ASSESSMENT OF TERRESTRIAL NATURALLY OCCURRING RADIOACTIVE MATERIAL IN SOIL AND MINE TAILINGS OF AWO AND EDE, OSUN-STATE, NIGERIA.

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ABSTRACT

The levels of natural radioactivity of ^{40}K, ^{238}U and ^{232}Th in soils and mine tailings of Awo and Ede areas of Osun-state, Nigeria have been measured using a portable, advanced survey meter 992 Fluke Victoreen Gamma Scout Model for in-situ investigation and a 1 inch by 1 inch Cesium Iodide detector system. The specific activities of ^{40}K varied from 316.67±14.23 Bq kg^{-1} to 1487.35±3.03 Bq kg^{-1} with a mean value of 620.05±9.57 Bq kg^{-1}; for 238U, 13.11±2.36 Bq kg^{-1} to 88.26±3.03 Bq kg^{-1} with a mean value of 27.50±3.14 Bq kg^{-1}; and for 232Th, 9.26±1.00 Bq kg^{-1} to 55.81±2.99 Bq kg^{-1} with a mean value of 18.26±2.17 Bq kg^{-1}. The mean values obtained for Absorbed Dose rate and External Hazard Index (Hex) were 48.12 nGyhr^{-1} and 0.26 respectively. Generally, with the exception of ^{40}K, the activity levels of the radionuclide investigated were low and within the world average values. The calculated hazard indices were lower than internationally recommended limits.

Keywords: Mining, NORMs, Hazards, Gamma Spectrometry

INTRODUCTION

Terrestrial naturally occurring radioactive material (NORM) consists of radioactive material that comes out of the Earth's crust and mantle, and where human activity results in increased radiological exposure. The materials may be original (such as uranium and thorium) or decay products thereof, forming part of characteristic decay chain series, or potassium-40. The two most important chains providing nuclides of significance in NORM are the thorium and uranium series. Another major source of terrestrial NORM is potassium 40 (K-40) (World Nuclear Association, 2013). Gamma radiation emitted from naturally occurring radionuclides, also called terrestrial background radiation, represents the main external source of irradiation of the human body. Human beings are exposed outdoors to the natural terrestrial radiation that originates predominantly from the upper 30 cm of the soil (Obed *et al.*, 2005). Soil serves as a direct source of radionuclides leading to the contamination of all agricultural products (IAEA, 1989). The radioactivity of soils is determined by the radionuclides in the parent rock and the nature of processes involved in the formation of the soils. These radionuclides occur at different levels and are not uniformly distributed in soils and rocks and hence the knowledge of their distribution will play an important role in radiation protection and measurements (UNSCEAR, 1993).

Awo-Ede area is underlain by mineralized pegmatites rich in various rare earth metals, semi-precious gems and accessory economic minerals (NGSA, 2011). These minerals have NORM associated with them. Mining brings these materials into the precincts of people and often in fine highly mobile particulate forms. Hitherto, no information on previous assessment of radioactivity in soils in the area. It is important therefore to study the level of these radiations and its implication on miners and residents of Awo-Ede area. The data generated in this study will provide an up-to-date knowledge of the radiation levels due to specific naturally occurring radionuclides in the soils and mine tailings within the study area and also provide information about the hazards associated with these radiation levels and this will be valuable for environmental assessment and useful for regulatory bodies.

Description of the Study Area

The study area covers Awo and Ede towns which lie within Egbedore and Ede (Ede North and Ede

South) Local Government Areas respectively, Osun State, Nigeria. It covers an areal extent of 75.70 km^2 defined within latitudes 007º 44^1 N and 007º 47^1 N and longitudes 004º 24^1 E and 004º 30^1 E (NGSA, 2011). Generally, accessibility to and within the two towns is through the existing roads, footpaths and tracks. The study area lies within the humid tropical zone whose climate is characterized by alternating periods of raining season and dry season. The raining season starts from March, with heavy rainfall in May-June-July and a dry break in August followed by heavy rains in September-October. Annual rainfall amounts to about 150 cm, while the temperature varies from 24°C to 27°C and the relative humidity is about 80% (Iloeje, 1981). The vegetation falls within tropical rain forest with abundant evergreen trees like cocoa, oil palm, kolanut and a host of deciduous ones like *Iroko*, Mahogany, *Afara* etc. Due to extensive logging, farming and annual bush burning, there are wide spread grasslands particularly in the northern part of the study area.

