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Heavy metals in water, soils and plants in riparian wetlands in the Pearl River Estuary, South China

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Abstract

Samples collected from sites of 26 for water, soils and two native plants (Scirpus tripueter Linn. and Cyperus malaccensis Lam.) in riparian wetlands were analyzed to investigate the distribution of heavy metals (Cd, Cr, Cu, Ni, Pb and Zn) in Panyu-Nansha area of Pearl River estuary. The results indicated that concentrations of heavy metals among three compartments were in the order: soils >plants >water and no obvious correlations were found between in soils and water, water and plants. Pb is the only metal accumulated in both plants that correlated with its concentrations in soils. The weak or lack correlations among metals in water, soils and plants suggest that other factors existed influence the metal uptake and storage in plants other than absorbing from soils and water. The plants had the same trend in metal accumulation that was Cd >Zn >Cu >Ni >Cr >Pb. The translocation factors showed that metals accumulation was mostly occurred in roots for these two plants. Compared to the other heavy metals, Cd seemed to be much more hazardous. Principal Component analysis and Cluster analysis were used to analyze the relevance of different metals and identify the major sources. The results showed two factors dominated the metals variability (83.4% of total variance) that Cd and Pb, were dominated by PC1 whereas Cr, Cu and Ni charged by another factors and Zn was affected by both two components. Analysis of CA for the sampling sites showed that among all of anthropogenic pollutions, industrial wastewater was major sources of heavy metals especially for Cd, Cr, Cu and Zn in the PRE.

Keywords: Riparian wetlands; Heavy metals; Pearl River estuary; Panyu-Nansha area

1. Introduction

Heavy metals are serious pollutants due to their toxicity, persistence in natural conditions and ability to be incorporated into food chains [1-4]. Wetlands, in particular riparian wetlands, generally are recognized to be important sinks as filters, retaining heavy metals that can have toxic effects on biota [5]. In riparian wetlands, heavy metals exist mainly in water, in soils and in plants or other organisms. For example, previous studies have reported that riparian wetland have a large capacity to accumulate heavy metals [6-12]. Arias et al. [9] investigated metal (Cd,

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Na, Mn, Pb, Cu, Fe and Zn) contamination levels in soils of riparian wetlands of the Conchos watershed in Chihuahua, Mexico and found that the quality of soils is above desirable levels. However, the metal distribution in these compartments can also transport among different compartments [13, 14]. Another reports have also found there are some connections between dissolved and particulate metals in river water such as Shulkin and Bogdanova [15] analyzed the extent of Zn, Cd, Pb and Cu release from the land-derived solids to estuarine and coastal water and found that Zn, Cd and Cu have considerable additional input of dissolved forms because of metals released from river suspended matters to water. However, the rapid development of industrialization, urbanization and agricultural practices has been threatening the ecological health of riparian wetlands [16]. In recent years, the contamination of aquatic systems has become a problem of great concern throughout the world.

Panyu-Nansha Area (PNA), located in south of Guangdong province, is the end of land area which the Pearl River flows through and into the South China Sea. The Pearl River is the third largest river in China. In the latest two decades, the rapid economic growth and urban development has led to excessive release of wastewater into the Pearl River and its tributaries [17]. With an important sediment load of 80×106 tons/a, the Pearl River, which contains relatively high metal concentrations among the Chinese rivers, produced a large amount of particulate metal transport to the South China Sea [7, 18]. Studies on contamination of heavy metals in the PRE have increased dramatically in last years [19-29]. However, few studies focused on the distribution of heavy metals in riparian wetlands and relationships among water, soils and plants in this region.

The objectives of this paper are to: (1) investigate the distribution and enrichment of heavy metals in water, soils and plants of riparian wetlands; (2) analyze the correlations of heavy metal distributions among these compartments.

