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The assessment on the heavy metal pollution and health risks in the Liujiang River under the Xijiang River region

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ABSTRACT

The research aimed to investigate on the quality of the Liujiang River in Guangxi Province. Concentrations of heavy metals, including Cu, Zn, Pb, Cd, As, Ni, Sb, and Tl, in the Liujiang River water were analyzed for studying the distribution and risks of heavy metal pollutants in this area. The methods of Nemerow comprehensive index and the risk assessment model for health were used to evaluate the water quality and the health risk severity caused by heavy metals in the Liujiang River. The results demonstrated that the water quality of each of the 9 tested points met the concentration requirement for heavy metals in the grade II. The water samples collected on tested points on the Liujiang River were clean and proved not to be polluted by heavy metals. According to the collected data, the concentrations of heavy metals As and Zn were lower than Cd, Pb, and Cu in the Liujiang River, but still lower than the maximum acceptable level of 5.0×10^{-5} a^{-1}. The water quality in comprehensive pollution index and health risk value of heavy metals changed consistently. The quality of the upstream was much better based on the results. As, as the major pollutant of the health risk of Liujiang river, should be managed more strictly for preventing and controlling environmental risk in the Liujiang River.

Keywords: Liujiang River; Heavy metals; Nemerow comprehensive index method; Health risks

1. Introduction

As economy develops, the pollution also increases. Together with the increasing industrialization and the shortage of clean fresh water resources, water pollution becomes worse. Since “The Twelfth Five Year Plan”, heavy metal pollution becomes increasingly severe in China. The heavy metal pollution is mainly caused by the discharge of industrial, agricultural, or domestic sewage, the release of polluted river sediments, and the atmospheric sedimentation. As heavy metals can enrich easily, microbes from the natural environment can hardly realize degradation. Finally, through the food chain, heavy metals can enter the human body and directly or indirectly jeopardize human health [1–4]. Therefore, heavy metal pollution and health risk assessment on water environment are urgently needed.

Since the 20th century, people started research on the water pollution. China’s environmental protection departments mainly adopted average method, single-factor evaluation method, and comprehensive pollution index for

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water quality. The limitations of these methods which use big data are obvious. When the water quality changes violently, the evaluation results are not satisfactory [5–8]. With the development of environmental statistics, fuzzy mathematics method, Bayesian method, multivariate statistical method, gray evaluation method, and neural network method are also used for evaluating water quality [9–16]. Many documents recorded a variety of mathematical methods for assessing water quality. Ru uses the single-factor pollution evaluation and the inner merlot pollution index to evaluate the water quality in the Tibetan Yamdrok Tso river valley [17]. Lu uses the fuzzy matron model. Shen used the gray correlation analysis method to evaluate the water quality of the Danjiangkou River basin [18,19]. Pang used analytic hierarchy process to comprehensively evaluate the water quality of the middle section in the South-to-North Water Diversion Group [20]. Back-Propagation artificial neural network is used by Yang to forecast and provide early warning for the eutrophication of main living supply and drinking water sources in Zhengzhou [21]. Yang studied the Wenyu River in Beijing [22].

The results show that the main quality index method on polluted water is more suitable for assessing water quality of the rivers with poor water quality because it can quantify the differences of water quality between different rivers. Hu assessed the water quality of the underground river in a Guilin village [23]. Li conducted a research on seven evaluation methods, including single-factor assessment, Nemerow index, average pollution index, fuzzy comprehensive evaluation, gray correlation analysis, matter element extension evaluation, and comprehensive water quality index [24]. The evaluation correlation coefficient of these methods is about 0.7, which is relevant with each other and complementary. Given the requirements for environmental management and the advantages and disadvantages of each evaluation method, single-factor evaluation method and comprehensive water quality index method are used together for water environment quality evaluation. Wang showed that the method of Nemerow index is suitable for controlling the water pollution, and it is applicable to the study on changing groundwater quality [25]. Jia used secondary enrichment coefficient method to evaluate heavy metal pollution of sediments [26]. The results from Jia's research show that due to the unstable discharge of water in the reservoir, the sediment in the reservoir is continuously transported to the front part of the dam. The sediment adsorbs more heavy metals, so the accumulation coefficient of heavy metal in the front of the dam increases [27].

