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Determination of Activity Concentrations in Selected Rock Samples from Quarry Sites in Ibadan, Nigeria

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ABSTRACT

Measurement of natural radioactivity in rock is very important to determine the amount of change in the natural background activity with time as a result of any radioactive releases. It was very important to monitor radioactive substances released to the environment for proper environmental protection. The aim of this work was to investigate the concentration of radioactive substance in selected rock samples in a quarry site in Ibadan for the purpose of safety assessment. Fifteen rock samples, each of mass 1kg were collected from the quarry site in Ibadan and then hammered into pieces before being crushed into very fine grain. The desired samples were then stored in polythene sachets for 28 days so as to reach secular equilibrium. Samples were thereafter taken to the Nuclear Physics Laboratory for counting and analysis. The gamma analysis was performed on a 76×76 mm Sodium Iodide (Thallium doped) NaI (TL) scintillation counter detector photomultiplier. Results obtained showed that the ranges of the activity concentrations of ^{40}K, ^{238}U, ^{232}Th in the rock samples are 87.05±8.46 to 174.45±2.56, 8.25±1.45 to 21.35±1.25 and 1.02±0.32 to 10.63±0.71 Bq kg^{-1}. The average concentrations of ^{40}K, ^{238}U, ^{232}Th are 126.79±3.36, 12.34±0.91 and 3.78±0.49 Bq kg^{-1}. Activity concentrations obtained were less than that of United Nations Scientific Committee on Effects of Atomic Radiation Report (UNSCEAR) report 2000 which indicated the worldwide activity concentration of ^{40}K, ^{238}U and ^{232}Th within the ranges 140-850, 17-60 and 11-64 Bq kg^{-1} and mean 400, 35 and 30 Bq kg^{-1}, respectively.

Key words: Natural radioactivity, activity concentration, secular equilibrium, rock samples, gamma analysis

INTRODUCTION

Exposure to ionizing radiation from natural sources is a continuous and unavoidable feature of life. Human beings are exposed to natural background radiation every day from the ground, building materials, air, food, outer space and even elements in their own bodies. Gamma radiation emitted from primordial radionuclides and their progenies were one of the main external sources of radiation exposure to the humans[1]. More than 60 radionuclides can be found in nature and they can be placed in three general categories i.e., Primordial-formed before the creation of the earth, cosmogenic-formed as a result of cosmic ray interactions and Human produced- enhanced or formed due to human actions[2]. Natural radioactivity can be found, in low concentrations, throughout the geology of the planet. It was naturally occurring in many rocks, soils and waterways. Phosphate rocks were one of the matrices that contain higher levels of natural radioactivity than the background. As the world population continues to grow,

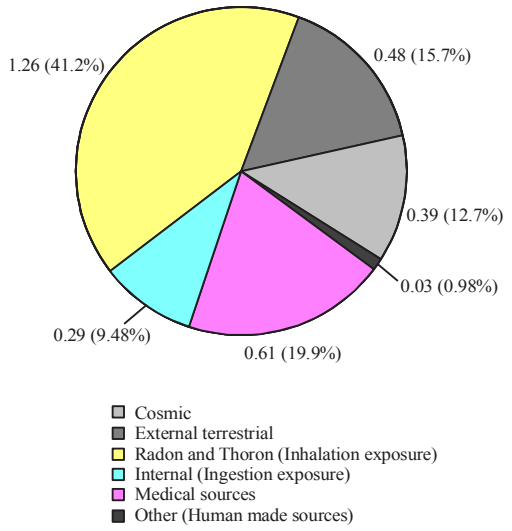

Fig. 1: Typical annual human exposures (mSv) to all sources of radiation worldwide

so does the world's need for food. The increased demands of food production had led to a steady increase in the use of phosphate fertilizers[3]. Artificial radiations originate mainly from fallout resulting from the nuclear weapons trial, airborne release from nuclear establishments and reactor accidents. Radioactive dust was released into the atmosphere, carried widely and later fall gradually to the earth surface or washed down in rain and eventually settles on land or in the sediments of water bodies. Therefore due to the natural and artificial processes radionuclides may accumulate and become concentrated in selected areas of environment[4]. According to UNSCEAR[5] report of 2000, the average annual dose from natural sources of radiations was about 2.4 mSv and this accounts for about 79% of total human exposure worldwide (Fig. 1).

