



# Attempt of basin-scale sediment quality standard establishment for heavy metals in coastal rivers



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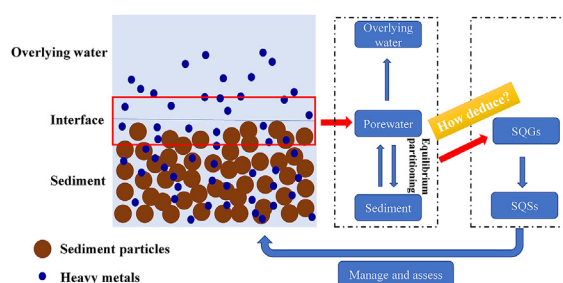
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## HIGHLIGHTS

- Standards of marine sediment and soil quality were inapplicable to river sediments.
- Basin-scale heavy metal SQSs could be used in river sediment management.
- Metal partition between sediment and porewater was affected by outer input.
- Evaluated results confirmed basin-scale heavy metal SQSs were feasible.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Heavy metal sediment quality standards (SQSs) derived from sediment quality guidelines (SQGs) are crucial in risk evaluation and environmental management. However, the establishment of SQSs is quite complex, especially for heavy metals. This study attempted to establish basin-scale SQSs for Cd, Cu, Pb and Zn based on SQGs combined with water quality standards in two coastal rivers in North China, named Jiaolai River (JL) and Jiahe River (JR), respectively. The spatial distribution, fraction, partition coefficients and environmental risk of heavy metals in sediments-porewater were investigated. The results showed that most heavy metals in sediments in JH were higher than those in JL, however, in the porewater, it exhibited an opposite trend. The geochemical fraction showed that most heavy metals in sediments were dominated by residual fraction. The partition of heavy metals between sediment and porewater were mainly affected by both sediment and porewater properties, and exogenous input of heavy metals. Contamination factors showed that Cd in sediment posed high pollution degree; the interstitial water criteria toxicity units and Nemerow Indexes suggested that heavy metal toxicities in porewater were low. The basin-scale heavy metal SQGs were calculated based on porewater quality derived from surface water quality standards using the modified equilibrium partitioning approach. The basin-scale heavy metal SQGs was classified with different grades to deduce the SQSs. Evaluated results of heavy metals in sediments based on SQSs showed lower potential bio-toxic effects in two rivers. In total, basin-scale SQGs for heavy metals were feasible for basin-scale SQSs establishment in coastal rivers.

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## 1. Introduction

Heavy metal pollution in aquatic systems has been a global environmental problem due to heavy metal biotoxicity, non-biodegradability, persistence, and bio-enrichment in food webs (Liu et al., 2018a). When entering the water column, heavy metals are prone to scavenging by suspended particulate matter and are subsequently absorbed and deposited into the sediments (Zhang et al., 2017). However, sediment-associated heavy metals can be released again due to depositional environment changes (Zhang et al., 2018; Liu et al., 2019). Therefore, the actual contamination assessment of heavy metals in sediments is crucial for the environment, aquatic organisms, and human safety.

Sediment quality standards (SQSs) are criteria for sediment quality assessment and in response to regulatory programs in the legal system. Presently, there are no national SQSs for heavy metal presence in rivers for numerous countries. Accordingly, some practical problems fail to be solved, such as quantification of dredging, evaluation of treatment and remediation, and definition of sediment function. In China, although the Marine Sediment Quality (GB18668-2002) divides sediments into three levels based on environmental quality, this standard is not applicable for terrestrial sediments due to the differences of sediment properties and specific protection objectives. Similarly, the Environmental Quality Standard for Soils (GB15618-1995) still is not applicable for sediments due to different conditions; nevertheless, this standard is chronically referenced for sediment management (Chen et al., 2016; Zang et al., 2017).

Generally, SQSs can be induced by corresponding sediment quality guidelines (SQGs), which are widely applied in pollutant control. Heavy metal SQGs are related to properties, bioavailable fractions in sediments, and protection objectives (Chen et al., 2007). Thus, variable sediment properties and different protection objectives result in great difficulties to establish SQGs based on waters on a large regional scale (Gao et al., 2015). Therefore, basin-scale SQGs would be more promising and reasonable. Moreover, a watershed always includes different water function zones based on different protection objectives. Accordingly, the thresholds of heavy metal SQGs in different water function zones are different. Therefore, it is crucial to establish heavy metal SQGs for the establishment of SQSs based on different water function zones. The equilibrium partitioning approach (EqPA) is a preferred method for establishing

heavy metal SQGs based on the thermodynamic equilibrium between sediments and porewater (US EPA, 1989; Liu et al., 2017). This method makes full use of the water quality standards and directly introduces the bioavailability of heavy metals in overlying water into the SQGs (Burton, 2002). Meanwhile, the application of water quality standards in porewater could provide scientific information for water environment management (Huo et al., 2013).

Coastal rivers generally have special geomorphological characteristics resulting from the effects of land-sea interaction. Moreover, coastal rivers always have been taken responsibilities of land-based pollutant discharge, especially for heavy metals (Islam et al., 2018). Therefore, establishing heavy metal SQGs and SQSs of coastal rivers is of great significance to the marine environmental protection. The aims of this study are to (i) investigate the spatial and fraction distribution of Cd, Cu, Pb, and Zn in coastal river sediments; (ii) identify the partition behavior of heavy metals between sediments and porewater; (iii) assess heavy metal contamination in sediments through porewater; (iv) attempt to establish heavy metal SQSs for coastal rivers based on SQGs combined with water quality standards.