2. Materials and methods

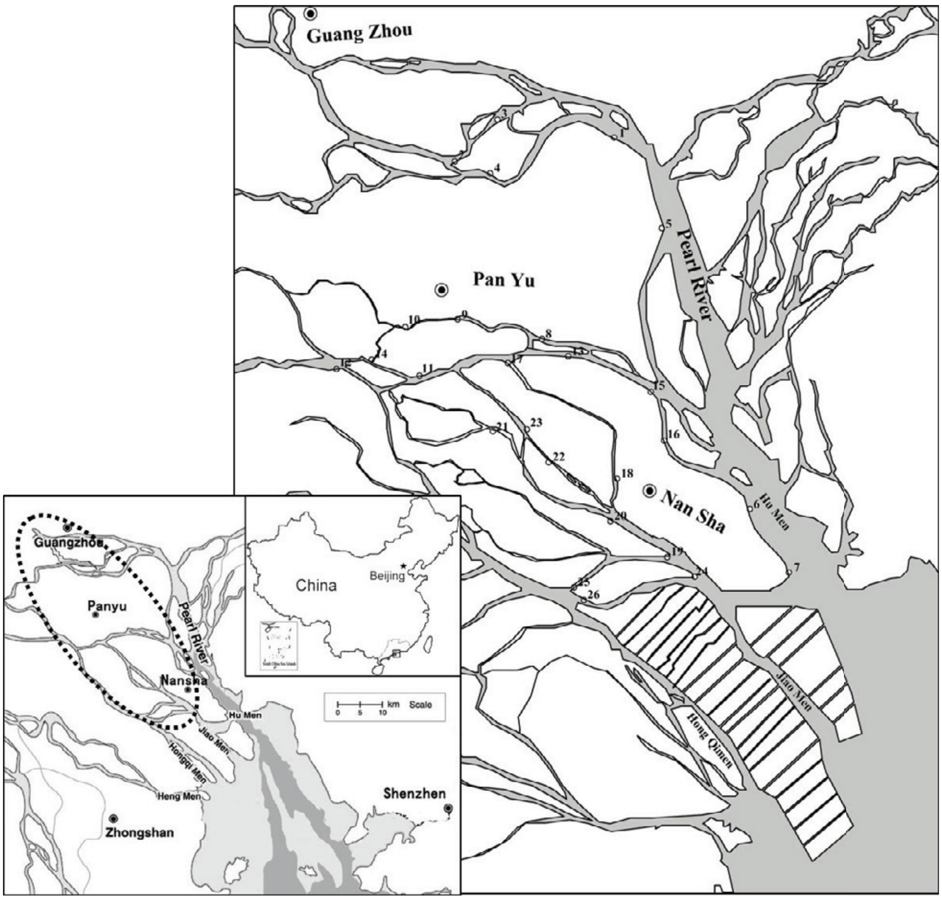

Fig. 1. Location of sampling sites in the riparian wetlands of PNA in the PRE

2.1. Sampling and sample preparation

The samples were collected in riparian wetlands during March and April 2009. The sampling sites were selected according to the spatial distribution of riparian wetlands in the PNA of PRE as shown in Fig. 1.

The vertical change of water quality in riparian wetlands is not very significant due to the shallow water with average depth of 0.5-1.5 m. Thus the water samples were collected at the depth of 0.5 m below the surface and stored in 500 ml polyethylene bottles precleaned with deionized water and rinsed with the sample to be collected from different sites. The water samples were filtered through 0.45μm Millipore filters and acidified to pH <2 using concentrated nitric acid and then stored in the dark at 4℃. The concentrations of heavy metals were measured by inductively coupled plasma-atomic emission spectrometry (ICP-AES).

