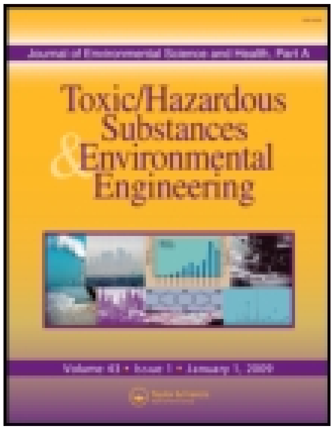

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Spatial and temporal variation of heavy metal risk and source in sediments of Dongting Lake wetland, mid-south China

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Surface sediments of Dongting Lake wetland were collected from ten sites to investigate variation trend, risk and sources of heavy metal distribution in dry seasons of 2011~2013. The three-year mean concentrations (mg/kg) of Cr, Cu, Pb, Cd, Hg and As were 91.33, 36.27, 54.82, 4.39, 0.19 and 25.67, respectively, which were all higher than the corresponding background values. Sediment quality guidelines (SQGs) and Geo-accumulation index (I_{geo}) were used for the assessment of pollution level of heavy metals. The pollution risk of Cd, Hg and As were great and that of Cr needed urgent attention because of its obvious increase. Pollution load index (*PLI*) and geographic information system (GIS) methods were conducted to assess spatial and temporal variation of heavy metal contamination. Results confirmed an increased contamination contribution inflow from Xiang River. Multivariate statistical analyses were applied to identify contribution sources of heavy metal, which showed anthropogenic origin mainly from mining, smelting, chemical industry and agricultural activity.

Keywords: Heavy metal, risk, sediment, spatial and temporal variation, source apportionment.

Introduction

In recent years, heavy metal contamination in the environment has been attracting a great deal of world-wide attention due to their toxicity, persistence, wide sources and non-biodegradable properties.[1-3] Their accumulation in the ecosystem and food chain will have a significant impact on human health.[4-7] Heavy metals have low solubility in water, and they usually get absorbed and accumulate in sediments.[8] In spite of their stability, heavy metals can transfer from sediment to aquatic environment with certain disturbances such as frequent resuspension of sediments by wind waves, trawling and large vessels.[9,10] Sediments can act as carriers and sinks for heavy metals.[11,12] The assessment of metal pollution status in the aquatic environment is primarily through the corresponding lake sediment analysis.[13]

Heavy metals enter the aquatic environment via several pathways of natural and anthropogenic sources, including erosion and weathering of the parent rock, industrial activities, agricultural activities, vehicle exhausts, sewage sludge, atmospheric precipitation and so on.[14, 15] Numerous studies show that the origins of heavy metals in the environment are primarily anthropogenic sources. Source apportionment of environmental pollutants is the foundation of pollution control.[16] Considerable studies have been focused on heavy metal contamination risk and source in sediments all over the world.[17-23] However, little information is available on investigating for years to analyze the variation trend of contamination risk and source of heavy metal in wetland sediments.

The variation of the contaminated area, contaminant and its origin can provide specific guidance for pollution control and remediation. Source apportionment of environmental pollutants is of great significance for pollution control. Dongting Lake is the second largest freshwater lake in China. However, research focusing on source apportionment of heavy metal in sediments of Dongting Lake wetland has been ignored. As rapid urbanization and industrialization appear around metropolitan areas in developing countries, ecosystem has been disturbed by anthropogenic activities easily.[24] Previous studies[25] mainly forced on landscape structure, habitat and human disturbance on birds, but information on studies carried out for ecological health risk of wetland heavy metal to

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migrants is scarce. In this article, the risk of heavy metal in sediments of Dongting Lake wetland to human health and migrants came up for discussion.

In this article, we investigated for the first time three-year variations of heavy metal contamination risk and source in sediments of Dongting Lake wetland. Multivariate statistical analysis and geographic information system (GIS) techniques were combined as a preliminary survey. Besides, the risk of heavy metal in sediments of Dongting Lake wetland to human health and migrant came up for discussion. Therefore, the present study was carried out to meet the following objectives: (1) to analyze pollution status of six heavy metals (Cr, Cu, Pb, Cd, Hg and As) in sediments by sediment quality guidelines (SQGs) and geo-accumulation index (I_{geo}); (2) to assess spatial and temporal variation of heavy metal contamination using both pollution load index (*PLI*) and GIS; (3) to identify their origins combining Pearson's correlation with factor analysis (FA); and (4) to qualitatively predict risk to human health and migrant.

