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Determination of radioactivity level and hazard assessment of unconsolidated sand and shale soil samples from petroleum oil field at Oredo (Benin, Niger Delta-Nigeria)

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ABSTRACT

Activity concentration of naturally occurring radionuclides in soil from Oredo oil field in Benin city, Edo State Nigeria have been measured using gamma-ray spectroscopy. A total of sixty-four samples were collected in the field. The range of average activity concentration of ^{226}Ra, ^{232}Th and ^{40}K were found to be 3.80 to 16.28 Bq kg^{-1}, 11.69 to 62.48 Bq kg^{-1} and 47.75 to 406.42 Bq kg^{-1} respectively. The average absorbed dose rates ranged from 17.50 to 51.33 nGy h^{-1} with an overall mean of 46.75 nGy h^{-1}, while the average annual effective dose rates ranged from 21.46 to 62. 92 µSv y^{-1} with an overall mean of 46.30 µSv y^{-1}. The overall average of external hazard index was 0.22. All the samples have radium equivalent activities lower than the limit (370 Bq kg^{-1}) set by UNSCEAR. The overall average annual gonadal dose associated with the field does not exceed permissible levels by UNSCEAR.

Keywords: Soil; Spectrometry; Activity concentration; Hazard assessment.

INTRODUCTION

Nigeria has one of the world's largest marshy-lands called Niger Delta. This covers 20000 Sq. km. However, this area is facing severe threat from man through environmental pollution by oil spills and gas flaring [1]. A lot of multinational oil companies do carry out oil exploration and production activities within the area. These led to refine, storage and transportation of large amounts of crude petroleum through this marshy lands to the adjacent near shore habitat [1]. The amount of radioactivity in soil varies widely and is a source of continuous exposure of human kinds to terrestrial radioactivity. Exposure to terrestrial radioactivity from soil depends on many factors such as types of soil, uses [2] most-especially oil exploration. Activities involving in oil and gas exploration and production had been shown to contain naturally occurring radioactive materials (NORM) from uranium, radium, thorium and their daughters [3] and lead. Geologists have recognized the presence of aforementioned radionuclides since the early 1930 and use it as a method of prospecting deposits. Wastes such as mineral scale inside pipes [4] sludge's, contaminated equipment or components produced waters are produced during exploration [5]. Also according to United State Environmental Protection Agency (USEPA), filed surveys have shown that petroleum pipe scale originating from oil production may have very high ^{226}Ra concentration, and on disposal exposes the environment to associated radioactive contamination [6]. Exposure to NORM by humans occurs in different pathways. Population at risk from this exposure includes workers at equipment, cleaning of facilities and oil field workers through inhalation

of ^{222}Rn; a decay product of ^{226}Ra [7]. More also pathways of concern to the general public are through water ingestion and radon inhalation [8] due to transportation of soil from oil and gas producing areas to other regions through various geomorphologic processes such as erosion, flooding, leaching and eluviation. However information on the radioactive content of soils from Oredo oil field is not available. This study therefore determined the presence, concentrations and hazards potentials of natural radionuclides in soils from Oredo oil field in Benin Niger Delta.

MATERIALS AND METHODS

2.1 STUDY AREA AND ITS GEOLOGY

Oredo is one of the three Local Governments in Benin metropolis in Edo State [9]). Benin as the capital of Edo lies between latitudes 6^0 20^1N and 6^0 31^1N and between longitudes 5^0 35^1 East and 5^0 41^1 East of Greenwich meridian. By its tropical location, it has a temperature of about 27^0C and annual rainfall of over 2000mm [10]. Oredo has a total land area of about 317.08 km^2 and a population of 344671 [11]. The formations found in the area are mostly unconsolidated sands and shale [12]. The oil field is located in approximately 40 km south east of Benin City and lies within the oil prolific belt of Niger Delta-Nigeria [12].

2.2 SAMPLING AND SAMPLE COLLECTION

The area was divided into eight (8) plots and a total of sixty four samples were collected to a depth of 10 cm. The samples were properly labeled catalogued and brought to the radiation laboratory at the Centre for Research and Development (CERD) at Obafemi Awolowo University Ile-Ife, Nigeria for processing before analysis.