## 2. Materials and methods

### 2.1. Study area

Both Jiaolai River (JL) and Jiahe River (JH) are typical coastal rivers located in the Shandong Peninsula (China) (Fig. 1), an area dominated by foothills. The length of the JH is ~140 km, and the total watershed area spans 2296 km<sup>2</sup>. The JL stretches for ~130 km, and covers an area of 5478.6 km<sup>2</sup>. Taking Yaojia Village, Pingdu City as a watershed, the JL flows southward into the Jiaozhou Bay and northward into the Laizhou Bay. The average annual runoffs of JL and JH are  $2.53 \times 10^9$  and  $2.81 \times 10^9$  m<sup>3</sup>, respectively. There are several industrial zones (chemical engineering, electronics, and mechanical manufacturing) located on both sides of JL, causing industrial waste discharge along the river. As irrigated agriculture is along the JL, the no-point source pollution (chemical fertilizers and pesticides) also threatens the river environment. The JH is also mainly used for agricultural irrigation, while it has a lower number of industries along both banks of the river compared to JL. However, with the development of industries and agriculture, the two rivers have been partially polluted by industry-agriculture wastes.

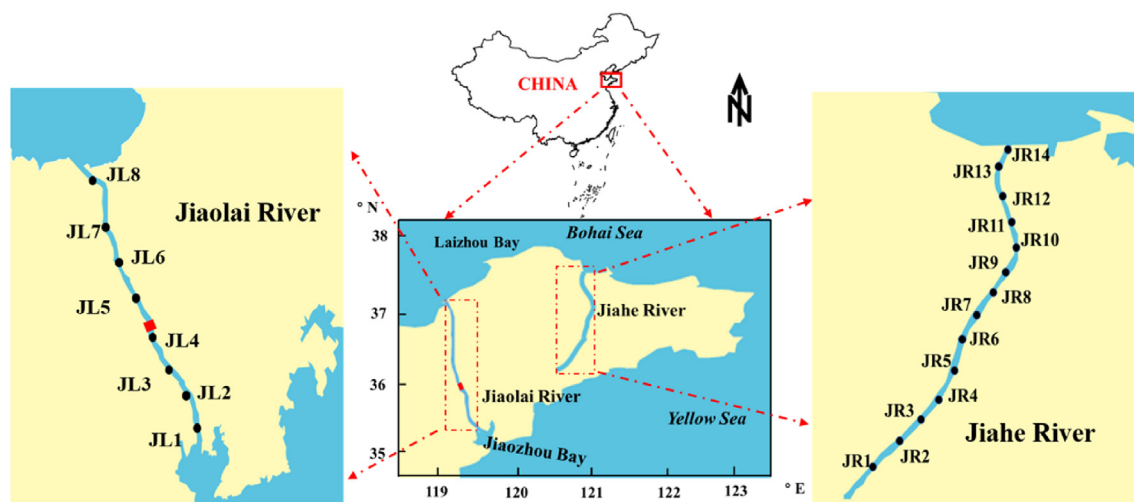


Fig. 1. Location of sampling sites.

## 2.2. Sampling and pretreatment

Eight (JL1–JL8) and fourteen (JH1–JH14) sediment samples were collected from the two rivers in October 2018 (Fig. 1). Transportation, pretreatment, and preservation of samples were strictly conducted according to standard methods (CENPA, 2002). Sampling sites were recognized by a GPS. Surface sediments (0–10 cm) were collected by a Peterson grab sampler and stored at 4 °C. For the pretreatment, a portion of each sediment was freeze-dried, ground, and passed through a 100 µm nylon mesh to obtain consistent physical properties for heavy metals and total organic carbon (TOC). A portion of no-ground sediment was destined for particle size detection. The porewater was obtained by centrifuging (6000 rpm, 30 min), and then filtered with a 0.45 µm cellulose acetate membrane (Han et al., 2014). Every 5 mL volume of filtered porewater was acidified with 0.2 mL 1 M nitric acid to ensure pH < 2, and stored at 4 °C until index determination.

## 2.3. Analytical produce

Dissolved Cd, Cu, Pb, and Zn in porewater were analyzed using inductively coupled plasma-mass spectrometry (ICP-MS, PerkinElmer NexION 300Q, USA). For total heavy metal concentration analysis, the pretreated sediments (0.2500 g ± 0.0001) were transferred into Teflon tubes and digested with a mixed solution of HNO<sub>3</sub> + HF + HClO<sub>4</sub> (8: 5: 2, v/v) in a microwave digestion instrument (Tank Basic, Sineo Microwave Chemistry Technology Co., LTD, China) and then analyzed by ICP-MS. The particle sizes were tested by a laser particle size analyzer (Malvern Mastersizer, 2000F, UK). The TOC was measured using a TOC analyzer (Vario Micro cube, German). Acid volatile sulfides (AVS) were analyzed following a modified cold diffusion procedure (Sheng et al., 2015), where an aliquot of sediment (~10 g wet weight) was purged for 1 h with 15 mL of 9 M HCl under N<sub>2</sub> flux. The S concentrations for AVS trapped in ZnS were measured using iodometric titration. The supernatant of a 1: 5 (w/v) suspension extracted with ultra-pure water was employed for the determination of electric conductance (EC), pH, and salinity of sediments.

The heavy metal fraction analysis was conducted using modified Bureau Communautaire de Référence (BCR) sequential extraction procedure (Rauret et al., 1999). The metal fraction was divided into the acid-soluble fraction (F1), reducible fraction (F2), oxidizable fraction (F3), and residual fraction (F4); determination details are listed in Table S1.