Guangxi is rich in heavy metals, such as lead and zinc, so Cd and As are two major types of heavy metal pollution in Guangxi. The Cd pollution accident occurred in the Longjiang River, a secondary tributary in the upper reaches of the Liujiang River in 2012. This is one of the most serious pollution incidents for heavy metals since the founding of the People's Republic of China. It poses a serious threat to the drinking water safety for the citizens of the Liuzhou City. Liuzhou is not only the largest industrial city in Guangxi but also the place of "three kinds of wastes" in Guangxi Zhuang Autonomous Region. The Liujiang River, as a branch of the Xijiang River, is 245 km long outside the Liuzhou. It is the second longest tributary of Longjiang which flows to the lower reaches with the Luoqing River flowing to the upper reaches of Liujiang River. Liujiang River supplies about 92% of water for the production and domestic needs. Also, it is a main outlet way for the direct or indirect discharge. In this research, in order to fully understand the status quo in the Liujiang River water environment, prevent and control heavy metal pollution, restore the ecosystem, and provide reference for environmental risk management on water, the concentration of heavy metals in the Liujiang River was measured and analyzed. Also, heavy metals, including Cu, Zn, Pb, Cd, As, Ni, Sb, and Tl, in the surface water of Liujiang River were monitored and analyzed. Then, Nemerow index method and health risk assessment were used to evaluate the Liujiang River water quality and health risk caused by heavy metals in water, the concentration of heavy metals, distributions, contamination severity, potential hazards, and risk levels of heavy medals in the Liujiang River.

2. Materials and methods

2.1. The selection of the tested site

According to the population distribution, the distribution of the dry waterway branches, the position of pollutant discharge outlet, and other factors, 9 tested sites were selected for survey along the Liujiang River in July 2016 (Fig. 1). The sites are as follows: Ximen Cliff (S1), Lutang (S2), Luowei (S3), Bainiao Beach (S4), Maoer Mountain (S5), Shabao Beach (S6), Shilong Terminal 2 (S7), Shilong Terminal 1 (S8), and Chedu Marina Stadium (S9).

2.2. Sample collection and analysis

2.2.1. Collection of water samples

Samples were taken according to *the Technical Specifications Requirements for Monitoring of Surface Water and Waste Water* (HJ/T 91-2002). The water samples were collected and kept still for 30 min. Then, they were filtered by 0.45 μm filter membrane. Finally, the filtered samples were placed in the bottle. Concentrated nitric acid was added to the site until the pH value was less than 2. The blank test samples in the whole program were collected, and the samples accounted for not less than 10% of the total number of samples. All these water samples were sealed in accordance with the requirement of *Monitoring and Analysis Methods of Water and Sewage* (4th Ed.). Then, these samples were properly preserved in refrigerator at 4°C after being brought back to the laboratory.

2.2.2. Sample analysis

Samples were analyzed according to the relevant standards. For analysis, inductively coupled plasma mass spectrometry (Agilent7700E ICP-MS) was applied. In order to ensure the stability and accuracy of the instrument, the standard samples were used in each determination process. The measurement value on samples was within the 95% confidence interval of the true value, the recovery rate of the sample was 95%–105%, and the relative deviation of the parallel sample was kept within 10%. In addition, not any program blank test results were detected. It can be said that all the results met the requirements for controlling quality.

