



APPLICATION OF ISOTOPE TECHNIQUES TO GROUNDWATER POLLUTION RESEARCH FOR XIANGSHAN URANIUM ORE FIELD, CHINA

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Abstract - *The investigation of groundwater pollution due to uranium deposits focused on the most important uranium metallogenic area--Zhoujiashan district of Xiangshan uranium orefield, China. Groundwater collected from five completed exploration boreholes in the area is regarded as the pollution source and is traced and analysed by using isotope as well as radio-hydrochemical techniques. In addition, the pollution situation of a small uranium ore pile for heap-leaching and a big uranium ore open pit are monitored by the same techniques. It has been experimentally proven that the uranium concentration and the uranium isotope ratio $^{234}\text{U}/^{238}\text{U}$ in natural waters are two sensitive indicators of radioactive pollution in natural waters. It was concluded that under present conditions, exploration of uranium deposits may not cause serious groundwater pollution of radioactive elements (U,Ra,Rn and Th), however, it is difficult to avoid the serious surface water pollution coming from the exploitation of uranium ore by a big open pit.*

1. INTRODUCTION

Radioactive pollution is a special kind of pollution which may be originated from various natural and artificial nuclides. Radioactive elements such as uranium, radium and radon are widely distributed in nature. As the diffusion rate of radium in water is extremely low and it is easy to be adsorbed by various solid materials (such as soil and different colloidal materials), radium does not migrate in most cases, very far from its source.

Radon is an inert gas which exists in atomic form. It can not enter the structure of minerals or chemical compounds nor be transported in natural water. Moreover, the half life of radon isotope ^{222}Rn is only 3.825d, so in natural conditions, it is not able to migrate far from its production place. The radioactive element thorium is not distributed as commonly as uranium and its concentration in natural water is quite low (about 0.5~1.0 $\mu\text{g}/\text{l}$). As viewed from geochemistry, most of thorium compounds are stable, weak in volatility and difficult to be dissolved in water. Especially since thorium in aqueous solution does not take part in redox reaction, it is impossible for it to cause obvious radioactive pollution to surface water drainage [1].

The radioactive element uranium is a kind of valence-changeable element. In an oxidizing medium or by the action of acidic or intensely alkaline groundwater, it can be changed from U^{4+} to U^{6+} uranyl cation (UO_2^{2+}) and dissolved in water. In a reducing medium, however, it is transformed from U^{6+} into U^{4+} and precipitated. Most of uranium minerals in nature, therefore, are difficult to dissolve in natural water (pH=7). Thus, uranium

concentration in natural water is one of the most important recognizable indicator of radioactive pollution [2].

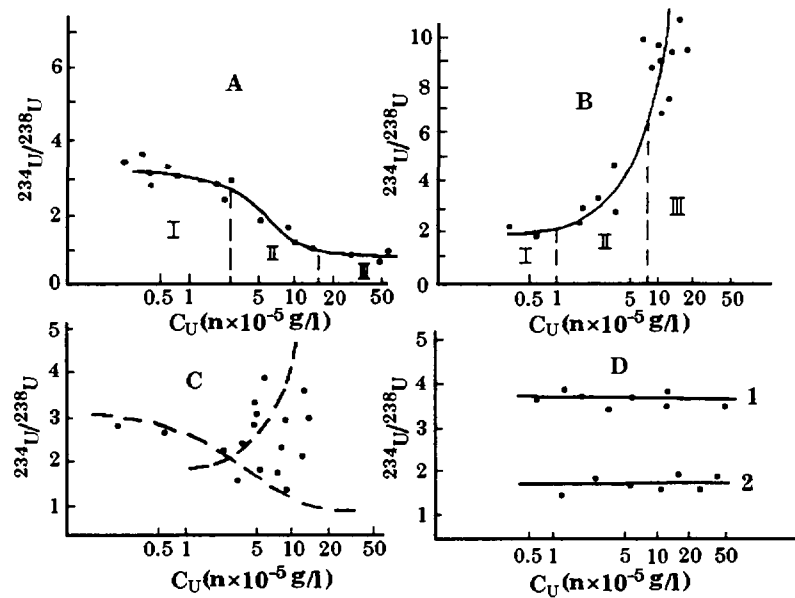


Fig.1 Relationship between uranium isotope ratio and uranium concentration in groundwater

A -Water halo of hydrothermal type of uranium deposit; B-Water halo of impregnated uranium molybdenum deposit; C-Water halo of uranium-phosphorus deposit in metamorphic rock; D-False anomaly halo: 1) Water halo in granite; 2) water halo in Tertiary sandstone [3].

