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Full Length Research Paper

Activity concentrations of ^{40}K, ^{232}Th, ^{226}Ra and radiation exposure levels in the Tabaka soapstone quarries of the Kisii Region, Kenya

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A radiological study was carried out in the soapstone quarries of Tabaka region of Kisii district in the Southern Nyanza province, Kenya, where soapstone has been mined and used as a carving medium for hundreds of years. To achieve this, 14 soil and rock samples collected from five quarries were analyzed using high-resolution gamma-ray spectroscopy. The activity concentrations of ^{232}Th, ^{40}K and ^{226}Ra in the samples as well as other radiological parameters were determined. These ranged from 38.6 to 271.7 Bqkg^{-1} for ^{232}Th, 43.1 to 360 Bqkg^{-1} for ^{226}Ra, and 245 to 1780 Bqkg^{-1} for ^{40}K. The absorbed dose rates were measured 1metre above the ground at each quarry using a Canberra radiagem 2000 model. The average absorbed dose rate for the five quarries was found to be 541.4 nGyh^{-1} 1m above the ground level while the calculated total absorbed dose rates were found to average 177.6 nGyh^{-1} below the surface. This is about 4 times higher than the world average of 43 nGyh^{-1}. Assuming a 40% occupancy factor, the corresponding annual effective dose rates due to the radionuclides ^{232}Th, ^{226}Ra and ^{40}K in the quarries ranged from 0.215 to 0.875 mSvy^{-1}, with a mean of 0.44 mSvy^{-1}. This corresponds to an excess lifetime cancer risk of 0.07%. The internal and external hazard indices (1.03 and 1.27, respectively) were found to be more than unity, hence slightly exceeding the permissible limits set by the International Commission on Radiation Protection (ICRP). The annual effective dose rate in the quarries was found to be 0.44 mSvy-1 and is less than the 1 mSvy^{-1} upper limit recommended for the public by the ICRP.

Key words: Soapstone, quarries, spectroscopy, absorbed dose, hazard index.

INTRODUCTION

Human beings have always been exposed to ionizing radiations of natural origin, namely terrestrial and extra-terrestrial radiation. Radiation of extra-terrestrial origin is from high energy cosmic ray particles and at sea level it is about 30 nGyh^{-1} (UNSCEAR Report, 2000), while that of terrestrial origin is due to the presence of naturally occurring radionuclides; mainly potassium (^{40}K), rubidium and the radionuclides in the decay chains of thorium (^{232}Th) and uranium (^{238}U). These radionuclides have half-lifes comparable to the age of the earth. Gamma radiation from these radionuclides represents the main external source of irradiation of the human body. Natural radioactivity in geological materials, mainly rocks and soil, comes from ^{232}Th and ^{238}U series and natural ^{40}K. Artificial radionuclides such as ^{137}Cs which result from weapon testing and the Chernobyl nuclear accident could also be present (UNSCEAR, 2000). The levels due to the terrestrial background radiation are related to the types of rock from which the soils originate. Higher radiation levels are associated to igneous rocks such as granite and lower levels with sedimentary rocks. There are some exceptions however, since some shales and phosphate

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Abbreviations: ICRP, International Commission on Radiation Protection; **NRPL,** National Radiation Protection Laboratory; **HPGe,** high purity germanium; **FWHM,** full width at half maximum; **MCA,** multichannel analyzer; **AED,** annual effective dose;H_{ex}, external hazard index;H_{in}, internal hazard index.

Table 1. Quarry identity and depth of sample collection.

Quarry name	Orenge	Nyaberi	Ouma	Barongo	British
Depths at which samples were collected (m)	5.0, 4.0, 0.3	7.0, 4.0	8.0, 4.0, 0.3	8.0, 4.0, 0.3	15.0, 4.0, 0.3

Table 2. Spectroscopic parameters employed for quantification (Tsai et al., 2008).

Element	Emitter nuclide	Half-life of nuclide	Gamma-ray energy (keV)	Absolute emission probability of gamma decay (%)
^{226}Ra	^{214}Bi	19.90 min	609.31	46.30
	^{214}Pb	26.80 min	351.92	37.20
^{232}Th	^{212}Bi	60.55 min	727.17	11.80
	^{212}Pb	10.64 h	238.63	44.60
	^{228}Ac	6.25 h	911.60	27.70
^{40}K		1.3×10^9 y	1460.81	10.67

rocks have a relatively high content of radionuclides (NCRP Report, 1993).

