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Spatial Distribution, Risk Index, and Correlation of Heavy Metals in the Chuhe River (Yangtze Tributary): Preliminary Research Analysis of Surface Water and Sediment Contamination

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Abstract: This comprehensive study aimed to evaluate the water quality and sediment contamination in the Chuhe River in Nanjing. The spatial assessment of 10 samples collected in September highlighted that, in surface water, Copper (Cu) > Nickel (Ni) > Zinc (Zn) > Chromium (Cr) > Lead (Pb) > Arsenic (As) > Cadmium (Cd) > Mercury (Hg), whereas in sediments, Zn > Cr > Cu > Pb > Ni > As > Cd > Hg. The coefficient of variation (CV) for Ni and Zn in surface water was >15, whereas As, Cu, Pb, and Ni had a CV that was higher than 15 in sediments, indicating variability in contamination sources. The Pollution Load Index values ranged between 2.16 and 3.05, reflecting varying contamination levels across samples. The Geoaccumulation Index data also showed moderate-to-considerable contamination, especially for elements such as Cd and Cu. Correlation analyses in water and sediments unearthed significant relationships, with notable links between Cu and Pb in the water and strong correlations between As and Cu and between Cr and Ni in sediments. In sediments, Total Nitrogen and Phosphorus were significantly correlated with As, Cu, Pb, and Ni. The Potential Ecological Response Index for sediments indicated that they are at medium to high risk (307.47 ± 33.17) and could be potentially detrimental to aquatic life in the tributary. The tributary, influenced by agricultural runoff, residential areas, and other anthropogenic activities, showed that despite Nemerow pollution index values for water samples being below 1, sediment analysis indicated areas of concern. Principal Component Analysis (PCA) was conducted to identify the potential sources of heavy metal contamination. In surface water, shared negative loadings on PC 1 (60.11%) indicated a unified influence, likely from agricultural runoff, while PC 2 (14.26%) revealed additional complexities. Sediments exhibited a unique signature on PC 1 (67.05%), associated with cumulative agricultural impacts, with PC 2 (18.08%) providing insights into nuanced factors, such as sediment composition and dynamic interactions. These findings offer a complete insight into the Chuhe River tributary's condition, underlining the urgency for ongoing monitoring and potential remediation measures.

Keywords: Yangtze tributary; metal concentrations; Nemerow pollution index; Geoaccumulation Index (Igeo); Pollution Load Index (PLI)

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1. Introduction

Heavy metal pollution in freshwater ecosystems is a universally pressing concern, with the ramifications being especially pronounced in regions undergoing rapid industrial and agricultural expansion. Despite their natural existence in the environment, these freshwater ecosystems serve diverse functions, including water supply, agricultural irrigation, flood

control, climate regulation, aquaculture, tourism, and biodiversity preservation [1]. Despite exponential economic growth, China faces escalating heavy metal concentrations in its freshwater sources, primarily attributed to urbanization, industrial activities, and modern agricultural practices [2,3].

Historically, natural traces of heavy metals in Chinese freshwater systems have been attributed to the country's rich and diverse geological features. However, research has highlighted that, in contemporary times, several water bodies are recording concentrations that dangerously surpass national safety thresholds [4,5]. Such alarming trends are not isolated but echo across industrial hubs, such as the Yangtze River Economic Belt and the Pearl River Delta. Here, industries, agriculture, and expanding urban sprawls intersect, producing a cocktail of pollutants that find their way into the aquatic systems [6–8].

China's extensive network of lakes and rivers, spanning 82,642 km^2, intertwines with the nation's socioeconomic and cultural fabric, serving agricultural, industrial, and recreational purposes. However, heavy metal discharges, intensified by agricultural activities, pose a significant threat. Chemical fertilizers, pesticides, and herbicides contribute to metal leaching into water systems, with Cadmium from phosphate fertilizers and Arsenic from pesticides being prominent concerns [9–12]. These contaminants, upon entering water bodies, often undergo intricate interactions. Research has shown that they can bind with organic runoffs from agricultural landscapes, potentially amplifying their toxicity and influencing their behavior in aquatic habitats [13–15]. Multiple researchers have outlined that peak agricultural seasons, coinciding with monsoons, can substantially influx these metals into rivers and lakes, creating episodic challenges for water management systems [16,17].

The complexity deepens as heavy metal dynamics in freshwater systems are non-linear, involving intricate interactions with organic runoffs and seasonal variations, particularly during peak agricultural seasons and monsoons. The Chuhe River in Nanjing exemplifies these challenges, as it experiences periodic heavy metal concentrations surging amidst urban and agricultural landscapes, reflecting the broader issues that Chinese rivers face [6,18–20].

In this context, we conducted an investigation into eight specific heavy metals—Arsenic (As), Mercury (Hg), Chromium (Cr), Nickel (Ni), Copper (Cu), Zinc (Zn), Cadmium (Cd), Lead (Pb), and Manganese (Mn)—in the surface water and sediment of the tributary connecting the Chuhe River and the Changjiang River. This study had three primary objectives: firstly, to elucidate the distribution patterns of these heavy metals within the water and sediment; secondly, to assess the extent of pollution and ecological risks associated with these heavy metals in the aforementioned aquatic environments; thirdly, to determine the relationship between the heavy metals and physicochemical parameters to identify the potential sources of the heavy metals. These comprehensive analyses and assessments are pivotal in showcasing this tributary's heavy metal pollution and ecological risk index issues.

2. Materials and Methods

2.1. Sampling Site

The sampling site, approximately 22 km long, is a tributary part of the Chuhe River that connects with the Yangtze River across Nanjing City. This tributary is severely exposed to five significant outlets that discharge agricultural runoff while serving as a water source for those agricultural lands. Residents were found to be swimming and fishing in this tributary. We collected 10 surface water samples (Table 1) in HDPE and glass bottles. The glass bottle samples were used for heavy metal analysis. Ten sediment samples were taken using a stainless-steel sediment sampler in the same location as the surface water sample. The sediment samples were stored in a ziplock bag and glass bottles. The samples were collected in triplicates. All samples were transported back to the lab within 2 h for physicochemical and heavy metal analysis. The samples were collected during September 2021.

Table 1. Sampling sites and the landmarks associated with them.

Sample	GPS	Landmark
1	32°14′55″ N 119°02′57″ E	Branching off from the Yangtze River
2	32°15′19″ N 119°01′48″ E	A pipe connecting to agricultural discharge is seen.
3	32°14′59″ N 119°00′32″ E	Normal
4	32°15′18″ N 119°59′23″ E	A pipe connecting to agricultural discharge is seen.
5	32°15′08″ N 119°57′32″ E	Normal
6	32°14′55″ N 119°56′02″ E	A pipe connecting to agricultural discharge is seen.
7	32°14′31″ N 119°54′57″ E	Normal
8	32°13′36″ N 118°55′20″ E	Closer to residential communities, exposed to household discharge.
9	32°12′33″ N 118°55′38″ E	Closer to residential communities, exposed to household discharge.
10	32°11′59″ N 119°56′17″ E	Reconnecting with Yangtze River.

2.2. Water Quality and Sediment Nutrient Assessment

The physicochemical parameters, such as pH, temperature, Total suspended solids (TSS), Chemical oxygen demand (COD), Total Nitrogen (TN), Total Phosphorus (TP), and Chlorophyll-α (Chlα), were measured based on the protocol mentioned by [21,22]. The values of organic matter (OM), TP, and TN in sediments were determined using the potassium dichromate volumetric method, molybdenum blue spectrophotometry, and the semi-micro-Kjeldahl method [23–25].

