



ASSESSMENT OF NATURAL RADIOACTIVITY LEVELS IN TAILINGS FROM LURAMBI ROSTERMAN GOLD MINE, KAKAMEGA COUNTY KENYA.

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ABSTRACT

Thirty samples were analyzed in the assessment of the activity concentration levels associated with naturally occurring radionuclides materials from Rosterman gold mine. Activity concentration with regard to gamma ray spectrometric analysis was $245 \pm 12.39 \text{ Bqkg}^{-1}$; $110 \pm 5.15 \text{ Bqkg}^{-1}$ and $84 \pm 4.23 \text{ Bqkg}^{-1}$ for ^{40}K , ^{232}Th and ^{238}U respectively. Averagely, the absorbed dose rate was $53.65 \pm 6.2 \text{ nGy}^{-1}$, the annual effective dose of $0.4 \pm 0.02 \text{ mSvy}^{-1}$ for indoor and $0.3 \pm 0.01 \text{ mSvy}^{-1}$ for outdoor were reported. The radiological parameters were averagely valued as $0.7 \pm 0.03 \text{ mSvy}^{-1}$, $0.8 \pm 0.04 \text{ mSvy}^{-1}$ and $278 \pm 13.08 \text{ Bq/Kg}$ for internal hazard index, external hazard index and radium equivalent respectively. All the radiological parameters were within the recommended permissible values. This therefore points out that gold mining at Rosterman has no significant radiological health implication on the miners and population around.

Subject Area: Nuclear and Radiation Physics

Key Words: NaI(TI) spectrometry, Radiological Parameters, Naturally Occurring Radioactive Materials.

1.0 Introduction

The radionuclides including ^{238}U , ^{232}Th , and ^{40}K are among the abundant natural radioisotopes detectable in soil and in other media of the Earth's crust (1). The associated exposure and natural radioactivity resultant from gamma radiations depend on the geographical conditions and appear at varied levels in the soil (2). Specific levels relate to the type of rock from which the soil originates. Igneous rocks like granitic and silic rocks are associated with higher radioactivity levels, also the lower levels with the sedimentary rocks except shale and phosphate rocks (3).

Cosmic radiations resulting from radiations from the sun and the stars, while terrestrial radionuclides (naturally occurring radioactive materials, NORMs) resulting from the Earth's soils and rocks are the main sources of natural radioactivity. Potassium – 40 and carbon – 14 in our bodies causes internal radiation exposure (4). Prolonged exposure to radiations often has detrimental effects on life. This results from the ionizing ability of radiation which impairs the normal functioning of cells (5). The DNA is damaged by radiations of low energy causing cellular death and mutagenesis; the process by which genetic information is modified by radiations. High radiation does kill the cells, while low radiations tend to alter genetic code (DNA) of the irradiated cells leading to birth defects and cancer diseases (6).

Mining is potentially a source of exposure to naturally occurring radioactive materials (7). Mining produces a lot of wastes (tailings or sediments), which causes pollution of soil over a large surface area, thus impacting negatively on human health and the environment (8). The decay series of ^{238}U , and ^{232}Th , together with ^{40}K radionuclide highly contribute to the rise in the concentration of NORMS in the environment. Thus, knowledge of ^{238}U and ^{232}Th decay activities and ^{40}K are useful in analysis of the absorption doses and evaluating the radiological hazards. The decay activity of these primordial radioisotopes progresses until a stable nuclide is formed (9). Use of phosphatic fertilizers, trace elements in agriculture and in medical practices are artificial activities that may lead to elevated radiations besides mining (5).

Due to the risk associated to these radiations, the international bodies like United Nations Scientific Committee on Effects of Atomic Radiations (UNSCEAR) and International

Commission on Radiation Protection (ICRP) have given the dose limit for the general public as 1 mSv annually, with a radium equivalence of 370Bq/kg (7). Extensive work has been done in many nations to assess the radiological impacts of technologically enhanced radioactivity on human and the environment. In Kenya for instance, previous radiological studies in rocks, sand, water and mining sediments show both low and high levels of environmental radioactivity (10). Most workers at the gold mine are oblivious of the problems that come with the correlation between NORM and mining. This study used NaI (TI) gamma ray detector to assess radioactivity levels in tailings. According to the radiological parameters, health implications due to exposure to the radiations resulting from the gold mine were assessed and found out to be at minimal levels.

2.0 MATERIALS AND METHODS.

2.1 Survey Area

The study is based at a gold mine situated in Western Kenya, Kakamega county. Rosterman gold mine site is located in Lurambi sub – county, 3.4 Km from Kakamega town. It is globally located at 0° 31.08 N, 34° 75.18E with approximately 420 km².

