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Distribution of Lithogenic Radionuclides in Soils and the Associated Gamma Radiation Dose Levels around Jos North, Plateau state, Nigeria

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ABSTRACT: Natural gamma radiation dose levels was investigated by field survey and laboratory based analysis using NaI(Tl) survey meter and gamma ray spectrometry system, respectively. The mean activity concentrations for the terrestrial radionuclides ^{238}U, ^{323}Th and ^{40}K were determined to be 84, 216 and 169 Bq kg^{-1}, respectively. The highest mean was estimated for the geological unit G1 (Older granite) and the lowest for geological unit G4 (Basalts, trachylite and rhyolite). The results indicate positive correlation (0.897) between the measured and the estimated dose rates whose mean values are 210 and 180 nGy h^{-1}, respectively. Among the radionuclides, ^{232}Th was found to be the highest (74%) contributor to the total terrestrial dose rate in the area. Distribution of activity concentrations of ^{238}U, ^{232}Th, ^{40}K and gamma dose rate for the study area were plotted inform of isodose map using ArcGIS software.

KEYWORDS: Lithogenic radionuclides; gamma dose rate; isodose map; Jos North.

I. INTRODUCTION

Environmental measurement of natural radioactivity levels has received a global interest for decades. This has led to a performance of extensive investigations and surveys in many countries of the world (UNSCEAR, 2000). The results of these studies can be useful in many ways, these include; to establish a references-data records, for epidemiological studies, for the assessment of public dose rates and for environmental routine monitoring of background radiations and also for appraising the influence of artificial radiations due to unprecedented nuclear accidents (Tzortzis et al., 2004). The world technological advancement and rapid population growth have exponentially increased the global energy demand. This coupled with the issue of global warming have lead many countries to think of installing nuclear power plant (NPP) to generate electricity to meet their energy demand. In the course of this, the world is at risk of radioactive contamination from the fallout which may occur as a result of the NPP accidents causing serious damage to human, non-human and the environment. Therefore, reference data on the levels of background radiation is required to assess possible changes in the environment. As a demonstration to this scenario, is the Fukushima Dai-ichi NPP accident in 2011. Before the incident, the normal average dose rate in Japan was estimated at 50 nGy h^{-1}(Furukawa and Shingaki, 2012) while after the incident, annual effective dose has raised to a maximum of 1000 mSv at location northwest of FDNPP (Endo et al., 2012). Moreover, radioactive fallout from the Chernobyl accident was detected in many parts of the world including Africa and in some parts of Nigeria (Farai and Jibiri, 2000). In this regard, baseline data-records is very important for many reasons as aforementioned.

Background radiation in the environment is mainly due to radioactivity from the terrestrial radionuclides within the earth crust, such as ^{40}K and the radioisotopes from the decay series of ^{238}U and ^{232}Th, which occur in trace quantities in all rocks and soil formations (Kurnaz, 2013). The main source of external exposure to human body is the gamma emitted from the radioisotopes, at times called terrestrial radiation. Exposure to gamma radiation due to natural radioactivity in a particular region is strongly related to the type of geological bedrocks and soils in which these radionuclides are contained; and the geographical condition of a particular region (Florou, 1992). Silica richer rocks, such igneous rocks are associated with higher radiation levels and lower levels with low-grade metamorphic and sedimentary rocks (Abba et al., 2017). For instance, igneous rock of granitic structure and its weathered soils are strongly enhanced in U and Th compared to basaltic and ultramafic composition (Tzortzis et al., 2004). This indicates that, soil is the good indicator for measuring the levels of radioactivity in the environment for the reasons of establishing baseline data for assessment of future radiation impact, radiation protection and exploration. Therefore, this work aimed to investigate the distribution of lithogenic radionuclides ^{238}U, ^{232}Th and ^{40}K in soil and to evaluate the corresponding gamma dos rates around Jos North. The results of this study will revealed the contribution of different lithological formations to the total dose rate for the area.

