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RESEARCH ARTICLE

Distribution and ecological risk assessment of heavy metals in sediments of Dajiuhu Lake Wetland in Shennongjia, China

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Abstract

The rapid development of modern society has resulted in discharge of large, heavy metal quantities into wetlands that have been continuously accumulating, causing severe pollution. Dajiuhu, located in the Shennongjia Forest District of Hubei Province in China, is a wetland of significant value internationally, serving as a model wetland ecosystem with heightened scientific research value. In this study, 27 surface sediment samples from nine sub-lakes in Dajiuhu were collected in August 2020. The concentrations of Cd, Cr, Cu, Ni, Pb, and Zn in the sediments were determined. The heavy metal occurrence and speciation characteristics were analyzed by an improved BCR (European Community Bureau of Reference) extraction method. Four methods were used to evaluate heavy metals' pollution degree and ecological risk. The possible source of heavy metals was inferred using correlation analysis and principal component analysis. The heavy metal content in the lake sediments of Dajiuhu wetland was from the highest to the lowest concentration as follows: Zn > Cr > Ni > Pb > Cu > Cd. The average Cd content exceeded the national nature reserve threshold values, while the other heavy metals measured were below their respective threshold values. However, due to the occurrence of Pb and Cd in different forms, they still pose certain pollution and ecological risk to the lake wetlands. On the other hand, Zn, Cr, Ni, and Cu do not pose an ecological risk in the lakes of the Dajiuhu wetland. The spatial distribution of heavy metal content in the nine sub-lakes did vary significantly. Regarding the heavy metal sources in the lake sediments, Ni, Cr, and Cu originate from natural factors, and Cd and Pb have mainly anthropogenic origins. In contrast, Zn has both natural and anthropogenic origins. This study provides further insights into the study of heavy metal pollution in lake wetlands. It provides a framework and a direction for managing heavy metal pollution in the Dajiuhu wetland.

Keywords Heavy metal · Sediments · Ecological risk · Lake · Wetland · Dajiuhu

Introduction

Wetland ecosystems are among the most productive natural ecosystems, providing many essential ecological functions, including water purification, flood storage regulation, and

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climate regulation and so on (Zhang et al. 2014). They also have an irreplaceable role, serving as habitats for endangered species. Due to economic development needs, many wetlands were converted into farmland in the middle and late twentieth centuries. In addition, under the circumstance of excessive resource exploitation and pollution, a large part of wetlands shrank, their biodiversity was reduced, and the corresponding ecological functions were weakened. The heavy metal contents in lake wetlands have become an important environmental index of wetland health, The control of heavy metal pollution is key for wetland protection and restoration. Heavy metals can enter the lakes from natural and anthropogenic sources. These include hydrological processes, natural erosion, atmospheric deposition, agricultural non-point source pollution, and industrial activities, both natural and man-made (Merciai et al. 2014; Zhang et al. 2021). Only a small part of the heavy metal percentage entering the lake is dissolved in the water, and most accumulates in the sediment, which functions like a "sink" (Ros et al. 2014).

Lake sediments are rich in organic matter, iron and manganese oxides, and secondary clay minerals, which can strongly adsorb heavy metal ions, resulting in a large number of pollutants accumulating on the sediment surface (Wang et al. 2018). At the same time, the changes in the physical and chemical properties of water (including temperature, pH, electrical conductivity, organic matter content, etc.) can lead to the re-release of heavy metals from the sediments into the overlying water, resulting in secondary lake water pollution. As a result, sediment is considered to be the most important pollution sink and a heavy metal source (Zhang et al. 2021). Sediments in lake wetlands provide food and habitat for fish and macrobenthos. Due to this, they are a key heavy metal source in the water environment and the food chain. Therefore, sediments are the best indicator to reflecting the heavy metal pollution status in lakes (Mamat et al. 2016). Heavy metal and organic pollutants coexist in many areas (Mattina et al. 2003; Zhang et al. 2010; Chen et al. 2020). Both pollutants can accumulate in sediments. As environmental pollutants, organophosphate esters (OPEs) can usually coexist with heavy metals, and there is an interaction between them (Hu et al. 2021). Heavy metals often affect the change of the absorption and transport behavior of organic chemicals (Lu et al. 2014b; Wang et al. 2017).

The heavy metal chemical form (metallic or organic) plays a key role in their environmental and ecological effects (Liang et al. 2017; Müller 1969; Zahra et al. 2014). Analysis of the heavy metal chemical and of the factors affecting them helps to explore the migration and transformation process of heavy metals in the soil and their potential threat to the ecological environment (Xia et al. 2020; Yuan et al. 2014; Zhao et al. 2013). Currently, the Tessier and the improved BCR (European Community Bureau of Reference) methods are widely used for the sequential extraction of heavy metals from sediments (Li et al. 2022). The Tessier method divides heavy metals into exchangeable, carbonate-bound, Fe/Mn oxide-bound, organic, and residual forms. The improved BCR method divides heavy metals into acid-soluble form (F1), reducible form (F2), oxidizable form (F3), and residual form (F4).

Dajiuhu is a world-famous human and biological reserve, a biodiversity protection demonstration area, and a national wetland park. It has great significance in the ecology research field towards the protection of the ecological environment. At the same time, the Dajiuhu wetland is the largest and best-preserved subalpine wetland in central China and is a key region for ensuring regional ecological security. The Dajiuhu wetland includes sphagnum bog, sleep vegetable bog, cattail bogs, numerous ponds, and lakes (Zhang and Cai 2003). In the previous century, a series of agricultural activities such as large-scale artificial reclamation, planting, and breeding took place, seriously damaging the wetland ecosystem of Dajiuhu and reducing the wetland area from 708 to 179 ha. After an ecological restoration project implementation, Dajiuhu has recovered nine sub-lakes, but the previous agricultural expansion still significantly impacts the ecosystem of the Dajiuhu (Li et al. 2017). Dajiuhu wetland has always been the focus of ecological and environmental research. Previous research projects have measured and analyzed the heavy metal content in the Dajiuhu wetland. The peat wetlands investigation found that atmospheric deposition resulted in Hg, Cu, and Pb pollution (Li et al. 2016; Liu et al. 2021). Atmospheric deposition has repeatedly been reported as the main source of Hg in wetland waters, which leads to excessive Hg and As in aquatic animals in the water (Chu et al. 2021; Ning et al. 2021). Thus, there is a significant occurrence of heavy metal pollution in the area, but additional research that systematically evaluates the pollution and ecological risk of the lake sediments is lacking.

