POLLUTANT DISTRIBUTIONS AND CONTROLS IN THE ENVIRONMENT

Risk analysis on heavy metal contamination in sediments of rivers flowing into Nansi Lake

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Abstract In order to understand the risk of heavy metals in sediments of the rivers flowing into Nansi Lake, 36 surface sediments were sampled from six rivers and seven heavy metals (Cr, Cu, Ni, Zn, As, Pb, and Cd) were determined. Potential ecological risk index (RI) of the six rivers showed significant differences: Xinxue River, Jiehe River, and Guangfu River were at medium potential risk, whereas the risk of Chengguo River was the lowest. Jiehe River, Xuesha River, and Jiangji River were meeting the medium potential risk at river mouths. Geo-accumulation index (I_{geo}) of the seven heavy metals revealed that the contamination of Cu and Cd was more serious than most other metals in the studied areas, whereas Cr in most sites of our study was not polluted. Moreover, correlation cluster analysis demonstrated that the contamination of Cu, Ni, and Zn in six rivers was mainly caused by local emissions, whereas that of As, Pb, and Cd might come from the external inputs in different forms. Consequently, the contamination of Cu and Cd and the potential risk in Xinxue River, Jiehe River, and Guangfu River as well as the local emissions should be given more attention to safeguard the water quality of Nansi Lake and the East Route Project of South to North Water Transfer.

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Introduction

Heavy metals play an important part in our life as most of them widely exist in the nature by being used and discharged in many industries (Rodriguez et al. 2009) or thrown away from electronic products in domestic waste (Hicks et al. 2005; Liu et al. 2006). Generally, heavy metals, from weathering of rocks and soils in natural geological evolution, have relatively low concentration in unpolluted environments (Liu et al. 2009; Zhao and Yan 1994). As in human activities and industrial development, heavy metals are increasingly accumulated in our environment, such as drainage of domestic wastewater or emissions of industrial process (Boller 1997). Because of their toxicity, abundance, and persistence (Giller et al. 1998; Duruibe et al. 2007; Li and Zhang 2012), the increasing heavy metals are threatening human health (Molina 2013; Zhao et al. 2012).

In the past 50 years, many researches have been conducted to reveal the influences of the sediment contamination to natural world and living things (Duruibe et al. 2007; Dauvin 2008). Sediments, accumulated over time on bottom of waters, not only act as the sources and the sinks of pollution but also the potential secondary sources of pollution to the overlying water column in aquatic environment (Harbison 1986; Palanques et al. 1995). Heavy metals would give rise to serious environment problems and damage human health through the food chain (Khan et al. 2008; O'Sullivan et al. 2012); thus, the contamination of heavy metals has attracted much attention in the management of river pollution (Varol 2011; Singare et al. 2012; Sunjog et al. 2012). For the research methods of heavy metals in sediments are improved gradually, many indices, such as geo-accumulation index (I_{geo}) (Muller 1969)

and potential ecological risk index (RI) (Hakanson 1980), have been widely applied to evaluate the risk of heavy metals in river sediments.

Nansi Lake is one of the biggest regulating and storing lakes in East Route Project of South to North Water Transfer. The water quality of Nansi Lake is directly related to the water safety of this transfer project. The risk assessment and polluted condition of heavy metals in sediments have a significant impact on whether the transferred water can be used as drinking water (Harguinteguy et al. 2013). So far, studies on the heavy metals in rivers flowing into Nansi Lake are few or just limited on one river (Li and Zhang 2012). Therefore, this study is very meaningful to understand the condition of heavy metal contamination on tributaries of Nansi Lake by and large.

In order to assess the potential risk of heavy metals in rivers of Nansi Lake and understand the sources of heavy metal contamination in river sediments, a total of 36 sediment samples of six rivers flowing into Nansi Lake were obtained to determine the concentrations of chromium (Cr), copper (Cu), nickel (Ni), zinc (Zn), arsenic (As), cadmium (Cd), and lead (Pb). The aims of this study are (1) to assess the polluted levels of sediments by analyzing the geo-accumulation index (I_{geo}) of the seven metals, (2) to evaluate the ecological risk of the six rivers by accounting the potential risk index (RI), and (3) to identify the main sources of heavy metal contamination by correlation and cluster analysis of I_{geo}.

