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Original Research

Assessing Exposure Hazards and Metal Analysis Resulting from Bauxite Samples Collected from a Saudi Arabian Mine

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Abstract

Since bauxite is a rock consisting of aluminum oxide, it is significant to measure natural radionuclide concentrations for occupational health purposes. The bauxite mine is located in the city of Az Zabirah in the Qassim region in Saudi Arabia. The radionuclide concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in bauxite samples were measured using a γ-ray spectrometer NaI (Tl). The average and range values of ^{226}Ra, ^{232}Th, and ^{40}K radioactivity concentrations were 83.7±0.3 (164.2-35.2), 107.3±1.3 (199.9-48.5), and 192.0±1.1 (487.1-24.6), Bq/kg respectively. These results were compared with published global limits of ^{226}Ra, ^{232}Th, and ^{40}K by (UNSCEAR, 2000; ICRP-60). The radiation hazard parameters were also calculated and compared with the recommended levels. There are no studies for the natural radioactivity in the bauxite mine in Az Zabirah, so these results are a start to establishing a database in this location. Furthermore, x-ray fluorescence (XRF) was used to determine the concentrations of elements and their oxides for the bauxite samples.

Keywords: bauxite, natural radioactivity, radiation hazard parameters

Introduction

Natural occurring radionuclides are present in many natural resources. Human activities may enhance concentrations of radionuclides and/or enhance potential exposure to naturally occurring radioactive material (NORM). Industrial residues containing radionuclides have been receiving considerable global attention because of the large amounts of NORM-containing wastes and the potential long-term risks of long-lived radionuclides [1]. They have become concentrated and exposed not only to the environment but also to mine workers. Actually, there are a number of authors who have published papers about radiation hazard assessment resulting from phosphate, oil, or gas production [2]. Although aluminum has been widely used in industry, there are few studies focusing on radiation risk from bauxite or red mud [3]. Table 1 shows the range of radionuclides concentrations in bauxite. The activity concentrations of ^{238}U and ^{232}Th were high because ^{238}U and ^{232}Th decay series depends on geological composition.

O'Connor [4-5] measured natural activity concentrations in bauxite, solid residues, and red mud resulting from alumina production in western Australia. ^{238}U concentration results were 120-350

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Table 1. Content of radionuclides in bauxite and red mud [9].

Radionuclide	Specific activity (Bq/Kg)	
	Bauxite	Red mud
^{238}U-^{226}Ra decay series	10-900	100-3,000
^{232}Th decay series	35-1,400	100-300
^{40}K	10-600	10-100

(bauxite), 5-200 (soil residues), and 150-600 (red mud). Furthermore, the activity concentrations of ^{232}Th were (450-1,050), (300-800) and (1,000-1,900) for bauxite, soil residues, and red mud, respectively. Radionuclide concentrations of red mud increase three times compared to the original bauxite mineral due to refined bauxite and produce alumina. The highest concentrations of uranium or thorium radionuclides are transferred to solid waste. However, little, if none, of their radionuclides are found in alumina. To reduce occupational exposure, many studies have reported that a percentage of red mud can be mixed with concrete or cement [6]. The absorbed dose of the mixed red mud should not exceed the limit determined by UNSCEAR (2000) [7]. Therefore, manufacturing operation reduces the radiation hazard parameters. Furthermore, Righi et al. [8] measured the natural radioactivity in bauxite. Their results showed that the activity concentrations of ^{238}U were between 500 and 600 Bq/ kg and from 400 to 450 Bq/ kg for ^{232}Th.

Ademola et al. [10] studied the activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K in bauxite ore in Nigeria using a NaI(Tl) detector. They concluded that the mean values of ^{226}Ra, ^{232}Th, and ^{40}K changed from 47±14 to 134±21. When high pure germanium and neutron activation methods were used for bauxite measurement, the results were 37 ±12 Bq.kg^{-1} for ^{238}U, 154 ±16 Bq.kg^{-1} for ^{232}Th, and 9.4 ±0.2 for ^{40}K [1]. The activity concentrations of bauxite samples in the Gulf of Corinth in Greece were determined using direct γ-ray spectroscopy [11]. The results showed that the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K were 150, 205, and 38, respectively. The average world activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K were 50, 50, and 500 Bq/Kg, respectively [7].