2.3. Heavy Metal Analysis

We measured eight heavy metals in surface water and sediment samples: Arsenic, Mercury, Chromium, Nickel, Copper, Zinc, Cadmium, Lead, and Manganese. We followed the pre-treatment protocol mentioned by [3,12] to prepare the samples for heavy metal quantification. All of the chemical reagents used were of a ≥99% purity grade. Upon collection, surface water samples were immediately acidified using nitric acid to attain a pH below 2, ensuring the stability of the metal concentrations. After this, filtration through a 0.45 µm membrane was performed to remove any particulates. For comprehensive metal analysis, 50 mL of the surface water sample underwent an acid digestion process, primarily using a blend of 4 mL nitric and 1 mL hydrochloric acids. After digestion, the samples were stored in a cool, dark place until the analysis time. Conversely, for sediment samples, they were oven-dried at temperatures around 35 °C. Post-drying, samples were sieved to segregate fine particles and then thoroughly grounded to achieve a fine, homogeneous powder consistency. The powdered sediment underwent acid digestion similar to surface water samples. Post-digestion, samples were filtered to eliminate any residual particulates, and the resultant solution was preserved until analysis. All heavy metals were quantified using Atomic Absorption Spectroscopy (ZEEnit 700P, Analytik Jena GmbH + Co. KG, Jena, Germany). The method was refined through the careful selection of optimal atomic lines for the following elements: Pb (283.3 nm), Hg (253.7 nm), Cd (228.8 nm), Zn (213.9 nm), As (193.7 nm), Cr (357.9 nm), Cu (324.7 nm), and Ni (232 nm).

2.4. Quality Assurance

Quality assurance for the sediment samples involved the use of certified reference materials (GBW07312) provided by the Institute of Geophysical and Geochemical Exploration, Chinese Academy of Geological Sciences [26]. To ensure accuracy, analytical reagent blanks were incorporated with every batch of digested samples and analyzed in the same manner to account for background interference. Each sample underwent triplicate measurements to evaluate both precision and accuracy. Results for the reference materials exhibited variabil-

ity within 10%, and the relative standard deviations (RSDs) for triplicate samples remained under 10%. The detection limits for various elements were as follows: 0.01 mg/Kg for Cd, 0.05 mg/Kg for Cu, 0.1 mg/Kg for Pb, 0.2 mg/Kg for Zn, 0.4 mg/Kg for Ni, 0.001 mg/Kg for Hg and As, and 0.25 mg/Kg for Cr (VI). Recovery rates for Cd, Cr (VI), Cu, Pb, Zn, Ni, Hg, and As ranged from 97% to 102.3%, 95.2% to 103.7%, 97.2% to 102.8%, 95.1% to 105.7%, 91.2% to 102.4%, 98.3% to 105.8%, 94.3% to 104.6%, and 95.4 to 104.9%, respectively.

2.5. Statistical Analysis

Analysis of variance (ANOVA) was performed to assess the distribution of heavy metals in surface water and sediment samples. Pearson correlation at a 95% confidence interval was performed using GraphPad Prism 10 to explore the relationship between physicochemical variables (pH, temperature, TSS, COD, TN, TP, and Chlα) and heavy metals in surface water samples, nutrient pollutants (TN, TP, and OM), and heavy metals in sediments and between heavy metals in surface water and sediment samples. Principal Component Analysis (PCA) was used to assess the interrelationships between heavy metal concentrations in surface water and sediment samples, aiming to elucidate potential sources of these metals. PCA transforms the original variables into a new set of uncorrelated variables termed principal components based on eigenvalue. This transformation facilitates dimensionality reduction while preserving the essential information inherent in the data. The coefficient of variation (CV) was calculated to assess each heavy metal variation among surface water and sediment samples. The bar graphs and CV were calculated and generated using Microsoft Excel 2019.

2.6. Heavy Metal Index Assessment

We calculated the Pollution Load Index (PLI), Nemerow pollution index, Geoaccumulation Index (Igeo), and Potential Ecological Risk Index (PERI) to interpret the impact of the heavy metals in sediment [27,28].

The Geoaccumulation Index (Igeo) was determined using the formula originally introduced by [29],

$$\text{Igeo} = log_2^{(\text{Cn}/1.5\text{Bn})}$$

where Cn represents the concentration of the metal (n) found in the sediment samples, Bn signifies the background value for the respective metal (n), and a factor of 1.5 accounts for background matrix adjustments owing to lithogenic factors, as outlined by [30] (Salomons and Förstner, 1984). The Igeo classification system proposed by [29] was adopted to assess the contamination level (Table S1). The sediment Bn value used for this study was adopted from [31] (Table S2).

2.7. Pollution Load Index (PLI)

It is a comprehensive indicator to evaluate the extent of soil contamination or sediments [4]. It was determined by taking the nth root of the product of contamination factor (CF) values according to the following formula:

$$\text{CF} = \text{Co}/\text{Cn}$$

Co represents the concentration of the metal (n) found in the sediment samples, and Bn signifies the background value for the respective metal (n) (Table S2) [9,31,32].

$$\text{PLI} = (\text{CF1} \times \text{CF2} \times \text{CF3} \times \text{CF4} \times \text{CF5} \times \text{CF6}. \ldots . \ldots \times \text{CFn})1/\text{n}$$

CF is the contamination factor, and "n" is the number of metals.

2.8. Potential Ecological Response Index (PERI or RI)

It provides the ecological risk of a freshwater or aquatic ecosystem that could harm aquatic organisms and overall health [17,18,33] (Table S4).

$$\text{Eri} = \text{CFn} \times \text{Tri}$$

Eri represents the single index of ecological risk factors, and Tri represents the toxic response factor.

Toxic response factor for the corresponding heavy metal (Table S3) is shown below.

The Potential Ecological Response Index (PERI or RI) is calculated based on the following formula:

$$\text{RI} = \sum \text{ERI}$$

2.9. Nemerow Pollution Index (NPI)

Several authors commonly used NPI to assess surface water quality based on five classes (Table S5) [34,35].

It was calculated based on the formula shown below:

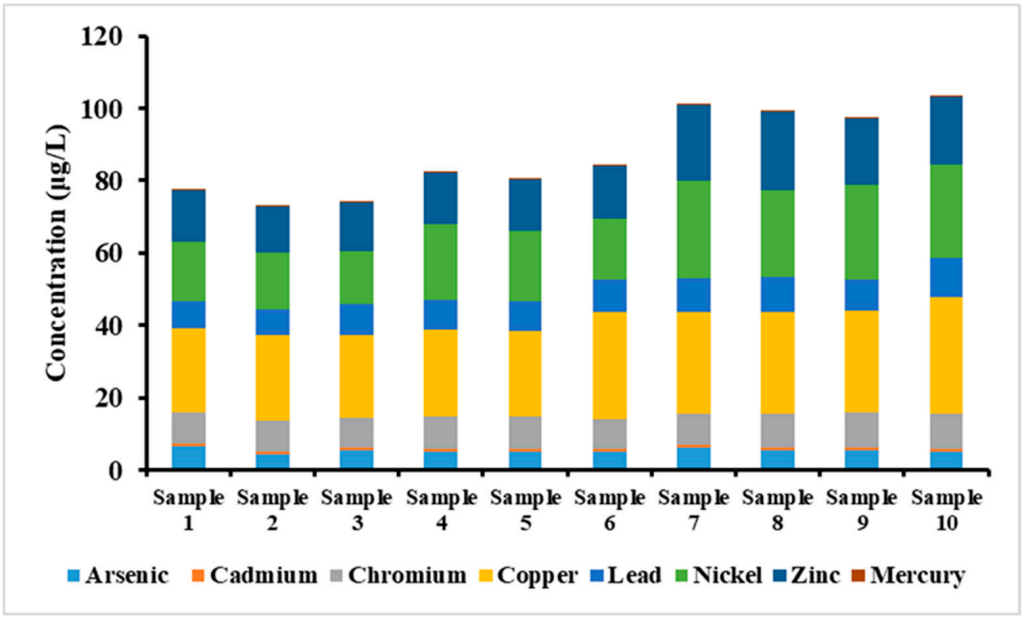
$$\text{NPI} = \sqrt{} (\max \text{Pi})2 + (\text{Pi})2/2,$$

where NPI is the Nemerow pollution index, max Pi is the maximum single pollution index among the pollutants in the water sample, and Pi is the average mean of single pollution indexes among all the pollutants in the water sample.

