


Article

Characterization and Risk Assessment of Nutrient and Heavy Metal Pollution in Surface Sediments of Representative Lakes in Yangxin County, China

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Abstract: Increased urbanization and industrialization globally have led to the widespread pollution of water bodies (e.g., lakes) by heavy metals (HMs) and nutrients. These pollutants accumulate in water and surface sediments, posing risks to both aquatic organisms and human health. In November 2022, surface sediment samples from three lakes—Lianhua Lake, Mati Lake, and North Lake—were collected to assess nutrient (nitrogen and phosphorous) and HM content. Total N (TN), total P (TP), and HM concentrations were analyzed. The pollution status was evaluated using comprehensive pollution index (FF) methods and the potential ecological risk index (RI) (E_r^i). The results were as follows: (1) Variations in nutrient and HM contents were observed among the three lakes. Lianhua Lake exhibited the highest average TN content (1600 mg/kg), while North Lake had the highest average TP content (2230 mg/kg). The average concentrations of Cd, Hg, and As in the surface sediment surpassed the soil background values of Hubei Province, reaching 1.41, 2.74, and 1.76 times the background values, respectively. Notably, Hg exceeded the standard in Lianhua Lake by 3.39 times, followed by North Lake (2.52 times) and Mati Lake (2.24 times). (2) The FF and potential E_r^i revealed that the average RI values for Mati Lake, North Lake, and Lianhua Lake were 106.88, 126.63, and 162.18, respectively. These indices categorized the ecological risk levels as moderate, while nutrient salts in the surface water reached a severe pollution level. (3) Correlation and PCA indicated that Cu, Pb, Cd, and Ni were linked to mineral smelting, aquaculture feed, and agricultural fertilizers. Hg and nutrient salts originated from atmospheric deposition of surrounding domestic waste water and traffic exhaust gases. Agricultural activities seemed to contribute to As concentration in the lakes, while Cr has its main origin in the weathering of the rock matrix.

Keywords: nitrogen; phosphorous; heavy metals; comprehensive pollution index; potential ecological risk index



Citation: Yang, X.; Leng, M.; Ge, X.; Wu, X.; Liu, H.; Lin, G.; Huang, Z.; Chen, Y. Characterization and Risk Assessment of Nutrient and Heavy Metal Pollution in Surface Sediments of Representative Lakes in Yangxin County, China. *Sustainability* **2024**, *16*, 2252. <https://doi.org/10.3390/su16062252>

Academic Editor: Changwoo Ahn

Received: 23 December 2023

Revised: 4 March 2024

Accepted: 6 March 2024

Published: 7 March 2024



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1. Introduction

Lake sediments play a crucial role in lake ecosystems, providing essential habitats for flora and fauna and serving as reservoirs for vital nutrients supporting lake biota. Additionally, lake sediments act as repositories for pollutants, acting as primary contributors to endogenous pollution within lakes [1]. Primary pollutants in sediments include heavy metals (HMs, e.g., Hg and Cd), excess nutrients (such as N and P), and insoluble organic compounds. HMs, known for their high toxicity, tendency to accumulate, and resistance to degradation, present unique challenges [2]. Upon entering water bodies, HMs undergo processes like adsorption, accumulation, and precipitation by suspended solids in the

water column, becoming enriched in sediments [3]. Changes in water body or sediment conditions can lead to the re-release of nutrients and HMs, causing secondary pollution [4]. Consequently, the concentration of HMs in sediments serves as a vital indicator for evaluating water body environmental quality. The accelerating processes of urbanization and industrialization contribute to heightened HM accumulation in sediments due to the discharge of domestic and industrial wastewater into urban water bodies, posing risks to human health through the food chain [5].

The pollution of water sediments is a global concern, with numerous scholars analyzing pollution status and water quality changes through sediment studies. In the Tigris River, Turkey, variations in sediment HM content and pollution levels were assessed to identify if waste discharged from an upstream Cu mine was the main contributor to water quality deterioration [6]. The sources of certain HMs in soil and sediment are intricately linked to urbanization, mineral exploitation, and smelting activities [7]. Studies in the Nadya mining area in Ukraine revealed a substantial correlation between chemical element distribution characteristics and anthropogenic loading intensity, categorizing mining as a hazardous source of environmental pollution. Consequently, mining operations were found to elevate HM concentrations in surrounding soils [8]. Urbanization's impact on environmental pollution was demonstrated through tracking HM levels in the atmosphere and soil in Croatia. Emissions from urban industrial and transportation sources led to the detection of toxic elements, including Ni and Pb, in suburban soils in prevalent wind directions [9].

The distribution of HMs in the sediments of Chao Lake, China, aligns with the traffic line, attributing the highest pollution contribution to traffic sources. Urbanization has also been identified as a factor influencing the extent of HM pollution in sediments [10]. Besides HMs, lakes also face widespread contamination by nutrient salts. Internal pollution in the sediments of Honghu, China, exhibits severity, with notable spatial and temporal variations in the release of N and P [11]. In Houguan Lake, China, water body eutrophication influences the oxidation–reduction potential, exacerbating HM pollution in sediments and contributing to water pollution [12]. This illustrates the prevalent complex pollution of sediments by both nutrients and HMs in lakes and other water bodies [13].

Various evaluation methods exist for nutrients in sediments, including assessments of organic C and organic N and comprehensive pollution index evaluations [14]. Similarly, diverse evaluation methods are available for HMs, encompassing pollution load, geoaccumulation, and potential ecological risk indices (RIs) (E_r^I) [15,16]. Statistical methods, such as correlation, principal component, and cluster analyses, are primarily employed to determine the origin of HMs in sediments [17,18].

Yangxin County, situated in southeastern Hubei Province, China, lies in the middle reaches of the Yangtze River, bordered to the east by the Yangtze River and featuring numerous shallow-water lakes. The region has witnessed significant mineral resource developments and metal smelting activities [19]. While prior research by Rong [20] and Zhao [21] has delved into the sediments of Wanghu Lake in Yangxin County, studies of the pollution of surface sediments in typical lakes in this area remain limited. This study investigated nutrient salts and HMs in the surface sediments of three representative lakes in Yangxin County—Lianhua Lake, Mati Lake, and North Lake. The aim of this study was to offer a theoretical reference for pollution control and the treatment of lakes in this region, as well as in the middle and lower reaches of the Yangtze River, by analyzing pollution distribution characteristics and evaluating ecological risk.

2. Materials and Methods

2.1. Study Area

Situated in the southeast of Hubei Province, in the middle reaches of the Yangtze River (114°43'~115°30' E, 29°30'~30°09' N), Yangxin County features numerous shallow lakes (Figure 1). Considering geographical location, nature, type, and importance, Lianhua Lake, Mati Lake and North Lake were chosen as representative lakes for this study. The

region experiences a subtropical monsoon climate, characterized by cold winters and hot summers, with an average annual temperature of 16.8 °C. The central urban area has a population of 204,800.

