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Assessment of Radiation Levels and Potential Radiological Hazards in the Soils of Riruwai Mining Area, North-Western Nigeria

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ABSTRACT

This research aimed to assess the levels of natural radionuclides in the soil of the Riruwai mining area and to evaluate the radiological hazards associated with them. A total of sixty soil samples were collected from four sampling sites during the dry and wet seasons. The concentrations of uranium-238 (238U), thorium-232 (232Th), and potassium-40 (40K) were measured using an HPGe gamma-ray spectrometer, and the measured concentrations were used to estimate the internal and external hazards and gamma dose rate. The results of the analysis indicated that the concentrations of 238U and 232Th in all sampling sites and seasons were higher than the world average. The radionuclide levels in the soils showed a significant seasonal variation, with higher concentrations obtained during the dry season. The correlation analysis revealed a very strong positive relationship between all the radionuclides, which indicates a common origin (parent material). The gamma dose rate computations revealed that only active and abandoned mining sites have a mean gamma dose rate greater than the tolerable limit of 59.00 nGy/h. The internal and external radiological hazards assessments showed that only the active mining sites had the potential to be hazardous to human health. The researchers, therefore, recommend that comprehensive monitoring of mining operations and public education about the dangers of radiation exposure in the study area be carried out.

تقييم مستويات الإشعاع والمخاطر الإشعاعية المحتملة في تربة منطقة ريرواي للتعدين، شمال غرب نيجيريا

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الكلمات المفتاحية:
معدل جرعة جاما
تعدين
االخطار اإلشعاعية
النويدات المشعة
ريرواي

الملخص

وكان الهدف من هذا البحث هو تقييم مستويات النويدات المشعة الطبيعية في تربة منطقة التعدين في ريرواي وتقييم المخاطر الإشعاعية المرتبطة بها. تم جمع ما مجموعه ستين عينة تربة من أربعة مواقع لأخذ العينات خلال المواسم الجافة والرطبة. تم قياس تركيزات اليورانيوم-238 (U238) والثوريوم-232 (Th232) والبوتاسيوم-40 (K40) باستخدام مطياف أشعة غاما HPGe، وتم استخدام التركيزات المقاسة لتقدير المخاطر الداخلية والخارجية ومعدل جرعة غاماأشارت نتائج التحليل إلى أن تركيزات U238 و Th232 في جميع مواقع ومواسم أخذ العينات كانت أعلى من المتوسط العالمي. وأظهرت مستويات النويدات المشعة في التربة تباينا موسميا كبيرا، مع الحصول على تركيزات أعلى خلال موسم الجفاف. وأظهرت مستويات النويدات المشعة في التربة تباينا موسميا كبيرا، مع الحصول على تركيزات أعلى خلال موسم الجفاف. كشف تحليل

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الارتباط عن وجود علاقة إيجابية قوية للغاية بين جميع النويدات المشعة ، مما يشير إلى أصل مشترك (المادة الأم). وكشفت حسابات معدل جرعة غاما أن مواقع التعدين النشطة والمهجورة هي وحدها التي لديها متوسط معدل جرعة غاما أكبر من الحد المسموح به البالغ 59.00 نانوغرام/ساعة. وأظهرت تقييمات المخاطر الإشعاعية الداخلية والخارجية أن مواقع التعدين النشطة هي وحدها التي يمكن أن تكون خطرة على صحة الإنسان. لذلك يوصي الباحثون بإجراء مراقبة شاملة لعمليات التعدين وتثقيف الجمهور حول مخاطر التعرض للإشعاع في منطقة الدراسة.

Introduction

Radioactivity is a natural phenomenon evidenced by the release of ionizing radiation that can be found in various quantities throughout the environment [1]. Radioactivity has become an important environmental concern around the world and has become a topic of great concern over the years due to its significant adverse effects on human health [2, 3]. Environmental radioactivity is mostly caused by cosmic rays and naturally occurring radionuclides found in the environmental compartments (water, soil, rock, and air). Radionuclides are responsible for around 86 % of the effective human dosage [3, 4]. These radionuclides consist predominately of the ^{238}U series, the ^{232}Th series, and the ^{40}K series, together with their decay products [5, 6]. Radiations emitted by radioactive materials and associated progenies are one of the most significant external sources of radiation exposure for human beings [7, 8]. Leukemia, a cancer of the blood cells, is one of the most common types of radiation-induced diseases. Excessive exposure to radionuclides can cause damage to the bone marrow, where blood cells are produced, leading to abnormal production of white blood cells. Symptoms of leukemia include weakness, fatigue, fever, and bleeding [9]. Aside from their possible ionizing impacts, radionuclides can be hazardous, undergo bio-concentration and bioaccumulation, and unfavorably affect human and ecological health [10]. The concentrations of radionuclides are extensively distributed across the earth's ecosystem and are largely determined by geology, soil properties, relative humidity, organic matter, soil pH, and geographical factors [4].

Soil is a vital component of the natural environment, and its quality is essential for sustaining Life on Earth. However, soil can also be vulnerable to radioactive pollution, which highlights the importance of monitoring and maintaining soil well-being [11]. The impacts of radioactive pollution on soil can be significant, leading to ecosystem disturbances and potential health risks for humans and animals [4]. The ability of soil to retain radionuclides is primarily due to its chemical properties, such as the presence of clay minerals and organic matter, which act as adsorption sites for radionuclide ions [12]. Soil acts as a channel for transferring radionuclides to plants, animals, and other environmental compartments; hence, it is the basic indicator of the radiological contamination of the environment [13]. The concentration of naturally occurring radioactive materials is generally low when undisturbed in the soil, but anthropogenic activities like mining and smelting processes have the potential to modify or increase their levels in the soil [14, 15]. Radionuclides in soils are absorbed by humans through the food chain via air dispersal, gravity settlement, plant absorption, and a variety of other geological factors [7]. The amount of radioactivity in the soil is affected not only by soil type and usage but also by seasonal variations [16]. It has been reported that radionuclide concentrations are found to be higher in the dry season compared to the wet season [17].

