RESEARCH ARTICLE

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Health risk assessment of heavy metals in the soil-water-rice system around the Xiazhuang uranium mine, China

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Abstract
Heavy-metal pollutants in the soil and surface water of agricultural areas accumulate in rice and may have adverse effects on the health of consumers. In this study, we determined the levels of heavy-metal contamination in agricultural areas around the Xiazhuang uranium deposits in northern Guangdong Province, China, using equidistant sampling methods along a river near the mine tailings. The pH values of all the water samples were determined. The heavy-metal concentrations in water, bottom sludge, and rice were measured. The extent of contamination was evaluated by calculating Nemerow's pollution index, contamination factor, and hazard quotient. The result shows U transferred from mine to soil and rice with irrigation water. The main pollutants and their pollution indices in soil are U (6.31), Th (4.02), Pb (2.52), Cd (2.36), Zn (1.52), and Mn (1.39). The rice grain can hardly enrich U and Th but were susceptible to Cr and Ni. The contamination factors (CFs) of the pollutants in rice grain are Cr (1.98) and Ni (3.09). The hazard quotient (HQ) shows that Cu (HQ > 1) could pose potential risks for humans upon long-term consumption of the rice.

Keywords : Heavy metal · Uranium · Rice · Soil · Water · Contamination assessment

Introduction

With the continuous development of industrial and mining exploration, various kinds of heavy-metal pollutants caused by mining have been observed in soil and crops. In China, many provinces have found that the contents of heavy metals, including Cu, Cr, Cd, Pb, Mn, and Hg, exceed the safe standard levels in crops. Notably, the Pb and Cd contents are much higher than the accepted levels (Cao et al. 2010; Yanchun et al. 2012; Liao et al. 2016; Ping et al. 2009; Ji-yun et al. 2016). In recent years, elevated uranium contamination levels have been found in some agricultural areas in China, and such toxic pollution in soil and crops is serious (Ping-hui et al. 2009; Gao-yang et al. 2015). Studies have shown that metal mineral mining, smelting, and residual waste runoff in mining areas

lead to heavy-metal pollutants in the surrounding soil and water which are difficult to deal with. The contamination of water is affecting not only soil and crops but also other activities such as aquaculture and poultry farming recreation (Kwon et al. 2017; Obiora et al. 2016; Sipter et al. 2008; Yu-Jing et al. 2004; Taylor and Francis Ltd. 2006; Ali et al. 2012). To date, about 10 million hectares of arable land are contaminated by heavy metals in China, while 2.1667 million hectares are contaminated by irrigation water containing heavy-metal pollutants. More than one tenth of all arable land has been polluted in China. According to statistics from various environmental protection departments, in China, 12 million tons of crops are polluted by heavy metals each year. This significant amount of contamination is a serious threat to public health.

Heavy-metal elements have chemical toxicity. Studies have shown that excessive heavy-metal concentrations (particularly Cd, Pb, Hg, Mn, Ni, and Cr) in the human body can cause heavy-metal poisoning, leading to many types of malignant diseases (Chen et al. 1999; Gillis et al. 2012; Prozialeck et al. 2008; Nemmiche et al. 2007). The radioactive toxicity of heavy metals, such as U and Th, can cause damage to organs and is often carcinogenic (L'Azou et al. 2002). At present, 23 provinces have uranium prospecting and mining activities in

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China. Therefore, it is extremely important to research soil and crop heavy-metal contamination near uranium ore districts. The Xiazhuang uranium ore field is one of the most famous uranium mines in China, with 18 uranium deposits and a large number of uranium-mineralized districts around it. It contains a variety of heavy metals, and mining activity has lasted for more than 50 years. In uranium mining, with hydrometallurgical ore smelting, waste residue constantly accumulated in the minine area. The waste residue undergone long-term weathering and eluviating produced lots of heavy metal pollutants which contaminated the river and steady transferred downstream. Contaminants have accumulated in the surrounding soil and water, and these have become long-term issues. Residents of Xiazhuang eat rice cultivated near the uranium mine tailings, and paddy field irrigation water is abstracted from the river downstream of the mine tailing dump. The paddy field crops may be more susceptible to heavy metal with polluted irrigation water, and heavy metals may be passed through the soil-water-rice route and then into the human body (Ali and Gupta 2006; Banerjee et al. 2016).