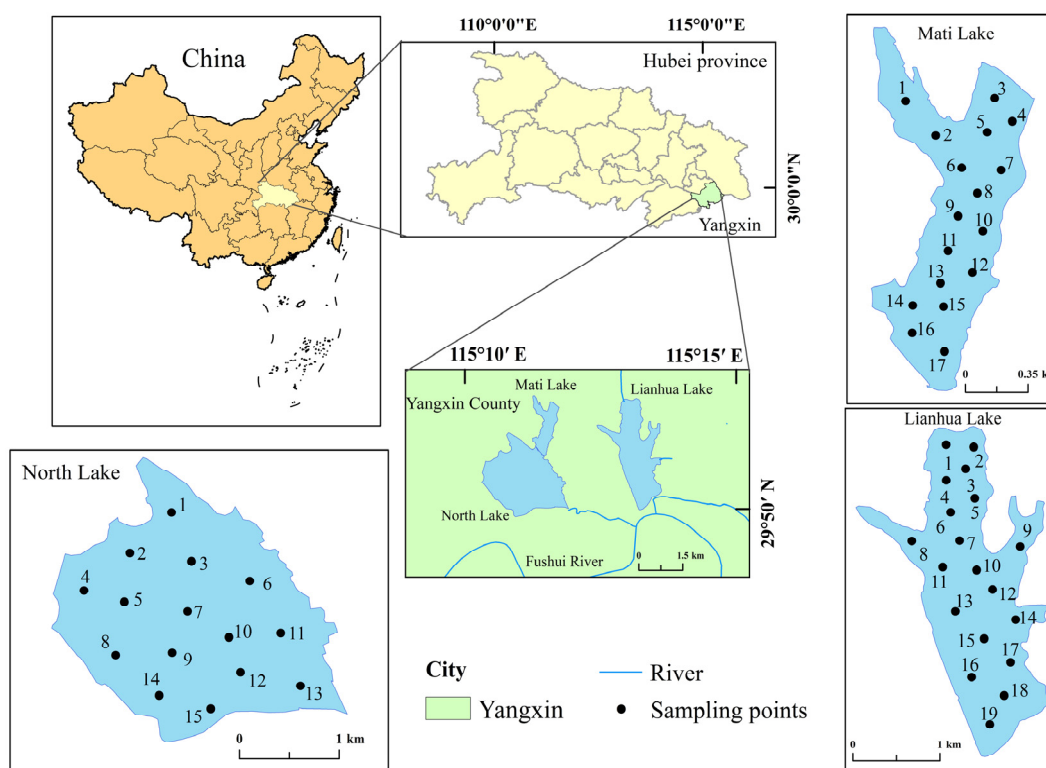


Figure 1. Map of the study area. Considering geographical location, nature, type, and importance, Lianhua Lake, Mati Lake and North Lake were chosen as representative lakes for this study.

Lianhua Lake: Located in the heart of Yangxin County, Lianhua Lake is significantly impacted by human activities. As the largest urban lake in the county, it has a water surface area of 3.15 km², with an average depth of 3.2 m. The lake's water flows into the Fu River. Rainwater discharge inlets are predominantly found on the west side, with sewage discharge inlets and combined sewer overflow outlets on the western side set to be redirected by 2021.

Mati Lake: Situated in the northwestern suburbs of the old Yangxin County, Mati Lake covers an approximate water area of 0.7 km². It can be divided into upper and lower lakes through connecting channels and sluice gates. Despite its relatively small size, its proximity to an old urban area has subjected it to prolonged human influence.

North Lake: Found to the southwest of Mati Lake, North Lake spans an area of about 3.4 km² with an average water depth of 3.85 m. This rural lake, surrounded by a sparse population, witnessed aquaculture activities until 2017. All three lakes have storage and irrigation functions, while Lianhua Lake and Mati Lake have also function as landscape entertainment, while North Lake serves a fish breeding purpose [22].

2.2. Sampling and Analytical Methods

Considering the size and morphological characteristics of the three lakes, a total of 19, 17, and 15 sampling sites were chosen in Lianhua Lake, Mati Lake, and North Lake, respectively (Figure 1). Sediment samples were collected in November 2020. Surface sediment samples (0–15 cm in depth) were collected by employing a Peterson grab sampler and carefully stored in clean polyethylene bags, refrigerated, and transported to the laboratory. Subsequently, the samples underwent manual filtration, drying, and grinding for the determination of nutrient salts and HMs.

Sediment total N (TN) was analyzed through semi-microanalysis using the Fully Automatic Autoanalyzer SKD-1000 from Shanghai Puou Instrument Co., Ltd. (Shanghai, China) [23]. Total P (TP) was determined by utilizing molybdenum antimony spectrophotometry with a UV-Vis spectrophotometer from Shanghai Yuanxi Instrument Co., Ltd. (Shanghai, China) [24]. Sediment Hg and As were determined through microwave digestion/atomic fluorescence spectrometry using an SH-AFS2200 fluorescence spectrophotometer from Qingdao Juchuang Huaye Instrument Co., Ltd. (Qingdao, China) [25]. The HMs Cr, Pb, Cd, Cu, Ni, and Zn were analyzed with aqua regia extraction inductively coupled plasma mass spectrometry using an inductively coupled plasma mass spectrometer from Jiangsu Tianrui Instrument Co., Ltd. (Kunshan, China) [26]. Parallel samples were prepared for each sampling site, ensuring an analysis error of less than 5%. The final result represented the average value of the parallel samples.

2.3. Data Processing and Analysis

Data preprocessing was carried out using Excel 2010 (Microsoft Corp., Redmond, WA, USA), and Origin 2021 (Origin Labs Inc., Northampton, MA, USA) was employed for correlation analysis and assessing the significance of the differences. Before the difference analysis, the normality and homogeneity of variance were tested using the Shapiro–Wilk and Levene methods, respectively ($p > 0.05$). Following analysis of variance, multiple comparisons were conducted using the Scheffé method, with the significance level set at $p < 0.05$. Pearson’s correlation analysis and principal component analysis (PCA) were performed to explore the relationship between nutrients and HMs. Both the Bartlett sphericity test and the Kaiser–Meyer–Olkin measure value were calculated to verify the suitability of data for PCA. To enhance the interpretability of the principal components, the varimax normalization algorithm was applied to rotate the components, and rotated data analysis was selected. These analyses were performed using SPSS 26.0 (IBM, Armonk, NY, USA). ArcGIS (Environmental Systems Research Institute, Inc., Redlands, CA, USA) was used to plot sampling points, and the spatial distribution characteristics of sediment nutrient salts and comprehensive potential E_r^i for HMs were analyzed using inverse distance weighting interpolation (IDW).

2.4. Environmental Risk Assessment Methods

2.4.1. Nutrient Pollution Evaluation Method

The assessment of nutrient salt pollution in the surface sediments of the lakes in Yangxin County utilized the single-factor pollution index and comprehensive pollution index methods [27]. The formulae applied were defined as follows:

$$S_j = C_j / C_s \quad (1)$$

$$FF = \sqrt{\frac{F^2 + F_{Max}^2}{2}} \quad (2)$$

where S_j is the single pollutant pollution index of the j th pollutant, C_j is the measured content of the j th pollutant, C_s is the environmental quality standard value of the pollutant, and FF is the comprehensive pollution index of nutrient salts. TN and TP pose the lowest ecological risks to sediments at 550 and 600 mg/kg, respectively, following the guidelines issued by the Ministry of Environment and Energy of Ontario, Canada [28]. F represents the average value of the pollution indices, (average values of S_{TN} and S_{TP}), and F_{Max} represents the maximum single-pollutant pollution index (maximum values of S_{TN} and S_{TP}). The relationship between the FF value and the pollution evaluation grade is outlined in Table 1.

Table 1. Classification of comprehensive pollution degree in sediments.