To the best of our knowledge, this is the first study to comprehensively investigate and analyze multiple heavy metal contamination in the sediments from all lakes of Dajiuhu, assessing the ecological risks. The following studies were investigated in this study: (1) the concentrations and morphological characteristics of Cd, Cr, Cu, Ni, Pb, and Zn in the sediments of Dajiuhu lake wetland; (2) the sediment heavy metal pollution and ecological risk were evaluated using an potential ecological risk index, a geo-accumulation index, a risk assessment code, and a pollution factor approach; (3) the total amount of heavy metals in the sediments and their possible sources were analyzed. This study supplements the current research on heavy metal pollution of subalpine lake wetlands in China with a multitude of heavy metal pollution data and environmental characteristics. It, further, provides insights into heavy metal pollution remediation of the Dajiuhu wetland.

Materials and methods

Study area

The Dajiuhu wetland (31°27′ ~ 31°33′ N, 109°56′ ~ 110°11′ E) is located at the western end of Shennongjia National Prak in Hubei Province of China, at the watershed between the Yangtze River and the Han River. Dajiuhu basin covers an area of 40.5 km^2, with an average elevation of 1730 m, and is surrounded by mountains of 2200 ~ 2600 m (Zhang Zhiqi et al. 2015). In 2014, Dajiuhu Wetland Management Bureau began to build a reservoir on the upper reaches of Lake 1 to store water and regulate the water surface of the lower reaches, with an area of about 300 hm^2 in the wet season and 200 hm^2 in the dry season (Li et al. 2017). Nine lakes of different sizes are formed in the basin. The surface of the nine sub-lakes gradually decreases from south to north. The sub-lakes are connected with each other by ditches. The water flows from Lake 1 to Lake 9 and eventually flows to the dark river through the downfall hole.

Sediment collection and analysis

In August 2020, a grab dredger was used to collect sediment samples from Lake 1 to Lake 9. Three sampling sites were set up in each sub-lake, with a total of 27 surface (0–10 cm) sediment samples collected (Fig. 1). The collected samples were stored in a polyethylene bags in a portable refrigerator (4 ℃) and transported back to the laboratory for further processing. Weeds, gravel, plant roots, and other impurities were picked out. Then, the samples were placed into a centrifugal tube and freeze-dried in a vacuum freeze dryer at – 70 ℃ to constant weight. They were thoroughly grounded in an agate mortar through a 200-mesh plastic sieve. The processed samples were sealed and stored in a 4 ℃ refrigerator to be used for the subsequent measurements.

The microwave digestion method (Tuzen et al. 2004) was used to determine the total metal element amount in the sediments. The BCR four-step extraction method (Nemati et al. 2011) was used to identify the fractionation of heavy metals. Inductively coupled plasma mass spectrometry (ICP-MS, NexION 350, PerkinElmer, America) was used to determine the heavy metal content of each sample. The detail parameters of ICP-MS are shown in Table S1.

Assessment of heavy metal pollution

Potential ecological risk index

The potential ecological risk index (PERI) (Hakanson 1980) evaluates the risk of heavy metals in sediments based on their characteristics, migration, and environmental transformation. The calculation formula of the single heavy metal potential ecological risk (E^i_r) and composite heavy metal potential ecological risk (RI) (Chen et al. 2016; Hu et al. 2013) is as follows:

$$E_{fi} = T_i \times C_i / C_{bk} \tag{1}$$

$$RI = \sum_{i=1}^{n} E_{fi} \tag{2}$$

where, T_i represents the toxicity coefficient of heavy metal i (the toxicity coefficient of Cd, Cr, Cu, Ni, Pb, and Zn are 30, 5, 5, 5, 1, respectively), C_{bk} is the environmental background value of a heavy metal, C_i is the actual test value of heavy metal i. The potential ecological risk levels of heavy metals in sediments are shown in Table S2.

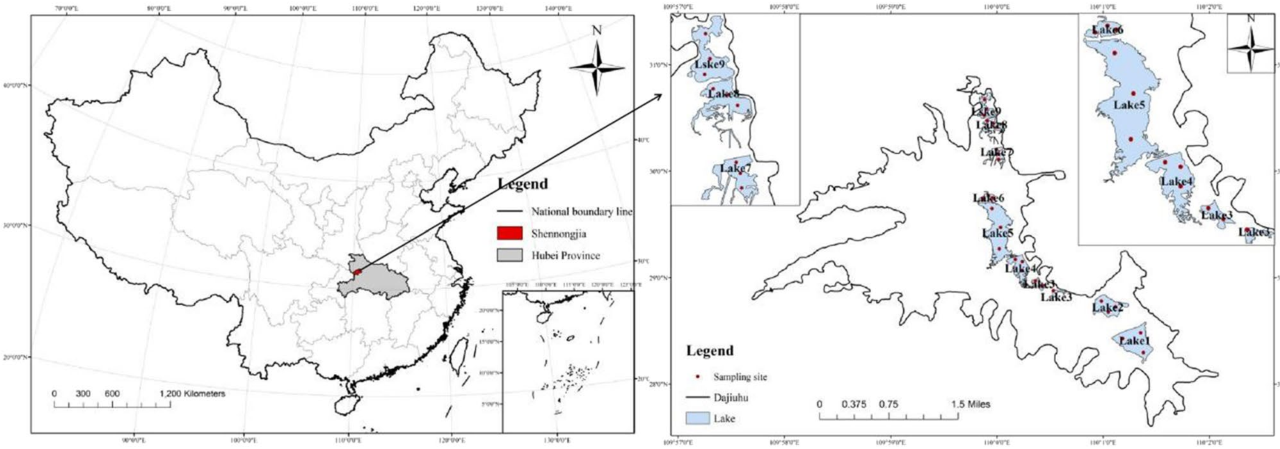

Fig. 1 Map showing the locations of the study area and sampling points

Geo-accumulation index

The geological accumulation index (I_{geo}) method (Müller 1969) is used as a quantitative index to evaluate the heavy metal pollution degree in sediments. In addition to the environmental background value and the influence of human activities, it also considers factors causing fluctuations in the environmental background value due to diagenesis. I_{geo} is the geological accumulation index, calculated by the following formula:

$$I_{\text{geo}} = \log_2\left[\frac{C_i}{K \times B_i}\right] \tag{3}$$

where C_i is the measured value of heavy metal i in sediments, B_i is the environmental background value of heavy metal i, and k is the modified index. Considering that rock geology in variability in different regions may cause a change in background value (Zahra et al. 2014), the general value is $k = 1.5$. The evaluation criteria for the pollution grade of the geological accumulation index method are listed in Table S3.