2.3 SAMPLE PREPARATION, PROCESSING AND PACKING

Soil samples were dried at room temperature to constant weight and sundry at 25± 2^0C to remove the moisture content. The samples were also oven dried at a temperature of 105^0C [13]. The removal of moisture took care of self-absorption in each of the sample. The dried samples were pulverized into fine grains so as to increase the total emission area [14] and passed through a sieve of mesh size 200 µm, so that clay and mineral particle may homogenize [2]. Samples were sealed in air tight plastic containers of diameter 6.5cm that could seat in the detector head. Samples were left for more than one month in order to allow secular equilibrium between ^{226}Ra and ^{222}Rn and its decay products before counting by gamma-ray spectrometry.

2.4 RADIOACTIVE MEASUREMENT

Gamma-ray spectroscopy method was employed for the measurements of the radioactivity in the samples and the procedures of this method as described in literature were followed [15]. A 76 mm x 76 mm NaI (Tl) detector crystal couple to a Canberra series 10 plus multichannel analyser (MCA) through a photomultiplier tube was used for the radioactivity measurement. IAEA-375 reference material was employed for the efficiency calibration of the system. The specific activities of the measured radionuclides are in good agreement with the reference values. The activity concentration of ^{232}Th was determined by the mean of the specific activities of ^{208}Tl, ^{212}Pb and ^{228}Ac and the activity concentration of ^{226}Ra was the mean specific activities due to gamma energies of ^{214}Pb and ^{214}Bi [2], ^{40}K was measured directly using the 1.460 MeV photopeak. Each sample was counted for 10 hrs. [16] in order to reach ± 5 % of analytical accuracy of measurements.

RESULTS AND DISCUSSION

3.1 ACTIVITY CONCENTRATION

The average concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the oil area are shown in Table 1 and Fig 1. The average activity concentrations range for ^{226}Ra, ^{232}Th and ^{40}K are 3.80-16.28 Bq kg^{-1} with an overall average 12.94 ± 3 Bq kg^{-1}, 11.69-62.43 Bq kg^{-1}with an overall average 35.69 ± 4 Bq kg^{-1} and 47.75-406.42 Bq kg^{-1} with an overall average 199.04 ± 4 Bq kg^{-1}, respectively. The concentration of ^{40}K accounts for approximately 80 % of the total gamma activity of the soil samples. This indicates that the specific activity due to ^{40}K is the largest contributor to the total activity for all soil samples. The results in Table 1 reveal that the overall activity concentrations of these radionuclides are in conformity with the worldwide concentrations.

3.2 ABSORBED DOSE RATE (D)

Neglecting the contribution of radionuclides such as ^{137}Cs, ^{90}Sr and ^{235}U decay series due to their little contribution, absorbed dose rate is calculated based on the concentrations of three measured naturally occurring radionuclides.

Based on the facts that radiation exposure pathways involved inhalation of radioactive pollutants, it is imperative to calculate absorbed dose rate. This is done by using the direct relationship between terrestrial gamma radiation and radionuclide concentrations at 1 m above ground as proposed by Beck [17] and UNSCEAR [18]:

$$D(nGy\ h^{-1}) = 0.427A_{Ra} + 0.662A_{Th} + 0.0432A_K \quad (1)$$

Where A_{Ra}, A_{Th}, A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1}, respectively.

The absorbed dose rates in air for the field investigated are listed in column 4 of Table 1. The average absorbed dose rates at Oredo oil field for soil samples varied from 17.50-51.33 $nGy\ h^{-1}$ with an overall average 37.75 $nGy\ h^{-1}$ (Fig 3). This value is lower than the population- weighted average value of global primordial radiation of 55 $nGy\ h^{-1}$ [18, 19].

3.3 RADIUM EQUIVALENT

Activity mass concentration due to ^{226}Ra, ^{232}Th and ^{40}K is not uniformly distributed throughout in the soil [2]. This non-uniformity in respect of exposure to radiation has been defined in terms of radium equivalent activity (Ra_{eq}). This is a single quantity that takes into consideration the radiation hazard associated with ^{226}Ra, ^{232}Th and ^{40}K. It is calculated as [20, 21].

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (2)$$

Where A_{Ra}, A_{Th}, A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1}, respectively.

