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Increasing trends in heavy metal risks in the Caohai Lake sediments from 2011 to 2022

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A B S T R A C T

The accumulation of heavy metals (HMs) in lakes can pose a serious risk to aquatic ecosystems. Caohai Lake is the largest plateau freshwater lake in Guizhou Province, China. However, the spatial and temporal dynamics and ecological risks of HM contamination in Caohai sediments in recent years remain unclear. Here, we analyzed the HM content of sediments in Caohai Lake in 2022 and collected historical (2011–2021) data from published articles to explore the spatiotemporal distribution characteristics, toxicity risks and potential sources of HMs. The mean concentrations (2011–2022) of Ni, As, Cd, Zn, Hg and Pb in the Caohai Lake sediments were all higher than the background values, and the values of As (32.0 mg/kg), Cd (14.19 mg/kg), Pb (87.6 mg/kg) and Zn (406.2 mg/kg) were 1.9, 4.0, 2.4 and 1.3 times their probable effect level (PEL) values, respectively. Sediment As and Cd concentrations in Caohai Lake showed significant decreasing and increasing trends from 2011 to 2022, with mutation points occurring in 2018 and 2020, respectively. The Caohai Lake sediment aquatic life risk index (TRI) for HMs showed a gradual increase until 2019, when it reached a sudden change point, leading to a very high toxicity risk in 2020, with Cd contributing the most to the TRI (43.66). Cd, Pb and Zn, the main pollutants in the sediments of Caohai Lake, originate from traffic, mining and agricultural sources. Our integrated results demonstrated the severity of HM pollution in Caohai Lake and can be helpful for formulating reasonable pollution prevention and control measures.

1. Introduction

Lake sediments are an important part of aquatic ecosystems and a key indicator of aquatic environmental quality (Jara-Marini et al., 2008). In recent years, heavy metal (HM) pollution, which causes the quality of the aquatic environment to deteriorate, has become an increasingly prominent problem that has attracted much attention (Karaouzas et al., 2021; Li et al., 2021a; Xie et al., 2022). HMs are highly toxic, nondegradable and easily enriched and can enter organisms through the food chain, thus threatening lake ecosystems and human health (Hu et al., 2016). HM pollution in lake sediments has received much attention, with the main research directions including the temporal and spatial distribution characteristics of HM contents, occurrence forms, ecological toxicity risk assessment, and source apportionment (Liu et al., 2014; Hillman et al., 2015; Cheng et al., 2022). HM contamination in sediments has become more complex, shifting from monotypic to complex contamination scenarios; As, Cd, Cu, Hg, Pb, Ni and Zn are the most studied HMs (Hu et al., 2013a).

The current methods for evaluating the levels of and potential risks from HM contamination in sediments include the geoaccumulation index method, toxicity risk index, hazard index, potential ecological risk index method and secondary phase compared with the primary value method (Pekey et al., 2004; Muller, 1969; Hakanson, 1980; Chernoff, 1973). The geoaccumulation index method considers not only human influences but also geochemical background values and the influence of diagenesis on the background values; therefore, the Igeo index is more objective for assessing the ecological risk of HMs (Loska et al., 2004). The toxicity risk index (TRI), which combines the threshold level (TEL) and the probable effect level (PEL), is more suitable for assessing toxicity risk, as it considers both the acute toxicity and persistent chronic toxicity of the contaminant of interest; thus, it is more reliable for assessing HM-related health risks (Zhang et al., 2016; Li et al., 2019a). In addition, other studies consider HM sources and sinks, thus integrating pollution sources and sediment properties associated with sediment risks. The sediment source aggregation class (SLISA-SCA) was used to divide the lake into different risk control areas, providing a more objective spatial

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view of HM risk control (Li et al., 2022). Because lakes play an important role in providing water, recharging groundwater and regulating the climate, a study of the distribution characteristics and ecological risks associated with HMs in lake sediments can serve as a reference for obtaining safe drinking water from lakes (Messager et al., 2016).

Caohai Lake is a typical karst plateau freshwater lake in Guizhou Province and an important wetland in the National Wetland Protection Plan. In recent years, industrial and agricultural development around Caohai Lake and domestic sewage discharge have severely damaged the stability of the Caohai wetland ecosystem (Hu et al., 2020). A survey of plants in the coastal wetlands of Caohai Lake found that the average content of Cd in the plants was 1.9 mg kg^{-1}, which was outside the normal range (0.2–0.8 mg kg^{-1}). Some plants also had phytotoxic levels (100–400 mg kg^{-1}) of Zn (specifically 125 mg kg^{-1}) (Xia et al., 2021), and HMs in sediments were a primary source of HMs in plants. Studies on HMs in Caohai Lake sediments have been widely reported in the last decade. For example, a study in 2021 (Zhu et al., 2021) found that the mean Cd concentrations in Caohai Lake sediments was 33 times higher than those in a study from 2013 (Zhang et al., 2013). Zhu et al. (2021) and Zhao et al. (2019) found that Cd was the main HM related to potential ecological hazards in Caohai Lake, that the carcinogenic risk exceeded acceptable levels and that it was the main element associated with the carcinogenic risk. In addition, the concentration of Zn in Caohai Lake sediments was 1.8 to 8.8 times the background value, representing heavy pollution (Hu et al., 2017). In addition to anthropogenic factors, HM inputs are influenced by topography, season, and environment (pH and redox potential), and lake sediment HM concentrations can show spatial and temporal variations (Zhu et al., 2021). As evidenced by the above studies, many scholars have studied HMs in Caohai Lake sediments. However, in the context of increasing human activities, the temporal and spatial dynamics of HMs in Caohai Lake sediments are still unclear. Therefore, an integrated analysis of HM pollution in the Caohai Lake sediments is needed.

Integrative analysis is a statistical method used to comprehensively analyze large amounts of analytical data from numerous studies and integrate the results (Moher et al., 2015). Integrative analysis was first used in medicine and is often used as a reference in clinical treatment decisions (Simpson and Pearson, 1904). In recent years, integrative analysis has also been applied to environmental pollution data to enable a comprehensive analysis of pollution. For example, Li et al. (2021a) assessed the ecological risk of HMs in Chinese lake sediments by collecting published literature analysis, and Niu et al. (2020) evaluated the contamination and sources of HM contamination in surface sediments of Lake Taihu in the last 20 years. Duan et al. (2016) integrated published data and analyzed the spatial distribution characteristics of HMs in Chinese agricultural soils. However, as integrative analysis has not developed into a unified analytical process for environmental pollution assessment, the method of collecting and reanalyzing pollutant data from relevant publications is currently more commonly used to characterize the pollution status of the study area.

The integration and analysis of published data is important for studying environmental pollution when continuous monitoring data are not available (Niu et al., 2020). As the largest plateau freshwater lake in Guizhou, China, Caohai Lake has suffered from the degradation of its submerged plants in recent years; an ecological risk assessment of Caohai Lake can provide a reference for its ecological restoration. In this study, due to the lack of long-term continuous monitoring in Caohai Lake, integrated analyses are proposed for assessing the spatial and temporal characteristics of heavy metals in the sediments of Caohai Lake, as well as their potential risks and sources. We integrated the historical data of HMs in Caohai Lake sediments and analyzed the spatial and temporal variation characteristics of HMs via spatial interpolation and the Mann–Kendall method. In addition, we analyzed the potential risk of HMs in the Caohai Lake sediments via the index of geo-accumulation (Igeo) and the potential ecological risk method and analyzed the sources of HMs in the sediments via principal component analysis (PCA). The research results can provide a reference for the prevention and control of HM pollution in Caohai Lake.

