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Background Radioactivity in the Sediments of Some Rivers and Streams in Akoko, Southwestern, Nigeria and Their Radiological Effects

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Abstract: The background radioactivity in sediment samples collected mainly from the edge of streams and rivers during dry season in Akoko in Ondo state southwestern Nigeria have been determined using a gamma spectrometry technique in this study. The activity concentration measured in various sediment samples was found to vary from 142.57±5.14 to 839.28±20.45 Bq kg^{-1} for ^{40}K from 9.40±0.41 to 52.71±0.97 Bq kg^{-1} for ^{226}Ra and from 24.87±1.29 to 301.14±3.60 Bq kg^{-1} for ^{232}Th. No artificial radionuclide was detected in the samples. The radiological effect of the natural radioactivity in the sediment samples was assessed using the radium equivalent activity and the absorbed dose terms. The calculated mean values of radium equivalent activity and absorbed dose rate in air obtained are 140.54 Bq kg^{-1} and 68.42 nGy h^{-1} respectively and the effective dose posed to humans when the sediments are exposed was obtained to be 0.08 mSv y^{-1}.

Key words: Radioactivity, sediments, gamma spectrometry, radium equivalent, absorbed dose, rivers

INTRODUCTION

Radioactivity occurs naturally in both the sea water and in the ocean sediment. Radioactivity in the sea water is fairly uniform geographically and is dominated by the naturally occurring isotope ^{40}K (potassium-40) (Nagaya and Saiki, 1967; McDonald, 1992). Unlike sea water, sediment radiation levels vary with sediment type and location. The major source of natural radionuclides in sediment results from weathering and recycling of terrestrial minerals and rocks that give rise to ^{40}k, ^{232}Th, ^{235}U and ^{238}U. The later three decay naturally to produce other important radioactive isotopes of elements; including radium, radon, polonium and lead (Topcuoglu *et al*., 2001; Qureshi, 2002; Mantazul *et al*., 1999; Ajayi, 2000).

Higher quantities of radioactive materials can be introduced into the environment in several ways. These include, for example, atmospheric or underground nuclear weapon testing, accidents at nuclear installations (such as the Chernobyl accident of 1886) and related global fallout, radioactivity transfer from other areas by seawater movements and through biota, coastal and inland nuclear power plants, nuclear submarines and radioactive waste dumping operations . Most of the radioactivity deposited on surface soils is washed by rains and drained through rivers to the oceans. Part of the ground deposited activity is adsorbed in the soils and percolates with the underground waters to the oceans (Akram *et al*., 2006).

Radionuclides reaching the ocean become part of the marine ecosystem (water, sediments, biota) and may transfer through seawater-sediment-biota interface to human beings. The spread and accumulation of radioactive substances-water and sediments raise many problems concerning the safety of biotic life, the marine food chain and ultimately consumers of the seafood (Pearson and Frangipane, 1975; IAEA, 1989). Exposure to ionizing radiation is generally regarded undesirable at all levels although no harmful effects are confirmed to result from low level exposures (CBEIR, 1980).

The study of radioactivity in rocks and sediments in various locations around the world had been published by many researchers (Ajayi, 2000; Ajayi and Kuforiji, 2001) and the results obtained had been used to estimate the level of natural background radiation to which humans are exposed to in such locations (IAEA, 1989).

It is therefore, essential that the radioactivity concentration in river and stream sediments often exposed during low tide conditions in an environment is assessed for their radiological implications on the fishermen and members of the public going to the rivers to fetch water. The focus of this work is to quantify the distribution of radionuclides in the sediments in the shallow streams of the study area and to assess the radiological impacts on human health in the region.

MATERIALS AND METHODS

Study area: The study area (Akoko land) falls under the northern part of Ondo state in the southwestern part of

the geopolitical zones of Nigeria. It lies between longitudes 5°30' and 6°50' east of greenwich meridian and latitudes 7°20' and 7°45' north of the equator as shown in Fig. 1.

It lies also in the heart of the Precambrian crystalline basement rock units with indications of metamorphism and Pan African mobile belt between the West African and Congo Craton (Rahaman, 1976, 1988; Oshin and Rahaman, 1986). The area is underlain by basement migmatites, gneisses, schists, quarzites, granites, basic and ultrabasic rocks which form rugged hills and rolling plains with the rocks assuming batholithic dimensions and forming impressive outcrops which tower few hundreds of metres above the surrounding lowlands.

