Investigation of the Heavy Metal Contamination of the Sediments from the Yellow River Wetland Nature Reserve of Zhengzhou, China

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Abstract

Background: Heavy metal pollution in the sediment of the Yellow River draws wide attention in the recent years. The Yellow River Wetland Nature Reserve of Zhengzhou is one of the major wetlands of the river and located at the beginning of the lower reach. In this article, we aimed to investigate the degree and the sources of the metal pollution in the reserve.

Methods: Metals as Cu, Pb, Cr, Cd and Mn in the sediment were monitored using flame atomic absorption spectrometry. The index of geo-accumulation (I_{geo}) and the modified degree of contamination (mC_d) were developed to evaluate individual metal pollution and overall enrichment impact of the elements.

Results: Compared with sediment quality guidelines, the effect of Cr and Pb are more serious than others. I_{geo} values show Pb pollution are moderate at the Xinzhai, Langchenggang and Nansutan sites, and mC_d analysis indicate the whole contamination at the Wantan, Langchenggang and Nansutan sites was low. Principal component analysis indicated that the first factor was Cu, Mn and Cd, mainly from soil erosion and the irrational use of phosphate fertilizers; the second Pb from fossil fuel burning; and the third Cr from weathering process.

Conclusion: We conclude that Pb contamination is serious in the reserve, and the main sources of the metal are crude oil consumption and coal combustion of the brick kilns around. We also draw a conclusion that it is vital to evaluate contamination degree with both individual elements and overall average.

Keywords: The Yellow River, Wetland, Sediment, Heavy metal pollution, China

Introduction

Sediments are the main sink for pollutants and have been recognized as the important indicators of water contamination (1), e.g. metals discharged. In the aquatic environment, heavy metals tend to be incorporated into the bottom sediments (2-4), and can be released by various processes under favourable conditions. Thereby metals reach the aquatic life community and human beings through food chain and cause great concern because of the potential health risks to the local inhabitants (1). Many studies have documented metal contamination in sediment for ecosystem quality assessment and widespread incidence of metal contamination in sediment (5-8).

The Yellow River, the second longest river in China, is a famous sediment-laden river. Heavy metal pollution in the sediment draws wide attention in the recent years (3, 9-12). At the Lanzhou reach of the upper Yellow River, the

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levels of heavy metals (Cd, Pb, Cr, Cu, and Zn) are proposed to influence the abundance and structure of nematode communities (9). In the middle stretch of the Yellow River, e.g. Inner Mongolia, the heavy metals mentioned above in the bed mud are evaluated to be in the non- to mid-pollution condition, and the moderate ecological risk of Cd, Cu, Pb and Cr are proposed in this area (12).

The lower reach of the Yellow River, from Huayuankou in Henan Province to the estuary in Shandong Province, is an important area in terms of agriculture, ecology and petroleum development (13). Since the 1960s, a series of large reservoirs were built in the upper and middle reaches of this river, which causes the accumulation of the silt in the lower reach and the forming of the hanging river (i.e., a river channel elevated above its floodplain) (14). In a sense, the sediment contamination has more serious impact on the organisms than the water in the lower reach of the Yellow River.

The Yellow River Wetland Nature Reserve of Zhengzhou is located at the beginning of the lower reach, on the north of Zhengzhou city. It is one of the major wetlands of the river, and also locates in the centre migratory route of the birds in Asia. This area has a large population, and the wetland landscape is challenged by the expanding demands of the land use due to local economic development. The expanding land use has significant effects on the natural ecosystem and leads to the accumulation of the heavy metals in the soil (15). Both Chen et al. (10) and Zhang et al. (16) focused on the whole lower reach of the Yellow River, and revealed the forms and the mobile of the heavy metals: A large proportion of trace metals remains in the primary phase (residual phase: fixed within the crystalline lattice); among the secondary mineral phases, Cu is mainly associated with organic matter and amorphous iron oxides; the major chemical forms for Pb are with iron and manganese oxides; the non-residual form of Mn exists mainly as carbonates and oxides; most Cd is mainly associated with the cryptocrystalline

manganese oxide, reactive iron oxides and carbonate; and Cr mainly exists in the oxide forms.

