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In-situ gamma-ray survey of rare-earth tailings dams – A case study in Baotou and Bayan Obo Districts, China

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ABSTRACT

An in-situ gamma-ray spectrometer survey with a scintillation detector of NaI(Tl) (Φ75 mm × 75 mm) was carried out in the Baotou and Bayan Obo Districts in order to estimate the levels of natural radionuclides near rare-earth (RE) tailings dams. In the RE tailings dam of Baotou, the mean concentrations of ^{238}U and ^{232}Th were 3.0 ± 1.0 mg/kg (range: 1.9–4.6 mg/kg) and 321 ± 31 mg/kg (range: 294–355 mg/kg), respectively. In the Bayan Obo tailings dam, the mean concentrations of ^{238}U and ^{232}Th were 5.7 ± 0.5 mg/kg (range: 5.3–6.1 mg/kg) and 276 ± 0.5 mg/kg (range: 275.5–276.3 mg/kg), respectively. The average ^{232}Th concentrations in the mining areas of the Bayan Obo Mine and the living areas of the Bayan Obo Town were 18.7 ± 7.5 and 26.2 ± 9.1 mg/kg, respectively. The ^{232}Th concentration recorded in the tailings dams was much higher than the global average (7.44 mg/kg). Our investigation shows that the ^{232}Th concentration in the tailings in the Baotou dam was 34.6 times greater than that in the local soil (in Guyang County); the average concentrations of ^{232}Th in the soil in the Baotou District and Bayan Obo Districts were about 1.35 and 2.82 times greater, respectively, than that in the soil in Guyang County. Based on our results, the highest estimated effective dose due to gamma irradiation was 1.15 mSv per year, estimated from the data observed in the Baotou tailings dams. The results of this preliminary study indicate the potential importance of radioactivity in RE tailings dams and that remedial measures may be required.

1. Introduction

China owns the largest deposit of rare earth (RE) RE resources in the world. In the well-known RE 'capital' of the world – Bayan Obo, the RE mineral industrial reserves account for more than 90% of the total reserves of China and with as many as 170 types of minerals and 71 elements having been identified (IGCAS, 1988; Smith et al., 2015; Yang et al., 2011; Zhang, 2004).

The Bayan Obo Fe–RE–Nb deposit is located in Inner Mongolia and was discovered in 1927. The RE mineralization is mainly concentrated in a dolomite unit (H8) of the Mesoproterozoic Bayan Obo Group. Dolomite in the main ore-bearing unit (H8f) occurs with magnetite, hematite, monazite, bastnaesite, and parisite, and so on. Fine-grained monazite occurs as fracture fillings in dolomite (Lai and Yang, 2013; Yang et al., 2004; Yang et al., 2011). The major value elements include Fe, light REs, Nb and Th. The RE resources in high grade reserves have been estimated at 48 mt at 6 wt.% RE_2O_3 (Drew et al., 1990); these are hosted in dolomite marble or in complex "banded ores". Lower grade resources hosted in massive magnetite and hematite have been estimated at 750 mt at 4.1 wt.% RE_2O_3 (Chao et al., 1997; Fan et al., 2003; Smith et al., 2015).

Baotou Iron & Steel (Group) Co. Ltd. (BIS), located at western Baotou and established in 1950s, is an important iron and steel industrial base and the largest RE industrial base in China. The Bayan Obo Mine, which belongs to BIS, is located in the north of Baotou City, approximately 150 km from the urban area of Baotou. Since 1950s the iron ores mined from the Bayan Obo open pit have been transported to Baotou by railway for the production of iron and steel because there is no water source in Bayan Obo areas.

The dressing plant of BIS has become one of the world first

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large-scale processing factories, with processing ore of 12,000,000 t per year. In the dressing plant, “weak magnetic separation – strong magnetic separation – reverse flotation” and “magnetic separation-reverse flotation” have been developed and used for iron oxide ore and magnetite ore, respectively. After extracting Fe and Nb, the tailings are directly discharged into the tailings dam at Baotou via special pipelines. Many valuable elements in the tailings have not been recovered because of complexity of mineral compositions in the Bayan Obo ores (Zhang, 2004).

