

Heavy metals in the riverbed surface sediment of the Yellow River, China

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Abstract One hundred and eleven riverbed surface sediment (RSS) samples were collected to determine the heavy metal concentration throughout the Inner Mongolia reach of the Yellow River (IMYR), which has been subjected to rapid economic and industrial development over the past several decades. Comprehensive analysis of heavy metal contamination, including the enrichment factor, geo-accumulation index, contamination factor, pollution load index, risk index, principal component analysis (PCA), hierarchical cluster analysis (HCA), and Pearson correlation analysis, was performed. The results demonstrated that a low ecological risk with a moderate level of heavy metal contamination was present in the IMYR due to the risk index (RI) being less than 150 and the pollution load index (PLI) being above 1, and the averaged concentrations of Co, Cr, Cu, Mn, Ni, Ti, V, and Zn in the RSS, with standard deviations, were 144 ± 69 , 77.91 ± 39.28 , 22.95 ± 7.67 , 596 ± 151 , 28.50 ± 8.01 , 3793 ± 487 , 69.11 ± 18.44 , and 50.19 ± 19.26 mg kg⁻¹, respectively. PCA, HCA, and Pearson correlation analysis revealed that most of the RSS was heavily contaminated with Zn, Ni, and Cu, due to the influence of anthropogenic activities; moder-

ately contaminated with Ti, Mn, V and Cr because of the dual influence of anthropogenic activities and nature; and slightly to not contaminated with Co because it occurs mainly in the bordering desert areas.

Keywords The Yellow River · Surface sediment · Heavy metal · Comprehensive analysis

Introduction

With the rapid development of the economy and industry, heavy metal contamination in aquatic ecosystems is becoming a worldwide environmental problem because a disproportionate amount of wastewater is being discharged into surface water bodies (Woitke et al. 2003; Singh et al. 2004; Sakan et al. 2009; Song et al. 2010; Hang et al. 2009; Yu et al. 2011; Zheng et al. 2008). Heavy metals tend to be trapped in aquatic environments and to accumulate in sediments during the process of adsorption, hydrolyzation, and precipitation (Loska and Wiechula 2003). The contaminated sediments can act as secondary sources of pollution to the overlying water column in the river (Shiple et al. 2011; Varol 2011), and mechanical disturbance of the sediments can increase the risk of contaminant release when they are re-suspended (Chen et al. 2007a). Heavy metals in aquatic environments are becoming a source of grave concern (Owens et al. 2005; Liu et al. 2009; Zhang et al. 2009; Varol 2011; Xu et al. 2014) because of their toxicity, persistence in the environment (Li et al. 2013; Karlsson et al. 2010; Khan et al. 2013), transport through flowing water, and subsequent accumulation in the bodies of aquatic microorganisms, flora, and fauna, which may, in turn, enter the human food chain and cause a host of health problems (Chen et al. 2007b; Guan et al. 2016; Li et al. 2013; Mendoza-Carranza et al. 2016).

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In China, historical information on heavy metal contamination in riverbed surface sediments (RSS) mainly focused on industrialized areas such as the deltas of the Yangtze River, Yellow River, and Pearl River (Zhong et al. 2011; Zhang et al. 2009; Bai et al. 2012), with only a few studies dedicated to the upper reaches of the Yellow River (Ma et al. 2016). The Inner Mongolia reach of the Yellow River, located at the end of the upper reach of the Yellow River, is in the central region of Mongolia, and it supplies the water to its neighboring cities. The districts bordering the Inner Mongolia reach of the Yellow River (IMYR) are also important energy bases and major grain producing areas in northwest China. With social and economic development, the cities bordering the IMYR, such as Bayan Nur and Baotou, have been rapidly developing industries such as electroplating, metallurgy, leather dyestuffs, mining, chemical engineering, and power generation, giving rise to contaminant sources and the continuous discharge of heavy metals in the main channel of the Yellow River (Li and Zhang 2010). Most of the heavy metals are absorbed by the bed materials, and because of the fast flowing stream and discontinuously inputted sewage, monitoring the quality of the water body alone may result in misleading and underestimated stream contamination levels (Varol 2011). In addition, heavy metals in the sediments of Chinese rivers are not included in the monitoring list (China's Ministry of Environmental Protection 2008). Hence, studying this aspect would help establish pollutant loading reduction goals, protect the ecological environment, and preserve public health in the Yellow River basin. The main objectives of the study were the following:

- (1) To determine the magnitude and spatial variation of the contemporary deposition and storage of the total sediment-associated heavy metals on the riverbed of the IMYR and to compare these values with estimates of the contamination levels with different assessment methods.
- (2) To determine the sources of sediment-associated heavy metals in the main channel systems of the IMYR and to explain the metal contamination in the study area.

This study not only provides valuable information related to the heavy metals in the RSS of the IMYR but also serves as a resource for local and regional environmental management and water resource planning authorities.

Materials and methods

Study area

The focus of this study is the Inner Mongolia reach of the Yellow River, which loops south near the city of Bayan Nur,

flows east to the city of Togtoh, and stays in the Inner Mongolia Autonomous Area for 500 km. It belongs to the upper to middle reaches of the Yellow River in terms of its geology (Fig. 1). The Yellow River flows through the northern border of the Hobq Desert (Yang 2003), and along the southern bank of this section, 10 tributaries named the “Ten Great Gullies” flow through the Hobq Desert (Fig. 1). This basin is an important energy base and a primary grain producing area in north China; it also plays a decisive role in both the industrial and agricultural economies. The cities along the Yellow River, such as Bayan Nur, Urad Front Banner, Baotou, and Dalad Banner, are rapidly developing their metallurgy, leather dyestuffs, mining, chemical engineering, and power generation industries; especially, Baotou is one of the major aluminous manufacturing bases of China. In the process, drainage from mines releases heavy metal ions, including Cu, Mn, V, Zn, and Ni, into the Yellow River from both active mines and abandoned sulfide tailing dumps, which are likely to cause environmental problems related to both water and sediment quality (Huang et al. 2007; Li and Zhang 2010; Fu et al. 2014). The cities along the Yellow River, such as Bayan Nur, Urad Front Banner, Baotou, and Dalad Banner, are rapidly developing their metallurgy, leather dyestuffs, mining, chemical engineering, and power generation industries. This reach of the Yellow River is on the fringe of the East Asian monsoon belt, and it has a continental climate and a low and unevenly distributed annual precipitation (150–363 mm). The duration of sunlight is long (≥ 10 °C accumulated temperature 3004~3515 °C), and evaporation is intense (the annual mean evaporation of 1939–3482 mm is as much as 10–12 times the precipitation) (Yang 2003).