However, further investigation to cover the entire reserve is required to understand the environmental impacts in the reserve quantitatively and qualitatively. The main objectives of the present study are to assess the extent and degree of metals along the study region, and the origin of these metals.

Materials and Methods

Study Region

The study region is located between 113°27′–114°11′ E longitude and 34°48′–35°6′ N latitude, on the axis of the Yellow River Alluvial Fan, and is covered by mainly loose deposits. The river flows on the modern alluvial deposits and the river bed is 3 m (in some places even 10 m) above the surface of the closely related flooding plane. The study area lies close to the Loess Plateau. The soil type in this region is loosely silt, thin-grained sand and clay, overlying the Quaternary sediments. In the reserve, there are dominated with different flora in wild land, e.g. *Imperata cylindrica. Var. major*, *Cynodon dactylon*, *Glycine soja* and Tamarix chinensis; and some other places are used for the farmland, e.g. *Triticum aestivum* and *Brassica campestris*. Three sites were selected to represent the wild land: the Nanguotou (NGT), the Xinzhuang (XZU) and the Nansutan (NST); and four sites to represent the farmland: the Madu (MD), the Wantan (WT), the Xinzhai (XZA), and the Langchenggang (LCG) (Fig. 1). The LCG site also represents a special condition, because hundreds brick kilns scattered around it.

Sampling, Preparation and Analysis

The sediment samples were collected with stainless steel containers and wrapped in the polyethylene plastic bags during March – May in 2008. All of the samples were extracted at both top horizon (5-20 cm deep from the surface)

and sub-horizon (20-40 cm deep from the surface). Four replicate samples were taken at each site to reduce the probability of random performance.

Sediments were air dried at room temperature, sieved through a 2 mm nylon mesh to remove the coarse debris, then ground using a mortar and pestle until all particles passed through 100-mesh nylon sieve. 0.5 g of each sample was digested twice with 5 ml of 3:1:1 HCl: HNO_3: HF, followed by the addition of 5 ml aqua regia (HNO_3: HCl = 1:3) in capped Teflon tubes. Then a small amount of nitric acid was added intermittently to digest the sediment completely until the supernatant became clear. Digested solution was diluted in 1 % HNO_3 (v/v) to 50 ml, and the final solution was passed through a 0.45 μm membrane filter for future analysis.

All glassware used were cleaned with 15% HNO_3 (v/v), followed with repeated rinsing using deionised water. Heavy metal concentrations of Cu, Pb, Cd, Cr, and Mn were analyzed by flame atomic absorption spectrometry (FAAS) with air-acetylene flame. Concentrations of metals were given relative to dry mass (DM).

Evaluation of Contamination Degree of Heavy Metals

The index of geo-accumulation (I_{geo}) and the modified degree of contamination (mC_d) were simple measures to quantify metal accumulation in contaminated sediments in terrestrial, aquatic and marine environments, and have been widely applied for evaluating individual metal pollution and overall enrichment impact of groups of pollutions in sediments [17-19]. The geoaccumulation index (I_{geo}) is expressed as follows [20]:

$$I_{geo} = \log_2 \frac{C_n}{1.5 B_n}$$

Where C_n is the measured concentration of the heavy metal in the sediments; B_n is the background or pristine value of the element. Different geochemical background may result in the variation of heavy metal pollution information

in different parts. Therefore the local geochemical background levels of Henan province [21] is used in the assessment. The constant 1.5 is the background matrix correction factor due to lithogenic effects [17-18, 22]. The contamination level may be classified descriptively for increasing I_{geo} values (I_{geo} class 0: $I_{geo} \leq 0$, uncontaminated (UC); class 1: $I_{geo} < 1$, uncontaminated to moderately contaminated (UMC); class 2: $I_{geo} < 2$, moderately contaminated (MC); class 3: $I_{geo} < 3$, moderately to strongly (MS); class 4: $I_{geo} < 4$, strongly contaminated (SC); class 5: $I_{geo} < 5$, strongly to extremely contaminated (SEC); class 6: $I_{geo} > 5$, extremely contaminated (EC)) [23].

