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Radioactivity Assessment of ^{40}K, ^{238}U and ^{232}Th in Surface Soil Samples of Igbokoda, Southwest of Nigeria

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Abstract

Natural radionuclides are found in soil and are capable of disintegrating leading to the release of ionizing radiations that can have harmful effects on individuals exposed to them most especially when exceeding the recommended global limits of radiological parameters. Assessment of activity concentrations of ^{40}K, ^{238}U and ^{232}Th in surface soil samples from different locations at Igbokoda where crude oil exploration was taking place, at Ondo State in Nigeria, had been determined by gamma spectrometry using NaI (TI) detector coupled with a pre-amplifier base connected to a multiple channel analyzer (MCA) which was used to calculate the radiological indicators. 10 samples were collected from the study area putting into consideration the densely populated parts where crude oil exploration and drilling its wells were taking place. The mean activity concentrations of ^{40}K, ^{238}U and ^{232}Th obtained from the soil samples were 494.64 ± 10.46 , 19.76 ± 3.09 and 31.98 ± 5.10 Bq kg^{-1}, respectively. The mean external hazard index (H_{ex}) and mean internal hazard index (H_{in}) for all the soil samples were 0.2836 and 0.3370, respectively, while the mean radium equivalent activity was 104.92 Bq kg^{-1}. The mean absorbed dose rate value was 49.68 nGy hr^{-1} with mean annual effective dose equivalent of 0.0610 mSv yr^{-1}. The mean excess lifetime cancer risk for outdoor exposure was 0.2132 x 10^{-3}. The values of the radiological parameters: mean external and internal hazard indices, mean radium equivalent activity, mean absorbed dose rate, mean annual effective dose equivalent and mean excess lifetime cancer risk were within the recommended limits of 1.0 Bq kg^{-1}, 370 Bq kg^{-1}, 55.00 nGy hr^{-1}, 1.0 mSv yr^{-1} and 1.45 x 10^{-3}, respectively.

Keywords: Activity concentration, Exposure, Health implication, surface soil.

Introduction

Human beings are continuously exposed to ionizing radiation from Naturally Occurring Radioactive Materials (NORM). NORM existing in soil could pose potential health risk (Wilson 1993), especially when aided by natural processes such as weathering deposition and wind erosion (Elles et al. 1997). The artificial sources of radionuclides are largely due to medical and industrial activities. Studies on radiation levels and radionuclides distribution in the environment provide vital radiological baseline information. Such information is essential in estimating human exposure from natural and man-made sources of radiation and necessary in establishing rules and regulations relating to radiation protection (Quindos et al. 1994).

The Earth is naturally radioactive, and about 90% of human radiation exposure arises from natural sources such as cosmic radiation, exposure to radon gas and terrestrial radionuclides (Lee et al. 2004). There are many sources of radiation and radioactivity in the environment. The earth and atmosphere contain varied levels from natural radionuclides such as ^{238}U and ^{232}Th decay chains as well as singly occurring types such as

^{40}K. Soil features, geological formations, and human activities related to radiation and radioactivity are important factors enhancing the background levels of natural radiation (Sujo et al. 2004). The continual enhancement of these radionuclides in the environment may be attributed to several factors such as the successive utilization of phosphate fertilizer, burning of fossil fuels (crude oil and coal), mining and milling operations, and building materials. Ingesting and inhaling such radionuclides contribute significantly to the radiation doses that people receive (Martínez 1989). In addition, mining and milling of both nuclear and non-nuclear materials may cause significant environmental and occupational radiological hazards. Typically, NORM in commercial and industrial products has the potential to expose workers and members of the public to various degrees of nuclear radiations. The main external source of radiation exposure to human is the gamma radiation emitted by naturally occurring radioisotopes, also called terrestrial environmental radiation (UNSCEAR 1993, UNSCEAR 2000). These radioisotopes, such as ^{40}K and the radionuclide from the ^{232}Th and ^{238}U series and their decay products, exist at trace levels in all ground formations.