As the increment of radioactive element concentration in natural water may result from different factors, both true and false anomalous halos in water are consequently generated. The determination of the isotope contents or the activity ratio of the radioactive elements is one of the most effective methods to recognize the genesis of radioactive anomalies in water and to discriminate the false anomalies from the true ones. Experiments show that the activity ratio of $^{234}\text{U}/^{238}\text{U}$ in water gradually decreases with increase of uranium concentration as the water coming from non-mineralized rocks enters into uranium orebodies. When ^{234}U and ^{238}U in water are in equilibrium state, the activity ratio $^{234}\text{U}/^{238}\text{U}$ becomes 1 [3,4]. Curves A and B of Fig. 1 show the behavior of water haloes of hydrothermal and impregnated uranium molybdenum deposits, respectively. With the increase of U-concentration in water, the activity ratio $^{234}\text{U}/^{238}\text{U}$ gradually decreases in the case of hydrothermal U-deposits and rapidly increases in the second case (Fig. 1B). Curve C represents the analytical results of the activity ratio $^{234}\text{U}/^{238}\text{U}$ in the dispersion halos in water of a uranium-phosphorus deposit in metamorphic rock. This figure shows that the variation of activity ratio $^{234}\text{U}/^{238}\text{U}$ is bigger and resembles the superimposition of curves A and B. Curve D shows false uranium anomalies in water of two districts, in which the activity ratios $^{234}\text{U}/^{238}\text{U}$ are all situated at the background level. In short, when the uranium concentration in water reaches the anomalous value and its activity ratio $^{234}\text{U}/^{238}\text{U}$ is simultaneously near 1, it indicates the occurrence of the hydrothermal uranium mineralization process. When the activity ratio $^{234}\text{U}/^{238}\text{U}$ in water of sedimentary and metamorphic rock area is 5-10 times higher than the background, it means that impregnated uranium deposits exist.

It has also been discovered that the activity ratio $^{226}\text{Ra}/^{228}\text{Ra}$ in water of non-mineralized igneous rocks and sedimentary rocks is generally less than 0.5. However, the ratio in water of uranium ore is equal to 2 or greater. The activity ratio $^{230}\text{Th}/^{232}\text{Th}$ in water of non-mineralized rocks is, in general, less than 5, but that in water associated with uranium ore may increase up to several hundred accompanied by an increment of ^{230}Th concentration in water (Fig.2,3) [3].

