



Heavy metals in soils and sediments from Dongting Lake in China: occurrence, sources, and spatial distribution by multivariate statistical analysis

Yaxin Zhang^{1,2} · Ye Tian^{1,2} · Maocai Shen^{1,2} · Guangming Zeng^{1,2}

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Abstract

Heavy metal contamination in soils/sediments and its impact on human health and ecological environment have aroused wide concerns. Our study investigated 30 samples of soils and sediments around Dongting Lake to analyze the concentration of As, Cd, Cr, Cu, Fe, Mn, Ni, Pb, and Zn in the samples and to distinguish the natural and anthropogenic sources. Also, the relationship between heavy metals and the physicochemical properties of samples was studied by multivariate statistical analysis. Concentration of Cd at most sampling sites were more than five times that of national environmental quality standard for soil in China (GB 15618-1995), and Pb and Zn levels exceeded one to two times. Moreover, Cr in the soil was higher than the national environmental quality standards for one to two times while in sediment was lower than the national standard. The investigation revealed that the accumulations of As, Cd, Mn, and Pb in the soils, and sediments were affected apparently by anthropogenic activities; however, Cr, Fe, and Ni levels were impacted by parent materials. Human activities around Dongting Lake mainly consisted of industrial activities, mining and smelting, sewage discharges, fossil fuel combustion, and agricultural chemicals. The spatial distribution of heavy metal in soil followed the rule of geographical gradient, whereas in sediments, it was significantly affected by the river basins and human activities. The result of principal component analysis (PCA) demonstrated that heavy metals in soils were associated with pH and total phosphorus (TP), while in sediments, As, Cr, Fe, and Ni were closely associated with cation exchange capacity (CEC) and pH, where Pb, Zn, and Cd were associated with total nitrogen (TN), TP, total carbon (TC), moisture content (MC), soil organic matter (SOM), and ignition lost (IL). Our research provides comprehensive approaches to better understand the potential sources and the fate of contaminants in lakeshore soils and sediments.

Keywords Soil/sediment heavy metals pollution · Pollution sources · Spatial distribution · Multivariate statistical analysis

Introduction

The accumulation of heavy metal in soils and sediments of surface is gaining concern around the world. The potential

hazard for ecological system and human health has arisen with increasing emission of heavy metal pollutants into the environment over the past decades (Dou et al. 2013; Nobi et al. 2010). Metals do not stay in the soluble form for a long time when the wastewater with heavy metals enters the surface water and tends to precipitate in the sediments (Alonso Castillo et al. 2013; Zhao et al. 2014). Immersing adjacent soil is the most basic natural transport in the process of supplying heavy metals to sediments (Kabatapendias and Pendias 2011; Kalnejais et al. 2010). Therefore, pollution degree of heavy metals in soil and sediment is considered as a reliable indicator to assess aquatic environment pollution (Looi et al. 2015; Satapathy and Panda 2015; Xu et al. 2014). Their occurrences in lakeshore areas are primarily derived from two sources: the parent materials and anthropogenic activities. Man-made

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✉ Yaxin Zhang
zhang_yx@hnu.edu.cn

¹ College of Environmental Science and Engineering, Hunan University, Changsha 410082, People's Republic of China

² Key Laboratory of Environmental Biology and Pollution Control (Hunan University), Ministry of Education, Changsha 410082, People's Republic of China

origins play an important role in the level of heavy metals in lakeshore environments, which include industrial activities (Davis et al. 2009), mining (Navarrete et al. 2007; Susaya et al. 2010), smelting (Puschenreiter et al. 2005), fossil fuel combustion, agricultural chemicals, and sewage discharges (Gao et al. 2015; Rahmanpour et al. 2014).

Dongting Lake, as the second largest freshwater lake of China, is a tourist area that is heavily disturbed by anthropogenic activities and is surrounded by farms, fisheries, shipyards, factories, and chemical plants. In recent years, there have been few researches on heavy metal pollution in the soil and sediment around Dongting Lake (Li et al. 2013). Therefore, the study of heavy metals in sediment and soil along Dongting Lake is very representative and research significance. It is necessary to investigate heavy metal concentration in the soils/sediments surrounding the shores of Dongting Lake in order to determine the status quo of soil/sediment quality and to prepare baseline data for future research. In addition, the mobility and toxicity of heavy metal in soil/sediment not only depend on their concentration, physicochemical properties, and their associations, but also on some environment conditions as like redox potential, pH, biological action, and formation of chelates (Guillén et al. 2012). Therefore, it not only requires extensive monitoring of the occurrence of heavy metals in the soil and sediment but also on surrounding environment conditions and the physicochemical properties for further studies. In this study, multivariate statistical analysis methods, such as principal component analysis (PCA) and hierarchical cluster analysis (HCA), are used to analyze the physicochemical properties and heavy

metal data for soil and sediment samples and to distinguish natural from man-made resources for heavy metals (Yuan et al. 2013; Yuan et al. 2014).

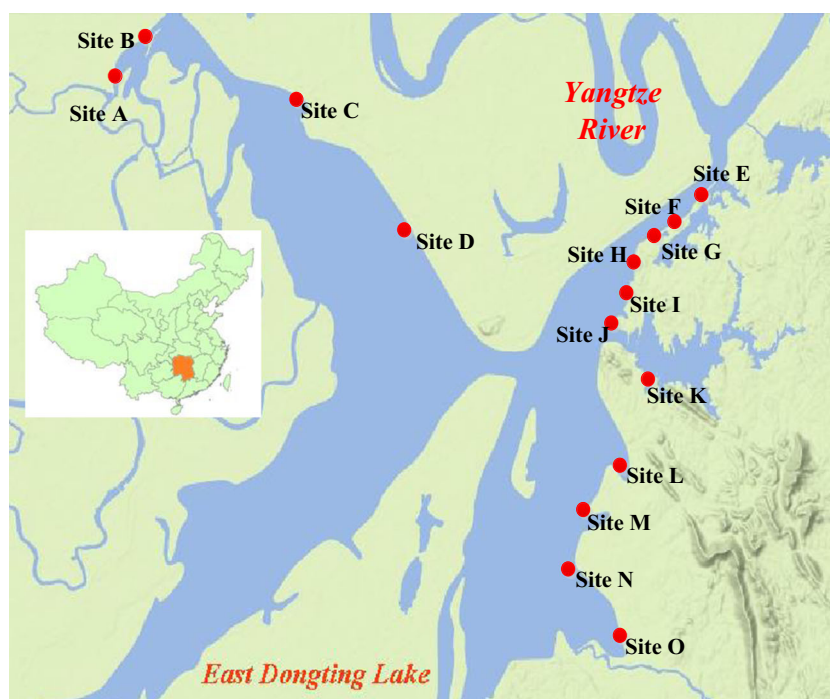
The main objectives of the present work are to study (i) the occurrence of heavy metal in soil/sediment, (ii) the main source of heavy metal in soil/sediment, (iii) the spatial distribution of heavy metals, and (iv) the relationship between the physicochemical properties and heavy metals. The study data and results of multivariate statistical methods can be used to monitor, manage, and control heavy metal pollution in soil/sediment.

