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Radiological Analysis of Suitability of Kitui South Limestone for use as Building Material

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ABSTRACT

Indoor exposure arises from the soils from which the building stands and the building materials used for construction. This is because all building materials contain certain levels of natural radionuclides ^{238}U, ^{232}Th, and ^{40}K. Limestone is the most commonly used building material in Kenya due to its availability and low cost. The limestone from Kitui South has not been mined to process for use but limestone mining and processing plants were being set by the time of this study. This study therefore aimed at evaluating the suitability of the limestone for use as a building material before its mining and processing begins. The activity concentrations of the limestone samples were determined and the radium equivalent, external hazard index, gamma activity index and alpha index evaluated. The results showed that the radium equivalent activity for all the samples was below the recommend limit of 370Bqkg^{-1} and all the radiation hazard indices were below a unit. Therefore, the limestone can be used as building material or for manufacture of building materials without any restrictions.

Keywords: Limestone, Radiation Hazard, Radionuclide, Kitui South
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INTRODUCTION

Human beings are exposed to both outdoor and indoor ionizing radiations from natural sources such as the primordial radionuclides: ^{238}U, ^{232}Th and ^{40}K and cosmic radiations incident on the earth. Indoor exposure may be due to radiations emitted from the ground where the building stands or from building materials used in construction of the building. Construction material can therefore cause substantial radiation exposure if they contain elevated levels of naturally occurring radionuclides. Most building materials derived from rocks and soils, which always contain natural radionuclides of Uranium (^{238}U), and Thorium (^{232}Th) series. The radioactive isotope of Potassium (^{40}K), which give both external and internal radiation exposure to the inhabitants of dwellings built with such materials(Dhanya Balakrishnan, Abraham, Rajagopalan, & Jojo, 2012) in which the gamma radiation arising from the walls, floors and ceilings, and Radon and Thoron and their progeny are the major sources of radiation exposures. Previous studies around the world have indicated the presence of natural radionucliudes in different types of building materials: Marbles in Pakistani(Aslam, Orfi, Khan, & Jabbar, 2002), concrete blocks from Nigeria in Ibadan (Ademola & Oguneletu, 2005), Portland cement used in Nigeria (Ademola, 2008). A report by UNSCEAR (United Nations Scientific Committee On The Effects Of Atomic Radiation, 2000) indicated that building materials that may be of radiological significance include marl, blast furnace slag, fly ash, Portland clinker and anhydrate (in cement industry) and clay (in ceramic industry).

A large number of studies on the level of radioactivity in building materials have done in various parts of the world. In Algeria, a study on the radioactivity in building materials commonly used in the country showed that radium equivalent activities for all the building were all below the criterion limit of gamma-radiation dose (1.5 mSv yr^{-1}) (Amrani & Tahtat, 2001). In another study in India, the radioactivity levels in building materials commonly used locally. In all the materials the radioactivity levels were below the recommended levels except for black stone and red stone which were found to be likely to enrich the indoor exposure if used as building materials (Gupta & Chauhan, 2011). In China a recent study on the radiological implications of use building materials on the indoor exposure showed that he values of external hazard index (Hex) and the internal hazard indexes (Hin) are less than unity implying that the building materials could be used safely for construction of houses (Lu, Li, Yang, & Zhao, 2013). In Kenya, a number of studies have been done to determine the radioactivity levels of building materials in high background radiation areas of Lambwe East (Achola, Patel, Mustapha, & Angeyo, 2012), along the Kenyan Coast (Hashim, Rathore, Kinyua, & Mustapha, 2004), Mrima hills (Mustapha, Patel, & Rathore, 1999) and Homa Bay (Mustapha et al., 1999). The knowledge of the natural radioactivity in limestone is important because it is widely used as a building material as well as a raw material in the manufacture of commonly used building materials such as concrete blocks, cement etc. In Kenya, Portland cement is used for making building blocks, concrete and for plastering mud walled buildings in the rural areas. In towns and cities, buildings are constructed with blocks and cement. Portland cement being one of the lowest cost materials used in construction over the last century throughout the world (Mahmoud, 2007), it is the most commonly used building material in Kenya. Limestone deposits of Kitui South District, Kenya have attracted a number of industries that are in the process of setting mining and processing plants within the area. Despite this, no study has been carried out to establish the radioactivity levels in these limestones thus the risk of elevated exposures in case the radionuclide levels are higher than the recommended limits. This paper reports the radioactivity levels of the limestone samples and the radiation hazard indices associated with the Kitui South Limestone from the three major deposits of Ndulukuni, Kituvwi and Mwanyani (See Figure 1) in a view to determining its suitability for use as a building material or raw material for the manufacture of other building materials.

