

# THE IMPACT OF OIL AND GAS INDUSTRY TO THE NATURAL RADIOACTIVITY DISTRIBUTION IN THE DELTA REGION OF NIGERIA

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## Abstract

Delineation of natural radionuclides and the possibility of contaminating the natural environment by the extensive usage of radioactive materials in the delta region of Nigeria have been carried out by employing in situ gamma ray spectroscopy using NaI(Tl) detector. The mean activity concentrations for the respective radionuclides:  $^{40}\text{K}$ ,  $^{238}\text{U}$  and  $^{232}\text{Th}$  are  $34.8 \pm 20.4 \text{ Bq kg}^{-1}$ ,  $16.2 \pm 3.7 \text{ Bq kg}^{-1}$  and  $24.4 \pm 4.7 \text{ Bq kg}^{-1}$ . Areas with oil extraction activities have activity concentration values greater than areas without any known oil extraction activity in the region.

## INTRODUCTION

Oil and gas industry is a multi-faceted industry that includes the construction, exploration, production, and downstream and marketing sectors. In most of these sectors, radioactive materials and radiation generators are used on a large scale. The applications of radioactive materials in both the onshore and offshore oil and gas industry include industrial radiography, automated ionizing radiation gauges, well logging, use of radiotracers, mapping and evaluation of geological formations and the extraction of other natural hydrocarbon resources. The environment is constantly at the risk of pollution from the use of these radiation sources. There have been cases of lost radiographic sources resulting in deaths of workers and members of the public (IAEA, 2001). In view of the extensive usage of ionizing radiations both in the downstream and upstream sectors of oil and gas industry in the region, significant exposure to this radiation can lead to a potentially serious hazard to any individual member of the public either working or living around such facilities. Improper handling of gauge sources, well logging sources, radiotracers, spillage and releases of radioactive materials from pressurized systems, unauthorized disposal of waste, damage or rupture to source encapsulation, emergencies and natural disasters like fire, explosion and transportation accidents are some of the potential contamination that can lead to extremely large internal and external doses to the exposed individual of the public. In the possible event of environmental contamination by radioactive materials from any of these applications, there is always the need to assess the extent of contamination against some reference level, that is the baseline data of natural radioactivity. An in-situ gamma spectrometric survey of the type being embarked upon in this work is one way to assess the environment for possible contamination. Environmental radionuclides can be conveniently identified and their contributions to the total exposure rate determined using in-situ gamma ray spectrometry (Terada et al, 1980;

Gogolak and Endrulat, 1981; Murith et al, 1988; Miller and Shebell, 1993; Miller et al, 1994; Benke and Kearfott, 1997).

### **The geology of the delta region**

The geological map of the Niger Delta complex is shown in Fig 1. Three major depositional (sedimentary) environments (marine, mixed and continental) are observable. Based on the sedimentary environmental classification, the three rocks formations used in describing the subsurface sedimentary sequences are the Benin, Agbada and Akata formations (Short and Stauble, 1967; Frankl and Cordry, 1967; Whiteman, 1982; Merki, 1972; Hospers, 1971; Walcott, 1972). These formations form the bulk of the deltaic (Tertiary) sediments of the Niger Delta Complex as shown in Fig 2. The Niger Delta complex is a site of cyclic sedimentation; the complete cycle consists of thin fossiliferous transgressive marine sand followed by an off-lap sequence of marine shale and laminated fluviomarine sediments (Whiteman, 1982). The Benin formation (coastal plain sand) as shown in Fig 2, consists of coarse to medium sand, subordinate silt and clay lenses. The off-lap sequence general description of this formation indicates that it consists of continental fluvial gravels and sands. It was also observed that both disseminated and bedded lignite occur within the Benin formation and that the formation is partly marine, deltaic, estuarine lagoonal and fluviolacustrine in origin. It was found that the continuous shale with occasional sand bodies within the Benin formation is the Afam clay and it is the most extensive of the group of “clay fill” structures known from the Niger Delta Complex (Whiteman, 1982). The Agbada formation underlies the Benin formation and forms the second of the three strongly diachronous Niger Delta Complex formations. The formation consists mainly of alternations of sands, sandstones and siltstones. The sandy part constitutes the main hydrocarbon reservoirs in the Niger Delta oilfields (Lambert et

