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Research Article

Surface Water Contamination by Uranium Mining/Milling Activities in Northern Guangdong Province, China

The northern region of Guangdong Province, China, has suffered from the extensive mining/milling of uranium for several decades. In this study, surface waters in the region were analyzed by inductively coupled plasma optical emission spectrometry (ICP-OES) for the concentrations of uranium (U), thorium (Th), and non-radioactive metals (Fe, Mn, Mg, Li, Co, Cu, Ni, and Zn). Results showed highly elevated concentrations of the studied radionuclides and metals in the discharged effluents and the tailing seepage of the U mining/milling sites. Radionuclide and heavy metal concentrations were also observed to be overall enhanced in the recipient stream that collected the discharged effluents from the industrial site, compared to the control streams, and rivers with no impacts from the U mining/milling sites. They displayed significant spatial variations and a general decrease downstream away from upper point-source discharges of the industrial site. In addition, obvious positive correlations were found between U and Th, Fe, Zn, Li, and Co ($R^2 > 0.93$, $n = 28$) in the studied water samples, which suggest for an identical source and transport pathway of these elements. In combination with present surface water chemistry and chemical compositions of uraniumiferous minerals, the elevation of the analyzed elements in the recipient stream most likely arose from the liquid effluents, processing water, and acid drainage from the U mining/milling facilities. The dispersion of radionuclides and hazardous metals is actually limited to a small area at present, but some potential risk should not be negligible for local ecosystem. The results indicate that environmental remediation work is required to implement and future cleaner production technology should be oriented to avoid wide dispersion of radioactivity and non-radioactive hazards in U mining/milling sites.

Keywords: Heavy metal; Radionuclide; Water pollution

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1 Introduction

The recent earthquake-tsunami that induced nuclear crisis in eastern Japan Nuclear Power Plant in 2011 seriously aroused world-wide public concerns on the radionuclide contamination in the environment. However, very little is known on the radioactive and environmental hazards behind the booming exploration of uranium resources in China during the past several decades. Similar to the extraction of other mineral resources, the extensive extraction of uranium (U) ores has produced enormous quantities of wastes [1–3]. Numerous waste-rock piles and slime dams have been generated and

disposed randomly, sometimes within local communities, with little consideration for the surrounding environment and water resources [3, 4]. Large amounts of lean U ore have been excavated, leaving various non-target elements exposed to the surface. Inherent to this, abandoned uranium mine wastewater, dumps, and tailings may become sources of not only radioactive pollutants (i.e., U and daughter elements) but also heavy metals (e.g., Cu, Ni, and Zn), which are potentially toxic to the environment [5]. Among them, U and its compounds, due to the combination of their large range of chemotoxic and radiotoxic properties, are highly reactive and may cause progressive or irreversible renal injury that may lead to kidney failure and death in some acute cases [3–7]. Therefore, special attention should be paid to the hazard potential of U and its compounds because of their exceptionally high mobility in the aquatic environment [5–7].

A rural area, located in the northern part of Guangdong Province, South China, bears a low grade granite uranium ore. The local U mining/milling industry commenced several decades ago [1].

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Abbreviations: DWL, drinking water limit; ICP-OES, inductively coupled plasma optical emission spectrometry

Sulfuric acid was used to dissolve and oxidize the uranium. The spent acids and tailings were piped from slurry to unlined ponds [8]. An estimated 0.1 million tons of tailings and 0.7 million gallons of liquid effluent were disposed into a 20-acre square, constituting an eco-environmental problem of extraordinary spatial dimensions. Until now, previous works mainly focused on measurements of radioactivity, but few studies on radionuclide contamination in this area have been carried out. Significant radiometric anomalies were detected during airborne radiometric surveys over mining sites in this area. The radiometric studies revealed that high gamma-activities emanating from immobile daughters such as ^{226}Ra of the uranium decay series in the tailing ponds pose serious threats to the nearby environment as a result of dust dispersion [8–10].

One of the main contributions of this study stems from the fact that most of the attention has been focused on the emissions of radon into the atmosphere during the past decades, but little attention was paid on their releases into the water bodies. Therewith, the aim of this article is to examine the contamination level, to identify the sources and the spatial distribution of radioactive elements (U, Th) and non-radioactive metals (Fe, Mn, Mg, Li, Co, Cu, Ni, and Zn) in surface water in the local area and to assess the potential risks arising from them.

