

Effect of overlying water pH, dissolved oxygen and temperature on heavy metal release from river sediments under laboratory conditions

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Abstract: The heavy metal release experiments were conducted in the laboratory to examine the effects of 3 factors – pH, dissolved oxygen (DO), and temperature on the metal release from sediments taken from the Huangpu River. The metal concentrations in the dry sediments ranged from 0.030 to 0.296 mg g⁻¹ for Cr, 0.021 to 0.097 mg g⁻¹ for Ni, 0.014 to 0.219 mg g⁻¹ for Cu, 0.035 mg to 0.521 mg g⁻¹ for Zn, 0.0002 to 0.001 mg g⁻¹ for Cd and 0.023 to 0.089 mg g⁻¹ for Pb. Most of the metals found in the sediments were in the form of residual fraction, the exchangeable fraction consisted of only a small portion of total metals. The average dissolved metal concentrations in the overlying water during the 13-day period under different conditions were ranging from 0.82 to 1.93 µg L⁻¹ for Cr, 1.08 to 4.19 µg L⁻¹ for Ni, 40.79 to 82.28 µg L⁻¹ for Cu, 20.30 to 29.96 µg L⁻¹ for Zn, 1.57 to 4.07 µg L⁻¹ for Cd, and 22.26 to 75.50 µg L⁻¹ for Pb, respectively. Statistical interpretation of the data indicated that pH (7, 8, 9), dissolved oxygen DO (1.0 and 5.0 mg L⁻¹) and temperature (4, 16, 25°C) had no significant effects on the heavy metal release under the studied conditions. Cu and Pb had the highest release flux, while Cd, Pb and Cu had higher mobility. The main factors controlling the metals release might be the inherent characters of metals and sediments.

Introduction

Heavy metals are ubiquitously distributed in various forms in the environment. In a river system, the heavy metals may be present in sediments, water and organisms, their distribution can change among different compartments dynamically through complicated biogeochemical exchanges under the influences of anthropogenic activities as well as the natural impacts (Zhang et al. 2009, Pokorny et al. 2015).

Sediments in river systems are products of soil erosion and rock weathering, they are heterogeneous assemblages of multitudinous sorbent phases such as organic matter, oxide, carbonates and other minerals, acting as the most important repository and sinks for various contaminants that entered the river environment (Yuan et al. 2014, Ridgway and Shimmield 2002, Yu et al. 2011, Zhang et al. 2014). Under the influence of human activities, large amounts of uncontrolled heavy metal inputs from urban and industrial sources entered the mainstream and tributaries of rivers and accumulated in the sediments (Yang et al. 2009). As environmental conditions alter, heavy metals in the sediments can be released into the water source. The release of trace heavy metals from contaminated sediments may increase their concentrations in the overlying water to undesirable levels at the local scale that affect the environment and ecosystem health due to its characteristics of persistence and toxicity (Payán et al. 2012).

Besides the metal content, the chemical species of the heavy metals often change simultaneously and exhibit different physical and chemical behaviors in terms of chemical interaction, mobility, bio-availability, and potential toxicity (Wang et al. 2014, Ibragimow et al. 2013). The flux and speciation of metals released from sediments are affected by many factors, such as pH, salinity, dissolved oxygen (DO), suspended solids, sediment particle size, etc. (Butler 2009, Atkinson et al. 2007).

It would be useful to predict the changes in metal availability and the fate of the metals, or to evaluate whether these sediments will become a residual source of metals to the river, if the effect of changes in chemical parameters on the release of heavy metals from river sediments was understood. Moreover, the fate and toxicity of heavy metals are greatly dependent on their partitioning between the sediment particles and the pore waters, dissolved heavy metals in the waters are more bio available and toxic than particulate metals (Atkinson et al. 2007).

This research investigated the influence of the overlying water parameters pH, DO and temperature on heavy metal release from a typical river sediments in east China over a 13-day period, and focused on the heavy metals of Cr, Cd, Cu, Ni, Pb and Zn, which are of primary concern for their toxicity and prevalent distribution. The research was conducted in the laboratory under controlled environment.

Materials and methods

Sediments sampling and characterization

The sediments for heavy metal release experiments were taken from a typical East China river and its tributaries. Surface sediments (depth 0–10 cm) were collected from 3 different sites (S1: 31°20'N, 121°22'E; S2: 31°05'N, 121°58'E; S3: 30°58'N, 121°18'E) within 2–3 m of the shoreline. Among the 3 sampling sites, S1 is located in an industrial district; S2 is located in the upstream and is one of the drinking water sources; and S3 is in an agricultural area. At each site approximately 2 Kg sediments were collected by a Stainless Sampler. At the same sampling sites, the water parameters of temperature, pH, oxidation reduction potential (ORP), electrical conductivity (EC), total dissolved solids (TDS) and DO were measured in situ with a portable water analyzer (WTW, Multi3410, Germany).

The collected sediments were sealed in polyethene bottles and labled, kept in an ice box during transportation, and stored in the dark under -20°C in the laboratory. Before use, the sediment was dried at 50°C to remove water, homogenized and passed through a 40 mesh sieve (pore diameter 0.42 mm) to remove large debris. The organic matter was approximately quantified by measuring the total volatile solid (TVS) after burning at 600°C. The particle size distribution (PSD) was analyzed using a Laser Size Analyzer (Microtrac S3500, USA) and the data was used as reported by the analyzer.

