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RADIATION EXPOSURE LEVELS ASSOCIATED WITH GOLD MINING IN SAKWA WAGUSU AREA, BONDO DISTRICT, KENYA.

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ABSTRACT
The concentrations and distribution of natural radionuclides in soils and water from around Sakwa Wagusu Area, Kenya were investigated with an aim of evaluating the environmental radioactivity and radiological health hazard. These radionuclides are a source of radiation exposure which has been documented to have detrimental health effects for populations living in high background radiation area. Thirty nine rock and soil samples were collected from ten sites of Sakwa Wagusu area, Bondo district to measure their natural radioactivity concentrations due to ^{226}Ra, ^{232}Th and ^{40}K radionuclides. Measurements were done by use of gamma spectrometry method with a high purity germanium (HPGe) detector. The calculated activities for ^{226}Ra ranged from 7.2 – 113.8 Bqkg^{-1} (mean: 44.2Bqkg^{-1}), for ^{232}Th ranged from 4.6 – 100.7 Bqkg^{-1}(mean: 40.3Bqkg^{-1}) and for ^{40}K ranged from 119.3 – 1611.8Bqkg^{-1} (mean: 639.6Bqkg^{-1}). The mean measured absorbed dose rate for the areas under study, measured at 1m above the surface was 141.6nGyh^{-1} while the calculated total absorbed dose rates were found to average 69.8nGyh^{-1} below the surface. To evaluate the radiological hazard of the natural radioactivity, the radium equivalent activity, the external hazard index, internal hazard index, the absorbed dose rate and the effective dose rate have been calculated and compared with internationally approved values. The radium equivalent activity values of all samples are lower than the limit of 370Bqkg^{-1}. The values of the external and internal indices are less than unity. The calculated mean outdoor effective dose rate was 0.17mSvy^{-1} and is less than 1mSvy^{-1} upper limit recommended for the public by ICRP. An excess lifetime cancer risk of 0.02% was achieved with an assumption of 30% occupancy factor, the risk factor of 0.04 per Sv (ICRP, 2008) and the mean annual effective dose rate of 0.17mSvy^{-1}.

Keywords: Natural radioactivity, Gold, Gamma-ray Spectrometry, Sakwa Wagusu, Activity.

INTRODUCTION
Radionuclides have always been present in the natural environment. The main natural contributors to external exposure from gamma-radiation are the uranium and thorium series, together with potassium 40 (^{40}K) wherever present in small quantities in the earth and in the building materials. Long-lived radioactive elements such as uranium, thorium and potassium and any of their decay products, such as radium and radon are examples of Naturally Occurring

Radioactive Materials (NORM). These elements have always been present in the earth's crust and atmosphere.

Throughout the history of life on earth, organisms have been continuously exposed to cosmic rays in the atmosphere, and from naturally occurring radionuclides which are ubiquitously distributed in all living and non-living components of the biosphere (Wicker and Schults, 1982). A wide range of activity concentrations in a wide variety of materials is reported (IAEA-TECDOC-1660). Examples of ores that have been found to be associated with elevated radionuclide concentrations include those of uranium, tin, tantalum, niobium, rare earths, aluminium, copper, gold and phosphate. The mining and processing of these ores can lead to further increases in radionuclide concentrations in the products, by-products or residues (IAEA-TECDOC-1660).

Natural atmospheric radioactivity is determined by the concentration of radon and its daughter elements in air. Radon is formed in the decay chain of naturally occurring primordial radionuclides ^{238}U, ^{232}Th and ^{235}U present in the earth's crust. When radon is formed, it may diffuse from the rocks and soils to enter the atmosphere. The extent to which radon will diffuse into the atmosphere depends on the geological (type of rock or soil matrix, water content) and meteorological (atmospheric temperature, pressure) factors (Schery *et al.*, 1998; Nazaroff, 1992; Schubert *et al*., 2002; Sannappa *et al.*, 1997, 1999; UNSCER, 1993). Radioactivity is due to alpha (α), beta (β) and gamma (γ) radiation from the unstable isotopes in the composition. For the most part, minerals that contain potassium (K), uranium (U) and thorium (Th) are radioactive.

Sakwa Wagusu goldmine area employs many people, both men and women. The mining is done manually by digging using hoes, mattock and spade and when hard rocks are reached, blasting is done. As the pits are dug, underground water is detected which is then pumped out and used for panning. Women can be seen panning around the mines. Gamma-ray spectrometer was used to find the concentration of natural radionuclides ^{226}Ra, ^{232}Th and ^{40}K in the collected samples. The radium equivalent activity, the external and internal indices, the absorbed dose rate, and the effective dose rates were calculated and compared with internationally approved values.