The average values of Ra_{eq} varied from 36.85-110.98 Bq kg^{-1} with an overall of 79.30 Bq kg^{-1} as seen in column 6 of Table 1. The estimated overall average value of Ra_{eq} in the present work is lower than the 370 Bq kg^{-1}[8]. Comparing this value with values from other countries of the world, it is observed that the overall average is lower than the measured values for all other countries (Table 2).

3.4 EXTERNAL HAZARD INDEX (H_{ex})

The external hazard index (H_{ex}) is another parameter of interest. It is calculated by a model proposed by Krieger as:

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (3)$$

It must not exceed unity for the radiation hazard to be negligible [22]. The mean results obtained are presented in Table1. The results range from 0.11-0.30 with an overall average of 0.22. The overall average is found to be < 1 for the study area.

3.5 ANNUAL GONADAL DOSE EQUIVALENT (AGDE)

UNSCEAR considered the activity of bone marrow and bone surface cells as the organ of interest when estimating dose equivalent [18, 19]. Therefore, AGDE due to the specific activities of ^{226}Ra, ^{232}Th and ^{40}K was calculated using the following relation [19, 22]:

$$AGDE(\mu Sv\ y^{-1}) = 3.09A_{Ra} + 4.18A_{Th} + 0.314A_K \quad (4)$$

The average values of *AGDE* are presented in Table 1. The values varied from 115.2-334.0 $\mu Sv\ y^{-1}$ (Fig 2) with an overall average found to be 214.3 $\mu v\ y^{-1}$. The overall average is lower than 2398 $\mu Sv\ y^{-1}$ obtained at Eastern Desert of Egypt [23].

3.6 ANNUAL EFFECTIVE DOSE EQUIVALENT (AEDE)

Applying the conversion factor of 0.7 Sv Gy^{-1}, which convert absorbed dose in air to human effective dose and using an outdoor occupancy factor of 0.2 as recommended by UNSCEAR [8], the average annual effective dose due to gamma-radiation from these terrestrial sources at Oredo oil field can be assessed. However, since people because of socio-economic reasons would always be in the field, 0.2 outdoor occupancy factor was used in this study. The annual effective dose equivalent was calculated from following relation:

$$AEDE(\mu Sv\ y^{-1}) = D(nGy\ h^{-1}) \times 8760(h\ y^{-1}) \times 0.2 \times 0.7(Sv\ y^{-1}) \times 10^{-3} \quad 5$$

The results of the calculation are given in Table 1. The mean *AEDE* values varied from 21.46-62.95 $\mu Sv\ y^{-1}$ with an overall average 46.3$\mu Sv\ y^{-1}$. The overall average is lower than 70 $\mu Sv\ y^{-1}$ (world averaged) [18]. The summary of the aforementioned radiation hazard parameters are presented in Fig 2.

CONCLUSION

The activity concentrations of primordial radionuclides in soil of Oredo oil field in Benin, Edo-State Nigeria have been studied in this work. The results obtained were used for estimation of the potential health risk in the area. Hazard parameters were found to be within the range specified by UNSCEAR [8] report. Therefore, it is suffices to say that soil samples from the environment will not pose any significant radiological threat to the populace.

