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Effects of Seasonal Change on the Levels of Geogenic Radionuclides in Sand and Rocks from Tyaa River deposit in Kitui County

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ABSTRACT

A total of 56 samples of raw building materials (26 rocks and 30 sand samples) each weighing 500g were randomly sampled along the riverine for the alternate rainy and dry seasons. Gamma-ray spectrometric analysis of rocks sampled during the rainy season reports an average activity concentration of ^{226}Ra, ^{232}Th and ^{40}K of 22±1.1 BqKg^{-1}, 46±2.3 BqKg$^{-1,}$ and 659±33 BqKg^{-1} respectively, while the sand collected during the same season revealed a mean activity of 27±1.4 BqKg^{-1}, 49±2.5 BqKg^{-1}and 824±41 BqKg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K respectively. A repeat study during dry season reported higher activities for ^{226}Ra, ^{232}Th, and ^{40}K for most samples. The corresponding dose rates and radiological indices estimated from specific activities of ^{226}Ra, ^{232}Th and ^{40}K showed an upward trend as seasons changed from rainy to dry. However, both seasons reported radiation doses below the permissible limits

Keywords: Construction sand, Natural radioactivity, gamma spectrometer, Dry season, Tyaa River

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INTRODUCTION

The omnipresence of ionizing radiations from natural decay cascades has led to constant exposure of the entire biota on earth. The intensity of energy from natural radiation sources reaching the living organisms on the crust is dependent primarily on the geographical condition and geology of the parent rock. The nature of the origin of the underlying stratum determines the minerals present in soils and rocks we interact with (Varley & Flowers, 1998). The spread of the natural radionuclides like ^{238}U, ^{232}Th, and ^{40}K in the earth's crust may be influenced by weathering, hydrology which affects leaching, biochemical and chemical interactions (Karakelle et al., 2002). The Earth's crust is made up of three classes of rocks namely sedimentary, metamorphic and igneous rock

which have been formed through different mechanisms, which is inferred for the differences in mineralogical concentrations in each type (Tzortzis, Svoukis, & Tsertos, 2004). Documentations from radiometric surveys reflect consistent trends of elevated levels of ^{40}K, ^{238}U, and ^{232}Th in rocks of igneous origin (UNSCEAR, 2000).

Radiations from primordial sources contribute to 87% of the human population exposure (UNSCEAR, 2000). The major pathways through which radionuclides or their deleterious decay products gain entry to biological systems include; inhalation of radon gas or dust particles with radionuclides attached on them, or through food chains. During mineral uptake by the plant's roots, the palatable products of crops may harbor radionuclides notably ^{40}K (IAEA. & IAEA., 2010). Therefore, human consumption of food grown on regions with potentially elevated levels of background radionuclides may facilitate radionuclides assess to human biological systems (Malanca, Gaidolfi, Pessina, & Dallara, 1996).

Owing to the long half-life of ^{238}U, ^{232}Th, and ^{40}K, some habitats at present have sufficient levels of gamma-emitting radionuclides to be associated with the clinical effects. The variability in levels of natural radionuclides besides regional geology depends on soil or rock porosity and solubility of the latter radionuclide which facilitates lateral or horizontal mobility of radionuclides through the soil profile due to leaching or by redistribution of sediments bearing the radionuclides through surface runoff causing their diversification or clustering changing their distribution pattern. As such a concentration level of radionuclides may change each time the hydration levels of rock and soils changes. The solubility of ^{238}U facilitates its mobility through the soil profile as it forms complex ions and migrates to lower soil horizons(Cowart & Burnett, 1994).

The main objective of this research was to determine the seasonal fluctuations on the level of terrestrial background radiation due to changes in hydration levels of soils and rocks from a seasonal river in Kitui County. The sand and rock samples were collected from the same points for two consecutive rainy and dry seasons. The samples were prepared and analyzed using Lead shielded HPGe detector with necessary electronics coupled with relevant software. Significant fluctuations in radioactivity levels of rocks and sand within the surveyed riverine length will spur more researches upstream and in the surrounding areas. This is because sand in the river-bed has weathered from different backgrounds, mixed thoroughly during transportation to and along the river course which is the lowest point. The findings from this work will be useful for tracking future changes in the level of background radiations, which is important for human and environmental protection.

