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Risk assessment of heavy metals in the surface sediment at the drinking water source of the Xiangjiang River in South China

Zhifeng Huang^{1,2}, Chengyou Liu¹, Xingru Zhao^{1*} , Jing Dong¹ and Binghui Zheng^{1*}

Abstract

Background: The Xiangjiang River is an important drinking water resource for the Hunan Province of China. It is crucial to ascertain the pollution status, influencing factors, ecological risks, and possible sources of heavy metals in the sediments of the Xiangjiang River. Sediment is both a source and a sink of heavy metals in aquatic ecosystems. In this study, surface sediment was collected from the Zhuzhou Reach of the Xiangjiang River and eight heavy metals were investigated.

Results: In all sediment samples, all eight heavy metals were detected and their average concentration fell in the order of Zn > Pb > As > Cu > Cr > Ni > Cd > Co. Assessment shows extremely serious Cd pollution and a very high potential ecological risk from Cd. According to correlation analysis and principal component analysis (PCA), As, Cu, Ni, Pb, and Zn originate from industrial wastewater and mineral smelting activities, whereas Co, Cr, and Ni come from natural sources. Redundancy analysis (RDA) reveals that the organic matter content and the particle size of the sediment have a certain influence on the enrichment of heavy metals.

Conclusion: Among all eight examined heavy metals in the surveyed area, the content of Zn, Pb, and As is the highest, and that of Cd and Co is the lowest. Despite a low level of absolute content, the Cd in sediment already renders a high ecological risk and thus calls for urgent attention. Anthropogenic activities are the main source of heavy metals in the sediment. The distribution of heavy metals may also be influenced by sediment properties. The results provide guidance for controlling heavy metal pollution and protecting drinking water sources in the Xiangjiang River.

Keywords: Heavy metal, Risk assessment, Sediment, Distribution, The Xiangjiang River

Background

Heavy metals are a pressing concern in terms of their pollution in aquatic ecosystems because of their persistence, environmental toxicity, bioaccumulation, etc. [1–4]. Aquatic environments (e.g., lakes [5], rivers [6], reservoirs [7], and wetlands [8]) receive heavy metals in untreated or inadequately treated wastewater from domestic, industrial, and agricultural sources. Pollution

from heavy metals is inflicting rivers worldwide [9, 10], especially in developing countries [11, 12].

As an important component in riverine ecosystems, sediment serves as both a sink and a source of heavy metals [13, 14]. Most heavy metals quickly deposit into the sediment after entering rivers, and are much more concentrated in the sediment than in the water body of riverine systems [15, 16]. Conversely, when the physicochemical or hydrological conditions change, heavy metals in the sediment may desorb or resuspend to cause secondary pollution in the water body [17, 18]. The accumulation of heavy metals in the sediment directly affects benthic organisms and also influences many other organisms through the food web [19, 20],

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and endangers the wellbeing of the aquatic ecosystem. Therefore, it is of great importance to assess and understand the distribution and accumulation of heavy metals in sediment.

The Xiangjiang River is a major tributary of the Yangtze River and a key river in South China. It flows through the Hunan Province and supports many densely populated cities, providing services to both industry and agriculture [21, 22]. Unfortunately, the Xiangjiang River has become one of the most seriously heavy metal-polluted rivers in China over the past few decades because of the wastewater discharge from mining and metallurgical industries carrying heavy metals [23]. Although many researchers have studied heavy metal pollution in the Xiangjiang River [24–26], a systematic study is still lacking to associate the sediment with the distribution, characteristics, risk assessment, possible sources, and impact of heavy metals.

The Zhuzhou Reach of the Xiangjiang River is a typical area polluted by heavy metals [27]. In this work, we collected 30 surface sediment samples from the Zhuzhou Reach of the Xiangjiang River in order to (1) characterize the occurrence and distribution of eight heavy metals (Cd, As, Pb, Zn, Cu, Ni, Cr, and Co) in the sediment; (2) assess the pollution status and the ecological risk of the heavy metals; (3) identify possible sources of heavy metal pollution from principal component analysis (PCA) and Pearson’s correlation analysis, and (4) use redundancy analysis (RDA) to analyze the sediment characteristics that influence heavy metal distribution. The results illuminate on the environmental behavior of heavy metals and provide a reference for controlling pollution and ecological remediation in nearby areas and similar riverine systems.