Regarding EDXRF, Yatkin et al. [12] compared the results of energy dispersive x-ray fluorescence (EDXRF) and inductively coupled plasma mass spectrometry (ICP-MS) for element concentrations in bauxite. These elements were lead (Pb), arsenic (As), nickel (Ni), cadmium (Cd), copper (Cu), chromium (Cr), and zinc (Zn). Other elements such as aluminum (Al), calcium (Ca), iron (Fe), magnesium (Mg), silicon (Si), chloride (Cl), potassium (K), sulphur (S), manganese (Mn), molybdenum (Mo), cobalt (Co), strontium (Sr), bromide (Br), titanium (Ti), tin (Sn), and antimony (Sb) were also tested. They concluded that there was good agreement in element analysis between ICP-MS and EDXRF for most elements, and the uncertainty in element concentrations was low.

The objective of the present work is to determine Al content in the bauxite mine in Az Zabirah using ARL Quant'X (Thermo Scientific Inc, USA) EDXRF and compare our results with similar studies in other countries. Furthermore, this study aims to measure the natural radioactivity levels of ^{226}Ra, ^{232}Th, and ^{40}K in the samples using γ-ray spectrometry and calculate the radiation hazard parameters such as radium equivalent activity (Raeq), external hazard index (Hex), and absorbed dose rate.

Experimental

Study Area

Az Zabirah (N27056', E43043') is located in the northeastern Qassim region (Fig. 1) [13], and it has a bauxite mine. It has a laterite profile because it is rich in iron and aluminum. It is more than 105 km into the scarp face of an early Cretaceous feature. The estimated total reserves are 101.8 million tons. Of course, the laterite development could be older than Oligocene. The thickness of bauxite is approximately 8.5 m described [13]. Az Zabirah is in the cuesta region of central Saudi Arabia.

Sampling and Sample Preparation

Forty samples of the bauxite were collected from Az Ziberah in the Qassim region. The samples were ground and sieved using a standard set of sieves. Weighed samples were put in polyethylene bottles of 350 cm^3 volume. The bottles were completely sealed for more than one month to allow radioactive equilibrium to be reached. This step was necessary to ensure that radon was removed from the volume [14-16]. Each powdered sample was homogenized using an electric shaker [17].

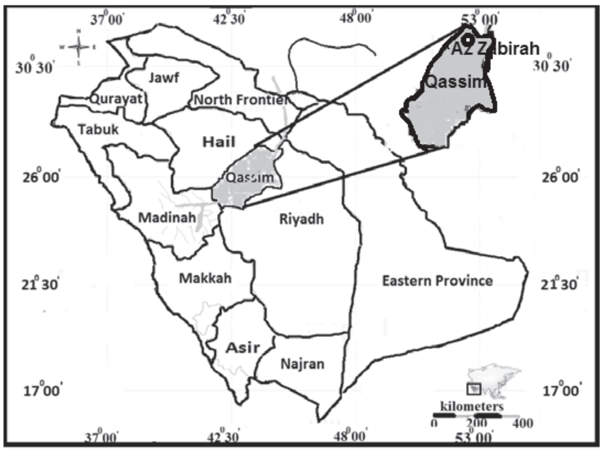

Fig. 1. Location of Az Zabirah in Saudi Arabia.

γ-ray Spectrometer *NaI(Tl)*

A NaI(Tl) detector was used to measure natural radionuclide activity. It was calibrated using a known source such as ^{60}Co and ^{137}Cs point sources. In order to calculate radionuclide activity, the concentration for each gamma-ray photo-peak relies on the secular equilibrium between parents and daughters in the samples:

$$A = \frac{N_P \times 100}{e \times \eta \times m} \quad (1)$$

…where Np is the net count rate (cps), e is the abundance of the γ-peak in a radionuclide, m is the mass of sample, and η is the measured efficiency for each gamma-ray peak observed for the same number of channels.