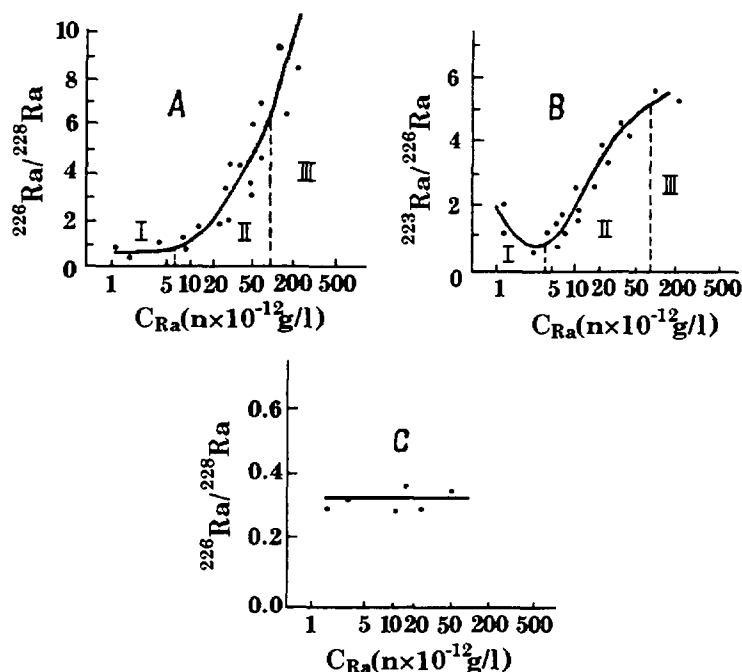


Fig.2 Relationship between radium isotope ratio and radium concentration in groundwater. A,B-Water halo of uranium deposit; C-Water halo of false anomaly in granite. [3]

In summary, the recognition criteria for a radioactive anomaly in groundwater is as follows: $^{234}\text{U}/^{238}\text{U} \geq 1$, $^{226}\text{Ra}/^{228}\text{Ra} > \text{or} = 2$ and $^{230}\text{Th}/^{232}\text{Th} > 20$.

These parameters can also be used as the basic data for the study of radioactive pollution of groundwater (Table 1).

Table 1. Obvious anomaly and obvious normal value of radioactive isotopic ratio in anomalous water [3]

Isotope ratio	Igneous rock		Sedimentary and metamorphic rock	
	Obvious anomaly	Obvious normal value	Obvious anomaly	Obvious normal value
$^{234}\text{U}/^{238}\text{U}$	≥ 1.0	$> \text{or} = 3$	$> \text{or} = 5$	-
$^{226}\text{Ra}/^{228}\text{Ra}$	$> \text{or} = 2$	$< \text{or} = 0.5$	$> \text{or} = 2$	$< \text{or} = 0.5$
$^{230}\text{Th}/^{232}\text{Th}$	> 20	< 1	> 20	< 1

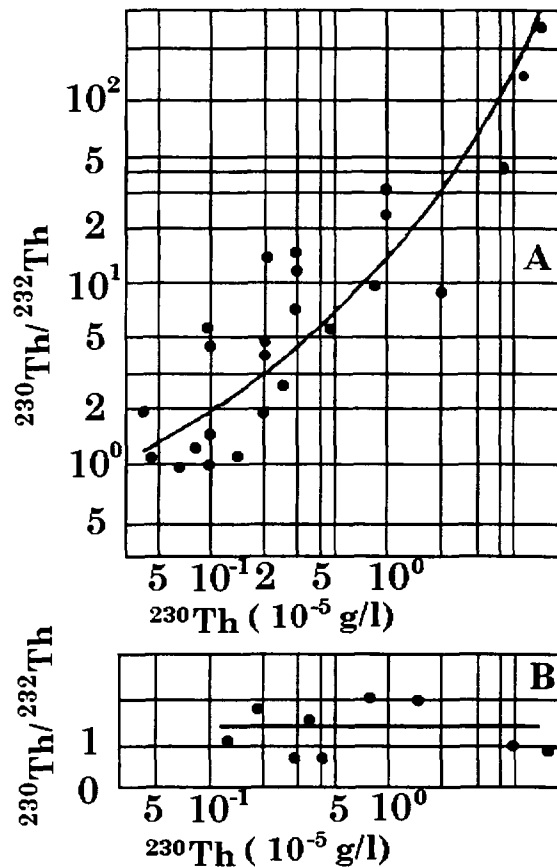


Fig.3 Relationship between activity ratio $^{230}\text{Th}/^{232}\text{Th}$ and ^{230}Th concentration in natural water
 A--Water halo of uranium deposit; B--Water in granite. [3]

2. PRESENT INVESTIGATIONS

2.1. Description of the study area

The Xiangshan uranium orefield is one of the largest U-orefield in China, containing many uranium deposits in the Mesozoic acidic volcanics. It is situated in a volcanic basin (the Xiangshan basin) which is located in the southwestern part of the Ganhang tectonic belt in Jiangxi province, China (Fig. 4).