Materials and methods

Study area and sample collection

The Xiangjiang River is the second largest tributary of the Yangtze River and an important drinking water source in South China. The main channel of the river is 856 km long, and its basin covers 94,721 km² and supports >30 million residents of the Hunan Province [28]. It flows from south to north en route 6 major cities in Hunan, i.e., Yongzhou, Hengyang, Zhuzhou, Xiangtan, Changsha, and Yueyang, and finally joins the Yangtze River via the Dongting Lake. The Hunan Province is known as the “Nonferrous Metal Village” because of its abundant mineral resources (e.g., Cd, Zn, Pb, Cu, etc. [29]). However, the mining and smelting of nonferrous metals over the past years caused severe heavy metal pollution to the Xiangjiang River. The city of Zhuzhou at the east of the Hunan Province is at the lower reaches of the Xiangjiang River. The area is rich in mineral resources and active in mining, ore smelting, and other major industrial practices. Therefore, we chose the Zhuzhou Reach of the Xiangjiang River for pollution evaluation and risk assessment. At the upstream of the surveyed area, there are many companies producing Zn, Pb, As, and alloys as well as other companies that carry out comprehensive recovery of Cu, Au, Ag, Bi, Te, Cd, In, and other rare metals. In this work, eight target heavy metals (Cd, As, Pb, Zn, Cu, Ni, Cr, and Co) were chosen for evaluation after examining the actual situation of the study area and consulting literatures on heavy metals in the Xiangjiang River [21, 25, 28].

Ten sampling transects (S1–S10) were selected from upstream to downstream in the investigated area (Fig. 1). Surface sediment samples (1 kg) were collected in August, 2011 with a core sampler at the north side, in the middle, and at the south side of the river from the

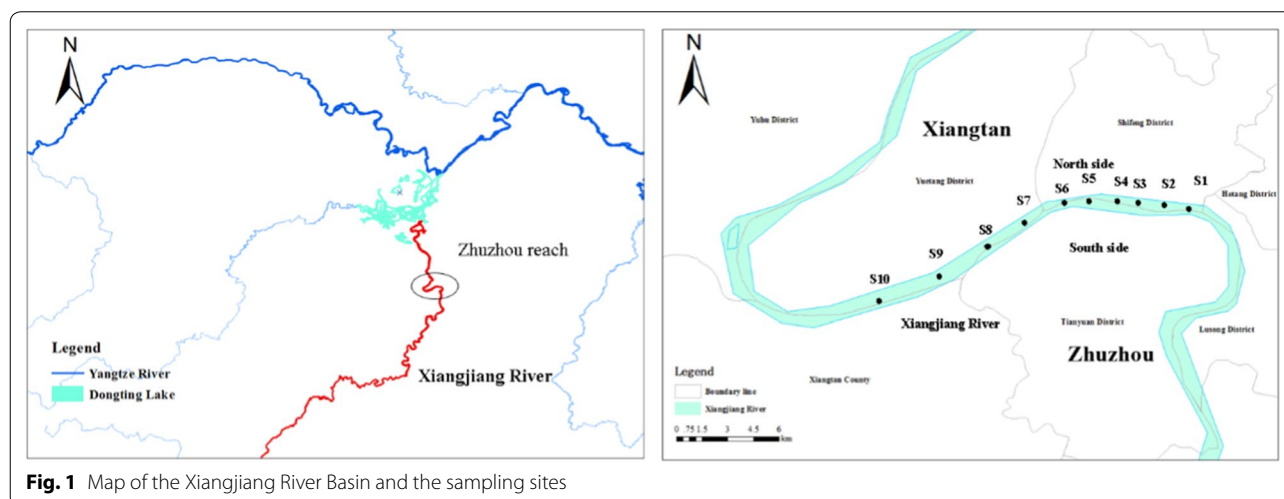


Fig. 1 Map of the Xiangjiang River Basin and the sampling sites

top 5 cm layer of the riverbed. Each sample consists of a mixture of three samples from one sampling site. The collections from each transect were stored in acid-rinsed polyethylene plastic bags and transported immediately to the laboratory for storage at $-20\text{ }^{\circ}\text{C}$ until further analysis, all sediment samples were analyzed within a week.

Sample treatment and analysis

Heavy metals in the sediment samples were determined at the Chinese Academy of Environmental Sciences according to published protocols [30, 31]. Briefly, crude sediment samples were freeze-dried, finely ground, homogenized, and sieved (100 mesh, $150\text{ }\mu\text{m}$). The prepared sample (0.05 g) was digested with mixed acid (5 mL HCl, 3 mL HNO_3 , 7 mL HF, 0.25 mL HClO_4) in a Teflon beaker. After the digestion, the dissolved sample was diluted with 2% HNO_3 , and the concentrations of heavy metals were analyzed using inductively coupled plasma mass spectrometry (ICP-MS, Agilent 7500 series, USA). The recoveries of standard reference metals were 90–110%. For more details, refer to Lin et al. [32]. The content of organic matters (OM) in the sediment was determined according to the standard methods for soil analysis [33]. Grain size was measured by a laser scattering particle size distribution analyzer (LA-300, Horiba, Kyoto, Japan) to determine the median sediment size (D_{50}) and the fine particle ($<63\text{ }\mu\text{m}$) content.

