RESEARCH ARTICLE

Heavy metal toxicity, ecological risk assessment, and pollution sources in a hydropower reservoir

Imali Kaushalya Herath¹ [·](http://orcid.org/0000-0003-1675-2330) Shengjun Wu1 · Maohua Ma1 · Huang Ping1

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Abstract

Heavy metal (HM) toxicity, ecological risk, and pollution sources were analyzed using the pollution indexing and statistical methods in the Three Gorges Reservoir (TGR). The average concentration of HM increased in the order of $Cr < Ni < As <$ Cd < Cu < Mn < Pb < Zn < Al < Fe during the recharge period and Cd < Cr < Ni < As < Cu < Pb < Mn < Zn < Al < Fe during the discharge period. Signifcant spatial variations of Pb, Zn, Cd, As, Mn, Ni, Cr, and Cu were observed at the upstream and downstream sampling sites. Pb sharply increased during the recharge period, ranges (6.93 –148.62 μg/L) and exceeded WHO and USPEA standards limit. HPI, HEI, Cd, WPI indicated low pollution and moderate pollution with the strong infuence of Pb and Cd in the discharge and recharging period, respectively. HTML values are below the permissible toxicity load except for Pb. The Pb toxicity removal percentage is 56.47%, suggesting that the lead's toxicity level is high in TGR and requires the removal process. Ecological risk index values indicated that pollution is low in TGR. The potential ecological risk indexes (RI) of 9.07 and 31.60 were obtained for the discharge and recharge period, respectively, indicating low potential ecological risk from heavy metals in TGR. However, RI values revealed that (Pb, Cd, As Cr Ni, Cu Zn, and Mn) were the most ecological risk HMs in TGR. A signifcant ecological risk of Pb and Cd distribution was observed across the TGR. Multivariate statistical results found that Pb, Cd, Zn, Mn, Ni, As, Cr, Cu mainly originate from industrial wastewater, mining, metals processing, and agricultural runoff. Fe and Al were mainly from bedrock weathering. Pb, Cd, Zn HMs are a threat to the reservoir ecosystem. This study delivered a current status of HM pollution, toxicity, ecological risk, and pollution sources, indicating a vital insight into HM pollution and water security management in the Three Gorges Reservoir.

Keywords Heavy metals · Ecological risk · Toxicity load · Pollution indices · Source apportion · Hydropower reservoir

Introduction

Heavy metal (HM) pollution in the aquatic systems is a severe environmental issue due to their biomagnifcation, toxicity, and excessive accumulation (Zhao et al. [2019;](#page-17-0) Ke et al. [2017;](#page-16-0) Ma et al. [2016\)](#page-16-1). Aquatic habitats can mainly relate to a certain degree of contamination, but severe pollution can damage fauna and fora (Ghorab [2018](#page-16-2)). Due to

Responsible Editor: Xianliang Yi

 \boxtimes Imali Kaushalya Herath imalikaushalya@yahoo.com

 \boxtimes Shengjun Wu wsj@cigit.ac.cn

Key Laboratory of Reservoir Aquatic Environment, Chongqing Institute of Green and Intelligent Technology, Chinese Academy of Sciences, Chongqing, China

the long persistence of HM in a water body, it tends to have bioaccumulation and biomagnifcation in the food chain. Being non-degradable, HM continuously accumulates in the water bodies, sediments, and food chain, leading to harmful effects on human and the ecosystem health (Weber et al. [2013;](#page-16-3) Yin et al. [2018\)](#page-17-1). Heavy metals can be derived from natural and anthropogenic sources. Natural sources mainly consist of rock weathering, atmospheric deposits, and rainfall. Anthropogenic sources include mining, sewage discharge, fertilizer, industrial waste, etc. (Zhou et al. [2020\)](#page-17-2). Past studies noted that heavy metals derived from anthropogenic sources were high in the largest rivers and reservoirs in the world (Romano et al. [2018;](#page-16-4) Ghorab [2018](#page-16-2); Zhou et al. [2020;](#page-17-2) Steinnes and Lierhagen [2018\)](#page-16-5). According to WHO and USEPA, heavy metal concentrations were above the threshold limits in developing countries than the developed countries (Zhou et al. [2020\)](#page-17-2).

Three Gorges Reservoir (TGR) is known as the world's largest hydropower reservoir, located in the upper reach of the Yangtze river in China, is paid global attention as the China's vital water resource in power generation, food control, transportation, irrigation, etc. (Wang et al. [2018](#page-16-6); Li et al. [2013](#page-16-7)). Hence, the reservoir area was undergone rapid development activities, industries, population and settlements expansion, which led to severe heavy metal pollution in terms of water, sediments, and soil in the reservoir (Zhu et al. [2019;](#page-17-3) Ye et al. [2019](#page-17-4); Zhaoyong et al. [2015](#page-17-5)). Industrial and residential discharge, sewage, pesticides, and fertilizer are often released into the TGR water (Zhou et al. [2020;](#page-17-2) Bing et al. [2016](#page-16-8)). Previous studies highlighted that As, Cd, Cr, Cu, Pb, and Zn increased and more probability to exceed the standards level 50, 1, 10, 100, 10, and 50 μ g/L, respectively, in the reservoir (Bing et al. [2016](#page-16-8); Zhu et al. [2015](#page-17-6), [2019](#page-17-3); Chen et al. [2011\)](#page-16-9). TGR dam operations altered the hydrologic and sedimentation process, which directly afects the reservoir's heavy metal distribution and behavior (Zhu et al. [2019\)](#page-17-3). Heavy metals accumulate in the reservoir riparian zone during the high water level (175 m) and periodically transform and dissolve in the reservoir water (Cheng et al. [2017](#page-16-10)). Thus, water level fuctuation may cause HM enrichment and pose a potential threat to water security for humans and ecosystems (Ma et al. [2016](#page-16-1)). Therefore, as TGR provides over 30 million people and ecosystems, the status of heavy metal pollution, degree of contamination, toxicity load, and ecological risk assessment are required to perform frequently in the reservoir. Previously, many studies were focused on the pre and post dam HM distribution in the TGR, and researches were conducted on either mainstream or tributaries such as Chongqing city, Badong to Yichang and Yibin, Luzhou (Wang and Zhang [2018](#page-16-11); Gao et al. [2016](#page-16-12); Ma et al. [2016](#page-16-1)) and limited or discontinuous to a specifc period. Adequate information is available on the spatial–temporal characteristics of heavy metals at diferent water levels operation periods in the reservoir. Pollution indexing, sources, and health risk assessment studies were also widely done to quantify the potential impact of HM pollution on human health. However, the ecological risk assessment of heavy metals in surface water in TGR under diferent reservoir operation periods remained scarce.