The Xiangshan uranium orefield has been developed for more than 30 years and much geological and hydrogeological research work has been carried out. Results of the above work are used as the foundation for the investigation of groundwater pollution in this area [5-10].

The basement of the Xiangshan basin consists of Sinian biotite-quartz schist, phyllite and coal-bearing formation. The cover is composed of Upper Jurassic volcanics which are divided into the lower part--the Daguding Formation and the upper part--the Ehuling Formation. The former one mainly consists of sandstone, siltstone, welded tuff and rhyodacite. The latter is made of sandy conglomerate, siltstone, welded tuff and thickly-bedded porphyroclastic lava. The whole volcanic series dips at about 20° to 30° towards the

centre of the Xiangshan basin. Collapse structures are well developed including ring fractures, downcast flexure and interlayered fissures.

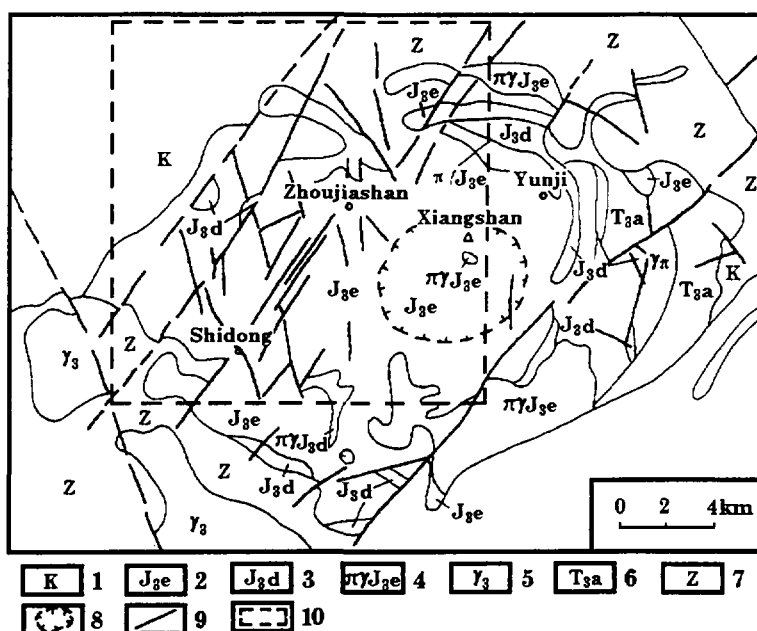


Fig.4. Schematic geological map of Xiangshan uranium orefield

1.Red sandy conglomerate, Cretaceous. 2.Ehuling Formation, Upper Jurassic. 3.Daguding Formation, Upper Jurassic. 4.Sub-porphyrific granite, Ehuling Formation, Upper Jurassic. 5.Granite. 6.Sandy conglomerate and sandstone, Upper Triassic. 7.Sinian metamorphics. 8.Volcanic ring collapse structure. 9.Fracture structure. 10.Boundary line of the investigated area.

Two main types of uranium mineralization have been recognized in the Xiangshan uranium orefield: One is the sodium metasomasis type with a mineralization age of 115.2 ± 0.5 Ma, developed in the eastern and the northern part of the Xiangshan uranium orefield. Another is the fluorite-hydromica type with a mineralization age of 97.6 ± 7.6 Ma, developed in its western part. The above two mineralization ages were calculated from U-Pb isochron of pitchblendes [6]. The detailed research indicates that the main ions in the ore-forming solution of sodium metasomasis uranium deposits are Na^+ and Ca^{2+} , HCO_3^- and minor Cl, originally alkaline which later on evolved to an acidic solution. The ore-forming solution of fluorite-hydromica uranium deposits are dominantly Na^+ , Ca^{2+} , HCO_3^- with minor F^- , SO_4^{2-} , and Cl. The ore-forming solution was originally acidic to weakly acidic which evolved into alkaline during metallogenic process.