Materials and methods

Study area

Dongting Lake, covering an area of 2820 km^2, is in the middle and lower reaches of Yangtze River. Due to reclaiming land from lakes since the 1960s, Dongting Lake has shrunk from the first largest freshwater lake to the second. With its horseshoe of northern exposure, Dongting Lake is located in northern Hunan province at latitudes 28°30′~29°38′N and longitudes 112°18′~113°15′E (Fig. 1). It is characterized by subtropical monsoon climate, the annual precipitation is about 1100–1400 mm, mainly between April and June. The mean depth is 6–7 m, about 18 days in the hydrology cycle. Wet season lasts from May to October, and dry season runs from November to March.

The Three Gorges Dam impounds during wet season and then supplies water for Dongting Lake during dry season. Dongting Lake has a special hydrological characteristic that lake will appear when the water level rises during wet season while wetland soil will appear when water level falls during dry season. Such emerged soil and plant in dry seasons have a significant ecological value for waterfowl. Dongting Lake, which is regarded as "Nonferrous Metal Village" with rich mineral resources, is the main receiving water of the river system in Hunan province. There are primarily four inflowing rivers including Yuan River, Xiang River, Zi River and Li River but only one outlet.[26] Thus, the sediment deposition is in large quantity with 70% sediment been trapped. The mean annual sediment depth is 0.018 m.[27] The major anthropogenic activities in this region are agriculture, tourism around the lake and large petrochemical enterprises.

Sampling

In dry season of 2011~2013, surface sediments were collected from ten sites of Dongting Lake wetland (Fig. 1). The sampling sites were determined by hand-held Global Positioning System (GPS) position indicator. Five samples of the top 5cm surface sediments were collected from each site, refrigerated with a polyethylene bags for measuring. Although all sampling sites were located in the margins, they would be in the lake when in wet season because of the special hydrological characteristic of Dongting Lake. Therefore, samples could represent the whole lake.

Analytical methods and quality control

Sediments were dried in an oven until constant weight, ground gently, sieved with 200 mesh sieve for homogenization. Precise 0.1000g preprocessed samples were weighted and transferred to airtight Teflon vessels, added by HNO_3 and $HClO_4$ for digestion. The pretreatment procedure and detection method for Cr, Cu, Pb, Cd and As by inductively coupled plasma-mass spectrometry (ICP-MAS) follows the Chinese DZ/T0223-2001. Besides, 0.3000g preprocessed samples were accurately weighted, using mixed acid system for digestion, and then added the stannous chloride solution. Atomic fluorescence spectrometry (AFS) was used to analyze Hg.

Quality assurance and quality control were assessed using duplicates, method blanks, and national first level standard materials (GBW07345, Qingdao Institute of Marine Geology) for sediments. Considering the determination accuracy and precision of the samples, repeat analysis and standard sample analysis were conducted with repetitive rate of 10%. The analysis results were reliable when repeat sample analysis error was less than 5%. The results met the quality control requirement of The Technical Specification for Soil Environmental Monitoring (HJ/T 166-2004).

Sediment quality guidelines (SQGs)

SQGs have been developed for freshwater ecosystems. These synthesized guidelines consist of a threshold effect level (TEL) below which harmful effects on sediment-dwelling organisms were not expected and a probable effect level (PEL) above which harmful effects were expected to occur frequently.[28] They are widely used to screen sediment contamination by comparing sediment contaminant concentration with the corresponding quality guideline.[29, 30]

Geo-accumulation index (I_{geo})

I_{geo} index was introduced by Müller (1969).[31] It is a geochemical criterion to evaluate heavy metal pollution in

Fig. 1. Study area and sampling sites of surface sediments along Dongting Lake wetland.

sediments. The I_{geo} enables the assessment of contamination by comparing current and preindustrial concentrations. The I_{geo} for sediments was calculated by the equation:

$$I_{geo} = log_2(C_n/1.5B_n) \quad (1)$$

where C_n is the measured concentration of element (n), mg/kg and B_n is the geochemical background value of element (n), mg/kg. In the equation, average values are used and 1.5 is the factor used for lithologic variations of trace metals. According to Hunan Environmental Quality Report (2011), background values of heavy metal contents in sediments of Dongting Lake were used as B_n for calculation. The level classification of I_{geo} is defined as: unpolluted (<0), unpolluted to moderately polluted (0–1), moderately polluted (1–2), moderately to strongly polluted (2–3), strongly polluted (>3).