Mining has been identified as a possible source of naturally occurring radionuclide exposure. Workers in the mining and mineral processing sectors are exposed to a variety of radiation hazards, including external gamma radiation from ores, inhalation of dust containing long-lived alpha-emitting radionuclides, and inhalation of radon decay products [4]. Mining operations cause naturally occurring radioactive materials to pre-concentrate and accumulate in mine tailings and water bodies, which eventually expose people to high levels of radiation [8, 18]. Due to the health hazards connected with naturally occurring radioactive material exposure and inhalation of radon's short-lived decay products, international and governmental organizations have taken stringent steps to limit such exposure [4]. The assessment of radioactivity levels in the environment is absolutely essential in order to protect public health and the environment from the detrimental effects of radiation [19].

In Nigeria, the majority of mining activities are performed by small-scale artisanal miners (ASM); these local miners have little to no awareness of the environmental and health impacts of mining operations [20]. Unsophisticated mining practices expose miners and their surroundings to excessive amounts of radionuclides. Nigeria is estimated to have more than 1218 abandoned mine sites, ponds, and quarries that need to be reclaimed [21]. One of these abandoned sites is Riruwai, which is an old mining community that housed the first and largest underground tin mine in Nigeria. Large-scale mining began in 1979 when nearly nine hundred tons of tin and zinc ores were produced every day. The mining operations were officially stopped after 5 years of continuous activity and have been reopened by artisanal and illegal miners thereafter [22]. After the stoppage of mining, no remediation of the site has been carried out yet. The area has been recognized as one of Nigeria's biggest prospective uranium mining zones, with about two hundred million tons of rock containing over half a million tons of niobium and uranium [23]. A reconnaissance survey by the researcher has confirmed that several mining sites are located close to settlements and farmland where people are engaged in farming and other activities, and the artisanal miners in the study area worked long hours without adequate safety wear; some miners even stay overnight and eat at mining sites. The mining has resulted in the accumulation of various tailings heaps that have been randomly dispersed over large areas. These tailings are reprocessed and used as building materials, and the land is being used by locals for farming [24]. Communities living in this area are constantly in danger of being exposed to ionizing radiation emitted by natural radionuclides found in rocks, soils, and dust. Previous research from all over the world reported that people living near mining areas are at a significantly higher risk of being exposed to ionization radiation [25-28]. To reduce the negative health effects of radioactive intake and bridge the knowledge gap, it is critical to measure the level of radionuclide presence in soils. At present, and based on our findings, there is a scarcity of available data on this type of study in the study area. This has created the urgency of carrying out an in-depth investigation that will provide detailed radiological information and pave the way for assessing any future environmental contamination due to radionuclides. It is envisioned that this study will lay the groundwork for long-term management and restoration of the study area.

Materials and Methods

Description of the study area

Riruwai is a town in Kano State, Doguwa Local Government Area, in northern Nigeria. It is located between latitudes 10°43"97"N and 10°45'01"N and longitudes 8°43"3"E and 8°47'39"E, and it spans an area of 129 km^2 (Figure 1). According to the 2006 census, Riruwai has a population of 150,645 people [29]. Riruwai is one of Nigeria's newer granite complexes. It is bordered by metamorphic and calc-alkaline meta-igneous rocks that range in age from the Precambrian to the Cambrian [30]. Riruwai is principally a mining community. Mining on a large scale began in 1979, with roughly 900 tons of Zn-Sn ore produced each day. The mining operations were shut down after five years of operation. However, artisanal and small-scale mining operations continue in the region [22].

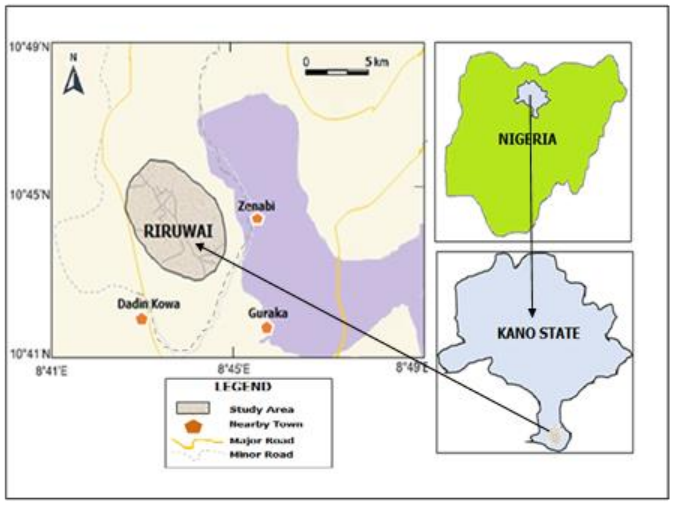

Fig. 1: Geographical Map of the Study Area

Soil Sampling and Collection

Sampling was carried out in two distinct seasons; the dry season (February and May 2020) and the wet season (July and August 2020). Sixty surface soil samples were collected from the top 0–20 cm layer at four sampling locations, including 18 from active mining sites, 12 from abandoned mine sites, 21 from farmlands, and 9 from control sites, using a steel soil Auger. Three (3) sub-samples were obtained at each sampling location within a 5-meter radius of the site in order to obtain representative and uniform samples. These sub-samples were then combined to form a composite sample. 1.00 kg of the mixed samples were selected by quartering, stored in a polyethylene bag, labeled, and transported to a laboratory for analysis [31].

Measurement of Radioactivity Concentrations

After the removal of extraneous materials such as stones, leaves, and roots, the collected soil samples were dried in an oven at 105 °C, ground, and sieved through 60 meshes to obtain homogeneous particles. 300 kg of samples were transferred into empty cylindrical plastic containers of uniform size (75 mm height by 70 mm diameter), sealed, and allowed to stay for 30 days in order to attain secular equilibrium between parent and daughter ^{238}U and ^{232}Th [32, 33]. Before the measurements of the sample, an empty container in similar conditions was used to record the background gamma radiation. This value was subtracted from the gamma-ray spectra of the samples examined in order to rectify the net peak areas for the counts and calculate radioactivity levels accurately. The activity concentrations of the radionuclide in each sample were measured using an ultra-low background High Purity Germanium (HPGe) gamma-ray spectrometer.

The gamma-ray spectrometer comprises a co-axial HPGe detector with a relative efficiency of 50.00% equipped with a 32-k multichannel analyzer. The data acquisition and analysis were performed with the use of Genie 2000 software (Canberra, USA). The detector contained 10.00 cm of lead shielding to reduce the background radiation. The activity of ^{238}U was computed as the mean of the activities obtained from the gamma-ray lines ^{214}Pb (295.30 and 352.00 keV) and ^{212}Bi (609.50, 1120.20, and 1764.5 keV) isotopes. The activity of ^{232}Th was obtained by the gamma-ray lines of ^{228}Ac (911.1 and 968.80 keV), and the activity of ^{40}K was obtained from the gamma-ray line of 1460.80 keV.

