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ORIGINAL PAPER

Assessment of radionuclide distribution and associated radiological hazards for soils and beach sediments of Akwa Ibom Coastline, southern Nigeria

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Abstract

In this study, the activity concentrations of natural radionuclides in soil and beach sediment samples collected from 15 randomly but uniformly distributed locations in the coastal area of Akwa Ibom (southern Nigeria) were measured using hyper pure germanium (HPGe) detector. The activity concentrations of ^{238}U, ^{232}Th and ^{40}K varied from 9 to 44 Bq/kg, 3 to 72 Bq/kg and 35 to 250 Bq/kg respectively. These results are below the 1000, 1000 and 4000 Bq/kg upper limits of safety for ^{238}U, ^{232}Th and ^{40}K respectively. Except ^{238}U, ^{232}Th and ^{40}K show lower standard deviations than their mean values, which indicate that soils and sediments have high degree of uniformity. The spatial distribution of radionuclide concentrations is nearly uniform except at locations where carbonaceous materials and clay soils exist. Mean value of Th/U ratio across the area was ≤ 7, typical of reducing condition in the depositional environment, thus suggesting enrichment of carbonaceous materials and clays. Additionally, the concentration of ^{40}K was observed to be slightly higher in locations where anthropic activities are prevalent. Apart from excess lifetime cancer risk (average concentration of 0.80×10^{-3}), the average concentrations of other radiological parameters: radium equivalent (83 Bq/kg), absorbed dose rate (39 nGy/h), indoor annual effective dose equivalent (0.18 mSv/year), outdoor annual effective dose equivalent (0.05 mSv/y) and annual gonadal dose equivalent (259 μSv/y), are below permissible limits. Hence, the soils and sediments are safe and suitable for all purposes.

Keywords Radionuclides · Radionuclide ratios · Radiological parameter · Beach sediments · Soils

Introduction

Radioactivity from radionuclides such as ^{238}U, ^{232}Th and ^{40}K and their decay products present in soils and rocks have over time posed serious environmental hazard to both man and a variety of animal species in their natural environments. Uranium and thorium series progenies (i.e., ^{226}Ra, ^{222}Rn ^{218}Po, ^{214}Pb, ^{214}Bi and ^{214}Po) and (i.e., ^{228}Ra, ^{220}Rn ^{216}Po, ^{212}Pb, ^{212}Bi and ^{212}Po) respectively, and ^{40}K are the most concerned. Studies have shown that about 96% of the total radiation received on the earth surface originates from natural sources, while about 4% is emitted by artificial sources (Chougaonkar et al. 2003). Natural radionuclides that yield constant activity originate from terrestrial (e.g., weathering of granitic rocks) and cosmogenic sources (Nizam et al. 2013; Issa and Alaseri 2015; Khuntong et al. 2015). Thus, rock forming minerals contain radionuclides that are natural in origin (Szarlowicz et al. 2019). High radiation levels are attributable to the presence of some radionuclide-bearing accessory minerals, e.g., monazite, zircon, allanite, apatite, sphene, mica and feldspars (Kannan et al. 2002; Ramola et al. 2011; Hannan and Nguyen 2013; Sivakumar 2014); colloidal iron; and manganese oxides and hydroxides (McKee 2008; Jurina et al. 2013; Raghu et al. 2015; Akpan et al. 2016). Artificial radioactivity originates from anthropogenic activities such as nuclear weapon testing programmes and accidental discharge from nuclear power plants that release radionuclides into the environment. Also, excessive application of phosphate fertilizers in agricultural soils have been reported to cause increased artificial radioactivity in soils (Todorović et al. 2015). Additionally, increased activity concentration due to industrial activities such as heavy metal mining, oil exploitation, processing and transportation activities have also been reported (Carvalho et al. 2007, 2009; United Nations

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Environmental Programme, UNEP 2011). These activities are prevalent within the study area, hence, the need for routine assessment. Soils and sediments have been identified as potential repository for radionuclides and other types of contaminants (Szarlowicz et al. 2019). Accumulation of radionuclides in terrestrial-marine and marine environments portends grave danger to both aquatic and human health. Environmental factors such as soil redox potential, clay-sized soil fraction, degree of weathering, concentration of contaminants, organic matter content, pH, temperature fluxes and precipitation have been identified as elements that controls bioavailability, uptake and transfer of these radionuclides in soils and different food chains (Ribeiro et al. 2018; Saint-Fort 2018). These processes are governed by the physical and chemical characteristics of the ecosystem. However, the transfer mechanism of radionuclides into biological systems is analogous to how these systems obtain nutrient from soil, water and food (Saint-Fort 2018). When present in sediments, radionuclides pose exposure risk due to their increased mobility and high solubility (Nizam et al. 2013; Momčilović et al. 2013; Issa and Alaseri 2015; Ravisankar et al. 2015). Radionuclides are retained in soils, which serve as interface that connects other components of the physical environment such as air and water (Chen et al. 1999). Radionuclides in soils and sediments can be transmitted either directly or in solution to animals, plants and the other components of the environment through inhalation, injection and ingestion where it accumulates in the host to harmful levels (Binesh et al. 2010; Raghu et al. 2015). The risk factor due to radioactive decay of ^{238}U and ^{232}Th series including ^{40}K is the release of gamma radiations that can pose external hazard to the surrounding or internal via inhalation and ingestion of radon and its decay products by man and biota (United Nations Scientific Committee on the Effects of Atomic Radiation, UNSCEAR 1988; Khalifa and El Arabi 2005; Povinec and Hirose 2015). High incidences of some chronic diseases (e.g., lung and kidney cancers, acute leucopoenia and anaemia) have been attributed to ionizing radiations from radioisotopes (Taskin et al. 2009). More so, ionizing radiation can result in mutation due to destruction of DNA (Busby 2013).