2 Materials and methods

2.1 Site description

The studied U-mineralized field is situated in the eastern portion of the Guidong intrusion in Northern Guangdong Province, China [1]. The intrusion was assumed to be a composed one, which was emplaced in various stages (from ~184 to 143 Ma) during the Yanshanian orogeny [11]. The mineralization is hosted mainly in medium-grained, porphyritic biotite granite, and to a lesser extent, in marginal muscovite microgranite. The contact zones of the pluton underwent the contact metamorphism at different degree. In the northeast and the east, the wall rocks are composed of Cambrian-Ordovician low metamorphic sandstone, slate, and carbonaceous slate, while those in the south are mainly Devonian-Carboniferous sandstone and carbonate rock [11]. The U tenors bear an average of 8–9 mg/kg U in Cambrian-Ordovician rocks and 5–13 mg/kg in Devonian-Carboniferous strata [1]. The ore field covers an area around 407 km², with approximately 50 000 residents, partially in small villages or scattered over the country [11]. Main economic activities comprise poorly developed farming and cattle breeding, and the main products are paddy, sugar cane, and soybean. The climate is subtropical-monsoonal with annual precipitation of 1600 mm. Seasonal precipitation ranges from a few hundred millimeters (October–February) to nearly 1000 mm (April–August). Mean temperatures in the year vary from 14 to 29°C, and the relative air humidity varies from 60 to 70% [8, 12]. The area is dusty and covers with rare vegetation. Typical vegetation consists of pine trees, shrubby trees, *Dicranopteris linearis* and *Phragmites australis* [8].

2.2 Reagents

All glassware and vessels were soaked in a 0.2 mol/L nitric acid solution for 24 h and subsequently rinsed with deionized water. Reagents were of super-pure grade. Suprapure nitric acid were purchased from the Merck Company (Darmstadt, Germany). All analysis

was carried out using ultra-pure water (18.25 M Ω cm) from a Microprocessor Automatic Water Still 400 (Suntex Instruments Co., Taipei, China).

2.3 Sampling

The tailing ponds and associated infrastructure, such as return water dams, pipelines, and metallurgical plants, constituted a multitude of sources of U migration. In addition, the chemical leaching of U from tailings and subsequent waterborne transport, as a dissolved phase into subjacent aquifers and nearby streams, formed another source of mining-related water pollution.

Water samples were collected from the local area through two sampling periods. In the first period (May, 2010), samples were taken from seven locations. Thus, seven surface water samples were collected accordingly, including three types of process wastewater (original process water, S1; heap leaching water, S2; perched water, S3), tailings seepage (S4), tap water (S7), river water 20 km (S5) and 40 km (S6) away from the uranium facilities. In the second sampling period (November 2010), more extensive investigation was achieved by collecting various surface waters around the U mining/milling site, including the tailing pond (pipelined effluent, W1; tailing seepage, W2), the mining and milling site (boreholes pit water, W3; decanting void water, W4; furrow water, W5), and the heap leaching site (heap leaching water, W6; heap leaching effluent, W7; ditch water, W8; pond water, W9, W10, and W11). In order to better understand the impact of the mining/milling activities, selected surface water samples (W12–W21) were investigated from aquifer systems in the proximity of the industrial site, which included a stream flowing through the tailing footprints, distributaries, and natural streams near or distal to the contamination sources (W12–W20, Fig. 1) and a tap water (W21). All the surface water samples (approximately 1 L for each) were collected manually with plastic containers from the top surface. The samples were filtered through 0.45- μm filters at site, and then preserved with ultrapure HNO₃ to acidify the samples to pH 2 and stored in coolers ($\leq 8^\circ\text{C}$) until analysis.

2.4 Instrumental analysis

All determinations were performed by means of inductively coupled plasma optical emission spectrometry (ICP-OES) (Optima 7000 DV, PerkinElmer, Inc., Waltham, USA). The operating conditions of the instrument are summarized in Tab. 1.