Risk assessment code

A risk assessment code (RAC) (Liang et al. 2017) was proposed as different heavy metals have different binding forces on sediments. The RAC value represents the proportion of extracted weak acid salts in the total heavy metal concentrations, used to evaluate heavy metal activation risk and migration and their potential to enter the food chain (Nemati et al. 2011). The evaluation criteria of the risk assessment codes are as follows: RAC < 1% corresponds to no risk, 1% < RAC < 10% to low risk, 11% < RAC < 30% to moderate risk, 31% < RAC < 50% to high risk, and RAC > 50% corresponds to extremely high risk respectively.

Pollution factor method

The pollution factor method is used to calculate the ratio of acid-soluble form, reducible form, and oxidizable form to the residual state, which is often used to describe the heavy metal's residence time and mobility of heavy metals in sediments. A greater C_f value corresponds to a shorter, residence time. Higher mobility represents a greater the potential threat to the environment, the stronger the mobility (Zhao et al. 2013). The pollution factor values are calculated according to the following formula:

$$C_f^i = C_{\text{bio}}/C_{\text{res}} \tag{4}$$

$$C_f = \sum_{i=1}^{n} C_f^i \tag{5}$$

where C_f^i is a single pollution factor; C_f is the comprehensive pollution factor; C_{bio} is the heavy metal concentrations after the first three steps of morphological analysis; C_{res} is the value of the fourth step (residual state) of a metal morphological analysis.

Quality control and statistical analysis

The teflon utensils and glass containers used in the experiment were soaked in 20% nitric acid solution for more than 24 h. The measurement samples were arranged with blank controls in batches to reduce the error. According to the number of experimental samples in each batch, 10 ~ 15% reference control samples were used. The relative standard deviation of the control samples did not exceed 10%. For Cd, Cr, Cu, Ni, Pb, and Zn detection, national reference materials were added for quality control, and the recoveries were 92.78 ~ 99.52%.

Pearson correlation analysis and principal component analysis were performed using SPSS 26.0 to investigate the correlations among the HMs (Deng et al. 2021). Maps of the study area locations and sampling sites were created using ArcGIS 10.2. Statistical graphs and figures were prepared using Origin 2021B.

Results and discussion

Concentrations and speciation characteristics of heavy metals in the sediments

The average content in the lake sediments of Dajiuhu of the six heavy metals was as follows: Zn > Cr > Ni > Pb > Cu > Cd. As the study location is a biodiversity conservation area and a national wetland park, it needs to meet the Level 1 standards of the Soil Environment Quality Risk Control Standard for Soil contamination of Agriculture Land of China (GB 15618–1995, Table 1). Pb concentration did not exceed the standard value at all sampling points. However, Cd exceeded the standard value at all sampling points, with its average content being threefold higher than the standard values (Fig. 2 and Table 1). Thus, Cd was the main heavy metal pollutant in the sediments of different areas (Xiao et al. 2019b; Hu et al. 2019). The average content of Zn exceeded the standard values, with only sampling points S3 and S27 being below the standards. The average values of the other elements were within the standard values, except for Cr and Ni, exceeding the standard values in S28 and S25 and Cu in S22 and S23 sampling points. Overall, the concentrations of heavy metals did not change significantly with the hydrological flow direction. Therefore, heavy metal content was affected by organic matter content, seasonal variations,

Table 1 Heavy metal contents in surface sediments of Dajiuhu Lake Wetland compared with the heavy metal contents in sediments of other high altitude lakes (mg/kg)

	Cd	Cr	Cu	Ni	Pb	Zn	Location	References
Dajiuhu Lake wetland, China	0.607	78.211	26.382	33.257	29.533	114.087	Hubei Province, China	This study
Caohai wetland	3.20	116	24	55	50	142	Yunnan-Guizhou Plateau, China	(Xia et al. 2020)
Ximen Co Lake	0.6	57.3	44.2	30.2	49.1	146.5	Tibetan Plateau, China	(Yuan et al. 2014)
Dianchi Lake	1.20	80.04	122.39	117.59	126.71	201.37	Yunnan Province, China	(Zhang et al. 2014)
Erhai Lake	1.10	103.8	63.1	52.2	47.4	109	Yunnan Province, China	(Lin et al. 2016)
Hamatai Lake	0.80	61.6	15.22	16.40	5.32	33.16	Nei Monggol Autonomous Region, China	(Liu et al. 2019)
Yangzonghai Lake	0.52	NA	NA	57.80	37.26	107.64	Yunnan Province, China	(Zhu et al. 2017)
Panchpokhari Lake	0.07	0.69	1.83	1.96	1.08	12.74	Langtang National Park, Central Nepal	(Raut et al. 2017)
BG*	0.11	54.0	20.0	22.0	22.0	65.0		
Class**	0.20	90	35	40	35	100		GB15618-1995

*The background (BG) value was taken from the average background values of chemical elements in stream sediments of China (2016)