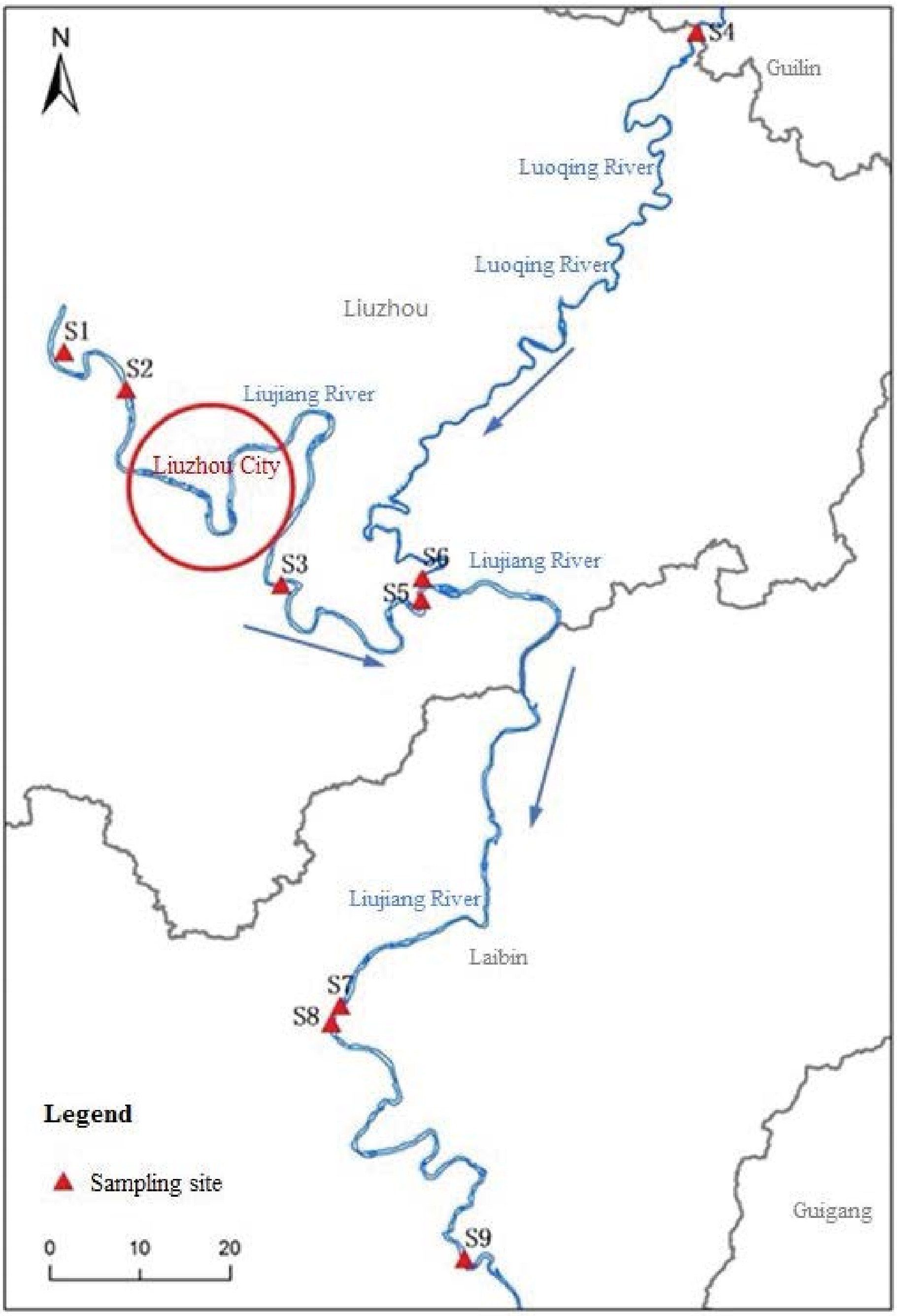

Fig. 1. Tested sites for the Liujiang River.

2.3. Evaluation model on water environment quality

2.3.1. Single-factor assessment

Single-factor evaluation can help determine main heavy metal pollutants and the harm degree. The single-factor evaluation can be generally expressed as pollution index, that is, the ratio of the measured value on heavy metal type to the corresponding evaluation standard value.

$$P_i = \frac{C_i}{S_i} \tag{1}$$

Here P_i refers to the pollution index of the heavy metal element *i*; C_i is the actual concentration of heavy metal *i*; and S_i means Grade-III standard value of heavy metal *i* based on the *Environment Quality Standards for Surface.*

2.3.2. Nemerow index evaluation method

The Nemerow index evaluation method is featured by the simple and clear mathematical process [28,29]. It is a comprehensive method for evaluating various parameters in the water bodies. The Nemerow index combines single-factor evaluation method, the extreme value, and the maximum and minimum pollution degrees. The calculation formula is as follows:

$$P_c = \sqrt{\frac{(P_i)^2 + (P_{i\max})^2}{2}} \quad (2)$$

$$\bar{p}_i = \frac{1}{n}\sum_{i=1}^{n} P_i \quad (3)$$

$$P_i = \frac{C_i}{S_i} \quad (4)$$

In Eq. (2), P_c is comprehensive pollution index; $P_{i\max}$, the maximum value for the contamination index of heavy metals; and $\bar{P}_i$, the mean value of all pollution index from heavy metals. In Eq. (3), P_i is the pollution index of the heavy metal element *i*. The meaning of each letter in Eq. (4) is identical to Eq. (1) above. Table 1 is the Nemerow index level.