The concentrations of the 6 studied heavy metals in the sediments were measured as reported (Li et al. 2015), briefly, 0.1 g of each sediments was digested with 5.0 mL nitric acid, hydrofluoric acid and hydrogen peroxide (volume ratio 3:1:1) in a TOPEX (PreeKem) closed-vessel microwave digestion system. After digestion, the digests were diluted with deionized water supplied by Milli-Q Advantage system to a final volume of 100 mL and centrifuged for 20 min under 4000 rpm. The heavy metals concentrations in the centrifuged solutions were quantified by an Inductively Coupled Plasma Mass Spectrometry (ICP-MS, NexION 300, Perkin Elmer, United States). The operation conditions were: RF power: 1100 w, plasma gas flow rate: 15 L min⁻¹, nebulizer gas flow rate: 0.93 L min⁻¹, auxiliary gas flow rate: 1.2 L min⁻¹, time of sweeps: 20 msec, number of replicates: 3 (Li et al. 2015).

The speciation of the 6 heavy metals in the dried sediments was performed by sequential extraction procedures (A. Tessier 1979). This is designed to separate metals into 5 operationally defined fractions: Exchangeable fraction (EXC); Carbonate bound fraction (CARB); Fe-Mn oxide bound fraction (Fe-Mn); Organic matter bound fraction (OM) and Residual fraction (RES). The heavy metal concentrations in the filtered solutions from each step were detected with ICP-MS as described before.

Heavy metal release

Constructed chambers (15 cm × 10 cm × 20 cm deep) were used to conduct heavy metal release experiments. The whole chamber was covered with aluminum foil (Fig. 1). Homogenized sediment was placed at the bottom of the chamber with a depth of 5.0 cm and 2.0 L deionized water was added as overlying water. For each pH, DO and temperature levels, a single chamber was prepared.

To assess the effect of initial overlying water pH on heavy metal release from river sediments, three pH values of 7, 8 and

9 were employed; adjustments of pH were made by addition of 1.0 M NaOH, and the pH values were monitored by a pH meter (PHS-2C, Leici Company, Shanghai, China). During the pH experiments, the overlying waters were kept at 16°C and DO at 1.0 mg L⁻¹.

The effect of overlying water DO was investigated at two concentrations: 1.0 mg L⁻¹ and 5.0 mg L⁻¹, the DO was maintained by intermittently purging nitrogen gas or stirring gently, and the DO values were monitored by a DO measure meter (JPB-607A, Leici Company, Shanghai, China). During DO experiments, the overlying waters were kept at 16°C and pH 8.

To investigate the effect of temperature on heavy metal release from river sediments, three temperatures of 4°C, 16°C and 25°C were studied and the temperatures were controlled by water bath. During temperature experiments, the overlying waters were kept at pH 8 and DO at 1.0 mg L⁻¹.

At last the release of the 6 heavy metals from 3 river sediments under the actual field conditions was studied. The pH, DO and temperature levels were set as that measured in situ at the sampling sites.

Every day a 5.0 mL overlying water sample was taken with a 5.0-mL pipette and filtered with 0.45 µm cellulose nitrate membrane syringe filters from 3.0 cm above the sediment top in each chamber, the water samples were immediately acidized and subjected to analysis by ICP-MS to measure the heavy metal concentrations. Moreover, the pH, DO and temperature of the overlying waters were also monitored daily over the 13-day test period.

For quality control, each measurement was replicated for 3 times, and the average value was reported as the final data. All measurements including pH, DO, temperature, ORP, EC, TDS, TVS, PSD in the replicate samples were within 10% relative percent deviation. All reported measurements were within the linear calibration curve.

Statistical analysis

Statistical interpretation of the experimental data by analysis of variance (ANOVA) was performed using Microsoft Excel 2000 to evaluate the statistically significant effects of different factors on heavy metals release.

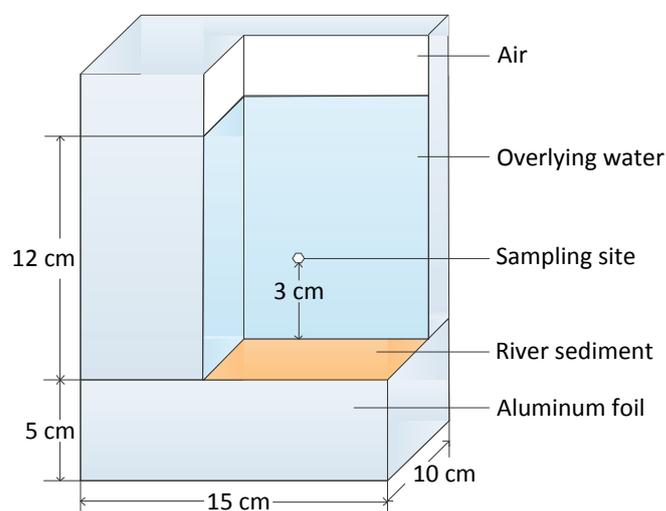


Fig. 1. The experimental setup for heavy metal release

Results and discussion

Characterization of sediment

The characteristics of river water and sediments often exhibit some properties of various sources such as anthropogenic inputs and geological matrix (Yuan et al. 2014), sometimes they could reveal the conditions of the surrounding water environment.

