

Gamma ray spectrometric analysis and assessment of radiation hazards in soils of Mbeere North region, Kenya

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Abstract

This study used gamma ray spectrometry to determine the radiological safety of construction soil sampled randomly from Mbeere North region, Kenya. The mean activity concentration of ^{232}Th , ^{238}U , and ^{40}K was $149.7 \pm 2.8 \text{ Bqkg}^{-1}$, $88.3 \pm 2.4 \text{ Bqkg}^{-1}$, and $490 \pm 35 \text{ Bqkg}^{-1}$, respectively. These averages exceed the world average for all the radionuclides. The radionuclides were non-uniformly distributed, with higher concentrations along the slopes and on the feet of the hills. The mean absorbed dose rate, indoor and outdoor annual effective dose, radium equivalent, external hazard index, and internal hazard index were $157.9 \pm 4.4 \text{ nGh}^{-1}$, $0.58 \pm 0.02 \text{ mSvy}^{-1}$, $0.39 \pm 0.01 \text{ mSvy}^{-1}$, $340.7 \pm 9.2 \text{ Bqkg}^{-1}$, 0.92 ± 0.02 and 1.14 ± 0.03 , respectively. Among the radiation safety indicators, only the average internal hazard index exceeded slightly the acceptable safe limit. Therefore, soils of Mbeere North region are radiologically safe for use in brick making and construction of human habitats.

Introduction

Primordial radionuclides and their progenies are naturally present on the surface of the earth. The abundance of these radionuclides is increased by extrusion of earth's crust materials through volcanism and anthropogenic activities such as mining. The dominant terrestrial radionuclides that constitute most of the extruded materials are ^{238}U , ^{232}Th , and ^{40}K and their progenies [1]. High energy photons released as a result of radioactive decay of these radionuclides have the potential to ionize atoms of the body cells that they impinge. The result of such interactions may have either stochastic or deterministic effects depending on the energy of the photons, type of body cell, and duration of exposure among other factors [2]. The spatial distribution of the geogenic radionuclides largely depends on the erosion of sediments weathered from lithological units containing the radionuclides. These radionuclides together with radon contribute above 87% of the average radiation dose absorbed by human cells [3].

In central eastern Kenya, most houses are built using calcined bricks casted from surface clays, shales, or fireclays. Such bricks are a cheaper alternative to quarry blocks that are only available from far areas in upper eastern and central Kenya. Despite the heavy use of soil from this region as a construction material, limited information is available regarding its radiation safety. Besides, studies have reported high cancer prevalence in eastern Kenya, but the predisposing factors are unknown [4–7].

This study evaluated the concentration of natural radionuclides (^{238}U , ^{232}Th , and ^{40}K) and radiation hazards associated with exposure to soils of Mbeere North region, Kenya. Soils sampled mainly from brick-making sites in the region were analyzed using a lead-shielded NaI(Tl) gamma ray spectrometer. The radiation risk associated with external exposure, inhalation, or ingestion of radionuclides present in the soil was evaluated using estimates of annual effective doses (AEDs) and radiation hazard indices determined from the activity concentration of natural radionuclides in the soil.

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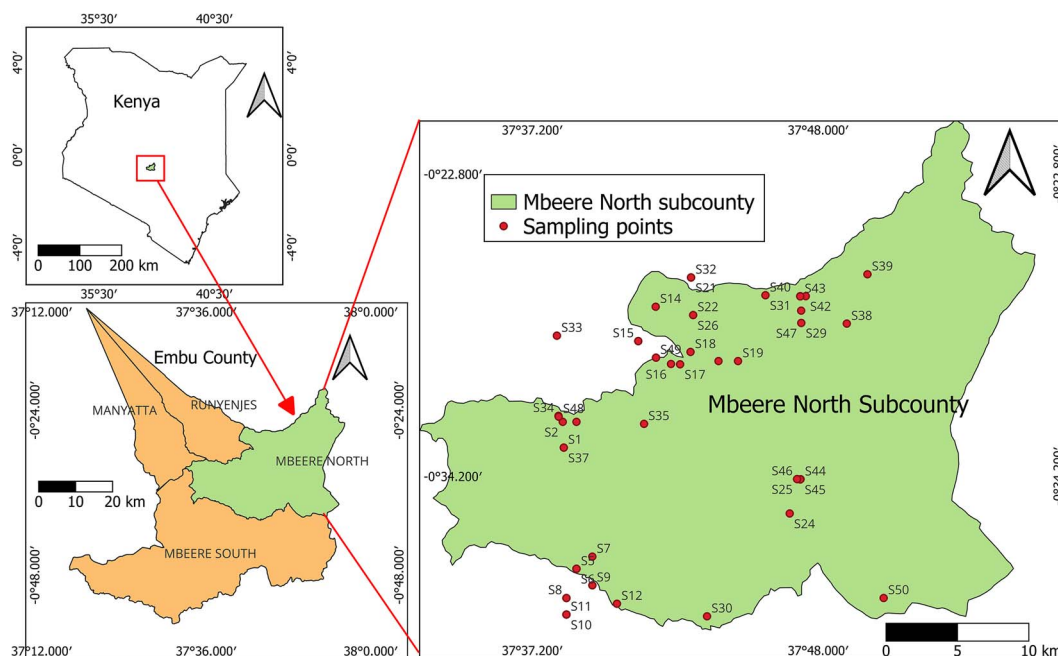


