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Effects of Physical and Chemical Properties on Natural Radionuclides Level in Soil of Quarry Sites in Ogun State, Nigeria

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Abstract: Activity concentration of Naturally Occurring Radionuclides (NOR) in soil samples were measured using NaI (Tl) scintillation detector. The soil samples were collected from twenty quarry sites across four Local government areas of Ogun state, Nigeria. The impacts of particle size distribution, organic carbon and matter as well as pH values of the soil samples, on the concentration of the radionuclides were determined using Pearson correlation and analysis of variance statistical methods. The activity concentration of potassium (^{40}K), uranium (^{228}U) and thorium (^{232}Th) in the soil ranged from 6.48-801.56, 2.04-90.12 and 2.60-89.23 Bq kg^{-1}, respectively. The result further revealed that there is no positive correlation between physical and chemical parameters and the radionuclides concentration in the soil samples. Whereas there are strong and positive correlation between the radionuclides concentrations.

Key words: Activity concentration, quarry site, soil, Nigeria

INTRODUCTION

The radionuclides in man's environment are from various sources (Hove, 1993; Larmash, 1983). The naturally occurring radionuclides are uranium (^{228}U), thorium (^{232}Th) and potassium (^{40}K), Carlson *et al.* (2003); and Mena *et al.* (1982) reported that naturally occurring radionuclides present in soils are not uniformly distributed hence knowledge of their distribution in soil plays an important role in radiation protection. The distribution of the naturally occurring radionuclides is influenced by factors such as chemical properties of the nuclides, physical and chemical properties of the soil as well as physical properties of the ecosystem (Risika and Nuccetelli, 2001; Ahmed, 2005).

Sources of ionizing radiation found in soil results from the rock from which the soil is derived and their levels vary (El-Aydarous, 2007). Natural radionuclides found at different depths of soil include ^{226}Ra, ^{232}Th and ^{40}K (Cox and Fankhauser, 1984; Sujo *et al.*, 2004; Manigandan, 2009).

MATERIALS AND METHODS

Study area: The study area is within latitude 6° 40^{1}N and 7° 40^{1}N and Longitudes 3° 20^{1}E and 4° 00^{1}E. The Local Government Areas (LGAs) that fall within the study area include Abeokuta North, Abeokuta South, Odeda and Obafemi Owode. The quarries considered for this research are distributed within these LGAs (Fig. 1). A total of one hundred soil samples were collected within and radially around the quarry sites. Each sample was collected by pitting the soil to depth of interest in each location, air dried and sieved with a <2 mm mesh sieve and then packed in a nylon made of non- radioactive material.

Measurement technique: The pH was measured by adding 2 g of dried soil sample into 50 mL beaker and 20 mL of distilled water was gradually added and left for 10 min to facilitate water movement through the soil (Agbenin, 1995). The mixture was later stirred occasionally with a glass rod and allowed to stand for 30 min. The pH meter (JENWAY 3150 MODEL) was standardized with buffer pH 4 and 9 solutions. The stabilized pH value was recorded from the display on the meter.

The Organic carbon content in soil samples was determined by the chromic acid oxidation method (Walkley and Black, 1934) and converted to organic matter by multiplying the percentage organic carbon by a factor of 1.729 (Black, 1965).

The particle size analysis was carried out by hydrometer method using sodium hexametaphosphate $(NaPO_3)_8$ as the dispersant (Bouyoucos, 1951).