The standard solutions of U and Th were prepared from ICP Single-element Standard Solution of U and Th (Ultra Scientific, Kingston, USA), respectively, and those for metal elements (Fe, Mn, Mg, Li, Co, Cu, Ni, and Zn) were prepared from ICP multi-element standard solution IV (Merck, Darmstadt, Germany). Ultra-pure water acidified with suprapure nitric acid was used as the calibration blank and for all dilutions. The detection limits were calculated as an average of three times the standard deviation (SD) of ten replicate analyses of a procedural blank, and they were determined as 0.01 mg/L for U, Th, and Mg, 0.0005 mg/L for Fe, Co, Li, and Ni, 0.0002 mg/L for Mn and Zn, and 0.0015 mg/L for Cu. The quality control standards were run at selected intervals to ensure consistent instrument performance over the length of the analysis. The data are expressed as the means \pm SD of three replicate measurements (Tab. 2 and Fig. 2).

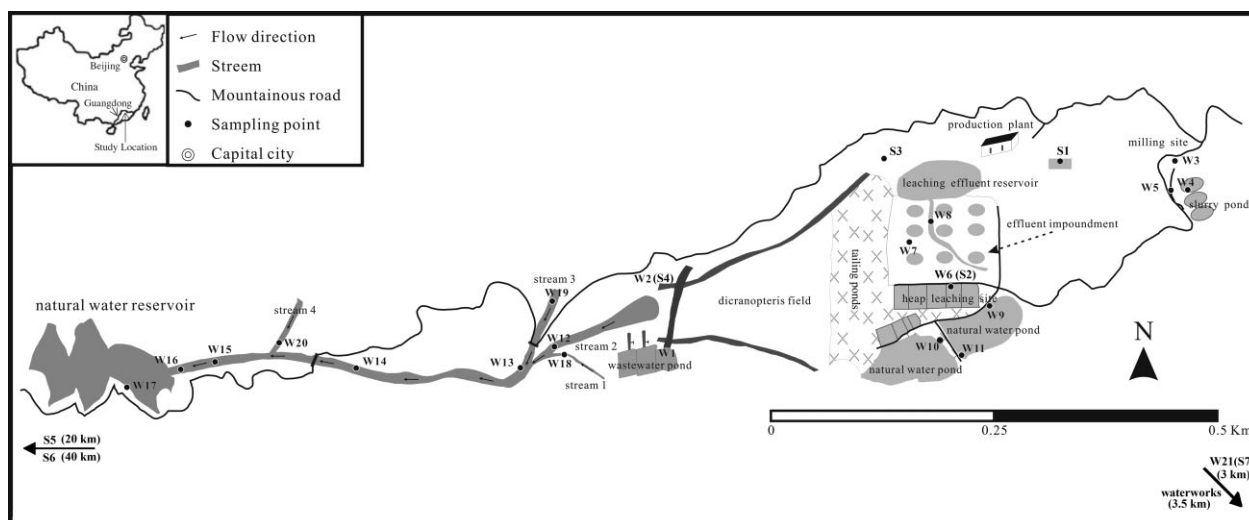


Figure 1. Map showing the sampling sites.

3 Results and discussion

3.1 U concentrations in surface waters

The data for U, Th, and other metal concentrations in the studied surface water samples were summarized in Tab. 2. The U and Th concentrations in water samples were found to vary from below the detection limit to 349.1 ± 32.1 mg/L and from below the detection limit to 17.33 ± 0.20 mg/L, respectively. As recommended by various health and environmental protection agencies, the safe limit of U in drinking water for human beings ranges from 0.002 to 0.05 mg/L [13–16]. During the first sampling period, the dissolved U concentrations of the process wastewater near the tailing ponds (S1–S3) were overall elevated, as compared to the drinking water limit (DWL). The tailing seepage (S4) which directly flows to a stream (stream 2) showed a high U concentration, exceeding the DWL by an order of over 100. Dissolved Th concentrations were comparatively low, since Th compounds are hardly soluble and easily precipitated in the solid phase [17]. Surface water from 20 to 40 km downstream of the uranium facilities (S5 and S6) generally exhibited low U and Th concentrations. It may be explained by the dilution effect of flushing water or due to the adsorption and enrichment by various solid mediums. Apart from this, the mountain torrents during site sampling may also contribute to enough dilution.