Sediment sampling and analytical procedure

Thirty-seven sampling sites were set up in the riverbed along the main stream of the Yellow River (Fig. 1). For each site, to make the samples more representative, we randomly took three parallel underwater samples approximately 15 m away from the bank using a homemade cylindrical steel sampler (approximately 10 cm in diameter), plastic valve bags, and tying cordage, and 111 samples of the riverbed surface sediment were obtained. All samples were analyzed for heavy metal concentration at the Key Laboratory of Western China's Environmental Systems (Ministry of Education), Lanzhou University. The analysis procedures were identical to those described by Guan et al. (2014) and Pan et al. (2015). Sample preparation involved air drying the samples and grinding them to yield grain sizes smaller than 75 μm . Up to 4 g of sample was weighed and poured into the center of a column apparatus with boric acid, and the apparatus was pressurized

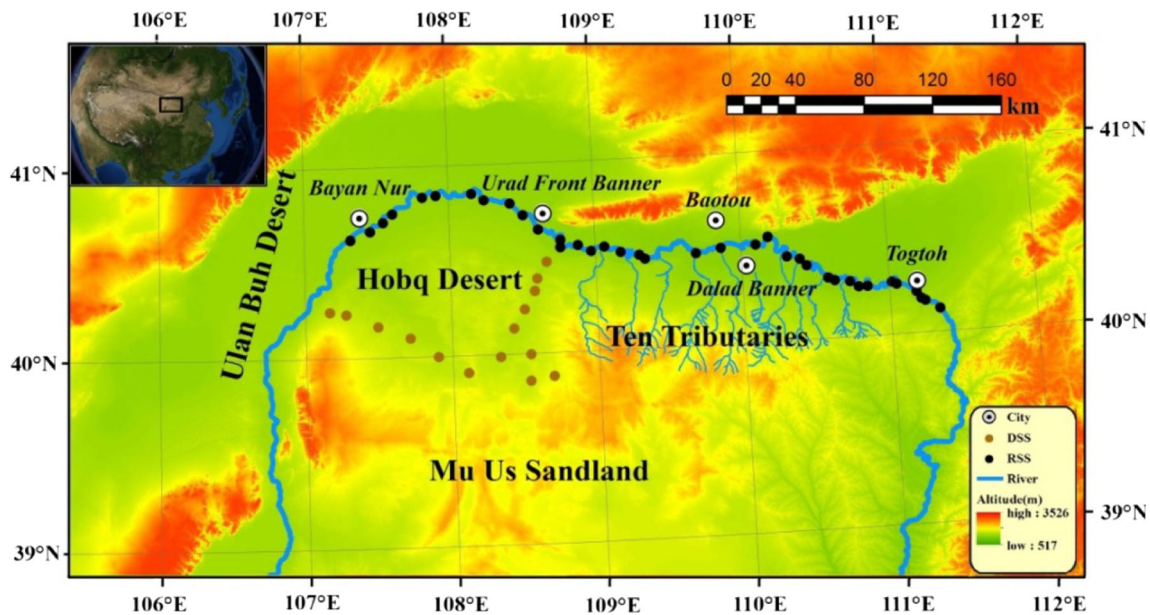


Fig. 1 Site location map of the IMYR. The *black solid circles* represent the sampling sites for the surface sediment of the riverbed (RSS, $n = 37$), and the *brown solid circles* represent the sampling sites for the surface sediments of the deserts (DSS, $n = 15$)

to 30 t m^{-2} for 20 s. Contamination during analysis may distort the result. To avoid potential contamination, all collected samples were sealed in clean plastic bags. The grinding container was rinsed with distilled water and subsequently air dried before use. The column apparatus for sample compaction was precleaned using absorbent cotton and alcohol and then air dried. The processed sample, measuring approximately 4 cm in diameter and 8 mm in thickness, was analyzed using a Philips Panalytical Magix PW2403 X-ray fluorescence (XRF) spectrometer (Holland) at ambient temperatures and pressures (approximately $20 \text{ }^{\circ}\text{C}$, 85 kPa). A calibration curve was developed using 16 Chinese National Standard soil reference samples (GSS-1 to GSS-16), 12 fluvial sediment reference samples (GSD-1 to GSD-12) and five rock reference samples (GSR-1 to GSR-5). The reproducibility of the element measurements was evaluated by repeat analysis using the National Standard soil reference sample GSS-8, with analytical uncertainties of $<3 \%$ for major elements and $<5 \%$ for trace elements. Detection limits (calculated on the basis of 10 determinations of the blanks as three times the standard deviation of the blank) in the sediment tests were 0.01 mg kg^{-1} for Co, Cr, Cu, Mn, Ni, Ti, V, and Zn and 0.01% for Al. The analytical results are reported in oxide compound form, apart from the trace elements which are given in elemental form.

Risk assessment methods of sediment contamination

In this paper, the background value of each metal comes from the 15 surface sediment samples from the Hobq Desert at preindustrial levels (Fig. 1; Table 3). The background values utilized were 279 mg kg^{-1} for Co, 36.19 mg kg^{-1} for Cr,

1.90 mg kg^{-1} for Cu, 330 mg kg^{-1} for Mn, 3.80 mg kg^{-1} for Ni, 2277 mg kg^{-1} for Ti, 37.41 mg kg^{-1} for V, 7.51 mg kg^{-1} for Zn, and 5.77% for Al. Approaches such as enrichment factor (EF), geo-accumulation index (I_{geo}), and contamination factor (CF) have been widely used to study heavy metal contamination and sources in river, estuarine, and coastal environments (Santos Bermejo et al. 2003; Owens et al. 2005; Zhang et al. 2009; Zahra et al. 2014). Pollution load index (PLI) and potential ecological risk index (RI) represent the sensitivity of various biological communities to toxic substances and illustrate the sediment's environmental quality and potential ecological risk caused by heavy metals (Fernandes et al. 1997; Huang et al. 2016; Mohammad et al. 2010; Yang et al. 2009; Varol 2011).

Enrichment factor

In our case, we used the metal EF as an index to evaluate anthropogenic influence on the sediments (Baptista et al. 2000; Gao and Chen 2012; Han et al. 2006; Zhang et al. 2009; Varol 2011; Xu et al. 2014; Resongles et al. 2014). EF is commonly defined as the observed metal to aluminum (Al) ratio in the sample of interest divided by the background metal/Al ratio (Owens et al. 2005; Zhang et al. 2009; Tam and Wong 2000; Zahra et al. 2014) because Al is one of the most abundant elements on the earth and usually has no contamination concerns (Zhang et al. 2009). In addition, the aluminum averaged concentration in the surface sediments of the IMYR (5.66) is less than its background value (5.77), with a small standard deviation (0.86) and coefficient of variance (0.15; Table 3). Hence, Al has been identified as the

normalized element in this paper (formula 1). Mathematically, EF is expressed as:

$$EF = \frac{(C/Al)_{Sample}}{(C/Al)_{Background}} \quad (1)$$

where $(C/Al)_{Sample}$ is the metal to Al ratio in the sample of interest, and $(C/Al)_{Background}$ is the natural background value of metal to Al ratio. EF provides a classification system for the degree of pollution (Table 1).

