Distribution of heavy metals in the water column, suspended particulate matters and the sediment under hydrodynamic conditions using an annular flume

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Abstract

Sediment resuspension plays an important role in the transport and fate of heavy metals in the aquatic environment. In the present study, the release and binding forms of Cr, Cu, Zn, Pb under hydrodynamic conditions were investigated using an annular flume. Two sediments located at YLZ and GBD from Liangshui River, Beijing were resuspended for 10 hr at 0.159 and 0.267 m/sec, respectively. The concentrations of suspended particulate matters of YLZ were higher than those of GBD during resuspension, indicating that the former sediment is more sensitive to the velocity. Cr in the dissolved phase stayed nearly constant at about 2.25 and 1.84 $\mu\text{g/L}$ for YLZ and GBD, respectively, due to the high percentage of its stable binding fractions in both sediments, while Cu, Zn, and Pb showed a fast release in the initial period of time. However, their concentrations in SPM generally decreased with time and were higher at the lower velocity of 0.159 m/sec, which resulted from the entrainment and depressing effect of larger size particles with lower heavy metal content, commonly referred to as the “particle concentration effect”. In addition, the binding form and heavy metal fractions were also found to vary during the resuspension event. A decrease in the sulphide/organic matters bounded form in GBD sediment was observed, whereas no visible changes were perceived in YLZ site samples. This phenomenon is due to the oxidation of heavy metal-sulphide binding forms, which originated from its high acid volatile sulphide content in GBD sediment.

Key words: heavy metals; sediment resuspension; suspended particulate matters; annular flume

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Introduction

Nowadays, heavy metals originating from human activities are frequently detected in sediments and the water column (Akçay et al., 2003; Olivares-Rieumont et al., 2005). In the aquatic environment, heavy metals are usually distributed in several ways as follows: water-soluble species, colloids, suspended forms and sedimentary phases (Peng et al., 2009). In some cases, more than 99% of the heavy metals entering into rivers can be stored in the sediment in various binding forms. However, the heavy metals accumulated in the sediment may not be permanently sequestered. When events such as storms, ship traffic or remedial dredging occur, resuspension of sediments can potentially lead to the release of heavy metals from sediments into the water column where they pose a threat to water quality and the aquatic ecosystems (Je et al., 2007). Therefore, it is of great importance to investigate the transport and distribution of heavy metals in the water column, suspended particulate matters, and sediments during resuspension events.

Some experiments have been conducted to study factors affecting heavy metal release, including water column

conditions and physicochemical properties of sediments. Salinity, pH, and dissolved oxygen (DO) of the water column have been found to influence metal desorption (Atkinson et al., 2007). In addition, the acid volatile sulfide (AVS) and grain sizes of the sediments appear to regulate the quantity of heavy metals released (Mucha et al., 2004; van Griethuysen et al., 2002). So far, however, little is known about the partitioning of heavy metals between the particulate and filterable phases caused by resuspension events, and the environmental conditions in previous studies were not realistically simulated. Thus, it is important to apply simulation device representative of rivers and estuaries to study heavy metal partitioning between the water column and sediments.

To date, an increasing number of studies (Kelderman and Osman, 2007; Xue et al., 2006) have focused on heavy metal fractions in sediments, which should provide more information about environmental impacts on bioavailability. However, in these investigations, the hydrodynamic conditions were not adequately taken into consideration. Most of them only applied full stirring of waters using oscillating grids or stirrers, which do not occur in nature. Hence, the rate of heavy metal entrainment is likely

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overestimated or restricted. Thus, knowledge about the heavy metal fractions in sediment under hydrodynamic conditions is vital to achieve better understanding of their speciation transformation and distribution in the water-sediment interface.

Several erosion chambers and flumes have been used to measure sediment transport with increasing bottom stress (Jonsson et al., 2006; Simpson et al., 1998). Among these devices, annular flumes have been widely used to investigate various physical, chemical and biological factors (e.g., particle size, organic matter, water content, and macrofauna) affecting the stability and erodibility of natural cohesive sediments (Widdows et al., 1998). One advantage of the annular flume is that the constant channel geometry and infinite flow length result in a fully developed boundary layer above the sediment (Amos et al., 1992). In addition, unlike a straight re-circulating flume, the annular flume is more suited to investigate the cohesive sediment dynamics, as the flocculated suspensions are not disrupted by re-circulating pumps and filters (Manning et al., 2007). Therefore, we applied the annular flume to simulate river conditions in the present study.