Rank Division	S _{TN}	S _{TP}	FF	Evaluation Level
I	S _{TN} < 1.0	S _{TP} < 0.5	FF < 1.0	Clean
II	1.0 ≤ S _{TN} < 1.5	0.5 ≤ S _{TP} < 1.0	1.0 ≤ FF < 1.5	Light pollution
III	1.5 ≤ S _{TN} < 2.0	1.0 ≤ S _{TP} < 1.5	1.5 ≤ FF < 2.0	Moderate pollution
IV	S _{TN} ≥ 2.0	S _{TP} ≥ 1.5	FF ≥ 2.0	Heavy pollution

2.4.2. HM Pollution Evaluation Method

In 1980, Swedish scholar Hakanson introduced the potential E_r^i method [29] to assess the potential ecological harm caused by HMs, considering their biological, toxicological, and sedimentary characteristics. This approach reflects the comprehensive impact of multiple HM pollutants and the potential influence of sedimentary HMs on the environment [30].

The calculation formula for the single-factor pollution parameter C_f^i was defined as follows:

$$C_f^i = C_s^i / C_n^i \quad (3)$$

The formula for the single-factor potential ecological risk coefficient (E_r^i) was defined as follows:

$$E_r^i = T_r^i \times C_f^i \quad (4)$$

The potential E_r^i was calculated using the following formula:

$$RI = \sum_r^i E_r^i \quad (5)$$

In these formulas, C_s^i is the measured concentration value of HM i in the sediment (mg/kg). C_n^i is the reference value of HM i in the background sediment (mg/kg). T_r^i is the ecotoxicity response coefficient of HM i , and RI is the comprehensive E_r^i of n types of HMs in the sediment, representing the sum of the E_r^i values [31,32]. The background values and toxicity response coefficients of the soils in Hubei Province are detailed in Table 2 [33].

Table 2. Background values and toxicity response coefficients of soils in Hubei province.

Element	Cd	Hg	As	Pb	Cr	Cu	Ni	Zn
Toxicity response coefficient	2	30	10	5	30	5	5	1
Soil background value	0.17	0.08	26.7	12.3	86	30.7	37.3	83.6

Hakanson's classification criteria, originally based on the toxicity coefficients of Polychlorinated Biphenyls (PCB), Hg, Cd, Pb, As, Cr, Cu, and Zn, were adjusted to accommodate the eight pollutants considered in the present study, ensuring accurate potential ecological risk evaluation results [34]. The modified classification boundaries are detailed in Table 3.

Table 3. A comparison of Hakanson's classification standards and the improved classification standards in this study.

Evaluation Level	Hakanson's Classification Standards		Improved Classification Standards in This Study	
	E_r^i	RI	E_r^i	RI
I, Low	$E_r^i < 40$	RI < 150	$E_r^i < 30$	RI < 100
II, Moderate	$40 \leq E_r^i < 80$	$150 \leq RI < 300$	$30 \leq E_r^i < 60$	$100 \leq RI < 200$
III, High	$80 \leq E_r^i < 160$	$300 \leq RI < 600$	$60 \leq E_r^i < 120$	$200 \leq RI < 300$
IV, Very high	$160 \leq E_r^i < 320$	RI ≥ 600	$120 \leq E_r^i < 240$	RI ≥ 300
V, Extreme	$E_r^i \geq 320$		$E_r^i \geq 240$	

3. Results

3.1. Distribution Characteristics of Nutrient Salts in Surface Sediments

The TN content in the sediments of the three lakes ranged from 220 to 3670 mg/kg, averaging 2270 mg/kg. North Lake had a slightly higher average TN content (2520 mg/kg) compared to Lianhua Lake (2100 mg/kg) and Mati Lake (2230 mg/kg). Variations in TN content were observed among samples from each lake, with Mati Lake showing the largest coefficient of variation (CV) at 32.17%, ranging from 220 to 3120 mg/kg. Lianhua Lake displayed a TN content ranging from 1210 to 3670 mg/kg, with a CV of 30.41%, and North Lake exhibited a range from 1820 to 3220 mg/kg, with a CV of 19.66%.

The average TP content decreased as follows: Lianhua Lake (1600 mg/kg) > Mati Lake (1430 mg/kg) > North Lake (1360 mg/kg). TP contents in different samples from the lakes ranged from 820 to 1940 mg/kg, averaging 1470 mg/kg. A significant difference in TP content was noted between Lianhua Lake and North Lake ($p < 0.05$). Lianhua Lake's TP content ranged from 1090 to 1940 mg/kg (CV: 16%), while that of North Lake ranged from 1820 to 3220 mg/kg (CV: 23.58%). No significant differences were observed between Mati and Lianhua lakes ($p < 0.05$), with Mati Lake's TP content ranging from 990 to 1770 mg/kg (CV: 16.84%). From a spatial distribution perspective (Figure 2), the southeastern part of Lianhua Lake exhibited lower TN contents, while higher contents of TP were predominant in the northeast. In Mati Lake, a high TN content was predominant in the northern part, whereas the TP content was more scattered across the sampling sites. In North Lake, high contents of both TN and TP were found in the northern and southern parts.

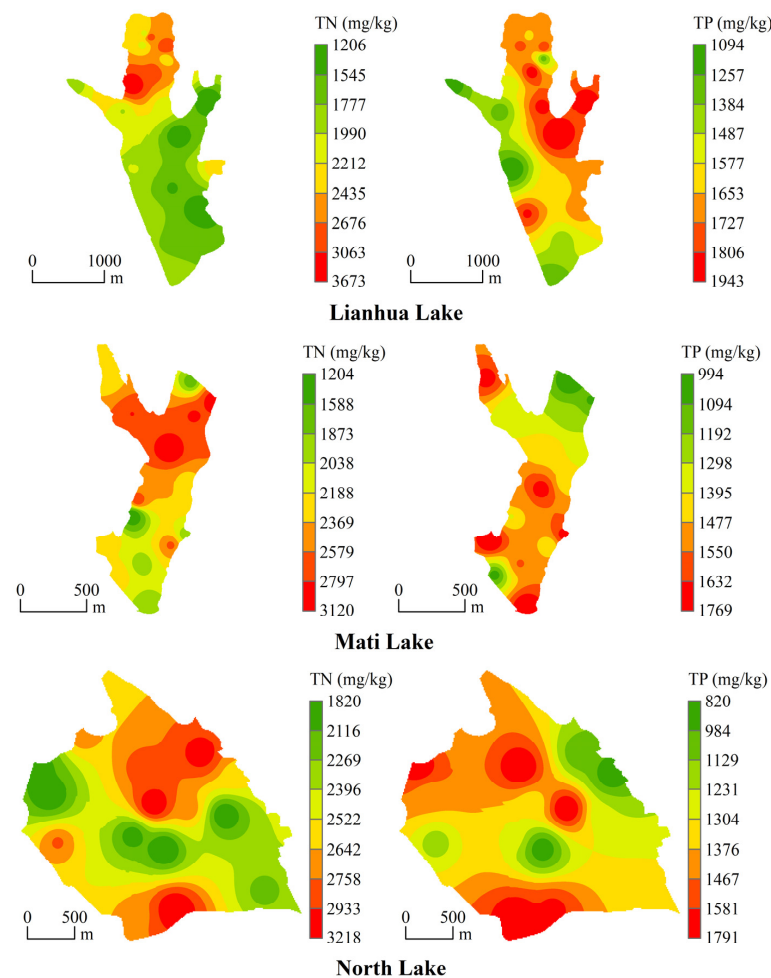


Figure 2. Spatial distributions of TN and TP contents in the surface sediment. The interpolation map illustrates the variations in nutrient content across the lake.

3.2. Distribution of HMs Characteristics in Surface Sediments

Lianhua Lake exhibited the highest average contents of Hg, As, and Zn in surface sediments, with the Zn concentration being approximately 1.5 times higher than the one registered for North Lake (Table 4). North Lake had the highest average Cr concentration, and the average concentrations of other HM elements were relatively similar in the three systems.