MATERIALS AND METHODS:

Sample Collection and Preparation

A total of 39 soil and rock samples were collected from four main areas (Wagusu market, Abimbo, Nyang'oma and Nango) comprising ten mines at different heights from the ground as shown in table 1 below. At each height, a sample of about 800g was collected in a straight line along that particular height. Figure 1 shows the main areas of study.

After collection, rock and soil samples were dried and then crushed separately into powder form. The crushed powder were then sieved through a 0.6mm mesh sieve, dried in an oven at 100°C for 24 hours, completely removing moisture from the samples. Each sample was then weighed and packed in special polyethylene plastic containers and tightly closed for about 4 weeks which was a sufficient time required to attain a state of secular radioactive equilibrium after their progeny (Karahan *et al.,* 2000, Kinyua *et al.,* 2011). After this period, the samples were then

taken for spectroscopic analysis. Both the field and laboratory aspect of this project were carried out between August, 2012 and March, 2013.

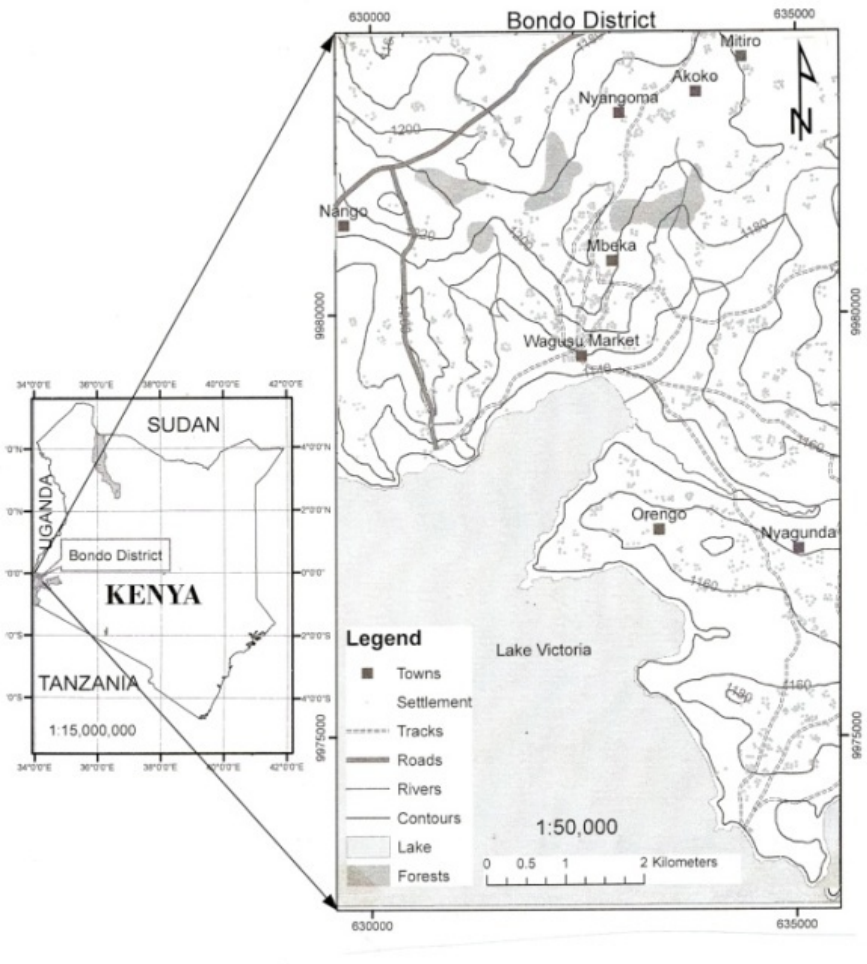


Figure 1: Location of the Study Area and Sampling Areas.

Table 1: Location, site, depth of rock and soil samples collection, average activity concentration of sites and average activity concentration of Wagusu market, Abimbo, Nyang'oma and Nango Areas.