al, 1992). According to Whiteman (1982), the formation consists of sands in which grain size increases upwards and that the gamma radiation from natural radionuclides in the upper layer of the formation is often higher than in the underlying marine clays. The sandstones or sands are very coarse to very fine grain. The sands are often poorly sorted except where sand grades into shale. Lignite streaks and limonite are common but limonite coated sand grains and feldspars, which characterize the Benin formation, are rare. In the Agbada formation, shaliness increases downwards and the formation passes gradually into the Akata shale formation. The basal major time transgressive lithological unit of the Niger Delta Complex is the Akata formation. This is composed mainly of marine shale but contains sandy and silty beds. It consists of dark uniform shale, especially in the upper part and it is sandy and silty as it grades into the Agbada formation. The formation is known to be the main source rock for the Cenozoic Niger Delta Complex oil and gas (Braide, 1987).

## MATERIALS AND METHODS

The Niger Delta region comprises of thick rain forest and swamps with annual rainfall of about 1800 mm. In view of the swampy nature of the region, the area remains inaccessible for most of the year. The present studies were conducted in the month of March, which is the peak of the dry season in the area when most parts could be accessed with minimal difficulty. In all, 175 sites in 32 communities were surveyed and the distribution was such that all communities having one oil operational facility or the other in the area that were accessible were covered. A few communities were chosen in the region where oil activities are not known to be taking place. Such areas were included to serve as reference points for the study. The locations are indicated with numbers 1 to 32 in the map in Fig 1. Table 1 gives the actual names of the communities with oil operational activities. The sites with no activities going on are given in the Table 2. A

survey site was usually an open space devoid of vegetations and structures that could cause significant deviations. Depending on the terrain and accessibility, sites were between 250 m and 500 m apart. In situ gamma spectroscopy system, in which an unshielded detector was placed at 1 m above the ground level, was employed. A field spectrum was acquired using a 7.6 cm x 7.6 cm NaI(Tl) detector that was coupled to a Canberra series 10 plus Multichannel Analyzer (MCA) (Model No 1104) through a preamplifier base. The output signals from the NaI(Tl) detector were passed to the MCA through co-axial cables of about 5 m. The same length of cable carries high voltage necessary to bias the PMT of the detector from the MCA. The MCA operates on a rechargeable Cd-cell battery, which lasted for about 8 hours when fully charged. A standby 12V car battery was used whenever the in-built batteries run down in the field. The recharging of the in-built battery was done overnight for the following day work. The resolution of the detector is about 8% at 0.662 MeV line of  $^{137}\text{Cs}$ . Despite the poor resolution, which is normally associated with NaI(Tl) detectors, the  $^{40}\text{K}$ ,  $^{238}\text{U}$  and  $^{232}\text{Th}$  peaks of the spectrum acquired at each site were reasonably clean and strong with very low continuum. The peaks considered in this work are the 1.460 MeV peak for  $^{40}\text{K}$ , 1.760 MeV peak from  $^{214}\text{Bi}$  for  $^{238}\text{U}$  and 2.614 MeV peak from  $^{208}\text{Tl}$  for  $^{232}\text{Th}$ . The 0.662 MeV was used to investigate possible contamination of the soil by  $^{137}\text{Cs}$ . In view of the high temperature associated with the tropical environment and the poor condition of roads in the surveyed areas, efforts were made to avoid thermal fluctuations and mechanical damage to the detector. This was achieved by placing the detector in a fully cushioned wooden box and was exposed only during measurements. A routine check for possible drift in peak position was usually carried out before and after each measurement by placing a  $^{137}\text{Cs}$  point source under the detector.