Recently, studies on natural radionuclide origin and behaviour (Michalik et al. 2013; Bondareva et al. 2017; Szarlowicz et al. 2019), spatial distribution in soil, sediments, water and plants (El-Gamal et al. 2007; Manigandan and Manikandan 2008; Navas et al. 2011; Ćujić et al. 2015), food crops (Jibiri et al. 2007) and impact of phosphate ores and fertilizers on soils (El-Taher and Abdelhalim 2013; Hassan et al. 2017; Boumala et al. 2018; Bramki et al. 2018; Ugolini et al. 2020) have dominated many scientific fora. Reports of radionuclide enrichment in some sea foods and snails also exist (Musthafa and Krishnamoorthy 2012; Manigandan et al. 2015). Smičiklas and Šljivić-Ivanović (2016) assessed radioactive contaminants in soils and suggested possible remediation strategies such as stabilization and chemical extraction techniques. Research show that beach sediments and sands often used in building houses contain certain levels of naturally occurring radioactive materials resulting from primordial radionuclide of cosmic origin (UNSCEAR 1982; Powell et al. 2007; Newman et al. 2008). Activity concentration of natural radionuclide in soils and sediments is dependent on rock type from which these materials occurred prior to weathering (Ravisankar et al. 2015). Issa and Alaseri (2015) reported that activity concentration of radionuclide in building materials could be of geologic origin and can also result from geochemical alterations of those materials. Radiological assessment of activity concentration is essential for monitoring and evaluating radioactive hazards, setting standards and guidelines necessary for environmental management and remediation (Porcelli and Baskaran 2011). An understanding of the abundance of radionuclides in marine sediments is useful in determining source of sediments, dynamics, transport mechanism and environmental reconstruction (Sondi et al. 1995; Baggoura 1997; Powell et al. 2007; Matisoff 2014). It can also be used to assess radiation risk on those who make their living around the beach and the rural dwellers that make contact with these sediments on a daily basis. Several researchers have devoted interest in the investigation of natural radioisotopes in coastal sediments (Amekudzie et al. 2011; Tari et al. 2013; SureshGandhi et al. 2014; Ravisankar et al. 2015; Issa and Alaseri 2015;). Reconnaissance investigations have been conducted in coastal, terrestrial-marine and marine environments to quantify the spatial variability of radionuclides in sands in India (Kannan et al. 2002; Sivakumar 2014; Raghu et al. 2015), Nigeria (Tchokossa et al. 2012; Akpan et al. 2016) and China (Lu et al. 2008; Wang et al. 2011). However, investigation of this sort has not been conducted around the Akwa Ibom coastline. The aim of this study is to assess the activity levels and radiologic effects resulting from exposure to radionuclides present in coastal soils and sediments of Akwa Ibom, southern Nigeria.

Physiography and geology

Site description

The study area is located between longitudes 7° 30′ and 8° 30′ E and latitudes 4° 25′ and 4° 40′ N of the equator along the coastline of Akwa Ibom, southern Nigeria. Five coastal enclaves comprising Oron, Eket, Ibeno, Eastern Obolo and Ikot Abasi were covered in the study (Fig. 1). The area is within the equatorial climatic zone that is characterized by two seasons—rainy (March–October) and dry (November–February). The dry season usually begins with gradual reduction in rainfall intensity occasioned by the arrival of a southward heading Sahara Desert-borne tropical continental air mass, which blows across the area. The rainy season normally starts when the northward heading moisture-laden trans-Atlantic air mass blows cross the area. Generally, the area around these coastal communities consists of beach ridge

