Natural Radioactivity in the Soil Samples of Coastal Southern Nigeria.

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Abstract

Studies on the gamma radiation level and the radionuclide distribution in the soils of the coastal region of Nigeria were carried out. The purpose of this study is to provide a baseline data on the radiation level and the distribution of some naturally occurring radionuclides in these coastal areas where a lot of oil exploration activities are taking place. The external gamma absorbed dose rates were measured using a high-resolution, low-background HPGe detector coupled to an EG & G ORTEC multi-channel analyzer. The activity of ^{40}K was found to vary between 7.049 and 586.69 KBq kg^{-1} with a mean value of 281.5 KBq kg^{-1}; that of ^{226}Ra varies between 4.103 and 91.293 KBq kg^{-1} with a mean value of 34.96 KBq kg^{-1}; while that of ^{232}Th was found to vary from 0.07 to 16.13 KBq kg^{-1} with a mean value of 9.16 KBq kg^{-1}. The contributions of ^{40}K, ^{226}Ra and ^{232}Th to the gamma absorbed dose rate were obtained to be 35.4%, 47.3% and 17.3% respectively. The mean values of the gamma absorbed dose rates obtained from the activity concentrations were compared with the literature values obtained for other coastal environments and also with the world average. In addition, the correlation between ^{40}K and ^{226}Ra, ^{40}K and ^{232}Th and ^{226}Ra and ^{232}Th were computed from results of the activity concentrations of these naturally occurring radionuclides. The correlation between these radionuclides varied from State to State with the strongest correlation occurring between ^{232}Th and ^{40}K in Akwa-Ibom State while the weakest correlation occurs between ^{232}Th and ^{226}Ra in Bayelsa State. The results are discussed in the paper.

Keywords: ^{40}K, ^{226}Ra, ^{232}Th, naturally occurring radionuclides, radioactivity, soil, environmental.

Introduction

For many years now, nuclear activities have been on the increase worldwide, which have led to an increase in the risks of abuse or accidents. These have thereby posed as having adverse health effects on human life. The measurement of the levels of the different components of radiation present in the environment at a given time can therefore be used to assess the risk to a particular population. These components include natural background radiation due to naturally occurring radioactive materials (NORM); and those of cosmic radiation of galactic origin. Baseline information on the spatial variation of radionuclide concentration in soils is necessary to evaluate any change induced by humans in the future (Meriwether et al., 1985). The primary source of radiation received by humans is from the store of natural radioactivity in soils, which derive essentially from naturally occurring radioactive materials (specifically ^{40}K and the radionuclides of ^{238}U and ^{232}Th decay series).

The coastal areas of Nigeria comprising of Lagos, Ogun, Ondo, Akwa-Ibom, Bayelsa, delta and Rivers State are believed to account for about 5% of the world's oil and gas production (Reijers et al., 1996). This present work is however based on Niger-Delta region comprising of Ondo, Delta, Bayelsa, Rivers and Akwa-Ibom States where a lot of oil exploration activities are being carried out (Figure 1). Thus there is need for surveillance on the radiation level of the region to assess the impact of these activities on the environment, and to know the level of the human exposure to technologically-enhanced as well as man-made radiation. The concentrations of the naturally occurring radionuclides were measured in the soil samples of these areas and the resultant dose rates in air due to these radionuclides were calculated. The results obtained were compared with literature values obtained for other environs (Table 1).

Materials and Method

The external gamma dose rates in air have been obtained from the radioactivity concentrations of the radionuclides in the soils of these coastal States. Soil samples were taken at a depth of about 0.15 m at points that were open, undisturbed, level ground surfaces free from sheltering vegetation and run-off during heavy rain and away from public roads and buildings.