## 2.4. Quality assurance and quality control

All determinations were conducted in accordance with strict quality control. Procedure blank samples and reagent blanks were applied across the entire process. The precision of analysis for dissolved heavy metals in porewater was validated through heavy metal solution standards with control, and recoveries were within 92%–107%. For total and fractionation analysis, sediment standard reference materials GBW 40376 and GBW 07311 (GSD-11) were used for quality assurance. The recoveries and relative deviations were 90%–108% and <8%, respectively.

## 2.5. Heavy metal SQGs calculation

The EqPA is a popular method for establishing SQGs of heavy metals based on the thermodynamic equilibrium between sediment and porewater (US EPA, 1989; Liu et al., 2017). According to the theory of EqPA, water quality standards can be applied to porewater contaminants; the SQG values can be defined by the

concentration of contaminants in the sediment that was in equilibrium with the concentrations of porewater quality standards derived from surface water standards (Chen et al., 2007). Considering the bioavailability and mobility of heavy metals, the SQGs were calculated with normalized EqPA as follows (Gao et al., 2015):

$$SQG = K_p \times WQC_i + M_{R,i} + M_{AVS,i} \quad (1)$$

$$SQG = K_p \times WQC_i + M_{R,i} \quad (2)$$

$$K_p = C_T \times (1 - A\%) / C_{i,w} \quad (3)$$

$$M_{AVS,i} = AVS \times M_i \times C_{T,i} / \sum_{i=1}^{n=5} C_{T,i} \quad (4)$$

where  $K_p$  is the partitioning coefficient between sediments and porewater;  $WQC_i$  are the corresponding water quality standards from the optimized environmental quality standards for surface water of China (EQSSW) (CSEPA, 2002), which match the classification of water quality standards of porewater based on different water function zones (Table 1);  $M_{R,i}$  is the residual fraction concentration of heavy metal in sediments (mg kg<sup>-1</sup>);  $M_{AVS,i}$  is the concentration of heavy metals (Cu, Cd, Zn, and Pb) associated with AVS in sediments (µmol g<sup>-1</sup>);  $C_T$  is the total concentration of heavy metals in sediments (mg kg<sup>-1</sup>);  $A$  is the percentage of residual heavy metals in the sediments (%); and  $C_{i,w}$  is the concentration of heavy metals in porewater (µg L<sup>-1</sup>).

## 2.6. Assessment of heavy metal contamination

### 2.6.1. Interstitial water criteria toxicity units

The toxicity level of porewater was assessed using the method of interstitial water criteria toxicity units (IWCTU) and Nemerow indexes (NI) (Tang et al., 2016), which were calculated as follows:

$$IWCTU_{Me} = [Me]_{i,w} / FCV_{Me} \quad (5)$$

**Table 1**

The classification of original and optimized quality standards for sediment porewater (mg L<sup>-1</sup>).

Threshold	Cd	Cu	Pb	Zn
Original standards for surface water				
I	0.001	0.01	0.01	0.05
II	0.005	1.0	0.01	1.0
III	0.005	1.0	0.05	1.0
IV	0.005	1.0	0.05	2.0
V	0.01	1.0	0.10	2.0
Optimized standards for sediment porewater				
I	0.001	0.01	0.01	0.05
II	0.005	0.5	0.05	1.0
III	0.01	1.0	0.10	2.0

**For surface water:** Grade I: it is applicable to drinking source water and national nature reserves; II: the first-class protection area of surface water source area of centralized drinking water, habitat of rare aquatic organisms, fishery and shrimp production field, rope bait field of larvae and juveniles; III: secondary protection areas of surface water sources, wintering grounds of fish and shrimp, migration passages, aquaculture areas and other fishery waters and swimming areas of centralized drinking water; IV: general industrial water use areas and recreational water areas where human body is not in direct contact; V: agricultural water use areas and waters with general landscape requirements.

**For sediment porewater:** I: the applicable area is same as Grade I of surface water; II: the applicable area is same as Grade II and III of surface water; III: the area is same as above Grade IV and V.

$$NI = \left\{ \left[ (IWCTU)_{max}^2 + (IWCTU)_{mean}^2 \right] / 2 \right\}^{1/2} \quad (6)$$

where  $[M_e]_{i,w}$  is the concentration of dissolved heavy metal  $i$  in porewater ( $\mu\text{g L}^{-1}$ ), and  $FCV_{M_e}$  represents the final chronic value of heavy metals ( $\mu\text{g L}^{-1}$ ) (US EPA, 2002, 2005). If  $IWCTU > 1$ , biota may suffer from potential toxicity. According to the NI values, the porewater quality can be classified into five classes, as shown in Table S2.

#### 2.6.2. Pollution load index and contamination factor

The pollution load index (PLI) and contamination factor (CF) were obtained using the following formulas:

$$CF = C_{M_e} / C_B \quad (7)$$

$$PLI = \sqrt[n]{(CF_1 \times CF_2 \times \dots \times CF_n)} \quad (8)$$

where CF is the contamination factor based on the proportion of heavy metal concentration ( $C_{M_e}$ ) to its corresponding background value ( $C_B$ ) ( $\text{mg kg}^{-1}$ ) (Shi et al., 2016), and it is interpreted according to four degrees of contamination, as presented in Table S2; PLI indicates the comprehensive contamination of measured heavy metals in sediments;  $n$  represents the number of selected heavy metals.