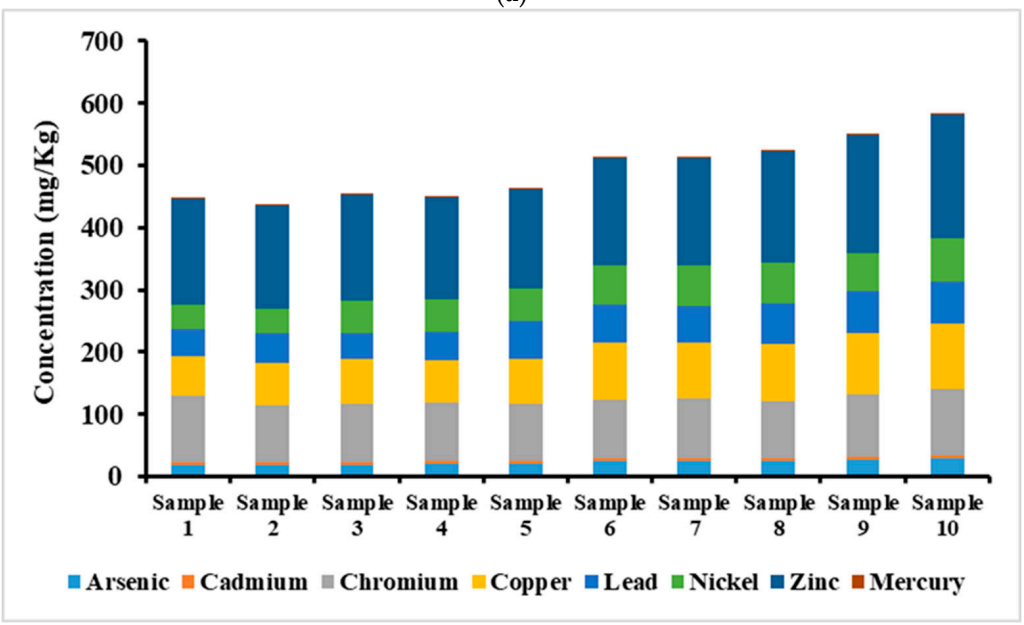
3. Results and Discussion

3.1. Spatial Assessment of Heavy Metals in Surface Water and Sediment

The concentration of heavy metals in surface water among all samples indicated Cu (26.9 ± 2.3) and Ni (21.2 ± 3.9) as the most dominant heavy metals (Figure 1a), whereas in sediments, it was Zn (174.3 ± 10.9) and Cr (97.05 ± 5.4) (Figure 1b). The data suggest a noticeable increase in the concentrations of most heavy metals in sediments and surface water from Sample 1 to Sample 10, indicating a possible contamination gradient across the sampled area. Notably, the levels of Cu and Ni for surface water and As and Cd for sediments increased significantly from Sample 1 to Sample 10. Heavy metal concentrations, including Arsenic, Cadmium, Copper, Nickel, Mercury, and Zinc, exhibited notable differences across the surface water samples, as indicated by the analysis of variance (ANOVA) results ($p < 0.05$) (Table 2). Heavy metal concentrations, including Arsenic, Chromium, Copper, and Zinc, exhibited notable differences across the sediment samples, as indicated by the analysis of variance (ANOVA) results ($p < 0.05$) (Table 3). These variations suggest spatial heterogeneity in heavy metal distribution across the sampled area. This could indicate an external source of contamination, possibly due to anthropogenic activities, such as industrial discharges, agricultural runoff, or improper disposal of hazardous waste [2,36,37]. The dominance of Cu and Ni could be due to stormwater and agricultural runoff, particularly from roads, and the use of copper-containing pesticides and fertilizers in agriculture can lead to Copper in river water. Nickel can also be found in certain fertilizers [2,25,38]. Since this tributary is exposed to agricultural runoff, this could possibly explain why Cu and Ni were found to be dominant.



(**a**)



(**b**)

Figure 1. (**a**) Heavy metal concentrations in surface water. (**b**) Heavy metal concentrations in sediments.

The hierarchy of heavy metal concentration in surface water is as follows: Cu > Ni > Zn > Cr > Pb > As > Cd > Hg. For sediments, it is the following: Zn > Cr > Cu > Ni > Pb > As > Cd > Hg. A medium coefficient of variation (CV) was observed for Ni (21.7) and Zn (19.5) in surface water (Figure 2), while for sediments, it was observed for Cu (16.88), Ni (17.9), Pb (16.2), and As (15.58) (Figure 3). The medium variations of heavy metal concentrations found in the surface water and sediments indicated potential variations in their origins, possibly influenced significantly by human or anthropogenic activities [39,40]. All the other heavy metals had a CV of <15, indicating no minor or significant changes in heavy metal variation.

Table 2. Results from ANOVA display the distribution of a heavy metal among all surface water samples. Groups with the same letter are not statistically significant.

Surface Water	Range (Sample 1 to Sample 10)	Mean ± SD (Mean of Combining Data Sample 1 to Sample 10)
Arsenic	4.066~6.900	5.452 ± 0.658 e
Cadmium	0.600~0.980	0.712 ± 0.079 f
Copper	21.790~32.880	26.276 ± 3.416 a
Lead	6.652~11.791	8.688 ± 1.214 d
Chromium	7.746~10.433	8.934 ± 0.709 d
Nickel	12.694~29.244	20.747 ± 4.877 b
Mercury	0.150~0.215	0.177 ± 0.150 f
Zinc	11.675~24.330	16.316 ± 3.496 c
pH	7.3~8.1	7.6 ± 0.2 C
Temperature	15.0~15.7	15.4 ± 0.2 B
TSS	22.0~46.0	36.1 ± 6.18 A
TN	2.1~3.3	2.69 ± 0.29 D
COD	32.6~45.6	36.91 ± 3.80 A
TP	0.13~0.18	0.15 ± 0.01 E
Chlα	2.4~3.8	3.07 ± 0.38 D

Table 3. Results from ANOVA to display the significance of a heavy metal among all sediment samples. Groups with the same letter are not statistically significant.