Table 1. Optimized instrumental conditions of ICP-OES

Parameters	Value/Type
Plasma frequency	40.68 MHz
RF power	1300 W
Plasma flow rate	15 L/min
Auxiliary gas flow rate	0.2 L/min
Nebulizer gas flow rate	0.8 L/min
Peristaltic pump	1.5 mL/min
Sample pump rate	1.5 mL/min
Nebulizer type	GemCone
Spray chamber type	Ryton
Integration time	10 s
No. of replicate	3

Given to the above findings, more extensive investigations were carried out during the second sampling period. As shown in Tab. 2, U concentration in the tailing seepage (W2) decreased to less than a half of that in S4 sampled in the 1st sampling period. A plausible explanation is the weak leaching effects due to limited water volumes in the dry season. In the milling site, the highest U level (8.7 ± 0.076 mg/L) prevailed in the decanting void water (W3), followed by 2.26 ± 0.011 mg/L in boreholes pit water (W4) and 1.702 ± 0.011 mg/L in furrow water (W5). Exceptionally elevated U level (84.6 ± 7.06 mg/L) was exhibited in the heap leaching water (W6). It subsequently flowed through a series of several settling ponds to allow for uraniferous particles to settle out, but only approximately two thirds were directly injected into the product line, while around one third was retained in the sediment of pond. All of this directly caused a high U concentration (7.237 ± 0.052 mg/L) in effluents from the heap leaching site (W7). Similarly, the ditch water (W8) from the heap leaching site contained U at over 20 times the DWL. Even the pond water samples (W9–W11) showed generally elevated U concentrations, varying from 0.238 ± 0.002 to 1.373 ± 0.007 mg/L. This may arise from the contamination of natural pond water by heap leaching effluent. Similarly, relatively high concentrations of U (0.338 ± 0.667 mg/L) were found in the pond water nearby a former heap leaching site in Germany [18].

Analyzing the spatial distribution of U levels of stream 2 (Tab. 2) that directly collects various discharged effluents from the mining/milling activities and tailing seepages, distinct differences were noticeable along the course of the stream. By far the highest in-stream U concentrations (1.781 ± 0.009 mg/L) occurred in W12, the headwater of stream 2, further illustrating the higher potential of the uranium mining/milling facilities for direct stream pollution. This peak was followed by a significant drop to about one eighth of the concentration, most likely owing to dilution by non-U-polluted rural runoff or chemical immobilization of U in some solid mediums. It is worth to note that at this constant U level (0.073 – 0.316 mg/L), a part of water from the stream 2 entered a pipeline which conveyed the stream across the de-watered compartments directly into some farmlands for irrigation. This U level, though not extremely high, was still well above the DWL (0.050 mg/L) [16]. Comparable U levels were also found in the surface water at identical

Table 2. Concentrations of U, Th, and non-radioactive metals (mg/L) in the surface water samples from the studied uranium mining/milling area