All reagents and solvents used in the sediment analysis were of analytical grade. The analytical procedure conformed to the certified standards of GBW07309 (China Stream Sediment Reference Materials). The results were within the acceptable level of uncertainty given by certified values.

Pollution assessment of heavy metals

Geoaccumulation

The geoaccumulation index (I_{geo}) formulated in 1969 is now one of the most widely used measures to evaluate heavy metal pollution in sediment because it can directly reflect the enrichment of exogenous heavy metals and provides consistent and comparable data [34, 35]. It is calculated as follows:

$$I_{\text{geo}} = \log_2 \left(\frac{C_{\text{sample}}}{1.5C_{\text{background}}} \right), \quad (1)$$

where C_{sample} is the concentration of the heavy metal in the sediment sample and $C_{\text{background}}$ is the geochemical background concentration (mg kg^{-1}) of the heavy metal. A background matrix correction factor of 1.5 is adopted as a coefficient to compensate for weathering and lithogenic effects. Table 2 lists the background concentration of the investigated heavy metals in the Dongting

Lake sediments [36]. Pollution can be classified into seven classes based on the I_{geo} value: Class 0 ($I_{\text{geo}} \leq 0$), practically unpolluted; Class 1 ($0 < I_{\text{geo}} \leq 1$), unpolluted to moderately polluted; Class 2 ($1 < I_{\text{geo}} \leq 2$), moderately polluted; Class 3 ($2 < I_{\text{geo}} \leq 3$), moderately to heavily polluted; Class 4 ($3 < I_{\text{geo}} \leq 4$), heavily polluted; Class 5 ($4 < I_{\text{geo}} \leq 5$), heavily to extremely polluted, and Class 6 ($I_{\text{geo}} > 5$), extremely polluted [37].

Assessment of potential ecological risk

The potential ecological risk index (RI) formulated by Hakanson in 1980 was used here to quantify the level of ecological risk of heavy metals in sediment [38]. The RI assesses the combined ecological and environmental toxicity to provide an overall evaluation of the potential risks of heavy metal pollution, and is calculated as follows:

$$E_r^i = T_r^i \frac{C_{\text{sample}}}{C_{\text{background}}}, \quad (2)$$

$$\text{RI} = \sum E_r^i, \quad (3)$$

where E_r^i and T_r^i are the potential ecological risk factor and the toxic response factor of the heavy metal, respectively, and C_{sample} and $C_{\text{background}}$ are the measured and background concentration. The T_r^i values for Co, Cd, Ni, Cr, Cu, As, Pb, and Zn, all taken from literature, are 5, 30, 5, 2, 5, 10, 5, and 1, respectively [39]. Table 1 lists the criteria for E_r^i and RI.

Statistical analysis

PCA and RDA were performed using Canoco 5.0 (Biometris, Netherlands). Pearson's correlation analysis was conducted using SPSS 20.0 (SPSS Inc., Chicago, IL, USA). Data analyses and statistical tests were performed using Microsoft Excel 2010.

Results and discussion

Occurrence and abundance of heavy metals in surface sediment

Table 2 summarizes the minimum, maximum, and average concentrations (represented by dry weight) of the eight heavy metals in the surface sediment samples from the Zhuzhou Reach of the Xiangjiang River. All eight heavy metals were detected in all sediment samples. The concentration of Cr, Co, Ni, Cu, Zn, As, Cd, and Pb ranged in 23.11–94.84, 5.26–25.24, 16.58–53.58, 20.54–159.3, 58.24–629.60, 34.74–186.80, 2.10–62.59, and 38.19–246.20 mg kg^{-1} , respectively. The average concentration ranked in the order of Zn (257.17 mg kg^{-1}) > Pb (102.52 mg kg^{-1}) > As (98.38 mg kg^{-1}) > Cu (71.29 mg kg^{-1}) > Cr (59.71 mg kg^{-1}) > Ni (36.29 mg kg^{-1}) > Cd (23.31 mg kg^{-1}) > Co (16.97 mg kg^{-1}). With the sediment of the Dongting

Table 1 Levels of potential ecological risk for heavy metal in sediments

Range of E_i^j value	Level of single metal ecological risk	Range of RI value	Level of comprehensive potential ecological risk
$E_i^j < 40$	Low	$RI < 150$	Low
$40 \leq E_i^j < 80$	Moderate	$150 \leq RI < 300$	Moderate
$80 \leq E_i^j < 160$	Considerable	$300 \leq RI < 600$	Considerable
$160 \leq E_i^j < 320$	High	$600 \leq RI$	High
$320 \leq E_i^j$	Very high		

