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Natural Radioactivity Levels and Some Elements Concentrations in Bauxite Ore Deposit of Az Zobirah Mine-Al Qassim - Saudi Arabia

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Abstract **Original Research Article**

The activity concentrations of (NORM) radionuclides ^{226}Ra, ^{232}Th and ^{40}K were determined in Bauxite of AzZobirah mine–AlQassim Saudi Arabia **using High Pure Germanium detector (HPGe)**. Bauxite samples concentrations ranged from 72.40±0.07 to 227.21±0.14, from 119.70±0.20 to 310.30 ± 0.10 and from 81.00±0.11to 119.00±0.01 and **the average were** 67.44, 171.77 and 96.94 Bq/ kg for ^{226}Ra,^{232}Th and ^{40}K **respectively.**, **respectively.** ^{226}Ra and ^{232}Th average values are higher than the world value 50 and 50 Bq/Kg, while for ^{40}K average is less than the world value 500 ^{226} Bq/ kg. ^{226}Ra concentrations are less than of ^{232}Th, this variations refer to the change and nature in the geological processes in this area such as the ^{226}Ra being more soluble in water. The radiological Radiation hazard indices (Raeq), (D), (AEDE), (H_{ex}), (H_{in}) were found higher than the world values. **By AA Analysis,** The concentration average values for nine elements (Al, Ca, Fe, K, Mg, Bi, Pb, Th, U) showed that, the major element is Al (20.50%), minor element is Fe (5.34%), trace elements Ca, K and Mg are (0.05, 0.04 and 0.43) % respectively. Bi is not detected, Pb, Th and U are trace elements with low concentration values (0.0006, 0.0043 and 0.0027) respectively. Bauxite has been most used in industry for producing aluminium. So, the level of pollution, the risk of the activity concentrations of (NORM) and the unacceptable limits levels of health hazard due to natural radioactivity in this area must be taken into consideration. The data will be saved as database to control the possible change in the environment area due to human activities in future.

Keywords: Bauxite ores, γ-ray spectrometry, Atomic Absorption Spectroscopy, bauxite residues hazards.

1. INTRODUCTION

Bauxite is a rock formed from a reddish clay material called laterite soil (a soil and a rock type rich in iron and aluminium), it is most found in tropical or subtropical regions (usually found near the surface of terrain). Bauxite is primarily comprised of aluminium oxide compounds (alumina, Al_2O_3.), silica (Silicon dioxide- SiO_2), iron oxides and titanium dioxide (TiO_2). It is a rock composed when laterite soils are leached of silica and other soluble materials in a wet tropical or subtropical climate (IDA VALETON, 1972). Bauxite does not have a specific composition. It is a mixture of hydrous aluminium oxides, aluminium hydroxides, clay minerals, and insoluble materials (quartz, hematite, magnetite, siderite, and goethite). Also, aluminum minerals in bauxite is gibbsite ($Al(OH)_3$), boehimate (AlO (OH)) (Hobart M. King 2018). Approximately 28000(Mt) million metric tons of Bauxite word resources are estimated (Allison Britt, 2017), 90% of the world bauxite production is used for making alumina ($A1_2O_3$), (SAM H *et al.*, 1936). Bauxite deposits originated from weathering or soil formation with an abundant of aluminium. Bauxite is used in the abrasives, chemical, and refractory industries. The igneous, metamorphic, or sedimentary rocks are the parent rocks of Bauxite **(Kusuma K. N.,** 2012). Bauxite refineries produce alumina (aluminium oxide), which is used to create aluminum metal. Bauxite is also used to manufacture other industrial products, such as abrasives, cement and chemicals. The most frequently-occurring radionuclides (238U- 226Ra, 232Th and 40k) and their decay progenies are found in bauxite and its processing residuals. These long-lived radioactive material (NORM) have been receiving considerable global attention because of their potential long-term risks (Cuccia V *et al.*, 2011). They have become concentrated and exposed not only to the environment but also to human workers in manufacturers, water suppliers, or mines, so the radiological impact resulting from bauxite or red mud are studied and it is important to measure the radionuclides concentrations to determine the health effect (Adel G.E., Abbady *et al.*, 2005). This study aims to measure the natural radioactivity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in the bauxite ores deposit site in Az Zabirah mine using two techniques: (γ-ray spectrometry) to calculate the radiation hazard parameters and estimate the radiological parameters risk, and (Atomic Absorption Spectroscopy) to evaluate the chemical elements concentrations. Results will help establishing a guideline for the environment protection from bauxite residues hazards.

