

Original Research

Distribution Characteristics, Pollution Assessment, and Source Identification of Heavy Metals in Sediments of Wetland Lakes

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Abstract

The aims of current study are to assess the levels of heavy metals (Cr, Cu, Ni, Fe, Zn, Co, Pb, and Cd) in sediments of lakes in Yangtze wetland and discuss the relationships between the sources and human activities. A total of 54 samples covering seven lakes along the Yangtze route in Anqing were selected. The concentrations of metals in lake sediments were ($\text{mg}\cdot\text{kg}^{-1}$): Cr, 4.08~12.58; Cu, 22.40~74.36; Ni, 29.89~142.17; Fe, 22899.20~50956.40; Zn, 102.31~242.04; Co, 8.35~26.89; Pb, 23.38~88.77; and Cd, 0.29~2.95. The situation of Xizi Lake was the most serious among the seven lakes investigated. Geoaccumulation index (I_{geo}) analysis showed that overall risk of heavy metals in sediments was approximately in the order: $\text{Cd} > \text{Pb} = \text{Zn} = \text{Cu} = \text{Ni} > \text{Fe} > \text{Co} = \text{Cr}$. The results of principal component analysis (PCA) suggested the main sources of Cd, Cu, Zn, Ni, and Pb on PC 1 were fuel combustion, metal smelting, industrial manufacturing, and other human activities, while Fe on PC 2 originated from rock weathering and other geochemical processes.

Keywords: heavy metals, Yangtze wetland, sediments, pollution assessment, principal component analysis

Introduction

Heavy metals are toxic pollutants with recalcitrance, bio-poisoness, biomagnification, and bioconcentration. They have directly or indirectly subjected human health to increased stress, and given sudden or persistent risks to ecosystems [1-3]. Heavy metals have been added to aquatic systems from natural or anthropogenic sources. They are released to water and generally are bound to particular matter, which eventually settles down and becomes incorporated into sediments [4-5]. The content

of heavy metals in the sediments has usually been considered as one of the important characterizations of water environmental quality. Therefore, the study on the pollution characteristics of heavy metals in the sediments of one region has great significance on aquatic resources protection, human health, and regional economic development.

There is a typical humid climate and dense population in the middle-lower area of China's Yangtze River, as well as developed industry. The lakes in this region receive massive surface runoff, domestic sewage, and effluent discharged from industry. On the other hand, these lakes are also important aquaculture waters while pollution levels of heavy metals in their sediments directly relates

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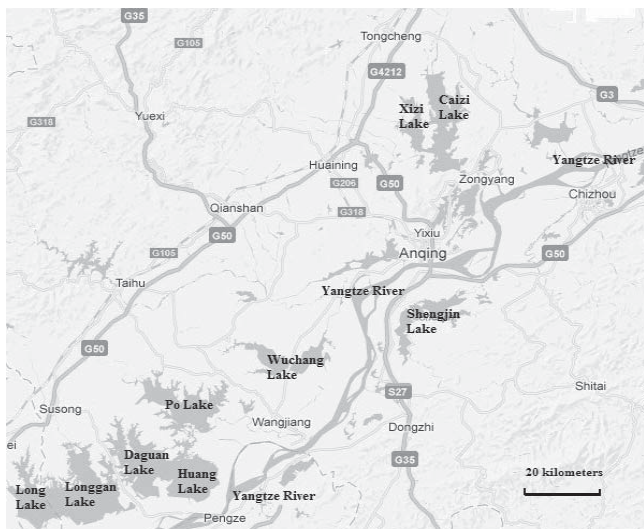


Fig. 1. Map of site in current study around Yangtze wetland.

to the quality and safety of aquatic products. Jian et al. found that the average contents of Cu, Zn, and Pb in the sediments of Poyang Lake were significantly higher than those of the local soil background values and the national average [6]. Zhu et al. concluded that the severity of pollution was determined as: South Dongting Lake > East Dongting Lake > West Dongting Lake > Datong Lake > Chenglingji after the system research of heavy metals in the sediments of Dongting Lake [7]. Moreover, the heavy metals contamination of South Lake was slight [8]. The study by Liu et al. indicated that the pollution by Pb and Zn in Ink Lake increased promptly with rapid industrialization and urban growth [9].