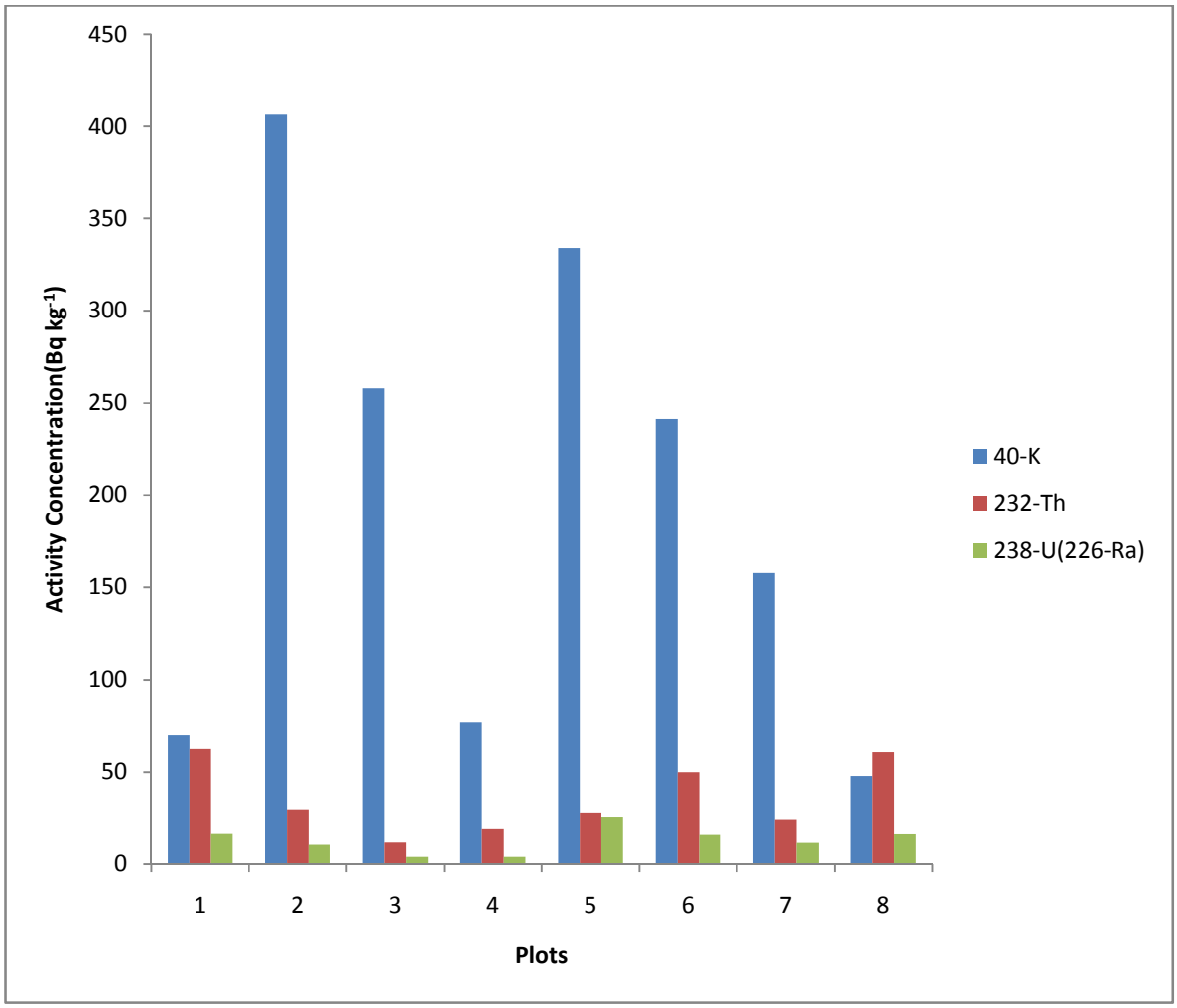

Fig 1. The concentration variation of ^{40}K, ^{232}Th and ^{226}Ra in the collected soil samples