2. MATERIAL AND METHODS

2.1. *Study area*

Mining bauxite: Bauxite is normally found near the surface of land and, it can be strip-mined economically and for environmental conservation efforts, the land is cleared before the mining by storing the topsoil , so it can be replaced after the bauxite is broken up and taken out of the mine to get the aluminium to the a refinery. When mining is completed, the topsoil is replaced and the area undergoes a restoration process (The Aluminium association). An average of 80 percent of the land mined for bauxite is returned to its native ecosystem. Topsoil from the mining site is stored so it can be replaced during the rehabilitation process. The Bauxite samples were collected from the study area which is located in the stable shelf in which the marine sedimentary rocks dominate the Arabian Platform (Fig. 1). Bauxite deposits are near Az Zabirah (27° 55′ N and 43° 41′ E) in the northern part of Saudi Arabia, 180 km to the north of Buraydah (USGS, 2014). The lower boundary of the Az Zabirah bauxite is along an angular unconformity where the Late Cretaceous rocks (Wasia and Aruma formations) overly the clastic sediments of Late Triassic, Early Middle Jurassic, and Early Cretaceous age. According to (A M Gray, 1982) and (A. C. Bowden, 1981), the bauxite profile extends as outcrops for approximately 125 km.

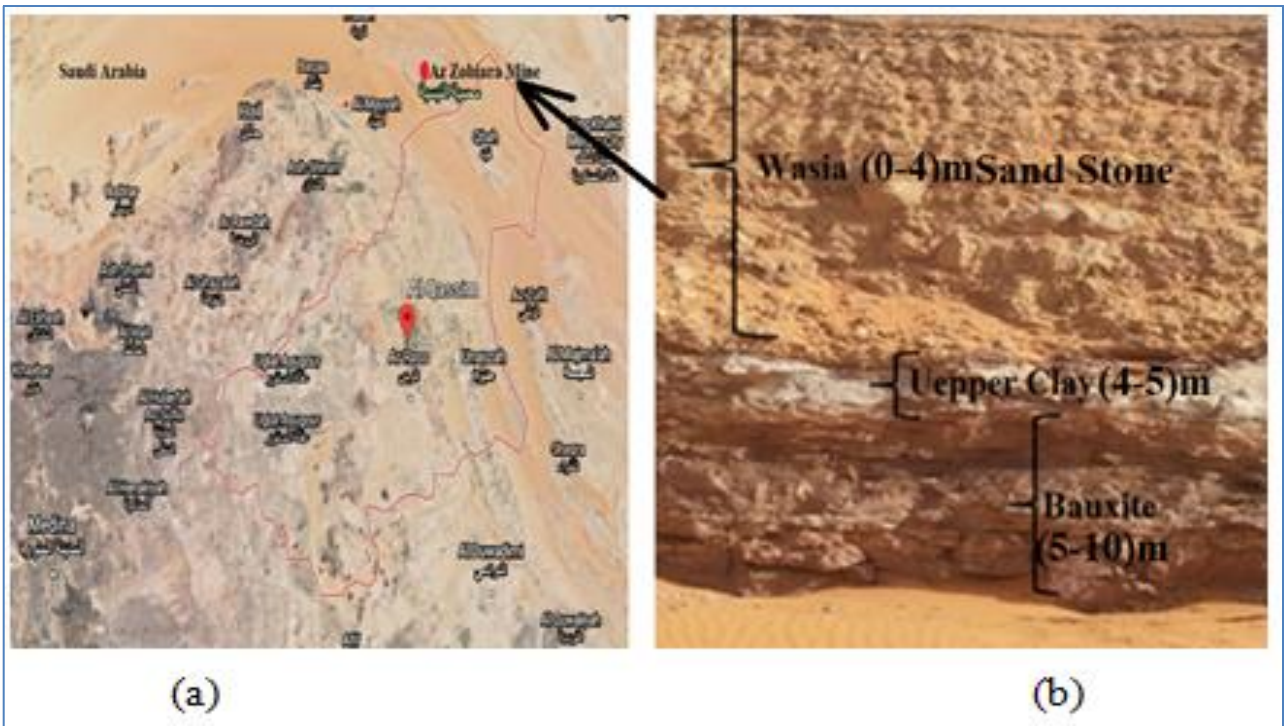

Fig-1 (a): Az Zabirah bauxite deposits (27° 55′ N and 43° 41′ E) AlQassim, Saudi Arabia, (b): Bauxite layer (5-10m) topped with a clay layer (4-5m) and Sand Stone layer (0-4m).

