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A new method for identifying potential hazardous areas of heavy metal pollution in sediments

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ABSTRACT

The combined effect of pollution source discharge and sediment adsorption leads to the rapid enrichment of heavy metals and other pollutants in lake sediments, which poses a serious threat to the lake ecosystem. Accurately identifying the risk areas of heavy metals in sediments is the key to lake sediment pollution control. Taking Taihu Lake as the study area, combined with the ecological risk status of heavy metals in sediments, the spatial clustering characteristics of pollution sources and the clustering information of sediment attributes, a potential toxic risk area identification method based on sediment source aggregation class (SLISA-SCA) was established. Through the source analysis of heavy metals in sediments, heavy metals such as Cr, Mn, Cu and Zn in Taihu Lake sediments were identified to have originated from natural sources and were subsequently disturbed by human activities to a certain extent. Cd was found to be strongly affected by human activities, and almost all Taihu Lake sediments were affected to varying degrees. In addition, the anthropogenic sources of heavy metals show high concentration clustering characteristics in the lake bay. By K-means cluster analysis of sediment attributes, three significant differences were obtained, which were determined as potential high pollution risk areas, potential medium risk areas and potential low risk areas, and the proportions were 5.6%, 27.6% and 66.8%, respectively. The SLISA-SCA model established in this study, from the perspective of source sinks, comprehensively considers the risks caused by pollution sources and sediment attributes to sediments and divides Taihu Lake into five different risk control areas (high-risk control area, potential high-risk control area, potential risk control area, potential low-risk control area and low-risk control area). This study identified areas with different levels of heavy metal pollution in Taihu Lake sediments, proposes corresponding treatment measures, and provides a scientific and systematic method and technology for the pollution management of other river and lake sediments in the world.

1. Introduction

Heavy metals are typical inorganic pollutants produced in the process of social and economic development (Kaur et al., 2022) and have carcinogenic, teratogenic, mutagenic and other effects on the human body. These pollutants are widely distributed in the environment and pose a serious threat to ecological security and human health (Esmaeili et al., 2022). Lake sediments are in a closed environment, which will make heavy metals and other pollutants more durable. When the lake environment was disturbed, various pollutants in sediments will be released again, posing a greater threat to the surrounding ecological environment and human health (Tao et al., 2019). There are more than 1400 large lakes in the world, which are important sources of water for human beings, and their river basin was also an important area for human social and economic development. Lake pollution is closely related to the production and life of the people in this region. The contradiction of global freshwater resources is becoming increasingly acute, which is also a fatal threat to mankind (Xiao et al., 2019). If the polluted areas in the lake can be identified and the areas with different degrees of pollution can be divided, it will help to accurately control lake

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pollution and improve the efficiency of lake pollution control. However, there is a lack of systematic analysis methods for the risk analysis and risk area identification of heavy metals and other pollutants in lake sediments.

Exploring heavy metal pollution in sediments from pollution sources is an important prerequisite for the study of lake ecological risk. With the rapid development of China's industrial economy, the scale and production capacity of various industries, such as the coal, building materials, metallurgy and chemical industries, are also soaring rapidly. The combustion of petroleum fossil fuels and the incomplete utilization of raw materials have inevitably led to the migration of heavy metals and other pollutants into the environment (Long et al., 2021). Studies have shown that the average daily production in China was 1.01×10^7 tons of heavy metal-containing solid waste in 2020 (China Statistical Yearbook, 2020), and the source discharge of pollutants has directly and seriously threatened ecological security. Therefore, exploring the source and spatial distribution characteristics of heavy metals in lake sediments has important scientific guiding significance. Ti is relatively stable and difficult to migrate, so it is often used as a reference element to correct the migration of other heavy metals (Wan et al., 2016). The anthropogenic-natural (AN) model can eliminate the impact of heavy metal migration and accurately and quantitatively analyse the anthropogenic and natural sources of heavy metals (Li et al., 2018). Compared with other pollutant source analysis methods, the isotope method needs to test the isotope data of each heavy metal, which requires considerable capital and manpower. Source analytic receptor models, such as the positive definite matrix method, have rotational uncertainty and human subjectivity, which will bring great errors to the analytical results. Kriging spatial interpolation can smooth the spatial variation in pollutants, so it is often used to explore the distribution of pollutants in sediments (Ahn et al., 2019). In fact, spatial anomalies are very common in sediments. As industry and population gather in some areas, this may lead to point or line pollution of pollutants in nearby rivers and lakes (Khiari et al., 2021; Vega-Herrera et al., 2022). Therefore, more attention must be paid to the cluster distribution pattern of the lake area.

Lake sediments are an important sink of pollutants. Sediment properties, such as total organic carbon (TOC) and clay, can adsorb pollutants such as heavy metals. The adsorbed pollutants can be stored in sediments more permanently, therefore increasing the potential risk of pollutants in sediments (González-Gaya et al., 2019; Berrojalbiz et al., 2011). Studies have shown that the higher the content of clay and organic matter in sediments is, the higher the concentration of pollutants. However, in the current sediment risk research, the impact of sediment attributes on risk assessment is often neglected, and few studies include sediment attributes such as clay and organic matter content in the pollutant risk assessment system. The main reason for this phenomenon is that the sediment properties cannot have a threshold similar to heavy metals and other pollutants (Feng et al., 2017), which leads to the fact that the potential risk of sediment properties cannot be well evaluated. K-means clustering belongs to the classification method of unsupervised learning. It combines the data of different categories into the same category, with the advantages of simplification and efficiency. It has been applied to the risk analysis of different pollutants (Morais et al., 2021; Schwarz et al., 2022).

Early studies generally used the risk assessment methods of heavy metal pollution, such as the enrichment index, geoaccumulation index and potential ecological risk index (Hakanson, 1980; Muller, 1969; Cai et al., 2015). To some extent, these methods accurately identify whether the sample points are polluted and the degree of pollution, and these methods are still used today (Aguilera et al., 2021). With the development and update of technology, geographic information technology has been widely used in the spatial prediction of pollution risk, which extends the previous point pollution risk assessment to surface pollution risk assessment, makes the evaluation results clearer and more specific, and makes substantial progress in the risk assessment of heavy metal pollutionm (Wu et al., 2019). As research on heavy metal pollution in sediments has formed a relatively perfect system, relevant research experts have generally reached a consensus: the pollution of heavy metals in sediments is jointly affected by the intensity (source) of anthropogenic emissions and the strong pot (sink) of sediment adsorption capacity. However, in the current research on the identification of heavy metal risk areas (Jia et al., 2020), few studies reasonably include the emission of pollution sources and the adsorption factors of collection areas in the evaluation system, which reduces the practical application value of these risk area identification methods.