Materials and methods

Sampling collection

Dongting Lake, as the second largest freshwater lake of China ($27^{\circ}39' \sim 29^{\circ}51'N$, $111^{\circ}19' \sim 113^{\circ}34'E$) with a watershed area of 2820 km^2 and the maximum depth of 30.80 m, is located by the south of the middle reaches of the Yangtze River. The study area is with intensive agricultural activities and some industries. The sampling sites are presented in Fig. 1 and more details are listed in Table S1. Surface soil and sediment samples of Dongting Lake were taken in October 2016. All 15 sites was collected for both soil and sediment samples. The soil samples were gathered to a depth of 10 cm within 2 m^2 for each sampling site which was stored in polyethylene bags individually. Sediments were collected within 2 m^2 collection locations at each sampling site, where the top layer (1–2 cm

Fig. 1 Map of sampling sites



depth) was removed in order to avoid interference and also gathered to a depth of 10 cm. Then, sediments were packed into a 2-L clean amber glass bottle with Teflon-lined caps. Then, all of the samples were sent to the laboratory and stored in a 4 °C environment. The soil and sediment samples were air-dried, grounded to 2 mm sieved, and homogenized, which were then stored for determination of the heavy metals and the physicochemical properties.

Statistical analysis

Statistical analysis in this paper was carried out using SPSS 23.0 (IBM SPSS Inc.). Principal components analysis (PCA) is crucial to ascertain the governing factors in the intricate initial dataset and classify the dataset into a PC depending on the values of a group of variables (Navarrete et al. 2017). The hierarchical cluster analysis (HCA) was accomplished on account of the heavy metal dataset of 15 studied samples. Two-dimensional HCA heat map was composed by a heat map with two dendrograms connected by colored mosaics, and the color map was on behalf of the intension of each metal for each sampling location (Li et al. 2015; Mcgregor et al. 2012).

Analytical methods and quality control

The physicochemical properties of soil and sediment were studied in this study, including pH, soil organic matter (SOM), electrical conductivity (EC), moisture content (MC), cation exchange capacity (CEC), total nitrogen (TN), total phosphorus (TP), total carbon (TC), and ignition lost (IL). The pH and EC were measured in a 1:2.5 and 1:5 ratio of sample to distilled water suspension respectively. SOM was analyzed using an oil bath-K₂Cr₂O₇ titration (Lin et al. 2017). The CEC of soil/sediment was studied by sulfuric acid barium chloride (H₂SO₄-BaCl₂) force exchange method. The method for determining total nitrogen in soils/sediments is semi-micro Kjeldahl, according to the agricultural industry criteria (NY/T 53-1987). Total phosphorus was determined by flame spectrometry following wet digestion with H₂SO₄ and HClO₄ (Lin et al. 2017). Soil and sediment samples were digested with a

commixture of HCl, HNO₃, HF, and HClO₄ in a polyvinyl fluoride couple by graphite furnace microwave digestion instrument (HJ 832-2017). Then, the digestion products were used to measure the concentration of nine heavy metals (Cr, Cd, As, Fe, Cu, Ni, Mn, Pb, Zn) by inductively coupled plasma optical emission spectrometer (PerkinElmer Optima 5300DV). All analyses were done in three sets of parallel samples and values were reported as mean.

Quality control of metal analysis was ascertained via reagent blanks, repeated experiment, and standard reference materials (ESS-3, GSBZ50013-88, from National Environmental Monitoring Center of China). The confidence level of the analyzed metals in standard reference material was 90%, and the relative standard deviation of repeated measurement was less than 10%. The limits of detection (LODs) were 0.053, 0.0027, 0.0071, 0.0054, 0.0062, 0.0016, 0.015, 0.042, and 0.0059 mg/L, respectively, for Cd, Cr, As, Fe, Mn, Ni, Cu, Pb, and Zn.

Results and discussion

Heavy metal contents

Soil

The descriptive statistical parameters of nine heavy metal contents in soils (*n* = 15) are summarized in Table 1. These results measured by Kolmogorov-Smirnov (K-S) test (*p* > 0.05) demonstrated that the concentration of As has a non-normal distribution, and those of Cd, Cr, Cu, Fe, Mn, Ni, Pb, and Zn have a normal distribution. After logarithmic transformation, the data of As appeared to follow a normal distribution. The mean concentration of heavy metals in soil (mg/kg) followed a decreasing order: Fe (25,699.0 ± 0.0062) > Mn (630.6 ± 0.0016) > Zn (138.8 ± 0.0059) > Cr (93.9 ± 0.0071) > Ni (40.4 ± 0.015) > Pb (39.3 ± 0.042) > Cu (34.3 ± 0.0054) > As (25.9 ± 0.053) > Cd (1.82 ± 0.0027). According to the national environmental quality standard for soil (GB 15618-1995), the mean concentrations of As, Cd, Cr, Ni, Pb, and Zn exceeded the background value in soil

Table 1 Heavy metal contents in soil samples (mg/kg, *n* = 15)