II. MATERIALS AND METHOD

A) THE STUDY AREA

Jos North local government area (LGA) of Plateau state is geographically located on Jos Plateau in the north central region of Nigeria between the latitudes of $8^{0}75^{'}20^{"}$ and $8^{0}96^{'}19^{"}$ north of the equator and longitudes of $9^{0}80^{'}00^{"}$ and $10^{0}02^{'}07^{"}$ of Greenwich meridian. The population and land mass of the Study area are 23456 and 5678 km^3, respectively. Geology of the study area is classified into three main categories; the Basement complex, Younger granites and volcanic rocks. Since these rocks formations are characterised with enhanced concentration of natural radionuclides (Coker et al., 2013; Sciocchetti et al., 1988), measurement of background radiation measurements is of interest for this area. Previous studies in other parts of the region have shown relatively higher background radiation (Ademola, 2008; Farai et al., 2006; Ibeanu, 2003; Jibiri et al., 2007a) compared to other regions of Nigeria (Jibiri, 2001; Jibiri and Bankole, 2006; Obed et al., 2005). The bedrock formations of the study area is presented Figure 1.

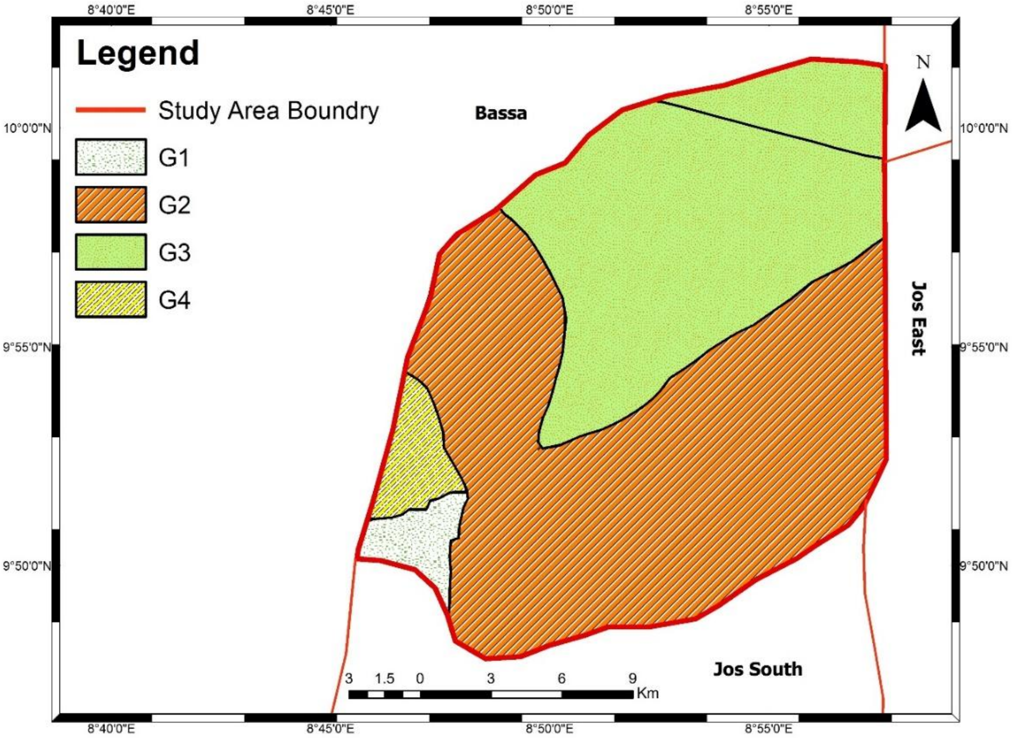

Fig. 1. Lithological formations of Jos North

B) FIELD RADIOMETRIC SURVEY AND SOIL SAMPLING.

