Potential Characteristics and Ecological Risk Assessment of Heavy Metals in Sediments of the Three Gorges Reservoir

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Abstract: In order to understand the spatial distribution and ecological risk of heavy metal pollution in the sediments of the Three Gorges Reservoir, surface sediment samples in six typical tributaries and estuaries were analyzed. The heavy metal content in sediment was detected by ICP-MS (PE Elan DRC II, PerkinElmer, Waltham, MA, USA) after digestion by mixed acid. The data were evaluated and analyzed using multiple methods, including the potential ecological risk index, the geoaccumulation index (I_{geo}), and cluster analysis. The results show that the tributary surface sediments were polluted by heavy metals to a certain extent. Cu, Zn, Pb, and Cr have been enriched in sediments in recent years. The results of I_{geo} show that the pollution degree is as follows: Pb > Cu > Zn > Cr > Cd > Ni > As > Hg. According to the potential ecological risk index, the grades of Yunyang, Xiaojiang, and Xinjin are all medium risk, and Cd has the highest contribution rate. Combined with field investigation data, cluster analysis, and correlation analysis, we conclude that Cu, Zn, and Cr are likely to come from agricultural chemical fertilizers and industrial sewage, while Pb mainly comes from the discharge of ship fuel.

Keywords: sediments; the Three Gorges Reservoir; heavy metals; geoaccumulation index

1. Introduction

Due to the influences of human utilization, industry, and urban development, most water ecosystems have been polluted by heavy metals to varying degrees [1]. Heavy metals are typical pollutants with the characteristics of harmful persistence, significant biological toxicity, and non-degradability, and they are harmful to nature [2,3]. After entering the water environment, heavy metals can quickly deposit into the sediments through complex physical, chemical and biological processes [4,5]. However, with the change of environmental conditions at the sediment–water interface, or due to biological disturbance or resuspension, heavy metals in sediments will be released into...
the water environment. This will cause “secondary pollution” in the water and worsen the water quality [6,7]. Heavy metal pollution in sediments such as the Yangtze River, Hong Kong mangroves, and Dianchi Lake has been a subject of previous research [8–10]. Sediments play the roles of sources and sinks in the migration and transformation of heavy metals in water environments, which is an important basis for the evaluation of heavy metal pollution [11,12].

The Three Gorges Reservoir is the world’s largest water conservancy project. Not only does it have economic and social benefits, such as flood control, power generation, and shipping, but it also facilitates the development and effective use of hydropower resources in the Yangtze River. Moreover, it is conducive to the economic development and ecological protection of the Three Gorges Reservoir environment [13]. Since the impoundment of the Three Gorges Reservoir, the flow velocity in the reservoir has slowed down. As the water level rises, the ability of water body to transfer heavy metals gradually weakens, which has led to continuous accumulation of heavy metals in the sediments of the Three Gorges Reservoir [14]. Related studies have shown that the surface sediments of the Three Gorges Reservoir have been polluted by heavy metals [15,16]. In addition, Yan found that the concentration of heavy metals in estuaries is relatively higher than that in the main stream [17]. There are 32 primary branches of different sizes distributed in the heart of the Three Gorges Reservoir. Therefore, the input pollution of the tributary is also an important factor in the accumulation of heavy metals in sediments in the Three Gorges Reservoir.

Researcher analyzed the concentrations of heavy metals in the sediments of the main stream of the Three Gorges Reservoir in different periods [18,19], but there have been few studies on the tributary sediments. Therefore, this study analyzed the heavy metal (Ni, Zn, Cu, Cd, Cr, Pb, As, Hg) contents of surface sediments in the six important tributaries of the Three Gorges Reservoir and their estuary. The geoaccumulation index and the potential ecological risk index are important methods for evaluating and analyzing the heavy metal content in sediments, as they can intuitively reflect the impact of human activities on heavy metals and the impact of heavy metals on the environment. Thus, this study used these assessment methods to evaluate heavy metal pollution, and cluster analysis and correlation analysis were used to carry out pollution traceability analysis. The results can provide a scientific basis for the management governance and protection of the ecological environment of the Three Gorges Reservoir.