The investigated area is located in the western part of the Xiangshan uranium orefield with an area of about 260 km^2 . The area has a subtropical, humid rainy climate. The mean annual temperature of this area is about 18°C , the highest in summer at 39.6°C and the lowest in winter at -9.2°C . The annual rainfall in the area is from 1500mm to 2200mm while the annual evaporation capacity is from 1200mm to 1600mm. The area belongs to a middle-low mountain area with moderate erosion, denudation and faulting. The area exhibits a higher relief in the southeast and a lower one in the northwest with significant elevation difference, steep mountains and deeply-dissected valleys leading to conditions which encourage surface runoff. The main surface drainage system in this area includes a series of brooklets, such as: Shidong-brooklet, Shutong-brooklet and Zhoujiashan-brooklet which flow over the Gongxi river (Fig. 5).

One of the main discharge patterns of the groundwater is the overflow of spring water including the overflow of water from the completed exploration boreholes. In the investigated area, the overflow water merges into the Gongxi river via corresponding brooklets. Thus, groundwater pollution, especially radioactive pollution which results from the overflow water of the completed exploration boreholes in the district with rich uranium deposits can be traced in brooklets and rivers. Rainwater passing through the uranium ore pile or the uranium ore open pit may transport radioactive pollutants into brooks, then to the main river.

2.2. Field sampling

In order to investigate groundwater pollution that results from the exploration of uranium deposits and to compare that with the surface water pollution, 15 water samples were collected. Among them, there are 5 borehole water samples (1, 4, 7, 9, 10), 8 brooklet water samples (2, 3, 5, 8, 11, 12, 13, 14) and 2 river water samples (6, 15). The surface water samples 14, 15 were taken from the north of a quite big uranium ore open pit. The brooklet water sample 12 is situated near a small uranium ore pile for heap-leaching. Each sample was measured for radioactivity, hydrochemistry and stable isotopes. Temperature of water samples and air were measured in the field while the geological and hydrogeological characteristics were described.

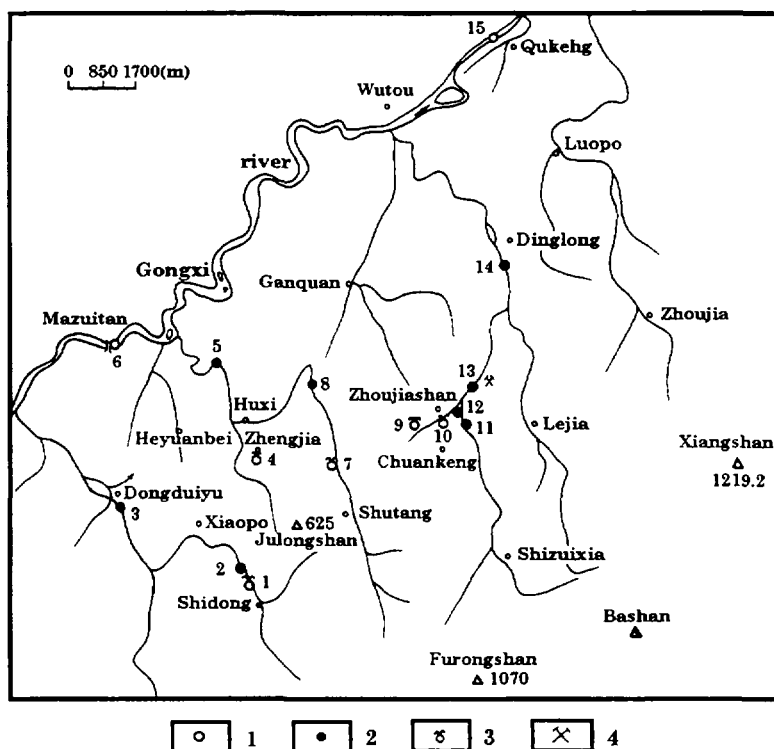


Fig. 5 Location of sampling points

1. River site. 2. Brooklet site. 3. Completed exploration borehole site. 4. Uranium ore open pit site.

2.3. Laboratory analysis

The 15 water samples were measured for $K^+ + Na^+$, Ca^{2+} , Mg^{2+} , HCO_3^- , SO_4^{2-} , Cl^- ; U, Th, Ra, Rn; M(or TDS), δ^2H , $\delta^{18}O$ and pH as well as the ratios of the radioactive isotopes $^{234}U/^{238}U$, $^{230}Th/^{232}Th$. As the half life of radon isotope ^{222}Rn is only 3.825d, radon concentrations were measured by the emanation ionization method in the same day of the field sampling.