A portable NaI(Tl) scintillation survey meter was used to carryout radiometric survey of gamma dose rates randomly at 96 locations covering the entire lithological formations of the study area. Certain number of measurements was conducted for each formation. The instrument has been widely used because of its linear response and wider coverage in detecting most of gamma energies from natural sources between the range of 40 keV and 1.2 MeV(Ramli, 1997). Measurements were taken in air for two minutes at 1 m above the ground and the gamma dose rates were recorded in mRh^{-1} and then converted to nGy h^{-1} using a conversion factor of 8.7 (Saleh et al., 2013a). Survey locations were recorded with the aid of Global Positioning System (GPS) device, Garmin eTrex 10 as shown in Fig. 2. Undisturbed locations and far away from public structures and other facilities were chosen for the measurements, so that the dose rates measured represent true natural radiation (Farai and Jibiri, 2000).

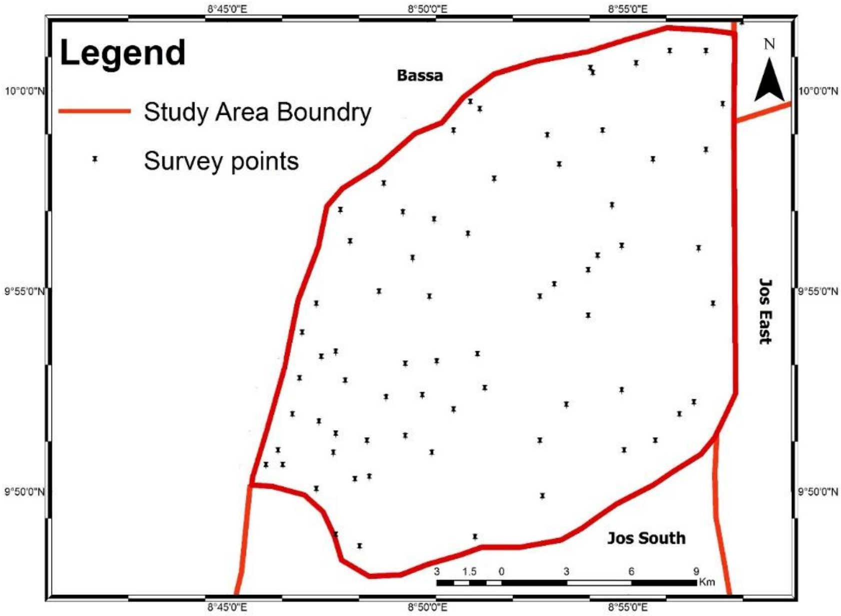

Fig. 2. Survey points

Field radiation survey meter respond to both cosmic and terrestrial gamma radiations and it is not possible to know the proportion caused by these sources. Therefore, laboratory analysis of soil samples from the area was performed to determine the relative contribution to dose rates due to terrestrial radionuclides ^{238}U, ^{232}Th and ^{40}K. Thus, gamma radiation dose rates of Jos North was also investigated by soil sampling at 38 different locations at the depth of 0-30cm across the formations for the determination of the radioactivity concentration of ^{238}U, ^{232}Th and ^{40}K. About 2 kg of soil were collected for each location in an open and uncultivated land and far away from public structures to avoid contribution from artificial sources. The samples were cleared of unwanted materials and debris. Later packed in a labelled plastic bags and thereafter transported to laboratory for preparation. Sampling locations were also recorded using GPS device. All loose soils were completely dried to constant weight in an oven overnight and then homogenized by sieving for grain size of less than 2 mm. About 500 ml volume of each sample was weighted and sealed in a standard Marinelli beakers and stored for one month to establish secular equilibrium for ^{238}U and ^{232}Th with their respective progeny (Arogunjo et al., 2009; Lee et al., 2009).

C) GAMMA-SPECTROMETRY

The measurements of soil samples were carried out using gamma spectrometry counting system at nuclear laboratory, Universiti Teknologi Malaysia. Specific activities concentration of ^{238}U, and ^{232}Th were determined through the activities of their decay products on the assumption that ^{238}U and ^{232}Th series are in the state of secular equilibrium with their decay products, while ^{40}K from its energy peak of 1461.8 keV. To avoid gamma spectra with large statistical error, the counting time of 43200 s was used. Genie 2000 software was used to analyse the spectrum. For the concentration of ^{238}U and ^{232}Th, the following gamma energy transition lines were used; ^{238}U: ^{214}Pb (295.21 and 352 keV) and ^{214}Bi (609 and 1120.29 keV), ^{232}Th: ^{212}Pb (238.6 keV), ^{208}Tl (583.1 keV) and ^{228}Ac (911.2 keV). Certified sources supplied by IAEA (SL-2, S-14, S-16) were used to test the performance, to ascertain the efficiency and energy calibration of the system. Periodic efficiency and energy calibrations were also perform to check for quality assurance. An empty Marinelli beaker under the same manner and geometry was counted for the same length of time to strip out the net peak area of the radiation due to background contribution.