2.4. Water environment health risk assessment model

At present, the health risk assessment model recommended by the U.S. Environmental Protection Agency is used for evaluating the water environment at home and abroad [30]. Health risk assessment on water environment is mainly aimed at detecting harmful substances on human bodies in the water environment and contained chemical carcinogenic and noncarcinogenic pollutants [31–37].

Health harm model for the chemical carcinogenic pollutants through the drinking water is as below:

$$R_c = \frac{(-D_i \times q_i)}{\text{ED}} \quad (5)$$

If the result is $R_c > 0.01$, the calculation for the concentration is as follows:

$$R_c = \frac{[1 - \exp(-D_i \times q_i)]}{\text{ED}} \quad (6)$$

R_c is per capita annual risk of chemical carcinogen *c* obtained from the drinking water, a^{-1}; D_m is daily average exposure dose per body weight of chemical carcinogen *c* from the drinking water, mg kg^{-1} d^{-1}; ED is per capita lifetime (per capita lifetime of Liuzhou is 76), a.

The formula for the health harm model on noncarcinogenic pollutants obtained from drinking water is as below:

$$R_n = \frac{\left(\frac{D_m}{\text{RFD}_n}\right)}{\text{ED}} \times 10^{-6} \quad (7)$$

In formula 6, R_n is per capita annual carcinogenic risk caused by the noncarcinogen *n*, a^{-1}; RFD_n is daily average exposure dose per body weight of noncarcinogen *n* obtained from the drinking water, mg (kg^{-1} d^{-1}). Daily average exposure dose D_m per body weight from the drinking water is calculated as below:

$$D_m = \frac{Q \times C_m}{W} \quad (8)$$

Q is daily average drinking amount of adult (suggested value of 2.2), L; C_m is actual concentration of chemical carcinogen or noncarcinogen, mg L^{-1}; *W* is per capita body weight (suggested value of 70), kg.
According to classification system compiled by the IARC and WHO, As and Cd are chemical carcinogens, with the carcinogenic intensity coefficients of 15 and 6.1; Zn, Pb, and Cu, as chemical noncarcinogens, have carcinogenic intensity coefficients of 3×10^{-1}, 1.4×10^{-3}, and 5×10^{-3}, respectively.

3. Results and analysis

3.1. Distribution of heavy metal content at different test points

Table 2 is the concentrations of heavy metals in the surface water of the Liujiang River. The surface water of the Liujiang River sections was tested. Based on the second and third grades for standards on the surface water quality (GB3838 – 2002), the heavy metal concentration conforms to the quality standards for the second grade of the surface water [38–40]. In general, heavy metals in the Liujiang River do not exceed the required amount.

From the results of the correlation analysis of heavy metals in Table 3, it can be seen that the concentrations for different heavy metals are correlated in the Liujiang River. As, Sb, and Tl had significant correlation with each other, and Pb and Cd were significantly correlated [41–43]. It is inferred that As, Sb, Tl, Pb, and Cd came from similar pollution sources.

From Tables 4 and 5, it can be seen that 3 major components account for 85.008% of sources for 8 heavy metals in the surface water of the Liujiang River. When the rate of component 1 is 42.49%, the positive load of As, Sb, and Tl increases. When the contribution rate of component 2 is 23.041%, the positive load of Pb, Cd, and Ni goes up. When the contribution rate of the component 3 is 19.477%, the positive load of Cu becomes higher and the negative load of Zn is higher. Three-dimensional factor load diagram of various heavy metal pollutants are shown in Fig. 2.

