies have dealt with the release of heavy metals in the remobilization of contaminated sediments under simulated field conditions. To date, the behavior of heavy metals during sediment resuspension is not fully understood. Therefore, an investigation concerning the partitioning of heavy metals between particulate and dissolved phases in water column is significant.

Currently, there are two main types of devices used to simulate the sediment resuspension process. One is the particle entrainment simulator (PES) designed by Tsai and Lick [4]; the other is flume tank [5]. Among flume tanks, annular flume has been extensively used to investigate various physical, chemical and biological factors which may influence the stability and erodibility of natural cohesive sediments [6–10]. One of the major advantages of annular flume

\*Corresponding author (email: [xpge@rcees.ac.cn](mailto:xpge@rcees.ac.cn))

is its fully developed boundary layer above the sediment due to the constant channel geometry and infinite flow length [11], converting the length of straight flume into criterion that satisfies lead-time and long distance during particle flocculation and sedimentation [6]. In addition, annular flume is more suited to investigating the cohesive sediment dynamics, as the flocculated suspensions are not disrupted by re-circulating pumps and filters [12], and this is beneficial to the measurement of SPM characteristics.

The aim of the present study was to evaluate the effects of different current velocities on the mobilization and distribution of heavy metals using an annular flume during sediment resuspension. Four heavy metals (Cr, Ni, Cu, and Pb) were investigated to determine how current velocity influences their remobilization and distribution between the particulate and dissolved phases, enabling a better understanding of the release of heavy metals from sediment to provide practical guidance for the riverine sediment management.

## 1 Materials and methods

### 1.1 Description of the annular flume

The annular flume is composed of acrylic materials with a diameter of 500 mm (outer) and 300 mm (inner), resulting in a 100 mm channel width with a total bed area of 0.13 m<sup>2</sup>. The maximum water depth is 350 mm, corresponding to a total flume volume of 44 L. Flows in the annular flume are induced by a top rotating lid equipped with four paddles spaced equidistantly (Figure 1). Water may be sampled from six different heights (100, 150, 200, 250, 300, 350 mm from the base) via 6 mm bore tubes fixed in the outer wall of the flume.

### 1.2 Sampling

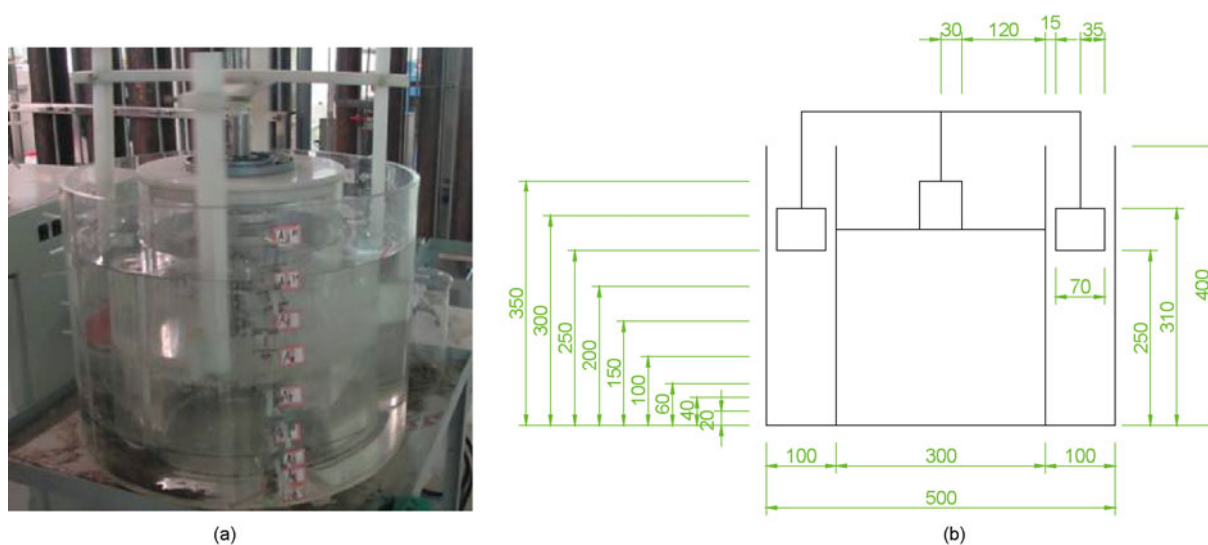
Sediment samples were collected at point YLZ (116°46'41.6"E, 39°47'43.3"N) using a Van Veen stainless steel grab sampler from a depth of < 10 cm in the Liangshui River, Beijing (Figure 2), then the samples were relocated to the laboratory in air-sealed plastic bags. Stones, branches and other solid materials in the sediments were carefully removed, and the sediments were then thoroughly mixed before being introduced into the flume.

