


Distribution and health risk assessment of dissolved heavy metals in the Three Gorges Reservoir, China (section in the main urban area of Chongqing)

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Abstract The Three Gorges Project (TGP) is the largest hydropower station ever built in the world. A better understanding of the concentrations of heavy metals in the aquatic environment of the Three Gorges Reservoir (TGR) is crucial for national drinking water security and sustainable ecosystem development. To thoroughly investigate the impact of heavy metals on water quality after the impoundment to the maximum level of 175 m in the TGR, the concentrations of the dissolved heavy metals (Cr, Cu, Zn, Cd, Pb, As) were measured in April and August 2015, by inductively coupled plasma mass spectrometry (ICP-MS). (1) Except Zn and Pb, most of the heavy metal concentrations in the water of the TGR reached the level of the National Surface Water Environmental Quality Standards (GB3838-2002) I of China, revealing that the water quality of the TGR was good overall. (2) There were significant positive correlations among the concentrations of Cu, As, and Cd, revealing that they may exhibit similar geochemical behaviors. (3) The spatial distribution of the heavy metal concentrations was diverse and complex. The Zn concentration obviously increased in the rainy season from upstream to downstream in the Yangtze

River, while the other heavy metals exhibited no significant changes in their concentrations. The distribution characteristics of the heavy metal concentrations on both sides and the middle of the river were different at different sites. (4) The health risk of the six elements was assessed through a human health risk assessment (HHRA), and the assessment results were lower than the maximum acceptable risk level designed by the US EPA and International Commission on Radiological Protection (ICRP). The HHRA model in the aquatic environment revealed that the risk of non-carcinogenic heavy metals (Cu, Zn, and Pb) was at a negligible risk level of 10^{-11} – 10^{-9} a⁻¹. At all the study sites, the risk of carcinogenic heavy metals (Cr, Cd, and As) was higher than the risk of non-carcinogenic heavy metals. As was the most important risk factor, followed by Cr. The results of this study hold great significance for a timely understanding of the changing water quality for affected departments to ensure the health of the residents in the TGR area.

Keywords Three Gorges Reservoir · Water quality · Dissolved heavy metal · Distribution · Health risk assessment

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Introduction

The Three Gorges Reservoir (TGR) is the largest hydropower station in the world, with a designed and installed capacity of 1820×10^4 kW and a total reservoir capacity of 393×10^8 m³ (Plateau 2006). Since construction was completed in 2009, the Three Gorges Project (TGP) has resulted in significant benefits in terms of flood control, power generation, and shipping but has also had a profound impact on the environment. With the success of impoundment in TGR, the backwater of the TGR is about 660-km long and forms a typical river-type reservoir from the Three Gorges Dam up to Jiangjin City in

the Chongqing Municipality (Bing et al. 2016). Under slow flow conditions, contaminants cannot be discharged and consequently accumulate in the reservoir (Li et al. 2006; Wei et al. 2016), which deteriorates the water quality, posing a serious threat to the health of humans drinking water from the TGR. During all phases of construction until the TGP was completed, the water quality safety of the TGR attracted much attention from all sectors of society (Yi et al. 2011; An et al. 2015) and heavy metal contamination was particularly important.

Heavy metal contamination elicits attention due to its inherent toxicity, persistence, irreversibility, concealment, bioaccumulation, and non-degradability (Arnason and Fletcher 2003; Audry et al. 2004; Bibi et al. 2007; Ye et al. 2011; Xiao et al. 2012; Cobbina et al. 2015; Pejman et al. 2015; Wei et al. 2016). The main sources of metals in water are the weathering of minerals; atmospheric precipitation; and anthropogenic activities such as urbanization, industrialization, and agriculture (Demirak et al. 2006; Kavcar et al. 2009). Heavy metals accumulate in sediments and suspended solids after entering the aquatic environment (Huang 1995), as well as in the soil of the hydro-fluctuation belt around the TGR. On the other hand, a portion of heavy metals may be conditionally released into the water column through disturbances, such as hydrological, physical, chemical, and biological activities, to recontaminate the environment, making the sediments a potential pollutant source to the water column (Tessier et al. 1979; Cuong and Obbard 2006; Arain et al. 2008; Wei et al. 2016). A portion of heavy metals enters aquatic organisms and is absorbed by humans through the food chain, which is a serious threat to human health. In excess of the maximum limit required for human health, some elements that are essential for the human body can also damage human health (Sundaray et al. 2012). As a result, problems in environmental science related to the existing states, distribution, migration, destination, and influence on human health of heavy metals were concerned by the international research community because of the complex chemical behavior and ecological effects of these contaminants (Nriagu 1992, Nriagu 1996; Saager et al. 1992; Cotté-Krief et al. 2002; González-Macias et al. 2006; Li and Zhang 2010; Li et al. 2008).

Human health risk assessment (HHRA), widely used in environmental pollution assessment, is a method of quantifying the impact of pollution on human health and determining whether the environment is safe or not (Joseph et al. 2015; Wu et al. 2016; Xu et al. 2016). HHRA is currently applied mainly to polycyclic aromatic hydrocarbons in organisms and fishes in the aquatic environment. Potential ecological risk assessment has been widely used for the sediments and water-level fluctuating zone of the TGR widely (Wang et al. 2012; Zhang et al. 2014; An et al. 2015), and an HHRA of heavy metals in the water of the TGR is a lesser concern.

The heavy metal contaminations in the water of the TGR has been of concern for many years before 2010,

while few investigations have been thoroughly conducted after the water was impounded to 175 m, the final designated water level. The detailed status, such as the concentration, spatial variation, and HHRA, of heavy metals in the water of the TGR needs to be determined during the different impoundment periods. The primary objectives of this study were to (1) quantify the concentrations of six heavy metals (Cr, Cu, Zn, Cd, Pb, As) and their contamination degree, (2) determine the spatial and temporal distribution characteristics of these metals, and (3) evaluate the risk of these heavy metals to human health. Ultimately, this study is important in protecting the ecological environment and health safety of residents in the TGR area.

Sampling and analysis

Study area

The TGR is the area flooded by water due to the construction of the TGP in the Yangtze River drainage basin. The TGR (105° 44'~111° 39' E, 28° 32'~31° 44' N), with a total area of 55,742 km² and a water surface area of 1862 km² that accounts for 3.44 % of the total area, is located in the western Hubei province and the middle eastern region of Chongqing city, China (Fig. 1). Its total capacity, total reservoir length, average width, and average depth are 3.93×10^{11} m³, 660 km, 1.1 km, and 90 m, respectively. The terrain is complex and mountainous. The reservoir is located in a subtropical monsoon climate zone with an annual average precipitation of 1140~1200 mm, of which 50~65 % falls from June to September (Chen 2014).

Construction on the TGP began in 1994 and was completed in 2009. Since completion, the TGR has been operated by a scheduling mode of “store clean water and discharge muddy water.” The TGR is released to its base level of 145 m for flood control at the end of May and early June, and the water level is maintained at 145 m during the flood season of June to September. After the flood season in October, the reservoir is impounded to the final designated water level of 175 m, where it is maintained until May of the next year (Wang et al. 2011; Xiong et al. 2013; Tang et al. 2014). Hence, the water level of the TGR is at the lowest in the flood season (June to September) and at the highest level in the dry season (October to May).