Table 1
Nemerow index level

	Water quality classification and grades				
	Clean	Slightly polluted	Polluted	Moderately polluted	Seriously polluted
Nemerow index level	$P_c < 0.74$	$0.74 \le P_c < 0.92$	$0.92 \le P_c < 1$	$1 \le P_c < 1.73$	$P_c \ge 1.73$

Table 2
The concentrations of heavy metals in the surface water of the Liujiang River (μg L^{-1})

Serial number	Name of section	Cu	Zn	Pb	Cd	As	Ni	Sb	Tl
S1	Ximen Cliff	0.86	1.93	0.70	0.06	2.62	0.20	4.53	0.02
S2	Lutang	0.46	ND	0.54	0.03	2.47	0.20	6.86	0.03
S3	Luowei	0.77	8.10	0.59	0.03	2.83	0.63	8.11	0.03
S4	Bainiao Beach	1.12	ND	0.42	0.02	1.02	0.49	1.06	0.01
S5	Maoer Mountain	1.01	2.31	1.27	0.15	3.62	0.65	4.78	0.03
S6	Shabao Beach	1.75	ND	0.47	0.03	1.61	0.51	1.08	0.01
S7	Shilong Terminal 2	0.78	11.81	0.26	0.09	2.15	0.67	2.91	ND
S8	Shilong Terminal 1	0.47	7.96	0.93	0.08	1.69	0.73	3.08	ND
S9	Chedu Marina Stadium	0.26	31.20	0.21	ND	1.80	0.33	1.42	ND
Minimum		0.26	0.00	0.21	0.00	1.02	0.20	1.06	0.00
Maximum		1.75	31.20	1.27	0.15	3.62	0.73	8.11	0.03
Average		0.83	7.03	0.60	0.06	2.20	0.49	3.76	0.02
Standard deviation		0.44	10.04	0.33	0.05	0.78	0.20	2.53	0.01
Coefficient of variation		0.53	1.43	0.56	0.85	0.35	0.41	0.67	0.91
GB3838-2002 I grade		10	50	10	1	50	—	—	—
GB3838-2002 II grade		1,000	1,000	10	5	50	—	—	—
GB3838-2002 III grade		1,000	1,000	50	5	50	—	—	—
Standard limits of specific items for centralized drinking water sources		—	—	—	—	—	20	5	0.1
Method detection limit		0.09	0.8	0.07	0.06	0.09	0.07	0.07	0.01

Note: ND means "have not been detected".

Table 3
The correlation analysis on heavy metals in water in the surface water of the Liujiang River

Heavy metal	Cu	Zn	Pb	Cd	As	Ni	Sb	Tl
Cu	1	–0.598	0.081	–0.011	–0.124	0.186	–0.316	0.003
Zn		1	–0.452	–0.105	–0.139	–0.007	–0.25	–0.457
Pb			1	0.717*	0.611	0.318	0.347	0.496
Cd				1	0.626	0.481	0.102	0.16
As					1	0.029	0.727*	0.782*
Ni						1	–0.084	–0.216
Sb							1	0.851**
Tl								1

* When the confidence (double measure) is 0.05, the correlation is significant.
** When the confidence (double measure) is 0.1, the correlation is significant.

Table 4
Total variance of principal component analysis on heavy metals in surface water

Components	Initial eigenvalue			Extract square sum and load			Rotation square and load		
	Eigenvalue	Analytic variance	Cumulative variances	Eigenvalue	Analytic variance	Cumulative variances	Eigenvalue	Analytic variance	Cumulative variances
1	3.399	42.49	42.49	3.399	42.49	42.49	3.011	37.643	37.643
2	1.843	23.041	65.531	1.843	23.041	65.531	2.078	25.976	63.619
3	1.558	19.477	85.008	1.558	19.477	85.008	1.711	21.389	85.008
4	0.571	7.136	92.144						
5	0.411	5.134	97.278						
6	0.173	2.163	99.44						
7	0.045	0.559	99.999						
8	5.05E-05	0.001	100						

Table 5
Factor loading matrix for principal component analysis on heavy metals in surface water