In this study, ten heavy metals were evaluated for reservoir recharge and discharge period using pollution indexing methods (HPI), HEI, Cd, WPI, which are formulated to assess the overall water quality of the aquatic systems. For further details, refer methods section. Heavy metal toxicity load (HMTL) used in the study that aggregates the toxicity level of HM in the water body is responsible for health impact and predict the required removal percentage of individual heavy metals from the water bodies (Dai et al. [2018;](#page-16-13) Zhao et al. [2011](#page-17-7); Tiang et al. [2016\)](#page-16-14). Ecological risk assessments (RI) were used for pollution control due to the high concentration of HM in sediments and their subsequent release into the water bodies and harmful to ecosystem health (Ke et al. [2017](#page-16-0); Ma et al. [2016](#page-16-1)). Multivariate statistical methods were used to identify the sources of HM pollution (Fu et al. [2014](#page-16-15); Zhang et al. [2018](#page-17-8)). GIS was used to show the spatial distribution of heavy metals' patterns in TGR. Thus, as main objectives of this study are (1) to analyze spatial–temporal characteristics, distribution and pollution status, and toxicity loads for heavy metals in TGR; (2) to quantify the potential ecological risk of heavy metals in the reservoir; and (3) to source identifcation for HM pollution in TGR.

Materials and methods

Description of the study area

The TGR (29° 16′–32° 25′N, 106° 50′–113°–85′E) is located upper reach of the Yangtze river (Fig. [1](#page-2-0)) and formed the narrow V-shaped valley (Three Gorges) covering from Chongqing to Yichang. The total surface area is $55,742 \text{ km}^2$, and the length, width, and depth of the reservoir are 660 km, 1.1 km, and 90 m, respectively (Ma et al. [2016;](#page-16-1) Bing et al. [2016](#page-16-8)). The average annual infow of TGR is about 13,085 m³/s, and dam discharge is about 12,214 m³/s (Zhao et al. [2019](#page-17-0); Zhu et al. [2015\)](#page-17-6). The water level changes seasonally between 145 m discharge and 175 m recharge. The reservoir belongs to the humid subtropical monsoon climate zone. The average annual temperature ranges between 15 and 19 °C, while the annual average rainfall is about 1250 mm with significant intra and inter-annual shifts. About 75% of the local annual rainfall occurs from May to September. The annual wind speed is 1.2 m/s (Ma et al. [2016](#page-16-1); Huang et al. [2017](#page-17-9)). The land use pattern in the reservoir is shown in Fig. [1](#page-2-0). The geology structure of the Three Gorges Reservoir is underlain by sedimentary, carbonate, limestone, dolomite, and clastic rocks. Coal strata are enriched in sulfdes and are inter-bedded with carbonates, especially in the southern basin (Chetelat et al. [2008\)](#page-16-16).

Sampling and analysis

Using 30 sampling sites along the Three Gorges Reservoir and downstream (after the dam), 60 water samples were collected for heavy metal analysis during the recharge period (November) water level at 175 m and discharge period (August) water level at 145 m. Water samples were grabbed at least 45 cm below the water surface and 75 m away from the river bank for analyzing the heavy metals of Pb, Cd, Cu, Cr, Zn, Mn, Ni, As, Fe, and Al. The samples were fltered through the disposable 0.45-μm cellulose nitrate syringe flters and acidifed with $(HNO₃)$. All samples were filled without headspace and

Fig. 1 Study area map for Three Gorges reservoir, sampling sites, and land use pattern (Agricultural forest 42%, Forest land 39.4%, Grasslands 9.5%, water bodies 5.5%, construction lands 3.2%, and abandoned lands 0.4%)

further sealed with a Paraflm®. Heavy metals were determined by inductively coupled plasma mass spectrometry (ICP-MS), Thermo ICapQ (Thermo-Fisher Scientifc Inc., Bremen, Germany).

Water pollution assessment indices

Heavy metal pollution index

The heavy metal pollution index (HPI) evaluates and assesses the overall water quality for each heavy metal (Ke et al. [2017](#page-16-0)). It indicates the combined infuence of water quality (Reza and Singh [2010\)](#page-17-10).

$$
W_i = K/S_i \tag{1}
$$

where Wi indicate unit weightage and Si represent the standard for *i*th parameter, while k is the constant of proportionality

$$
Q_i = 100V_i/S_i \tag{2}
$$

Qi gives ith parameter sub-index, V*i* offers the observed value of the ith parameter in μg/l, and *Si* is the standard or permissible limit for the ith parameter.

$$
HPI = \frac{\sum_{i=1}^{i=n} (Q_i \times w_i)}{\sum_{i=1}^{i=n} w_i}
$$
 (3)

 W_i represents the unit weight of *i*th parameters, Q_i is the sub-index of the *i*th parameter, *n* is the number of parameters. The weighted arithmetic index has been used to calculate HPI (Ma et al. [2016;](#page-16-1) Ke et al. [2017](#page-16-0)).

Heavy metal evaluation index

Heavy metals evaluation is determined to assess the water quality relative to heavy metals concentration (Kumar et al. [2019](#page-16-17)). HEI grade the accumulation rate of each heavy metal of the overall water quality. The index value between 0 and 1 refects each quality concern's relative/subjective importance and is inversely proportional to each metal's standard. Heavy metal evaluation index (HEI) provides overall water quality concerning each heavy metal and is computed as follows (Ma et al. [2016;](#page-16-1) Zhou et al. [2020\)](#page-17-2):

$$
HEI = \sum_{i=1}^{n} \left(\frac{M_i}{MAC_i} \right)
$$
 (4)

where MAC_i is the observed value and maximum permissible amount of the *i*th heavy metal. Categories water quality based on HEI: $<$ 10 for low, 10–20 for moderate, and $>$ 20 are high pollutions (Zhou et al. [2020](#page-17-2)).

Degree of contamination

The degree of contamination (C_d) indicates the accumulated detrimental effect of the heavy metals in water

(Kumar et al. [2019;](#page-16-17) Ma et al. [2016\)](#page-16-1)

$$
C_d = \sum_{i=1}^{n} C f_i
$$
\n⁽⁵⁾

$$
Cf_i = \frac{M_i}{MAC_i} - 1\tag{6}
$$

C*f*i is the pollution factor for the ith HM; Contamination degree explains: < 1 for low, 1–3 for moderate, and > 3 for high pollutions (Kumar et al. [2019](#page-16-17); Ma et al. [2016](#page-16-1)).