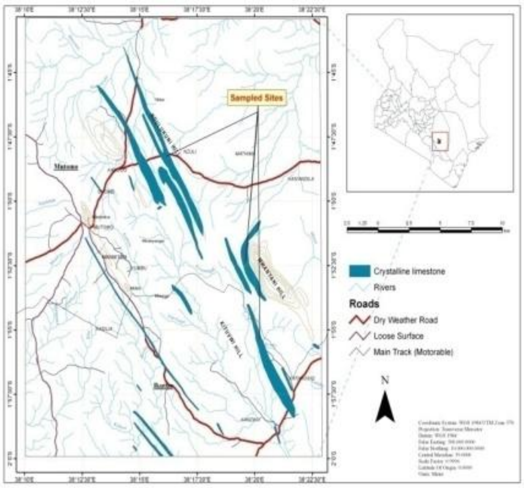

Fig.1 Simplified geological map of Kitui South limestone deposit areas (Waswa, 2011)

MATERIALS AND METHODS

The total number of 45 samples, each weighing between 200g and 400g were collected consisting of 15 samples from each of the three deposits in the region. The limestone samples collected from limestone rock outcrops in Kituvwi, Ndulukuni and Mwanyani. All the samples where pulverized and packed into sample polythene bags ensuring that the sample bags completely filled in order to ensure even distribution of ^{222}Rn and put into plastic containers of the same the volume. The plastic containers where sealed with aluminium foil tightly for 30 days for the radionuclides to attain secular equilibrium (Mohanty, Sengupta, Das, Vijayan, & Saha, 2004). The analysis of samples for activity concentration of radionuclides was performed using hyper pure Germanium (HPGe) detector spectrometry with active volume of 144 cm^{-3} and external diameter of 76 mm. The detector had an efficiency of 31.6% at 1.8 keV. Each sample placed on the detector that enclosed in a thick walled hollow cylinder of lead to shield it from background radiations. Prior to the sample measurement, the environmental gamma ray background inside the laboratory was determined using a similar empty plastic container under identical measurement conditions and subtracted from the measured gamma ray spectra of each sample. Each measured spectrum was analyzed offline by dedicated software PCA3 from Canberra Canada. The detector was energy calibrated using standard source (SRM-1) supplied by the International Atomic Energy Association (IAEA). The activity concentrations of the radionuclides ^{226}Ra was determined from the photopeak of ^{214}Pb (351.9 keV) and ^{214}Bi (609.31 keV) and that of ^{232}Th by the photo-peak of ^{212}Pb (238.63 keV) and ^{228}Ac (911.21 keV). The activity concentration of ^{40}K was determined by its 1460.8 keV photopeak (Jibiri & Emelue, 2008).

RADIUM EQUIVALENT ACTIVITY (Ra_{eq})

This refers to weighted sum of the activities of ^{226}Ra, ^{232}Th and ^{40}K based on the assumption that 1Bq/Kg of ^{226}Ra, 0.7 Bq/Kg of ^{232}Th and 13 Bq/Kg of ^{40}K deliver the same gamma dose rate (Beretka & Matthew, 1985). The Ra_{eq} calculated for the three regions in order to compare the specific activities of the limestone samples from the three different regions. Using equation (1) (Beretka & Matthew, 1985).

$$Ra_{eq} = C_{Ra} + 1.423C_{Th} + 0.077C_K \qquad (1)$$

Where C_{Ra}, $C_{Th,}$and C_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K respectively.

EXTERNAL HAZARD INDEX (H_{ex})

This a quantity used to assess the radiological suitability of a building material. According to ICRP the upper maximum limit radiation dose that arises from building material is 1.5 mSv/y (Mountford & Temperton, 1992). Using a model suggested by (Krieger, 1981) it is calculated that,

$$H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \qquad (2)$$

Where all the variables have same meaning as in equation(1)

GAMMA ACTIVITY INDEX (I_γ)

This is another radiation quantity, which used for controlling the radioactivity of a building material based on controls and exemption levels. In terms of radiation protection, any effective dose rates that exceed 1mSv/y taken into account. Based on this, it has recommended that the excess dose rates offered by building materials above the outdoor effective dose rates should range between 0.3 - 1mSv/y. The activity concentration index also called the Gamma Activity Index, used to determine whether this dose criterion is met (Commission, 1999) and is obtained as;

$$I_\gamma = \frac{C_{Ra}}{300} + \frac{C_{Th}}{200} + \frac{C_K}{3000} \qquad (3)$$

Where all variables retain their meanings

ALPHA INDEX

This is the criterion used for the assessment of excess internal exposure due to alpha radiation originating from the inhalation of radon gas and is defined as;

$$I_\alpha = \frac{C_{Ra}}{200} \quad (4)$$

According to RPA, the recommended exemption level and the upper limit of ^{226}Ra in building materials are 100 Bq/kg and 200Bq/Kg respectively (Strålehygiejne & Säteilyturvakeskus, 1986). When the activity concentration of ^{226}Ra exceeds 200 Bq/Kg it is possible that the radon exhalation from this material exceeds 200 Bq/m^3 and unlikely when the concentration of ^{226}Ra is less than 100 Bq/Kg (Xinwei, Lingqing, Xiaodan, Leipeng, & Gelian, 2006). A ^{226}Ra activity of 200 Bq/Kg gives an alpha index of 1(Tufail & Hamid, 2007).