2. Materials and methods

2.1. Study area

The Caohai Basin is located in southwestern Weining County, Guizhou Province (104°10′–104°20′E, 26°47′–26°52′N), which is part of the Yangtze River Basin. The water supply mainly comes from precipitation and groundwater. The total area of the basin is 96 km^2, with an average altitude of 2172 m. With a water area of 25 km^2, Caohai Lake is the largest natural freshwater lake in Guizhou Province and one of the three largest highland lakes in China, with an average water depth of no more than 1.5 m. The average annual precipitation in Caohai Lake is 950 mm, the average annual sunshine is 1805 h, and the annual average temperature is 10.5 °C, corresponding to a subtropical plateau monsoon climate (Zhang et al., 2013). The Caohai Lake wetland is rich in plant species and is one of the main wintering habitats and resting places for migratory birds in the southwest; black-necked cranes and gray cranes come to the region every winter to feed (Xia et al., 2021).

2.2. Data collection

The Web of Science, Google Scholar and China National Knowledge Infrastructure (CNKI) databases were used to collect data on HMs in the Caohai Lake sediments. The search terms used in these databases were “heavy metals”, “sediment” and “Caohai Lake” or “plateau lake” or “karst lake” in Guizhou, and the searches covered studies from 2011 to 2020 (Zhang et al., 2013, 2014; Song et al., 2016; Liu et al., 2016; Yin et al., 2020; Peng, 2018; Hu et al., 2017; Wu et al., 2019; Xia et al.,2020; Zhao et al., 2019; Fan et al., 2021; Fan, 2021; An et al.,2022; Zhu et al., 2021). A total of 56 publications and 153 data records from 14 publications were selected (Fig. S1, Table S1). In 2022, we collected 15 sediment samples from Caohai Lake for the analysis of HM concentrations; therefore, a total of 168 points of HM data were obtained for this study (Table S1). The following criteria were used to screen data from the literature: (1) the selected publications involved surveys of surface sediments throughout Caohai Lake; (2) the studies included a clear number of survey points, coordinates, HM concentrations and periods for the survey; and (3) the digested sediments were analyzed by single or mixed acid digestion with strict quality control and assurance. The greater the amount of data collected for each element was, the more representative the calculated concentrations. An element could have been investigated several times during the same year, so we used a weighted average to represent the concentration values for that year.

2.3. Sediment sampling and analysis

To obtain the latest HM concentration data for the Caohai Lake sediments, we set up 15 sampling sites in Caohai Lake in March 2022. Surface sediments were collected in situ by using a grapple bottom sampler, preserved at low temperatures and transported to the laboratory for freeze-drying. Approximately 0.05 g of sediment sample was weighed into an ablation tube, 5 mL of an ablation reagent (HF:H_2O_2:HNO_3 = 2:1:1) was added, and then the sample was placed in a microwave ablator (Ethos UP, Milestone, Italy) for 4 h. After cooling, 1 mL of the supernatant was collected and diluted to 10 mL. The HM concentration was determined using an inductively coupled plasma emission spectrometer (ICP–OES, iCAP6000, Thermo Fisher, USA). Analytical precision was monitored using standard reference material (GSB04-1767-2004). Each sample was analyzed three times, and the recovery of each HM was calculated. The recovery of HMs in this study ranged from 88.7 % to 112.4 %.

2.4. Assessment method

2.4.1. Contamination assessment—I_{geo}

I_{geo} is a geochemical indicator proposed by Muller (1969) and used to identify the impact of anthropogenic activities on the environment via a comparison of present HM concentrations with preindustrial period ones (Praveena et al., 2008). It was calculated as follows:

$$I_{geo} = \log_2\left(\frac{C_n}{1.5 \times B_n}\right) \tag{1}$$

where C_n is the measured content of HMs, mg kg^{-1}; B_n is the geochemical background.

value of the HM, and a constant of 1.5 represents natural fluctuations in the environment and very small human activity influences (Loska et al., 2004; Lu et al., 2009). The specific classification of I_{geo} is shown in Table S2.

2.4.2. Aquatic life risk—TRI

The toxicity risk assessment of contaminants consists of two components, the TEL and PEL, and the TRI, which combines the TEL and PEL; it widely used to assess the potential toxicity risks of HMs to aquatic organisms. In this study, the TRI for sediments was calculated using TEL and PEL values from the literature (Li et al., 2019b):

$$\mathrm{TRI} = \sum_{i=1}^{n} TRI_i = \sqrt{\frac{\left(S_s^i/S_{PEL}^i\right)^2 + \left(S_s^i/S_{TEL}^i\right)^2}{2}} \tag{2}$$

where S_s^i is the sediment HM content (mg kg^{-1}), S_{TEL}^i is the TEL value and S_{PEL}^i is the PEL value for each metal (mg kg^{-1}). The TRI consists of 5 grades or classes based on Gao et al. (2018): none (<5), low (5–10), moderate (10–15), considerable (15–20) and very high toxicity risk (>20).

2.5. Data statistics processing

2.5.1. Trend analysis

The Mann–Kendall statistical test is a nonparametric statistical test that has been recommended and is widely used by the World Meteorological Organization (WMO) to test the significance of both time series trends and mutation tests (Li et al., 2021b). The Mann–Kendall test does not require the sample to follow a certain distribution and is not affected by a few outliers, has a high degree of quantification, and has a wide range of detection; it can be used to test the significance of trends in time series as well as for abrupt change analysis. Assuming that the time series of HMs are x_1, x_2…, x_n, the basic principle of the Mann–Kendall trend test can be expressed as follows (Gocic and Trajkovic, 2013; Huang et al., 2022):

$$s_i = \sum_{i=1}^{n-1} \sum_{j=i+1}^{n} sgn\left(x_i - x_j\right) \tag{3}$$

$$sgn\left(x_i - x_j\right) = \begin{cases} 1, & x_i - x_j > 0 \\ 0, & x_i - x_j = 0 \\ -1, & x_i - x_j < 0 \end{cases} \tag{4}$$

$$Var(S) = \frac{n \times (n-1)(2n+5)}{18} \tag{5}$$

$$Z_c = \begin{cases} \dfrac{S-1}{\sqrt{Var(s)}}, & \text{if } S > 0 \\ 0, & \text{if } S = 0 \\ \dfrac{S+1}{\sqrt{Var(s)}}, & \text{if } S < 0 \end{cases} \tag{6}$$

$$\beta = median\left(\frac{x_j - x_i}{j - i}\right) \ \forall 1 \le i \le j \le n \tag{7}$$

where s obeys a normal distribution, sgn is the sign function and β is the slope. Within the confidence interval, the trend is significant if $|Z_c| \ge Z_{(1-\alpha/2)}$. If $\beta > 0$, which indicates an upward trend of climate elements and vice versa.