### 1.3 Resuspension experiments

Sediments were placed into the flume to a depth of 6 cm. In order to fill the flume with water without disturbing the sediment, a sheet of plastic was carefully placed over the surface and water was gently pumped onto the sheet, which then gradually floated off the sediment surface. During resuspension, the current velocity was increased from 0.15 to 0.35 m/s with 0.05 m/s increments imposed for each experiment. The rotation was held for 30 min. After each rotation rate, water samples were collected at a height of 20 cm from the flume base using plastic containers, and replacement water was added after sampling to maintain the volume.

### 1.4 Analytical methods

Particle size analysis was performed using a Mastersizer 2000 (Malvern Co., UK). Acid volatile sulfide (AVS) was determined by a method adapted from a previous study [13]. Whole sediment and water samples containing SPM were digested using a CEM model MARS X. A combination of concentrated acids (i.e. 3 mL HF, 2 mL HCl and 5 mL HNO<sub>3</sub>) was added to each sample (0.2 g or 5 mL for the



**Figure 1** Setup for the annular flume. (a) Photo; (b) schematic diagram with size specifications (mm).



**Figure 2** Map of Liangshui River showing the sampling sites. YLZ (L8): sampling site.

sediments and water samples, respectively). Heavy metal concentrations (Cr, Ni, Cu and Pb) were quantified using an Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES) or Inductively Coupled Plasma Mass Spectrometry (ICP-MS) based on their concentrations. Acid-soluble metal concentrations were determined using the methods adopted by previous studies [14,15], which reflects the labile fraction of heavy metals in SPM. Procedures for total organic carbon (TOC) analysis were as follows: homogenization-dried samples were acidified with 21% H<sub>3</sub>PO<sub>4</sub> to remove carbonates, then analyzed with a TOC Analyzer (Apollo 9000, USA). Blank and triple samples were taken for heavy metal analysis in order to obtain accurate results.

## 2 Results and discussion

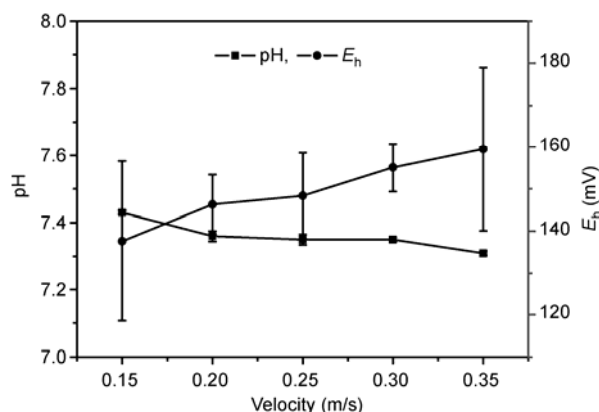
### 2.1 Sediment and overlying water characteristics

The characteristics of sediment are shown in Table 1. The sediment was mainly composed of silt-and sand-sized particles (94.3%), with minor cohesive clay fractions (5.7%). The acid volatile sulphide in the sediment was very low with only 0.35 μmol/g, indicating that the sulphide bound metal fraction would have little influence on the release of heavy metals.

Overlying water parameters (pH and *E<sub>h</sub>*) during sediment resuspension are shown in Figure 3. It is found that overlying water pH fluctuated between 7.31–7.43 with no significant change, which could be ascribed to the strong pH buffer capacity in the sediment and overlying water system. However, the *E<sub>h</sub>* value for the overlying water increased from 137.6 (0.15 m/s) to 159.5 mV (0.35 m/s). It is possible that a higher velocity would lead to stronger disturbance in the water column, resulting in the increase of oxygen content in the overlying water. In addition, with the velocity changing from 0.15 to 0.35 m/s, SPM increased from 165 to 4220 mg/L, nearly as much as 25 times. Furthermore, there is a

**Table 1** Physicochemical characteristics of sediments from Point YLZ

AVS (μmol/g)	TOC (g/kg)	Grain size (%)			Metals (mg/kg dry weight)			
		Sand	Silt	Clay	Cr	Ni	Cu	Pb
0.35	95.47	22.1	72.2	5.7	25.7	51.1	113.9	17.6