Measurement of natural radioactivity is crucial in implementing precautionary measures whenever the source is found to exceed the recommended limit. The present study aims at investigating naturally occurring radioactive elements and exposure levels to ionizing radiation at the Tabaka soapstone quarries in Kisii district, Kenya. The mining is done by men using hoes, pick axes, machetes, shovels and iron rods. The men dig pits approximately 5 to 30 m deep and diameter 50 to 75 m to excavate the soapstone. No machinery is used. The results are of general interest since soapstone carvings are used as building and ornamental material.

MATERIALS AND METHODS

Sample preparation

A total of 14 samples were collected from the five main quarries named Orenge, Nyaberi, Ouma, Barongo and British as shown in Table 1 below.

After collection, the rock and soil samples were separately crushed into powder form and sieved through a 0.6 mm mesh sieve. They were then dried in an oven at 80°C for 24 h, completely removing water from the samples. A mass of 200 g of each sample was packed in special gas tight polyethylene plastic containers then closed and tightly sealed using cellotape. The containers were labeled appropriately and then kept for 30 days. After this period all the decay products in the ^{232}Th series and ^{226}Ra sub-series were in radioactive equilibrium with their daughters. The samples were then taken for gamma spectrometric analysis at the Kenya National Radiation Protection Laboratory (NRPL).

Activity concentration measurements

The concentrations of radionuclides (^{226}Ra, ^{232}Th and ^{40}K) in each sample were determined using a high purity germanium (HPGe) gamma ray spectrometer consisting of a p-type intrinsic germanium coaxial detector (ORTEC model 7450) mounted vertically. The detector had a relative efficiency of 33 % and full width at half maximum (FWHM) of 2.0 keV energy resolution for the 1332 keV gamma ray line of ^{60}Co. The detector was connected to a Canberra multichannel analyzer (MCA) with apex software, that allowed data acquisition, display of gamma-spectra, analysis of the gamma-spectra and storage of the results in memory channels. The MCA was interfaced with a computer and a printer. The detector was housed inside a 10 cm thick lead shield internally lined with 2 mm Cu foils. The foils provided an efficient suppression of background gamma radiation present within the laboratory. Each sample was run for 43200 s.

The activity concentration for each radionuclide in the measured samples was determined using the total energy under respective peaks after applying appropriate factors for peak efficiency, gamma intensity of the radionuclide and weight of the sample. For example ^{40}K activity was determined from its 1460.81 keV γ-line. The analysis of results was performed using Microsoft Excel software.

Radionuclide identification reports

The ^{226}Ra activities for samples assumed to be in radioactive equilibrium were estimated from ^{214}Pb (351.92 keV) and ^{214}Bi (609.31 keV). The gamma-ray energies of ^{212}Bi, ^{212}Pb and ^{228}Ac were used to estimate activity of ^{232}Th. The activity concentrations of ^{40}K were measured directly by its own gamma rays (1460.81 keV). Details of the employed spectroscopic parameters are shown in Table 2.

Absorbed dose

Two approaches are used to estimate the external doses that result from deposition of radionuclides in soil surfaces: direct measurements and calculations based on radionuclide deposition densities.

Measurement of absorbed dose rates in air

Absorbed dose rates in air were measured 1m above the surface at

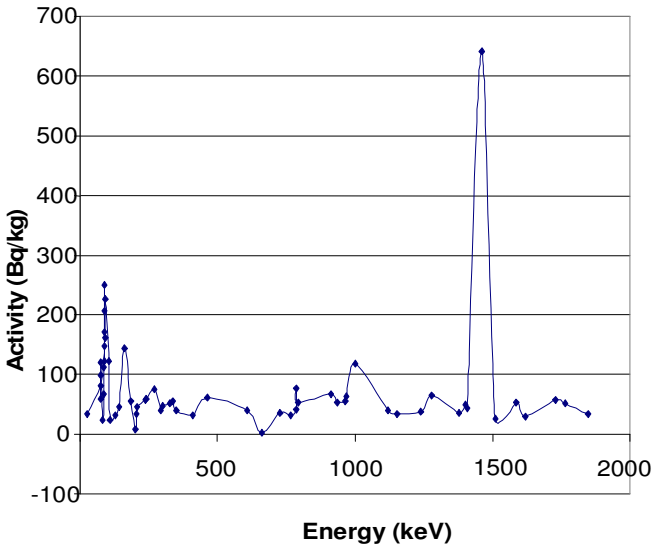

Figure 1. A γ-ray spectrum for a sample from Barongo quarry collected at a depth of 3 m.

