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Evaluation of NORM and Dose Assessment in an Aluminium Industry in Nigeria

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ABSTRACT

The activity concentrations of natural radionuclides ^{226}Ra, ^{232}Th and ^{40}K in bauxite ore, alumina, dross tailing, aluminium scraps and soil samples collected from an aluminium industry in Nigeria were determined by gamma ray spectroscopy method. The mean values of ^{226}Ra, ^{232}Th and ^{40}K content of the samples ranged from 16 ± 6 (alumina) to 31 ± 10 (scrap), 41 ± 0.12 (scrap) to 134 ± 21 (bauxite) and 47 ± 14 (bauxite) to 354 ± 8 (scrap) Bq·kg^{-1}, respectively. The mean activity concentrations of ^{226}Ra and ^{40}K in all the samples are lower than the world average for soil while ^{232}Th is higher with the exception of alumina and scrap. As a measure of radiation hazard to the occupational workers and the members of the public, the radium equivalent activities and external gamma dose rates due to the radionuclides at 1 m above ground surface were calculated. The radium equivalent activities which varied between 88 ± 10 (alumina) and 222 ± 34 (bauxite) Bq·kg^{-1} are within the safety recommended limit of 370 Bq·kg^{-1}. The mean annual effective doses calculated from the absorbed dose rates in air were between 54 ± 6 (alumina) and 134 ± 20 (bauxite) μSv·y^{-1}, which is lower than the 1 mSv·y^{-1} recommended for the general public. The annual gonadal dose equivalent of all the samples with the exception of alumina was higher than the world average for soil.

Keywords: NORM; Gamma Radiation Dose; Radiation Hazards; Aluminium

1. Introduction

Natural radioactivity is a common phenomenon that is as old as the age of the planet earth. Radioactive elements occur naturally in rocks, soils and water in varying concentrations [1]. They give rise to a natural radiation background that varies by approximately two orders of magnitude over the surface of the earth, but in most situations this exposure is not amenable to control [1]. Most of the essentials of man come from the earth crust with the exception of air. Dependence on these essentials such as soil, water, rock and minerals poses some natural radiation treats to man.

Enhanced levels of naturally occurring radionuclides may be associated with certain natural materials, minerals and other resources. Exploitation of these resources for the production of consumer items may lead to further enhancement of the radioactivity at concentrations above normal in the products, by-products, residues or waste arising from the industrial process. Raw materials used in industries may contain naturally occurring radioactive materials (NORM). The most important are the ^{238}U and ^{232}Th and their decay products as well as ^{40}K [2-5]. The raw materials vary in activity concentrations depending on the region of origin [6]. The mining, transportation and processing of the raw materials may lead to the release of radionuclides into the environment and the distribution of the radionuclides in products and waste thereby giving rise to radiological hazards in workplaces and in the environment.

Aluminium is the most abundant metallic element and the third most abundant element in the earth's crust after oxygen and silicon [7]. The chief source of aluminium is bauxite ore which contains about 30% to 50% hydrated aluminium oxide ($Al_2O_3 \cdot 2H_2O$ with some impurities like iron and clay). This may contain significant activity concentrations due to either or both ^{238}U and ^{232}Th, depending on the ore mineralogy. As a result of this, aluminium industries are considered as industries with a potential radiological impact on people and the environment [8-10].

The Aluminium Smelter Company of Nigeria (ALSCON) is the nation's premier smelter plant. It is located

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at Ikot-Abasi in Akwa Ibom State, in the southeastern part of Nigeria. Aluminium also occurs with other elements as silicates. Common silicates that contain aluminium are feldspar, muscovite mica, kaolin, fuller's carts etc. Even though kaolin is found in abundance in Benue State, the north-central of Nigeria as well as in Akwa Ibom State, the southern part of the country, the industry depends solely on imported raw materials (bauxite and alumina) for the production of aluminium. This is because the silicates are complex and not normally used for aluminium production and besides, the silicates are not economical because of the large contents of clay and iron haemitite in them.

Work activites in which radiation exposure of workers and members of the public is increased due to the presence of NORM are receiving attention from regulatory authorities. To the best of our knowledge, there is no study on radiological aspect of the aluminium industry in Nigeria. Therefore, the present study seeks to investigate the level of natural radionuclides in the raw materials used for the production of aluminium in Nigeria and soil samples from the surrounding area of the industry. The health effects that such radionuclides may have on the population are also assessed.