From 1958 to 2004, about 12.5 million tons of RE resources have been produced from Bayan Obo ore, of which about 2 million tons is lost in the process of mining, smelting and storage (a loss rate of about 15%). The RE recovery process captures approximately 1.2 million tons of RE (with a utilization rate of <10%). The remaining 9.3 million tons of tailings are discharged into the Baotou tailing dam (Xu and Shi, 2005).

Radioactive contamination caused by the activities at Bayan Obo mine and Baotou tailings dam has been an issue of concern for the Chinese government since the 1980s. An epidemiological study on the possible combined effects of thorium dioxide (ThO_2) and Th-containing RE iron ore dust was conducted out in the Bayan Obo Mine from 1983 to 2001. In this investigation, the average ^{232}Th lung burden, which was calculated using 1270 measurements taken from 751 miners exposed to dust, was 1.58 Bq and whereas the maximum value was 11.1Bq. These values were obviously related to the conditions in which the miners worked (Chen et al., 1985, 1996; Chen and Cheng, 1998; Cheng and Chen, 2007).

The evaluation of environmental ionizing radiation dose rate has become a major area of research. In Baotou, the outdoor air absorbed dose rate is not high. The background value was reported as 64.2 ± 13.2 nGy/h, with a range of 30.9–152 nGy/h. However, in the Baotou dam the average dose rate was 400 ± 160 nGy/h, with a range of 53.4–2430 nGy/h. In Bayan Obo Town, the average dose rate varied from 59.6 to 181 nGy/h with a mean of 100 ± 19 nGy/h; in the mining areas of Bayan Obo, the maximum value was 1350 nGy/h with an average of 160 ± 120 nGy/h (Wang et al., 2001). The approximate levels of dose rate in Baotou and in Bayan Obo were reported (Li et al., 1998; Miao and Li, 1998; Miao et al., 2001; Bai et al., 2001). In addition, Liu et al. (2010) reported indoor dose rate levels in Baotou and in the miners' working places in Bayan Obo Mine: the average dose rate was 150 nGy/h with a range of 120–283 nGy/h.

In recent years, several researchers have studied Th contamination in the soil near the Baotou dam and Bayan Obo Mine. The Th average concentration in the surface soil samples collected from Bayan Obo Mine was reported as 12.8 mg/kg with a range from 3.43 to 59.1 mg/kg. Only two samples, which were collected from locations near the tailings, had higher Th concentrations compared with the samples collected from the background areas (Li et al., 2014a). The average concentrations for Th in the soil (with sampling depths from 0 to 40 cm) collected at the locations less than 1 km from the tailings dam were 120 ± 140 Bq/kg and 57 ± 23 Bq/kg at distances larger than 1 km from the tailings dam, with ranges of 33–483 Bq/kg and 39–89 Bq/kg respectively. The Th activity concentration in the tailings powder was as high as 520 Bq/kg (Wang et al., 2009). However, the ^{232}Th activity concentration was as low as 33.5–59.7 Bq/kg in the newly constructed tailings dam in the West Ore Mine in Bayan Obo (Dong, 2013).

The dose rate levels in Baotou are mainly based on the aforementioned data, but very few studies have investigated ^{232}Th activity concentration in the soil near the tailings dams and the Bayan Obo Mine.

In order to identify radioactive contamination sources and their distribution status in the Baotou dam and Bayan Obo Mine areas, a survey by airborne gamma-ray spectrometry was carried out in 2005–2006; it covered a total area of 1900 km^2, with 644 km^2 situated in the Bayan Obo Mine area and 1275 km^2 in the Baotou urban area. The interval of the measured lines was 500 m but this was altered to 250 m above the tailings and mining areas. Wu et al. (2011) reported that the contaminated areas of the tailings dams, open-pit mining and slag were delineated based on the spectrum data. The estimated dose rate near the Baotou tailings dam was between 650 and 1320 nGy/h; in the Bayan Obo Mine, the dose rates ranged from 450 to 800 nGy/h (Wu et al., 2011). However, the activity concentrations of ^{232}Th in soil/rock or tailings were not reported in his paper.