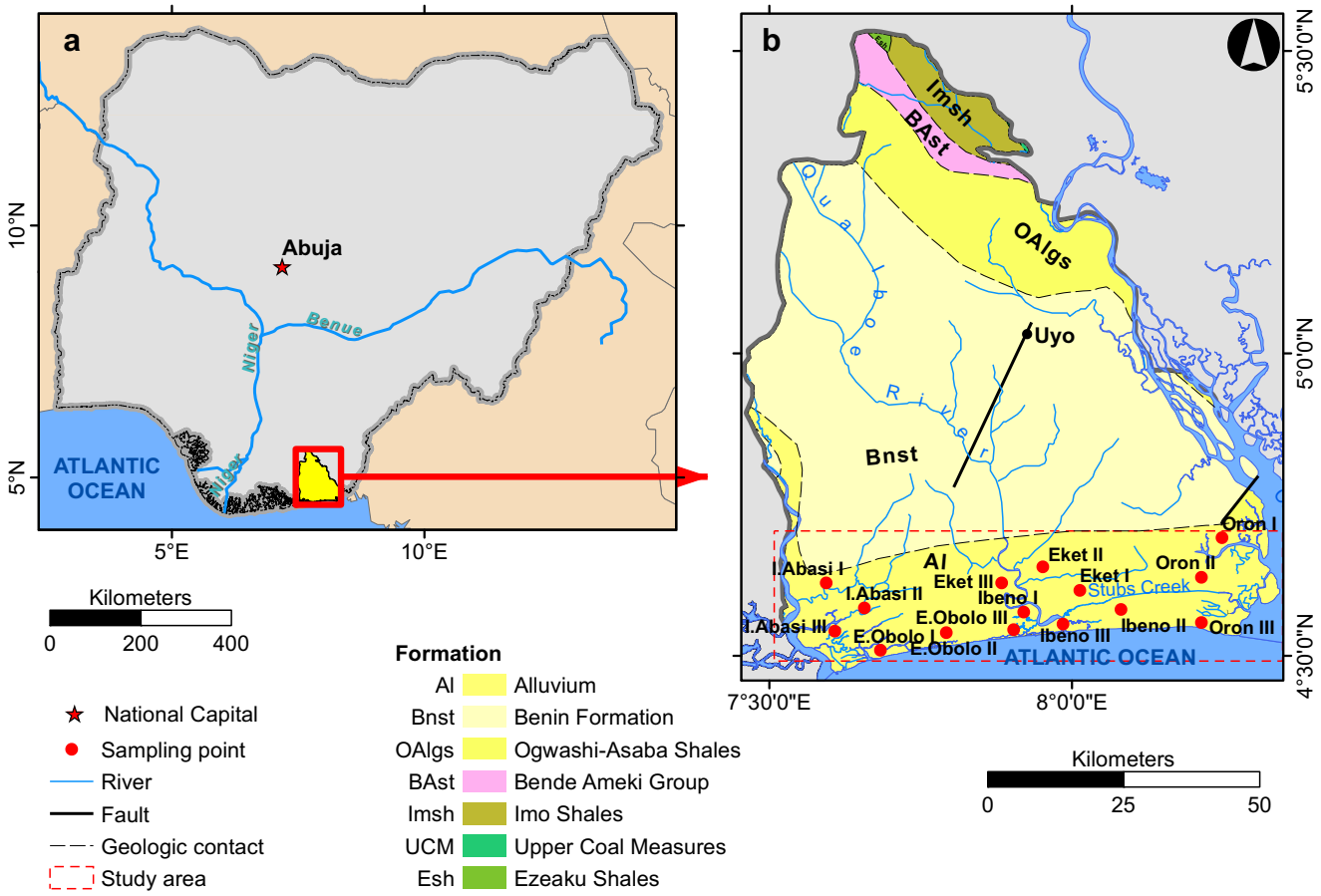

Fig. 1 Map of Nigeria showing the location of Akwa Ibom State (**A**) and generalized geological map of Akwa Ibom State showing the sampling points along the coastline (marked with red dotted lines) and Atlantic Ocean (**B**)

complexes, mangrove swamps of southern Nigeria and landward, gently undulating coastal plain. The beach ridge complex forms parts of the agricultural lands reserved for farming by the local people. Within the farming areas, varieties of crops and vegetables are cultivated biannually and both organic and inorganic chemicals (mainly nitrogeneous fertilizers), and herbicides are applied sometimes to enhance soil fertility and crop yield. Besides farming, the area is the hub of oil exploration, exploitation and refining activities and these anthropic activities have resulted in serious environmental challenges leading to increased activity concentration in soils (UNEP 2011). Other prevailing human activities, which can also enhance heavy metal and radionuclide concentrations in beach sediments, include marine transportation, fishing, mining and boat making (Tam and Wong 2000; Li et al. 2007; Ravisankar et al. 2015).

Geology

The study area is composed of Late Tertiary-Holocene sediments. The coastal and border-lines of the estuaries and rivers like Qua Iboe and Imo are surrounded by alluvial sediments that connect the river bodies with the Coastal Plain Sands in the off-coast areas. These bank sediments consist mainly of clays, silts and silty-sands that are fine to medium and well sorted. Farther away from the coast, the sedimentary succession grades into the Coastal Plain Sands. Basically, the sediment characteristics determine the nature of soils and are primarily influenced by environmental factors such as climatic conditions, topography and land use practices. Soils in the area are fragile, acidic and low in native fertility and are classified based on taxonomy into entisols and inceptisols (Udoh et al. 2013). At some isolated locations some distance away from the beach mouth but far removed from the hinterland, abundant deposits of shales, limestones and gravels exist. The general characteristics of the sediments in terms of colour is white and indicates the abundance of monocrystalline quartzitic minerals. However, in some areas especially zones of terrigeous influxes, sediments are dominantly grayish, probably due to the prevalence of oxidation-reduction reactions (Stein et al. 1996; Johnston et al. 2010). Organic debris transported and deposited by tidal and flooding activities around rivers and streams, also results in the darkening of coastal sediments.

Blackening of sediments is typically the character of zircon- and monazite-enriched terrigeneous sediments. Comparatively, monazite has large concentrations of ^{232}Th and ^{238}U and lower amounts of ^{40}K (Alam et al. 1999; Mohanty et al. 2004; Abdel-Halim and Saleh 2016). The bottom sediments are characterized by sand fractions of biogenic provenance as indicated by their characteristic low organic carbon contents (Martins et al. 2012; Avinash et al. 2016). These processes including chemical composition of sediments, physical parameters such as grain size, sediment stack height and soil/sediment type, and anthropogenic activities can influence the distribution and concentration of radionuclides in coastal sediments.

Materials and methods

Sampling and sample preparation

Composite sediment samples were collected from 15 locations along the Akwa Ibom coastal area in the late dry to early rainy seasons (January–April) of 2016 using plastic spatula. This sampling period was considered optimal because loose sediments transported by surface run-off and deposited in the area might have settled by the period of sampling and thus, exist in their true ecological and textural conditions (Raghu et al. 2015). In each LGA, three samples comprising sandy and argilaceous (fine sand and clay) materials were collected. The plastic spatula used in collecting the samples was always washed and dried before use in other locations. Before sampling, the upper 1 cm of the soil/sediment dominated by materials such as dead leaves and wood fragments was removed and samples were collected within the upper 2 cm soil layer since most anthropogenic and other biogenic contaminants settle within this depth (Krishna and Govil 2007). At each sampling site, a hand-held Garmin global position system, GPSmap76 model was used in measuring geographic coordinates. The samples were stored in black polyethylene bags and later transported to the laboratory. The soils and sediments were oven dried at a temperature of 120 °C to remove moisture and later pulverized using mortar and pestle. The soil samples were seived using a 200-mm mesh in order to remove wood fragments, high angularity components like stones, pebbles and other macro-materials.