Water pollution methods WPI

The water pollution index is applied to all kinds of water use and management. The WPI index explains the relative number with the minimum acceptable limit for each heavy metal supporting pollution control. The water pollution index (WPI) indicated how polluted water needed to be diluted to the permissible limits (WHO [2017](#page-16-18)).

$$
WPI_i = \frac{M_{i-}Min_i}{R_i} \tag{7}
$$

where *Min*_i Indicate minimum permissible limit, and R_i represents the range of the allowable limits values for heavy metal were taken from (WHO [2017\)](#page-16-18).

Heavy metals toxicity load

Heavy metal toxic load (HMTL) calculates the total toxic load for each heavy metal in water that affects human health. It gives warning to water resources management authority what percentage of water treatment is required to remove heavy metal toxicity for human use. HMTL is calculated by multiplying the measured values of heavy metals with their hazard intensity.

$$
HMTL = \sum_{i=1}^{n} C \times HIS
$$
 (8)

C is the heavy metal concentration, *n* is the number of heavy metals, and HIS represents the hazard intensity score (ATSD [2019\)](#page-16-19).

Ecological risk assessment

The ecological risk index (RI) is a standard method to estimate heavy metals' environmental risk in the water bodies and quantify the potential damage from heavy metal pollution by combining ecological toxicity (Huang et al. [2021](#page-16-20)):

$$
Cf = \frac{C_{\text{sample}}}{C_{\text{background}}}
$$
 (9)

where C_f^i is the contamination factor of the element *i*, *C*sample and *C*backgrounds represent measured and background values, and \vec{F}_r^i and \vec{T}_r^i are the potential ecological risk factor and the toxic response factor of heavy metal, respectively. The T_r^i values for Pb, Cd, Cr, Cu, Zn, Mn, As, and Ni 5, 30, 2, 5, 1, 1, 10, 5 respectively.

$$
E_r^i = T_r^i \times C_f^i \tag{10}
$$

$$
RI = \sum E_r^i \tag{11}
$$

Multivariate statistical methods

Pearson's correlation, cluster analysis (CA), and principal component analysis (PCA) were adopted to detect the associations and origins of heavy metal sources in this study. Further details of these statistical methods can be obtained from (Chen et al. [2011;](#page-16-9) Yang et al. [2017](#page-17-11)). All data were processed using standard commercial software such as SPSS 20.0, Origin Pro 21.0, and GIS.

Results and discussion

Spatial and temporal distribution of heavy metals

Descriptive statistical data of HM concentration during the recharge and discharge period in the Three Gorges Reservoir were presented in Table [1](#page-4-0) with threshold concentrations of heavy metals as per WHO (World Health Organization) and UNEPA (United Nation Environment Protection Agency) standards. Average concentration of 10 heavy metals were ranked in the order of $Cd < Cr < Ni < As < Cu < Pb < Mn <$ Zn< Al<Fe, 0.51 μg/L, 1.03 μg/L, 1.25 μg/L, 2.97 μg/L, 3.49, 6.84 μg/L, 31.06 μg/L, 58.66 μg/L, 227.19 μg/L, and 296.74 μg/L, respectively in the discharge period. Al is above the standard limit of WHO and USEPA, and the rest of heavy metals are within the standard limit. While,

	Discharge period μ g/L					Recharge period μ g/L						
Metals	Ave	Max	Min	SD	CV%	Ave	Max	Min	SD	CV%	WHO	USEPA
Pb	6.84	15.17	2.06	3.91	57.23	34.46	148.62	6.93	32.36	93.91	10	15
C _d	0.51	2.66	0.11	0.51	99.14	2.95	8.28	0.65	2.01	68.35	3	5
Cr	1.03	5.27	0.52	0.85	82.96	1.19	2.54	0.59	0.51	42.98	50	100
Cu	3.49	10.70	2.07	1.78	51.04	4.03	10.80	1.40	1.97	48.86	2000	1300
Zn	58.66	158.80	25.15	30.12	51.35	53.49	92.93	26.02	13.74	25.68	1000	1000
Mn	31.06	121.19	7.68	27.19	87.55	14.50	67.33	2.87	14.76	101.77	100	50
As	2.97	6.35	1.97	0.84	28.17	1.84	7.03	0.20	1.52	82.55	10	10
Ni	1.25	2.91	0.74	0.46	37.12	1.30	3.15	0.56	0.60	46.37	20	15
Fe	296.74	1405.13	61.96	295.10	99.45	376.71	1726.38	94.41	350.90	93.15	300	300
Al	227.19	899.27	69.74	211.18	92.95	317.61	847.40	187.45	148.40	46.72	200	200

Table 1 Total heavy metals concentration in μg/L during the discharge and recharge phases of the Three Gorges reservoir in 2017/2018 and concentration of heavy metals in water per WHO and USEPA

the average concentration of the recharge period is in the order of $Cr < Ni < As < Cd < Cu < Mn < Pb < Zn < Al < F$ e, 1.19 μg/L, 1.30 μg/L, 1.84 μg/L, 2.95 μg/L, 4.03 μg/L, 14.50 μg/L, 34.46 μg/L, 317.61 μg/L, 376.71 μg/L, respectively. The coefficient variation (CV) of the HM showed Fe (99.45%) exhibited maximum variation followed by Cd (99.14%) while the minimum variation was the As (28.17%) during the discharge period. In the recharge period, maximum CV was observed from Mn (101.77%) and Pb (93.91%), while minimum CV value was obtained from Zn (25.65%). Pb concentration varies from 2.06 to 15.17 μ g/L in discharge the period, and the maximum value is above the standard limit of WHO and USEPA. Pb concentration drastically increased during the recharge period, ranging from 6.93 to 148.62 μg/L, and the maximum value of Pb is 14 times above the standard limit of WHO. A recent study also noted that Pb concentration in TGR exceeded the standard limit of WHO and UNEPA (Zhao et al. [2019;](#page-17-0) Gao et al. [2016\)](#page-16-12). The average Pb concentration of other reservoirs in southern China is signifcantly lower than TGR, about 0.22 μg/L in recharge and 0.67 μg/L discharge period (Wang et al. [2018\)](#page-16-6). Yangtze River obtained 55.1 μg/L while Perl River (0.08 μg/L), Poyang Lake (2.74 μg/L), and Taihu Lake (1.97 μg/L) sharply dropped Pb contents (Wang et al. [2018](#page-16-6)). Pb concentration was sharply increased in TGR compared to other water sources in China. A global study of heavy metals found that Pb and Al concentrations exceeded the standard of WHO and USEPA in North America, and Cd, Zn, Ni, Al, Mn, and As contents were above the standard threshold in Europe (Zhou et al. [2020\)](#page-17-2). This study found that Pb, Cu, Cd, Cr Ni As, and Mn slightly increased during discharge and recharge periods compared to the study of Zhao et al. [2017.](#page-17-12) Cd concentration of TGR varies from 0.11 to 8.28 μg/L and increases in the recharge period exceeding the threshold limit. Yangtze River reported 4.7 μg/L in 2015 but within the standard (Wang and Zhang [2018](#page-16-11)). Southern reservoirs