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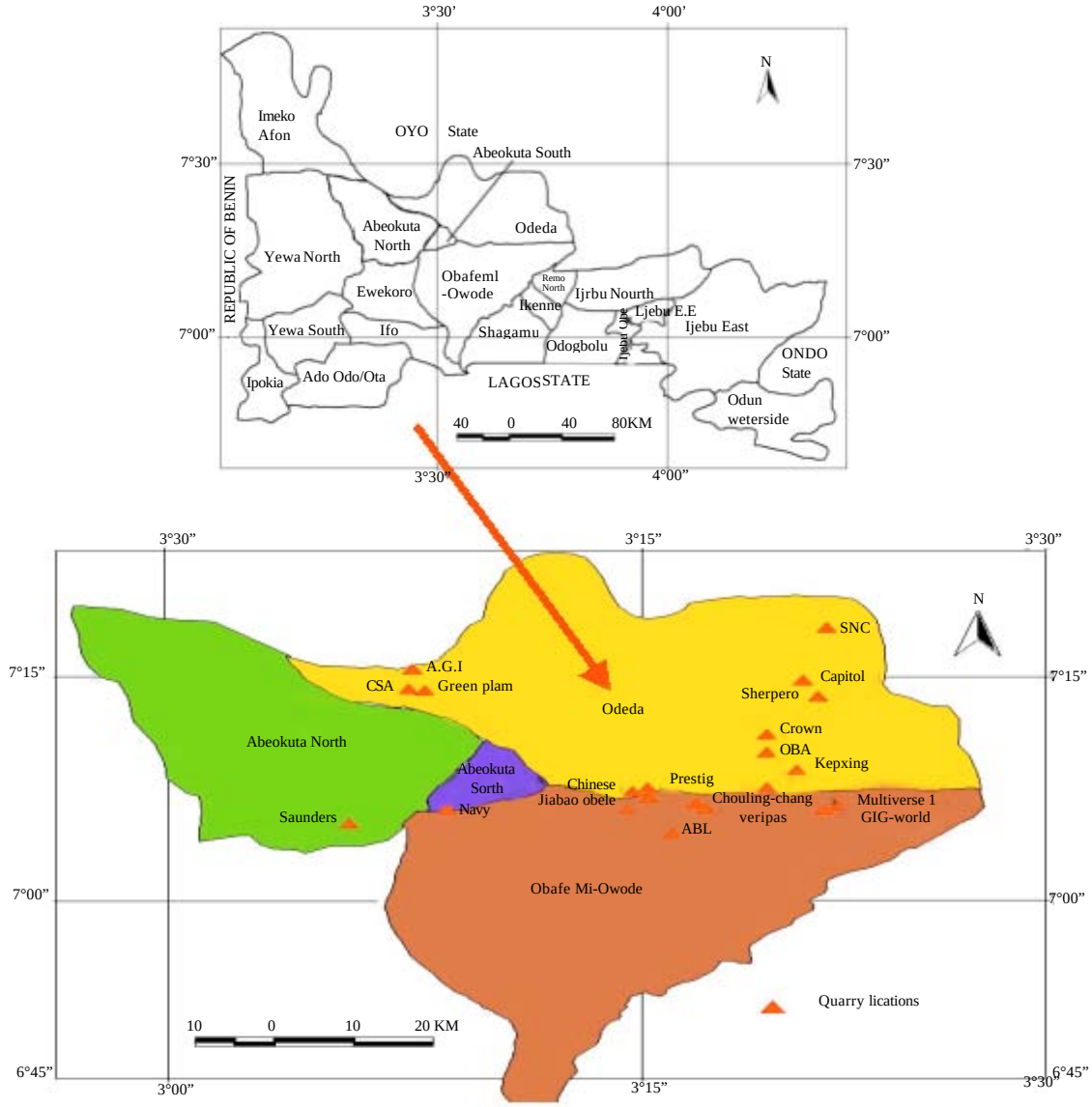

Fig. 1: Location map of the study area

The gamma spectrometric system used in the determination of natural radionuclide contents of the soil samples consists of a 7.6×7.6 cm NaI (Tl) scintillation detector (Model Bircom) and a multichannel spectroscopic analyzer (Canberra series 10). The detector which interfaced with the electronic system through 50 W coaxial cable, is placed in a lead castle to reduce signals from external background radiation. The Multi Channel Analyzer (MCA) electronic system consists of an internal spectroscopic amplifier and a 100 mHz Wilkinson type of Analogue to Digital Converter. The NaI (Tl) scintillation detector adopted for the study has been used by many researchers (Olomo *et al*., 1994; Ajayi and Ajayi, 1999; Tchokossa *et al*., 2011; Okedeyi *et al*., 2012).

Three Regions of Interests were created for the purpose of this research using the channel numbers corresponding to their gamma ray energies. The background radiation due to the naturally occurring radionuclides in the environment around the detector was measured by using an empty plastic container; the empty plastic container was counted in the same manner as the soil samples for the same counting time (36000 sec). ^{40}K content of the samples was obtained from the intensity of 1.461 MeV gamma ray peak followed by the ^{238}U content

from 1.761 MeV peak of ^{214}Bi and the ^{232}Th content from the 2.614 MeV gamma ray peak from ^{208}Tl. The mean specific activity per kilogram of dried mass of the samples at the beginning of the measurement in this study was calculated from the relation:

$$\frac{A_s}{A_{st}} = \frac{N_s}{N_{st}}$$

where, A_s = Specific activity concentration of radionuclides (Bq Kg^{-1}) in unknown sample. A_{st} = Specific activity concentration of radionuclides (Bq kg^{-1}) in reference Material, N_s = Net count rate under region of interest for unknown sample and N_{st} = Net count rate under region of interest for reference material.