Table 2 Concentration of heavy metals (mg kg⁻¹) in the sediment of the Xiangjiang River and of other rivers in South China

	Cr	Co	Ni	Cu	Zn	As	Cd	Pb	Refs.
Xiangjiang River									
Zhuzhou Reach (n = 30)	23.11	5.26	16.58	20.54	58.24	34.74	2.1	38.19	This study
Minimum									
Zhuzhou Reach	94.84	25.24	53.58	159.3	629.60	186.80	62.59	246.20	
Maximum									
Zhuzhou Reach	59.71	16.97	36.29	71.29	257.17	98.38	23.31	102.52	
Average									
Yongzhou Reach (n = 8)	129.25	30.88	107.38	51.58	71.85	51.90	6.825	93.80	[28]
Hengyang Reach (n = 8)	54.59	–	–	112.1	659.7	135.2	21.66	359.4	[40]
Changsha Reach (n = 12)	95.10	19.45	48.70	73.13	465.7	–	13.63	140.10	[28]
Jinjiang River (n = 8)	7.63	1.88	2.80	7.24	48.09	40.96	0.07	32.79	[41]
Hanjiang River (n = 6)	16.24	3.39	12.94	9.28	62.47	59.48	0.10	26.97	[41]
Yangtze River (n = 34)	82.87	–	32.85	25.14	82.92	9.31	0.17	25.13	[42]
Dongting Lake (background)	44	10.3	21.2	20	83.3	15	0.33	23.3	[36]

Lake taken as the background and a reference point, the concentration of the heavy metals is 1.65–70.64 times higher in the sediment of the Zhuzhou Reach, thus indicating remarkable pollution. In particular, Zn and Pb have higher concentration than other heavy metals, which is consistent with the Pb–Zn smelting activities around the Zhuzhou City. The lower concentration of other heavy metals is probably related to a less amount of discharge.

Table 2 also summarizes the concentration of the examined heavy metals in other reaches of the Xiangjiang River and in other rivers in South China [28, 36, 40–42]. The distribution of heavy metals is clearly different among different reaches of the Xiangjiang River (including Yongzhou, Hengyang, Zhuzhou, and Changsha). The Yongzhou Reach is the upstream and has the lowest concentration of heavy metals (except for Cr) in sediment [28]. The Hengyang Reach is located between Yongzhou and Zhuzhou and has the highest concentration of Cu, Zn, As, and Pb in sediment [40]. For the Zhuzhou Reach, the concentration of Co, Ni in the sediment is generally lower, but the concentration of Cd is the highest. The

Changsha Reach is the downstream and does not have a heavy metal with particularly high or low concentration [28]. The distinct distribution of different heavy metals may be associated with specific local mining activities. However, the sediment of the Xiangjiang River is clearly more polluted by heavy metals than the sediment of other rivers in South China, including the Jinjiang River, Hanjiang River, and Yangtze River [41, 42]. The concentration of Zn seems to be the highest among the examined heavy metals for all rivers, possibly due to regional Zn production. This high level of Zn is not specific to Hunan. In fact, for the past 20 years in China, Zn has an annual production higher than all seven other heavy metals except Cu [43].

Spatial distribution of heavy metals

Figure 2 illustrates the concentration of the heavy metals at S1–S10. The total concentration of heavy metals gradually increases as the water flows through the Zhuzhou Reach and peaks at S6 (1042.00 mg kg⁻¹), then suddenly decreases sharply at S7 to only 280.33 mg kg⁻¹, but rises again to about 650 mg kg⁻¹ at the downstream

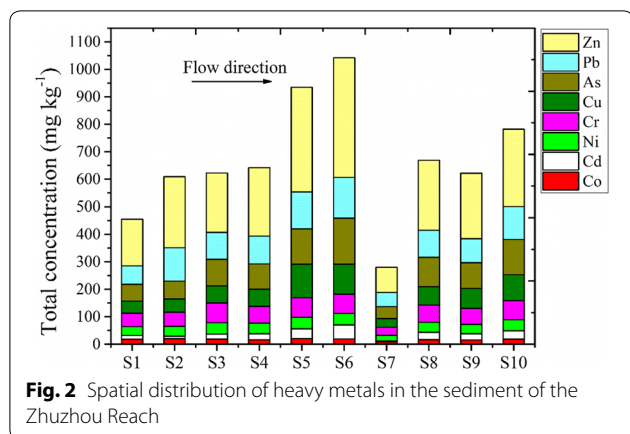


Fig. 2 Spatial distribution of heavy metals in the sediment of the Zhuzhou Reach

sites S8–S10. Zn has the highest concentration at S6 ($435.84 \text{ mg kg}^{-1}$) and then the next highest at S5 ($127.04 \text{ mg kg}^{-1}$). There is no significant difference in the concentrations of heavy metals within the upstream (S1–S4) and within the downstream (S8–S10), but there is a significant difference within the midstream (S5–S7).