each quarry using a hand held survey meter Canberra-radiagem 2000 model with a reading range of up to 100 μSvh^{-1}. The absorbed dose rates in air in nGyh^{-1} were computed from the dose rates in μSvh^{-1} as measured in the field using the conversion coefficient factor of 0.7 SvGy^{-1} as recommended by UNSCEAR, 2000.

Calculation of absorbed dose rate from measured activity concentrations

Radiation emitted by a radioactive substance is absorbed by any material it encounters. UNSCEAR (2000) has given the dose conversion factors for converting the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K into dose (nGyh^{-1} per Bqkg^{-1}) as 0.427, 0.662 and 0.043, respectively. Using these factors, the total absorbed dose rate in air is calculated as given in the Equation (1) (UNSCEAR, 2000).

$$D = (0.427C_{Ra}+0.662C_{Th} + 0.043C_K)\ nGyh^{-1}, \quad (1)$$

where C_{Ra}, C_{Th} and C_K are the activity concentrations (Bqkg^{-1}) of radium, thorium and potassium, respectively in the samples.

Calculation of annual effective dose

To estimate the annual effective dose (AED), took into account the conversion coefficient, (0.7 SvGy^{-1}) from the absorbed dose in air to effective dose and the outdoor occupancy factor (~ 20 %) (UNSCEAR, 2000). The average fraction of time spent indoor and outdoor (occupancy factors) in Kenya are 0.6 and 0.4, respectively (Mustapha, 1999). The world average indoor and outdoor occupancy factors are 0.8 and 0.2, respectively (UNSCEAR, 2000). The effective dose rate in units of mSvy^{-1} was estimated using the formula (UNSCEAR, 1998);

$$\text{Effective dose rate} = \text{dose rate} \times 24\ h \times 365\ \text{days} \times 0.4 \times 0.7\ SvGy^{-1} \times 10^{-6} \quad (2)$$

External hazard index (H_{ex})

Radiation exposure due to ^{226}Ra, ^{232}Th and ^{40}K may be external. This hazard, defined in terms of external hazard index or outdoor radiation hazard index and denoted by H_{ex}, can be calculated using the equation (Beretka and Mathew 1985):

$$H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \quad (3)$$

where C_{Ra}, C_{Th} and C_K are activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively in Bqkg^{-1}. The value of this index should be less than 1 mSvy^{-1} in order for the radiation hazard to be considered acceptable to the public (Beretka and Mathew, 1985).

Internal hazard index (H_{in})

The internal hazard index (H_{in}) gives the internal exposure to carcinogenic radon and is given by the formula (Beretkab and Mathew, 1985):

$$H_{in} = \frac{C_{Ra}}{185} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \quad (4)$$

where C_{Ra}, C_{Th} and C_K are activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively, in Bqkg^{-1}. The value of this index should be less than 1 mSvy^{-1} in order for the radiation hazard to have negligible hazardous effects to the respiratory organs of the public (Beretka and Mathew, 1985).

RESULTS AND DISCUSSION

γ-ray spectrum and nuclide identification

Various spectra that were generated from various samples from during spectrometric analysis were used to identify various radio nuclides present in the sample. A typical γ-ray spectrum is shown in Figure 1.

Based on the spectroscopic parameters employed for quantification (Table 2) and the gamma ray spectra for various samples (such as the one in Figure 1), several radionuclides were identified in the samples. Nuclide identification reports for all quarries were compiled and the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K were isolated as in Table 3.

The general activity of ^{226}Ra, ^{232}Th and ^{40}K in the collected samples were in the range of 43.1-360, 38.6-271.7 and 307.0-1780 Bqkg^{-1}, respectively. There was no particular relationship between activity of the radionuclides and depth of the point of collection of the sample. This could be attributed to the fact that there was mixing of rock debris during refilling of the quarries for formation of new soapstone which is again excavated after 5-10 years.