RESULTS AND DISCUSSION

The range of the physical and chemical properties of the sampled soil in the 20 quarry sites were given in Table 1. The result revealed that Multiverse-2 quarry had the highest percentages of silt with values ranging from 10.00-26.00% while the lowest silt contents were recorded at Veritas quarry (3.00-4.16%). Similarly, ABL, CSA, Multiverse-2, CNC, Sheperd, Crown, Oba and Jiabao quarry sites recorded clay contents ranging from 9.60-30.00% whereas, Fam quarry (9.20-13.20%) and Chinese-prestige quarry (11.20-13.20%) had the lowest percentages of clay in the sampled soil.

The result also showed that Multiverse-2, capitol, CNC, Shouling-Shang, AGI, Chinese-prestige, Sheperd, Veritas, Fam and Green-palm quarry sites had sand contents ranging from 57.80-88% while Oba quarry (71.60-79.00%) and Multiverse quarry (54.00-80.00%) obtained the lowest percentages of sand. The organic carbon contents of the sampled soil indicated that Chinese-prestige, Crown, AGI, Jiabao, Capitol, Shouling-Shang, Multiverse and Fam quarry sites recorded values ranging from 0.14 -6.26% whereas, Navy quarry (0.01-0.64%), CSA quarry (0.62-1.31%) and sheperd quarry (0.71-1.57%) had the lowest percentages of organic carbon.

Table 1 also indicated that Jiabao, AGI, Multiverse, Capitol, Crown, Fam and Oba quarries had organic matter contents ranging from 0.24-4.83% while Navy quarry (0.01-0.28%), CSA quarry (0.62-1.31%) and sheperd quarry (0.07-1.59%) recorded lowest percentages of organic matter in sampled soil. Similarly, Multiverse, Navy, Kepxing, Sheperd, Saunder, AGI, Oba and Crown quarry sites had pH values ranging from 4.12-8.02 while Jiabao quarry (5.38-5.69), ABL quarry (4.96-6.12), CSA quarry (4.04-6.22), Green-palm quarry (4.99-6.33) and Veritas quarry sites (5.25-6.44) recorded varied pH readings over the 20 quarry sites.

The sand concentration varied among the quarry sites with Capital and Shouling-Shang quarries recording the highest percentages of sand (84.40%), other quarry sites had statistically close percentages. The highest organic carbon content was recorded at Chinese-Prestige quarry (2.07%), while the highest organic matter content was obtained in Jiabao quarry (3.75%). However, the pH values of the soil samples showed that most of the quarry sites were acidic with highest acidic concentration recorded in Chinese-Prestige quarry (3.80), Navy quarry (7.13) and Multiverse quarry (7.26) were alkaline. This finding is in agreement with previous

Table 1: Range of the physical and chemical properties of the soil samples in the 20 quarry sites

Quarry name	Silt (%)	Clay (%)	Sand (%)	Organic carbon (%)	Organic matter (%)	pH
OBA	8.00-14.00	12.40-18.00	1.60-79.60	0.98-1.92	1.69-3.31	5.75-6.69
AGI	0.60-13.60	12.60-17.60	68.80-85.80	0.14-2.79	0.24-4.83	4.12-7.11
Crown	5.00-16.60	12.00-19.40	64.00-83.00	0.22-5.68	0.38-3.55	1.52-6.90
Navy	6.00-12.40	11.20-15.20	74.80-82.80	0.01-0.64	0.01-0.28	6.50-7.81
Kepxing	5.60-11.00	11.60-16.60	74.80-82.80	0.01-1.42	0.21.2.45	6.23-7.76
CSA	3.60-10.60	14.00-23.00	66.40-82.40	0.36-0.76	0.62-1.31	4.04-6.22
Green-palm	2.60-7.60	12.40-14.80	79.20-83.60	0.36-1.40	0.62-2.42	4.99-6.33
FAM	2.00-7.00	9.20-13.20	76.80-83.80	0.34-2.05	0.59-3.55	5.96-6.80
Saunders	5.20-14.00	14.40-17.40	69.60-81.00	0.22-1.64	0.38-2.83	5.31-7.19
Sheperd	2.40-9.00	9.60-21.60	57.80-84.80	0.04-0.92	0.07-1.59	6.34-7.31
ABL	2.00-19.20	10.40-30.00	55.00-87.60	0.20-1.90	0.35-3.27	4.96-6.12
Veritas	3.00-4.60	12.00-15.00	80.40-84.20	0.96-1.38	1.66-2.38	5.25-6.46
Capitol	2.00-8.60	9.60-14.40	76.80-86.80	0.66-2.22	1.14-3.59	6.23-6.88
CNC	2.00-7.60	11.20-21.60	70.80-86.80	0.54-1.28	0.11-2.21	4.86-6.81
Shouling-shang	2.00-8.60	9.60-14.60	76.80-86.80	0.66-2.22	1.14-2.8	6.23-6.88
Multiverse-2	10.00-26.00	10.00-22.00	54.00-80.00	0.88-1.38	1.10-2.38	5.75-6.62
Multiverse	2.00-8.00	10.00-15.40	76.60-88.00	0.66-2.09	1.14-3.62	6.72-8.02
JiabAo	7.00-12.00	10.80-18.40	73.60-82.80	0.99-2.40	1.76-8.00	5.38-5.69
Gio-world	4.00-9.40	10.80-15.20	78.20-83.80	0.44-1.38	0.76-2.38	6.02-6.85
Chinese-prestige	2.60-7.6	11.20-13.20	81.20-85.80	0.14-6.26	0.24-3.07	0.59-6.95