The main part of the TGR area is located in Chongqing. With rapid urbanization and industrialization, the garbage and domestic sewage from Chongqing are directly discharged to the TGR, and the concentrations of Cu, Zn, and Cd are higher due to anthropogenic inputs (Wei et al. 2016). These pollutants may affect the water quality in the TGR.

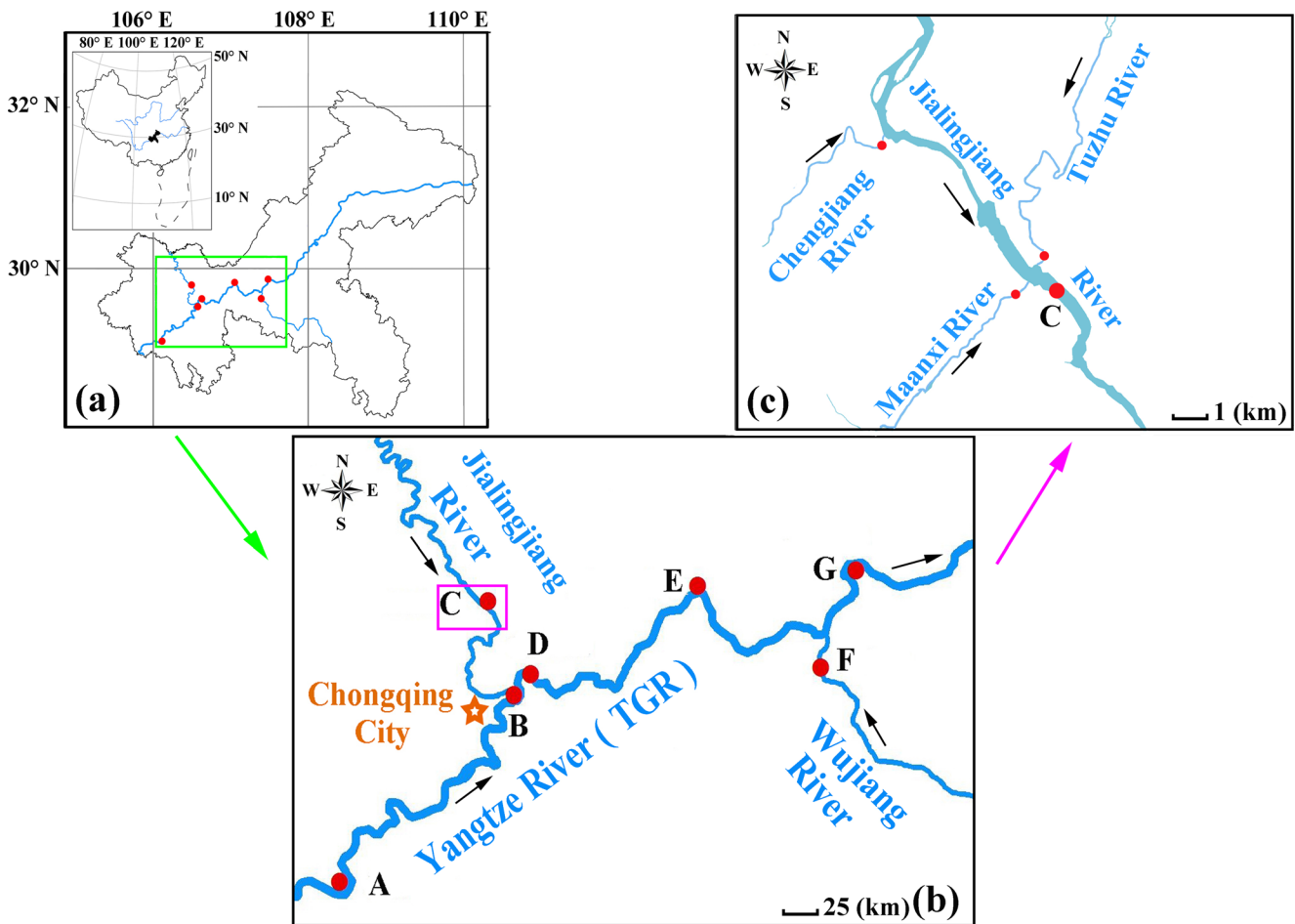


Fig. 1 Distribution of the sampling sites in the TGR. **a** Location of the sampling sites in Chongqing. **b** Distribution of the sampling sites in the Yangtze River drainage basin (TGR); red dots A–G represent Jiangjin, Wanglongmen, Cuntan, Changshou, Nantuo, Beibei, and Fuling, respectively; the pentagram and black arrows represent Chongqing City and the

flow direction of the river, respectively. **c** Distribution of the sampling sites in the Jialingjiang River basin in Beibei; the red dots represent the sampling points, C represents the sampling point where the water was collected from the Jialingjiang River in Beibei

Water sampling and analytical methods

Because of the seasonal change of water level (section “Study area”), 48 and 38 water samples were collected in April and August 2015, respectively, at seven sites (Jiangjin, Wanglongmen, Beibei, Cuntan, Changshou, Fuling, and Nantuo) in the TGR area (Fig. 1b). The water samples collected in April and August represent the dry and rainy seasons, respectively. At each sampling site, water samples were collected in the river where the water flow was obvious near the right and left banks (approximately 5 m from the bank) to represent the right and left water samples, respectively. Water was also collected in the middle of the flow of the stream to represent the middle water sample. At the same time, water samples were collected at depths of 0.5 m (surface layer), 5 m (upper layer), and 15 m (deep layer) from the surface water to the riverbed to represent the surface, upper, and deep water samples (Fig. 2), respectively. Polymer polyethylene bottles and caps were boiled for 5 h in a 1:5 volume of nitric

acid, flushed with ultrapure water, allowed to dry naturally in an ultra clean room (Bench, class 100 clean), and packaged in

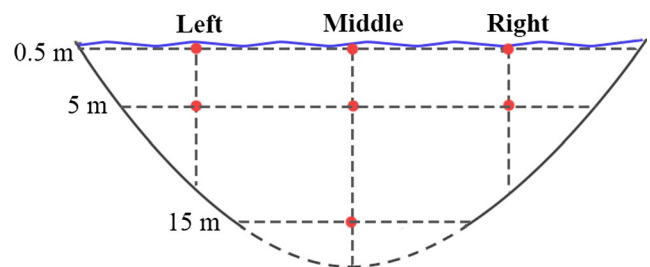


Fig. 2 Transect of sampling points in the river. Samples from every station were collected at sites located approximately 5 m from the right bank, the middle of the flow of the river, and approximately 5 m from the left bank (marked with R, M, and L, respectively) along a transect. Water samples along the water column were collected at depths of 0.5 m (surface layer), 5 m (upper layer), and 15 m (deep layer) at each site, marked by S, U, and D, respectively. Because the depth and flow rate of the river were different, the number of water samples was different at different sites

polyethylene bags before use. At the sampling sites, the samples were filtered through pre-washed 0.45- μm Millipore nitrocellulose filters, and the bottles were washed with the filtrate; the filtrate was then added into the polyethylene bottles and acidified to pH <2 by adding the trace level ultrapure HNO_3 (1:1) (J.T. Baker, Ultrex II Ultrapure Reagent); the bottles were sealed for transport back to the laboratory, where they were stored under refrigerated conditions until analysis. The water temperature, pH, and conductivity were measured by a Multi 3430 portable multi-parameter water quality analyzer (Germany WTW), which has a resolution of 0.1 $^\circ\text{C}$, 0.001 pH unit, and 1 $\mu\text{S}/\text{cm}$.

