Original Research

Analysis of Heavy Metal Sources and Health Risk Assessment of Typical Coal Mine Collapsed Lakes in Huaibei Coalfield, Anhui Province, China

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Abstract

Heavy metals in the collapsed lake of Zhuxianzhuang Mine in the typical coal mine of Huaibei Coalfield were selected as the research object. Differences, sources and human health risks of heavy metals such as Cu, Cd, Cr, Ni, Pb, V, Zn and Mn in four seasons of lake water were analyzed. Among them, the content of V element was the highest in all four seasons (1.80~7.04mg/l), while the content of Cd was the lowest (0.03~1.92mg/l). In addition, the elements of the super background value are: V, Ni, Cu, and Cr, wherein the V content exceeds the background value multiple of 4.3 to 12.93. Principal component analysis (PCA) results show that the three principal components share 81.6% of the variance contribution rate. Finally, the results of lake water health risk assessment showed that the risk of non-carcinogenicity of 8 heavy metals was: Cu>Cd>Cr>Ni>Pb>V>Zn>Mn. The Cr element is the main carcinogenic risk element in the lake water of the study area, and the risk value is concentrated at 1.16E-08~7.54E-07. Among the exposed population, the hazard index (HI) of Mn was 49.21 times that of adults. In general, the hazard index value is <1, and the cancer risk is within acceptable levels (1E-06). But from the perspective of public health and safety, we need to maintain further observation and research on these elements.

Keywords: coal mine collapsed lake, heavy metal element, health risk assessment, Huaibei Coalfield

Introduction

Coal mining worldwide is deteriorating the local mining environment in different ways [1-2]. Ground

subsidence, air pollution, vegetation pollution and water pollution have become important environmental threats caused by coal mining activities [3-4]. Among them, environmental damage caused by ground subsidence is one of the problems to be solved [5-6]. As of 2011, the ground subsidence area caused by coal mining activities in China reached 8000 km² [7]. Large-scale ground subsidence forms a coal mine collapsed lake under the

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recharge of atmospheric precipitation, surface water and shallow groundwater [8]. Therefore, the collapsed lake is a special surface water body formed by the nonartificial excavation unique to the mining area, and the current status of the collapsed lake water environment quality has received extensive attention [9]. With the development of ecotoxicological methods such as bioassays, the pollution status of heavy metal elements with toxic accumulation and long-lasting properties on natural waters is severe [10-12]. In the current research situation, the main research contents of coal mining subsidence lakes focus on water quality assessment, water body repair, and water management. For example, Chen (2019) and other selected heavy metal elements in six collapsed waters of Huainan coalfield for health risk assessment [13]; Xie et al. (2013) analyzed the water quality of the collapsed waters in Huaibei, and proposed a variety of ecological restoration methods. However, such research is lacking in the Huaibei coalfield where the collapsed lake area is relatively wide (about 300 km²) [14].

In Huaibei coalfield, the Zhuxianzhuang coal mine has a long history of mining. Lake water body formed 25 to 35 years ago, the water depth is 3~7 m, and the environmental geochemical characteristics of heavy metal elements are typical. To this end, the two collapsed lakes of Zhuxianzhuang Mine in Huaibei Coalfield were selected as the research object. The research objectives include: (1) demonstrate the distribution characteristics of heavy metal elements in the two water bodies in the four seasons, (2) explain the source of heavy metal elements in water, (3) calculate the health risk value of heavy metal elements for adults and children, and (4) provide reference for the utilization of the water body in the collapsed lake of Huaibei coalfield.

Methods and Materials

Study Area and Sampling

Zhuxianzhuang Mine is situated in Suzhou City, northern Anhui Province, and was put into operation in April 1983. Mine field range: 117°05'37.6"E~117°09'23"E, 33°33'31.4"N~33°39'37.6"N. The area of the minefield is 26.3 km², and the current annual coal production capacity is 2.45 million tons. After more than 30 years of mining, the collapsed area is about 11 km² [15]. In this study, the collapsed lakes are formed on the surface of the second and fourth mining areas, respectively. In order to facilitate the analysis, according to the collapsed time, two lakes are named as collapsed lake-I (CL-I) and the collapsed lake-II (CL-II). There are many fish culture zones and phytoplankton growth in the collapsed lake. In addition, many pieces of coal gangue and fly ash are accumulated along the lakeshore. The study area and sampling location are shown in Fig. 1.