The count rate was subtracted from an empty polystyrene container using the same manner as measured samples to obtain the net count rate. The values of e and η for each isotope used to calculate the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K are shown in Table 2. The measuring time for gamma-ray spectra was 43,200 s [15, 18-22].

X-ray Fluorescence (XRF)

We used an ARL Quant'X (EDXRF) manufactured by Thermo Scientific Inc, USA for elemental analysis. It can analyze elements ranging from ^{22}Na to heavy elements such as ^{238}U. A material can be excited when it collides with high-energy x-rays. The anode material of an x-ray tube consists of rhodium (whose symbol is Rh and atomic number 45), and the voltage changes between 4 and 50 kV. The detector is made from semiconductor Si(Li) material. It is a cylindrical shape with 15 mm^2 cylindrical area and 3.5 mm cylindrical depth. Table 3 shows a list of filters used to decrease the x-ray energy to the correct excitation bandwidth of the elements to be analyzed [12].

Table 3. Operational parameters of EDXRF.

Beam filter	Beam filter	Live time (sec)	Element
Cu thick	50	240	Sn, Sb
Cu thin	50	800	Mo, Cd
Pd thick	30	1,600	As, Br, Sr, Pb
Pd medium	20	960	Cu, Zn
Pd thin	16	600	Fe, Co, Ni, Mn
Aluminum	12	1,000	Ti, V, Cr
Cellulose	8	200	S, Cl, K, Ca
No filter	4	100	Mg, Al, Si

Results and Discussion

Activity Concentrations of ^{226}Ra, ^{232}Th, and ^{40}K

The activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in the bauxite samples in Az Zabirah are shown in Table 4. According to the recommended reference level [7], the activity concentration should be less than 50, 50, and 500 Bq/kg for ^{226}Ra,^{232}Th, and ^{40}K, respectively. The average values of ^{226}Ra and ^{232}Th in the bauxite samples were higher than the recommended reference level (Table 4). However, the activity concentrations of ^{40}K were lower than the recommended reference level [7]. Table 5 shows the comparison between the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in the bauxite samples in this study with those by other investigations in different locations around the world. The average value of ^{226}Ra in this study was higher than the average value of ^{226}Ra measured in India, Guinean, the Gulf of Itea in Greece, and Brazil. However, our result was lower than for Greece, Antikyra Bay, Hungary, China, and Turkey. Regarding the result of ^{232}Th, our result was lower than all countries. However, ^{40}K was the highest value. Figure 2 illustrates the correlations between ^{226}Ra, ^{232}Th, and ^{40}K. We noted good correlation between ^{226}Ra and ^{232}Th and between ^{226}Ra and ^{40}K, with correlation coefficients of 0.99 and 0.95, respectively.

Table 2. Gamma rays and their related isotopes used to calculate the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K.

Nuclide	Isotopes	Energy (KeV)	Measured efficiency for each gamma-ray peak (η) (%)	Abundance of γ-peak in a radionuclide (e)
^{226}Ra	^{214}Pb	351.90	37	0.21
	^{214}Bi	609.30	46	0.12
	^{214}Bi	1,120.30	15	0.08
	^{214}Bi	1,764.5	15.9	0.067
^{232}Th	^{228}Ac	911.10	29	0.09
	^{212}Pb	238.60	44	0.25
^{40}K	^{40}K	1,460.00	11	0.07

Table 4. Activity concentrations (^{226}Ra, ^{232}Th, and ^{40}K) in the bauxite samples in the study area.