P_{350} extractive chromatography was used to determine the radioactive isotopic ratios $^{234}U/^{238}U$ and $^{230}Th/^{232}Th$ of natural water samples. Five liters of water sample are taken, then acidified using concentrated nitric acid (HNO_3). Ferric chloride ($FeCl_3$) and ammonium chloride (NH_4Cl) used as carriers are added, then ammonia (NH_4OH) is added to adjust the pH to a value of 8-9, for the coprecipitation of ferric hydroxide with uranium and thorium hydroxide. The precipitate is dissolved by using concentrated nitric acid, then a mixed solution containing 15% HNO_3 and 15% $Al(NO_3)_3 \cdot 9H_2O$ is prepared for chromatography. After the mixed solution is passed through the P_{350} teflon support column, uranium and thorium are purified, then thorium is mixed with the hydrochloric acid (HCl) and the uranium with the sodium fluoride (NaF). The mixture is thereafter separately electro-deposited on different stainless steel disks. After the disks are rinsed and dried, they are measured with α -spectrometry. The net counts in the energy region of the isotopes of U and Th in the same spectrum are compared, then the ratios of $^{234}U/^{238}U$ and $^{230}Th/^{232}Th$ of natural water samples are obtained [11, 12].

Hydrogen isotope samples were prepared using the metal uranium method. The hydrogen gas was collected in special tubes and measured in a Finnigan-MAT251 gas isotope mass-spectrometer. The precision of the method is about $\pm 1\%$. The CO_2-H_2O equilibrium method is used to measure the ratio of oxygen isotope. The precision of the method is about $\pm 0.2\%$.

3. RESULT AND DISCUSSION

In March 1997, 15 water samples in the investigated area were collected for studying the radioactive pollution caused by the exploration and the exploitation of uranium deposits. Tables 2, 3, 4 show the hydrochemical results, the concentrations of U, Th, Ra and Rn, the activity ratios ($^{234}U/^{238}U$ and $^{230}Th/^{232}Th$) and the stable isotopic contents (δ^2H and $\delta^{18}O$). According to these results, some main points can be obtained as follows.

Table 2. shows that the concentrations of the cations $K^+ + Na^+$, Ca^{2+} (especially $K^+ + Na^+$) and the anion HCO_3^- of five borehole water samples (1, 4, 7, 9, 10) are obviously higher than those of other surface water samples. This fact indicates that the five borehole water samples (1, 4, 7, 9, 10) are in close space relationship with the ores of fluorite-hydromica uranium deposits. As the brooklet water sample 12 is near a small uranium ore pile for heap leaching, the concentrations of the cations Ca^{2+} , Mg^{2+} and the anion SO_4^{2-} are obviously higher than those of other surface water samples, while the concentrations of cations $K^+ + Na^+$ and anions HCO_3^- and Cl^- are all close to zero with pH equal to 5.2, the lowest in the 15 water samples collected in the investigated area. Apparently, the the small uranium ore pile for heap leaching was affected by sulphuric acid. Although the brooklet water sample 13 is not far from the brooklet water 12, as it is shown in Fig. 5, its concentrations of cations $K^+ + Na^+$, Ca^{2+} , Mg^{2+} and anions HCO_3^- , SO_4^{2-} , Cl^- show no

obvious difference with those of other surface water samples. This fact indicates that the brooklet water sample 13 has not been affected by the uranium ore pile.