Figure 1. Map of Mbeere North region showing the sampling points.

Materials and methods

Geology of Mbeere North region, Embu County, Kenya

Test samples were collected from brick-making sites in Mbeere north region within a zone of about 777 km² bounded by latitudes 3° 15' South and 3° 62' South, and longitudes 37° 25' East and 38° 17' East. In the area, bricks are cast out of indiscriminate collection of soil from the banks of river Thiba, Tana, and Ena and on the foot of Kiang'ombe, Kirigo, and Kamarandi hills. The hills are reported to contain calcaceous, gneiss, and granite rocks as well as traces of graphite, diamonds, migmatites, pegmites, iron ore, mica, gemstone, and quartz [8, 9]. Surface soils in the area are rich in sediments from the rocks due to semi-arid weathering conditions [9]. Figure 1 shows the location and the distribution of the sampling points.

Sample collection and preparation

Fifty samples of soil were collected randomly from various brick-making sites in Mbeere North region, Kenya. From each sampled site, three 350 g samples were scooped from a depth of about 10 cm within an area of 1 m². The samples were then mixed to form a single representative sample weighing about 1 kg which was then packed in polythene bags for further processing. Extraneous materials like plant parts, pebbles, and stones were removed from the samples using a sieve or handpicking where possible. The precise location

of each sampling site was recorded using a global positioning system. The geographical distribution of the points is shown in Fig. 1.

The soil samples were pulverized and then sieved through a 1 mm mesh to yield a fine powder. Later, the soils were dried in an oven set at 110 °C for 8 h in order to expel moisture [10]. The dry samples were then packed in 250 cm³ plastic containers, weighed, and hermetically sealed. Before counting, the sealed samples were left uninterrupted for 30 days to allow the short-lived progenies to reach secular equilibrium [11].

Detector calibration and data acquisition

A NaI(Tl) spectrometer and its associated electronics were used to identify and quantify the gamma ray emitting radionuclides present in the soil samples. The detector crystal was placed at the center of a 10 mm thick lead shield, which acted as a filter of background radiations. Before counting the decay rate of radionuclides in the soil samples, the detector was calibrated using an International Atomic Energy Agency (IAEA)-certified reference sample comprising of ⁶⁰Co, ¹³⁷Cs, and ²⁴¹Am. This calibration certificate was considered appropriate because the energy range of its constituent radionuclides (59.5 keV for ²⁴¹Am to 1332 keV for ⁶⁰Co) adequately spans the range of the radionuclides of interest. Test samples were placed on the chamber of the detector crystal and counted for 28 800 s. The region of interest from the centroid of the spectra

of ^{212}Pb at 239 keV, ^{214}Pb at 351 keV, and ^{40}K at 1460 keV was used to calculate the activity concentration of ^{238}U , ^{232}Th , and ^{40}K , respectively. A background correction was performed on the count of each soil sample to yield its net count.

Results and discussions

Activity concentration of ^{238}U , ^{232}Th , and ^{40}K in soil and *Catha endullis*

The activity concentration (A_s) is the rate of decay of a unit mass of a radioactive sample. Equation (1) was used to compute the activity concentration for ^{238}U , ^{232}Th , and ^{40}K in the soil samples and the results are presented in Fig. 2.

$$A_s = \frac{N}{mt\rho\varepsilon} \quad (1)$$

where N , m , t , ρ , and ε are, respectively, the net gamma ray count of each radionuclide, mass of the soil sample, counting live-time, gamma emission probability, and the counting efficiency of the NaI(Tl) detector.