Location	Site Name	Depth of Sample Collection (m)	Average Activity Concentration ($Bqkg^{-1}$)			Average Activity Concentration ($Bqkg^{-1}$)		
			^{226}Ra	^{232}Th	^{40}K	^{226}Ra	^{232}Th	^{40}K
Wagusu Market	K'Onduru 3	0.2, 2.0, 5.0, 15.0, 30.0	35.0	20.0	401.4			
	K'Onduru 4	0.3, 2.0, 8.0, 15.0	25.6	27.0	406.0			
	K'Opiyo	0.3, 5.0, 7.0, 15.0, 25.0	47.8	41.8	612.1	35.4	28.6	438.4
	K'Otieno Manyala	0.2, 1.0, 3.5, 8.0	33.0	25.6	334.2			
Abimbo	Ka'Nyasembo	0.2, 2.0, 5.0, 7.0, 35.0	34.5	27.4	568.4	36.0	29.9	575.5
	K'Obonyo	0.3, 5.0, 7.0, 15.0, 25.0	37.4	32.3	582.6			
Nyang'oma	K'Ondoro	0.1, 1.5, 4.0, 9.0	71.4	43.1	733.3			
	Ka'John	0.1, 1.5, 6.5	26.7	35.0	682.1	44.2	40.7	647.7
	K'Okumu	0.03, 1.0	34.6	44.1	527.6			
Nango	K'Ogoch	0.3, 6.0	61.2	61.9	896.6	61.2	61.9	896.6

Activity Concentration of Natural Radionuclide and Absorbed Doses

Measurement of activity concentrations was performed with high-purity germanium (HPGe) gamma-ray detector with $144mm^3$ active volume and outside diameter of 76mm. The detector has an efficiency of 31.6% and a resolution of 1.8keV. Each sample was put in a malineri beaker of $500cm^3$ and filled up to the same level as the certified reference standard soil (IAEA –RGK-1, IAEA-RGU-1, IAEA-RGTh-1 and IAEA-375 soil) then placed in a lead shielded detector. A counting time of between 22000 – 179000 seconds was adopted. A soil sample (IAEA –RGK-1, IAEA-RGU-1, IAEA-RGTh-1 and IAEA-375 soil) was analysed for the method of validation and spectrometer calibration using PCA 2 (Version 1.00 software).

The activity concentration of each radionuclide was then calculated using the comparative method. For each sample, there were five significant gamma lines, ^{40}K line, $^{214}P_b$ and $^{214}B_i$ lines from ^{238}U and $^{212}P_b$ and $^{228}A_c$ lines from ^{232}Th. The activity of ^{40}K was evaluated from 1461keV gamma line while the activity of ^{238}U from 352keV and 609keV gamma lines of $^{214}P_b$ and $^{214}B_i$ respectively and that of ^{232}Th from 238keV and 912keV gamma lines of $^{212}P_b$ and $^{228}A_c$ respectively.

Absorbed Dose

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

Measurement of Absorbed Dose Rate in Air

The absorbed dose rates in air at 1m above the ground surface were measured for each pit mine using a hand-held survey meter Thermo Scientific, FH 40G-L10 Radiameter with a reading range of $10nSvh^{-1}$ – $100mSvh^{-1}$. The absorbed dose rates in air in $nGyh^{-1}$ were computed from

the dose rates in $nSvh^{-1}$ as measured in the field using the conversion coefficient factor of $0.7SvGy^{-1}$ as recommended by UNSCEAR, 2008.

Radiation Hazard Indices

The total absorbed dose rates (D) due to gamma radiations in air of the naturally occurring radionuclides ^{226}Ra, ^{232}Th and ^{40}K were calculated based on guidelines provided by UNSCEAR 2000. Assumption was that the contributions from other naturally occurring radionuclides were insignificant. D can therefore be calculated from equation (1):

$$D\ (nGy.h^{-1}) = 0.427A_{Ra} + 0.662A_{Th} + 0.043A_K \quad \textbf{(1)}$$

Where A_{Ra}, A_{Th} and A_K are the activity concentrations ($Bqkg^{-1}$) of radium, thorium and potassium respectively in the samples, and 0.427, 0.662 and 0.043 are the dose conversion factors for converting the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K into dose ($nGy.h^{-1}$ per $Bqkg^{-1}$) as given by UNSCEAR 2000.

To estimate the annual effective dose rates, the conversion coefficient from absorbed dose in air to effective dose ($0.7Sv.Gy^{-1}$) and outdoor occupancy factor ($0.2Sv.Gy^{-1}$) proposed by UNSCEAR 2008 are used. In Kenya, the average of time spent indoor and outdoor (occupancy factors) 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, 2008). Therefore, the effective dose rate in units of $mSvy^{-1}$ was estimated using the formula (UNSCEAR, 1998);

$$Effective\ dose\ rate = Dose\ rate\ (D) \times 8760h.y^{-1} \times 0.4 \times 0.7\ Sv.Gy^{-1} \times 10^{-6} \quad (2)$$

The most widely used radiation hazard index, radium equivalent activity (Ra_{eq}), is calculated using the formula as (Berekta *et al.,* 1985):

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_K \quad (3)$$

A widely used hazard index, reflecting the external exposure, called the external hazard index, H_{ex}, is defined as (Harb *et al.,* 2010):

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (4)$$

In addition to external hazard index, radon and its short-lived products are also hazardous to the respiratory organs. The internal exposure to radon and its daughter products is quantified by the internal hazard index, H_{in}, which is given by the equation (Harb *et al.,* 2010):

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (5)$$

The values of the indices (H_{ex} and H_{in}) must be less than unity for the radiation hazard to be negligible.