The aim of this work was to investigate the concentration of radioactive substance in some rock samples in a quarry site in Ibadan for purpose of safety assessment and recommendations.

MATERIALS AND METHODS

The knowledge of the absorbed dose rates at environmental level was necessary in determination of stochastic effects[6]. The study of the distribution of the primordial radionuclides (^{238}U, ^{232}Th and ^{40}K) and their daughters allows the understanding of the radiological implications of these elements due to the γ-ray exposure of the body[7]. Every rock contains natural radionuclides such as Potassium, Thorium and Uranium and the radioactivity levels depend strongly on the type of rock. Table 1 shows the concentrations of radioactivity in some major rock types. The amount of heat generated was proportional to the rate of decay and the amount of radioactive material present at the time[8].

Table 1: Concentrations (Bq kg^{-1}) of radioactivity in major rock types

Rock type	K-40	Rb-87	Th-232	U-238
Igneous rocks	300	30	10-15	7-10
Basalt (Average)				
Sedimentary rocks	800	110	50	40
Shale sandstones	<300	<40	25	10
Beach sands (unconsolidated)	70	8	8	25
Average	850	100	44	36

Radioactive elements found in rock and soil from the earth makes their way into the bodies through the water we drink, food we eat, air we breathe it contains them. These naturally occurring radioisotopes such as carbon-14, potassiurn-40, Thorium-232, Uranium-238, Polonium-218 and Tritium (Hydrogen-3) expose us to radiation from within the bodies. The largest contributor to the daily exposure of radiation was the natural radioactivity and the major form of natural radiation was Radon gas. Radon-222 was a naturally occurring decay product of Uranium-238 which was commonly found in rocks and soils[9]. Table 1 below gives a picture of radioactivity concentrations in major rocks according to the National Council of Radiation Protection.

Description and geology of study area: Ibadan is located in south-western Nigeria in the south-eastern part of Oyo State at about 119 km (74 miles) northeast of Lagos and 120 km (75 miles) east of the Nigerian international border with the Republic of Benin. It lies completely within the tropical forest zone but close to the boundary between the forest and the derived savannah. The city ranges in elevation from 150 m in the valley area, to 275 m above sea level on the major north-south ridge which crosses the central part of the city. The city covers a total area of 3,080 km^2 (the largest in Nigeria). It has a tropical wet and dry climate with a lengthy wet season and relatively constant temperatures throughout the course of the year. Ibadan's wet season runs from March through October, though August sees somewhat of a lull in precipitation. This lull nearly divides the wet season into two different wet seasons. November to February forms the city's dry season, during which Ibadan experiences the typical West African harmattan. The mean total rainfall for Ibadan was 1420.06 mm, falling in approximately 109 days. There were two peaks for rainfall, June and September. The mean maximum temperature is 26.46°C, minimum 21.42°C and the

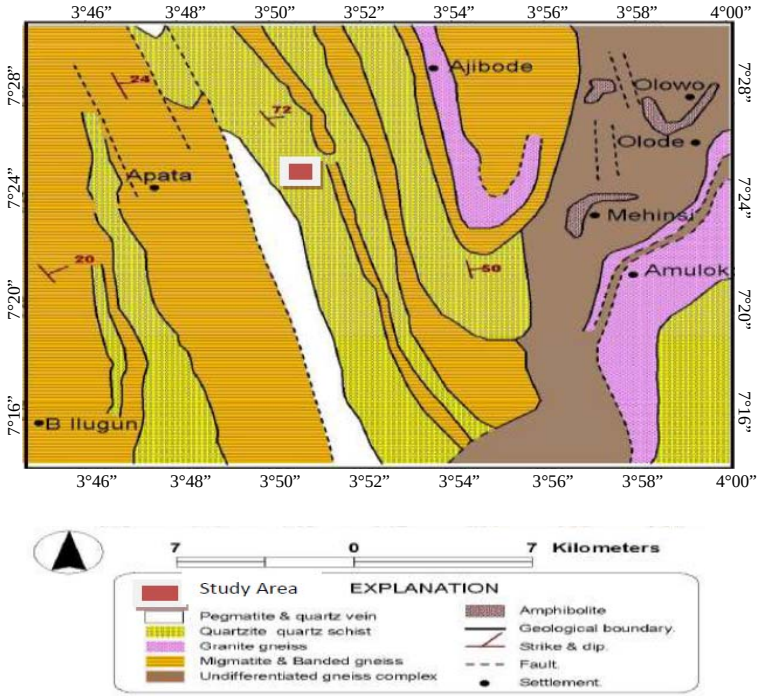

Fig. 2: Geological map of Ibadan showing the quarry site (After Okunlola *et al.*[10])