Table 4. HM contents in surface sediments in Lianhua Lake, Mati Lake and North Lake. Values correspond to the average concentration \pm standard deviation (SD) (mg/kg).

	Cd	Hg	As	Pb	Cr	Cu	Ni	Zn
Lianhua Lake	0.23 \pm 0.19	0.27 \pm 0.14	80.53 \pm 55.82	8.21 \pm 3.81	39.78 \pm 11.52	26.41 \pm 15.48	30.10 \pm 8.49	107.37 \pm 178.35
Mati Lake	0.22 \pm 0.17	0.18 \pm 0.08	20.74 \pm 10.77	8.76 \pm 3.46	43.88 \pm 11.42	29.44 \pm 14.17	35.23 \pm 8.55	74.24 \pm 23.94
North Lake	0.27 \pm 0.22	0.20 \pm 0.06	34.46 \pm 46.58	8.8 \pm 4.51	64.6 \pm 102.04	30.31 \pm 17.88	31.53 \pm 9.81	69.06 \pm 25.10

Notes: The unit for HM content is mg/kg.

The coefficients of variation for the eight HMs in the three lakes ranged from 24% to 166%. Zn in Lianhua Lake had the highest CV at 166%, followed by Cd with 84%. Mati Lake also showed a relatively high CV for Cd at 77%. As and Cr in North Lake had coefficients of variation of 135% and 157%, respectively, indicating significant differences in the spatial distributions of the various HMs in the three lakes.

Comparing the HM concentrations in the surface sediments of the study area to the background values of the soil in Hubei Province, the average concentrations of Cd, Hg, and As were 1.4, 2.7, and 1.8 times higher, respectively. Lianhua Lake exhibited higher Hg contamination, with an exceedance factor of 3.4. North Lake and Mati Lake also had high levels of Hg contamination, with exceedance factors of 2.5 and 2.2, respectively, suggesting that Hg is the most significant HM for the pollution in the surface sediments of the study area.

3.3. Environmental Quality Evaluation of Surface Sediments

3.3.1. Evaluation of Nutrient (TN and TP) Pollution

In accordance with the environmental quality evaluation criteria for sediments established by the Ontario Ministry of the Environment, Canada, the lowest observable effect levels for TP and TN were set at 600 and 550 mg/kg, respectively. These levels represent the tolerance concentrations for most benthic organisms [35]. Severe effect levels (SELs) for TP and TN were established at 2000 and 4800 mg/kg, respectively, indicating higher nutrient contents that could harm benthic organisms at higher levels. A comparison of nutrient content in the sediments of Lianhua, Mati, and North lakes with these standards revealed that all sampling sites exhibited concentrations of TN and TP below the lowest level of ecological toxicity. Furthermore, these concentrations were lower than those of the SELs. Consequently, sedimentary nutrient salts in Lianhua, Mati, and North lakes pose relatively low ecological risks.

The single pollution index of STN in the three lakes predominantly fell into categories III and IV (Figure 3). The percentage of severe TN pollution in North Lake (73.33%) was slightly higher than the ones registered in both the Mati Lake (70.29%) and Lianhua Lake (47.37%). In all three lakes, the TP content in the surface sediments surpassed that of TN. The single pollution index STP was categorized as IV, indicating 100% severe TP pollution across all sampling sites.

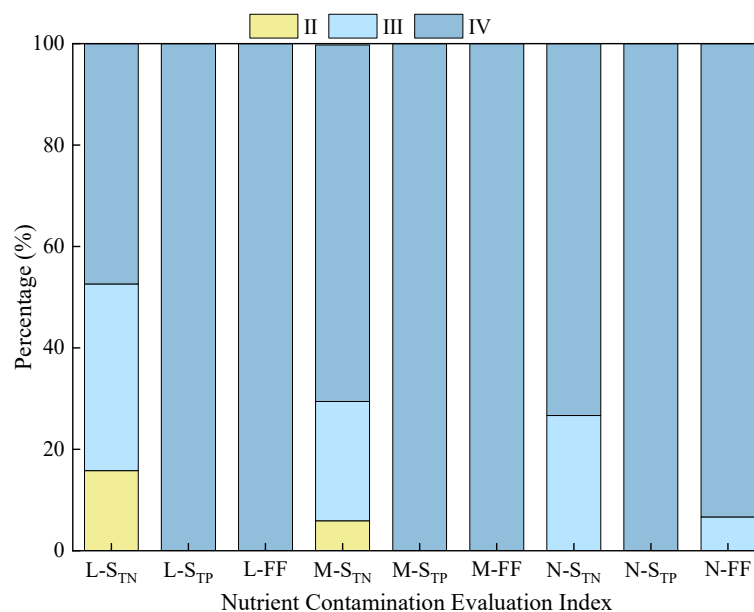


Figure 3. Evaluation of the single pollution index of sedimentary nutrient salts. Evaluation of variations in S_{TP} , S_{TN} , and FF among lakes. The graphical representation of changes in the proportion of pollution levels intuitively reflects the overall pollution status of each lake. TN TP pollution deserves more attention compared to TP TN pollution. Abbreviations: L, Lianhua Lake; M, Mati Lake; N, North Lake; S_{TN} , single pollution index of TN in the sediments of lakes; S_{TP} , single pollution index of TP; FF, comprehensive pollution index of nutrients.

The distribution of FF followed this order: Lianhua Lake (3.41) > Mati Lake (3.17) > North Lake (3.12). Pollution was most severe in Mati Lake and Lianhua Lake, with all sampling sites designated as category IV. North Lake exhibited relatively lower pollution compared to the other systems, with the highest level being category III. The specific pollution indices of nutrient salts in the sediments of the three lakes are detailed in Table 5.

Table 5. Grading of comprehensive pollution index of nutrient salts in surface sediments.

	Degree of Pollution Classification		
	Lianhua Lake	Mati Lake	North Lake
S_{TN}	2.10	2.34	2.52
S_{TP}	3.81	3.41	3.24
FF	3.41	3.17	3.12
Pollution grade (based on the grades in Table 1)	Severe pollution	Severe pollution	Severe pollution