Therefore, by using in-situ gamma ray spectrometry in Baotou City we carried out a preliminary survey with the following aims:

(1) To identify the properties and distribution of radioactive contamination sources based on the measured ^{238}U and ^{232}Th concentrations, i.e., to understand the level and distribution of U and Th.
(2) To identify the level and distribution of ^{232}Th in the dam and the regions near the dam.
(3) To investigate some hot spots caused by RE processing enterprises, and to provide an example for the application of radioactive supervision in the production and processing of mineral products.

2. Materials and methods

2.1. Study area

The survey area was located near Baotou City of Inner Mongolia, northern China, at 41°20′~42°40′N, 109°50′~111°25′E. Baotou City has a total area of 27,768 km^2 and population of 2.735 million. It has a semi-arid temperate continental monsoon climate.

In the Bayan Obo deposit, the exposed strata are mainly clastic rocks of the Mesoproterozoic Bayan Obo group, shale, carbonate sedimentary formation. The Bayan Obo giant RE deposit, hosted in the massive dolomite marble in the Bayan Obo Group, occurs in one of the syncline ores. To the north of the ore body, a complete sequence of the Bayan Obo Group is exposed in the Kuangou anticline, which is developed on the Paleoproterozoic basement rocks with a distinct angular unconformity. The ore-hosting dolomite marble, which is covered by K-rich slate (H9 term), was generally considered a component of the Bayan Obo Group. It is known as H8 term. The low grade clastic sequences of the Bayan Obo Group represent the sedimentary units deposited within the Bayan Obo marginal rift, which correlates with the Mesoproterozoic continental breakup event of the North China Craton.

The Bayan Obo RE deposit is composed of three RE ore bodies: Main Ore body, East Ore body, and West Ore body (Fig.1a). The Main Ore body and East Ore body are distributed in between the boundary of ore-hosting dolomite marble and Bayan Obo group K-rich slate. The West Ore body, which includes many small ore bodies, occurs mainly in the massive dolomite marble. The Main Ore body and East Ore body occur as large lenses. The ores are distributed along a west-east striking belt (Fig. 1). From south to north, they are defined as four groups, namely the riebeckite type, aegirine type, massive type, and banded type (Yang et al., 2011).

After the extraction of Fe and Nb, the tailings are directly discharged into the tailings dam through special pipelines. Large quantities of RE and Nb tailings are discarded during the processing of Bayan Obo ore, which results in a low utilization ratio (10%) of the RE in Bayan Obo ore and even a complete waste of the Nb (Ma et al., 2009).