2.2. *Sample collection, preparations and measurement*

Bauxite samples were collected, crushed, air-dried and sieved for homogenizes, then each sample transferred to standard measuring container polyethylene Marinelli beakers. Each container was filled up by samples (0.55Kg), sealed, and kept for two months in order to achieve the radioactive equilibrium between ^{226}Ra - ^{222}Rn and their progenies. The gamma-rays of the samples were measured by high-resolution gamma spectrometer based on a coaxial P- type high-purity germanium (HPGe) detector (Canberra Model number GC2520), with efficiency of 25% and energy resolution of 2 keV FWHM for the 1332 keV line of ^{60}Co, and (16k) MCA card with software Gamma (Gennie 2000) was used for Gamma acquisition and data analysis in our nuclear lab Jeddah university. The detector is boarded inside a thick lead shield to reduce the background. Each sample was counted for 28800 s. The background was measured many times at the same conditions of the measurement. The system was calibrated for energy and efficiency (IAEA, 2018). ^{226}Ra activity concentrations were evaluated using gamma-ray lines of its related isotopes, ^{214}Pb (352 keV) and ^{214}Bi (609.31, 1120.27, 1764.49 keV). For ^{232}Th, gamma ray lines of ^{212}Pb (238. 583.1KeV) and ^{228}Ac (338.42, 911.16, 964.6, 968.97 KeV) were used to measure the activity concentrations. The activity concentrations of ^{40}K were determined by using 1460.8 keV gamma ray line.

2.3 Calculations

2.3.1 Activity concentration

The radionuclide activity concentration in Bq/kg for each gamma ray line was calculated using the equation (Amrani, D., M. Tahtat, 2001):

$$A = \frac{C}{m\,\beta\,\varepsilon} \quad (1)$$

Where: C is the net peak area of specific gamma ray energy (count per second), m is the mass of the samples in (kg), β is the transition probability of gamma-decay, ε is the detector absolute efficiency at the specific gamma-ray energy.

Exposure to radiation has been defined in terms of the radiological risk indices such as radium equivalent activity Ra_{eq} in Bq/kg which is calculated from equation (UNSCEAR. 1999):

$$Ra_{eq} = CRa + (CTh \times 1.43) + (CK \times 0.077) \quad (2)$$

Where: C_{Ra}, C_{Th} and C_K are the concentrations in Bq/kg dry weight for radium, thorium and potassium respectively.

2.3.2 Absorbed dose rate, Effective dose and external hazard index

The absorbed dose rate (nGy/h) in air at 1 m above the ground surface due to the activity concentrations of the radionuclides ^{226}Ra, ^{232}Th and ^{40}K (Bq/ kg) dry weight was calculated using the equation (UNSCEAR, 2000).

$$D\ (nGy/h) = 0.462C_{Ra} + 0.604C_{Th} + 0.0417C_K \quad (3)$$

Where: C_{Ra}, C_{Th} and C_K are the concentrations in Bq/kg for radium, thorium and potassium respectively. The absorbed doses D (nGy/h) were converted to annual effective dose equivalent D_{eff} (mSv/ y) in air was calculated by (UNSCEAR 2000) using the dose conversion factor of 0.7 Sv/Gy and the outdoor occupancy factor of 0.2 (people spend about 20% of their life outdoor), the Annual Effective Dose (in mSv/y) received by population is calculated by equation:

$$D_{eff}\ (mSv/y) = D\ (nGy/h) \times 8{,}766\ h \times 0.7(Sv/Gy) \times 0.2 \times 10^{-6} \quad (4)$$