The drainage pattern is trellis, dendritic and radial River Ose is the major river that drains the southeast of the area. Also, smaller tributaries and seasonal streams drains the entire region. The climate is tropical, with 2 sharp seasons: the dry season and the wet season. The wet season commences in April and ends in early October while the dry season which is really hot and dry, occurs between late October and early April. The average amount of rainfall lies between 1480 and 2500 mm, the relative humidity also is 60-85% in a year while the temperature ranges between 28 and 32°C.

Fourteen sediment samples each weighing about 1kg were collected from the fourteen main and ecologically significant rivers and streams from the Akoko region to check their radionuclide composition. The sediment samples were collected mainly from the edge of the streams and rivers during dry season in 2006. During the season, the sediments are were exposed because the streams were almost dried up. Each sample was oven dried at a temperature of 105°C until a constant weight was attained, indicating the lowest water conten (Bellia *et al*., 1998). They were then pulverized, weighed and packed each into an empty radon impermeable cylindrical plastic container of uniform size, which is comparable to that of the detector. They were sealed for about 24 days to allow Ra-226(^{238}U) and ^{228}Ac(^{232}Th) and their respective progenies to reach secular equilibrium (Ajayi *et al*., 2007). The samples were later subjected to gamma spectroscopy to determine the activities of their constituent radionuclides.

Experimental set up: Gamma spectrometry measurements on the samples were carried out with coaxial-type Germanium detectors (Canberra Industries Inc.) of 50% relative efficiency and having a resolution of 2.4 kev at 1.33 Mev. The system was set up to cover about 2 Mev. Photon energy ranges over 4 k channels. The detectors are properly shielded in lead castles (Ajayi *et al*., 2007).

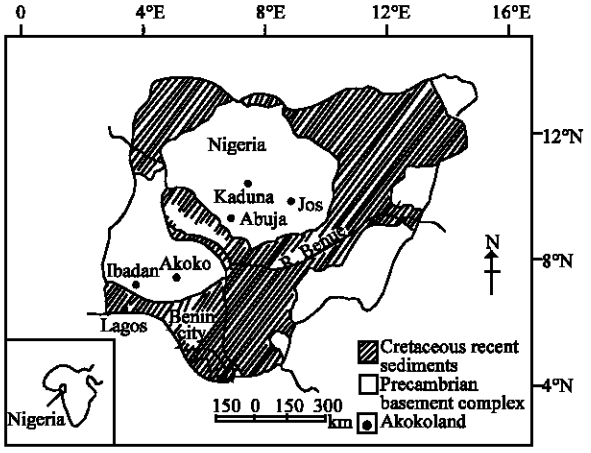

Fig. 1: Map of Nigeria showing Akokoland-the study area

Calibration of the measuring systems had been carried out using certified reference standard for various radionuclides. Spectra analyses were performed with the Genie 2K spectrometry software, version 2.1 (Canberra Industries Inc.). A library of radionulides, which contained the energy of the characteristic gamma peaks for each of nuclides analyzed and their corresponding emission probabilities was built from the data sullied in the software. The characteristics gamma peak energies of the various nuclides were used for their identification. The samples were counted for 24 h each to achieve minimum counting error. Specific activities of the radionuclides were expressed in Bqkg^{-1} of dry mass of the samples and corrected for the time elapsed since the sample collection in the sampling site.

Calculation of radiological effects from the natural radioactivity in the sediments: The radiological or health effect of the natural radioactivity in the sediment samples was assessed using the radium equivalent activity (Ra_{eq}) term. Ra_{eq} is a common index used to estimate the activity concentration of the three radionuclides-^{238}U (in equilibrium with its daughter product, ^{226}Ra , ^{232}Th (in equilibrium with its daughter product, ^{228}Ac) and ^{40}K . The term takes into account the radiation hazards associated with the radionuclides in terms of the external gamma dose and internal dose due to radon. Radium equivalent (Ra_{eq}) is calculated by using the following relation (Beretka and Mathew, 1985; Venturini and Nisti, 1997).