However, there is little information on the heavy metals contamination of the lakes in the Yangtze wetlands in Anqing city of Anhui Province. Herein, the levels and distribution characteristics of eight heavy metals in freshly deposited sediments of several lakes of the middle-lower Yangtze wetland were evaluated, as well as the ecological risk.

Principal component analysis (PCA) has been used to estimate the source of heavy metals contamination by many researchers [10-12]. PCA was a multivariate statistical method to pick out less representative variables in many original correlation indicators by dimension reduction and linear transformation. In this study, PCA was used to explain the composition characteristics of heavy metals in sediments of Xizi Lake and explore the internal relationships among the sources.

Materials and Methods

Sampling

Samples were collected from September to November 2011 in seven lakes (Daguang, Huang, Wuchang, Shengjin, Xizi, Po, and Gangyao) along the Yangtze route in Anqing (Fig. 1). Eleven samples of freshly deposited

upper sediment layers were collected from Daguang, seven samples from South Huang, eight from Wuchang, five from Shengjin, six from Xizi, 13 from Po, and four from Gangyao. Two parallels were set up for each sample. The sediment samples were collected using a homemade bamboo-bumpless gravitational sampler and sealed in pre-cleaned polyethylene (PE) bags and then stored at 4°C. GPS was used to locate when collecting.

Chemical Analyses

After weathering, the samples were crushed, ground, and sieved. The test methods of indicators are shown in Table 1. Each sample was duplicated, and the average was reported.

Table 1. Test methods of heavy metals.

Heavy metal	Test method	Standard
Cr	Flame atomic absorption spectrophotometry (FAAS)	HJ 491-2009 [32]
Cd, Pb	Graphite furnace atomic absorption spectrophotometry (GFAAS)	GB/T 17141-1997 [33]
Ni, Co	FAAS	GB/T 17139-1997 [34]
Cu, Zn	FAAS	GB/T 17138-1997 [35]
Fe	Diethylenetriamine pentaacetic acid (DTPA) extraction method	NY/T 890-2004 [36]

Table 2. Index of geoaccumulation and series of degree.

Program	I_{geo}						
	≤ 0	(0, 1)	[1, 2)	[2, 3)	[3, 4)	[4, 5)	≥ 5
Grade	0	1	2	3	4	5	6
Pollution	Unpolluted	Mild	Mild to moderate	Moderate	Moderate to strong	Strong	Very strong

Risk Assessment Methods

Geoaccumulation Index (I_{geo}) analysis

Geoaccumulation index (I_{geo}) was introduced by Muller [13] in order to determine and define metal contamination in sediments by comparing current concentrations with pre-industrial levels. The geoaccumulation index (I_{geo}) is defined by the following equation:

$$I_{geo} = \log 2[C_n / (K \cdot B_n)]$$

... where C_n is the measured concentration of the examined metal (n) in the sediment and B_n is the geochemical background concentration of the metal (n). K is the background matrix correction factor due to lithogenic effects, which generally is 1.5 [14]. The geoaccumulation index is divided into seven grades that show how pollution levels are enhanced. In this study, element background values in soil of Anhui Province [15, 16] were referred to as geochemical background (B_n): Cr = 66 mg/kg, Cu = 20.0 mg/kg, Ni = 29.8 mg/kg, Zn = 62.0 mg/kg, Co = 16.3 mg/kg, Pb = 26.0 mg/kg, Cd = 0.33 mg/kg, and Fe=28600.00 mg/kg. The corresponding relationship between geoaccumulation index and pollution degree are shown in Table 2.

Potential Ecological Risk Index (RI) Analysis

The method was proposed in 1980 by Hakanson [17]. Toxicity and pollution sensitivity of heavy metals and the difference of regional background values were considered to give the quantitative classification of potential ecological risk of heavy metal elements. Potential ecological risk index is one of the most widely used methods in heavy metal evaluation at home and abroad [18, 19]. The Potential ecological risk index (RI) is defined by the following equation:

Table 3. Potential ecological risk factor and index of sediment heavy metals and corresponding pollution grades of potential ecological risk.