Sample number	Activity concentrations		
	^{226}Ra	^{232}Th	^{40}k
1	37.2	54.1	30.0
2	35.4	52.2	30.1
3	35.4	50.7	24.6
4	42.3	58.9	35.0
5	49.5	70.1	46.8
6	45.9	62.6	44.9
7	53.7	73.5	51.1
8	52.1	72.5	50.2
9	45.1	64.6	59.0
10	77.9	112.9	96.4
11	61.4	84.6	52.6
12	65.6	95.4	71.0
13	61.3	86.2	77.8
14	60.7	84.5	67.3
15	51.2	71.0	62.8
16	58.4	80.7	84.5
17	56.8	77.5	84.3
18	152.2	178.2	444.5
19	139.4	166.1	419.5
20	159.4	199.9	443.7
21	148.4	178.1	423.2
23	164.2	197.2	487.1
24	161.2	198.1	460.6
25	103.3	127.6	298.2
26	101.9	124.5	288.9
27	40.6	54.9	74.1
28	51.9	67.2	92.6
29	35.3	48.5	61.8
30	54.5	75.6	96.2
31	54.8	74.0	101.8
32	161.5	198.4	461.3
33	104.1	128.4	168.4
34	100.7	123.9	292.6
35	99.1	120.9	283.6
36	81.2	97.6	233.6
37	86.4	105.1	245.1
38	88.1	106.9	286.1
39	139.0	169.3	409.9
40	148.0	191.3	446.0
Average	83.7	107.3	192.0

Radiation hazard parameters can be estimated using indices such as radium equivalent, absorbed dose, and the annual effective dose. The distribution of ^{226}Ra, ^{232}Th, and ^{40}K in samples are not uniform. To obtain the same gamma dose rate, the activity concentration from the three radionuclides are assumed to be 370 Bq·kg^{-1} from ^{226}R, 259 Bq·kg^{-1} from ^{232}Th, and 4810 Bq·kg^{-1} from ^{40}K. This is the definition of radium equivalent and is given as Equation 2:

$$Ra_{eq} = A_{Ra} + 1.43\times A_{Th} + 0.077\times A_{k} \quad (2)$$

…where A_{Ra}, A_{Th}, and A_{K} are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in Bq·kg^{-1}, respectively. Regarding radiological hazard, the maximum Raeq in the bauxite samples must be less than 370 Bq kg^{-1} for safe use. The calculated Raeq results obtained from bauxite samples are presented in Fig. 3a). The radium equivalent results ranged from 109.4±10 to 483.8 ±34 Bq·kg^{-1}, and mean value was 251.9. The average value was in the safety recommended limit. These results were comparable with the results obtained from bauxite in Nigeria (189-262 Bq·kg^{-1} with mean value 222 Bq·kg^{-1}) [17]. Furthermore, the absorbed dose rate in air at 1 m above the ground surface was calculated at different sampling points using Equation 3:

$$D_{R}\ (nG\ h^{-1}) = 0.427A_{Ra} + 0.623A_{Th} + 0.043A_{K} \quad (3)$$

The values varied from 48.0 to 215.3 nGy/h and the average value was 111.5 nGy/h in the Az Zabirah bauxite mine as shown in Fig. 3a). The average obtained value calculated based on Equation (3) was higher than the worldwide average value of 60.0 nGy/h [7]. The average effective dose value at which populations are exposed to gamma rays in the Az Zabirah mine was 0.14 mSv/year. The obtained average value was lower than the recommended level (1.0 mSv/year) [7]. The calculation value of the absorbed dose rate in air at 1 m above ground level and the annual effective dose to which a population may likely be exposed at different sampling

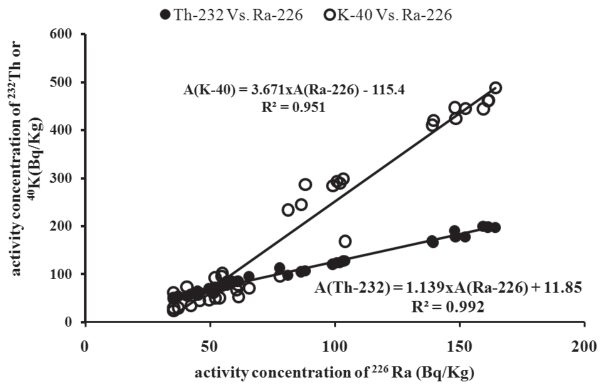

Fig. 2. Correlations between ^{226}Ra and ^{232}Th (●) and ^{226}Ra and ^{40}K (○) in the bauxite samples in Qassim Province, Saudi Arabia.