## RESULTS AND DISCUSSION

### **Distribution of radionuclides activities in the region**

The mean of the several measurements made in each location using Zombori et al (1983) conversion factors was taken as the activity concentration value for the location and are presented in Table 3. The activity concentrations of  $^{40}\text{K}$  within the region ranged from  $13.4 \pm 0.7 \text{ Bq kg}^{-1}$  at Igbokoda to  $127.8 \pm 0.8 \text{ Bq kg}^{-1}$  in Calabar. The activity concentrations of  $^{238}\text{U}$  within the region ranged from  $10.8 \pm 2.0 \text{ Bq kg}^{-1}$  at Imo river flow station to  $26.7 \pm 1.5 \text{ Bq kg}^{-1}$  at Otujeremi. The activity concentrations of  $^{232}\text{Th}$  ranged from  $11.0 \pm 2.2 \text{ Bq kg}^{-1}$  at Ovu-Kokori to  $30.5 \pm 1.9 \text{ Bq kg}^{-1}$  at Ekulama. The mean activity concentrations for  $^{40}\text{K}$ ,  $^{238}\text{U}$  and  $^{232}\text{Th}$  are  $34.8 \pm 20.4 \text{ Bq kg}^{-1}$ ,  $16.2 \pm 3.7 \text{ Bq kg}^{-1}$  and  $24.4 \pm 4.7 \text{ Bq kg}^{-1}$ , respectively.

Generally, the three radionuclides could be said to be uniformly distributed in the region since 90.6%, 71.9% and 78.9% of the activity concentrations of  $^{40}\text{K}$ ,  $^{238}\text{U}$  and  $^{232}\text{Th}$ , respectively, lie within  $\pm 1\sigma$  of their mean values. The mean value of the activity concentration of  $^{40}\text{K}$  in the region is 46.1% of the combined activity concentration values of the three radionuclides while  $^{238}\text{U}$  and  $^{232}\text{Th}$  account for 21.5% and 32.4% respectively. The mean activity concentration values of  $^{40}\text{K}$ ,  $^{238}\text{U}$  and  $^{232}\text{Th}$  in the region were found to be lower than the earth's crustal mean of between  $37.8 \text{ Bq kg}^{-1}$  and  $49 \text{ Bq kg}^{-1}$  for  $^{238}\text{U}$  and about  $60 \text{ Bq kg}^{-1}$  for  $^{232}\text{Th}$  (Taylor, 1964). This can be attributed to the fact that the geology of the Niger Delta complex is made up of mainly sedimentary rocks that include sandstones, siltstones, marine shale, lignite, limestone, feldspars, etc. Since the weathering of rocks contribute to the formation of soil and that softer sedimentary rocks will have significant effects in this regard.

The activity concentration for locations with known oil extraction activities ranged from 16.4 Bq kg<sup>-1</sup> to 57.5 Bq kg<sup>-1</sup> with a mean of 33.23 ± 11.24 Bq kg<sup>-1</sup>, 10.8 Bq kg<sup>-1</sup> to 26.7 Bq kg<sup>-1</sup> with a mean of 16.37 ± 3.92 Bq kg<sup>-1</sup> and 11.0 Bq kg<sup>-1</sup> to 30.5 Bq kg<sup>-1</sup> with a mean of 25.17 ± 4.27 Bq kg<sup>-1</sup> for the respective radionuclides: <sup>40</sup>K, <sup>238</sup>U and <sup>232</sup>Th. The activity concentration for locations without any known oil extraction activities ranged from 13.4 Bq kg<sup>-1</sup> to 127.8 Bq kg<sup>-1</sup> with a mean of 41.08 ± 18.62 Bq kg<sup>-1</sup>, 10.8 Bq kg<sup>-1</sup> to 19.1 Bq kg<sup>-1</sup> with a mean of 15.40 ± 3.22 Bq kg<sup>-1</sup> and 13.9 Bq kg<sup>-1</sup> to 25.1 Bq kg<sup>-1</sup> with a mean of 20.04 ± 4.45 Bq kg<sup>-1</sup> for the respective radionuclides: <sup>40</sup>K, <sup>238</sup>U and <sup>232</sup>Th. The error stated in this work is the standard deviation, which gives the extent of spatial variation in the region. It was observed in the work that the locations with known oil activities have activity concentration values between 6.3% and 25.6% higher than areas without any known oil extraction activities. This indicates the extent to which the radiation burden of the areas with oil extraction activities has been increased. The implication of this is that the natural baseline level of radiation in the area has been disturbed due to the various activities of oil and gas industries in the area.