E_r^i	<40	[40, 80)	[80, 160)	[160, 320)	≥ 320
RI	<150	[150, 300)	[300, 600)	≥ 600	-
Grade	1	2	3	4	5
	Low	Moderate	High	Higher	The highest

$$RI = \sum_{i=1}^n E_r^i = \sum_{i=1}^n T_r^i \times C_f^i = \sum_{i=1}^n T_r^i \times \frac{C^i}{C_r^i}$$

... where E_r^i is the potential ecological risk index of the examined metal (i), T_r^i is the toxic response coefficient of the metal (i), C_f^i is the pollution index of a certain metal (i) (enrichment factor), C^i is the measured concentration of the metal (i) in the sediment, and C_r^i is the geochemical background concentration of the metal (i) – which is the same as the previous method. The toxic response coefficients of the metals were determined as follows: Cr = 2, Cu = 5, Ni = 5, Zn = 1, Co = 5, Pb = 5, and Cd = 30 [19-21]. The standards of the potential ecological risks of metals in sediments are shown in Table 3.

Statistical Analyses

One-Way AVOVA was applied to analyze the significant differences among sampling stations for different metal levels. T-test was also performed to identify the significant correlation of the metals. Pearson correlation matrix and principal component analysis were both calculated for different metals in sediments to trace the common sources of pollutants. SPSS statistical package (Windows 17.0) was used for data analyses while Origin 8.0 was used to draw.

Results and Discussion

Concentrations of Metals in Sediments

Concentrations of metals in seven lake sediments are given in Fig. 2. Cu, Ni, Fe, Zn, Pb, and Cd levels were relatively higher than the element background values in soil of Anhui Province, and the multiples were 2.5, 2.8, 1.4, 2.8, 2.1, and 4.7, respectively. Especially the concentration of Cd was most prominent, indicating that Cd was the most serious pollutant. The difference among sampling locations was statistically significant (ANOVA) for levels of metals (Fig. 2). The highest average concentrations of Cr, Cu, Fe, Zn, and Co appeared in Xizi Lake, as well as Ni, Pb, and Cd, respectively, and the maximum appeared in Po, Shengjin, and Wuchang lakes. The concentrations of the measured metals except Cr exceeded the standard of Chinese first-class soil (GB15618-1995 [22]). The situation of Xizi was the most serious in the average concentration's view.