Geo-accumulation index

The I_{geo} is another classical assessment model that enables evaluation of metal pollution in sediments by comparing current concentrations with preindustrial levels (Chabukdhara et al. 2012; Christophoridis et al. 2009; Zhang et al. 2009; Varol 2011; Zahra et al. 2014). The I_{geo} is defined by the following formula:

$$I_{geo} = \log_2 \left(\frac{C_{Sample}}{k \times C_{Background}} \right) \quad (2)$$

where C_{Sample} is the measured content of the metal, $C_{Background}$ is the corresponding background content of the metal, k is the

background matrix correction factor introduced to account for possible differences in the background values due to lithospheric effects, and the constant is used due to potential variations in the baseline date ($k = 1.5$; Lu et al. 2009; Bhuiyan et al. 2010; Varol 2011). I_{geo} provides a classification system for the degree of pollution (Table 1).

Contamination factor

Sediment contamination was also assessed using the contamination factor and degree. In the version suggested by Hakanson (1980), they assess the sediment contamination by using the concentrations in the surface desert sediments at preindustrial levels as reference points.

$$CF = \frac{C_{Sample}}{C_{Background}} \quad (3)$$

where C_{Sample} is the mean content of the metal from the sampling site, and $C_{Background}$ is the preindustrial concentration of the individual metal. Hakanson (1980) defines four categories of CF, shown in Table 1.

Table 1 Evaluation criteria of different risk assessment methods

Assessment methods	Level	Values	Comprehensive assessment level
Enrichment factor (EF)	1	$EF \leq 2$	Deficiency to minimal enrichment
	2	$2 < EF \leq 5$	Moderate enrichment
	3	$5 < EF \leq 20$	Significant enrichment
	4	$20 < EF \leq 40$	Very high enrichment
	5	$EF > 40$	Extremely high enrichment
	0	$I_{geo} \leq 0$	Practically uncontaminated
Geo-accumulation index (I_{geo})	1	$0 < I_{geo} \leq 1$	Uncontaminated to moderately contaminated
	2	$1 < I_{geo} \leq 2$	Moderately contaminated
	3	$2 < I_{geo} \leq 3$	Moderately to strongly contaminated
	4	$3 < I_{geo} \leq 4$	Strongly contaminated
	5	$4 < I_{geo} \leq 5$	Strongly to very strongly contaminated
	6	$I_{geo} < 5$	Very strongly contaminated
Contamination factor (CF)	1	$CF < 1$	Low contamination factor
	2	$1 < CF \leq 3$	Moderate contamination factor
	3	$3 < CF \leq 6$	Considerable contamination factor
	4	$CF > 6$	Very high contamination factor
Ecological risk index (RI)	1	$RI \leq 150$	Low ecological risk
	2	$150 < RI \leq 300$	Moderate ecological risk
	3	$300 < RI \leq 600$	High ecological risk
	4	$RI > 600$	Significantly high ecological risk
	1	$PLI \leq 1$	Uncontaminated by heavy metals
	2	$PLI < 1$	Contaminated by heavy metals

Pollution load index

The PLI was proposed by Tomlinson et al. (1980), and for the entire sampling site, PLI has been determined as the n th root of the product of the n C_f^i :

$$PLI = \sqrt[n]{C_{f1}^i \times C_{f2}^i \times C_{f3}^i \times \cdots \times C_{fn}^i} \quad (4)$$

where C_f^i is the single element pollution factor, the values were calculated using Formula 3. This empirical index provides a simple and comparative means for assessing the level of metal pollution (Mohammad et al. 2010; Varol 2011). If the PLI is larger than 1, the sediment is considered to be contaminated by heavy metals (Table 1). However, if the PLI is smaller than 1, the sediment is considered to be uncontaminated by heavy metals (Varol 2011; Fujita et al. 2014; Table 1).

Potential ecological risk index

The potential ecological risk method was developed by Hakanson (1980). The RI was introduced to assess the degree of contamination of trace metals in the sediments. The formulas for calculating the RI are as follows:

$$E_r^i = T_r^i \times C_f^i = T_r^i \times (C_s^i / C_n^i) \quad (5)$$

$$RI = \sum_{i=1}^n E_r^i \quad (6)$$

where C_s^i is the content of the element in the sample, C_n^i is the background value of the element, C_f^i is the single element pollution factor, E_r^i is the RI of an individual element, and T_r^i is the biological toxicity factor of an individual element, which are defined for Ti, Mn, Zn, V, Cr, Cu, Ni, and Co as 1, 1, 1, 2, 2, 5, 5, and 5, respectively (Yi et al., 2011; Zhang et al. 2014). RI is the sum of E_r^i . Table 1 shows the factor standard for different levels.

Multivariate analysis

Multivariate analyses of principal component analysis (PCA) and hierarchical cluster analysis (HCA) were used in this study. All experimental data performed in multivariate analysis was standardized through z -scale transformation to avoid misclassification due to wide differences in data dimensionality (Varol 2011; Gao and Chen 2012). PCA can convert vast raw data into minority unrelated variables and explain the original data's variance without losing much of the primary information. PCA with Varimax rotation of standardized component loadings was conducted for the deriving factors, and PCs with an eigenvalue above 1 were retained (Li and Zhang 2010; Liu et al. 2003; Huang et al. 2013). Kaiser–Meyer–Olkin (KMO) and Bartlett's sphericity tests were performed

to examine the suitability of the data for PCA; KMO was used to determine the correlation of variables. A high value (close to 1) indicates that the principal component can be useful; KMO = 0.860 in this study. Bartlett's test of sphericity determines whether a correlation matrix is an identity matrix, which may indicate that the variables are unrelated, that is, a significance level of <0.05 is fit for PCA (Varol 2011); the significance level = 0 in this study.