With the rapid development of industrial economy, the demand for fresh water resources has increased sharply, and the contradiction of water resources has become increasingly serious. As an important reservoir of fresh water, the environmental quality of lakes is very important. This study comprehensively considered the risk status of pollutant source–sinks and takes Taihu Lake as the study area. The main research purposes are as follows: (1) to quantitatively analyse the sources of heavy metals in sedimentary materials and their spatial cluster distribution patterns; (2) to explore the spatial clustering characteristics and potential risk status of sediment attributes in Taihu Lake; and (3) to establish a comprehensive risk area identification method of heavy metals in sediments based on source–sink.

2. Materials and methods

2.1. Sample collection and processing methods

Taihu Lake is located in the subtropical zone, with a mild and humid climate. The lake covers an area of 2427.8 km^2, with hills and mountains in the west and southwest and plains and water networks in the east. The water system of Taihu Lake flows from west to east, with an average annual runoff of 7.5×10^{10} m^3 and a water storage capacity of 4.4×10^{10} m^3, which is an important freshwater resource in the Yangtze River Delta region of China. The average depth of Taihu Lake is approximately 1.8 m, which makes Taihu Lake a shallow lake. Taihu Lake Basin is one of the largest comprehensive industrial bases in China, and industrial production technology and equipment have a good foundation. The industrial category structure is electronics, machinery, chemistry, metallurgy, textile and food, and the industrial output value of these six industries accounts for more than 85% of the total industrial output value. A stainless steel grab was used to collect sediment samples from Taihu Lake at an average interval of 3 km, and the sample collection density was appropriately increased in the area close to the shore and in the lake bay (Fig. 1). The first 2 cm of each sediment sample was selected and stored in a polyethylene self-sealing bag, and the sample number was marked on the bag. The 63 surface sediment samples collected were placed in an incubator with dry ice and transported back to the laboratory. Freeze-drying All sediment samples were freeze-dried, and foreign substances such as shells and plant roots were removed. The sediment particle size was measured by a Mastersize 2010 laser particle size metre produced by Malvern Company in the UK. The organic matter in the sediment was determined by the potassium dichromate volumetric method and external heating method. Heavy metals in sediments were determined by inductively coupled plasma emission spectrometry (ICP–OES) and inductively coupled plasma–mass spectrometry (ICP–MS). The specific operation steps are in the supplementary materials.

2.2. Quantitative source analysis

For the source analysis of heavy metals in sediments, taking Ti as the reference element can eliminate the impact of heavy metal migration, eliminate noise, and more accurately and sensitively analyse the contribution of anthropogenic sources and natural sources (Wan et al., 2016). The specific calculation formula is as follows:

$$[M]_{anthropogenic} = [M]_{sample} - [M]_{natural} \tag{1}$$

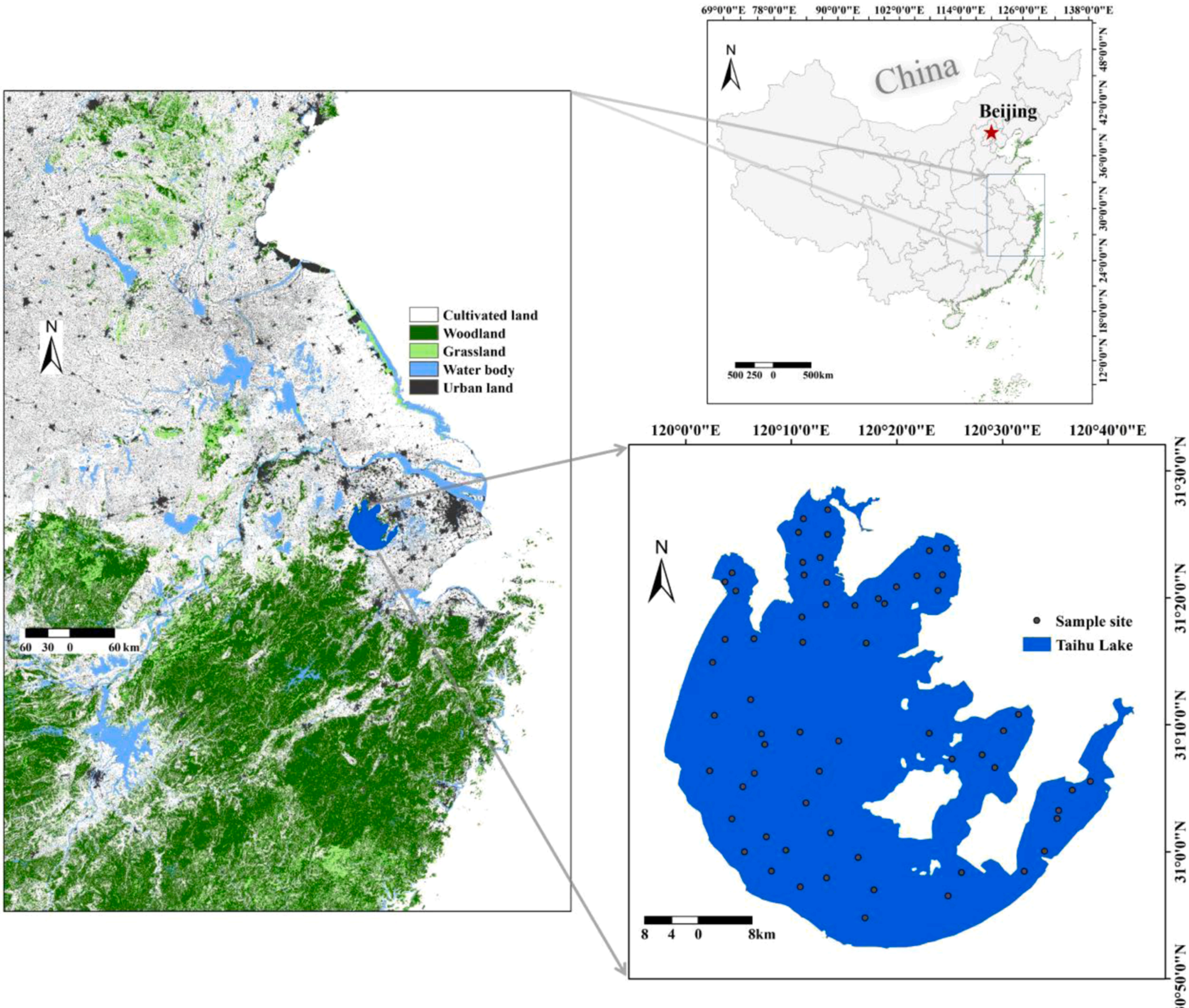

Fig. 1. Study area and sampling map.

$$[\mathrm{M}]_{\mathrm{natural}} = [\mathrm{Ti}]_{\mathrm{sample}} \times [\mathrm{M}]_{\mathrm{background}} \Big/ [\mathrm{Ti}]_{\mathrm{background}} \tag{2}$$

$$\mathrm{R}_{\mathrm{anthropogenic}} = [\mathrm{M}]_{\mathrm{anthropogenic}} \Big/ [\mathrm{M}]_{\mathrm{sample}} \tag{3}$$

In Formulas (1) and (2), $[\mathrm{M}]_{\mathrm{anthropogenic}}$, $[\mathrm{M}]_{\mathrm{natural}}$ and $[\mathrm{M}]_{\mathrm{sample}}$ represent the anthropogenic, natural and total concentrations of metal M in sediment samples, respectively. $[\mathrm{Ti}]_{\mathrm{background}}$ and $[\mathrm{Ti}]_{\mathrm{sample}}$ represent the natural background value and total concentration of Ti in sediments, respectively. $[\mathrm{M}]_{\mathrm{background}}$ indicates the concentration of metal M in the natural background material. $\mathrm{R}_{\mathrm{anthropogenic}}$ is the anthropogenic source contribution rates. The background values of elements in Taihu Lake sediments are selected from Ref. Li et al. (2018).