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

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

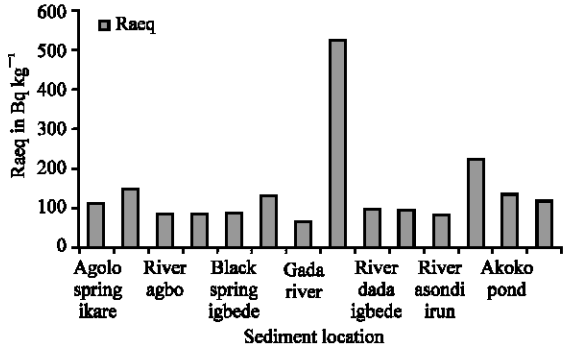

Fig. 2: The variation of the radium equivalent generated by the natural radioactivity in the sediments

absorbed dose rate, D, due to the mean activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K was calculated, in nGy h^{-1} using the formula (Tahir *et al.*, 2005; UNSCEAR, 2000; IAEA, 1991).

$$D = 0.427A_{Ra} + 0.662A_{Th} + 0.0432A_{K}$$

where, A_{Ra}, A_{Th} and A_{K} are the mean activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively. It is noteworthy that this equation is derived to calculate the absorbed dose rates in air for a height of 1 m above the ground surface. The variation of Ra_{eq} in the sediments due to their natural radioactivity is depicted in Fig. 2.

The annual outdoor dose equivalent to humans from the measured natural radioactivity when the sediments are exposed during the season is calculated using the relation below taking into account the conversion factor of 0.7 Sv Gy^{-1} and an outdoor occupancy factor of 0.2 (IAEA, 1993, 1996):

$$H = D\ (nGy\backslash h) \times 8760\ (h) \times 0.2 \times 0.7\ (Sv\backslash Gy)$$

where, H is the effective dose equivalent in mSv and D is the total absorbed dose rate in air in (nGy h^{-1}).

RESULTS AND DISCUSSION

The results of analysis of activity concentrations of ^{226}Ra, ^{228}Ra and ^{40}K radionuclides in sediment samples for the different stream of the study area are presented in Table 1. Activity is reported in Bq kg^{-1} on the basis of the sediment's dry weight. The overall measured activity concentrations range from from 142.57±5.14 to 839.28±20.45 Bq kg^{-1} for ^{40}K from 9.40±0.41 to 52.71±0.97 Bq kg^{-1} for ^{226}Ra and from 24.87±1.29 to 301.14±3.60 Bq kg^{-1} for ^{232}Th. The mean activity concentrations of ^{226}Ra, ^{228}Ac and ^{40}K radionuclides are 17.42±0.17, 57.67±1.59 and 527.86±3.86 Bq kg^{-1}, respectively. The calculated mean values of radium equivalent activity was 140.54 Bq kg^{-1}, the radiological parameters of absorbed dose rate in air and effective dose generated by the natural radioactivity of the sediments were calculated to be 68.42 nGy h^{-1} and 0.08 mSv y^{-1}, respectively. No artificial radionuclides of any radiological interest were detected in any of the sediments. A graphical presentation for the mean activity levels of the three natural radionuclides found in the sediments is shown in Fig. 3.

The levels of activity concentrations determined for Akoko river sediments have been compared with the results quoted in the literature for the marine coastal sediments for other regions of the world. The results of the sediment samples from off the Sindh coast has been found to vary from 15.93±5.22 to 30.53±4.70 Bq kg^{-1} for ^{226}Ra, from 11.72±1.22 to 33.94±1.86 Bq kg^{-1} for ^{228}Ra and from 295.22±32.83-748.47±28.75 Bq kg^{-1} for ^{40}K (Akram *et al.*, 2006). Cox's Bazar, Sea Beach, Bay of Bengal, are ^{226}Ra (13.88–25.17 Bq kg^{-1}) and ^{40}K (382.92–651.89 Bq kg^{-1}) (Akram *et al.*, 1997; Abbady, 2004).

The value of ^{226}Ra for Pacific Ocean sediments is 21.4 Bq kg^{-1} (IAEA, 1991). The activity levels for Irish Sea sediments are ^{226}Ra (23.90 Bq kg^{-1}), ^{228}Ra (36.70 Bq kg^{-1}) and ^{40}K (560.0 Bq kg^{-1}) (IAEA, 1993). The activity values for ^{226}Ra, ^{228}Ra and ^{40}K for Indian Ocean sediments are 13.80, 26.70 and 297.0 Bq kg^{-1}, respectively (Abbady, 2004). The range of ^{40}K is 118.28-608.21 Bq kg^{-1} for the coast of Chittagong, Bay of Bengal (Ajayi and Ajayi, 1999). The levels of natural radionuclides in sediments from the Turkish coast, Black Sea, are ^{232}Th (17-37 Bq kg^{-1}) and ^{40}K (301-833 Bq kg^{-1}) (Topcuoglu *et al.*, 2001).