The sample S10 showed the highest concentration of ^{232}Th and ^{238}U and S25 the highest concentration of ^{40}K , while S27, reported the lowest concentration of ^{232}Th and ^{238}U and S28 reported the lowest concentration ^{40}K . Interestingly, the sites S11, S46, S42, and S43 which neighbor the areas with extreme activity concentration did not show comparable levels of radionuclide activity. Therefore, Mbeere North region has a non-uniform spatial distribution of the radionuclides tested. This could be ascribed to the abrupt geology changes observed in the area as evidenced by the rapid changes in elevation and terrain. Such changes vary the nature and distribution of lithographic units in basement systems. Sites S10, S14, S22, S33, S34, S35, S39, S45 and S46 lie along the slopes or on the feet of the hills in the area. These sites reported radionuclide concentrations much higher than the regional average. This indicated that soil along the slopes or on the feet of the hills were contaminated with radionuclides present in the sediments that were eroded from the granite and gneiss rich Kiang'ombe, Kirigo, and Kamarandi hills.

Unlike ^{40}K , the trend in activity concentration level of ^{232}Th in majority of the sites mirrors that of ^{238}U . This implies that ^{232}Th and ^{238}U , whose origin is primarily the earth's crust, have a common source. Compared against the world averages activity concentration, 96%, 96%, and 52% of the samples exceeded the world average activity concentration of 33, 45, and 420 Bq per kg for ^{232}Th , ^{238}U , and ^{40}K , respectively [12, 13].

The 50 samples had an average activity concentration of $149.7 \pm 2.8 \text{ Bqkg}^{-1}$, $88.3 \pm 2.4 \text{ Bqkg}^{-1}$,

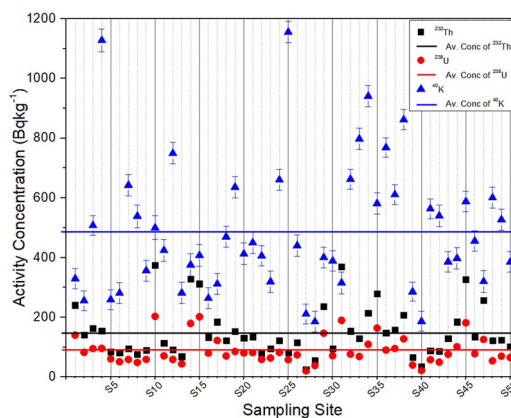


Figure 2. Activity concentration of ^{232}Th , ^{238}U , and ^{40}K in soil. The horizontal lines show the average concentration.

and $490 \pm 35 \text{ Bqkg}^{-1}$ for ^{232}Th , ^{238}U , and ^{40}K , respectively. Clearly, the average activity concentration exceeded the world average. The high concentration levels of ^{232}Th and ^{238}U are postulated to result from erosion of weathered sediments of granite and gneiss rocks that are abundant in the neighbouring Kiang'ombe, Kirigo, and Kamarandi hills. High activity concentration of ^{40}K could be a result of heavy use of phosphate fertilizers in cultivation of *Catha endullis* (khat) and other crops grown in the area. As shown in Table 1, the activity concentration determined in this study compares well with findings reported in other studies in Kenya, and especially in Embu County, Meru County, and the neighbouring Tharaka Nithi County.

Radium equivalent

Radium equivalent (Ra_{eq}) is an estimate of the amount of radiation energy accumulated by a unit mass of matter when exposed to a sample with multiple nuclides. The total specific radium activity from the multi-nuclide samples was estimated using the weighted contribution factor of 1.426 for ^{232}Th , and 0.0792 for ^{40}K according to Equation (2) [2, 20].

$$Ra_{eq} = A_{Ra} + 1.426A_{Th} + 0.0792A_K \quad (2)$$

where A represents the activity concentration, and the subscripts denotes the corresponding radionuclide. Table 2 and Fig. 3 show the results for the range and average Ra_{eq} of the soil samples.