Materials and methods

Study area and sample collection

Nansi Lake, which is located in Jining, Shandong Province, is an influx of 53 tributaries and generally flows from north toward south. Six rivers flowing into eastern of Nansi Lake were chosen as sampling stations since the eastern part of Nansi Lake has been more serious polluted from anthropogenic activities than the western part (Wu and Zhang 2011). The six rivers are Guangfu River, Jiehe River, Chengguo River, Xinxue River, Xuesha River, and Jiangji River from north to south (Fig. 1). Guangfu River is the longest (48 km) with the most drainage area (1367 km^2) and Jiangji River is the shortest (5 km) with the least drainage area (54 km^2) among the six rivers (Zhang 2011).

For heavy metals mostly accumulated in surface sediments (Yadav et al. 2002), every six samples were collected from surface sediments (0–10 cm depth) of Guangfu River (116° 37′ 15.84″ N–116° 37′ 33.18″ N, 35° 18′ 52.38″ E–35° 16′ 43.14″ E), Jiehe River (116° 50′ 53.28″ N–116° 51′ 51.78″ N, 35° 6′ 10.26″ E–35° 7′ 31.86″ E), Chengguo River (116° 57′ 14.40″ N–116° 57′ 59.04″ N, 34° 55′ 36.96″ E–34° 56′ 40.80″ E), Xinxue River (117° 8′ 33.78″ N–117° 9′ 1.83″ N, 34° 44′

2.40″ E–34° 45′ 44.82″ E), Xuesha River (117° 11′ 18.00″ N– 117° 11′ 27.24″ N, 34° 43′ 23.64″ E–34° 45′ 2.52″ E), and Jiangji River (117° 11′ 37.92″ N–117° 12′ 16.80″ N, 34° 42′ 51.42″ E–34° 43′ 55.56″ E), respectively, in April 2013. Three samples were collected on either side along each river from the river mouth of Nansi Lake, and the spacing of each two samples at one side of rivers was 1 km. All locations were identified by global positioning system and detailed sampling sites were shown in Fig. 1.

All samples were collected by a hand shovel covered by rubber to prevent the content of heavy metals in samples from being influenced by the sampling kits (Li et al. 2007a, b). And the samples were quickly transferred into sealed bags for lab analysis.

Sample analysis

Prior to elements analysis, sediment moisture content was measured by drying certain samples in oven at 105 °C until constant weight. Sediment samples were ground and then passed through a 0.149-mm nylon sieve after air-dried, and finally, 36 samples of 0.3000 g were weighed for the experimental operation, which include digestion in the PTFE crucibles with a mixture of 10 ml HNO$_3$, 5 ml HF, and 2 ml HClO$_4$ (Song and Sun 2014). The final residues were dissolved in 1 ml HNO$_3$ and diluted to volumetric flask of 25 ml.

The concentrations of Cr, Cu, Ni, and Zn were determined by inductively coupled plasma atomic emission spectrometry (ICP-AES; THERMO, IRIS Advantage). Element of As was measured by atomic fluorescence spectrometry (AFS; Beijing Titan Instruments, AFS-933). Sediment samples were analyzed for Cd and Pb by using graphite furnace atomic absorption spectrometry (GFAAS; American PE Company, AAnalyst 800). All glassware used were cleaned by soaking in 10 % dilute acid for at least 24 h and rinsed abundantly in deionized water before use.

Geo-accumulation index (I_{geo})

According to the previous reports, the geo-accumulation index I_{geo} was widely used in the assessment of heavy metal contamination in soils, sediments, and urban road dusts (Wei and Yang 2010; Li et al. 2013a, b). I_{geo} value can be interpreted as follows:

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

where C_n represents the determined concentration of heavy metal n, and B_n is the background value of the element n in local sediments. The average background values for the surface sediments of Yellow River were adopted for the background values in our study (Cr, 60 mg/kg; Cu, 13 mg/kg; Ni,