Pollution load index (PLI)

Contamination factor (*CF*) is the ratio of measured concentration by background value. For the entire sampling site, *PLI* has been determined as the *n*th root of the product of the *n CF*:

$$CF = C_n/B_n \quad (2)$$

$$PLI = (CF_1 \times CF_2 \times CF_3 \times \ldots \times CF_n)^{1/n} \quad (3)$$

This empirical index provides a simple, comparative means for assessing the level of heavy metal pollution. When $PLI > 1$, it means that pollution exists; otherwise, if $PLI < 1$, there is no metal pollution.[32]

Multivariate statistical and GIS-based analyses

In this study, relationships among the variables considered were tested using Pearson's correlation with statistical significance at $P < 0.05$ and $P < 0.01$. FA was applied on standardized data. FA was performed using principal components analysis (PCA) extraction method and VARIMAX rotation. PCA was performed to identify the sources of elements in terms of the correlation among elements in sediment samples. All the statistical analyses were performed using SPSS 18.0 for Windows.

The inverse distance weighted (IDW) interpolation method with weighting power of 2.0 was used to elucidate spatial variations of heavy metals based on ArcGIS 10.0 software. IDW assumes that the predictions are a linear combination of available data, and greater weighting values are assigned to values closer to the interpolated point. The statistical results of IDW interpolation showed a good agreement between measured line and predicted line with the acceptable mean error (<0.05) and Root-Mean-Square (<0.05).

Results and discussion

Sediment contamination characteristics

The mean contents of Cr, Cu, Pb, Cd, Hg and As in three years were presented in Table 1, along with background values in surface sediments of Dongting Lake wetland and guide values of freshwater sediment quality of China. The

Table 1. Summary statistics of heavy metal concentrations in sediments from Dongting Lake wetland, compared with guide values of China and SQGs (mg/kg, dry weight).

	Cr	*Cu*	*Pb*	*Cd*	*Hg*	*As*
2011						
Min	34.90	19.00	34.90	0.05	0.04	10.33
Max	101.00	58.00	120.90	11.00	0.34	56.76
Mean	75.20	34.12	58.49	2.55	0.17	26.76
S.D. [a]	19.46	11.26	25.26	3.29	0.11	16.36
CV [b] (%)	25.88	33.01	43.19	128.71	64.84	61.15
2012						
Min	35.00	21.00	21.00	0.60	0.04	12.80
Max	78.60	70.90	89.00	19.40	0.58	83.70
Mean	67.06	38.22	57.56	3.85	0.20	23.46
S.D. [a]	13.40	14.16	28.73	5.60	0.15	21.33
CV [b] (%)	19.98	37.05	49.91	145.34	75.26	90.94
2013						
Min	79.80	17.90	16.90	0.20	0.08	10.40
Max	199.00	54.80	95.80	20.70	0.64	66.30
Mean	131.74	36.46	48.41	6.75	0.22	26.80
S.D. [a]	38.28	12.56	27.12	6.86	0.16	19.43
CV [b] (%)	29.06	34.46	56.01	101.55	75.11	72.49
Mean of 3 years	91.33	36.27	54.82	4.39	0.19	25.67
Background [c]	83.92	25.00	27.75	0.23	0.07	13.41
TEL	37.3	35.7	35	0.60	0.17	5.9
PEL	90	197	91.3	3.53	0.49	17

[a]S.D.: standard deviation.
[b]CV: coefficient of variation.
[c]Background: According to the Environment Quality Report of Hunan Province (2011).

concentrations of all elements were higher than their corresponding background values, especially Cd and Hg. Cd and Cr in 2013 increased obviously with approximately twice as high as that of 2012. The mean concentrations of all elements exceeded TEL and Cd, As and Cr were higher than PEL.

The results indicated that Cd, As and Cr were likely to result in harmful effects on sediment-dwelling organisms which were expected to occur frequently. In addition, Hg presented relatively elevated concentrations at S2, which were closed to the estuary of Yuan River and consistent with previous research.[33] Yuan River spans Guizhou province where the resource and production of Hg are the first in Asia and the third in the world.