The soil samples used in this study were collected at each site for 0 to 30 cm depth and the exact location was recorded with GPS. All of soil samples were air-dried at room temperature immediately after collection and sieved through a 2-mm nylon sieve to remove coarse debris. For each sample, 30 g sub-sample of soils was ground with an agate mortar until all particles passed a 100-mesh nylon sieve. A 0.1000 g sub-sample of dried and homogenized soil was accurately weighed and digested with 2 ml HNO_3, 1 ml $HClO_4$ and 5 ml HF at a temperature of 90±190 ℃ for 16 h. The residue was then dissolved in 2 ml of 4 mol/L HCl and diluted to 10 ml with deionized water and analyzed for a number of heavy metals (Cd, Cr, Cu, Ni, Pb and Zn) by inductively coupled plasma-atomic emission spectrometry (ICP-AES).

The two plants (*Scirpus tripueter Linn.* and *Cyperus malaccensis Lam.*) were sampled as representative in these riparian wetlands. For all samples, the roots, and the aboveground parts were separated and stored in the zipped polyethylene bags, respectively. The samples were washed thoroughly with tap water and rinsed with deionized water, then dried at 70℃ for 24 h in an oven. After the measurement of dry weights, the samples were ground into fine powder in an agate mortar. Precise weigh of each sample (0.2000 g) were used to prepare the solution of digestion reaction. A mixture of concentrated HNO_3 (2 ml) at 65% and H_2O_2 (1 ml) at 30% was used for digestion reaction. After cooling to the room temperature, the residue was diluted with deionized water to 10 ml and analyzed for metals by ICP-AES.

For quality control analysis, standard reference soil material (GBW07401, GSS-1) and plant material (GBW08513) from the National Research Center for Standards in China were used to verify the accuracy of metal analysis. The recovery rates of heavy metals in the standard reference material were around 91-97%. Reagent blanks were also employed to detect potential contamination during the digestion and analytical procedure.

2.2. Statistical analysis

In the study, statistical analysis of data was performed using Microsoft Office Excel and the computing package called Statistical Package for Social Science (SPSS 16.0 for Windows, SPSS Inc., IL, U.S.A.). Nonparametric test was used to determine any significant difference in metal contents among different sample sites at the level of $p \leq 0.05$. Correlations among metal concentrations in the water, soils and plants samples were evaluated using Pearson correlation coefficients. Both Principal Component Analysis (PCA) and Cluster Analysis (CA) were conducted to investigate associations of different sample sites and assess the major sources of metals in riparian wetlands.

3. Results

3.1. Concentrations of heavy metals in river water

Table 1 summarizes the total mean concentrations and standard deviation of Cd, Cr, Cu, Ni, Pb and Zn in river water from riparian wetlands of PNA. The mean concentrations of Cu, Zn and Cr were lower than the threshold of class I regulated in the Environmental Quality Standards for Surface Water in China (EQSSW) [30] which was set to protect the water quality in national nature reserves. The concentrations of Cd and Pb were little higher than the class I threshold. In comparison with other natural water, although the metal concentrations in our study are relatively high [31, 32], the concentrations are still at a safe level for aquatic life, except for Pb.

Table 1. Concentrations (mean ± standard deviation) of heavy metals in the water (mg/L).*

Wetlands	Cd	Cr	Cu	Ni	Pb	Zn
RWs	0.0029 ± 0.0014	0.0085 ± 0.0029	0.0016 ± 0.0006	0.0125 ± 0.0031	0.0128 ± 0.0111	0.0089 ± 0.0080
I Class	≤0.001	≤0.01	≤0.01	NS	≤0.01	≤0.05
WQC	0.005	1	0.01	0.1	0.1	0.1
Darwin Harbor	≤0.0001		0.0003	0.0002	≤0.0001	0.0001
Bing Bong Coast	≤0.0001		0.0003	0.0002	≤0.0001	0.0001
Nhue River		0.007	0.004	0.007	0.004	0.011

* RWs mean riparian wetlands in PNA; I Class means class I standard regulated in the Environmental Quality Standards for Surface Water in China; WQC means Water Quality Criteria for the protection of aquatic life in China; Darwin Harbor & Bing Bong Coast [31]; Nhue River [32].