Fig. 1 The sampling sites in six rivers, and the spacing of each two samples at either side of the rivers was 1 km
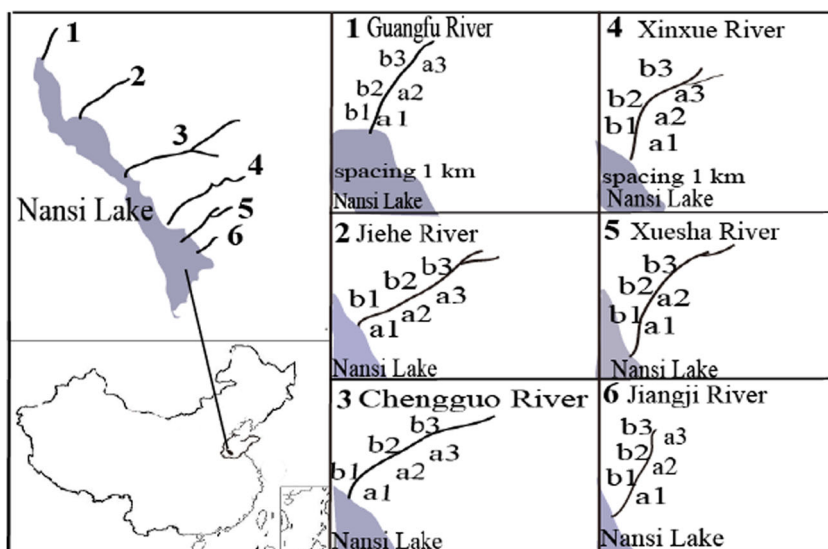
20 mg/kg; Zn, 40 mg/kg; As, 7.5 mg/kg; Pb, 15 mg/kg; Cd, 0.077 mg/kg; Zhao and Yan 1994). The constant value 1.5 was introduced to minimize the effect of possible variations in the background values, as recommended in the previous reports (Wang et al. 2013; Thuong et al. 2013). Evaluation classification and contamination level for I_{geo} were that $I_{geo} \leq 0$, unpolluted; $0 < I_{geo} \leq 1$, from unpolluted to moderately polluted; $1 < I_{geo} \leq 2$, moderately polluted; and $2 < I_{geo} \leq 3$, from moderately polluted to strongly polluted.

Potential ecological risk index (RI)

RI has been introduced by Hakanson (1980) to assess the sum of contamination factors in sediments. The equation for calculating the RI value was proposed, according to toxicity of heavy metals and response to the environment:

$$RI = \sum_{i=1}^{n} E_r^i = \sum_{i=1}^{n} T_r^i * C_f^i$$

$$C_f^i = \frac{c_n^i}{c_0^i}$$

where c_n^i is the concentration of any metal in samples, and c_0^i is the corresponding reference value of the metal. E_r^i is the monomial potential ecological risk factor, and T_r^i is the toxic-response factor (Cr, 2; Cu, 5; Ni, 5; Zn, 1; As, 10; Pb, 5; Cd, 30). In our study, c_0^i is the

Fig. 2 The distribution patterns of mean RI in six rivers (Duncan test). *The bars sharing the same letter* have no difference at *p<0.05*
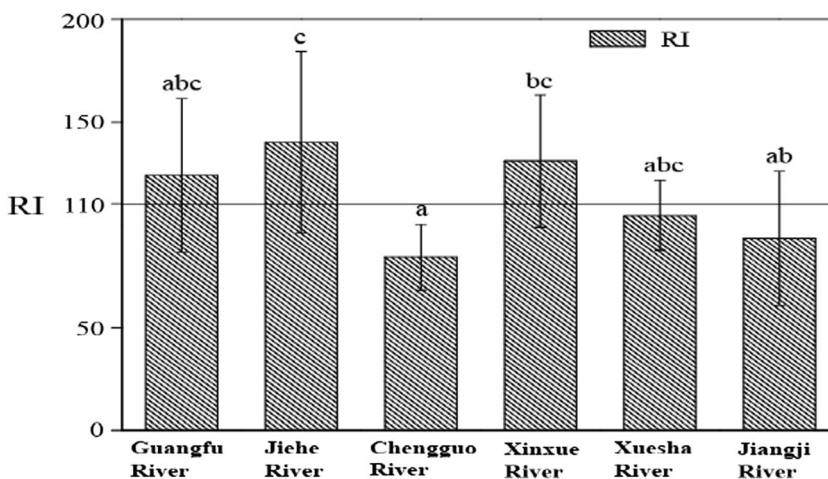
background value of Yellow River sediments, which is equal to B_n (Zhao and Yan 1994). RI is the risk factor that illustrates the potential ecological risk caused by certain contamination. Since the number of pollutants considered in our study was different from that of Hakanson (1980), the risk rank and evaluation classification were adjusted (RI<110: low risk; 110≤RI<220: medium risk; 220≤RI<440: high risk; RI≥440: very high risk).