RESULTS AND DISCUSSION

The specific activity concentrations of ^{238}U series (^{226}Ra), ^{232}Th series (^{232}Th) as well as ^{40}K, expressed in $Bqkg^{-1}$ for samples obtained from Wagusu market, Abimbo, Nyang'oma and Nango areas are presented in Table 1. It can be seen from Table 1 that the average range of ^{226}Ra values was 25.6 to $47.8Bqkg^{-1}$, 34.5 to $37.4Bqkg^{-1}$, 26.7 to $71.4Bqkg^{-1}$, and $61.2Bqkg^{-1}$ respectively. The average value for ^{232}Th ranged from 20.0 to $41.8Bqkg^{-1}$, 27.4 to $32.3Bqkg^{-1}$, 35.0 to $44.1Bqkg^{-1}$ and $61.9Bqkg^{-1}$ respectively and for ^{40}K, the average values ranged from 334.2 to $612.1Bqkg^{-1}$, 568.4 to $582.6Bqkg^{-1}$, 527.6 to $733.3Bqkg^{-1}$ and $896.6Bqkg^{-1}$ respectively for the same sites. There was no particular relationship between activity of the radionuclides and depth of the point of collection of the samples. From table 1, it can be noticed that ^{40}K had the highest activity concentration.

The average values of the activity of ^{40}K were higher than the worldwide recommended value of $400Bqkg^{-1}$ (UNSCEAR, 2008) in all the areas. The average values of ^{226}Ra and ^{232}Th were within the worldwide allowable limits for Wagusu market and Abimbo areas and higher than the worldwide recommended values of $37Bqkg^{-1}$ and $33Bqkg^{-1}$ (UNSCEAR, 2008) for Nyang'oma and Nango areas respectively. Figure 2 shows comparisons between the average activity concentrations of the radionuclides ^{40}K, ^{226}Ra and ^{232}Th in the obtained samples.

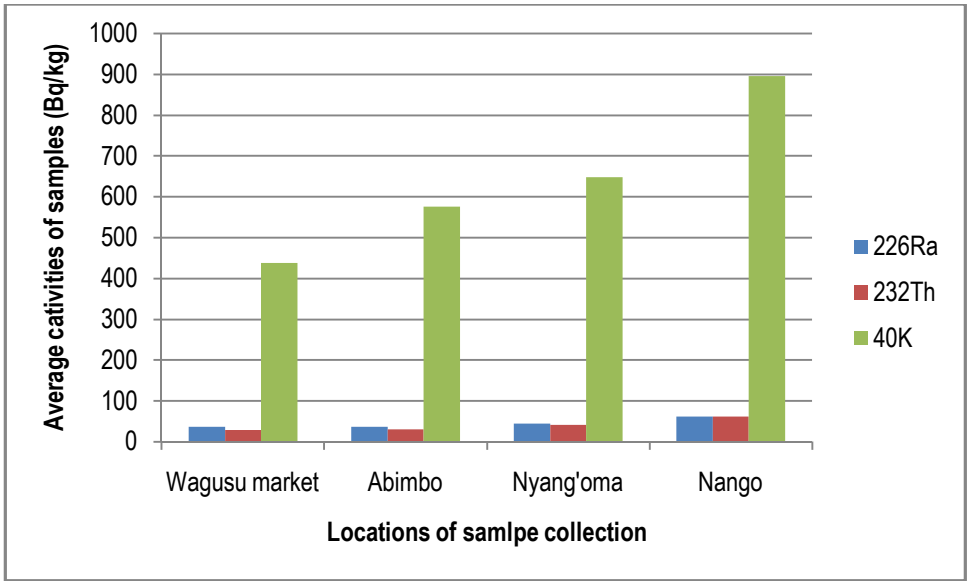


Figure 2: Average Value of Sample Activities Due to ^{226}Ra, ^{232}Th and ^{40}K in All Area under Investigation

Comparison of Activity Concentrations with Similar Studies

The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in soil and rock samples from studied area was compared with those from similar investigations in other countries and a summary results given in table 2.