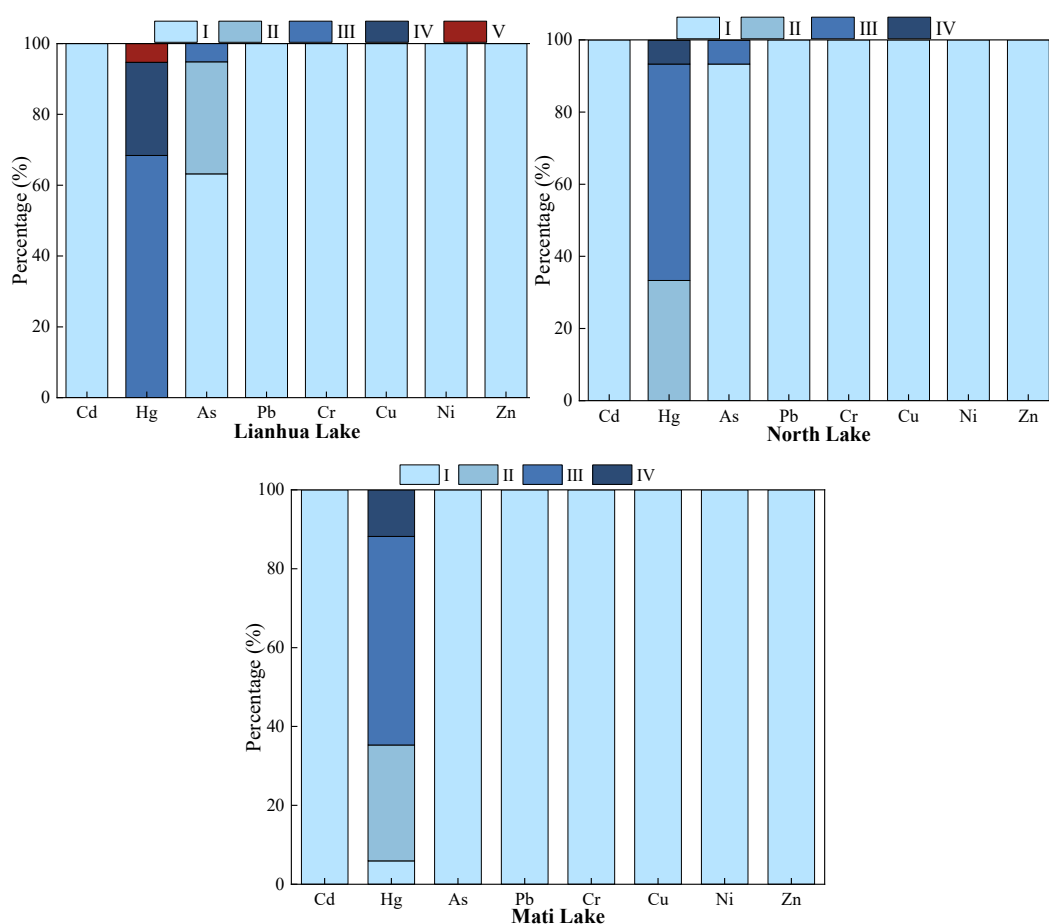
3.3.2. Evaluation of Heavy Metal Pollution in Sediments

Table 6 illustrates that Hg had a relatively high ecological risk in Lianhua Lake, Mati Lake, and North Lake, with ecological contribution rates of 63.3%, 62.9%, and 58.9%, respectively. The E_r^i value for As in the sediments of Lianhua Lake was 30.16, indicating a moderate ecological risk. The individual E_r^i values of the HMs in the three lakes were generally consistent, with the most significant pollution arising from Hg, followed by As. Other elements in each lake exhibited low individual E_r^i , emphasizing that Hg and As are the primary contributors to ecological hazards in the surface sediments in the studied lakes.

Table 6. Potential ecological risk parameters of HMs in the sediments of Lianhua Lake, Mati Lake, and North Lake.

Lake	E_r^i								RI
	Cd	Hg	As	Pb	Cr	Cu	Ni	Zn	
Lianhua Lake	2.51	102.67	30.16	3.34	13.88	4.30	4.04	1.28	162.18
Mati Lake	2.51	67.32	7.77	3.56	15.31	4.79	4.72	0.89	106.87
North Lake	2.97	74.65	12.91	3.58	22.53	4.94	4.23	0.83	126.62

In the North Lake, 100% of the sampling points exhibited medium and high 279 ecological risk levels for Hg, while 6.6% of the sampling sites had a higher ecological risk level for As. For Mati Lake, 94.1% of the sampling sites demonstrated medium and high ecological risk levels for Hg, with approximately 52.9% at the higher ecological risk level and approximately 11.8% at the high ecological risk level. In Lianhua Lake, Hg was present at 100% of the sampling points at high and above ecological risk levels. Among these, approximately 68.4% were at the high ecological risk level, approximately 26.3% were at the higher ecological risk level, and approximately 5.3% were at the extremely high ecological risk level. The proportions of the single-factor potential ecological risk coefficients of metals in the sediments of the three lakes are presented in Figure 4.

**Figure 4.** Proportion of ecological risk coefficient (E_r^i) pollution in sediment HMs; I to V represent increasing levels of potential ecological risk, ranging from mild to severe.

The distribution of the RI of HMs in the surface sediments of the Yangxin County lakes is depicted in Figure 5. The comprehensive RI of the Mati Lake sediments was the lowest at 54.78, whereas that of North Lake sediments was the highest, reaching a maximum value

of 266.58. Lianhua Lake and North Lake had 5.2% and 6.2% of the sampling points at higher ecological risk levels, respectively, while the remaining points were categorized as medium and below. The average RI values for Mati Lake, North Lake, and Lianhua Lake were 106.88, 126.63, and 162.18, respectively, indicating moderate ecological risk levels.

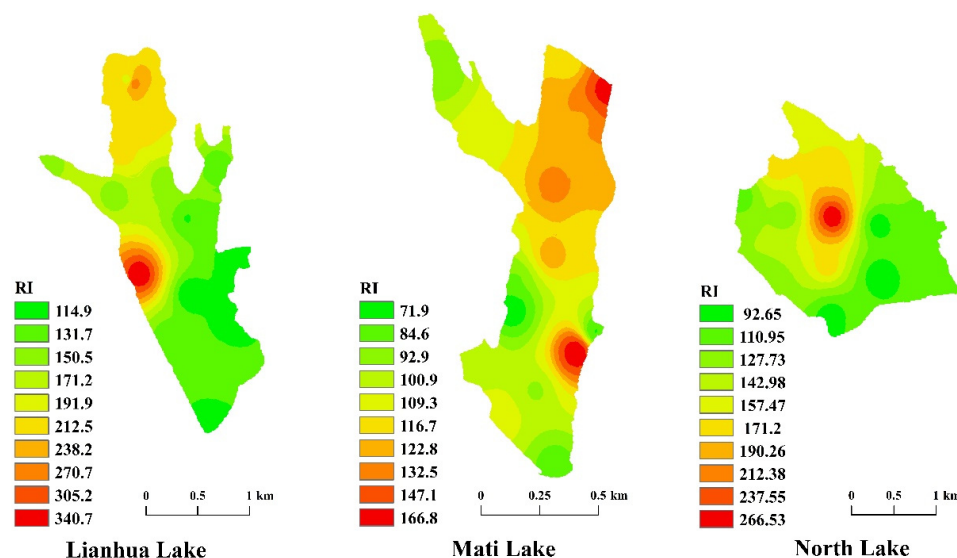


Figure 5. Spatial distribution of the potential E_r^i of HMs in the sediments of typical lakes in Yangxin County. RI, comprehensive potential E_r^i for HMs.

3.4. Relation between Nutrient Salts and HM

Pearson's correlation analysis of the three lakes revealed variations in the correlations between nutrients and HMs (Figure 6) [36]. In Lianhua Lake, there were highly significant correlations between Hg, Cd, TN, and TP, suggesting a potential common nutrient source for Hg and Cd in the sediment. In Mati Lake, the strongest correlation was observed between Ni and Cr ($p < 0.01$, $r = 0.940$), followed by significant correlations between Zn and Cd and Pb, Cr, Ni, and Cu (Figure 6). The North Lake sediments exhibited the highest correlations between Cd and Pb ($p < 0.01$, $r = 0.968$) and Cu and Zn ($p < 0.01$, $r = 0.969$). Additionally, a relatively strong correlation was observed between Ni and Zn, Cu, Pb, and Cd. Notably, the correlation between TN and TP was not significant, suggesting distinct sources for N and P.

The findings indicate the complex relationships between nutrient salts and HMs in the sediments of the three lakes, highlighting the importance of understanding their interactions for effective environmental management.