Table 5. Comparison of natural radioactivity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in the bauxite samples between the present study and published papers in other countries around the world.

Country	Description	Activity concentration (Bqkg^{-1})			References
		^{226}Ra	^{232}Th	^{40}K	
Saudi Arabia, Az Zabirah	bauxite	83.7	107.3	192.0	Present study
Guinea	Bauxite	51.2	227	-----	[23]
India	Bauxite	70.2	499	----	[23]
Brazil	Bauxite	64	154	9.4	[1]
Greece, Gulf of Itea	Bauxite	72.8	185	43	[24]
Greece, Antikyra Bay	Bauxite	150	205	28.3	[11]
Hungary	Bauxite	419.0	256.0	47.0	[25]
China	Bauxite	370.0	400.0	63.0	[8]
Turkey	Bauxite	15.5-405	19.9-414	12.8-111.6	[26]

points in these study sites are shown in Fig. 4. The hazard index parameters calculated to assess the level of risk to which the population may be exposed to the terrestrial radiation from bauxite are shown in Fig. 3b). The obtained average values of the external hazard index (Hex) and the internal hazard index (Hin) were 0.7 and 0.9, respectively. The obtained average values of both hazard indices were lower than the recommended level [7].

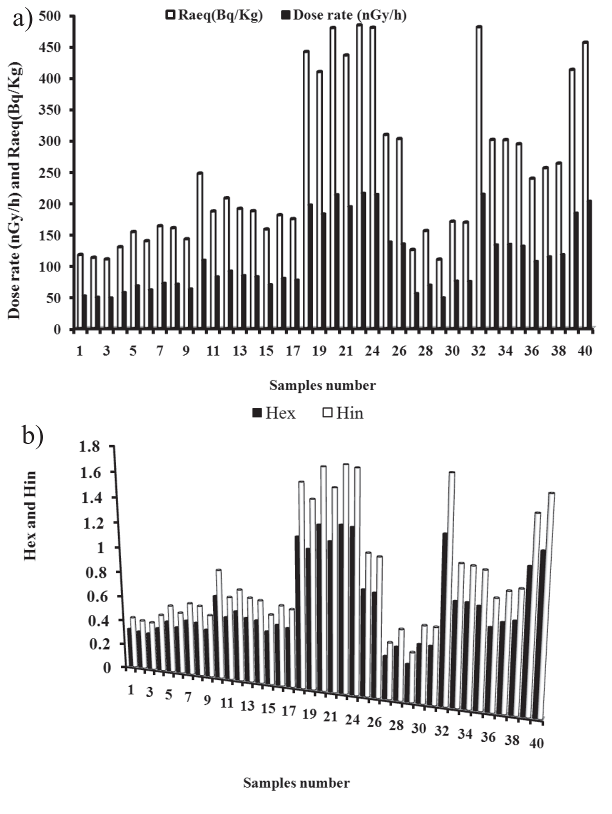

Fig. 3. a) Air-absorbed dose rates and radium equivalent calculated for bauxite samples collected from Az Zabirah in Qassim Province and b) calculated hazard index parameters, external index (Hex), and internal index (Hin) collected from the mine.

Chemical Analysis by EDXRF

Energy dispersive x-ray fluorescence analysis (ARL Quant'X EDXRF) was used to determine major and minor element concentrations and their oxides in the bauxite samples as shown in Table 6. Aluminum oxide (Al_2O_3) was the highest value, ranging between 63.3% and 40%, and the average percentage value was 56.7%. This result agrees with other studies [27-28]. Impurities, humidity, and location from which the bauxite was mined have a significant effect on the element's composition [3].