Table 2: Comparison of Natural Radioactivity Levels in Soil Samples under Investigation with Those in Other Countries

Country	Activity concentration ($Bqkg^{-1}$)			References
	^{226}Ra	^{232}Th	^{40}K	
Saudi Arabia, Taif	23.8	18.6	162.8	A. El-Aydarous
Syrian	20	20	270	UNSCEAR, 2000
Turkey (Istanbul)	21	37	342	Karahan *et al.*
Cyprus	7.1	5	104.6	Tzortzis *et al.*
Denmark	17	19	460	UNSCEAR, 2000
Canada (Saskatchewan)	19	8	480	Kiss, J *et al.*
Tafila/Jordan	35	18	320	Abu-Haija
World average	37	33	400	UNSCEAR, 2008
Kenya (Sakwa Wagusu area)	40.7	35.8	574.4	Present work

Absorbed Dose Rates

The measured absorbed dose rates (averages) in air measured 1m above the surface at each pit mine are as represented in Table 3. The mean measured absorbed dose rate for the ten pit mines was 141.6**$nGyh^{-1}$** against the world average value of 60**$nGyh^{-1}$** (UNSCEAR, 2000).

Table 3 also gives the average absorbed dose rates as measured 1m above the surface at each location.

For clarity purposes, these values of average measured absorbed dose for the areas under study are presented in a bar graph of Figure 3a. Figure 3b shows these averages as calculated from the activities after analysis.

Table 3: Average absorbed dose rates as measured 1m above the surface at each pit mine and average absorbed dose rates as measured 1m above the surface at each location.

Location	Site name	Average Absorbed dose rate ($nGyh^{-1}$)	Average Absorbed dose rate ($nGyh^{-1}$)
Wagusu market	K'Onduru	118.9	124.4
	K'Opiyo	122.9	
	K'Otieno Manyala	131.4	
Nango	K'Ogoch	139.3	151.8
	Ka'Nyakwaka	164.3	
Abimbo	K'Obonyo	138.6	138.7
	Ka'Nyasembo	138.7	
Nyang'oma	K'Ondoro	195.7	151.3
	K'Okumu	117.7	
	Ka'John	140.6	

Table 4 shows calculated absorbed dose rates due to ^{232}Th, ^{226}Ra and ^{40}K and the total average absorbed dose rate for all the sites. The gamma dose rates due to naturally occurring terrestrial

radionuclides ^{232}Th, ^{226}Ra and ^{40}K were calculated based on their activities in soil samples, determined by gamma-ray spectrometry.

From Table 4, the total average absorbed gamma dose rates due to the radionuclides ^{232}Th, ^{226}Ra and ^{40}K were 47.2, 55.2, 71.2 and 105.6nGyh^{-1} for Wagusu market, Abimbo, Nyang'oma and Nango areas respectively. Therefore, the calculated average absorbed dose rate of 69.8nGyh^{-1} was higher than the world average value of 54nGyh^{-1} (UNSCEAR, 2000) in all the areas.

Table 5 shows the average values of annual effective dose (mSvy^{-1}), external hazard index (H_{ex}) internal hazard index (H_{in}) and radium equivalent activity (Bqkg^{-1}) of the studied soil samples from the sites.

From table 5, the estimated mean annual effective dose was 0.11, 0.14, 0.17 and 026mSvy^{-1} for Wagusu market, Abimbo, Nyang'oma and Nango areas respectively. The external hazard index (H_{ex}) was calculated and the mean value was 0.26, 0.31, 0.37 and 0.59 for Wagusu market, Abimbo, Nyang'oma and Nango areas respectively. The internal hazard index (H_{in}) was also calculated, the mean value was 0.33, 0.37, 0.46 and 0.76 for Wagusu market, Abimbo, Nyang'oma and Nango areas respectively. Radium equivalent activity (Ra_{eq}) was also calculated and the mean value was 96.9, 112.0, 135.2 and 218.6Bqkg^{-1} for Wagusu market, Abimbo, Nyang'oma and Nango areas respectively.

From table 7, it can be observed that:

- The values of annual effective dose were lower than the worldwide value of 1mSvy^{-1}. (ICRP, 2000). To achieve these values, equation 2 was used.
- The values of external hazard index (H_{ex}) and internal hazard index (H_{in}) are lower than acceptable limit of unity. These values were computed from equations 4 and 5 respectively.
- Radium equivalent (Ra_{eq}) values for the studied areas (computed from equation 3) were found to vary from 96.4 to 218.6Bqkg^{-1}, with an average of 140.7Bqkg^{-1}. These values were lower than the internationally accepted value of 370Bqkg^{-1} (Lu Xinwei *et al*., 2006).