Table 1 shows the characteristics of the overlying water and sediments of the studied river. The pH values of the overlying water in the 3 sites fell in neutral to alkaline range. During sediment collection, the overlying waters had temperatures of 15–17°C, and pH of 7.3–9.7. The overlying water in S2 had the highest DO value of 6.10 mg L⁻¹ indicating relatively clean water quality. S1 water had the lowest DO of 0.27 mg L⁻¹ and the highest TDS value, which possibly indicated more *anthropogenic discharge*. The organic matter contents of the 3 sediments ranged from 39.03 to 44.73 mg g⁻¹. The particle sizes and distribution of the 3 sediments indicated that there were no significant diversities in particle size distribution, they comprised mainly of clay and silt, as well as a small amount of fine sand and very fine sand, of which clay accounted for 27.9 to 33.2%, while silt accounted for 61.7 to 65.3% based on the logarithmic particle size method proposed by Udden and Wentworth (Liu et al. 2014, Udden 1914, Wentworth 1922).

Effect of pH on heavy metal release from sediments

S1 is located in an industrial district; it has the highest metal concentrations. Sediments from S1 were used to investigate the heavy metal release as a function of initial water pH, DO and temperature over time. Heavy metal concentrations in the bulk dry S1 sediment were 0.296 mg g⁻¹ Cr, 0.097 mg g⁻¹ Ni, 0.219 mg g⁻¹ Cu, 0.521 mg g⁻¹ Zn, 0.001 mg g⁻¹ Cd, and 0.089 mg g⁻¹ Pb.

Many researchers reported that at low pH the metal release was more obvious than at middle or high pH, however, the low pH instances are very rare in natural environment (Butler 2009, Watmough et al. 2007, Pérez-Esteban et al. 2013, Yang et al. 2006, Jing et al. 2007). So in this study, only middle and high pH effects were investigated. The release of metals was observed under all 3 studied pH values, in most of the cases, the heavy metal release near pH 9 was less than that of pH~7 and pH~8, but the differences were not distinct (Fig. 2). The dissolved Cr, Ni, and Zn concentrations in the overlying water had a rising tendency during the 13 days, while dissolved Cu, Cd and Pb were fluctuating, which might represent a repetitious metal release-sequestration procedure and this trend indicated higher metal mobilization potential of the three metals.

The release of metals often reflected its mobility; in this research, the average dissolved metal concentrations in the overlying water for Cr, Ni, Cu, Zn, Cd and Pb under pH 8 were 1.22, 3.24, 51.13, 23.29, 2.29 and 60.98 µg L⁻¹, respectively. This sequence did not exactly correspond to their concentrations

Table 1. Selected characteristics of the studied river sediments and water

	S1	S2	S3
Water			
Temperature (°C)	16.0	17.3	15.0
pH	8.2	9.7	7.3
ORP (mV)	362.4	150.0	218.0
EC (ms cm ⁻¹)	0.959	0.694	0.641
TDS (mg L ⁻¹)	0.967	0.451	0.417
DO (mg L ⁻¹)	0.27	6.10	4.85
Sediment			
Organic matter (mg g ⁻¹)	44.34	39.03	44.73
particle size (µm)			
D ₅	1.08	0.91	4.96
D ₅₀	16.38	16.58	15.92
D ₉₅	66.07	81.32	63.41
D _{av}	23.03	29.88	22.14
Clay: <5 µm (%)	29.8	27.9	33.2
Silt: 5–50 µm (%)	64.5	65.3	61.7
Very fine sand: 50–100 µm (%)	4.75	3.67	4.35
Fine sand: 100–250 µm (%)	0.94	1.74	0.76
Medium sand: 250–500 µm (%)	0.0012	1.35	
Coarse sand: 500–1000 µm (%)		0.03	
Very coarse sand: 1000–2000 µm (%)			

D₅: 5% of the total particles have a particle size less than this value. D₅₀: Median particle diameter. D₉₅: 95% of the total particles have a particle size less than this value. D_{av}: average particle diameter