The concentration of heavy metals is the highest at S6, possibly due to the discharge of local pollution sources. Other factors such as the grain size and the OM content of sediment can also affect the concentration of heavy metals. For instance, sediment with a higher content of OM adsorbs more heavy metals [44]. Indeed, Additional file 1: Table S1 shows that the OM content of S6 is the highest among all sites. In fact, S6 is located in the middle of the surveyed area and there is a sandbar between S6 and S7. This geographic feature may increase the deposition of particles carrying heavy metals into the sediment of S6.

Most heavy metals have slightly higher concentration in the middle of the river than on the south side, and all heavy metals have significantly higher concentrations on the north side (Fig. 3). In particular, the concentration of Zn and Pb in the north is nearly twice as much as in the middle and the south. The asymmetrical concentration can be associated with the nonferrous metal mining and smelting plants in the north that produce Zn, Pb, and alloys and discharge substantial amount of wastewater.

Assessment of heavy metal pollution

Many measures have been used in literature to assess heavy metal pollution in sediment, e.g., the geoaccumulation index, the potential ecological risk index, the enrichment factor, etc. [45–47]. In this study, we used I_{geo} and RI to assess heavy metal pollution in the sediment of the Zhuzhou Reach with reference to the concentration of Dongting Lake sediments as the background.

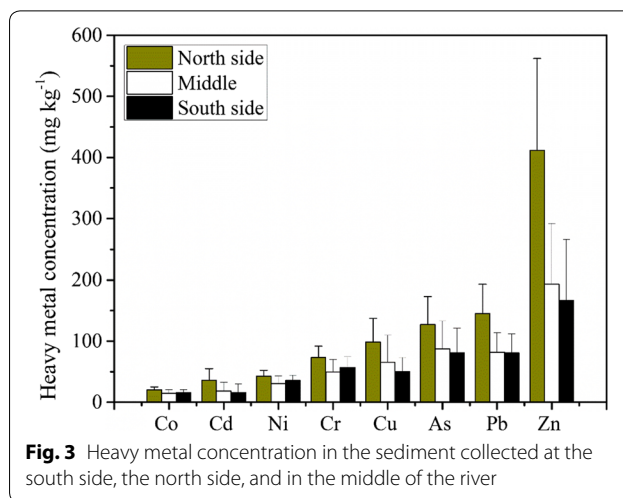


Fig. 3 Heavy metal concentration in the sediment collected at the south side, the north side, and in the middle of the river