The modified degree of contamination (mC_d) is given below [17, 24]:

$$C_f^i = \frac{C_n}{B_n}$$

$$mC_d = \frac{\sum_{i=1}^{i=n} C_f^i}{n}$$

Where n is the number of analysed elements; i is the number of pollutants; C_f^i is the contamination factor. For classification and description of the modified degree of contamination, the gradation are proposed [17]: $mC_d < 1.5$, nil to very low degree of pollution; $1.5 \leq mC_d < 2$, low degree of pollution; $2 \leq mC_d < 4$, moderate degree of pollution; $4 \leq mC_d < 8$, high degree of pollution; $8 \leq mC_d < 16$, very high degree of pollution; $16 \leq mC_d < 32$, extremely high degree of pollution; $mC_d > 32$, ultra high degree of pollution.

Statistical Analysis

Statistical analysis of data was performed using the software package SPSS 13.0. Significant differences between means were tested by one-way Analysis of Variance (ANOVA). The original variables were made to clarify the loadings of five heavy metals at the regional scale. Differences at 0.95 confidence level ($P < 0.05$) were considered significant. To interpret the relations between heavy metals in geochemical

processes, correlation analysis was followed by Spearman correlation with 2-tailed test and principal component analysis (PCA) with none rotated method.

Results

Table 1 shows that the concentration values of Pb, Cr, Mn and Cd are higher in most of the samples than the LBL values. The highest value of Cr is 265.22 mg kg^{-1} in sub-horizon sediments of wild land at the NGT site, which is about four times of the LBL of Cr (63.80 mg kg^{-1}). Only one mean value of Cr (51.27 mg kg^{-1} at the MD site) is lower than the LBL. The peak value of Mn contents is 3113.7 mg kg^{-1} in the sample from top-horizon sediment at the XZA site, about five times of LBL of Mn (579.0 mg kg^{-1}). Only at the XZU site, the Mn concentration value (489.89 mg kg^{-1}) is lower than the LBL, and is also significantly lower than the highest mean (1220.58 mg kg^{-1}) at the XZA site. The highest average value of Cd is recorded at the LCG site (0.19 mg kg^{-1}), then at the NST site (0.15 mg kg^{-1}), and the WT site (0.13 mg kg^{-1}); all of them are significantly higher than that at the XZU site (0.03 mg kg^{-1}). Cd concentrations in the farmland are higher than that in the wild land. The mean concentration value of Pb peaks at the NST site (61.94 mg kg^{-1}), then at the LCG (43.47 mg kg^{-1}), both significantly different from the other sites. Along the study region, the contamination of Pb began to occur mainly from the XZA site (40.03 mg kg^{-1}). The mean concentration of Pb in the wild land is higher than that in the farmland. The concentrations of Cu exceed the local background level in Henan Province (LBL) of Cu (19.70 mg kg^{-1}) at the NGT and the WT sites (27.05 and 23.15 mg kg^{-1}, respectively), and, at the other sites, are slightly lower.

Details of the I_{geo} values for the individual elements at different sites in the reserve are in Table 2. The negative I_{geo} values in the table are the results of relatively low levels of contamination for some metals in some sites and the background variability factor (1.5) in the I_{geo} equation. There is no or only very light pollution for Cu, Cr, Pb, Mn and Cd in the study region with the exception of moderate pollution of Pb at the XZA (1.03), the NST (1.66) and the LCG (1.15) sites.