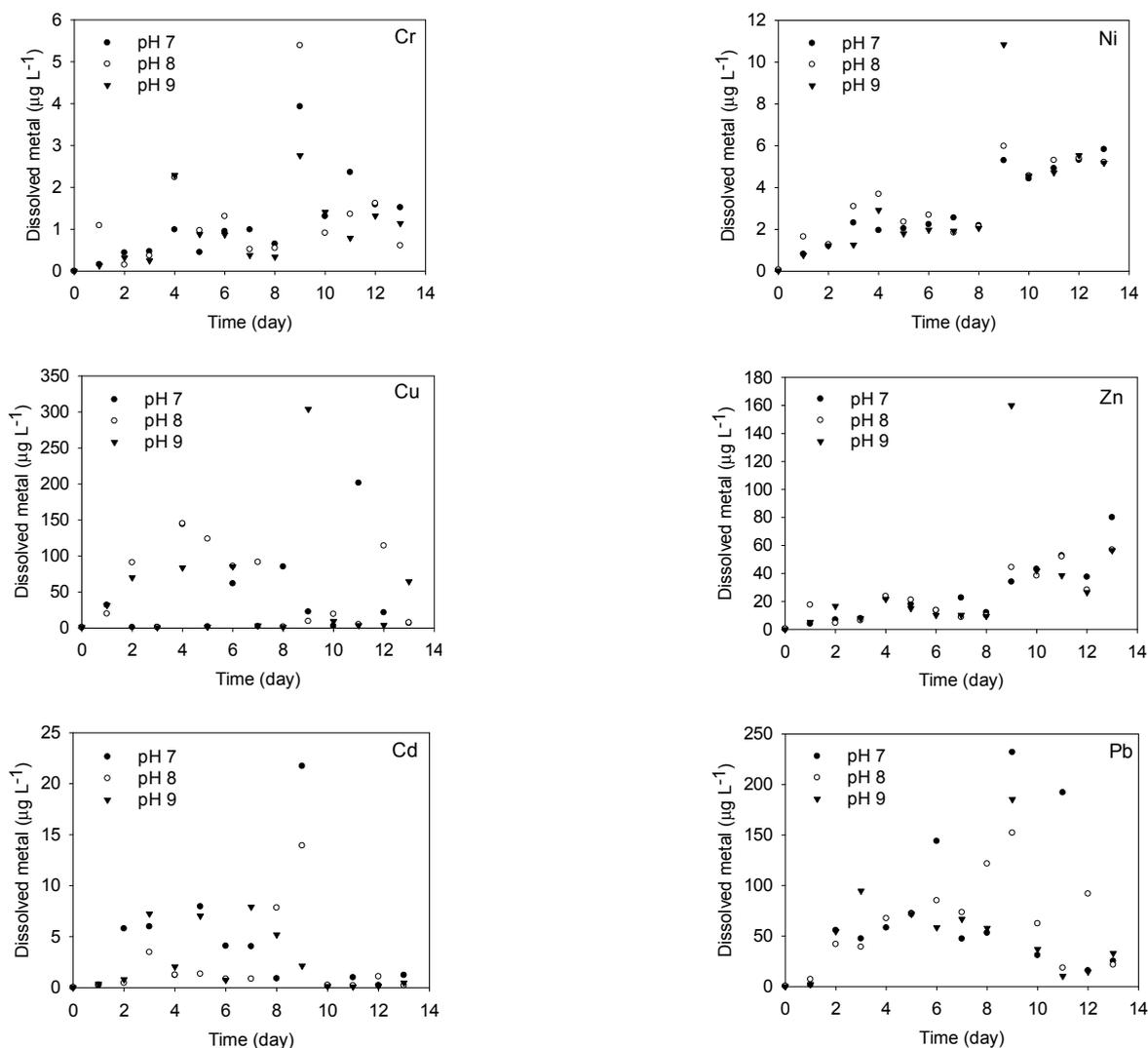


Fig. 2. Concentrations of dissolved metals in the overlying waters over the 13 days at different pH. Temperature: 16°C , DO: 1.0 mg L^{-1}

in the sediment. By comparing the ratio of dissolved metal concentrations in the overlying water to metal concentrations in the sediment, it was found that Cd had the highest value, which might indicate a higher mobility, followed by Pb, then Cu, while Cr had the lowest value, thus implying a lower mobility. This finding was consistent with some previous reports (Usman 2008, Covelo et al. 2007, Equeenuddin et al. 2013, Helios Rybicka et al. 1995).

The metal release mechanism includes desorption, distribution, diffusion, dissolution, ionic exchange, etc. In the pH range of 7–9, the release of heavy metals was usually difficult to occur, there were several reasons to explain this phenomenon: (1) most of the metals existed in the form of insoluble precipitation under neutral pH; (2) the zeta potential of the sediment became more negative under higher pH, thus the specific adsorption stem from the existence of surface charge enhanced accordingly, which inhibited the metal release (Yuan et al. 2007); (3) there was less competition of protons for adsorbing sites in the sediment at higher pH (Yang et al. 2006).

Metal speciation in the sediment was another factor that affected the metal release. Previous studies found that the fraction of metal existing in exchangeable form have the highest mobility. Carbonate bound and Fe-Mn oxide bound

metal fractions were sensitive to pH changes and more likely to mobilize and release from the sediment under low pH (Pérez-Esteban et al. 2013, Equeenuddin et al. 2013). Under the middle to alkaline pH in this research, most of the metals were partitioned into carbonate and Fe-Mn oxyhydroxides, thus the released metals in the overlying water should mostly come from exchangeable fraction. Table 2 shows the percentage distribution of speciation of the 6 heavy metals in S1 sediment. It indicates that the exchangeable fraction accounted for only about 0.06 to 2.63% of all the metals in the sediment, while the residual fraction, which is the most stable one, accounted for about 51.50 to 86.45% of all the metals in the sediment. These reasons contributed to the low release flux for all the 6 studies metals.

Effect of DO on heavy metal release from sediments

In the DO experiment, the pH of overlying water was 8 and the temperature was 16°C (Fig. 3). Generally, the dissolved heavy metals concentrations under low DO of 1.0 mg L^{-1} were slightly higher than those under high DO of 5.0 mg L^{-1} . For instance, in the 13-day period, the dissolved Cr concentrations ranged from 0.15 to $5.39 \mu\text{g L}^{-1}$ with an average value of $1.22 \mu\text{g L}^{-1}$ when DO was maintained at approximately 1.0 mg L^{-1} . At DO of 5.0 mg L^{-1} , the dissolved Cr concentrations ranged