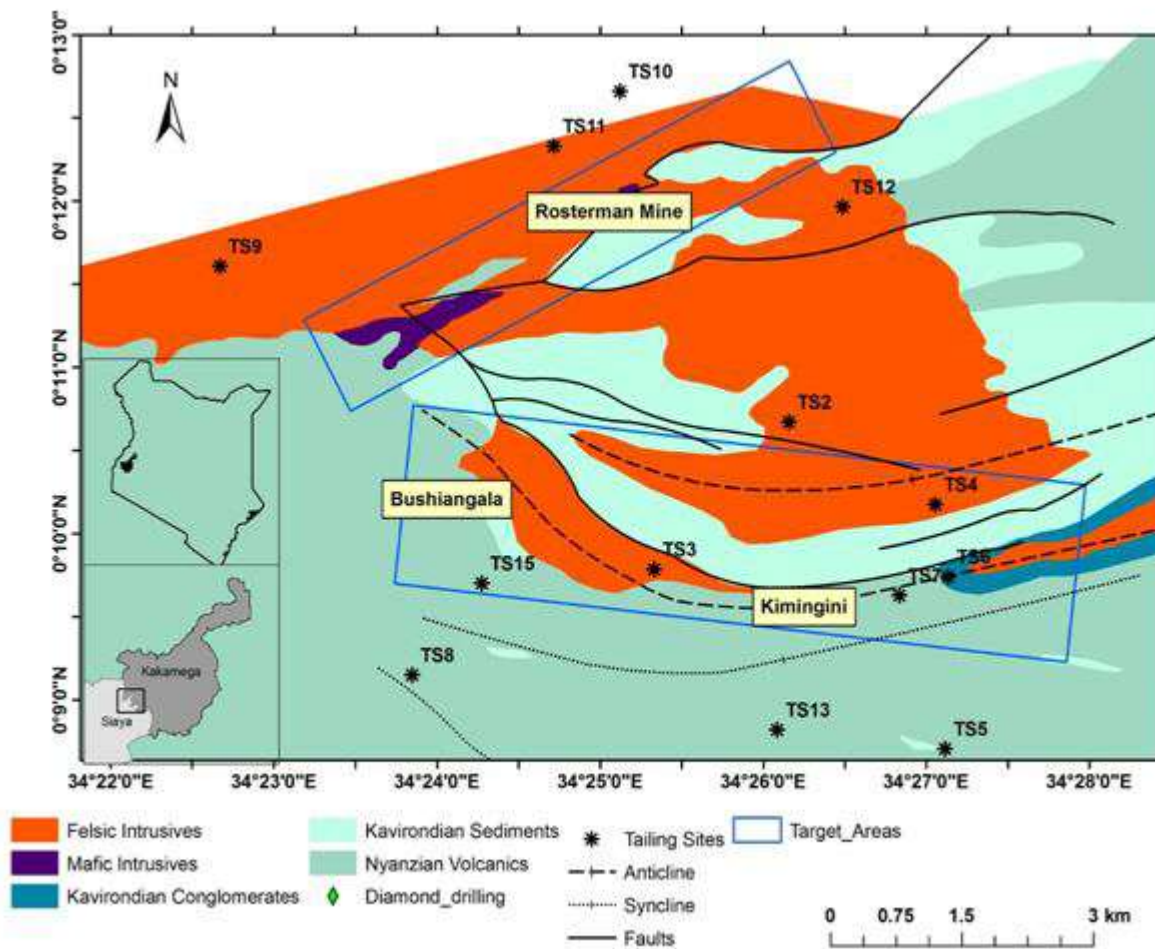


Figure. 2.1: Map of Rosterman Gold Mine Site

Approximately Lurambi has 65, 121 households. Currently, the population may have risen as a result of the increased birth rates as well as increased job opportunities because of the presence of the Rosterman gold mine.

2.2 Sample Preparation

There was sampling of thirty tailing wastes, these were randomly collected from Rosterman gold mining site, Lurambi sub – county, Kakamega county, Western Kenya. Each sample was collected at a depth of 50 cm and of net weight of 0.3 Kg. samples were then packed into 500g plastic bottles temporarily. These bottles had the following information, date of collection, sample identification number and tunnel number. These were then sealed to avoid leakage and external contamination. These samples were then dried using ovens independently and then

ground to ensure homogeneity. These samples were then sieved with a 2 mm mesh wire and then packed in 300g plastic containers. These containers were water and air tight. They were kept for a duration of at least one month in order to establish secular equilibrium among some progenies of ^{232}Th and ^{238}U series 11.