### **Distribution of radionuclide activities with rock formation in the region**

The activity concentration of <sup>40</sup>K has been found to account for the highest percentage of 46.1% compared to the 21.5% and 32.4% for <sup>238</sup>U and <sup>232</sup>Th respectively. This is consistent with Ahrens (1957) who noted that the activity of <sup>40</sup>K in sedimentary rocks depends upon the relative amounts of feldspar, mica and clay minerals that make up the mineral-aggregate sediments. The high percentage of <sup>40</sup>K could be attributed to the presence of feldspar and clay that characterized the Benin formation (Whiteman, 1982). Rogland and Rogers (1961) observed that most of the thorium in sedimentary rocks like sandstones is carried by quartz and feldspar. Hence, the thorium/uranium ratio, which

was found to be greater than unity in all the locations surveyed, may be due to the presence of sandstones and feldspar rock types in the Benin formation. The natural radiation level of the area is expected to depend mostly on the Benin formation (0 –2 km thick), which is the upper stratum of the three formations in the Niger Delta region. Since the in - situ method employed in this work is effective in measuring radioactivity in the topsoil up to a depth of 30 cm, radiation from the Agbada and Akata formations will be excluded except in locations near the sea where the Benin formation thins out as shown in Fig 2.

#### **Variation of radionuclide activities with mean distance from the sea.**

The delta area of Nigeria is of swampy vegetation and the swampy nature of the region increases toward the sea. The locations studied in this region have been grouped into five based on their estimated distance from the sea using the map of the area shown in Fig. 1 and are presented in Table 4. The knowledge of the radionuclide distribution with the mean distance of each group from the sea (MDS) will enable us to determine distribution pattern for the radionuclides in the area. The first group consists of those locations with MDS between 0 to 25 km. Twelve locations fall into this group representing 37.5% of the total location. The second group consists of locations with MDS between 26 – 50 km. Eight locations fall into this group representing 25% of the total location. The third group consists of locations with MDS between 51 - 75 km. Three locations representing 9.4% of the total locations fall into this group. The fourth group consists of locations with MDS between 76 - 100 km. Two locations fall into this group representing 6.2% of the total locations. The fifth group consists of locations with MDS between 101 – 125 km. Seven locations fall into this group representing 21.9% of the total locations. The above results are represented in the graph shown in Fig 2. The results indicate no direct pattern of variation for  $^{40}\text{K}$  the activity concentration with the

mean distance from the sea. For the  $^{238}\text{U}$ , the activity concentration could be said to decrease with increasing MDS up to 75km while  $^{232}\text{Th}$  activity increases with increasing MDS up to 75km, above this distance, no direct pattern of variation could be deduced. The data in Table 4 were fitted with polynomial equations for the respective radionuclides:  $^{40}\text{K}$ ,  $^{238}\text{U}$  and  $^{232}\text{Th}$  by employing the curve fitting software of the statistical tool pack of excel 2000 as:

$$A_{\text{K}} = -1 \times 10^{-6} \mathbf{D}^4 + 0.0002 \mathbf{D}^3 - 0.0054 \mathbf{D}^2 - 0.2013 \mathbf{D} + 38.319 \quad (\mathbf{R}^2 = 1) \quad 1$$

$$A_{\text{U}} = -3 \times 10^{-6} \mathbf{D}^4 + 0.0006 \mathbf{D}^3 - 0.0443 \mathbf{D}^2 + 1.137 \mathbf{D} + 7.9937 \quad (\mathbf{R}^2 = 1) \quad 2$$

$$A_{\text{Th}} = -2 \times 10^{-5} \mathbf{D}^3 + 0.0021 \mathbf{D}^2 - 0.0417 \mathbf{D} + 25.093 \quad (\mathbf{R}^2 = 0.99) \quad 3$$

where  $A_{\text{K}}$ ,  $A_{\text{U}}$  and  $A_{\text{Th}}$  are the activity concentrations for the respective radionuclides and  $\mathbf{D}$  is the mean distance from the sea. The coefficients of determination obtained for equations (1) – (3) show that the polynomial relationship between the activity concentration and the mean distance from the sea is appropriate.