	As	Cd	Cr	Cu	Fe	Mn	Ni	Pb	Zn
Mean	25.9	1.82	93.9	34.7	25,699	630.9	40.4	39.3	138.8
Minimum	14.5	0.6	65.7	19.7	21,214	1.976	26.0	20.7	77.6
Maximum	54.4	4.9	121.5	57.7	33,713	1129.0	54.0	76.0	251.3
SD	11.22	1.168	18.37	8.829	3345	258.4	8.08	15.31	45.68
CV (%)	43.33	64.19	19.56	25.44	13.02	40.96	20.00	38.95	32.91
K-S test	0.008	0.200	0.200	0.200	0.200	0.200	0.200	0.200	0.200

SD standard deviation, CV coefficient of variation

of natural conditions. Concentrations of As, Cd, Ni, Pb, and Zn in sites F, G, H, and I (Table S1) were much higher than their respective mean concentration, and the concentration of Cr was higher in sites D, E, G, H, I, and K. Moreover, the coefficient of variation (CV) represented the degree of variation in a set of data, and there was a high CV of these elements (As, Cd, Cu, Mn, Pb, and Zn) accounting for the strong inhomogeneity in heavy metal contamination (Chandrasekaran and Ravisankar 2015; Ramos et al. 1994). The heterogeneity may be generated by the extrinsic factors such as agricultural, industrial, and human activities and atmospheric deposition. The low CV values for Cr, Fe, and Ni indicated a weak variation in the contents across sampling sites, and their concentrations in soil were almost constant. In sum, the accumulation and persistence of As, Cd, Cr, Ni, Pb, and Zn in soils from Dongting Lake have reached a serious level compared with the national standard (GB 15618-1995), and values of Cd, As, Mn, Cu, Pb, and Zn were highly susceptible to anthropogenic activities (Bella et al. 2013; Ungureanu et al. 2016; Wu et al. 2010).

Sediment

The heavy metal content in sediments of the sampling sites and standard statistical analysis (mean, K-S test, SD, CV) was consistent with the soils performed in Table 2. The Kolmogorov-Smirnov (K-S) test for sediment was carried out a result that all heavy metals have a normal distribution after logarithmic transformation except for As. The analysis of the 9 heavy metal contents in the 15 studied sites was discussed as follows. The mean concentrations of As, Cd, Cr, Cu, Fe, Mn, Ni, Pb, and Zn were determined to be 4.5, 0.82, 70.24, 30.21, 25,853, 781, 33.5, 34.11, and 121.6 mg/kg, respectively. Concentration of Cd in sites E, F, G, H, I, J, K, and L (Table S1) was far more than the national environmental quality standard for soil (GB 15618-1995), and higher concentrations of Pb and Zn were observed in sites E, G, I, and K (Table S1), whereas the concentrations of Cr, Fe, and Ni were significantly higher in sites A, C, D, I, and M (Table S1). The CVs reflected the variability of heavy metal in the collected samples (Luo et al. 2012; Yuan et al. 2014). Through the

report of Han et al. (Han et al. 2006), the CVs of heavy metals derived from natural sources were comparatively lower than that of heavy metals dominated by artificial sources. The high CV values for As, Cd, Mn, and Pb indicated that the contents of these elements in sediment vary strongly. On the other hand, the low CV value for Cr, Cu, Fe, Ni, and Zn suggested that these elements have a relatively stable variation in sediments. Therefore, these statistics suggested that anthropogenic activities and disturbances made a major influence in the accumulation and persistence of As, Cd, Mn, and Pb in sediments (Argyrazi and Kelepertzis 2014). We found a similar conclusion in the study of Wu et al. (2010).

Source and spatial distribution of heavy metals

Two-dimensional HCA

Hierarchical cluster analysis (HCA) is a powerful tool designed to investigate the categories within the initial datasets based on similarities and distinctions (Liao et al. 2016; Shan et al. 2010). Samples with semblable degree of heavy metal pollution or heavy metals (variables) with similar source would be clustered into the same group. Prior to HCA, concentrations of nine heavy metals in the 15 studied locations should be standardized as follows (García et al., 2004; Shi et al. 2012; Xu et al. 2013):

$$z_{ij} = \frac{(c_{ij} - c_i^0)}{sd_i} \quad (1)$$

where z_{ij} is the standardized concentration of the i th heavy metal in the j th sampling site, c_{ij} is the original concentration of the i th heavy metal in the j th sampling site, c_i^0 is the average concentration of the i th heavy metal, and sd_i is the standard deviation of the i th heavy metal.

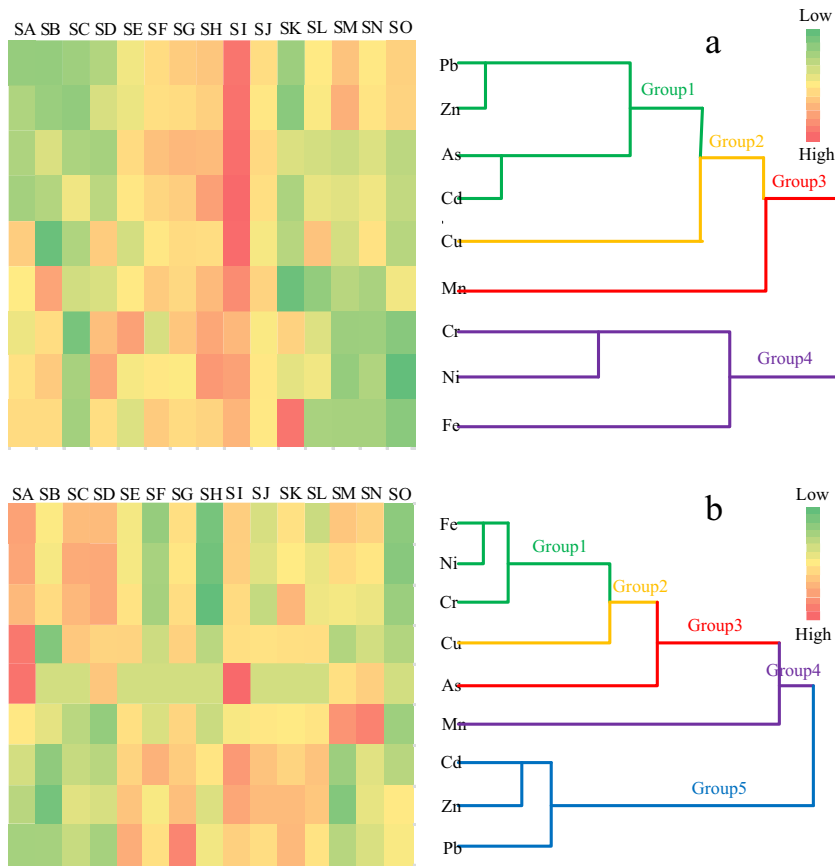
Heat map combined with HCA was performed with squared Euclidean distance and completed linkage clustering to reveal the clustering methods of the samples and variables based on heavy metal contents (Zahra et al. 2014). In this study, a two-dimensional HCA heat map was applied to the complete dataset of heavy metals and is given in Figs. 2 and 3.