#### 2.6.3. Mean sediment quality guideline quotient (mSQG-Q)

The mSQG-Q proposed by Long and MacDonald (1998) has been a widely applied method (Cheng and Yap, 2015; Liu et al., 2017). It considers the bioavailability and bio-toxic effects of heavy metals, and its credibility, sensitivity, and robustness are higher than those of other methods (Caeiro et al., 2015). The mSQG-Q was obtained through computing mean quotients for multiple heavy metals by following formula:

$$\text{mSQG} - Q = \sum_{i=1}^n (C_i / \text{SQG}_i) / n \quad (9)$$

where  $C_i$  represents the concentration of heavy metal  $i$  in sediments ( $\text{mg kg}^{-1}$ );  $n$  is the number of heavy metals;  $\text{SQG}_i$  is the SQG of heavy metal  $i$  ( $\text{mg kg}^{-1}$ ). Based on mSQG-Q values, the sediments' quality is defined as:  $\text{mSQG} - Q \leq 0.1$ , unimpacted or lowest potential bio-toxic effects;  $0.1 < \text{mSQG} - Q < 1$ , moderate impact potential bio-toxic effects;  $\text{mSQG} - Q \geq 1$ , high impact potential bio-toxic effects.

### 2.7. Statistical analysis

The data was statistically analyzed using SPSS software. Data normality was detected by the Kolmogorov-Smirnov (K-S) test. Correlations were calculated with Pearson correlation analysis, and a  $P < 0.05$  was regarded as a significant correlation level.

## 3. Results and discussion

### 3.1. The physicochemical properties and heavy metal distribution of sediments

The sediments' properties are shown in Table S3. The pH of sediments in JL and JH ranged from 4.78 to 7.90 and 7.73 to 8.20, respectively. The low pH in JL was related to industrial acid wastewater discharge along river. The mean values of salinity and EC in sediments were 1.0‰ and  $1.8 \text{ mS cm}^{-1}$  in JL, 1.3‰ and

$2.3 \text{ mS cm}^{-1}$  in JH, respectively. The coefficient of variation (CV) of EC and salinity in JH was higher than those in JL, indicating a greater variation of these parameters along JH. With regard to particle size, sediments were all dominated by sand, followed by silt and clay, which was primarily due to rainwater erosion and transport effects. The average TOC concentration in JH was 0.94% (0.65%–1.97%), and high values were primarily observed in the middle reach, where the slow water current contributed to organic matter deposition. Compared to JH, TOC in JL was low (0.25%), which was attributed to frequent artificial disturbance of sediments, such as dredging and sand excavation. Furthermore, the AVS in two rivers was uneven based on CV analysis, because it was greatly affected by TOC, redox condition, and sulfate reducing bacteria (Allen et al., 1993). As shown in Table S3, the inhomogeneity of TOC in JH and JL was partially responsible for this phenomenon. Moreover, the heavy metal SQGs was closely related to the AVS in sediments, because AVS could combine with sulfophilic metals (Cd, Cu, Ni, Pb, and Zn) to form insoluble metal sulfides, significantly reducing their bioavailability (Toro et al., 1991).

As shown in Table S4, the average total heavy metal concentrations ( $\text{mg kg}^{-1}$ ) in sediments in JL decreased according the following sequence: Zn ( $62.59 \pm 49.56$ ) > Pb ( $14.05 \pm 3.62$ ) > Cu ( $10.57 \pm 2.66$ ) > Cd ( $0.33 \pm 0.08$ ). The CV values of Cd, Cu, and Pb were low (<25%, Table S4), exhibiting a homogeneous distribution in JL. A different spatial distribution was observed for Zn (CV 79.18%), indicating that Zn was variable at different sites. The mean total heavy metal concentrations ( $\text{mg kg}^{-1}$ ) in JH followed the order: Zn ( $98.96 \pm 45.61$ ) > Cu ( $34.08 \pm 10.81$ ) > Pb ( $30.90 \pm 6.75$ ) > Cd ( $0.61 \pm 0.67$ ) (Table S4), which indicated a great difference compared to JL. In particular, the Cd and Zn in JH 13 and JH14 were approximately three times higher than at other sites (Fig. 2(a)), which was probably due to their location at the estuary, where heavy metals were more easily scavenged (Pan and Wang, 2012). Moreover, regarding CV values, the highest variation was found in Cd (109.80%), whereas Pb exhibited relatively a homogeneous distribution (21.83%).

Usually, grain size and TOC are major factors for the regulation of heavy metal spatial distribution in sediments (Ma et al., 2019). According to Pearson's correlation analysis (Table S5), the TOC exhibited a significant positive correlation with Zn ( $P < 0.05$ ) in JL, suggesting the distribution of Zn was controlled by TOC in this area. The fine grain size (silt + clay) of sediments in JH was only significantly correlated with Pb ( $P < 0.05$ ), indicating that the distribution of Pb in sediments in JH was controlled by grain size (Soliman et al., 2019). Furthermore, the grain size and TOC showed a weak correlation with most heavy metals in this study. This phenomenon suggested that heavy metals in the sediment were mainly contaminated by point sources (such as industrial and municipal wastewater) (Neyestani et al., 2016).

Geochemical fractions of heavy metals in JL are portrayed in Fig. 2(a). Among the four fractions, the F1 represents the bioavailable fraction, and a sum of F1, F2, and F3 is considered as the mobile fraction (Rauret et al., 1999). All heavy metals were dominated by the F4, on average, and approximately 48% of Zn, 72% of Cu, 79% of Cd, and 65% of Pb fall into this fraction. The high percentage of F4 showed a minor ecological risk posed to biota. Only Zn was presented at a high proportion at F1 (23%), which indicated high potential ecological risk, while the remaining heavy metals occupied minor proportions (<7%). Higher percentages of Pb (28%) and Zn (21%) were found in F2. Generally, all heavy metals accounted for the lower proportions (<10%) in F3 at all sites. Overall, average proportions of mobile fractions of heavy metals in sediments in JL occurred in the descending order: Zn (52%) > Pb (35%) > Cu (28%) > Cd (20%).