The activity concentrations of the naturally occurring radionuclides in these soil samples were determined by gamma spectrometric analyses using a p-type coaxial HPGe detector with an efficiency of 20% and a resolution of 1.86 keV at 1.33 MeV. The gamma ray spectrum was recorded using a PC-based 4096 channel analyzer and processed using the SPAN 5.0 software (Wang L., 1995). The detector was housed

inside a massive lead shielding comprising of 100 mm thickness of Lead, 3 mm thickness of Cadmium and 0.7 mm thick Copper. The detector was calibrated using the IAEA quality assurance reference standard sources. These standard sources had energies covering the energy range required for the measurements to be taken i.e. between 0.662 and 2.615 MeV. The efficiency of the detector was also determined experimentally using the standard sources. The lower detection limits of 95% confidence level for 10 hrs of counting time and 200 g of standard sample weight were found to be 578.40 $Bqkg^{-1}$ for ^{40}K; 20.90 $Bqkg^{-1}$ for ^{226}Ra; and 10.47 $Bqkg^{-1}$ for ^{232}Th, representing the specific activities of these radionuclides in the standard sample.

Each of the 279 soil samples collected from 81 different locations, mainly close to the coast, was taken to the laboratory for processing. The samples were dried at a laboratory temperature of 27 ^{0}C, and relative humidity conditions, for 72 hrs. The dried soil was then pulverized to a particle size not greater than 2 mm mesh (Myrick et al., 1983); with the larger particles discarded. Each of the soil samples was then stored in an air-tight container for a minimum of 30 d to allow for radioactive equilibrium of ^{222}Rn and ^{220}Ra with their corresponding progeny. The containers were sealed carefully using cellophane tape to avoid the escape of gaseous radon and thoron from the sample.

The samples were then counted in containers identical to that of the mixed standard sample obtained from the Environmental Measurement Laboratory, U.S.A. This mixed calibration sample contained certified concentrations of the radionuclides of interest i.e. ^{40}K, ^{232}Th and ^{226}Ra. Thus the same counting geometry was maintained for a period of 10 h to minimize the counting error. The activities of the samples were determined using the total net counts under the selected photo-peaks, the measured photo-peak efficiency, gamma intensity, and weight of the sample (Narayana et al., 1993). The activity of ^{40}K was evaluated from the 1.461 MeV photo-peak, the activity of ^{226}Ra from the 1.761 MeV photo-peak and that of ^{232}Th from the 2.615 MeV photo-peak. From the concentrations of the naturally occurring radionuclides in the soil, the absorbed dose rates in air were estimated. The conversion factors used to compute the absorbed dose rate in air per unit of specific activity concentration in soil were 0.0414 $nGyh^{-1}$ for ^{40}K; 0.461 $nGyh^{-1}$ for ^{226}Ra; and 0.623 $nGyh^{-1}$ for ^{232}Th (UNSCEAR, 1993).

Results and Discussion

External Gamma Dose Rates in Air

The external gamma dose rates in air have been obtained from the activity concentration values of the naturally occurring radionuclides. The values of the gamma dose rates range from (2.229 ± 0.081) nGy h^{-1} to (63.758 ± 0.706) nGy h^{-1} with a median value of 33.01 nGy h^{-1}. Lower dose rates were obtained in sandy soils along the beach in certain locations while high values were also recorded for some locations with granitic soils. However, there was no regular pattern regarding these locations in respect of distances

from the coast. The high dose rates obtained in some river mouths may be traced to the enrichment of certain radioactive materials in these locations due to mechanical sorting as a result of wind and wave action.

The external gamma dose rates in air in these coastal areas of Nigeria are compared with the values reported for other environs, in Table 2. It is observed that these values are comparable with the values obtained for the coastal areas of a number of other countries. However, these values are orders of magnitude higher when compared to the dose rates in high background areas.

Activities of Individual Radionuclides

The radioactivity concentrations of the soil samples of these coastal areas of Nigeria are given in Table 2. These values are found to vary significantly from place to place. The activity concentration of ^{40}K varies from (7.049 ± 0.706) kBq kg^{-1} in Offot, Akwa- Ibom State, to (586.69 ± 12.991) kBq kg^{-1} in Gbaranfor Island, Bayelsa State with a median value of 283.28 kBq kg^{-1}; the activity concentration of $^{238}U(^{226}Ra)$ varies from (4.103 ± 0.069) kBq kg^{-1} again in Offot, Akwa Ibom State to (91.293 ±1.081) kBq kg^{-1} in Ugbo, Delta State with a median value of 33.761 kBq kg^{-1}, while the activity concentration of ^{232}Th varies from (0.073 ± 0.003) kBq kg^{-1} in Offot to (16.13 ± 0.088) kBq kg^{-1} in Sagbama, Bayelsa State with a median value of 9.171 kBq kg^{-1}.