Both the Bartlett sphericity test ($0.00 < 0.05$) and the Kaiser–Meyer–Olkin measure value > 0.5 indicated that nutrients and HMs were suitable for PCA.

In Lianhua Lake, the two first axes explained 64.0% of the total variance. Additionally, there was a significant correlation ($p < 0.01$) between Cu, Pb, and Cr, indicating similar spatial patterns. High concentrations of HMs in Lianhua Lake were mainly distributed in the southern and northwestern parts of the lake. In Mati Lake and North Lake, Zn had a higher loading in PC1, suggesting a relatively consistent source of HMs in these three lakes, potentially originating from natural sources or similar anthropogenic activities like mining and traffic emissions.

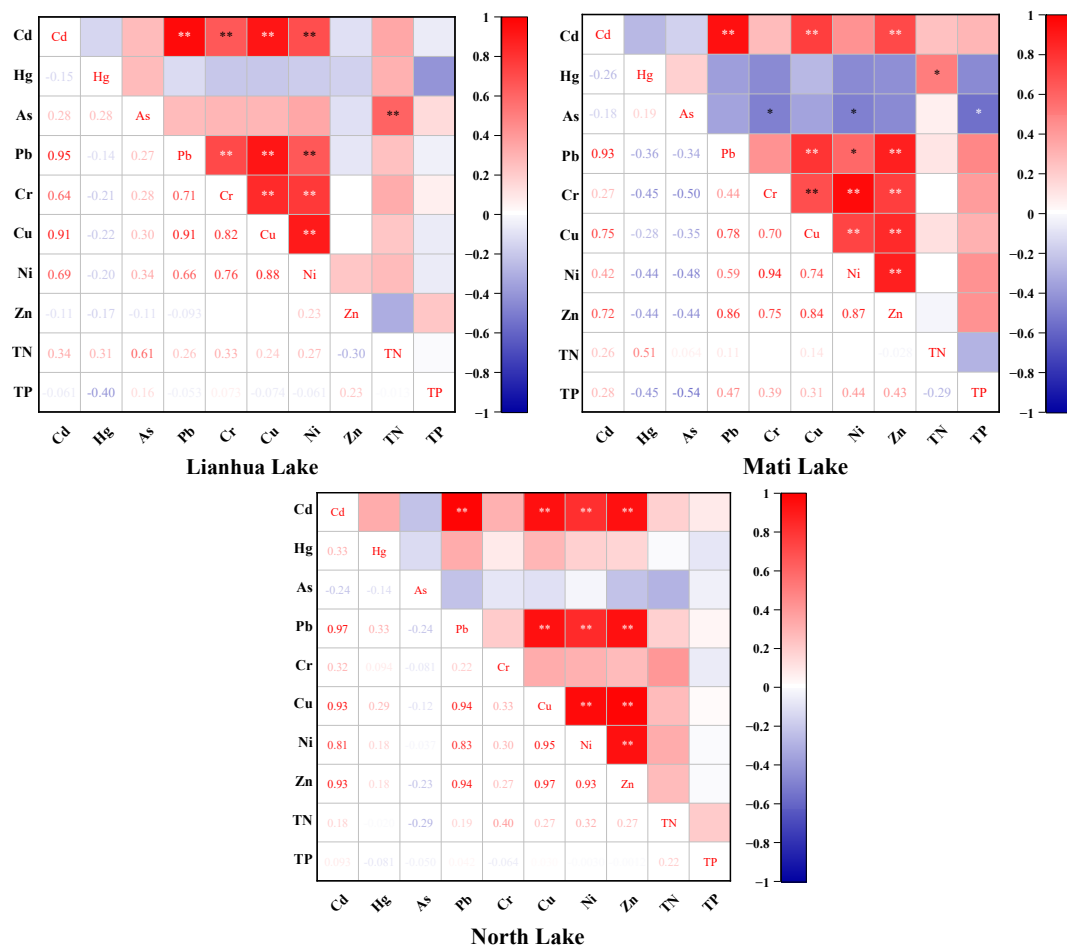


Figure 6. Pearson's correlation between nutrient salts (TN and TP) and HMs present in the surface sediments in Lianhua Lake, Mati Lake, and North Lake. TN, total N; TP, total P. Note: * $p < 0.05$; ** $p < 0.01$.

4. Discussion

4.1. Analysis of Nutrient Salt and HM Distribution Characteristics

This study indicated that a portion of the N and P present in the surface sediments in the Lianhua Lake, Mati Lake and North Lake is potentially derived from external inputs, while the other portion originates from aquatic organisms [6]. Lianhua Lake had the highest comprehensive nutrient pollution index, followed by Mati Lake, which may be related to the locations of the lakes. Lianhua Lake is located in a newly developed urban area in Yangxin County. In recent years, with the migration of the population and the development of buildings, point source pollution from production and domestic emissions has increased. Although wastewater treatment plants have been constructed near urban areas, the incomplete coverage of wastewater treatment network has resulted in an inability to intercept all domestic wastewater [22]. Additionally, in the coastal areas of the lakes in Yangxin County, there are large areas where combined sewer systems are used for wastewater discharge. There are 15 inlets directly discharged into Lianhua Lake and 11 inlets directly discharged into Mati Lake, all of which inevitably affect water quality.

Mati Lake is located in the old urban area of Yangxin County, where the surrounding non-point source pollution is severe. In addition to the direct discharge of domestic sewage into the lake along its shore, there are significant instances of garbage dumping and feedlot farming that contribute to non-point source pollution. Moreover, the vegetation along the shore is relatively uniform, leading to a fragile ecosystem where natural restoration from environmental pollution is challenging to achieve.

Although North Lake is located on the outskirts of the city, with no large residential areas nearby and only some farmland distributed around it, there used to be privately contracted fish farming activities before 2018, involving the use of high-density organic fertilizers. Research has shown that in aquaculture, fish and other organisms can only absorb 30% of the P in their feed, whereas the rest remains in the water or sediment in free or particulate forms [37]. The high TN content in the eastern part of North Lake may be related to coastal fish farming using fencing. This was similar to the findings of Yang et al. [38].

Excessive nutrient levels in the water of the lakes are closely associated with aquatic plants. The periodic decay and decomposition of aquatic plants increase the accumulation of nutrients in the sediment, leading to endogenous nutrient contribution to the water body. When aquatic plants die, their remains settle on the bottom of the lake. During the decomposition process, organic matter breaks down into dissolved nutrients such as N and P, which are then released into the water [39].

The high TN and TP contents in the northern part of Lianhua Lake were largely due to the proximity of the lakeside boardwalk to the park. To enhance the scenic beauty of the lake for tourism purposes, park managers plant aquatic plants, such as lotuses and water lilies, thereby increasing the richness of the lake's landscape. Similar observations have been made in Taihu Lake in China, where the withering of large aquatic plants was observed to impact the increase in nutrients in the water [40]. Additionally, researchers have found that the root systems of aquatic plants regulate the secretion of oxygen and other substances that influence the transport and transformation of P [41]. Therefore, aquatic plants should be harvested before they wither and decay to reduce the accumulation of plant residues in sediments.

The sources of HMs in lakes, in general, include background HMs from the watershed matrix and anthropogenic pollution emissions, with the latter being the primary source [42]. Owing to the inherent characteristics of HMs, which are persistent and difficult to degrade, they tend to accumulate easily in the aquatic sediments, leading to water, sediment, and living organism contamination [43]. The accumulation of HMs is related to local economic development, pollution emissions, and the status of underground resource reserves [44].

The E_r^i results for the HMs in the studied lakes were generally consistent, with the Hg pollution being the most severe, followed by As. This indicates that the high risk of HMs in the watershed was mainly related to Hg and As.