Comparison of the results obtained with those of above indicates that the mean activity concentrations of ^{226}Ra in Akoko sediments were higher than those of the sediments of the and Sindh coast, Cox's Bazar, Sea beach. But the mean activity concentration of the radionuclide was lower than the values for Pacific ocean and the Irish sea. The Indian Ocean has ^{226}Ra concentration less than the value for Akoko stream sediments. It should however be noted that the sediments of Akoko streams are precipitated by fresh water while those from the seas and oceans are from salt water. This may account for the slight differences in their natural radioactivity contents.

The annual effective dose for Ra_{eq} of 370 Bq kg^{-1} corresponds to the dose limit of 1.0 mSv for the general population (Ajayi and Ajayi, 1999). Earth surface materials for which radium equivalent activity concentration

Table 1: Activity concentration of ^{226}Ra, ^{228}Ra and ^{40}K in sediment samples, radium equivalent activities (Ra_{eq}) and total air absorbed gamma radiation dose rates (D) at 1.0 m above the surface for the Akoko river sediments

Sediment number	Sediment name	Activity concentration (Bq kg^{-1})			Ra_{eq} (Bq kg^{-1})	D (nGy h^{-1})
		^{40}K	^{226}Ra	^{228}Ac	^{238}U	^{232}Th
1	Agolo spring Ikare	698.60±15.90	15.34±0.58	30.23±1.17	112.36	56.74
2	Arinko stream	839.28±20.45	16.73±0.82	45.89±1.87	146.98	73.78
3	River Agbo	383.28±9.24	9.40±0.41	31.98±1.15	84.64	41.74
4	Egbe dam	556.44±14.63	7.64±0.80	24.87±1.29	86.05	43.76
5	Black spring Igbede	142.57±5.14	16.58±0.60	40.41±1.47	85.34	39.99
6	Awara dam Ikare	937.21±19.09	12.53±0.59	31.04±1.29	129.08	66.38
7	Gada river Ikare	177.94±6.07	18.32±0.49	22.38±0.86	64.02	30.33
8	River Afelumo	495.12±10.50	52.71±0.97	301.14±3.60	521.46	243.25
9	River Dada	534.38±15.99	11.96±0.54	27.95±1.07	93.08	46.69
10	Imeran stream, Ikare	562.92±15.64	10.27±0.39	25.46±0.78	90.02	45.56
11	River Asondi, Irun	394.44±12.44	12.42±0.51	28.44±1.04	83.46	41.17
12	Old gada river, Erusu	550.61±15.20	24.14±0.71	109.51±2.09	223.14	106.58
13	Akoko pond	602.46±18.14	19.28±0.73	48.09±1.39	134.43	66.09
14	River Oyinmo, Ikare	514.85±14.27	16.56±0.50	40.01±1.00	113.42	55.79
Mean		527.86±12.79	17.42±0.68	57.67±2.19	140.54	68.42