Major mineral compositions of the Bayan Obo tailings are listed

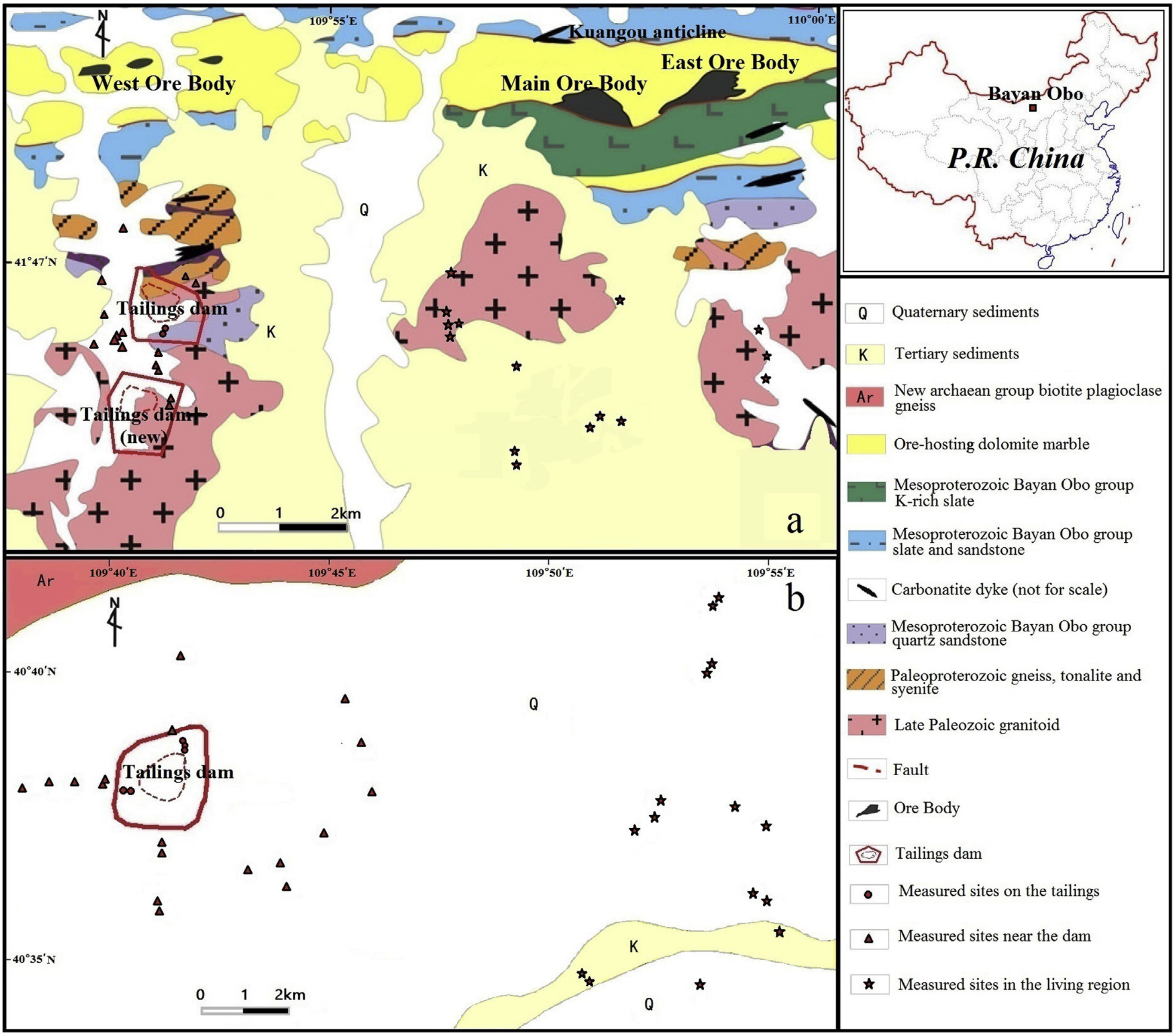

Fig. 1. Geological sketch map of the Bayan Obo area. Modified after (Yang et al., 2011).

in Table 1. The tailings with a particle size from 245 to 104 μm accounted for 30.3% of the composition, whereas those with particle sizes from 104 to 44 μm accounted for 69.7%, respectively (Li et al., 2014b).

Table 1
Major mineral compositions in the tailings (Li et al., 2014b).

Composition	Concentration (%)
Fluorite	20.62
Magnetite and hematite	19.47
Amphibole, pyroxene	14.75
Carbonatite	11.26
Bastnaesite	6.83
Quartz, feldspar	6.38
Biotite	6.07
Apatite	3.76
Monazite	3.56
Barite	2.04
Pyrite	1.95
Others	3.41

Therefore, in order to investigate the influence of the tailings dams on the environment, two district were chosen. In Baotou District, 38 sites were measured (Fig. 2a); in Bayan Obo District, 32 sites were measured (Fig. 2b).

2.2. Instruments and equipment

A portable gamma-ray spectrometer (Model DigiDART) with a NaI(Tl) (Φ75 mm × 75 mm) scintillation detector (1024 Channels) was employed in this research. The spectrometer was stabilized by an automatic gain control and had an energy resolution of 7.43% at 662 keV.