The fraction partitions in JH were less uniform than those in JL.



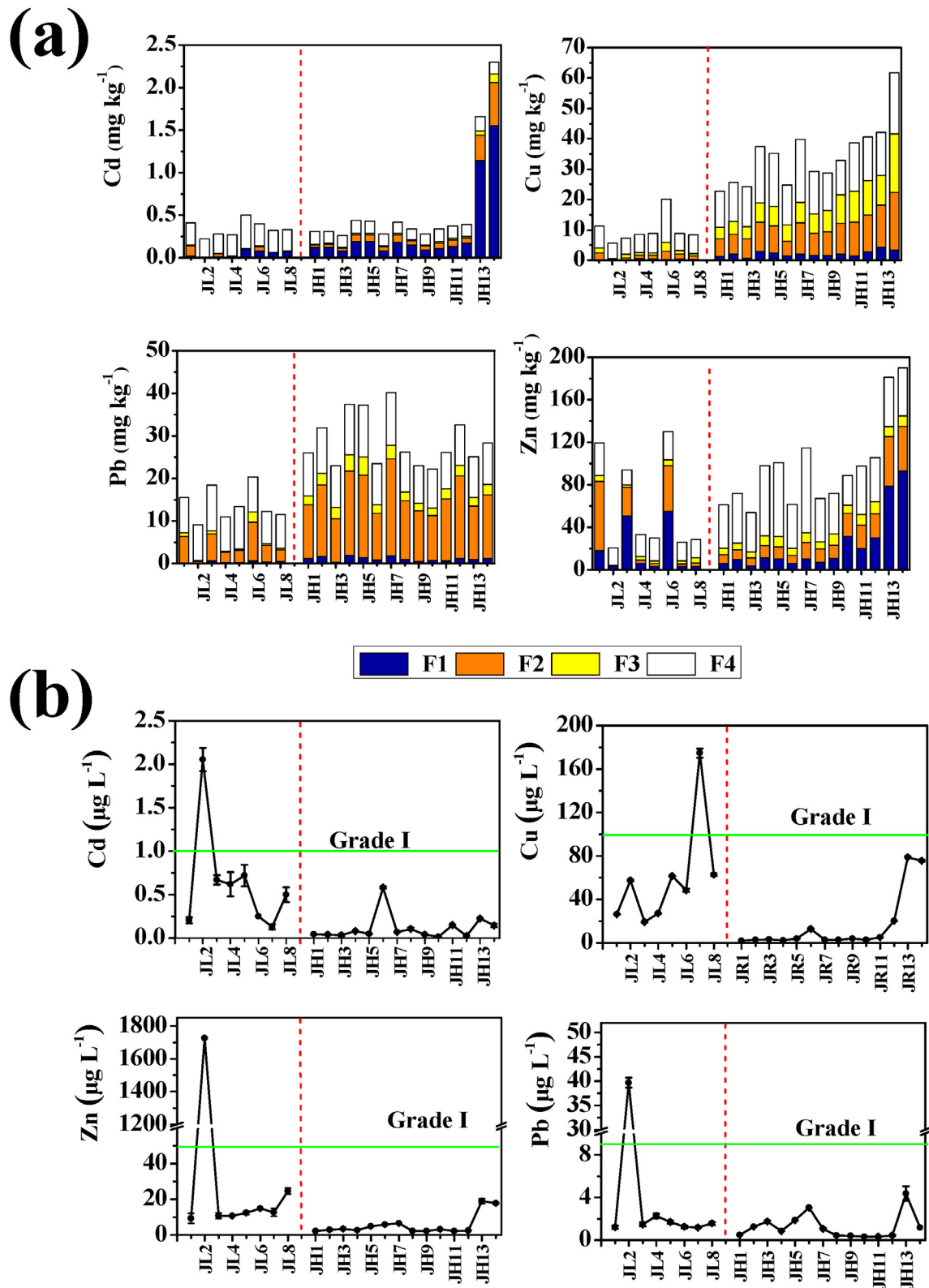


Fig. 2. Total and different chemical fraction concentrations of heavy metals in sediments (a); heavy metal concentrations in porewater in JL and JH (b).

The Cu showed the highest percentages of F4 (45%), while F2 and F3 also accounted for higher proportions (>20%). For Pb, F2 was the dominated fraction (52%), followed by F4 (36%), F3 (9%), and F1 (4%). This result was consistent with previous reports (Morillo et al.,

2004; Cu and Lin, 2016), suggesting that Pb was mainly associated with F2. Notably, both Pb (<4%) and Cu (<6%) had low percentages in F1 at all sites. Furthermore, the dominant fraction of Cd was F1 (43%), and a similar result was found in the surface sediments from

the Jinjiang River (Liu et al., 2018b) and Jiaozhou Bay, China (Lin et al., 2016), indicating that Cd was primarily associated with anthropogenic sources (Alves et al., 2018). Similar to Cu, the dominated fraction of Zn was also F4 (54%), meanwhile, F2 accounted for a high proportion (19%). For the mobility sequence in sediments in JH, Pb showed the highest proportion (64%), followed by Cd (63%), Cu (55%), and Zn (46%).