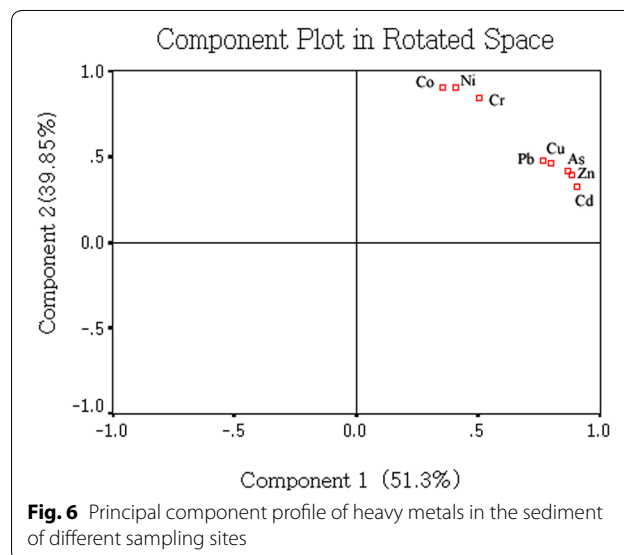
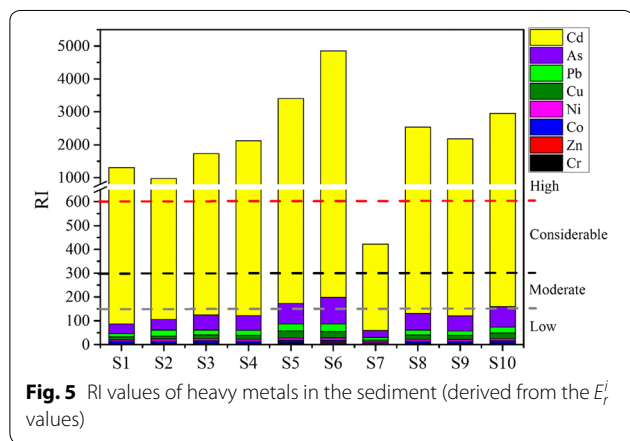
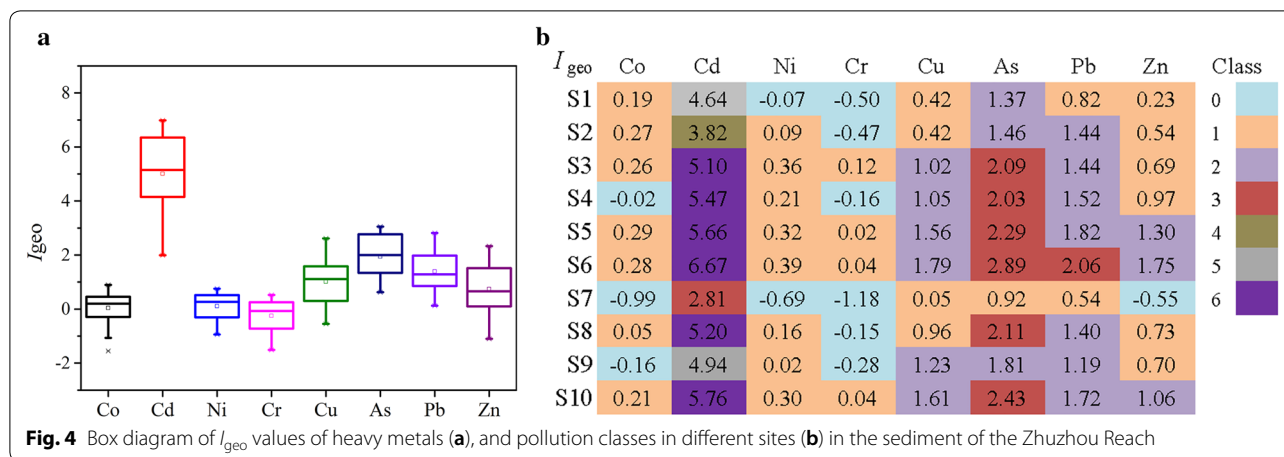
Assessment of I_{geo}

Figure 4a shows a boxplot of the average I_{geo} values for the eighty heavy metals, which can be ranked in descending order as follows: Cd (5.01) > As (1.94) > Pb (1.40) > Zn (0.74) > Ni (0.11) > Co (0.04) > Cu (0.43) > Cr (−0.26). The pollution levels of the metals are thus segmented into 4 classes according to the I_{geo} values. That is, the Zhuzhou Reach is extremely polluted by Cd (Class 6), moderately to heavily polluted by As and Pb (Class 2), unpolluted to moderately polluted by Zn, Ni, Co, and Cu (Class 1), and practically unpolluted by Cr (Class 0).

The different colors in Fig. 4b denote the class of I_{geo} for each heavy metal at S1–S10. The pollution of Cd is highly variable geographically, from Class 3 (S7) up to Class 6 (S3–S6, S8, S10). The pollution of As and Pb varies from Class 1 to Class 3, and the pollution of all other metals stay within two neighboring classes. The pollution of Cr, Co, and Ni appears to be mild (Class 0 Cr for 60%, Class 1 Co for 70%, Class 1 Ni for 80% of the 10 sites). Pollution is more severe midstream (S5–S7) and downstream (S8–S10) than upstream (S1–S4).

Assessment of potential ecological risk

Additional file 1: Table S2 lists the calculated E_r^i values of the heavy metals, from which the RI values were derived (Fig. 5). The RI value of the whole surveyed area ranged from 422.31 to 4854.41, with an average of 2246.64, thus indicating high ecological risk (Table 1) from heavy metal pollution. The sediment of S7 has $RI > 300$, and all others even have $RI > 600$ (Fig. 5). Among the tested metals, Cd creates very high ecological risk to the sediment at all sites ($E_r^i > 320$). The level of single metal ecological risk of As is the lowest at S7 ($E_r^i = 28.84$), considerable at S5, S6, and S10, and moderate at all other sites. Other heavy



metals (Pb, Cu, Ni, Co, Zn, and Cr) have low ecological risk in all sediments ($E_r^i < 40$).

Identification of the sources of heavy metals

Analyzing the sources of heavy metals can help understand their distribution. Principal components analysis (PCA) and Pearson’s correlation analysis were used to analyze the relationship and source of the heavy metals [28, 48].