Sediment	Range (from Sample 1 to Sample 10)	Mean ± SD (Mean of Combining Data Sample 1 to Sample 10)
Arsenic	17.650~29.700	22.925 ± 3.845 e
Cadmium	3.100~4.970	3.873 ± 0.463 f
Chromium	85.79~112.820	97.06 ± 7.625 b
Copper	60.120~111.800	81.350 ± 14.708 c
Mercury	0.062~0.095	0.075 ± 0.009 f
Nickel	35.290~73.800	57.175 ± 11.346 d
Lead	38.260~72.450	56.932 ± 10.757 d
Zinc	152.200~207.340	174.352 ± 12.745 a
TN	1184.0~2360.0	1811.1 ± 364.6 A
TP	633.0~870.0	742.4 ± 87.4 B
OM	2.6~4.8	3.49 ± 0.72 C

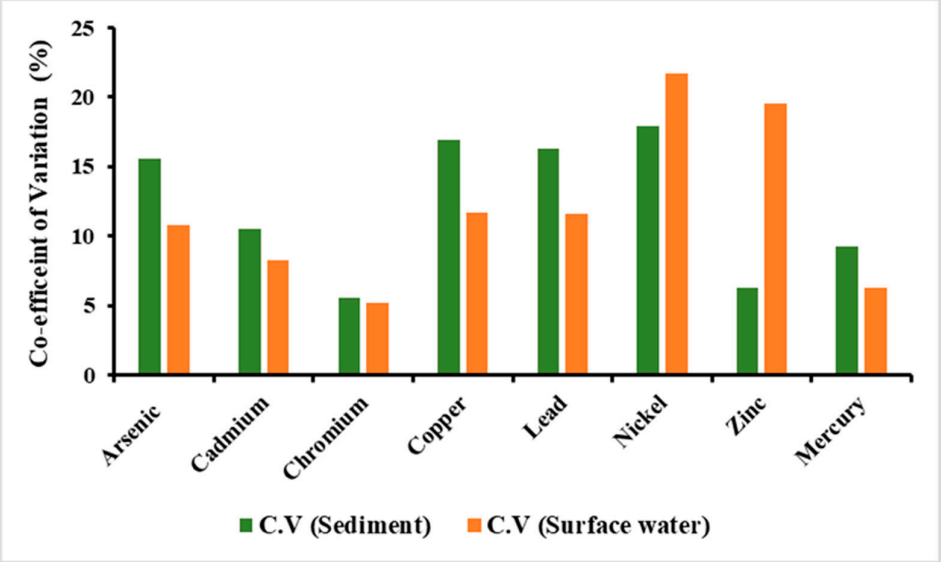


Figure 2. Coefficient of variation among the samples in surface water and sediments.

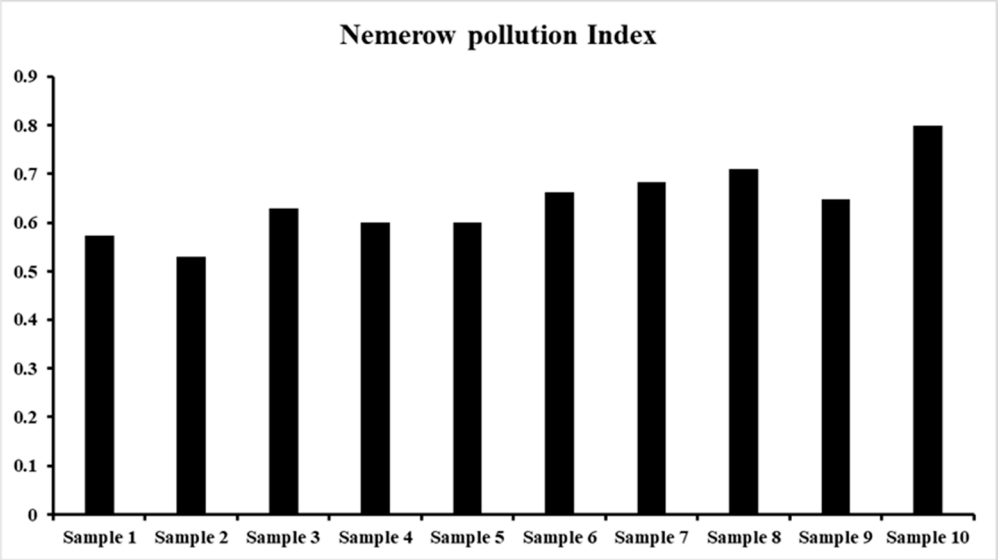


Figure 3. Nemerow pollution index in the surface waters of the tributary.

All the mean concentrations of heavy metals in surface water were lower than the Chinese heavy metal guidelines [41] and World Health Organization (WHO) guidelines, while for sediments, all the mean metal concentrations were higher than the heavy metal guidelines [31]. The heavy metal concentrations of this tributary were compared with existing literature to identify the current pollution status in surface water and sediments.

Regarding surface water, we compared the heavy metals to other freshwater ecosystems, such as Poyang, Chishui River, Dongting, Taihu, Chaohu, Hongze, Hong, Daye, Dianchi, and specific areas of the Yangtze River [10,42,43]. The mean concentration of As (5.44 ± 0.58 mg/L) in our surface water was similar to Chao and Taihu Lakes, and it was slightly higher than the standard value observed in the Yangtze River (3.4–4.7 mg/L). Likewise, most of our heavy metal values were similar or in range with the Chaohu, Taihu, and Poyang lakes, indicating that heavy metal pollution in surface waters is a massive problem most freshwater ecosystems face [44]. Most of these lakes are prone to agricultural runoffs, discharges from wastewater treatment plants, and anthropogenic activities [3,9,10]. However, we did observe that our heavy metal concentrations for surface water were higher than the concentrations found in the middle and lower parts of the Yangtze River [7,45]; we believe that this could be due to the exposure of this tributary to an excessive agricultural runoff that caused elevated levels of heavy metals, as this was also observed by [38,46].

The sediment heavy metal concentrations were compared with Dianchi, Taihu, Haohu, Dongting, Poyang, Yangtze, Haihe River, Yellow River, Pearl River, Honghu, Donghu, and Daye lakes [6,11,47,48]. Unlike surface water, the sediment metal concentrations were within range. Even though the values were within range, they were still higher than the guideline values, indicating that the sediments are exposed and vulnerable to diverse contamination sources, including industrial, agricultural, and anthropogenic activities [14]. The observation of sediment metal concentrations being higher than standard guidelines has been commonly observed by other studies that have also explored lakes and river sediments for metals [46–48]. The heavy metal concentration in this tributary indicates that government intervention with stricter guidelines and policies for lakes and rivers is highly required, and removing or reducing the heavy metal concentration could benefit the ecosystem.

3.2. Heavy Metal Assessment through Various Indices

An investigation of surface water quality through the analysis of the Nemerow pollution index was conducted on a tributary in Nanjing in September (Figure 3). The indices showcased a fluctuating range from 0.5309 to 0.7988. Three sample points (Samples 8–10)

illustrated indices surpassing 0.7, inferring a minor pollution level at these locations. The scenario implies a possibility of specific pollution sources or events near or upstream of these points. The diverse pollution levels within the tributary could emanate from various sources, given its exposure to agricultural, residential, and anthropogenic activities. Elevated indices, especially in Samples 8–10, might be attributed to agricultural runoff, a common concern in surface water pollution, especially following rainfall events, due to the washing away of fertilizers and pesticides into water bodies [49]. September's climate context might be pivotal, as late summer and early autumn can introduce climatic variables influencing runoff and pollution levels. Residential communities adjacent to the tributary can also significantly contribute to the pollution indices observed. Domestic waste, sewage mismanagement, and everyday activities can directly or indirectly introduce contaminants into the water body. Anthropogenic activities, such as industrial operations or recreational uses, might also adversely impact the water quality [50]. Illegal discharges, inadequate waste management, or non-compliance with environmental regulations exacerbate pollution issues in urban water bodies. Given the multifaceted potential sources, managing and mitigating pollution in the tributary necessitates an integrated approach. Employing biological remediation, enforcing stringent regulations on pollutant discharges, and community awareness programs might reduce current pollution levels and safeguard the tributary against future pollution threats.