The stripping method as recommended by IAEA (1989) was used for determining ^{238}U, ^{232}Th and ^{40}K activity concentrations in soil and rocks. Stripping ratios are defined as the ratio of the number of counts due to a nuclide in other windows against the number of counts in the window for the nuclide. The ratios were

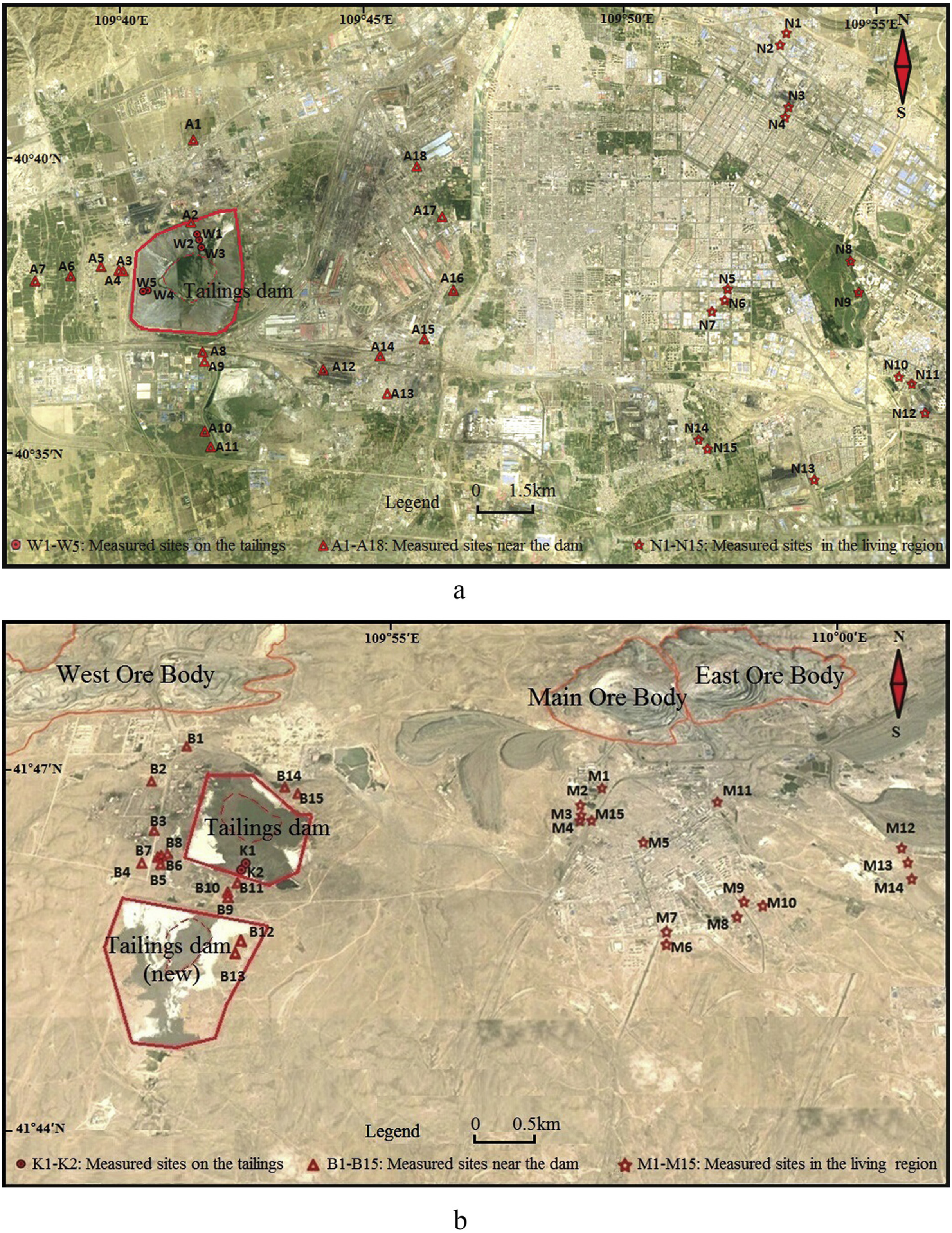

Fig. 2. Map showing the sites of in-situ gamma-ray spectrometer survey at Baotou District (a) and Bayan Obo District (b).

confirmed from five calibration pads.