relative humidity was 74.55%. Ibadan is a region in the Southwestern part of Nigeria is dominated by rock types such as granite and granitic schist of the metase dimentary series, banded gneiss and granite gneiss, augen gneiss and migmatite complex[10]. After Okunlola *et al.*[10] showing the Ibadan quarry site as shown in Fig. 2. Quartzite outcrops occur as ridges with relatively high elevation and are commonly schistose in form. Their strike line runs in the north-south direction between 3400 and 3500 consistently dipping eastwards with characteristic crosscutting features[11].

METHODOLOGY

Fifteen rock samples each weighing about 1 kg was collected from five different locations in the study area. The rocks were hewed out of the masses with the use of a geological hammer. Figure 3 shows the rock samples collected at the quarry site. The whole 1 kg of rock was first hammered into smaller pieces, before going through the rock-crushing machine, Pulveriser and ball-mill. Finer grains-more powdered-finer grains were achieved. The powdered samples were then passed through a fine-aperture mesh screen (2 mm in diameter), in order to have fine grained powdered samples of desired specification. The desired samples were hand-packed and stored in

Fig. 3: Samples of rocks collected from investigated area

taken down to the Nuclear Physics Laboratory, Obafemi Awolowo University, Ile-Ife for counting and analysis.

Procedure of counting and analysis: The gamma analysis was performed on a 76×76 mm Sodium Iodide (Thallium doped) NaI (TL) scintillation counter detector photomultiplier. The output of the detector was connected to a Canberra series 10 plus portable multichannel analyzer (MCA) which recorded the gamma spectra of the rock samples as well as background radiation. The counting time for background and each sample

was 2 h (7,200 sec). Thirty five grams of each meshed rock sample was weighted out and transferred to a 0.6 L capacity container for gamma analysis. Cross contamination was avoided by maintaining utmost cleanliness while counting each sample. The detector system was calibrated for a radionuclide concentration estimation using the same counting time for Uranium (U-238), Thorium (Th-232) and Potassium (K-40) standards spelt out by IAEA.

UNSCEAR has given the dose conversion factors for converting the activity concentrations of K-40, U-238 and Th-232 into dose (nGy h^{-1} per Bq kg^{-1}) as 0.427, 0.662 and 0.043, respectively. Using these factors, the total absorbed dose rate in air is calculated as given in equation below:

$$D = 0.042A_k+0.428A_U+0.666A_{Th} \quad (1)$$

where, A_k, A_U and A_{Th} are the activity concentration for K-40, U-238 and Th-232. Substituting the values of the activity concentration for K-40, U-238 and Th-232 into Eq. 1 above gives the mean absorbed dose rate due to the three primordial radionuclides as 0.013±0.001 μGy h^{-1} for the five locations. The collective effective dose equivalent S_E was calculated for the regions using:

$$S_E = NH \quad (2)$$

where, N is the population of the regions (i.e., all locations) and H is the mean annual effective dose equivalent to individuals. A conversion factor of 0.7 Sv Gy^{-1} was used to convert the absorbed dose rate to human effective dose equivalent with an outdoor occupancy of 20% on the absorbed dose rate to obtain as the mean annual effective dose equivalent in the region using Eq. 2.

RESULTS AND DISCUSSION

The Table 2 displays the Mean Values of Radioactive Concentration due to K-40, U-238 and Th -232 and highest activity concentration for K-40, U-238 and Th-232 was observed at Ogunmakin South, Idi Igba and Abanla (151.35±3.62, 17.45±0.89 and 5.35±0.74 Bq kg^{-1}), respectively, while the lowest activity concentrations due to K-40, U-238 and Th-232 is observed at Idi Igba, Idi Ayunre and Ogunmakin North (95.71±4.71, 9.81±1.01 and 1.90±0.30 Bq kg^{-1}), respectively. The average activity concentrations for K-40, U-238 and Th-232 in the present study were lower than worldwide average for these radionuclides in rock samples.