**Class is the level 1 standard about the China Environmental Quality Standard for Soils (GB 15618–1995)

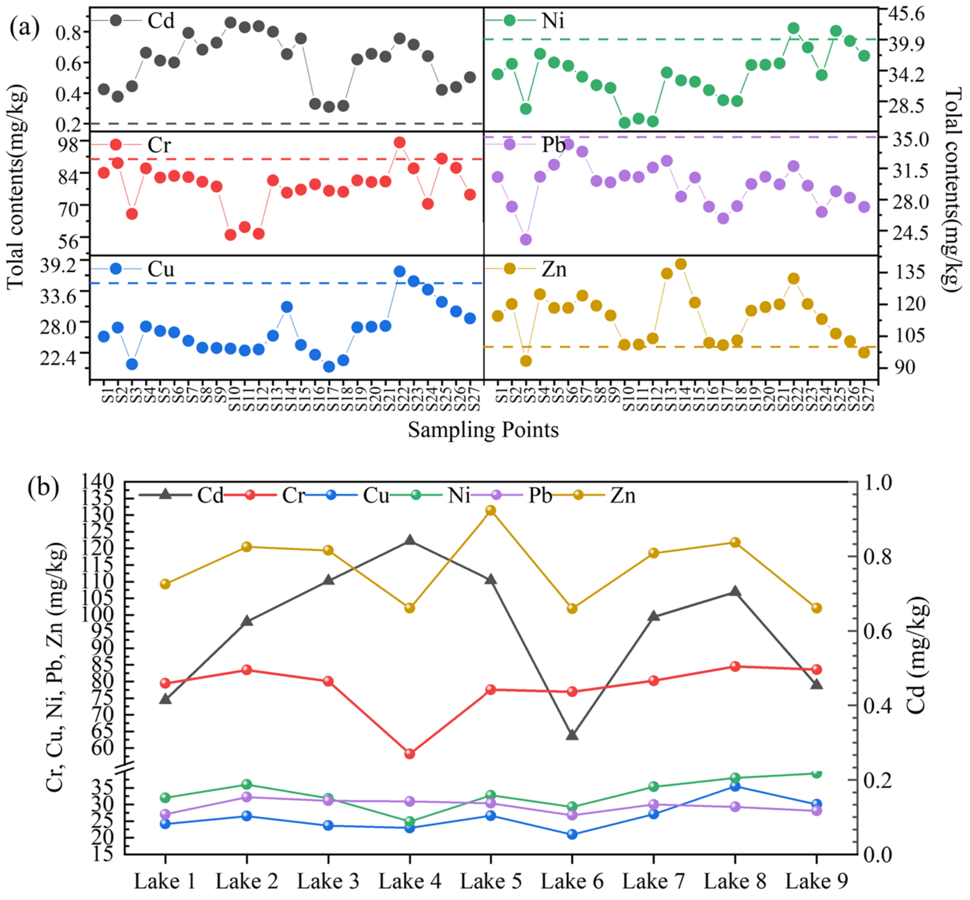

Fig. 2 Heavy metal contents in the sediments of Dajiuhu Lake. (**a**) The heavy metal content in each sampling points, the dotted line is the Environmental quality standard for soils (GB 15618–1995). (**b**) The average content of the heavy metals in each sub-lake (Lake 1–Lake 9)

pH changes, and redox conditions (Xiao et al. 2022). Additionally, external conditions change can affect heavy metal content in sediments when mineralization, desorption, dissolution, and release occur (Hu et al. 2019). Zn and Cr exhibited an obvious decreasing trend in Lake 4, and a similar decreasing trend was observed for Zn and Cd in Lake 6. Due to the changes in hydraulic conditions and human factors, the concentration of heavy metals may increase. Although the nine sub-lake systems of Dajiuhu are connected with ditches, the heavy metal content in lake water may change due to precipitation and surface runoff. Furthermore, heavy metals can easily combine with organic matter. Under such conditions, heavy metals' mobility will be reduced, resulting in a higher concentration of heavy metals in sediments (Xiao et al. 2019b).

The heavy metal contents in the sediments of high-altitude lakes in different regions are listed in Table 1. The heavy metal content of heavy metals in the sediments of Dajiuhu Lake was higher than that of Panchpokhari Lake. The contents of all heavy metals in the area of study were lower than those in Dianchi Lake. The heavy metal content in the wetland region directly connected to rivers was higher than in regions that were indirectly connected to rivers (Zhang et al. 2012), The Dajiuhu lakes are part of the wetland that is indirectly connected to rivers. Only Cu and Zn were higher than Caohai Wetland and Erhai Lake, respectively. In the Hamatai Lake, only Cd concentration was higher than the standard values. River or flood inputs can cause high concentrations of Cd in sediments (Tang et al. 2010a, b; Xiao 2019b). The average concentrations of Ni and Pb in Yangzonghai Lake were higher than those in Dajiuhu Lake Wetland; the Pb accumulation in sediments may be related to soil texture and metal mobility (Bai et al. 2011). The Cd, Cr, and Ni contents in the sediments of Dajiuhu Lake were higher than Ximen Co Lake. The differences in heavy metal contents in different research areas may be caused by the joint influence of natural factors and human activities (Xia et al. 2020). In addition, environmental conditions change, and the structure and source of heavy metals in different study areas might change, eventually affecting heavy metal enrichment.

The heavy metal morphology can affect their mobility and bioavailability (Huang et al. 2013). The persistence of heavy metals in sediments is mainly affected by morphology (Zhang et al. 2019). The forms of Cr, Cu, Ni, and Zn in the sediments of Dajiuhu were F4 > F3 > F2 > F1 (Fig. 3). The heavy metals were mainly in the residual form, indicating that they were stable under natural conditions with low migration and transformation ability (Chen et al. 2016). Only the form of Cr was the same across all sub-lakes and the whole lake. On the other hand, the distribution form of the other heavy metals was different in the sub-lakes compared to the whole lake. The metal ions in acid-soluble forms, such as Cr and Ni, are easily to be released from sediments and absorbed by aquatic organisms under certain conditions (Bi et al. 2007). However, Cr and Ni concentrations were comparatively low. Thus, Cr and Ni had a small impact on the ecological risk in the lakes of Dajiuhu. Furthermore, although the proportion of soluble acid form of Cu and Zn was small on overage, elevated concentrations were measured in Lake 3, Lake 4, and Lake 5. Thus, Cu and Zn may cause ecological risks to lakes in Dajiuhu.

The Cd form varied greatly in the different sub-lakes, but it was mainly in its oxidizable form in the whole lake. The activities of aquatic organisms and organic wastewater discharge produce oxidizable forms of heavy metals. Heavy metals migrating under strong oxidative conditions might underline a certain ecological risk of Cd in lake sediments. It should be noted that the ratio of Cd acid soluble form was higher compared to the other elements, especially in Lake 2, Lake 3, and Lake 4. In those lakes, Cd mainly occurred in acid-soluble form. In this form, Cd will generally be absorbed in clay and humus and then absorbed by aquatic plants through migration and transformation, endangering aquatic ecosystems (Wang et al. 2019). This can cause the greatest biological and ecological harm in the region.