The samples were hermetically stored in labelled cylindrical containers with dimensions of about 6.5 cm (diameter) and 3.5 cm (height). Other information captured on the containers include date of acquisition, name and position coordinate of sampling site and net weight. The containers were tightly sealed and left for 4 weeks so that ^{226}Ra and ^{228}Ra and their short-lived daughters can attain secular radioactive equilibrium. Under this condition of secular equilibrium, the activity concentration of ^{226}Ra corresponds to the activity concentration of ^{238}U (Bochiolo et al. 2012). All measurements were performed using a p-type coaxial hyper pure germanium gamma ray detector available at the National Institute of Radiation Protection and Research, University of Ibadan, Nigeria. The measuring system has low background shield, 25% efficiency and 2 keV resolution at 1332 keV gamma line of ^{60}Co. The energy efficiency of the detector was determined by first counting and analysing the activities of various multi-gamma radiation emitting standard sources (e.g., ^{241}Am, ^{109}Cd, ^{60}Co, ^{139}Ce, ^{88}Y and potassium chloride standard solution) with energies between 60 and 1836 keV in equivalent geometry. The spectrum of the gamma rays was recorded using a PC-based 8192 channel analyser and processed using Genie-2000 software (Canberra, USA). Self absorption and coincidence summing corrections were performed using the Laboratory Sourceless Calibration System (LabSOCS) incorporated into the Genie-2000 software. The Geometry Composer tool in the software was used during sample geometry modelling and efficiency calibration file generation. The efficiency for the specified energies were estimated by LabSOCS based on Monte Carlo N-Particle modelling code after physical parameters such as sample container, absorber matrix and specific source-to-detector distance have been specified.

The samples were placed in the detector and counted independently for a minimum period of 1 day (86,400 s). The activities of primordial radionuclides were determined from the gamma ray 609 keV line from ^{214}Bi peak, while ^{232}Th was computed from 911 keV gamma radiation from ^{228}Ac. The activity of ^{40}K was evaluated from the peak energy of 1461 keV from ^{40}K. The specific activities of the various radionuclides in each sample were obtained from the net counts at peak emissions by removing background counts and correcting for photopeak efficiency, gamma intensity of the radionuclide and weight of the sample (Kannan et al. 2002). The activity concentration of the radionuclides in each sample, A (Bq/kg), was obtained from Eq. 1

$$A\ (\mathrm{Bq/kg}) = \frac{\frac{N_s}{t_s} - \frac{N_b}{t_s}}{P_\gamma \times \varepsilon(E) \times K_{\mathrm{sc}} \times K_{\mathrm{sa}} \times K_{\mathrm{dc}} \times M_s} \quad (1)$$

where $\frac{N_s}{t_s}$ and $\frac{N_b}{t_s}$ are counts per second of radionuclide in the sample and radionuclide in the background respectively, P_γ is the number of gamma radiations per disintegration of the nuclide at energy E (i.e., the emission probability of gamma decay), t_s is the counting time in seconds, $\varepsilon(E)$ is the full energy peak efficiency, K_{sc} is the cascade summing correction, K_{sa} is the correction factor for self attenuation, K_{dc} is the decay correction factor and M_s is the mass of the sample (kg). The values of the various radiological parameters were determined by computation from the observed activities of radionuclides.

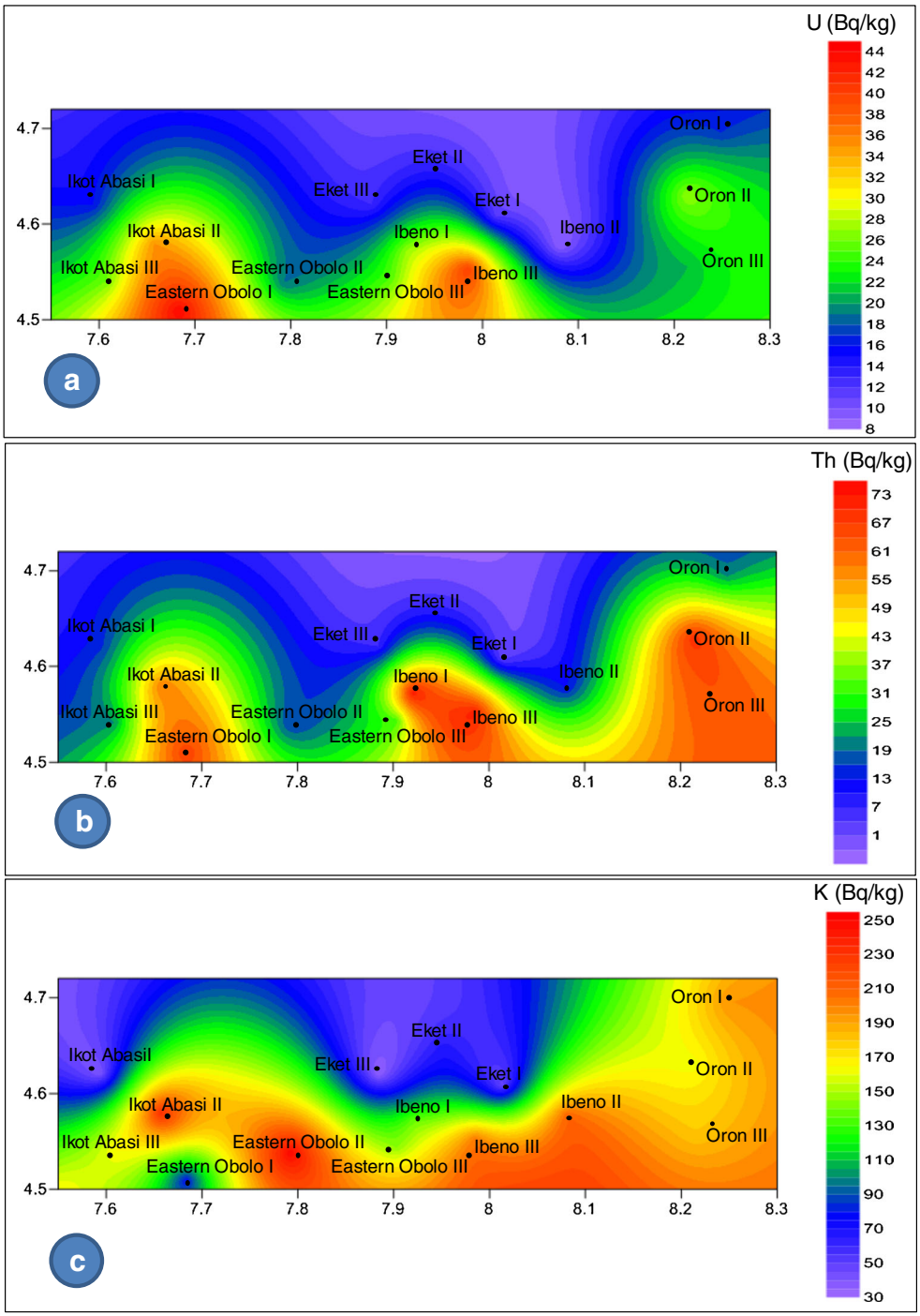

Fig. 2 Isoactivity concentration maps for uranium (**A**), thorium (**B**), and potassium (**C**) along the Akwa Ibom coastline