Table 2 Heavy metal contents in sediment samples (mg/kg; $n = 15$)

	As	Cd	Cr	Cu	Fe	Mn	Ni	Pb	Zn
Mean	4.5	0.82	70.24	30.21	25,853	781	33.5	34.11	121.6
Minimum	6.4	0.3	51.2	20.2	21,040	490.1	22.1	20.3	79.7
Maximum	25.9	1.8	83.3	45.2	30,100	1245	42.8	59.7	155.3
SD	7.752	0.561	9.264	6.096	2703	207.5	6.17	11.73	23.13
CV (%)	172.3	68.4	13.2	20.2	10.5	26.6	18.4	34.4	19.0
K-S test	0.000	0.200	0.200	0.183	0.200	0.081	0.200	0.200	0.190

SD standard deviation, CV coefficient of variation

Fig. 2 Results of two-dimensional HCA heat map for nine heavy metals in soils (a) and sediments (b). In heat map, the deep-slight color gradient presents the concentrations of individual heavy metals of highest to lowest intensity. The HCA was based on heavy metal dataset of 135 values showing significant differences with $p < 0.05$. Euclidean correlation was used for samples and heavy metal clustering. The S followed by letter corresponds to the samples



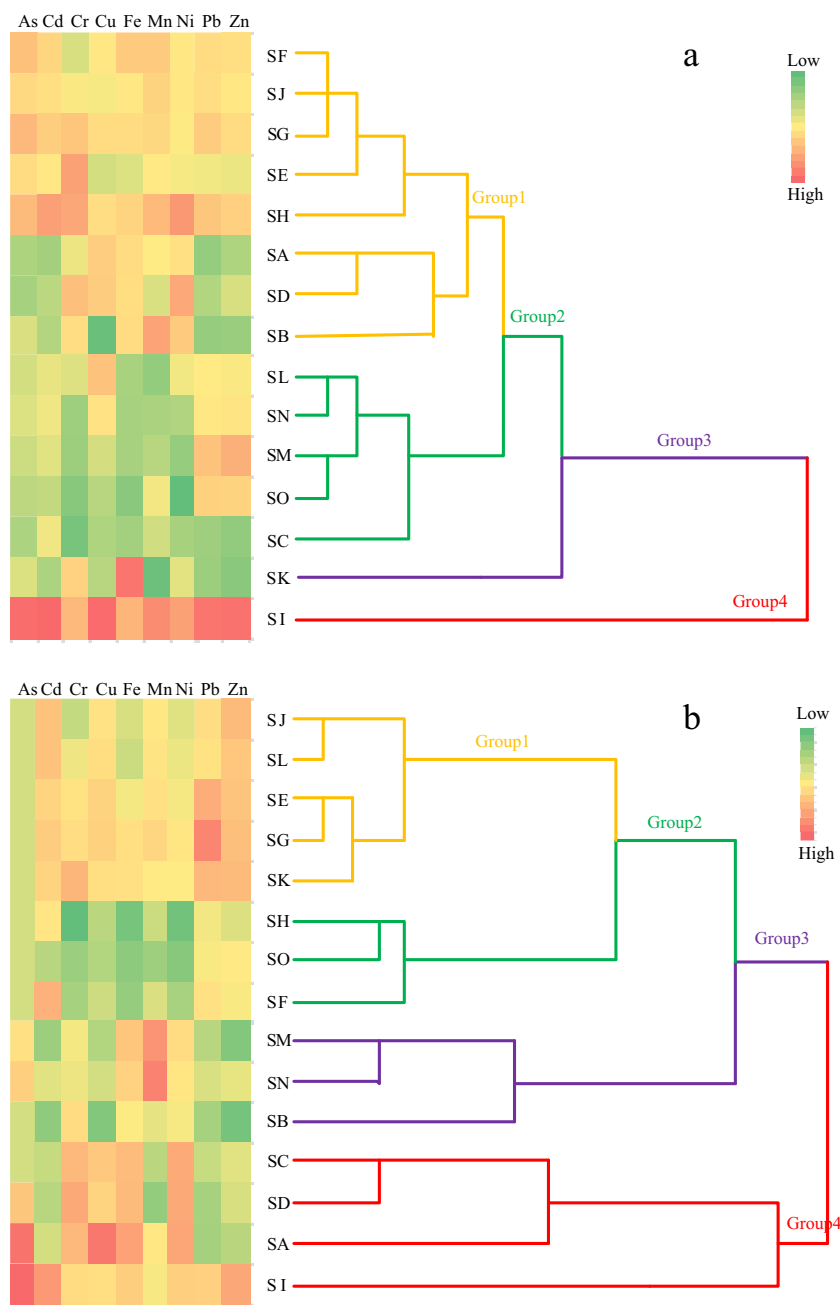
Source of heavy metals

The dendrogram associated with clustering of the variables for soils is exhibited in Fig. 2a. As shown, Pb, Zn, As, and Cd clustered together in group 1; Cr, Ni, and Fe clustered in group 4; and Cu and Mn were divided into group 2 and group 3, respectively. Heavy metals clustered in the same group may have a similar source. Many researches indicated that Pb, Zn, As, and Cd approximately originated from anthropogenic factors such as vehicle exhaust emissions, the disposal of household refuse, application of phosphate fertilizers and pesticide, and so on (Shan et al. 2010; Ungureanu et al. 2016). As shown in Fig. 2a, heavy metals contained in group 1 were clearly concentrated in sites J, I, H, and G. Combined with the surroundings of the sampling sites, sites I, H, and G were situated on the downstream of site J where the sewage treatment plant is located. Moreover, there were some factories such as harbor, concrete plants, shipping company, and chemical company around sites I, H, and G. Heavy metals in the wastewater tend to precipitate in sediments and the immersing adjacent soil is the most basic natural transport in the process of supplying heavy metals to sediments (Kabatapendias and Pendias 2011; Zhao et al. 2014). Therefore, the high level of heavy metals in group 1 was greatly impacted by these factories and domestic wastewater. Except for geogenic and pedogenic

sources, utilization of the fertilizers also had a great effect on the content of Ni in surface soils (Ungureanu et al. 2016). So, the fisheries and farms near sites L and M probably played an effect on the concentration of Ni in the downstream sampling sites (sites J, I, H, and G). Different from Pb, Zn, and Cd, Cr mainly came from natural sources, so the parent materials may also control the concentration of Cr in soils (Ungureanu et al. 2016). In this study, we can get a similar conclusion on Fe. Cu and Mn were clustered independently of the other elements, and concentration of Cu and Mn was consistent with the geographical gradient rule. Therefore, the cluster of Cu and Mn seems to be associated with geogenic and pedogenic sources. In addition, the distribution of copper in soils was similar to heavy metals in group 1, so the copper in soils may also originate from anthropogenic activities, which was used as an antifouling agent for paint, agriculture, animal, and human waste (Bella et al. 2013).

The result of HCA in Fig. 2b showed that heavy metals in sediments can be mainly divided into five groups: (1) Fe, Ni, and Cr; (2) Cu; (3) As; (4) Mn; and (5) Cd, Zn, and Pb. On the result of two-dimensional HCA, heavy metals clustered in the same group showed a completely similar regularity of distribution. So, we can deduce that heavy metals which were clustered in each group came from the same source. According to Fig. 2b, the method of clustering in sediments was similar