Estimation of Radiological Hazards

Gamma Dose Rate (GDR)

The GDR (nGy/h) was computed using the following relation:

$$GDR = 0.462 \times C_{U-238} + 0.604 \times C_{Th-232} + 0.462 \times C_{K-40} \quad 1$$

Where: $C_{U\text{-}238}$, $C_{Th\text{-}232}$ and $C_{K\text{-}40}$ are the activity concentrations of ^{238}U, ^{232}Th and ^{40}K respectively in soils [4].

Internal Hazard Index (H_{in})

The internal radiation exposure is computed using the following equation:

$$H_{in} = \frac{C_U}{185} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \quad 2$$

Where, C_U, C_{Th} and C_K are the measured concentrations of ^{238}U, ^{232}Th and ^{40}K respectively in Bq/kg [4]

External Hazard Index (H_{ex})

The external hazard (H_{ex}) index is another radiation hazard index defined by Beretka and Mathew [34] to evaluate the indoor radiation dose rate due to the external exposure to gamma radiation from the natural radionuclides in the building materials of dwellings. The external hazard index (H_{ex}) is used to quantify the radiation hazard caused by gamma-ray exposure. Hex was calculated using equation (3).

$$H_{ex} = \frac{C_U}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \quad 3$$

Where, C_U, C_{Th} and C_K are the measured concentrations of ^{238}U, ^{232}Th and ^{40}K respectively in Bq/kg. The H_{in} and H_{ex} value greater than one may cause significant health hazards [4].

Quality control and statistical analyses

All data were treated as the mean ± standard deviation of three replicates. The calibration of the HPGe gamma-ray spectrometer was carried out using International Atomic Energy Agency (IAEA) standard reference materials, i.e., RGU-1, RGTh-1 and RGK-1, for the quantitative determination of ^{238}U, ^{232}Th, and ^{40}K of the soil samples, respectively. One-way analysis of variance (ANOVA), independent t-test, and Pearson correlation analysis was performed using SPSS 23.0 (SPSS Inc., Chicago, USA). The ANOVA was used to compare radionuclide contents from the different sampling sites, and statistical significance was established using the Tukey HSD post-hoc test at $p \leq 0.05$ levels. The independent t-test was used to identify whether there are significant differences in radionuclide concentrations between dry and wet seasons, while correlation analysis was employed to identify the relationship between radionuclides in the soils. All the graphs were plotted using Microsoft Excel 2013.

Results and Discussion

Results

The concentrations of ^{238}U, ^{232}Th and ^{40}K in the soils of the Riruwai mining area during the dry and wet seasons are presented in Tables 1 and 2.

Table 1: Radionuclide Concentrations (Bq/kg) in Soils of Riruwai Mining Area during the Dry Season

Dry Season					
Radionuclides	Sampling Location				UNSCEAR (2000)
	RSSI	RSS2	RSS3	RSS4	
^{238}U	134.77^{a} ± 2.15	98.51^{a} ± 1.30	56.11^{c} ± 1.11	27.92^{d} ± 0.97	35.00
^{232}Th	110.66^{a} ± 1.34	80.91^{b} ± 0.60	38.51^{c} ± 1.45	18.41^{d} ± 1.34	35.00
^{40}K	296.87^{a} ± 3.93	239.15^{b} ± 1.18	157.03^{c} ± 1.37	84.13^{d} ± 1.51	400.00

Values are mean ± standard deviation (n = 3). RSS1 = active mining sites, RSS2 = abandoned mining sites, RSS3 = farmland, RSS4 = control site. The values on the same row with the same superscript letters are statistically the same ($p > 0.05$), whereas values on the same row with different superscript letters are statistically different ($p \leq 0.05$) as revealed by one-way ANOVA and the Tukey HSD post-hoc test.

Table 2: Radionuclide Concentrations (Bq/kg) in Soils of Riruwai Mining Area during the Wet Season

Radionuclides	Sampling Location				UNSCEAR (2000)
	RSS1	RSS2	RSS3	RSS4	
^{238}U	97.16^{a} ± 1.16	75.21^{a} ± 0.75	49.40^{b} ± 1.53	20.13^{b} ± 0.12	35.00

^{232}Th	85.09^{a} ± 1.12	73.94^{b} ± 1.76	37.56^{c} ± 2.39	15.38^{d} ± 0.96	35.00
^{40}K	243.83^{a} ± 1.31	205.71^{b} ± 2.10	129.03^{c} ± 2.09	66.45^{d} ± 1.02	400.00

Values are mean ± standard deviation (n = 3). RSS1 = active mining sites, RSS2 = abandoned mining sites, RSS3 = farmland, RSS4 = control site. The values on the same row with the same superscript letters are statistically the same ($p > 0.05$), whereas values on the same row with different superscript letters are statistically different ($p \leq 0.05$) as revealed by one-way ANOVA and the Tukey HSD post-hoc test.

The concentrations of uranium-238 as shown in Tables 1 and 2 ranged from 27.92 (control site) to 134.77 Bq/kg (active mining site), such that 56.11 Bq/kg (farmland) and 98.51 Bq/kg (abandoned mining site) were found between the lowest and highest experimental values in the dry season. Levels of uranium-238 in active mining sites (134.77 Bq/kg), abandoned mining sites (98.51 Bq/kg), and farmland (56.11 Bq/kg) were found to be greater than the threshold value of 35.00 Bq/kg recommended by UNSCEAR [4]. The control value of 27.92 Bq/kg is, however, lower than the threshold value. During the wet season, the concentrations of uranium-238 ranged from 20.13 Bq/kg (control site) to 97.16 Bq/kg (active mining site). The levels of 37.56 mg/kg (farmland) and 98.51 Bq/kg (abandoned mining sites) fell between the lowest and highest values of uranium-238 determined. Except for the control site, the levels of uranium-238 in all the sampling locations were above the UNSCEAR [4] threshold limit. A one-way ANOVA revealed a significant difference in radionuclide concentrations across the four sampling locations. Orosun et al. [35] observed a similarly high concentration of uranium-238 above the UNSCEAR [4] recommended limit when they studied the natural radionuclides and radiological risk assessment of a granite mining field in Asa, North Central Nigeria. High mean uranium-238 concentrations of 78.00 22 ± 3.00 Bq/kg, comparable to this study, were also reported in soils around the Arufu mining area in Taraba State, North-Eastern Nigeria [36]. Moreover, a comparable value of 79.3 Bq/kg was also recorded by Yang and Sun [37] when they measured the natural radioactivity of the surface soil in Guangdong, China. Uranium exposure may cause health risks because of its chemotoxicity and radiotoxicity. Many epidemiological and laboratory studies have demonstrated that environmental and occupational uranium exposure can induce multifarious health problems. For example, when Zhivin et al. [38] investigated the health impacts of occupational uranium exposure among veterans of the Persian Gulf and Balkan wars, they observed some signs of an elevated risk of lung cancer among uranium-processing workers.