Results and discussion

Activity concentrations of ^{238}U, ^{232}Th and ^{40}K in beach sediments

The spatial distribution of activity levels of the primordial radionuclides (^{238}U, ^{232}Th and ^{40}K) in coastal soils and beach sediments along Akwa Ibom Coastlnie is shown in Fig. 2, while the box-whisker plot (Fig. 3) shows the basic statistical description of the radionuclides and radiologic parameters (Table 1). Activity concentrations of ^{40}K, with respective minimum and maximum concentrations of 35 ± 8 Bq/kg at Eket III and 250 ± 6 Bq/kg at Eastern Obolo II (average of 145 ± 6 Bq/kg), is the highest for all the radionuclides (Fig. 2). The range of activity concentrations for ^{232}Th is from 3 ± 1 Bq/kg at Eket I to 72 ± 9 Bk/kg at Ibeno I (average of 36 ± 2 Bq/kg). Activity concentrations of ^{238}U vary between 9 ± 1 Bq/kg (Ibeno II) and 44 ± 6 Bq/kg (Eastern Obolo I) (average of

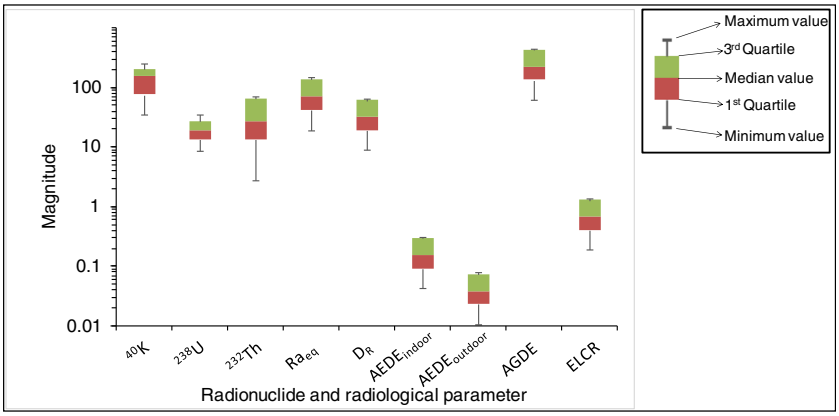

Fig. 3 A box-whisker plot showing the distribution of radionuclide and calculated radiological parameters

23 ± 3 Bq/kg) (Fig. 2). The average abundances of ^{238}U and ^{40}K are comparatively lower than world average values of 30 and 400 Bq/kg, respectively, while ^{232}Th was slightly higher than the worldwide average of 35 Bq/kg (UNSCEAR 2000). However, activity levels of ^{238}U in most clays (e.g., Ibeno III and Eastern Obolo I) are higher than the world average. Similarly, activity levels of ^{232}Th exceed the world average at Oron II, Ibeno I, and III, Eastern Obolo I and Ikot Abasi II. Generally, activity concentrations of radionuclides in the area increased southwards, towards the Atlantic Ocean (Fig. 2). Results of activity concentrations obtained from this investigation were compared with UNSCEAR (2000) and results from other countries (Table 2). These results portray the beach sediments as originating from migmatic rocks like granites, rhyolites, andesites and pegmatites, which were weathered, transported and deposited at their present location (Chabaux et al. 2003; Örgün et al. 2007). Granitic and metamorphic rocks occur extensively in the adjoining Precambrian Oban Massif and can be the provenance of these sediments (Rahaman et al. 1981; Okpara et al. 2014).

Along the coastline, radionclide distribution is not even but rather vary widely with spatial location (Fig. 2), geology and probably the extent of antropogenic activity (El Mamoney and Khater 2004; Akpan et al. 2016). The pattern of ^{40}K and ^{232}Th

Table 1 Radionuclide concentrations and basic statistics of coastal soils and beach sediments in Akwa Ibom

Location	Sample type	Longitude (°E)	Latitude (°N)	^{238}U (Bq/kg)	^{232}Th (Bq/kg)	^{40}K (Bq/kg)
Oron I	Medium sands	8.2482	4.6955	17 ± 3	21 ± 3	195 ± 3
Oron II	Clay	8.2139	4.6303	27 ± 5	69 ± 1	163 ± 3
Oron III	Fine sands	8.2143	4.5554	21 ± 2	62 ± 2	181 ± 8
Ibeno I	Fine–medium sands	7.9205	4.5723	27 ± 7	72 ± 9	137 ± 9
Ibeno II	Clayey sand	8.0811	4.5771	9 ± 1	13 ± 2	209 ± 4
Ibeno III	Clays	7.9854	4.5529	40 ± 2	71 ± 1	202 ± 7
Eket I	Fine sands	8.0132	4.6085	12 ± 2	3 ± 1	48 ± 6
Eket II	Fine sands	7.9519	4.6472	15 ± 3	12 ± 2	68 ± 5
Eket III	Fine-medium sands	7.8842	4.6206	13 ± 2	7 ± 2	35 ± 8
Eastern Obolo I	Clays	7.6836	4.5094	44 ± 6	67 ± 1	98 ± 7
Eastern Obolo II	Medium sands	7.7924	4.5384	19 ± 2	16 ± 2	250 ± 6
Eastern Obolo III	Medium sands	7.9036	4.5433	25 ± 2	33 ± 3	145 ± 5
Ikot Abasi I	Fine sands	7.5942	4.6206	16 ± 3	15 ± 2	41 ± 3
Ikot Abasi II	Clays	7.6571	4.5795	35 ± 5	58 ± 1	233 ± 5
Ikot Abasi III	Fine sands	7.6087	4.5409	26 ± 4	22 ± 2	165 ± 5
Minimum				9 ± 1	3 ± 1	35 ± 8
Maximum				44 ± 6	72 ± 9	250 ± 6
Mean				23 ± 3	36 ± 2	145 ± 6
Standard deviation				29.67	26.78	71.29
Skewness				3.16	0.31	(0.32)
Kurtosis				11.07	(1.86)	(1.19)