The pollution level of Cd and Hg has generally increased during the three years studied (Fig. 2). Cd was in high pollution status, followed by Hg. As was in a moderate level. Cu presented no pollution risk with all negative values of I_{geo}. Cr showed a low value of I_{geo} with its high background value and increasing concentration. According to I_{geo}, average pollution level decreased following the order Cd > Hg > Pb > As > Cu > Cr. Compared with the SQGs approach, there were similar assessment results for Cd. The results were consistent with previous studies.[33–35] In general, the pollution risk of Cd, Hg and As were great and that of Cr needed urgent attention because of its obvious increase. According to coefficient of variation, the distribution of Cd in sediments among the sites was relatively more uneven, followed by Hg and As. Cr showed a small spatial differentiation.

A comparison of heavy metal contents in sediments of Dongting Lake wetland with other largest four freshwater

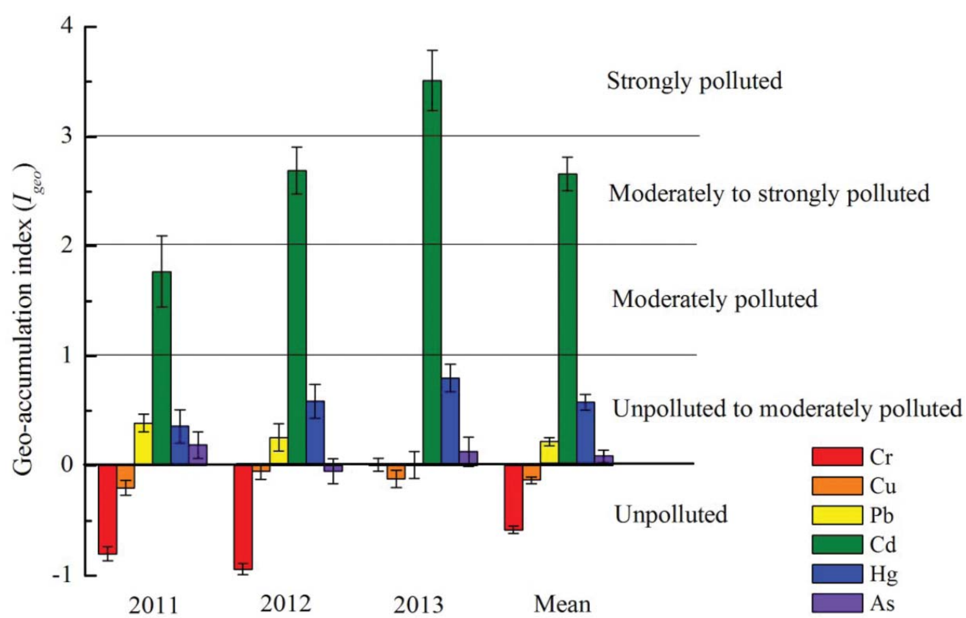

Fig. 2. Geo-accumulation index (I_{geo}) of heavy metals in sediments from Dongting Lake wetland each year.

lakes in China was shown in Table 2.[7,36–39] It revealed that the mean contents of Cr, Pb, Cd, Hg and As in Dongting Lake wetland were all higher than those in other four lakes, only Cu content was lower than Taihu Lake. Therefore, more attention is urgently needed on pollution management and control of Dongting Lake wetland.

Table 2. Comparison of heavy metal contents in sediments of Dongting Lake with previous studies and other largest four freshwater lakes in China (mg/kg).

	Cr	*Cu*	*Pb*	*Cd*	*Hg*	*As*	*Reference*
Dongting Lake	74.75	52.38	127.05	10.86	—	175.4	[35]
Dongting Lake	88.29	47.48	60.99	4.65	0.157	29.71	[33]
Dongting Lake	91.33	36.27	54.82	4.39	0.19	25.67	This study
Chaohu Lake	80.1	26.2	49.8	0.43	0.18	8.02	[36,37]
Poyang Lake	70.77	27.71	48.67	0.56	0.08	10.67	[38]
Taihu Lake	56.2	36.7	51.8	0.94	0.11	13.5	[7]
Hongze Lake	57.59	34.99	18.82	3.24	0.07	23.67	[39]

Spatial risk assessment of heavy metal

Spatial risk maps of heavy metal in Dongting Lake were shown in Figures 3a–c. In 2011, pollution risk mainly existed at S9, the lower reach of Dongting Lake. It might be affected by effluent from industries and chemical enterprises of Yueyang City. Yueyang City has rich mineral resources with more than 200 spots. Famous enterprises include Changling Refining & Chemical Company, Baling Petrochemical Crop, Huaneng power plant and Tiger Forest & Paper Group. In addition, Yueyang Harbor is the largest harbor in China's inland regions and one of the world's top 50 harbors. Hydraulic regime, deposition turbulence and inputs from the ships (cargo, paints and oil spill) can affect the accumulation of pollutants.[40] S4 and S5 presented a moderate risk.