Therefore, natural environmental radioactivity and the associated external exposure due to gamma radiation depend primarily on the geological and geographical conditions, and appear at different levels in the soils of each different geological region (UNSCEAR 1993, UNSCEAR 2000). However, it has been observed that the types and concentrations of radionuclides vary considerably depending on the soil types. The effects of the radiation emitted by different radionuclides depend on the over lining soil material (thickness and type), its chelating agents and physico -chemical properties (Belivermis et al. 2009). Investigation has shown that natural radioactivity and the associated exposure due to gamma radiation depend primarily on soil type (Belivermis et al. 2009). The interaction of ionizing radiation with the human body leads to various biological effects which may later show up as clinical symptoms (ICRP 2007). Najam and Younis (2015) studied the natural radionuclides in the soil samples of selected regions at Nineveh province in Iraq and health implications to individuals in the study area, the average absorbed dose rate, average radium equivalent value, and average internal and external hazard indices were obtained to be 48.91 nGy hr^{-1}, 89.41 Bq kg^{-1}, 0.329 Bq kg^{-1} and 0.238Bq kg^{-1}, respectively, and all the values were within the recommended limits globally. Also, the study of the activity concentrations of radionuclides in soil and water samples in Eagle, Atlas and rock cement companies in Port Harcourt was carried out by Avwiri (2005). Soil and water samples collected from the respective locations were analyzed using the gamma-ray spectrometer. The mean absorbed dose rates in the soil samples were 49.27, 45.21 and 42.33 nGy hr^{-1} for Eagle, Atlas and Rock cements companies, respectively while the values for water samples were 22.16, 20.75 and 19.37 nGy hr^{-1} for the respective companies. Mean annual effective dose equivalents of 0.18 and 0.39 mSv yr^{-1} were obtained for the water and soil samples, respectively. The results obtained by Avwiri (2005) were lower than the International Commission on Radiological Protection (ICRP 2007) maximum permitted limit of 1.0 mSv yr^{-1} and therefore, had no significant radiological health burden on the environment and the populace. Also, Sowole (2014) studied the activity concentrations of radionuclides in surface soil samples of major markets in Sagamu and obtained the highest activity concentrations of ^{40}K to be 1274.26 ± 4.26 Bq kg^{-1} and ^{238}U to be 40.72 ± 3.12 Bq kg^{-1}, both from Falawo market surface soil samples while that of ^{232}Th was obtained from Sabo market with values of 115.62 ± 16.39 Bq kg^{-1}. The mean external hazard index (H_{ex}) was calculated to be 0.616 Bq kg^{-1} and mean internal hazard index (H_{in}) was 0.691 Bq kg^{-1} for all the soil samples from Falawo market, and that of Awolowo market were 0.566 Bq

kg^{-1} for mean external hazard index (H_{ex}) and 0.634 Bq kg^{-1} for mean internal hazard index (H_{in}). Also for Sabo market the mean values were calculated to be 0.594 Bq kg^{-1} for external hazard index (H_{ex}) and 0.658 Bq kg^{-1} for internal hazard index (H_{in}). All the values obtained were less than 1.0 Bq kg^{-1} as recommended by International Commission on Radiological Protection (ICRP 2007) and therefore have no negative radiological health implication to the people within the markets and their environs.

The objective of this work however was to measure the activity concentrations of ^{40}K, ^{238}U and ^{232}Th in soil samples of Igbokoda in Ilaje local government area, Ondo State, Southwest of Nigeria, and determine the radiological hazard parameters on human beings due to exposure to these naturally occurring radionuclides. The measured activity concentrations of these natural radionuclides were used to estimate the internal and external hazard indices, radium equivalent activity, absorbed dose rate in air and other radiological risk parameters to individuals within the study area.