The study area is characterized by a low relief and generally low-lying dissecting peneplain. The main landforms present are east-west trending ridges that rise up to 10-20 m above the surrounding plains. The pegmatite bodies in Awo are located in two prominent low undulating hills approximately 1.8 km apart. The first hill is located about 1.2 km east of Awo Township, with an elevation of about 10 m above the surrounding. The second hill lies about 1km southwards with an elevation of about 12 m above the surrounding (NGSA, 2011). The two hills are intensely weathered, with thick soil profile that supports cultivation of cassava and yam in adjoining area. The areas are drained by river Osun and its tributaries which flow southwards (Fig. 1). This is believed to be naturally controlled by fractures and the relief of the area.

The study area is underlain by pegmatites. The Nigerian pegmatites evolve during the time span of 600-530 Ma (Matheis and Caen-Vanchette, 1983), which indicates formation during the period of Pan-African magmatism. Pegmatites are formed from granitic magmas in the final stages of crystallization. These pegmatites, if mineralized contains varying proportions of minerals such as Beryl, Tourmaline, Tantalite and Columbite. The metals recovered from pegmatite are Niobium, Tin, Thorium and Uranium. Pegmatites can either be simple pegmatite or complex pegmatite (NGSA, 2011). Simple pegmatites are those pegmatites that are mainly not mineralized and they contain silicate minerals such as feldspars, quartz and muscovite, while complex pegmatites are those pegmatites that are mineralized and contain the minerals of economic importance which include gemstones such as beryl and ruby, molybdenite, tin, ores such as uraninite and thorite. The ores of uranium and thorium coexist with these gemstones in the pegmatites and they contain NORM which pose health hazards to human beings during gemstone mining and mining activities in general (NGSA, 2011). The pegmatites of Awo-Ede area have been classified as mineralized pegmatites rich in varieties of rare earth metals and semi-precious gem. They are composed of white Na-rich plagioclase, quartz, (Smoky and milky crystals), and books of muscovite in hand specimen. Accessory economic minerals are black tourmaline, tantalite, columbite, green beryl, rock crystal and mica (NGSA, 2011). The host rocks of pegmatite of Ede area are migmatitic gneisses and biotite muscovite schist. Wide spread tourmaline (boron) and muscovite in the pegmatites of Ede area presumably reflects metasedimentary or sedimentary precursor. There is significant occurrence of black tourmaline, garnet, muscovite, biotite and minor amount of beryl. From these, the pegmatites in Ede area have affinity for muscovite-rare element class.

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MATERIALS AND METHODS

Thirty samples comprising of soils (27) and mine tailings (3) were collected within the study area considering factors such as geology, location of mine sites, drainage paths, population density, agricultural areas and commercial centres. The actual sampling points were determined with the use of Global Positioning System. A survey meter 992 Fluke Victoreen Gamma Scout Model was employed in surveying the radioactivity in-situ before taking sample in these areas.

Soil samples (core) were taken (with a coring tool at depths between 0-15 cm) randomly at each corner of identified 1 m^2 area and at the center and composed to make a single sample

(composite) (IAEA, 1989). This is to ensure that the samples are representative of the sites from where they were taken. Each sample was labeled appropriately to avoid mix up. The samples collected were air-dried, pulverized and weighed. The plastic containers used as sample containers were washed with distilled water rinsed with dilute HCl and air-dried to avoid contamination. As part of measures to obtain a reliable result, the containers used have the same geometry with that of the background and also already prepared standard sample, which had the same base as the detector surface area. Each empty container with its cover was weighed by the use of an electronic balance. Then each of the pulverized samples was placed in each container, labeled and reweighed. The difference in masses is the mass of the sample. The containers were covered, labeled, and sealed (air tight) for a minimum of 28 days, for the sample to attain secular equilibrium and to avoid the escape of Ra-226 gas.

The gamma ray spectrometric analysis was carried out using a 1 inch by 1 inch Cesium Iodide (CsI) detector at the Biological Trace Element Research Laboratory, Department of Physics, Obafemi Awolowo University, Ile-Ife. The detector was enclosed in a thick lead shield. The energy and efficiency calibration were done using an IAEA-375 reference soil standard. Each sample was counted for 10 hours. The photopeaks observed with regularity belonging to those headed by the non-series ^{40}K, ^{232}Th and ^{238}U-series. Commonly used relations were used to calculate the radiation hazard indices. Spatial distribution maps for the hazard indices were generated using Oasis Montaj Software.