In this study, PCA was performed based on the determined concentrations of heavy metals with varimax rotation. The Kaiser–Meyer–Olkin (KMO) test gives 0.76 (>0.7) and Bartlett test gives 397.58 (df=28, $p < 0.001$), both indicating strong correlation among variables and confirming that PCA can be used to reduce the dimensionality of variables. In the PCA results (Fig. 6 and Additional file 1: Figure S1), PC1 accounts for 51.30% of the variation, and its representative congeners include Cd, Zn, As, Cu, and Pb, implying a common source that is possibly exogenous discharge [24, 49]. This speculation is further supported by the Pearson’s

correlation analysis (Fig. 7), in which all heavy metals are significantly positively correlated with each other ($r > 0.60$, $p < 0.01$). Note that Pb and Zn, the main pollutants of the current Pb–Zn mining and smelting industry in China [50, 51], have a particularly high correlation ($r = 0.95$, $p < 0.01$). The PC2 accounts for 39.85% of the variation, and its representative congeners include Co, Ni, and Cr. The PCA results also indicate that at sites 5, the sediment at north side is affected by Cd, Zn, As, Cu, Pb (Additional file 1: Figure S1), while the sediment at the middle of river is mainly affected by Co, Ni, and Cr. It may be due to the effects of hydrodynamics or other sources, in the pollution assessment, Cr, Ni, and Co all have lower I_{geo} and lower E_r^i . Therefore, they may have been derived from natural processes such as soil erosion and rock weathering. This hypothesis is also supported by previous study [28, 46].

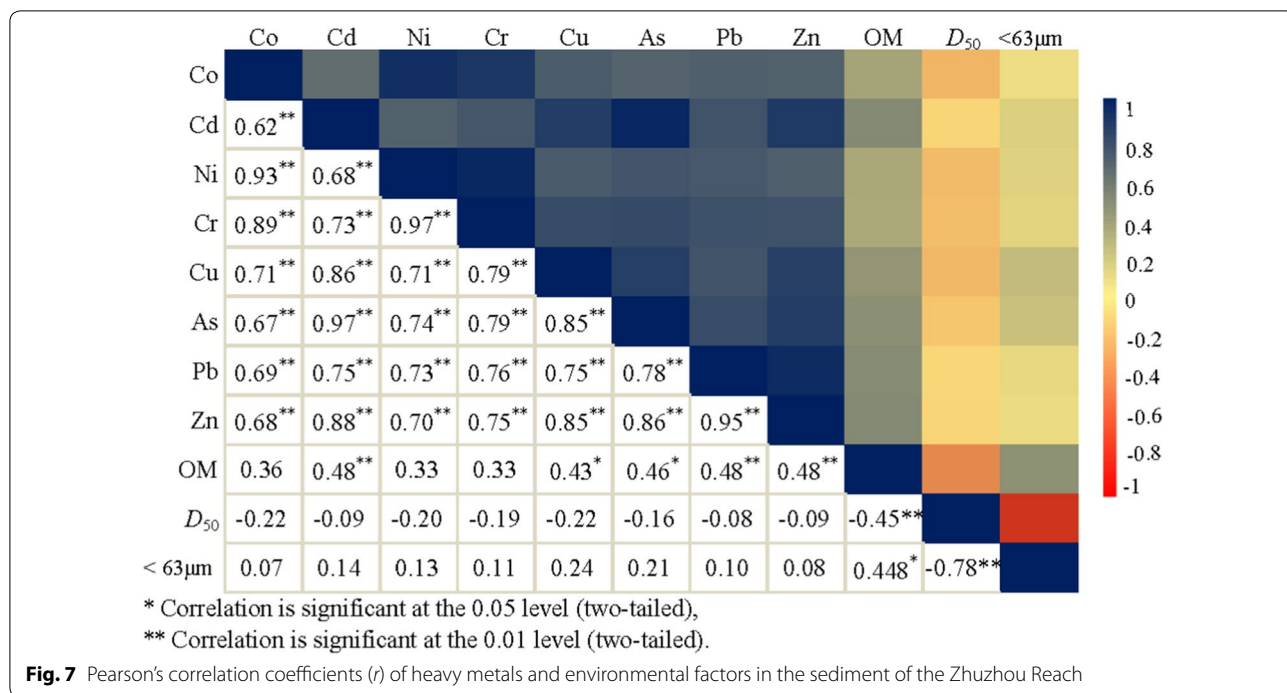


Fig. 7 Pearson's correlation coefficients (*r*) of heavy metals and environmental factors in the sediment of the Zhuzhou Reach