Table 2: Mean values of radioactive concentration due to K-40, U-238 and Th-232 in Bq kg^{-1}

		Activity concentration (Bq kg^{-1})				
Rock samples	Locations	K-40	U-238	Th-232	Dose rate (mSv h^{-1}) for g	
A1	Ogunmakin south	152.24±4.53	15.29±0.24	10.63±0.71	0.17	0.16
A2	Ogunmakin south	127.35±3.78	12.02±0.76	2.15±0.86	0.18	0.19
A3	Ogunmakin south	174.45±2.56	13.43±0.84	1.02±0.32	0.16	0.15
	Mean deviation	151.35±3.62	13.58±0.61	4.60±0.63	0.17	0.17
B1	Idi Ayunre	132.84±3.71	10.94±1.42	1.25±0.52	0.14	0.13
B2	Idi Ayunre	129.04±2.68	9.56±0.64	2.54±0.09	0.17	0.16
B3	Idi Ayunre	109.65±1.47	8.94±0.98	5.32±0.92	0.19	0.18
	Mean deviation	123.84±2.62	9.81±1.01	3.04±0.51	0.17	0.16
C1	Idi Agba	94.65±1.04	11.83±0.47	4.25±0.13	0.17	0.16
C2	Idi Agba	105.43±4.64	19.16±0.94	1.94±0.52	0.15	0.14
C3	Idi Agba	87.05±8.46	21.35±1.25	5.78±0.22	0.13	0.14
	Mean deviation	95.71±4.71	17.45±0.89	3.99±0.29	0.15	0.145
D1	Abanla	97.65±4.52	11.34±0.68	6.25±0.95	0.12	0.13
D2	Abanla	106.83±3.85	10.14±0.75	6.38±0.86	0.18	0.17
D3	Abanla	143.54±3.92	9.76±1.68	3.42±0.42	0.17	0.18
	Mean deviation	116.01±4.10	10.41±1.04	5.35±0.74	0.157	0.16
E1	Ogunmakin North	160.32±1.59	8.25±1.46	1.64±0.25	0.14	0.15
E2	Ogunmakin North	155.35±2.00	14.38±0.72	2.25±0.36	0.16	0.17
E3	Ogunmakin North	125.54±1.77	8.73±0.94	1.82±0.28	0.11	0.12
	Mean deviation	147.07±1.79	10.45±1.04	1.90±0.30	0.14	0.15
		Total average of value of activity concentration			Dose rate (mSv h^{-1}) for g	
		K-40	U-238	Th-232	Before lab.	After lab.
		126.79±3.36	12.34±0.91	3.78±0.49	0.15	0.15

Table 3: Comparison of radioactivity of rock samples with other areas of the world

Country	^{238}U (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	^{40}K (Bq kg^{-1})	References
Egypt (Wadi Karim)	14-227	10.5-183	2299-7356	EL Arabi[17]
Brazil	2.9-9087	1.4-3834	132-10230	Anjos *et al.*[18]
Cyprus	1-588	1-906	50-1606	Tzortzis *et al.*[19]
China	20.4	30.1	1009.5	Lu and Zhang[20]
India	9.1-24.1	3.7-12.7	101.8-264.1	Patra *et al.*[21]

Studies in the past have shown that naturally occurring radionuclides were trapped in the earth's crust during the formation of the parent rocks. The subsequently ended in soils forming part of rock cycle[12]. Shittu *et al.*[13] determined the risk associated with naturally occurring radioactive materials at selected quarry site in Abuja, Nigeria. Analysis of their measurement showed that the amount of Uranium (^{238}U), Thorium (^{232}Th) and Potassium (^{40}K) were present in the rocks are 74.74±5.67, 199.23±43.30 and 1021.27±7.14 Bq kg^{-1}, respectively. In measuring the activity concentrations of rocks from quarry sites in Abeokuta, South-Western Nigeria, it was also found out that the mean concentration of ^{40}K, ^{238}U and ^{232}Th are 124.85±24.05, 27.804±10.10 and 20.22±8.2, respectively[14].

In the article titled The 226Ra, 232Th and 40K contents in the Abakaliki baked shale construction materials and their potential radiological risk to public health, southeastern Nigeria, Celestine Okogbue and Mathias Nweke found out that the mean activity concentrations of 226Ra, 232Th and 40K were 37.57±2.86, 67.97±1.70 and 552.02±2.92 Bq kg^{-1}, respectively, higher than the world average activity concentrations[15].