2.3 Instrument Calibration

NaI(Tl) gamma ray spectrometer was used to determine the radionuclides of interest and their corresponding abundance 12. The calibration was done by the standard materials obtained from the international atomic energy agency whose activities are 4900 Bq/Kg for RGU-1, 3280 Bq/Kg for RG Th-1 and 13400 Bq/Kg for RGK-1 13. Two tailing samples were run each day for a live time of eight minutes each. The determination of the activity concentration of ^{232}Th and ^{226}Ra was done assuming a secular equilibrium with their decay products. To determine the concentrations of ^{238}U series, the gamma energy transitions of 609.2 keV for ^{214}Bi and 351.9 keV for ^{214}Pb were used, while gamma energy transition of 911 keV of ^{228}Ac and ^{208}Tl were used to determine the concentrations of ^{232}Th series. The activity concentration of ^{40}K was determined from a photo peak of 1460keV.

3.0 RESULTS AND DISCUSSION

3.1 Activity Concentration of ^{238}U , ^{232}Th and ^{40}K in Tailing Samples

The mean activity concentrations for each tailing sample were calculated using equation 3.1, 14

$$A_c = \frac{N_D}{p \cdot \eta \cdot m} \quad (3.1)$$

Where A_c is the specific activity concentration in Bqkg^{-1} for each sample, N_D is the net count rate at energy E_γ , p is the gamma-ray emission probability for a transition at energy E_γ , η is the photo peak detection efficiency at specific gamma-ray energy E_γ and m is the mass of the tailing sample in kg.

The results for the activity concentration of the three radionuclides (^{232}U and ^{40}K) were then compared as shown in Figure 3.1 below.

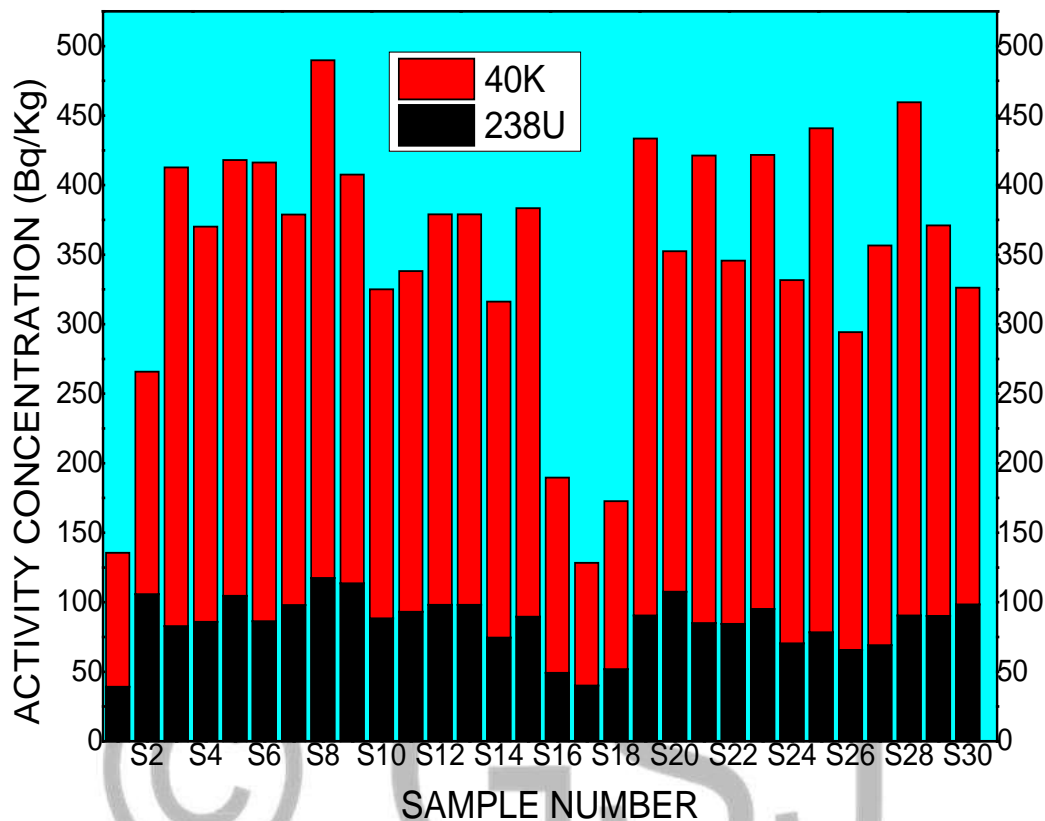


Figure 3.1: Comparison of activity concentration of ⁴⁰K (Potassium-40), and ²³⁸U (Uranium-238) in the collected tailing sample at Rosterman gold mine.