Assessment of Radiation Hazard

Absorbed Gamma Radiation Dose Rate (D): Effects of gamma radiation are normally expressed in terms of the absorbed dose rate in air, which originate from radioactive sources in the soil. The activity concentrations in soil correspond to the total absorbed dose rate in air at 1 m above the ground level. This was calculated using the following equation (Beck *et al.*, 1972).

$$\mathbf{D = 0.042A_c(K) + 0.429A_c(U) + 0.666A_c(Th)} \quad \ldots\ldots\ldots\ldots\ldots\ldots (1)$$

Where,

D = Absorbed dose rate in air measured in nGyhr^{-1}

A_c (K) = Activity Concentration of Potassium in Bqkg^{-1}

A_c (U) = Activity Concentration of Uranium in Bqkg^{-1}

A_c (Th) = Activity Concentration of Thorium in Bqkg^{-1}

Annual Effective/Equivalent Dose (De): The annual effective dose received by the population was calculated using the following equation (UNSCEAR, 2000) with a conversion factor of 0.7 Sv/Gy (ICRP, 1988).

Equivalent Dose (De) = D x Conversion factor x 1 x 365days x 24hrs x 10^{-6} (mSv/yr)...... (2)

External Radiation Hazard Index (Hex): The decay of naturally occurring radionuclides in soils produces an external radiation field to which all human beings are exposed. This was computed using the following equation (Alaamer, 2008).

$$Hex = \frac{A_U}{370} + \frac{A_{Th}}{259} + \frac{A_k}{4810} \quad (3)$$

Where A_U, A_{Th}, and A_k are the activity concentrations of ^{238}U, ^{232}Th, and ^{40}k in Bqkg^{-1} respectively

RESULTS AND DISCUSSION

Radioactivity Contents

The activity concentrations, in Bq kg^{-1}, of the selected radionuclide of ^{238}U, ^{232}Th, and ^{40}K in the 30 soil and mine tailings samples are presented in Table 1. In soil, the specific activity concentrations for ^{238}U varied from 13.11±2.36 Bq kg^{-1} to 88.26±3.03 Bq kg^{-1} with a mean value of 26.44±9.42 Bq kg^{-1}; for ^{232}Th, 9.26±1.00 Bq kg^{-1} to 55.80±2.99 Bq kg^{-1} with a mean value of 17.67±1.68 Bq kg^{-1}; and for ^{40}K, 316.67±14.23 Bq kg^{-1} to 1487.35±3.03 Bq kg^{-1} with a mean value of 595.61±8.44 Bq kg^{-1}. In mine tailings, the corresponding concentrations were 20.56±4.63 Bq kg^{-1} to 50.54±17.37 Bq kg^{-1} with a mean value of 37.04±7.28 Bq kg^{-1} for ^{238}U, 13.69±3.71 Bq kg^{-1} to 31.68±16.64 Bq kg^{-1} with a mean value of 23.54±2.50 Bq kg^{-1} for ^{232}Th and 535.89±15.03 Bq

kg^{-1} to 1039.14±14.70 Bq kg^{-1} with a mean value of 840.04±19.79 Bq kg^{-1} for ^{40}K.

Table 1: Radioactivity Concentrations of Radionuclides in the Soils and Mine Tailings of Awo-Ede Areas (Bq kg^{-1})

	SAMPLE CODE	^{40}K	^{238}U (^{214}Pb)	^{232}Th (^{208}Tl)
SOIL (n=27)	**S1**	316.67±14.23	19.36±13.79	12.87±2.96
	S2	498.40±9.04	22.98±11.62	15.40±10.83
	S3	394.95±11.41	17.52±5.24	12.52±3.32
	S4	490.06±9.20	27.84±9.59	17.98±9.28
	S5	1487.35±3.03	88.26±3.03	55.80±2.99
	S6	563.66±7.10	23.22±11.50	15.54±10.73
	S7	548.91±8.21	26.27±10.16	17.79±9.38
	S8	784.45±5.75	33.27±8.03	20.49±8.14
	S9	740.04±6.09	35.97±7.42	24.68±6.75
	S10	515.11±8.75	24.12±11.07	15.20±10.89
	S11	423.75±10.64	24.72±10.80	16.96±9.83
	S12	589.85±7.64	25.85±10.33	18.52±9.01
	S13	550.39±8.19	21.58±12.37	15.32±10.89
	S14	336.76±13.38	18.47±14.46	11.80±4.13
	S15	574.21±7.85	20.85±12.81	14.13±11.81
	S16	500.09±9.01	22.14±12.06	14.18±11.76
	S17	411.84±10.94	14.67±8.20	10.15±1.43
	S18	919.38±4.90	32.68±8.17	22.42±7.44
	S19	465.95±9.67	19.57±13.64	13.20±2.63
	S20	656.69±6.86	17.55±5.21	12.45±3.40
	S21	381.54±11.81	17.45±5.30	11.73±4.22
	S22	590.95±7.63	36.39±7.34	24.19±6.89
	DP1	710.88±6.34	21.78±12.26	15.37±10.86
	DP2	805.33±5.60	33.74±7.91	23.27±7.17
	DP3	725.68±6.21	29.39±9.08	18.44±9.04
	DP4	417.97±10.78	13.11±2.36	9.26±1.00
	DP5	680.46±6.62	24.99±10.68	17.42±9.57
	RANGE	316.67 - 1487.35	13.11- 88.26	9.26 - 55.80
	MEAN	595.61±8.44	26.44±9.42	17.67±1.68
MINE	**MT1**	535.89±15.03	20.56±4.63	13.69±8.71
TAILINGS (n=3)	**MT2**	945.07±16.16	50.45±17.37	31.86±16.64
	MT3	1039.14±14.70	40.11±21.85	25.07±2.15
	RANGE	535.89-1039.14	20.56-50.45	13.69-31.86
	MEAN	840.04±19.79	37.04±7.28	23.54±2.50