D) ACTIVITY CONCENTRATION

The specific activity concentration in Bq kg^{-1} of ^{238}U, ^{232}Th and ^{40}K in soil samples were computed using the net peak area count after background corrections in each photo peak. The expression used is given by Jibiri et al. (2007b).

$$C(Bqkg^{-1}) = \frac{C_n}{\varepsilon M_s \rho_y} \qquad (1)$$

where, C_n is the number of counting under the photo peak of each radionuclide, γ is gamma ray detecting efficiency, P_γ absolute transition probability of the gamma ray and M_s is mass of the each soil sample (kg).

E) ESTIMATION OF GAMMA DOSE RATE IN AIR

Gamma dose rate in air 1 m above soil surface was estimated from the activity concentration of the radionuclides. Assuming the contribution from the radionuclide ^{137}Cs is considered negligible, absorbed gamma dose rate in air for the uniform distribution of ^{226}Ra, ^{232}Th and ^{40}K was estimated on the basis of guidelines provided by UNSCEAR (2008) using the expression given below:

$$D\ (nGy\ h^{-1}) = (0.621C_{Th}+0.462C_{Ra}+0.0417C_K) \qquad (2)$$

where, 0.621 nGy/h, 0.462 nGy/h and 0.0417 nGy/h are conversion factors for ^{232}Th, ^{238}U and ^{40}K, respectively from activity concentration to gamma dose rate and C_{Th}, C_U and C_K are the average activity concentrations of ^{232}Th, ^{226}Ra and ^{40}K in Bq/kg, respectively.

F) PLOTTING OF ISODOSE MAP

The data set for the 96 dose rates measurements and the coordinates for the survey points and that of the soil sampling locations were used to plot an isodose map for the area which represents the distribution of terrestrial radionuclides ^{238}U, ^{232}Th and ^{40}K and the levels of external exposure rate due to natural sources gamma radiation. Kriging technique was adopted for the isodose mapping. Non-biased linear estimation of data set on dose rate has been provided by ordinary Kriging extrapolation technique (Sanusi et al., 2014). The World Geodetic System (WGS84) was used to define the coordinates. The Kriging technique was applied using ArcGIS software version 10.3 Adjustments were made to take care of the lithological boundaries (Ramli et al., 2005).

III. RESULTS AND DISCUSSION

The summary of basic statistics of radiometric for the outdoor gamma dose rates for the 96 survey points is presented in Table 2. The dose rates values ranged between 128 and 874 nG h^{-1} with overall mean value of 210 nGy h^{-1}. Fifty percent of the measured dose rates was found to be ranged between 252 and 327 nGy h^{-1} as shown by the distribution curve in Figure 3. The mean value is by 72% greater than the global average of 59 nGy h^{-1}(UNSCEAR, 2000).

Table 2. Summary of the basic statistics for external gamma dose rates.

Statistics	Dose rate (nGy h^{-1})
Mean	210
Range	128 – 874
Std. Error	12
Std. Dev.	128
Median	253
Mode	252
95% conf. interval for mean	205-288
World average value	59