The measured concentrations of thorium-232 varied from 18.41 (control site) to 110.66 Bq/kg (active mining site) during the dry season. The concentrations of 38.51 Bq/kg (farmland) and 80.91 Bq/kg (abandoned mining site) lay between the lowest and highest thorium-232 values obtained. The concentrations of thorium-232 in active mining sites, abandoned mining sites, and farmland were above the recommended value of 35.00 Bq/kg set by UNSCEAR [4]. The concentration of 18.41 Bq/kg in the control site was, however, below the recommended value. During the wet season, the concentrations of thorium-232 (Bq/kg) ranged from 15.38 (control site) to 85.09 (active mining site), when 37.56 Bq/kg (farmland) and 73.94 Bq/kg (abandoned mining site) fell between the extreme experimental values. The experimental values of thorium-232 in the present study were slightly higher than the reported literature values of 67.00 ± 1.00 Bq/kg determined in soils around gold mines in Zamfara State, Northwest Nigeria [36], but comparable to the average thorium-232 value of 154 ± 56 Bq/kg measured in farmland soil from Bukuru, Jos, North- Central Nigeria by Jibiri et al. [39]. Thorium is a well-known radioactive and chemically hazardous environmental pollutant. Continuous thorium exposure may increase the chance of developing lung and liver illnesses as well as lung and pancreatic cancer [37].

The concentration of potassium-40 spread from 84.13 to 296.87 Bq/kg during the dry season. The highest concentration (296.87 Bq/kg) was observed at the active mining site, while the lowest concentration (84.13 Bq/kg) was found at the control site. The experimental values of 157.03 Bq/kg (farmland) and 239.15 Bq/kg (abandoned mining site) fell between the lowest and highest observed concentrations. During the wet season, the levels of potassium-40 varied from 66.45 to 243.83 Bq/kg, such that 129.03 Bq/kg (farmland) and 205.71 Bq/kg (abandoned mining sites) fell between the lowest and highest observed values.

The measured concentrations of potassium-40 in all the sampling sites and seasons were lower than the UNSCEAR [4] threshold concentration of 400.00 Bq/kg. The experimental values of potassium-40 were comparable to the literature value of 277.81 ± 5.77 Bq/kg reported by Beogo et al. [40] and lower than the value of 570.91 ± 6.0 reported by Orosun et al. [35]. Laniyan and Adewumi [36] also found a potassium-40 content of 314.11 Bq/kg in the soils of the Arufu mining area, Taraba State, North-East Nigeria, which was relatively similar to the experimental values of 296.87 Bq/kg and 239.15 Bq/kg found in active and abandoned mining sites, respectively, during the dry season. Similarly, a mean concentration of 218.93 Bq/kg, close to that of the present study, was also reported in the farmland of Barkin Ladi, Plateau State, Nigeria [41]. Anthropogenic activities such as mining and smelting have resulted in an increase in potassium-40 radionuclides in the environment. According to reports, exposure to high concentrations of potassium-40 poses serious health concerns [42].

Correlation Analysis

Table 3 shows the results of Pearson's correlation analysis of radionuclides (^{238}U, ^{232}Th and ^{40}K) in the soils of the Riruwai mining area during the dry and wet seasons.

Table 3: Correlation between Radionuclides of Soil during the Dry and Wet Seasons

Dry season				Wet season			
	^{238}U	^{232}Th	^{40}K		^{238}U	^{232}Th	^{40}K
^{238}U	1			^{238}U	1		
^{232}Th	0.999	1		^{232}Th	0.987	1	
^{40}K	0.994	0.991	1	^{40}K	0.996	0.997	1

The ^{238}U, ^{232}Th and ^{40}K exhibit very strong positive correlations with one another during the dry and rainy seasons.

Gamma Dose Rate

Figure 2 shows the values of gamma dose rate (GDR) in active mining sites (RSS1), abandoned mining sites (RSS2), farmland (RSS3), and control sites (RSS4) during the dry and wet seasons.

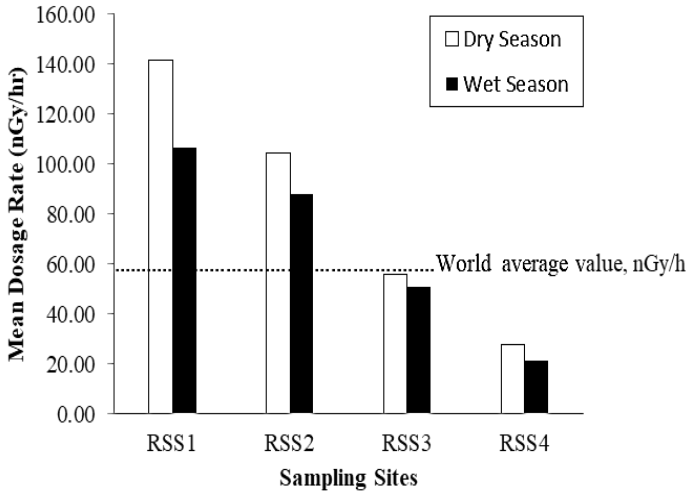

Fig. 2: Gamma dose rate (GDR) in soils of the Riruwai mining area during the dry season wet seasons

The values of GDR in RSS1, RSS2, RSS3, and RSS4 are 141.48, 104.35, 55.73, and 27.53 nGy/h, respectively. During the wet seasons, the values of GDR are 106.45, 87.98, 50.89, and 21.36 nGy/h for RSS1, RSS2, RSS3, and RSS4, respectively. GDR levels in RSS2 and RSS2 were significantly higger than the global average of 59.00 nGy/h in both seasons, whereas values in RSS3 and RSS4 were lower than the global average value. Sanusi et al. [43] and Mukhtar et al. [44] both

observed comparable higher GDR values of 182 nGy/h and 116 nGy/h, respectively.