2.3. Local indicators of spatial association

The global Moran index method was proposed by Patrick, an Australian statistician, in 1950. It is an important statistical index used to study the spatial autocorrelation of a region. The so-called spatial autocorrelation, similar to time series autocorrelation, refers to the overall spatial correlation of the spatial attributes of a spatial variable at different locations in the study area and the spatial attributes of its surrounding adjacent locations. Because of its harsh and complex requirements, Anselin proposed local indicators of spatial association (LISA) in 1995. The methodology holds that for the subregions centred on any spatial elements in the study area, the autocorrelation statistical index can be calculated to express the spatial correlation in different subregions to better analyse its distribution law (Anselin, 1995). The specific calculation formula of LISA is as follows:

$$C_j = \left[\left(x_j - \overline{x}\right) * \sum_{i=1, i \neq j}^{n} w_{i,j} * \left(x_i - \overline{x}\right) \right] \Bigg/ \frac{\sum_{i=1, i \neq j}^{n} \left(x_i - \overline{x}\right)^2}{n-1} \tag{4}$$

In Formula (4), $C_j$is the local Moran index corresponding to the j-th sample, $w_{i,j}$ represents the spatial weight value between sample i and sample j, and n is the total number of samples of variable x. LISA analysis was completed by geoda software (http://geodacentre.github.io/).

2.4. K-means clustering

The basic principle of the traditional K-means clustering algorithm is that first, the user selects K scenes as the initial clustering centre in advance, where k is the number of typical scenes expected at the end of clustering (Vasilaki et al., 2018). Then, the Euclidean distance from each remaining sample scene to the initial cluster centre was calculated, and the remaining sample scenes were classified into their adjacent initial cluster centres to form a cluster according to the proximity principle. Finally, the average Euclidean distance of scenes in K clusters was calculated, and the value of the average Euclidean distance of each cluster was taken as the new cluster centre. Therefore, the process of scene compression was completed by using the traditional K-means clustering algorithm until the new cluster centre and the old cluster centre were no longer changed and the cluster centre was output. The flow chart of scene compression is shown in Fig. S1 By calculating the distance between each sample and each cluster centre, the k-means algorithm marks the samples according to the principle of nearest distance (Peng et al., 2018; Gautam et al., 2022):

$$dij = \sqrt{\sum_{k=1}^{p} \left(x_{ik} - x_{jk}\right)^2} \tag{5}$$

where each micro trip has P parameters, $x_{ik}$and $x_{jk}$are the k characteristics of samples i and j, and $d_{ij}$is the distance of each micro trip. If the same sample data are clustered repeatedly, the appropriate K value should produce the same or similar clustering results; i.e., stability was regarded as an indicator of whether the K value is appropriate. According to the clustering stability test results, it was finally determined that all driving units were grouped into three categories. K-means clustering analysis was carried out in SPSS 26.0 (Statistical Graphics Crop, Princeton, USA).

2.5. Establishment of the SLISA-SCA model

The SLISA-SCA model was mainly composed of two parts (Fig. 2): first, the source of heavy metals in sediments was used to quantitatively analyse the contribution of human activities in each region of Taihu Lake, cluster analyse its spatial distribution, and evaluate the ecological risk of heavy metals. The second was the attribute of the heavy metal sink area, which was classified by K-means clustering, and its potential risk status was explored. Finally, based on the source sink risk analysis of heavy metals in sediments, a conceptual model was established to partition the risk of heavy metal pollution in Taihu Lake.

3. Results and discussion

3.1. Characteristics of heavy metals

The spatial distributions of the concentrations of heavy metals such as Pb, Cr, Zn, Cu and Mn in sediments are similar (Fig. 3a). The concentration was relatively high in the lake bay area, which was basically concentrated in Zhushan Bay, Meiliang Bay and Gonghu Bay. In the centre and southeast of Taihu Lake, the concentration of heavy metals was relatively low. The concentration of the heavy metal Cd was the highest near the west bank of Taihu Lake, higher in Zhushan Bay and Gonghu Bay, and lower in the centre and southeast of Taihu Lake. The area close to the shore and the lake bay area were most affected by human activities, and the corresponding pollutant concentration will be higher. The flow direction of Taihu Lake (from west to east) and the special handicraft industry of coastal cities affect the spatial distribution of heavy metals to a great extent. Combined with the ecological risk analysis of heavy metals (Fig. 4), it was found that the areas with pollution risk of heavy metals in Taihu Lake sediments were also basically distributed in Lake Bay areas, especially Zhushan Bay. The pollution risk areas of different heavy metals vary greatly. The areas with pollution risks of the heavy metals Cr, Mn, Ni, Cu, Zn, Cd and Pb accounted for 34.9%, 7.9%, 6.3%, 15.9%, 4.8%, 96.8% and 66.7%, respectively. Among them, Cd pollution in the sediments of Taihu Lake was the most serious, which basically covers the whole Taihu Lake. This situation required management to pay enough attention and take relevant pollution control measures immediately.

Investigations and studies have shown that Yixing City on the West Bank of Taihu Lake, with the reputation of “ceramic capital”, produces a large number of ceramic products year round and sells them at home and abroad. However, raw materials for ceramic production, such as gehuang and cadmium red, contained a large amount of Cd. In the process of ceramic production, they inevitably enter the lake environment through atmospheric dust, wastewater infiltration and waste

