Elemental analysis and radiation hazards parameters of bauxite located in Saudi Arabia

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Abstract. Since Bauxite has been widely used in industry and in scientific investigations for producing Aluminum, it is important to measure the radionuclides concentrations to determine the health effect. The Bauxite mine is located in Az Zabirah city in Saudi Arabia. The concentrations of the radionuclides in the bauxite samples were measured using γ-ray spectrometer NaI (Tl). The average and range values of the concentrations of $^{226}$Ra, $^{232}$Th and $^{40}$K were 102.2 (141.1-62.7), 156.3 (202.8-102.8) and 116.8 (191.7-48.9) Bq/kg respectively. These results were compared with the reported ranges in the literature from other locations around the world. The radiation hazard parameters; radium equivalent activity, annual dose, external hazard were also calculated and compared with the recommended levels by International Commission on Radiological Protection (ICRP-60) and united nations scientific committee on the effects of atomic radiation UNSCEAR reports. There are no studies for the natural radioactivity in the bauxite mine in Az Zabirah city, so these results are a start to establishing a database in this location.

1. Introduction

Natural occurring radionuclides are present in many natural resources. Human activities may enhance the concentrations of radionuclides and/or enhance the potential of exposure to naturally occurring radioactive material (NORM). The 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 human workers in manufacturers, water suppliers, or mines. Actually there are numbers of authors studied the radiological impact resulting from phosphate production and oil or gas production [2]. However Aluminum productions have been widely used in industry and in scientific investigations (bauxite is a rock consisting of aluminum oxide) but the number of studies on the radiological impact resulting from bauxite or red mud (the waste and tail material from primary aluminum production) were low [3]. Table 1 shows the range of radioactivity concentrations in the bauxite. The activity concentration is high because of $^{238}$U and $^{232}$Th decay series, depending on the geological composition. O’Connor [4, 5] measured natural activity concentration in the bauxite, solid residues and red mud resulting from alumina production in Western Australia. $^{238}$U activity concentration results were 120 – 350 (bauxite) 5 – 200 (soil residues) and 150 – 600 (red mud). Furthermore, the activity concentrations of $^{232}$Th were (450 – 1050), (300 - 800) and (1000 –
1900) for bauxite, soil residues and red mud respectively. The radionuclide concentration of red mud increases three times compared to the original bauxite mineral due to refine bauxite and produce alumina. The most concentration of uranium or thorium radionuclides is transferred to the solid waste. However, little, if none, of the radionuclides from them are found in the alumina. To reduce the occupational exposure, many studies 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 to 500 and 600 Bq/ kg and from 400 to 450 Bq/ kg for $^{232}$Th.

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

| Radionuclide          | Specific activity (Bq/Kg) |
|-----------------------|---------------------------|
| $^{238}$U-$^{226}$Ra decay series | 10-900                    |
| $^{232}$Th decay series | 35-1400                    |
| $^{40}$K              | 10-600                     |
| Bauxite               | 100-3000                   |
| Red soil              | 100-300                    |

Ademola et al. [10] studied the activity concentration of $^{226}$Ra, $^{232}$Th and $^{40}$K using bauxite ore in Nigeria country. They used NaI(Tl) detector for measurements. They concluded that the activity concentrations were between 134 ± 21 and 47 ± 14. Using High pure germanium and neutron activation methods for bauxite measurement, the results were 37±12 Bq.kg$^{-1}$ for $^{238}$U and 154±16 Bq.kg$^{-1}$ for $^{232}$Th [1]. The activity concentrations of the bauxite samples in the Gulf of Corinth, Greece country were determined using direct γ-ray spectroscopy by Papatheodorou et al.[11]. They 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.

The present work aims to i) measure the natural radioactivity level of ($^{226}$Ra, $^{232}$Th and $^{40}$K) in samples of bauxite in Az Zabirah mine, Saudi Arabia by using γ-ray spectrometry. ii) calculate the radiological parameters such as radium equivalent activity Raeq, external hazard index Hex and absorbed dose rate which is related to the external γ-dose rate. iii) Compare the results of activity concentrations and radiation hazards with similar studies carried out in other countries and UNSCEAR data.

2. Materials and methods

2.1 Study area
Az Zabirah (N27°056', E43°043’) is located on the northeast of Buraidah in Qassim region. It is 180 km from Buraydah. It has a bauxite mine. It is indicated as laterite profile because of rich in iron and aluminum. It is over a distance of 105 km in 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 by Vincent [12]. Az Zabirah is in the cuesta region of central Saudi Arabia.