All sediment sampled locations exhibited PLI values considerably exceeding 1.0 and reaching values as high as 3.04, indicating the existence of concerning levels of pollution across all tested sites (Figure 4). The range of PLI from 2.16 (Sample 2) to a strikingly elevated 3.05 (Sample 10) confirms a ubiquitous contamination presence and intriguingly suggests a gradient or potentially variable sources of contaminative input across the sampled sites. Dissecting the variability and progressive increment in PLI values from Sample 1 to Sample 10 could be emblematic of a spatial relation to contamination sources. Considering the observable trend, we believe that the potential contaminative activities—such as agricultural runoffs and domestic waste outputs—could be the reason for these elevated PLI values. PLI values in this tributary were as high as three, indicating a high level of environmental quality degradation and potential risk to aquatic organisms [51,52]. Elevated metal contents in sediments could result in bioaccumulation within aquatic organisms, thus infiltrating the food chain, consequently impacting higher trophic levels, and potentially posing threats to human health [53]. The observed escalation in PLI warrants a profound exploration into the bioavailability of these contaminants and potential bioaccumulation within local aquatic biota.

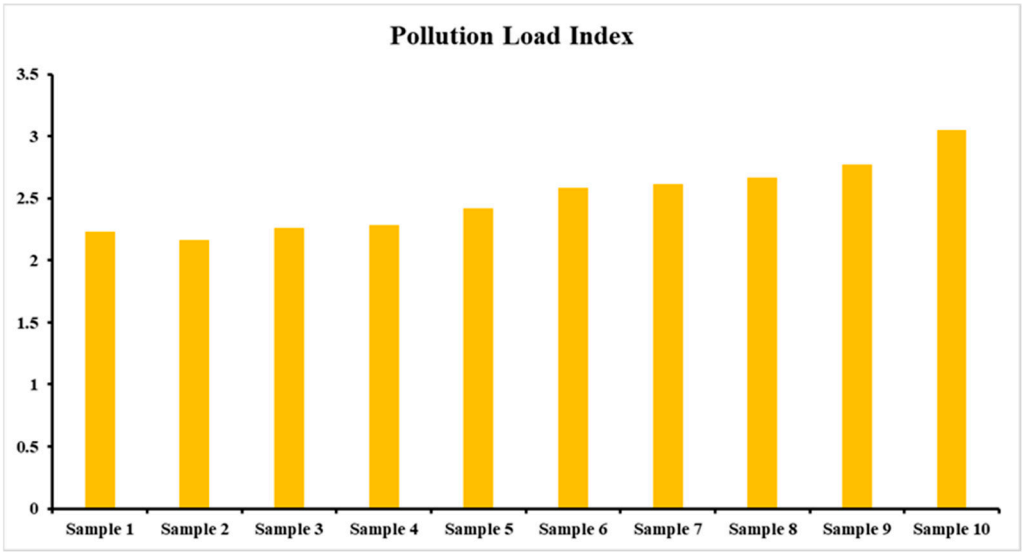


Figure 4. Nemerow pollution index among sediment samples of the tributary.

The consistently elevated Igeo values for specific heavy metals, particularly Arsenic and Cadmium, across various sediment samples indicate a pervasive contamination issue (Table 4). These findings are comparable to national and global contexts, where anthropogenic activities, notably agricultural runoff, anthropogenic activities, domestic discharge, and industrial emissions, have notoriously facilitated the percolation of heavy metals into aquatic ecosystems [1,54,55]. Arsenic, particularly noted for its toxicity, bioaccumulation, and potential to propagate through trophic levels [56], emerges as a conspicuous concern given its consistently high Igeo values, implying a pressing need for strategic intervention. A conspicuous disparity in the Igeo values among various metals underscores the complex contamination and sedimentation dynamics. Mercury, for instance, consistently manifested negative Igeo values across all samples, emblematic of comparatively unpolluted conditions. Contrasting sedimentation and dilution mechanisms might be influential, as proposed by [57], who emphasized the role of sediment particle size, organic matter, and redox conditions in modulating Hg distribution and bioavailability in sediments. The elevated levels of certain metals, such as Cd and Pb, even at localized hotspots such as Sample 10, warrant a comprehensive evaluation considering their ecotoxicological implications. Cd, renowned for its nephrotoxic, carcinogenic, and mutagenic properties [46], and Pb, noted for its neurological and developmental toxicity, elevate the ecological and health risk spectrums associated with the investigated tributary [58]. Zonal differences in heavy metal accumulation, evidenced by the spatial variability in Igeo values, suggest divergent contamination sources or transport mechanisms. Investigating the disparate contaminant profiles and scrutinizing the potential sources and pathways using isotopic fingerprinting or spatial distribution modeling, akin to methodologies adopted by [59], can discernibly elucidate contamination origins and trajectories. Implementing rigorous pollution source controls, enforcing stringent discharge norms, and adopting sustainable land-use practices will mitigate further contamination. Furthermore, community-centric approaches that amalgamate local knowledge, stakeholder engagement, and participative management will bolster the effectiveness and inclusivity of contamination management strategies.

Table 4. Geoaccumulation index values among sediment samples in the tributary.

Igeo	Arsenic	Cadmium	Chromium	Copper	Lead	Nickel	Zinc	Mercury
Sample 1	1.05	2.04	1.44	1.39	−0.06	0.69	−0.07	−1.902
Sample 2	1.06	1.92	1.24	1.49	0.01	0.71	−0.11	−2.13
Sample 3	1.05	2.03	1.27	1.57	−0.13	1.09	−0.06	−2.10
Sample 4	1.22	2.06	1.25	1.50	0.02	1.09	−0.12	−2.19
Sample 5	1.22	2.10	1.24	1.57	0.40	1.10	−0.17	−1.97
Sample 6	1.46	1.93	1.28	1.91	0.39	1.43	−0.06	−2.09
Sample 7	1.45	2.07	1.29	1.90	0.37	1.43	−0.05	−2.05
Sample 8	1.47	2.11	1.24	1.91	0.49	1.47	−0.02	−2.06
Sample 9	1.60	2.22	1.36	2.03	0.52	1.35	0.08	−2.1
Sample 10	1.68	2.44	1.46	2.10	0.58	1.53	0.13	−1.76

The Ecological Risk Index (Ei) and Potential Ecological Response Index (Ri) illuminated a noticeable presence and an alarming level of certain heavy metals across all sampling sites (Figure 5). Remarkably, Cd showcased elevated Ei values across all sites, reaching 245.7 in Sample 10. Similarly, As and Hg also demonstrated pronounced Ei, although not as critically elevated as Cd. A perceivable augmenting trend was discerned in Ri values, cascading from 280.94 (Sample 1) to 386.68 (Sample 10), potentially suggesting an augmenting ecological risk along the sampled path. The infiltration of heavy metals, notably Cd, As, and Hg, in the tributary sediments underscores a tangible ecological problem, revealing a scenario of pronounced metal contamination. The significantly high Ei values for Cd

across all sampling sites burgeon above the typical range observed in uncontaminated sites, rendering it an element of critical concern. The previous literature documented Cd as a potent toxicant, even at minimal concentrations, imposing severe repercussions on aquatic life and potentially spiraling up the trophic chain [38,60]. As and Hg also disturbed the sediments with notably high Ei values, entailing potential detrimental consequences. Arsenic is renowned for its non-threshold toxicity, culminating in severe ecological ramifications [56]. Concurrently, Hg's potential for bioaccumulation and biomagnification poses tangible threats to aquatic ecosystems and human health via seafood consumption [42]. The tendency of Ri to escalate from sample sites 1 to 10 warrants an exploration into potential contamination sources or a contamination gradient, even though it could be potentially stated that consistent agricultural and anthropogenic activity may have caused the values to increase from Samples 1 to 10. However, it would be ideal for establishing spatially resolved risk assessments and considering the hydrodynamic and sedimentary processes, potentially facilitating deciphering contamination dispersion mechanisms.