from 0.31 to 2.20 $\mu\text{g L}^{-1}$ with an average value of 0.83 $\mu\text{g L}^{-1}$. For Ni, the average dissolved metal concentration at low DO was 3.24 $\mu\text{g L}^{-1}$, and at high DO this value was 2.91 $\mu\text{g L}^{-1}$. For Zn, the average dissolved metal concentrations were 23.29 and 21.03 $\mu\text{g L}^{-1}$ at low and high DO concentrations, respectively. The exception was Cu, which exhibited a contrary trend of having a lower average dissolved concentration of 51.14 $\mu\text{g L}^{-1}$ at low DO concentration and a higher dissolved concentration of 61.49 $\mu\text{g L}^{-1}$ at high DO concentration. These results were consistent with several other studies (Atkinson et al. 2007, Ho et al. 2012). Atkinson et al. 2007 explained the

reason to be related with the slower oxidative precipitation rate and scavenging of the dissolved heavy metal ions by iron and manganese hydroxide formed from Fe and Mn cations simultaneously released from pore waters under low DO (Atkinson et al. 2007, Santana-Casiano et al. 2004). At high DO, oxygen penetration was stronger, Fe and Mn oxidative precipitation was more rapid, so the generated hydroxide or oxide would adsorb more dissolved heavy metals, thus leading to reduced metals release. For Cu, it became more labile after oxidation under high DO, which caused its faster release (Ho et al. 2012).

Table 2. Percentage distribution of different heavy metals species in S1 sediment

	EXC (%)	CARB (%)	Fe-Mn (%)	OM (%)	RES (%)
Cr	0.06	0.04	6.39	7.07	86.45
Ni	2.63	0.03	14.66	13.60	69.08
Cu	0.30	4.47	0.35	43.38	51.50
Zn	0.27	0.01	21.63	8.23	69.87
Cd	0.74	0.15	5.37	7.42	86.32
Pb	0.32	0.39	11.62	11.47	76.20

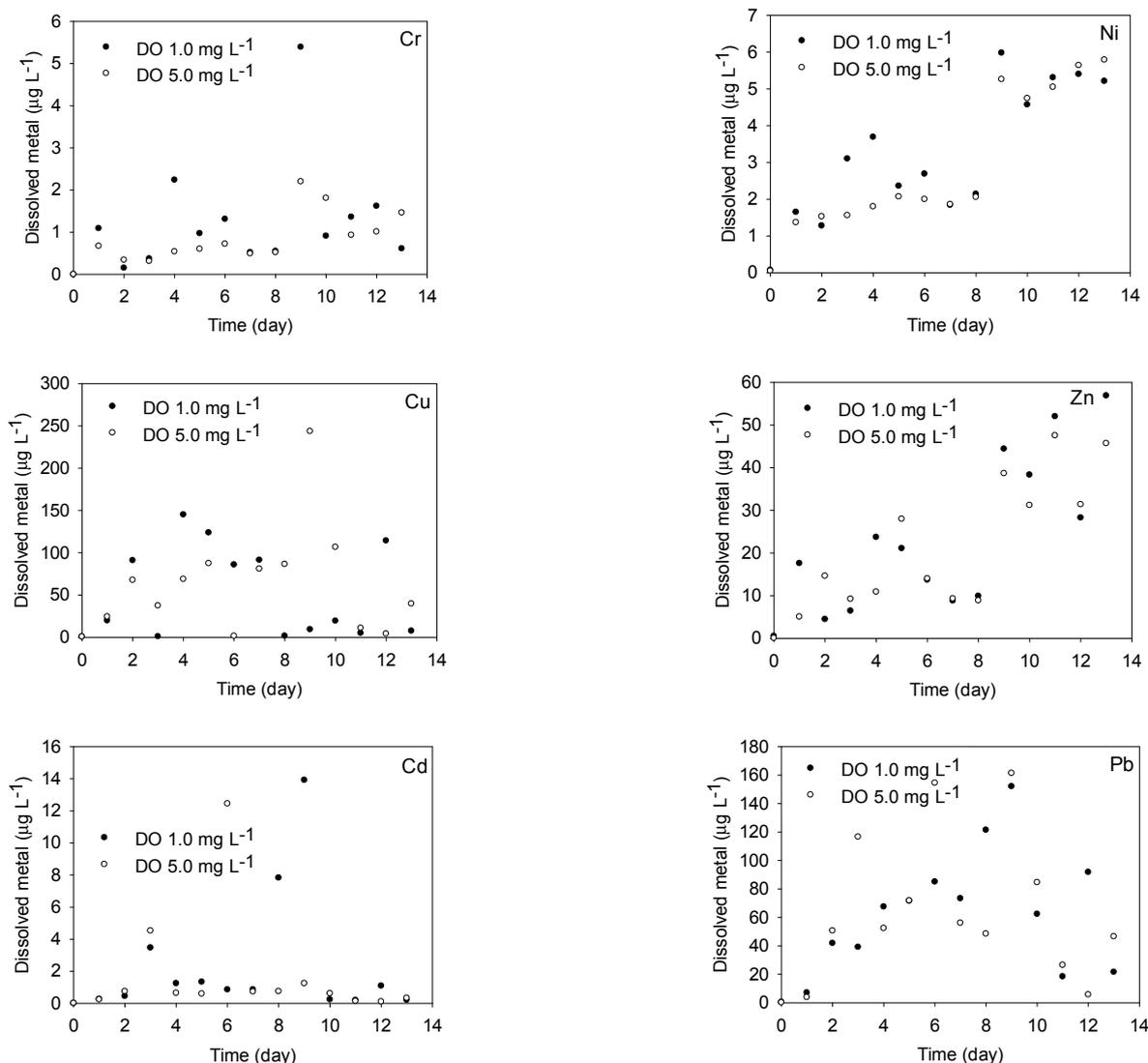


Fig. 3. Concentrations of dissolved metals in the overlying waters over 13 days at different DO. Temperature: 16°C, pH: 8

Effect of temperature on heavy metal release from sediments

In temperate zones, the water parameters such as pH, DO and salinity do not fluctuate considerably, but the water temperature changes wildly with the seasons. Although some research reported that the heavy metals transportation changed with temperature variation (Lourino-Cabana et al., Green-Ruiz et al. 2008), there was also some research reported that the influence of temperature on metal transportation was not evident (Aston et al. 2010, Biesuz et al. 1998, Zhang et al. 2013).