3.2. Concentrations of heavy metals in soils

The concentrations of heavy metals in soils from different riparian wetlands are presented in Table 2. As shown in Table 1 and 2, the concentrations of heavy metals in soils were far higher than those in water and showed enrichment of metals in the riparian soils. The metal concentrations in soils were higher than the thresholds of Class I regulated in the Environmental Quality Standard for Soils in China (EQSS) [33] which was set to represent the natural background levels of soils. Whereas most of metal concentrations were lower than the class II threshold which was set for general agricultural and grazing land and the soil quality pose no harm to the plants and environment. However, the amounts of Cd were significant higher than the values of class II and obvious contaminated in soils.

Table 2. Concentrations (mean ± standard deviation) of heavy metals in the soils (mg/kg).*

Wetlands	Cd	Cr	Cu	Ni	Pb	Zn
RWs	2.38 ± 1.87	109.7 ± 25.73	65.36 ± 31.26	50.56 ± 16.64	79.27 ± 23.88	244.42 ± 83.1
EQSS I Class	≤0.2	≤90	≤35	≤40	≤35	≤100
EQSS II Class	≤0.3	≤200	≤100	≤50	≤300	≤250
Guangzhou	7.2	23.8	49.3	42.1	167.7	267.7
Mudflat	4.22	79.9	68.2	60.4	32.2	311.1

* RWs mean riparian wetlands in PNA; EQSS means the Environmental Quality Standards for Soils in China; Guangzhou [25]; Mudflat in PRE [22].

3.3. Concentrations of heavy metals in plants

In our survey, the two plants Scirpus tripueter and Cyperus malaccensis are the majority communities of emergent aquatic plants in riparian wetlands in the PRE. The metal concentrations in the plants from riparian wetlands are listed in Fig. 2. The metal concentrations of Cu, Ni and Zn in Cyperus malaccensis were higher than those in Scirpus tripueter at most of sample sites. For two plants, Zn was the mostly accumulated in the plants, followed by Cu, Cr, Ni and Pb and the least was Cd.

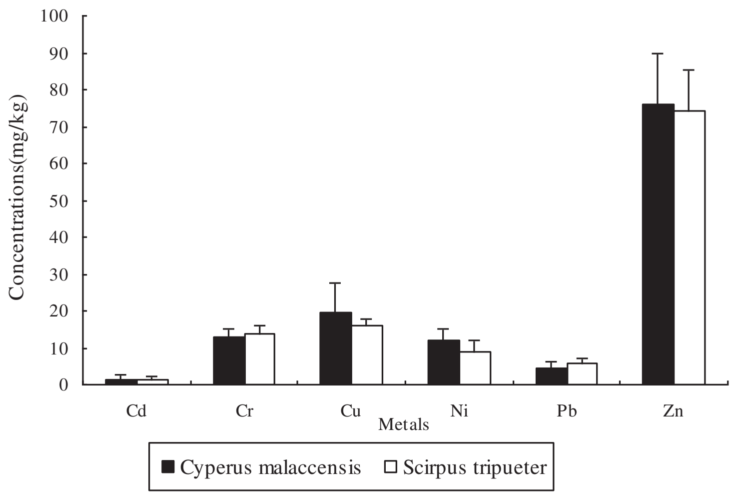

Fig. 2. Concentrations (mean ± standard deviation) of heavy metals in the plants.