The HCA is a classification process using different categories or clusters of data sets; it is an unsupervised classification procedure that involves measuring either the distance or the similarity between variables (Gao and Chen 2012), and it is a widely used sorting method (Ikem et al. 2011; Li and Zhang 2010; Varol 2011; Gao and Chen 2012). HCA is a multivariate technique whose primary purpose is to classify objects of a system into categories or clusters based on their similarities, and the objective is to find an optimal grouping where the observations or objects within each cluster are similar, but the clusters are dissimilar from each other (Ikem et al. 2011; Islam et al. 2015; Paramasivam et al. 2015). The dendrogram visually displays the order in which parameters or variables combine to form clusters with similar properties. Q-model hierarchical agglomerative HCA was performed in the normalized data set through squared Euclidean distances as a measure of similarity and Ward's method to obtain dendrograms. Stations in the same cluster have a similar contamination source (Singh et al. 2004; Chen and Huang et al. 2007; Li and Zhang 2010). All multivariate analyses were completed in SPSS 20.0 for Windows.

Results and discussion

Sediment pollution survey and assessment

Descriptive statistical analysis

A preliminary statistical analysis of metal contamination in the study area has been performed, and the results show that the elements of maximum and minimum averaged concentration were Ti and Cu, respectively (Table 2). The sequence of the average metal contents was Ti > Mn > Co > Cr > V > Zn > Ni > Cu in RSS, which was similar to that of the desert, Ti > Mn > Co > V > Cr > Zn > Ni > Cu (Table 2), indicating that some of the metal concentrations in the surface sediments of the study area fluctuated due to anthropogenic activities and most of the elements are naturally occurring. Compared with the background value, the sequence of the ratio was Cu > Ni > Zn > 6 > Cr > 2 > V > Mn > Ti > 1 > Al > Co. Co and Al were not contaminated by anthropogenic means because their averaged concentrations were less than their BVs (Table 2), and the

Table 2 Descriptive statistical analysis of nine elements in the IMYR

	Co	Cr	Cu	Mn	Ni	Ti	V	Zn	Al
Max	339	295.43	44.15	1035	51.95	4985	121.70	104.70	7.90
Min	34	42.53	10.40	335	13.20	2064	26.93	15.80	3.95
Mean	144	77.91	22.95	596	28.50	3793	69.11	50.19	5.66
SD	69	39.28	7.67	151	8.01	487	18.44	19.26	0.86
CV	0.48	0.50	0.33	0.25	0.28	0.13	0.27	0.38	0.15
BG	279	36.19	1.90	330	3.80	2277	37.41	7.51	5.77
Mean/BG	0.51	2.15	12.08	1.81	7.50	1.67	1.85	6.68	0.98

Co, Cr, Cu, Mn, Ni, Ti, V, and Zn are trace elements (mg/kg), and Al is the major element (%)
SD standard deviation, *CV* coefficient of variation, *BG* background

average contents of Cu, Ni, and Zn were 12.08, 7.50, and 6.68 times their BVs, respectively (Table 2), indicating contamination by humans. The average concentrations of Cu, Ni, and Zn in the RSS of the IMYR were higher than those for the upper continent (Wedepohl 1995) and even higher than those for the sediments from the south of Tegger Desert located near the study area (10.37, 14.99, and 42.66 mg/kg for Cu, Ni, and Zn; Guan et al. 2014). However, the surface sediments from northern Mexico in semi-arid environment contained higher Ni, Cu, and Zn (69.5 ± 15.5 , 400.5 ± 15.8 , and $78.8 \pm 6.5 \text{ mg kg}^{-1}$, respectively) than the RSS of the IMYR, due to mining activities (Meza-Figueroa et al. 2009).

Assessment of the degree of heavy metal contamination

The results from this study in the IMYR showed that significant enrichments ($5 < EF \leq 20$) of Cu, Ni, and Zn were found in 100, 97, and 92 % of the sampling sites, respectively, suggesting that these metal contaminations were currently a major concern. It is evidenced by Nriagu that industrial activities such as smelting of steel and non-ferrous metals are important sources for anthropogenic heavy metals (Nriagu et al. 1979). Inner Mongolia is known for its industries including steel and rare earth industries, which greatly promote the increase of its gross domestic product (Liu and Liu, 2013). The higher EF and I_{geo} values for Cu, Ni, and Zn in sediment samples from Inner Mongolia may be from steel industries and rare earth industries. Moreover, a significant enrichment of Cr was found in site 3 (Fig. 2a), indicating that Cr contamination could be correlated to local point sources. The averaged enrichment factors of Cr (2.25 ± 1.37), Mn (1.82 ± 0.21), V (1.86 ± 0.25), and Ti (1.72 ± 0.24) were found to range from 1.5 to 2 in the IMYR, suggesting that these metal contaminations appeared to be moderate in some localized areas (the number of sampling sites with EF values larger than 2, which for Cr, Mn, V, and Ti are 14, 9, 9 and 3, respectively; Fig. 2a). The contamination level of heavy

metals in the RSS of the IMYR was reflected by the EF values in the following ranking: $Cu > Ni > Zn > Cr > Mn > V > Ti$. Moreover, the percentages of the I_{geo} indexes that reached the moderate or contaminated level ($I_{geo} > 1$) in all of the RSS sites were 97 % for Cu, 92.5 % for Ni, 89 % for Zn, 2.7 % for Cr, 2.7 % for Mn, 2.7 % for V, and 0 % for Ti (Fig. 3a–c). Assessment of the sediment integrated pollution degree showed that the EF evaluated result is in concordance with the I_{geo} index result (Figs. 2a and 3a). Especially noteworthy is that the significant contamination of Cu, Zn, Mn, and V was observed at the downstream of site 20 (Baotou; $EF_{Mn}, V > 2$; $I_{geo} (Cu, Zn) > 2$; Figs. 2b, c and 3b, c), suggesting that the contamination at site 20 may be due to anthropogenic inputs that consist of the metals mentioned above. In addition, the I_{geo} indexes of Co and Al were all negative, indicating that contamination by Al and Co has not occurred in the study area (Fig. 3a), and this result is similar to that by Guan et al. (2016).