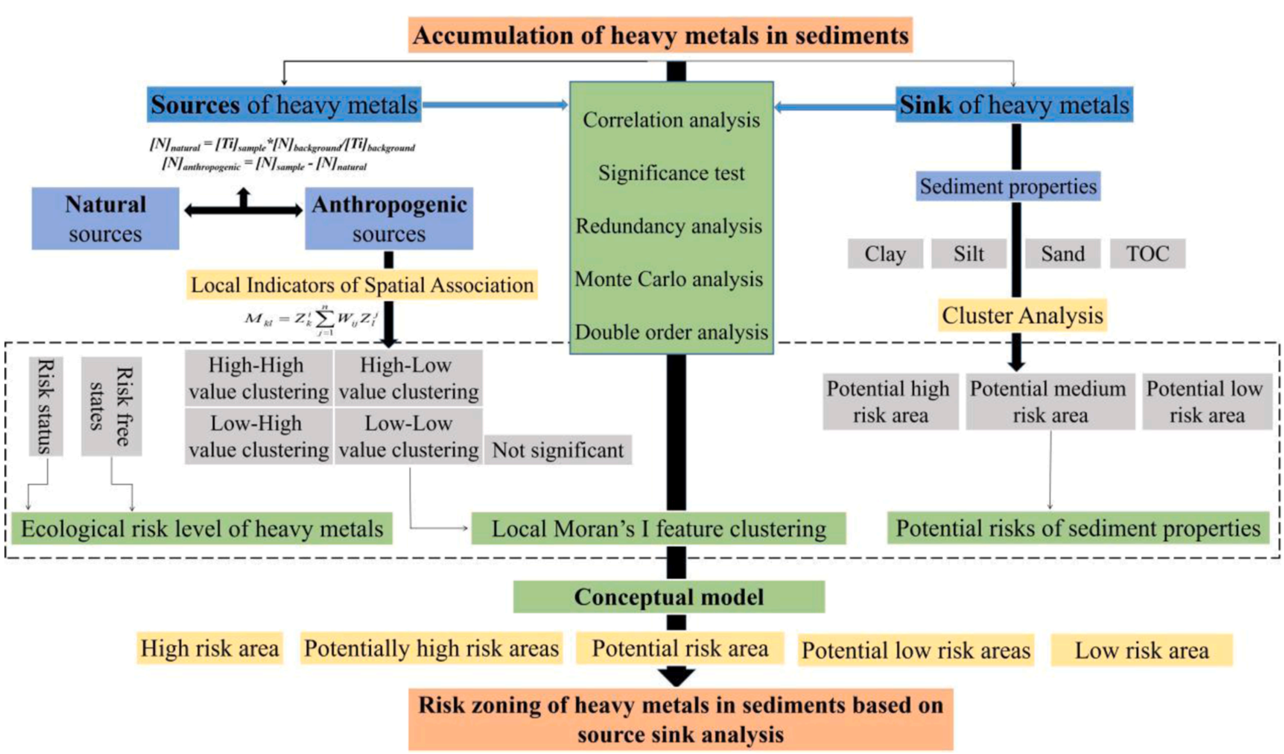

Fig. 2. Flow chart of heavy metal risk identification in sediments based on source-sink analysis.

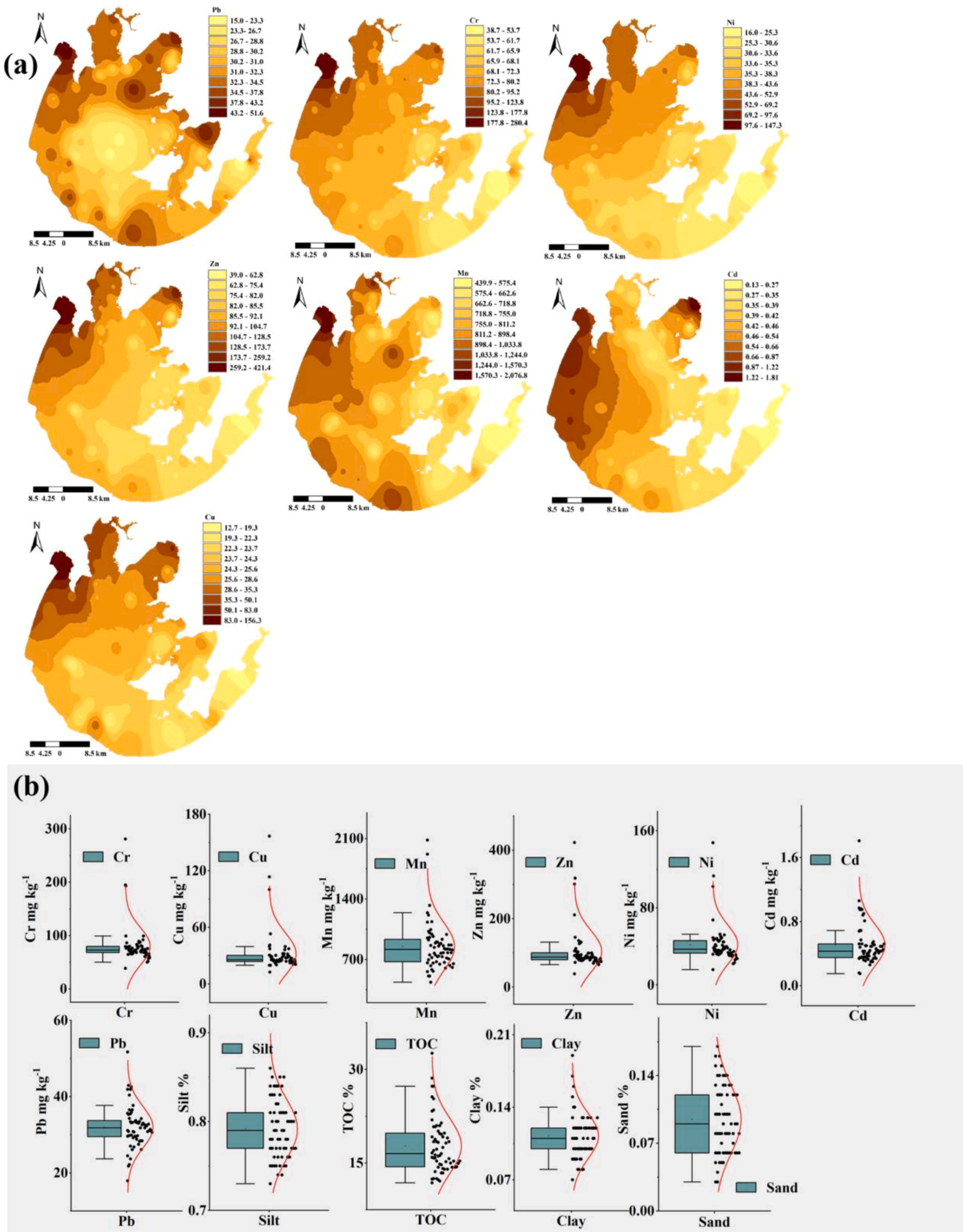

Fig. 3. Spatial distribution (a) and statistical characteristics (b) of heavy metal concentrations in sediments of Taihu Lake.

transportation, which poses a great threat to the ecological security of Taihu Lake. Studies have shown that the ceramic industry in Yixing has caused serious Cd pollution to the surrounding soil, sediment and lake water (Liao et al., 2015). Compared with the concentrations of heavy metals in the sediments of other lakes at home and abroad (Table 1), Cr, Mn and Cu in the sediments of Taihu Lake were at a medium level, while the heavy metals Pb and Cd are at a high level. The environmental management department should pay attention to this situation, and