Although in 2012, S5 was the most serious contamination area, which was located at the estuary of Xiang River. Xiang River is one of the most serious heavy metal contaminated rivers in China. Many industries for mining and smelting of metals are located along Xiang River, such as Valin Xiangtan Iron and Steel Co., Ltd, Zhuzhou Smelter Group and Shuikoushan Mining Bureau.[31,41] In 2013, S5 and S6 presented a high pollution level, with S9 and S10 in moderate level. Variation of special risk revealed that the contamination of Dongting Lake contributed by Xiang River became more and more obvious.

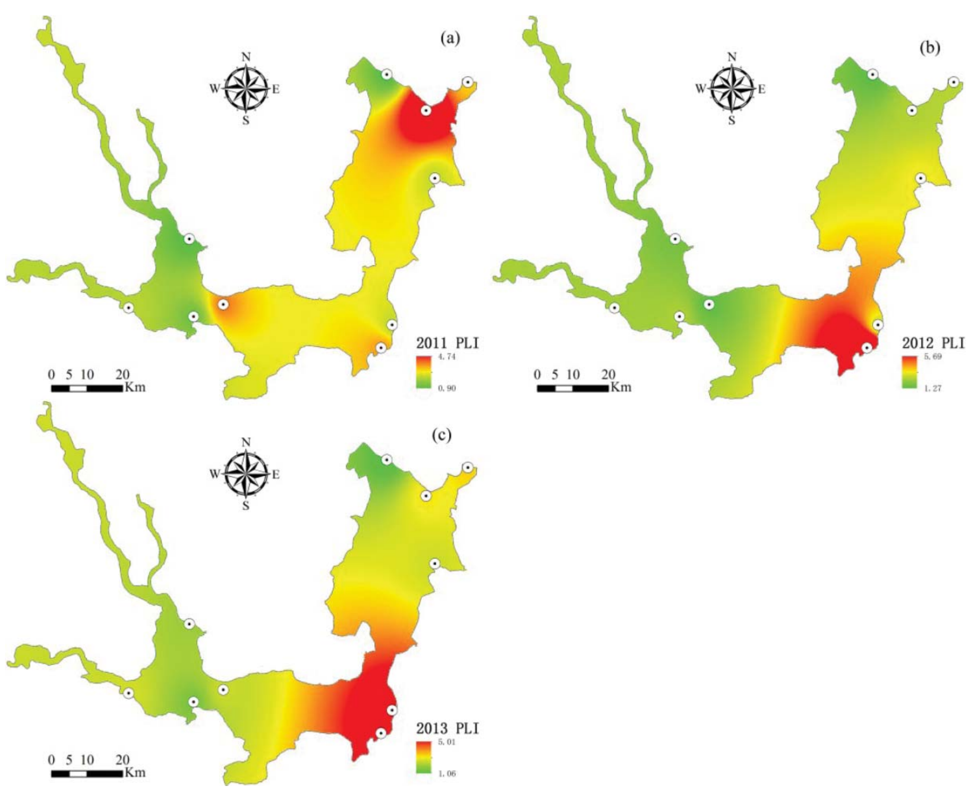

Fig. 3. Spatial distribution maps using pollution load index (*PLI*) for heavy metals in sediments of Dongting Lake wetland: (a) for 2011, (b) for 2012 and (c) for 2013.

S8 was at a relatively low level of pollution risk during the studied three years, which was located in core area of East Dongting Lake National Nature Reserve. Hunan province had transformed the ecological environment and carried out closed-off management in this area since 2006. Measures for eco-environmental modification were mainly as follows: creating different water level gradient such as grass land and mudflats for birds habitat in dry season, enriching species diversity and restoring damaged wetlands ecosystem. To 2012, closed-off management had been implemented all over the core area.