2. Materials and Methods

2.1. Sample Collection

Samples were collected in May 2019 from 12 sampling points along 6 important tributaries and estuaries of the Three Gorges Reservoir (Figure 1). Each sampling point was georeferenced by a portable GPS device. The tributary sampling points were selected to avoid human factors and the main stream sampling points were selected at a distance after the merging of the tributaries to avoid the impact of human factors on the samples. Three representative surface sediment samples were collected by the Peterson grab dredger and mixed evenly. Then, the larger impurities such as gravel, garbage, and plant roots were picked up. After being sealed in a bag, sediment samples were refrigerated in a freezer at −4 °C and transported back to the laboratory for analysis.
2.2. Laboratory Analysis

After natural air-drying, sediment samples were ground and crushed by an agate mortar and sifted by 200 meshes. The sediment samples were accurately weighed to 0.2 g using the method of coning and quartering, and then they were digested on an electric hot plate by mixed acid digestion combined with HNO₃ and HClO₄. After digestion, the volume was fixed to 100 mL with ultrapure water. The concentrations of Ni, Zn, Cu, Cd, Cr, and Pb in the digestion solution were determined by inductively coupled plasma mass spectrometry (ICP-MS, PE Elan DRC II, PerkinElmer, Waltham, MA, USA), and the concentrations of As and Hg were determined by atomic absorption spectrometry (AFS-8500, Beijing HaiGuang instrument Co., Ltd., Beijing, China) [20].

During sample pretreatment and testing, no less than three parallel samples and sediment reference materials (GBW07305) were set to control the accuracy of the entire process, respectively. The relative error for the metal element parallel samples was less than 10%. The recovery rate of heavy metals was 95–110%, and the content of each metal element in the experimental blank was below the detection limit of the instrument.

2.3. Pollution Degree Evaluation Methods

2.3.1. Geoaccumulation Index (Igeo)

The geoaccumulation index (Igeo) is widely used to quantitatively evaluate heavy metal pollution in soil or stream sediment [21]. Through a comparison of the relationship between the content of heavy metals in soil or stream sediment and their geochemical background values, this method can directly reflect the influences of natural and human activities on heavy metal pollution. The calculation formula is as follows:

\[ I_{geo} = \log_2 \left( \frac{C_i}{kB_i} \right) \] (1)

In the formula, Igeo is the geoaccumulation index and Ci and Bi are the measured values and selected background values of heavy metals, respectively. k is a constant, generally 1.5, which may cause a change in background values by diagenesis. The background value of this paper refers to the average content of heavy metals in the sediments of the Yangtze River in the 1990s [18]. According to the value of Igeo, the pollution degree can be divided into 7 grades, from no pollution to serious pollution (Table 1).
Table 1. The classification of I_{geo} and the heavy metal pollution degree.

| Project | I_{geo} |
|---------|---------|
| Series  | Degree  |
| <0      | No pollution |
| 0-1     | Mild |
| 1-2     | Partial |
| 2-3     | Moderate |
| 3-4     | Partial Heavy |
| 4-5     | Heavy |
| 5-6     | Serious |

2.3.2. Potential Ecological Risk Index

The potential ecological risk index which comprehensively considers four influencing factors—the type, content, toxicity level of pollutants in sediments, and the sensitivity of water to heavy metal pollution was proposed by Swedish scholar Lars Hakanson [22]. This method not only reflects the impact of heavy metal pollution on the environment, but also evaluates the potential ecological risk of heavy metal pollution [20]. The calculation formula is as follows:

\[
RI = \sum_{i=1}^{n} E_i T_i C_i B_i \tag{2}
\]

In the formula, RI refers to the comprehensive potential ecological risk index of heavy metals in sediments. \(E_i\) is the potential ecological risk index of a single factor \(i\). \(T_i\) refers to the toxicity response coefficient of a single pollutant (the values of Ni, Cu, Zn, Pb, Cd, Cr, As, and Hg are 5, 5, 1, 5, 30, 2, 10, and 40). \(C_i\) is the pollution index of a single heavy metal. \(C_i\) is the measured value of heavy metals, and \(B_i\) is the background value of the selected heavy metals. To aid with comparison, this background value also refers to the average concentration of heavy metals in the sediments of the Yangtze River in the 1990s [18]. The reference values of the heavy metal potential risk index and the comprehensive potential risk index are shown in Table 2 [23].