## CONCLUSION

In view of the oil extraction activities in the delta region of Nigeria where radioactive sources have been in use for both the upstream and downstream operations, activity concentration values of natural radionuclides have been determined for this region. It was observed that the locations with known oil activities have activity concentration values between 6.3% and 25.6% higher than areas without any known oil extraction activities. The implication of this is that the radioactivity level of the region has been enhanced by the oil extraction activities in the area. However, no artificial radionuclide was detected in the surveyed area. The distribution of the radionuclides in the region shows no direct pattern of variation, which suggested that the variation of

natural radionuclides depend, to a large extent on the local geology of a particular environment.

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Table 1: Locations of areas with and without oil activities

Location No.	Communities	Oil Industries (Activity)
1	Okitipupa	None
2	Igbokoda	None
3	Mahin	Shell-Bp (Oil well)
4	Warri River Flow Station	Shell-Bp (Oil well)
5	Ovu-kokori	Shell-Bp (Oil well)
6	Otorugu	Shell-Bp (Oil well)
7	Otujeremi	Shell-Bp (Oil well)
8	Evwreni	Shell-Bp (Oil well)
9	Oroni	Shell-Bp (Oil well)
10	Uzede West	Shell-Bp (Oil well)
11	Uzede East	Shell-Bp (Oil well)
12	Opukushi	Shell-Bp (Oil well)
13	Sagbama	Shell-Bp (Oil well)
14	Okoso-logan	None
15	Oloibiri	Shell-Bp (Oil well)
16	Soku	Shell-Bp (Oil well)
17	Ekulama	Shell-Bp (Oil well)
18	Krakama	Shell-Bp (Oil well)
19	Abonnema Wharf	Shell-Bp (Oil well)
20	Elelenwo	Shell-Bp (Oil well)
21	Obigbo	Shell-Bp (Oil well)
22	Imo River Flow Station	Shell-Bp (Oil well)
23	Obunku	Shell-Bp (Oil well)
24	Afam	Shell-Bp (Oil well)
25	Ibibio	Shell-Bp (Oil well)
26	Ibeno	Mobil
27	Qua Ibo River Flow Station	Mobil
28	QIT (Mobil Platf.)	Mobil (Oil Platform)
29	Ifiayong	None
30	Eket	Mobil
31	Uyo	None
32	Calabar	None

Table 2: The mean specific activities of  $^{40}\text{K}$ ,  $^{238}\text{U}$  and  $^{232}\text{Th}$  for the region