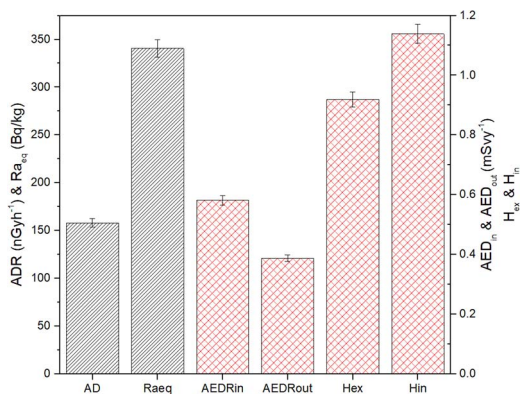
The values of Ra_{eq} varied from 73.2 ± 8.1 to 776 ± 10 . The mean radium equivalent for the 50 samples was $340.7 \pm 9.2 \text{ Bqkg}^{-1}$, which is below the recommended safe limit of 370 Bqkg^{-1} . Therefore, use of soil for construction and brick making does not pose risks of exposure to harmful radiation.

Table 1. Comparison of activities in soil in this study with similar studies.

Sampling area	^{238}U (Bqkg $^{-1}$)	^{232}Th (Bqkg $^{-1}$)	^{40}K (Bqkg $^{-1}$)	References
Mbeere North, Kenya	87 ± 13	149.7 ± 7.2	490 ± 13	This study
Igembe South, Kenya	98.7 ± 2.3	191.8 ± 2.8	267.3 ± 43.8	[14]
Kakamega, Kenya	80 ± 4	200 ± 10	968 ± 48	[15]
Tharaka Nithi, Kenya	53 ± 3	96 ± 4	1069 ± 46	[16]
Embu County, Kenya	33 ± 5	22 ± 11	344 ± 12	[17]
Pakistan	26	39	485	[18]
Imo State, Nigeria	4.15	1.64	134.13	[19]
World average	33	45	420	[13]

Table 2. Mean values and range of and radiation hazard indices and radiation dose.

Parameter	Range	Mean
Absorbed dose (nGyh $^{-1}$), Radium equivalent (Bq per kg)	(34.5 ± 3.9)—(355.9 ± 5.1)	157.9 ± 4.4
Indoor annual effective dose (mSvy $^{-1}$)	(0.13 ± 0.01)—(1.31 ± 0.02)	0.58 ± 0.02
Outdoor annual effective dose (mSvy $^{-1}$)	(0.08 ± 0.01)—(0.87 ± 0.01)	0.39 ± 0.01
External hazard index	(0.20 ± 0.02)—(2.10 ± 0.03)	0.92 ± 0.02
Internal hazard index	(0.25 ± 0.03)—(2.65 ± 0.04)	1.14 ± 0.03

Figure 3. A summary of the mean values of absorbed dose rate (ADR), indoor annual effective dose (AED_{in}), outdoor annual effective dose (AED_{out}), internal hazard index (H_{in}), and external hazard index (H_{ex}).

Absorbed dose rate

The amount of energy absorbed by a unit mass of matter impinged by the ionizing radiation from the soil samples was estimated using Equation (3) [2, 21].

$$ADR = 0.427A_U + 0.662A_{Th} + 0.043A_K \quad (3)$$

Figure 3 and Table 2 show that the absorbed dose ranged from 34.5 ± 3.9 to 355.9 ± 5.1 nGyh $^{-1}$ with a mean of 157.9 ± 4.4. About 96% of the samples

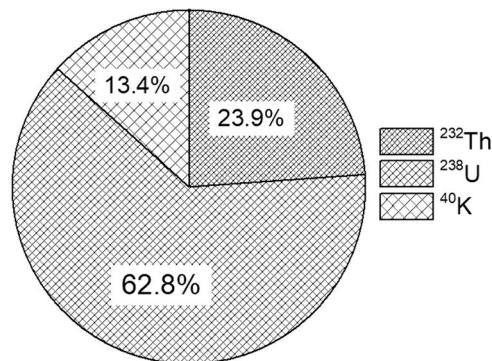


Figure 4. Percentage contribution of the radionuclides to the absorbed dose rate.

had an absorbed dose higher than the world average of 57 nGyh $^{-1}$. However, all the sampled sites had an absorbed dose of less than the safe limit of 1500 nGyh $^{-1}$ recommended by UNSCEAR, 2010 [22]. This confirms that the soils in the Mbeere North region, Kenya are radiologically safe for use in construction of human dwellings.

The percentage contribution of each of the radionuclides to the absorbed dose was computed and the results are presented in Fig. 4.

The dose contribution value of 62.76% indicates that the radiation effect of ^{232}Th is dominant. The dose contribution of the radionuclides of earth's crust origin

is more pronounced than that of ^{40}K whose origin is mainly anthropogenic activities.