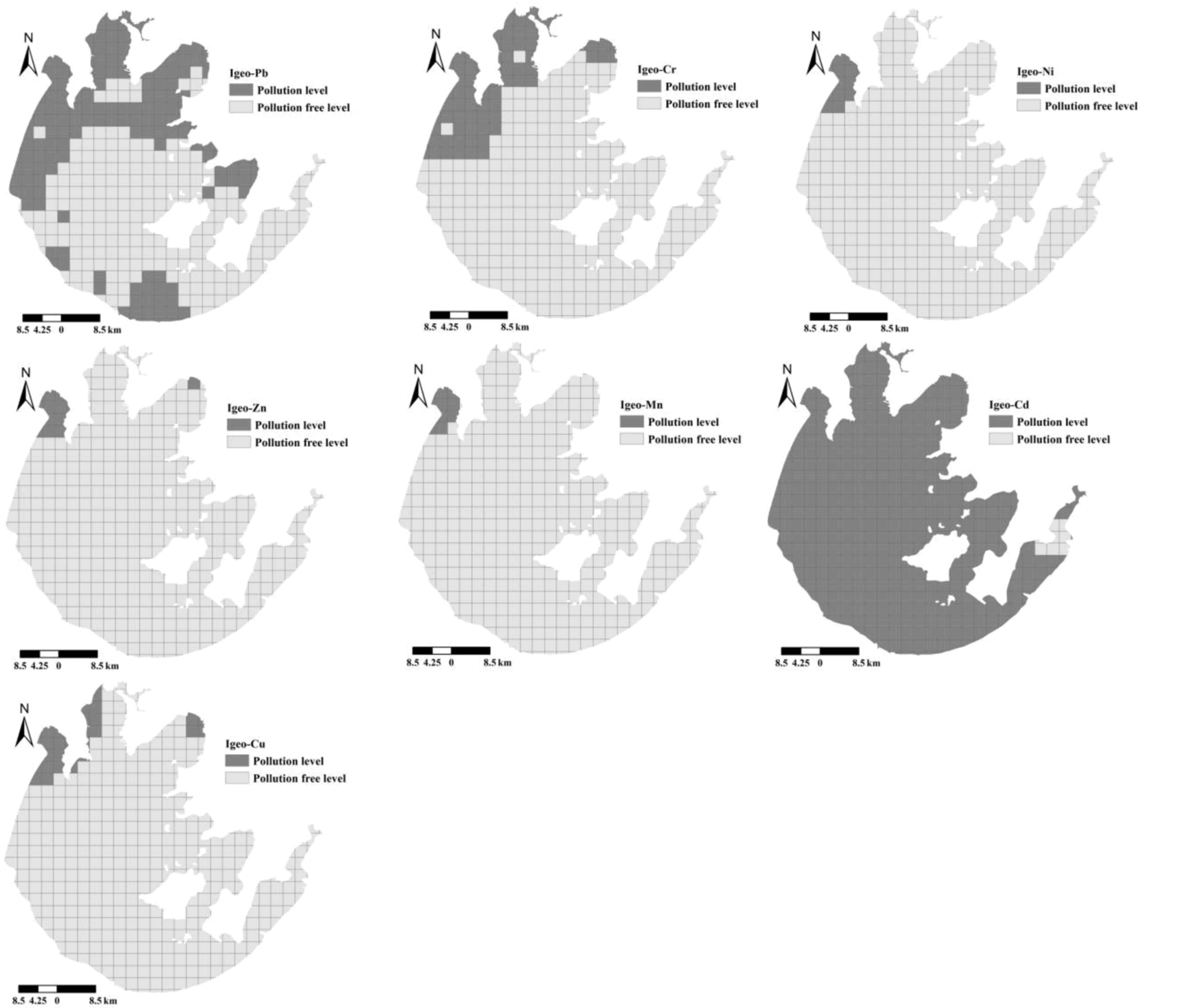

Fig. 4. Risk of heavy metals in sediments.

Table 1
Concentrations of heavy metals in sediments of Taihu Lake and other lakes (mg kg^{-1}).

	Cr		Mn		Ni		Cu		Refs.
	Mean	Range	Mean	Range	Mean	Range	Mean	Range	
This study	80.0	38.4~281.0	850	438~2081	41.9	15.8~147.6	31.7	12.6~156.7	
Laizhou Bay							10.6	6.2~14.4	Liu et al. (2019)
Bohai							34.1	20.2~60.7	Liu et al. (2019)
Yellow River	51.7	40.1~69.2			26.5	19.8~39.6	18.5	13.5~24.7	Liu et al. (2019)
Yueqing Bay	61.5						34.2		Yao et la. (2021)
Bellandur wetland, Indian		33.9~199.4				15.1~138.4		105~1147.8	Ramachandra et al. (2018)
East Sea-Byeong, Korea	73.9		1589		37.4		32.7		Kim et al. (2021)

	Zn		Cd		Pb		Ti		Refs.
	Mean	Range	Mean	Range	Mean	Range	Mean	Range	
This study	104.6	38.7~422.3	0.51	0.15~1.81	32.0	17.9~51.7	4324	3894~4862	
Laizhou Bay	62.6	13.0~112.2	0.33	0.21~0.47	14.1	10.0~20.4			Liu et al., 2019; Liu and Fan, 2019
Bohai	100.0	53.0~197.4	0.61	0.25~2.53	30.9	24.3~43.4			Liu et al. (2019)
Yellow River	62.2	40.6~84.2	0.15	0.09~0.28	21.6	18.1~28.4			Liu et al. (2019)
Yueqing Bay	107.3		0.12		28.4				Yao et la. (2021)
Bellandur wetland, Indian		125.7~2001		1.6~55.3		31.2~308.2			Ramachandra et al. (2018)
East Sea-Byeong ocean dumping site, Korea	116.0		0.31		33.9				Kim et al. (2021)

clarifying the sources of Pb and Cd in the sediments of Taihu Lake has become the primary task of pollution control.

3.2. Sources and spatial clustering characteristics of heavy metals

The sources of heavy metals can also be divided into natural sources and anthropogenic sources. Natural heavy metals of Taihu mainly come from soil erosion, nineral weathering and biogeochemical cycles (Li et al., 2018). Anthropogenic sources of Taihu mainly come from industrial, agricultural production and life processes (Liu et al., 2021; Niu et al., 2020). Different heavy metals may have the same or similar pollution sources or may come from different pollution sources. To explore the source characteristics of heavy metals in Taihu Lake sediments, the correlation and significance between different heavy metals were analysed (Table S3). The heavy metals Cr, Mn, Ni, Cu, Zn, Cd and Pb in Taihu Lake sediments are significantly correlated at the level of 0.01, and the correlation between the heavy metals Cr, Mn, Ni, Cu and Zn is more than 0.8 (R^2 value is 0.64). This indicated that there were similar pollution sources of heavy metals in the sediments of Taihu Lake, and some heavy metals may have the same pollution sources. In contrast, the correlation between the heavy metal Cd and other elements in Taihu Lake sediments was relatively weak, maintained at approximately 0.5 (R^2 value was approximately 0.25), which indicated that in addition to the similar pollution sources between Cd and other elements, there were also their own unique pollution sources. These special pollution sources were different from other elements, and the heavy metal Pb also had similar characteristics to Cd in pollution sources.