Heavy metals	Factor load matrix			Post rotation factor load matrix		
	PC1	PC2	PC3	PC1	PC2	PC3
Cu	0.008	0.645	–0.666	–0.237	0.079	0.893
Zn	–0.475	–0.341	0.736	–0.324	–0.043	–0.881
Pb	0.808	0.371	0.117	0.509	0.691	0.261
Cd	0.615	0.49	0.509	0.242	0.904	–0.036
As	0.904	–0.173	0.201	0.847	0.405	–0.079
Ni	0.14	0.705	0.408	–0.264	0.780	0.067
Sb	0.762	–0.521	–0.046	0.919	–0.039	–0.088
Tl	0.851	–0.369	–0.302	0.949	–0.036	0.221

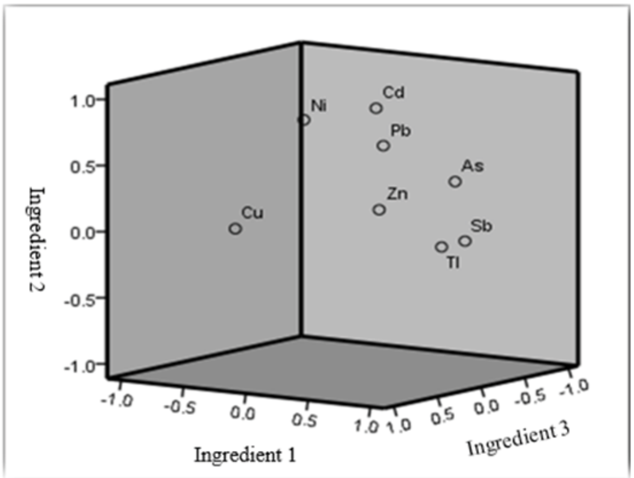

Fig. 2. Three-dimensional factor load diagram of various heavy metal pollutants.

3.2. Evaluation on heavy metal pollution at different sampling points

The standard values are set up according to Grade-III for the water quality standard of the *Environment Quality Standards for Surface Water* (GB 3838 - 2002) and the limits for specific items for centralized drinking water sources. The measured values for heavy metals in each tested point are calculated by the single pollution index and the Nemerow comprehensive index methods. The results are shown in Table 6. The Table 6 shows that the pollution index P_i for a single heavy metal element is more than 0.74 except for Sb in S2 and S3, and all of the other elements in the rest of points are less than 0.74. In addition, according to comprehensive pollution index P_c, S2 and S3 have a value greater than 0.74. If the water quality ranges from 1 to 1.73, it can be regarded as moderately polluted place. The comprehensive pollution index of less than 0.74 is regarded as noncontamination and clean water.

3.3. Health risk assessment on heavy metals at different tested points

Based on health risk assessment model, cancer intensity coefficient, and determination of concentrations of heavy metals in the Liujiang River, the health risk caused by the drinking water ways at each tested point in the Liujiang River can be calculated. The calculation results are shown in Table 7. Three-dimensional factor load diagram of various heavy metal pollutants are shown in Fig. 1.

It can be seen from Table 7 that the health risk of heavy metals is as follows As > Zn > Cd > Cu > Pb. The concentrations of carcinogens As and Cd through drinking water ranges from

Table 6
Heavy metal pollution index and the level of water quality in the Liujiang River

Monitoring point position	P_i								P_c	Pollution degree
	Cu	Zn	Pb	Cd	As	Ni	Sb	Tl		
S1	0.00086	0.0019	0.0140	0.0120	0.0524	0.0100	0.9060	0.21	0.65	Clean
S2	0.00046	0.0004	0.0108	0.006	0.0494	0.0100	1.3720	0.34	0.98	Slight pollution
S3	0.00077	0.0081	0.0118	0.006	0.0566	0.0315	1.6220	0.29	1.16	Moderate pollution
S4	0.00112	0.0004	0.0084	0.004	0.0204	0.0245	0.2120	0.09	0.15	Clean
S5	0.00101	0.0023	0.0254	0.0300	0.0724	0.0325	0.9560	0.32	0.69	Clean
S6	0.00175	0.0004	0.0094	0.0060	0.0322	0.0255	0.2160	0.14	0.16	Clean
S7	0.00078	0.0118	0.0051	0.0189	0.04291	0.0333	0.58111	0.05	0.42	Clean
S8	0.00047	0.0079	0.0186	0.0161	0.03379	0.0364	0.61553	0.05	0.44	Clean
S9	0.00026	0.0312	0.0043	0.0060	0.0359	0.0164	0.28452	0.05	0.20	Clean

Note: the location below the detection limit is calculated by half of the detection limit.