The Mann–Kendall abrupt change test is calculated as follows (Gocic and Trajkovic, 2013; Huang et al., 2022):

$$d_k = \sum_{i=1}^{k} y_i \ \ (1 \le k \le n) \tag{8}$$

$$r_i = \begin{cases} 1, & x_i > x_j \\ 0, & x_i \le x_j \end{cases} \quad (j = 1, 2, \cdots, i) \tag{9}$$

Under the assumption of random independence of the time series, we define the statistic as follows:

$$UF_k = \frac{d_k - E(d_k)}{\sqrt{Var(d_k)}} \ \ (1 \le k \le n) \tag{10}$$

where$UF_k = 0$, and $E(d_k)$ and $Var(d_k)$ are the mean and variance of d_k, respectively, where x_1, x_2…, x_n are independent of each other and have the same continuous distribution. They can be calculated by the following equations:

$$E(d_k) = \frac{k(k-1)}{4} \tag{11}$$

$$Var(d_k) = \frac{k(k-1)(2k+5)}{72} \ \ (1 \le k \le n) \tag{12}$$

After the abrupt change test, the UF_i and UB_i graphs were plotted. If $UF_i > 0$, then the series trends upward and vice versa; when UF_i and UB_i exceed the confidence interval, there is a clear upward or downward trend. If the two curves intersect and the point of intersection is between the confidence intervals, then the point of intersection can be regarded as the abrupt change point of series x. Additionally, to calculate the effect of year on the variation in the HM content, we used linear regression analysis to explore the correlation between the year and the HM content. The Mann–Kendall statistical test was performed using the "cpm" package in R statistical software (version 4.2.2).

2.5.2. Spatial analysis

The inverse distance weighting method is a type of exact interpolation, where the weighted average is calculated by using the distance between the interpolated point and a known point as a weight so that the closer the sample point is to the interpolated point, the greater the weight given (Chaplot et al., 2006). The basic principle of the inverse distance weighting method is expressed as follows:

$$Z(x) = \frac{\sum_{i=1}^{n} w_i(x) z_i}{\sum_{j=1}^{n} w_j(x)} \tag{13}$$

$$w_i(x) = \frac{1}{d(x, x_i)^p} \tag{14}$$

where $Z(x)$ is the estimated value of the interpolated point x; z_i is the concentration value of a known sampling point; n is the number of known points used in the interpolation process; w_i is the weight value; d is the distance between the known point x_i and the interpolated point x; and p is the power of the distance (the general value is 2), with p significantly affecting the accuracy of the interpolation results (Habibi et al., 2017). Based on the principles of proximity and similarity, when the distance between the 2 interpolation points in study is shorter, their properties are more similar, and conversely, if they are farther away, they are less similar. Therefore, the values of the interpolated points in the study area are doubly influenced by the values of the sample points in the area and their weights. In this study, ArcGIS 10.2 was used to map the distribution of sediment sampling sites and perform spatial interpolation analysis of HMs in Caohai Lake.

2.5.3. Source analysis

We identified potential sources of HMs in Caohai Lake sediments by using PCA, a multivariate statistical method that applies projections to downscale the data (Thurston and Spengler, 1985; Chow and Watson, 2002). Furthermore, we assumed that these common factors lead to covariation of apparent variables, thus enabling the identification of potential structures and extraction of common factors from the observed items that lead to variable covariance (Hou et al., 2014). Prior to data analysis, we ensured that the Kaiser-Meyer-Olkin (KMO) value was greater than 0.5, that Bartlett's sphericity test was significantly less than 0.05 and that the eigenvalues of the extracted principal component factors were all greater than 1.

3. Results and discussion

3.1. Concentration and distribution

3.1.1. Concentration

The historical mean contents of Ni, As, Cd, Zn, Hg and Pb in the Caohai Lake sediments were 1.04, 1.6, 21.5, 3.7, 2.5 and 4.1 times the background values, respectively (Table 1), and the average concentrations of Cr and Cu were below their background values (CEMS, 1990). The coefficient of variation (CV) of Cd was the largest (113.7 %), those of Cr, Hg, Pb, Ni and Zn ranged from 55.5 to 78.7 %, and those of the other elements ranged from 11.9 to 37.2 %. This result indicated that there was strong spatial variation in the Cd concentration. As, Cd, Cr, Hg, Pb, Ni and Zn exceeded the threshold effect level (TEL), and Cr, Hg and Ni were above the TEL but below the probable effect level (PEL); As, Cd, Pb and Zn were above the PEL, with values 1.9, 4.0, 2.4 and 1.3 times greater than the PEL, respectively (Ma et al., 2013; MacDonald et al., 2000).

Caohai Lake is in the lake region of the Yunnan-Guizhou Plateau; this region of Yunnan Province has many other plateau lakes, of which Dianchi, Erhai and Yilong Lakes are representative. We collected HM data from the sediments of Dianchi Lake (Table 1). The concentration of HMs in the sediments of Dianchi Lake exceeded the background values for soil elements in Yunnan Province; the data also indicated a relatively high enrichment of HMs in Yunnan Dianchi Lake (CEMS, 1990). The average Cd content of the Caohai Lake sediments was 5.65 times higher than that of Dianchi, but all other element concentrations were lower than those of Dianchi (Liu, 2012; Meng, 2016). These changes in HM concentrations began with rapid urbanization and industrialization in the 1970 s; by 2014, the resident population in the basin reached 4.06 million. The long-term discharge of domestic and industrial wastewaters may have led to HM sediment enrichment (Liu, 2012; Meng, 2016). Compared with Dianchi Lake, the Caohai Lake watershed is less densely populated but is impacted by pollutant emissions from agriculture and industrial sources.

The sediment As, Cd, Pb and Zn contents of Caohai Lake were all higher than those of Hongfeng Lake and were 1.08, 18.43, 2.44 and 2.86 times higher than those of Hongfeng Lake, respectively (Liu et al., 2010). Compared with the Puding Reservoir, the contents of the other six elements in Caohai Lake were relatively low, except for Cd and Hg (Tang et al., 2017). Although the concentrations of some HMs in Hongfeng Lake were lower than those in Caohai Lake, the average concentrations were higher than the background values (except for Cr); this situation may be related to the presence of large mercury mines, coal mines, zinc mines and many factories around the reservoir (Liu et al., 2010). Due to interception by gates and dams, the slow-flowing water in the reservoir has a long residence time, which promotes the accumulation of pollutants and leads to a high concentration of HM pollution in the sediments of the Puding Reservoir (Tang et al., 2017). The Caohai Lake sediments also showed higher enrichment contents for As, Cd, Hg, Pb, Ni and Zn, especially Cd and Zn, compared with the average values for Chinese lake sediments (Li et al., 2021a). In summary, the historical mean contents of Ni, As, Cd, Zn, Hg and Pb in the Caohai Lake sediments were above background values; compared with sediments in the lakes in the same and adjacent provinces, those in the plateau lake areas and those on a national scale, the sediments of Caohai Lake had relatively high degrees of enrichment in HMs, among which Cd, Pb, and Zn need more attention.