Lake water sampling was implemented according to Standard of Surface Water and Sewage Detection Technology HJ/T91-2002 [16]. The sampling was divided into four stages: summer (July 2017), autumn (October 2017), winter (January 2018), and spring (May 2018), and the sampling period was 1 year. Before sampling, the raw water at 0.5 m below the water surface was cleaned and the polyethylene



Fig. 1. Map of the study area and sampling locations.

sampling bottle (2.5L) was loaded three times before sampling. After retrieval, it was filtered by 0.45m microporous membrane and adjusted by high-grade pure concentrated nitric acid for pH reserve. A total of 28 samples were taken from the two lakes, and the sampling points were positioned using handheld GPS. After collecting freshwater samples, we added nitric acid of guaranteed reagent (65-68%) reagent to digest a small amount of organic matter. Then we added nitric acid (2%) to maintain an acidic environment for testing an inductively coupled plasma mass spectrometer (ICP-MS) (Agilent 7500cx) selected to test the elements of V, Cr, Mn, Ni, Cu, Zn, Cd and Pb. In order to ensure the authenticity of the experimental data, the average values of the three experimental data were taken as the experimental results, and the error of the experimental analysis was controlled to within 5%. The test data of the standard sample was very consistent with the authentication value, which shows the accuracy of the test.

Research Method

Principal Component Analysis

PCA is a method for characterizing the information carried by all variables with a small number of indicators according to actual needs and used for heavy metal source analysis in this study [17]. Before the analysis, in order to compress the data scale and make the data more stable, all variables in the original data were subjected to logarithmic transformation that can be calculated according to Equation (1). Software analysis was done using statistical product and service solutions (SPSS).

$$y_{ij} = logc_{ij} - \frac{1}{p} \sum_{i=l}^{p} logc_{ij} (i = 1, 2..., m, j = 1, 2..., p)$$
(1)

...where c_{ij} was the raw measurement of the *j*th indicator of the *i*th water sample(mg/L), and y_{ij} was the x_{ii} after the logarithmization process.

Human Health Risk Assessment

Heavy metals affected human health by causing cancer and non-cancer diseases in human organs [18-19]. Routes entering the human body were dermal absorption, direct intake and inhalation [20-21]. Combined with the actual situation of this study, there were multiple fish culture areas in the waters and they were not used as local drinking water sources. Therefore, the risk of human health caused by heavy metal elements passing through the food chain was the most common exposure route [22]. Based on the risk criteria recommended by the United States Environmental Protection Agency (USEPA) 2004 [23], the daily exposure dose (ADD_{ingestion}) of food ingested in water was calculated according to formula (2):

$$ADD_{ingestion} = \frac{C_i \times I_R \times BCF \times EF \times ED}{B_W \times AT}$$
(2)

...where C_i denoted the test value of heavy metal elements in each water sample ($\mu g/L$) and I_{R} denoted the daily average fish consumption of the human body. The average daily fish consumption of an adult was 0.0065 kg [24]. The average daily fish consumption of children was 0.0019kg [25]; BCF denoted fish bioaccumulation coefficient (mg/kg/d), where 2.7 for V, 16 for Cr, 47 for Ni, 200 for Cu, 47for Zn, 81for Cd, 49 for Pb [26], and 0.34 for Mn [27]; EF was the exposure frequency (d/a) and the value was 350; ED was the exposure period (a), adult was 30 [28], and child was 6 [29]; Bw was the average body weight (kg), the average adult weight was 56.8, and the average child weight was 15.9; AT was the average time for carcinogenic or non-carcinogenic (d), the average time for carcinogenesis was 25550 [28], the average time for non-cancer in adults was 10950, and the average time for non-carcinogenesis in children was 2190 [29].