Data analysis

All data were analyzed by using SPSS (version 21.0). ANOVA was used to determine the significant differences among the six rivers and analyze the differences in both sides of different rivers. Bivariate correlation was done in the relationship of seven heavy metals, and cluster analysis was also implemented in seven heavy metals. OriginPro (version 8.0)

Table 1 RI and I_{geo} of the six rivers in Nansi Lake

Rivers	Sites	RI	I_{geo} (Cr)	I_{geo} (Cu)	I_{geo} (Ni)	I_{geo} (Zn)	I_{geo} (As)	I_{geo} (Pb)	I_{geo} (Cd)
Guangfu River	a1	96.72	0.08	*1.28*	0.57	0.62	−0.08	0.36	−0.24
	a2	109.52	0.18	*1.11*	0.63	0.72	0.67	0.56	−0.14
	a3	*193.86*	−0.17	0.71	0.31	0.45	0.03	0.43	*1.66*
	b1	*138.27*	−0.15	0.86	0.21	0.84	−0.53	0.35	*1.01*
	b2	108.71	−0.05	0.86	0.27	0.49	0.45	0.15	0.21
	b3	96.81	−0.34	0.56	0.24	0.46	−0.37	0.21	0.21
Jiehe River	a1	*179.23*	0.23	*1.31*	0.66	0.83	0.33	0.31	*1.35*
	a2	*159.59*	−0.25	0.72	0.16	0.46	0.17	0.44	*1.25*
	a3	109.01	−0.33	0.65	0.12	0.19	0.08	0.41	0.39
	b1	*134.64*	−0.37	0.46	−0.06	−0.07	0.19	0.30	0.95
	b2	*186.57*	−0.05	0.87	0.38	0.31	0.50	0.77	*1.47*
	b3	71.60	−0.33	0.37	−0.19	−0.07	−0.46	0.32	−0.53
Chengguo River	a1	87.32	−0.28	0.81	0.04	0.20	−0.77	0.46	−0.07
	a2	103.47	−0.40	0.97	0.28	0.54	−0.96	0.39	0.36
	a3	77.97	−0.39	0.67	0.23	0.34	−1.23	−0.10	−0.18
	b1	74.41	−0.08	0.93	−0.06	0.35	−0.29	−0.05	−0.65
	b2	61.38	−0.40	0.29	−0.25	−0.39	−0.66	0.04	−0.83
	b3	100.38	−0.23	*1.00*	0.21	0.64	−0.80	0.02	0.30
Xinxue River	a1	*111.64*	−0.10	0.94	0.43	0.47	0.53	−0.03	0.24
	a2	*183.34*	−0.51	0.50	−0.23	0.04	*1.11*	−0.20	*1.46*
	a3	*154.40*	−0.11	*1.30*	0.18	0.96	0.32	0.21	*1.07*
	b1	106.30	−0.33	0.73	0.14	0.49	0.66	−0.10	0.16
	b2	101.56	−0.26	0.78	0.09	0.30	0.69	0.14	−0.03
	b3	*128.98*	−0.51	0.73	−0.74	0.52	0.13	−0.12	0.91
Xuesha River	a1	*115.37*	0.02	*1.06*	0.53	0.53	0.36	0.16	0.32
	a2	94.61	−0.28	0.68	0.26	0.17	−0.21	0.13	0.10
	a3	108.81	−0.79	0.20	0.27	−0.02	−0.73	−0.65	0.72
	b1	*130.70*	−0.53	0.45	0.65	0.28	−0.58	−0.15	0.97
	b2	84.93	−0.47	0.39	0.15	0.10	−0.27	−0.17	−0.03
	b3	93.04	−0.12	0.77	0.41	0.38	0.06	0.06	−0.10
Jiangji River	a1	*143.32*	−0.32	0.85	−0.09	0.32	−0.44	0.11	*1.14*
	a2	*123.59*	−0.10	0.94	0.65	0.61	−0.71	−0.17	0.76
	a3	66.51	−0.24	0.73	0.23	0.20	−0.39	−0.13	−0.99
	b1	73.77	0.03	*1.06*	0.60	0.60	−0.26	−0.37	−0.97
	b2	87.43	0.12	*1.41*	0.56	*1.06*	0.07	−0.11	−0.71
	b3	65.90	−0.46	0.42	0.13	−0.03	−0.53	−0.44	−0.68
Mean		*112.88*	0.23	0.79	0.22	0.39	−0.11	0.10	0.30

The medium polluted values were in italics

and Adobe Illustrator (version 16.0.0) were used to draw the graphs.