The research of heavy-metal contamination in paddy caused by U and Th is scarce in China. Current research regarding the heavy-metal pollution of food crops has mainly concentrated on Cr, Cu, Pb, and Zn, and most studies on plants were conducted under laboratory conditions. The research in laboratory often focuses on a specific element in plant, but the growth of rice in field is affected by various elements and factors such as soil pH, temperature, air condition, water, and fertilizer. So the conclusion of this paper provides some suggestion for more practical rice-growing status. This study therefore focuses on the North Guangdong Xiazhuang uranium mine area researching the rice in natural habitats. In addition to heavy-metal concentrations, pH values in the irrigation water, bottom sludge, rice, and paddy soil near the uranium mining area were measured to evaluate the contamination in soil, water, and crops.

Materials and methods

Study area

The study area is located in Weng Yuan County, northern Guangdong Province, China. The Xiazhuang uranium deposits lie on the eastern Guidong granite complex mass, with an area of about 185 km^2. It is the main uranium ore field in the deposits. This area contains 32 types of mineral resources, including an abundance of U, Fe, Pb, Zn, Cu, and W. The main crop grown in the county is rice. The area has a subtropical monsoon climate type, with warm, rainy weather. The landform is mainly mountainous terrain and basin. The prevailing soil types are red soils and acid soils, with high vegetation coverage.

Sample collection

The sampling area is shown in Fig. 1. There is a river about 3.2 km in length between Wengyuan County and the mine smelter. The rice planting area lies beside the river, which is used as a source of irrigation water for paddy fields. The village is about 1.9 km from the mines, while the paddy fields are about 2.2 km away. In this study, in order to reduce the interference caused by different sample spacing, we collected 500-mL water samples and 500-g bottom sludge samples from the river in plastic bottles every 300 m. The pH of all water samples was measured. Each soil sample (15 cm × 15 cm at a depth of 0 to 10 cm in the paddy field) was stored in a plastic bag. Rice samples were selected from mature plants. The full rice root, leaf, and rice grain were collected from fresh plants before storage in plastic bags. We totally collected 30 soil samples, 30 rice samples (including rice grain, root, and leaves; each of them collected 30 samples), 6 water samples, and 8 bottom sludge samples (the bottom of the river)