In all quarries, ^{40}K had the highest activity concentration.

Table 3. Activity of radio nuclides ^{232}Th, ^{226}Ra and ^{40}K in the various samples.

Quarry name	Depth of sample collection point (m)	Activity (Bqkg^{-1})		
		^{232}Th	^{226}Ra	^{40}K
Orenge	0.3	147.7	72.6	587.0
	4	84.8	43.1	1780.0
	5	38.6	52.2	922.0
Nyaberi	4	263.3	75.8	307.0
	7	100.2	52.5	1020.0
Ouma	1	120.8	49.9	887.0
	4	93.9	63.3	813.0
	8	258.4	360.0	744.0
Barongo	0.3	15.8	78.2	641.0
	4	172.2	61.0	245.0
	8	92.7	177.9	1090.0
British	0.3	210.4	72.7	534.0
	4	271.7	82.0	284.0
	15	236.5	65.8	403.0

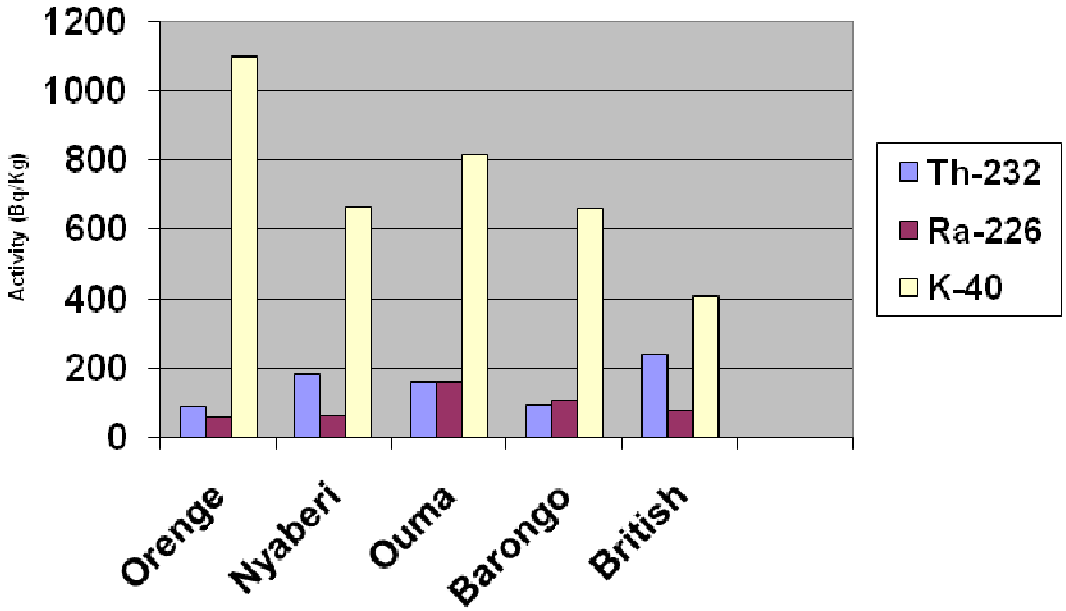

Figure 2. Bar graph showing average activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th from the five quarries.

All the average values were higher than the world wide average activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th which are 400, 35 and 30 Bqkg^{-1}, respectively (UNSCEAR, 2000).

Potassium activity varied widely between 245 and 1780 Bqkg^{-1} due to heterogeneous soil characteristics. Figure 2 shows comparisons between the average activity concentrations of the radionuclides ^{40}K, ^{226}Ra and ^{232}Th in the samples retrieved from the five quarries.

The variation of natural radioactivity levels at different sampling sites was due to the variation of concentrations of radionuclides in the geological formations. The younger granites represent the highest elevation while the older rock is relatively low. The Presence of such high radioactivity in younger granites may be attributed to the presence of relatively increased amount of accessory minerals such as zircon, iron oxides, fluorite and other radioactive related minerals. These minerals play an

Table 4. Absorbed dose rates measured 1 m above the surface at each quarry.

Quarry name	Absorbed dose rates ($nGyh^{-1}$)
Orenge	483
Nyaberi	480
Ouma	557
Barongo	590
British	597

Table 5. Calculated absorbed dose rates ($nGyh^{-1}$) of the samples.