The concentrations of six elements (Cr, Cu, Zn, Cd, Pb, As) were measured in the samples by inductively coupled plasma mass spectrometry (ICP-MS, British GV Instruments), with a test precision of within 5 %, in the State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences.

To examine the difference in the horizontal distribution of the heavy metal concentrations in the water of Jialingjiang in Beibei, the authors collected supplementary water samples in October 2015, from the following tributaries of Jialingjiang: Tuzhu River, Chengjiang River, and Maanxi River (Fig. 1c).

Data analysis and evaluation methodology

Humans can be exposed to heavy metals via three main pathways: direct ingestion, inhalation through the mouth and nose, and dermal absorption through the skin (Caussy et al. 2003; US EPA 2004; Sekhar et al. 2005; De Miguel et al. 2007; Wu et al. 2009). Gastric and intestinal areas are exposed directly to the harmful effects of heavy metals by drinking polluted water. In addition, the skin, respiratory system, and lungs can be harmed by heavy metals in the atmosphere. However, ingestion is the main route of exposure for humans because breathing and skin contact are less affected by heavy metal pollutants compared with food sources (Caceres et al. 2005; Xu et al. 2006; Beckett et al. 2007; Kavcar et al. 2009).

HHRA, developed in the 1980s, can evaluate water quality as well as quantitatively express the risk posed by pollutants to the human body. According to the characteristics of pollutants, an environmental HHRA for water can be divided into an evaluation model for carcinogens and an evaluation model for non-carcinogens (Pan 1991; US EPA 1987, 1989, 2004; Zhang et al. 2009).

The carcinogenic risks are evaluated by Eq. (1):

$$R_c = \sum R_i^c = \sum [1 - \exp(-D_i q_i)] / 70 \quad (1)$$

where R_i^c is the average years of cancer risk of the genetically toxic substance i by direct ingestion (a^{-1}), D_i is the unit body quality daily exposure dose of the genetically toxic substance i by direct ingestion ($\text{mg}/(\text{kg day})$), q_i is the carcinogenic factor

of the genetically toxic substance i by direct ingestion ($\text{mg}/(\text{kg day})$), and 70 is the average human lifespan (a).

Non-carcinogenic risks are evaluated by Eq. (2):

$$R_n = \sum R_i^n = \sum (D_i / \text{Rf}D_i) \times 10^{-6} / 70 \quad (2)$$

where R_i^n is the average years of cancer risk value of the non-carcinogen i by direct ingestion (a^{-1}), D_i is the unit body quality daily exposure dose of the non-carcinogen i by direct ingestion ($\text{mg}/(\text{kg day})$), $\text{Rf}D_i$ is the carcinogenic factor of the non-carcinogen i by direct ingestion ($\text{mg}/(\text{kg day})$), and 70 is the average human lifespan (a).

The equation for the unit body quality daily exposure dose (D_i) for drinking is as follows:

$$D_i = 2.2 \times C_i / 70 \quad (3)$$

where 2.2 is the average daily water consumption for adults (L), and C_i is the measured concentrations of heavy metals in the water ($\mu\text{g}/\text{L}$).

Assuming that there is no antagonistic or synergistic relationship between the toxic effects of the heavy metals on human health, the total health risks of heavy metals to the human body caused by drinking are evaluated by Eq. (4):

$$R = R_c + R_n \quad (4)$$

This paper only discusses the risk posed by heavy metals to the human body through drinking water. According to the classification system compiled by the International Agency for Research on Cancer (IARC) and the World Health Organization (WHO), Cr, Cd, and As are carcinogens, whereas Cu, Zn, and Pb are non-carcinogens. The intensity coefficients of the carcinogens and the reference doses of the non-carcinogens are shown in Table 1.

Results and discussion

Concentrations of heavy metals

The minimum, maximum, mean, standard deviation, and variation coefficient of the heavy metal concentrations in water samples from different depths and offshore distances collected

Table 1 The values of model parameters q_i and $\text{Rf}D_i$ (cite from Sun et al. 2009)

Carcinogens	By drinking q_i [$\text{mg}/(\text{kg}\cdot\text{d})$]	Non-carcinogens	By drinking $\text{Rf}D_i$ [$\text{mg}/(\text{kg}\cdot\text{d})$]
Cr	41	Cu	5×10^{-3}
Cd	6.1	Zn	0.3
As	15	Pb	1.4×10^{-3}

at each site in the dry and rainy seasons were calculated and are compared with the National Surface Water Environmental Quality Standards (GB3838-2002) in Table 2. The average concentrations of dissolved heavy metals in the water of the TGR in the dry season follow the sequence Zn > As > Cu > Pb > Cr > Cd, and the concentrations of Zn and As were 128.030 and 2.566 µg/L, respectively. In contrast, the average concentration of dissolved heavy metals in rainy season lay in the sequence Zn > Cu > Pb > As > Cr > Cd. The concentration of Zn was the highest in both the rainy and dry seasons. Compared with the dry season, the concentration of Pb increased; the concentration of As decreased; and the Cu, Cr, and Cd concentrations changed little in the rainy season. The variation coefficient of heavy metals in the water of the TGR was always small, which shows that the difference in the heavy metal concentrations among the seven sampling sites was small (Sun et al. 2009).

The heavy metal concentrations did not exceed the standard of the National Surface Water Environmental Quality Standards (GB3838-2002) III in China in either the dry or rainy season. The concentrations of Cr, Cu, As, and Cd reached class I of the national water standard, and the concentration of Zn reached the class II standard. However, the concentration of Pb reached the class III standard in the rainy season. The results of Qiao et al. (2007) indicated that the Yangtze River has been polluted by Zn and Pb due to anthropogenic contributions over the past 20 years, in

comparison with data from the 1980s. As shown in Table 2, the concentration of Zn varied from 16.562 to 586.172 and 24.157 to 723.150 µg/L in the dry and rainy seasons, respectively.

The proportion of water samples with concentrations of heavy metals that exceeded the national class I standard in the dry and rainy seasons were approximately 65 and 45 %, respectively. These results illustrate that Zn may be the important contaminant in the study area. However, overall, the six elements in the TGR all meet the standard. The sand content and suspended solids increased in the reservoir with the increase of precipitation in August. Suspended matter is the main carrier of heavy metals in water and determines the migration, transformation, destination and biological effects of heavy metals in the aquatic environment (Qin et al. 2015). As a result, the sand content is one of the contributors to the higher heavy metal concentration in the rainy season compared to that in the dry season at the vast majority of sites.