Location	$^{40}\text{K}$ (Bq.kg $^{-1}$ )	$^{238}\text{U}$ (Bq.kg $^{-1}$ )	$^{232}\text{Th}$ (Bq.kg $^{-1}$ )
1	19.5 ± 6.3	17.2 ± 4.3	13.9 ± 3.8
2	13.4 ± 0.7	10.8 ± 0.7	17.2 ± 2.0
3	16.4 ± 5.2	12.1 ± 1.0	18.0 ± 1.8
4	44.3 ± 3.2	24.4 ± 4.3	22.8 ± 1.6
5	28.5 ± 2.6	11.6 ± 2.0	11.0 ± 2.2
6	53.2 ± 8.0	22.1 ± 3.4	28.2 ± 4.5
7	57.5 ± 7.1	26.7 ± 1.5	29.0 ± 3.3
8	39.7 ± 7.5	18.4 ± 1.6	23.1 ± 3.1
9	41.7 ± 6.5	18.0 ± 1.4	25.9 ± 2.9
10	25.1 ± 3.4	13.1 ± 4.0	19.9 ± 1.8
11	27.5 ± 3.6	11.7 ± 2.1	18.3 ± 1.9
12	43.7 ± 3.4	15.7 ± 3.9	27.4 ± 2.8
13	49.4 ± 4.4	14.7 ± 4.7	25.5 ± 3.9
14	22.8 ± 13.0	13.7 ± 5.6	21.6 ± 4.1
15	48.8 ± 4.4	18.0 ± 1.4	27.7 ± 2.9
16	36.3 ± 5.2	15.5 ± 2.8	26.7 ± 1.8
17	42.5 ± 2.9	14.8 ± 2.6	30.5 ± 1.9
18	36.3 ± 6.0	13.9 ± 6.3	29.7 ± 0.9
19	36.1 ± 3.0	15.7 ± 2.1	26.5 ± 2.5
20	28.1 ± 2.6	14.2 ± 3.9	29.0 ± 2.9
21	32.7 ± 8.5	16.2 ± 2.8	26.0 ± 3.0
22	18.8 ± 2.2	10.8 ± 2.0	28.0 ± 1.8
23	24.9 ± 7.4	12.5 ± 3.9	27.2 ± 1.9
24	26.0 ± 5.0	13.4 ± 4.0	26.4 ± 2.6
25	36.5 ± 4.8	17.9 ± 3.2	28.0 ± 2.3
26	24.6 ± 4.3	21.1 ± 2.5	23.9 ± 2.8
27	21.2 ± 1.5	18.3 ± 2.7	29.8 ± 3.1
28	26.0 ± 3.9	21.2 ± 4.0	24.4 ± 1.9
29	22.7 ± 4.5	15.2 ± 2.5	23.6 ± 2.1
30	20.9 ± 7.3	15.6 ± 3.9	24.1 ± 2.1
31	21.9 ± 6.7	16.2 ± 4.0	22.4 ± 3.4
32	127.8 ± 0.8	19.1 ± 6.4	25.1 ± 10.5
Mean ± SD	34.8 ± 20.4	16.2 ± 3.7	24.4 ± 4.7

Table 3: The mean specific activities with mean distance from the sea

Location (km)		$^{40}\text{K}$ (Bq kg $^{-1}$ )		$^{238}\text{U}$ (Bq kg $^{-1}$ )		$^{232}\text{Th}$ (Bq kg $^{-1}$ )	
Range	MDS	Range	Mean	Range	Mean	Range	Mean
0 – 25	12.5	13.4 – 127.8	35.3± 29.2	10.8 – 21.2	16.4± 3.1	17.2 – 30.5	24.9± 4.1
26 – 50	37.5	19.5 – 43.7	30.9± 7.3	12.5 – 17.2	15.1± 1.5	13.9 – 29.0	25.4± 4.4
51 – 75	62.5	18.8 – 49.4	33.6± 12.5	10.8 – 16.2	13.9± 2.3	25.5 – 28.0	26.5± 1.1
76 – 100	87.5	22.8 – 57.5	40.2± 17.4	13.7 – 26.7	20.2± 6.5	21.6 – 29.0	25.3± 3.7
101 – 125	112.5	25.1 – 57.5	37.1± 9.6	11.6 – 26.7	16.6± 4.3	11.0 – 29.0	21.3± 5.3