The comparison of measured activity concentrations of ^{238}U, ^{232}Th, and ^{40}K in soils and mine tailings within the study areas with values reported in soils worldwide (Elham and Arash, 2011) and contents in processed mine tailings from a mine site located in the Jos tin fields of Plateau State, Nigeria is presented in Tables 2 and 3 respectively. Radionuclide concentrations in the mine tailings were compared with the National Radiation Protection Department (NRPD) maximum permissible limits. It was found that the measured activity concentrations of the 3 selected naturally occurring radionuclide in the soil within this study areas were lower than most of the reported values from other countries as well as the world average values with the exception of ^{40}K (Table 2). This could be due to differences in the geological and geographical conditions (UNSCEAR, 2000). Likewise, the activity concentrations of radionuclides in tailing samples from mines in this study were lower when compared with that of Jos tin mines and average values given by NRPD (Table 2). In order to find out to what extent these radionuclides can exist together at a particular location, a correlation studies was carried between each pair of the radionuclides. The regression carried out was between K-40 and Th-232, K-40 and U-238, and U-238 and Th-232.

Table 2: Comparison with Mean Activity Concentration Values for some Radionuclides in Soil Samples from some Countries and UNSCEAR Report [12]

Country	Sample Type	Mean Activity Concentration (Bq kg^{-1})		
		^{238}U	^{232}Th	^{40}K
Yugoslavia	**Soil**	39.3	53	454
Turkey	**Soil**	39	-	643
Brazil	**Soil**	-	50	704
Pakistan	**Soil**	38	59	570
India	**Soil**	32	-	152.2
UNSCEAR (2000)	**Soil**	35	30	400
This Study	**Soil**	26.4	17.7	596

Table 3: National Radiation Protection Department (NRPD) Maximum Permissible Limits for ^{40}K, ^{238}U and ^{232}Th in Tailings (Adapted from Mangset and Sheyin [17]).

Activity concentration Bq kg^{-1})	(NRPD	This Study (n = 3)
^{40}K	11000	
^{238}U	1800	37.04 ±14.62
^{232}Th	900	23.54 ±9.17

The correlation co-efficient (R^2) derived for the pairs were 0.63, 0.55 and 0.99 respectively. The scatter plots and the correlation co-efficient calculated for each pair is shown in Figures 1 to 3. It is evident from the figures that correlations are not very perfectly strong between K-40 and Th-232, K-40 and U-238 but was perfectly strong between U-238 and Th-232.This trend between U-238 and Th-232 has been similarly reported by Jibiri *et al.*, (2011). This implies that the correlation between the pair indicates that an individual result for any one of the radionuclide concentrations is therefore, a good predictor of the individual values for the other. The results shown in Table 1 further indicate that in both soil and mine tailings the mean value of $^{40}K > ^{238}U > ^{232}Th$. The high abundance of K-40 in the soils and tailings is mainly due to the geochemical characteristics of rare metal pegmatite underlying the areas. This is consistent with the earlier findings of Ahrens (1957) and Okunlola (2005) that soils formed from rare metal pegmatites have a high concentration of K-40 due to the presence of minerals such as k-feldspars, mica (probably muscovite) and other clay minerals (such as illite) present in them.