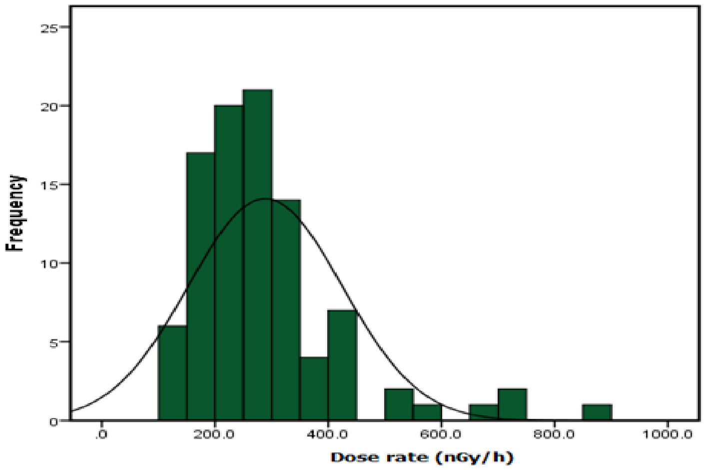

Fig. 3. Dose rate frequency distribution curve.

The mean activity concentration of the terrestrial radionuclides and their corresponding dose rate for each geological unit are presented in Table 3. The determined activity concentrations of ^{238}U, ^{232}Th and ^{40}K in the soil samples ranges from 25 to 300, 62 to 595 and 42 to 1040 Bq kg^{-1}, respectively. The mean values are respectively 84, 216 and 169 for ^{238}U, ^{232}Th and ^{40}K. The mean values are 84% and 64% greater than the corresponding world averages of 35 and 30 Bq kg^{-1}, respectively, while ^{40}K is 58% lower than the world average of 400 Bq kg^{-1}. The mean values are comparatively higher than that obtained by Nada et al. (2009) in Egypt and lower than that reported by (Masok et al., 2015) in the same region.

The estimated outdoor gamma dose rates based on the laboratory analysis of the soil samples is in the range of 134 nGy h^{-1} observed with the lithological unit G4 (Basalts, trachylite and rhyolite) and 254 nGy h^{-1} estimated on the lithogical unit G1 (Older granite) with overall mean value of 180 nGy h^{-1}. Among the terrestrial radionuclides, ^{232}Th has the highest contribution of 74%, this could be attributed to the presence of cassiterite (tin ore) and columbite in the area (Olise et al., 2010) which is believed to contain significant amount of radioactive minerals such as monazite and zircon (UNSCEAR, 2008). Soils containing monazite, with high concentrations of thorium, have been reported to be common in areas around the region (Jibiri et al., 2007b; Oresegun and Babalola, 1990). The mean dose rate for the study is found to be three time the global average of 59 nGy h^{-1}(UNSCEAR, 2000). The median difference between the measured and calculated dose rates is 30 nGy h^{-1}, being the cumulative contributions from cosmic radiation (30 nGy h^{-1}) (Furukawa and Shingaki, 2012), this suggest the accuracy of our measurements. The results obtained for this work are compared with those from different parts of the world in Table 4. The concentration of lithogenic radionuclides distribution of ^{238}U, ^{232}Th and ^{40}K are shown in Figures 4a, 4b and 4c, respectively.

Table 3. Mean activity concentration of ^{238}U, ^{232}Th, ^{40}K and the estimated dose rate for each lithological unit.

Geology	Mean activity concentration (Bq kg^{-1})			Mean dose rate (nGy h^{-1})	Contribution of radionuclide to dose rate (%)		
	^{238}U	^{232}Th	^{40}K		^{238}U	^{232}Th	^{40}K
G1	105	320	180	254	19	78	3
G2	69	210	160	169	18	77	5
G3	98	180	139	162	28	69	3
G4	65	155	197	134	22	72	6
Mean	84	216	169	180	22	74	4

Table 4. Activity concentrations of natural radionuclides and associated dose rates from other parts of the world are compared with the present study.