The objective was to investigate the release and distribution of heavy metals (Cr, Cu, Zn and Pb) in the sediment under hydrodynamic conditions using an annular flume. For a comparison, two different sediments collected from the Liangshui River, Beijing were resuspended at the velocity of 0.159 and 0.267 m/sec, respectively, which would commonly occur in many aquatic environmental conditions. Results from this study may provide insight into how the contaminated sediments behave during resuspension events, which would establish a scientific basis for river sediment management.

1 Materials and methods

1.1 Description of the annular flume

The annular flume is made of acrylic material with 500 mm (outer) and 300 mm (inner) in diameter and has an annular test channel 100 mm wide and 350 mm deep, resulting in a maximum water volume of 44 L (Fig. 1).

Fig. 1 Setup for the annular flume. (a) photo; (b) schematic diagram with size specifications (unit: mm).

Flow in the annular flume is induced by a top rotating lid equipped with four evenly spaced plexiglass paddles. The acceleration and lid speed are controlled by a motor. Water can be sampled from six different heights (100, 150, 200, 250, 300, and 350 mm from the base) via 6 mm bore tubes in the outer wall of the flume. In the present study, only two of the above sampling outlets (100 mm (down) and 350 mm (up)) were used.

1.2 Sediment sampling

Liangshui River, Beijing, a tributary stream down to the Northern Canal into the Haihe River, was employed for the field study. As a flood control and drainage-taking river system, the river pools the municipal wastewater and sewage outfalls along the waterway, and sluice gate facilities are constructed on the river bank for sewage-irrigation in the southeastern Tongzhou area. In the present investigation, two sampling sites were used, as shown in Fig. 2. One was GBD (116°34'28.9"E, 39°46'17.8"N), located downstream in the sewer drainage canal of GBD (Gaobeidian Municipal Wastewater Treatment Plant) and in the catchment of the Yizhuang High-Tech Zone; The other was YLZ (116°46'41.6"E, 39°47'43.3"N), which is near the entrance of the Liangshui River into the Northern Canal at the Yulinzhuang flood gate.

A Van Veen stainless steel grab sampler was employed to collect the sediment samples from a depth of < 10 cm

Fig. 2 Map of the Liangshui River showing the sampling sites GBD and YLZ.

in the river bed, and then they were taken to the laboratory in air-tight 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 experiment

The annular flume was used to perform the sediment resuspension. Sediments were placed into the flume to a depth of 6 cm. In order to fill the flume with deionized water without disturbing the sediment, a sheet of plastic film was carefully put on the sediment surface and water was gently pumped onto the sheet, which then gradually floated off the sediment surface. Samples were withdrawn and the water column characteristics (pH, Eh, suspended particulate matters (SPM) concentrations) were measured at 2, 4, 6, 8, 10 hr, respectively. These samples were collected at 10 and 35 cm height from the flume base using plastic containers. Replacement water was added after each sampling to maintain constant water volume in the system. In addition, surface sediments in the annular flume were also sampled after 10 hr resuspension in order to evaluate the change of heavy metals fraction.

1.4 Analytical methods

For SPMs analysis, 30 mL of water samples were immediately vacuum filtered using a preweighed glass fiber filter to separate the water and SPM. Particle size analysis was performed with a Mastersizer 2000 system (Malvern, UK). Acid volatile sulphide (AVS) was determined according to a previous study (Fang et al., 2005). Total organic carbon (TOC) contents of sediments were determined using an Apollo 9000 system (Teledyne, USA).

Whole sediments and water samples containing SPM were digested in Teflon vessels using a CEM model MARS X system. A mixture of concentrated acids (3 mL HF, 2 mL HCl and 5 mL HNO₃) was added to each sample (0.2 g or 5 mL). Heavy metal concentrations (Cr, Ni, Cu and Pb) were quantified by Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES) or Inductively Coupled Plasma Mass Spectrometry (ICP-MS) based on their concentrations. Standard solutions of Cr, Ni, Cu and Pb at 1000 mg/L were obtained from the National Center of Analysis and Testing for Nonferrous Metals and Electronic Materials. Blank and triplicate samples were taken to insure the accuracy.

A sequential extraction procedure described by Tessier (1976) was used for metal fractionation in this study. Briefly, it was performed as follows: (1) F1: exchangeable, by extraction with 8 mL MgCl₂ (1.0 mol/L, pH = 7.0) at room temperature for 1 hr; (2) F2: bound to carbonates, by extracting the residue from step 1 with NaAc (1.0 mol/L)

adjusted to pH = 5.0 with acetic acid (HAc); (3) F3: bound to iron and manganese oxides, by extracting the residue from step 2 with NH₄Cl (0.04 mol/L); (4) F4: bound to sulphide/organic matters, by oxidation of the residue from step 3 with H₂O₂ (30%, pH = 2.0) and HNO₃ (0.02 mol/L) with intermittent heating, followed by extraction with 3.2 mol/L NH₄Ac; (5) F5: residual, by digestion of the residue from step 4 with 65% HNO₃.