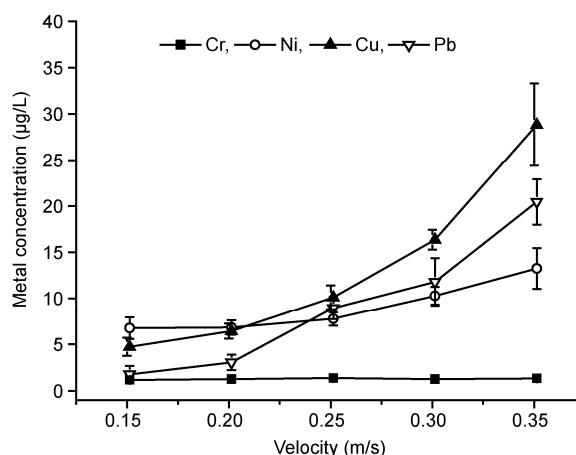


**Figure 3** Variations of pH and *E<sub>h</sub>* in the overlying water during sediment resuspension.

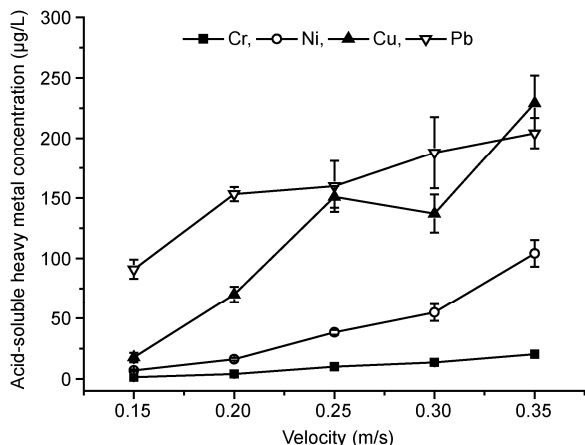
negative correlation between current velocity and TOC of SPM ( $R^2=0.97$ ,  $P<0.05$ ). TOC of SPM decreased from 96.79 g/kg at 0.20 m/s to 84.77 g/kg at 0.35 m/s, due to the increase in proportion of larger particles, which contained lower TOC per unit mass [3].

### 2.2 Distribution of heavy metals in resuspension experiments

(i) Heavy metals in dissolved phase. The variations in the concentration of dissolved heavy metals with the simulated velocity are shown in Figure 4. Chromium content in the dissolved phase remained nearly constant at approximately 1.3 μg/L during the resuspension. Other heavy metals (Ni, Cu, and Pb) increased from 6.76, 4.74, 1.77 μg/L (0.15 m/s) to 13.24, 28.84, 20.45 μg/L (0.35 m/s), respectively. The variation of dissolved heavy metal concentrations may be due to the following reasons: (1) Part of the heavy metals in SPM changed from stable phase to exchangeable phase, supported by the increase of the concentration of acid-soluble heavy metals during the resuspension event (Figure 5). In addition, as the velocity increased, particle-



**Figure 4** Variations in dissolved heavy metals with simulated velocity.



**Figure 5** Variations of acid-soluble heavy metals in water column with the simulated velocity.

particle collision would become more frequent, facilitating the desorption of the labile fraction of heavy metals in SPM; (2) Particles which are smaller than  $0.45 \mu\text{m}$  (the size of filter membrane in this study) entrained into the overlying water increased (Table 2). According to the previous study [16], finer particles tend to enrich more heavy metals than larger ones. Therefore, it is possible that heavy metals adsorbed onto particles (smaller than  $0.45 \mu\text{m}$ ) would result in an increase in dissolved phase, due to the fact that their passing through the filter membrane would be calculated as the dissolved fraction.

(ii) Acid-soluble heavy metals. The variations of acid-soluble heavy metal concentrations with the simulated velocity are shown in Figure 5. Acid-soluble heavy metal contents reflect the labile fraction of heavy metals in SPM. It is found that acid-soluble heavy metal fractions generally showed an increase as the velocity increased. Cr, Ni, Pb, and Cu increased from 1.35, 6.88, 90.92, and  $17.62 \mu\text{g/L}$  (0.15 m/s) to 20.26, 104.12, 204.17, and  $229.17 \mu\text{g/L}$  (0.35 m/s), respectively, except for Cu at 0.30 m/s. It could be attributed to the sharp rise in SPM concentration during the resuspension, increasing from  $165 \text{ mg/L}$  (0.15 m/s) to  $4220 \text{ mg/L}$  (0.35 m/s) (Table 2), leading to an increase in the acid-soluble fraction per volume. Another possible reason is that heavy metal speciation in SPM changed from stable phase to labile phase during the resuspension event,