In this study, the spectrometer detector was calibrated in the Radiometric Exploration Methodology Station of Nuclear Industry in Shijiazhuang, Hebei Province, China. Five calibration pads were applied to determine stripping ratios. Each pad comprised a cylinder (2.20 m in diameter 0.50 m in height) enclosed within a thin iron sheet (on the sides and bottom side), and an epoxy resin sealing (on the top side). After calculating the universal peak count rates of ^{40}K, ^{214}Bi and ^{208}Tl, with energies of 1.46 MeV, 1.76 MeV and 2.62 MeV respectively, in the characteristic gamma-ray energy range, an inverse matrix solution spectral method was used to acquire the conversion factors and the concentrations of ^{40}K, ^{238}U, and ^{232}Th via (Eq. (1)).

$$\begin{bmatrix} I_1 \\ I_2 \\ I_3 \end{bmatrix} = \begin{bmatrix} a_{11} & a_{12} & a_{13} \\ a_{21} & a_{22} & a_{23} \\ a_{31} & a_{32} & a_{33} \end{bmatrix} \begin{bmatrix} C_K \\ C_U \\ C_{Th} \end{bmatrix} \quad (1)$$

where I_1, I_2 and I_3 are the counting rate of ^{40}K, ^{214}Bi and ^{208}Tl respectively after deduction of the background counting rate, in cps; a_{11}, a_{12} … a_{33} are the conversion factor; C_K, C_U and C_{Th} are the mass concentrations of ^{40}K, ^{238}U and ^{232}Th, respectively, in the soil or rocks, in percent for ^{40}K and mg/kg for ^{238}U, and ^{232}Th. From Eq. (1), the concentration of ^{40}K and the equivalent concentration of ^{238}U and ^{232}Th can be calculated.

2.3. Field measurements

The measuring sites including the tailings dams and residential regions were designed on the GIS map. The sites were chosen as the following:

(1) The measurement points were as close as possible to the tailings dam from the east and west direction, and from the north and south direction. Where possible, some points were measured inside of the tailings dam.
(2) Additional measurements were added during a field survey if radioactive contamination was discovered.
(3) The measurement points in the living area were mainly distributed in the areas that were covered with a lawn.

The tailings dams, whether in Baotou or Bayan Obo, were naked, i.e., they were without cover materials or vegetation. In most of the central of the dam, the tailings were flooded with water; however at the edge regions the tailings dams were dry or semi-dry. Unfortunately, some of the designed points could not be measured because the sites were located in BIS, where were forbidden to entry.

In total, five sites (w1, w2, w3, w4 and w5) were measured on the tailings inside the tailings dam, with 18 points (coded as A1 to A18) around BIS and 15 (coded as N1 to N15) in the living regions in Baotou District (Fig. 2a). In the Bayan Obo District, two sites (marked as K1 and K2) were located inside of the tailings dam, with 15 (coded as B1 to B15) around the tailings dam and 15 (coded as M1 to M15) in living regions. Measurement points B12 and B13 were situated inside a new tailings dam as shown as in Fig. 2b. The overview of the Baotou tailings dam was shown as in Fig. 3.

The detector of the gamma-ray spectrometer was placed on the surface of ground, which corresponds to the calibration condition. Every measurement site was located using a portable GPS. The sites were chosen to have flat terrain surfaces with relatively homogeneous radioactivity. No high buildings were situated within 10 m around the sites. Counting time was normally 300 s at each site. The measurement procedures used in the field conformed to the Technique Regulation of Gamma-Ray Spectrometry on the Ground-DZ/T 0205–1999, which was issued by The Ministry of Land and Resources of China (Wang et al., 2000).

3. Results and discussion

3.1. Baotou District

The concentrations of ^{238}U, ^{232}Th, and ^{40}K in surface media at the measured sites in Baotou District is presented in Table 2. The results can be summarized as follows:

Fig. 3. Photograph of the Baotou tailing dam.