Gbadebo[16] determined the concentrations of natural radionuclidess in the crystalline bedrocks and soils of the selected abandoned quarries in Abeokuta. The results of the radionuclide from rocks show concentration of ^{40}K, ^{238}U, ^{232}Th and as 201.36±5.87, 39.17±4.45 and 15.09±14.06, respectively. The variations and patterns of activity in the rocks considered in this work agree with those mentioned.

The variations of natural radioactivity levels at different sampling areas were due to the variation of concentrations of these elements in the geological formations. The concentrations of the above mentioned radionuclides were compared with that of other author's for rock in Table 3.

As could be seen, among the U-238 concentrations, average value goes along with U-238 concentrations of India, Eygpt and Cyprus and lower than average U-238 concentrations of Brazil[18], China and Turkey (Eskişehir). Among the Th-232 concentrations, Th-232 concentrations value goes along with Egypt[17], Cyprus and India and lower than that of China[20], Brazil and Turkey (Eskişehir). For K-40 concentrations, value goes along with that of India and Cyprus[21] and lowers than the others[19].

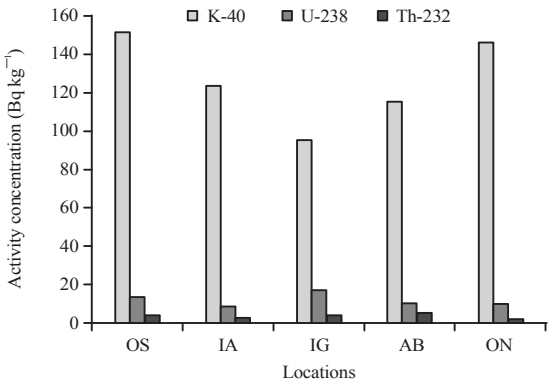

Fig. 4: Activity concentration due to K-40, U-238 and Th-232, OS: Ogunmakin South, IA: Idi Ayunre, IG: Idi Igba, AB: Abanla, ON: Ogunmakin North

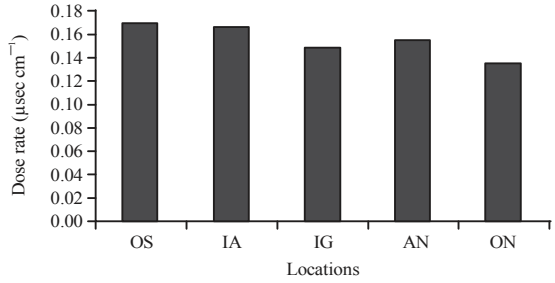

Fig. 5: Dose rate of rock samples before laboratory analysis

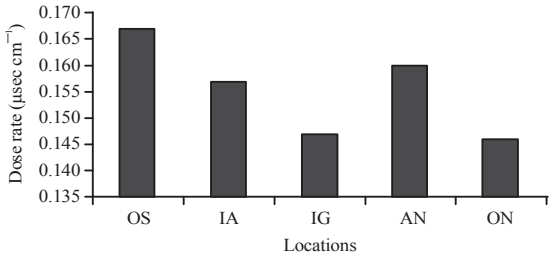

Fig. 6: Dose rate of rock samples after laboratory analysis

Figure 4 is the bar chart of the activity concentration due to K-40, U-238 and Th-232. Figure 5 and 6 show the dose rate of rock samples before and after laboratory analyses while Fig. 7 is their comparison.

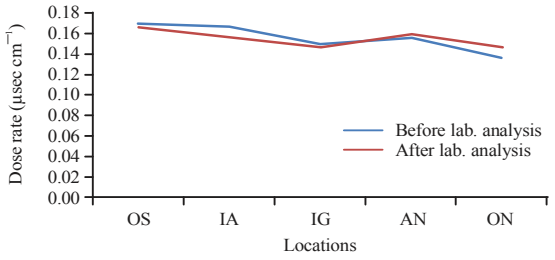

Fig. 7: Comparison of dose rate measurement before and after laboratory analysis

CONCLUSION

By analyzing the results it was concluded that the natural radiation in an area can be measured by analyzing the rock samples from that area. Activity concentrations obtained are less than that of UNSCEAR report 2000 which indicated the worldwide activity concentration of ^{40}K, ^{238}U and ^{232}Th within the ranges 140-850, 17-60 and 11-64 Bq kg^{-1} and mean 400, 35 and 30 Bq kg^{-1}, respectively. However, proper routine monitoring of the level of activity concentration was required for proper environmental protection.

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