Intermetal relationships and source apportionment of heavy metal contamination

Correlation analysis of heavy metals was shown in Table 3. Loadings of the principal components (PCs) for three years were shown in Figures 4a–4c. Three PCs were extracted in each year explaining 94.6%, 97.3% and 88.0% of the cumulative variance, respectively.

For 2011, Cd had a strong association with other elements except for Hg, indicating that Cd had extensive sources. In addition, there were significant correlations between Cu and Cr as well as Cu and Pb. It implied that they might have a common source. PC1, explaining 41.3% of the total variance, had strong loadings on As, Pb, Cd. PC1 could be agricultural nonpoint source pollution and effluent from mining. Statistics showed that about 1.7 million tons of pesticides and fertilizers were used in Dongting Lake basin each year. Pesticides contain Cr, Cu, Hg, As and fertilizers also contain Cd, Cr, Cu, Pb, etc.

Moreover, only a small percentage of applied fertilizer can be absorbed by the crops.[15,42] It can cause contamination through agricultural runoff. PC2, explaining 29.3% of the total variance, had strong loading on Cr. Possible sources could be industrial point sources pollution from Yueyang City referring to PLI in 2011, effluent from chromate production, electroplating industries, tanning, mining and rubber raw material. PC3, which explained 24.0% of the total variance, was dominated by Hg. It might be burning of fossil fuels.

For 2012, Cd showed strong correlations with As and Hg. As showed significant correlations with Cu and Hg. PC1 (52.5% of the total variance) was dominated mainly by As, Hg, Cd. Possible sources could be agricultural runoff and mining. PC2 (23.7% of the total variance) was dominated mainly by Cr, and the origin could be from industrial point source pollution. PC3 (21.1% of the total variance) was dominated mainly by Pb, which was primarily brought by Xiang River. As there were many chemical industries and smelting plants along Xiang River, PC3 might be mining and burning of fossil fuels.

For 2013, As and Pb showed a strong correlated relationship. Cd showed no obvious relativity with other elements. PC1, PC2 and PC3 accounted for 32.7% with strong loading of Hg, 32.5% with strong loading of Cr and 22.8% with strong loading of Cd, respectively. Possible sources of Hg and Cr was similar to those mentioned for 2011. Cd might have comprehensive sources such as pesticides, fertilizers, mining, smelting and chemical industry.

Table 3. Pearson correlation matrix for heavy metal concentration in sediments from Dongting Lake wetland.

	Cr	*Cu*	*Pb*	*Cd*	*Hg*	*As*
2011						
Cr	1					
Cu	0.808 [a]	1				
Pb	0.502	0.819 [a]	1			
Cd	0.416	0.820 [a]	0.855 [a]	1		
Hg	0.386	0.682 [b]	0.470	0.756 [b]	1	
As	0.255	0.609	0.724 [b]	0.790 [a]	0.470	1
2012						
Cr	1					
Cu	0.696 [b]	1				
Pb	0.363	0.561	1			
Cd	0.185	0.751 [b]	0.326	1		
Hg	0.258	0.634 [b]	0.058	0.900 [a]	1	
As	0.235	0.817 [a]	0.428	0.963 [a]	0.834 [a]	1
2013						
Cr	1					
Cu	0.525	1				
Pb	0.617	0.731 [b]	1			
Cd	0.254	0.462	0.171	1		
Hg	0.335	0.363	0.529	−0.074	1	
As	0.650 [b]	0.699 [b]	0.935 [a]	0.208	0.642 [b]	1

[a]Correlation is significant at the 0.01 level (2-tailed).
[b]Correlation is significant at the 0.05 level (2-tailed).

Qualitative prediction of risk to human health and migrant

With change of the condition, heavy metal in sediments can be released into the water environment, especially in bioavailable forms. One function of Dongting Lake is farm irrigation. Some heavy metals such as Cd and Pb can accumulate in large amount in crops, some animals and fish can also accumulate heavy metals in tissues.[24,43] As Dongting Lake is an important commodity grain and fishing base in China, its products can enter the food chain and then threaten human health. In addition, the density of population around Dongting Lake is relatively higher compared to the average of Hunan province, so the heavy metal contamination has a higher potential risk to human health of residents in the region.[44]

Dongting Lake wetland, including South Dongting Lake Wetland and Waterfowl National Nature Reserve, West Dongting Lake Provincial Nature Reserve and East Dongting Lake National Nature Reserve, has great significance for Asian-Pacific bird inhabit, migration and wintering. Because of the special hydrological characteristic of