Table 2. Ecological risk index and grades of risk intensity.

| Single Pollutant Pollution Index \(E_i\) | Potential Ecological Risk Index RI |
|----------------------------------------|-----------------------------------|
| Threshold Interval | Degree | Threshold Interval | Degree |
| \(E_i < 40\) | Low | \(RI < 150\) | Low |
| \(40 \leq E_i < 80\) | Middle | \(150 \leq RI < 300\) | Middle |
| \(80 \leq E_i < 160\) | High | \(300 \leq RI < 600\) | High |
| \(160 \leq E_i < 320\) | Very high | \(600 \leq RI < 1200\) | Very high |
| \(E_i \geq 320\) | Extremely high | \(RI \geq 1200\) | Extremely high |

2.3.3. Data Processing

The ArcGIS 10.5 (Environmental Systems Research Institute, Inc., Redlands, CA, USA) was used to draw a schematic diagram of the sampling points, and Excel 2016 (Microsoft, Redmond, WA, USA) and Origin 9.0 (OriginLab, Northampton, MA, USA) were used to process and map the heavy metal content. The data of the heavy metal content were analyzed by cluster analysis and principal component analysis using SPSS 25.0 (International Business Machines Corporation, Armonk, NY, USA).

3. Results

3.1. The Concentration Distribution Characteristics of Heavy Metals in the Surface Sediments of the Three Gorges Reservoir

Due to the lack of base quality criteria for heavy metals in sediments in China, the average concentration of heavy metals in the sediments of the Yangtze River in the 1990s and the national second-level standard of soil quality (GB15618-2008) in China were used as background values.

Figure 2 shows that the concentrations of Ni and As barely differed among different sampling sites. The concentrations of Cu, Zn, and Pb in the tributary were significantly higher than those in the
main stream, and the concentrations of these elements in Xiaojiang, Longxi River, and Wushan were higher than in the other sampling sites. Compared with the tributary, the concentration of Cr in the main stream was significantly lower, while the concentration of Hg in Yunyang and Xinjin was higher, and there was no significant difference in other sampling sites.

Figure 2. Cont.
Figure 2. Concentration maps of heavy metals in surface sediments from the Three Gorges Reservoir.

Table 3 shows that compared with the background values of heavy metal concentrations in the sediments of the Yangtze River in the 1990s, the average values of Ni, Cu, Zn, Pb, Cd, Cr, and As were relatively higher. Only the average value of Hg was lower. The highest concentrations of Cu, Zn, Pb, and Cr were respectively 9.6, 5.51, 14.62, and 2.26 times the background values of the Yangtze River sediments. Compared with the national second-level standard of soil quality in China (GB15618-2008), different proportions of Cu, Zn, Pb, and Cd in the surface sediments of the Three Gorges Reservoir exceeded the standard. The Pb content in the different sites was higher than the standard by 66.67%. In contrast with the standard limit of 80 mg/kg, the concentration in Wushan was 394.7 mg/kg. The coefficients of variation for Cr and Ni in the surface sediments of the Three Gorges Reservoir were small, while those of Cu, Zn, Pb, Cd, Hg, and As (37.63–87.68%) were all greater than 36%, indicating a high level of variation [24]. In particular, the coefficients of variation of Cu, Pb, and Hg were all greater than 80%, indicating that the concentrations of these three elements vary greatly at different sampling points, and their distribution is uneven in different tributaries.
Table 3. Distribution of heavy metals in sediments of the Three Gorges Reservoir.