In this study, chemical carcinogens Cd and Cr were selected for carcinogenic health risk assessment models [30-31], and chemical carcinogens also had non-carcinogenic effects [32-33]. Therefore, chemical non-carcinogenic elements (Cr, Cd, Cu, V, Zn, Mn, Ni, Pb) were selected by non-carcinogenic health risk assessment models. The calculation formulas of the two models were as shown in Equations (3), (4), and (5):

$$HQ_{ingestion} = \frac{ADD_{ingestion}}{RfD}$$
(3)

$$HI = \sum HQ_{ingestion} \tag{4}$$

$$CR = ADD_{ingestion} \times SF$$
 (5)

...where $HQ_{ingestion}$ was a hazard quotient and HI is the hazard index. When HI>1, the non-carcinogenic risk value brought by heavy metal elements should be considered [23]; RfD denotes daily average reference exposure dose (µg/kg/d), in this study, 3 for Cr, 0.4 for Cd, 20 for Ni, 40 for Cu, 300 for Zn, 20 for V, 3.6 for Pb, and 520 for Mn [26]; *SF* was a carcinogenic slope factor ((mg/(kg·d))-1), 0.5 for Cr, and 0.38 for Cd.

	Spring			Summer			Autumn			Winter		
	Max.	Min.	Ave.	Max.	Min.	Ave.	Max.	Min.	Ave.	Max.	Min.	Ave.
V	4.93	1.80	3.35	7.04	2.66	4.51	4.78	3.00	3.87	5.25	1.88	3.13
Cr	0.17	0.13	0.14	0.07	0.03	0.05	1.52	1.11	1.33	1.92	1.08	1.40
Mn	2.44	0.22	0.80	1.47	0.23	0.71	0.23	0.12	0.17	1.79	0.36	0.98
Ni	1.47	1.03	1.23	1.66	0.41	0.84	0.58	0.23	0.40	0.85	0.64	0.77
Cu	0.88	0.13	0.45	1.29	0.36	0.79	0.42	0.27	0.34	0.75	0.37	0.54
Zn	0.69	0.16	0.34	1.89	0.46	1.10	1.59	1.04	1.28	1.46	0.92	1.18
Cd	0.02	0.001	0.01	0.06	0.02	0.04	0.05	0.02	0.03	0.05	0.03	0.03
Pb	0.05	0.01	0.02	0.16	0.03	0.08	0.12	0.04	0.07	0.08	0.02	0.05

Table 1. Statistical characteristic analysis of heavy metal contents in collapsed lake (µg/L).

Results and Discussion

Seasonal Content Characteristics of Heavy Metals

Predecessors have accumulated a lot of research results on the abundance and distribution characteristics of heavy metals [34-36]. First, the two collapsed lakes as a whole are described as the seasonal variation characteristics of their heavy metal concentrations (Table 1). The order of the average content of metal elements in lake water is spring: V>Ni> Mn>Cu>Cr>Zn>Pb>Cd, summer: V>Zn>Ni>Cu>Mn> Pb>Cr>Cd, autumn: V>Cr>Zn>Ni>Cu>Mn>Pb>Cd, and winter: V>Cr>Zn>Mn>Ni>Cu>Pb>Cd. It can be seen from the above that the order of the weight metal content of the collapsed lake water was different in each season. It was worth noting that V has the highest content in the four seasons, and Cd has the lowest content in the four seasons, reflecting that the content of these two elements was less affected by seasonal changes. The contents of Ni, Cu and Pb elements

were consistent in the four seasons, all of which are Ni>Cu>Pb. The obvious seasonal difference was the Cr element, which is low in spring and summer, but it increases significantly in autumn and winter and was only lower than V. In addition, the average value of heavy metal content in spring and summer in the study area was compared with the water body in the collapsed area of Panyi Coal Mine in Huainan, 150km away. The order of the metal element mean content is spring: Cu>Pb>Cd>Cr>Ni>Zn [37] and summer: Mn>Zn>Cu>Ni>Cr>Pb>Cd [38]. Comparison showed that the order of heavy metal content in spring and summer in Huainan collapsed lake was significantly different from that in the study area. And high content difference of Cu, Pb, Mn and other elements may be related to the mixing of industrial and domestic wastewater in the surface river (Ni River) of Panyi Coal Mine.

Secondly, we compared the seasonal differences in heavy metal content between the two collapsed lakes. In Fig. 2, the left side of the red dotted line is collapsed lake-I (*CL-I*), and the right side is collapsed lake-II



Fig. 2. Seasonal comparison of the average heavy metal contents in two collapsed lakes.

	V	Cr	Mn	Ni	Cu	Zn	Cd	Pb
V	1							
Cr	-0.463	1						
Mn	0.171	-0.51	1					
Ni	0.4	-0.486	0.653	1				
Cu	0.155	-0.693	0.224	0.46	1			
Zn	-0.391	0.183	-0.589	-0.697	-0.222	1		
Cd	-0.433	-0.049	-0.375	-0.372	0.25	0.253	1	
Pb	0.244	-0.096	-0.348	-0.546	-0.28	0.456	-0.169	1

Table 2. Correlation matrix of eight heavy metals.