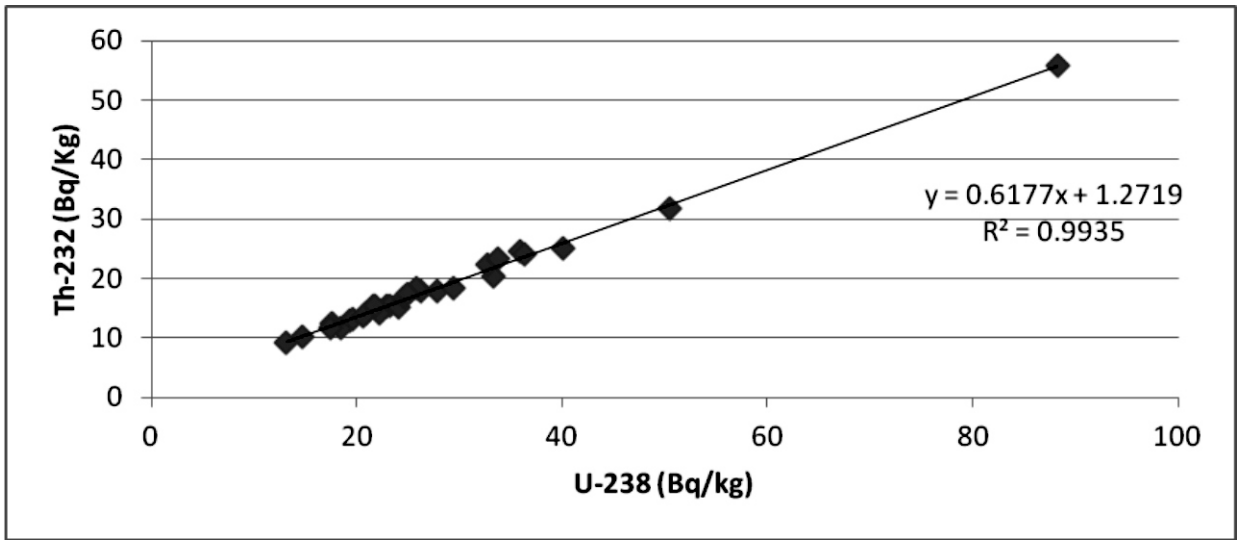

Figure 1: Scatter Diagram of Th-232 and U-238 in the Soil and Mine Tailing Samples from Awo and Ede Areas.

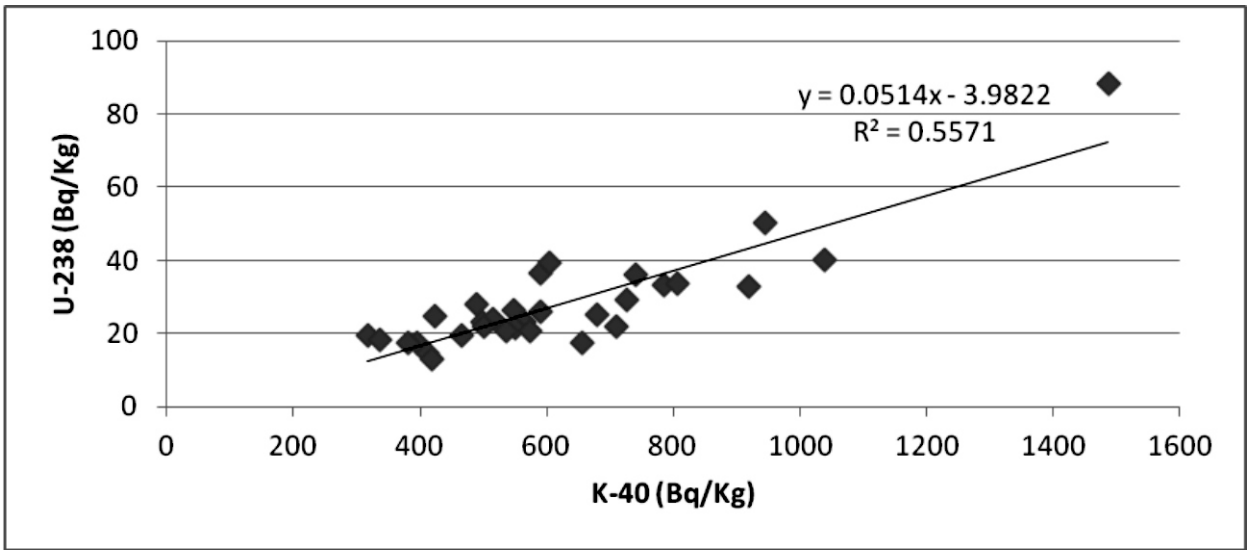

Figure 2: Scatter Diagram of U-238 and K-40 in the Soil and Mine Tailing Samples from Awo and Ede Areas.