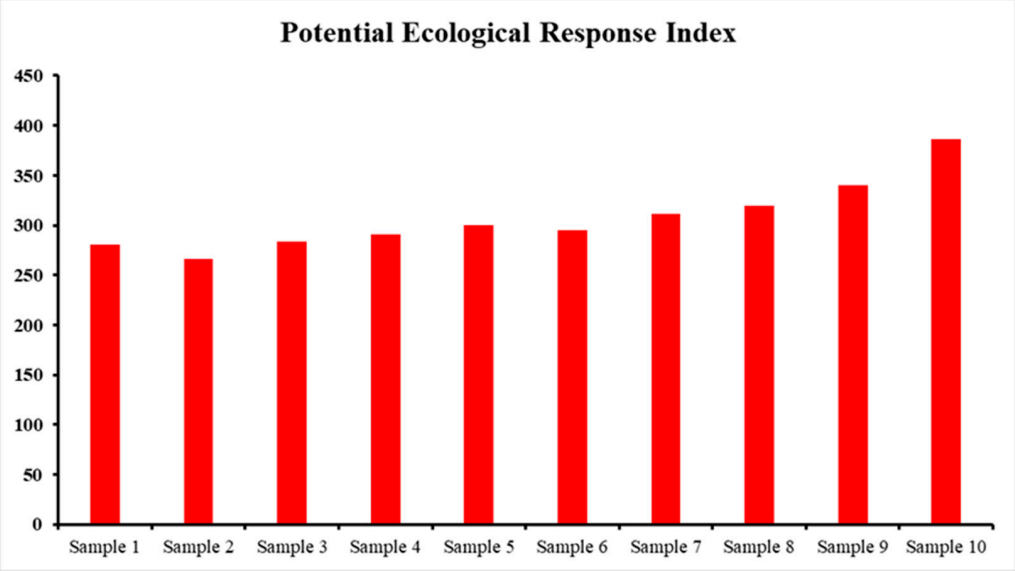


Figure 5. Potential Ecological Risk Index among sediment samples in the tributary.

3.3. Relationship between Physicochemical Parameters and Heavy Metals in Surface Water and Sediments

Pearson correlation analysis was performed to identify potential pollution sources that could cause the presence of heavy metals in surface water and sediments. The analysis of surface water samples revealed intriguing relationships between various heavy metals, shedding light on potential pollution sources and the interplay of these metals in the environment (Figure 6). A notable positive correlation was observed between Cd and Cu (r = 0.73 *). This finding suggests that these two heavy metals may share a common source or be transported via similar pathways. The presence of Cd and Cu in the water samples could be attributed to agricultural activity processes that release these metals into the surrounding environment [61]. Elevated levels of Cadmium and Copper can potentially contribute to water quality degradation, leading to adverse effects on aquatic ecosystems and human health. We observed a strong positive correlation between Cr and Ni (r = 0.89 ****). Such a high correlation coefficient indicates a tightly interwoven connection between these heavy metals. Previous studies have shown that their presence in the water may pose significant risks to water quality, including potential toxicity to aquatic life. Cr and Ni are often associated with industrial discharges, raising concerns about the extent of contamination from these sources and the potential impacts on water quality [62]. We observed Zn and Pb (r = 0.68 *) to be positively correlated, implying that these two metals may co-occur in the surface water samples. Anthropogenic sources, such as agricul-

tural runoff, likely contribute to Zn and Pb concentrations in the tributary [63]. Mercury negatively correlated with pH (r = −0.34), indicating an inverse relationship between pH and Hg concentration. This correlation underscores the influence of pH on the behavior and availability of mercury in aquatic ecosystems. pH levels can enhance the bioavailability and toxicity of Hg, which has implications for both water quality and ecosystem health [53]. A negative correlation was observed between Cd and pH (r = −0.06). This suggests that the slight alkalinity of water may play a role in influencing the concentration and behavior of Cadmium. Moderate pH levels can increase the solubility and mobility of certain heavy metals, including Cadmium, potentially exacerbating their impact on water quality [64]. Copper exhibited a significant positive correlation with temperature (r = 0.73 *), implying a connection between these two parameters. It is well-established that temperature can influence the solubility and reactivity of Copper in aquatic systems. As water temperature increases, Copper's availability and potential to impact water quality may also rise [39]. Zinc positively correlated with COD (r = 0.63 *), suggesting that organic pollution may contribute to the presence of Zinc in the water. Organic matter can be a carrier for certain heavy metals, such as Zinc, potentially impacting water quality and aquatic ecosystems [65]. Chromium positively correlates with TN (r = 0.22); elevated TN concentrations are often associated with agricultural runoff, which may have fertilizers. Since agricultural runoff combines organic matter (rich in N) and fertilizers (rich in Cr), it may explain their positive correlation. Lead significantly correlates with TSS (r = 0.49), implying that high levels of suspended solids in water may facilitate the transport of lead particles. Mercury exhibits a robust positive correlation with Total Phosphorus (TP) (r = 0.83 **). This finding suggests that water's Phosphorus levels can influence mercury's behavior and bioavailability. Elevated TP concentrations may exacerbate mercury contamination since elevated levels indicate eutrophication, and since this tributary is exposed to anthropogenic pollution, it could explain the reason behind the strong correlation [66].

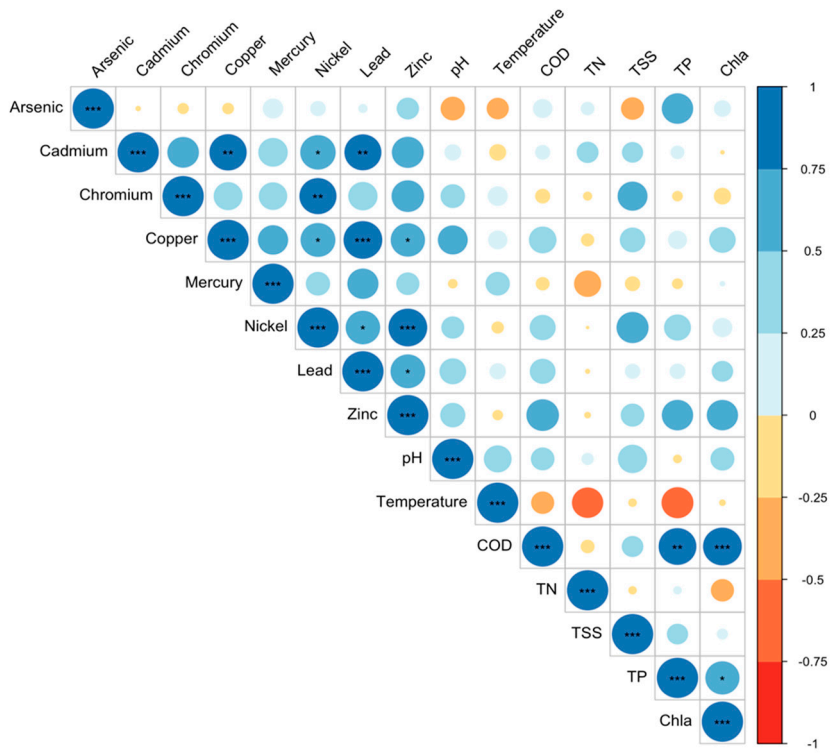


Figure 6. Pearson correlation between heavy metals and nutrient parameters in surface water. "* means $p \leq 0.05$", "** means $p \leq 0.01$", "*** means $p \leq 0.001$".