in China showed low Cd levels compared to TGR (Wang et al. [2018](#page-16-6)). Zn and Mn concentrations vary from 25.15 to 158.80 μg/L and from 2.87 to 121.19 μg/L, respectively, over the low and high water levels. The As level varied from 0.20 to 7.01 μg/L over dam operation and noted an increasing trend during the reservoir recharge. Zn and As concentration signifcantly increased in the Yangtze River over the last 20 years due to anthropogenic contribution (Zhao et al. [2019](#page-17-0)). The highest Cr content, 2.53 μg/L, was found during the recharge, whereas the highest Cr value of 5.26 μg/L was found during the discharge period. The highest Cu values were found (10.79 μ g/L, 10.68 μ g/L) during the recharge and discharge period, respectively. In reservoir water fuctuation periods, the aluminum concentration varies within (69.85–898.24 μg/L), and an increasing trend was observed in the discharge period. However, Cr, Cu, As, Ni, and Zn were not above the standard level and lower than the Han River and Danjiakou Reservoir in the upper Yangtze (Zhao et al. [2019\)](#page-17-0). Globally, these heavy metals values in TGR are greater than the standard values in South America, Asia, and Africa (Zhou et al. [2020\)](#page-17-2). Overall, temporally the heavy metal contents of Pb, Cd, Cr, Cu, Ni, Fe, and Al increased in the recharge period, while Zn, Mn, and As levels increased in the discharge period.

The heavy metals of Pb, Cd, Zn, Mn, As, Ni, Cr, Cu, and Al concentration were mapped for spatial distribution along the mainstream from upstream (Chongqing) to downstream (after Yichang) in TGR, as shown in Fig. [2](#page-5-0). These heavy metals showed signifcant regional variation across the TGR basin, mainly upstream and centralized urban areas, that might correlate with industrial and agricultural productions. The highest values of Pb were found at the upstream sites of the TGR (CQ, CS, FL) during the discharge period. In contrast, the recharge period sharply increases at the downstream sampling sites of the reservoir (NH, SS, and HG). The early study mentioned that Pb pollution of the

Fig. 2 Spatial distribution of heavy metals in Three Gorges reservoir

Yangtze basin is due to automobile emissions, batteries, and lead mining (Ma et al. [2016\)](#page-16-1). Cd concentration fuctuates across the reservoir basin from upstream to downstream; the highest cadmium concentration was found at the Chongqing site in both water fuctuation periods in TGR. Zn concentration increased from the middle of the reservoir (YD, WZ, YY, MG, FG) to downstream sites (TGD, TGU, YH, JZ) in the recharge phases and the highest Zn concentration was

Fig. 2 (continued)

observed at Chongqing and Changshou during the discharge period. These results are similar to previous studies, which noted that the Chongqing city constantly increased Zn, Cd, and Pb (Zhao et al. [2017](#page-17-12), [2019](#page-17-0)). Heavy metal variation in the upper reach of the Three Gorges reservoir mainly depends on the polluted wastewater from industries, agricultural and residential sectors (Gao et al. [2016\)](#page-16-12). Upstream sampling sites Chongqing, Fuling, and Badong, recorded an

Fig. 2 (continued)

increasing trend of Pb, Cd, Zn, Mn, As, Ni, Cu, Cr heavy metals. These results are compatible with reported studies that found Pb and Zn pollution increased at Chongqing and Yangtze mainstream since 1980 (Zhao et al. [2017\)](#page-17-12). Mn, As, and Ni (Fig. [2](#page-5-0)) also obtained the highest values at the upstream (Chongqing) and downstream sites after the reservoir dam during the recharge period. Chongqing is the largest urban center and industrial hub in southwest China;

therefore, sewage discharge from Chongqing city is often released to the upstream sites of TGR (Chen et al. [2011](#page-16-9); Deyerling et al. [2014;](#page-16-21) Ma et al. [2016](#page-16-1)). Chemical fertilizer and pesticide 7% and 9% migrate to the reservoir via drainage pipelines, subsurface flow, and surface runoff (Gao et al. [2016](#page-16-12)). Previous studies reported that the mainstream of TGR recorded a high concentration of Pb, Cu, Cd, Cr Ni As, and Mn (Zhao et al. [2019\)](#page-17-0). This study observed that Zn, Mn, and As concentration is higher in the discharge period (wet) while Pb, Cr, Cu, Cd, Ni, and Al increase the recharge period (dry). The water quality of the main stem in TGR varies according to the water level of the reservoir. Low dilution capacity in the water samples and high sedimentation from suspended particles lead to high concentrations of heavy metals in the water (Zhao et al. [2017\)](#page-17-12). We have noticed that heavy metals concentration at the downstream sampling sites were decreased slightly, especially after the dam; the possible reason might be that suspended solid particles mainly deposited by dam interception. Heavy metals are prone to be adsorbed on the surface of the sediments; thus, most of the heavy metals migrate to the sediments deposits, resulting in low heavy metals concentration of surface

water downstream of TGR (Zhao et al. [2019](#page-17-0)). On the other hand, downstream of the reservoir area, bedrock materials from Wushan to Badong are mainly limestone. The natural background values of these heavy metals are high; hence, heavy metals downstream of TGR may be related to natural or geological factors (Li et al. [2019](#page-16-22)).