Fig. 3 Results of two-dimensional HCA heat map for 15 soils (a) and 15 sediments (b). In heat map, the deep-slight color gradient presents the concentrations of individual heavy metals of highest to lowest intensity. The HCA was based on heavy metal dataset of 135 values showing significant differences with $p < 0.05$. Euclidean correlation was used for samples and heavy metal clustering. The S followed by letter corresponds to the samples



with that in soils to a large extent. Therefore, based on the above analysis, we can get a conclusion that the nature of the source of heavy metals in sediments was analogical with that in soils.

Spatial distribution of heavy metals

Four main soil sample clusters are explored in Fig. 3a (group 1 in yellow, group 2 in green, group 3 in purple, and group 4 in red), and soils from sites D to H and sites A, B, and J were clustered in group 1; those from sites L to O and site C were clustered in group 2; and those from site K and site I were

clustered in group 3 and group 4, respectively. Therefore, the pollutant characteristics might be similar within each sample cluster. The heat map also explored the spatial distribution of individual heavy metal in soils in different sample groups. Concentration of heavy metal in soils in group 2 indicated a lower level in the four clusters accounting for that there was an entrance of Xiangjiang to Dongting Lake and some of its tributaries around the sites in group 2. Furthermore, the sample sites in group 2 were far from residential and industrial suburb so that there was a lower level disturbance of human activities. In group 3, site K exhibited a lower content of heavy metals except Fe, and the concentration of Fe in site K was the

highest among all sites resulting from the red earth in site K. On the other hand, a high level of heavy metals in soils was indicated in group 1 and group 4. Moreover, group 4 referred to site I, which was characterized by the highest level of almost all heavy metals since site I was located at a low-lying area and there were some factories such as harbor service, concrete company, and sewage treatment plant in the upstream of site I. Heavy metal contamination caused by anthropogenic activities was more likely to be migration and transformation (Han et al. 2006). Based on the spatial distribution of heavy metals in soils, we can deduce a geographical gradient that the accumulation of heavy metals downstream was greater than upstream because of the impact of anthropogenic activities around the study area.

The result of sediment clustering is illustrated in Fig. 3b, which allowed the identification of four groups of samples based on their similarities. It indicated that they have a similar extent of heavy metal contamination in each group. The heat map can provide intuitive information on the variance of heavy metal concentrations which followed a decreasing order: group 4 (red) > group 1 (yellow) > group 3 (purple) > group 2 (green). There were some farms and fisheries around sites A, C, and D (group 4), so the high level of heavy metals in group 4 is probably affected by the pesticides and fish feed. Some port companies and concrete plants were located in the upper reaches of site I (group 4) which may give rise to the deposition of heavy metals in site I. There were many factories such as shipping company, harbor, and chemical company around sites E, G, and K (group 1), resulting in higher background values in these places. Concentrations of Pb and Zn in group 1 were significantly higher than in other groups probably owing to the anthropogenic activities such as vehicle exhaust emissions, disposal of household refuse, application of phosphate fertilizers and pesticide, and so on (Shan et al. 2010; Ungureanu et al. 2016). On the other hand, sites M, N, and O were situated on the upstream and it was the entrance of Xiangjiang flowing into Dongting Lake, so there were not plenty heavy metals to deposit. Therefore, it generated a lower concentration of heavy metals in group 2 and group 3. However, the concentration of Mn in sites M and N was higher than in the other sites, and according to the result in the “Source of heavy metals” section, we can get a deduction that the content of Mn in sediments is probably associated with the parent materials.