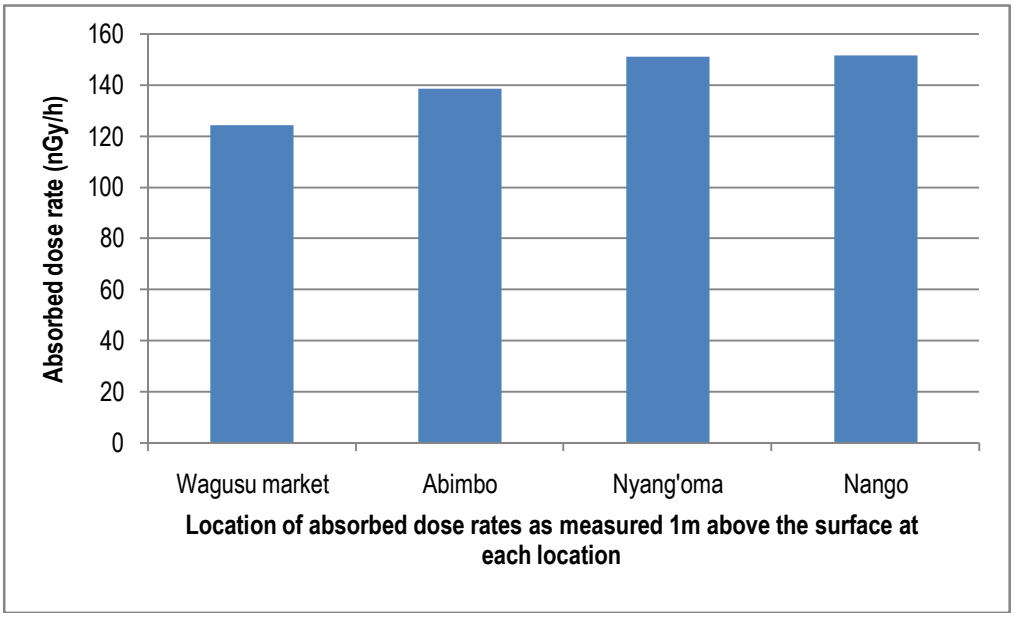
Assuming that the mine workers work in the mines for 30years, the mean annual effective dose rate is 0.17mSvy^{-1} and the risk factor is 0.04 per Sv (ICRP, 2008), then the excess lifetime cancer risk is 0.02%.

Figure 1: Location of the study area and sampling areas.

Figure 2: Shows average values of sample activities due to ^{226}Ra, ^{232}Th and ^{40}K in all area under investigation.

Figure 3: **(a)** Shows average absorbed dose rates as measured 1m above the surface at each Location
(b) Average absorbed dose rates as calculated at each location.

Figure 4: The average values of:
(a) Annual effective dose
(**b)** Radium equivalent activity
(c) External and internal hazard index in all the area under investigation.



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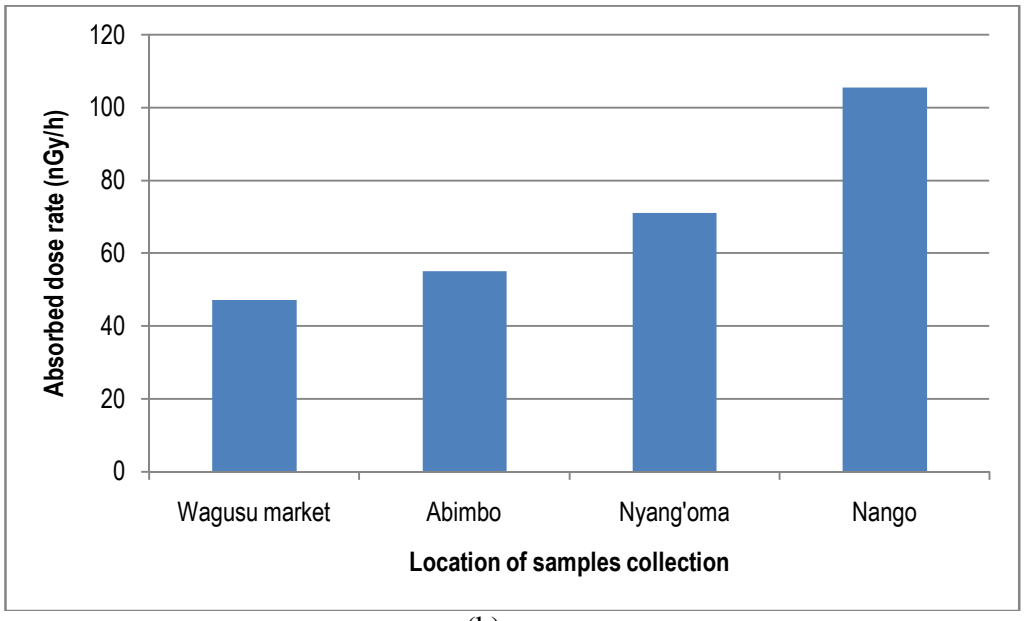


(b).