Internal hazard (H_{in}) and external hazard (H_{ex})

The values of internal (H_{in}) and external (H_{ex}) hazards in active mining, abandoned mining, farmland, and control sites during the dry and wet seasons are presented in Figure 3.

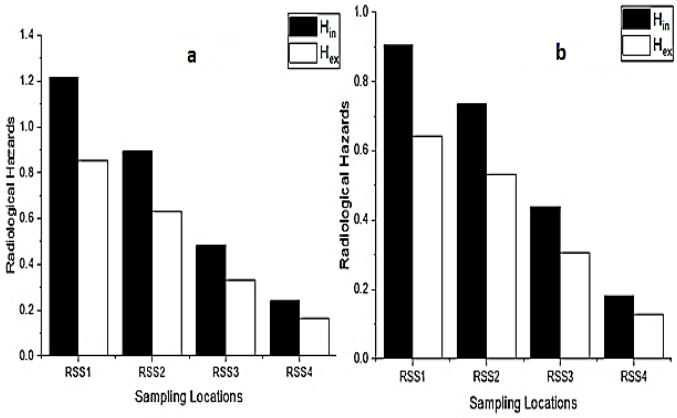

Fig. 3: Internal (H_{in}) and External (H_{ex}) Hazards of Radionuclides in Soils during (a) dry Season (b) Wet season

Discussion

Mining activities have contributed to the distribution and uptake of radionuclides in the soil. This has led to an increase in their concentrations in the environment and increased human health risks. The findings of this study revealed that the concentrations of ^{238}U, ^{232}Th, and ^{40}K varied significantly ($p \leq 0.05$) along the sampling locations, with levels of ^{238}U and ^{232}Th above the UNSCEAR [4] recommended limits in all the sampling locations except for the control sites. Orosun et al. [35] observed similar high concentrations of ^{238}U and ^{232}Th above the UNSCEAR [4] recommended limit when they studied the natural radionuclides and radiological risk assessment of a granite mining field in Asa, North Central Nigeria. Higher concentrations of those radionuclides above the threshold limits were also reported in soils around gold mines in Zamfara State, Northern Nigeria [36]. Moreover, Cao et al. [45] reported similar higher concentrations of ^{238}U and ^{232}Th above the threshold limits when they studied the distribution pattern of radionuclides in the soils of mainland China. Higher concentrations of ^{238}U and ^{232}Th above the threshold limit in active mining sites, abandoned mining sites, and farmland could be attributed to the fact that mining activities and farming Mining involve the excavation of deep soil, which exposes natural radionuclides and their corresponding decay products that were already buried beneath the ground to the earth's surface, hence increasing the radiation level of the impacted area [41, 46]. Underlying geology could be another factor that would result in an elevated level of ^{238}U and ^{232}Th in an environment. It has been documented that environments with granitic geological formations are linked to higher gamma amount of radiation rates because of the substantial silica content and radioactive materials of granite rock, whereas locations with sedimentary rock origins have a lower terrestrial gamma radiation dose rate [4]. The study area is geologically characterized by several types of underlying granitic rock formations. Therefore, the high level of radionuclides found in the area could be attributed to the geological formations beneath it. Moreover, the variation of radionuclide concentrations across the sampling locations could be due to a number of factors, including geology, soil type and features, and the application of fertilizer [47]. For example, the application of fertilizer has been reported to increase the accumulation of radionuclides in the soil [48]. The existence of the uranium series and associated decay products in phosphate fertilizers is well documented [47]. Aközcan et al. [48] reported that the radioactivity levels of ^{238}U, ^{232}Th, and ^{40}K have increased with fertilization applications when they studied dose rates and seasonal variations of ^{238}U, ^{232}Th, ^{226}Ra, ^{40}K, and ^{137}Cs radionuclides in soils along the Thrace, Turkey.

The concentrations of radionuclides in the soil are seasonally varied, with higher values obtained during the dry seasons. The difference was, however, statistically not significant, as obtained from an independent t-test result. In all the sampling locations, the concentrations of ^{238}U, ^{232}Th, and ^{40}K were significantly higher during the dry season. High concentrations of radionuclides during the dry season could be attributed to the release of radionuclides because, in the dry season, gases move upward, carrying radionuclides with them and thereby increasing ionization near the soil surface [16]. The decrease in radionuclide levels during the wet season could be due to high soil water content, as a result of reduced rates of gas diffusion. This characteristic influences radiation spread in soils, where water molecules likely form a thin physical shield against gamma-ray detection during the rainy season, which disappears during the dry season [49]. Tchorz-Trzeciakiewicz et al. [50] confirmed that higher water content in the soil during the rainy season could restrict the emanation of gamma rays and hence, lower the concentration of radionuclides.

The radionuclides show very strong positive correlations with one another in both the dry and rainy seasons. This demonstrates that these radionuclides may have come from similar natural sources (parent materials). Several studies have confirmed that naturally occurring radionuclides are originated from parent material that is, granitic rocks [37, 51].

The results of GDR show that RSS1 and RSS2 have higher values than the world average value of 59.00 nGy/h in both dry and wet seasons. This demonstrates that the soils from the aforementioned locations have a very high radiation rate and are therefore not suitable for farming and other anthropogenic activities. The GDR values are found to be significantly higher in the dry season compared to the wet season in all the sampling locations. Lower GDR values during the wet season could be due to the high moisture of the soil, which decreases the emanation of gamma radiation [50]. Melintescu et al. [52] and Barbosa et al. [53] reported similar low GDR values during the wet season compared to the dry season.

The internal hazard (H_{in}) and external hazard (H_{ex}) are used to evaluate the radiological hazards in this study. The H_{in} values of radionuclides are less than one in all sampling sites and seasons, with the exception of active mining sites. This demonstrates that only the soil from active mining sites may be hazardous to human health. H_{in} seeks to assess the internal exposure to radon and its short-lived progenies, which are perilous to the respiratory organs. For safety precautions, the H_{in} should be less than or equal to one [54, 55]. The H_{ex} of values of radionuclides at all the sampling sites and seasons is less than one. Similar observations were recorded by Alomari et al. [13], Orosun et al. [35], and Hassan et al. [56]. The estimated values of H_{ex} suggest that the soils from the Riruwai mining area may be used for building and construction purposes. H_{ex} is used as a radiation exposure index to assess the indoor radiation dose rate in relation to external exposure to gamma radiation from natural radionuclides [39, 57, 58].