The concentrations of Cr, Cu, Zn, As, and Cd did not exceed the average global background values (Klavinš et al. 2000), the Sanitary Standard for Drinking Water in China (GB5749-2006) (Ministry of Health 2006), the Guidelines for Drinking Water Quality (the Third Edition) developed by the WHO (2006), or the Drinking Water Quality Standard in the USA (2006), whereas the concentration of Pb essentially met the standards except in two samples collected in the rainy season (Tables 2 and 3). In addition, the concentrations of Cr,

Table 2 The concentrations of dissolved heavy metals in the water of TGR

Season	Heavy metals	Min (µg/L)	Max (µg/L)	Mean (µg/L)	S. D (µg/L)	Coefficient of variation	Standard values for I ^a (µg/L)	Standard values for III ^b (µg/L)	Over standard ^c rate (%)
Dry season (April)	Cr	0.210	0.865	0.490	0.133	0.274	10	50	0.00
	Cu	0.936	3.260	1.735	0.420	0.245	10	1000	0.00
	Zn	16.862	586.172	128.030	146.213	1.154	50	1000	64.58
	As	1.844	3.071	2.566	0.258	0.102	50	50	0.00
	Cd	0.013	0.085	0.045	0.015	0.331	1	5	0.00
	Pb	0.139	4.140	0.526	0.641	1.231	10	50	0.00
Rainy season (August)	Cr	0.216	0.841	0.453	0.123	0.272	10	50	0.00
	Cu	0.529	4.165	1.744	0.718	0.411	10	1000	0.00
	Zn	24.157	732.510	128.548	173.799	1.352	50	1000	44.74
	As	0.680	3.515	1.544	0.543	0.352	50	50	0.00
	Cd	0.003	0.225	0.031	0.046	1.457	1	5	0.00
	Pb	0.207	21.669	1.637	4.616	2.820	10	50	0.00

^a National Surface Water Environmental Quality Standards (GB3838-2002), China. Standard I is mainly suitable for the waters as the source water and national nature reserve

^b National Surface Water Environmental Quality Standards (GB3838-2002), China. Standard III is mainly suitable for centralized drinking water, the second protection zones for surface water source, wintering grounds, swimming travel channel for fish and shrimp, and other aquaculture, fishery waters, and swimming zones

^c The over standard rate refers to the percentage of the number of water samples in which the concentration of heavy metals is higher than the standard values in the total number of samples (standard sample number/total number of samples). Taking National Surface Water Environmental Quality Standards (GB3838-2002) I, China, as the reference standard

Table 3 The concentrations of dissolved heavy metals in TGR, and compared with the concentrations in other studies and guidelines (unit in $\mu\text{g/L}$)

Sites or guidelines	Time	Cr	Cu	Zn	As	Cd	Pb
Three Gorges Reservoir (this study)	Dry season	0.49	1.74	128.03	2.57	0.05	0.53
	Rainy season	0.45	1.74	128.55	1.54	0.03	1.64
Han River (Li and Zhang 2010)	Dry season	5.89	7.76	–	10.74	3.21	7.37
	Rainy season	10.32	18.97	–	17.58	1.38	11.02
Danjiangkou Reservoir (Li et al. 2008)	–	6.29	13.32	2.02	11.08	1.17	10.59
World average background values (Klavinš 2000)	–	1	–	–	–	0.08	–
China (Ministry of Health 2007)	–	50	1000	1000	10	5	10
WHO (2006)	–	50	2000	–	10	3	10
US EPA (2006)	MCL	100	1300	–	10	5	15
	MCLG	100	1300	–	0	5	0

MCL maximum contaminant level, MCLG maximum contaminant level goal

Cu, As, Cd, and Pb in the water of the TGR was significantly lower than that in the upper reaches of the Han River (Li and Zhang 2010), whether in the dry or rainy season. Except for Zn, the heavy metal concentrations were lower than that in the Danjiangkou Reservoir (Li et al. 2008).

Correlation analysis of heavy metal concentrations with pH and EC

Changes in the physical and chemical characteristics of the aquatic environment may influence the solubility and migration of heavy metals (Gundersen and Steinnes 2003). Therefore, in order to understand the relationship between the heavy metals with each other and with the electrical conductivity (EC) and pH of the water, the values of these

variables at the time of sampling in April and August of 2015 were analyzed by Pearson correlation analysis to yield a correlation coefficient matrix (Table 4).

The correlation of the concentrations of the six heavy metals with the pH and EC were weak in both the dry and rainy seasons, suggesting that the pH and EC were not the main factors influencing the heavy metals, but maybe affected their concentrations in water in combination with other factors.

In August, the correlation between Cd and Pb was 0.948 ($P \leq 0.01$), suggesting that the sources of the two are almost identical. In addition, the concentrations of Cd, Pb, As, and Zn were positively correlated with each other ($R = 0.634\text{--}0.948$, $P \leq 0.01$), indicating that the geochemical behavior was similar among these four elements and they may have a certain

Table 4 Correlations of the concentrations of dissolved heavy metals in TGR and their relationships with pH and EC

Month	Element	Cr	Cu	Zn	As	Cd	Pb	pH	EC
April	Cr	1							
	Cu	0.658**	1						
	Zn	0.096	0.164	1					
	As	0.321*	0.613**	−0.211	1				
	Cd	0.491**	0.653**	0.139	0.596**	1			
	Pb	0.604**	0.498**	0.235	0.306*	0.398**	1		
	pH	−0.183	−0.383**	−0.037	−0.231	−0.350*	−0.114	1	
	EC	0.089	0.060	−0.043	−0.066	0.350*	−0.081	−0.384**	1
August	Cr	1							
	Cu	0.349*	1						
	Zn	0.052	0.598**	1					
	As	0.300	0.837**	0.634**	1				
	Cd	0.084	0.699**	0.776**	0.812**	1			
	Pb	0.010	0.490**	0.760**	0.725**	0.948**	1		
	pH	0.067	−0.319	−0.331*	−0.460**	−0.490**	−0.460**	1	
	EC	−0.003	−0.387*	−0.350*	−0.678**	−0.477**	−0.489**	0.633**	1

* $P \leq 0.05$; ** $P \leq 0.01$

homology (Song et al. 2011). In April and August, Cu, As, and Cd were significantly positively correlated ($R = 0.596\text{--}0.837$, $P \leq 0.01$). The correlation coefficients between Cr and the other elements were relatively low, except with Cu and Pb in April, indicating that the source of Cr is different from the source of the other elements.

In general, the correlation of the heavy metal concentrations in the rainy season was significantly enhanced compared with that in dry season, which may have been due to a large amount of precipitation and surface runoff carrying the heavy metal particles from the land and air inflow reservoir and a substantial increase in the content of suspended particulate matter, an important carrier of heavy metal elements (Avigliano and Schenone 2015). These conditions may have been favorable to the increased heavy metal concentrations in the rainy season.

Spatial and temporal distribution characteristics of heavy metals

Distribution characteristics of heavy metals at different sites

Because the physical and chemical properties of the elements are different, the spatial characteristics of their concentrations also differ with temporal variations in time, changes in the aquatic environment, variability in the spatial pattern of the river, and altering human activities. To compare the differences in the heavy metal concentrations at the different sites, the average values of heavy metals in multiple samples (left, middle, and right of the reservoir/river and different depths of the water profile, Fig. 2) collected from each site (sites A–G, Fig. 1) were calculated and used to represent the average concentration of heavy metals at each site (Fig. 3).