Results

Potential ecological risk index (RI)

The six rivers showed differences in the distribution patterns of RI. The RI of 14 sites (38.9 % of the total sampling sites) had exceeded 110 (Table 1). According to the risk ranked by RI, these 14 sites were at medium potential risk. As described in Fig. 2, the potential risk of the six rivers could be sorted as Jiehe River>Xinxue River>Guangfu River>Xuesha River>Jiangji River>Chengguo River. Chengguo River, Jiangji River, and Xuesha River had low potential risk, whereas Jiehe River, Xinxue River, and Guangfu River had medium potential risk (Fig. 2). The medium potential risk of three rivers (Jiehe River, Xuesha River, and Jiangji River) was mainly at the river mouths, whereas the medium potential risk at a3 site of Guangfu River (RI=193.86) was far from the river mouth.

Geo-accumulation index (I_{geo}) of heavy metals

To understand the influences of seven heavy metals to the six rivers, I_{geo} and its distribution patterns of seven heavy metals in the six rivers of Nansi Lake were calculated and showed in Table 1 and Fig. 3. In contrast to the evaluation classification of I_{geo}, I_{geo} of Cr showed that Cr in 30 sediment samples (83.8 %) were not polluted and that the other six sites (16.2 %) were under medium pollution. I_{geo} of Cu showed that Cu in 24 sampling sites (77.8 %) were under medium

pollution and that the other eight sites (22.2 %) were in medium pollution. I_{geo} of Cd showed that Cd in 15 sampling sites (41.7 %) were not polluted, 13 sites (36.1 %) were under medium pollution, and 8 sites (22.2 %) were in medium pollution, respectively. Cu, Ni, Zn, and Cd in the six rivers, Pb in Guangfu River and Jiehe River, and As in Jiehe River and Xinxue River were mostly under moderate pollution level (Table 1).

Correlation cluster analysis

Correlation analysis and cluster analysis (Table 2 and Fig. 4) were applied to explore the sources of seven metals in six rivers flowing into Nansi Lake in our study. Significant correlations and the same cluster existed among Cr, Cu, Ni, and Zn ($p<0.01$). Pb and As had significant correlations with Cr, respectively ($p<0.05$), whereas Cd was not significantly correlated with other metals.

Table 2 Spearman's correlations of heavy metals ($n=36$)

	Cr	Cu	Ni	Zn	As	Pb	Cd
Cr	1						
Cu	0.840**	1					
Ni	0.577**	0.525**	1				
Zn	0.675**	0.867**	0.549**	1			
As	0.373*	0.257	0.052	0.168	1		
Pb	0.364*	0.267	0.045	0.192	0.210	1	
Cd	−0.097	0.019	0.030	0.127	0.249	0.291	1

*Correlation is significant at 0.05 level (two-tailed); **correlation is significant at 0.01 level (two-tailed)