Sampling period	Sample	Description	U	Th	Fe	Mn	Mg	Cu	Zn	Co	Ni	Li
May, 2010	S1	Original process water	245.200 ± 29.400	10.210 ± 0.170	2264 ± 134	493.700 ± 19.700	1497 ± 20	4.656 ± 0.049	24.570 ± 1.470	2.539 ± 0.020	2.134 ± 0.154	11.700 ± 0.190
	S2	Heap leaching water	349.100 ± 32.100	17.330 ± 0.200	3060 ± 190	455.800 ± 30.500	1829 ± 35	5.784 ± 0.102	31.840 ± 0.700	2.963 ± 0.010	2.591 ± 0.106	15.300 ± 1.620
	S3	Perched water	0.776 ± 0.001	0.016 ± 0.001	1.258 ± 0.010	4.962 ± 0.010	15.630 ± 0.300	0.036 ± 0.001	0.526 ± 0.006	0.045 ± 0.001	0.029 ± 0.001	0.192 ± 0.004
	S4	Tailings seepage	5.002 ± 0.085	0.170 ± 0.002	23.180 ± 1.070	23.070 ± 0.050	98.990 ± 8.810	0.092 ± 0.001	1.016 ± 0.003	0.088 ± 0.001	0.061 ± 0.001	0.579 ± 0.001
	S5	River water ^{a)}	ND ^{f)}	ND	0.214 ± 0.003	0.342 ± 0.001	2.216 ± 0.011	0.001 ± 0.001	0.007 ± 0.001	0.003 ± 0.001	ND	0.002 ± 0.001
	S6	River water ^{b)}	ND	ND	0.049 ± 0.003	ND	1.238 ± 0.004	ND	ND	ND	ND	0.001
Nov., 2010	S7	Tap water	0.001	ND	0.035 ± 0.001	ND	0.367 ± 0.002	ND	0.165 ± 0.001	ND	ND	0.001
	W1	Pipelined effluent	2.091 ± 0.016	0.030 ± 0.002	0.820 ± 0.003	30.050 ± 2.890	133.800 ± 2.000	0.032 ± 0.001	1.217 ± 0.040	0.146 ± 0.001	1.004 ± 0.024	0.064 ± 0.019
	W2	Tailings seepage	1.863 ± 0.017	ND	0.410 ± 0.015	35.550 ± 2.240	148.800 ± 1.100	0.009 ± 0.001	0.444 ± 0.014	0.042 ± 0.002	0.019 ± 0.001	0.214 ± 0.004
	W3	Boreholes pit water	8.700 ± 0.076	0.800 ± 0.017	0.470 ± 0.008	0.620 ± 0.005	11.600 ± 0.250	0.026 ± 0.001	0.009 ± 0.001	ND	ND	0.060 ± 0.001
	W4	Decanting void water	2.260 ± 0.011	0.072 ± 0.001	0.930 ± 0.002	0.810 ± 0.003	8.690 ± 0.035	0.011 ± 0.001	0.081 ± 0.001	0.009 ± 0.001	0.007 ± 0.001	0.080 ± 0.001
	W5	Furrow water	1.702 ± 0.011	0.056 ± 0.001	1.460 ± 0.018	1.580 ± 0.004	5.030 ± 0.023	0.007 ± 0.001	0.051 ± 0.001	ND	ND	0.041 ± 0.001
	W6	Heap leaching water	84.600 ± 7.060	5 ± 0.037	383.800 ± 10.400	841 ± 44.800	266 ± 1	5.969 ± 0.024	12.650 ± 0.300	1.798 ± 0.009	1.950 ± 0.002	1.465 ± 0.001
	W7	Heap leaching effluent	7.237 ± 0.052	0.235 ± 0.003	10.700 ± 0.140	69.450 ± 7.990	314.600 ± 3.100	0.401 ± 0.006	1.471 ± 0.021	0.211 ± 0.001	0.155 ± 0.006	2.117 ± 0.064