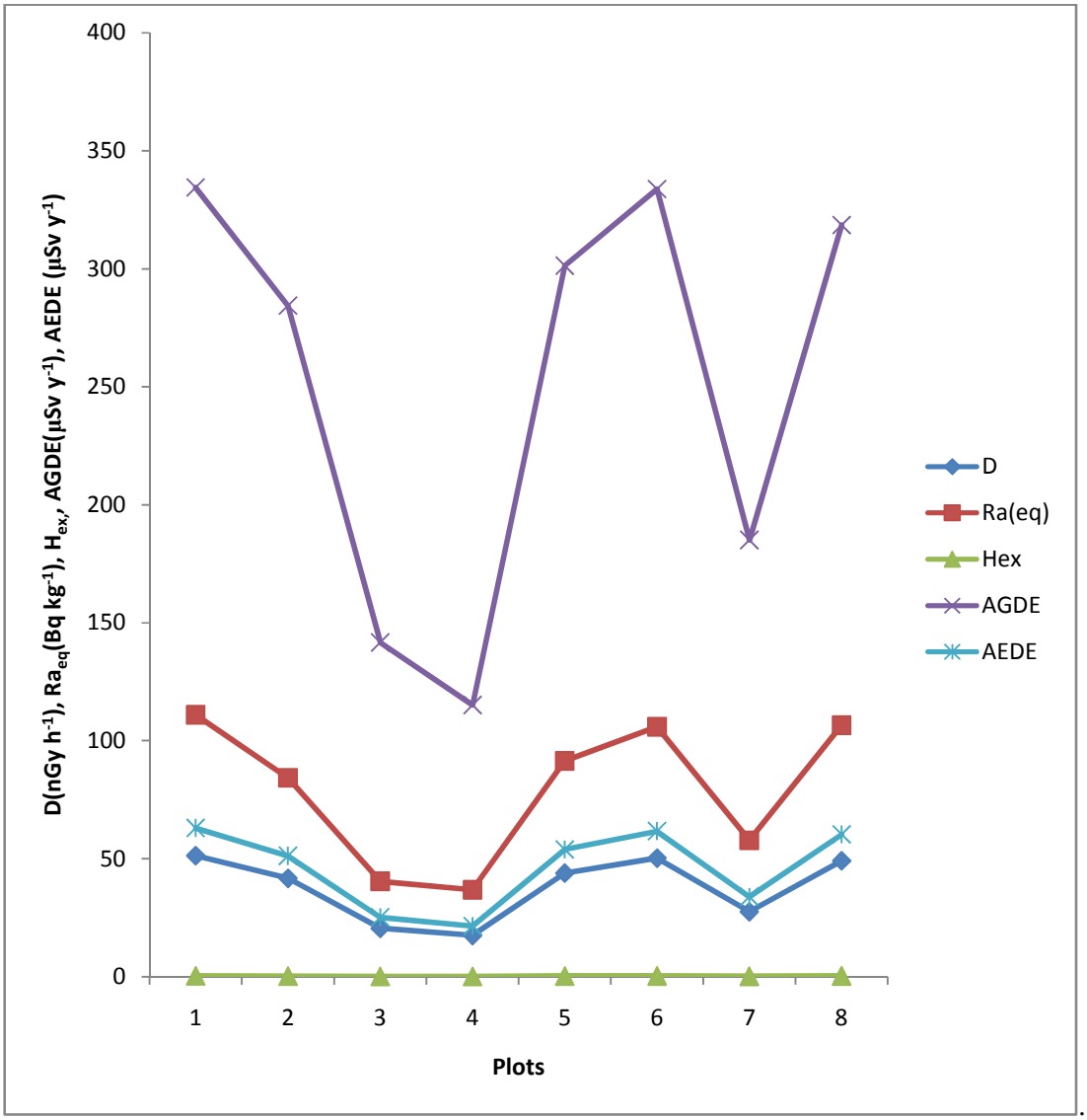

Fig 2: Distribution of absorbed dose rate, radium equivalent, external hazard index, annual gonadal equivalent and annual effective dose rates

Table 1 Average radioactivity concentrations, absorbed dose rates, radium equivalent, external hazard index, annual gonadal dose equivalent and annual effective dose rates

Plot	^{40}K±σ Bq kg^{-1}	^{232}Th±σ Bq kg^{-1}	^{226}Ra±σ Bq kg^{-1}	D nGy h^{-1}	Raeq Bq kg^{-1}	Hex	AGDE µSv y^{-1}	AEDE µSv y^{-1}
1	69.84±3	62.48±3	16.28±3	51.33	110.98	0.30	334.40	62.95
2	406.42±5	29.76±4	10.47±2	41.73	84.32	0.23	284.36	51.18
3	258.17±5	11.69±3	3.80±2	20.51	40.40	0.11	141.67	25.15
4	76.77±4	18.98±4	3.80±2	17.50	36.85	0.09	115.18	21.46
5	334.04±5	27.96±5	25.75±4	43.94	91.45	0.25	301.33	53.89
6	241.59±4	50.02±5	15.81±3	50.30	105.94	0.29	333.80	61.69
7	157.72±4	23.89±4	11.54±3	27.56	57.85	0.16	185.04	33.80
8	47.75±3	60.73±3	16.07±3	49.13	106.59	0.29	318.50	60.25
Overall Average	199.04±4	35.69±4	12.94±3	37.75	79.30	0.22	214.29	46.30
Worldwide Average*	580	40	40	55	370			460

Table 2 Overall average radium equivalent from Oredo compared with different countries

Country	Ra_{eq} (Bq kg^{-1})
Eastern Desert Egypt	493.8 [23]
Eskisehir, Turkey	366.9 [24]
Xiazhuang, China	266.0 [25]
Northern, Jordan	103.1 [22]
Rize, Turkey	166.3 [19]
Oredo, Nigeria	79.3 present study

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