Correlation of variables in bold is greater than 0.6

(CL-II). For heavy metals (V, Cr, Mn, Cu, Zn, Pb), content comparison is: CL-I>CL-II. For heavy metals (Ni, Cd), content comparison is: CL-II>CL-I. On the whole, the heavy metal content of the CL-I was higher than that of the CL-II, which may be related to the surrounding area of the collapsed lake-I, which is affected by domestic sewage or garbage. In terms of the trend of heavy metal seasons, the two collapsed lakes have higher consistency (Fig. 2). For example, for V and Pb the trend of change is increased in autumn and winter in spring and summer; on the contrary, the trend of Cr changes in spring and summer to decrease in autumn and winter, while the trend of Cd content in seasons increases gradually, because the metal content of the two collapsed lakes was much lower than the drinking water quality guidance issued by the World Health Organization (WHO) in 2011 and the groundwater national standard announced by the National Quality Supervision Bureau in 2017 [39-40]. The Yangtze River was selected as the area providing background values in the study [41]. Fig. 2, the elements of the above background value are V and Ni, and partly Cd, Cr, Cu and Pb. Among them, the background value of V in the four collapsed lakes is 8.45~12.93 and 4.23~6.54. It is worth noting that the Cr content was lower than the background value in spring and summer, and about 3.7 times higher than the background value in autumn and winter.

Heavy Metal Element Source Analysis

PCA was often used to understand the correlation and source of various heavy metal elements in water [42]. From the above, regarding the four seasons of heavy metals in the two water bodies, the changes were more consistent, so the heavy metal source analysis was carried out as a whole. Determining the correlation of each heavy metal was a necessary step before the data was subjected to principal component analysis. The correlation of heavy metal elements after "logarithmic" pretreatment is mostly greater than 0.3 (Table 2). Studies had shown that when the correlation of most variables was greater than 0.3, the results of principal component analysis were more reliable [43].

Three principal components (PCs) (Fig. 3) were extracted from eight heavy metal elements, which together account for 81.6% of the variance contribution rate, so they were used to discuss the sources of each heavy metal. The variance contribution rate of principal components 1 (PC1) was 42.3%, and the high load variables were Mn and Ni. The source was mainly soil formation and rock weathering. In addition, the first principal component of the variable was often related to human activities, so coal resource combustion, automobile exhaust and industrial waste emissions were the sources of the siderophile elements Mn and Ni [44]. The variance contribution rate of principal components 2 (PC2) was 20.6%, and the high load variables were Pb and V. Combined with the V element content in



Fig. 3. Principal component analysis (PCA) for dissolved heavy metals in collapsed lakes.

Risk	Spr (N =	ing = 10)		nmer = 6)	Autu (N =		Winter (N = 6)					
	Adult	Child	Adult	Child	Adult	Child	Adult	Child				
	Carcinogenic Risk(CR) ^a											
Cr	5.38E-08	5.62E-08	1.71E-08	1.79E-08	4.99E-07	5.22E-07	5.28E-07	5.51E-07				
Cd	1.88E-08	1.96E-08	5.46E-08	5.70E-08	4.55E-08	4.76E-08	5.03E-08	5.25E-08				
Hazard Quotient(HQ) ^a												
Cr	8.38E-05	8.75E-05	2.67E-05	2.79E-05	7.77E-04	8.11E-04	8.21E-04	5.15E-03				
Cd	2.88E-04	3.01E-04	8.38E-04	8.75E-04	6.99E-04	7.30E-04	7.72E-04	4.84E-03				
Mn	5.73E-08	2.82E-06	5.07E-08	2.49E-06	1.25E-08	6.15E-07	7.00E-08	2.06E-05				
Ni	3.16E-04	3.30E-04	2.16E-04	2.26E-04	1.04E-04	1.08E-04	1.99E-04	1.25E-03				
Cu	1.96E-03	2.04E-03	3.47E-03	3.62E-03	1.49E-03	1.56E-03	2.37E-03	1.48E-02				
Zn	5.76E-06	6.02E-06	1.88E-05	1.97E-05	2.20E-05	2.30E-05	2.03E-05	1.27E-04				
V	4.97E-05	5.19E-05	6.68E-05	6.98E-05	5.73E-05	5.98E-05	4.64E-05	2.91E-04				
Pb	2.65E-05	2.77E-05	1.13E-04	1.18E-04	1.04E-04	1.08E-04	7.04E-05	4.41E-04				
HIª	2.73E-03	4.75E-03	3.26E-03	4.29E-03	2.85E-03	4.96E-03	3.40E-03	2.69E-02				

Table 3. Carcinogenic and non-carcinogenic risk values of heavy metals in the intake pathway.