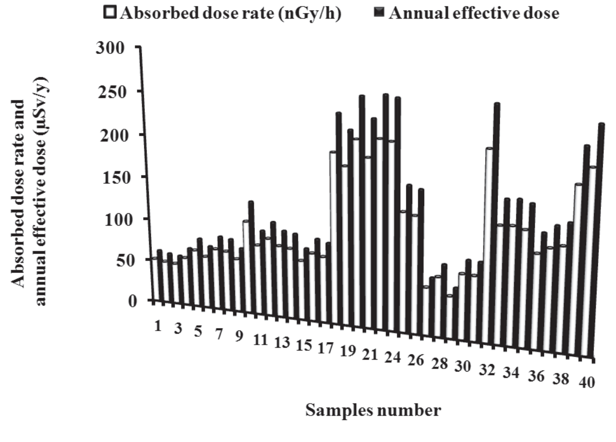

Fig. 4. Air-absorbed dose rates and annual effective doses calculated for the bauxite mine samples collected from Az Zabirah in Qassim Province.

Table 6. Chemical composition of bauxite samples collected from Az Zabirah mine in Qassim Province, Saudi Arabia.

Sample number	Element oxide (%)												
	Al_2O_3	SiO_2	TiO_2	Fe_2O_3	Na_2O	MgO	SO_3	CaO	ZrO_2	SrO	Cr_2O_3	MnO	Nb_2O_5
1	40,1	39.8	4.5	2.6	----	7.7	3.0	1.6	0.4	0.02	0.02	0.01	0.03
2	40.8	38.1	4.7	3.0	----	8.2	3.1	1.7	0.4	0.03	0.02	0.02	0.03
3	58.7	26.1	5.2	6.1	---	---	2.3	0.7	0.5	0.3	0.06	0.04	0.04
4	63.0	25.0	4.3	4.8	---	1.9	1.0	0.5	0.4	0.1	---	0.03	0.03
5	42.1	31.0	2.9	2.1	11.2	7.7	1.6	1.0	0.3	0.05	-----	0.01	0.02
6	27.4	25.9	3.0	1.8	32.6	5.4	2.3	1.3	0.3	0.02	-----	---	0.01
7	57.0	24.8	5.0	7.1	---	2.0	2.5	0.6	0.6	0.3	----	---	0.05
8	63.3	22.2	6.7	4.1	-----	-----	----	2.5	0.9	0.07	0.05	0.03	0.04
9	62.3	25.8	4.6	4.5	----	1.7	----	0.4	0.5	0.1	----	0.02	0.03
10	58.6	20.3	5.9	3.2	7.0	1.8	----	1.9	0.8	0.05	-----	0.02	0.03
11	55.5	26.0	4.1	3.7	4.8	1.7	2.1	1.4	0.4	0.1	0.04	0.02	0.03
12	59.0	22.3	3.6	3.7	7.8	1.6	1.0	0.4	0.3	0.08	0.04	0.02	0.02
13	57.5	27.5	4.6	4.0	----	2.0	----	1.5	0.5	0.15	0.04	0.03	0.03
14	56.2	23.9	4.2	4.6	6.6	1.7	1.4	0.6	0.4	0.2	0.05	0.02	0.03
16	61.6	25.1	4.9	5.5	----	----	1.7	0.5	0.4	0.2	0.05	0.03	0.04
17	56.6	25.2	3.9	3.0	6.1	1.8	1.3	1.4	0.5	0.1	0.04	0.03	-----
18	61.9	24.7	4.4	5.1	----	1.7	1.1	0.5	0.4	0.2	-----	0.03	0.03
19	58.0	31.0	4.3	2.9	----	1.7	1.2	0.4	0.4	0.1	0.03	----	0.03
20	60.0	28.8	3.7	4.7	----	1.9	---	0.3	0.3	0.06	----	0.03	0.02
21	61.8	23.6	4.9	3.6	----	1.9	1.5	1.7	0.6	0.3	-----	----	0.03
22	58.8	21.5	3.8	4.2	7.7	1.7	1.4	0.4	0.3	0.08	-----	0.02	0.02
23	60.7	26.5	4.7	5.3	----	----	1.6	0.6	0.4	0.2	0.04	0.03	0.03
24	60.5	28.7	4.8	3.9	----	----	----	1.4	0.6	----	----	0.03	0.04
25	40.8	35.6	3.8	2.2	5.1	8.2	2.5	1.4	0.3	0.0	0.0	----	0.0
26	58.5	26.3	4.7	6.3	----	----	2.8	0.6	0.4	0.2	----	0.0	----
27	42.1	34.1	4.0	2.4	4.6	8.3	2.4	1.7	0.3	0.0	----	0.0	0.0
28	59.0	27.1	4.9	5.4	----	----	2.2	0.6	0.5	0.2	0.0	----	0.0
29	60.4	20.1	5.3	3.2	6.1	2.1	----	1.9	0.7	----	0.0	0.0	0.0
30	60.4	23.6	3.7	3.6	6.3	1.5	----	0.3	0.3	0.1	0.0	----	0.0
31	59.8	22.2	3.8	3.4	8.0	1.8	----	0.5	0.3	0.1	----	0.0	0.0
32	42.7	32.9	3.6	3.2	14.3	----	1.9	0.9	0.3	0.1	----	----	0.0
33	61.9	24.4	6.8	3.6	----	----	----	2.3	0.9	0.1	----	0.0	0.0
34	57.7	28.3	4.2	3.7	----	1.9	2.1	1.4	0.4	0.1	----	0.0	0.0
35	61.6	27.6	3.9	2.6	----	2.0	----	1.4	0.4	0.1	----	0.0	0.0
36	55.9	24.2	3.6	2.9	8.0	1.8	1.8	1.3	0.3	0.1	----	0.0	0.0
37	56.8	29.3	4.2	4.6	----	1.9	1.8	0.7	0.4	0.2	0.0	0.0	0.0
38	58.1	29.4	4.4	4.9	----	----	1.9	0.7	0.4	0.2	0.1	0.0	0.0
39	60.5	28.4	3.7	4.4	----	2.0	----	0.6	0.3	0.1	----	0.0	0.0