Fig. 3 The distribution patterns of I_{geo} of seven heavy metals in six rivers (Duncan test). *1* Guangfu River, *2* Jiehe River, *3* Chengguo River, *4* Xinxue River, *5* Xuesha River, *6* Jiangji River. *The bars sharing the same letter have no difference at $p<0.05$*
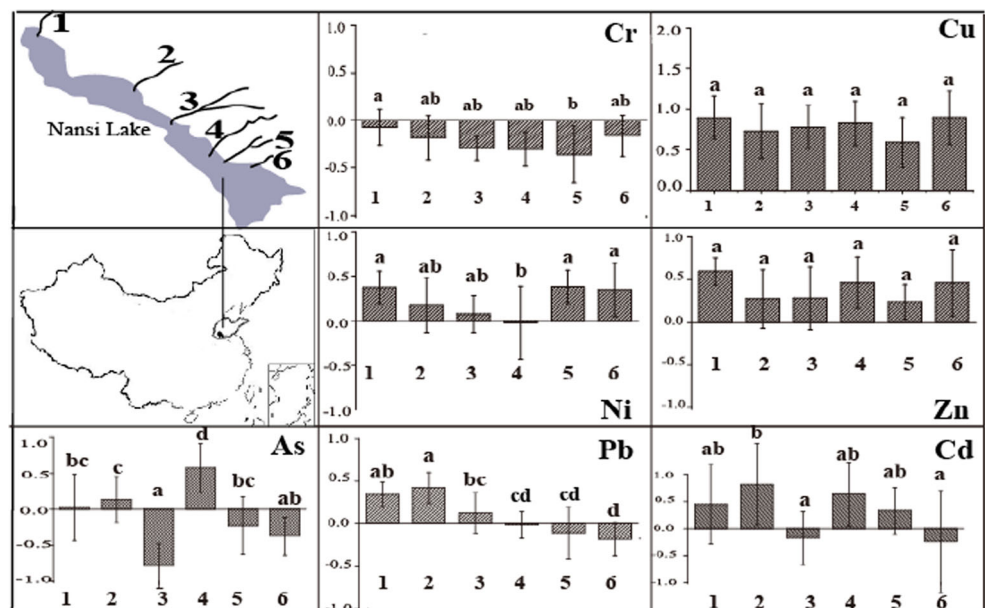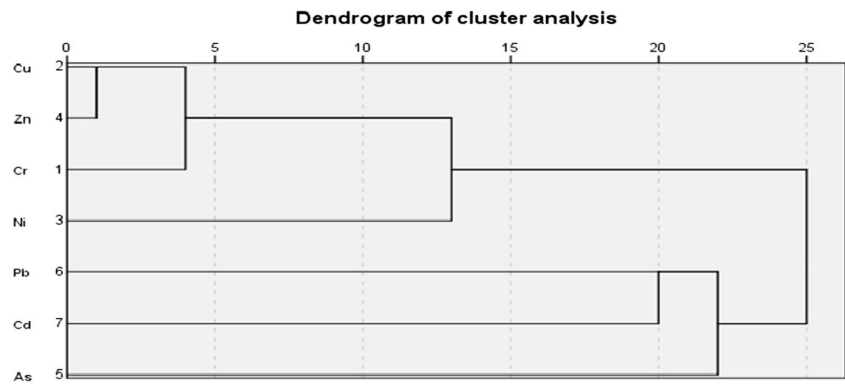
Fig. 4 The cluster analysis on I_{geo} of heavy metals (Cu, Zn, Cr, Ni, Pb, Cd, As) in six rivers

Discussion

Potential ecological risk index (RI) has been widely used to evaluate the ecological risk of sediments in rivers or lakes by considering various harmful elements (Liu et al. 2009; Huang et al. 2009; Yi et al. 2011). In our study, the relatively high risk at river mouths revealed that the accumulations of heavy metals in sediments were inclined to rise in the process of river waters flowing into the Nansi Lake smoothly. This finding agreed with the previous reports of Chen et al. (2007) and Yi et al. (2012), and it might help us detect and amend pollution effectively. However, the medium potential risk in a3 site of

Guangfu River mostly resulted from the excess contamination of Cd.

RI is one approach to understand the extent of river pollution in our study, while geo-accumulation index (I_{geo}) could be very helpful to analyze the influences of seven heavy metals to the six rivers and Nansi Lake, which is one of the biggest regulating and storing lakes in East Route Project of South to North Water Transfer. The contamination of Cr, Cu, Ni, and Zn was correlated, and the pollution level of Cu was the highest, followed by Zn ($p<0.01$), whereas that of Cd was the highest among Pb, As, and Cd. Comparing our research on heavy metal contamination to other relative reports of China

Table 3　The status of heavy metal contamination in sediments of many main rivers or lakes of China