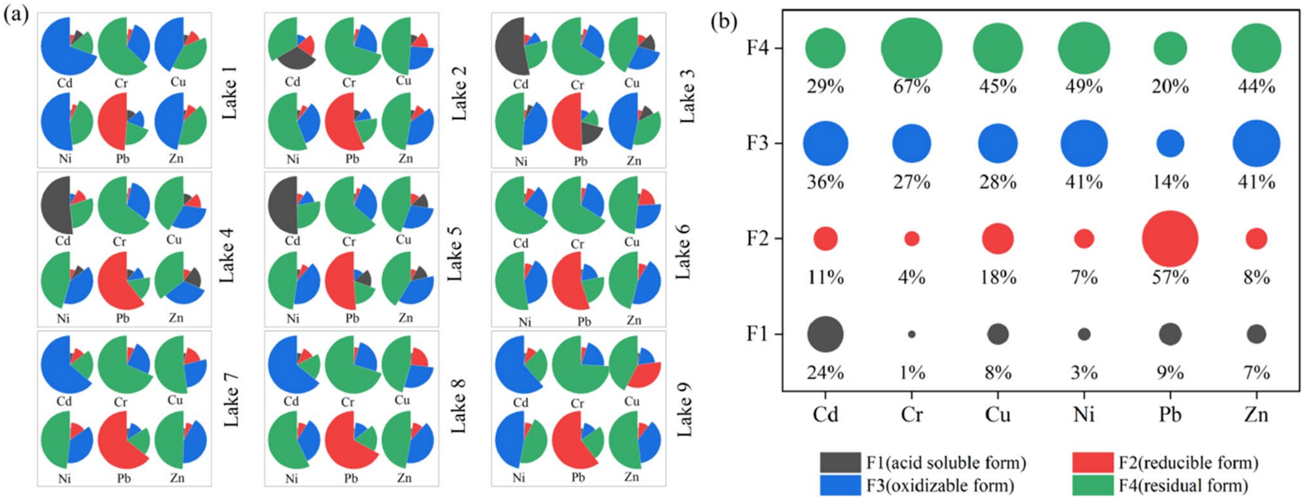

Fig. 3 The form of heavy metal in the sediments of Dajiuhu Lake. (**a**) The morphological distribution of different heavy metals in each sub-lake (Lake 1–Lake 9). (**b**) The average form distribution of the six heavy metals

The Pb form in Lake 3 and Lake 5 was different compared to the other regions (F2 > F4 > F3 > F1). Furthermore, the proportion of its reducible form was the highest (48.77 ~ 66.97%). As the reducible form can be easily converted to the acid-soluble form under anoxic conditions (Bi et al. 2007), and lake sediments are prone to anoxic conditions, Pb could have a negative environmental and ecological impact on Dajiuhu Lake. Additionally, the Pb acid soluble form content in Lake 3 and Lake 5 was 20.00% and 16.45%, respectively. This is also considered as high, thus having a great potential risk to the ecological environment.

Assessment of heavy metal pollution in sediments

The potential ecological risk index can more intuitively illustrate the ecological risk of various heavy metals in the different lakes through heat maps. According to the heat map, among the nine sub-lakes of Dajiuhu, Lake 4 (RI = 254.81) had the highest ecological risk, Lake 1 (RI = 141.55) and Lake 6 (RI = 113.28) had low ecological risk, while the other six sub-lakes (RI = 155.86 ~ 230.85) had moderate ecological risk. The ecological risks of the remaining heavy metals measured in the nine lakes were relatively lower. Therefore, Cd results in significant environmental risks in all lakes. The single potential ecological risks in Lake 1, Lake 6, and Lake 9 were 113.05, 86.61, and 123.72, respectively, which are considerable potential ecological risks. Meanwhile, the single potential ecological risks in the remaining six lakes ranged from 170.31 to 229.43, corresponding to high potential risks (Fig. 4). Cd is the main contributor to the potential ecological risk of heavy metals in Dajiuhu Lake that mainly comes from Cd, which is consistent with other studies on lakes in China (Lin et al. 2016; Wang et al. 2021). This is a result of Cd's highest toxicity coefficient even though its content is low.

The average I_{geo} index of the six heavy metals was Cd > Zn > Ni > Cr > Cu > Pb. The average pollution degree of Cd in the lake wetland sediments was moderate, ranging from mild to moderate pollution across different sampling points. This was similar to the findings in Lijiang and Huixian wetlands in China (Xiao et al. 2019a, b; Xiao et al. 2021). Cd in sediments usually corresponded to moderate to heavy pollution levels (Zhao et al. 2017). The average pollution degree in the sediment was mild, while some sampling sites were Zn and Ni pollution-free. In terms of Cr, Cu, and Pb, most sediment samples were pollution-free on average, with the exception of some samples being slightly polluted (Fig. 5a). Therefore, the patterns of heavy metal pollution differed across the lakes. Notably, Lake 8 and Lake 9 were polluted by five heavy metals to different degrees, and Lake 2 was polluted by four heavy metals (Fig. 5b). Pollution by multiple heavy metals can be influenced by many factors, such as urbanization, transportation, and industrial emissions (Asfandyar et al. 2020). It is speculated that although Lake 8 and Lake 9 are newly developed lakes, their hydrological flow may lead to serious sediment pollution compared to other lakes. Besides, Lake 8 is adjacent to provincial Highway 282, which can result in vehicle exhaust and other