Sample preparation and analytical methods

The soil samples were ground after air-drying and sifted through a 100-mesh sieve. The concentrations of Ca, P, S, Fe, Zn, Cu, Pb, and Cd were measured by atomic absorption spectrophotometry (ALS Minerals-ALS Chemex, AFS3100). Total heavy-metal concentration was determined in 200-g soil samples after aqua regia and perchloric acid digestion. The rice samples were ground after air-drying and sifted through an 80-mesh sieve. The prepared rice samples (leaves and rice) were cold digested for approximately 8 h in nitric acid, then heated for 3 h in a graphite heating block. The samples were subsequently cooled and brought up to volume with HCl. The resulting solution was mixed thoroughly and analyzed by inductively coupled plasma-atomic emission spectrometry (ICP-AES) and inductively coupled plasma-mass spectrometry (ICP-MS). The analytical results were corrected for inter-element spectral interferences.
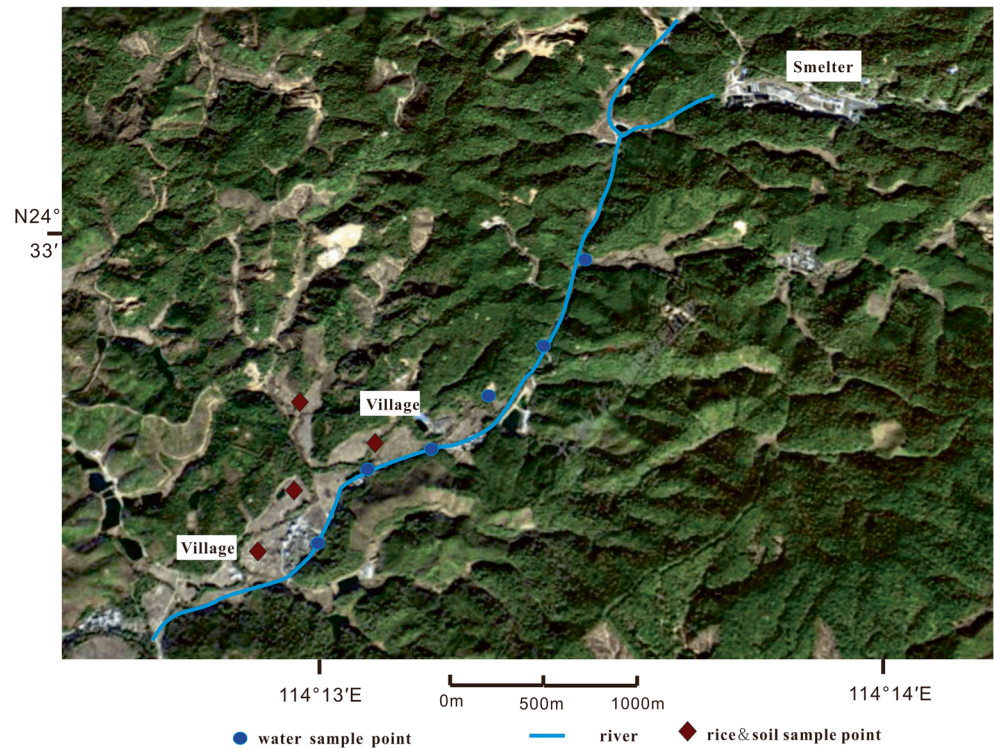
Data analysis methods

Contamination factor

The contamination factor (CF) is the ratio obtained by dividing the concentration of each heavy metal in the samples by the baseline or background value. The contamination factor was established according to the following equation:

$$CF = \left[C_{\text{heavy metal}} / C_{\text{baseline}} \right]. \tag{1}$$

Fig. 1 Sampling location map of the study area

Nemerow's pollution index

For soil samples with a variety of heavy-metal pollutants, Nemerow's pollution index can evaluate the comprehensive pollution status of samples for all heavy metals (Chen 2010).

Nemerow's pollution index can be calculated by Eq. (3):

$$P = \sqrt{\left[(Pa)^2 + Pi\mathrm{max}2\right]/2},$$
(3)

where P is the Nemerow pollution index, P_a is the average single factor pollution index, and Pi_{max} is the maximum single factor pollution index. The Nemerow index assessment standard is summarized in Table 2.

Enrichment factor

In this study, the enrichment factor (EF) was calculated to assess the enrichment ability of each heavy metal in rice.

For rice samples, the heavy-metal content evaluation standard referred to the National Food Safety Standards (MOH 2017). The degree of contamination for each CF was divided into 4 levels in Table 1 (Liu et al. 2005).

Single factor index

The single factor index was applied to assess the pollution degree of one pollutant in soil (Chen 2010). The index for a single pollutant was calculated by Eq. (2):

$$[P_i = C_i/S_i].$$
(2)

P_i is the single pollution index; C_i (mg/kg) represents the measured average concentration of heavy metals; S_i (mg/kg) is the standard value of the pollutants. For soil samples, we used the background value of soil in Guangdong Province as the standard value.