In April, the difference in the concentrations (Cr, Cu, As, Cd, and Pb) at each site was small, and their concentrations in the water of Beibei and Fuling were significantly lower than that at the other stations. This result shows that the concentrations of Cr, Cu, As, Cd, and Pb in the tributaries, i.e., the Jialingjiang and Wujiang Rivers, were lower than in the mainstream Yangtze River in the dry season. The difference in the Zn concentration among the seven sites was the greatest (54.792~204.105 $\mu\text{g/L}$), and the concentration of Zn in the tributaries was higher than that in the mainstream Yangtze River, except at the Jiangjin site. The change in the heavy metal concentrations, except Zn, was small at the five sites in the mainstream Yangtze River in the dry season. Heavy metals in the river will migrate from upstream to downstream, causing the concentration of heavy metals in the upstream water to be lower than that downstream (Sekhar et al. 2005).

The concentration of heavy metals at the last site in the downstream stretch of the Yangtze River, i.e., Nantuo, was slightly smaller than that at the first site in the upstream stretch, i.e., Jiangjin, in April. This should be attributed to

the dilution effect and the existence of settling processes which can accumulate the heavy metals into the sediments (Li et al. 2008). Firstly, the concentration of heavy metals (except Zn), both in Beibei (Jialingjiang River) and Fuling (Wujiang River), is lower than that in the sites on the Yangtze River, in April (Fig. 3). So, the two tributaries can dilute the concentration of heavy metals in the Yangtze River (TGR). Secondly, the disturbance from surface runoff is small because of less precipitation in the dry season (April in this study). And the altitude difference between the upstream and the downstream of the TGR is small and the flow rate is slow, because of the high water level of the TGR in the dry season. All the conditions mentioned above are conducive to the deposition of the heavy metals and result in the decrease of heavy metal concentrations in the water of the downstream.

In August, the concentration of heavy metals in Fuling was lower than that at the other sites, revealing that the concentration of heavy metals in the water samples collected from the Wujiang River in the rainy season was lower than that in the Jialingjiang and Yangtze Rivers. The concentration of heavy metals, except Cr, was highest in Beibei, which explains why the concentrations of Cu, Zn, As, Cd, and Pb in the Jialingjiang River were higher than that in the Yangtze and Wujiang Rivers. The concentration of Zn obviously increased from Jiangjin to Nantuo in the Yangtze River, suggesting that the accumulation of Zn is stronger than the other heavy metals.

The horizontal distribution characteristics of the concentration of heavy metals at each site

The average concentrations of heavy metals in the water samples collected from the left bank, middle of the river, and right bank were mapped (Figs. 4 and 5). During dry season, in Jiangjin, the concentration of As in the middle of the river was lower than that in the left and right banks, while other heavy metals were opposite; in Wanglongmen, the concentration of Zn increased from the right to left bank, but the other heavy metals were higher in the middle of the river than on both sides; in Beibei, the concentration of As increased from the left to right bank, in contrast to the other heavy metals; at the Cuntan site, the heavy metal concentrations in the middle of the river was higher than that on both sides, except for Cr; in Changshou, the concentrations of all the heavy metals were higher in the middle of the river than on both sides; the variation of the heavy metal concentration was complex in Fuling and Nantuo. Overall, the fluctuation of the As concentration was the smallest in the left bank, middle of the river and right bank at all sites.

During the rainy season, the difference in the Zn concentration in the water samples was small between the left bank, middle of the river, and right bank in Jiangjin, Wanglongmen, Cuntan, Changshou, and Fuling but was significant in Beibei

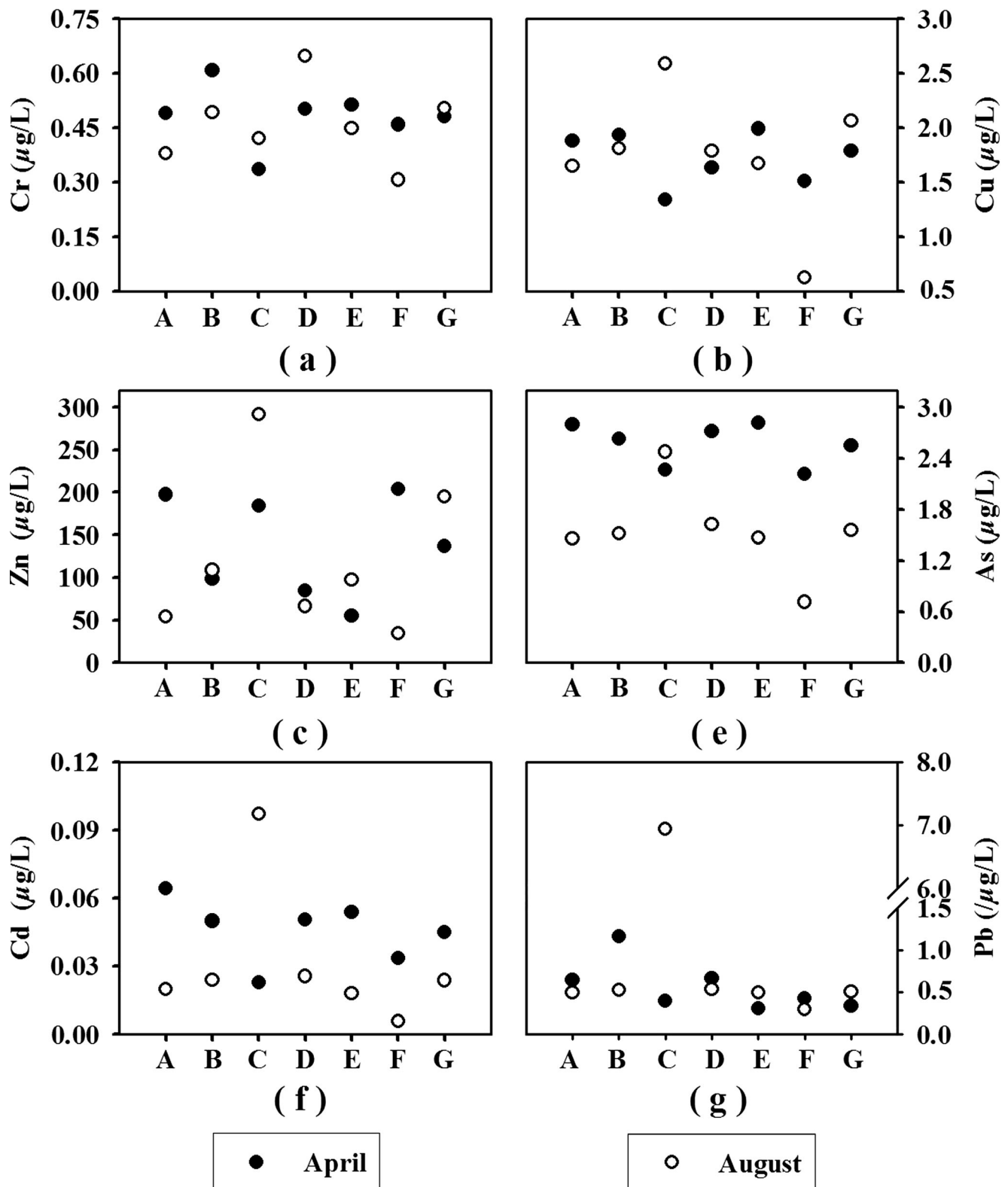


Fig. 3 Spatial distribution of heavy metal concentrations. a–f is the concentration distribution of Cr, Cu, Zn, As, Cd, and Pb, respectively; filled circles and empty circles represent the heavy metal concentrations in