For overall enrichment impact of the elements, mC$_d$ can present integrated assessment of the pollutions in the sediments. Table 3 shows the degrees of contamination are low at the WT (1.57), NST (1.80) and LCG sites (1.74), and nil to very low at the other sites. The mC$_d$ data suggest light anthropogenic impact in all sites. The main contamination factor at the WT site is Cr, whose C$_f$ is 2.58, and at NST and LCG sites are Pb and Cd. It is vital to evaluate contamination degree with both individual elements and overall average.

To investigate and determine possible sources of the heavy metals in the sediments, Spearman correlation with 2-tailed test is applied to the values of the metal concentrations data set. Table 4 shows that the relationship between Cu and Cd ($r = 0.471$) is significant ($P < 0.05$), which suggests that these heavy metals in sediments have common sources, or mutual dependence and/or similar behaviour during transport, or is subject to certain factors control. The significant probability between Mn and Cu ($r = 0.403$) is 0.070, and Cd ($r = 0.394$) is 0.078.

Further examination of the sources of the heavy metals was made with principal component analysis (PCA) applied to data reduction. Table 5 shows that three factors explain a relatively large extent of the total variance (80.32 %) of the five variables used in the analysis. Cu, Cd and Mn were showed an association with the first factor (variance 30.67 %). Elemental correlation also shows that three heavy metals have significant or nearly significant relationships. That is to say these metals have similar sources. Pb shows an association with the second factor (variance 26.61 %), and the large load of the third factor (variance 23.03 %) is Cr.

Table 1: Mean, standard deviation, minimum and maximum value of heavy metal concentrations in sediments (mean ± S.D) at seven sampling sties ($P < 0.05$) (mg kg-1 dry weight)

	Cu	Cr	Pb	Mn	Cd
NGT	27.05 ± 9.36^{c}* (19.01-37.3)	94.71 ± 71.84^{ab} (31.72-172.93)	22.53 ± 14.56^{ab} (7.89-37.0)	751.0 ± 147.1^{ab} (634.3-916.3)	0.10 ± 0.06^{ab} (0.04-0.16)
MD	14.65 ± 0.41^{b} (14.18-14.92)	51.26 ± 25.46^{a} (36.24-80.66)	6.23 ± 0.31^{a} (5.88-6.47)	879.6 ± 45.5^{ab} (828.0-913.5)	0.04 ± 0.02^{ab} (0.03-0.06)
WT	23.15 ± 2.80^{bc} (20.35-25.94)	164.89 ± 3.72^{b} (161.2-168.67)	10.11 ± 3.87^{a} (6.24-13.97)	1031.6 ± 29.1^{ab} (1002.5-1061)	0.13 ± 0.03^{ab} (0.10-0.15)
XZA	9.26 ± 3.72^{ab} (5.17-12.43)	68.69 ± 22.65^{a} (46.45-91.72)	40.03 ± 4.77^{b} (35.15-44.69)	1220.6 ± 658.3^{b} (566.3-1882.7)	0.06 ± 0.02^{ab} (0.05-0.08)
XZU	4.83 ± 3.50^{a} (1.33-8.32)	97.55 ± 3.49^{ab} (94.06-101.03)	36.15 ± 1.75^{b} (34.40-37.89)	489.9 ± 72.2^{a} (417.7-562.1)	0.03 ± 0.01^{a} (0.02-0.03)
NST	14.55 ± 4.63^{b} (9.92-19.17)	113.47 ± 11.99^{ab} (100.25-123.7)	61.94 ± 12.56^{c} (49.38-74.5)	678.5 ± 67.6^{ab} (610.8-746.1)	0.15 ± 0.05^{ab} (0.10-0.20)
LCG	14.81 ± 2.41^{b} (12.58-17.37)	98.70 ± 74.66^{ab} (20.20-168.82)	43.47 ± 23.10^{bc} (20.4-66.6)	853.7 ± 422.3^{ab} (514.2-1326.6)	0.19 ± 0.10^{b} (0.03-0.39)
Average	15.47 ± 8.18	98.47 ± 48.70	31.49 ± 21.11	843.56 ± 422.31	0.10 ± 0.09
Different depth (top-horizon, n=28; sub-horizon, n=28)					
Top-horizon	18.53 ± 7.66 (8.28-36.82)	97.55 ± 38.83 (31.50-130.11)	30.52 ± 23.76 (5.88-74.50)	921.1 ± 684.1 (343.2-3113.7)	0.09 ± 0.10 (0.02-0.39)
Sub-horizon	12.69 ± 10.03 (0.20-37.81)	99.78 ± 80.88 (13.36-265.22)	30.69 ± 20.45 (5.88-71.28)	804.2 ± 350.2 (417.7-1790.6)	0.09 ± 0.11 (0.02-0.39)
Tillage condition (wild land, n=12; farmland, n=16)					
Wild land	15.48 ± 11.11 (1.33-37.32)	101.91 ± 37.50 (31.72-172.96)	40.20 ± 19.84 (7.89-74.5)	639.9 ± 146.6 (417.7-916.3)	0.09 ± 0.07 (0.02-0.20)
Farmland	15.47 ± 5.65 (5.17-25.94)	95.88 ± 57.21 (20.20-168.82)	24.96 ± 20.37 (5.88-66.57)	996.4 ± 367.5 (514.2-1882.7)	0.11 ± 0.10 (0.03-0.39)
LBL[#]	19.70	63.80	19.60	579.0	0.07