The calculated CF values for all nine metals are shown in Fig. 4. Overall, the CF for all metals shows the descending order of $Cu > Ni > Zn > Cr > Mn > V > Ti > Al > Co$. The average CF values of Cu, Ni, Zn, Cr, Mn, V, Ti, Al, and Co were 12.08 ± 4.04 , 7.50 ± 2.11 , 6.68 ± 2.57 , 2.15 ± 1.09 , 1.81 ± 0.46 , 1.85 ± 0.49 , 1.67 ± 0.21 , 0.98 ± 0.15 , and 0.51 ± 0.25 , respectively. The CF for Cu was in the range of “high contamination” for all sites except site 24. Risk levels for all other metals are as follows. High contamination: Ni 89.19 %, Zn 51.35 %, and Cr 2.70 %. Significant contamination: Ni 10.81 %, Zn 43.24 %, Mn 2.70 %, and V 2.70 %. Moderate contamination: Zn 5.41 %, Cr 97.30 %, Mn 97.30 %, V 94.60 %, Ti 97.30 %, and Al 35.14 % (Fig. 4). A high value of CF_{Co} was observed at site 1 despite low contamination at all other sites (Fig. 4). Because the IMYR is bordered by the Ulan Buh Desert and the Hobq Desert (Fig. 1), the aeolian sand with high Co concentration from the bordering deserts is transported into the riverbed during the windy seasons (especially the spring and winter), and sand dunes may encroach onto the floodplain or even into the

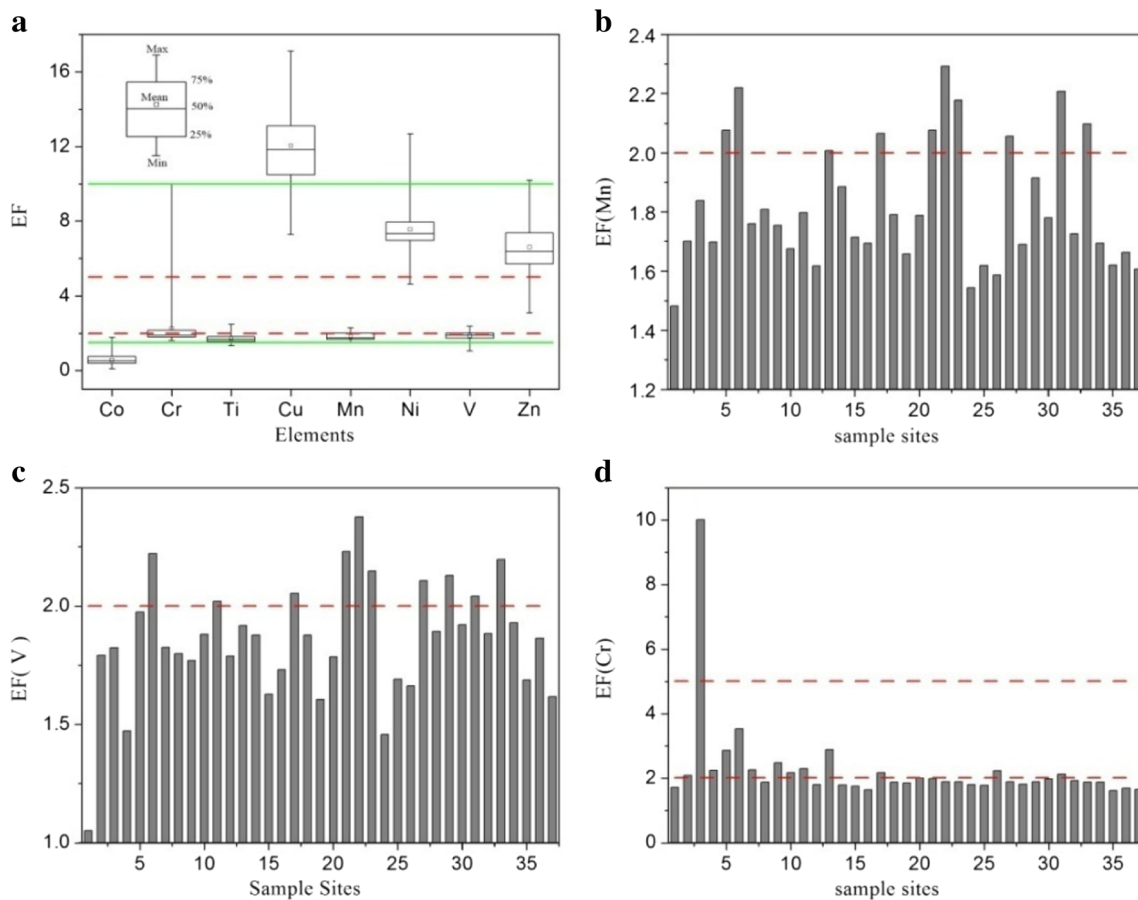


Fig. 2 Enrichment factor (EF) value of heavy metals in the RSS of the IMYR, China (a), and the EF values of Mn (b), V (c), and Cr (d) in each sampling site present in b, c, and d, respectively

riverbed, which increases the Co concentration of the RSS in the IMYR (Pan et al. 2015; Guan et al. 2016). High Cr contamination was observed at site 3 due to industrial effluents and solid wastes from heavy industries (such as the mining and smelting of non-ferrous metals, chemical production, and lead–acid battery production, as well as the leather and dyeing industry) in Bayan Nur (Fig. 4). The highest concentrations of Mn, Ni, V, Zn, and Cu occurred at site 23, and the metal concentrations in the RSS of the sites 21, 22, 27, 29, 31, and 33 were higher than those measured on average (Fig. 4), which were ascribed to the influence of industrial activities (steel and machine manufacture, metallurgy, and production of rare earth materials) in bordering cities such as Baotou and Dalad Banner (Huang et al. 2007; Li and Zhang 2010; Fig. 1).

Assessment of the degree of ecological risk and pollution loading

The PLI values ranged from 1.63 to 3.57, with an average of 2.65, and the RI values ranged from 29.75 to 104.63, with an average of 58.34 (Fig. 5), indicating that a low ecological risk with a moderate level of heavy metal contamination was

present in the IMYR. Liu et al. (2005) reported that the PLI values for agricultural soils with sewage irrigation in Beijing, China ranged from 2.39 to 3.43, with an average of 3.06. Yi et al. (2011) have registered an average RI value for the Yangtze River as 94.31 (range 36–222.1). Yin et al. (2011) have calculated RI values for Taihu Lake ranging from 41.5 to 655, with an average of 188. The RI value, taking the geochemical background and national soil background as reference, ranged from 9.5 to 209.09, with an average of 46.71, and it ranged from 31.56 to 1583.92, with an average of 267.42, for Luan River, China, respectively (Liu et al. 2009). The present PLI range is lower than the values for Beijing, and the present RI range is lower than the values for Luan River, Taihu Lake, and Yangtze River. In the present study, sites 3, 17, 21–23, 31, and 33 have high PLI and RI values due to the solid wastes and wastewater from the cities bordering the IMYR, which may dominate the enhanced polluted metal distribution (Figs. 1 and 5).