Materials and Methods

Surface soil samples were collected randomly at different locations within Igbokoda study area (Figure 1). Ten (10) samples were collected putting into consideration areas that were densely populated in the study area. The samples were taken from four (4) cardinal directions (north, west, east and south) at a distance of not less than 300.0 m apart for all the samples, and each sample was collected from 100.0 cm^2 within the surface depth of 0.0 to 30.0 cm into the ground. The samples were put in different containers and taken to the laboratory to oven dry for about seventy two hours (72 hrs) at a temperature of about 110 °C at a constant weight and relative humidity of about 70% as recommended by IAEA (1989). Each dried soil sample was crushed and sieved using a 2 mm mesh screen. The dried samples were then packed 150.0 g by mass in labeled cylindrical plastic containers of uniform base diameter of 5.0 cm which could sit on the 7.6 cm by 7.6 cm NaI (Tl) detector. The plastic containers were tightly covered, sealed and left for 28 days prior to counting, for attainment of secular equilibrium between ^{238}U and ^{232}Th and their respective progenies (Papaefthymiou et al. 2007).

The method of gamma spectrometry was adopted for the analysis of the samples collected in order to obtain the activity concentrations of ^{40}K, ^{238}U and ^{232}Th. The spectrometer used was a Canberra lead shielded 7.6 cm $\times$ 7.6 cm NaI (Tl) detector coupled to a multichannel analyzer (MCA) through a preamplifier base. The system was calibrated using standard point sources of gamma emitting isotopes. The resolution of the detector was about 10% at 0.662 MeV of ^{137}Cs. The value was good enough for NaI detector to distinguish the gamma ray energies of most radionuclides in samples. For the analyses of ^{40}K, ^{238}U and ^{232}Th, the photo peak regions of ^{40}K (1.46 MeV), ^{214}Bi (1.76 MeV) and ^{208}TI (2.615 MeV) were used, respectively for the collected ten surface soil samples. The cylindrical plastic containers holding the samples were put to sit on the high geometry 7.6 cm $\times$ 7.6 cm NaI (TI) detector. High level shielding against the environmental background radiation was achieved by counting in a Canberra 10 cm thick lead castle. The counting of each sample was done for 10 hours. Spectral analyses were performed and the areas under the photo-peaks of ^{40}K, ^{238}U and ^{232}Th were analyzed using the multichannel analyzer system. The activity concentrations of the radionuclides were calculated based on the measured efficiency of the detector and the net count rate under each photopeak over a period of 10 hours using equation 1 (IAEA 1989).

$$A = \frac{N(E\gamma)}{\varepsilon(E\gamma)I\gamma Mtc} \qquad 1$$

Where: $N(E_\gamma)$ = Net peak area of the radionuclide of interest, $\varepsilon(E_\gamma)$ = Efficiency of the detector for the γ-energy of interest, I_γ = Intensity per decay for the γ- energy of interest, M = Mass of the sample, t_c = Total counting time in seconds (36000 s).