Several studies found that increased temperature resulted in a higher maximum sorption of metals by minerals (Echeverría et al. 2003, Echeverría et al. 2005). On the other hand, the temperature might also affect the bacterial activities, DO, oxidation-reduction reaction rate and molecules diffusion rate, which might pose contrary impact on metal release, thus the impact of temperature on heavy metal release is complex and uncertain.

In the presented study, as shown in Fig.4, taking Cr for example, the average dissolved metal concentrations were 1.17, 1.22 and 0.87 $\mu\text{g L}^{-1}$ at 4, 16 and 25°C, respectively. For Zn, the average dissolved metal concentrations were 22.49 and 23.29 and 20.30 $\mu\text{g L}^{-1}$ at 4, 16 and 25°C, respectively. In brief summary, in the temperature range of 4–25°C, no significant

difference in heavy metal release was noted, which indicated a weak temperature-dependence.

Heavy metal release from river sediments under field conditions

The release of the 6 heavy metals from river sediments under the actual field conditions (Table 1) of the 3 sampling sites was investigated. The sediments from the 3 sites had different metals concentration levels, in the bulk dry S2 sediment, the metals concentrations were 0.030 mg g^{-1} Cr, 0.021 mg g^{-1} Ni, 0.014 mg g^{-1} Cu, 0.035 mg g^{-1} Zn, 0.0002 mg g^{-1} Cd, and 0.023 mg g^{-1} Pb. In the bulk dry S3 sediment, the metals concentrations were 0.167 mg g^{-1} Cr, 0.086 mg g^{-1} Ni, 0.157 mg g^{-1} Cu, 0.367 mg g^{-1} Zn, 0.0006 mg g^{-1} Cd, and 0.041 mg g^{-1} Pb. The results in Fig. 5 show that among the 3 sites, S2 sediment had the lowest metals contents, yet it did not generate the lowest metals release. Actually, the final dissolved concentrations of each metal in the overlying water were very close among the 3 sites, which means the metals release flux had no significant relationship with their concentrations in the bulk sediments. The main factors controlling the metals release might lie in relevant metals properties such as the distribution coefficient, ionic radii, atomic weight, hydrolysis