Heavy metal sources of the IMYR

Because heavy metals in the RSS of the IMYR have now been shown to have a low ecological risk with a moderate level of

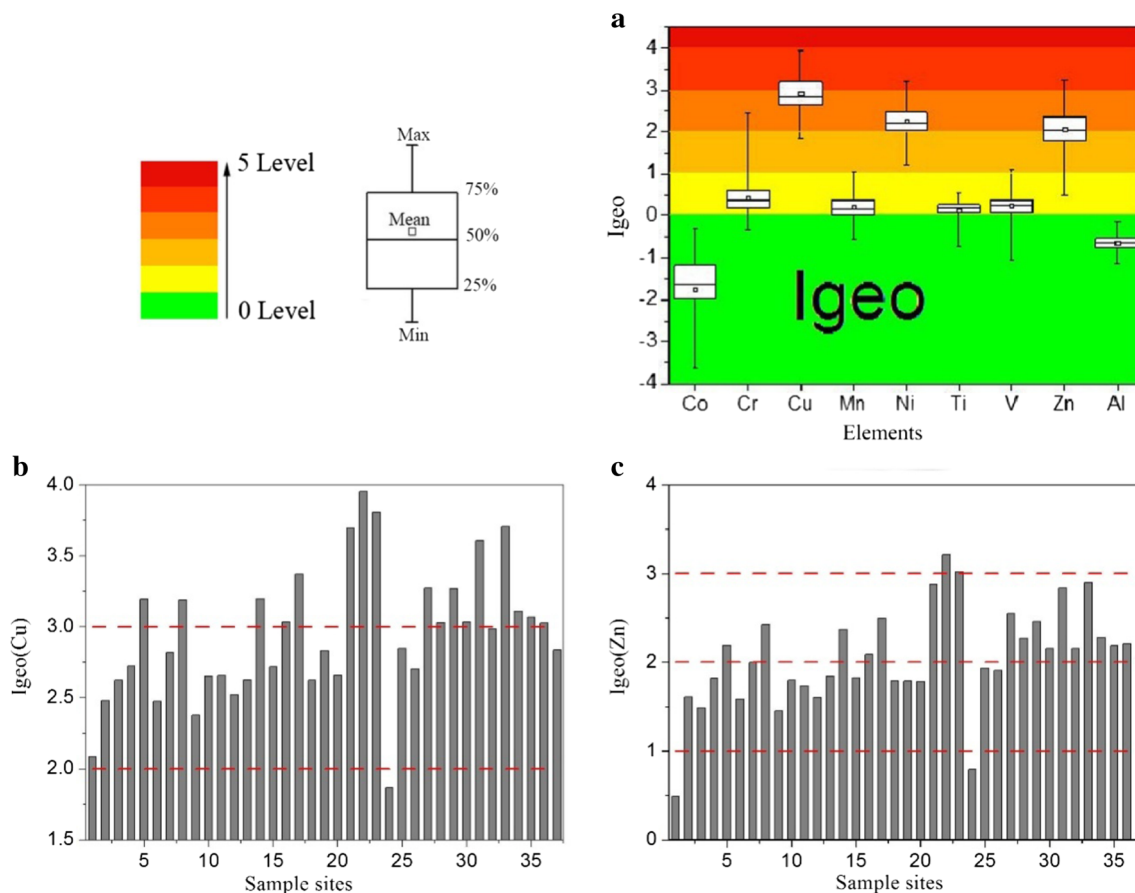


Fig. 3 Geoaccumulation index (I_{geo}) value of f heavy metals in the RSS of the IMYR, China (a), and the I_{geo} values of Cu (b) and Zn (c) in each sampling site present in b and c, respectively

contamination, it is important to analyze and control the sources of pollution. Heavy metals in sediments often exhibit complex interrelationships due to the disturbance of the original concentration of the parent materials and contamination by human activities (Chen et al. 2007a; Li and Zhang 2010; Islam et al. 2015; Paramasivam et al. 2015). PCA, HCA, and Pearson correlation analysis were conducted in the present study to determine the sources of heavy metals and the causes of their heavy metal contamination (Huang et al. 2013; Liu et al. 2003; Loska and Wiechula 2003; Quan et al. 2014; Singh et al. 2004; Zahra et al. 2014; Varol 2011; Zheng et al. 2008).

Source analysis based on PCA

To further examine the extent of metal contamination in the study area, we performed PCA analysis on the metals. PCA is performed on the correlation matrix between different parameters followed by Varimax rotation. It gives two PCs with eigenvalues >1, explaining 87.51 % of the total variance (Table 3). PC1 accounted for 73.49 % of the total variance and is mainly characterized by high positive loadings of variables such as Cu, Mn, V, Zn, Al, Ti, and Co. Moreover, a moderately positive loading of variables, which is present in

PC2 (Ni), also presents a significant positive loading in PC1. The second PC accounted for 14.02 % of the total variance, which consists only of Cr with high positive loading (Fig. 6; Table 3). In PC1, the loading coefficients of Cu, Mn, V, Zn, Al, and Ni were larger than 0.85, implying perhaps a common source (Han et al. 2006). In this case, the Ti loading (0.639) is not as high as the loadings of the other elements of the group and is separated from the other elements in PC1 (Fig. 6a; Table 3), which may, therefore, imply quasi-independent behavior within the group (Han et al. 2006). Co is the only element with negative loading in the PC loading plot, suggesting that the source of Co in the RSS is different from the other metals, and it can be classed into a separate group. Overall, the heavy metals in the RSS of the IMYR were classed into four groups by PCA as follows: group 1 consisted of Cu, Mn, V, Zn, Al, and Ni; groups 2, 3, and 4 consisted only of Ti, Co, and Cr, respectively.

Source analysis based on HCA

Based on the information derived from the PCA above, we performed HCA on the data. Distance metrics are based on the Wards method, using squared Euclidean distance. As shown