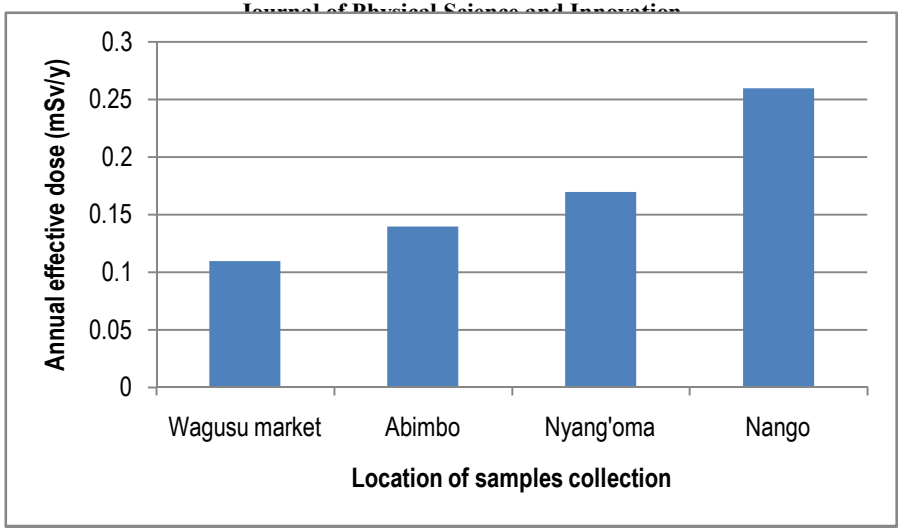
Figure 3: **(a) Average absorbed dose rates as measured 1m above the surface at each location,**
(b) Average absorbed dose rates as calculated at each location.

Table 4: Calculated Absorbed Dose Rates ($nGyh^{-1}$) of the Samples from Locations

Location	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 Average Absorbed Dose Rate ($nGyh^{-1}$)
	Range	Average	Range	Average	Range	Average	
Wagusu Market	4.0 – 36.5	17.2	3.1 – 25.0	11.1	5.1 – 45.3	18.9	47.2
Abimbo	3.0 – 47.5	19.8	4.2 – 25.2	10.7	7.5 – 49.5	24.7	55.2
Nyang'oma	5.4 – 66.7	27.0	3.6 – 48.6	16.4	12.2 – 69.3	27.8	71.2
Nango	27.6 – 54.3	40.9	19.7 – 32.5	26.1	38.0 – 39.1	38.6	105.6

Table 5: Annual Effective Dose Rates, External (H_{ex}), Internal (H_{in}) Hazard Indices and Radium Equivalent Activity (Ra_{eq}) As Per Locations

Location	Annual Effective Dose Rate ($mSvy^{-1}$)		External Hazard Index, (H_{ex})		Internal Hazard Index, (H_{in})		Radium Equivalent Activity, (Ra_{eq}) in ($Bqkg^{-1}$)	
	Range	Average	Range	Average	Range	Average	Range	Average
Wagusu Market	0.02 – 0.26	0.11	0.04 – 0.57	0.26	0.04 – 0.64	0.33	14.1 – 212.5	96.94
Abimbo	0.03 – 0.27	0.14	0.06 – 0.62	0.31	0.06 – 0.78	0.37	20.7 – 230.3	112.02
Nyang'oma	0.07 – 0.33	0.17	0.16 – 0.80	0.37	0.18 – 1.10	0.46	58.5 – 294.5	135.15
Nango	0.21 – 0.31	0.26	0.47 – 0.71	0.59	0.60 – 0.91	0.76	175.8 – 261.5	218.63