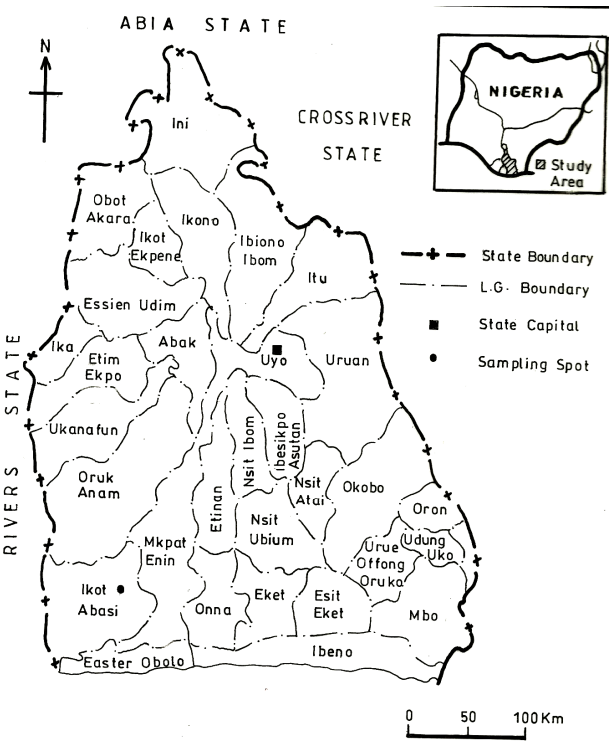

Figure 1. The map of Nigeria showing the sampling spot.

2. Materials and Methods

A total of forty-four samples were collected from ALSCON for this study. They include bauxite ore (4 samples), alumina (10 samples), dross tailing (14 samples), aluminium scraps (2 samples) and soil (14 samples). The soil samples were taken around the dross at distances of about 100 cm apart. The scrap samples were collected from the aluminium scraps obtained during production and processing of aluminium into other products like ingots and billet before it is recycled. Limited number of samples was collected due to restriction by the industry involved. The map of Nigeria showing the sampling spot is shown in **Figure 1**.

The samples were air dried and then oven dried. The dried samples were grinded in order to achieve homogenization; they were sieved through a 0.20 mm mesh sieve. A mass of 200 g each of the samples was weighed and packed into plastic container, hermetically sealed and stored for a minimum of one month before measurement. This is to prevent the escape of gaseous ^{222}Rn and ^{220}Rn from the samples and ensure the attainment of secular equilibrium [11].

The activity concentrations of the natural radionuclides were measured using a 7.6 cm × 7.6 cm NaI(Tl) detector (model No. 802-series) by Canberra Inc. The detector is coupled to series 10 plus Multi Channel Analyzer (MCA) (model No. 1104) through a preamplifier base. The detector is placed in a lead shield. The detector has a resolution of about 8% at 0.662 MeV of ^{137}Cs. This is capable of distinguishing the gamma ray energies considered during measurements. The ^{226}Ra and ^{232}Th content were determined from the photopeaks of ^{214}Bi (1.764 MeV) and ^{208}Tl (2.614 MeV), respectively. The potassium content of the sample was determined from photopeak of ^{40}K (1.460 MeV). These peaks are clean, reasonably strong with very low continuum and were considered good enough because of poor resolution of the NaI(Tl) detector used in this work. The counting time for each sample was 10 hours.

Energy calibration was done to ensure that a relationship exist between the peak position of the spectrum and the corresponding gamma-ray energy. Energy calibration is achieved by measuring the spectrum of a source emitting gamma-rays of known energies and comparing the same with the measured peak positions. The calibration of the system was carried out using gamma sources of known energies for student laboratory experiments from Nucleus Inc., Oak Ridge, TN, USA. The peak area of each radionuclide was computed from the memory of the MCA using an algorithm which subtracts counts due to Compton scattering of higher peaks and other background effects from the total area.