EXPERIMENTAL TECHNIQUES

GEOLOGY OF THE STUDY AREA

The surveyed length of the Tyaa River (2 km) crosses the Thika-Garissa Highway Bridge situated approximately 800 m from Mwingi town which is 97.6 km from Kitui County capital. Mwingi town is globally located 0º56' S, 38º 03' E and the altitude of the sampled length lies between 203 m -1378 m above sea level. Kitui County lies within the Central Eastern part of the Mozambique Belt Segment (Nyamai, 2003). Geological studies from the region report the presence of the three classes of rocks with an abundance of metamorphic rocks within the riverine and surrounding grounds. Metamorphosed granites rocks are closely associated with Quartz-monzonites which are usually scratch-resistant and due to their hardness, they are usually preferred for internal cladding and flooring in building sectors (Tzortzis et al., 2004). The seasonal river studied deposits large volumes of sand along the studied length due to the meandering river course that slows the speed of river facilitating the sand deposition. During the dry season, local and non-local residents heavily mine building materials from this river for building purposes.

SAMPLE COLLECTION AND PROCESSING

The samples were collected during consecutive rainy and dry seasons. A total of 40 geological samples (20 rocks and 20 sand samples) each weighing 0.5 kg were randomly collected during the rainy season and the GPS locations of the sampling points recorded.

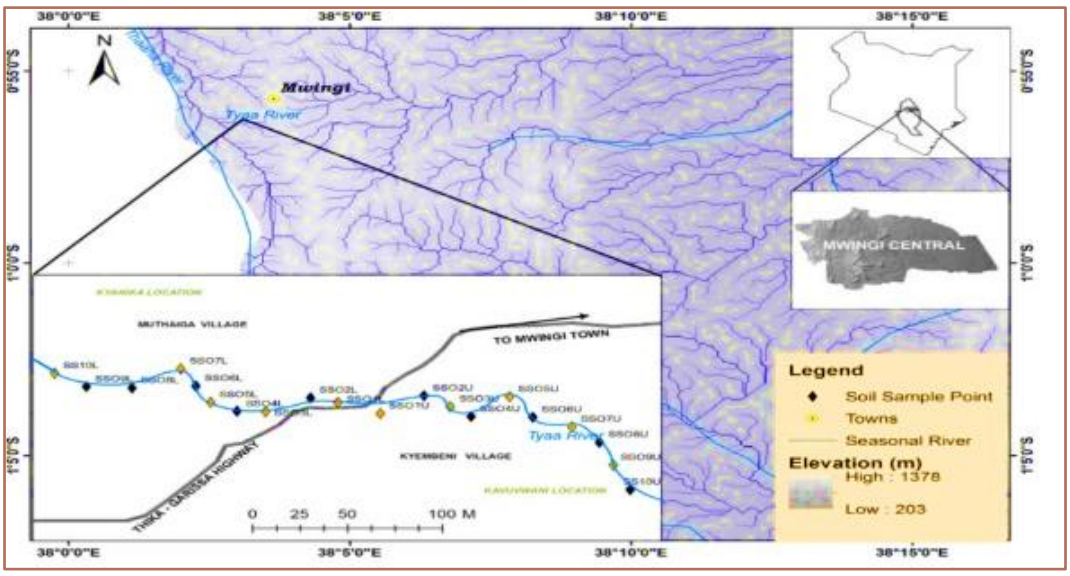

Fig 1 Map of the sampled river profile

Table 1 GPS coordinates for the sampled locations for the rock and sand samples during the consecutive rainy and dry season

Sample ID	Upper stream		Lower stream	
S01	0° 56' 17.3 S	38° 02' 36.3 E	0° 56' 21.5 S	38° 02' 34.8 E
S02	0° 56' 12.2 S	38° 02' 35.6 E	0° 56' 21.1 S	38° 02' 32.6 E
S03	0° 56' 21.0 S	38° 02' 40.9 E	0° 56' 45.3S	38° 02' 28.8 E
S04	0° 56' 22.5 S	38° 02' 43.9 E	0° 56' 19.5S	38° 02' 31.6 E
S05	0° 56' 21.6 S	38° 02' 41.7 E	0° 56' 24.7S	38° 02' 32.0 E
S06	0° 56' 20.6 S	38° 02' 43.3 E	0° 56' 21.1S	38° 02' 35.1 E
S07	0° 56' 21.0 S	38° 02' 42.8 E	0° 56' 19.5S	38° 02' 34.3 E
S08	0° 56' 21.8 S	38° 02' 45.5 E	0° 56' 27.0S	38° 02' 37.5 E
S09	0° 56' 22.5 S	38° 02' 45.7 E	0° 56' 27.1S	38° 02' 37.5 E
S10	0° 56' 24.6 S	38° 02' 47.1 E	0° 56' 19.8S	38° 02' 30.6 E