Table 1 The degree of contamination for each contamination factor

Contamination level	CF	Degree of contamination
1	$P_i < 1$	None
2	$1 \leq P_i < 3$	Moderate contamination
3	$3 \leq P_i < 6$	Considerable contamination
4	$P_i > 6$	Heavy contamination

Table 2 Grading standard for Nemerow's index of heavy metals

Pollution level	P	Pollution condition
1	$P \leq 1$	Non-polluting
2	$1 < P < 2$	Mild pollution
3	$2 < P < 3$	Moderate pollution
4	$P > 3$	Heavy pollution

Table 3 Heavy-metal contamination: soils

| Elements	Range	Ave (27 samples)	Median	CV	Background value of soil elements (mg/kg)	
	(mg/kg)				Values in China	Values in Guangdong
U	7.80–134.00	34.69	18.8	1.05	2.72	5.5
Th	26.1–70.80	51.47	49.35	0.26	12.8	–
As	2.30–6.00	3.64	3.3	0.28	9.6	8.9
Cd	0.03–0.30	0.13	0.14	0.54	0.08	0.06
Cr	4.00–28.00	15.75	16	0.35	53.9	–
Co	4.50–10.00	6.82	6.35	0.24	11.6	7
Cu	3.50–16.70	10.51	11.45	0.36	20.7	17
Mn	200.00–652.00	386.5	411.5	0.39	540	279
Ni	2.30–10.80	7.39	8	0.38	24.9	14.4
Pb	67.90–128.50	90.63	87.25	0.17	23.5	36
Zn	37.00–123.00	71.69	74	0.31	68	47.3

The EF can identify the heavy metals with the most uptake and accumulation in rice. The EF was calculated according to Eq. (4) (Taylor 1964; Zhang et al. 2009)

$$EF = \left[(metal/Al)_{sample} / (metal/Al)_{background} \right]. \quad (4)$$

This study uses the hazard quotient (HQ) to assess the potential risk of each heavy-metal pollutant. The HQ was calculated by Eq. (5):

$$HQ = C \times \frac{IR}{MRL \times BW}. \quad (5)$$

where C is the concentration of a heavy-metal pollutant in the rice, IR are the factors in Chinese dietary guidelines for daily per capita rice intake, MRL denotes the minimal risk level, and BW denotes adult body mass (Noli and Tsamos 2016; ATSDR 2016; NHFPC 2016; USEPA 2005). A potential risk is indicated if HQ > 1, while HQ < 1 is deemed to be safe.
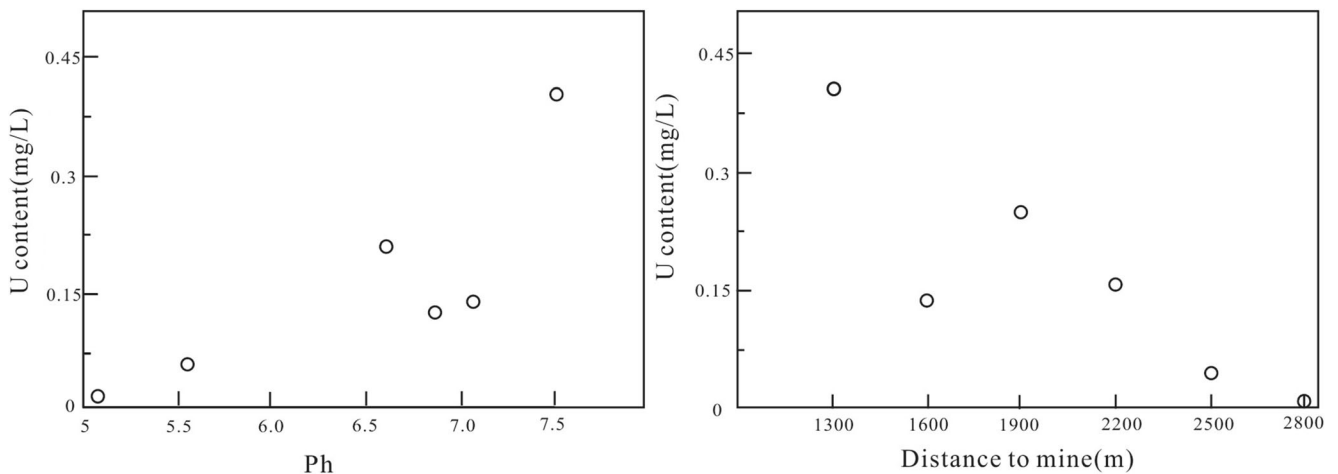
Fig. 2 The relationships between U and sampling distance from the mine as well as water sample pH values