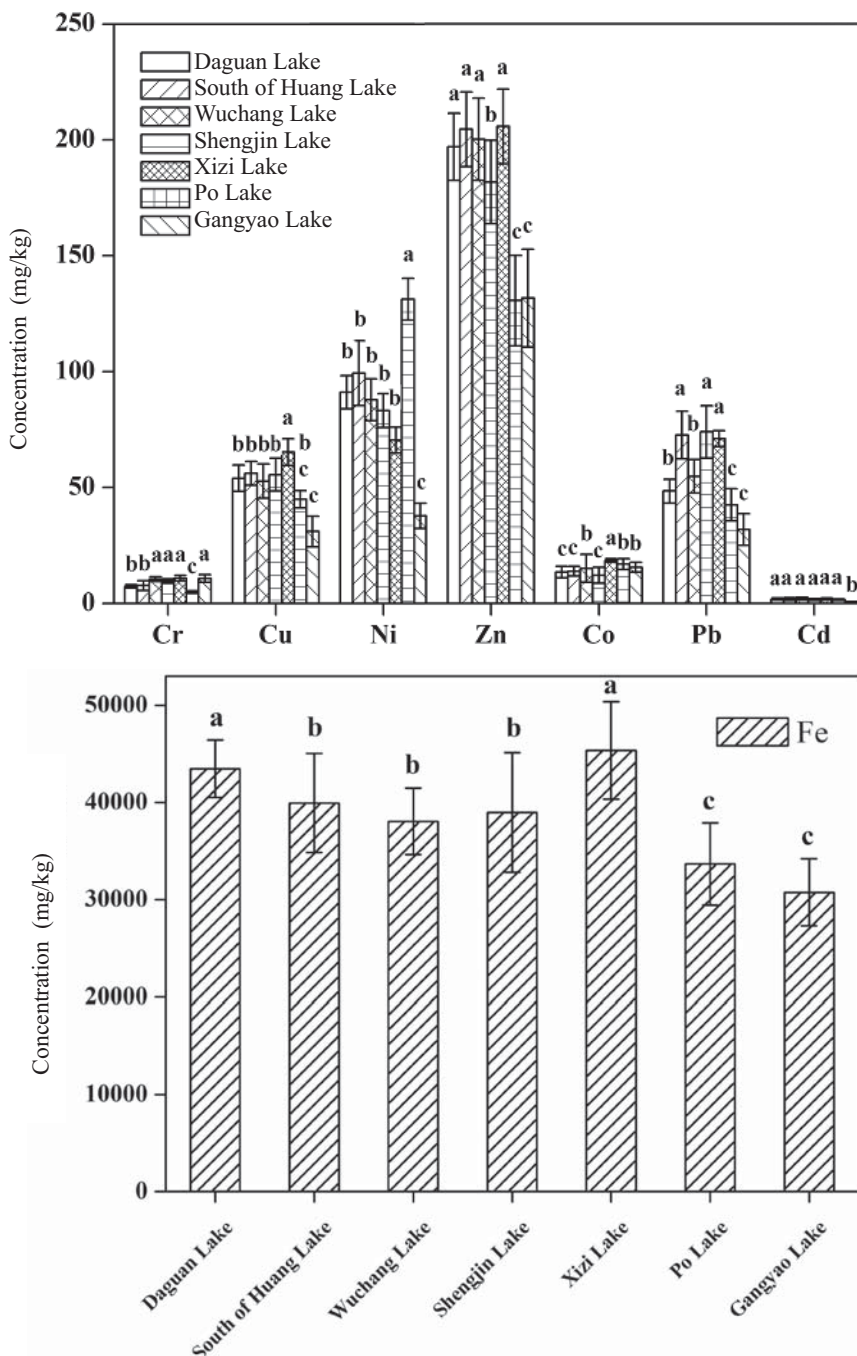


Fig. 2. Concentrations of heavy metals in sediments of five lakes. a, b, and c, mean value followed by different lowercase letters is statistically different (ANOVA; t-test, p<0.05).

There is a close relationship among the four interlocking lakes around the Yangtze: Huang, Dagan, Long, and Longgan. The water of Dagan Lake flows in the direction of Longgan and Huang lakes due to its higher position. In addition, Xizi and Caizi also are interlinked. The industrial emissions were considered to be the main source of heavy metals in sediments. The pollution statuses of the lakes were in connection with their geographical locations. Concentrations of seven heavy metals in the Huang sediments (except Fe and Cd) were higher than in Dagan, which may

be because of the difference in altitude between the two connected lakes, with Huang being lower. Similarly, some heavy metals of Dagan ran to Longgan due to Dagan's higher position. Qingcao and Wuchang are interlinked, and the water of Wuchang Lake was accepted by Qingcao Lake, resulting in Wuchang's light pollution. Similarly, the waters of Baitu and Caizi led to serious pollution of heavy metals in Xizi. However, Gangyao suffered from the slightest pollution due to its geographical position, which included a lack of urbanization and industrialization.

Table 4. Concentrations of heavy metals in sediments of other lakes.

Lake	Heavy metal (mg/kg)							
	Cr	Cu	Ni	Fe	Zn	Co	Pb	Cd
Chao Lake [37]	37.48*	17.29	–	39855	114.79	–	15.13	0.18
Dongting Lake [10]	100.97	69.38	–	–	113.73	–	35.78	15.20
Poyang Lake [38]	28.05	61.53	–	–	194.11	–	48.17	1.540
Moshui Lake [39]	213.00	93.10	57.30	–	1264	–	101.50	1.12
Tai Lake [24]	–	21.45	28.30	–	74.52	–	–	–

* Mean.

Table 5. The interval indexes of geo-accumulation and rankings of heavy metals pollution in sediments of seven lakes.