Quarry name	Dose rates due to ^{232}Th ($nGyh^{-1}$)	Dose rates due to ^{226}Ra ($nGyh^{-1}$)	Dose rates due to ^{40}K ($nGyh^{-1}$)	Total absorbed dose rates ($nGyh^{-1}$)
Orenge	59.8	23.9	47.1	130.8
Nyaberi	120.3	27.4	28.5	176.2
Ouma	104.4	67.6	35.0	207.0
Barongo	93.2	45.1	28.3	166.6
British	158.6	31.4	17.5	207.5

important role in controlling the distribution of uranium and thorium. Zircon usually contains uranium and thorium concentration ranging from 0.01 to 0.19% and 1 to 2%, respectively (Cuney et al., 1987). Uranium in iron oxides is trapped by adsorption (Speer et al., 1981).

Absorbed dose rates

The measured absorbed dose rates in air measured 1 m above the surface at each quarry are presented in Table 4. The mean measured absorbed dose rate for the five quarries was 541.4 $nGyh^{-1}$. This is about nine times higher than the world average value of 60 $nGyh^{-1}$ (UNSCEAR, 2000).

The absorbed dose rates due to terrestrial gamma rays 1m above the ground were calculated using Equation (1). Other radionuclides such as ^{137}Cs, ^{235}U, ^{231}Th, ^{211}Bi and ^{22}Na were neglected because they contribute very little to the total dose rates from environmental background (Kocher and Sjoreen, 1985). The results of the calculations are presented in Table 5.

The calculated average total absorbed dose rates due to the presence of ^{226}Ra, ^{232}Th and ^{40}K in each of the quarries were 130.8, 176.2, 207.0, 166.6 and 207.5 $nGyh^{-1}$ for Orenge, Nyaberi, Ouma, Barongo and British quarries, respectively. The calculated average absorbed dose rate for the five soapstone quarries was found to be 177.6 $nGyh^{-1}$. This is about 4 times higher than the world average of 43 $nGyh^{-1}$ (UNSCEAR, 2000). The measured absorbed dose rates in air were much higher than the calculated absorbed dose rates for all the quarries. This may be attributed to the fact that the measured absorbed dose rates may not have come wholly from the sampled rock. They could have originated from other rocks and soils on the surface. Also the measured absorbed dose rates may include the dose due to terrestrial sources**.** The contribution by each of the radionuclides ^{226}Ra, ^{232}Th and ^{40}K to the total absorbed dose rate at the quarries were 25% (46.0 $nGyh^{-1}$), 58% (106.3 $nGyh^{-1}$) and 17% (31.5 $nGyh^{-1}$) respectively as illustrated in Figure 3.

A comparison between absorbed dose rates at Tabaka soapstone quarries and absorbed dose rates at other places in the world was done. Details of the comparison are presented in Table 6.

Annual effective dose rates, external (H_{ex}) and internal (H_{in}) hazard indices

The external hazard index was calculated using Equation (3). The average external hazard index from all the samples was 1.03. This average value is greater than the acceptable average value of unity (ICRP, 2000).

The internal exposure to ^{222}Rn and its radioactive progeny is controlled by the internal hazard index (H_{in}) which is given by Equation (4). The average internal hazard index for all the samples is 1.27. For safe use of a material in the construction of human dwellings, H_{in} should be less than unity (ICRP, 2000).

The mean annual effective dose rate in all the samples is 0.44 $mSvy^{-1}$. The value is more than the average annual effective dose rate (5.62 μSvy^{-1}) received by artisanal gold miners at Osiri, Macalder, Mikei and Masara gold mines in South Nyanza (Odumo, 2009). Assuming that the quarry workers work in the quarries for 40 years, and the risk factor is 0.04 per Sv (ICRP, 2008), the excess lifetime cancer risk is 0.07%. The average