### 3.2. Heavy metal concentrations and contamination in porewater

The heavy metal concentrations in porewater are presented in Fig. 2(b). Mean concentrations ( $\mu\text{g L}^{-1}$ ) in JL were in the following descending order: Zn ( $227.72 \pm 605.55$ ) > Cu ( $59.74 \pm 49.51$ ) > Pb ( $6.30 \pm 13.49$ ) > Cd ( $0.64 \pm 0.61$ ). Cd, Pb, and Zn concentrations in JL were higher than Grade I of the EQSSW (CSEPA, 2002) at site JL2, including Cu at site JL7. The average values ( $\mu\text{g L}^{-1}$ ) in JH were in the descending order of: Cu ( $15.68 \pm 26.58$ ) > Zn ( $5.53 \pm 5.65$ ) > Pb ( $1.27 \pm 1.18$ ) > Cd ( $0.11 \pm 0.15$ ). Compared to JL, ranges and average values in JH were low, and all metals at all sites satisfied Grade I.

Generally, heavy metals weakly associated (mobile fractions) with sediments are prone to their re-release into porewater (Ji et al., 2018). In this study, although heavy metals in porewater in JL were higher than those in JH, the total content of heavy metals and their mobile fractions in sediments of JL was lower than that in JH (Fig. 2(a)). This phenomenon was probably related to the external input of heavy metals with industrial wastewater discharge along the river, resulting in high concentrations in porewater. Furthermore, lower TOC and coarse sediments (grain size) in JL (Table S3) were also not favorable to trap heavy metals from porewater (Ma et al., 2019). Additionally, the lower pH in JL could also promote the release of heavy metals due to cation exchange and competitive adsorption increase (Yao et al., 2016; Wang et al., 2018).

Heavy metals in porewater represent the bioavailable part in sediments, which can be considered as essential indicators of toxic effects (Tang et al., 2016). The IWCTU can reflect the degree of heavy metal contamination in the porewater. In Table S6, the heavy metals in porewater in JL exhibited a low contamination degree on the whole. IWCTU values > 1 were only found at Zn (JL2) and Cu (JL7), posing long-term toxicity effects to aquatic organisms. For the NI, high values were observed at JL2 (1.25) and JL7 (1.70), suggesting suffering from a slight contamination degree. The NI values at other sites were below 1, implying no impact at these sites. In JH, the IWCTU values were <1 except for Cu at JH12–JH14. The NI < 1 was observed at JH1–JH11, indicating no adverse impacts at these sites. However, the NI values in JH13 and JH14 were 3.89 and 3.69, respectively, indicating strong contamination levels. The values of the IWCTU and NI indicated that although several heavy metals posed some adverse effects to biota, the pollution and toxicity of heavy metals in porewater was low at most sites.

### 3.3. Distribution relationships of heavy metals in porewater and sediments

Heavy metals consistently exhibit a migration trend from sediments to porewater, affected by their physicochemical properties and environmental factors (Ji et al., 2018). In this study, the ratio of total concentrations of heavy metals in sediments and porewater, as the partition coefficient ( $K_d$ ), was used to determine the heavy metal migration ability. The calculation was expressed as:

$$K_d = C_s / C_w \quad (10)$$

where  $C_s$  ( $\mu\text{g g}^{-1}$ ) and  $C_w$  ( $\mu\text{g L}^{-1}$ ) are heavy metal contents in sediments and porewater at equilibrium (Hierro et al., 2014). The lower  $K_d$  values indicate that heavy metals in sediments easily

**Table 2**

The partition coefficients of heavy metals between sediments and porewater.

	Cd	Cu	Pb	Zn
JL				
Min	0.11	0.05	0.23	0.01
Max	2.46	0.42	16.13	4.11
Mean	1.05	0.24	8.97	3.30
JH				
Min	0.43	0.56	6.18	10.10
Max	21.25	15.31	83.80	48.73
Mean	8.52	7.60	43.34	25.51

migrate to porewater (Ji et al., 2018).

The  $K_d$  values in JL and JH are summarized in Table 2. Generally, the heavy metals occupied by higher mobile fraction in sediments would exhibit lower  $K_d$  values, as they are easily released into porewater (Ji et al., 2018). However, an unusual phenomenon was observed for Zn and Pb in JL. The mobile fraction concentrations of Zn and Pb were higher in JH, however, their  $K_d$  values were also higher. The partition equilibrium of heavy metals between sediments and porewater usually depends on complexant properties in porewater, external heavy metals import, and sediment composition, except for mobile fractions (Wang et al., 2018). The reason of this phenomenon in this study was due to external Pb and Zn discharge into porewater (as discussed in Section 3.2), which altered the original partition equilibrium between sediments and porewater. Similar results were also observed for Pb and Zn in JH. Generally, the partition coefficients ( $K_d$ ) of heavy metals in JH were higher than those in JL. This was because heavy metals in porewater in JL were higher than those in JH, while in sediments they were lower than JH.

### 3.4. Heavy metal SQGs

Generally, there are many water functional zones in a specific watershed, and to ensure this, heavy metal SQGs need to be classified into different grades to avoid the effects of heavy metal release. In this study, the grade classification of heavy metal SQGs was set based on optimized EQSSW for porewater (Table 1). The original heavy metal SQGs of the same grade varied greatly among different sites, especially for Zn (e.g., 68.49–1610.73  $\text{mg kg}^{-1}$ ) in both JL and JH. Apart from differences in the chemical fractionation and concentration of heavy metals, variations of AVS, pH, and TOC of sediments were also responsible for the large differences in heavy metal SQGs of the same grade. For example, heavy metal SQGs can be greatly affected by AVS normalization, and AVS concentrations were variable at different sites (Table S3), resulting in differences between sites. Therefore, to ensure the accuracy of heavy metal SQGs establishment, some outliers of SQGs were eliminated at several sites, and subsequently the geometric average value of heavy metal SQGs at rest sites was calculated to obtain basin-scale SQGs (Table 3). Some great variations of SQGs between different metals were also observed. This was due to different water quality standards and partitioning coefficients (Chen et al., 2007). Similarly, all heavy metal SQGs exhibited large differences between JL and JH (Table 3). Moreover, the same heavy metal SQGs between different grades also exhibited large differences. Hence, it was not feasible to establish SQGs for rivers on a large scale, and establishment for basin-scale heavy metal SQGs is deemed more realistic and reasonable.