| Element | Minimum | Maximum | Average | Standard Deviation | Coefficient of Variation (%) | Over-Standard Rate (%) | Background Value of Yangtze River Sediment [18] | National Second-Level Standard for Soil Quality a |
|---------|---------|---------|---------|-------------------|----------------------------|------------------------|-----------------------------------------------|-----------------------------------------------|
|         | mg/kg   | mg/kg   | mg/kg   |                   |                            |                        | mg/kg                                         | mg/kg                                         |
| Ni      | 20.45   | 48.40   | 35.17   | 8.36              | 23.77                      | 0                      | 33                                            | 90                                            |
| Cu      | 25.46   | 336.0   | 97.72   | 82.84             | 84.78                      | 33.33                  | 35                                            | 100                                           |
| Zn      | 87.77   | 430.2   | 177.0   | 108.4             | 61.24                      | 16.67                  | 78                                            | 250                                           |
| Pb      | 25.83   | 394.7   | 114.1   | 95.30             | 83.53                      | 66.67                  | 27                                            | 80                                            |
| Cd      | 0.10    | 0.57    | 0.33    | 0.16              | 46.69                      | 16.67                  | 0.25                                          | 0.50                                          |
| Cr      | 87.40   | 185.1   | 140.3   | 38.62             | 27.54                      | 0                      | 82                                            | 300                                           |
| As      | 5.25    | 18.29   | 10.08   | 3.80              | 37.63                      | 0                      | 9.6                                           | 25                                            |
| Hg      | 0.01    | 0.15    | 0.05    | 0.04              | 87.68                      | 0                      | 0.08                                          | 0.50                                          |

a The national second-level standard of soil quality in China (GB15618-2008).
3.2. Evaluation of Pollution Degree of Heavy Metals in the Surface Sediments of the Three Gorges Reservoir

3.2.1. Geoaccumulation Index (Igeo)

Figure 3 shows the geoaccumulation index box chart of Ni, Cu, Zn, Pb, Cd, Cr, As, and Hg. It can be seen that the geoaccumulation index of Ni in each sampling site was less than 0, indicating no pollution. One sampling site of As was between 0 and 1, indicating mild pollution. Although 41.66% and 16.67% of sampling site of Cd and Hg were mild pollution, the average value was less than 0. This means that their pollution degree in the Three Gorges reservoir is no pollution. According to the Igeo grade and heavy metal pollution degree in Table 1, the Pb in the surface sediments of the Three Gorges Reservoir was found to be up to 3.28, indicating partial heavy pollution. This result is consistent with the concentration analysis. Although some of the sampling sites of Cu, Zn, and Cr indicated mild or partial moderate pollution, their average geoaccumulation index was found to be between 0 and 1, indicating mild pollution. However, the average geoaccumulation index of Pb was found to be 1.16, and most of the sampling sites were partial moderate pollution or above. The result shows that the heavy metal pollution of Pb in the sediments of the Three Gorges Reservoir has been becoming more and more serious. Pb is an indicator of industrial pollution, which mainly comes from the combustion of coal and crude oil, motor vehicle exhaust emissions, and so on [25]. To sum up, the pollution degree of heavy metals in the surface sediments of the Three Gorges Reservoir is as follows: Pb > Cu > Zn > Cr > Cd > Ni > As > Hg.

![Figure 3. Geoaccumulation index of heavy metals in Three Gorges Reservoir.](image)

3.2.2. Potential Ecological Risk Index

The potential ecological risk index of heavy metals in sediments of the Three Gorges Reservoir is listed in Table 4. According to the data, the average values of all heavy metals were lower than 40, indicating that all heavy metals in the Three Gorges Reservoir are present at low pollutant levels. Additionally, the order of potential ecological risk index values of heavy metals was Cd > Hg > Pb > Cu > As > Ni > Cr > Zn. In addition, the maximum values of Pb, Cd, and Cu were found to be between 40 and 80, indicating that they are all at low pollution levels in these sampling sites. The result is similar to that obtained with the geoaccumulation index.
Table 4. Potential ecological risk assessment results of heavy metals in sediments of the Three Gorges Reservoir.

| Project | Potential Ecological Risk Index Ei |
|---------|----------------------------------|
| Ni      | Cu | Zn | Pb  | Cd | Cr | As | Hg  |
| Average | 5.33 | 13.96 | 2.27 | 21.13 | 39.85 | 3.42 | 10.50 | 25.38 |
| Minimum | 3.10 | 3.64 | 1.13 | 4.78 | 11.77 | 2.13 | 5.47 | 3.63 |
| Maximum | 7.33 | 48.00 | 5.52 | 73.08 | 68.30 | 4.51 | 19.05 | 75.29 |