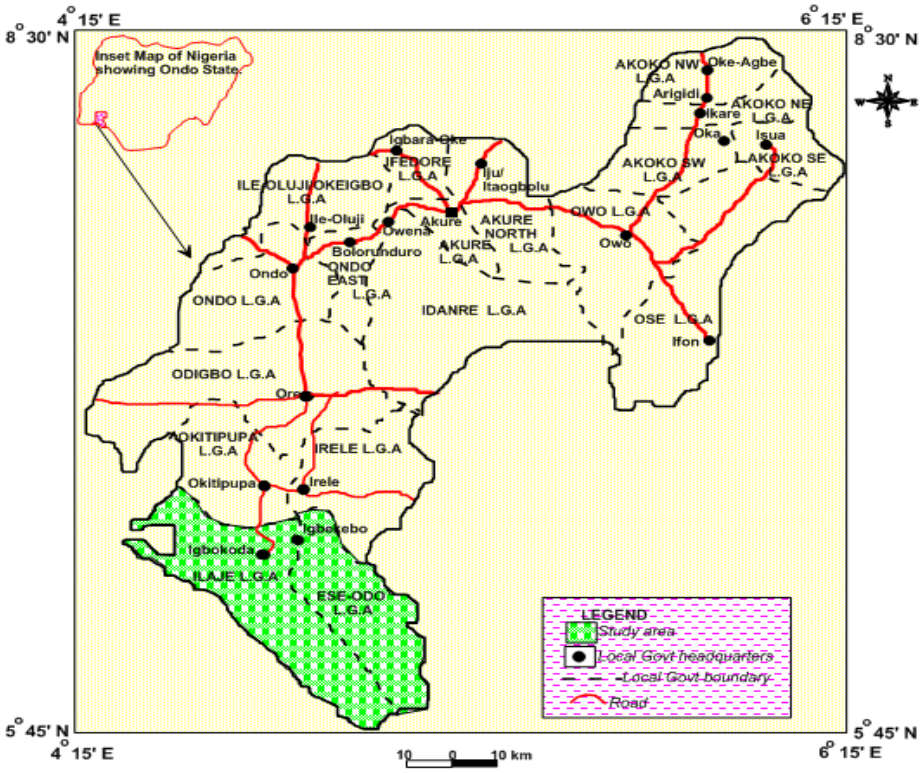

Figure 1: Map of Igbokoda study area in Ondo State (Source: Oparinde and Ojo 2014).

The radiological indicators which were parameters used to assess the radiological risks to individuals exposed to the soil samples were: radium equivalent activity (Ra_{eq}), internal hazard index (H_{in}), external hazard index (H_{ex}), annual effective dose equivalent (AEDE), excess lifetime cancer risk (ELCR), annual gonadal equivalent dose (AGED) and representative gamma index (Iyr).

***Radium equivalent activity (Ra_{eq})*:** This was calculated by the equation described by Beretka and Mathew (1985) and Yang et al. (2005) as indicated by equation 2.

$$Ra_{eq} = \frac{10}{130} C_k + \frac{10}{7} C_{Th} + C_{Ra} \qquad 2$$

Where: C_{Ra}, C_{Th} and C_k were the activity concentrations in Bq kg^{-1} of ^{226}Ra, ^{232}Th and ^{40}K, respectively.

***External hazard index (H_{ex})*:** Commonly used to evaluate radiation dose rate due to external exposure to gamma radiation from natural radionuclide in soil samples as reported by Hamzah et al. (2008) was presented in equation 3.

$$H_{ex} = \frac{Ck}{4810} + \frac{CTh}{259} + \frac{CRa}{370} \qquad 3$$

***Internal hazard index (H_{in})*:** H_{in} is a parameter for estimating the negative effect of radioactive materials on lungs and other respiratory organs. The risk internal exposure due to the natural radionuclides: ^{40}K, ^{226}Ra and

^{232}Th can be assessed from the value of H_{in} using equation 4.

$$H_{in} = \frac{Ck}{4810} + \frac{CTh}{259} + \frac{CRa}{185} \quad 4$$

***Absorbed dose rate (D)*:** The absorbed dose rate in air for external gamma radiation at about 1.0 m above the ground from the natural radionuclides D (nGy hr^{-1}) was given by UNSCEAR (2000):

$$D\ (nGy\ hr^{-1}) = 0.0417C_k + 0.604C_{Th} + 0.462C_{Ra} \quad 5$$

The absorbed dose rate in air was used to estimate the annual outdoor effective dose equivalent for individuals. The effective dose conversion factor was taken to be 0.7 Sv.Gy yr^{-1} and an outdoor occupancy factor of 0.2 with the annual occupancy time approximately 8760 hr yr^{-1}.

***Annual effective dose equivalent (AEDE)*:** This was estimated using equation 6 as expressed in UNSCEAR (2000).

$$AEDE\ (mSv\ yr^{-1}) = D\ (nGy\ hr^{-1}) \times 8760\ (hr\ yr^{-1}) \times 0.2 \times 0.7\ (Sv\ Gy^{-1}) \times 10^{-6} \quad 6$$

***Excess lifetime cancer risk (ELCR)*:** This was calculated based on the values of the annual outdoor effective dose equivalent as in Table 2 using the equation:

$$ELCR = AEDE \times LE \times RF \quad 7$$

where LE is life expectancy taken to be 70 years and RF is fatal risk factor per sievert which is 0.05 (ICRP 2007). According to UNSCEAR (1988) activity concentrations of radionuclides and their effects on bone marrow and the bone surface cells are considered as organs of interest.