Evaluation of the pollution status by using pollution indices in the Three Gorges reservoir

Heavy metal pollution index

This study calculated the heavy metal pollution index (HPI) for Pb, Cd, Cr, Cu, Zn, Mn, As, Ni, and Fe and Al in discharge and recharge periods, as shown in Table [2](#page-8-0). HPI values can be assigned into three categories such as low pollution index $(<19$), medium pollution index $(19-38)$, and high pollution index (< 38) (Huang et al. [2021](#page-16-20)). In this study, HPI values were obtained 22.76 and 66.17 for the discharge and recharge period, respectively. Thus, the discharge period indicated medium pollution while the recharge period showed heavy pollution in the reservoir. However,

Table 2 Heavy metal pollution index for heavy metals in surface water in Three Gorges Reservoir during water regulation phases (Discharge and Recharge)

Heavy Metals	Average $(\mu g/L)$	Standard values for heavy met- als by USEPA (Si)		Sub index (Qi)		Unit weight $(Wi = K/Si)$	
Discharge period							
Pb	6.84	15	45.6		0.141		6.4
Cd	0.51	5	10.3		0.422		4.3
Cr	1.03	100	1.0		0.021		0.0
Cu	3.49	1300	0.3		0.002		0.0
Zn	58.66	1000	5.9		0.002		0.0
Mn	31.06	50	62.1		0.042		2.6
As	2.97	10	29.7		0.211		6.3
Ni	1.25	15	8.3		0.141		1.2
Fe	296.74	300	98.9		0.007		0.7
\mathbf{Al}	227.19	200	113.6		0.011		1.2
	$\sum Wi = 1$	ΣW i x Q i = 22.75602	$HPI = 22.76$				
Recharge period							
Pb	34.46	15		229.7		0.141	32.4
C _d	2.95	5		58.9		0.422	24.9
Cr	1.19	100		1.2		0.021	$0.0\,$
Cu	4.03	1300		0.3		0.002	0.0
Zn	53.49	1000		5.3		0.002	$0.0\,$
Mn	14.50	50		29.0		0.042	1.2
As	1.84	10		18.4		0.211	3.9
Ni	1.30	15		8.7		0.141	1.2
Fe	376.71	300		125.6		0.007	0.9
\mathbf{Al}	317.61	200		158.8		0.01	1.7
	ΣW i = 1		ΣW i x Q i = 66.1678	$HPI = 66.17$			

the critical threshold value is 100, revealing that the surface water of TGR was not critically polluted over both water fuctuation phases of the reservoir. The weightage value of Pb (32.4) and Cd (24.9) were very high during the recharge period, resulting in high HPI values in the recharge period. Thus Pb and Cd concentration highly contributed to HPI calculations. HPI values of this study were higher than the previous study (Ma et al. [2016\)](#page-16-1); they obtained that HPI values of 2.02 recharge period (October 2014) and 3.57 discharge period (January 2015). Thus, it is noted that within four years, pollution levels of the reservoir were increased signifcantly. Prolonged water retention time in TGR and pollutants transfer from the input rivers such as Yangtze, Jialing, Wujiang contributed to the high pollution during the impoundment period of TGR (Ma et al. [2016](#page-16-1)). However still, TGR water was not above the critical pollution zone.

Heavy metal evaluation index

Heavy metal evaluation index (HEI) results were summa-rized in Table [3](#page-9-0). HEI values explain that < 10 , low pollution, 10–20 moderate, and > 20 high pollutions. In this study, the calculated HEI values were 2.89 and 5.59 discharge and recharge periods, respectively, indicating low water pollution in TGR. However, further digging into the HEI results, we noted that the highest values of M_i/MAC 4.46 and 2.36 of Pb concentration were found for discharge and recharge

period, respectively. These results indicate that Pb concentration mainly contributed to the high HEI values of TGR. The existing studies showed that calculated mean HEI values were 4.58 and 3.48 for the recharge period and discharge period in TGR during 2014–2015 (Ma et al. [2016\)](#page-16-1). Compared to this study, HEI values of existing literature noted that the main input rivers of Yangtze, Jialing, and Wujiang were high, indicating that most HM pollutants migrate from those rivers to the TGR (Zhao et al. [2019\)](#page-17-0).

Degree of contamination

The degree of contamination (C_d) of the reservoir water was analyzed in Table [3.](#page-9-0) The results of C_d can be interpreted as \lt 1 low pollution, 1–3 moderate pollution, and $>$ 3 high pollutions. The degree of contamination (C^d) in TGR, for the discharge and recharge period, was obtained 6.11 and 3.41 respectively, showed greater than three, suggesting that water is highly polluted in the discharge and recharge period. These results indicated pollution level was high in the discharge period, and the results were compatible with (Ma et al. [2016\)](#page-16-1). The Cf values of the C_d analysis showed that heavy metals that contributed to the pollution were in order of $Cu > Zn > Cr > As Ni > Cd > Pb > Mn$ in the discharge period. In contrast, the recharge period Pb > Cu and Zn > $Cr > Fe > Mn HM$ contributed for Cd values. However, it is noted that Pb becomes the most contaminant metal in the

Table 3 Heavy metal evaluation index (HEI), and degree of contamination (Cd) in [water](#page-10-0) [pollution index](#page-10-0) (WPI) Three Gorges reservoir

recharge period. The previous study obtained C_d results of − 4.42 and − 5.52 for the water impoundment and release periods, respectively. It is also mentioned that Pb concentration contributed to high reservoir contamination (Ma et al. [2016](#page-16-1)).

Water pollution index

The WPI results in TGR are summarized in Table [3.](#page-9-0) The results interpretation mentioned that 0–1 is acceptable for a specific purpose, > 1 how many times the polluted water should be diluted for acceptable range, and $\lt 0$ is water deficient in the factor. The results showed that Pb (3.45), followed by Fe (1.26), contributed to the high HM pollution in surface water in TGR during the recharge period. The tested water with a Pb value of 34.46 μg/L needs to dilute with 3.45 μg/L to return to the acceptable range. Similarly, tested water of Fe value of 376.71 μg/L should be diluted with 1.26 μg/L acceptable range. Cd, Mn, and As within 0–1 water quality is acceptable for specifc use. While Cr, Cu, Zn, and Ni were less than 0, suggesting a defcient factor in the recharge period. WPI indicated that Fe, Pb, Mn As, and Cd were within the 0–1 acceptable for human use in the discharge period. Cr, Cu, Zn, and Ni were less than 0 indicated deficient factors heavy metals in TGR. There were no heavy metals found > 1 in the discharge period. Though the calculated C_d , HPI and HEI, and WPI may induce different results, water pollution evaluation deduced from various methods was comparable. Overall, this study found that Pb, Cd concentration highly contributed to the heavy metal pollution during the water impoundment period (recharge) of the Three Gorges Reservoir.