(1) The distribution of ^{232}Th concentration showed obvious regularity. ^{232}Th was concentrated in tailings in the Baotou Dam. The ^{232}Th concentration at the sites inside of the tailings dam ranged from 294 to 355 mg/kg, with a mean value of 321 ± 31 mg/kg. The distribution of ^{232}Th was uniformly in the tailings. Site A2 was located on the slope of the western berm of the dam and the ^{232}Th concentrations at this site reached 43.2 mg/kg. This site may be contaminated by the RE slag, resulting in ^{232}Th concentrations higher than the other sites.
(2) The concentrations of ^{238}U and ^{40}K in the tailings ranged from 1.9 to 4.6 mg/kg and 2.6 to 3.2%, with means of 3.02 ± 1.04 mg/kg and 2.9 ± 0.3%, respectively. This suggests that ^{238}U is not a contaminant element in the tailings.
(3) The ^{232}Th contamination was limited to the inside of the tailings dam, but ^{232}Th concentrations are increasing in the soil in Baotou City day by day. ^{232}Th concentrations were 13.4 mg/kg and 12.6 mg/kg in the soil near BIS and in the living area, respectively. These values are approximately 1.5 times greater than the average globe value for soil.
(4) A hot spot (Site N15) was identified in the Eastern zone of Baotou District. This site was located in the vicinity of an abandoned RE processing workshop. The measured ^{232}Th concentration on a heap of slag reached 180 mg/kg. The material components of slag need to be further analyzed. We noted that people were not working in the workshop; it appeared that this workshop had been closed.
(5) The ^{238}U concentration in the soil in Baotou City had not increased above background. It was 2.22 mg/kg in the soil near BIS and 2.88 mg/kg in the living area in Baotou.

3.2. Bayan Obo District

The in-situ gamma-ray spectrometry measurement data for the Bayan Obo District is divided into five groups based on the type of the ground medium; the results are listed in Table 3.

Table 3 and Fig. 2 show the following results:

(1) ^{232}Th is the most important radioactive contaminant element in the tailings in Bayan Obo District, but the concentration was a little lower than that recorded in Baotou District. This difference may be because the tailings are from

Table 2
Measured concentration of U, Th and K by in-situ gamma-ray spectrometry in Baotou District.

Nuclides	Statistics	On the tailings	On the berm of the dam	Near the dam	Near the living zones	Slag in a workshop
U (mg/kg)	Average	3.02	1.3	2.22	2.88	10.7
	SD	1.04		0.37	1.64	
	Min.	1.9		1.5	1.9	
	Max.	4.6		2.9	7.9	
Th (mg/kg)	Average	321	43.2	13.4	12.6	180
	SD	30.7		4.66	4.80	
	Min.	294		8.3	8.0	
	Max.	355		24.0	27.1	
K (%)	Average	2.86	1.9	1.9	2.24	1.6
	SD	0.27		0.34	0.75	
	Min.	2.6		1.0	0.9	
	Max.	3.2		2.3	3.7	

Table 3
Measured concentration of U, Th and K by in-situ gamma-ray spectrometry in Bayan Obo District.

Nuclides	Statistics	Mining zone	On the tailings	Waste disposal sites	Near the dam	Near the living zones	
						Pavement	Soil (coal[a])
U (mg/kg)	Average	1.78	5.70	0.97	1.92	2.33	2.93 (2.27)
	SD	1.13	0.57	0.21	0.58	0.47	0.15
	Min.	0.5	5.3	0.8	0.9	1.8	2.8
	Max.	2.9	6.1	1.2	2.7	2.7	3.1
Th (mg/kg)	Average	18.7	276	17.2	42.0	13.5	26.2 (60.4)
	SD	7.54	0.57	1.99	39.6	4.15	9.05
	Min.	11.2	276	16.0	16.1	9.4	20.1
	Max.	31.2	276	19.5	166	17.7	36.6
K (%)	Average	3.00	2.05	2.97	3.01	3.07	3.13 (2.99)
	SD	1.42	0.07	0.58	0.51	0.72	0.25
	Min.	0.6	2.0	2.3	2.2	2.6	2.9
	Max.	4.2	2.1	3.3	4.2	3.9	3.4

[a] The measured data on a heap of coal.

different ore. The tailings in Bayan Obo District come mainly from the West Ore Body, whereas the tailings in Baotou District come from the Main Ore Body and the East Ore Body. Thus, ^{232}Th concentrations differ according to ore type.

(2) The concentration of ^{232}Th was not high in the mining zones, including the waste disposal sites near the East Ore Body. The average ^{232}Th concentrations were 18.7 mg/kg in the mining zones and 17.2 mg/kg (Sites M12, M13, and M14) in the waste disposal sites.