Table 3 shows that the concentrations of radioactive elements radium and thorium of the 15 water samples collected in the investigated area (except the brooklet water sample 12) show no obvious regular variation. This phenomenon might be closely related to the radioactive hydrochemical features of radioactive elements radium and thorium as mentioned above. The concentrations of radioactive elements uranium and radon of five borehole water samples (1, 4, 7, 9, 10) and the brooklet water sample 12 are all obviously higher than those of other surface water samples, which reveals that the above anomalous concentrations rapidly disappear as soon as the waters coming from above five boreholes (1, 4, 7, 9, 10) and the brooklet water 12 flow into a brook or a river. In other words, no obvious radioactive pollution caused by them would occur in this area. As the locations of borehole water samples (1, 4) are slightly far from the uranium rich Zhoujiashan mineralization district than those of borehole water samples (7, 9, 10), as it is shown in Fig. 5, it is reasonable that the concentrations of radioactive elements uranium and radon (especially uranium) of borehole water samples (1, 4) are less than those of borehole water samples (7, 9, 10). The variation of uranium concentrations of brooklet water samples (13, 14) is quite significant, while their Ra, Th and Rn concentrations show no obvious change, which once more indicates that the concentration of uranium in water is one of the most important indicator for evaluating the radioactive pollution.

Table 4. shows that the activity ratios $^{234}\text{U}/^{238}\text{U}$ (1.07, 0.98, 1.14) of borehole water samples (7, 9, 10), respectively are closer to 1 than those of borehole water samples (1,4) which have ratios of (1.83, 1.54) respectively. This reveals that the borehole water samples (7,9,10) are closer to the equilibrium state of ^{234}U and ^{238}U in water than the borehole water samples (1,4). This phenomenon is also coincident with the fact that the locations of borehole water samples (7, 9, 10) are closer to the uranium rich Zhoujiashan mineralization district than those of borehole water samples (1,4). The activity ratios $^{234}\text{U}/^{238}\text{U}$ of brooklet water samples (12, 13, 14) are 0.99, 1.08, 0.98 respectively. This fact indicates that the brooklet water sample 12 and 14 are closer to the equilibrium state of ^{234}U and ^{238}U in water than the brooklet water sample 13, because the former is closer to a small uranium ore pile for heap leaching than the latter. The uranium concentration of sample 14 is $130 \times 10^{-7}(\text{g/l})$ which is much higher than the background value, indicating that it is difficult to avoid serious surface water pollution coming from the rainwater passing through a big uranium ore open pit. There is no a regular distribution of thorium activity ratio $^{230}\text{Th}/^{232}\text{Th}$ of the 15 water samples. This fact shows that the activity ratio $^{230}\text{Th}/^{232}\text{Th}$ in water can only be used as a supplemental recognition parameter for evaluating the radioactive pollution.

4. CONCLUSIONS

It has been theoretically and practically proven that the uranium concentration in natural waters is one of the most important indicators for evaluating the radioactive pollution as compared with other parameters, such as concentrations of radium, radon and thorium. In addition, the activity ratio of $^{234}\text{U}/^{238}\text{U}$ in water is a sensitive indicator for evaluating the natural water pollution. Activity ratios $^{226}\text{Ra}/^{228}\text{Ra}$ and $^{230}\text{Th}/^{232}\text{Th}$ in water can only be used as the supplementary indicators.

As most of surface waters are near neutral, the exploration of uranium deposits may cause no obvious groundwater pollution by radioactive elements (U, Ra, Rn and Th). However, it is difficult to avoid the serious surface water pollution coming from the exploitation of uranium ore by a quite big open pit.