Annual effective dose

The cumulative radiation dose received due to exposure to radiation in a year was estimated using the AED. Using the estimates of house occupancy factors in Kenya (60% indoors and 40% outdoors) that were proposed by Mustapha *et al.* [23] and the radiation dose conversion factor of 0.7 Sv per Gy [2], estimates of indoor and outdoor AEDs were determined using Equations (4) and (5) [23–25].

$$AED_{in} = ADR \left(n\text{Gy}h^{-1} \right) \times 8760 \left(by^{-1} \right) \times 0.6 \times 0.7 \left(Sv\text{Gy}^{-1} \right) \times 10^{-6} \quad (4)$$

$$AED_{out} = ADR \left(n\text{Gy}h^{-1} \right) \times 8760 \left(by^{-1} \right) \times 0.4 \times 0.7 \left(Sv\text{Gy}^{-1} \right) \times 10^{-6} \quad (5)$$

The analytical results for range and mean AED_{in} and AED_{out} are presented in Fig. 3 and Table 2.

The soil sample S10 had the highest values of both AED_{in} and AED_{out} of 1.31 ± 0.02 and 0.87 ± 0.01 mSvy^{-1} , while the sample S27 recorded the lowest values of 0.13 ± 0.01 and 0.08 ± 0.01 mSvy^{-1} , respectively. Out of all the samples, 88% recorded a value of AED_{in} lower than the safe limit of 1 mSvy^{-1} . However, all soil samples had AED_{out} lower than the acceptable safe limit. The average values of AED_{in} and AED_{out} were 0.58 ± 0.02 and 0.39 ± 0.01 mSvy^{-1} respectively. Therefore, human habitations constructed using soil from the study region does not expose users to harmful health effects due to indoor or outdoor exposure to gamma radiations.

External and internal hazard indices

The potential danger posed by radionuclides that are inhaled or ingested was estimated using internal hazard index (H_{in}), while the effect of radionuclides that impinge on the skin was estimated using external hazard index (H_{ex}). Both external and internal hazard indices must be lower than 1 (equivalent to Ra_{eq} of 370 Bqkg^{-1}) for radiation to have unnoticeable effects on the general population. Equation (6) was used to estimate these parameters, and the results are summarized in Table 2 and presented in Fig. 3 [12, 20].

$$H_{ex} = \frac{A_{RA}}{370} + \frac{A_{Tb}}{259} + \frac{A_K}{4810} \quad (6)$$

$$H_{in} = \frac{A_{RA}}{185} + \frac{A_{Tb}}{259} + \frac{A_K}{4810} \quad (7)$$

H_{ex} and H_{in} were highest at site S10 with 2.10 ± 0.03 and 2.65 ± 0.04 and lowest in site S27 with 0.20 ± 0.02 and 0.25 ± 0.03 , respectively, for the hazard indices. The mean values H_{ex} and H_{in} were 0.92 ± 0.02 and 1.14 ± 0.03 , respectively. Unlike H_{ex} , the average value of H_{in} exceeded slightly the acceptable safe limit. Nonetheless, other indicators were within the acceptable safe limits. Therefore, soil from Mbeere North region does not pose any danger of internal or external radiation exposure when used as a material for construction of human habitats.

Conclusion

This study quantified the levels of primordial radionuclides in soils from Mbeere North region, Kenya. A Thallium-doped Na(I) gamma ray spectrometer was used to identify and measure the concentration of ^{232}Th , ^{238}U , and ^{40}K in 50 soil samples collected from different locations of the region. The decay rate in the samples was measured and analyzed to determine the activity concentration, radium equivalent, absorbed dose rate, indoor and outdoor AEDs, and internal and external hazard indices. The test results suggest that primordial radionuclides were nonuniformly distributed across the sampling site with higher concentrations on the foot and slopes of the hills in the region. The average activity concentrations of the radionuclides exceeded the world average. Most of the sampled soils had an absorbed dose higher than the world average of 57 nGyh^{-1} but lower than the recommended safe limit of 1500 nGyh^{-1} . Nearly all the soil samples had an indoor effective dose lower than the acceptable safe limit. The outdoor AED for the entire region was lower than the acceptable safe limit of 1 mSvy^{-1} . Among the radiation safety indicators, only the average internal hazard index exceeded slightly the acceptable safe limit. Based on the analysis of the data collected, soils of Mbeere North region are radiologically safe for use in brick making and construction of human habitats.

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