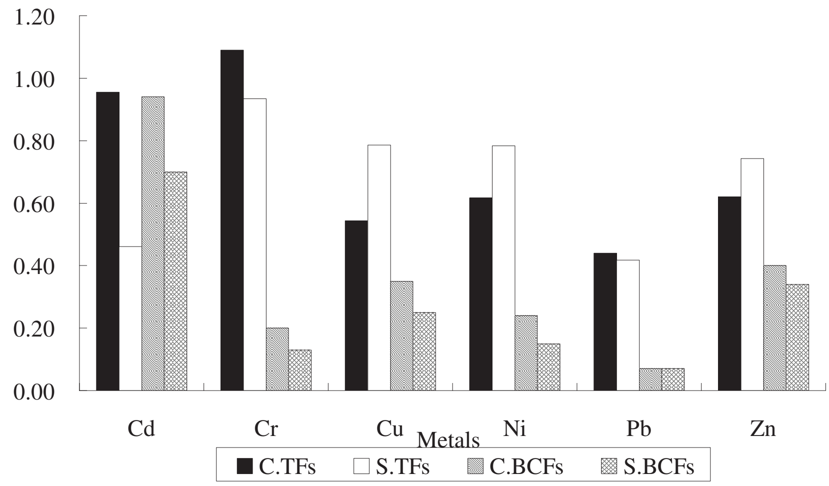

Fig. 3. Translocation factors and Bioaccumulation factors of heavy metals in the plants. C.TFs and C.BCFs represent the translocation and bioaccumulation factors in Cyperus malaccensis, repectively; S.TFs and S.BCFs represent the translocation and bioaccumulation factors in Scirpus tripuete, repectively.

In our study, translocation factors (TFs) that the ratio of the metal concentration in the aboveground parts to that in roots were calculated and presented in Fig. 3 [34]. The TFs for metals were different between the two plants and were also different among each sampling site. Most of values were less than 1, except for Cr that the highest TF value was 1.09 for Cr in *Cyperus malaccensis*.

3.4. Heavy metals distribution among water, soils and plants

As shown in the Table 1-2 and Fig. 2, the distribution of metals among water, soils and plants in different sample sites showed a similar trend as the following: soils >plants >water. The bioaccumulation factors (BCFs) that the ratio of metal concentration in the plants to that in the soils were calculated and listed in Fig. 3. The BCFs for different metals in the two plants were in the following order: Cd >Zn >Cu >Ni >Cr >Pb.

The correlations of metals among the water, soils and plants were analyzed in our study. The correlations between the soils and water, plants and water were not obvious. There were also no significant correlations between the soils and plants for Cd, Cr, Cu, Ni and Zn. The correlation coefficient for the concentrations of Pb was 0.52 with a significance level of 0.01 (2-tailed) between the plants and the soils.

4. Discussion

4.1. Concentrations of heavy metals in river water

In our study, the highest concentration among all of metals was Pb with range of 0.0128 mg/L. This result was different from the finding reported by Cheung et al. [25] that Zn was the most amount in the river samples, however, they also pointed out that the process of urban storm runoff carrying the Pb deposits into the river water appears to elevate the concentrations of Pb. On the other hand, the atmospheric depositions such as automobile exhausts and gasoline containing Pb may be other major sources of Pb in the region. According to the results reported by Wong et al. [26], atmospheric depositions of Cu, Cr, Pb and Zn were serious on the surface environment in this region.

Compared with other riparian wetlands water, the water in our study is higher enriched by Cd, Cu, Ni, Pb and Zn [31]. It is the reason that the discharge of domestic wastewater resulted in the higher metal concentrations [25]. The concentrations of Cu in all of samples exceed the range from 0.0005 to 0.001 mg/L which is the normal Cu levels in uncontaminated fresh waters [35]. The elevated concentrations of Cu may be attributed to the enrichment of Cu in the suspended matter, as [15] pointed out that even at background Cu concentrations in the river suspended matter can release considerable amounts of dissolved Cu into river water.

4.2. Concentrations of heavy metals in soils

As shown in Table 2, the concentrations of metal displayed a wide variation as reflected by the large standard deviation values, suggesting the spatial variation of metals is obvious [36]. The Zn concentration was the highest whereas the lowest concentration was Cd. This is agreement with the result reported by Gupta et al. [37] that Zn was maximally concentrated in riverine sediments collected from Ganges at Allahabad and the Cd was the least enrichment. However, when compared with the Cd concentration in water, the concentrations in soils is very high and far higher than the maximum permitted limit of class III in EQSS (1mg/kg). This is the reason that Cd is released into environment by some industries such as paints, alloys and then to be combined with the sediment and particles [38], moreover, Cd could be easily absorbed by plants and then released and enriched to the soils through the plant cycling [10]. This agrees with BCF of Cd in plants as shown in Fig. 3.