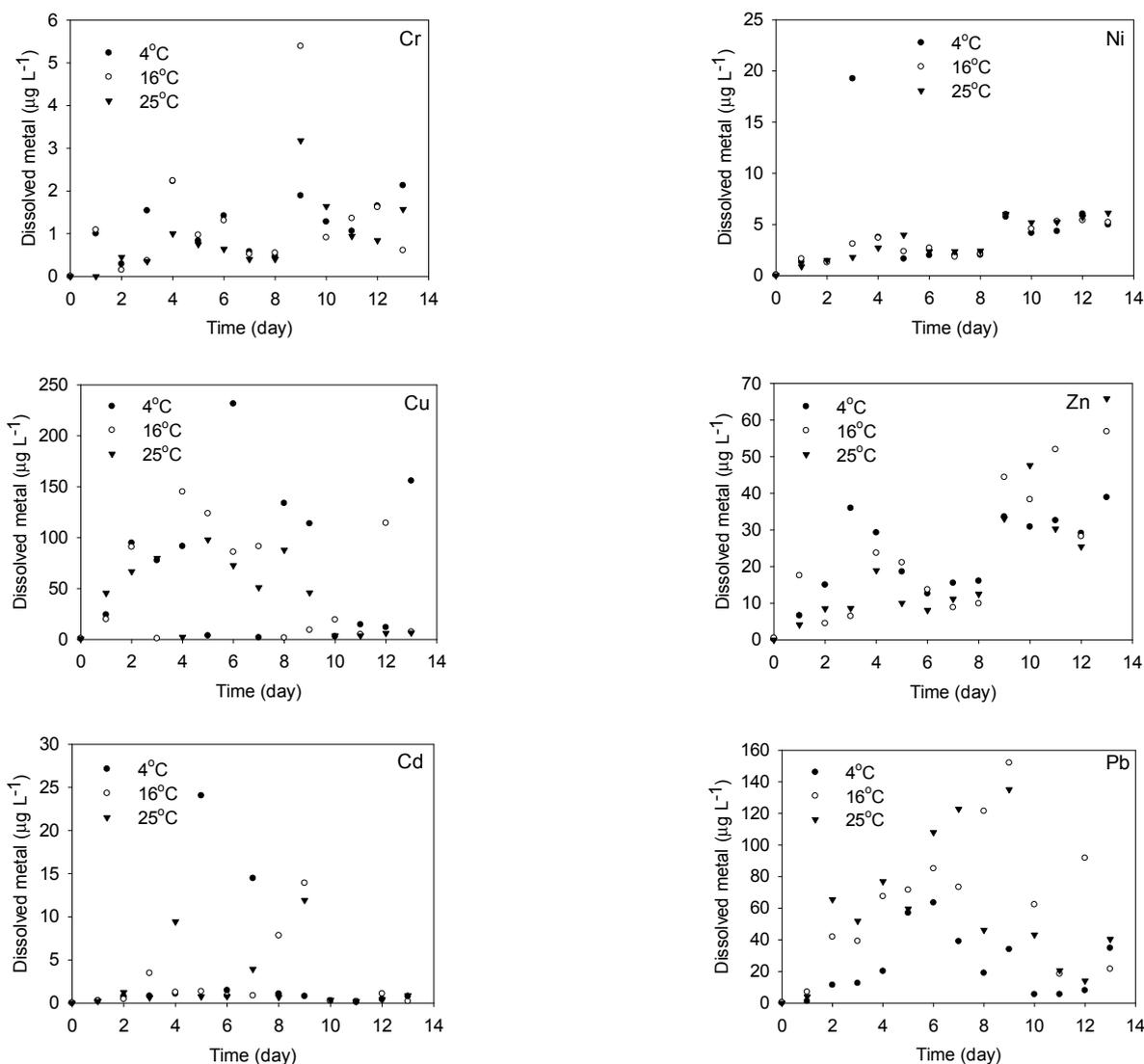


Fig. 4. Concentrations of dissolved metals in the overlying waters during 13 days at different temperatures. DO: 1.0 mg L^{-1} , pH: 8

constant, electonegativity, amphotermism, etc., or other properties of sediments and overlying water such as the organic content, salinity, etc. (Usman 2008, Antoniadis et al. 2007). For example, if metal adsorption was electrostatic, metal ions of lower ionic radii would be more strongly adsorbed, thus exhibiting higher mobility; or if oxidation occurred, the metal fractionation would be modified, thus leading to changed metal mobility.

Compared with researchers working on similar problems, this study mainly concerned the heavy metal release from river sediments in a highly industrialized and commercial big city under mild natural conditions, the 3 selected parameters all fell in a narrow range that was commonly found in surrounding environment. Moreover, very limited research has been done to examine the heavy metal release from the natural river sediment in this city, thus the result provided some valuable information to let people understand the heavy metal distribution in normal situation, and be able to consult local water quality criteria for basic environmental assessment. There are many related studies carried out all over the world that could be referred to, such as those in Australia, USA, India, South Korea, Belgium, and Vietnam, etc. However, these researches mainly focused on metal release from sediments under more or less extreme environments. For example, Atkinson (Atkinson et al. 2007)

studied the release of metals from marine sediments with very high metal concentrations (86, 240, 700, and 3000 mg kg⁻¹ dry weight for Cd, Cu, Pb and Zn). Equeenuddin (Equeenuddin et al. 2013) studied the metal behavior in sediment of an acid mine drainage stream, and the sediments had very low pH of 2.5 and very high electrical conductivity of 1816 μ S/cm. Ho (Ho et al. 2012) studied the metal release from the dredged sediments with the pH range from very low to very high – 2 to 11. Payán (Payán et al. 2012) studied the metal release from contaminated estuarine sediment in a very special case – when subjecting to CO₂ leakages from Carbon Capture and Storage sites.

To better understand the changes of all the measured parameters, the distribution of the dissolved metal concentrations in the overlying water was compared after the longest time – 13 days of experiment and the results were shown in Fig. 6.

Statistical analysis

To assess the statistical significance of the effects due to different factors on the heavy metal release, ANOVA tests were conducted. The statistical summary for the main effects on each metal release is shown in Table 3. Statistically significant effects are indicated by p -value < 0.01 and F -value > F -critical. Generally it could be concluded that under the