Considered in Fig. 1 and according to the result of two-dimensional HCA for samples (Fig. 3), the spatial distribution of heavy metal in soil was different from that in sediments. In soils, the accumulation of heavy metal in downstream was greater than in upstream, which conformed to the geographical gradient. But there were many factors influencing the accumulation and migration of heavy metal in sediment such as aquaculture, agricultural irrigation, domestic water, water transportation, and tourism (Liao et al. 2016). There was no obvious role

of heavy metal distribution in sediments, while those sites, including sites E, G, I, and J, with high level of heavy metals in sediments were consistent with that in soils. Combined with the environment around the sampling sites, we can get a deduction that the sites with intensive human activities usually have a high level of heavy metal accumulation.

Relationship between physicochemical properties and heavy metals

Principal components analysis

As a multivariate analytical technique, factor analysis exported factors which were subaggregates of unrelated variables that interpreted the variance indicated in source dataset (Yidana et al. 2008). Composition in PCA indicated the common and unique variance of the variables, which was a variable-focused method that generated total variance of variables and the correlations with all components (Yidana et al. 2008). The accumulation and distribution of heavy metal in soils were affected not only by their natural and anthropogenic sources but also by the primary soil properties (Tian et al. 2016). Therefore, in this study, we applied PCA to the dataset of the concentration of each metal and selected physicochemical properties in all samples from Dongting Lake to extract the main factors that correspond to various origins of variation in data. Understanding the correlation between heavy metals (As, Cd, Cr, Cu, Fe, Mn, Ni, Pb, and Zn) and physicochemical properties (TN, TC, TP, SOM, CEC, MC, pH, IL, and EC) gives insights into the fate and behavior of heavy metal in sediment and soil.

Factors affecting heavy metal content in soil

All soils studied and the selected physicochemical properties were grouped into two PCs, PC 1 with a relative inertia of 31% had a positive correlation with TN, TC, TP, SOM, and IL, respectively, whereas Fe contributed to negative loadings (Fig. 4). The secondary main constituent had a relative inertia of 29.7%, ascertained by As, Cd, Mn, Ni, Cu, Zn, and Pb positively, respectively. Rotation PCA loading matrix and variable communalities for soils are arranged at Table S2. The concentration of Cr in soils was shown near to the pH of soils, indicating significant correlations between them. Meanwhile, soil pH correlated with As, Mn, Ni, and Fe. The result of PCA (Fig. 4) indicated that pH played an important effect on the concentration of heavy metal in soil. Additionally, TP revealed a certain correlation with Cu, Pb, and Zn, which demonstrated that the anthropogenic activities have an effect on the heavy metals in soils. However, there was no significant relationship among TN, TC, SOM, IL, CEC, EC, and MC in soils and heavy metals.

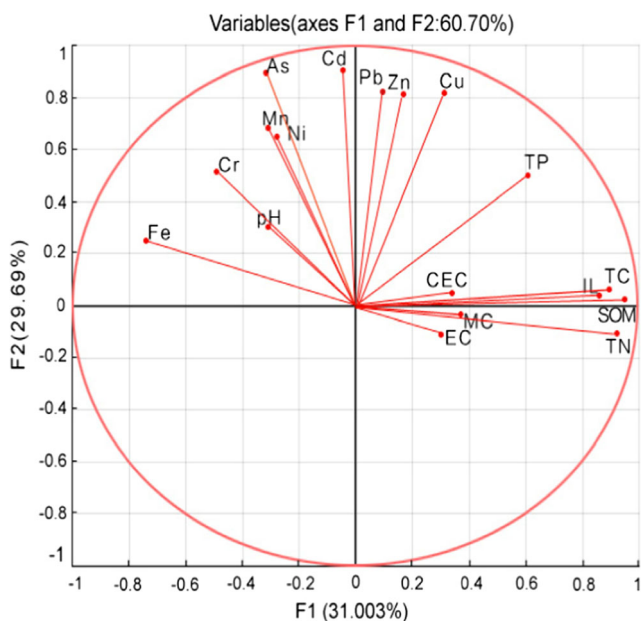


Fig. 4 Plots of the first and second principal components (PCs) extracted from the principal component analysis (PCA) of all selected soil properties and heavy metals after varimax factor rotation. The first PC is the line through the data points along which the variance is maximized

Factors affecting heavy metal content in sediment

Factor loadings and variable communalities of physicochemical properties and metal concentration in the sediments are exhibited in Table S3, and plots of the two principal components (PCs) extracted from the PCA of the entire dataset about sediment are shown in Fig. 5. All variables were divided into two PCs which explained 28 and 24% of the total variance, respectively (Table S3). In the dataset, PC 1 accounted for the largest proportion of the overall variance and PC 2 occupied the largest proportion of the residual variance and so on (Chandrasekaran and Ravisankar 2015). The result of PCA demonstrated a significant correlation of As, Cr, Fe, and Ni with CEC and pH (Fig. 5). In addition, there was a semblable conclusion with the result of HCA in the “Source of heavy metals” section. Figure 5 exhibited an obvious correlation of Pb, Zn, and Cd with TN, TP, TC, MC, SOM, and IL. Therefore, the result of PCA further illustrated that the application of phosphate and nitrogen fertilizers was the dominant factor contributing to the accumulation of Cd, Pb, and Zn in sediments, which was similar with the result of Nicholson et al. (2003). These results indicated strong affinity of TP to Pb, Zn, and Cd in sediments, implying that Olsen-P can potentially immobilize heavy metals in soils and can debase the possibility of plant absorption, as discussed by Bolan et al. (2003). The content of Olsen-P in sediments reflected the level of heavy metals persisted in sediments for a long time (Navarrete et al. 2017). Moreover, organic matter is one of the paramount factors for impacting the mobility, accumulation, and bioavailability of heavy metal in sediment. SOM in