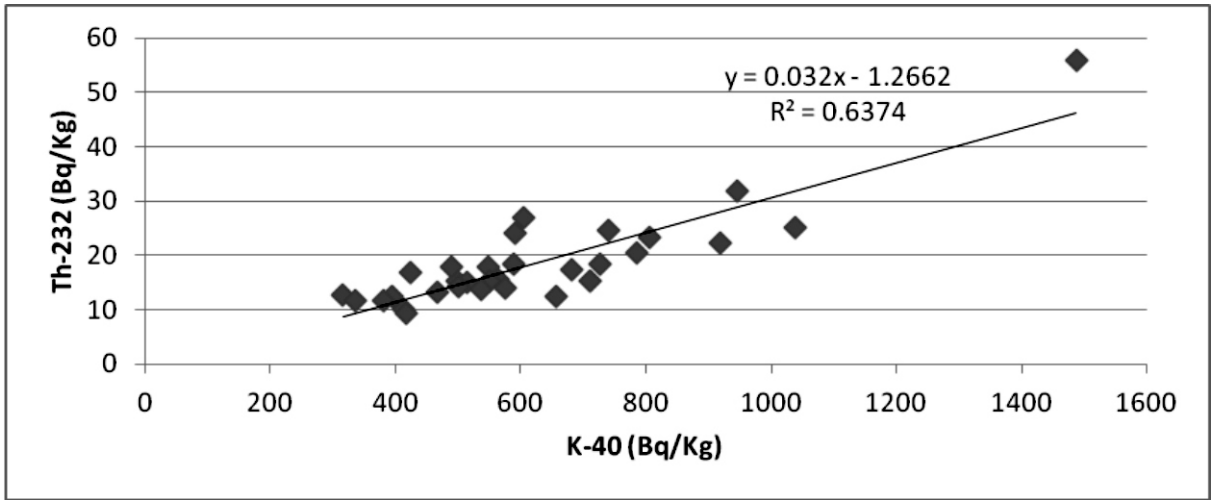

Figure 3: Scatter Diagram of Th-232 and K-40 in the Soil and Mine Tailing Samples from Awo and Ede Areas.

Assessment of Radiation Hazards

The values of air absorbed gamma radiation dose rate (D), annual effective dose (De), and external radiation hazard index (Hex) were 48.12 nGyhr^{-1}, 0.42 mSvyr^{-1} and 0.26, respectively in soils and 66.85 nGyhr^{-1}, 0.59 mSvyr^{-1} and 0.37 respectively in the mine tailings. It was observed that the mean values computed for D, De, and Hex in both soil and mine tailings were lower than the values reported worldwide (Table 4). This study has shown that radiation hazard in Awo-Ede areas is low. Maps were generated to show the spatial distribution of these calculated quantities as they occurred in the study area (Figs. 4, 5 and 6). The spatial distribution maps however showed existence of hotspots sometimes in excess of the acceptable limit of 60 nGyhr^{-1} for the absorbed dose. The hotspots were around the mine sites and drainage paths.

Table 4: Comparison of Radiation Risk Quantities With Threshold Limits.

		Absorbed Dose Rate (nGyhr^{-1})	Effective Dose Rate (mSvyr^{-1})	External Radiation Hazard Index
Soil	**Range**	29.35-137.49	0.26-1.20	0.16-0.76
	Mean	48.12	0.42	0.26
Mine tailings	**Range**	40.45-77.55	0.35-0.72	0.22-0.46
	Mean	66.85	0.59	0.37
Threshold Limits	**UNSCEAR (2000)**	60	-	<1
	ICRP (2000)	-	1	-

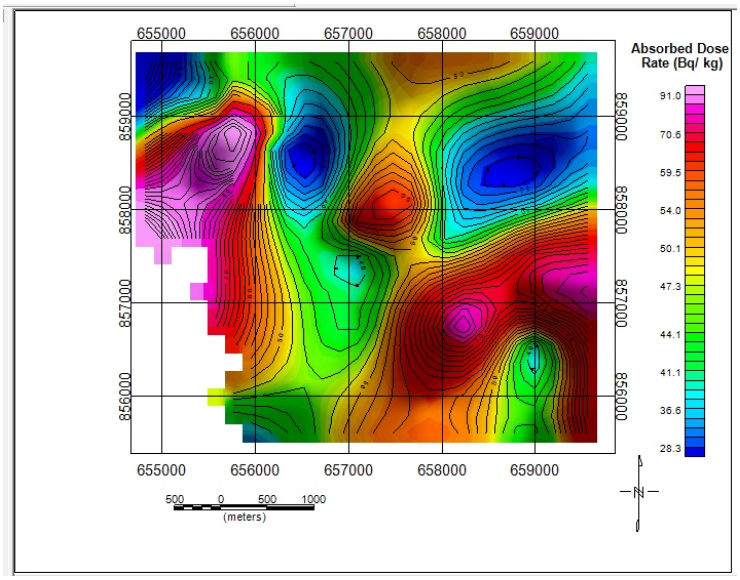

Figure 4: Distribution Map of Absorbed Dose Rate within Awo-Ede Areas.