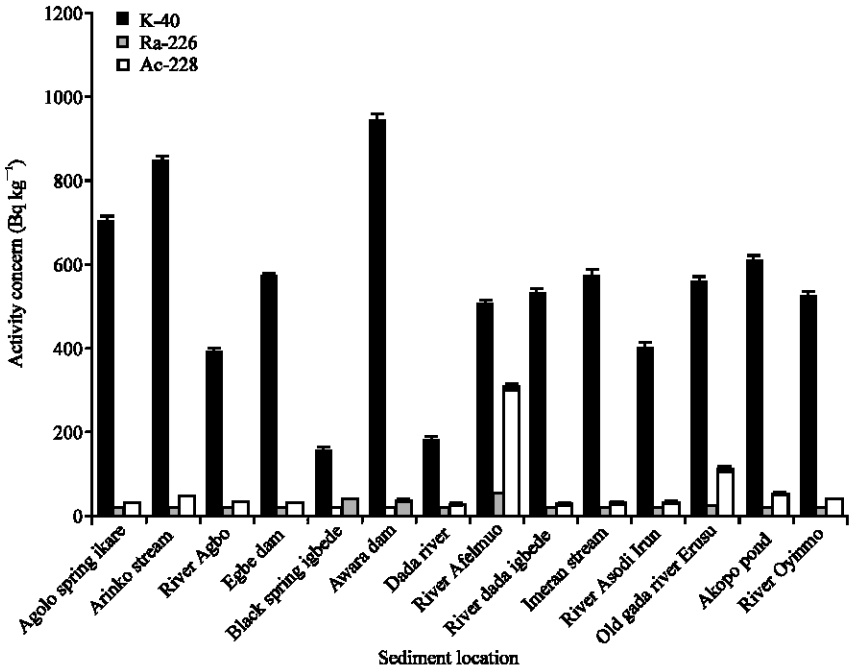

Fig. 3: The variation of the concentration of the three radionuclides in the sediments

exceeds 370 Bq kg^{-1} may pose radiation hazards. The Ra_{eq} activity for Akoko sediments of the Akoko streams is in the range 64.02-521.46 BqKg^{-1} with an overall mean value of 140.54 Bq kg^{-1}. The measured mean value of Ra_{eq} was found to be much below the value of 370 Bq kg^{-1}.

The absorbed dose rates determined in air at 1.0 m above the ground surface as a result of gamma radiation of the detected natural radionuclides in the sediments for various streams and rivers are given in Table 2. The results show that the dose rate varied from 30.33-243.25 nGy h^{-1} with an overall mean value of 68 nGy h^{-1}. The mean values of the air absorbed dose rates are in the normal range (Jibiri and Farai, 1998; UNSCEAR, 1977).

As the values of absorbed dose are calculated for an air medium, these values will be even lower because of the radiation attenuation from overlying water columns.. It should be noted that in the present study, the sediment samples were are collected mainly from the edge of the streams and rivers at extreme dry season conditions.

During dry season conditions, the sediments are often exposed because the rivers and streams dry up. The range of annual effective doses to humans resulting from the activity concentration of natural radionuclides in

sediments during exposure is 0.04-0.08 mSv y^{-1}, with an overall mean value of 0.06 mSv y^{+} . These values are less than the average external gamma dose of 0.48 mSv y^{-1} from natural radiation sources of terrestrial origin (UNSCEAR, 1988). The effective dose is also below the 0.41 mSv y^{-1} estimated as the world average by UNSCEAR implying that the sediments are free from radioactive contamination.

Even if not exposed, these sediments in submerged conditions are within the reach of a fair population of bathing chidren and fishermen, who spend a considerable time in this region for purposes like recreation activities, bathing and fishing, etc. The radiation effect on the exposed population will however be reduced according to the inverse square law of radiation transmittance since the water above the sediments will act as protective layer over the natural radioactivity generated by the river sediments.

CONCLUSION

It is concluded from this study that the mean activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in sediment samples from different shallow rivers of Akoko area are 17.42±0.17, 57.67±1.59 and 527.86±3.86 Bq kg^{-1} respectively and are comparable to the results for other coasts of the world. The calculated radiological parameters of radium equivalent activity and the absorbed dose terms resulting from the natural radioactivity in the sediment samples showed that the mean value of radium equivalent activity was 140.54 Bq kg^{-1} while absorbed dose rate obtained was 68.42 nGy h^{-1}.

During dry season conditions when the sediments are often exposed the range of annual effective doses to humans resulting from the activity concentration of natural radionuclides in sediments during the exposure is 0.04-0.08 mSv y^{-1}, with an overall mean value of 0.06 mSv y^{-1}. These values are less than the average external gamma dose of 0.48 mSv y^{-1} from natural radiation sources of terrestrial origin. The effective dose is also below the 0.41 mSv y^{-1} estimated as the world average by UNSCEAR implying that the sediments are free from radioactive contamination.

It is therefore, concluded that the activity concentration of the natural radionuclides present in the sediment are not high enough to contaminate the river water or the fishes and so no harmful radiation effects are posed to the public going to the river side for recreation or the fishermen involved in their activities in the area as a result of the radioactivity of the river sediments.

The data presented in this study will serve as baseline information on radionuclide concentration in shallow river sediments of the study area as no data is presently available to the knowledge of the authors. The data will also be useful for detecting any pollution inventories from unusual radiological events in the territorial waters of the study area.

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