Table 2 Comparison of activity concentration of Akwa Ibom coastal area with results from other countries of the world

S. no	Country	^{238}U (Bq/kg)	^{232}Th (Bq/kg)	^{40}K (Bq/kg)	References
1	Oman	12–23	11–25	223–535	Zare et al. (2012)
2	Albania	8–27	13–40	266–675	Tsabaris et al. (2007)
3	Italy	42–70	31–37	410–475	Doretti et al. (1992)
4	Algeria	11–25	6–32	56–607	Benamar et al. (1997)
5	Greece	29–110	19–88	152–1593	Florou and Kriditis (1992)
6	Spain	77–6401	12–63	-	Lozano et al. (2002)
7	Egypt	4–35	3–30	112–313	El-Gamal et al. (2007)
8	United States	11–74	13–186	386–1047	Powell et al. (2007)
9	Iran	2–4	15–22	180–465	Tari et al. (2013)
10	South Africa	4–213	5–33	34–209	Newman et al. (2008)
11	Brazil	-	-	12–1029	Ribeiro et al. (2018)
12	India	2–37	2–644	301–449	Ravisankar et al. (2015)
13	Worldwide	16–110	11–64	140–850	UNSCEAR (2000)
14	Present study	9–44	3–72	35–250	

enrichments show that both radionuclides are closely related to sediment type. Around the river bank, where organic matter-enriched sediments (clays and fine sands) are dominant, relatively high ^{40}K activity concentrations were observed (Fig. 2). The increasing abundance of ^{40}K along the east and east-western portions suggests that their enrichments may be associated with anthropic activities such as hydrocarbon exploitation activities, industrial wastewater disposal, oil spillage and marine transportation. The basic statistical description of radionuclide enrichment pattern (Table 1) indicates that the standard deviations of ^{232}Th (28.78) and ^{40}K (71.29) are lower than their mean values of 36 ± 2 and 145 ± 6 Bq/kg respectively, while that of ^{238}U (29.67) was observed to be slightly higher than its mean value of 23 ± 3 Bq/kg. These observations indicate that the concentration of radionuclides in soils and sediments have high degree of uniformity in terms of ^{232}Th and ^{40}K activity concentrations and low degree of uniformity with respect to ^{238}U distribution (Gupta 2001). The low degree of uniformity could be due to clay soils, which show high mean activity concentration (2.52 ppm) of ^{238}U and Th/U ratio of ~ 7.0 (Table 3) typical of marine sediment (Klaja and Dudek 2016). Additionally, the low uniformity (i.e., measure of spatial variation of the activity concentration) of ^{238}U represented by the large value of standard deviation than the mean indicates the influence of physical and geochemical processes on the accumulation of radionuclides in the sediments. Although these sediments were sampled spatially, there exists strong affinity with regard to time of deposition. Skewness is a measure of asymmetry or lack of symmetry in the shape of a frequency distribution (Ravisankar et al. 2015). The peak of a frequency distribution can be positively or negatively skewed (Gupta 2001). ^{238}U and ^{232}Th radionuclides were positively skewed, which indicates that their distributions are asymmetric. However, ^{40}K displayed lack of symmetry due to its negative skewness. The kurtosis procedure that measures the peakedness of the curve and consequently internal sorting or distribution of the data was employed. Zero value of kurtosis represents normal curve or mesokurtic distribution. Positive values indicate more peaked normal curve or leptokurtic distribution, while negative values (i.e., less normal curve) represent platykurtic distribution (Gupta 2001). It was observed from the study that the distribution pattern of ^{238}U is leptokurtic, while ^{232}Th and ^{40}K followed a platykurtic pattern.

Table 3 Mean composition of radionuclides in Akwa Ibom coastal soils and their ratios

Soil type	Radionuclides			Radionuclide ratios		
	^{238}U (ppm)	^{232}Th (ppm)	^{40}K (%)	U/Th	U/K	Th/U
Clays	2.52	16.30	0.56	0.15	4.51	6.46
Clayey sands	0.72	3.12	0.67	0.23	1.07	4.33
Sands	1.35	6.20	0.41	0.22	3.28	4.61

Relative abundance or depletion of radionuclides in the sediments, assessed using Th/U ratios, was employed in the study to assess the content of organic matter present in the sediments. The mean value of Th/U ratio across the area is ≤ 7 indicating that the depositional environment is in a reducing condition (Klaja and Dudek 2016). This observation suggests enrichment of carbonaceous materials and clays in the area (Aswathanarayana 1985; Bodin et al. 2011). These results correlate well with U/Th and U/K ranges of sedimentary rocks as reported by Galbraith and Saunders (1983).