It can be seen from Figure 4 that the potential ecological risk index grades of Yuyang, Xiaojiang, and Xinjin were all found to be medium risk, and the highest contribution rate of Cd was 38.88%, 37.29%, and 14.73%, respectively. Combined with the data shown in Table 4, we can draw the conclusion that the risk of Cd pollution in the Three Gorges Reservoir should be taken seriously into account.

![Potential ecological risk assessment results of heavy metals in sediments of the Three Gorges Reservoir.](image)

Figure 4. Evaluation results using the comprehensive potential ecological risk index.

4. Discussion

4.1. Distribution of Heavy Metal Pollution in Surface Sediments of the Three Gorges Reservoir

According to the data shown in Figure 2, the concentration of heavy metals in the tributary surface sediments of the Three Gorges Reservoir was found to be higher than that in the estuary, which is consistent with previous studies. The tributary is narrower and slower than the main stream. The heavy metals are more likely to be deposited in the sediment, which leads to the water quality in tributaries often being worse than that in the main streams. In addition, this study found that the concentration of Pb at the Wushan site reached 394.64 mg/kg, which is much higher than the national second-level standard of soil quality of 80 mg/kg. Combined with the environmental status of the sampling sites, we can see that Wushan is the river mouth of the Daning River. There are tourism wharfs known as the Little Three Gorges in the downstream area, where all kinds of cruise ships converge. A large amount of Pb produced by fuel combustion is deposited in the sediment, which leads to the serious excessive concentration of Pb at the Wushan site.

In addition, it is clear from Figure 2 that the pollution of Cu and Cr in the sampling site of the Xiaojiang River is serious. It may have been caused by the construction of a small hydropower station in the upper reaches of the Xiaojiang River. The impoundment of the hydropower station deposits sediment in the water, leading to a trend of accumulating heavy metals.
Compared with the early research in the Three Gorges Reservoir (Table 5), the concentrations of Cu, Zn, Pb, and Cr in sediments have tended to be enriched in recent years. The contents of Ni and As were similar to those in 1999, indicating that these two heavy metals have undergone little change in sediments. In addition, the contents of Cd and Hg were lower than those found in related studies, which may have been caused by the Western development policy. Wanzhou has gradually become a good deep-water port in the Three Gorges reservoir, and the rapid development of the salt chemical industry, food processing, and building materials industry has changed the concentrations of heavy metals in sediments in the Three Gorges Reservoir, causing them to exceed the standard.

Table 5. Temporal variation of heavy metals in sediments in the Three Gorges Reservoir over the last 20 years (mg/kg).

| Year   | Ni    | Cu    | Zn    | Pb    | Cd    | Cr    | As    | Hg    |
|--------|-------|-------|-------|-------|-------|-------|-------|-------|
| 1999 [18] | 36.94 | 62.54 | 160.6 | 25.71 | 0.27  | 145.2 | 2.78  | 0.42  |
| 2001 [26] | 34.80 | 35.35 | 90.29 | 53.41 | 0.18  | 63.01 | 5.80  | -     |
| 2008 [19] | 46.81 | 76.03 | 137.6 | 59.40 | 0.75  | 86.31 | 18.07 | 0.11  |
| 2010 [27] | 40.20 | 40.80 | 93.00 | 14.70 | 0.66  | 75.20 | -     | -     |
| 2017 [16] | 44.45 | 61.24 | 164.5 | 60.42 | 0.98  | 100.4 | -     | -     |
| This study | 35.17 | 97.72 | 177.0 | 114.1 | 0.33  | 140.3 | 10.08 | 0.05  |