* Means with the same letter in the same column are not different based on least significant difference;
# Local background level in Henan Province (EMC 1990).

Table 2: Index of geoaccumulation (I_{geo}) and contamination level for mean metal concentrations in the sediments

Sites	Cu	Cr	Pb	Mn	Cd
NGT	-0.13(UC)	-0.01(UC)	0.20(UMC)	-0.21(UC)	-0.12(UC)
MD	-1.01(UC)	-0.90(UC)	-1.65(UC)	0.02(UMC)	-1.28(UC)
WT	-0.35(UC)	0.78(UMC)	-0.96(UC)	0.25(UMC)	0.27(UMC)
XZA	-1.67(UC)	-0.48(UC)	1.03(MC)	0.49(UMC)	-0.65(UC)
XZU	-2.61(UC)	0.03(UMC)	0.88(UMC)	-0.83(UC)	-1.98(UC)
NST	-1.02(UC)	0.25(UMC)	1.66(MC)	-0.36(UC)	0.51(UMC)
LCG	-1.00(UC)	0.04(UMC)	1.15(MC)	-0.02(UC)	0.86(UMC)

Table 3: Modified degree of contamination (mC_d) and contamination factors (C_f)

Sites	Contamination factors (C_f)					sumC$_f$	mC$_d$
	Cu	Cr	Pb	Mn	Cd		
NGT	1.37	1.48	1.15	1.30	1.38	6.69	1.34
MD	0.74	0.80	0.32	1.52	0.62	4.00	0.80
WT	1.17	2.58	0.52	1.78	1.81	7.87	1.57
XZA	0.47	1.08	2.04	2.11	0.95	6.65	1.33
XZU	0.25	1.53	1.84	0.85	0.38	4.85	0.97
NST	0.74	1.78	3.16	1.17	2.14	8.99	1.80
LCG	0.75	1.55	2.22	1.47	2.71	8.71	1.74

Table 4: Correlation coefficient between different metals in sediments of the Spearman correlation with 2-tailed test (significant probabilities are given in parenthesis)

	Cu	Cr	Pb	Mn	Cd
Cu	1.000				
Cr	0.209 (0.363)	1.000			
Pb	-0.357(0.112)	0.069(0.767)	1.000		
Mn	0.403(0.070)	-0.236(0.302)	-0.182(0.430)	1.000	
Cd	0.471*(0.031)	0.227(0.323)	0.381(0.089)	0.394(0.078)	1.000