Table 1 shows the GPS coordinates for the sampled points while figure 1 is a map showing the sampled profile of the river. The same points were re-sampled to the iso-levels for a total of 16 samples during the dry season. This was meant to hold constant factors which might affect the levels of natural radioactivity in surveyed locations. Each raw sample was provisionally packed into a plastic bottle well tagged with sample ID, date of collection and GPS of sampled point. The plastic bottles were all sealed to avoid cross-contamination. The sand samples were air dried independently and sieved to micro-scale.

Dry rock samples were ground each at the time and a mass of 250g of each sample transferred into a clean standard plastic container of capacity 300 ml with the same geometry as those holding the various standard reference materials supplied by IAEA 2007 used in this work. The latter was stored for at least one month to allow ^{232}Th and ^{226}Ra assume secular equilibrium with their short-lived daughter progenies (Ahmed et al., 2014).

CALIBRATION AND DATA ACQUISITION

A gamma-ray spectrometer using an HPGe detector was used to determine the radioactive species of interest and their corresponding abundance. To ensure resolved peaks for the significant energy peaks, the detector was maintained in liquid Nitrogen Dewer with liquid nitrogen at -196 °C. The detection efficiency of the gamma-ray spectrometer was determined regularly using equation 1. The energy calibration was obtained from a linear fit of four points obtained from a standard mixture containing ^{241}Am, ^{137}Cs, and ^{60}C. The standard sample was counted for 500 seconds for proper statistics in the same geometry as the samples under investigation.

Two geological samples were run for a Live time of 30,000 Seconds, which made it possible to run two samples per day. Spectra for the reference and geological samples were collected and stored for offline analysis. Upon the assumption that the radioisotopes of interest present and their decay daughters were at secular equilibrium, the activity concentration of ^{226}Ra was estimated from ^{214}Pb and ^{214}Bi gamma lines at 351 keV and 609 keV respectively. ^{232}Th activity concentration was estimated from the average emissions of ^{212}Pb and ^{228}Ac gamma lines at 238 keV and 911 keV respectively, and the relative abundance of ^{40}K was estimated from its single photo-peak at 1460 keV.

$$\eta = \frac{N_D}{p.A_c.m} \quad (1)$$

Where; N_D is the net source count rate, p is the gamma decay transition probability, $\boldsymbol{m}$ is the mass (Kg) of the geological sample. A_c- is the nuclide individual's activity concentration (BqKg^{-1}) for the reference sample.

RESULTS AND DISCUSSION

SPECIFIC ACTIVITY CONCENTRATION

The specific activity concentrations of ^{238}U, ^{232}Th, and ^{40}K in sand and rocks were evaluated separately using the analytical equation 2 (Ebaid, 2010) and the results showing the range and average activity of sand and rocks tabulated in table 2 & 3.

$$A_c = \frac{N_D}{p.\eta.m} \quad (2)$$

Where: A_c is the specific activity concentration for a sample in BqKg^{-1}, N_D is the net count rate energy E_γ, p is the emission probability for gamma-ray transition energy E_γ, η is the detection efficiency (photopeak) at particular gamma-ray energy E_γ, m is the sample mass in kg.

Table.2 Mean activity concentration (Bq/Kg) of ^{226}Ra, ^{232}Th and ^{40}K for the sand samples collected upper and lower Tyaa River.