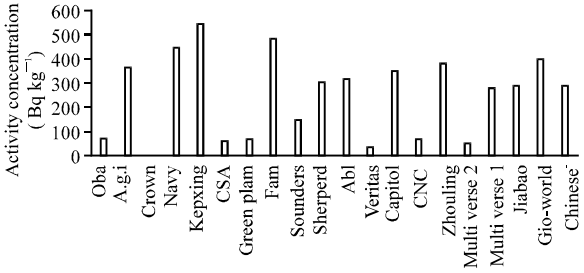

Fig. 2: Activity concentration of ^{40}K

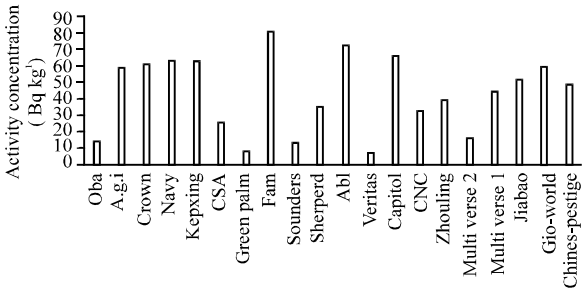

Fig. 3: Activity concentration of ^{238}U

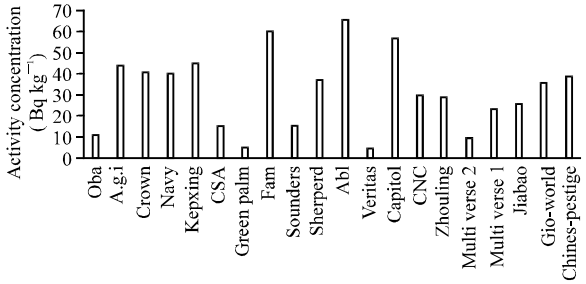

Fig. 4: Activity concentration of ^{232}Th

studies by Gbadebo (2011) which reported pH of 3.3-6.1 in the soil of an abandon quarry site in Abeokuta.

The range and mean of the radionuclide activity concentration in the soil (Fig. 2) revealed that Kepxing quarry had the highest ^{40}K concentration of 401.50-801.5 Bq kg^{-1} with average of 543.63 Bq kg^{-1}, while Gio-world quarry 266.38-502.35 Bq kg^{-1} with mean of 58.81 Bq kg^{-1}), Multiverse-2 quarry (6.90-85.82 Bq kg^{-1} with mean of 49.19 Bq kg^{-1}), Veritas quarry (8.77-57.29 Bq kg^{-1} with the mean of 32.13 Bq kg^{-1}) and Crown quarry sites (199.87-501.83 Bq kg^{-1} with mean of 31.90 Bq kg^{-1}) recorded lowest concentration of ^{40}K.

Figure 3 showed that Fam, ABL, Capitol, Navy, Kepxing and Crown quarry sites recorded 71.09-90.12, 57.98-88.54, 45.25-87.09, 38.74-72 .89, 46.40-70.45 and 49.45-72.54 Bq kg^{-1} with the average values of 79.00, 71.83, 65.79, 63.04, 62.26 and 61.45 Bq kg^{-1}, respectively of ^{238}U concentration, while Veritas quarry site (2.04-11.92 Bq kg^{-1} with mean of 7..59 Bq kg^{-1}) had the lowest activity concentration of ^{238}U.

The result also showed that Gio-World quarry site recorded the highest concentration of ^{232}Th with value ranging from 18.83-52.88 Bq kg^{-1} and an average of 65.05 Bq kg^{-1}, while Green-palm quarry site (3.56 -6.98 Bq kg^{-1} with mean of 5.17 Bq kg^{-1}) and Veritas quarry site (2.60- 6.37 Bq kg^{-1} with average of 4.55 Bq kg^{-1}) obtained the lowest concentration of ^{232}Th in the soil samples as shown in Fig. 4.