**Table 2** SPM characteristics at different velocities

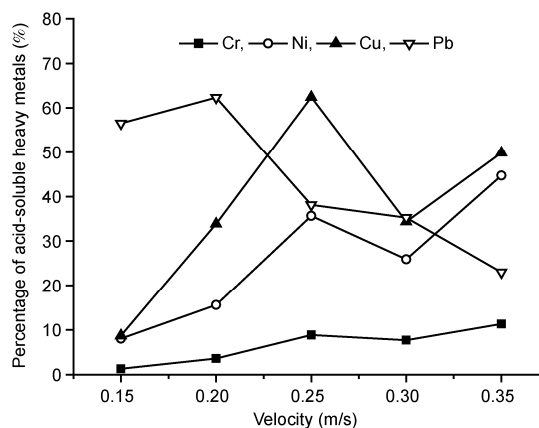
Velocity (m/s)	SPM (mg/L)	S/S/C (%) <sup>a)</sup>	TOC (g/kg)
0.15	$165 \pm 25$	8.7/26.1/65.2	ND <sup>b)</sup>
0.20	$307 \pm 10$	10.1/25.7/64.2	$96.8 \pm 1.5$
0.25	$1120 \pm 113$	10.7/22.6/66.7	$90.3 \pm 1.9$
0.30	$2368 \pm 69$	11.1/20.5/68.4	$89.2 \pm 0.9$
0.35	$4220 \pm 380$	13.1/17.6/69.3	$84.8 \pm 3.3$

a) S/S/C (%) represents the percentages of sand, silt, and clay, respectively; b) ND, not detected.

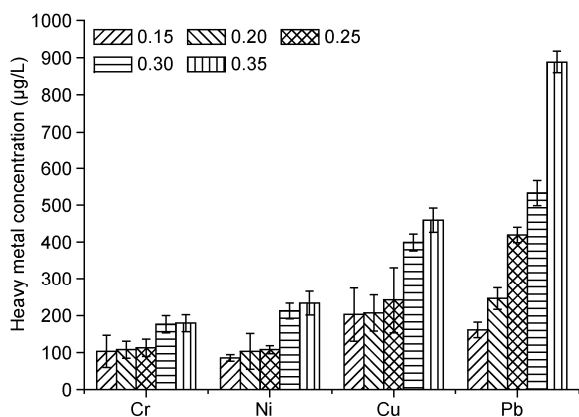
supported by the fact that the TOC content in SPM decreased as velocity increased (Table 2).

The percentage of acid-soluble fraction relative to the total heavy metals in SPM on volume normalization is shown in Figure 6. It can be seen that the percentage for individual acid-soluble metal fraction varied during the resuspension. Ni and Cr generally showed an increase as the velocity rose, which is similar to the previous study [14], indicating that part of heavy metal speciations in SPM transformed from stable phase to labile phase during resuspension. Cu and Pb, however, showed an increase at first and then decreased as the velocity continued to increase. This may be due to the fact that the speciation of Cu and Pb changed from stable to exchangeable forms at the first stage, and then were adsorbed and co-precipitated by the Fe and Mn hydroxide/oxides which were caused by the rapid oxidation of Fe(II) and Mn(II) at the second process [17].

(iii) Heavy metal concentrations in SPM. The change of heavy metal concentrations in SPM on volume normalization is shown in Figure 7. Heavy metal concentrations in the particulate phase increased by approximately 2–6 times during the resuspension event. Among these heavy metals, Pb exhibited the greatest increase from  $160.8$  (0.15 m/s) to  $888.6 \mu\text{g/L}$  (0.35 m/s), nearly 6 fold. The concentrations of Ni, Cr and Cu increased from 85.5, 103.3, and  $202.6 \mu\text{g/L}$  (0.15 m/s) to 232.6, 178.8, and  $459.7 \mu\text{g/L}$  (0.35 m/s), respectively, only about 2–3 fold. This phenomenon was in accordance with the change of the concentrations of SPM in the overlying water as the velocity increased (Table 2). That heavy metal concentrations are proportional to the velocity would suggest that more attention should be given to the sediments that were historically contaminated with heavy metals. Heavy metal release into the overlying water during sediment resuspension may exceed the aquatic criteria, leading to water quality deterioration. Previous studies [18,19] have noted that the increases in heavy metals during resuspension events are due to the entrainment of particles under turbulent conditions.



**Figure 6** Changes in acid-soluble metal fractions as the percentage of total heavy metals in SPM on volume normalization with the simulated velocity.