Yangxin County is an important area for mineral resource development in China, and over the past 20 years, there have been a large number of mineral developments and associated transportation activities. Hg originates mainly from the combustion of petroleum products and pesticides, accumulating in sediments through atmospheric deposition and the surface runoff of industrial and transportation waste gases and wastewater [30]. The areas surrounding Lianhua Lake and Mati Lake, characterized by the busiest traffic routes in the urban area of Yangxin County, have witnessed long-term emissions of vehicle exhaust gases settling into lake water through atmospheric deposition [31]. The distribution of roads was closely related to the distribution of Pb, Cu, and Cr in the sediments of Lianhua Lake, confirming that Pb originates mainly from vehicle exhaust emissions and metal smelting emissions, while Cu and Cr are indicative pollutants from fossil fuel combustion [45].

In the northern part of North Lake, pollution mainly originates from agricultural cultivation and aquaculture. Fertilizers often contain HMs, such as Hg, Cd, As, and Pb [46]. These HMs seep into the lake through surface and underground runoff, causing non-point source pollution. Some studies have indicated that the migration capabilities of different HMs are variable. HMs with higher migration capabilities can enter rivers and lakes with surface water flow, whereas those with lower migration capabilities can transfer only small amounts of elements to water or sediment [47]. Pollution in North Lake mainly occurs at the center of the lake, displaying a decreasing trend from the center towards the periphery. Similar trends were observed in Lianhua and Mati lakes. This may be related to the low migration capability of As, as it tends to accumulate in the central areas of lakes and

gradually diminishes towards the outskirts [48,49]. However, the migration of HMs is closely related to the environmental pH, microbial activity, and redox reactions [13].

Aquaculture activities in North Lake, Lianhua Lake, and Mati Lake are essential economic sources for the residents [22]. Feed used in aquaculture contains HMs such as Cd, Cu, and Zn. A large amount of untreated feed is directly released into the water and sediments, and through absorption and excretion by aquatic organisms, these HMs are deposited directly or indirectly into the water, leading to their accumulation [47]. Due to the relatively few inflowing rivers in North Lake and the stable sedimentary environment within it, HMs tend to accumulate in the central part of the lakes.

4.2. Analysis of Pollution Sources

The correlations obtained via PCA (Figure 7) for the three lakes confirmed that Cu, Pb, Cd, and Ni undergo similar geochemical processes and multiple elements contribute to the pollution of the same environment. Except for Cd, the concentrations of the other HMs did not exceed the background values of the soil HMs in Hubei Province [15].

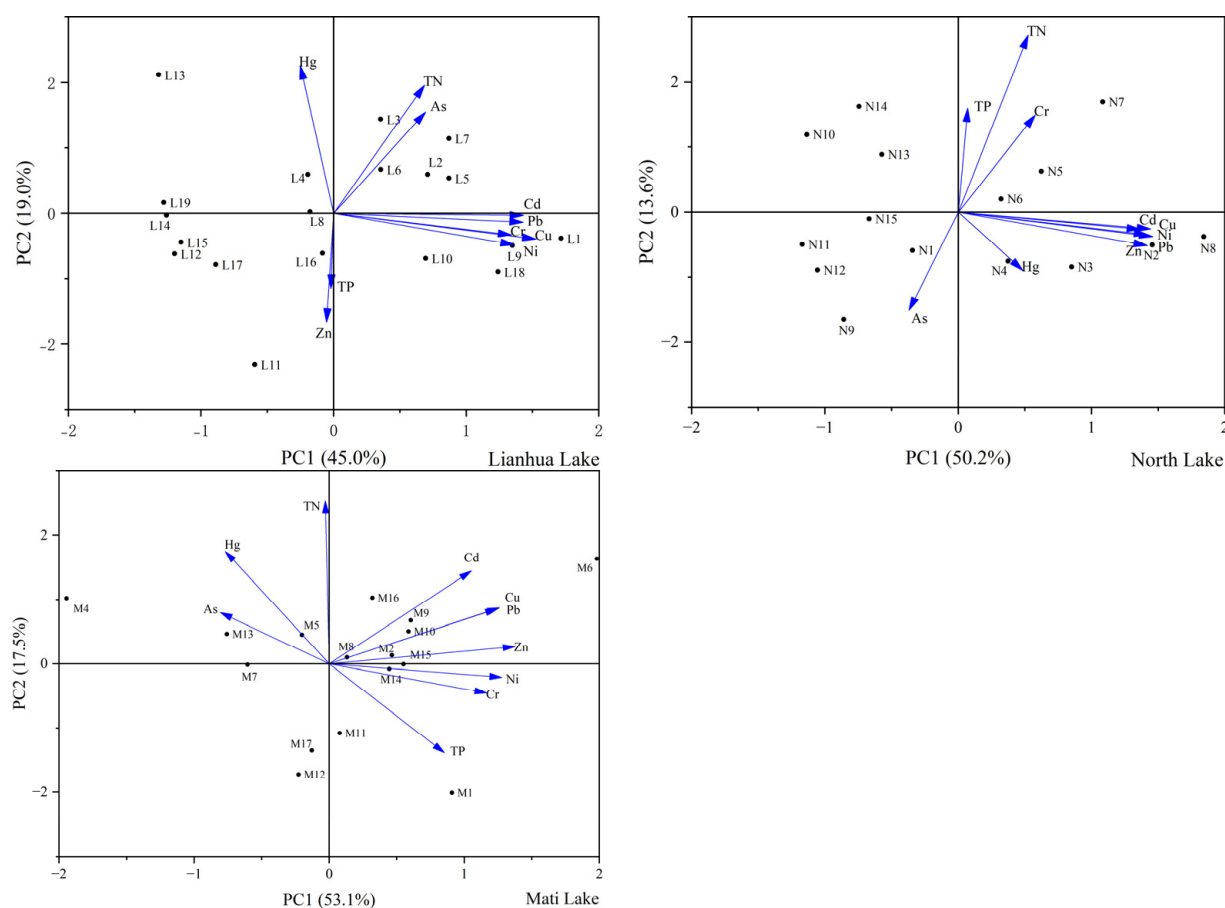


Figure 7. Principal component analysis of TN, TP, and HMs in sediments. Abbreviations: L, Lianhua Lake; M, Mati Lake; N, North Lake.

Yangxin County has a long history of Cu mining, with remnants of human excavations of Cu mines dating back to the Warring States period still preserved today. In 2020, a Cu smelting plant with a capacity of 400,000 t was established in Yangxin County, making Cu the primary element for mineral development in this area [35]. Studies have shown that fishmeal fertilizers used in fish feed contain large amounts of Cd elements [50]. Some researchers believe that Cd is associated with the use of fertilizers in agricultural production [49]. Agricultural activities are important sources of Pb, Ni, and Zn [40]. Agricultural land is still distributed in the northern part of North Lake; therefore, it is believed that the

primary sources of the first principal component in these three lakes are anthropogenic activities, such as mining and metallurgical industries; aquaculture feed; and agricultural fertilizers.