For the calculation of individual radionuclide, the detection efficiency E_p of the system was determined. The detection efficiency at the constant geometry of counting is defined as [12,13];

$$E_p = \frac{A}{tCYm} \quad (1)$$

where A is the net area under the photopeak, C (Bq·kg^{-1}) is the activity concentration of a reference sample of mass m (kg) counted for a time t (s) and Y is the gamma radiation yield. The efficiency was determined using a reference source of known activity concentrations for each of the radionuclides prepared from Rocketdyne Laboratories, California USA, which is traceable to a mixed standard gamma source (No. 48772-356) by Analytic Inc., Atlanta, Georgia. The activity concentrations of each radionuclide in the samples were obtained by relating the detection efficiency to the net area under each photopeak.

3. Results and Discussion

The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K are shown in **Table 1**. The mean activity concentrations range from 16 ± 6 (alumina) to 31 ± 10 (scrap), 41 ± 0.1 (scrap) to 134 ± 21 (bauxite) and 47 ± 14 (bauxite) to 354 ± 8 (scrap) for ^{226}Ra, ^{232}Th and ^{40}K, respectively. The errors quoted are the standard deviations of the mean values. The mean activity concentrations of ^{226}Ra and ^{40}K in all of the samples are less than the world average for soil, 33 and 420 Bq·kg^{-1}, respectively while ^{232}Th is higher than the world average, 45 Bq·kg^{-1}, with the exception of alumina and scrap [1]. **Figure 2** presents a bar chat of the mean activity concentrations of the three radionuclides. Some reported concentration ranges of radioactivity in bauxite ore are given as 10 - 9000 Bq·kg^{-1} for uranium series radionuclides, 35 - 1400 Bq·kg^{-1} for thorium series radionuclides and 10 - 600 Bq·kg^{-1} for ^{40}K [9]. Comparison of these with this study shows that the results obtained fall within the lower ranges.

The radiological risk was assessed by calculating the 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 compare the activity concentrations and the radiological effect of the three radionuclides, a common index is used. The widely used index is the radium equivalent activity, Ra_{eq} [2,14]. The Ra_{eq} had been defined assuming that 370 Bq·kg^{-1} of ^{226}Ra, 259 Bq·kg^{-1} of ^{232}Th and 4810 Bq·kg^{-1} of ^{40}K produce the same gamma dose rate and is given as [2,15,16];

$$Ra_{eq} = C_{\mathrm{Ra}} + 1.43C_{\mathrm{Th}} + 0.077C_{\mathrm{K}} \quad (2)$$

where C_{Ra}, C_{Th} and C_{K} are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq·kg^{-1}, respectively. As far as radiological hazard is concerned, the safety criterion for materials containing these three radionuclides is 370 Bq·kg^{-1} [17]. The results obtained for Ra_{eq} are presented in **Table 2**. The mean values of the radium equivalent activity ranged from 88 ± 10 to 222 ± 34 Bq·kg^{-1}. These fall within the safety recommended limit of 370 Bq·kg^{-1}.

The absorbed dose rate in air at a height of 1 m above the ground due to ^{226}Ra, ^{232}Th and ^{40}K was calculated using the formula [1,18];

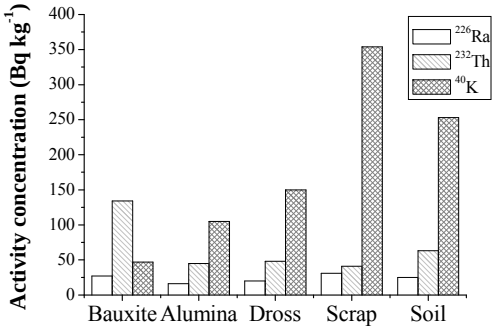

Figure 2. Mean activity concentrations of natural radionuclides in samples.

$$\mathrm{D(nGy{\cdot}h^{-1})} = 0.462C_{\mathrm{Ra}} + 0.604C_{\mathrm{Th}} + 0.0417C_{\mathrm{K}} \quad (3)$$

where C_{Ra}, C_{Th} and C_{K} are as defined in Equation (1). The mean absorbed dose rates due to the three radionuclides ranged from 39 ± 5 nGy·h^{-1} (alumina) to 95 ± 4 nGy·h^{-1} (bauxite). The mean absorbed dose rate for soil samples in the surroundings of the industry is 60 ± 8 nGy·h^{-1}. This corresponds to the estimated world average value for soil 60 nGy·h^{-1} [1]. All other samples had a mean value less than the world average except bauxite which had 95 ± 14 nGy·h^{-1}.