To further determine the types of pollution sources and their contribution of heavy metals in sediments, we analysed the sources of heavy metals in sediments of Taihu Lake (Table S4). According to the analysis of the heavy metal source analysis model, the sources of heavy metals were divided into natural sources and anthropogenic sources. Basically, the natural source variation range of each heavy metal was narrow and relatively stable, and the value was small, which was in line with the content characteristics of natural source elements, indicating that the source analysis results have high reliability. Compared with the natural source concentration of heavy metals in the sediment of Taihu Lake, the anthropogenic source concentration of heavy metals varies widely, which was several to dozens of times that of its natural source. The coefficient of variation of the heavy metals Cr, Mn, Ni and Cu exceeded 100%, indicating that they have been strongly disturbed by the external environment. The anthropogenic source contribution rates of Cr, Mn, Ni, Cu, Zn and Pb in sediments were 34.2%, 11.7%, 11.9%, 24.5%, 12.0% and 34.1%, respectively, which indicated that the environment of Taihu Lake has been seriously disturbed by human beings. Most of the heavy metal sources in the sediments of Taihu Lake contribute 74.4% of the Cd (Fig. 5c). This phenomenon needs to be given enough attention. Heavy metals in Taihu Lake have been obviously and strongly disturbed by human beings. Taihu Lake Basin is the largest comprehensive industrial base in China, and its electronics, machinery, chemistry, metallurgy, textile and other industries account for more than three quarters of its total industrial output value. As we all know,

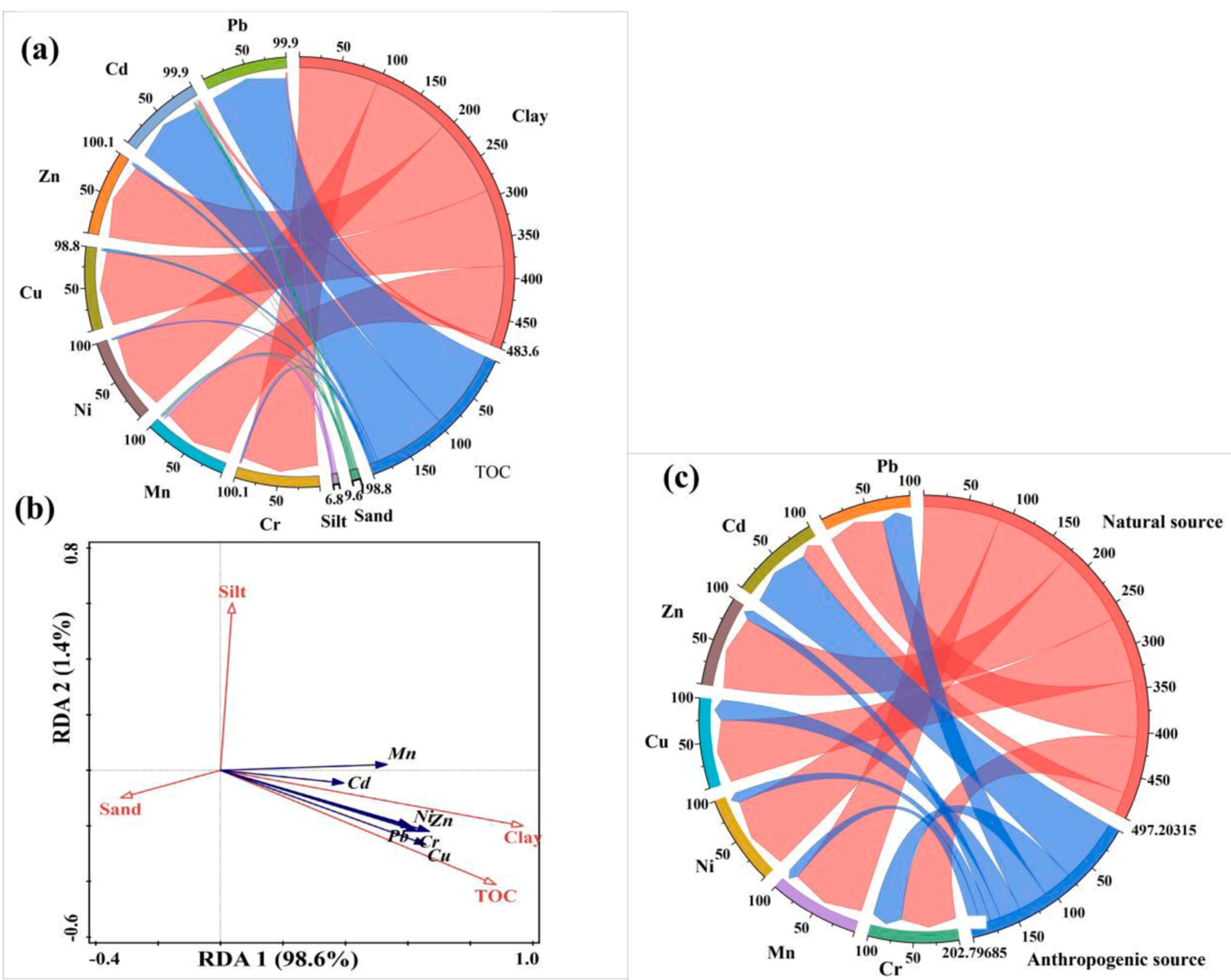

Fig. 5. Influence degree of sediment particle size and TOC on heavy metals (a), redundancy analysis of sediment particle size and TOC and heavy metals (b), contribution of pollution sources to heavy metals (c).

electronics, metallurgy, chemistry and other industries are important sources of heavy metal pollution (Avellaneda et al., 2022). In the process of production and processing, it is inevitable to discharge waste rich in heavy metals into the surrounding environment.

3.3. Cluster analysis of sink sediment attributes

The properties of sediments in the collection area can significantly affect the enrichment of pollutants to a certain extent. The sediment attribute data of Taihu Lake were tested and described (Table S2): the average concentration of sediment TOC was 17.7 mg kg^{-1}, with a variation range of 11.8~32.6 mg kg^{-1}. The average proportion of clay particles was 11.3%, and the variation range was 7~19%. The average proportion of silt was 79.2%, and the variation range was 73~86%. The average proportion of sand is 9.5%, and the variation range is 3~17%. The high content of TOC in sediments was mainly distributed in Zhushan Bay, Meiliang Bay, Gonghu Bay and East Taihu Lake (Fig. S2a), and the TOC content is relatively low in the southern, western and central areas of Taiyuan. The clay content showed similar spatial distribution characteristics to TOC. The content of sand and gravel was mainly concentrated in the middle and high sand and gravel areas in western Taihu Lake and southern Taihu Lake. Five years ago, the total organic carbon content in Taihu Lake sediments was 6.86 mg g^{-1}, and the clay content was 11.0%. In contrast, the content of clay particles in Taihu Lake sediments has not changed, while the content of total organic matter has increased significantly, which indicates that the accumulation of organic matter in Taihu Lake sediments has been significantly affected in recent years. The correlation between different properties of sediments and various heavy metals is obvious (Table S3). There was a significant positive correlation between TOC and clay content and various heavy metals, while there was no significant correlation between gravel and silt content and various heavy metals. By taking the concentration indices of heavy metals in sediments as species factors and sediment particle size, TOC and other indices as environmental factors, redundancy analysis was carried out. The analysis results are shown in Fig. 5a and 5b. The contents of TOC and clay particles in sediments of Taihu Lake have significant effects on all heavy metals, and their influence degrees reach 28.6% and 69.2%, respectively, which largely determines the enrichment capacity of heavy metals in sediments. The content of sediment silt and sand has a weak effect on the enrichment of heavy metals.