Country	^{238}U (Bq kg^{-1})	^{323}Th (Bq kg^{-1})	^{40}K (Bq kg^{-1})	Dose rate (nGy h^{-1})	Reference
China	38.5	56.6	584	124	(Yang et al., 2005)
Ghana	23	31	143	35	(Darko et al., 2015)
Nigeria	39	58	218	63	(Jibiri, N. 2001).
Egypt	65	23	146	44	(Nada et al., 2009)
Turkey	50	42	643	77	(Kurnaz et al., 2007)
Serbia	2.5	9.4	1.60	7.0	(Mandić et al., 2010).
USA	40	35	370	47	(UNSCEAR, 2000)
Spain	32	38	470	76	(UNSCEAR, 2000)
Malaysia	78	188	286	179	(Saleh et al., 2014)
Japan	33	28	310	53	(UNSCEAR, 2000)
Belgium	26	27	380	43	(UNSCEAR, 2000)
Iran	28	22	640	71	(UNSCEAR, 2000)
India	29	64	400	56	(UNSCEAR, 2000)
Hong Kong	59	95	530	87	(UNSCEAR, 2000)
Jos North	84	216	169	180	Present study
World	35	30	400	59	(UNSCEAR, 2000)

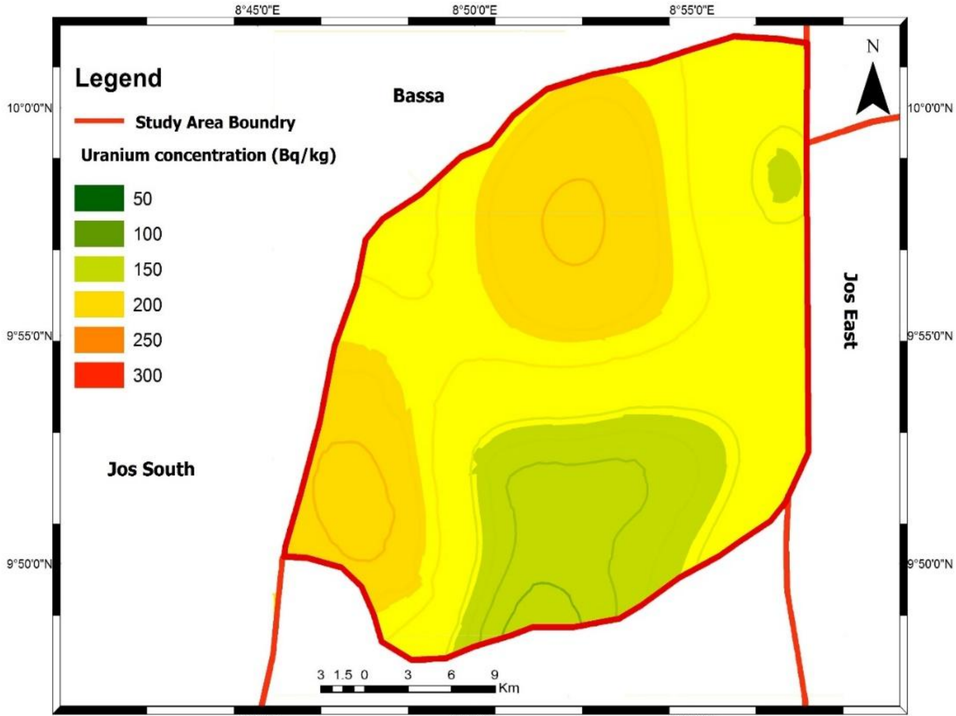

Fig. 4a. Distribution of activity concentration of ^{238}U

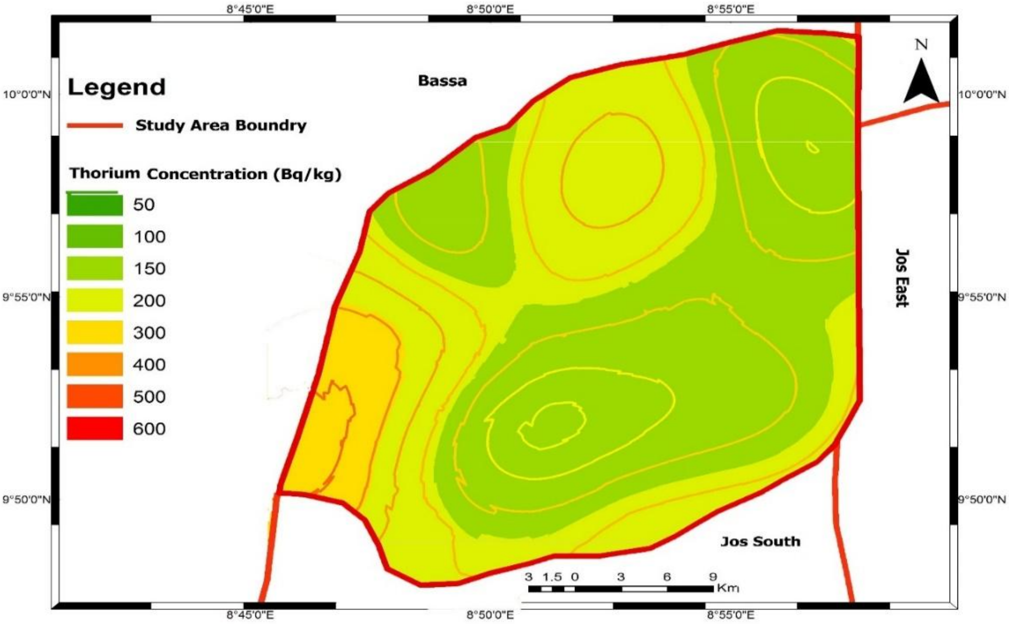

Fig. 4b. Distribution of activity concentration of ^{232}Th