Conclusion

The natural radioactivity levels of ^{238}U, ^{232}Th and ^{40}K were measured, and the results of the analysis revealed that the concentrations of ^{238}U and ^{232}Th in all sampling sites and seasons were higher than the world value, while the concentrations of ^{40}K were lower than the world average value. The levels of radionuclides in the soils show a significant seasonal variation, with higher concentrations obtained during the dry season. According to the internal and external radiological risk assessments, only active mining sites have the potential to be hazardous to human health, and the soils in the study area could be used for farming and construction. The information obtained in this study would serve as baseline radiological data for future reference in the study area and similar locations.

References

[1] Beogo, C.E., Cisse, O.I., Zougmore, F. (2022). Assessment of Radiological Hazards from Soil Samples in the Northeastern area of Burkina Faso., SN Appl. Sci., **4**, 73. DOI: 10.1007/s42452-022-04960-x.

[2] Gulan, L., Milenkovic, B., Zeremski, T., Milic, G., & Vuckovic, B. (2017). Persistent Organic Pollutants, Heavy Metals and Radioactivity in the Urban Soil of Priština City,

Kosovo and Metohija., Chemosphere., **171**, 415–426. DOI: 10.1016/j.chemosphere.2016.12.064.

[3] Filgueiras, R. A., Silva, A. X., Ribeiro, F. C. A., Lauria, D. C., Viglio, E. P. (2020). Baseline, Mapping and Dose Estimation of Natural Radioactivity in Soils of the Brazilian State of Alagoas. Radiat. Phys. Chem., **167**, 108332. DOI: 10.1016/j.radphyschem.2019.05.022.

[4] UNSCEAR, 2000. Sources, effects and risks of ionizing radiation. United Nations Scientific Committee on the effects of Atomic Radiation. Exposures from natural sources, 2000 report to General Assembly, Annex B. New York, United Nations Publications.

[5] Durusoy, A., Yildirim, M. (2017). Determination of Radioactivity Concentrations in Soil Samples and Dose Assessment for Rize Province, Turkey. J. Radiat. Res. Appl. Sci., **10**(4), 348–352. DOI: 10.1016/j.jrras.2017.09.005.

[6] Guagliardi, I., Caloiero, T., Infusino, E., Callegari, G., Ricca, N. (2021). Environmental Estimation of Radiation Equivalent Dose Rates in Soils and Waters of Northern Calabria (Italy). Geofluids, **1**, 1-8.

[7] Ademola, A.K., Bello, A.K., Adejumobi, A.C. (2014). Determination of Natural Radioactivity and Hazard in Soil Samples in and Around Gold Mining Area in Itagunmodi, South Western, Nigeria. J. Radiat. Res. Appl. Sci., **7**, 249 55. DOI: 10.1016/j.jrras.2014.06.001.

[8] Ladan, S.C., Mustapha, I.M., Hassan, A., Aliyu, M.S., Tukur, S.R. (2022). Assessment of Radioactivity Concentration Level in Soil Samples of some Gold Mining Areas of Shiroro, Niger State, Nigeria. J. Radiat. Nucl. Appl., **7**(2), 1-6.

[9] Faanu, A., Adukpo, O.K., Tettey-Larbi, H. Lawluvi, D. O. Kpeglo, E. O., Darko, G. et al. (2016). Natural Radioactivity Levels in Soils, Rocks and Water at a Mining Concession of Perseus Gold Mine and Surrounding Towns in Central Region of Ghana. Springer Plus, **5**, 98. DOI: 10.1186/s40064-016-1716-5.

[10] Hassona, R. K., Sam, A. K., Osman, O. I., Sirelkhatim, D. A., LaRosa, J. (2008). Assessment of Committed Effective Dose Due to Consumption of Red Sea Coral Reef Fishes Collected From the Local Market (Sudan). Sci. Total Environ, **393**(3), 214-218. DOI: 10.1016/j.scitotenv.2007.12.014.

[11] Baborowski, M., Büttner, O., Morgenstern, P., Jancke, T., Westrich, B. (2012). Spatial Variability of Metal Pollution in Groyne Fields of the Middle Elbe-Implications for Sediment Monitoring. Environ. Pollut., **167**, 115-123. DOI: 10.1016/j.envpol.2012.03.046.

[12] El-Gamal, H., Maher., Hussien, T., Saleh, E.E. (2019). Evaluation of Natural Radioactivity Levels in Soil and Various Foodstuffs from Delta Abyan, Yemen. J. Radiat. Res. Appl. Sci., **12**(1), 226–233. DOI:10.1080/16878507.2019.1646523.

[13] Alomari, A.H., Saleh, M.A., Hashim, S., Alsayaheen, A.. Abukashabeh, A. (2019). Statistical Relationship between Activity Concentrations of Radionuclides ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs and Geological Formations in Surface Soil of Jordan. Isotopes Environ. Health Stud. **55**(2), 211-226. DOI: 10.1080/10256016.2019.1581776.

[14] Gulan, L., Milenkovic, B., Stajic, J.M., Vuckovic, B., Krstic, D., Zeremskic, T. et al. (2013). Correlation between Radioactivity Levels and Heavy Metal Content in the Soils of the North Kosovska Mitrovica Environment. Environ. Sci. Process. Impacts, **15**, 1735.

[15] Gbadamosi, M. R., Abayomi, A. A., Afolabi, T. A., Adegboye, M. A., Bakare, H. O. and Banjoko, O.O. et al. (2021). Pollution Sources Identification, Health, and Radiological Risk Assessment of Naturally Occurring Radioisotopes and Heavy Metals in Waste DumpSites in Ijebu-Ode, Ogun State, Southwest Nigeria. Environ. Forensics, **22**, 1-18. DOI: 10.1080/15275922.2021.2006365.

[16] Guagliardi, I., Rovella, N., Apollaro, C., Bloise, A., De Rosa, R., Scarciglia, F. et al. (2016). Modelling Seasonal Variations of Natural Radioactivity in Soils: A Case Study in Southern Italy. J. Earth Syst. Sci., **125**, 1569–1578. DOI: 10.1007/s12040-016-0758-y.