3.1.2. Temporal changes

To investigate the dynamic trends of the HM content in the Caohai Lake sediments and its relationship with time, we used the Mann–Kendall test and regression analysis; the results are shown in Fig. 1. Trend analysis showed that the As and Ni concentrations in the Caohai Lake sediment had a decreasing trend (Fig. 1), but only As had a significant trend (-1.58 mg kg^{-1} $year^{-1}$); additionally, the As concentrations were highest in 2013 (17.55 mg kg^{-1}) and then decreased, reaching their lowest level in 2019 (8.48 mg kg^{-1}). We analyzed the Statistical Yearbook of Wining County (https://www.gzweining.gov.cn) from 2005 to 2020 and found that in 2013, Wining County had the highest agricultural fertilizer application, industrial solid waste generation and industrial solid waste emissions, while after 2013, these indicators showed a decreasing trend (Table S3), so we believe that this change in trend is related to the influence of agriculture and industry.

Table 1
Statistical description of HMs in sediments and recommended limits.

Lake	Parameter	HMs (mg/kg)								References
		As	Cd	Cr	Cu	Hg	Pb	Ni	Zn	
Caohai	Min.	6.0	0.28	9.5	13.4	0.04	9.7	13.3	6.8	This study
	Max.	283.6	69.30	142.0	82.3	1.22	272.5	180.7	1821.2	
	Median	16.1	7.47	58.7	25.2	0.39	65.2	37.3	305.1	
	Mean	32.0	14.19	66.8	26.7	0.41	87.6	40.2	406.2	
	CV (%)	11.9	113.7	55.5	37.2	65.0	69.9	59.9	78.7	
	Kurtosis	0.23	2.02	−1.16	4.21	0.33	0.59	5.60	3.08	
	Skewness	−0.18	1.57	0.33	2.26	0.52	1.14	3.09	1.64	
	BGV	20.0	0.659	95.9	32.0	0.11	35.2	39.1	99.5	
	TEL	5.9	0.60	37.3	35.7	0.17	18.0	35.0	123.0	
	PEL	17.0	3.53	90.0	197.0	0.49	36.0	91.3	315.0	
Dianchi	Mean	97.3	2.51	93.8	366.6	0.66	183.5	—	514.6	Liu, 2012; Meng, 2016
Hongfeng		29.7	0.77	87.9	91.9	0.66	35.9	190.9	142.0	Liu et al., 2010
Puding Reservoir		48.7	3.90	151.8	150.3	0.23	284.9	82.1	546.9	Tang et al., 2017
China		32.4	0.67	73.41	43.4	0.20	46.28	37.3	140.4	Li et al., 2021a

Note: CV and BGV, coefficient of variation, and background value; TEL and PEL, threshold. effect level and probable effect level (MacDonald et al., 2000); —, no available data.

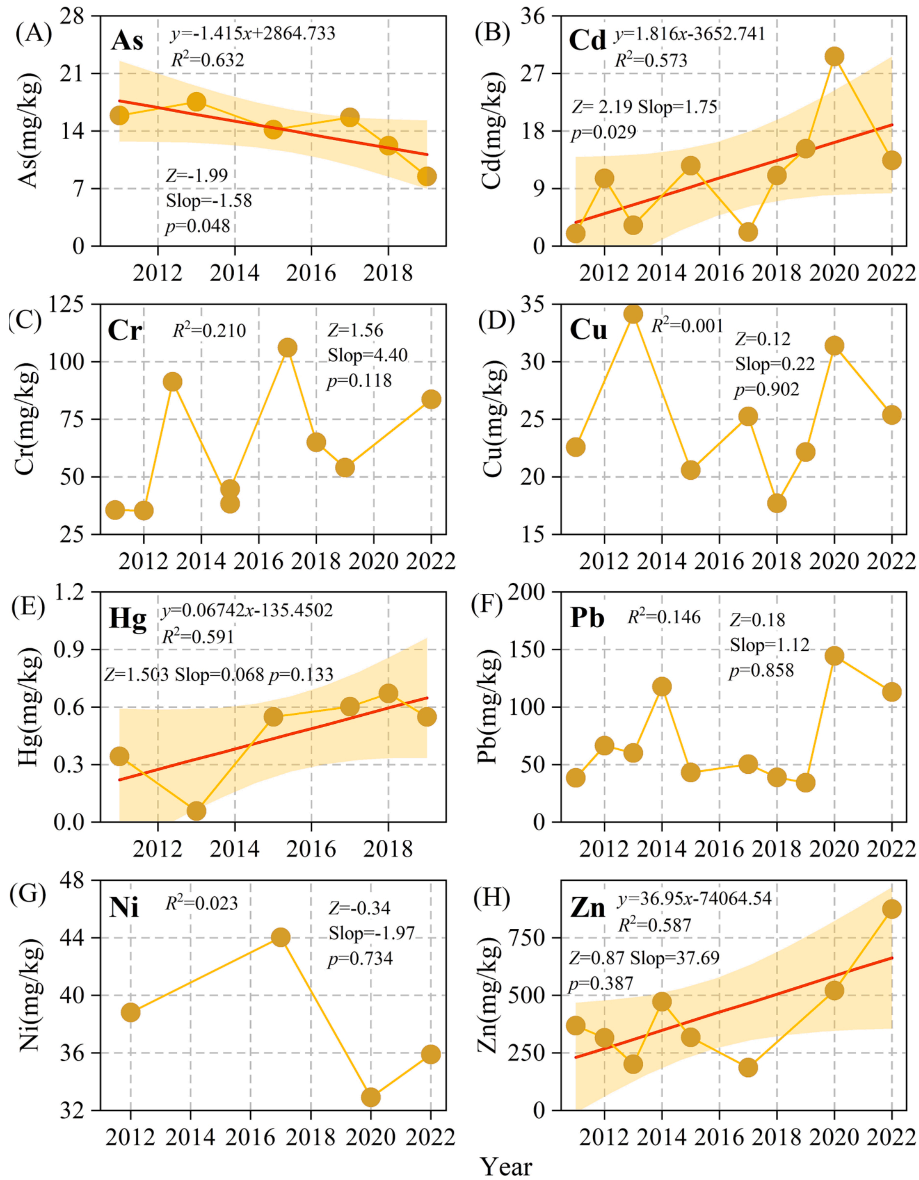

Fig. 1. Variation and relationship of HM concentrations in Caohai sediments over time. (A)–(G) represent As, Cd, Cr, Cu, Hg, Pb, Ni, and Zn, respectively.

Regression analysis showed that the As content in the Caohai Lake sediment was significantly negatively correlated with time and had an overall downward trend after peaking in 2013 (Fig. 1). The concentrations of the remaining six HMs in the sediment showed an increasing trend, with Cd significantly increasing at a rate of 1.75 mg kg^{-1} per year; the concentrations of the remaining HMs fluctuated with no significant increasing trend. The highest concentrations of Cr, Cu and Zn in sediment were found in 2017, 2013 and 2022, respectively, but Cd, Cu and Pb all showed high peaks in 2020 (Fig. 1). Meanwhile, the regression analysis showed that the Cd, Hg and Zn contents in the Caohai Lake sediments were significantly and positively correlated with time, with annual rates of increase of 1.816, 0.06742 and 36.95 mg kg^{-1}, respectively, similar to the results of the Mann–Kendall test analysis. The contents of Cr, Cu, Pb and Ni in sediments peaked in 2017, 2013, 2020 and 2017, respectively, but no significant correlation was found between the concentrations of these elements and time.