The large variations of heavy metal SQGs values compared to other studies (Table 3) were mainly ascribed to the different SQG derivation methods and water quality guidelines, apart from sediment properties and bioavailability of heavy metals. Moreover,

**Table 3**Comparison of basin-scale SQGs between study area and area of other studies (mg kg<sup>-1</sup>).

	Cd	Cu	Pb	Zn	References
JL					
SQGs-I <sup>a</sup>	3.82	62.94	293.20	413.19	This study
SQGs-II <sup>b</sup>	5.16	67.33	409.70	1606.48	
SQGs-III <sup>c</sup>	10.65	136.55	536.70	2677.16	
JH					
SQGs-I <sup>a</sup>	6.56	147.13	453.97	817.00	This study
SQGs-II <sup>b</sup>	22.25	295.55	1264.41	10,113.93	
SQGs-III <sup>c</sup>	41.42	3159.97	2264.57	19,879.93	
Dianchi Lake (China)	5.56	53.8	67.8	106.4	Chen et al. (2007)
Xiangjiang River (China)	3.56	92.28	102.79	1495.09	Liu et al. (2017)
Chaohu Lake (China)	23.90	56.95	362.93	74.68	Huo et al. (2013)
Yangtze River (China)	8.35	73.05	197.41	4120.12	Gao et al. (2015)
UK	7.68	34	32.68	191.4	Webster and Ridgway (1994)
Probable effect concentration <sup>d</sup>	4.98	149	128	459	MacDonald et al. (2000)
Risk screening value <sup>e</sup>	0.8	200	240	300	Ministry of Ecological and Environment of China, 2018
CMSQS-I	0.50	35.0	60.0	150.0	CSBTS China State Bureau of Quality and Technical Supervision, 2002
CMSQS-II	1.50	100.0	130.0	350.0	
CMSQS-III	5.00	200.0	250.0	600.0	

a, b and c: their application areas are sediments from water function zones which applied to the Grade I, II and III porewater quality standards, respectively; d: probable effect concentration below which adverse effects are not expected to occur; e: risk screening value for soil (paddy field) below which contamination of agricultural land are not expected to occur.

different protection objectives, the non-uniformity of testing environmental factors, and acquiring biological effects also could cause different SQGs (Chen et al., 2007). In JL and JH, the heavy metal SQGs were also variable compared to CMSQS (CSBTS, 2002) and the risk screening value (Ministry of Ecological and Environment of China, 2018), which indicated that standards of marine sediment and soil quality were not suitable for river

sediment management and control.

### 3.5. Application and feasibility of SQGs for assessment of heavy metal contamination

In this study, the calculated basin-scale heavy metal SQGs was tentatively considered as corresponding SQGs after normalization

**Table 4**

The mSQG-Q for heavy metals in sediments in JL and JH.

Sites	SQG-Q				mSQG-Q
	Cd	Cu	Pb	Zn	
JL1	0.11	0.18	0.05	0.25	0.15
JL2	0.06	0.09	0.03	0.04	0.05
JL3	0.07	0.12	0.06	0.23	0.12
JL4	0.07	0.14	0.04	0.08	0.08
JL5	0.13	0.14	0.04	0.07	0.10
JL6	0.10	0.32	0.07	0.31	0.20
JL7	0.09	0.14	0.04	0.06	0.08
JL8	0.08	0.13	0.04	0.07	0.08
Mean	0.09	0.16	0.05	0.14	
JH1	0.05	0.15	0.06	0.07	0.27
JH2	0.05	0.18	0.07	0.09	0.26
JH3	0.04	0.17	0.05	0.07	0.16
JH4	0.07	0.26	0.08	0.12	0.61
JH5	0.07	0.24	0.08	0.12	0.61
JH6	0.04	0.17	0.05	0.08	0.59
JH7	0.06	0.27	0.09	0.14	0.65
JH8	0.05	0.20	0.06	0.08	0.59
JH9	0.04	0.20	0.05	0.09	0.51
JH10	0.05	0.22	0.05	0.10	0.66
JH11	0.05	0.26	0.06	0.12	0.75
JH12	0.06	0.28	0.07	0.13	0.86
JH13	0.25	0.28	0.06	0.22	0.46
JH14	0.35	0.43	0.06	0.23	0.48
Mean	0.09	0.24	0.06	0.12	

The mSQG-Q:  : unimpacted or lowest potential bio-toxic effects;  : moderate impact potential

bio-toxic effects.

and optimization, and the corresponding SQSs were employed to assess heavy metal contamination with the method of mSQG-Q. The evaluation results of mSQG-Q for heavy metals in sediments are shown in Table 4. In JL, all heavy metals were below their SQSs-I (SQG-Q < 1) at all sites. This demonstrated that heavy metal pollution in sediments in JL was low. For mSQG-Q, the SQSs-I was applied to assess the heavy metal risk in sediments, because it was calculated based on Grade I of EQSSW, which could effectively protect most aquatic organisms (CSEPA, 2002). Results showed that Zn and Cu were the major contributors to sediment contamination, while Pb exhibited lower level. The mSQG-Q values at sites JL1, JL2, and JL6 ranged from 0.1 to 1, implying that their potential combined bio-toxic effect levels were moderate. The mSQG-Q values at other sites were below 0.1, presenting unimpacted or lowest potential bio-toxic effects. For JH, all heavy metals were lower than SQS-I<sub>JH</sub> values, which indicated lower pollution levels. The mSQG-Q values ranged from 0.1 to 1, which showed moderate impact potential bio-toxic effects.