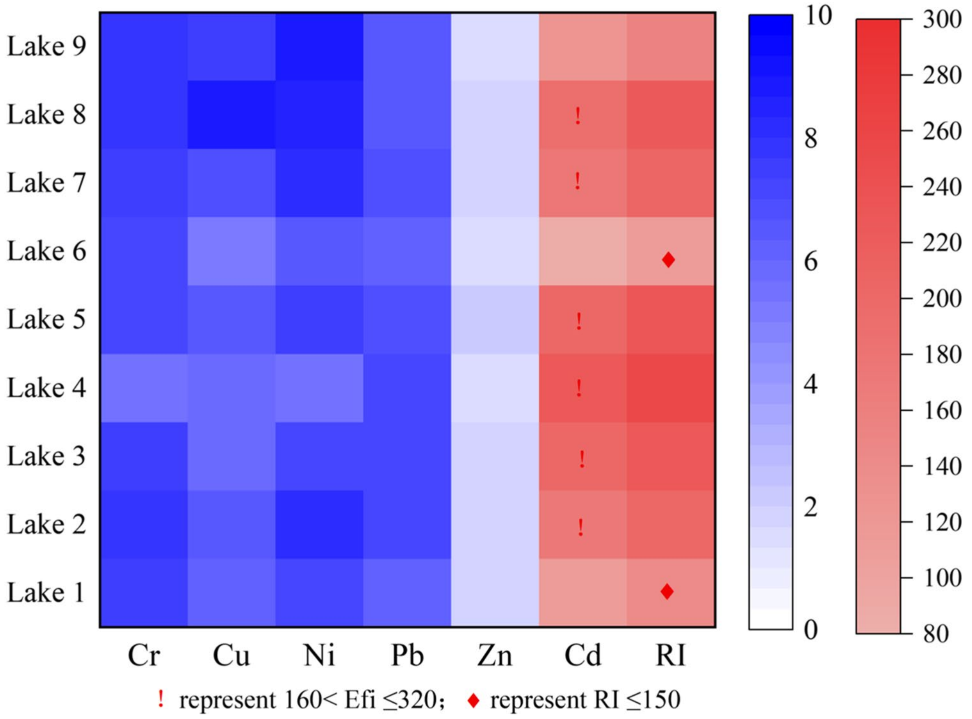

Fig. 4 The potential ecological risk index of heavy metal in each sub-lake of Dajiuhu Lake (Lake 1–Lake 9)

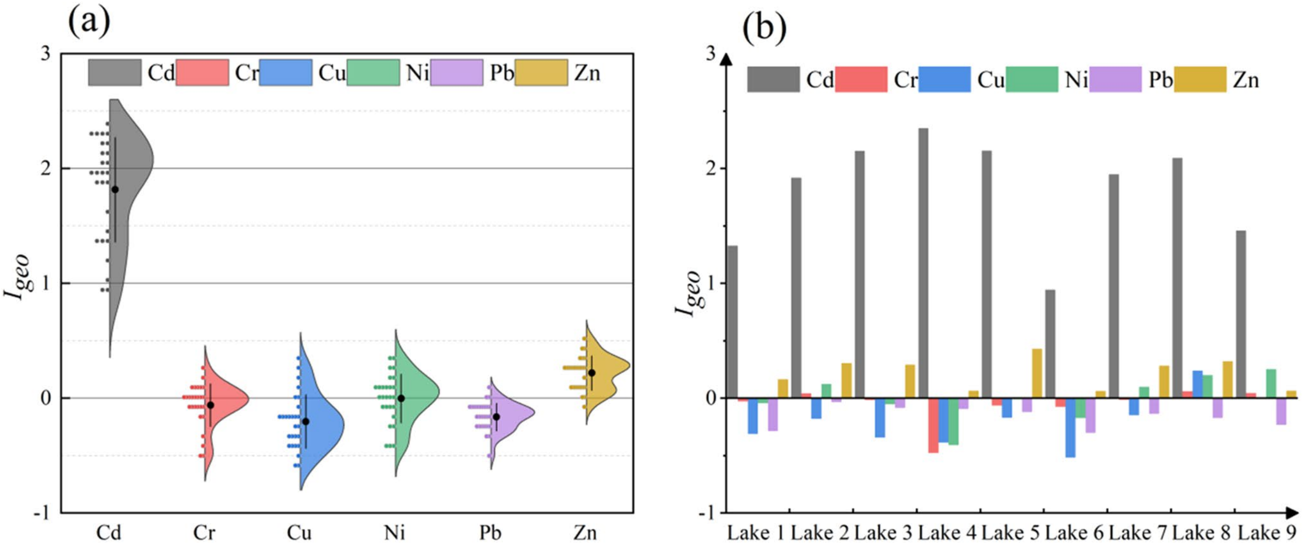

Fig. 5 The geological accumulation risk index of heavy metal in the sediments of Dajiuhu Lake. (**a**) The average value and value distribution of Igeo of different heavy metals in all sediment samples. (**b**) The mean value of Igeo of six heavy metals in different lakes (Lake 1–Lake 9)

man-made pollution. Furthermore, road dust can directly enter water bodies or sediments under precipitation and erosion, causing pollution and posing potential ecological risks to aquatic ecosystems (Wong et al. 2006). In addition, Lake 2 is in the proximity of a parking point for sightseeing vehicles, which might contribute to higher heavy metal pollution (Asfandyar et al. 2020). Residents mainly lived near Lake 2 before ecological migration. Thus, the more intensive human activities might have affected heavy metal concentration (Shahab et al. 2020; Deng et al. 2021).

The acid-soluble Form (F1) is the most unstable and potentially bioavailable form and can be assessed by the BCR sequential extraction method. Therefore RAC has been proven to be useful for assessing the mobility and potential release risk of heavy metals (Ke et al. 2017; Liang et al. 2017). The Cd ecological risk was higher than that of the other five heavy metals assessed. Cd exhibited a medium–high risk in the five sub-lakes, with Lake 3 exhibiting the highest risk. The ecological risk of Cu and Pb in the five sub-lakes was moderate. Ni exhibited a moderate risk only in Lake 4. The ecological risk of Zn was moderate in the three sub-lakes. The ecological risk of Cr in all lakes was low (Fig. 6a). Furthermore, heavy metals such as Cu, Cd, and Zn were more bioavailable and more