	W8	Ditch water	0.326 ± 0.005	ND	0.040 ± 0.001	94.230 ± 5.770	334.400 ± 2.200	0.006 ± 0.001	0.414 ± 0.006	0.086 ± 0.003	0.062 ± 0.001	1.808 ± 0.011
	W9	Pond water	1.373 ± 0.007	ND	6.810 ± 0.043	89.280 ± 7.070	316.900 ± 5.161	0.024 ± 0.001	0.492 ± 0.006	0.092 ± 0.001	0.068 ± 0.001	1.358 ± 0.034
	W10	Pond water	0.238 ± 0.002	ND	0.310 ± 0.008	79.620 ± 2.630	309.600 ± 3.400	0.015 ± 0.001	0.308 ± 0.012	0.069 ± 0.004	0.036 ± 0.001	1.726 ± 0.015
	W11	Pond water	0.353 ± 0.007	ND	4.580 ± 0.132	156.100 ± 11.300	655.900 ± 1.800	0.101 ± 0.003	0.929 ± 0.025	0.240 ± 0.002	0.234 ± 0.015	2.206 ± 0.024
	W12	Headwater of stream 2	1.781 ± 0.009	0.007 ± 0.001	0.110 ± 0.001	36.970 ± 6.080	166.500 ± 2.300	0.008 ± 0.001	0.260 ± 0.002	0.030 ± 0.001	0.014 ± 0.001	0.216 ± 0.002
	W13	Water of stream 2	0.112 ± 0.002	0.005 ± 0.001	0.270 ± 0.002	1.970 ± 0.006	8.380 ± 0.024	ND	0.022 ± 0.001	0.001 ± 0.001	ND	0.044 ± 0.001
	W14	Water of stream 2	0.316 ± 0.001	0.006 ± 0.001	0.420 ± 0.003	6.190 ± 0.095	27.510 ± 0.950	0.006 ± 0.001	0.173 ± 0.001	0.018 ± 0.001	0.008 ± 0.001	0.123 ± 0.002
W15	Water of stream 2	0.207 ± 0.002	0.004 ± 0.001	0.530 ± 0.029	5.340 ± 0.079	22.610 ± 0.120	ND	0.038 ± 0.001	0.002 ± 0.001	ND	0.052 ± 0.001	
W16	Water of stream 2	0.073 ± 0.001	0.004 ± 0.001	0.120 ± 0.001	1.650 ± 0.004	7.020 ± 0.066	0.001 ± 0.001	0.047 ± 0.001	0.003 ± 0.001	ND	0.055 ± 0.001	
W17	Reservoir water ^{c)}	0.003 ± 0.001	0.006 ± 0.001	0.650 ± 0.004	0.200 ± 0.001	0.870 ± 0.008	ND	0.007 ± 0.001	ND	ND	0.036 ± 0.001	
W18	Water of stream 1	ND	0.006 ± 0.001	0.010 ± 0.003	0.010 ± 0.001	0.410 ± 0.002	ND	0.02 ± 0.001	ND	ND	0.036 ± 0.001	
W19	Water of stream 3	ND	0.005 ± 0.001	0.010 ± 0.002	0.010 ± 0.001	1.470 ± 0.013	ND	ND	ND	ND	0.035 ± 0.001	
W20	Water of stream 4	ND	0.006 ± 0.001	0.080 ± 0.001	0.020 ± 0.001	0.350 ± 0.001	ND	ND	ND	ND	0.035 ± 0.001	
W21	Tap water	0.064 ± 0.002	0.008 ± 0.001	1.330 ± 0.001	1.020 ± 0.005	4.150 ± 0.025	0.003 ± 0.001	0.834 ± 0.003	ND	ND	0.047 ± 0.001	
	DWL ^{d)}	0.050	0.100	0.300	0.100	NG ^{g)}	1	1	2	1	0.020	NG ^{g)}
	Background value ^{e)}	0.001	0.001	0.085	0.045	NG	0.006	0.008	0.001	0.001	0.008	0.002