Lake	$I_{geo}/$ grade	Cr	Cu	Ni	Fe	Zn	Co	Pb	Cd
Daguan Lake	I_{geo}	[-4.07,-3.57]	[0.65,1.18]	[0.85,1.22]	[-0.20,0.16]	[0.84,1.21]	[-1.31,-0.49]	[0.09,0.57]	[1.14,2.36]
	grade*	0	2	2	1	2	0	1	3
South of Huang Lake	I_{geo}	[-4.23,-3.21]	[0.72,1.11]	[0.72,1.37]	[-0.32,0.16]	[1.04,1.33]	[-1.00,-0.44]	[0.41,1.08]	[0.89,2.53]
	grade	0	2	2	1	2	0	2	3
Wuchang Lake	I_{geo}	[-3.38,-2.99]	[0.56,1.14]	[0.73,1.21]	[-0.40,-0.03]	[1.00,1.38]	[-1.29,0.14]	[0.31,0.72]	[1.10,2.57]
	grade	0	2	2	0	2	1	1	3
Shengjin Lake	I_{geo}	[-3.52,-3.20]	[0.60,1.06]	[0.78,1.08]	[-0.46,0.11]	[0.81,1.18]	[-1.55,-0.64]	[0.62,1.19]	[1.25,2.19]
	grade	0	2	2	1	2	0	2	3
Xizi Lake	I_{geo}	[-3.37,-3.00]	[0.96,1.31]	[0.48,0.79]	[-0.15,0.25]	[0.92,1.24]	[-0.52,-0.36]	[0.74,0.94]	[1.24,2.20]
	grade	0	2	1	1	2	0	1	3
Po Lake	I_{geo}	[-4.60,-4.07]	[0.42,0.76]	[1.43,1.67]	[-0.91,-0.11]	[0.17,0.76]	[-0.80,-0.19]	[-0.27,0.33]	[1.18,1.91]
	grade	0	1	2	0	1	0	1	2
Gangyao Lake	I_{geo}	[-3.52,-3.04]	[-0.42,0.33]	[-0.58,-0.10]	[-0.72,-0.33]	[0.14,0.68]	[-0.95,-0.48]	[-0.74,-0.01]	[-0.77,0.67]
	grade	0	1	0	0	1	0	0	1

* The results are identified according to the maximum membership degree law, the same in Table 6.

Compared with the contents of heavy metals in sediments of other lakes in the middle-lower Yangtze area, the contents of Cr in seven lakes were relatively low; however, the contents of Cu in seven lakes were higher than that of Chao and Tai lakes, and the contents of Ni exceeded Moshui and Tai. The contents of Fe were similar to those of Chao, as well as Zn and Pb. Concentrations of Cd were similar to other lakes except for East Dongtong (Table 4).

Environmental Risk Assessment of Heavy Metals

In order to characterize and quantify enrichment features and the pollution status of heavy metals in sediments of seven lakes, efforts were made to evaluate the risks of heavy metals.

Geoaccumulation Index (I_{geo}) Analysis

I_{geo} can be used as a reference to estimate the extent of heavy metal pollution in an aquatic system. I_{geo} values of different lakes are shown in Table 5. I_{geo} values in this study were less than zero for Cr and Co (except Wuchang Lake), less than one for Fe, less than two for Cu, Ni, Zn, and Pb, and less than three for Cd. Results thus suggested “unpolluted” with Cr and Co; “mild” pollution by Fe; “mild to moderate” pollution of Cu, Ni, Zn, and Pb; and “moderate” pollution by Cd. The pollution by heavy metals in seven lakes was similar to each other according to I_{geo} . Overall risk of heavy metals in sediments was approximately of the order: Cd > Pb = Zn = Cu = Ni > Fe > Co = Cr

Table 6. The interval indexes of potential ecological risk and rankings of heavy metals pollution in sediments of seven lakes.

Lake	E_r^i / grade	Cr	Cu	Ni	Zn	Co	Pb	Cd
Daguan Lake	E_r^i	[0.18,0.26]	[11.75,16.97]	[13.51,17.49]	[2.69,3.46]	[3.03,5.34]	[7.96,11.15]	[99.09,231.82]
	grade	1	1	1	1	1	1	4
South of Huang Lake	E_r^i	[0.16,0.33]	[12.31,16.13]	[12.40,19.25]	[3.08,3.75]	[3.75,5.52]	[9.93,15.83]	[83.64,260.00]
	grade	1	1	1	1	1	1	4
Wuchang Lake	E_r^i	[0.29,0.38]	[11.03,16.48]	[12.46,17.33]	[3.00,3.90]	[3.06,8.25]	[9.27,12.39]	[96.36,268.18]
	grade	1	1	1	1	1	1	4
Shengjin Lake	E_r^i	[0.26,0.33]	[11.36,15.57]	[12.82,15.82]	[2.62,3.39]	[2.56,4.82]	[11.50,17.07]	[107.27,205.45]
	grade	1	1	1	1	1	1	4
Xizi Lake	E_r^i	[0.29,0.38]	[14.54,18.59]	[10.45,12.87]	[2.84,3.53]	[5.22,5.86]	[12.54,14.37]	[106.36,206.36]
	grade	1	1	1	1	1	1	4
Po Lake	E_r^i	[0.12,0.18]	[10.02,12.73]	[20.22,23.85]	[1.69,2.54]	[4.31,6.58]	[6.26,9.45]	[101.82,169.09]
	grade	1	1	1	1	1	1	4
Gangyao Lake	E_r^i	[0.26,0.36]	[5.6,9.43]	[5.02,7.01]	[1.65,2.41]	[3.87,5.38]	[4.50,7.43]	[26.36,71.82]
	grade	1	1	1	1	1	1	2