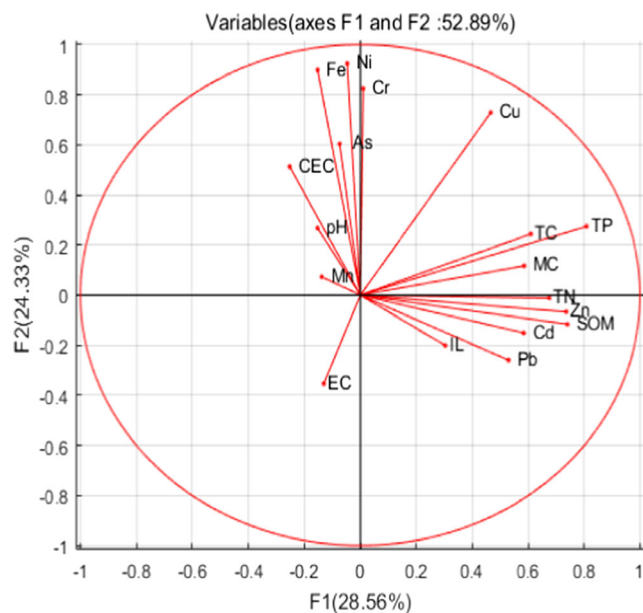


Fig. 5 Plots of the first and second principal components (PCs) extracted from the principal component analysis (PCA) of all selected sediment properties and heavy metals after varimax factor rotation. The first PC is the line through the data points along which the variance is maximized

sediments provided additional binding sites to heavy metal and improved the stability and durability of heavy metal (Navarrete et al. 2017).

Conclusion

The selected physicochemical properties and concentration of heavy metals in sediment and soil samples collected from sites around the Dongting Lake were determined to evaluate the pollution status by different analytical method. The descriptive statistical parameters combined with the result of multivariate statistical analysis revealed that the accumulations of As, Cd, Mn, and Pb in soils and sediments were effected apparently by anthropogenic activities, whereas Ni, Fe, and Cr levels were dominated by parent materials. In addition, the concentration of Cu and Mn in soils probably associated with geogenic and pedogenic sources. Furthermore, there is a geographical gradient that concentration of heavy metals in downstream was greater than in upstream in soils, but there is no similar conclusion in sediments probably because the clay fraction and hydrodynamic conditions have strong impact on the spatial distribution of metal in sediments. In the result of PCA, pH played an important effect on the concentration of heavy metals and TP revealed a certain correlation with Cu, Pb, and Zn, whereas there was no discernible effect of other properties. And for sediments, higher levels of heavy metals have been found in sediments of estuaries of rivers that have been severely affected by human activity. In addition, PCA for sediments suggested that CEC and pH played an important

effect on the accumulation of As, Cr, Fe, and Ni and there is an obvious correlation between Pb, Zn, and Cd and TN, TP, TC, MC, SOM, and IL. Multivariate data analysis is an efficient method in researching the occurrence, source, and spatial distribution of heavy metal in soil and sediment, which is vastly beneficial to assess the risks of heavy metal to ecological environment and human health.