April and August, respectively; A–G represent the sampling sites of Jiangjin, Wanglongmen, Beibei, Cuntan, Changshou, Fuling, and Nantuo, respectively

and Nantuo, for which the middle of the river was significantly lower and higher, respectively, than that on both sides; the

concentrations of As, Cd, and Pb in the middle of the river were lower than that on both sides in Beibei, but their

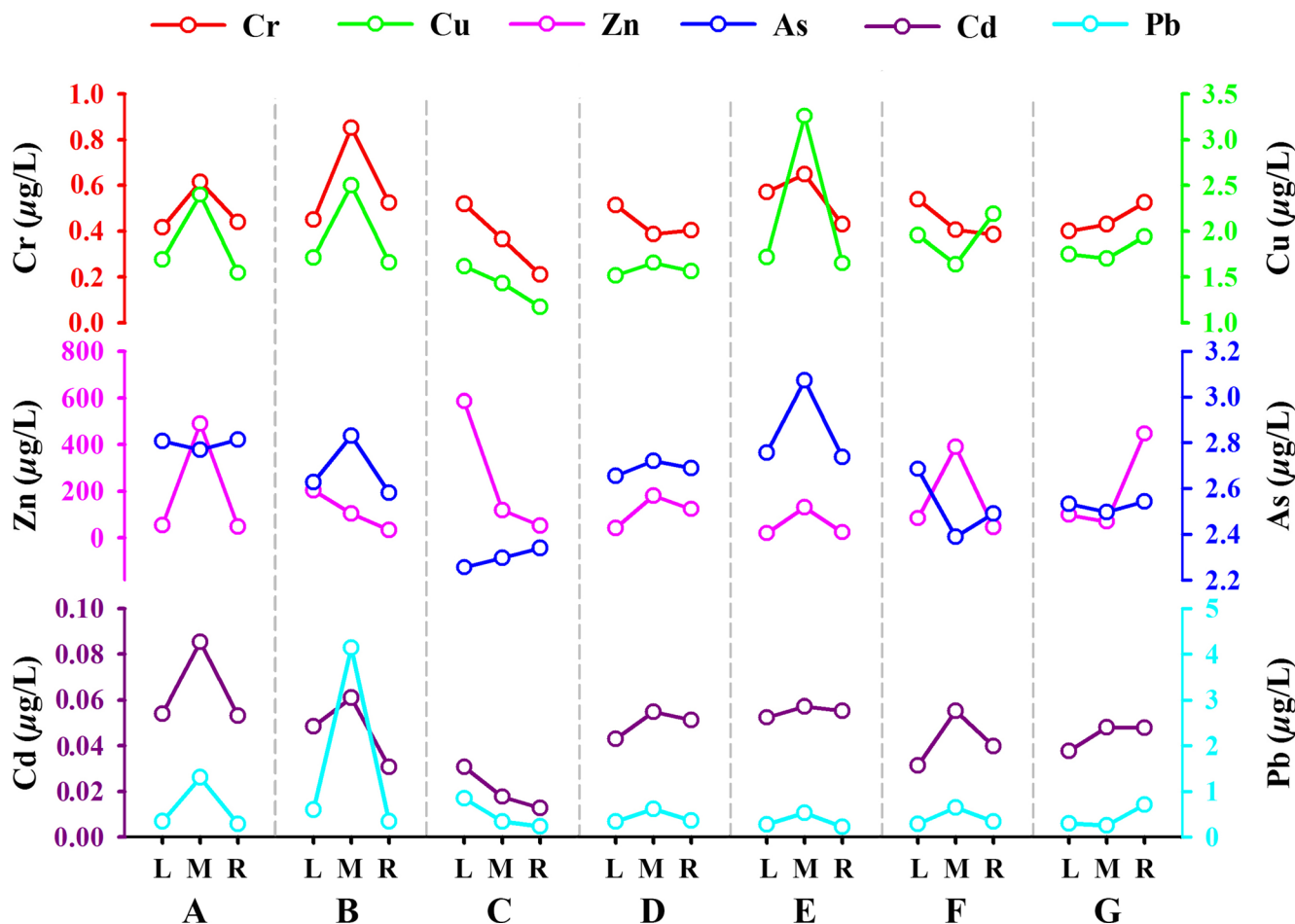


Fig. 4 Spatial distribution of the heavy metal concentrations in the left bank, middle, and right bank in the dry season. A–G represent Jiangjin, Wanglongmen, Beibei, Cuntan, Changshou, Fuling, and Nantuo,

respectively; *L*, *M*, and *R* represent the samples collected from the left bank, the middle, and the right bank of the river, respectively

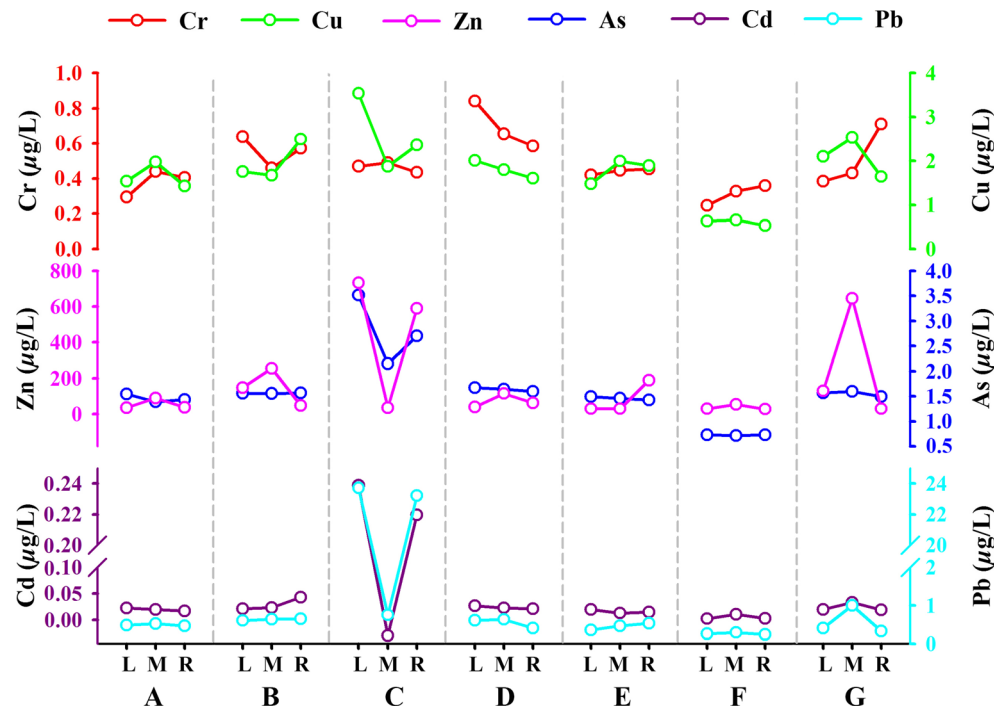
concentrations were not notably different among the left bank, middle of the river, and right bank at the other sites; the variation in the Cu concentration in the left bank, middle of the river, and right bank was consistent with Cr in Jiangjin, Wanglongmen, and Cuntan.