The concentrations of Cd, Pb and Zn in these riparian wetlands are significantly lower than those in Guangzhou city riverine sediment. It is apparently attributed to the decrease of disturbances by industrial or municipal activities because the sampling sites in our study mostly located in the rural area. Whereas compared with the mudflat in PRE, Cr and Pb are generally higher in our study and the other metals were obvious lower than those concentrations in mudflat [22]. It may imply the increase of accumulations of Cr and Pb occurred in recent years.

In order to further identify the relationships between different metals and its corresponding origins, principal component analysis (PCA) and cluster analysis (CA) are conducted in our study. PCA and CA could provide some indications for association of heavy metals and give some information about the origins of contamination [39].

The results of PCA reflect that Cd, Cr, Cu, Ni, Pb and Zn are dominated by two principal components which accounted for 83.4% of the total variance explains. As shown in Table 3, Cd and Pb are associated with the high values in the first component whereas Cr, Cu and Ni are greater in the second component. The concentrations of Zn are simultaneously controlled by both first and second component although the first component was more important. Many previous studies have proved that good associations of different metals indicated similar sources of pollution [17,18,40].

Table 3. The rotated component matrices for heavy metals in PCA.*

Rotated Component Matrix		
	Component	
	1	2
Cd	**0.953**	0.069
Cr	-0.037	**0.717**
Cu	0.463	**0.820**
Ni	0.254	**0.898**
Pb	**0.917**	0.135
Zn	0.813	0.545

Based on CA as shown in Fig. 4 and 5, the sampling sites are classified into three clusters using a criteria value of rescaled distance between 10 and 15. The sampling sites of Cluster A locate in the riparian of Pearl River which mainly flowed through Guangzhou (Fig. 1). It could be inferred that the contaminations of metals in these sites come from the industrial activities because 0.4, 1.7, 43, 705 and 10304 tons of Cd, Pb, Cr, Cu and Zn were annually released into Pearl River from Guangzhou city, respectively [25]. Cluster C only contains one sampling site that was site 5 with higher concentrations of Cr, Cu, Ni and Zn which locate in the downstream of a chemical factory; moreover, there are medium-sized ports for marine transports. Thus the metal concentrations are affected by both industrial wastewater and leakage or emissions of petrol used by vessels [32]. The rest sites belong to Cluster B, which has relatively lower concentrations of heavy metals. Field observation reveals that these sites are most remote and little disturbances from industry, busy marine transports and high density of human settlements. The accumulations of metals mainly come from upstream river inputs and atmospheric depositions [2, 41].

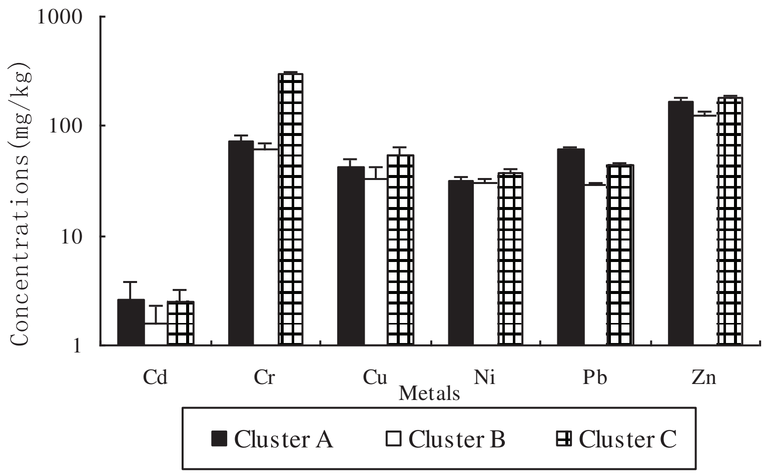

Fig. 4. Statistic analysis of metal concentrations in each cluster in CA.