Radionuclide	Rainy season	Dry season
^{226}Ra	27±1.4	30±1.5
^{232}Th	49±2.5	54±2.7
^{40}K	824±41	850±43

Generally, ^{226}Ra, ^{232}Th, and ^{40}K reported higher mean activity concentration during the dry season compared to the rainy season as presented by table 2. The activities of ^{226}Ra, ^{232}Th, and ^{40}K were higher in sand compared to rock samples. Higher activity concentration in the sand was attributed to high clay and silt contents in sand samples (Baeza et al., 1995). The activity concentration of ^{40}K in the sand within the sampled profile of the river was twice the world's mean of 400 Bq/Kg (UNSCEAR, 2000). The elevated levels of ^{226}Ra, ^{232}Th, and ^{40}K during the dry season were attributed to change in mineral accessories of sand as a result of deposition of new sand rich in ^{226}Ra, ^{232}Th, and ^{40}K minerals along the river course. This is possible since more sand was deposited along the waterway before the river dried, and after collection of samples analyzed for the rainy season. Deposition of sand minerals with high organic contents could also be a reason for high levels of radionuclides like ^{238}U reflected during the dry season (Valkovic, 2000). Figure 2 compares ^{226}Ra concentration in sand samples collected from the same points for consecutive rainy and dry seasons. The red line shows the world's average value of 33 Bq/Kg of ^{226}Ra (UNSCEAR., 2008). ^{232}Th in the sand exceeded the world's average of 30 Bq/Kg for both seasons (UNSCEAR, 2000).

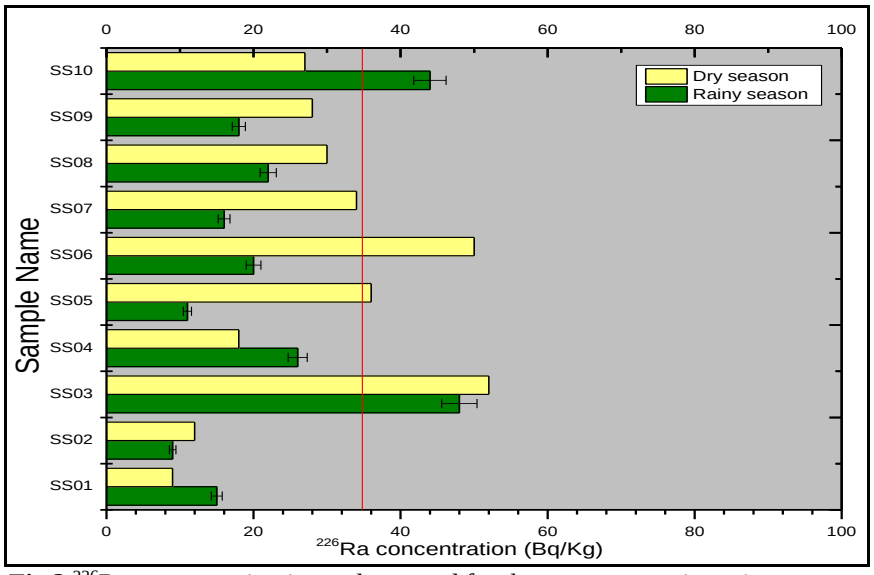

Fig.2 ^{226}Ra concentration in sand reported for the two consecutive rainy seasons

Table.3 Statistical summary for activity concentration (Bq/Kg) of ^{238}U, ^{232}Th and ^{40}K for rock samples collected upper and lower riverine

	Rainy season		Dry season	
Radioisotope	**Range**	**Average**	**Range**	**Average**
^{226}Ra	6±0.34 to 37±1.9	22±1.1	18±0.93 to 25±1.28	21±1.1
^{232}Th	14±0.71 to 119±5.9	46±2.3	30±1.51 to 98±4.93	58±2.9
^{40}K	94±4.74 to 1112±55	659±33	368±18.43 to 1389±69.46	975±49

The concentration and the spread of ^{226}Ra in rock samples were close to homogeneous for the consecutive rainy and dry seasons. A higher average activity concentration of ^{232}Th and ^{40}K in rocks from sampled river profile was reported during the dry season (table 3). High levels of ^{40}K in rocks were attributed to its high solubility which facilitates its crustal mobility(El Mamoney & Khater, 2004). The high abundance of ^{40}K was inferred to the presence of silicate minerals in the sampled rocks (Fujiyoshi & Sawamura, 2004). However, the mean values of ^{40}K, ^{238}U and ^{232}Th were far below the exemption level of 100,000 Bqkg^{-1} for ^{40}K and 1000 Bqkg^{-1} for ^{238}U and ^{232}Th (IAEA, 1996).