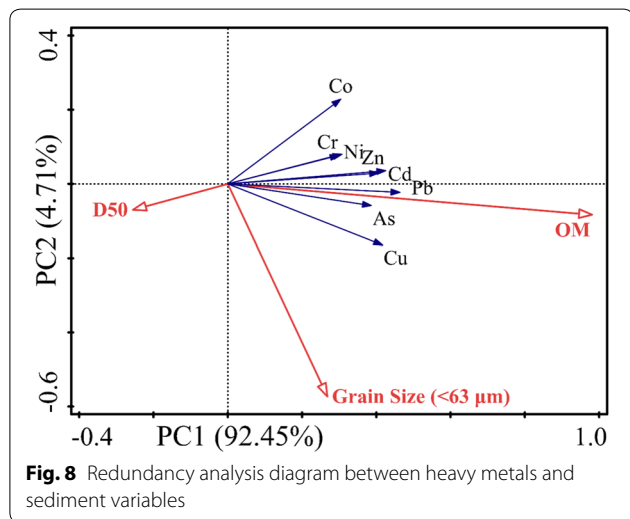


Fig. 8 Redundancy analysis diagram between heavy metals and sediment variables

Effects of environmental variables on the heavy metals

Many environmental factors affect the distribution of heavy metals in sediment. The redundancy analysis (RDA) was used here to further investigate the environmental variables that affect the distribution of heavy metal. The considered environmental variables include the organic matter content (OM), median size (D_{50}), and fine particle content (grain size <63 μ m) of the sediment samples (Additional file 1: Table S1). In the RDA result (Fig. 8), the angle between the environmental variable

and the heavy metal reflects their relevance. The concentrations of heavy metal are positively correlated to OM, i.e., sediment with relatively high OM tends to be more polluted by heavy metals. It was previously suggested that OM in sediment can adsorb heavy metals from the environment [52]. The grain size distribution of sediment is also an important factor that affects the enrichment of heavy metals [53]. Sediment particles can adsorb heavy metals by both physical and chemical processes. Physical adsorption is mainly related to the specific surface area of the sediment, and a smaller particle size of the sediment gives larger surface area and thus stronger adsorption capacity. Chemical adsorption is related to the active components contained in sediment particles, and the sediment tends to adsorb a greater amount of active components when it has smaller particle size [44, 54, 55]. Therefore, heavy metals are generally negatively correlated with D_{50} . The fine particle content (grain size <63 μ m) has influences on the heavy metal concentration. Figure 7 also shows the correlation between the environmental variables and the heavy metals.

Conclusions

Heavy metal pollutants in river sediment have been a serious environmental concern in aquatic ecosystems. This study examines eight heavy metals in the surface sediment of the Zhuzhou Reach of the Xiangjiang River. All eight heavy metals were detected at all sampling sites, with a concentration higher than the background value.

Their average concentration fell in the order of Zn > Pb > As > Cu > Cr > Ni > Cd > Co. Pollution appeared more serious in the midstream and on the northern side of the Xiangjiang River, possibly due to point source pollution nearby. Among the selected heavy metals, Cd presents very high ecological risk and should be carefully monitored and remediated. Most other heavy metals incur limited, if any, pollution according to the risk assessment. It was found from PCA and correlation analysis that Pb, Zn, As, Cu, and Cd are mainly originated from anthropogenic activities (industrial wastewater and mineral exploitation), whereas Cr, Co, and Ni can be attributed to the release from natural sources. Organic matter and grain size are the main factors affecting the distribution of heavy metals in the sediment, and heavy metals become enriched in the sediment when its organic matter is higher and particle size is smaller.

Supplementary information

Supplementary information accompanies this paper at <https://doi.org/10.1186/s12302-020-00305-w>.

Additional file 1: Table S1. Parameters of sediment samples. All values are given as mean values with standard deviation. **Table S2.** E_p^i values of heavy metals in sediment. All values are given as mean values with standard deviation. **Figure S1.** PCA score plot of heavy metals in the sediment of different sampling sites.

Abbreviations

PCA: Principal component analysis; RDA: Redundancy analysis; I_{geo} : Geoaccumulation index; RI: Potential ecological risk index; OM: Organic matters; D_{50} : Median size.

Acknowledgements

Not applicable.

Authors' contributions

ZH and XZ were involved in the experiments and manuscript writing. ZH was responsible for the data analysis. CL and JD collected samples. ZH and BZ designed the study. XZ and BZ contributed to correction of the manuscript. All authors read and approved the final manuscript.

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Availability of data and materials

The datasets obtained and analyzed in the current study are available from the corresponding author on reasonable request.

Ethics approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

Competing interests

The authors declare that they have no competing interests.

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