Where: D (nG/h) is the total air absorbed dose rate in the outdoor. 8,766 h is the number of hours in 1 year. 10^{-6} is conversion factor of nano and milli. To limit the annual external gamma-ray dose to 1.5 Gy for the samples. The internal exposure to ^{222}Rn and its radioactive progenies is controlled by the internal hazard index (H_{in}) rise to the emitted gamma rays for each sample, which is calculated by Equation (5) (Veiga R., *et al*., 2006):

$$H_{in} = C_{Ra}/185 + C_{TH}/259 + C_K/4810 \quad \leq 1 \quad (5)$$

The external hazard index (H_{ex}) due to the emitted gamma rays for each sample is given by the equation (El Aassy, *et al*. 2011):

$$H_{ex} = C_{Ra}/370 + C_{Th}/259 + C_K/4810 \leq 1 \quad (6)$$

2.3.3. Element concentrations

Atomic absorption is none destructive technique, it is the process which enables to determine an element's presence within a sample and to measures its concentration (Muhammad Akhyar Farrukh, 2011). In this study, atomic absorption spectrometry (AAS) uses for concentrations measurements of the elements Al, Ca, Fe, K, Mg, Bi, Pb, Th, U in Bauxite samples.

3. RESULT AND DISCUSSIONS

3.1. *Activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K*

The calculated activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K, in the bauxite ore deposit samples in Az Zabirah were stated in Table (1). In bauxite samples, the concentrations of ^{226}Ra and ^{232}Th (Bq/ kg) range values were 72.40 ± 0.07 to 227.21 ± 0.14 and 119.70 ± 0.20 to 310.30 ± 0.10) Bq/Kg, the mean activities of ^{226}Ra and ^{232}Th, (Bq/ kg) were (67.44 and 171.77), which they were higher than the recommended reference values (50 and 50), the activity concentration of ^{40}K was $81.00 \pm 0.11$to 119.00 ± 0.01 and mean was (96.94) (Bq/ kg which is less than the recommended reference value (500) . The results show that the concentration values of ^{226}Ra are less than those of ^{232}Th in Az Zabirah. This variations refer to the change and nature in the geological processes in this area such as the ^{226}Ra being more soluble in water.

Table-1: The specific activity concentrations in Bq/kg for Bauxite Samples measured by Gamma Spectroscopy

Sa. No / Discerption.	Location		Specific activities (Bq/ kg)		
			^{226}Ra	^{232}Th	^{40}K
B1/Pink	Bauxite Samples layer, Az Zabirah Mine, Saudi Arabia	N: 27– 54 - 39.1- E:43 – 42 - 47.3	72.40±0.07	119.70±0.20	98.00±0.40
B2/Paige			79.30±0.12	136.70±0.13	109.00±0.24
B3/Brown			79.40 ± 0.01	140.30±0.13	110.00±0.30
B4/Brown			84.80 ± 0.13	184.30±0.17	86.30±0.30
B5/L. Brown			90.20±0.12	198.30±0. 12	92.10±0.36
B6/ D. Red			91.43±0.31	168.20 ±0.29	93.20±0.30
B7/ D. Red			91.76 ±0.06	235.33±030	86.10±0.30
B8/Paige			92.00±0.02	128.70±0.01	119.00±0.01
B9/ Pink			92.80 ±0.20	148.43±0.10	93.20 ±0.43
B10/ Paige			93.20±0.11	133.00±0.12	96.00±0.30 ±
B11/ Paige			93.80±0.17	144.50±0.03	87.00±0.40
B12/ Paige			97.80 ± 0.10	307.00 ± 0.13	113.00 ± 0.40
B13/D. Pink			103.30±0.12	218.30±0.10	83.00±0.40
B14/D. Pink			110.75±0.11	145.25 ±0.04	96.20 ±0.44
B15/D. Pink			161.60 ± 0.11	310.30 ± 0.10	99.50 ± 0.44
B16/D. Pink			172.75 ± 0.13	259,25 ± 0.02	107.00 ± 0.50
B17/D .Red			227.21±0.14	213.30±0.23	81.00±o.11
Range			72.40±0.07 to 227.21±0.14	119.70±0.20 to 310.30 ± 0.10	81.00±0.11to 119.00±0.01
Average			**67.44**	**171.77**	**96.94**
(UNSCEAR, 2000)			**50**	**50**	**500**

The activity concentrations of (^{226}Ra, ^{232}Th, and ^{40} K) Bq/Kg, in the bauxite ore deposit mine samples(Az Zabirah were shown in Fig.2.. The bauxite concentrations of ^{226}Ra and ^{232}Th for all samples were greater than the global values (1.35% and 3.44%) except potassium ^{40} K samples (they were much less than the global values by 0.45%). This means that the level of pollution and the risk of the activity concentrations of (NORM) in this area must be taken into consideration.