[17] Maxwell, O., Olusegun, O.A., Joel, E.S., Ijeh I.B., Mary, A.T., Emeka, E.O. et al. (2021). Measurements of Seasonal Variations of Radioactivity Distributions in Riverine Soil Sediment of Ado-Odo Ota, South-West Nigeria: Probabilistic Approach Using Monte Carlo, Radiat. Prot. Dosimetry, **193**(2), 76–89. DOI:10.1093/rpd/ncab027.

[18] Innocent, A. J., Onimisi, M. Y., Jonah, S. A. (2013). Evaluation of Naturally Occurring Radionuclide Materials in Soil Samples Collected from Some Mining Sites in Zamfara State, Nigeria. British J. Appl. Sci. & Tech., **3**(4), 684-692. DOI:10.9734/BJAST/2013/3244.

[19] Botwe, B.O., Schirone, A., Delbono. I., Barsanti, M., Delfanti, R., Kelderman, P. et al. (2017). Radioactivity Concentrations and their Radiological Significance in Sediments of the Tema Harbour (Greater Accra, Ghana). J. Radiat. Res. Appl. Sci., **10**, 63-71. DOI: 10.1016/j.jrras.2016.12.002

[20] MMSD, 2017. Nigeria's Mining and Metal Sector. Federal Republic of Nigeria, Abuja, Nigeria.

[21] Mallo, I.I.Y. (2010). Environmental, Health and Socio-Economic Implications of Solid Minerals Mining in Nigeria. African Journal of Environmental Pollution and Health, **8**(1), 62-72.

[22] Attahiru, A. M. (2017). Assessment of Radiological Hazards around Riruwai tin mines, Kano State, North Western Nigeria. Unpublished doctoral dissertation. Ahmadu Bello University Zaria, Zaria, Kaduna State.

[23] Abdu, N. M., Aznan F. I., Nura, N. G. (2022). Annual Effective Dose Associated with Radioactivity in Drinking Water from Tin Mining Areas in North-Western Nigeria. J. Radiat. Res. Appl. Sci., **15**(3), 96–102. DOI: 10.1016/j.jrras.2022.06.008.

[24] Ismaila, A., Aliyu, A.S., Ibrahim, Y.V. (2022). Evaluation of Gamma Radiation Dose Level in Mining Sites of Riruwai, Kano, Nigeria. Commun. Physic. Sci., **8**(1), 101-108.

[25] Ibrahim, U., Akpa, T.C., Daniel, I.H. (2013). Assessment of Radioactivity Concentration in Soil of Some Mining Areas in Central Nasarawa State, Nigeria. Sci. World J., **8**(2), 1-12.

[26] Ahmad, A.Y., Al-Ghouti, M.A., AlSadig, I., Abu-Dieyeh, M. (2019). Vertical Distribution and Radiological Risk Assessment of ^{137}Cs and Natural Radionuclides in Soil Samples. Sci. Rep., **9**, 12196. DOI: 10.1038/s41598-019-48500-x.

[27] Akpanowo, M., Umaru, I., Iyakwari, S., Joshua, E.O., Yusuf, S., Ekong, G.B. (2020). Determination of Natural Radioactivity Levels and Radiological Hazards in Environmental Samples from Artisanal Mining Sites of Anka, North-West Nigeria. Sci. African, 10: 1-11. DOI:10.46266/njes.1009.

[28] Huang, Y.J., Yang, L.T., Zhao, F. Guo, G., Wu, L. (2021). Spatial Distribution and Characteristic of Radiological Hazard of the Paddy Field around Decommissioned Uranium Mine in Eastern China. J. Radioanal. Nucl. Chem., **327**, 789–799.

[29] NPC, 2006. Population and Housing Census, "Population Distribution by Sex, State, LGA and Senatorial District. Federal Republic of Nigeria.

[30] Olasehinde, A., Ashano, E.C., Singh, G.P. (2012). Analysis of Magnetic Anomaly over the Riruwai Younger Granite Ring Complex: A Geodynamic Implication. Cont. J. Earth Sci., **7**(1), 9 – 18. DOI:10.5707/cjearthsci.2012.7.1.9.18.

[31] Fan, S., Wang, X. (2017). Analysis and Assessment of Heavy Metals Pollution in Soils around Pb and Zn Smelter in Baoji City, Northwest China. Hum. Ecol. Risk Assess., **23**(5), 1099-1120. DOI:10.1080/10807039.2017.1300857.

[32] Boukhenfouf, W. and Boucenna, A. (2011). The Radioactivity Measurements in Soils and Fertilizers using Gamma Spectrometry Technique. J. Environ. Radioact., **102**(4), 336–339. DOI:10.1016/j.jenvrad.2011.01.006.

[33] Khandaker, M.U., Asaduzzaman, K., Nawi, S.M., Usman, A.R., Amin, Y.M., Daar, E. et al. (2015). Assessment of Radiation and Heavy Metals Risk due to the Dietary Intake of Marine Fishes (Rastrelliger Kanaguta) from Straits of Malacca. PLoS ONE Journal, **10**(6), 1-16. DOI:10.1371/journal.pone.0128790.

[34] Beretka, J., Matthew, P.J. (1985). Natural Radioactivity of Australian Building Materials, Industrial Wastes and By-Products. Health Phys, **48**, 87-95. DOI: 10.1097/00004032-198501000-00007.

[35] Orosun, M.M., Usikalub, M.R., Oyewumia, K.J., Adagunodob. T.A. (2019). Natural Radionuclides and Radiological Risk Assessment of Granite Mining Field in Asa, North-central Nigeria. Methods X, **6**, 2504–2514. DOI:10.1016/j.heliyon.2020.e04240.

[36] Laniyan, T.A., Adewumi, A.J. (2021). Health Risk Profile of Natural Radionuclides in Soils, Sediments, Tailings and Rocks Around Mining Sites in Nigeria. Environ. Earth Sci., **80**, 375. DOI:10.1007/s12665-021-09674-8.

[37] Yang, J., Sun, Y. (2022). Natural Radioactivity and Dose Assessment in Surface Soil from Guangdong, a High Background Radiation Province in China. J. Radiat. Res. Appl. Sci., **15**, 145–151. DOI: 10.1016/j.jrras.2022.01.019.

[38] Zhivin, S., Laurier, D., Guseva Canu, I. (2014). Health effects of occupational exposure to uranium: do physicochemical properties matter? Int J Radiat Biol., **90**(11),1104–1113.DOI:10.3109/09553002.2014.943849.