Heavy metal toxicity load

Heavy metals toxicity loads (HMTL) quantify that pollution load necessary to eliminate heavy metals from the water body for human use (Kumar et al. [2019\)](#page-16-17). HMTL evaluates the heavy metals concentration in the water bodies which impacts human health. It signals the responsible authority about the water resources required treatments to the standard consumption level. This study calculated HMTL for the toxic heavy metals of Pb, Cd, Cr, Cu, Zn, Ni, Mn As, and Al for the recharge and discharge phases in TGR, as shown in Table [4;](#page-10-1) in order to assess toxic pollutants load in the Three Gorges Reservoir that leads to non-carcinogenic health risk. Agency for Toxic Substances and Disease Registry (ATSDR) and USEPA are concerned about various materials found in considerable amounts at NPL sites, which have a potential threat to human health and provide efficient treatment and management methods (Saha and Paul [2018](#page-16-23); Huang et al. [2021\)](#page-16-20). Hazard intensity scores were obtained from (ATDSR [2019\)](#page-16-19). Permissible concentrations

Table 4 Heavy metal toxicity load (HMTL) of Three Gorges reservoir, the relative toxicity level of heavy metals responsible for human health hazards, and Heavy metal removal percentage to reduce pollution in the reservoir

Metals	HIS	HMTL µg/l HMTL permissible μ g/l		% HM removal
Discharge period				
Pb	1531	10,471.81	22,965	PLT
Cd	1318	678.07	39,540	PLT
Cr	1149	1183.67	57,450	PLT
Cu	805	2805.55	1,046,500	PLT
Zn	913	53,552.93	4,565,000	PLT
Mn	797	24,752.29	239,100	PLT
As	1676	4969.44	16,760	PLT
Ni	993	1238.46	69,510	PLT
Al	685	155,625.01	137,000	11.96
		HMTL 255,277.24 µg/l		
Recharge period				
Pb	1531	52,759.38	22,965	56.47
C _d	1318	3883.59	39,540	PLT
Cr	1149	1365.60	57,450	PLT
Cu	805	3243.13	1,046,500	PLT
Zn	913	48,839.15	4,565,000	PLT
Mn	797	11,558.11	239,100	PLT
As	1676	3077.02	16,760	PLT
Ni	993	1291.27	69,510	PLT
Al	685	217,561.45	137,000	37.02
		HMTL 343,578.70 μg/l		

Hazard intensity score (HIS) taken from ATSDR (2019)

were in µg/L Pb 15, Cd 30, Cr 50, Cu 1300, Zn 5000, Mn 300, As 10, Ni 70, and Al 200 (USEPA [2009](#page-16-24)). In the discharge period, the HMTL was 255,277.24 µg/L and ranges (678.08–155,625.01), whereas the HMTL 343,578.70 µg/l ranged (1291.27–217,561.45) from the recharge period. In the present study, HMTL is lower than the permissible toxicity load except for Pb and Al, revealing lower contamination. Lead and Aluminium toxicity removal percentage is 56.47% and 37.02%, respectively. It was suggested to proceed with the toxicity removal plans for Pb in TGR. Since TGR serves over 30 million population, the drinking water standard is crucial, and health risk from $Cr > As > Cd > Pb > Cu >$ Zn is slightly higher than the acceptable health risk in the TGR mainstream during the low water level period (Zhao et al. [2019\)](#page-17-0). Meanwhile, drinking water ingestion increased from 2 to 3 in orders of magnitude larger than dermal contact. A previous study for water bodies in southern China cited that heavy metals As, Cd, Sb, and Mn in the Xiangjiang River exceeded the HMTL than the permissible limit. It was suggested that toxic metals need to be removed for the Xiangjiang River to be a safe source of drinking water (Huang et al. 2021). These results help provide an efficient and heavy metal treatment management plan in the reservoir. Elimination of the excess heavy metal concentration in the water is essential for aquatic organisms, human health, and safety. One study highlighted that water bodies near industrial sites need more intensive treatment than those from residential areas. (Zhao et al. [2019\)](#page-17-0). HMTL support for water resource management authorities to launch suitable water quality treatment methods. Rather than implementing treatment measures, mitigation of pollutants entering the water body is a cost-efective method. Thus, identifying the sources of contaminants is vital in heavy metals studies.

Potential ecological risk index

The study of Hakanson ([1980](#page-16-25)) was proposed in a sedimentology perspective, the Potential Ecological Risk Index method, to assess heavy metal contamination, characteristics, and behavior. Furthermore, Hakanson's method uses to estimate the ecological efects of toxicology. This method includes a toxic response factor of each heavy metal contamination, a single contamination coefficient, a Potential Ecological Risk Index, and a comprehensive contamination measure. Hakanson's method was applied water samples in TGR to fnd out RI in HM contamination, and results were concluded in Table [5.](#page-11-0) According to contamination factor, results explain, $C_f^i < 1$, non contamination, $\geq 1 < 2$, Light, $\geq 2 < 3$ moderate and ≥ 3 ,

heavy pollution. The heavy metals in TGR, Cd, Cr, Cu, Zn, and Ni were within < 1 indicated non-contamination, while Pb, Mn, and As demonstrate ≥ 1 light contamination in the discharge period. While the recharge period, the contamination factor indicated that Pb is $\geq 2 < 3$ shows moderate pollution, and other HM showed light contamination. The ecological risk index E_r^i demonstrates the risks of various pollutants and pervasive impacts in the ecosystems (Huang et al. [2021](#page-16-20); Saha and Paul [2018\)](#page-16-23). In this study, ecological risk index values are shown in Table [5](#page-11-0) and explain the values' standards are $<$ 40, low $\geq 40 < 80$, moderate $\geq 80 < 100$, strong ≥ 320 , extremely high. And it is showed low contamination during the discharge and recharge period in TGR. The potential ecological risk delineated in Fig. [3](#page-12-0) shows RI values and the TGR sampling sites. Most sampling sites in TGR have a low ecological risk from heavy metal pollution. RI results for heavy metals in TGR are 9.07 and 31.60 during the discharge and recharge period, respectively, indicating low potential ecological risk from heavy metals in the Three Gorges Reservoir. However, the ecological risk index values' order of heavy metals Cd > Pb > As >Ni > Mn revealed that these heavy metals infuenced the ecological risk index over water fuctuation periods in the Three Gorges Reservoir. Spatially, Lead and Cadmium risk increase at downstream sites after the TGR dam (NG, NE, XZ, SS) and upstream (CQ and XT). These sites are located around agricultural and industrial lands (Huang et al. [2021;](#page-16-20) Zhu et al. [2019](#page-17-3)); thus, Pb and Cd distribution in the Three Gorgeous

and potential ecological risk index for heavy metals in the Three Gorges reservoir during recharge and discharge period