Potential Ecological Risk Index (RI) Analysis

RI is used to reflect the pollution levels of heavy metals in sediments and potential ecological dangers. *RI* values and grades of seven heavy metals in sediments of seven lakes are listed in Table 6. All grades of heavy metals were one while the grade of Cd was over 5, which indicated that the potential ecological risk of Cd was the highest with the peak at 231.82. On the contrary, other metals exhibited relatively lower risk. The high potential ecological risk of Cd may be due to the excessive content and high toxic response factor [19]. Other heavy metals were at low ecological risk because of lower coefficients and less content.

Correlation Analysis

To determine the common source of metals, a correlation matrix was calculated for heavy metals in sediments. Table 7 showed significant correlation among Cr, Cu, Ni, Fe, Zn, Co, Pb, and Cd. Cr showed a relationship with Zn ($p < 0.05$), and Cu showed a relationship with Cd ($p < 0.05$), Fe, Pb, and Zn ($p < 0.01$). Similarly, a relationship was found between Fe and Zn ($p < 0.01$), Pb ($p < 0.05$), and Zn and Pb ($p < 0.01$) and Cd ($p < 0.05$). Alternatively, no correlations were noted among Ni, Co, and other metals. But in all relationships, Only Pb and Cu showed a close relationship, which suggested similar geochemical properties and a common source, or combined pollution of these two metals [14, 23]. Although there is some

Table 7. Correlation coefficients of heavy metals in sediments of seven lakes.

	Cr	Cu	Ni	Fe	Zn	Co	Pb	Cd
Cr	1							
Cu	0.24	1						
Ni	-0.79	-0.02	1					
Fe	0.27	0.62**	-0.18	1				
Zn	0.50*	0.65**	-0.34	0.60**	1			
Co	-0.06	0.09	0.14	-0.13	-0.17	1		
Pb	0.35	0.70**	-0.09	0.51*	0.59**	-0.10	1	
Cd	0.01	0.54*	0.17	0.36	0.52*	-0.16	0.32	1

* $P < 0.05$, ** $P < 0.01$.

Table 8. Correlation coefficients of heavy metals in sediments of Xizi Lake.

	Cr	Cu	Ni	Fe	Zn	Co	Pb	Cd
Cr	1							
Cu	0.40	1						
Ni	-0.11	0.58	1					
Fe	-0.40	0.11	0.39	1				
Zn	-0.32	0.45	0.87**	0.18	1			
Co	-0.39	-0.71	-0.16	-0.48	0.05	1		
Pb	-0.41	0.35	0.79**	0.10	0.76**	0.26	1	
Cd	0.33	0.64*	0.30	-0.54	0.53	-0.12	0.27	1

* $P < 0.05$, ** $P < 0.01$.

correlation among the other metals, the related coefficients were too low.

A correlation matrix was also calculated for heavy metals in sediments of Xizi due to the most serious pollution (Table 8). As shown in Table 8, there was a close relationship among Ni, Zn, and Pb ($p < 0.01$). Cu also appeared in a relationship with Ni and Cd ($p < 0.05$). No correlations were found among other metals. These results demonstrated that Ni, Zn, and Pb had a common source as well as Cu, Ni, and Cd. Or they were able to interact to cause common pollution.