Comparison of Activities with other Coastal Environs

The activity concentration of each of these three radionuclides in the soil samples of coastal Nigeria are compared with literature values obtained from some other coastal environments of the world in Tables 3, 4 and 5.

From Table 3, we observe that for ^{40}K, the mean activity concentration in Coastal Nigeria is found to be higher than the world average and also much higher than values obtained in other coastal environments of the world (Narayana et al., 2001).

The mean activity of ^{226}Ra is also higher than the world average as indicated in Table 4. The values are again higher than those obtained in other coastal environs.

From Table 5, we observe that the concentration of ^{232}Th falls within the world range and the wide range of values reported for Coastal Karnataka in India, in United States, Nile Delta, Ireland and China. The higher values of activity concentrations of ^{226}Ra and ^{232}Th obtained for places like Sagbama and Famgbe in Bayelsa State; Ugbo in Delta State; Ilutitun and Ominla in Ondo State; may be traced to either laterite type of soil or the asphalt type of soil obtained in these locations.

Correlation between the Naturally Occurring Radionuclides

The correlations between ^{226}Ra and ^{40}K; ^{232}Th and ^{40}K; and between ^{226}Ra and ^{232}Th, were computed from the mean activity concentrations obtained.

It was observed that for Akwa-Ibom State, the correlation between ^{226}Ra and ^{40}K was weak at a correlation coefficient of 0.423. The correlation between ^{232}Th and ^{226}Ra is fair with a correlation coefficient of 0.5795, but the correlation between ^{232}Th and ^{40}K was high with a correlation coefficient of 0.814. The low correlation between ^{226}Ra, ^{232}Th and ^{40}K indicated that individual results for any one radionuclide concentration in each pair were not good predictors of individual values for the other. The correlation coefficient between ^{232}Th and ^{40}K as illustrated in Figure 2 was significant at 95% confidence level.

For Bayelsa State, the correlation coefficient between ^{226}Ra and ^{40}K was 0.44, that between ^{232}Th and ^{40}K is 0.33 with that between ^{232}Th and ^{226}Ra being 0.0073 which was the weakest (Figure 3).

For Delta State, the correlation coefficients between^{226}Ra and ^{40}K and between ^{232}Th and ^{226}Ra were 0.12 and 0.034 respectively which were extremely weak values. On the other hand, the correlation coefficient between ^{232}Th and ^{40}K was 0.62 which indicated a strong correlation between these two radionuclides which is significant (Figure 4).

In Ondo State, there was a weak correlation of 0.23 between ^{232}Th and ^{40}K, while for ^{40}K and ^{226}Ra; and for ^{232}Th and ^{226}Ra, the correlation coefficients were very low at 0.009 and 0.0115 respectively (Figure 5).

For Rivers State, the correlation between ^{40}K and ^{226}Ra was the weakest with a value of 0.25. The correlation between ^{232}Th and ^{40}K; and between ^{232}Th and ^{226}Ra has values of 0.61 and 0.41 respectively which are comparable with values obtained for Akwa-Ibom State (Figure 6).

Gamma absorbed dose rates

The range of the gamma dose rates for each of the States were computed from the activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th and the results are presented in Table 6. The absorbed dose rate due to ^{40}K varies between 0.292 and 24.289 nGy h^{-1} with a mean value of 333.37 nGy h^{-1}. The dose rate in air due to ^{226}Ra varies from 1.892 to 29.435 nGy h^{-1} with a mean value of 36.796 nGy h^{-1}. The dose rate due to ^{232}Th is in the range 0.044 to 10.049 nGy h^{-1} with a mean value of 10.234 nGy h^{-1}. The computed dose rates in air from this present study are found to be lower the world average values. The percentage contributions of ^{40}K, ^{226}Ra and ^{232}Th to the external dose rates in air are presented in Table 7. UNSCEAR (1982) has reported the world wide average values for the relative contributions to the gamma dose rates in air from ^{40}K, ^{226}Ra and ^{232}Th to be 35%, 25% and 40% respectively.