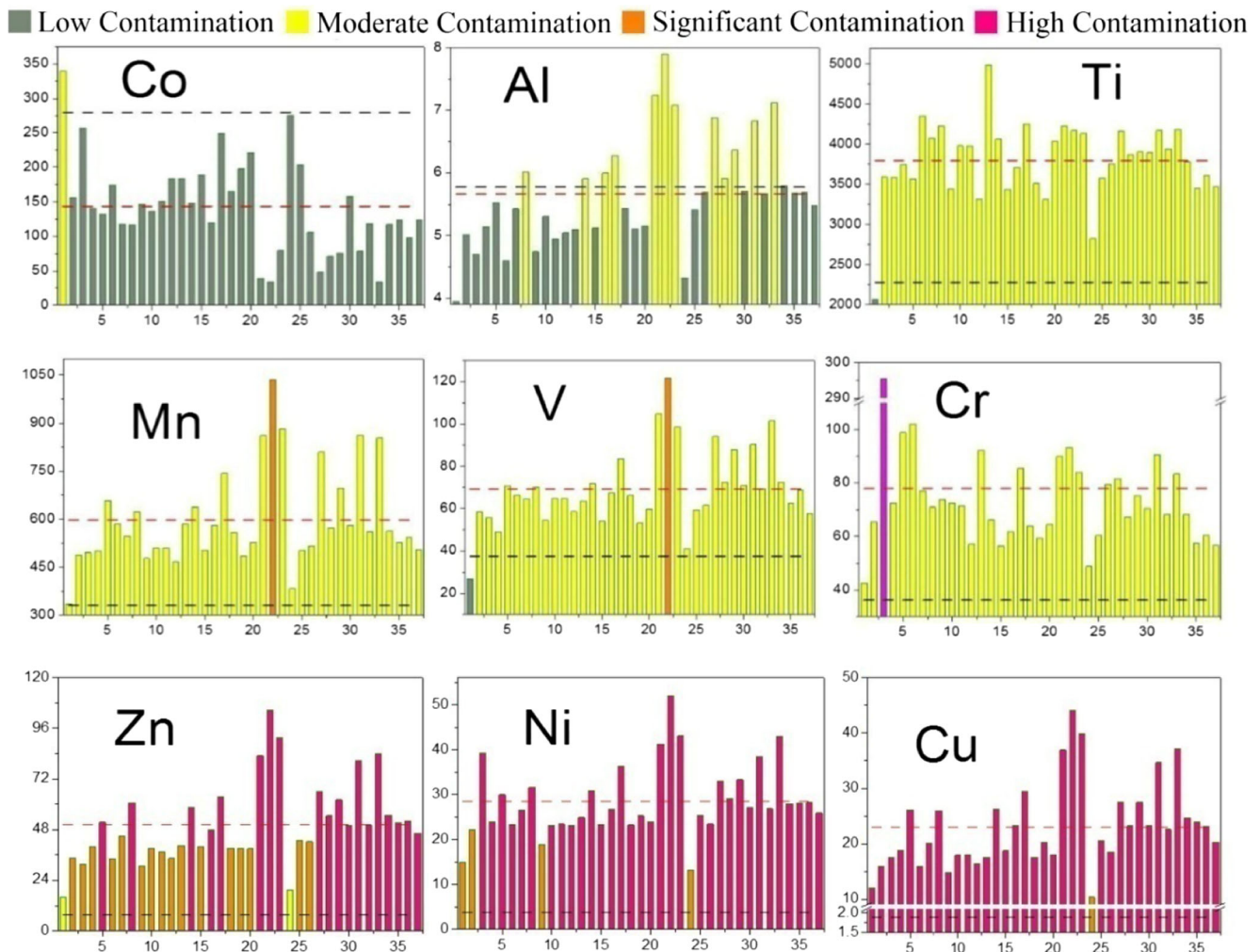


Fig. 4 The spatial distribution of heavy metal concentrations in the RSS of the IMYR and their evaluated contamination factors (CF). The red and black dashed lines reflect the average concentration and background value for each heavy metal, respectively

in Fig. 6b, the variables taken for this analysis are the same as for the PCA. In this dendrogram, nine elements in the RSS of the IMYR are grouped into five significant clusters based on the similarities between them. Cluster 1 consisted of Cu, Mn, V, Zn, and Al. Cluster 2 consisted of Ni, Ti, Co, and Cr. As the

similar source of Ni and the elements in cluster 1 (Han et al. 2006) and the distance between Ni and the elements in cluster 1 were apparently less than the distance between Ni and Ti, Co, and Cr, respectively, (Fig. 6b), Cu, Mn, V, Zn, Al, and Ni were classed into the same cluster. Clusters 3, 4, and 5 consisted only of Ti, Co, and Cr, respectively, due to the differences of their sources (Han et al. 2006).

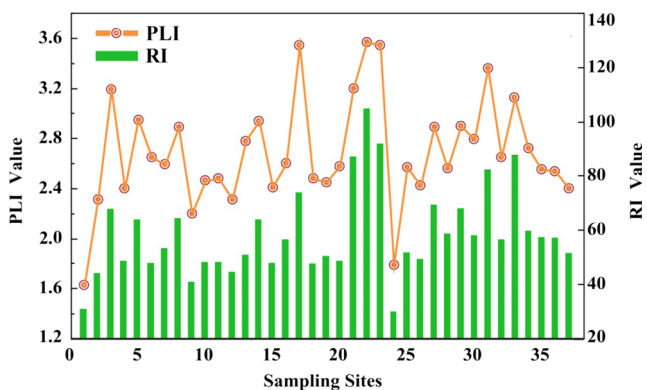


Fig. 5 Heavy metal potential ecological risk indexes and pollution loading indexes of the IMYR

Source analysis based on Pearson analysis

High correlation coefficients between different metals mean common sources, mutual dependence, and identical behavior during transport (Liu et al. 2003; Mohammad et al. 2010; Wang et al. 2016; Islam et al. 2015; Paramasivam et al. 2015; Zheng et al. 2008). The correlation coefficients between Cu, Mn, V, Zn, Al, Ni, and Ti were larger than 0.5 at the $p < 0.01$ level, indicating that these metals are associated with each other and may have common sources in the sediments (Table 3). Cu, Mn, V, Zn, and Al are significantly positively

Table 3 Statistical results of PCA and Pearson correlation analysis

Pearson correlation matrix of metals in the INIYR:

	Co	Cr	Ti	Cu	Mn	Ni	V	Zn	Al
Co	1								
Cr	0.090	1							
Ti	-0.547**	0.199	1						
Cu	-0.702**	0.082	0.502**	1					
Mn	-0.694**	0.143	0.624**	0.948**	1				
Ni	-0.601**	0.420**	0.540**	0.927**	0.904**				1
V	-0.773**	0.119	0.652**	0.929**	0.968**	0.889**	1		
Zn	-0.759**	0.045	0.564**	0.990**	0.957**	0.912**	0.950**	1	
Al	-0.798**	0.004	0.551**	0.957**	0.936**	0.871**	0.950**	0.976**	1

Principal component analysis of metals in the ININR:

Latent roots:

1	2	3	4	5	6	7	8	9
6.698	1.179	0.653	0.324	0.0766	0.034	0.023	0.009	0.004

Rotated loading matrix:

	1	2
Co	-0.826	0.193
Cr	0.026	0.982
Ti	0.639	0.242
Cu	0.964	0.067
Mn	0.962	0.138
Ni	0.884	0.407
V	0.975	0.099
Zn	0.986	0.027
Al	0.982	-0.030

Variance explained by rotated components:

1	2
6.614	1.262

Percent of total variance explained:

1	2
73.493	14.021

**Correlation is significant at the 0.01 level (two-tailed)

correlated (the correlation coefficients are larger than 0.9), which may suggest a common origin, while Ni formed the same group based on the significantly positive correlation of Ni, V, and Al (Ni vs. V for 0.889 and Ni vs. Al for 0.871). Although the correlation coefficients between Ti, Cu, Mn, V, Zn, Al, and Ni appeared significant (larger than 0.5), the values of these correlation coefficients are not as high as the other metals mentioned above, implying an independent source within a single group. Co is negatively correlated with the other metals, reflecting different sources of Co compared with the other elements. The absence of correlation among the metals suggests that the concentrations of these metals are not controlled by a single factor but by a combination of geochemical support phases and their mixed association (Islam et al. 2015; Paramasivam et al. 2015). Hence, in the present

study area, Cr is moderately to slightly correlated with the other metals, which is clustered for one group.