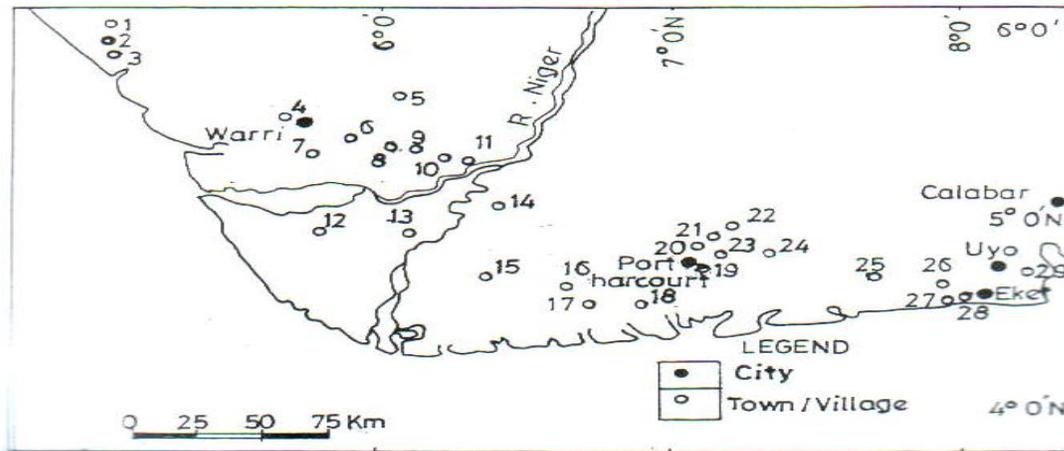


Fig. 1: Map showing locations in the region

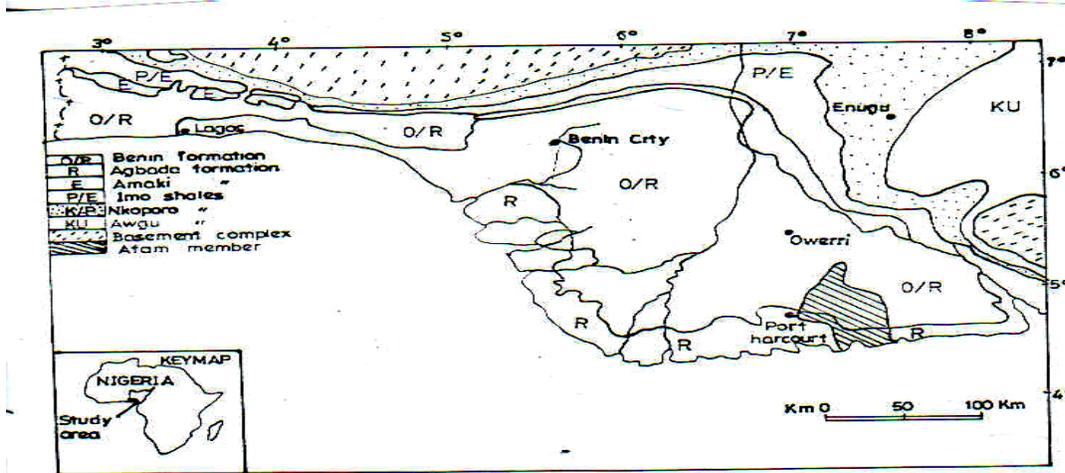


Fig. 2: The geological map of Niger Delta complex after Short and Stauble (1967)

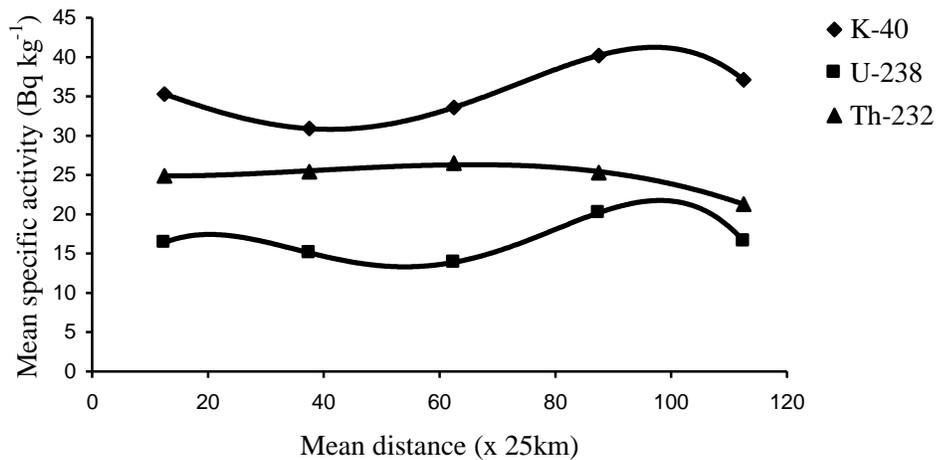


Fig 3: Variation of specific activities with mean distance from the sea