CONCLUSION

The absorbed dose rates in air in these coastal environments ranged from 2.228 to 73.52 nGy h^{-1} with a mean value of 37.14 nGy h^{-1}. From the world-wide average values of ^{40}K, ^{226}Ra and ^{232}Th, the total dose rate in air was computed to be 53.23 nGy h^{-1}. From the correlation analyses, it is observed that in Akwa-Ibom State, for a 100% presence of ^{40}K in the soils obtained, there is a 65% likelihood of the presence of ^{238}U and 90% likelihood of the presence of ^{232}Th. In Bayelsa, Delta and Rivers States there is a proportionate increase for the three radionuclides. On the other hand, for Ondo State the likelihood of the presence of ^{238}U decreases for an increase in the amount of ^{40}K and ^{232}Th. The gamma dose rate in air due to these naturally occurring radionuclides can be predicted from the local geology of the region.

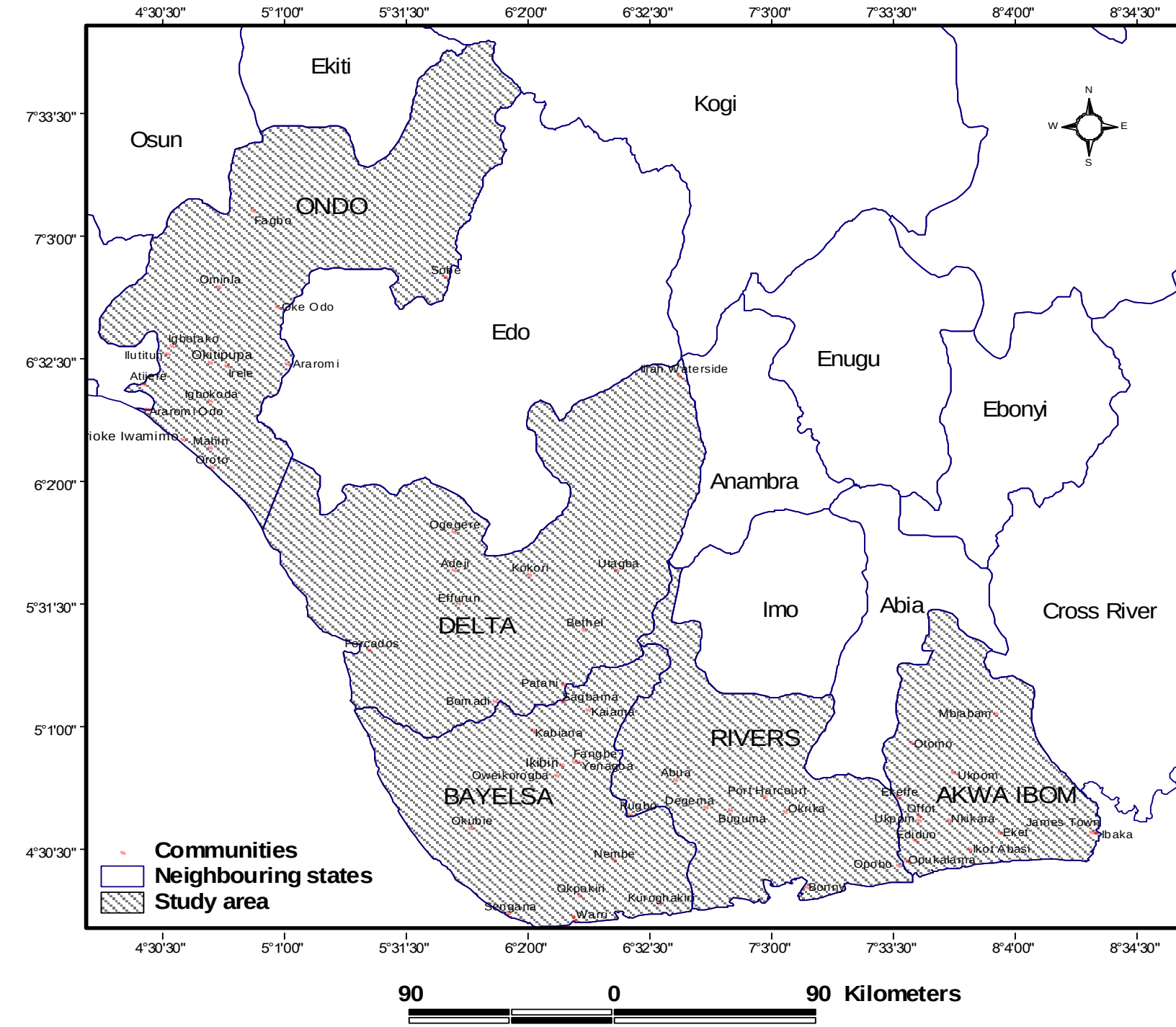

Figure 1: Map Showing the Niger-Delta Region

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Table 1: Comparison of ^{40}K, ^{226}Ra and ^{232}Th Activity Concentration Values obtained with those of other Coastal Areas.

Region	Mean Activity Values (Bqkg^{-1})			Reference
	^{40}K	^{226}Ra	^{232}Th	
Present Work	7.04 - 586.7 × 10^3 (283.3× 10^3)	4.103 – 91.293×10^3 (34.95× 10^3)	0.07 – 16.13 × 10^3 (9.16× 10^3)	
Canary Islands	141.6 – 1,489	7.3 – 104.0	11.6 – 110.5	Fernandez-Aldecoa et al., (1992)
Nile Delta	29 - 653	-	2.5 – 95.6	Ibrahiem et al., (1993)
Coastal Karnataka of South India	61.0 – 316.7	20.1 – 62.3	14.3 – 48.6	Narayana et al., (2001)