[39] Jibiri, N.N., Alausa, S.K., Farai, I.P. (2009). Radiological Hazard Indices due to Activity Concentrations of Natural Radionuclides in Farm Soils from Two High Background Radiation Areas in Nigeria. Int, J. Low Radiat., **6**(2), 79–95. DOI:10.1016/j.jenvrad.2006.12.011.

[40] Beogo, C.E., Cisse, O.I., Zougmore, F. (2022). Assessment of Radiological Hazards from Soil Samples in the Northeastern Area of Burkina Faso. SN Appl. Sci. **4**, 73. DOI: 10.1007/s42452-022-04960-x.

[41] Jwanbot, D.I., Izam, M.M., Nyam, G.G., John, H.N. (2013). Radionuclides Analysis of Some Soils and Food Crops in Barkin Ladi LGA, Plateau State-Nigeria. J. Environ. Earth Sci., **3**(3), 79-86.

[42] Aborisade M. A, Gbadebo A.M., Adedeji O.H, Okeyode I.C., Ajayi O.A. (2018). Excess lifetime Cancer risk and Radiation Pollution hazard indices in rocks and soil of some selected mining sites in Nasarawa State, Nigeria. AEgean J. Environ. Sci., **3** (2017-2018), 1-18.

[43] Sanusi, M. S. M., Ramli, A. T., Gabdo, H. T., Garba, N. N.,Heryanshah, A., Wagiran, H., et al. (2014). Isodose mapping of terrestrial gamma radiation dose rate of Selangorstate, Kuala Lumpur and Putrajaya, Malaysia. J. Environ. Radioact. **135**: 67–74. DOI:10.1016/j.jenvrad.2014.04.004.

[44] Mukhtar, A., Garba, N. N., Rabiu, N., Muneer, A. S., Bello,S., and Mayeen, U. K. (2021). Statistical analysis of terrestrial gamma radiation dose rates in relation to different geological formations and soil types of Katsina State, Nigeria. Int. J. Environ. Anal. Chem. **102**: 2970-2978. DOI: 10.1080/03067319.2021.1905806.

[45] Cao, L., Yang, Y., Zhang, Y., Zheng, Y., Yang, T. (2012). Distribution Pattern of Radionuclides in the Soil of Mainland China. J. East China Instit. Tech., **35**(2), 167–172.

[46] Rawlins, B. G., Scheib, C., Tyler, A. N., and Beamish, D. (2012). Optimal Mapping of Terrestrial Gamma Dose Rates Using Geological Parent Material and Aero Geophysical Survey Data. J. Environ. Monit., **14** (12), 3086–3093. DOI: 10.1039/c2em30563a.

[47] Belyaeva, O., Pyuskyulyan, K., Movsisyan, N., Saghatelyan, A., Carvalho, F. P. (2019). Natural Radioactivity in Urban Soils of Mining Centers in Armenia: Dose Rate and Risk Assessment. Chemosphere, **225**, 859–870. DOI: 10.1016/j.chemosphere.2019.03.057.

[48] Aközcan, S., Yılmaz, M., Ku¨lahc, F. (2013). Dose Rates and Seasonal Variations of 238U, 232Th, 226Ra, 40K and 137Cs Radionuclides in Soils along Thrace, Turkey. J. Radioanal. Nucl. Chem, **356**, 1-7. DOI: 10.1007/s10967-013-2730-5.

[49] Guagliardi, I., Apollaro, C., Scarciglia, F., De Rosa, R. (2013). Influence of Particle-Size on Geochemical Distribution of Stream Sediments in the Lese River Catchment, Southern Italy. Biotechnol. Agron. Soc. Environ, **17**, 43–55.

[50] Tchorz-Trzeciakiewicz, D.E., Kozłowska, B., Walencik-Łata, A. (2023). Seasonal Variations of Terrestrial Gamma Dose, Natural Radionuclides and Human Health. Chemosphere, **310**, 1-11. DOI:10.1016/j.chemosphere.2022.136908.

[51] Rangaswamy, D.R., Srilatha, M.C., Ningappa, C., Srinivasa, E., Sannappa, J. (2016). Measurement of Natural Radioactivity and Radiation Hazards Assessment in Rock Samples of Ramanagara and Tumkur districts, Karnataka, India. Environ. Earth Sci., **75**, 373 84. DOI:10.1007/s12665-015-5195-8.

[52] Melintescu, A., Chambers, S.D., Crawford, J., Williams, A.G., Zorila, B., Galeriu, D. (2018). Radon-222 related influence on ambient gamma dose. J. Environ. Radioact. **189**, 67–78. DOI: 10.1016/j.jenvrad.2018.03.012.

[53] Barbosa, S.M., Miranda, P., Azevedo, E.B. (2017). Short-term variability of gamma radiation at the ARM Eastern North Atlantic facility (Azores). J. Environ. Radioact. **172**, 218–231. DOI: 10.1016/j.jenvrad.2017.03.027.

[54] Fajkovic, H., Mikelic, I.L., Prohic, E. (2013). Vertical Distribution of K-40, Th-232 and Cs-137 Mass Activities in Lake Sediment (Vransko Lake, Croatia) and their Relationship with the Source Material and Sedimentation. J. Radioanal. Nucl. Chem., **295**, 2273– 228. DOI:10.1007/s10967-012-2332-7.

[55] Itodo, A. U., Edimeh, P. O., Eneji, I. S., Wuana, R. A. (2020). Radiological Impact Assessment of Mining on Soil, Water and Plant Samples from Okobo Coal Field, Nigeria. J. Geosci. Environ. Protect., **8**, 65-81. DOI:10.4236/gep.2020.85005.

[56] Hassan, M., Ngadda, Y.H., Adamu, A. (2020). Health Risk Assessment of Radionuclides in Soil and Sediments of some Selected Areas of Pindiga, Nigeria. J. Radiat. Nucl. Applic., **5**(2), 95-103. DOI:10.18576/jma/050203.

[57] Shittu, A., Aznan, F. I., Supiat, S. (2019). Radiological Characterization of Building Materials Used in Malaysia and Assessment of External and Internal Doses. Nucl. Sci. Tech., **30**, 46. DOI:10.1007/s41365-019-0569-3.

[58] Papastefanou, C., Stoulos, S., Manolopculou, M. (2005). The radioactivity of building materials. J. Radioanal. Nucl. Chem. **266**, 367–372. DOI: 10.1007/s10967-005-0918-z.