We also conducted an abrupt change test for changes in sediment HM concentrations in Caohai Lake by using the Mann–Kendall test (Fig. 2) and found that the UF and UB curves for As, Hg, Ni and Zn all had only one intersection; among them, the sediment As and Ni concentrations showed a decreasing trend from 2015 and 2018, respectively, but none of these trends were significant. Hg and Zn also showed a decreasing trend from 2015 and 2019, respectively, and again, the trends were not significant. The UF and UB curves for Cd intersected within the 95 % confidence interval, and both UF values were greater than 0. Therefore, we can determine that the Cd concentrations showed an increasing trend; furthermore, since UF > 1.96 after 2019, we can determine that the Cd in Caohai Lake sediments had a significant increasing trend after 2019. In addition, Cr fluctuated in a small range within the confidence interval of the UF and UB curves without a significant increase, while the changes in Cu and Pb showed no obvious pattern. The urban population of Wining County in 2020 (572,000) was 33,000 more than that in 2019 (539,000), which is two to three times the growth rate of previous years (Weining County Bureau of Statistics, 2017–2021). The accumulation of HMs in the Caohai Lake sediments was closely related to the surrounding industrial and agricultural activities and domestic sewage discharge. For example, more than 8,000 t of domestic sewage is directly discharged into Caohai Lake from Weining County every day (Long et al., 2021). In addition, HMs readily combine with phosphorus, and human activities in the villages surrounding Caohai Lake discharge more than 13 tons of phosphorus into Caohai Lake every year (Yang et al., 2019; Zhou et al., 2018).

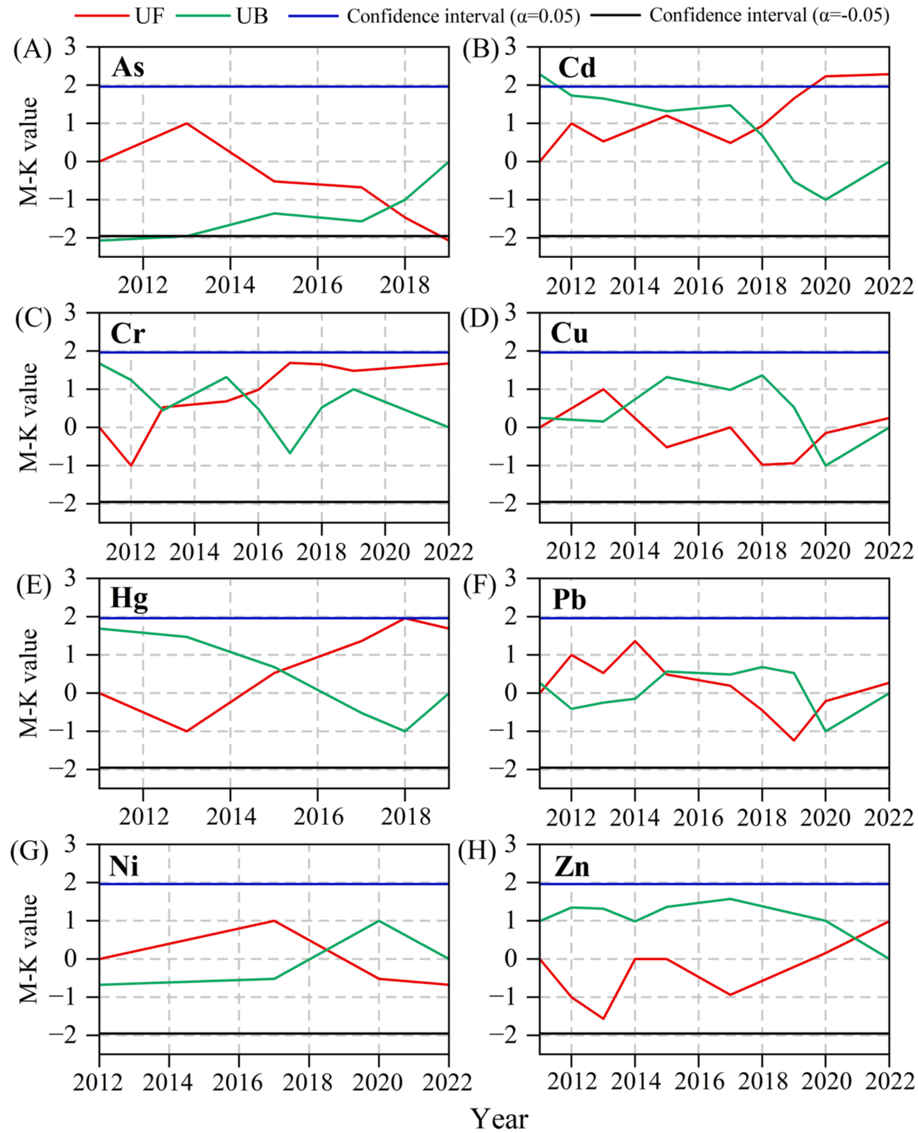

Fig. 2. Mutation M–K curves of the mean concentration of HMs from 2011 to 2022. (A)–(G) represent As, Cd, Cr, Cu, Hg, Pb, Ni, and Zn, respectively.

3.1.3. Spatial distribution

We used the inverse distance weighting method to spatially interpolate the HM concentrations in the Caohai Lake sediment; the results are shown in Fig. 3. The concentration of As in the central sediment of Caohai Lake was higher than the background value at most points, while the concentration around it was relatively low (Fig. 3A). The distribution characteristics of Cd, Pb and Zn in the surface sediments of Caohai Lake were similar, and more than 90 % of the sample sites had concentrations greater than background values (Fig. 3B, G, H). The wharf and county town of Weining are on the northern shore of Caohai Lake, which where tourists gather and where the waterway is located. Domestic sewage, traffic and tourism discharges from the county town are the main sources of pollution in this area (Zhang et al., 2013). On the southwest and southeast coasts, some farmland and domestic pollution emissions also surround the lake; furthermore, discharge from zinc smelting in Weining, Hezhang and other places causes high levels of Cd and Zn in these areas (Bi et al., 2007). The eastern lake area of Caohai Lake has a reducing environment dominated by water-holding plants, while the western part mostly has an oxidizing environment suitable for submerged plants (Cao, 2016); redox conditions are important factors influencing the transport and transformation of HMs. Cd from sediments is released under oxidizing conditions and remobilized under reducing conditions, while Fe-Mn oxides in the sediments resorb Cd^{2+} released from the oxidizing environment (Zhang et al., 2016), leading to the high Cd concentration in the sediments of the central-eastern lake layer. Similarly, the distribution characteristics of Zn and Pb are influenced by plant and sediment Fe and Mn. The concentrations of Cr and Cu in the Caohai Lake sediments were relatively low, and only a few areas along the coast and the L4 points had values higher than the background values (Fig. 3C, D). The points that had sediment Hg contents lower than the background value were scattered throughout the lake, mainly near L1, L4, L7, L12 and L14. The Ni contents in northwestern and southern Caohai Lake exceeded the background value (Fig. 3F).