Water samples were collected from a branch of the Jialingjiang River (Tuzhu River located on the left bank; Chengjiang and Maanxi Rivers located on the right bank) in Beibei, and the concentrations of heavy metals were measured and compared with the average concentrations of heavy metals in the water samples collected from the left bank, middle of the river, and right bank of the Jialingjiang River in Beibei (Table 5; Fig. 1c). The Tuzhu River’s inflow to the Jialingjiang has a high concentration of Cr (11.525 µg/L); the concentrations of Cu (3.226 µg/L) and Zn (459.096 µg/L) in the Chengjiang River that flowed into the Jialingjiang from the right bank were higher than that in the right bank of the Jialingjiang River; furthermore, the concentrations of Zn (317.988 µg/L) and As (2.967 µg/L) in the Maanxi River were slightly higher than those in the right bank of the Jialingjiang River. At the same time, strong human

activity was one cause of the high concentrations of heavy metals in the left and right banks. Due to the cultivation of crops on the banks, fertilizer and pesticide residues in the soil may enter the fluvial systems in surface runoff and interstitial water (Li et al. 2008). In addition, industrial wastewater and domestic wastewater are discharged into the Jialingjiang River by industrial enterprises and residents, which also result in water pollution. Because the flow rate of water close to the bank is significantly slower than that in the middle of river, water with high concentrations of heavy metals from the tributaries cannot flow away quickly from the bank, while the water located in the middle of the river can quickly change. As a consequence, the concentrations of heavy metals in the water of left and right banks were significantly higher than that in the middle of the Jialingjiang River in Beibei.

The horizontal distribution characteristics of the heavy metal concentrations in the water were intricate, with significant differences between the dry and rainy seasons at each site. The vertical distribution of heavy metals in the water did not show obvious regular changes, and the scale of the changes was small; therefore, this paper does not discuss these distributions in detail.

Fig. 5 Spatial distribution of the heavy metal concentrations in the left bank, middle, and right bank in the rainy season. A–G represent Jiangjin, Wanglongmen, Beibei, Cuntan, Changshou, Fuling, and Nantuo, respectively; L, M, and R represent the samples collected from the left bank, the middle, and the right bank of the river, respectively



HHRA

HHRA plays an important role in identifying the harm caused by heavy metals to human health and has been widely used in research on drinking water (Ma et al. 2007; Nur 2012; Spickett et al. 2012). The intake by the body is variable in health risk assessments. The risk posed by heavy metals to human health is high, considering the human intake of water that is increasingly polluted by heavy metals (Ma et al. 2007).

HHRA results

To reflect the overall concentration of heavy metals at a site, the average concentration of the heavy metals was calculated from water samples collected from different depths and

Table 5 Comparison of concentrations of dissolved heavy metals in Jialingjiang River and its branches in Beibei (Fig. 1c) (unit in $\mu\text{g/L}$)

Sample	Cr	Cu	Zn	As	Cd	Pb
BB-L	0.451	3.855	515.502	2.989	0.178	12.986
BB-M	0.433	1.955	46.526	2.091	0.014	0.318
BB-R	0.380	1.950	312.618	2.350	0.099	10.326
Tuzhu River	11.525	1.799	75.842	2.702	0.070	0.285
Chengjiang River	0.324	3.226	459.096	1.525	0.023	0.616
Maanxi River	0.282	1.694	317.988	2.967	0.014	0.564

BB-L, BB-M, and BB-R denoted the water samples collected from the left side, the middle, and the right side of Jialingjiang River, respectively

horizontal positions at the same site, and the annual potential risk of heavy metals to adults from drinking water (Table 6) was evaluated according to the HHRA model and evaluation parameters (Table 1). The maximum acceptable risk level and negligible risk level to the public were designed by part of the organizations (Table 7).

The health risk of carcinogenic heavy metals to adults through drinking water from the TGR followed a descending order of $\text{As} > \text{Cr} > \text{Cd}$ and accounted for total risk values of 46.55~64.89, 34.68~53.27, and 0.18~1.64 %, respectively. Thus, As was the major potential pollutant that produced the greatest risk to human health, followed by Cr.

Based on the total risk values at the seven sites, the risk levels of Cr and As were higher than the maximum acceptable risk level ($1 \times 10^{-6} a^{-1}$) given by the Swedish National Environmental Protection Board, the Holland Ministry of Construction and the Environment, and the Royal Society but were lower than the maximum acceptable risk level designated by the US EPA ($1 \times 10^{-4} a^{-1}$) (US EPA 1987) and the International Commission on Radiological Protection (ICRP; $5 \times 10^{-5} a^{-1}$). The risk of Cd was lower than the maximum acceptable risk levels recommended by the above institutions.

The HHRA indicated that the total health risk level of non-carcinogenic heavy metals was 10^{-9} ~ 10^{-10} , which is a negligible risk level and lower than the maximum acceptable risk level designated by the institutions in Table 7. The non-carcinogenic heavy metals rarely have harmful effects on human health. Compared with the carcinogenic heavy metals, the damage caused to adults by non-carcinogenic heavy

Table 6 The adults annual health risk values assessed for TGR water based on the dissolved heavy metals through drinking water (a^{-1})