DOSE RATES

Evaluation of absorbed gamma dose rates D_R (nGyh^{-1}) in air 1 m above the ground was done by converting specific activity concentration to absorbed dose using the suggested conversion factors of 0.427, 0.662 and 0.043 for ^{226}Ra, ^{232}Th and ^{40}K respectively(UNSCEAR, 2000). To estimate annual individual's total risks as a result of gamma radiation exposure, the absorbed dose was modified to Annual Effective Dose AEDR (mSvy^{-1}) using guidelines and estimated Kenyan occupancy factors for indoor and outdoor exposures (Mustapha, Patel, & Rathore, 1999). Dry season reported higher levels of absorbed dose, indoor and outdoor AEDR in sand and rocks for the majority of samples compared to samples collected during the rainy season. Figure 3 shows a variation of dose rates and hazard indices for the sand reported for the rainy and dry season.

RADIOLOGICAL HAZARD INDICES

The values for radium equivalent, internal and external hazard indices varied for each sample suggesting the non-uniform crustal distribution of natural radionuclides. The average values for radium equivalent, internal and external hazard indices for the rainy and dry seasons are present in table 4.

Table 4 The mean values for radiological indices for the sand and rock samples for the rainy and dry seasons

Parameter	**Sand**		**Rocks**		**Permissible level**
	Rainy	**Dry**	**Rainy**	**Dry**	
Radium equivalent (Ra_{eq})	154±7.74	181±9.05	112±5.61	206±10.3	370 Bq/Kg (UNSCEAR, 1988)
External hazard (H_{ex})	0.42±0.02	0.49±0.02	0.3±0.02	0.55±0.02	A unit (El-Taher & Al-Zahrani, 2014)
Internal hazard (H_{in})	0.47±0.02	0.58±0.03	0.35±0.02	0.52±0.61	A unit (UNSCEAR, 2000)

The potential radiological risks of exposure from primordial radioisotopes in rocks and sand from this river when used as building materials were assessed for the two consecutive rainy and dry seasons. The levels of radium equivalent (Ra_{eq}), the external hazard index (H_{ex}) and internal hazard indices (H_{in}) were estimated using respective analytical equations from (Xinwei, Lingqing, Xiaodan, Leipeng, & Gelian, 2005) and (Higgy, El-Tahawy, Abdel-Fattah, & Al-Akababwy, 2000) and (Beretka & Matthew, 1985) and respectively. Higher values for radiological hazard indices were reported during dry the season due to the corresponding higher activities reported during the dry season.

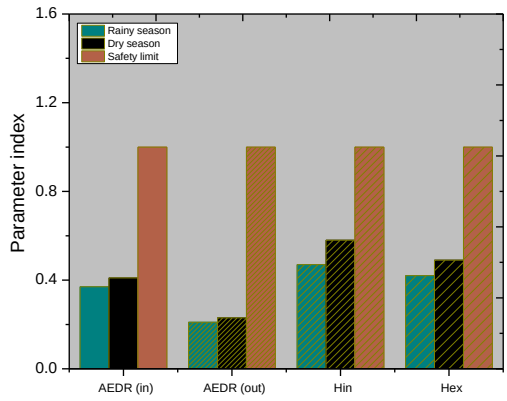

Fig.3 Annual Dose rate and radiation hazard indices for this work

The average Ra_{eq} was below a permissible limit of 370 KgBq^{-1} (ICRP., 2005), while H_{ex} and H_{in} were below a unit and thus the radiation exposure to the public due to the rocks and sand from this river was insignificant for both seasons. Indoor annual effective dose rate ($AEDR_{in}$), Outdoor annual effective dose rate ($AEDR_{out}$) and hazard indices are represented in Fig3.

CONCLUSIONS

The level of natural radioactivity has been evaluated for consecutive rainy and dry seasons. Geological samples collected during dry season reported higher radioactivity concentrations compared to samples collected during the rainy season. Higher activity in the sand during the dry season was attributed to changes in the level of the mineral content in sand due to deposition of new sand collected upstream. This suggests there could be a background upstream with elevated levels of ^{226}Ra, ^{232}Th and ^{40}K. Higher activity in most rocks samples reported during the dry season was associated with silt and mineral accessories trapped in rock grains during rainy season due to the porosity of rocks and solubility of the primordial radionuclides. Dry season reported higher doses rates compared to doses reported from samples collected during the rainy season. However, the indoor and outdoor AEDR were within the safety limits of 1mSvy-^{1} (ICRP., 2005) and generally, the sand and rocks were safe when used as building materials. Similar studies need to be repeated for several seasons to monitor the consistency in trends of radioactivity for the preceding seasons.

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