(3) The concentrations of ^{232}Th varied extensively in the vicinity of the tailings dam in Bayan Obo District. ^{232}Th concentrations at the sites near the tailings dam ranged from 16.1 to 166 mg/kg, with a mean of 42 ± 40 mg/kg. The maximum ^{232}Th concentration of 166 mg/kg was observed at a polluted area of Site B8 where the delivery pipe for the tailings was placed. At sites B5, B6, and B7, the ^{232}Th concentrations were 88.6, 49.0 and 49.0 mg/kg, respectively.

(4) Seven sites were situated in the living areas of miners at Bayan Obo Town. The distribution of ^{232}Th was not uniform in the town. The average ^{232}Th concentration was 26.2 ± 9.1 mg/kg, except Site M15. The maximum ^{232}Th concentration was at Site M15, which was a heap of coal. A higher concentration of ^{232}Th, 36.6 mg/kg, was observed at Site M10, which is located near a workshop. On the cement pavement, the average ^{232}Th concentration was 13.5 ± 4.2 mg/kg.

(5) ^{238}U and ^{40}K were distributed normally and uniformly in ore, tailings and in soil.

In addition, a background investigation was conducted at two sites in Guyan County, which is situated between Baotou District and Bayan Obo District. The concentration of ^{238}U, ^{232}Th, and ^{40}K in the soil ranged from 2.20 to 3.99 mg/kg (mean: 2.55 ± 0.25 mg/kg), 7.61 to 10.7 mg/kg (mean: 9.3 ± 1.1 mg/kg) and 2.04–2.29% (mean: 2.18 ± 0.07%), respectively.

Finally, the annual effective doses equivalent (AEDE) due to ^{232}Th, ^{238}U and ^{40}K was calculated (UNSCEAR, 1993). The average AEDE was 1.03 ± 0.10 mSv/y in the tailings dam in Baotou District and 0.90 ± 0.01 mSv/y in Bayan Obo District. The average AEDE for all sites was 0.12 ± 0.05 mSv/y in these two district excluding Site B8 (0.64 mSv/y), which was contaminated by ^{232}Th from the tailings in Bayan Obo District, and Site N15 (0.54 mSv/y), which was located at a workshop in Baotou District.

Our investigation results show that the ^{232}Th concentrations in the tailings at the Baotou dam were 35 times greater than that in the soil in Guyang County; whereas the average concentrations of ^{232}Th in the soil in Baotou District and Bayan Obo District were about 1.35 and 2.82 times greater, respectively, than that in Guyang County's soil. Our results also indicate that the increase in ^{232}Th concentration observed in the soil in Baotou City is mainly attributable to mining, ore dressing, smelting, and accumulation of tailings of Bayan Obo iron ore.

4. Conclusions and suggestions

Based on the data acquired by gamma-ray spectrometry in Baotou City, we can draw the following conclusions.

(1) The most important radioactive contamination ^{232}Th element in the soil at Baotou City and its presence can be attributed to mining, dressing, smelting, and accumulation of tailings of Bayan Obo iron ore. Monazite accounts for about

3.56% of the tailings' concentration and shows a uniform distribution in the tailings. The ^{232}Th concentration in the tailings was 35 times greater than that in the soil in Guyang County, which is located between Baotou District and Bayan Obo District.

(2) The ^{232}Th concentration in the soil in Baotou District and Bayan Obo District is increasing; currently, the average ^{232}Th concentration in the soil is 1.35 times greater than that in the soil in Guyang County. This increase may originate from the deposition of the tailings dust because there is no water or plants at the edge of the tailings dam.

(3) There is a potential problem of ^{232}Th contamination near the RE mineral processing sites.

In order to decrease the secondary contamination caused by tailings dust, we suggest the following:

(1) The water seal area of the dam in Baotou District should be increased and the tailings powder moisture concentration should be $\geq$ 8% to guarantee the safety of the tailings dam, especially in the dry and cold winter season.
(2) Trees should be planted outside the berm of the tailings dam and other plants should be planted inside the dam to minimize the spreading of dust caused by the wind.
(3) The ore processing techniques used to extract thorium from the Bayan Obo ore should be improved.

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