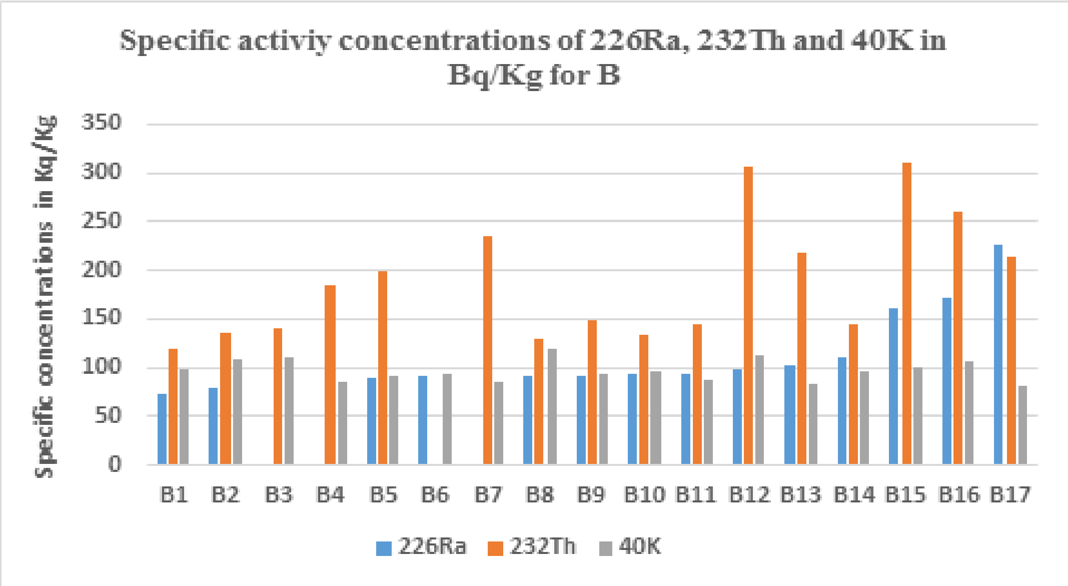

Fig-2: The specific activity concentrations of ^{226}Ra, ^{232}Th, and ^{40} K in Bq/kg for Bauxite samples in Az Zabirah Mine Sa. Arabia.

3.2. Radiation hazard indices

The radiological Radiation hazard indices (Radium Equivalent Activity (Raeq) , Absorbed Dose Rate in Air (D) , Annual Effective Dose Equivalent (AEDE), the external hazard index (H_{ex}), the internal hazard index (H_{ix}) are important to assess to population who are exposed to these radiation. The different radiation hazard indices in AzZabirah mine were calculated from the measured activity concentrations of three main radionuclides ^{226}Ra, ^{232}Th, and ^{40} K in the samples. The results of the radiological hazard were recorded in Table (2**).** For Bauxite samples, the average value for Radium equivalent (Ra_{eq}) was 653.2 Bq/Kg, this value was higher than the worldwide average value 370 Bq/Kg by 1.77 %. The obtained average value of the absorbed dose rate was 171.4 nGy/h, which was higher than the worldwide average value 65 nGy/h by 2.64%. The calculated effective dose average value Was 78.2 mSv/y, this value was higher than the safe limit of 1 mSv/y by 78.20 %. The average values of external index (Hex) and he internal hazard index (Hin) were 1.2 and 1.4, these average values slightly higher than the recommended average ≤ 1 by **(**UNSCREAN, 2000).

Table-2: The values of radium equivalent, absorbed dose, effective dose, external hazard and internal index for Bauxite Samples.