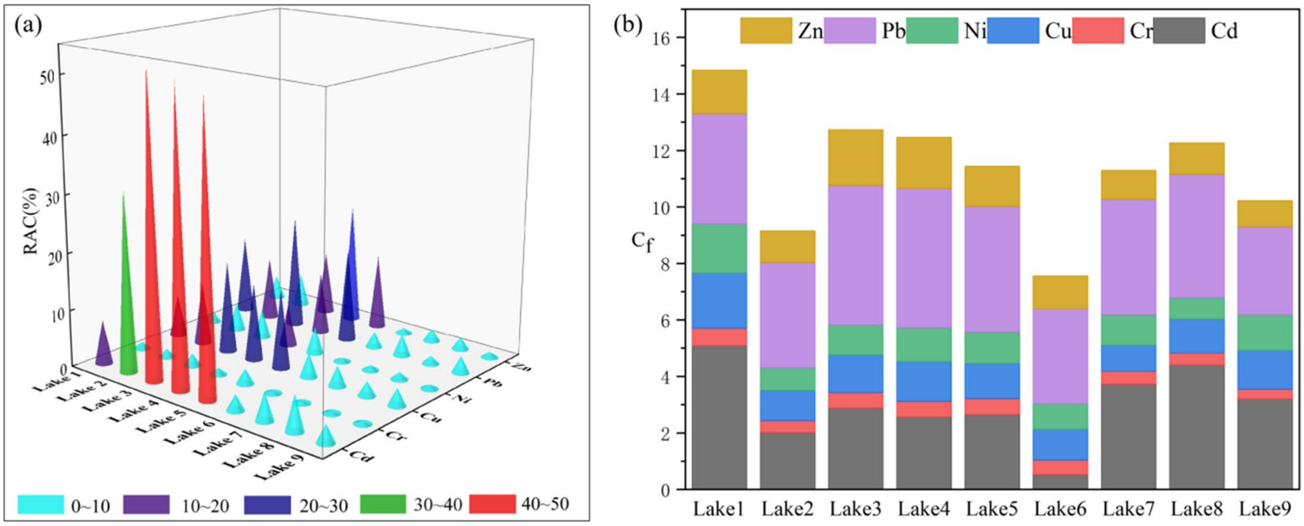

Fig. 6 Risk assessment of heavy metals in each sub-lake of Dajiuhu Lake based on forms. (**a**) The risk assessment code of heavy metal in the sediments of sub-lakes (Lake 1–Lake 9). (**b**) The pollution factor of heavy metal in the sediments of sub-lakes (Lake 1–Lake 9)

likely to be released from sediments into the water (Xu et al. 2016). Compared with other lakes, Lake 2 to Lake 5 exhibited a high potential bioavailability and high bioavailability risk. These four lakes are close to the highway based on their geographical location. At the same time, cruise ships can be found Lake 5. In contrast, Lake 6 to Lake 9 are far away from the scenic spot and less disturbed by tourism activities.

The pollution factor method can intuitively determine the heavy metal migration ability of sediments in Dajiuhu sediments. The stronger mobility of Cd and Pb in sediments relative to other heavy metals indicates that they are easily released in their residual form and can easily enrich the biological chains, thus being more harmful to the environment. Cr migration capacity and activity were the weakest among the six heavy metals, indicating that its potential pollution and environmental and ecological risks are low, consistent with the RAC evaluation results. Furthermore, although Cu, Ni, and Zn were less active, their migration capabilities are still considerable. Thus, their pollution risk should be further monitored. Among the different sub-lakes, the migration capacities ranged as follows: Lake 1 > Lake 3 > Lake 4 > Lake 2 > Lake 5 > Lake 7 > Lake 9 > Lake 2 > Lake 6. There were no significant differences observed in the migration capacity of the other seven lakes except for Lake 2 and Lake 6 (which were mainly affected by Cd and Pb) (Fig. 6b). Therefore, considering all our results, Lake 6 was the least polluted lake with the lowest ecological risk.

All the four assessment methods showed that Cd was the most serious heavy metal pollutant in the sediment of Dajiuhu lakes posing significant ecological risks. This was consistent with the results of numerous previous studies (Vu et al. 2017; Zhang et al. 2018). The high ecological risk of Cd indicates that it is more easily transferred in sediments and that human factors such as agriculture may be contributors to such risk of Cd (Yang et al. 2017). Furthermore, the pollution factor method indicated that Pb carried a significant environmental and ecological risk. This is because the pollution factor method takes into account various heavy metal forms, while RAC only considers the acid-soluble form. Therefore, we showed that the form of heavy metals in sediments plays a key role in their environmental effects, and the environmental effects of heavy metals were different due to different forms (Sastre et al. 2004). Therefore, the heavy metal weight cannot be regarded as the only criterion for evaluation of their environmental and ecological impact. In addition, previous studies have shown that different pollution and risk assessment methods result in inconsistent results (Xu et al. 2016). However, according to the nine sub-lakes, Lake 6 was the least polluted Lake with the lowest ecological risk.

Source analysis of heavy metals in sediments

Correlation analysis is often applied to study the heavy metal sources in sediments. The higher the correlation between heavy metal content, the higher the probability of a common source (Li et al. 2013). We found that there was a strong correlation between Cr, Cu, and Ni in the samples, with the correlation coefficient between Cr and Ni being 0.88. In samples from similar sources or transportation routes, the heavy metals will show a significant correlation (Cao et al. 2015; Xiao et al. 2019a, b), indicating that Cr, Cu, and Ni originated from a similar source. Notably, Cr, Cu, and Ni concentrations were not significantly different from the background values, indicating that they were mostly derived from weathering of rocks and parent materials and deposited in lake sediments (Xu et al. 2016). However, anthropogenic pollution such as fertilizers, traffic, and fossil fuel combustion could still cause pollution of Pb, Zn, and Cu in sediments through rainwater transportation (Tang et al. 2010b). The correlation between Cd and other heavy metals, except Pb, was weak, indicating that the sources of Cd were different compared to the other heavy metals (Fig. 7a).

Principal component analysis (PCA) can reveal the relationship and similar patterns between different elements by groups clustering across principal components (PC). Both correlation and principal component analyses can be used to trace the source of heavy metals (Han et al. 2006; Karim and Qureshi 2014; Zhang et al. 2016). Through the analysis of the two main principal components with a cumulative contribution rate of more than 80.8%, the contribution rate of the first main factor (PC1) was 49.5%, which mainly corresponded to Ni, Cr, Cu, and Zn. This indicated that the sources of the four heavy metals were similar and may have come from natural or anthropogenic causes (Han et al. 2006; Martín et al. 2006). Moreover, the four elements exhibited high correlations according to Pearson correlation analysis (Fig. 7b), indicating that they originated from similar sources. Our previous evaluation results showed Ni, Cr, Cu, and Zn had low pollution and ecological risks. Although the erosion was slow, the contents of Ni, Cr, Cu, and Zn in the strata flow into the sediments were high. When the heavy metal enrichment degree is not obvious, it indicates that they mainly originate from the natural geology, such as the weathering and deposition of parent rock (Chen et al. 2015; Xiao et al. 2019a, b). Different studies have confirmed that high concentrations of Ni and Cr are related to natural causes (Al-Hashim et al. 2021; Heidari et al. 2021; Xiao et al. 2019a, b). So PC1 reflected the source of medium metals under natural conditions.