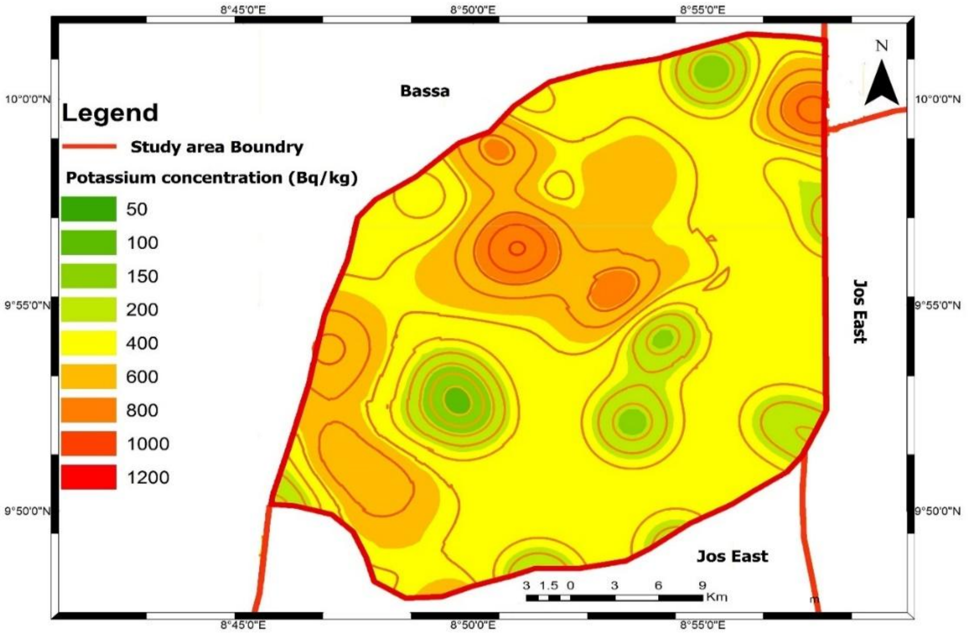

Fig. 4c. Distribution of activity concentration of ^{40}K

Enhanced concentrations of ^{238}U and ^{232}Th is observed in the north-western and south-eastern parts of the area while ^{40}K is observed to be elevated in the north-central and north-eastern parts. Exposure rate due gamma radiation dose is observed to be high in areas around the north-western and south-western parts as shown in Figure 5. Correlation between the measured and estimated dose rates was tested using linear regression plot (Figure 6). The graph revealed a positive correlation of 0.897 between the measured and estimated dose rate. The non-zero intercept (58.77 nGy h^{-1}) of the graph was assumed to be a contribution from cosmic rays and other terrestrial radionuclides. The inverse of the slope (0.964) was used to correct the measured dose rates by multiplying all the data set with this correction factor.