***Annual Gonadal Equivalent Dose (AGED)*:** Radiological estimation parameter for the people using the soil of the study area as building material was calculated using the equation:

$$AGED\ (mSv\ yr^{-1}) = 3.09\ C_{Ra} + 4.18\ C_{Th} + 0.314\ C_k$$

where C_{Ra}, C_{Th} and C_k were the activity concentrations in Bq kg^{-1} of ^{226}Ra, ^{232}Th and ^{40}K, respectively.

***Representative Gamma Index (Iyr)*:** Representative Gamma Index was used to estimate the γ-radiation hazard associated with the natural radionuclides in specific investigated samples. OECD (1979) expressed representative gamma index as:

$$I_{yr} = \frac{Ck}{1500} + \frac{CTh}{100} + \frac{CRa}{150}$$

This gamma index was also used to correlate the annual dose rate due to the excess external gamma radiation caused by superficial materials. It was a screening tool for identifying materials that might become of health concern when used for construction (Tufail et al. 2007). Values of I yr ≤ 1 correspond to annual effective doses of less than or equal to 1 mSv, while values of I yr ≤ 0.5 correspond to annual effective doses less or equal to 0.3 mSv (Turhan and Gündüz 2008).

Results and Discussion

The activity concentrations of ^{40}K from the surface soil samples ranged from 285.32 ± 10.65 Bq kg^{-1} to 698.65 ± 12.04 Bq kg^{-1} with a mean value of 494.64 Bq kg^{-1}, ^{238}U ranged from 2.85 ± 2.06 Bq kg^{-1} to 33.24 ± 3.22 Bq kg^{-1} with a mean value of 19.76 Bq kg^{-1} and ^{232}Th ranged from 11.58 ± 2.37 Bq kg^{-1} to 52.36 ± 9.96 Bq kg^{-1} with a mean value of 31.98 Bq kg^{-1} as shown in Table 1. ^{40}K had the highest activity concentration while ^{238}U recorded the lowest value. The activity concentrations were within the range of values for normal background area (Akinloye and Olomo 1995). Radium equivalent activity (Ra_{eq}) ranged from 59.97 Bq kg^{-1} to 138.27 Bq kg^{-1} with a mean value of 104.92 Bq kg^{-1} which was higher than the mean value of 89.41 Bq kg^{-1} obtained by Najam and Younis (2015) for Iraq, both were below the world limit of 370 Bq kg^{-1} (OECD 1979, Beretka and Mathew 1985). Furthermore, the study made by Akinloye et al. (2012) on surface soil of Ore

metropolis, Ondo State, Nigeria gave similar result for radium equivalent activity ranging from 33.39 Bq kg^{-1} to 85.07 Bq kg^{-1}; though lower than the results obtained in this work. Moreso, the external hazard index (H_{ex}) ranged from 0.1621 Bq kg^{-1} to 0.3737 Bq kg^{-1} with a mean of 0.2836 Bq kg^{-1} as shown in Table 2. Furthermore, internal hazard index (H_{in}) ranged from 0.1796 Bq kg^{-1} to 0.4635 Bq kg^{-1} with mean value of 0.3370 Bq kg^{-1}. The values were within the limit of safety value of 1.0 Bq kg^{-1} recommended by European Commission (1999), ICRP (2007) and UNSCEAR (2000).

Table 1: Activity concentrations of natural radionuclides in soil samples

Sample	Activity concentration (Bq kg^{-1})		
	^{40}K	^{238}U	^{232}Th
SSS_1	432.34 ± 6.24	16.35 ± 3.42	45.49 ± 5.02
SSS_2	534.17 ± 8.35	27.81 ± 6.25	26.72 ± 6.18
SSS_3	332.76 ± 7.62	6.47 ± 0.28	19.53 ± 3.82
SSS_4	448.06 ± 10.24	22.35 ± 3.44	52.36 ± 9.96
SSS_5	698.65 ± 12.04	16.17 ± 4.07	22.87 ± 3.24
SSS_6	462.71 ± 8.93	24.32 ± 1.43	36.24 ± 5.75
SSS_7	542.58 ± 10.65	2.85 ± 2.06	11.58 ± 2.37
SSS_8	584.43 ± 15.18	33.24 ± 3.22	42.05 ± 6.15
SSS_9	625.34 ± 14.71	19.05 ± 2.62	26.93 ± 4.96
SSS_{10}	285.32 ± 10.65	28.96 ± 4.14	36.02 ± 3.53
Range	**285.32 – 698.65**	**2.85 – 33.24**	**11.58 – 52.36**
Mean	**494.64**	**19.76**	**31.98**