Sample No.	Radiation hazards				
	Radium equivalent Ra_{eq} (Bq/Kg)	**Absorbed dose D (nGy/h)**	**Annual effective dose D_{eff} (mSv/y)**	**External index (H_{ex})**	**Internal index (H_{in})**
B1	429.83	109.70	48.41	0.68	0.88
B2	487.05	123.71	54.32	0.77	0.98
B3	497.60	126.04	55.18	0.78	1.09
B4	625.19	139.48	66.67	0.96	1.30
B5	671.52	166.02	71.44	1.03	1.27
B6	367.15	147.70	64.81	0.92	1.16
B7	778.06	189.50	80.36	1.18	1.54
B8	478.41	124.58	56.10	0.77	1.02
B9	531.58	136.09	60.70	0.84	1.09
B10	488.37	126.78	57.28	0.79	1.04
B11	720.47	219.22	116.04	1.37	2.16
B12	992.00	239.11	98.70	1.47	1.74
B13	740.42	183.68	79.65	1.14	1.42
B14	541.22	141.97	103.00	0.88	1.18
B15	1064.60	267.84	94.92	1.66	2.09
B16	931.00	239.93	108.69	1.50	1.96
B17	759.42	233.07	113.18	1.45	2.07
B. average	**653.2**	**171.4**	**78.2**	**1.1**	**1.4**
UNSCEAR	**370**	**65**	**1**	**≤ 1**	**≤1**

The analysis shows wide distributions of natural radionuclides in the samples, which may be due to the variety of geological formations in Bauxite deposit. Also, the levels of health hazard due to natural radioactivity were found to be higher than the acceptable limits. The data will be saved as database to control the possible change in the environment area due to human activities in future.

3.3. Elements Concentrations for B in (%) and in (ppm) measured by AAS.

Table (3). lists the results of the measurements for 17 Bauxite samples for nine elements (Al, Ca, Fe, K, Mg, Bi, Pb, Th, U) by atomic absorption spectroscopy (AAS). Results show that in bauxite samples, the major element is Al %, its concentrations were ranged from 19.07 % B15 to 27.05 % B1with Average value 20.50 %. The minor element is Fe %, its concentrations were ranged from 0.51% B7 to 15.51% B14 with average value 5.34 %. Trace elements are Ca, K and Mg respectively, their concentration values were ranged from 026-0.73, 0.01-0.09 and 0.13-1.64 % and the average value 0.05, 0.04 and 0.43 % respectively. Bi is not detected, while Pb, Th and U have very low concentration values, they were ranged 0.0003-0.0012, 0.0027-0.0077 and 0.0014-0.0042 %, and the average values 0.0006, 0.0043 and 0.0027 respectively. Bauxite ore contains 15%–25 Al (S.K. Haldar, 2020), Az Zabirah Mine contains 20.50 Al %, this percentage gives a promising outcome for economically mine extracting aluminum from the this mine. Fe is minor element with average 5.34%. Ca, K, Mg are trace elements and Pb, Th and U have very low contents.

Table-3: Elements Concentrations of Al, Ca, Fe, K, Mg in (%) and of Bi, Pb, Th, U in (% ppm) for Bauxite samples measured by AAS.