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References

- Alonso Castillo ML, Sánchez TI, Vereda AE, García dTA, Cano Pavón JM (2013) Bioavailability of heavy metals in water and sediments from a typical Mediterranean Bay (Málaga Bay, region of Andalucía, southern Spain). *Mar Pollut Bull* 76:427–434
- Argyaki A, Kelepertzis E (2014) Urban soil geochemistry in Athens, Greece: the importance of local geology in controlling the distribution of potentially harmful trace elements. *Sci Total Environ* 482–483:366–377
- Bella GD, Turco VL, Potorti AG, Rando R, Licata P, Dugo G (2013) Statistical analysis of heavy metals in *Cerastoderma edule* glaucum and *Venerupis aurea laeta* from Ganzirri Lake, Messina (Italy). *Environ Monit Assess* 185:7517–7525. <https://doi.org/10.1007/s10661-013-3116-4>
- Bolan NS, Adriano DC, Natesan R, Koo BJ (2003) Effects of organic amendments on the reduction and phytoavailability of chromate in mineral soil. *J Environ Qual* 32:120–128. <https://doi.org/10.2134/jeq2003.1200>
- Chandrasekaran A, Ravisanakar R (2015) Spatial distribution of physico-chemical properties and function of heavy metals in soils of Yelagiri hills, Tamilnadu by energy dispersive X-ray fluorescence spectroscopy (EDXRF) with statistical approach. *Spectrochimica Acta Part A Mol Biomol Spectrosc* 150:586–601
- Davis HT, Aelion CM, Mcdermott S, Lawson AB (2009) Identifying natural and anthropogenic sources of metals in urban and rural soils using GIS-based data, PCA, and spatial interpolation. *Environ Pollut* 157:2378–2385
- Dou Y, Li J, Zhao J, Hu B, Yang S (2013) Distribution, enrichment and source of heavy metals in surface sediments of the eastern Beibu Bay, South China Sea. *Mar Pollut Bull* 67:137–145
- Gao X, Zhuang W, Chen CT, Zhang Y (2015) Sediment quality of the SW coastal Laizhou Bay, Bohai Sea, China: a comprehensive assessment based on the analysis of heavy metals. *PLoS One* 10:e0122190. <https://doi.org/10.1371/journal.pone.0122190>
- García JH et al (2004) Characterization and implication of potential fugitive dust sources in the Paso del Norte region. *Sci Total Environ* 325:95–112
- Guillén MT, Delgado J, Albanese S, Nieto JM, Lima A, Vivo BD (2012) Heavy metals fractionation and multivariate statistical techniques to evaluate the environmental risk in soils of Huelva Township (SW Iberian Peninsula). *J Geochem Explor* 119–120:32–43
- Han YM, Du PX, Cao JJ, Posmentier ES (2006) Multivariate analysis of heavy metal contamination in urban dusts of Xi'an, Central China. *Sci Total Environ* 355:176–186
- Kabatapendias A, Pendas HK (2011) Trace elements in soils and plants. CRC Press,
- Kalnejais LH, Martin WR, Bothner MH (2010) The release of dissolved nutrients and metals from coastal sediments due to resuspension. *Mar Chem* 121:224–235
- Li F, Huang J, Zeng G, Yuan X, Li X, Liang J, Wang X, Tang X, Bai B (2013) Spatial risk assessment and sources identification of heavy metals in surface sediments from Dongting Lake, middle China. *J Geochem Explor* 132:75–83
- Li J, Jia C, Lu Y, Tang S, Shim H (2015) Multivariate analysis of heavy metal leaching from urban soils following simulated acid rain. *Microchem J* 122:89–95
- Liao J, Chen J, Ru X, Chen J, Wu H, Wei C (2016) Heavy metals in river surface sediments affected with multiple pollution sources, South China: distribution, enrichment and source apportionment. *J Geochem Explor* 176:9–19
- Lin Y, Han P, Huang Y, Yuan GL, Guo JX, Li J (2017) Source identification of potentially hazardous elements and their relationships with soil properties in agricultural soil of the Pinggu district of Beijing, China: multivariate statistical analysis and redundancy analysis. *J Geochem Explor* 173:110–118
- Looi LJ, Aris AZ, Yusoff FM, Hashim Z (2015) Mercury contamination in the estuaries and coastal sediments of the Strait of Malacca. *Environ Monitor Assess* 187:1–15
- Luo XS, Yu S, Zhu YG, Li XD (2012) Trace metal contamination in urban soils of China. *Sci Total Environ* 421–422:17–30
- Mcgregor LA, Gauchottelindsay C, Daéid NN, Thomas R, Kalin RM (2012) Multivariate statistical methods for the environmental forensic classification of coal tars from former manufactured gas plants. *Environ Sci Technol* 46:3744–3752. <https://doi.org/10.1021/es203708w>
- Navarrete IA, Asio VB, Jahn R, Tsutsuki K (2007) Characteristics and genesis of two strongly weathered soils in Samar, Philippines. *Soil Res* 45:153–163
- Navarrete IA, Gabiana CC, Dumo JR, Guzman MA, Valera NS, Espiritu EQ (2017) Heavy metal concentrations in soils and vegetation in urban areas of Quezon City, Philippines. *Environ Monitor Assess* 189(145):145. <https://doi.org/10.1007/s10661-017-5849-y>
- Nicholson FA, Smith SR, Alloway BJ, Carlton-Smith C, Chambers BJ (2003) An inventory of heavy metal input to agricultural soil in England and Wales. *Sci Total Environ* 311:205–219
- Nobi EP, Dilipan E, Thangaradjou T, Sivakumar K, Kannan L (2010) Geochemical and geo-statistical assessment of heavy metal concentration in the sediments of different coastal ecosystems of Andaman Islands, India. *Estuarine Coastal & Shelf Sci* 87:253–264
- Puschenreiter M, Horak O, Friesl W, Hartl W (2005) Low-cost agricultural measures to reduce heavy metal transfer into the food chain—a review. *Plant Soil Environ* 51:1–11
- Rahmanpour S, Ghorghani NF, Lotfi Ashtiyani SM (2014) Heavy metal in water and aquatic organisms from different intertidal ecosystems, Persian gulf. *Environ Monitor Assess* 186:5401–5409
- Ramos L, Hernandez LM, Gonzalez MJ (1994) Sequential fractionation of copper, lead, cadmium and zinc in soils from or near Doñana National Park. *J Environ Qual* 23:50–57
- Satapathy DR, Panda CR (2015) Spatio-temporal distribution of major and trace metals in estuarine sediments of Dhamra, Bay of Bengal, India—its environmental significance. *Environ Monitor Assess* 187(4133):4133. <https://doi.org/10.1007/s10661-014-4133-7>
- Shan W, Xia X, Lin C, Xi C, Zhou C (2010) Levels of arsenic and heavy metals in the rural soils of Beijing and their changes over the last two decades (1985–2008). *J Hazard Mater* 179:860–868
- Shi GL, Tian YZ, Guo CS, Feng YC, Xu J, Zhang Y (2012) Sediment-pore water partition of PAH source contributions to the Yellow River using two receptor models. *J Soils & Sediments* 12:1154–1163
- Susaya JP, Kihyun K, Asio VB, Chen ZS, Navarrete I (2010) Quantifying nickel in soils and plants in an ultramafic area in Philippines. *Environ Monitor Assess* 167:505–514
- Tian K, Hu W, Xing Z, Huang B, Jia M, Wan M (2016) Determination and evaluation of heavy metals in soils under two different greenhouse vegetable production systems in eastern China. *Chemosphere* 165:555–563

- Ungureanu T, Iancu GO, Pintilei M, Chicoş MM (2016) Spatial distribution and geochemistry of heavy metals in soils: a case study from the NE area of Vaslui county, Romania. *J Geochem Explor* 176:20–32
- Xu L, Wang T, Ni K, Liu S, Wang P, Xie S, Meng J, Zheng X, Lu Y (2014) Ecological risk assessment of arsenic and metals in surface sediments from estuarine and coastal areas of the southern Bohai Sea, China. *Human Ecol Risk Assess Int J* 20:388–401
- Xu J, Tian YZ, Zhang Y, Guo CS, Shi GL, Zhang CY, Feng YC (2013) Source apportionment of perfluorinated compounds (PFCs) in sediments: using three multivariate factor analysis receptor models. *J Hazard Mater* 260:483–488
- Yidana SM, Ophori D, Banoeng-Yakubo B (2008) A multivariate statistical analysis of surface water chemistry data—the Ankobra Basin, Ghana. *J Environ Manag* 86:80–87
- Yuan GL, Sun TH, Han P, Li J (2013) Environmental geochemical mapping and multivariate geostatistical analysis of heavy metals in topsoils of a closed steel smelter: Capital Iron & Steel Factory, Beijing, China. *J Geochem Explor* 130:15–21
- Yuan GL, Sun TH, Han P, Li J, Lang XX (2014) Source identification and ecological risk assessment of heavy metals in topsoil using environmental geochemical mapping: typical urban renewal area in Beijing, China. *J Geochem Explor* 136:40–47
- Zahra A, Hashmi MZ, Malik RN, Ahmed Z (2014) Enrichment and geoaccumulation of heavy metals and risk assessment of sediments of the Kurang Nallah—feeding tributary of the Rawal Lake reservoir, Pakistan. *Sci Total Environ* 470–471:925–933
- Zhao J, Hu B, Li J, Yang J, Bai F, Dou Y, Yin X (2014) One hundred-year sedimentary record of heavy metal accumulation in the southeastern Liaodong Bay of China. *Environ Earth Sci* 71:1073–1108