Table 4 Heavy-metal contamination: water

Element	U μg/L	Th mg/L	As	Cd	Cr	Co	Cu	Mn	Ni	Pb	Zn
Ave (6)	190.4	< 0.001	< 0.001	0.001	< 0.001	0.002	< 0.001	0.542	< 0.001	0.002	< 0.001
GB3838I	0.680	–	0.100	0.010	0.100	1.000	1.000	0.100	0.020	0.100	2.000

Table 5 Heavy-metal contamination: bottom sludge

Element	U mg/kg	Th	As	Cd	Cr	Co	Cu	Mn	Ni	Pb	Zn
Range	8.0–95.2	28.3–56.4	3.1–5.6	0.02–0.26	4.0–19.0	5.3–8.7	4.8–8.6	269.0–678	3.0–5.8	75.1–139	37.0–62
Ave (8)	66.5	39.6	4.48	0.18	9.25	7.25	6.38	521.25	4.33	106.93	51.75
Average in paddy	34.69	51.47	3.64	0.13	15.75	6.82	10.51	386.5	7.39	90.63	71.69

Results and discussion

Concentration of heavy-metal elements in paddy soil, water, and bottom sludge

Table 3 lists the concentration range and mean values of heavy-metal concentrations in soil. We used the background values for soil elements in Guangdong as reference values in Table 1 and the concentration of heavy metals in the surface water is shown in Table 4 (General Bureau of China National Environmental Protection 2002). The mean values of heavy-metal contents in water and bottom sludge are listed in Tables 2 and 3. The study measured the pH values of water, the U content, and statistical variations, in water (Fig. 2). From Table 1, all soil samples were contaminated with U, Th, and Pb. In total, 81.25% of samples contained Zn and Cd, and 56.25% of samples contained Mn. In decreasing order, the concentrations of heavy metals that exceeded stated allowable limits in soil were the following: U > Th > Pb > Cd > Zn > Mn.

The concentrations of As, Cr, Co, Cu, and Ni were low. Compared with other uranium mines in China, the concentrations of U, Th, Pb, and Zn were higher, while those of Cd and Mn were lower in Xiazhuang paddy soil (Ping et al. 2009; Gaoyang et al. 2015; Haribala and Wang 2016; Yan et al. 2016). According to Tables 2 and 3, the water near the rice planting area was contaminated with U and Mn. The concentration of U was 280 times the limit (GB5749-2006), and the Mn content was five times the surface water limit (GB3838-2002). U and Mn pollutants are dangerous due to their toxicity and bad biodegradability in water (ImranAli, 2010; Ali et al. 2009). The concentrations of U, Pb, and Mn in bottom sludge were higher than in soil (Table 5), which is expected in sediments from the mine that have been carried by surface water. Pb and Zn are associated with U in the Xiazhuang uranium mine, and the background U and Pb contents were higher than those in other mines in China, which may be one source of U and Pb pollution (Zou 2012). Research shows that Mn is difficult to be stable in the surface water with high oxygen contents and only

Table 6 Heavy-metal content of rice

	Range mg/kg	Ave (27)	Median	CV %	Food safety standard limit mg/kg
U	< 0.005	< 0.005	< 0.005	–	1.9
Th	0.002–0.015	0.006	0.004	0.8	1.2
As	0.06–0.27	0.154	0.145	0.495	0.2
Cd	0.008–0.33	0.071	0.031	1.527	0.2
Cr	0.6–3.4	1.975	1.95	0.552	1
Co	0.999–12.35	3.791	1.885	1.1	2.5
Cu	1.13–4.61	2.308	2.21	0.444	10
Mn	41–177	83.375	79	0.511	85
Ni	1.93–4.22	3.094	3.17	0.224	1
Pb	0.05–0.11	0.079	0.075	0.283	0.2
Zn	14.7–22.8	18.113	17.9	0.133	50