3.2. Pollution intensity

3.2.1. Lake scale

To explore the overall contamination of the Caohai Lake sediments, we analyzed the I_{geo} values of the collected data (Fig. 4). The I_{geo} values of As and Cr in the sediments were less than 0, indicating that these two elements were not present as pollutants in Caohai Lake, 3.7 % of the Cu points were slightly polluted, and the other points were not polluted. Points were slightly or moderately polluted with Ni at 9.9 % and 2.5 %, respectively. Cd was found in 12.5 % of the points with I_{geo} values

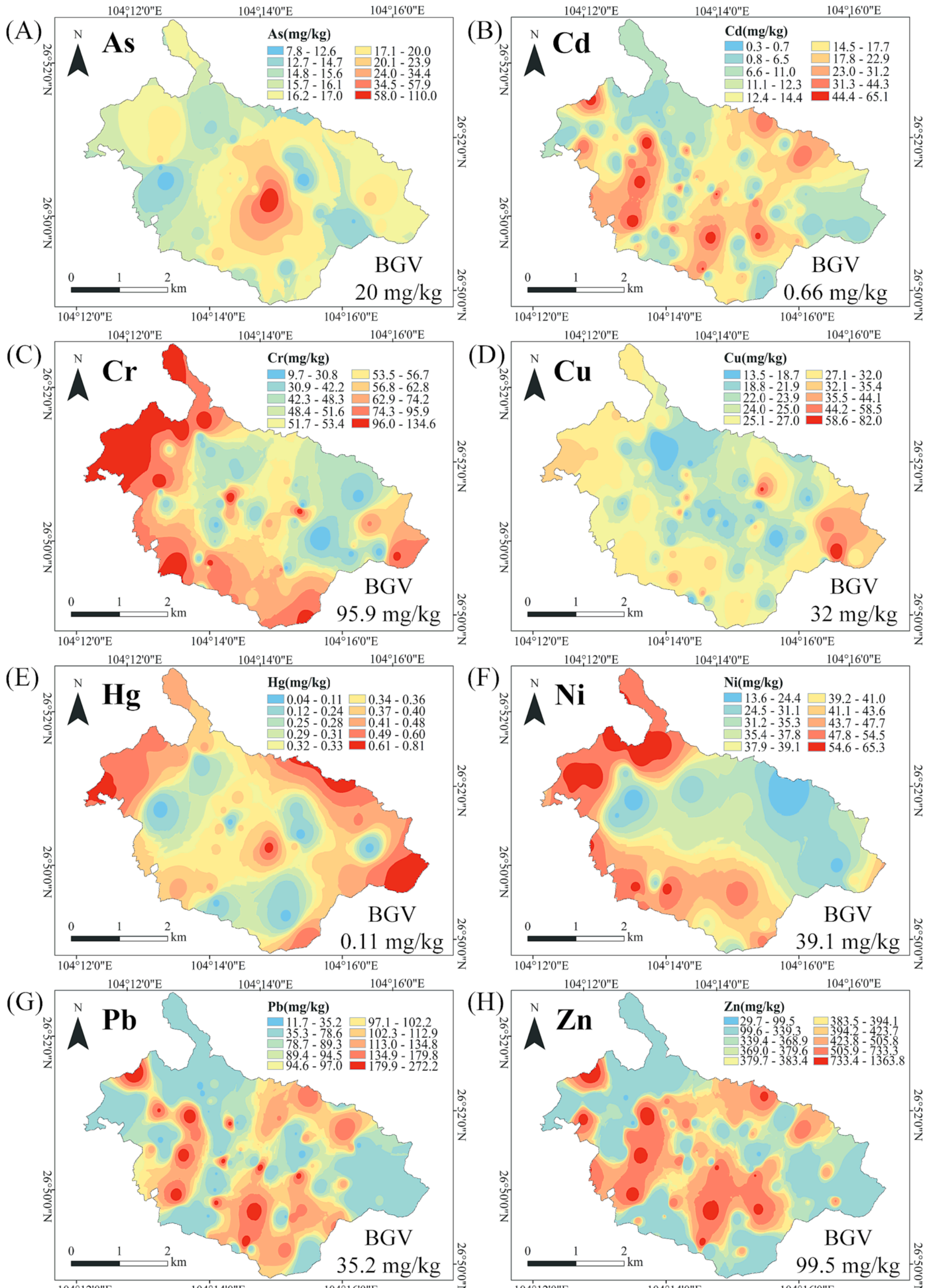

Fig. 3. Spatial distribution of sediment HMs in Caohai Lake. BGV: background values; (A)–(G) represent As, Cd, Cr, Cu, Hg, Pb, Ni, and Zn, respectively.

greater than 5, reaching extreme pollution, while the percentages of severe to extreme, severe, moderate to severe, moderate and light pollution were 20.3 %, 15.2 %, 14.6 %, 13.1 % and 10.1 %, respectively, with only 13.9 % uncontaminated. A total of 16.1 %, 51.8 % and 8.9 % of the points in the sediments were moderately to severely, moderately and lightly contaminated with Hg; 7.1 %, 25.0 % and 29.2 % were moderately to severely, moderately and lightly contaminated with elemental Pb; and 3.6 %, 16.9 %, 31.3 %, and 31.3 % of the points in the sediments were severely, moderately to severely, moderately and lightly contaminated with Zn. A total of 16.9 % of the points were not contaminated. In summary, Caohai Lake was less polluted by As, Cr, Cu, and Ni, slightly to moderately polluted with Hg, Pb, and Zn, and moderately to severely polluted with Cd.

3.2.2. *Temporal variation*

The I_{geo} analysis of sediment HMs from 2011 to 2022 indicated that As, Cr, Cu and Ni showed no contamination during this period (Fig. 5). The I_{geo} value for Cd fluctuated greatly, with three peaks in 2012, 2014

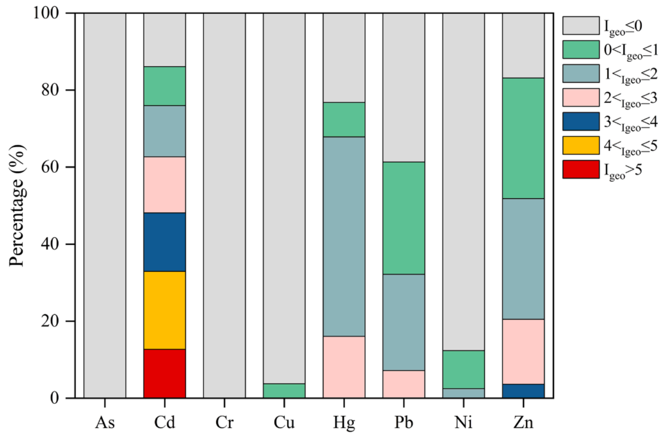

Fig. 4. The stacked columns of the proportion of HMs in the Caohai Lake sediments show different pollution intensities.

and 2020, and the peaks gradually increased, reaching severe to extreme pollution levels in 2020 ($I_{geo} = 4.64$). Hg had the lowest contamination in 2013 ($I_{geo} = -1.54$) and then showed a gradual increase, reaching its highest value in 2018 ($I_{geo} = 2.02$). Sediment contamination with Hg after 2019 could not be analyzed due to data limitations. Overall, Hg was at a moderate level of contamination during the period from 2015 to 2019. The I_{geo} value of sediment Pb fluctuated widely, but it reached its highest value in 2020 ($I_{geo} = 1.16$), indicating a mild level of pollution. The I_{geo} value of Zn in the sediments peaked in 2011 ($I_{geo} = 1.29$), 2014 ($I_{geo} = 1.23$) and 2022 ($I_{geo} = 2.41$), with mild to moderate pollution. The I_{geo} value of Zn after 2017 also exhibited a continuously increasing trend, with mild pollution to moderate pollution. Taken together, these results showed that Cd, Pb and Zn in the sediments were heavily to extremely, lightly and lightly polluted in 2020, respectively.