2 Results and discussion

2.1 Sediment characteristics

Sediment characteristics are shown in Table 1. Both sediments consisted primarily of silt and sand particles with a minor clay fraction. In addition, compared with GBD, the YLZ sediment has more fine particles (silt/clay fraction) and heavy metal content. However, the AVS content in YLZ was lower than that in GBD sediment. For both the sediments, the Cr and Pb contents in the interstitial water were below the detection limits.

2.2 Overlying water characteristics

The variation of overlying water parameters (pH, Eh and SPM concentration) during short time resuspension are shown in Fig. 3. While the pH values of the overlying water for GBD sediment stayed at about 7.08–7.21, those for YLZ fluctuated between 7.28 and 7.67. The pH values for both sediments showed no significant change, which could be ascribed to the strong pH buffer capacity in the overlying water/sediment interfacial systems. The pH values generally decreased with the velocity and kept stable with little change in the different water column layers. However, the Eh values were found to increase with the velocity, which may be due to the effect of DO (dissolved oxygen). The average SPM concentrations for YLZ sediment were 280 mg/L at 0.159 m/sec compared to 4500 mg/L at 0.267 m/sec, while they were 50 and 2100 mg/L, respectively for GBD sediment at the two velocities, indicating that YLZ sediment was more sensitive to the increase of velocity. This may be due to the high content of finer size particles (silt/clay fraction) in sediment YLZ (Table 1), which are more easily entrained into the water column during resuspension.

2.3 Heavy metal release into the overlying water

Changes in the concentration of heavy metals in the dissolved phase are plotted against time at the two different velocities and column layers, as shown in Fig. 4. The concentration of Cr in the overlying water stayed at about 2.25 and 1.84 µg/L, respectively, for both YLZ and GBD sediments with little variation during the short-term

Table 1 Physical and chemical characteristics of YLZ and GBD sediments

Sediments	AVS (µmol/g)	Grain size (%)			Metals (mg/kg dry weight)				Metals in interstitial water (µg/L)			
		Sand	Silt	Clay	Cr	Cu	Zn	Pb	Cr	Cu	Zn	Pb
YLZ	0.38 ± 0.04	23.9	71.8	4.3	27.4 ± 1.8	169.5 ± 16.5	351.6 ± 9.2	43.6 ± 4.2	ND	0.22 ± 0.04	0.43 ± 0.05	ND
GBD	56.7 ± 2.4	45.2	51.2	3.26	17.8 ± 1.7	40.1 ± 6.1	80.5 ± 6.0	13.2 ± 0.8	ND	0.14 ± 0.06	0.11 ± 0.04	ND

ND: not detectable.

Fig. 3 Monitored pH, Eh, suspended particulate matters (SPM) over time during short-term resuspension at the velocities of 0.159 and 0.267 m/sec, respectively. up: sampling outlet at 350 mm; down: sampling outlet at 100 mm. Note: SPM is plotted on a log scale.

resuspension event. This indicated that more stable binding forms of Cr existed in both sediments, which would be supported by the fact that the stable fractions of Cr (F3–F5) account for 73.74% and 73.06% of the overall Cr content in YLZ and GBD, respectively (Fig. 6). Cu, Zn, and Pb in the dissolved phases generally increased with time and were higher at 0.267 m/sec than at 0.159 m/sec. It is very likely that part of the Cu, Zn, and Pb speciation in SPM transformed from stable to labile fractions during resuspension, and particle-particle collision became more frequent at higher velocity, facilitating the desorption and release of labile fractions into the water column. The release of Cu, Zn and Pb at the two different velocities and column layers for both sediments after 2 hr was appropriately 26%–89%, 34%–85%, and 46%–65%, respectively with respect to the total heavy metals release after 10 hr of resuspension, indicating that heavy metals can be quickly released into the water column in this short period of time. Therefore, more attention should be paid to the issue of initial heavy metals release during short-term resuspension events.

Furthermore, concentrations of Cu, Zn and Pb in the upper part of the water column are generally a little higher than those in the lower part. It is assumed that concentrations of tiny colloidal particles smaller than 0.45 μm (the filter pore size used in this study) in the upper part of the water column are generally a little higher than those

in the lower part (Table 3). According to previous studies (Fukue et al., 2006), finer particles favor the entrainment of more heavy metals than the larger size ones on the basis of unit mass. Therefore, a considerable amount of heavy metals associated with these particles can pass through the filter membrane and would be calculated as the soluble fraction, leading to higher heavy metal concentrations in the upper part.