Radium equivalent activity

Radium equivalent activity (Ra_{eq}) was used in estimating the amount of radiation hazard in sediments from net activities of ^{238}U, ^{232}Th and ^{40}K (Beretka and Mathew 1985). Ra_{eq} was computed using Eq. 2

$$Ra_{eq}\left(\frac{Bq}{kg}\right) = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (2)$$

where A_{Ra}, A_{Th} and A_K are the net activity concentrations in Bq/kg of ^{238}U, ^{232}Th and ^{40}K respectively. Radiation risk assessed using Ra_{eq} procedure is externally attributed to gamma radiations and internally to radon and its progenies (UNSCEAR 2000). Studies have shown that ^{238}U, ^{232}Th and ^{40}K emit γ-dose at different rates even when present in the same amount in sediments (Raghu et al. 2015). It was assumed from Eq. 2 that 370 Bq/kg of ^{238}U, 259 Bq/kg of ^{232}Th and 4810 Bq/kg of ^{40}K produced γ-radiation doses at equal rate and radioactive equilibrium was uniquely attained in the ^{238}U and ^{232}Th series (Yu et al. 1992). Ra_{eq} observations in the study area (Figs. 3 and 4) range from 19 Bq/kg in Eket I to 170 Bq/kg in Ibeno I (average of 83 Bq/kg). This average is far below the permissible limit of 370 Bq/kg that is equivalent to an external dose of 1.5 mSv/year. Hence, these sediments cannot pose significant radiation hazards to the environment.

Absorbed dose rate

Absorbed gamma dose rate, D_R (nGy/h) at a distance of 1 m above the ground, is the amount of energy received from ionizing radiations absorbed per unit mass per unit time from matter (Raghu et al. 2015). The contributions of natural specific activities of ^{238}U, ^{232}Th and ^{40}K account for the total absorbed dose rate in air. There exist a direct relationship between radionuclide concentrations in beach sediments and terrestrial gamma radiation. Neglecting contributions from other radionuclides such as ^{235}U and its progenies, ^{137}Cr and

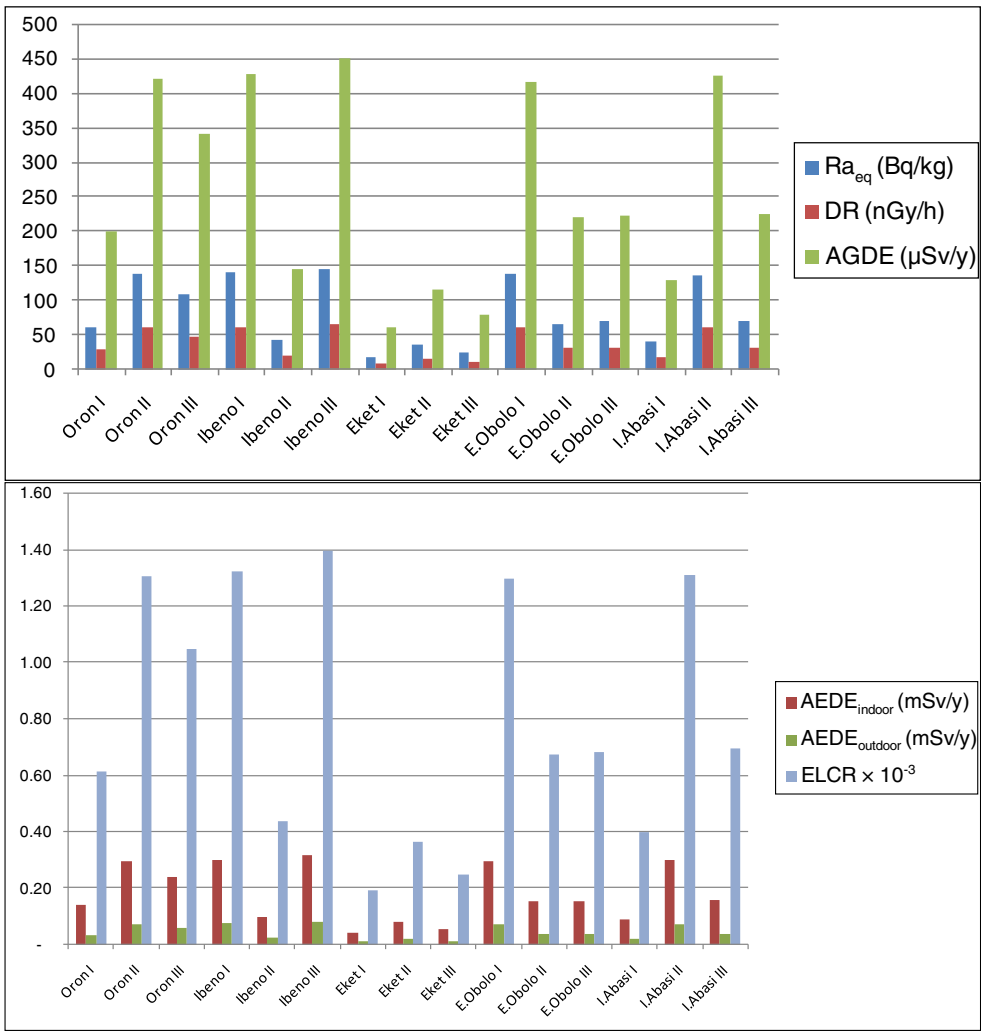

Fig. 4 Estimated radiological parameters of coastal area soils and sediments in Akwa Ibom. E. Obolo and I. Abasi are hereafter referred to as Eastern Obolo and Ikot Abasi respectively

^{90}Sr, since their total contributions are negligibly small, the outdoor air-absorbed dose rate resulting from terrestrial gamma radiations at 1 m above ground surface due to ^{238}U, ^{232}Th and ^{40}K was calculated from Eq. 3

$$D_R(\mathrm{nGy/h}) = 0.462A_{Ra} + 0.060A_{Th} + 0.042A_K \quad (3)$$

where 0.462, 0.0604 and 0.042 are dose conversion factors used in transforming the activity concentrations of ^{238}U, ^{232}Th and ^{40}K to their equivalent dose rates in nGy/h per Bq/kg (UNSCEAR 2000). D_R observations in the study area range between 10 nGy/h around Eket Beach I and 75 nGy/h in Ibeno Beach I (average of 39 nGy/h). These D_R observations are far below the safe limit, and the average value is 29% lower than the UNSCEAR (2000) stipulated safety standard value of 55 nGy/h.