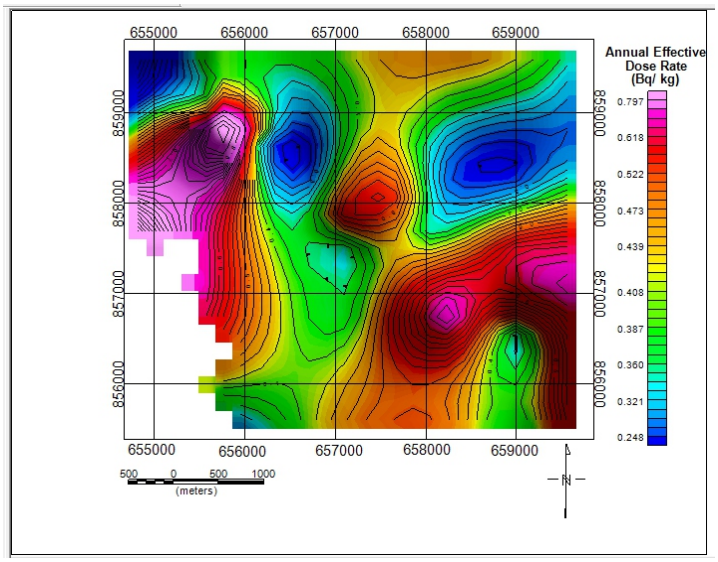

Figure 5: Distribution Map of Annual Effective Dose Rate within Awo-Ede Areas.

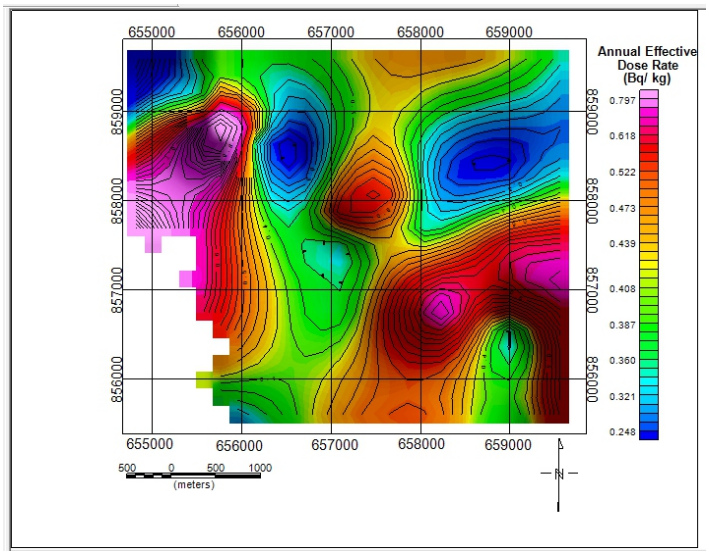

Figure 6: Distribution Map of External Hazard Index within Awo-Ede Areas.

Survey Meter Measurement

The result of the in-situ measurement using the survey meter is presented in Table 5. These values ranged from 0.02 $Svhr^{-1}$ to 0.11 $\mu Svhr^{-1}$ with an average value of 0.06 $\mu Svhr^{-1}$ in locations where there are piles of soils and 0.08 $\mu Svhr^{-1}$ to 0.09 $\mu Svhr^{-1}$ with an average value of 0.08 $\mu Svhr^{-1}$ in locations where there are piles of tailings. On the average, locations with the mine tailing piles have the highest value of dose rate. This could be as a result of concentration of ore mineral in the sampling location. Most of the values recorded for in-situ radioactivity measurement in soils and tailings from mines in Awo and Ede areas fell within the normal background radioactivity value of 0.11$\mu Svhr^{-1}$ (ICRP, 2007). This result is not conclusive since the survey meter does not indicate the activity of each radionuclide present. Comparing the dose-in-air measurement using a survey meter with that measured in the laboratory, the in-situ measurement result exhibited certain degree of about 50% agreement with the laboratory measurement data (Table 5). The reason for the differences may be due to difference in techniques, sampling time, and soil moisture variation. Assessment of the correlation between both results showed a weak positive relationship (Fig. 7) with no significant statistical difference in their average values.