The annual effective doses were calculated using a conversion coefficient of 0.7 Sv·Gy^{-1} for an absorbed dose in air to effective dose in human body [1]. The occupancy time was taken as the normal working hours in Nigeria which is 8 hours per day for five days in a week. For 50 working weeks per annum, the occupancy time is 2000 h·y^{-1}. Hence, the annual effective dose, H, is

$$\mathrm{H} = \mathrm{D} \times 10^{-9}\ \mathrm{(Gy{\cdot}h^{-1})} \times 0.7\ \mathrm{Sv{\cdot}Gy^{-1}} \times 2000\ \mathrm{h{\cdot}y^{-1}} \quad (4)$$

The average annual effective dose are 134 ± 20, 54 ± 6, 63 ± 13, 76 ± 7 and 84 ± 11 μSv·y^{-1}, respectively for bauxite, alumina, dross, scrap and soil. These are less than the 1 mSv·y^{-1} recommended for the public (non exposed workers) [19].

The organs of interest considered by UNSCEAR (1988), are the gonads, the active bone marrow and the bone surface cells [20]. Hence the annual gonadal dose equivalent (AGDE), due the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the samples was calculated using the following relation [21];

$$\mathrm{AGDE\ (\mu Sv{\cdot}y^{-1})} = 3.09C_{\mathrm{Ra}} + 4.18C_{\mathrm{Th}} + 0.314C_{\mathrm{K}} \quad (4)$$

The mean values of the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K were used to calculate the AGDE. The results obtained (**Table 2**), show that the AGDE from all the samples with the exception of alumina are higher than the world average value of 0.30 mSv·y^{-1} [21].

4. Conclusion

The activity concentrations of natural radionuclides in

Table 1. Activity concentrations of natural radionuclides in samples.

Sample	^{226}Ra		^{323}Th		^{40}K	
	Range	Mean ± Std	Range	Mean ± Std	Range	Mean ± Std
Bauxite (4)	19 - 36	27 ± 7	112 - 160	134 ± 21	28 - 59	47 ± 14
Alumina (10)	5 - 23	16 ± 6	35 - 50	45 ± 4	91 - 137	105 ± 16
Dross (14)	7 - 55	20 ± 13	40 - 61	48 ± 5	118 - 193	150 ± 20
Scrap (2)	24 - 38	31 ± 10	41	41 ± 0.12	348 - 360	354 ± 8
Soil (14)	16 - 35	25 ± 6	54 - 80	63 ± 7	152 - 442	253 ± 110

Table 2. Radium equivalent activity (Ra_{eq}), absorbed dose rate (D) annual effective dose (H) and annual gonadal dose equivalent (AGDE) of natural radionuclides in samples.

Sample	Ra_{eq} (Bq·kg^{-1})		D (nGy·h^{-1})		H (μSv·y^{-1})		AGDE (mSv·y^{-1})
	Range	Mean ± Std	Range	Mean ± Std	Range	Mean ± Std	
Bauxite (4)	189 - 262	222 ± 34	81 - 112	95 ± 14	114 - 157	134 ± 20	0.66
Alumina (10)	62 - 97	88 ± 10	27 - 43	39 ± 5	37 - 60	54 ± 6	0.27
Dross (14)	74 - 149	101 ± 20	33 - 67	45 ± 9	46 - 94	63 ± 13	0.31
Scrap (2)	109 - 124	117 ± 11	50 - 58	54 ± 5	71 - 81	76 ± 7	0.38
Soil (14)	116 - 177	134 ± 17	52 - 80	60 ± 8	72 - 112	84 ± 11	0.42

samples from an aluminium industry had been measured using gamma ray spectroscopy method. The radium equivalent activity (Ra_{eq}), absorbed dose rate in air and the annual effective dose were calculated from the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K. The results of the Ra_{eq} are within the recommended safety limit. The annual effective doses due to the samples are less than the recommended limit of 1 mSv·y^{-1} to the general public. Hence the workers and the public are not at risk as far as radiological hazard is concerned. However the annual gonadal dose equivalent was higher than the world average value with the exception of alumina.

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