The results of PCA and CA indicated that the metal accumulations of riparian wetlands of PNA in PRE are mainly caused by the disturbances of human activities. The results are in consistent with the findings reported by Ip et al. [42] that the increase in trace metals in the PRE sediments since 1970s may mainly come from anthropogenic sources of rapid development of industry and urbanization. The study identified anthropogenic activities as a major source of heavy metal contamination of riparian wetlands and the environment protection need for policy intervention in anthropogenic pollution management.

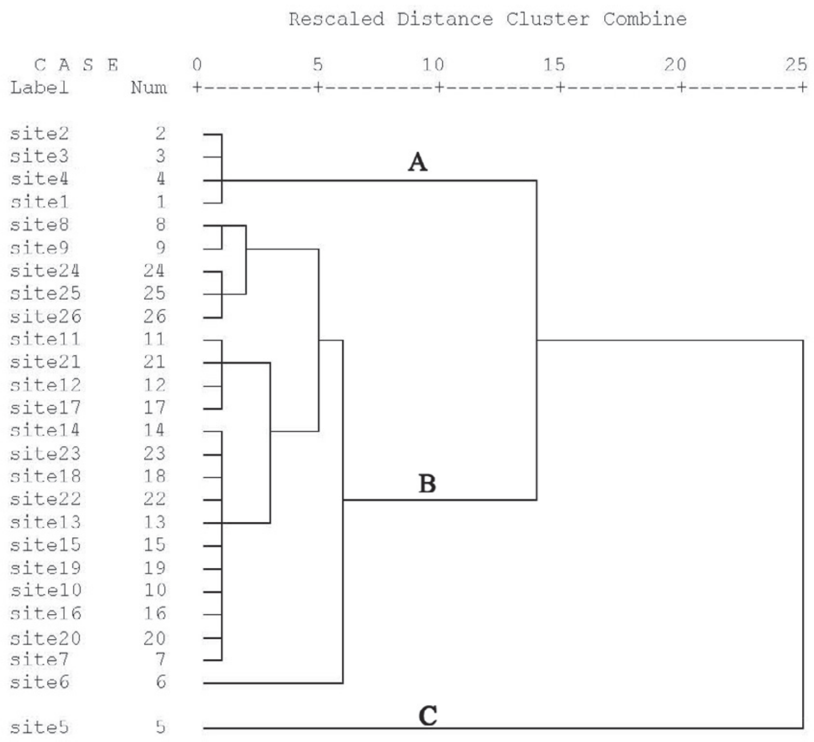

Fig. 5. Dendrogram of hierarchical cluster analysis of heavy metal concentrations in riparian wetlands of PNA.

4.3 Concentrations of heavy metals in plants

Although the metal concentrations were changed among different sampling sites, there are similar trends for different metal accumulations in the two plants with the maximum concentration of Zn, followed by Cu, Cr, Ni, Pb and Cd. The higher metal concentrations for Cu, Ni and Zn in *Cyperus malaccensis* suggest the stronger ability of *Cyperus malaccensis* to accumulate these metals from soils than that of *Scirpus tripueter*, whereas *Scirpus tripueter* has better capacity to accumulate Cd, Cr and Pb. The concentrations of Zn is the highest in the two plants among six metals and Cd is the lowest which are similar with the distribution trend of concentrations in soils, indicating that the concentration of metals in plants are associated with the concentration in soils, to some extent. Similar findings were reported by Zhang et al. [43] who studied the heavy metals in reeds collected from Hengshuihu Wetlands and found that the correlation coefficient for concentration of Cr between soils and reeds was 0.668 with a significance level of 0.01. Although Cd has the least accumulation among the metals, Cd is a highly toxic element for organisms [44] and the concentrations of Cd in normal plants from uncontaminated soils usually range from 0.05 to 0.2 mg/kg [45]. In our study, the range of Cd concentrations in the plants was 1.39-1.47 mg/kg, implying Cd contamination occurred in plants of riparian wetlands in the region.