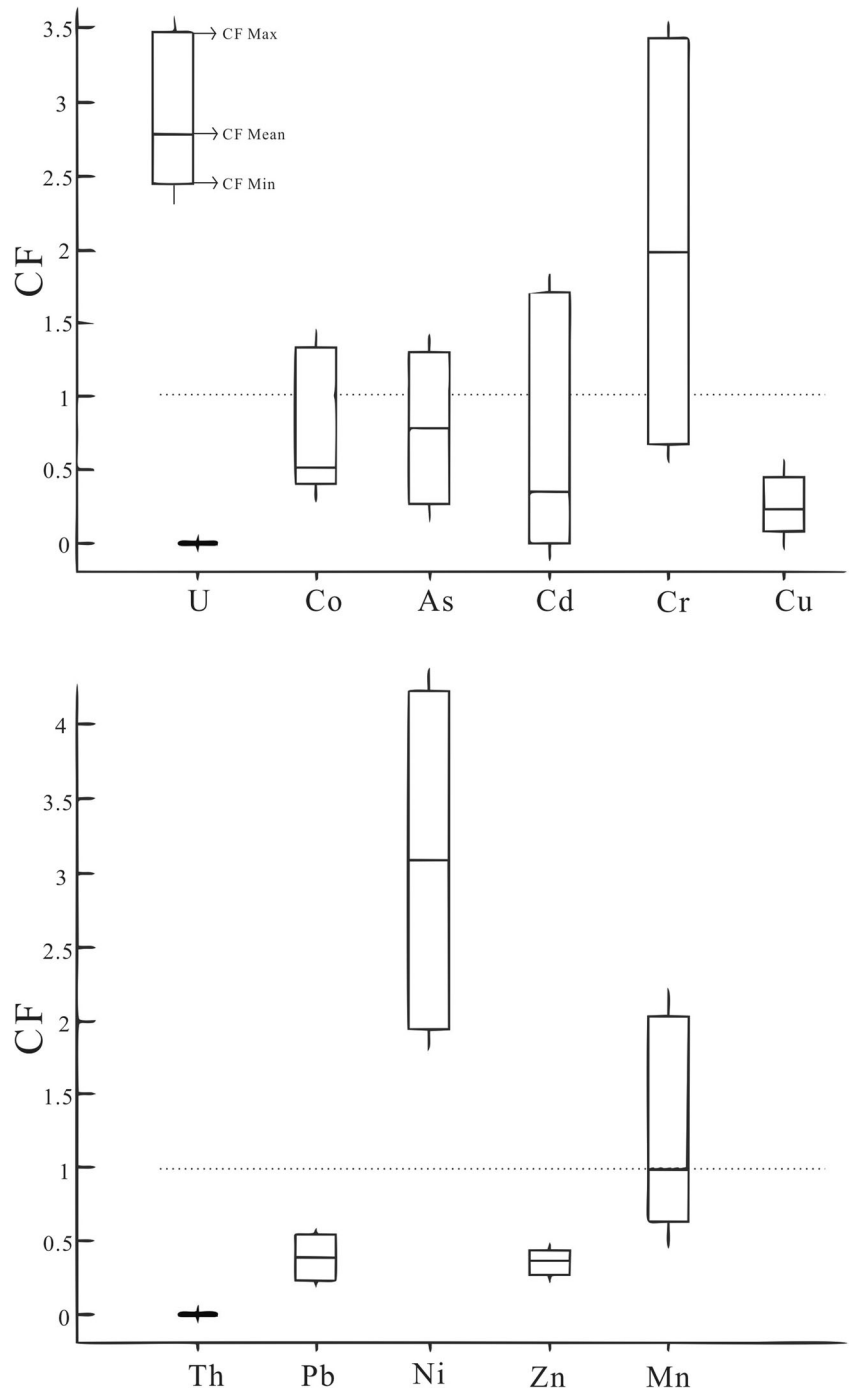**Table 7** Enrichment factors and heavy-metal content of root and leaf samples

	U	Th	As	Cd	Cr	Co	Cu	Mn	Ni	Pb	Zn
Value in rice	< 0.005	0.01	0.15	0.07	1.98	1.89	2.31	83.38	3.09	0.08	18.11
EF	0.31	0.69	1.17	3.42	4.05	9.96	3.05	1.98	7.02	0.58	2.49
Value in leaves	0.47	0.66	0.04	0.03	14.25	3.67	5.43	220	7.07	2.44	25.50
Value in root	10.25	35	4.06	0.34	47.60	62.40	24.30	460	41.70	49.60	135