a) 20 km distant from the site;
 b) 40 km distant from the site;
 c) the end of stream 2;
 d) DWL, drinking water limit of U, Th is from [16] and the others from [24];
 e) [25];
 f) ND, not detectable;
 g) NG, not given.

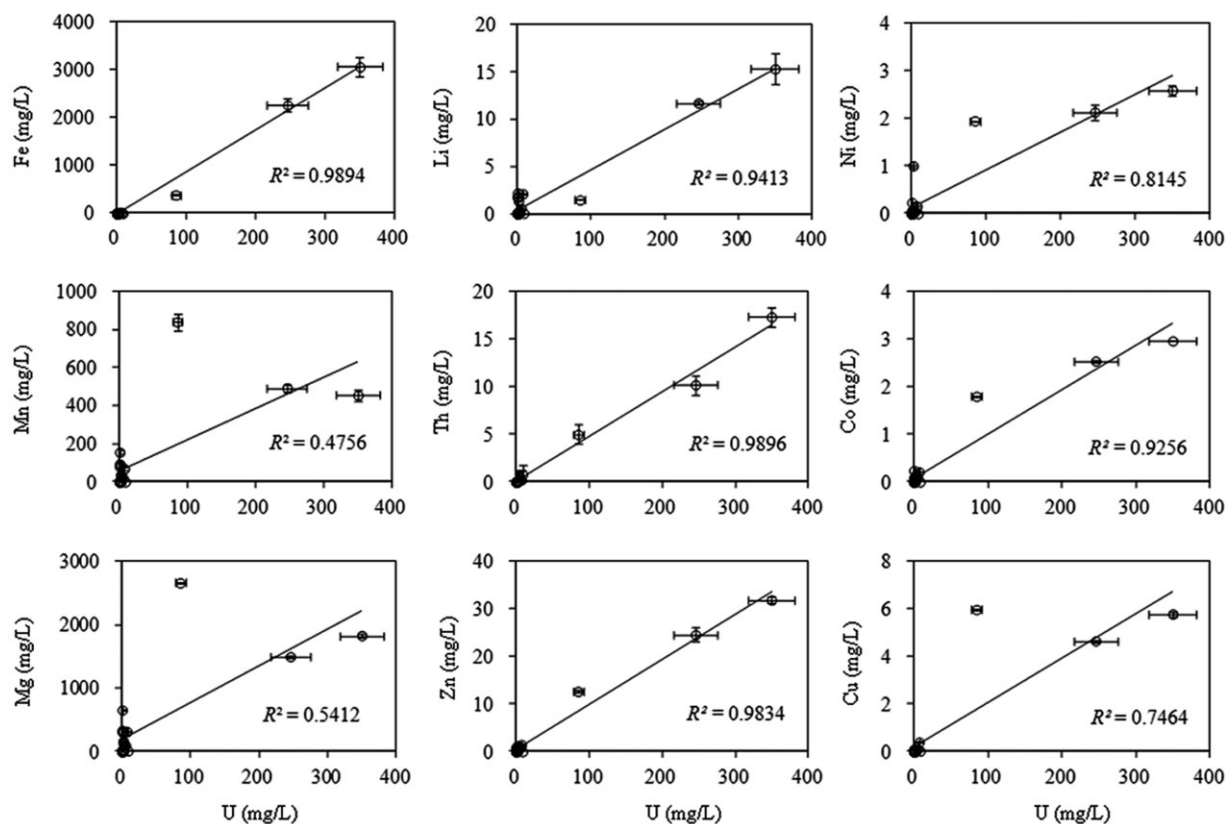


Figure 2. Correlations between U and other studied elements.

sampling sites. As reported, U concentrations ranged from 0.552 to 1.842 mg/L in the pond water at an abandoned uranium mine in Portugal [19]. The average concentration of U in the stream water amounted to 0.319 mg/L at a gold mining site in South Africa [20].

The farmlands that received the stream waters for irrigation thus might be at a risk of U contamination. Other exposure pathways discernable from the survey included the use of stream-water for irrigating garden vegetables (for own consumption) as well as for watering of livestock [18]. Apart from it, an even greater risk exists for the local residents using stream-water as the main or sole water resource. This is especially typical for inhabitants of informal settlements with no formal water supply, who were frequently observed to consume untreated water directly [21]. Besides, U concentrations were below the detection limit (0.001 mg/L) in the water samples from similar natural streams (i.e., stream 1, stream 3, and stream 4) nearby, which shared a common granitic geological environment with stream 2. These concentration levels were as well consistent with those (<0.001 mg/L) reported in the background river [22]. Therefore, we could further speculate that U contamination in stream 2 was not naturally occurring but induced from the mining/milling activities.

It is worthy to note that, in the 2nd sampling period, U concentration in the tap water (W21) rose to 0.064 ± 0.002 mg/L. This was slightly above the proposed guideline for drinking water of 0.05 mg/L in China (Tab. 2). According to the local residents, the tap water was exclusively provided by pumping the underground water. This led us to surmise that groundwater might be influenced

by U migration as well. As reported, U contamination of groundwater could be achieved by tailing seepages and flooding of mine void water [18–21, 23].

3.2 Non-radioactive metal concentrations in surface waters

The values for the concentrations of trace metals such as Fe, Mn, Mg, Li, Co, Cu, Ni, and Zn in the water samples are given in Tab. 2. The concentrations of Fe in the water samples lied in the range of 0.01–3060 mg/L, mostly much above the recommended level (0.3 mg/L) [24]. The concentrations of Mn in all the water samples overall highly exceeded the DWL (0.1 mg/L) [24]. In contrast, the contents of Cu, Zn, Co, and Ni in most of the water samples were below the safe limit. Extremely high dissolved Fe, Mn, and Mg concentrations were shown in the surface water samples collected from U mining/milling site, which bore significant enrichment of U and Th, e.g., at sites of S1, S2, and W1–W12.