Table 2. Results of hydrochemical simplification analysis of 15 water samples in the investigated area

Sample number	Ca ⁺⁺	Mg ²⁺	K ⁺ +Na ⁺	Cl ⁻	SO ₄ ²⁻	HCO ₃ ⁻	M (TDS)	pH	Temperature(°C)	
	(mg/l)	(mg/l)	(mg/l)	(mg/l)	(mg/l)	(mg/l)	(mg/l)	(Value)	Water	Air
1	24.05	8.27	22.75	4.25	8.64	118.81	186.77	6.5	16	23.5
2	4.81	1.22	12.80	7.10	1.92	37.34	65.19	7.2	17	22
3	3.61	1.95	12.00	7.10	6.72	29.24	60.62	7.3	19	13
4	4.00	1.21	74.75	7.10	5.76	177.57	270.39	7.4	19	22
5	2.40	1.46	10.75	4.96	6.72	23.70	49.99	6.2	17.5	14
6	3.61	1.70	13.75	9.93	3.84	31.12	63.95	5.5	15	20
7	0.80	0.73	84.00	5.67	2.88	153.72	247.80	7.2	15	26
8	1.60	1.44	12.00	7.45	3.84	23.79	50.12	6.4	15.5	13
9	36.07	3.13	27.00	5.67	10.56	168.74	251.17	7.0	12.5	18
10	28.09	7.68	27.25	4.96	6.72	173.96	248.66	7.2	12.5	24.5
11	1.60	1.44	11.75	5.67	8.64	20.14	49.24	5.4	13	12
12	96.99	13.13	0	0	34.56	0	144.68	5.2	12	11
13	1.60	1.44	9.25	4.96	4.80	20.14	42.19	6.0	13	12
14	1.20	1.68	10.00	5.67	3.84	21.96	44.35	6.0	19	15
15	2.80	2.40	21.00	24.11	6.72	21.96	78.99	5.8	20	16

Table 3. Radioactive element concentration of 15 water samples in the investigated area

Sample number	type of water sample	U	Th	Ra	Rn
		(×10 ⁻⁷) (g/l)	(×10 ⁻⁷) (g/l)	(×10 ⁻⁷) (g/l)	(Bq/l)
1	Borehole water	10.40	0.011	0.28	175.49
2	Brooklet water	6.50	0.204	0.10	2.42
3	Brooklet water	1.56	2.979	0.34	3.06
4	Borehole water	0.52	0.010	0.11	446.78
5	Brooklet water	0.26	2.750	1.00	3.19
6	River water	0.26	4.721	0.29	3.68
7	Borehole water	14.30	11.460	1.29	330.33
8	Brooklet water	0.52	0.071	9.25	3.58
9	Borehole water	14.30	47.020	0.95	320.14
10	Borehole water	59.80	5.270	2.44	803.45
11	Brooklet water	2.60	0.198	0.15	13.59
12	Brooklet water	7800.00	71.270	170.00	512.99
13	Brooklet water	0.26	4.580	1.07	4.63
14	Brooklet water	130.00	7.998	2.34	2.88
15	River water	130.00	4.354	0.29	2.26

Table 4. Radioactive isotope ratio and stable isotope composition of 15 water samples in the investigated area

Sample number	Type of water sample	Radioactive isotope ratio		Stable isotope composition(‰)	
		$^{234}\text{U}/^{238}\text{U}$	$^{230}\text{Th}/^{232}\text{Th}$	$\delta^2\text{H}(-\text{V-SMOW})$	$\delta^{18}\text{O}(-\text{V-SMOW})$
1	Borehole Water	1.83	549.4	-37.0	-7.02
2	Brooklet Water	1.16	85.9	-30.4	-6.38
3	Brooklet Water	1.45	6.05	-29.2	-6.33
4	Borehole Water	1.54	1547.9	-40.4	-7.08
5	Brooklet Water	1.03	7.45	-32.6	-6.90
6	River Water	1.33	0.65	-36.0	-6.07
7	Borehole Water	1.07	0.33	-48.7	-8.66
8	Brooklet Water	0.28	262.4	-41.2	-7.25
9	Borehole Water	0.98	7.17	-40.7	-7.42
10	Borehole Water	1.14	5.71	-37.2	-7.19
11	Brooklet Water	0.72	119.0	-46.3	-7.09
12	Brooklet Water	0.99	38.6	-40.4	-6.47
13	Brooklet Water	1.08	9.97	-47.6	-7.31
14	Brooklet Water	0.98	7.70	-50.7	-6.80
15	River Water	0.90	0.69	-37.6	-5.88

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