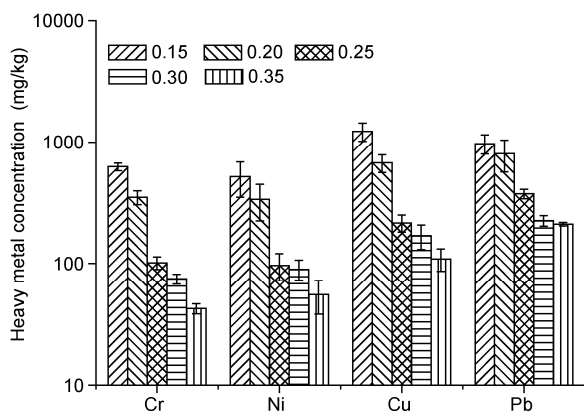


**Figure 7** Heavy metal concentration profiles on volume normalization versus velocity.

On the basis of mass normalization, however, the heavy metal concentrations decreased as the velocity increased (Figure 8). Cr, Ni, Cu and Pb diminished from 626.2, 518.3, 1228.1, and 974.5 to 42.4, 55.1, 108.9, and 210.6 mg/kg, respectively. These results are similar to previous results [14]. One possible explanation for this phenomenon was that at higher velocities, the entrainment of larger size particles contaminated with lower heavy metals diluted the contaminants, hence depressing their concentration in SPM. Furthermore, the sharp increase in SPM concentration from 165 to 4220 mg/L (Table 2) may also affect the partitioning of particulate heavy metals due to the “particle concentration effect”, which showed a decrease in metal adsorption onto SPM when SPM concentration in the water column increased [20–22]. Finally, heavy metal speciation in SPM may change from the stable phase to the labile phase during the resuspension event, supported by the fact that the TOC content in SPM decreased as the velocity increased (Table 2).

**2.3 Sediment-water partitioning of heavy metals**

The partitioning of heavy metals between dissolved and particulate phases is given by the distribution coefficient



**Figure 8** Heavy metal concentration profiles on mass normalization versus velocity. Note: the ordinate scale is logarithmic.

( $K_D$ ) as shown in eq. (1):

$$K_D = \frac{C_P}{C_S}, \tag{1}$$

where  $C_S$  is the concentration in dissolved phase (mg/L) and  $C_P$  stands for the concentration in particulate phase (mg/kg). In this study, the distribution coefficients between solid and liquid phases for heavy metals were calculated and listed in Table 3.

As can be seen from Table 3, the  $K_D$  values for heavy metals reduced as the velocity increased from 0.15 to 0.35 m/s during the resuspension, with the exception for Ni at 0.35 m/s, which was consistent with previous study [23]. One possible explanation for this phenomenon is that finer particles (silt/clay fraction), which enrich more heavy metals compared with sand fraction, decreased as the velocity increased (Table 2). Another possible reason is the change of TOC in SPM. According to previous studies [23],  $K_D$  values for heavy metals are typically high in organic-SPMs. In this study, the TOC of SPM decreased from 96.79 to 84.77 g/kg as the velocity increased from 0.2 to 0.35 m/s (Table 2), which is in accordance with the descending order of the  $K_D$  values.

For the heavy metals investigated, the partitioning coefficients were found to be in the order of  $Pb \approx$  or  $> Cr > Cu > Ni$ , which is generally consistent with the Irving-Williams rule [24]. The order remains unchanged as the velocity increases, and this important factor needs to be carefully considered when assessing the heavy metal bioavailability during sediment resuspension in the future.

**3 Conclusions**

Remobilization of heavy metals during sediment resuspension was investigated using an annular flume at velocities ranging from 0.15 to 0.35 m/s. SPM increased from 165 to 4220 mg/L, nearly as much as 25 fold. Soluble heavy metal concentrations generally showed an increase due to the desorption from SPM as the velocity increased. Acid-soluble heavy metal concentrations increased during the resuspension, indicating that part of the heavy metals in SPM transformed from the stable phase to the labile phase. Heavy metal concentrations in SPM on volume normalization

**Table 3** The log  $K_D$  values observed during annular flume resuspension events

Velocity (m/s)	Cr	Ni	Cu	Pb
0.15	5.73	4.88	5.41	5.74
0.20	5.45	4.69	5.02	5.42
0.25	4.87	4.09	4.33	4.62
0.30	4.77	3.94	4.01	4.28
0.35	3.38	4.74	3.58	4.01

displayed an increase of approximately 2–6 times. However, on the mass weighted basis, they decreased due to the “particle concentration effect”. The entrainment of larger particles with lower heavy metals into the overlying water at high velocity diluted the contaminants and depressed their concentrations in solid phase. The distribution coefficients ( $K_D$ ) of heavy metals decreased, due to fine particles (silt/clay fraction) decreasing as the velocity increased.

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