In particular, significantly elevated concentrations were displayed in S1, S2, and W3, amounting to 3060 ± 190 mg/L Fe, 493.7 ± 19.7 mg/L Mn, and 1829 ± 35 mg/L Mg, from hundreds to thousands order of magnitude higher than the drinking limit and background value (Tab. 2) [16, 24, 25]. To a lesser extent, an elevation of Li, Ni, Co, Cu, and Zn was observed in the samples in the U mining/milling process water and surface water from stream 2. Meanwhile, concentrations of the examined heavy metals were quite low in the natural streams

Table 3. Main element compositions of predominant minerals in the studied uranium mine

Predominant minerals in the studied uranium mine	Main element composition
Pitchblende	U ⁴⁺ , U ⁶⁺
Pyrite	Fe, (Co, Ni)
Hematite	Fe, (Mn, Mg, Co)
Chalcopyrite	Cu, Fe, (Ni)
Chalocite	Cu
Chlorite	Fe, Mg

The element in bracket means that the mineral includes it, but not substantially.

and rivers (for Fe ~0.01 mg/L, Mn ~0.01 mg/L, Li ~0.001 mg/L, Mg ~0.40 mg/L, and for Ni, Co, Cu, Zn below the detection limit), which represent the background values. Therewith, the impacts of the tailing ponds and the U mining/milling processing water on stream 2 could be visible, by means of a simple comparison of metal contents in the surface water samples investigated.

3.3 Source identification

Linear regression analysis was used to estimate the relationship between the concentrations of U and the other studied elements (Fe, Mn, Mg, Li, Th, Zn, Ni, Co, and Cu) at the sampling sites, and the results are plotted in Fig. 2. Significant positive correlations were found between U and Th, Fe, Zn, Li and Co ($R^2 = 0.93\text{--}0.99$). Some positive linear relationships were observed between U and Ni, Mn and Mg ($R^2 = 0.48\text{--}0.81$). This may be explained by an overall identical source and transport pathway of these elements. As reported, the uraniumiferous ores predominantly consist of pitchblende, pyrite, hematite, chalcopyrite, chalocite, sphalerite, and chlorite [11]. The extraction of U from the ores was achieved by means of a heap-leach process that composed of ore crushing and the addition of a sulfuric acid to dissolve and oxidize U, followed by precipitation with ammonium hydroxide solution. The liquid effluent generated during this procedure consisted of an acid solution with high concentrations of other elements dissolved along with U, such as Th, Fe, Cu, Zn, Li, Mn, and Mg.

Accordingly, based on the general chemical composition of such minerals (Tab. 3) and the present surface water chemistry, it is possible to surmise that U and the other element enhancement in stream 2 might arise from the liquid effluent emissions of the U mining/milling activities and the acid mine drainage of the waste (i.e., waste rock material, tailings dump). During the mining/milling procedure, the waste rock and spoil deposited in the void immediately adjacent to the active strip, came into direct contact with water, resulting in potential leaching of solutes. More importantly, the waterborne erosion of tailings could facilitate easy access of oxygen to the widely dispersed tailing materials, thereby causing sulfide oxidation (acid mine drainage) to occur readily and liberating U together with other elements directly into the stream 2.

4 Conclusions

Results from this preliminary environmental assessment indicate that concentrations of radionuclides (U, Th) and other non-radioactive metals (Fe, Mn, Mg, Li, Co, Cu, Ni, and Zn) in the U mining/

milling discharged effluents and tailing seepages largely or moderately exceeded the drinking water safe limit given by the Chinese National Guideline. Elevated concentrations of these elements were observed in some surface water samples from the recipient stream 2 of the waste effluent in the vicinity of the mining/milling facilities. Spatial distribution of the studied elements in the water of stream 2 showed an overall decreasing trend along the course from the industrial site to downstream. Concentrations of U in the surface water samples generally displayed strong positive linear relationships with those of other elements examined. By analyzing the present surface water chemistry and the chemical compositions of uraniumiferous minerals, it could be concluded that the elevation of U and the other elements in stream 2 were mainly contributed by the anthropogenic activities from the U mining and milling facilities. Water samples from other streams and rivers had much lower concentrations of the studied radionuclides and heavy metals, suggesting that the dispersal of radioactive waste and effluents from the mining and milling activities is actually limited to a small area, and that there is no immediate hazard to the off-site public at present.

However, the high level contaminations in part of the area still pose a potential of long-term risk to the public, given the existing environmental conditions with no effective remediation. Firstly, it is likely that U-polluted stream-water from the discharge point into the underlying aquifer could eventually affect surrounding river systems indirectly. Besides, a potential risk exists for local ecosystem through using this U-polluted stream-water for irrigation, watering, and direct consumption. Therefore, it is critical and important to further investigate the environmental quality of uranium tailing sites, and to improve the treatment of waste materials from the U mining and metallurgy.

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