The results of cluster and principal component analyses match well with the Pearson correlation analysis. From the overall statistical analyses, nine elements in the RSS of the IMYR were classed into four significant groups: group 1 consisted of Cu, Mn, V, Zn, and Al; and groups 2, 3, and 4 consisted only of Ti, Co, and Cr, respectively.

Identification of heavy metal contamination

Because the calculated I_{geo} and CF values of Al (-1.13 to -0.13 for I_{geo} and 0.68 to 1.37 for CF) indicate non-contamination to low contamination in the RSS of the IMYR, the aluminous abundance in the Earth’s crust is

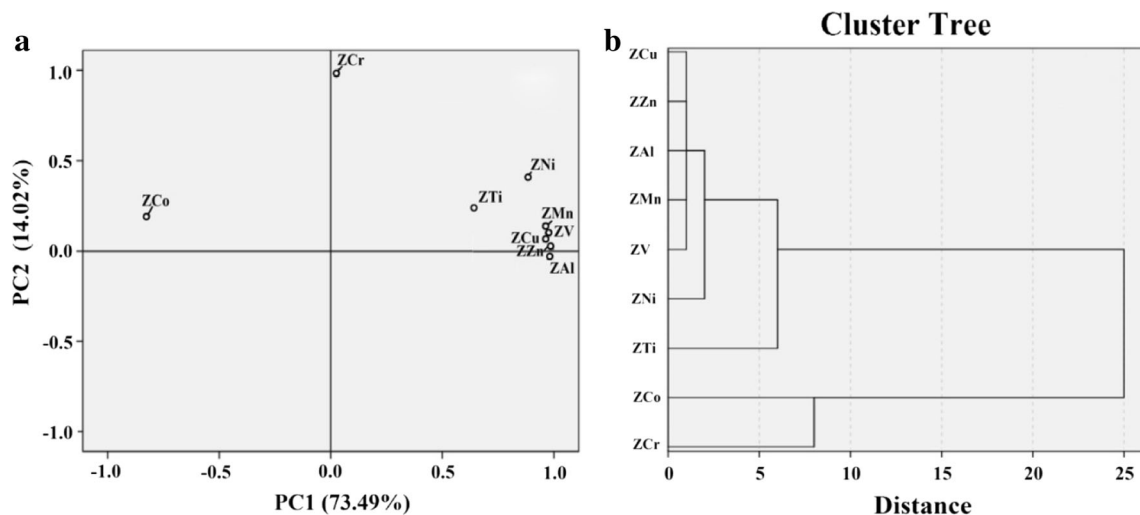


Fig. 6 Principal component (PC) loadings of the first two PCs of the heavy metals in the RSS of the IMYR (a). HCA shows the relevant association among the parameters (b). Distance metrics are based on the Ward's distance single linkage method (b)

relatively high (Pekey et al. 2004; Zhang et al. 2009; Li and Zhang 2010), which may indicate, therefore, that the Al comes predominantly from natural origins. For instance, the higher EF, I_{geo} , and CF values of Cu, Zn, and Ni indicate that these metals are the major contributors to sediment pollution and mainly controlled by anthropogenic activities such as non-ferrous metal mining and refining, manufacture and application of chemical phosphate fertilizers, organic fertilizers and pesticides, and waste disposal (Han et al. 2006; Bai et al. 2012; Wang et al. 2011). However, combined with rock weathering and soil formation (Huang et al. 2007; Li and Zhang 2010), the moderate contamination of Mn and V in the RSS was due to the dual influences of anthropogenic activities and natural origins. The Ti in the RSS of the IMYR faces a similar situation. Although Ti is an element that is widely applied in the petroleum, coal, metallurgy, and chemical industry (Mohammad et al. 2010), the CF, EF, and I_{geo} values of Ti showed only moderate contamination. The significant high contamination of Cr, observed in the areas bordering the Bayan Nur (sampling site 3; Figs. 2 and 4), might be due to the effects of mining, metal smelting and manufacturing in this city. However, the residual sampling sites only reach the moderate contamination level, which indicates natural sources of contamination (Fig. 3).

Among the sites, the range of CF, EF, and I_{geo} values for Co were 0.12 to 2.21, 0.09 to 1.77, and -3.62 to -0.31 , respectively, indicating an unpolluted to moderately polluted status of the RSS. Combined with the result of the descriptive statistical analysis that the average concentration of Co was less than its background value, the Co in the RSS of the IMYR was not due to anthropogenic activities. Further evidence shows that the surface sediments from the Ulan Buh Desert and the Hobq Desert also had high concentrations of Co (280–300 mg/kg; Guan et al. 2016; Pan et al. 2015), almost two

times that of the surface sediments in the riverbeds (143.52 ± 68.58 mg/kg), and they can be transported into the RSS by wind erosion and solifluction to increase the concentration of Co in the RSS.

Conclusion

In the present investigation, high PLI and RI values of heavy metals were presented in the urban areas bordering the Yellow River, which suggests that the RSS of the IMYR is moderately polluted (the average value of PLI is 2.65) by heavy metals and might create a low risk (the average value of RI is 58.34) to this riverine ecosystem. The concentrations of Zn, Ni, and Cu in the RSS of the IMYR were higher than their background values; heavy contamination of these metals was determined by the values of EF, I_{geo} , and CF, and this was due to the influence of anthropogenic activities. The moderate contamination of Ti, Mn, V, and Cr indicated by the values of I_{geo} and CF was partly ascribed to the influence of anthropogenic activities. The slight to no contamination with Co ($EF < 1.5$; $I_{geo} < 0$; $CF < 1$) shows that it is mainly from the bordering deserts. The Co and Al in the RSS were mainly from natural sources because the I_{geo} and CF values indicate non-contamination. However, the concentrations of Co in the RSS bordering the deserts were high due to the inputting of desert materials. The spatial distributions of Mn, Ni, V, Zn, and Cu concentrations were similar in that high concentrations occurred in the areas bordering the cities of Baotou and Dalad Banner. In addition, high concentrations of Cr and Ni also occurred in the area bordering Bayan Nur city, which suggests that industrial activities from these cities played a major role in the heavy metal contamination of these areas.

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