Note: N-samples; ameans CR, HQ, HI were average value of water samples.

Fig. 2, the source of analysis was industrial and domestic combustion of coals [45]. The variance contribution rate of principal components 3 (PC3) was 18.7%, and the high load variables were Cr and Cu. Studies have shown that there was a good spatial-temporal correlation between Cr and Cu in the soil of the mining area [46]. In combination with the content of Cr and Cu in Fig. 2, the consistent variation characteristics of the two collapsed lakes were consistent. It was speculated that the source of Cr was consistent with the source of Cu, which is affected by natural factors. It is speculated that the seasonal characteristics of Cr content in Fig. 2 may be related to the following: (1) atmospheric precipitation in the study area mainly exists in spring and summer, and atmospheric precipitation dilutes Cr in water and (2) spring and summer are the seasons for the growth of fry and phytoplankton. Cr is adsorbed into the fish or plant by the suspended matter in the water. Less fish eating and phytoplankton death in autumn and winter increases the Cr content in water.

Risk Assessment

Non-Carcinogenic Risk

Hazard quotient (HQ) was introduced to characterize the non-carcinogenic health risks of 8 heavy metals in humans under the intake route. Table 3 shows that children are exposed to health risks greater than adults in all four seasons. It should be noted that in winter lake water, the risk to children with Cr and Cd was higher, the values were: 5.15E-03, 4.84E-03, and children harmed by Mn was 49.21 times that of adults (Table 3). Overall, the non-carcinogenic risks of heavy metals were ranked as follows: Cu>Cd>Cr>Ni>Pb >V>Zn>Mn.

In terms of seasonal changes, the health risks of heavy metals in winter were highest. For example, the risk of Cu in adults and children in winter is 2.37E-03 and 1.48E-02 respectively. Secondly, the summer and winter health risks were greater than spring and autumn. This phenomenon was common in the two lakes, such as Cr, Mn, Ni, and V.



Fig. 4. Non-carcinogenic health risk assessment of two collapsed lakes in the study area.

Fig. 4 shows the seasonal mean hazard index (*HI*) for two lakes. Except for spring, the *HI* value of the *CL-I* in the other three seasons was higher than that of the *CL-II* (Fig. 4), reflecting the higher overall health risk of the *CL-I*, which is consistent with the discussion in Fig. 2. Secondly, the summer health risk was the biggest and the spring is the smallest, which is related to the difference in the content of each heavy metal in the four seasons.

Generally, the *HI* values of the two lakes are less than 1, which means that the non-carcinogenic risk caused by heavy metals is lower [23]. However, due to the persistence and toxicity accumulation of heavy metals, the potential non-carcinogenic health risks in lake water such as Cu, Cd, Cr and Ni increase with time.

Carcinogenic Risk

Cr and Cd were chemical carcinogens in this study. Cr and Cd had different carcinogenic health risks (CR) in adult and children through the intake of fish. In the exposed population, children were at higher carcinogenic risk than adults in four seasons (Fig. 5) [47].

On the time scale, there were different major carcinogenic risk heavy metals in different seasons. The factor for high carcinogenic risk in spring, autumn and winter was Cr, and Cd in summer. Specifically, unlike the non-carcinogenic risk of Cr, the seasonal order of Cr carcinogenic risk was: winter>autumn>spring>summer. The carcinogenic risk value of Cd was affected by high content in summer (Fig. 2), and the order was: summer>winter>autumn>spring. On a spatial scale, carcinogenic risk caused by Cr in *CL-I* in summer, autumn and winter was higher than that in *CL-II*. In four seasons, carcinogenic risk caused by Cd in *CL-II* was higher than that of *CL-II*.