Annual effective dose equivalent

The annual effective dose equivalent, AEDE (mSv/year), was computed based on the UNSCEAR (2000) standards, which stipulate that adults spend average of 20% of their time outdoor and so are exposed to air outside their living houses. The conversion coefficient from the absorbed dose in air was set at 0.7 Sv/Gy, while outdoor occupancy factor of 0.2 was used in converting dose rate to annual effective dose. The indoor and outdoor AEDE were determined from the computed values of D_R using equations 4 and 5 respectively as

$$\mathrm{AEDE}_{Indoor}(mSv/\mathrm{year}) = D_R(\mathrm{nGy/h}) \times 8760\ \mathrm{h} \times 0.8 \times 0.7(\mathrm{Sv/Gy}) \times 10^{-6} \quad (4)$$

$$\mathrm{AEDE}_{Outdoor}(mSv/\mathrm{year}) = D_R(\mathrm{nGy/h}) \times 8760 \times 0.2 \times 0.7(\mathrm{Sv/Gy}) \times 10^{-6} \quad (5)$$

where 8760 h is the total time (in hours) in 1 year. Observed minimum and maximum values of AEDE_{indoor} were 0.04 mSv/y at Eket I and 0.32 mSv/y at Ibeno III (average of 0.18 mSv/y) respectively. Corresponding values of $\mathrm{AEDE}_{outdoor}$ were 0.01 mSv/y at Eket I and 0.08 mSv/y at Ibeno I and III (average of 0.05 mSv/y) respectively. These values are lower than the 1 mSv/y benchmark value stipulated by UNSCEAR (2000). Thus, the results suggest that the environment is safe for habitation.

Annual gonadal dose equivalent

According to UNSCEAR (2010), the thyroid, lungs, bone marrow and surface, gonads and female breast constitute sensitive organs of interest in radiation studies. This is because these organs, in addition to their ability to store radionuclides, are usually the most affected parts of the body when exposed to ionizing radiations. The concept of annual gonadal dose equivalent (AGDE) was introduced by UNSCEAR for use in measuring the genetic significance of the total doses of radiation received by the reproductive organs in a year. Hence, the specific activities of ^{226}Ra, ^{232}Th and ^{40}K was used in assessing the effects of radiation on these sensitive organs of mammalian body. Thus, AGDE was estimated using Eq. 6

$$\mathrm{AGDE}(\mu Sv/y) = 3.09A_{Ra} + 4.18A\mathrm{Th} + 0.314A_K \quad (6)$$

Results obtained vary from 62 μSv/y obtained at Eket 1 to 450 μSv/y at Ibeno III (average of 259 μSv/y). The average observation is less than the recommended permissible limit of 300 μSv/y for soils (Zaidi et al. 1999) suggesting that these organs are not susceptible to being exposed to ionizing radiations. However, due to the high sensitivity of these organs and their capacities to continuously store small amounts of the radionuclides, the radionuclides can accumulate and their effects can manifest strongly later in life. This study reveals that the effects of natural radiation on sensitive organs of mammals within the coastal area are negligible. Comparatively, the average AGDE observed in this study is less than the results of other researchers like the 2398 μSv/y obtained by Arafa (2004) in Eastern Desert (Egypt) and 550.5 μSv/y obtained by Kurnaz et al. (2007) in Rize (Turkey).

Excess lifetime cancer risk

The excess lifetime cancer risk, ELCR, was evaluated using Eq. 7 (Taskin et al. 2009),

$$\mathrm{ELCR} = \mathrm{AEDE} \times \mathrm{LD} \times \mathrm{RF} \quad (7)$$

where AEDE is the total annual effective dose rate in mSv/y (estimated by adding AEDE_{indoor} and $\mathrm{AEDE}_{outdoor}$), LD is the lifetime duration (70 years) and RF is the risk factor in per sievert. For random effects, the International Commission for Radiological Protection stipulates a value of 0.05 for RF (also called fatal cancer risk per sievert) for the public (Taskin et al. 2009). ELCR observations vary from 0.19×10^{-3} at Eket I to 1.39×10^{-3} at Ibeno III. Average ELCR was observed to be 0.80×10^{-3}, which is higher than the world average (0.29×10^{-3}) reported by UNSCEAR (2010) and Taskin et al. (2009). These results reveal that cancer risk increases with exposure time.

Conclusion

The concentrations of primordial radionuclides in soils and sediments of Akwa Ibom coastal area varied from 9 to 44 (average of 25.46 Bq/kg) for ^{238}U, 3 to 72 Bq/kg (average of 34.52 Bq/kg) for ^{232}Th and 35 to 250 Bq/kg (average of 145 Bq/kg) for ^{40}K. These results were below the 1000, 1000 and 4000 Bq/kg upper limits of safety for ^{238}U, ^{232}Th and ^{40}K

respectively. ^{232}Th and ^{40}K, except ^{238}U, show lower standard deviation than their mean, which indicates that soils and sediments have high degree of uniformity. Their distributions were nearly uniform; although the concentrations of ^{40}K were relatively high in areas where anthropogenic activities like marine transport, hydrocarbon exploitation are prevalent hence, suggesting the contribution of these activities to radionuclide enrichment within the area. At some locations in Ibeno and Eastern Obolo ^{238}U activity concentration levels of clays were comparatively high, compared with their proportion in other locations and were also higher than the worldwide average. Relative abundance or depletion of radionuclides in the sediments show the mean value of Th/U ratio across the area to be ≤ 7 typical of reducing condition of depositional environment, thus suggesting enrichment of carbonaceous materials and clays. Except excess lifetime cancer risk, where average value (0.80×10^{-3}) is higher than world average value of 0.29×10^{-3}, average values of radiological parameters: radium equivalent (83 Bq/kg), absorbed dose rate (39 nGy/h), indoor annual effective dose equivalent (0.18 mSv/y), outdoor annual effective dose equivalent (0.05 mSv/y) and annual gonadal dose equivalent (259 μSv/y) were below international limits. These results show that the beach sediments are safe and thus, suitable for building residential places and other civil structures.

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