Note: SSS represents surface soil sample at 99% confidence interval

Table 2: Determined values of radiological indicators from surface soil samples

Sample	Ra_{eq} (Bq kg^{-1})	H_{in} (Bq kg^{-1})	H_{ex} (Bq kg^{-1})	D (nGy hr^{-1})	I_{yr} (mSv)	AGED (mSv yr^{-1})	AEDE (mSv yr^{-1})	ELCR x 10^{-3} (Outdoor)
SSS_1	114.59	0.3539	0.3097	53.06	0.8521	376.43	0.0651	0.2278
SSS_2	107.07	0.3645	0.2894	51.26	0.8087	365.35	0.0629	0.2200
SSS_3	59.97	0.1796	0.1621	28.66	0.4603	206.11	0.0352	0.1230
SSS_4	131.62	0.4161	0.3557	60.64	0.9713	428.62	0.0744	0.2603
SSS_5	102.58	0.3210	0.2773	50.42	0.8023	364.94	0.0618	0.2164
SSS_6	111.69	0.3676	0.3019	52.42	0.8330	371.92	0.0643	0.2250
SSS_7	75.42	0.2115	0.2038	36.98	0.5965	269.38	0.0454	0.1587
SSS_8	138.27	0.4635	0.3737	65.13	1.0317	461.99	0.0799	0.2796
SSS_9	105.66	0.3371	0.2856	51.16	0.8135	367.95	0.0628	0.2196
SSS_{10}	102.37	0.3549	0.2767	47.03	0.7435	329.64	0.0577	0.2019
Mean	**104.92**	**0.3370**	**0.2836**	**49.68**	**0.7913**	**354.23**	**0.0610**	**0.2132**

Note: The confidence of interval was 99%

The absorbed dose rate in air; ranged from 28.66 nGy hr^{-1} to 65.13 nGy hr^{-1} with a mean value of 49.68 nGy hr^{-1} which was less than the limit of 55.00 nGy hr^{-1} worldwide mean value as reported by UNSCEAR (2000). The Annual Gonadal Equivalent Doses **(**AGED) ranged from 206.11 Bq kg^{-1} to 461.99 Bq kg^{-1} with a mean of 354.23 Bq kg^{-1}. Representative Gamma Index (I_{yr}) ranged from 0.4603 to 1.0317 with mean value of 0.7913 in which the

values were within the recommended world limit of unity (ICRP 2007, Örgün et al. 2007) and the annual outdoor effective doses equivalent ranged from 0.0352 to 0.0799 mSv yr^{-1} with a mean value of 0.0610 mSv yr^{-1} which were below the limit of 1.0 mSv yr^{-1} (ICRP 2007). The excess lifetime cancer risk values for outdoor exposure ranged from 0.1230×10^{-3} to 0.2796×10^{-3} with mean value of 0.2132 x 10^{-3} which were below the world average of 1.45×10^{-3} (ICRP 2007, Qureshi et al. 2014). Similarly, Ramasamy et al. (2009) carried out the evaluation of (ELCR) in river sediments of Karnataka and Tamilnadu, India. The average of ELCR was found to be 0.20×10^{-3} which was less than the value obtained in this research work.

Conclusion

The radiological safety of the people within Igbokoda in Ondo State of Nigeria had been assessed and the results obtained for the radiological parameters were within the dose limits recommended. These showed that there were no significant radiological hazards to individuals from the exposure to natural radionuclides in surface soils from the study area. Also, crude oil exploration activity in the study area had shown that it had no negative radiological effect on the people in the study area. However, the results can serve as baseline for further research work to be done in future in line with the study area.

Conflict of Interest

The authors declare no conflict of interest.

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