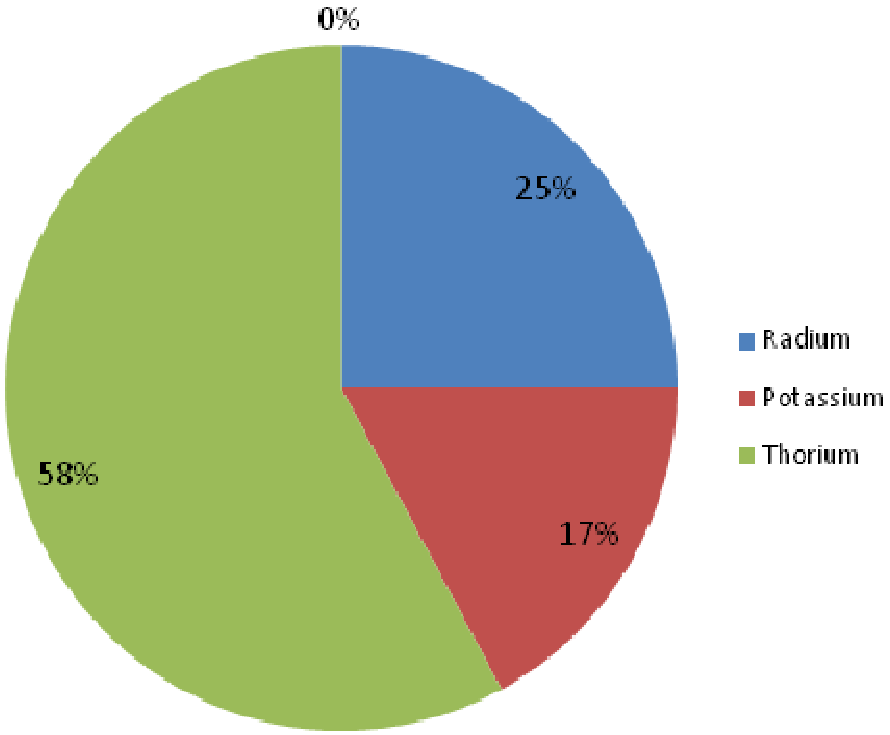

Figure 3. Percentage contributions to the total absorbed dose rates due to Thorium (^{232}Th), Radium (^{226}Ra) and Potassium (^{40}K) from the samples.

Table 6. Comparison of absorbed dose rates in Tabaka soapstone quarries with other areas of the world.

Country	Absorbed dose rates (nGyh^{-1})	References
Ruri hill, Kenya	949	Achola (2009)
Minas, Brazil	220	Malanka et al. (1993)
Firtina valley, Turkey	77	Kurnaz et al. (2007)
Xiazhuang, China	124	Yang et al. (2005)
Eskisehir, Turkey	167	Orgun et al. (2005)
Eastern desert, Egypt	448	Arafa (2004)
Tabaka, Kenya	178	Present study

value and all the annual effective dose rate values in Table 7 are less than 5.705 mSvy^{-1} obtained by Achola (2009) in a radiological survey carried out at Lambwe east location and are also less than 1 mSvy^{-1} which is the annual effective dose rate limit for the public exposure (ICRP, 2000). However care has to be taken since it is believed that radiation at any level poses a risk. The calculated values of annual effective dose rates, external and internal hazard indices are presented in Table 7.

Conclusions

The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K were all found to be above the world's average. In all quarries sampled, ^{40}K had the highest activity concentration. The measured average absorbed dose rate in air (541.4 nGyh^{-1}) at the soapstone quarries was 9 times higher than the world measured average (60 nGyh^{-1}) and 2.9 times higher than the calculated average.

The calculated average absorbed dose rate in air (177.6 nGyh^{-1}) due to gamma-ray emitters in the soapstone quarries was 4 times higher than the world average (43 nGyh^{-1}). Thorium and potassium contributed the highest and lowest values respectively to the average absorbed dose rates in the quarries. The averages for both the external (1.03) and internal (1.27) hazard indices exceeded a unity, the limit recommended by ICRP 2000. Therefore soapstone rock may not be suitable for construction of houses. The annual effective dose rate in

Table 7. Annual effective dose rate, external and internal hazard indices.

Quarry name	Average effective dose rate ($mSvy^{-1}$)	External hazard index (H_{ex})	Internal hazard index (H_{in})
Orenge	0.321	0.73	0.88
Nyaberi	0.432	1.02	1.19
Ouma	0.507	1.20	1.63
Barongo	0.408	0.97	1.25
British	0.509	1.21	1.41

the quarries (0.44 $mSvy^{-1}$) was less than 1 $mSvy^{-1}$, the limit recommended for the public (ICRP, 2000), hence soapstone products are safe to consumers.

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