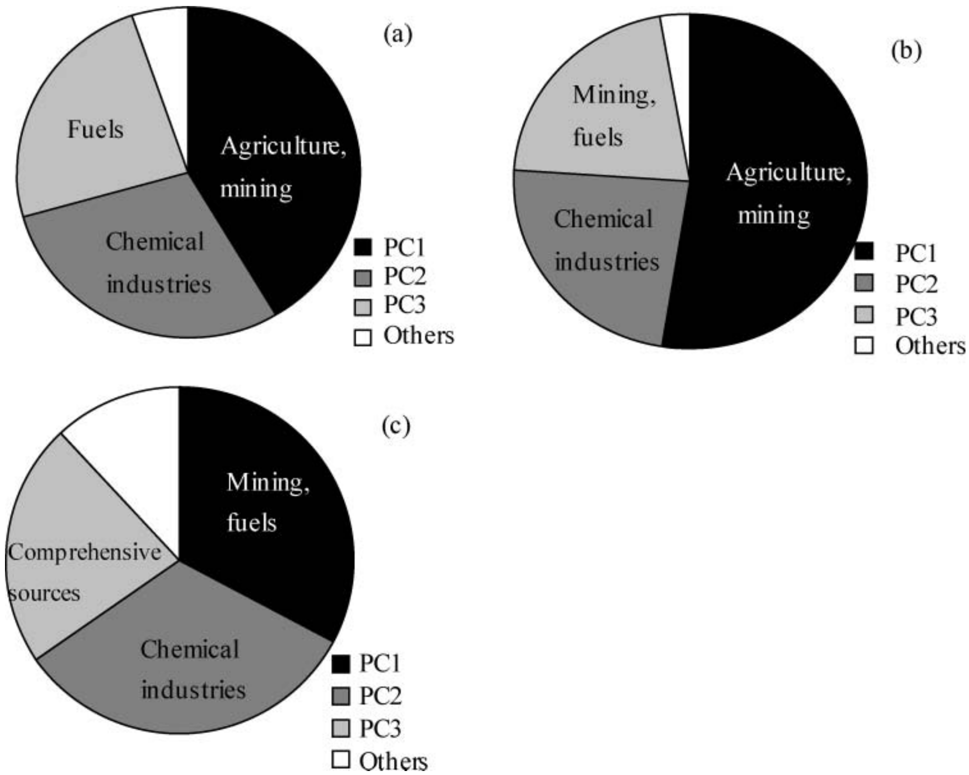

Fig. 4. Loadings of the PCs for heavy metals: (a) for 2011, (b) for 2012 and (c) for 2013.

Dongting Lake mentioned before, wetland soil will appear when the water level falls during dry season. Such place with high water content plays significant ecological roles. However, with heavy metal contamination, exposed plants enriched with heavy metal can provide food for waterfowl. When birds migrate from Dongting Lake to another place, they may cause pollution in the place through many ways such as droppings of migrants.

Conclusion

Three-year longitudinal assessment and analyses were conducted in the present study in sediments of Dongting Lake wetland during dry seasons. The investigations carried out revealed a pollution condition with all six heavy metals in different degrees. Cd and Hg were in high potential risk with considerable loadings and increased contents. It could be due to the increase of burning of fossil fuels, agricultural runoff, mining and smelting. Obvious increase of Cr concentration might be caused by effluent from chromate production, electroplating industries, mining, tanning and rubber raw materials. It could be a potential threat to agriculture, fishery, livestock and human health. Additionally, As was also at high risk according to SQGs. Contamination and spatial distribution analyses indicated an increased pollution contribution inflow from Xiang River.

The study suggested that agricultural non-point sources pollution existed during the studied period. With closed-off management and quit of industry point source pollution from Dongting Lake, heavy metal contamination mainly came from Xiang River, Yueyang City and Yuan River. Besides, heavy metal contamination of Dongting Lake wetland can bring risk to human health and migrants. The risk to wildlife needs quantitative analysis in future studies. The results have great significance in designing effective developing strategies to prevent and control further heavy metal pollution of the ecosystem in Dongting Lake wetland. Although the three-year variation of element concentration has been studied innovatively, there was a restrictive factor without research and analysis of sedimentation rates in the lake.

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