However, for sediments, most heavy metals were found to have a significant positive correlation with each other and also with TN and TP, apart from Cd (positive but

non-significant), Hg (positive but non-significant), and OM (positive but non-significant) (Figure 7). The strong positive correlations among critical heavy metals, As, Cu, Pb, and Ni, indicate a potential convergence of sources or similar behavior patterns, likely occurring because of anthropogenic influences or geological attributes intrinsic to the river basin [7]. This observed clustering of correlations emphasizes the interlinked nature of these heavy metals in the sediment matrix, highlighting the necessity for a complete approach to source identification. Conversely, the marked positive association between Cd and Zn suggests shared origins, possibly attributable to industrial activities or human activities [67]. A separate cluster emerges with Cr exhibiting robust correlations with Cu and Pb, implying a degree of co-release, a phenomenon that may have sources in agricultural discharges within the river's catchment area [68]. The distinctive behavior of Hg, displaying only moderate correlations with its heavy metal counterparts, apart from Cd and Cr, is noteworthy. This abnormality suggests differentiated sources or factors governing its mobility in the sediment, warranting a focused examination of its intricate transport mechanisms and contamination sources [40]. We observed positive associations between Total Nitrogen (TN) and Total Phosphorus (TP) with heavy metals, hinting at potential nutrient–metal interplays or shared inputs, unveiling yet another layer of complexity in the sediment's elemental composition [69]. Such associations underscore the interdependent nature of nutrient and heavy metal dynamics, contributing to the intricate ecological balance in river ecosystems. Organic matter (OM) positively correlated with multiple heavy metals, notably Chromium. This underscores the pivotal role of OM as a binding agent, influencing the retention and distribution of heavy metals within the sediment matrix [70]. These findings accentuate the multifaceted nature of heavy metal interactions and underscore the potential significance of organic matter content in sediments as a regulator of metal mobility.

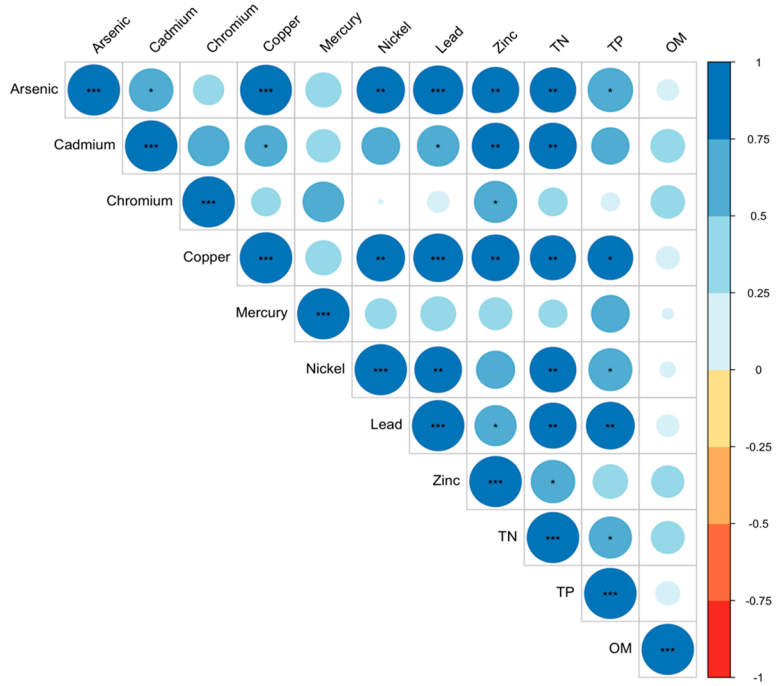


Figure 7. Pearson correlation between heavy metals and nutrient parameters in sediments. "* means $p \leq 0.05$", "** means $p \leq 0.01$", "*** means $p \leq 0.001$".

The exploration of heavy metal concentrations and environmental parameters in water samples yields critical insights into aquatic health and possible contamination sources. A robust Pearson correlation analysis among heavy metals (Arsenic, Cadmium, Chromium, Copper, Lead, Nickel, Zinc, and Mercury) and other environmental parameters (pH, Temperature, COD, TN, TSS, TP, and Chlα) exposes several noteworthy relationships. A profoundly strong correlation was observed between Nickel (Ni) and Chromium (Cr)

(r = 0.89 ****), suggesting their simultaneous presence and potential common source, which may be attributed to industrial discharge, given the ubiquity of these metals in several industries [71]. Simultaneously, the apparent strong association between Copper (Cu) and Lead (Pb) (r = 0.86 **), metals often conjointly found in electronic wastes and industrial emissions, denotes an imperative need for scrutinizing industrial activities occurring within the watershed [48].

Interestingly, the relationships between certain metals and nutrients hint towards the intricate and multifaceted dynamics within the water body. For example, a significant positive correlation between Zinc (Zn) and Total Phosphorus (TP) (r = 0.60 *) might stem from agricultural runoff, where Zn often originates from fertilizers and TP from both fertilizers and organic matter [72]. Additionally, high correlations between COD and TP (r = 0.83 **) and between COD and Chlα (r = 0.88) suggest a potential scenario where nutrient and organic matter enrichment is fostering eutrophication, which could detrimentally affect aquatic ecosystems [73]. Moreover, the relationships between heavy metals indicate possible synergistic toxicological effects on aquatic life, warranting a deeper dive into understanding the bioavailability and speciation of these metals in the water body. High concentrations and the coincidence of heavy metals may induce adverse effects beyond what might be anticipated from their concentrations [55,62]. Emphasizing further potential sources, the noted correlations may point towards distinct anthropogenic inputs. For instance, the associations between metals might signify influences of urban and industrial activities, while relationships between nutrients and certain metals might allude to agricultural influences. Such integrated insights into pollutant sources can serve as a baseline to develop and implement strategic monitoring and management approaches to safeguard aquatic ecosystems. From a broader perspective, the illustrated correlations and their implications underline the importance of adopting an integrative approach to understanding aquatic environmental chemistry, considering the role and impact of anthropogenic activities in shaping these chemical dynamics. Future studies should encompass a thorough spatial-temporal analysis to elucidate pollution hotspots and trend shifts and a rigorous risk assessment to effectively comprehend ecological and health implications.