3.3. Risk assessment

The TRI of HMs in the Caohai Lake sediments ranged from 11.89 (2011) to 43.66 (2020), with an average of 22.99, indicating that the

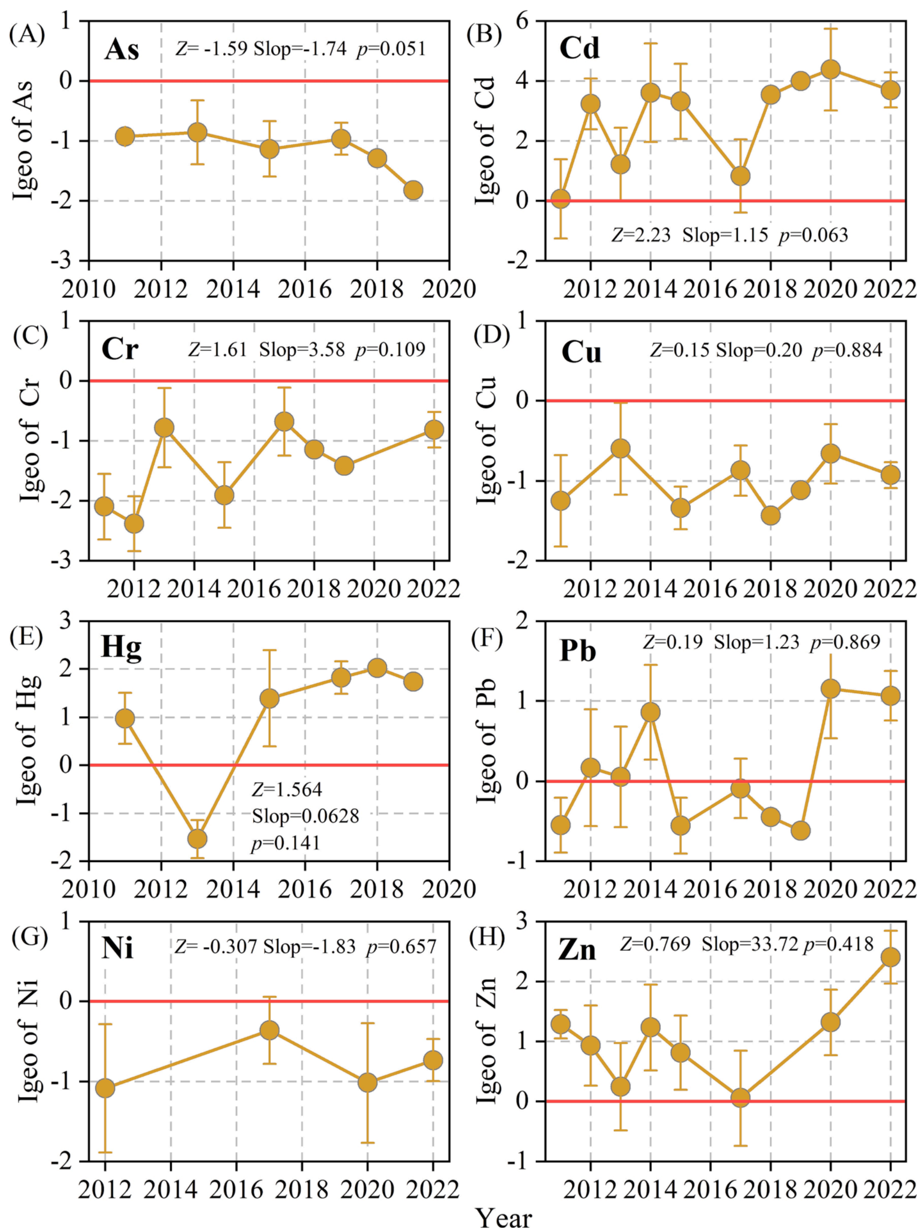

Fig. 5. Variation in the I_{geo} values of HMs in the Caohai Lake sediments. (A)–(G) represent As, Cd, Cr, Cu, Hg, Pb, Ni, and Zn, respectively.

Caohai Lake sediments showed a very high risk of toxicity (TRI > 20) (Fig. 6). The TRI values in 2011 (TRI = 11.89), 2012 (TRI = 18.27), 2013 (TRI = 12.87), and 2017 (TRI = 13.17) showed a moderate to considerable risk of toxicity (Fig. 6A). The TRI of the Caohai Lake sediments was greater than 20 in 2014, 2015 and after 2018, showing a very high toxicity risk. Pb and Cd contributed 7.60–16.91 % and 54.53–74.59 % to the TRI, respectively, and the cumulative contribution of these two elements was 69.34–90.56 %. These results indicated that Pb and Cd were the main contributors to the toxicity risk associated with the Caohai Lake sediments, which was mainly attributed to the low TEL value of Cd and high Cd concentration in the sediments. These findings highlight the rather high toxicity risk from metals in the Caohai Lake sediments and show that more attention should be given to Cd and Pb pollution. The Mann–Kendall trend analysis showed that the Caohai Lake sediment HM TRI increased from 2011 to 2022 (Z = 2.33, slope = 2.03), with the TRI reaching its highest value in 2020 (TRI = 43.7). From the Mann–Kendall abrupt change analysis, the UF and UB values for the Caohai Lake sediment HM TRI intersect within the confidence interval, indicating an abrupt change in ecological risk for HMs in Caohai Lake sediments in 2019 (Fig. 6). The rapid increase in population in 2019–2020 may be an important reason for the increased ecological risk of Caohai Lake sediment (Weining County Bureau of Statistics, 2017–2021). In addition, different amounts of HM data were collected in different years (8 elements in 2011, 4 in 2012 and 2014, 5 in 2020, 6 in 2013, 2018, 2022, and 7 in 2015 and 2019); the TRI is the sum of the risk indices of the different elements, so except for 2011, our calculation of HM TRI values in the different years is likely low and may underestimate the toxicity risk of the sediments.

3.4. Source analysis

We employed PCA to further identify potential sources of eight metals in the Caohai Lake sediments. Three principal components were selected according to the size of the eigenvalues (>1); they explained a total of 65.94 % of the variance (Table 2). PC1 (Cd, Pb, and Zn), PC2 (As, Cu, and Ni), and PC3 (Cr and Hg) accounted for 25.86 %, 21.77 %, and 18.31 % of the total variance, respectively. The loadings of Pb, Cd and Zn in PC1 were relatively high at 0.87, 0.83, and 0.72, respectively; studies have shown that Pb is a major marker of traffic emissions, as it mainly originates from fuel combustion in vehicles, vehicle engines and tire friction (Wang et al., 2019). Traffic roads are densely distributed along the Caohai Lake coast, thus potentially enabling large amounts of Pb to enter the lake, especially in the most congested northern counties. In addition, industrial emissions or wastewater discharges are major

Table 2
Principal component loading matrix.