Samples	Research sites	Heavy metals	Contaminated status	Relative references
1 (fishes)	Yangtze River	Cd, Cr, Cu, Hg, Pb, and Zn	*Cd* and Pb exceeded the permissible limits in several fishes	Yi et al. 2012
2 (sediments)	Yangtze River Delta, China	Fe, Mn, Zn, Cu, Pb, and Cd	*Cd*, Pb, and Cu were polluted severely	Lin et al. 2002
3 (sediments)	Pearl River Estuary	Zn, Ni, Cr, Cu, Pb, and Cd	*Cd* and Zn were polluted the most	Li et al. 2007a, b
4 (sediments)	Pearl River, Guangzhou	Cd, Cr, Cu, and Pb	*Cd* and Cu were polluted the most	Lv et al. 2009
5 (sediments)	Huaihe River	Cu, Pb, Zn, and Cr	Cu>Zn>Pb>Cr	Chen et al. 2006
6 (sediments)	Liaodong Bay (Liaohe River)	Zn, Cd, Cu, and Pb	*Cd*>Zn>Cu, Pb	Zhou et al. 2004
7 (sediments)	Songhua River	Cu, Pb, Zn, and Mn	Cu and Pb were polluted slightly	Zhang et al. 2010a, b
8 (sediments)	East China Sea	V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Mo, Sn, Cd, and Pb	The pollution of *Cd* and Pb was severe	Yuan et al. 2004
9 (sediments)	Quanzhou Bay	Cu, Zn, Pb, Cd, Cr, Ni, Hg, and As	*Cd* was polluted the most, Cu and Hg followed	Yu et al. 2008
10 (sediments)	Western Xiamen Bay	Cu, Pb, Zn, Cd, Cr, Ni, and Fe	Pb was polluted the most	Zhang et al. 2007
11 (sediments)	Dianchi Lake, Yunnan province	Cd, Cr, Cu, Pb, and Zn	*Cd*, Zn>Cu, Pb>Cr	Li et al. 2007a, b
12 (sediments)	Dongting Lake	Cr, Cu, Zn, Pb, Cd, As, and Hg	*Cd* exceeded the evaluated level	Li et al. 2013a, b
13 (sediments)	Hongze Lake	Cu, Zn, Pb, Cd, Cr, Hg, As, Fe, Al, and Mn	*Cd*>As>Cu>Cr>Zn>Hg>Pb	Yu et al. 2011
14 (sediments)	Hulun Lake	Cu, Zn, Pb, Cd, Cr, As, and Hg	*Cd* and As was polluted to certain extent	Zhang et al. 2010a, b
15 (sediments)	Tai Lake	Hg, Cd, Pb, As, Cu, Zn, and Cr	Hg>*Cd*>Pb>As>Cu>Zn>Cr	Xiang et al. 2006
16 (sediments)	Rivers flowing into Nansi Lake	Cu, Zn, Pb, Cd, Cr, Ni, and As	Cd and Cu were polluted severely, Zn and Ni followed	Our study

The severely polluted heavy metal was in italics

(Table 3), we found that most sediments had been suffering the similar heavy metal pollution: the contamination of Cd, Cu, Pb, and Zn was higher than other heavy metals, especially that of Cd, though those rivers or lakes differed in temperature, climate, or surroundings (Table 3). The similar trend might be associated with the development of economic and the industrial structure of economic development in China as China has been a country leading by secondary industry (Liang et al. 2009). So, we inferred that the anthropogenic discharge might be the main reason resulted in the contamination of heavy metals in sediments of China. Therefore, the key measure to fight against heavy metal pollution in sediments mostly rests with the change of industrial structure of economic development in China and the innovation of technology. The medium pollution of Cd in eight sites was probably due to the inappropriate use of fertilizers and pesticides because their distribution patterns in six rivers had no similarities. So, we could remedy it by ex situ extracting or separating method (Peng et al. 2009). For some widespread heavy metals, they could be remedied in situ remediation technology, such as sand cap (Theofanis et al. 2001) and phytoremediation (Suresh et al. 2004).

The sources tracing could help manage the risk of heavy metals. According to previous studies, correlation analysis and cluster analysis could profile the sources and affiliations of heavy metals (Tam and Wong 2000; Yang et al. 2009; Vukosav et al. 2014). In our study, the correlations and clusters of Cr, Cu, Ni, and Zn might suggest their homologous sources. Though the whole course of studied rivers had different surroundings, their local environment were similar at sampling stages, as many villages lay along those rivers and a lot of people there lived by aquiculture. As Cr was not polluted in most sampling sites, so we inferred that the contamination of Cu, Ni, and Zn was mainly caused by local emissions. As, Pb, and Cd were not significantly correlated with other metals ($p > 0.01$, Table 2), and they had various distribution patterns in the six rivers. So the contamination caused by As, Pb, and Cd might be of multisources and could be regarded as their external inputs in different forms. As previous reports showed, the main sources of As were mining and the oxidation of metals (Wang et al. 2006; Smedley et al. 2002), and that of Pb were coal burning and metallurgy activities (Komárek et al. 2008), while that of Cd were mining, smelting activities (Bi et al. 2009), fertilizers, and pesticides (Atafar et al. 2010). So our results indicated that the contamination of As, Pb, and Cd in the studied rivers might come from the related industries. Though the anthropogenic pollution plays a leading role in heavy metal pollution (such as industrial emissions, agricultural pollution, and domestic sewage, Birch et al. 1999; Pekey 2006), the specific sources of the seven heavy metals in the six rivers still need further investigation.

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