* Correlation is significant at the 0.05 level

Table 5: Factor load of the heavy metals in sediments

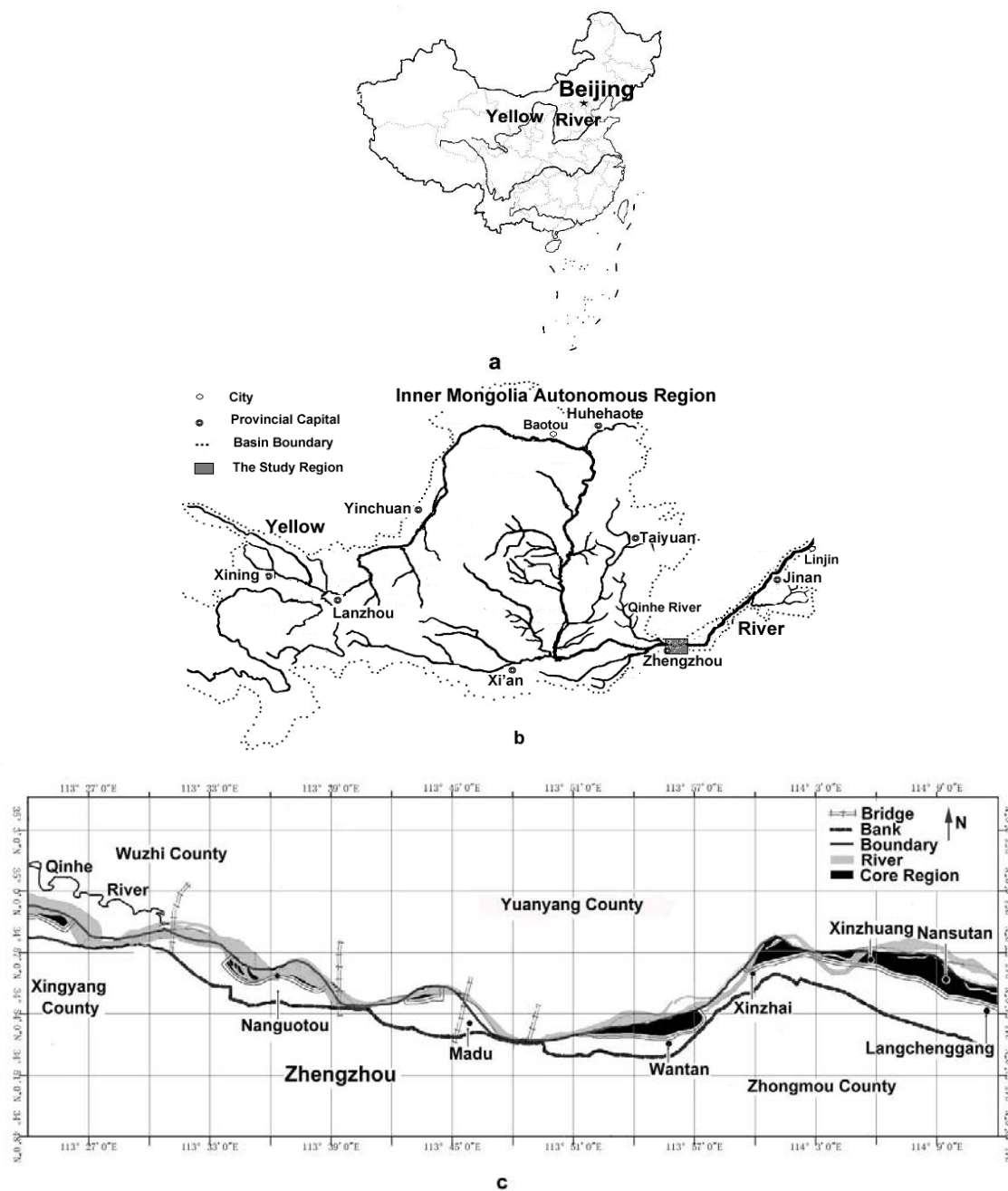| Metals	Component Matrix [a]		
	F1	F2	F3
Cu	0.588	-0.601	0.241
Cr	0.037	-0.279	0.848
Cd	0.845	0.338	0.124
Pb	0.200	0.869	0.305
Mn	0.657	-0.145	-0.516

a principal component analysis

Fig. 1: Map showing the research area and the geographical location of the sampling sites. a: the whole China map; b: a zoomed view of the Yellow River; c: detailed sample sites