HMs	PC1	PC2	PC3
As	−0.16	0.81	−0.14
Cd	0.83	−0.04	−0.15
Cr	−0.16	−0.47	0.54
Cu	0.19	0.77	0.21
Hg	−0.05	0.11	0.85
Pb	0.87	0.14	0.28
Ni	0.11	0.89	0.07
Zn	0.72	0.02	−0.38
Eigenvalues	2.07	1.50	1.10
Contribution rate (%)	25.86	21.77	18.31

sources of Pb and Zn (Facchinelli et al., 2011). Mining is also a major source of Cd and Zn, and the Caohai Basin is rich in mineral resources; many local zinc smelting sites have been developed in recent decades (Hu et al., 2017). In addition, Cd is associated with the long-term use of pesticides and fertilizers in agricultural production (Zhou et al., 2016), and a large amount of agricultural land is distributed along the coast of the Caohai Lake. Therefore, PC1 mainly characterizes the influence of traffic, mining and agricultural sources on the Cd content of the sediments. For PC2 (As, Cu, and Ni), Cu and Ni originate from urban pollution and industrial wastes, such as those from the electroplating industry (Islam et al., 2015; Li et al., 2009). They may also originate from pesticides, insecticides and fertilizers from agricultural activities (Amlund et al., 2012), but the concentrations of As, Cu and Ni were lower than the background values. The HMs associated with the parent soil composition are generally considered elements with low contamination, and the Igeo values showed that the concentrations of As, Cu and Ni did not reach the risk thresholds (Fig. 4). These results suggest that human activities have contributed little to the accumulation of these three metals in the Caohai Lake sediments. Therefore, PC2 represents the soil parent material, i.e., the natural source. For PC3, the loading values for Cr and Hg were relatively high. Cr may come from a variety of industrial sources, particularly the metal plating and treatment industries (Hou et al., 2017). Studies in the Pearl River Delta in southern China have shown that Cr in soils is mainly derived from rock weathering and soil-forming parent material, and similar results have been observed in other countries worldwide (Hu et al., 2013b; Guan et al., 2018). Hg in the environment mainly comes from activities such as coal-burning exhaust gases and automobile exhaust emissions (Huang et al., 2017). In summary, PC3 was defined as representing industrial sources.

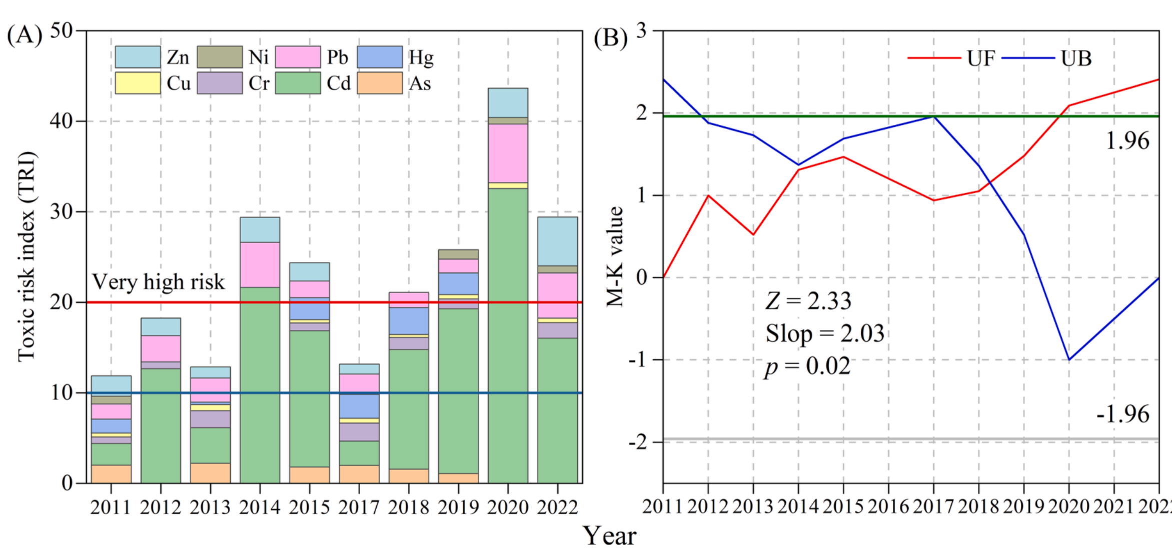

Fig. 6. Variation in the toxic risk index (TRI) of HMs in the Caohai Lake sediments and mutation M–K curves from 2011 to 2022.

4. Conclusions

In conclusion, in this study, the HM pollution, spatiotemporal distribution and potential risks in Caohai Lake sediments were assessed by collecting data from 2011 to 2022 in Caohai Lake, and several conclusions were drawn. (1) The historical mean concentrations of As, Cd, Pb and Zn in the Caohai Lake sediments were higher than the background values and PEL. (2) The Mann–Kendall statistical test showed that the concentrations of As and Ni in the Caohai Lake sediment had a decreasing trend (2011–2022), and the As concentration changed abruptly in 2018; the regression analysis revealed that Cd had a significant increasing trend of 1.75 mg kg^{-1} per year, and Cd concentrations increased significantly in 2020. (3) The inverse distance weighting analysis proved that the sediment concentrations of Cd, Hg and Zn were above background values at 90 % of the sample sites. (4) From the perspective of cumulative HM pollution, the Caohai Lake sediments were most polluted with Cd, with I_{geo} values showing moderate and heavy pollution that reached 75.9 % and 48.1 %, respectively. The I_{geo} values for Cd and Zn exceeded the light pollution level between 2011 and 2022. (5) The TRI for HMs in the Caohai Lake sediments increased from 2011 to 2022, reaching its highest value (43.66) in 2020. The Caohai Lake sediments exhibited a very high toxicity risk (TRI > 20), and the contribution of Cd to the TRI was 54.53–74.59 %. (6) Source analysis showed that the Cd, Zn and Pb in the Caohai Lake surface sediments were mainly from traffic, mining and agricultural sources; As, Cu and Ni were from natural sources; and Hg and Cr were from natural and industrial sources. To prevent and control HM pollution, more effective monitoring of the handling and discharge of traffic, industrial and domestic pollutants is needed.

5. Founding

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CRediT authorship contribution statement

Dianpeng Li: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Software, Writing – original draft. **Zhengjie Zhu:** Investigation, Methodology, Software, Writing – review & editing. **Xuecheng Cao:** Methodology, Data curation, Formal analysis, Writing – review & editing. **Tangwu Yang:** Data curation, Software, Formal analysis. **Shuqing An:** Supervision, Conceptualization, Writing – review & editing.

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.

Appendix A. Supplementary material

Supplementary material to this article can be found online at https://doi.org/10.1016/j.arabjc.2023.105543.

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