To investigate the feasibility of basin-scale heavy metal SQSs, a comparison between the evaluation results of mSQG-Q and CF (including PLI) was performed. The evaluation results of PLI and CF are illustrated in Table 5. In JL, the CF values of Cu and Pb were <1, implying low contamination levels. The CF of Cd and Zn ranged from 2.10 to 4.70 and 0.34 to 2.26, exhibiting a moderate to considerate and a low to moderate contamination degree, respectively. Overall, mean CF values for all heavy metals were: Cd > Zn > Pb > Cu. For PLI values, the heavy metal pollution level at sites JL6 and JL1 were high compared to other sites. For JH, the CF values of Cu ranged from 0.96 to 2.89, with a moderate contamination degree. Sediments in JH suffered from moderate pollution with Pb (1 < CF < 3). The Cd in JH exhibited the highest contamination degree, followed by Cu, Zn, and Pb. High PLI values were found in JH13 and JH14, and the lowest PLI value was observed in JH6. Overall, Cd was the major contributor to sediment pollution in JL and JH based on CF evaluation.

The evaluation results of CF and/or PLI were highly variable compared to mSQG-Q based on SQSs. The mSQG-Q showed little difference in the pollution levels among all sites, even presenting

**Table 5**  
The CF and PLI values of heavy metals in sediments in JL and JH.

Sites	CF				PLI
	Cd	Cu	Pb	Zn	
JL1	4.00	0.63	0.63	1.96	1.33
JL2	2.10	0.38	0.42	0.34	0.58
JL3	2.80	0.46	0.73	1.64	1.11
JL4	2.60	0.52	0.48	0.51	0.76
JL5	4.70	0.30	0.50	0.45	0.75
JL6	3.00	0.68	0.85	2.26	1.41
JL7	3.40	0.54	0.45	0.46	0.79
JL8	3.40	0.52	0.63	0.46	0.84
Mean	3.25	0.50	0.59	1.01	
JH1	3.00	0.96	1.08	0.91	1.30
JH2	3.30	1.20	1.33	1.09	1.55
JH3	3.10	1.10	1.01	0.85	1.31
JH4	4.40	1.74	1.73	1.63	2.15
JH5	4.50	1.83	1.66	1.57	2.15
JH6	2.50	1.13	1.03	0.97	1.30
JH7	5.00	1.82	1.81	1.77	2.32
JH8	3.10	1.23	1.16	1.07	1.47
JH9	2.70	1.35	1.03	1.16	1.44
JH10	3.40	1.61	1.01	1.50	1.70
JH11	3.60	1.75	1.20	1.73	1.90
JH12	3.90	1.98	1.49	1.82	2.14
JH13	17.30	2.11	1.12	3.10	3.36
JH14	25.30	2.89	1.36	3.18	4.22
Mean	6.08	1.62	1.29	1.60	

CF:  : low degree;  : moderate degree;  : considerate degree;  : very high degree.

the same contamination degree, whereas the results of CF and/or PLI showed pollution levels of sites that were largely different. This was mainly due to the differences in the evaluation principle between mSQG-Q and CF (PLI). Some conventional methods (e.g., CF and/or PLI) used background values to measure the pollution levels of heavy metals in sediments. However, background values in different areas always have large differences, resulting in inaccuracies of evaluation results. Moreover, the complexity and bioavailability of geochemical forms of heavy metals in sediments are also neglected by employing such methods. For SQSs, owing to the focus on bioavailability and toxicity levels of heavy metals in sediments, basin-scale SQSs can avoid the shortcomings of other conventional methods and reflect the actual pollution level of heavy metals in sediments.

Furthermore, the heavy metal SQSs derived from the corresponding SQGs could be applicable to assess corresponding sediment quality in different water function zones, which could facilitate effective management and remediation of heavy metal polluted sediments. Therefore, the basin-scale heavy metal SQGs based on water quality would be a reliable method for SQS establishment.

#### 4. Conclusions

The spatial distribution of heavy metals in sediments in JL and JH were highly variable. Most heavy metals in sediments were dominated by residual fraction, except for Pb and Cd in JH. The partition of heavy metals between sediments and porewater were influenced by sediment properties and input of external heavy metals. IWCTU results showed that only several sites might pose long-term toxicity effects to the aquatic organisms in two rivers. For heavy metal SQGs and SQSs, the basin-scale heavy metal SQGs can reasonably classify different grades of sediments in different water function zones, and deduce the corresponding SQSs after normalization and optimization. Moreover, the mSQG-Q based on SQSs showed that heavy metals posed a lower potential bio-toxic effects in both JL and JH. With the comparison of evaluation results based on CF, establishing basin-scale heavy metal SQSs in coastal rivers based on SQGs combined with water quality standards was a feasible task. Notably, the normalized and optimized heavy metal SQGs were directly assumed as SQSs in this study only from a scientific point of view. In reality, the heavy metal SQSs establishment needs to integrate various factors (e.g., technologies and costs). Thus, there has been still much work to be done for establishing heavy metal SQSs.

#### Author contribution section

Conceived and designed the experiments: Qunqun Liu, Yanqing Sheng. Performed the experiments: Qunqun Liu, Ming Jiang, Guoqiang Zhao, Changyu Li. Analyzed the data: Qunqun Liu. Contributed reagents/materials/analysis tools: Yanqing Sheng. All authors agree with the contribution and declare that they have no actual or potential competing financial interests.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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## Appendix A. Supplementary data

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