Figure 1. (a) The location map of Az Zabirah in Saudi Arabia and (b) the bauxite rocks in the location of the study area
2.2 Sampling and sample preparation
A total of 24 samples of the bauxite were collected for investigation from Az Ziberah in Qassim region. The samples were ground, homogenized 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 [13, 14, 15]. Each powdered sample was homogenized using an electric shaker [16].

2.3 The γ-ray spectrometer (NaI(Tl))
NaI(Tl) detector was used to measure the natural radionuclides activity (count rate in the environmental samples). It was calibrated using known source such as 60Co and 137Cs point sources. In order to calculate the radionuclide activity concentration (activity per unit mass) for each gamma-ray photo-peak rely on the secular equilibrium between parents and daughters in the samples, the equation 1 was used.

$$A = \frac{N_p \times 100}{e \times \eta \times m}$$  

where:

$N_p$ is the net count rate (cps)
$e$ is the abundance of the γ-peak in a radionuclide
$m$ is the mass of sample
$\eta$ 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 measuring time for gamma-ray spectra was 43200 s [14, 17, 18, 19, 20, 21].

3. Results and discussion

3.1 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 were shown in table 2. According to the recommended reference level by [7], the activity concentration should be less than 50, 50 and 500 Bq/kg for $^{226}$Ra, $^{232}$Th and $^{40}$K respectively [22]. The obtained results of $^{226}$Ra and $^{232}$Th in the bauxite samples were higher than the recommended reference level (table 2). However, the activity concentrations of $^{40}$K in the study were lower than the recommended reference level by [7]. Table 3 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 in the world. The average value of $^{226}$Ra in this study was higher than the average value of $^{226}$Ra measured in India, Guinean, Greece Gulf of Itea and Brazil. However, our result was lower than Greece, Antikyra Bay, Hungary, China and Turkey. Regarding to the result of $^{232}$Th, our result was lower than all countries except Brazil. However, $^{40}$K was the highest value. Figure 2 illustrates the correlations between $^{226}$Ra, $^{232}$Th and $^{40}$K. It is noted that a good correlation between $^{226}$Ra and $^{232}$Th and between $^{226}$Ra and $^{40}$K with correlation coefficients of 0.875 and 0.647, respectively.
Table 2. Activity concentrations ($^{226}\text{Ra}$, $^{232}\text{Th}$ and $^{40}\text{K}$) in the bauxite samples in the study area.

| Sample No | $^{226}\text{Ra}$ | $^{232}\text{Th}$ | $^{40}\text{K}$ |
|-----------|-------------------|-------------------|-----------------|
| 1         | 65.9              | 109.7             | 59.6            |
| 2         | 62.7              | 105.8             | 59.9            |
| 3         | 62.9              | 102.8             | 48.9            |
| 4         | 75.4              | 119.4             | 69.7            |
| 5         | 88.1              | 142.1             | 93.2            |
| 6         | 81.5              | 127.0             | 89.4            |
| 7         | 94.7              | 149.1             | 101.7           |
| 8         | 91.8              | 147.1             | 99.8            |
| 9         | 78.9              | 131.0             | 117.4           |
| 10        | 127.5             | 149.8             | 191.7           |
| 11        | 107.6             | 171.5             | 104.7           |
| 12        | 115.5             | 193.4             | 141.2           |
| 13        | 107.3             | 174.9             | 154.8           |
| 14        | 106.1             | 171.4             | 134.0           |
| 15        | 89.6              | 144.0             | 124.82          |
| 16        | 102.0             | 163.7             | 168.03          |
| 17        | 99.2              | 157.3             | 63.11           |
| 18        | 130.6             | 180.8             | 142.07          |
| 19        | 119.3             | 168.5             | 117.24          |
| 20        | 137.4             | 202.8             | 141.34          |
| 21        | 127.4             | 180.7             | 120.95          |
| 22        | 141.1             | 200.1             | 184.42          |
| 23        | 138.5             | 201.0             | 158.07          |
| Average   | 102.2             | 156.3             | 116.8           |

Table 3. Comparison of natural radioactivity concentration (Bqkg$^{-1}$) in the bauxite for present study with previous study reported from different countries of the world.