In Lianhua and Mati lakes, Hg and TN displayed high positive loadings on the second principal component, signifying a significant correlation, suggesting a common pollution source. The spatial distribution of Hg in the study area exhibited a clear decrease: a north-to-south concentration decrease in North Lake, higher concentrations in the east compared to the west in Mati Lake, and higher concentrations in the west than the east in Lianhua Lake. This pattern aligns precisely with the distribution of inlets from harbors and channels around the county seat. Research indicates that Hg, commonly found in fossil fuels, is significantly influenced by non-point source pollution [51], correlating with TN, another parameter affected by non-point source pollution. Considering the layout of surrounding industries, transportation routes, and older communities, it can be inferred that Hg and nutrients in the sediment originate from the atmospheric deposition of wastewater and traffic exhaust from the surrounding areas as they had relatively high positive loading, with no significant correlation with other elements (see PCA, Figure 7). Research indicates that As is primarily found in agricultural run-off and industrial effluents. Cr exhibited a relatively high positive loading in North Lake, with no significant correlation with other HMs, differing from Lianhua Lake and Mati Lake. Research also suggests that in the Pearl River Delta region of China, Cr in the soil primarily originates from the weathering of parent rock materials [52]; similar results have been obtained in other countries [53]. Therefore, it can be inferred that Cr in North Lake mainly originated from the soil parent material.

4.3. Comparison of Pollution in Surface Sediments of Different Areas

The sediments of typical lakes in Yangxin County exhibit a relatively high nutrient salt content. The average TP and TN contents in the sediments of the three typical lakes in Yangxin County were higher than those in the middle and lower reaches of the Yangtze River (Table 7), including Dongting Lake, Taihu Lake, Chizhou Lake, Hongze Lake, and Changtan Reservoir in Zhejiang (Table 8). They also exceeded levels in northern lakes such as Qinghai Lake and Shandong Swan Lake and the Tianjin Haihe River but were lower than those in Mulan Lake in Inner Mongolia, Tianmu Lake in Jiangsu, Dianchi Lake in Yunnan, and Wanghu Lake in the same region.

Table 7. A comparison of nutrient salt contents in surface sediments from different lakes and reservoirs.

Study Area	TN (mg/kg)	TP (mg/kg)	Source
Lianhua Lake	2100	1600	Current study
Mati Lake	2230	1430	Current study
North Lake	2520	1360	Current study
Dongting Lake	1029	697	[50]
Taihu Lake	1010	501	[40]
Chaohu Lake	1088	585	[54]
Hongze Lake	1020	580	[55]
Wuliangsuhai Lake	7910	1890	[56]
Qinghai Lake	1800	471	[57]
Swan Lake	850	350	[58]
Haihe River	1012	874	[59]
Tianmu Lake	2598	323	[60]
Changtan Lake	1740	490	[61]
Dianchi Lake	4910	2160	[52]
Wanghu Lake	3122	913	[20]

Table 8. Average HM contents in surface sediments of typical polluted lakes.

Study Area	Cd	Hg	As	Pb	Cr	Cu	Ni	Zn	Source
Lianhua Lake	0.23	0.27	80.54	8.21	39.79	26.41	30.11	107.37	Current study
North Lake	0.27	0.20	34.46	8.80	64.60	30.31	31.53	69.07	Current study
Mati Lake	0.23	0.18	20.74	8.76	43.88	29.44	35.24	74.24	Current study
Cihu Lake	2.48	-	-	177.26	66.39	127.86	33.34	383.42	[31]
Daye Lake	77.13	-	-	134.22	-	650.13	78.46	-	[45]
Wanghu Lake	0.7	-	-	37.1	119.6	62.6	-	165.2	[20]
Qingshan Lake	-	-	-	82.48	59.17	201.16	46.26	300.18	[62]
Dongting Lake	2.88	-	16.58	31.14	93.47	30.57	34.47	121.01	[50]
Daming Lake	0.24	-	5.18	42.89	62.06	46.73	19.37	138.71	[63]
Tangxun Lake	0.66	0.17	12.88	41.6	85.28	51.28	40.49	145.01	[64]
Hongze Lake	0.23	-	16.55	27.2	66.78	25.35	33.89	74.77	[55]
Poyang Lake	0.7	-	-	49.39	132.49	44.89	-	142.79	[65]
Yangcheng Lake	0.45	0.09	15.85	34.02	101.28	66.54	68.72	187.33	[37]
Taihu Lake	0.07	0.09	9.01	34.16	93.69	31.32	-	101.93	[40]
Wuliangsuha Lake	0.43	0.02	3.64	5.86	43.11	53.74	46.33	94.69	[56]

Compared with other lakes in China, the three lakes in Yangxin County exhibited higher concentrations of Hg and As; higher average concentrations of Cd, Cr, Cu, Zn, and Ni; and lower concentrations of Pb. The As content in Lianhua Lake was eight times that of Taihu Lake, and the Hg content was three times that of Taihu Lake.

HM pollution in water bodies is a significant environmental issue [66]. Due to their inherent biotoxicity, they can directly harm the cells and tissues of living organisms, impacting growth and reproduction, ultimately leading to a decrease in population and the decline of ecosystems [67]. As humans occupy the top of the food chain, toxins accumulate layer by layer along the food chain. The occurrence of Minamata disease in Japan illustrates this phenomenon, where local residents consume seafood contaminated with Hg and Cd, marking one of the earliest instances of disease-causing environmental pollution. Therefore, HM pollution of aquatic environments cannot be overlooked.

5. Conclusions

The comprehensive pollution index of nutrients (FF) in all three lakes reached severe pollution levels. The potential E_r^i of the three lakes indicated moderate pollution, with Hg showing the highest pollution level, followed by As and Cd. However, due to the numerous assessment indicators, a detailed analysis of the sources of each pollutant was not sufficiently conducted, like Ni and Zn. As a follow-up to the present study, it is proposed in the future to characterize the main sources of pollution in the area surrounding the lakes and assess the main chemical elements present in the water and sediments.

Geographical location significantly influenced HM pollution, with Lianhua Lake and Mati Lake, which were closer to towns, exhibiting more severe pollution, particularly with higher levels of elements such as Pb, Cu, and Hg. This could be attributed to traffic pollution, domestic sewage, and pesticide residues. In North Lake, the high nutrient content was mainly distributed in the northern and southern parts, corresponding to coastal aquaculture and crop cultivation activities. The stable sedimentary environment of North Lake led to concentrated HM pollution in the central part.

Based on the research results, the following recommendations are proposed for the sustainable development of lake water resources in the area:

1. Optimize sewage discharge pipelines in county towns, strengthen the construction of sewage treatment plants, and ensure that domestic sewage and industrial wastewater meet standards before discharge;
2. Implement regular clean-up of garbage and large aquatic plant residues in the water bodies;

3. Enforce strict control over the scale of aquaculture in the lake area and scientifically plan aquaculture density;
4. Prioritize ecological restoration efforts by constructing artificial wetlands in lake areas and planting adaptable aquatic plants with purification capabilities.

Author Contributions: Conceptualization, X.Y. and M.L.; methodology, X.Y.; software, H.L.; validation, X.Y., Z.H. and H.L.; formal analysis, X.Y.; investigation, Y.C.; resources, X.W. and X.G.; data curation, M.L.; writing—original draft preparation, X.Y.; writing—review and editing, X.G. and X.W.; supervision, G.L.; project administration, X.W. and X.G. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the Open Foundation of Hubei Key Laboratory of Pollutant Analysis and Reuse Technology (Hubei Normal University) (PA220103), the Open Foundation of Research Center for Transformation and Development of Resource depletion Cities (Hubei Normal University) (KF2023Z01), the Science and Technology Research Project of the Education Department of Hubei Province (Q20212505), and the College Students' Innovative Entrepreneurial Training Plan Program (S202310513064).

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: The datasets generated and/or analyzed during the current study are available from the corresponding author on request.

Conflicts of Interest: The authors declare no conflicts of interest.

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