Table 8 Contamination factors and hazard quotient for soil and rice

		U	Th	As	Cd	Cr	Co	Cu	Mn	Ni	Pb	Zn
Soil	P_i	6.31	4.02	0.41	2.36	0.29	0.97	0.62	1.39	0.51	2.52	1.52
	Nemerow index	4.65										
Rice	CF	–	0.01	0.77	0.35	1.98	0.754	0.23	0.98	3.09	0.39	0.36
	HQ	–	<0.01	0.15	0.71	–	–	1.14	–	–	0.11	–

Fig. 3 Contamination factors for heavy-metal contaminants in rice
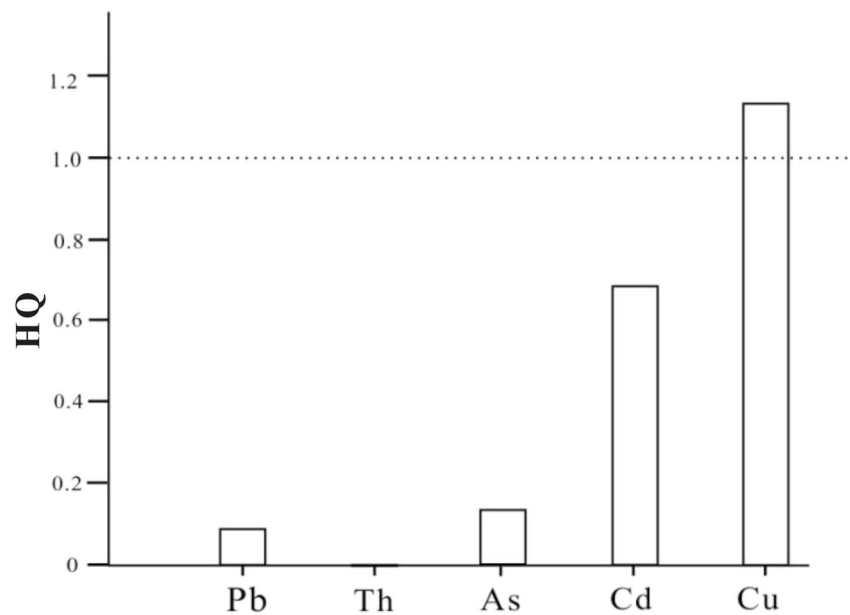
the process of groundwater recharge will enrich Mn in the surface water (Gantzer et al. 2009). Water with high pH values is suitable for U complex transfer, and acidic water leads to the precipitation of uranium in the bottom sludge (Runan and Jinshuang 1985; Song et al. 2014). The concentration of U was closely related to sampling distance from the mine, and the U content decreased with decreasing pH of water samples (Fig. 2). The U content in the sediment had a rising trend with sample sites gradually close to the mine, which indicates that U from mine waste is constantly enriched in the bottom sludge and U precipitation occurs continuously. The pH conditions provide the advantage of uranium to transformation and richness in the bottom sludge. Uranium is prevalent in Xiazhuang surface waters, and it contaminates surrounding soils by irrigation and permeation. Compared with U and Mn, cadmium, cobalt, and lead also existed in the surface water at the lower concentration. The high concentration of U, Mn, Cd, Co, and Pb in the bottom sludge proves that there is a transfer-precipitation process in water and sediment. On the contrary, As, Ni, Cu, Cr, and Zn are hard to be found in the surface water, and their contents in water sediment samples were lower than those in paddy soils.

As a result, there are two factors that contribute to the high concentrations of heavy metals in Xiazhuang paddy soils and water. One is the high soil background values of heavy metals in the mine area; U, Mn, Cd, Co, and Pb, through the surface water flow from the mine, which the villagers used to irrigate paddy soil, contaminated the paddy fields.