Table 6. Continued.

40	58.7	21.5	3.7	4.2	8.0	1.5	1.4	0.4	0.3	0.1	0.0	0.0	0.0
Mean	56.7	26.9	4.4	4.0	3.6	2.4	1.5	1.0	0.4	0.1	0.0	0.0	0.0
Max	63.3	39.8	6.8	7.1	14.3	8.3	3.1	2.5	0.9	0.3	0.1	0.0	0.1
Min	40.1	20.1	2.9	2.1	4.6	1.5	1.0	0.3	0.3	0.0	0.0	0.0	0.0

Conclusion

In order to estimate the radiation hazards resulting from a bauxite mine, it is important to determine radionuclide concentrations (^{226}Ra, ^{232}Th, ^{40}K). The method of gamma spectrometry has been used to measure the radioactivity concentration of 40 samples collected from Az Zabirah bauxite mine in Qassim Province, Saudi Arabia. From this study, the range and average values of the concentrations of ^{226}Ra, ^{232}Th, and ^{40}K were 83.7±0.3 (164.2-35.2), 107.3±1.3 (199.9-48.5), and 192.0±1.1 (487.1-24.6) Bq/kg, respectively. Overall, the study showed that the measured values for ^{226}Ra and ^{232}Th were higher than the recommended levels. The mean value of radium equivalent and total absorbed dose rate were 251.9 Bq/kg and 111.5 nGy.h^{-1}, respectively. The world average value of radium equivalent and absorbed dose are 370 Bq.kg^{-1} and 65 nGyh^{-1}. Annual effective gamma dose was lower than the world average. The internal and external hazard parameters in Az Zabirah were lower than the safe limit values used by UNSCEAR. The study area is a zone of high radiation levels, and it is possible to produce radiation hazards to the environment as well as the human health. However, this data may provide a general background level for the area studied and may serve as a guideline for future measurement and assessment of possible radiological risks to human health in this province.

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