The characteristics of sediment TOC and particle size affect or determine the enrichment capacity of heavy metals and other pollutants to a great extent and pose a potential threat to the ecological security of lakes. To clarify the combined characteristics of different attribute indices of sediments in Taihu Lake and classify the combined characteristics of different attributes, this study carried out cluster analysis (K-means clustering) on different attribute indices of sediments. In the K-means clustering algorithm, the number of categories K had an important impact on the accuracy of the clustering results, but usually the accurate value of K was unknown at first. If K was greater than the true value, the same class of data will be wrongly divided into multiple classes, which will lead to the fuzzy boundary of the clustering results. In contrast, merging different categories of data into the same category will reduce the compactness of the cluster. Therefore, clustering stability was usually used to determine the K value (Luxburg et al., 2010; Bendavid et al., 2007). The basic idea of this method was that if the same sample data wre clustered repeatedly, the appropriate K value should produce the same or similar clustering results; i.e., stability was regarded as the index of whether the K value is appropriate. According to the clustering stability test results, it was finally determined to cluster all driving units into three categories (Tables S6, S7). Because there was no change or only a small change in the cluster centre, convergence was realized. The maximum absolute coordinate change of any centre was 0. The current iteration was 7, and the minimum distance between the initial centres was 8.506.

According to the cluster analysis, there were significant differences among the three types of sediments (Table S7). The contents of TOC, clay, silt and sand of the first type of sediment were 23.9 g kg^{-1}, 14.2%, 78.0% and 8.7%, respectively. The contents of TOC, clay, silt and sand of the second type of sediment attribute were 19.5 g kg^{-1}, 11.8%, 80.7% and 8.7%, respectively. The contents of TOC, clay, silt and sand of the third type of sediment attribute were 15.2 g kg^{-1}, 10.3%, 78.8% and 9.3%, respectively. In summary, there were obvious differences in the attribute characteristics of the three types of sediments. The first type of sediment had the highest content of organic matter and clay, had a strong adsorption capacity for heavy metals and other pollutants and was basically distributed in the lake bay area (Fig. S2b). This area is strongly disturbed by human activities. Therefore, the area of the first type of sediment was determined to be a potential high pollution risk area. The content of organic matter and clay particles in the second type of sediment was relatively high, and the adsorption capacity of heavy metals and other pollutants was relatively strong, which was roughly distributed in the lake bay and the area near the shore. This area also receives relatively strong interference from human activities. Therefore, the area of the second type of sediment was determined to be a potential medium-risk area. The third type of sediment had the lowest content of organic matter and clay, the adsorption capacity of heavy metals and other pollutants was very small, and the content of gravel was the highest. It was mainly distributed in southern Taiyuan and the central area, and the interference from human activities was weak. Therefore, the area of the third type of sediment was determined as a potential low-risk area. The proportions of potential high-risk areas, medium-risk areas and low-risk areas of heavy metals in sediments of Taihu Lake are 5.6%, 27.6% and 66.8%, respectively.

3.4. Identification of potential risk areas

Combined with the sources of heavy metals in sediments and the attribute characteristics of sediments in the collection area, we established a new method to comprehensively evaluate the pollution risk status of heavy metals in sediments from the following three aspects: the ecological risk of heavy metals in sediments, the spatial clustering characteristics of anthropogenic sources of heavy metals, and the attribute clustering characteristics of sediments in the collection area. If the geoaccumulation index (Igeo, details are in the supplementary materials) was greater than 0, it indicated that there was an ecological risk of heavy metals in sediments. If the geoaccumulation index was less than 0, it indicated that there was no ecological risk of heavy metals in sediments. There were three spatial clustering characteristics of anthropogenic heavy metal concentrations: high-high, low-low and not significant. High-high means that each point in the area is surrounded by high concentration points of heavy metals, indicating that the area has high pollution risk. Low-low means that each point in the area is surrounded by low concentration points of heavy metals, indicating that the area has no pollution risk or low pollution risk. The nonsignificant area runs through the points of high concentration and low concentration, showing an irregular state. There may or may not be heavy metal pollution risk in this area. To some extent, the attribute characteristics of sediments in the collection area determine the enrichment capacity of pollutants and become a potential risk index. According to cluster analysis, the sediment attributes of Taihu Lake were divided into three cluster combinations: potential high-risk areas, potential risk areas and potential low-risk areas. According to the established comprehensive risk assessment method, the study divides the risk status of Taihu Lake into five types: high-risk control area, potential high-risk control area, potential risk control area, potential low-risk control area and low-risk control area. For example, if an area exceeds the ecological risk threshold of heavy metals, the spatial clustering characteristics of anthropogenic sources of heavy metals show a high-high area, or the clustering of sediment attributes shows potential risks, this area is the health risk control zone of heavy metal pollution. The classification of different levels of potential risk control areas is mainly based on the clustering characteristics of sediment attributes, and the special circumstances of the other two factors are also considered. The specific classification information and standards are shown in Table S8.

Combined with the established new comprehensive risk assessment method for sediments, this study divided the risk status of various heavy metals in Taihu Lake sediments (Fig. 6). The high-risk control areas of heavy metal Pb in sediments are mainly distributed in Zhushan Bay, Meiliang Bay and Gonghu Bay, and the potential high-risk control areas were mainly distributed in Xukou Bay and East Taihu Lake, accounting for 7.4% and 2.6%, respectively. Potential risk control areas and potential low-risk control areas were mainly distributed in the lake bay and near the lake shore, and low-risk control areas were mainly distributed in the centre of the lake, accounting for 35.9%, 25.6% and 28.5%, respectively. The comprehensive risk zoning of Sediment Heavy Metals Cr, Mn, Ni, Cu and Zn showed similar characteristics to Pb (Fig. S3). The high-risk control area of heavy metal Cd in sediments was mainly distributed in West Taihu Lake and Zhushan Bay, the potential risk control area covers almost the whole Taihu Lake, and the distribution area of the potential high-risk control area and low-risk control area was very small, accounting for 14.7%, 80.6%, 3.2% and 1.5%, respectively. Based on this pollution zoning, the environmental management department can take different governance measures according to different pollution conditions of heavy metals in sediments. For example, for high-risk and potentially high-risk control areas, measures such as regular dredging and algae removal can be taken to cut off the enrichment of heavy metals. For potential risk areas, we can strengthen the restriction intensity of lake operation activities in this area and observe sediment and water quality in real time. Environmental managers should recommend the generalization of domestic and industrial wastewater treatment and the implementation of equipment such as constructed wetlands aimed at depurating rainwaters flowing on streets, roads and pavements. In addition, regarding all measures to be taken in all areas of Lake Taihu, environmental managers should recommend the generalization of domestic and industrial wastewater treatment and the implementation of equipment such as constructed wetlands aimed at depurating rainwaters flowing on streets, roads and pavements.