Table 5: Survey Meter Reading in Air and Effective Dose Rate in Soil and Tailings Samples from Awo-Ede Areas.

Sample codes	Survey Meter readings ($\mu Svhr^{-1}$)	Effective Dose Rate ($\mu Svhr^{-1}$)
MT1	0.09	0.04
MT2	0.08	0.08
MT3	0.08	0.08
S1	0.05	0.03
S2	0.03	0.04
S3	0.04	0.03
S4	0.06	0.04
S5	0.07	0.14
S6	0.04	0.04
S7	0.02	0.05
S8	0.11	0.06
S9	0.03	0.06
S10	0.03	0.04
S11	0.04	0.04
S12	0.06	0.05
S13	0.06	0.04
S14	0.05	0.03
S15	0.05	0.04
S16	0.09	0.04
S17	0.08	0.03
S18	0.04	0.07
S19	0.05	0.04
S20	0.04	0.04
S21	0.08	0.03
S22	0.06	0.06
DP1	0.05	0.05
DP2	0.07	0.06
DP3	0.09	0.06
DP4	0.08	0.03
DP5	0.07	0.05
Average	0.06	0.05

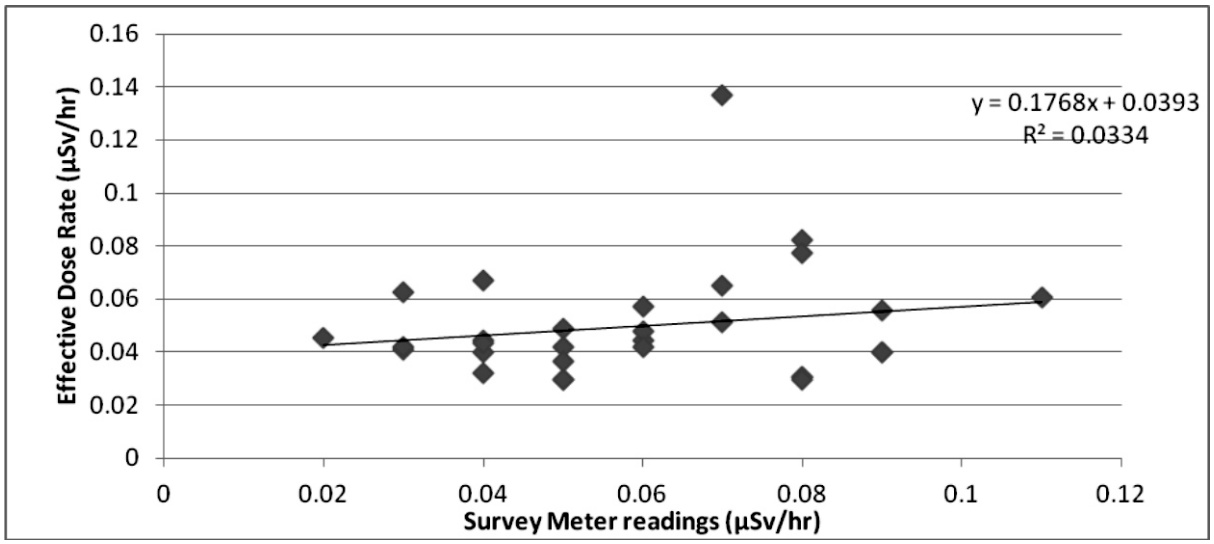

Figure 7: Scatter Diagram of Dose-in-air Measured by Survey Meter and that Measured in the Laboratory for Soil and Mine Tailing Samples from Awo and Ede Areas.

CONCLUSION

The activity concentrations of natural radionuclides have been determined in soil and mine tailing samples from Awo and Ede areas, Osun State, Nigeria using gamma-ray spectroscopy. The dose equivalents and external hazard index of the radionuclides in these soils and mine tailings were equally determined so as to assess the radiation risk to the bio-system. This prompted the in-situ survey of the radioactivity of the area and some samples were taken to the laboratory for gamma spectrometry analysis. Measured mean activity concentrations of the selected radionuclides were found to be less than the world average values. Calculated values of external radiation doses were also lower than world average values. The health impact assessment of the study area shows that the radiation hazard is low. However, some hot spots existed. Regular monitoring should be conducted to take care of unforeseen eventualities. The data and information generated here may be useful for establishing a comprehensive framework to investigate other areas and develop guidelines for monitoring and control of NORMs in the environment as a whole in Nigeria.

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