Element	Al	Fe	Ca	K	Mg	Bi	Pb		Th		U	
Units	%	%	%	%	%	ppm	%	ppm	%	ppm	%	ppm
DL.	**0.02**	**0.02**	**0.02**	**0.01**	**0.01**	**1.00**	**0.0002**	**2.00**	**0.0001**	**1.00**	**0.0005**	**5.00**
B1	19.52	4.39	0.30	0.04	0.050	ND	0.0021	20.71	0.0035	33.47	0.0005	5.14
B2	19.08	4.33	0.28	0.02	0.038	ND	0.0029	29.28	0.0037	36.92	0.0003	5.33
B3	19.28	3.19	0.19	0.05	0.051	ND	0.0036	35.47	0.0077	77.03	0.0012	12.16
B4	19.14	4.02	0.36	0.03	0.049	ND	0.0020	19.05	0.0035	35.18	0.0005	5.44
B5	19.52	4.39	0.30	0.04	0.050	ND	0.0021	20.71	0.0034	33.47	0.0005	5.14
B6	23.97	8.30	0.15	0.02	0.036	ND	0.0042	41.64	0.0059	58.76	0.0009	9.50
B7	19.57	0.51	0.62	0.03	0.048	ND	0.0032	32.00	0.0043	43.22	0.0006	6.14
B8	19.08	4.33	0.28	0.02	0.038	ND	0.0030	29.28	0.0037	36.92	0.0005	5.33
B9	20.27	0.52	0.13	0.01	0.061	ND	0.0024	24.13	0.0042	42.00	0.0006	5.53
B10	24.08	14.80	1.64	0.02	0.073	ND	0.0014	24.17	0.0056	55.45	0.0010	9.57
B11	20.23	4.54	0.32	0.09	0.029	ND	0.0030	30.05	0.0027	26.56	ND	ND
B12	20.73	5.49	0.68	0.09	0.026	ND	0.0021	20.75	0.0028	27.81	ND	ND
B13	19.14	4.02	0.36	0.03	0.049	ND	0.0019	19.05	0.0035	35.18	0.0005	5.44
B14	27.05	15.51	0.80	0.02	0.069	ND	0.0041	41.47	0.0071	70.96	0.0012	12.12
B15	19.07	3.96	0.23	0.04	0.034	ND	0.0027	27.09	0.0037	37.13	0.0005	5.08
B16	19.14	4.02	0.36	0.03	0.049	ND	0.0019	19.05	0.0035	35.18	0.0005	5.44
B17	19.52	4.39	0.30	0.04	0.050	ND	0.0020	20.71	0.0035	33.47	0.0005	5.14
Reange	**19.07-27.05**	**0.51-15.51**	**0.13-1.64**	**0.01-0.09**	**026-0.73**	**ND**	**0.0014-0.0042**	**---**	**0.0027-0.0077**	**---**	**0.0003-0.0012**	**---**
Average	**20.50**	**5.34**	**0.43**	**0.04**	**0.05**	**ND**	**0.0027**	**---**	**0.0043**	**---**	**0.0006**	**---**

Table-4: Comparison of activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K Bq/Kg in bauxite samples for present study with previous study reported from countries of the world

Country	Area	activity concentrations (Bq/Kg)			References
		^{226}Ra	^{232}Th	^{40}K	
Camroon	Volcain area	14	30	103	**M. Ngachin et al. 2008**
Syria	36°5-2°12`E 32°-37° 18`N	19	24	336	**Al Marsi et al. 2006**
Turkey	7 Geographical regions of Turkey	28	33	448.5	**Turhan et al. 2012**
Portugal	Uranium Mining	200	91	-	**Carvalho et al. 2007**
Eastern Germany	Ronneburg	370	45	620	**Winkelmann et al. 2001**
Brazil	Rio Grande do Norte	29.2	47.8	704	**Malanca 1996**
Saudi Arabia	**Az Zabirah**	**67.44**	**171.77**	**96.94**	**Present Work**
Word average	-	**33**	**45**	**422**	**UNSCEER 2008**

4. CONCLUSION

In bauxite samples in Az Zabirah, the average activities of ^{226}Ra and ^{232}Th, (Bq/ kg) were higher than the recommended reference values, while the average activity of ^{40}K (244.35) was less than the recommended reference value. Also, the results show that the concentration values of ^{226}Ra were less than those of ^{232}Th. This variations refer to the change and nature in the geological processes in this area such as the ^{226}Ra being more soluble in water. The mean values of radiological Radiation hazard indices Ra_{eq} Bq/Kg, D nGy/h, AEDE mSv/y, H_{ex} and H_{ix} in Bauxite samples were higher than the worldwide values. This means that the level of pollution and the risk of the activity concentrations of (NORM) and Radiation hazard indices in this area must be taken into consideration. The data will be saved as database to control the possible change in the environment area due to human activities in future and to determine background radiation level in order to evaluate the health hazards resulting. Atomic absorption spectrometry (AAS) uses for concentrations measurements of the nine elements Al, Ca, Fe, K, Mg, Bi, Pb, Th, U in Bauxite samples. Results show that, major element is Al, minor element is Fe, and Trace elements are (Ca, K and Mg) respectively. Bi is not detected, while (Pb, Th and U) have very low concentration values. Az-Zabirah Mine contains (20.50) % Al, this percentage gives a promising outcome for economically mine extracting aluminium.

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