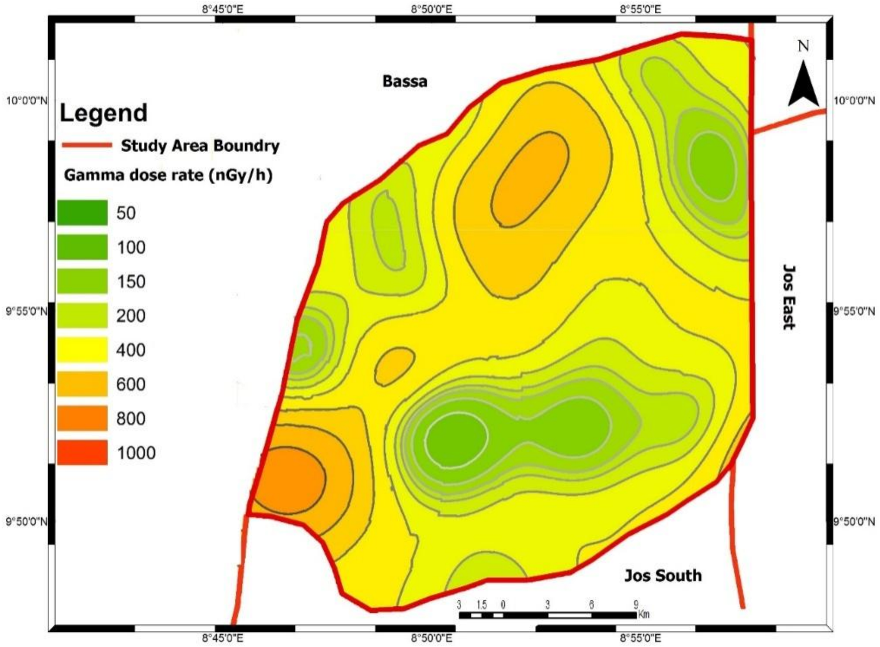

Fig. 5. Distribution of external gamma dose rate for Jos North

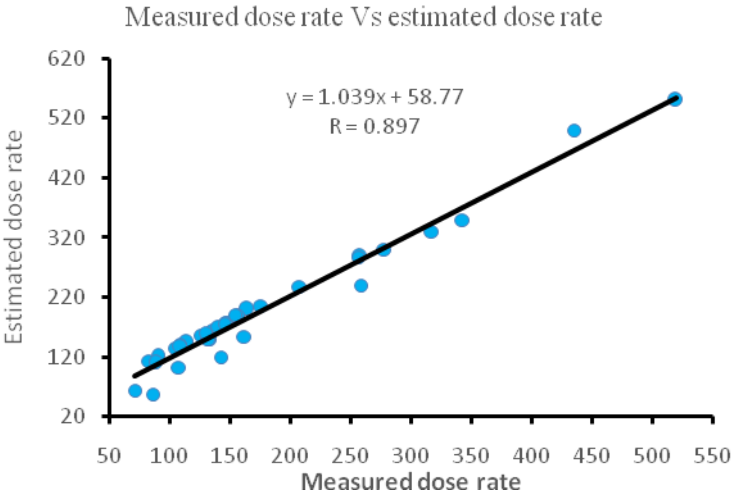

Fig. 6. Linear regression of measured dose rate vs estimated dose rate

IV. CONCLUSION

The mean value natural gamma radiation dose rates (cosmic and terrestrial) and terrestrial dose rate for Jos North are found to be 210 and 180 nGy h^{-1}, respectively. The levels of natural radiation exceed the norm value provided by UNSCEAR report (2000). The radioactivity in the soil samples were determined to be in the range of 25 to 300, 62 to 595 and 42 to 1040 Bq kg^{-1} for ^{238}U, ^{232}Th and ^{40}K with mean values of 84, 216 and 169 Bq kg^{-1}, respectively. ^{232}Th contributed the highest (74%) to the terrestrial gamma dose for the area. The data obtained in this study could be useful for mineral exploration and natural radioactivity mapping. The results could further be used to estimate public exposure risk due to natural sources of radiation.

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