| Country                   | Description       | Mean activity concentration (Bqkg$^{-1}$) | References |
|---------------------------|-------------------|------------------------------------------|------------|
| Saudi Arabia, Az Zabirah  | Bauxite           | $^{226}\text{Ra}$ 102.2 $^{232}\text{Th}$ 156.3 $^{40}\text{K}$ 116.8 | Present study |
| Guinean                   | Bauxite           | 51.25 $^{227}$ | [23] |
| India                     | Bauxite           | 70.2 $^{499}$ | [23] |
| Brazil                    | Bauxite           | 64 $^{154}$ | [11] |
| Greece, Gulf of Itea      | Bauxite           | 72.8 $^{185}$ | [24] |
| Greece, Antikyra Bay      | Bauxite           | 150 $^{205}$ | [11] |
| Hungary                   | Bauxite           | 419.0 $^{256.0}$ | [25] |
| China                     | Bauxite           | 370.0 $^{400.0}$ | [8] |
| Turkey                    | Bauxite           | 235.3 $^{243.4}$ | [26] |
Figure 2. Correlations between $^{226}$Ra with $^{232}$Th (●) and $^{226}$Ra with $^{40}$K (○) in the bauxite samples in Qassim province Saudi Arabia.

The radiological risk can be estimated using indices such as radium equivalent activity, the 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 assuming 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:

$$R_{a_{eq}} = A_{Ra} + 1.43 \times A_{Th} + 0.077 \times A_{K}$$  \hspace{1cm} (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 to radiological hazard, the maximum $Ra_{eq}$ in the bauxite samples must be less than 370 Bq kg$^{-1}$ for safe use. The radium equivalent results ranged from 213.7 ± 10 to 464.6 ± 34 Bq·kg$^{-1}$ and mean value was 340.2. The average value was in the safety recommended limit. These results were higher than the results obtained from bauxite in Nigeria (189-262 Bq·kg$^{-1}$) [10]. Furthermore, the absorbed dose rate in air at 1 m above the ground surface was calculated at different sampling points using equation 3.

$$D_{R} \text{ (nGy h}^{-1}) = 0.427ARa + 0.623A_{Th} + 0.043AK$$ \hspace{1cm} (3)

The values varied from 97.0 to 213.7 nGy/h and the mean value was 155.5 nGy/h in the Az Zabirah bauxite mine. The obtained average value based on Equation (3) was higher than the worldwide average value of 60.00 nGy/h [7]. The calculated effective dose to which population are exposed to gamma rays in the Az Zabirah mine ranged from 0.119 to 0.262 mSv/year with a mean of 0.19 mSv/year. The obtained average values in bauxite mine was lower than the safe limit of 1.00 mSv/year recommended by [7]. The calculation value of the absorbed dose rate in air at 1 m above the ground level and the annual effective dose to which population may likely be exposed at different sampling points in this study sites are shown in figure 3. The obtained values of external index ($H_{ex}$) ranged from 0.60 to 1.3 with an average of 0.9. The internal hazard index ($H_{in}$) values in the bauxite mine in Az Zabirah varied from 0.7 to 1.6 with a mean of 1.2. Comparing the obtained average values of both external and internal hazard indices in both areas, it can be seen that the obtained average values of the external hazard parameters in Az Zabirah was lower than the safe limits values except for the internal hazard index value was slightly higher than the recommended average values by [7].
4. Conclusion

It is important to determine background radiation level in order to evaluate the health hazards resulting from Az Zabirah bauxite mine. The method of gamma spectrometry has been used to measure the radioactivity concentration of 24 samples collected from Az Zabirah bauxite mine in Qassim Province, Saudi Arabia. From this study, the range and average values of the concentrations of $^{226}\text{Ra}$, $^{232}\text{Th}$ and $^{40}\text{K}$ were 102.2 (141.1-62.7), 156.3 (202.8-102.8) and 116.8 (191.7-48.9) Bq/kg respectively. Overall, the study showed that the measured values for $^{226}\text{Ra}$ and $^{232}\text{Th}$ were higher than the values in the worldwide soil. The mean values of radium equivalent and total absorbed dose rate were 340.2 Bq/kg and 155.5 nGy.h$^{-1}$ respectively. The world average values of radium equivalent and absorbed dose are 370 Bq.kg$^{-1}$ and 65 nGy.h$^{-1}$. The annual effective gamma doses was lower than the world’s average. The external hazard parameters in Az Zabirah were lower than the safe limits values. The internal hazard index value was slightly higher than the recommended average values by UNSCEAR. The study area is in the zones of high radiation level, 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 measurements and assessments of possible radiological risks to human health in this province.

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