Discussion

In the reserve, the average concentration value of Cu of all seven sites (15.47 mg kg^{-1}) is a little less than the result of Zhang et al. (16), while

that of Mn and Cr nearly double times. Though the contamination degrees are low, mC$_d$ data show that the main contamination factor at NST

and LCG sites is Cd. Because of the river's distinct loess series soil type and loess rock properties (10), soil erosion is an important source of the concentrations of Mn, Cu and Cd in the Yellow River. Silt has high ability in copper sorption, but within certain pH values, the precipitation proportion of Cu ions reduces with the sediment concentration increasing (25, 26). The pollution source of Cd in the farmland mainly also comes from the irrational use of phosphate fertilizers (15). For a large proportion of exchangeable and carbonate-bound Cd exist in the Yellow River sediment (10), the use of phosphate fertilizers should be reduced.

Comparisons with those of other studies along the Yellow River, the Lanzhou section (3, 9), the Inner Mongolia stretch (12), and the delta land formed in 2006 (27), the average concentration of Pb at Zhengzhou is the highest. This shows that the main source of Pb is at the beginning of the lower reach of the Yellow River. Chen et al. (10) also reported that a large proportion of exchangeable and carbonate-bound Pb exist in the sediment in this area. I_{geo} analysis indicates the moderate pollution of Pb at the XZA, the NST and the LCG sites, and mC_d results also showed Pb was one of the main contamination factors at NST and LCG sites.

Near the LCG site, there are hundreds brick kilns. Airborne trace metal levels were prior to industrializations (28). Atmospheric pollution, such as coal combustion of the brick kilns and consumption of crude oils, seriously effects the concentrations of Pb in the soil (29-31) and is one of the main reasons for the high environmental risk. The air pollution may also impact the environment of the NST site, only several kilometres west to the LCG site. Therefore, it is necessary to shut off the brick kilns and reduce the use of crude oils in the reserve.

In the study, the mC_d data show that the main contamination factor at the WT site is Cr, and correlation coefficients indicate that the relationships between Cr and others elements are weak. Zhang et al. (16) reported that 94.1 % percentage of Cr was in the residual fraction and the origin of Cr mainly depended on the weathering degree, controlled not by a single factor but by a combination of geochemical support phases and their mixed associations (32).

In conclusion, the concentration values of Pb, Cr, Mn and Cd in most of the sediment samples of the reserve are higher than the background values of Henan Province, while most concentrations of Cu are comparable. Soil erosion, irrational use of phosphate fertilizers, weathering process, and coal combustion of the brick kilns around, and crude oils consumption are the main sources of the heavy metals. For the evaluation of individual element contamination degree, Igeo values showed that the pollution of Pb at the XZA, the NST and the LCG sites were moderate, and others are uncontaminated to moderately contaminate; and of overall average, mCd suggested the LCG, the WT and the NST sites with low degree of contamination. Compared with the sediment quality guidelines, the enrichments of Cr and Pb are more toxic than other elements, while Cd and Cu have little harmful effects on the organisms. It is necessary to evaluate contamination degree with both individual elements and overall average, and with a focus on ecosystem health and protection of organisms in freshwater ecosystem.

Ethical considerations

Ethical issues (Including plagiarism, Informed Consent, misconduct, data fabrication and/or falsification, double publication and/or submission, redundancy, etc) have been completely observed by the authors.

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of the sediment samples. The authors declare that there is no conflict of interests.

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