The contribution rate of the second major factor (PC2) was 31.3%, which corresponded to Cd, Pb, and Zn. Zn may originate from the discharge of galvanizing industry. It is also commonly used in the production of batteries, magnetic

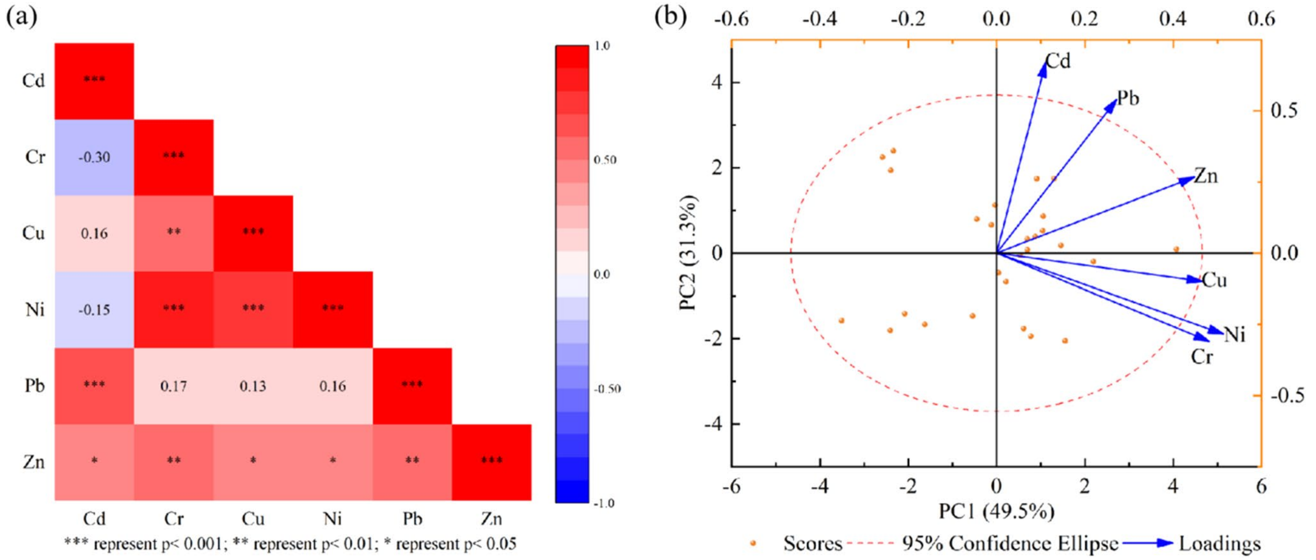

Fig. 7 The source analysis of heavy metal in the sediments of Dajiuhu Lake. (**a**) The correlation analysis of heavy metals in lake sediments, where red represents positive correlation and blue represents negative correlation. (**b**) The principal component analysis of heavy metals in lake sediments

materials, alloys, and pigments (Liu et al. 2019). Cd and Pb can originate from industrial wastewater, waste residue and mineral exploitation, wastewater generated by the electronics industry, automobile exhaust, vehicle tire friction, and ship navigation (Al-Khashman 2004). Cd discharge is also associated with metallurgy and casting industries (Islam et al. 2018; Liu et al. 2019). Based on the analysis of the geographical location analysis of Dajiuhu and the surrounding industrial profile, it can be concluded that there is no direct industrial pollution source in close proximity. Previous studies have shown that atmospheric deposition is the main source of heavy metals in Dajiuhu due to the fading of East Asian monsoon and heavy rainfall (Deng et al. 2014; Liu et al. 2021; Zhu et al. 2017), Similarly, in the Huixian wetland, atmospheric deposition is the second most important source of heavy metals (Lu et al. 2014a, b). Therefore, pollution sources caused by industrial production can only be deposited in lakes through atmospheric deposition or surface runoff formed with rainwater. Cd may be linked to traffic emissions and fossil fuel combustion (Heidari et al. 2021), similarly to Pb (Asfandyar et al., 2020). According to the sampling observation, sightseeing cars and cruise boats were prevalent in the touristic locations. Moreover, Lake 7 and Lake 8 were close to the provincial road, with a large traffic flow. Thus, tourism and traffic could be significant pollution sources (Chen et al. 2012; Xiao et al. 2021). Cd was a common heavy metal pollutant in fertilizers and pesticides, and Cd pollution in lake sediments was more severe than other heavy metals. Thus, it might be related to the early agricultural economic development of wetlands. Excessive use of pesticides and phosphate fertilizers before ecological migration caused non-point source Cd pollution (Wang and Zang 2014). This potentially lead to heavy metal pollution and enrichment along with other anthropogenic pollution (Lu et al. 2014a, b). Heavy metal pollution caused by agriculture in the twentieth century was concentrated in lake sediments due to rainfall. Therefore, PC2 reflected the anthropogenic pollution sources caused by atmospheric deposition, tourism, agriculture, and transportation. Other studies have similarly reported higher Cd, Pb, and Zn concentrations mainly due to anthropogenic sources (Dai et al. 2018; Nguyen et al. 2020; Zhang et al. 2018).

Conclusions

In this study, six heavy metal elements (Cr, Cd, Cu, Zn, Pb, and Ni) in sediments from nine lakes of the Dajiuhu wetland were studied. The main conclusions of this study were as follows: (1) The Pb content in the lake sediment samples of Dajiuhu did not exceed the national standard value. In contrast, Cd contents were above the national standard value in all samples. Cr, Cu, Zn, and Ni content exceeded the national standard values only in certain sampled areas. (2) Among the six heavy metals, Cd had the highest pollution potential and, together with Pb, was the main influencing element with a high potential ecological risk in the lake wetland of Dajiuhu. Lake 6 had the lowest pollution degree and ecological risk index among the nine lakes. In comparison, the remaining eight lakes were polluted to a certain extent or had certain ecological risks. (3) Ni, Cr, Cu, and Zn accumulation in sediments of lakes in Dajiuhu were a result of natural conditions. On the other hand, Cd, Pb, and Zn in sediments were mainly assumed to accumulate due to atmospheric deposition, traffic pollution, tourism pollution, and agricultural pollution. There are few studies on heavy metals' overall distribution and biotransformation process in Dajiuhu wetlands, and our study provides important insights into such processes and mechanisms, which can further be investigated in future studies.

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Author contribution All authors contributed to the study conception and design. Material preparation, data collection, and analysis were performed by JW, CY, YG, and YG. Project administration, funding acquisition, and supervision were performed by JG. Writing—review was performed by XY, DC, ZL, and SY. The first draft of the manuscript was written by JW, and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

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Data availability The datasets used and/or analyzed during the current study are available from the corresponding author on reasonable request.

Declarations

Ethical approval Not applicable.

Consent to participate Not applicable.

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