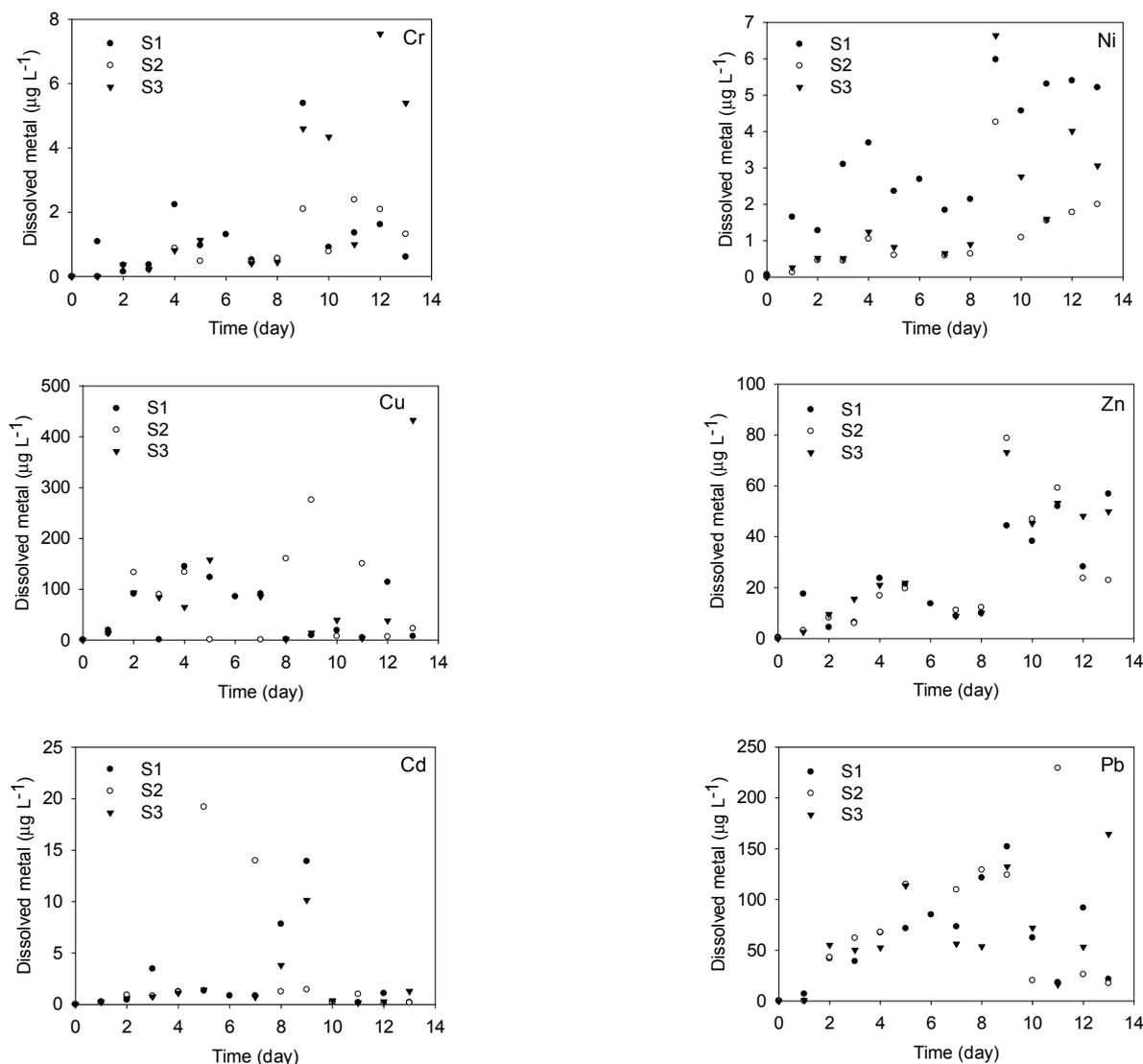


Fig. 5. Concentrations of dissolved metals in the overlying waters during the 13 days under field conditions of the sampling sites

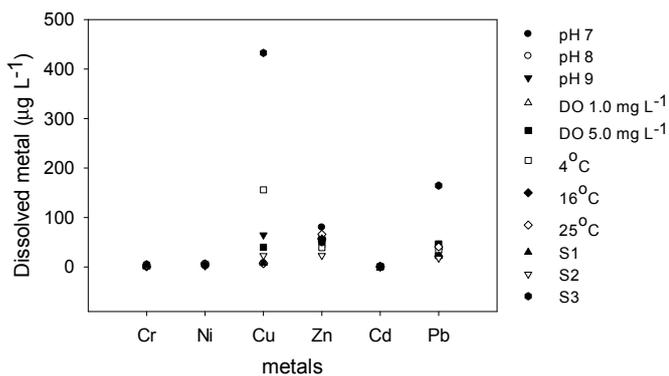


Fig. 6. Concentrations of dissolved metals in the overlying waters at the 13th day under different conditions

Table 3. Statistical summary for effects for each metal ($\alpha=0.05$)

		pH	DO	Temperature
Cr	F-value	1.24	2.04	1.27
	p-value	0.30	0.18	0.30
Ni	F-value	0.42	3.04	0.55
	p-value	0.66	0.10	0.58
Cu	F-value	0.07	0.20	0.96
	p-value	0.93	0.66	0.39
Zn	F-value	0.41	1.43	0.51
	p-value	0.67	0.25	0.61
Cd	F-value	1.00	0.21	0.21
	p-value	0.38	0.65	0.81
Pb	F-value	0.13	0.02	9.90
	p-value	0.88	0.88	0.006
	F-critical	3.37	4.67	3.37

given condition of the present study, there were no statistically significant effects of pH, DO and temperature on the release of each metal from the sediments. Partly because the ranges of the 3 parameters were within a narrow scope, another reason is that the dissolved metal concentrations in the overlying waters were very low, which might lead to wider error margin in detection, thus bring about less clearer tendency.

Conclusions

Most of the heavy metals existed in residual fraction form in the sediments, which ranged from 51.50% to 86.45%, heavy metals only in small part were in exchangeable fraction, which ranged from 0.06% to 2.63% for different heavy metals studied.

The heavy metals release from river sediments occurred during the whole 13-day period. The average dissolved metal concentrations in the overlying water during the 13 days ranged from several to tens micrograms per liter for the 6 target heavy metals. All the heavy metals had a low release flux, among them Cu and Pb had the highest release flux, while Cd, Pb and Cu were observed to have higher mobility than Zn, Ni and Cr,

thus might pose more threats to the surface water and living creature. According to statistical interpretation, the 3 factors of pH, DO and temperature had no significant effects on the heavy metal release under the studied conditions. Under the actual field conditions, the heavy metal release was similar to that under controlled conditions. The main factors that impact the metals release should mostly be the inherent properties of special metals, and other properties of overlying water and sediments.

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