Month	Site	R_{Cr}	R_{Cd}	R_{As}	R_{Cu}	R_{Zn}	R_{Pb}	R_C	R_n	R
April	Jiangjin	6.68×10^{-6}	1.74×10^{-7}	1.05×10^{-5}	1.69×10^{-10}	2.95×10^{-10}	2.07×10^{-10}	1.73×10^{-5}	6.71×10^{-10}	1.73×10^{-5}
	Wanglongmen	7.75×10^{-6}	1.36×10^{-7}	1.01×10^{-5}	1.74×10^{-10}	1.47×10^{-10}	3.72×10^{-10}	1.80×10^{-5}	6.92×10^{-10}	1.80×10^{-5}
	Beibei	5.01×10^{-6}	6.23×10^{-8}	9.38×10^{-6}	1.20×10^{-10}	2.76×10^{-10}	1.27×10^{-10}	1.45×10^{-5}	5.22×10^{-10}	1.45×10^{-5}
	Cuntan	6.79×10^{-6}	1.38×10^{-7}	1.03×10^{-5}	1.46×10^{-10}	1.26×10^{-10}	2.13×10^{-10}	1.73×10^{-5}	4.86×10^{-10}	1.73×10^{-5}
	Changshou	6.91×10^{-6}	1.46×10^{-7}	1.05×10^{-5}	1.79×10^{-10}	8.20×10^{-11}	9.87×10^{-11}	1.75×10^{-5}	3.59×10^{-10}	1.76×10^{-5}
	Fuling	6.38×10^{-6}	9.08×10^{-8}	9.26×10^{-6}	1.35×10^{-10}	3.05×10^{-10}	1.36×10^{-10}	1.57×10^{-5}	5.77×10^{-10}	1.57×10^{-5}
	Nantuo	6.59×10^{-6}	1.23×10^{-7}	9.99×10^{-6}	1.60×10^{-10}	2.05×10^{-10}	1.06×10^{-10}	1.67×10^{-5}	4.71×10^{-10}	1.67×10^{-5}
August	Jiangjin	5.53×10^{-6}	5.44×10^{-8}	7.10×10^{-6}	1.48×10^{-10}	8.09×10^{-11}	1.58×10^{-10}	1.27×10^{-5}	3.87×10^{-10}	1.27×10^{-5}
	Wanglongmen	6.72×10^{-6}	6.47×10^{-8}	7.29×10^{-6}	1.62×10^{-10}	1.63×10^{-10}	1.67×10^{-10}	1.41×10^{-5}	4.92×10^{-10}	1.41×10^{-5}
	Beibei	5.98×10^{-6}	2.63×10^{-7}	9.84×10^{-6}	2.32×10^{-10}	4.36×10^{-10}	2.53×10^{-9}	1.61×10^{-5}	3.19×10^{-9}	1.61×10^{-5}
	Cuntan	8.08×10^{-6}	6.97×10^{-8}	7.65×10^{-6}	1.60×10^{-10}	9.92×10^{-11}	1.70×10^{-10}	1.56×10^{-5}	4.29×10^{-10}	1.58×10^{-5}
	Changshou	6.27×10^{-6}	4.93×10^{-8}	7.12×10^{-6}	1.50×10^{-10}	1.46×10^{-10}	1.59×10^{-10}	1.34×10^{-5}	4.54×10^{-10}	1.34×10^{-5}
	Fuling	4.66×10^{-6}	1.56×10^{-8}	4.07×10^{-6}	5.59×10^{-11}	5.13×10^{-11}	9.31×10^{-11}	8.74×10^{-6}	2.00×10^{-10}	8.74×10^{-6}
	Nantuo	6.83×10^{-6}	6.47×10^{-8}	7.41×10^{-6}	1.86×10^{-10}	2.92×10^{-10}	1.61×10^{-10}	1.43×10^{-5}	6.38×10^{-10}	1.43×10^{-5}

metals through drinking water is slightly lower. In general, the risk of carcinogenic heavy metals is higher than the risk of non-carcinogenic heavy metals at each sampling site.

The temporal and spatial distributions of the health risk of heavy metals

In April, the order of the total health risk value of the heavy metals to adults through drinking water in the TGR was Wanglongmen > Changshou > Jiangjin = Cuntan > Nantuo > Fuling > Beibei. Therefore, the health risk values of the five sites in the Yangtze River were higher than those at the Fuling and Beibei sites. These results prove that the health risk of the heavy metals in the Yangtze was higher than that in its tributaries, where the contamination was slightly lower.

In August, the order of the total health risk value of the heavy metals to adults through drinking water in the TGR was Beibei > Cuntan > Wanglongmen > Changshou > Jiangjin > Fuling > Nantuo. Compared with that in April, the relative order of the seven sites had greatly changed, and Beibei posed the site with the greatest risk in the TGR. At the same time, the total health risk of most sites, except Beibei, was lower than that in April. The authors hypothesize that the industrial wastewater and agricultural chemicals discharged by humans were the most important sources of the heavy

metal pollution in Beibei. Because of the machine and equipment manufacturing factories, chemical industries, and agricultural land along the Jialingjiang River, industrial and agricultural wastewater would be discharged directly into the Jialingjiang River. In the rainy season (August), the heavy metals in the atmosphere and ground would be washed by rainwater and carried into the Jialingjiang River, resulting to the higher total health risk than that in the dry season.

In addition, Zhao et al. (2012) and Varol (2013) declared that the lowest concentration of heavy metal occurred in the reservoir water near the dam, and the highest concentration appeared in the middle part of the reservoir (Zhao et al. 2012). This is different to our observation. Although in April, the lowest concentrations appeared in the water of Nantuo, they are not consistently declined from the upstream to the lower stream (from Jiangjin to Nantuo), and the concentrations changed greatly in August. Our investigation showed there is no significant correlation between the heavy metal concentration and the pH and EC of the water. While the discharge and quality of tributaries influences on the water of mainstream and reservoir significantly. As the biggest developing country and in the process of rapid industrialization and urbanization, environmental protection has become one of the key issues for the sustainable development of China. Chongqing is one of the most rapid development areas in China, with density population, and accounts for 80 % of the discharge of the industrial waste water and 95 % of urban domestic sewage discharge to the TGR. Both the strict management on the tributaries and the discharge of urban waste water is essential to the protection of the water quality for the TGR.

Finally, the heavy metals accumulated in aquatic organisms and sediments may be ingested into the body or released into the reservoir again under appropriate conditions to cause potential contamination and hazards (Wei et al. 2016), even though the water quality of the reservoir is not significantly polluted by heavy metals at present. Furthermore, other

Table 7 The maximum acceptable risk level and negligible risk level recommended by some organizations (a^{-1}) (cite from Sun et al. 2009)

Nation or organization	Maximum acceptable risk level	Negligible risk level
Sweden	1×10^{-6}	–
Holland	1×10^{-6}	1×10^{-8}
England	1×10^{-6}	1×10^{-7}
US EPA	1×10^{-4}	–
ICRP	5×10^{-5}	–

exposure pathways than drinking were not considered, for example, direct ingestion of aquatic organisms, in this HHRA of heavy metals. As a result, the risk value in this study should be less than the actual risk value. The authors advocate that relevant government offices should continue their pollution control efforts and strengthen supervision and management in the TGR and neighboring regions in order to avoid the occurrence of serious pollution incidents. This study also provided fundamental data for further water quality monitoring work in the TGR and contributes to the monitoring and supervision of the dynamic variation in the water quality of the region.

Conclusions

(1) The concentrations of six dissolved heavy metals in the water of the TGR reached the standard of the National Surface Water Environmental Quality Standards (GB3838-2002) III in China, and Zn was the main potential contaminant in the dry and rainy seasons. Furthermore, the concentrations of the six heavy metals did not exceed the Sanitary Standard for Drinking Water in China (GB5749-2006), the Guidelines for Drinking Water Quality (Third Edition) developed by the WHO, or the Drinking Water Quality Standard (2006) of the USA, among other standards.

(2) There were significant positive correlations among Cu, As, and Cd, showing that they may have the same geochemical behavior. The correlation of the heavy metal concentrations in the rainy season was significantly greater than that in the dry season.

(3) The temporal and spatial variations of the heavy metal concentrations were diverse and complex. The spatial differences in the concentration of Zn were the most significant, whereas those of the other elements were relatively small. The concentrations of heavy metals did not evidently change from upstream to downstream. In the rainy season, there was no significant change in the concentrations of the heavy metals except Zn in the Yangtze River; the concentration of Zn was obviously increased. Due to the influences of many factors, the distribution characteristics of the heavy metal concentrations on both sides and the middle of the river were different at the different sites.

(4) The results of the aquatic environmental HHRA showed that the risk of the carcinogenic heavy metals was higher than the risk of the non-carcinogenic heavy metals at each sampling site. As posed the highest risk to human health in the TGR, followed by Cr. The HHRA results of the six heavy metals to adults via drinking water were lower than the maximum acceptable risk level designated by the US EPA and ICRP.

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