The activity concentration results from this study are presented in **Figure 3.1**. The crustal abundance of ⁴⁰K in the surveyed area was generally high compared to ²³²Th and ²³⁸U, which is a common geological occurrence in most of the crustal sediments **15**. The mean activity concentrations of ⁴⁰K, ²³²Th and ²³⁸U for the 30 tailing samples averaged were 245 ± 12.39 Bq/Kg, 110 ± 5.15 Bq/Kg and 84 ± 4.23 Bq/Kg respectively. The activity concentration of ²³⁸U and ²³²Th were above the world's average of 35 Bqkg^{-1} and 45 Bqkg^{-1} respectively, while that of ⁴⁰K was below the world's mean of 400 Bqkg^{-1} **16**. The reported averages were within the documented world's exemption levels of 1000 Bq/Kg for ²³²Th and ²³⁸U **17**.

3.2 Radium Equivalent (Ra_{eq}) in Tailing Samples

Determination of gamma-ray emission from different radionuclides of ^{238}U , ^{232}Th and ^{40}K was done using NaI(Tl) spectrometer was calculated using equation 3.2, 18.

$$Ra_{eq} = C_{Ra} + 1.423C_{Th} + 0.077C_K \quad (3.2)$$

Where C_{Ra} , C_{Th} and C_K are the specific activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K , respectively. The values for the radium equivalent are shown in figure 3.2.

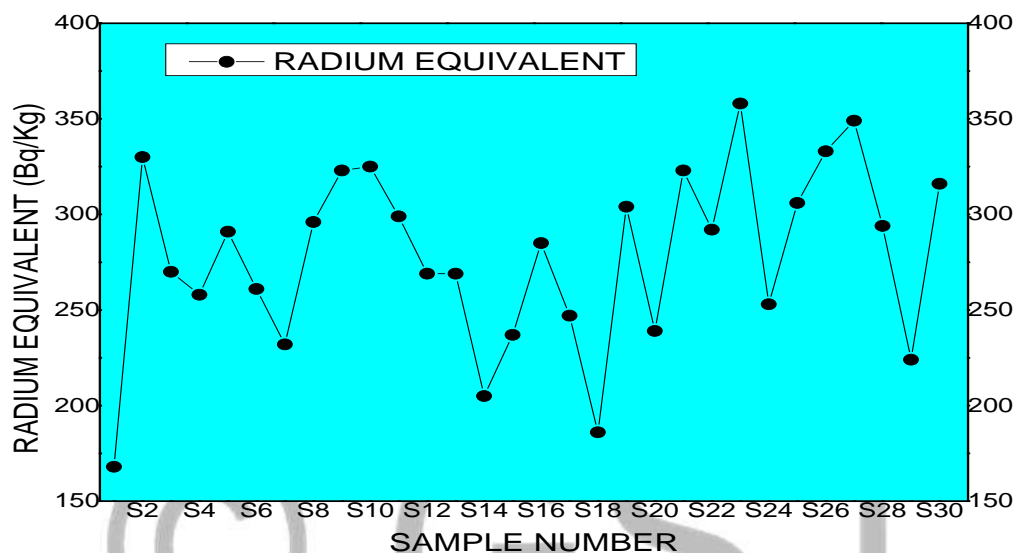


Figure 3.2: A Line Graph showing Radium Equivalent for the collected tailing samples at Rosterman gold mine.

The average and the ranges of radium equivalent in tailing samples are recorded in Table 3.1. The reported mean radium equivalent for all the tailing samples was $278 \pm 13.08 \text{ Bqkg}^{-1}$. Radium equivalent indices for all the samples were below the recommended level of 370 Bq/kg, 19, and thus the mining of gold at Rosterman has no radiological harm to the population carrying out artisanal mining.

3.3 Absorbed Dose Rate

Absorbed dose rate (D) which defines the energy absorbed per unit mass of the tissue was estimated from the activity concentration of ^{226}Ra , ^{232}Th and ^{40}K using equation 3.3 and conversion factors (nGy^{-1} per Bqkg^{-1}) of 0.427, 0.662 and 0.043 for ^{226}Ra , ^{232}Th and ^{40}K , respectively 20.

$$D(\text{nGyh}^{-1}) = 0.427C_{Ra} + 0.662C_{Th} + 0.043C_K \quad (3.3)$$

The activity to dose conversion factor of $(0.427)^{238}\text{U}$ is appropriate for ^{226}Ra since at secular equilibrium their decay rate is the same **21**.