Table 5 Ecological risk index

 T_r^i values were obtain from Håkanson, 1980, Background value for C_F^i taken from WHO, 2011

Fig. 3 Variation of potential ecological risk index in Three Gorges reservoir

Reservoir during recharge and discharge periods across all the sampling sites showed signifcant risk in this study. Previous studies also noted that Cd, Pb, As, Ni, and Mn have a serious ecological threat for the upper Yangtze sediments (Yi et al. [2016\)](#page-17-13). Other southern rivers also reported severe ecological risks from Pb and Cd (Huang et al. [2021](#page-16-20)). The heavy metals of Cd, Cr, Ni, Cu, Zn, As, and Pb have harmful impacts on the Zijiang River in South China (Zhang et al. [2018](#page-17-8)).

Source apportionment of heavy metal pollution in Three Gorges Reservoir

The source identifcation of the heavy metal pollutants in the aquatic system is essential for water security and management. Accumulation of heavy metals in the aquatic systems mainly serves from natural sources such as rock weathering, atmospheric deposition, and erosion surface runoff, while human activities contribute from agricultural runof, industrial, and residential infux (Wang et al. [2018](#page-16-6)). Continentally, heavy metal pollution occurred in Asia and Europe due to mineral weathering, manufacturing, and mining activities. In comparison, North America is responsible for agricultural fertilizer, pesticide, manufacturing, and mining industry. Africa mainly contributed to fertilizer and rock weathering. South America (rock weathering, sewage, mining and, agricultural fertilizer) (Zhou et al. 2020 ; Kumar et al. 2019). In this study, we applied multivariate analysis to identify HM sources in the Three Gorges Reservoir, compiling reliable statistical analysis such as correlation matrix, PCA, and CA (Zhang et al. [2014;](#page-17-14) Zhao et al. [2017,](#page-17-12) [2019\)](#page-17-0).

Correlation coefficient analysis for heavy metals in TGR

Correlation analysis for heavy metals reveals the metals' migration and origin (Wang et al. [2013](#page-17-15); Zhao et al. [2017](#page-17-12); Zhang et al. [2014\)](#page-17-14). If there is no correlation between the metals, then the metals are not controlled by a single factor (Ke et al. [2017](#page-16-0)). A positive correlation among the metals indicates possible interaction among them (Saha and Paul [2018](#page-16-23)). Pierson correlation applied for heavy metals in the TGR discharge and recharge period as shown in Table [6.](#page-13-0) The correlation result varies among the heavy metals based on the water fuctuation periods in the reservoir and the pollutant sources of the HM (Ma et al. [2016\)](#page-16-1). The main toxic heavy metals showed a signifcant positive correlation at *P* < 0.01 among Pb-Cd (0.814) and Pb-Zn (0.521), and a signifcant positive correlation at (*P* < 0.05) was observed for Pb, Mn, As, Ni during the discharge periods. These results indicated that Pb, Cd, Zn, Mn, As, Ni, Cr, and Cu have a similar origin during the water-releasing period of the reservoir. Previous studies noted that mineral mining, machinery, smelting industrial-based production in the upper reach of the reservoir are responsible for this heavy metal pollution (Zhao et al. [2019;](#page-17-0) Gao et al. [2016;](#page-16-12) Ma et al. [2016;](#page-16-1) Wang and Zhang [2018](#page-16-11)).

In the water impoundment period of the reservoir, a significant positive correlation coefficient was observed at *P* < 0.01 among Pb–Cd (0.895), Pb-Zn (0.485), Pb-As (0.575), Cd-Zn, and (0.631) and Cd-As (0.516). Cr positively correlates with Cu, Mn, As, Ni, Fe, and Al at *P* < 0.01. These results revealed that heavy metals might originate from similar sources or be impacted by the same factors. A previous study explained that mixed factors might be infuenced by several heavy metal pollution in the recharge period of TGR (Ma et al. [2016\)](#page-16-1). Here, we noticed that Pb, Cd, and Zn were same-origin during the recharge and discharge period. The correlation results are compatible with previous studies of heavy metals in the Three Gorges Reservoir (Zhao et al.

[2017](#page-17-12), [2019](#page-17-0); Ma et al. [2016;](#page-16-1) Gao et al. [2016](#page-16-12)). Heavy metals of Pb-Cd, Pb-Cr, Pb-Zn, and Pb-Cu signifcantly correlated in the Three Gorges Reservoir, suggesting that these heavy metals have compound contamination and homology (Zhao et al. [2019](#page-17-0)). However, it is complex to defne the sources of heavy metals only based on the correlation coefficients. Therefore, we applied PCA analysis to the heavy metals in the reservoir to support correlation results.

Principal components analysis

The PCA analysis results are shown in Table [7](#page-14-0) and Fig. [4](#page-14-1) with a rotated varimax diagram to identify the heavy metal sources in TGR. According to the results of PCA, two rotated components were extracted with eigenvalues > 1 , accounting for a cumulative 80.28% of the total variance during the recharge period. PC1 heavy loaded with Fe, Ni, Al, Mn, Cu, Cr, which explained 58.50% of the total variance, exhibited eigenvalues of 5.8, indicating the similar origin of heavy metals. PC2 explained 21.77% of the total variance and high loading of Pb, Cd, Zn, and As with eigenvalues of 2.1, revealing the same source or inputs of metal pollutants. PC1 group of metals, Fe, Ni, Al, Cr, Mn, and Cu, shared a strong signifcant positive correlation. According to our spatial distribution maps, Fig. [2](#page-5-0) clearly showed that these heavy metals obtained the increasing trend in the downstream side of the reservoir. Bedrock materials downstream of TGR from Wushan to Badong are mainly limestone. The natural background values of these heavy metals are high; hence, heavy metals downstream of TGR might be related to natural factors of bedrock weathering or geological factors (Li et al. [2019\)](#page-16-22). Therefore, we suggest that the PC1 group of heavy metals might create from natural sources and mining activities such as rock weathering. These results are further confrmed with the previous studies, indicating that heavy metals with high natural background values were mainly increased the downstream of TGR might origin from geological factors (Ma et al. [2016](#page-16-1); Gao et al. [2016](#page-16-12); Zhao