RESULTS AND DISCUSSION

The activity concentrations in Bqkg^{-1} of ^{238}U series (^{226}Ra), ^{232}Th and ^{40}K in limestone samples analysed from the three limestone deposits found within Kitui South District have been determined. The results are summarised in Table 1.

Table.1 Activity of primordial radionuclides in the limestone

Radionuclide	Activity Concentration	Kituvwi	Mwanyani	Ndulukuni
Uranium series (^{226}Ra)	Maximum	105.9	103.6	67.9
	Minimum	7.3	6.4	11.0
	Average	28.3±23.2	47.4 ± 35.6	32.2 ± 17.7
Potassium (^{40}K)	Maximum	356.0	481.4	664.7
	Minimum	49.4	38.8	56.1
	Average	95.6±73.2	142.6±127.5	87.4 ± 43.0
^{232}Th Series	BDL	BDL	BDL	BDL

BDL- (Below Detection Limits)

The results presented in Table 1 show that average activity concentrations of ^{226}Ra were low with the highest being in Mwanyani (47.4 Bqkg^{-1}) and potassium being highest in samples from Ndulukuni (56.1 Bqkg^{-1}) while the activity concentrations for ^{232}Th were below the detection limits for all the samples analysed.

The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K were thus all below the global averages of 50, 50 and 500 Bq/Kg respectively (United Nations. Scientific Committee on the Effects of Atomic Radiation, 2000). For assessment of the suitability of the limestone samples for use as building materials, the radiation hazard indices ($Ra_{eq}, H_{ex}, I_\gamma, and\ I_\alpha$) were calculated from the activity concentrations of the limestone samples from the different areas that were analysed. The results presented in Table 2.

Table.2 Radiation Hazard Indices for limestone samples analysed

Area	$Ra_{eq,}$	H_{ex}	I_γ	I_α
Kituvwi	35.66	0.10	0.13	0.14
Mwanyani	58.38	0.16	0.21	0.24
Ndulukuni	38.92	0.11	0.14	0.16

The results presented in Table 2 show that the average Ra_{eq} was highest for limestone samples from Mwanyani (58.38 Bqkg^{-1}).

For building materials, the Ra_{eq} should not exceed 370 Bq/Kg, which produces a radiation dose rate of 1.5 mGy/y. This implies that for all the samples analysed the radium equivalent activity was far below the maximum limit making the limestone radiological safe for use as a building material (Krieger, 1981).

The upper maximum limit radiation dose that arises from building material is 1.5 mSv/y for the building material considered safe and be used without restrictions (Icrp, 1991). For a material to be acceptable as a building material without any restriction, the H_{ex} must be less than unity, which implies a maximum value of Ra_{eq} less than 370 Bq/Kg. Here the highest H_{ex} was 0.16 for the samples from Mwanyani thus the limestone found safe for use as a building material. In terms of radiation protection, any effective dose rates that exceed 1mSv/y should be taking into account. Based on this, it has been recommended that the excess dose rates offered by building materials above the outdoor effective dose rates should range between 0.3 - 1mSv/y (Commission, 1999). The activity concentration index also called the gamma activity index used to determine whether this dose criterion is met (Commission, 1999). If the gamma index is equal or less than 1, then the material can be used as a building material without any restrictions in terms of radiological hazard. The Gamma activity index for the limestone in Kitui South found to range from 0.13 to 0.21 thus the limestone samples could be use without the risk of elevated indoor exposure.

According to RPA, the recommended exemption level and the upper limit of ^{226}Ra in building materials are 100 Bq/kg and 200 Bq/Kg respectively(Strålehygiejne & Säteilyturvakeskus, 1986). When the activity concentration of ^{226}Ra exceeds 200 Bq/Kg it is possible that the radon exhalation from this material exceeds 200 Bq/m^3 and unlikely when the concentration of ^{226}Ra is less than 100 Bq/Kg (Xinwei et al., 2006). A ^{226}Ra activity of 200 Bq/Kg gives an alpha index of 1(Tufail & Hamid, 2007). If I_γ is below the recommended value then radon concentration is less than 200 Bq/m3 and I_α is less than 1. The calculated I_α for all the limestone samples ranged from 0.14 to 0.24 with an average of 0.18, thus the radon exhalation from dwellings constructed with the limestone would cause indoor radon concentrations that are obviously less than 200 Bq/m^3.

CONCLUSIONS

The activity concentrations of limestone samples and radiation hazards from the three deposits in Kitui South district have been determined and compared with internationally agreed values in order to determine the radiological risk that could arise due to use of the limestone as building material. The values of the radium equivalent activity and the radiation hazards evaluated (External hazard index, Gamma activity index and alpha index) were all found to be below the recommended limits (< 1). The researcher concludes that the use of limestone from Kitui South as a building material does not pose any significant risk of elevated indoor exposure.

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