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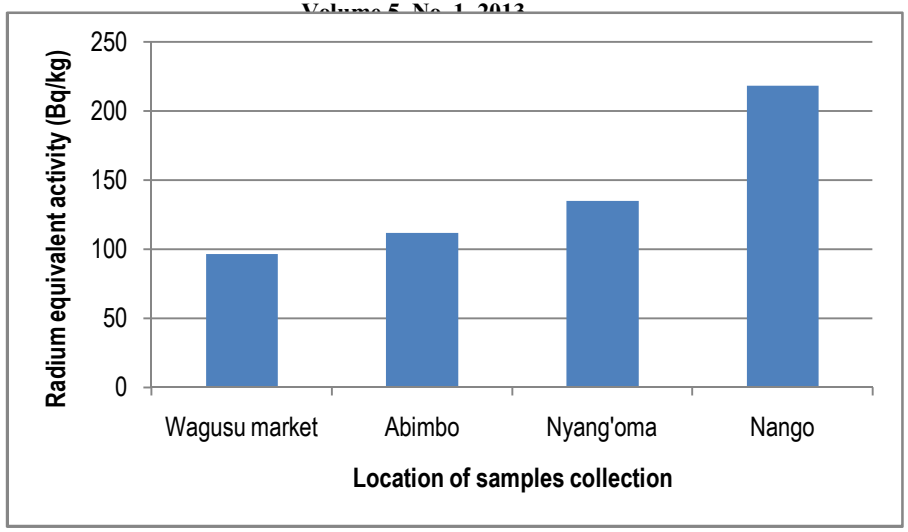

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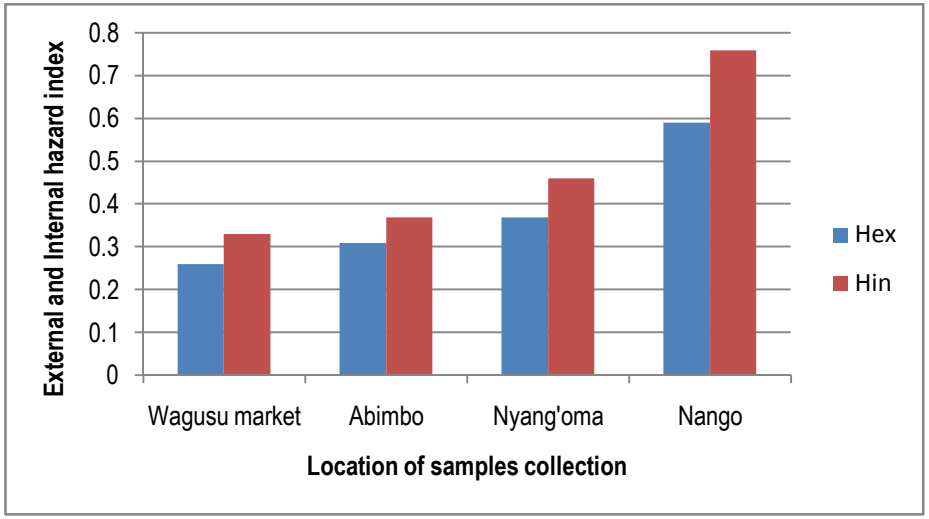

(c).

Figure 4: The Average Value of: (a) Annual Effective Dose (b) Radium Equivalent Activity (c) External and Internal Hazard Index in All the Area Under Investigation.

CONCLUSIONS

From the study, the average activity concentration for the area under study is 640.0, 44.2 and 40.3Bqkg^{-1} against the worldwide accepted limit of 400, 37 and 33Bqkg^{-1} for ^{40}K, ^{226}Ra and ^{232}Th (UNSCEAR, 2008) respectively.

The mean absorbed dose rate, measured 1m from the ground was 141.6nGyh^{-1}, while the mean calculated absorbed dose rate was 69.8nGyh^{-1}. The measured absorbed dose rate was higher than the calculated dose rate; a fact that could be attributed to the fact that calculated dose rate does not include cosmic rays These values were higher than the worldwide recommended values of 60nGyh^{-1} (UNSCEAR, 2000) and 54nGyh^{-1} (UNSCEAR, 2008) for measured and calculated absorbed dose rates respectively.

Since the average activity concentration for the area under study for ^{40}K, ^{226}Ra and ^{232}Th is more than the worldwide accepted limit and also the measured and calculated dose rate are higher than the worldwide dose rate; this area may therefore be characterized as a High Background Radiation Area (HBRA). The values of annual effective dose rate were lower than the worldwide value of 1mSvy^{-1}, which is the annual effective dose rate limit for the public exposure (ICRP, 2000).

Radium equivalent (Ra_{eq}) values for the studied areas varied from 96.9 to 218.6Bqkg^{-1}, with an average of 140.7Bqkg^{-1}. These values were lower than the internationally accepted value of 370Bqkg^{-1} (Lu Xinwei *et al*., 2006). The values of external hazard index (H_{ex}) and internal hazard index (H_{in}) are lower than acceptable limit of unity. The radiation risk from these soils can therefore be negligible and the soils and rocks here can be safely used in construction without posing any significant radiological threat to population.

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