Concentration of heavy-metal elements in rice

Tables 6 and 7 list the concentrations of heavy-metal elements in rice grain, root, and leaves and their enrichment factors. The

limiting values were referenced from national safety standards. Nemerow's index and the hazard quotient are listed in Table 6. All rice grain samples contained Ni and Cr, and 75% of rice grain samples contained concentrations that exceeded the limits. Co-concentrations exceeded the limit in 25% of rice grain samples (MOH 2017). Heavy-metal concentrations in rice grain decreased in the following order: Ni > Cr > Co. According to Table 7, the concentrations of U, As, Th, Cr, Mn, Ni, Pb, and Zn in the paddy were $C_{root} > C_{leaves} > C_{rice}$, while the Co contents in rice grain were higher than those in the leaves. In decreasing order, the enrichment factor (Noli and Tsamos 2016; Cejpkov et al. 2016) in rice was as follows: Co > Ni > Cr > Cd > Cu > Zn > Mn > As > Th > Pb > U. Except for Cd and Co, all of the heavy metals were enriched in the root and then transported into the rice grain and leaves. The enrichment factors of U, Th, Pb, and As were low, while those of Co, Cd, Cu, and Ni were high. This suggests that rice grain is much more susceptible to Co, Cd, Cr, Cu, and Ni contamination than other heavy metals. Comparing the achievements of other researchers that little uranium, thorium, and lead can enter into the fungus though their concentration were very high in soil (Kubrová et al. 2014; Kubrová and Borovička 2015), similar findings have emerged in rice. We found a small amount of uranium, thorium, and lead in the root, much less of them in leaves, but they barely existed in rice grain. It means uranium, thorium, and lead can hardly migrate in rice.

The pollutants in soil and rice were quite different. The contamination factors for soil and rice grains are listed in Table 8. The predominant pollutants in soil were U, Th, Pb, and Cd. Figure 3 shows that the predominant heavy-metal pollutants in rice grain were Cr and Ni. Ni pollution is much more serious than that caused by Cr. The hazard quotients of the less prevalent elements (Th, Cu, As, Cd, and Pb) are shown in Fig. 4; all

elements were shown to have HQ < 1, except for Cu (HQ > 1). This proves that Cu could be a potential health risk to long-term rice consumers in the study area. The two tables show the differences of U and Th contents in two paddy fields.

Conclusion

This study detected heavy-metal element contaminant levels in soil, rice, sediment, and surface water around the Xiazhuang uranium mine. The health risks of rice were also assessed.

For the water The average concentrations of U and Mn were 280 times and 5 times, respectively, greater than the national safety standard for surface water. The U concentration decreased about 0.075 mg/L every 300 m with increasing distance from the mine. The results indicate that the surface water used for irrigation caused U, Mn, Pb, and Cd pollution in soil but the water had no effect on rice pollution.

The main pollutants in the paddy soil are U, Th, and Pb. The contamination factors for heavy metals in the paddy fields, in decreasing order, were U (6.31) > Th (4.02) > Pb (2.52) > Cd (2.36) > Zn (1.52) > Mn (1.39) > Co (0.97) > Ni (0.51) > As (0.41) > Cr (0.29). The Nemerrow index of paddy is 4.65, which means that there is considerable contamination with multiple heavy metals in paddy.

For the rice Uranium, thorium, and lead can hardly enter into rice grain though they are the main pollutants in the soil. The predominant pollutants in rice grain were Ni and Cr. The paddy root had greater heavy-metal enrichment ability than the leaves, and Ni, Co, Cd, Cr, Cu, and Zn accumulated more easily than the other elements in the rice grain. The enrichment factors for Ni, Co, Cd, Cr, Cu, and Zn in rice grains were Cd (3.32), Cr (4.05), Co (9.96), Cu (3.05), Ni (1.96), and Zn (2.49). The hazard quotients for trace levels of heavy metals in rice grain were Pb (0.11), Th (< 0.01), As (0.15), Cd (0.71), and Cu (1.14), suggesting that Cu could pose potential risks for the human body upon long-term consumption of the rice.

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