In our study, the roots at 0-20 cm below ground parts were collected for the calculation of TFs which can reflect the ability of a plant in transferring a metal from roots to aboveground parts [34]. As is shown in Fig. 3, the most of TFs are less than 1 in the two plants, indicating that metals accumulated by these two plants were largely retained in roots. Amongst all the metals, the TFs for Cr in the two plant species are the highest (1.09 and 0.93 mg/kg, respectively), followed by Cd, Ni, Zn, Cu and Pb in *Cyperus malaccensis* and Cu, Ni, Zn, Cd and Pb in *Scirpus* *tripueter*, respectively. These indicate internal metal transportation also obviously varies among the plant species and the metal type.

4.4 Heavy metal distribution among water, soils and plants

Although the metal concentrations in soils are higher than those in water at the same sampling site, the correlations of metals between water and soils are weak according to Pearson correlation analysis. This is mainly because of high flow rate in these wetlands. Zhang et al. [43] also found that the concentrations of Pb and Cu in water have no correlations with those in soils due to the constantly flow of water in Hengshuihu wetlands.

The results in our study indicate that metals uptake by plants varied with the different of plant species. Moreover, there has no clear correlations among the metal concentrations in soils and plants ($P > 0.05$, 2-tailed) for most heavy metals in the present study. Similar results were reported by some previous studies, for example, Keller et al. [5] found that the concentrations of Ni, Cr, Zn, Cu and Pb in roots were not correlated to the concentrations in marsh sediments. Moreover, Pichtel et al. [46] were also found that Cd in plant tissue was poorly correlated with soil Cd concentration. All the results mentioned above suggest that other factors existed influence the metal uptake and storage in plants other than absorbing from soils. The concentrations of Pb in the plants indicate a positive correlation with the concentrations in soils with coefficient of 0.52 ($P<0.01$). This is similar with the findings reported by Cardwell et al. [47] that the increasing accumulation of Zn in aquatic macrophytes is positively correlated with the increasing sediment metal concentrations. Their researches indicated that there are more of less relationship in metal concentrations between plants and soils. These also can explain the fact that the concentrations of Zn and Cd were highest and lowest, respectively, in the two plants that agree with the concentrations in soils in our study. The possible correlations between concentrations in water and plants are also calculated. Metals didn't show the clear correlations between water and plants. These results are similar with the findings reported by Zhang et al. [43] that there were no correlations for Hg, As and Cu concentrations between water and reeds. The poor correlations for metal concentrations between plants and soils or plants and waters were found in our study, implying that the other factors such as air pollution including traffic emissions and the wastes discharged from industry affect the accumulation of metals in the plants.

5. Conclusions

The concentrations of heavy metals varied among water, soil and two main native aquatic plants (*Scirpus tripueter* and *Cyperus malaccensis*) collected from different riparian wetlands in PNA of PRE. Whereas the generally accumulation of metals was in the order soils > plants >water. For water, all metals, except for Pb, were below the safe values for aquatic life in China; Soil samples showed a clear accumulation for all metals when compared with the class I regulated in the EQSS; For plants, although BCFs and TFs showed the different abilities in both accumulation of metals from surrounding environments and transportation of the metals in the tissues of plants. *Cyperus malaccensis* has stronger ability of accumulation for Cu, Ni and Zn, whereas *Scirpus tripueter* can enrich more concentrations of Cd, Cr and Pb. No correlations were found between water and soils, water and plants, respectively, and amongst all metals, Pb is the only metal in plants which has a good correlation with the concentrations in soils. According to PCA and CA, the metal accumulations of Cd and Pb, Cr, Cu and Ni were originated similar sources, respectively, and anthropogenic activities were seriously contributed to the contamination of riparian wetlands of PNA in PRE.

Acknowledgements

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