2.4 Heavy metals in suspended particulates matter

Variations of heavy metal concentrations in SPM over time during short-term resuspension are plotted in Fig. 5. In addition, the enrichment of heavy metals in SPM relative to the whole sediment was investigated further. The enrichment factors (EFs) were calculated as follows:

$$EF = \left(\frac{Me_{SPM}^{2+}}{Me_{whole}^{2+}} \right) - 1 \quad (1)$$

where Me_{SPM}^{2+} (mg/kg) is the metal fraction in SPM and Me_{whole}^{2+} (mg/kg) is the metal concentration in the whole sediment (Yang et al., 2008). Calculated EF values after 10 hr of resuspension are given in Table 2.

As shown in Fig. 5, the concentrations of heavy metals in SPM per unit mass for both sediments generally decreased with the time and were a little higher in the

Table 2 Calculated EFs for Cr, Cu, Zn and Pb in SPM after 10 hr of resuspension at 0.159 and 0.267 m/sec velocities

	YLZ _{0.159}		YLZ _{0.267}		GBD _{0.159}		GBD _{0.267}	
	up	down	up	down	up	down	up	down
Cr	10.85	10.44	4.56	3.99	2.16	1.20	0.13	0.01
Cu	7.12	6.86	1.24	1.12	11.94	11.68	6.62	7.07
Zn	4.85	2.76	0.58	0.46	23.75	16.43	11.17	9.64
Pb	9.99	9.48	7.67	7.35	3.44	2.53	0.46	0.37

up: sampling outlet at 350 mm; down: sampling outlet at 100 mm.

Table 3 Grain size distribution of suspended particulate matter in overlying water during resuspension at 0.267 m/sec velocity

	Sand/Silt/Clay (%)				
	2 hr	4 hr	6 hr	8 hr	10 hr
YLZ _{up}	10.3/26.9/62.8	10.3/27.3/62.4	11.2/27.2/61.6	9.8/28.6/61.6	9.5/29.8/60.7
YLZ _{down}	16.7/31.5/51.8	16.9/33.2/49.9	15.3/32.8/51.9	15.6/34.7/49.7	13.7/37.3/49.0
GBD _{up}	15.3/23.9/60.8	15.7/24.1/60.2	15.1/25.9/59.0	14.6/23.9/61.5	14.1/26.2/59.7
GBD _{down}	17.8/21.7/60.5	18.1/22.1/59.8	15.9/23.8/60.3	16.4/25.7/57.9	16.1/24.3/59.6

Fig. 4 Changes of heavy metal concentrations in overlying water over time during short-term resuspension.

upper part than in the lower part of the water column. This result coincides with the ascending trend of heavy metals in the dissolved phase over time during the resuspension (Fig. 4). The law of mass conservation would here follow. It is possible that heavy metal speciation changing from

stable to labile fractions during resuspension facilitates desorption of heavy metals from SPMs. In addition, the percentage of finer size particles (clay/silt) in the upper part of the water column was higher than that in the lower part (Table 3), leading to more heavy metals enriched in the

Fig. 5 Variation of heavy metals in SPM during short-term resuspension.

SPMs of the upper part of the water column.

With increasing velocity, the heavy metal content in SPM decreased, which is consistent with the previous study (Bibby and Webster-Brown, 2005). A reasonable explanation for this observation is that the entrainment of larger size particles with lower contaminants into the overlying water at higher velocity diluted the heavy metal

concentration (Kim et al., 2004). Another possible reason may be due to their redistribution into the water column, resulting from the loss of adsorption sites on particles as the SPM concentration increased (Alkhatib and Berna, 2008), which is supported by the increase in soluble heavy metals at higher velocity as shown in Fig. 4.

EF values for heavy metals varied between sediments

Fig. 6 Heavy metal fractions in binding forms of F1–F5 for original sediments and after 10 hr of resuspension at the velocities of 0.159 and 0.267 m/sec, respectively.