At the level of carcinogenic risk, the carcinogenic risk of Cr was higher than that of Cd, which is 3199

consistent in three seasons (excluded summer) (Fig. 5). For adults and children, the carcinogenic risk range of Cr was 7.22E-07~1.16E-08; 7.54E-07~1.21E-08, and the range of Cd was 9.22E-08~1.31E-09; 9.63E-08~1.36E-09. It can be seen from the above that Cr is the main carcinogenic risk element in lake water, which is consistent with the main carcinogenic elements of shallow groundwater in the study area. Therefore, it is speculated that the lake water has a recharge relationship with shallow groundwater [18]. Finally, the maximum acceptable risk (1E-06) recommended by the Swedish Environmental Protection Agency was selected as the reference standard [48]. The results showed that the carcinogenic risk of all water samples in the two lakes is currently below the warning line (1E-06), but the cumulative carcinogenic risk of Cr needed attention. Therefore, we can take some water treatment measures to change the occurrence of high content of heavy metal elements in water [49-50].

Uncertainty Analysis

A comprehensive health risk assessment should include health risks in three ways: intake, skin contact, and respiratory inhalation. This study evaluated the health risks of eight heavy metal elements in water based on the intake route. Uncertainty factors still exist in this study, such as: (1) water area of the collapsed lake is large, and the health risks of heavy metals passing through the respiratory inhalation route are not considered; (2) heavy metal types are not comprehensive enough, resulting in health risk calculations less than actual results; and (3) due to the lack of background parameters in the study area, the model parameters used in the paper are all from the US Environmental Protection Agency (except for a small number of parameters). In summary, this paper links the use of lake water with the way that heavy metals are most likely to enter the human body (fish intake), which



Fig. 5. Carcinogenic health risk assessment of Cr and Cd in two collapsed lakes in the study area.

can provide a reference for related research by scholars in other regions.

Conclusions

Two collapsed lakes in Zhuxianzhuang Mine of Huaibei Coalfield were taken as examples. Twentyeight lake water samples were taken in spring, summer, autumn and winter, and 8 heavy metals (V, Cr, Ni, Cu, Zn, Mn, Cd, and Pb) in lake water were measured by ICP-MS. The difference of heavy metal content in each season was as follows – spring: V>Ni>Mn>Cu>Cr> Zn>Pb>Cd, summer: V>Zn>Ni>Cu>Mn>Pb>Cr>Cd, autumn: V>Cr>Zn>Ni>Cu>Mn>Pb>Cd, and winter: V>Cr>Zn>Mn>Ni>Cu>Pb>Cd. In addition, taking the Yangtze River water source area as the background values were: V, Ni, Cu, Cr, and the V content exceeds the background value multiple of 4.3~12.93.

Principal component analysis method was applied to the analysis of the source of heavy metal elements in the lake water, and the contribution rate of the three principal components extracted was 81.6%. Coal resource combustion, automobile exhaust and industrial waste emissions were sources of Mn and Ni. Highcontent element V source was mining or fossil energy burning. Because of the good space-time correlation between Cr and Cu, it was speculated that Cr and Cu have the same source, which are affected by natural factors.

Eight heavy metal elements were used as chemical non-carcinogens, and Cr and Cd were chemical carcinogens. The most common exposure pathways for heavy metals into the human body (fish intake route) were determined, and the risk of non-carcinogenic and carcinogenic health of the lake water was calculated. The results show that the HI of the lake is less than 1, namely the non-carcinogenic risk caused by heavy metal elements was still within the safety line. Secondly, the non-carcinogenic risk of adults and children brought about by 8 heavy metals was consistent. The change of hazard quotient was: Cu>Cd>Cr>Ni>Pb>V>Zn>Mn. Among the carcinogenic factors, Cr was the main carcinogenic risk element in lake water, and the risk value varies from 1.16E-08 to 7.54E-07, which has not exceeded the maximum acceptable risk range (1E-06). Among the exposed population, children were more threatened by health risks than adults. Among them, Mn has a child risk quotient of 49.21 times that of adults. Finally, there were differences in health risks in different seasons. Overall, spring health risks were lowest and winters are highest

In summary, although the risk of human health caused by ingestion of food in water is still within the safe range, the health risk values of elements such as Cu and Cr show that they are getting closer to the safety line. From the perspective of public health and safety, we need to maintain further observation and research on these elements.

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Conflicts of Interest

The authors declare no conflict of interest.

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