Table 7
The health risk values of carcinogens and noncarcinogens

Monitoring point	Carcinogens		Noncarcinogens			Total
	Cd	As	Pb	Cu	Zn	
S1	1.64E-07	1.76E-05	4.40E-10	1.93E-09	2.60E-07	1.81E-05
S2	8.22E-08	1.66E-05	3.39E-10	1.03E-09	5.39E-08	1.68E-05
S3	8.22E-08	1.90E-05	3.71E-10	1.73E-09	1.09E-06	2.02E-05
S4	5.48E-08	6.87E-06	2.64E-10	2.51E-09	5.39E-08	6.98E-06
S5	4.11E-07	2.44E-05	7.98E-10	2.27E-09	3.11E-07	2.51E-05
S6	8.22E-08	1.08E-05	2.95E-10	3.93E-09	5.39E-08	1.10E-05
S7	2.60E-07	1.44E-05	1.62E-10	1.75E-09	1.59E-06	1.63E-05
S8	2.21E-07	1.14E-05	5.86E-10	1.06E-09	1.07E-06	1.27E-05
S9	8.22E-08	1.21E-05	1.34E-10	5.72E-10	4.20E-06	1.64E-05
Average value	1.60E-07	1.48E-05	3.77E-10	1.87E-09	9.65E-07	1.59E-05

Note: The location below the detection limit is calculated by half of the detection limit.

10^{-5} to 10^{-8}, of which As is the largest, followed by Cd. All the health risk values in all the tested points were lower than the maximum acceptable risk level for radiation protection recommended by the International Commission (5×10^{-5} a^{-1}), but the health risk values caused by As at S1, S2, S3, S5, S6, S7, S8, and S9 exceeded the maximum recommended acceptable level, 1×10^{-5} a^{-1}, by the Netherlands Ministry of Construction and Environment, Swedish Environmental Protection Agency. It can be seen that As is a major pollutant in the Liujiang River environment and should be prioritized during the environmental risk management for the Liujiang River.

According to Table 7, the risk level for noncarcinogens Pb, Cu, and Zn is Zn > Cu > Pb; 10^{-9} to 10^{-10} a^{-1} of Cu and Pb will pose risks to human health. The health risk values of Cu, Pb, and Zn do not exceed the maximum acceptable values 1.0×10^{-5} a^{-1}, which is recommended by the Ministry of Construction and Environment of the Netherlands and the Swedish Environmental Protection Agency. The health risks caused by Cu, Pb, and Zn are not significant and will not pose significant hazard to human health.

4. Conclusions

- According to samples, the concentrations of the heavy metals Cu, Zn, Pb, Cd, As, Ni, Sb, and Tl in the surface water of each monitoring section in the Liujiang River Basin meet the requirements of class II in the Water Quality Standards for Surface Water Environmental Quality. The quantity of heavy metals in the Liujiang River meet the requirements.
- The risk level for noncarcinogens Pb, Cu, and Zn is Zn > Cu > Pb. The concentrations of carcinogens As and Cd through drinking water range from 10^{-5} to 10^{-8}, of which As is the largest, followed by Cd. All the health risk values in all the tested points were lower than the maximum acceptable risk level for radiation protection recommended by the International Commission (5×10^{-5} a^{-1}), but the health risk values caused by As at S1, S2, S3, S5, S6, S7, S8, and S9 exceeded the maximum recommended acceptable level, 1×10^{-5} a^{-1}, by the Netherlands Ministry of Construction and Environment, Swedish Environmental Protection Agency. It can be seen that As is a major pollutant in the Liujiang River environment and should be prioritized during the environmental risk management for the Liujiang River.
- The comprehensive pollution index of heavy metals in the Liujiang River is consistent with the water quality data reflected through the health risk value, both of which show that the results of the upstream are slightly better. Although the water quality of the Liujiang River is good, the effluent discharge of upstream enterprises at points S2 and S3 should be supervised.

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