Kiaga, South West coast of India	78.3 – 254.8	15.5 – 61.2	11.4 – 41.9	Karunakara et al., (2001)
World Range	100 - 700	2.96 – 140.6	7 – 50	UNSCEAR (1988)
World Average	440	30	34	McAulay and Moran (1988)

*Values given in parenthesis are the median values.

Table 2: Comparison of ^{40}K activity in soil

Activity in Bq kg^{-1}			
Present Work(×10^3)	Literature Values	Region	References
7.049 - 586.7	61.0 - 316.7	Coastal Karnataka, India	Narayana et al., (2001)
	34.78 – 245	Bombay	Lalit & Shuklah (1982)
	12 – 1,570	Greece	Anagaostakis et al., (1996)
	48 – 1,586	Spain	Baeza et al., (1992)
	580 – 760	Italy	Bellia et al., (1997)
	398	Louisiana	Delaune et al., (1986)
	141.6 – 1,489	Canary Islands	Fernandez-Aldecoa et al., (1992)
	29 – 453	Nile Delta	Ibrahiem et al., (1993)
	266 – 711	Canada	Kiss et al., (1988)
	670 – 1000	Finland	Klemola et al., (1991)
	289 – 703	Spain	Martinez-Aguirre & Garcia-Leon (1997)
	40 – 800	Ireland	McAulay and Moran (1988)
	440	World Average	McAulay and Moran (1988)
	100 - 700	World Range	UNSCEAR (1988)

Table 6: Range of Gamma Dose Rates

	Dose Range (nGy h^{-1})			
State	^{40}K	^{226}Ra	^{232}Th	Total Dose (nGy h^{-1})
Akwa-Ibom	0.292 - 18.053	1.892 - 19.152	0.044 - 7.208	2.228 - 44.413
Bayelsa	2.232 - 24.289	9.575 – 29.435	3.427 – 10.049	15.234 – 63.773
Delta	3.805 - 22.744	12.12 – 42.085	3.744 – 8.691	19.669 – 73.52
Ondo	2.289 – 18.716	5.5 – 28.61	1.028 – 9.731	8.817 – 57.057

Rivers	2.901 – 23.757	12.295 – 23.29	3.04 – 9.638	18.236 – 56.685