This result is similar to previous study by Belivermis *et al.* (2008) which recorded a mean value of 509, 22.3 and 24.7 Bq kg^{-1} for ^{40}K , ^{228}U and ^{232}Th and (Kilic *et al.*, 2008) that recorded 442.5, 22.3 and 26.6 Bq kg^{-1} but higher than Belivermis *et al.* (2010) that found an average value of 388, 27.7 and 32.5 Bq kg^{-1} activity concentration of the radionuclides in Istanbul soil.

The results of analysis of variance of this study indicated significant effect ($p \leq 0.01$ and 0.05) of all the parameters evaluated excepts organic carbon, thereby suggesting differences in physical and chemical properties and radionuclides among the twenty quarry sites investigated on soil sample (Table 2). Using Pearson two-tailed correlation coefficient (Table 3), a strong relationship was identified between silt and clay, sand and organic carbon and organic carbon and organic matter, conversely, strong negative relationship was observed between silt and sand, clay and sand or organic carbon. The finding also showed that there is no significant relationship between physical and chemical properties and the natural radionuclides. This is in contrary to earlier report by Believermis *et al.* (2010) which reported significant correlation between natural radionuclides and physical and chemical properties. Also, there was strong and positive relationship between ^{40}K and ^{238}U as well as similar positive strong relationship between ^{238}U and ^{232}Th indicating that increase in particular radionuclide lead to increase in other . The result further show showed a positive correlation between ^{40}K, ^{238}U and ^{232}Th in the sampled soil. This finding is similar to earlier report by Al-Jundi (2002) which observed positive correlation between ^{40}K and ^{232}Th in old phosphate soil of Jordan. While Valkovic (2000) reported that uranium concentration of the soil is related to soil organic matter. However, Foth (1984) reported that ^{40}K is present in mineral fraction rather than the organic fraction of the soil.

Table 2: Result of analysis of variance for the physical and chemical properties and radionuclides concentration evaluated in soil samples in the 20 quarry sites

		Means square value								
Source of variance	Df	Silt (%)	Clay (%)	Sand (%)	O/C (%)	O/M (%)	pH	^{40}K	^{238}U	^{232}Th
Intercept	1	5227.29	19143.49	615754.09	128.57	313.01	3775.98	6898990.79	183095.84	96469.25
Replication	4	9.52	14.77	9.13	0.20	0.74	0.60	4327.54	263.46	132.44
Quarry site	20	43.13**	14.45*	102.47**	1.22^{ns}	3.07**	1.84*	127640.06**	2502.04**	564.01**
Error	76	19.86	9.82	40.26	0.81	1.19	0.92	10059.22	172.41	130.42

** Significant at 1% probability level, *Significant at 5% probability level Ns: Not significant N.B: O/C: Organic carbon, O/M: Organic matter

Table 3: Pearson's correlation matrix showing interactions among physical and properties and radionuclides concentrations in soil samples in the 20 quarry sites

Properties	Silt (%)	Clay (%)	Sand (%)	O/C (%)	O/M (%)	pH	^{40}K	^{238}U	^{232}Th
Silts (%)	1	0.42**	-0.82**	-0.12	-0.02	0.12	-0.05	-0.15	0.12
Clay (%)		1	-0.68**	-0.17	-0.06	-0.15	-0.19	-0.15	-0.08
Sand (%)			1	0.02*	0.10	0.01	0.11	0.13	0.07
Organic carbon (%)				1	0.47**	-0.50**	-0.01	0.07	0.01
Organic matter (%)					1	0.18	-0.04	-0.01	-0.05
pH (Bq kg^{-1})						1	0.17	0.03	0.01
^{40}K (Bq kg^{-1})							1	0.71**	0.62**
^{238}U (Bq kg^{-1})								1	0.85**
^{232}Th (Bq kg^{-1})									1

**Correlation is significant at the 0.01 level (2-tailed) *Correlation is significant at the 0.05 level (2-tailed) N.B: O/C: Organic carbon, O/M: Organic matter

CONCLUSION

The study confirmed the presence of ^{40}K, ^{238}U and ^{232}Th in appreciable amount and that ^{40}K is the principal naturally occurring source of radiation in the study area. The abundance of ^{40}K in soil samples could be linked to its elevated concentrations in the rocks of the quarry sites. The activity concentrations observed are high in some of the quarry sites and could have radiological implications for population living around such quarries.

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