and treatments. For YLZ sediment, they followed the order of $\text{Cr} > \text{Pb} > \text{Cu} > \text{Zn}$ at 0.159 m/sec, and $\text{Pb} > \text{Cr} > \text{Cu} > \text{Zn}$ at 0.267 m/sec, respectively; while for GBD sediment, EF values at both velocities were in the following sequence: $\text{Zn} > \text{Cu} > \text{Pb} > \text{Cr}$. It was found that the EF values in GBD generally followed the Irving-Williams order of complex stability, in which Zn and Cu exhibit high complex stability and hence good enrichment capacity. Many factors may contribute to the magnitude of EF values for the heavy metals. Besides the velocity, the grain size distribution in the sediments is important to affect the suspended particles composition and size fractions entrained into the overlying water during the resuspension event, which determines the capacity of heavy metals taken up by SPM. The variation in grain size distribution (Tables 1 and 3) for both the sediments may explain why the EF values of Pb and Cr for YLZ are larger than those for GBD, because higher fractions of silt/clay of finer size were identified in the former sediment. In addition, the SPM concentrations in the water column were also a significant factor affecting heavy metals in the particulate phase. Previous studies (Shi et al., 1998; Lu and Allen, 2001) have shown a decrease in metal adsorption onto SPM for increasing SPM concentrations,

which is possibly due to the “particle concentration effect” that would be defined in adsorption thermodynamics as the declining trend in metal partition coefficients as the suspended particulate matters increase (Benoit and Rozan, 1999). In the present study, smaller EF values are obtained for both the sediments at 0.267 m/sec than those at 0.159 m/sec since larger SPM concentrations occurred in the former (Fig. 3).

2.5 Heavy metal fractions in sediments

The heavy metal speciation (Cr, Pb, Cu and Zn) of binding forms F1–F5 in the sediments during resuspension are shown in Fig. 6. It can be seen that the heavy metal fractions are related to the characteristics of the applied sediments and the applied velocity. The F1 fractions of labile form were low for YLZ, which is consistent with previous studies (Kelderman and Osman, 2007). For example, the F1 fraction of Cu was only 3.03% as the percentage of total content in YLZ sediment, while high F1 fractions of Cu and Pb (10.84% and 13.74%, respectively) were found in the GBD sample, which could be ascribed to the pollution events.

For GBD sediment, a decrease in the F4 phase after 10 hr of resuspension was observed, especially at the higher

velocity of 0.267 m/sec. However, no significant changes were found in the F4 fraction for the YLZ sample. This may be due to the high AVS content (56.7 $\mu\text{mol/g}$, Table 1) in the GBD sediment. According to the metal fraction procedure, the F4 fraction mainly originated from two kinds of binding forms, i.e. the sulphidic and the organic. From Fig. 3, it can be seen that Eh values increase with the velocity during the short-term resuspension, which may cause the oxidation of sulphide in GBD sediment, leading to the release of metal-sulphide in the F4 fraction. Since the AVS content in the YLZ sample was very low (0.38 $\mu\text{mol/g}$, Table 1), its F4 phase can be mainly ascribed to the organic binding forms, which may not be easily oxidized during the resuspension. This result is in accordance with the previous studies (Akçay et al., 2003; Morse and Rickard, 2004).

Furthermore, as the F4 fraction in GBD sediment decreased during the resuspension, the F3 form of Cr and the F2 phase for Pb, Cu and Zn all showed an increase with no visible changes in the F1 and F5 forms. This is indicative of the fact that heavy metals released through the above sulphide oxidation mechanism from the F4 fraction will be recombined in the loosely binding F2–F3 phases via the redistribution process.

As can be expected, the relative stable F5 (residual) fractions remained rather constant for both the sediments during resuspension. The F5 phases for Cr, Pb, Cu and Zn in YLZ sediment were 7.2, 13.4, 49.3, and 96.3 mg/kg, respectively, while they were 3.8, 1.5, 8.5, and 12.8 mg/kg in GBD sample. Since the F5 fraction represents the strong lithogenic binding form of heavy metals, the above results can be related with the geochemical processes of fairly unpolluted sediments.

3 Conclusions

Sediment resuspension plays an important role in the release of heavy metals in the aquatic environment. The results for hydrodynamic simulation with an annular flume show that the heavy metals Cu, Zn, and Pb in the dissolved phase increased with time and were found in higher concentrations at 0.267 m/sec than at 0.159 m/sec for both the YLZ and GBD sediments during the resuspension. A quick release of Cu, Zn and Pb into the overlying water column was obtained after 2 hr of resuspension. Heavy metal concentrations in SPMs were lower at the higher velocity of 0.267 m/sec, which can be ascribed to the “particle concentration effect”. A decrease in the F4 phase was found in GBD sediment due to the oxidative release of sulphide from the high AVS content during the resuspension, whereas the F4 fraction in the YLZ sample showed no significant changes because of the majority of organic binding forms in F4 speciation, which may not be easily oxidized. The increase in the F3 form of Cr and the F2 phase for Pb, Cu and Zn in GBD sediment implies that heavy metals released by the sulphide oxidation mechanism from the F4 fraction will be recombined via the redistribution process into the loosely binding F2–F3 phases.

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