4.2. Sources of Heavy Metals in the Three Gorges Reservoir

According to the analysis of the geoaccumulation index, Cu, Zn and Cr are present in mild pollution, while Pb is a partial moderate pollution. This is the same result as that found in the other studies in the Three Gorges Reservoir in previous years. This result indicates that Pb not only tends to be enriched, but it also causes some harm to the sediments of the Three Gorges Reservoir to a certain extent. Pb is indicative of industrial pollution and mainly comes from the combustion of coal and crude oil, motor vehicle exhaust emissions, and so on. Cd is indicative of agricultural pollution and is considered to be a landmark element for the use of chemical fertilizer [28–30]. A large amount of cultivated land was inundated when the Three Gorges reservoir impoundment reached 175 m, and the Yangtze River has been polluted by Cd for a long time [10]. This is the same as the results shown in this experimental analysis.

According to the analysis of the potential ecological risk index, the average number of heavy metals in sediments in the Three Gorges Reservoir is lower than 40, which indicates that they all low pollution levels. The difference between the two results is mainly due to the different emphasis of the two methods. The geoaccumulation index mainly focuses on the pollution status of heavy metals in water sediment, while the potential ecological risk index comprehensively considers four influencing factors: the type, content, toxicity level of pollutants in sediments, and the sensitivity of water to heavy metal pollution. Therefore, in the assessment of river heavy metal pollution, these methods must be organically combined in order to comprehensively and scientifically evaluate the level of heavy metal pollution [31]. Cd is the main contributor to pollution at the three sampling sites of Yunyang, Xiaojiang and Xinjin, which is consistent with the results of the previous analysis.

The metal elements in sediments are mainly affected by natural weathering and human activities in the basin. Additionally, the degree of influence is determined by the types of water metal elements present. The correlation analysis of the heavy metal content of different elements in sediment can assist us when determining the sources of pollution in the reservoir area. Consequently, this provides a certain reference for the treatment of heavy metal pollution in the reservoir.

Cluster analysis is a branch of mathematics that divides data into groups or classes according to certain rules. By analyzing the similarity and distance relationship between pollution factors, it provides assistance when preliminarily judging the types of pollution sources. The commonly used clustering methods are two-step clustering, K-means clustering and systematic clustering. In this study,
the systematic clustering method was used to analyze the heavy metal pollution sources of stream sediments in the Three Gorges Reservoir.

According to the Figure 5, the monitored heavy metals can be divided into three categories. Each category represents a kind of pollution source, and the heavy metals under this category represent similar pollution sources. The first category includes Cd, Hg, As, Ni, which are common in agricultural chemical fertilizers, pesticides, and other types of non-point source pollution, metallurgical manufacturing, house demolition, rock and geological changes, and so on. During the construction of the Three Gorges Reservoir, millions of people were moved and a large number of houses, cultivated land, and so on became submerged. Thus, a large number of these elements were released into the water. The second category includes Cu, Cr, and Zn, which are likely to come from the same pollution source. These three elements usually come from the cement industry, metallurgy, and the use of agricultural chemical fertilizers and pesticides. Combined with the environmental conditions of tributaries in the Three Gorges Reservoir, we can predict that the heavy metals in sediment mainly come from chemical fertilizers used in the reservoir area, sewage discharged by industry, and mining.

![Figure 5. Heavy metal spectrum of sediments in the Three Gorges Reservoir.](image)

5. Conclusions

An analysis of heavy metals in the surface water sediments of the Three Gorges Reservoir found that their concentrations are all affected by human factors to varying degrees. The concentrations of Cu, Zn, Pb, and Cr in the sediments have been enriched in recent years. Cu, Zn, and Cr are mild pollution, while the geoaccumulation index of Pb reaches 1.16, indicating partial moderate pollution. According to the comprehensive potential ecological risk index, the potential ecological risk index of each heavy metal is ranked Cd > Hg > Pb > Cu > As > Ni > Cr. The cluster analysis shows that three elements (Cu, Cr, and Zn) may come from the same pollution source, with the main sources being the cement industry, metallurgy, and the use of agricultural fertilizers and pesticides. In summary, Cu, Pb, and Zn are more harmful in the surface sediments of the Three Gorges Reservoir, and it is necessary to limit their sources. This analysis can provide a reference for the governance of the Three Gorges Reservoir.

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