The Principal Component Analysis (PCA) of heavy metal concentrations in surface water and sediments revealed intriguing patterns, offering insights into the nuanced interactions between these environmental matrices. Principal components with an eigenvalue greater than 1 were selected. In surface water (Table 5), the shared negative loadings of Mercury, Zinc, Chromium, Cadmium, Arsenic, Copper, Nickel, and Lead on PC 1 (explaining 60.11% of variance) suggest a unified influence, potentially originating from agricultural runoff. The fact that these heavy metals show similar behavior on PC 1 suggests a common source or process influencing their concentrations in surface water. We believe that this unified influence and the strong correlation shown in Pearson correlation may originate from agricultural runoff. Agricultural activities often involve the use of fertilizers, pesticides, and other chemicals, contributing to the presence of heavy metals in water bodies [72]. The dominance of PC 1 in explaining the variance strengthens the argument that the identified unified influence of heavy metals is a significant factor shaping the heavy metal composition of surface water. The contribution of PC 2 (14.26%) indicates additional complexities, possibly linked to variations in agricultural practices, land use, or seasonal fluctuations impacting the dynamics of heavy metals in the water. The positive loadings of Cadmium, Chromium, Copper, and Lead suggest that variations in these elements positively correlate in PC 2, indicating potential commonalities, such as land use or geochemical characteristics in their sources or behaviors distinct from the negative correlations observed in PC 1. This contrasts with the negative correlations observed in PC 1, indicating a nuanced differentiation in the environmental dynamics of these heavy metals. This nuanced differentiation implies subtle distinctions in their relationships, possibly influenced by factors unique to PC 1, contributing to a more comprehensive understanding of the complex interactions between these elements.

Table 5. PCA results of HMs in the surface water.

Variance	Component Loadings	
	PC 1	PC 2
Arsenic	−0.107289	−0.9809221
Cadmium	−0.8709873	0.1593387
Chromium	−0.754253	0.1214629
Copper	−0.8848301	0.18606383
Mercury	−0.6102366	−0.2304781
Nickel	−0.9011935	−0.0293826
Lead	−0.8909814	0.06455346
Zinc	−0.8419311	−0.2140067
Eigenvalue	4.80918575	1.14091955
Percentage of Variance (%)	60.1148219	14.2614944
Cumulative (%)	60.11482	74.37632

In sediments (Table 6), the positive loadings of Chromium, Nickel, Mercury, Arsenic, Cadmium, Copper, Lead, and Zinc on PC 1 (explaining 67.05% of variance) signify a unique sediment signature, likely reflecting the cumulative impact of agricultural activities. Furthermore, the contribution of PC 2 in sediments (18.08%) provides additional insights into variations not fully captured by PC 1. In PC 2, the substantial positive loading of Chromium implies a robust positive correlation with other heavy metals, suggesting potential common sources or shared environmental behaviors. This was in accordance with our findings from previous Pearson correlation analyses. The negative loadings of Nickel, Arsenic, Copper, and Lead indicate an inverse relationship, pointing to distinct patterns or sources compared with Chromium. Exploring PC 2 in sediments may unveil specific factors influencing heavy metal concentrations, such as sediment composition or dynamic interactions between the agricultural site and the tributary. Sediment composition, for example, can vary based on factors such as mineral content, organic matter, and particle size. The mineral composition of sediments can influence heavy metal concentrations. Certain minerals may adsorb or release heavy metals, impacting their availability in the sediment. The presence of organic matter in sediments can affect heavy metal mobility. Organic compounds may form complexes with heavy metals, influencing their solubility and retention in sediments [45]. Dynamic interactions between the agricultural site and the tributary could involve factors such as seasonal changes, water flow patterns, or land use practices that influence sedimentary processes and subsequently impact heavy metal concentrations.

Table 6. PCA results of HMs in the sediments.

Variance	Component Loadings	
	PC 1	PC 2
Arsenic	0.9605338	−0.2050113
Cadmium	0.8304608	0.2549575
Chromium	0.5239969	0.8363269
Copper	0.9496160	−0.2307083
Mercury	0.6006275	0.4253232
Nickel	0.8041798	−0.4949484
Lead	0.8872527	−0.3302491
Zinc	0.8836678	0.2281635
Eigenvalue	5.364178380	1.446698664
Percentage of Variance (%)	67.05222975	18.08373331
Cumulative (%)	67.05223	85.13596

The contrasting patterns in PC loadings between surface water and sediments emphasize the multifaceted nature of heavy metal dynamics in the tributary. Agricultural

runoff likely plays a pivotal role in influencing surface water quality, introducing metals that can be transported downstream. In sediments, the cumulative impact of these inputs and sedimentary processes contribute to a distinctive composition. The contrasting patterns between surface water and sediments suggest that diverse sources influence surface water. Negative loading values for all heavy metals surface water and positive loading for sediments in the tributary suggest unique origins not captured by Factor 1. This indicates potential contributions from seasonal changes, sediment deposition, and water flow movement [74]. In sediment samples, Factor 1 explained 67.05% of the total variance and was strongly associated with concentrations of Cr, Ni, Zn, and Cd, indicative of common sources. These metals are widely found in fertilizers and sewage discharges [3,75].

4. Conclusions

The comprehensive assessment of heavy metal contamination in the studied tributary's surface water and sediments reveals a complex and concerning scenario. The dominance of Cu and Ni in surface water and Zn and Cr in sediments, coupled with a noticeable increase in concentrations from Sample 1 to Sample 10, underscores a potential contamination gradient and the influence of external contamination sources, possibly linked to anthropogenic activities, such as industrial discharges and agricultural runoff. The hierarchical distribution of heavy metals and their correlation with various physicochemical parameters in both water and sediments elucidates potential sources and pathways of contamination. The strong correlations between certain heavy metals, notably between Cu and Pb, Ni and Cr, and Zn and TP, hint at shared origins or co-occurring release mechanisms, potentially tied to industrial activities, agricultural practices, and other anthropogenic influences. The elevated levels of heavy metals in sediments, surpassing guideline values, and the consistent elevation of PLI values across all sampled sites indicate a pervasive contamination issue and a potential risk to aquatic organisms and, by extension, the broader ecosystem. The elevated Ei values for Cd, As, and Hg and the increasing trend in Ri values from Sample 1 to Sample 10 further highlight the tangible ecological risk and suggest a contamination gradient or variable sources of contaminative input across the sampled sites. PCA uncovered intricate patterns in surface water and sediments, emphasizing the role of agricultural runoff in shaping water quality and sediment composition. The contrasting dynamics between matrices underscore the diverse nature of heavy metal influences. This study calls for continued exploration of PC 2 in sediments to unveil specific factors influencing heavy metal concentrations, contributing to a comprehensive understanding of environmental interactions in the tributary. The observed correlations and trends in heavy metal concentrations and ecological risk indices necessitate a multifaceted approach to managing and mitigating pollution in the tributary. This includes rigorous pollution source controls, stringent discharge norms, sustainable land-use practices, and community-centric approaches that amalgamate local knowledge, stakeholder engagement, and participative management.

Future research should encompass a broader temporal scale to decipher seasonal variations in heavy metal concentrations and distributions. Investigating these heavy metals' bioavailability and potential bioaccumulation within the aquatic biota will provide a more holistic view of the ecological impacts. Employing advanced techniques such as isotopic fingerprinting and spatial distribution modeling could elucidate contamination origins and trajectories more precisely.

Supplementary Materials: The following supporting information can be downloaded at https://www.mdpi.com/article/10.3390/app14020904/s1, Table S1. Classification of Igeo that represents various levels of pollution; Table S2. The background value for the respective metal; Table S3. Toxic response factor quotients for the studied heavy metals; Table S4. Various ER and RI index levels represent risks to the aquatic environment; Table S5. Water quality level determination based on the Nemerow pollution index method.

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