Table 6 Pierson correlation results for heavy metals in Three Gorgeous reservoirs during the discharge and recharge period

Discharge period

Table 7 Plot of loading of principal component analysis (PCA) results in discharge and recharge phases of the Three Gorges reservoir

Extraction method: a principal component analysis. Rotation method: varimax with Kaiser normalization

et al. [2017](#page-17-12)). The second factor, PC2, is loaded with Pb, Cd, Zn, and As. Zn was mainly produced from mining, machinery manufacturing, electroplating, smelting, etc. Cadmium is widely used in manufacturing industries, and more than 70% of the industrial fuel in Chongqing city comes from fossil fuels (Zhao et al. [2019](#page-17-0)). The lead was mainly derived from the water transportation facilities. Upon completing the construction of TGR, large amounts of ships were gathered in the reservoir, and fuel combustion and exhaust emission increased (Zhu et al. [2015,](#page-17-6) [2019](#page-17-3)). Waste and chemical discharge from the Pb industry and the electroplating industry are released to the reservoir (Wang and Zhang [2018](#page-16-11)). Arsenic mainly originates from agricultural lands with pesticides. Sichuan basin is the main hub for agriculture in southwest China, and TGR is located surrounding farmlands in the mountainous areas (Zhu et al. [2019](#page-17-3); Gao et al. [2016](#page-16-12)). Seasonally, pesticide and chemical fertilizers apply in these farmlands, and these agricultural discharges add to the reservoir (Ma et al. [2016\)](#page-16-1). Therefore, heavy metals of the PC2 group originate from industrial and agricultural pollution during the discharge period in TGR.

The rotated component matrix results of the recharge period in TGR are shown in Fig. [4](#page-14-1) and Table [7](#page-14-0). Threerotated component factors were extracted with eigenvalues>1, cumulative of 84.02% of the total variance. PC1 has strong positive loadings for Mn, Ni, Fe, and Cr, which explained 47.97% from total variance, and eigenvalues are 4.9.

PC2 has strong positive loading for Pb Cd, Zn, As which explained 25.95% of the total variance with 2.59 eigenvalues. PC3 has strong positive loading only for Cu, which explained 10.08% of the total variance and 1.01 eigenvalues. Thus, the PC1 (Mn, Ni, Fe, and Cr) group of heavy metals is mostly related to natural origin, as explained earlier, similar to the discharge period. PC2 (Pb Cd, Zn, As) in the Yangtze and TGR basin, these Zn, Pb, As, and Cd mainly

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derived from the industrial and agricultural discharges, including metal industry, mining activities. According to the spatial distribution of the heavy metal in TGR (Fig. [2](#page-5-0)), in the recharge period, highest values of Pb, Zn, As, and Cd was found at the upstream sites (Chongqing to Wangzhous), where main industries located in the reservoir basin. The results further confrmed with the previous studies (Zhao et al. [2019](#page-17-0); Ma et al. [2016;](#page-16-1) Zhu et al. [2019](#page-17-3); Gao et al. [2016](#page-16-12)). In comparison, PC3 strongly correlates with Cu metals, which mainly originated from the Cu mining industry, the Three Gorges Reservoir. (Zhao et al. [2019\)](#page-17-0).

Cluster analysis

Spatial cluster analysis (CA) for heavy metals was further applied to confrm heavy metal sources in the Three Gorges Reservoir, as shown in Fig. [5](#page-15-0) a and b discharge and recharge periods, respectively. CA provides a dendrogram clustered into two categories; cluster group 1 (Cr, Ni, Cd, Cu, As, Pb, Mn, and Zn) and cluster group 2 (Fe and Al) were obtained according to their similarities in the reservoir discharge period. It is clearly shown that cluster group 1 heavy metal originates from anthropogenic factors such as industrial waste discharge, mining activities, metal processing, and agricultural runoff pollution. Cluster group 2 mainly originates from natural factors during the discharge period in TGR. In contrast, in the recharge period, two cluster groups were extracted. Cluster group 1 (Cr, Ni, As, Cd, Cu, Mn, Pb, and Zn) and cluster group 2 (Fe and Al) showed similar results of the discharge period. Thus, cluster 1 heavy metals are associated with anthropogenic factors, and cluster 2 group metals are from natural origin. A small distance in the dendrogram shows a close relationship among the metals. The CA result is reasonably similar to PCA results on heavy metal source classifcation. Further studies suggested with land use pattern in TGR.

Conclusion

Heavy metal pollution and ecological risk were investigated for ten heavy metals during the recharge and discharge period in the Three Gorges Reservoir in China. The heavy metal concentration increased temporally in the order of $Cr < Ni < As < Cd < Cu < Mn < Ph < Zn < Al < Fe$ during the recharge period. While discharge period $Cd < Cr < Ni$ $<$ As $<$ Cu $<$ Pb $<$ Mn $<$ Zn $<$ Al $<$ Fe. Spatially, Pb, Zn, Cd, As, Mn, Ni, Cr, and Cu signifcantly varied at the upstream and downstream sampling sites. Pb concentration sharply increased during the recharge period, ranging from 6.93 to 148.62 μg/L, and exceeded WHO and USPEA standards limits for drinking water in the reservoir. HPI, HEI, C^d , WPI indicated low pollution and moderate pollution with Pb and Cd infuence in the discharge and recharging period, respectively. HTML values are below the permissible toxicity load except for Pb. The Pb toxicity removal percentage is 56.47% and is suggestive of the removal process for Pb in TGR. The calculated ecological risk index (RI) indicates that (Pb, Cd, As Ni, and Mn) and (Pb and Cd, Cr, Cu, and Zn) were the most ecological risk heavy metals during discharge and recharge period, respectively. Pb and Cd distribution was a signifcant risk across all the sampling sites in TGR. PCA, CA, and correlation results found that regardless of the water level fuctuation periods, Pb, Cd, Zn, Mn, Ni, As, Cr, Cu originate mainly from anthropogenic such as industrial wastewater, mining, metals processing, and agricultural runof. Fe and Al are mainly from the natural origin of bedrock weathering. The study offered comprehensive recent HM pollution status, toxicity loads, ecological risk assessment, and pollution sources, suggesting a vital insight into heavy metal pollution and water security management in Three Gorges Reservoir.

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