Principal Component Analysis (PCA)

Fig. 3 exhibits eight detected metals that were divided into two main components. PC 1 and PC 2 accounted for 51.247% and 28.790% of the total variables, respectively. Two main components cumulatively contributed 80.037 percent, which were able to reflect the vast majority of all data. It could be seen from Fig. 3 that the load values of Pb, Zn, Ni, Cd, and Cu were high on PC 1 as well as low on

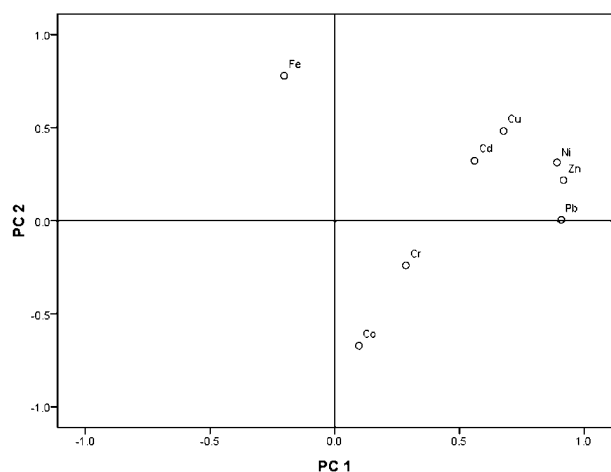


Fig. 3. Loading plots of the pollutants of Xizi Lake in the space defined by two components.

PC 2. This suggests that these metals were representative factors of PC 1. Similarly, Fe was a representative factor of PC 2 due to the high value on PC 2 and low on PC 1.

Two different sources of heavy metals were usually represented by PC 1 and PC 2 [24]. As discussed above, the concentrations of Cd, Cu, Zn, Co, and Pb in Xizi were higher than the others. And there were correlations among these elements. Xizi is located in the southeastern hinterland of Tongcheng with convenient transportation, superior position, dense population, and developed industry and commerce. There are a lot of packing, printing, machinery manufacturing, textile clothing, and other types of enterprises nearby and they put great pressure on Xizi, especially the developed electroplating industry. The sources of Pb were ore and gasoline combustion – effluents discharged by the lead industry [25] – and domestic sewage [26]. Cu was mainly due to the discharge of acidic wastewater by Taochong and Liantanhengshan copper mines nearby. On the other hand, waste incineration and industrial emissions were important sources of Cu and Zn [27]. Metal smelting was the major reason for Cd pollution. Additionally, alloy manufacturing and crude oil combustion would cause Cd and Ni [14, 28] pollution. In addition, the heavy metals in the eroded soil caused by land reclamation and excessive use might lead to pollution [29-30]. The five heavy metals on PC1 reflected the homoplastic sources. Therefore, PC 1 mainly reflected the contributions of fuel combustion, metal smelting, and manufacturing.

Concentrations of heavy metals in sediments of six lakes (except Gangyao) indicated that there was no obvious regional differentiation of distribution of Fe. The content was relatively close. Fe content in Xizi was a bit higher but not very different from the others. So we guessed that Fe was derived from rock weathering and other geochemical processes. Therefore, PC 2 reflected the metals pollution in sediments from natural processes [14].

Conclusions

1. The concentrations of Cu, Ni, Fe, Zn, Pb, and Cd in sediments of seven lakes in Yangtze wetland area were higher than the element background values in soil of Anhui Province. Seven metals contents exceeded the standard of Chinese first-class soil except for Cr. Cd caused the most serious pollution in seven lakes, while Xizi was the most polluted.
2. I_{geo} values of heavy metals in sediments of seven lakes indicated that the major pollutant in sediments was Cd, followed by Ni, Cu, Zn, and Pb. Co and Cr showed minimal ecological risk. Potential ecological risk index analysis illustrated that the risk of Cd was the highest and the others were in low danger. The two evaluation methods both confirmed that Cd was one of the relatively serious heavy metal pollutants in the Yangtze wetland sediments and even in Chinese lakes [31].
3. Correlation analyses showed that there was a close relationship between Pb and Cu in sediments of seven

lakes. The correlation coefficient was 0.70, which was the highest number, suggesting that the geochemical properties of the two metals were similar. No close relationship was noted among the other metals. In Xizi sediments, Ni showed a relationship with Zn, Cu, and Pb. Also, there was a correlation between Zn and Pb as well as Cu and Cd. The results suggested that there were similar sources of these heavy metals in Xizi.

- The principal component analysis of heavy metals in Xizi showed that the main sources of Cd, Cu, Zn, Ni, and Pb on PC 1 were fuel combustion, metal smelting, industrial manufacturing, and other human activities. Fe on PC 2 originated in rock weathering and other geochemical processes.

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