This method has been well applied in this study and addressed gaps for the study of classification methods for indicators lacking classification thresholds, such as sediment attributes. The newly established SLISA-SCA model combined the risk clustering characteristics of the sediment heavy metal source sink and its own pollution risk level to make a reasonable risk zoning for the study area, and the model reduced the uncertainty of risk identification and zoning caused by only using spatial interpolation. Compared with other technologies, the SLISA-SCA model has stronger reliability and a wider scope of application for risk identification and management (Table 2). However, there were still some problems in this model that need to be further improved: 1. The bioavailability of different forms of heavy metals in sediments varies greatly. If conditions permit, it is best to test the content of free and residual heavy metals in sediments to better evaluate the ecological risk of heavy metals in sediments; 2. Black carbon components such as char and soot have strong adsorption. These indicators can also be considered when classifying sediment attributes.

4. Conclusions

Lake sediments are in a closed environment, in which heavy metals and other pollutants are rapidly enriched. When the lake environment is disturbed, various pollutants in the sediment will be released again, posing a greater threat to the surrounding human health and ecological environment. The study of the risk of heavy metal pollution in the lake system is complex, and the heavy metals in the area with a great impact of human activities show a strong enrichment at this point, which will strengthen the spatial heterogeneity of heavy metals. LISA can overcome this defect and is a good method to identify specific pollutant hotspots. In addition, the TOC and clay properties of sediments have a strong ability to adsorb heavy metals and other pollutants, which can greatly increase the potential ecological risk of heavy metals in sediments.

This study used a model to analyse the anthropogenic and natural sources of heavy metals in Taihu Lake sediments and found that human activities significantly accelerated the enrichment of heavy metals in sediments. Using LISA and K-means clustering analysis, the spatial clustering characteristics of heavy metal elements and attribute characteristics in sediments are analysed, and different potential risk types caused by sediment attributes were identified. Based on this, the SLISA-SCA model established in this study, from the perspective of source sinks, comprehensively considers the risks caused by pollution sources and sediment attributes to sediments and divides Taihu Lake into a high-risk control area, potential high-risk control area, potential medium-risk control area, potential risk control area, potential low-risk control area and low-risk control area. The SLISA-SCA model reduced the uncertainty of risk identification and zoning and provides a more intuitive and objective spatial view of heavy metal risk management and control.

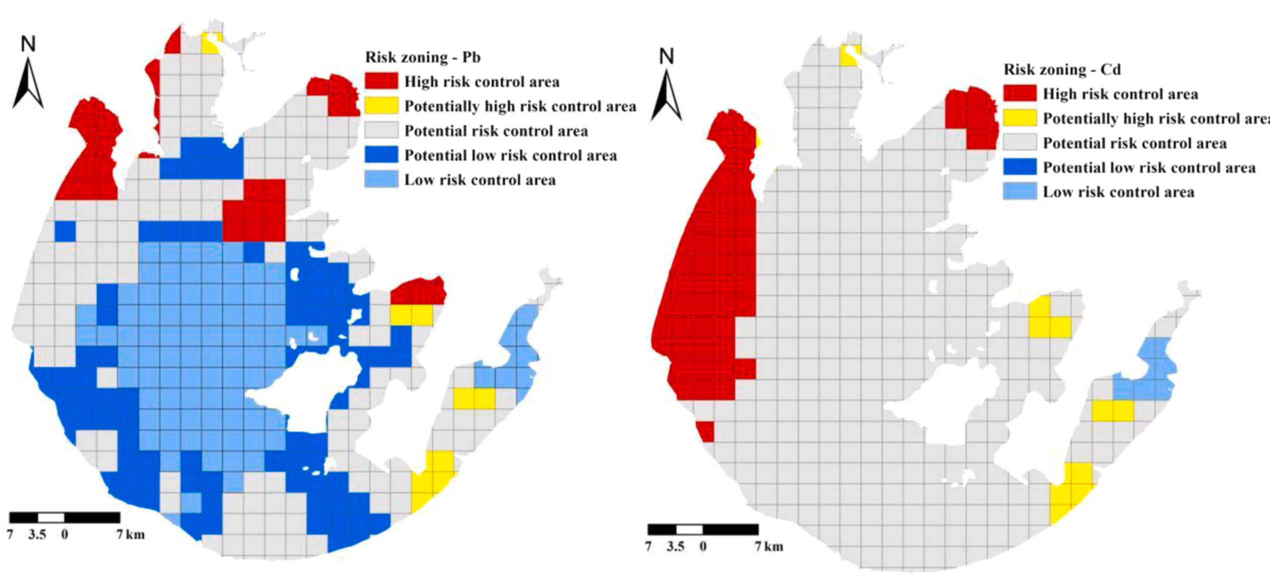

Fig. 6. Identification of heavy metal Pd and Cd risk areas in sediments based on new method analysis.

Table 2
Comparison of different risk management methods for heavy metals in sediments.

Method	Risk assessment	Spatial interpolation	Spatial clustering of HM pollution sources	Sediment attribute clustering	Risk hotspot (point or surface)	Risk zoning	Applicability	Fitness	Refs.
SLISA-SCA	√√	Yes	√√√	√√√	surface	Yes	√√√	√√√	This study
PIs	√√√	–	–	–	point	–	√	√	Feng et al. (2017)
SQGs	√√√	–	–	√√	point	–	√√	√√	Hjy et al. (2020)
ERI	√√	–	√√	–	point	–	√	√√	Niguse et al. (2018)
CCME-SeQI	√√√	Yes	–	–	surface	–	√	√√	Ahn et al. (2019)
AHH	√√	–	√	–	point	–	√	√	Ayoob et al. (2015)
BLISA-RA	√√	Yes	√√√	–	surface	Yes	√√	√√	L et al. (2021)
ERA	√√√	Yes	√√	–	surface	–	√√	√√	Zhai et al. (2021)

Note: √: fair; √√: good; √√√: excellent; –:not included; Applicability: the ability to play a specific role in the actual situation; Fitness: reliability and rationality of results.

Compared with other technologies, this model had stronger reliability and a wider scope of application for risk identification and management, and this research provides important technical and theoretical support for the management of river and lake pollution.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data Availability

Data will be made available on request.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.watres.2022.119065.

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