Table 3.1: Analytical results for all the collected tailing samples.

Parameter	Range	Average
Radium Equivalent (Bq/kg)	$168 \pm 8.42 - 358 \pm 17.9$	278 ± 13.04
Absorbed dose rate (nGy/h)	$38 \pm 4.98 - 78 \pm 9.17$	53.65 ± 6.2
AEDR Outdoor (mSv/y)	$0.10 \pm 0.00 - 0.40 \pm 0.02$	0.30 ± 0.01
AEDR Indoor (mSv/y)	$0.20 \pm 0.01 - 0.50 \pm 0.02$	0.40 ± 0.02
H_{ex}	$0.50 \pm 0.02 - 1.0 \pm 0.05$	0.80 ± 0.04
H_{in}	$0.30 \pm 0.02 - 1.10 \pm 0.04$	0.70 ± 0.03

The data in **table 3.1** shows that the absorbed dose from the tailing samples varied from $38 \pm 4.98 \text{ nGy}^{-1}$ to $78 \pm 9.17 \text{ nGy}^{-1}$ with an average of $55 \pm 6.2 \text{ nGy}^{-1}$. This is less than the world's average value of 60 nGy^{-1} **22**. The rise in absorbed dose rate was due to the high concentration of ^{40}K in the tailing samples of Rosterman gold mine.

3.4 Annual Effective Dose Rate

The annual effective dose rate (AEDR) was evaluated using equation **3.4, 23**

$$E = D \times T \times Q \times 10^{-6} \tag{3.4}$$

In this case E , D , T and Q are the AEDR, dose rate, occupancy time and dose-conversion factors, respectively. UNSCEAR recommends the use of occupancy factors of 0.2 and 0.8 for calculation of the global annual mean outdoor and indoor effective dose rates, respectively. However, in Kenya, most adults spend $\sim 40\%$ of their time outdoors and 60% indoors. Therefore, the constants 0.6 and 0.4 were used as the time fractions for the indoor and outdoor occupancy factors in determination of internal and external annual effective doses. Due to the difference in fractions of time spent indoors and outdoors, equation **3.4** was modified using occupancy factors of 0.6 and 0.4 for indoor and outdoor AEDR, respectively, to give equations **3.5** and **3.6, 24**.

$$E_{in}(mSvy^{-1}) = D(nGyh^{-1}) \times 8760(hy^{-1}) \times 0.6 \times 0.7(SvGy^{-1}) \times 10^{-6} \quad (3.5)$$

$$E_{out}(mSvy^{-1}) = D(nGyh^{-1}) \times 8760(hy^{-1}) \times 0.4 \times 0.7(SvGy^{-1}) \times 10^{-6} \quad (3.6)$$

Where E_{in} and E_{out} are the indoor and outdoor AEDR, respectively, D is the absorbed dose rate, 8760 hy^{-1} is the time expressed in hours for 1 year, $0.7 \text{ (SvGy}^{-1}\text{)}$ is the dose conversion factor and 0.6 and 0.4 are the indoor and outdoor occupancy factors, respectively. To estimate effective individual's annual exposure and evaluate total risks due to radiation and radionuclides intake, the absorbed dose was converted to the effective dose (AEDR) using mathematical construct suggested by International Commission on Radiation Protection (ICRP), 16. The findings for this work report a mean indoor AEDR for the tailing samples of $0.40 \pm 0.02 \text{ mSvy}^{-1}$, which is below the world's average of 0.41 mSvy^{-1} and the corresponding permissible limit of 1 mSvy^{-1} . The average outdoor AEDR was $0.3 \pm 0.01 \text{ mSvy}^{-1}$ (Table 3.1). The mean level of indoor AEDR suggests that the mining of gold from Rosterman poses no radiological health threat to population mining and residing at the mining site 25. The mean of outdoor AEDR was below the safety limit of 1 mSvy^{-1} , suggesting that the human population interacting with the tailings are safe from harmful effects associated with elevated doses of radiations.

3.5 Internal and external hazard indices

The external hazard index H_{ex} estimates the potential radiological hazard posed by different tailing samples. It is a dimensionless quantity and a safety criterion for materials used for building construction is that $H_{ex} \leq 1$, 2. In addition to the external hazard, 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} . The calculation of internal hazard index was done using equation 3.7, 26

$$H_{in} = \frac{C_{Ra}}{185} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \leq 1 \quad (3.7)$$

Where C_{Ra} , C_{Th} and C_K are the specific activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in BqKg^{-1} respectively.

Any material with H_{in} indices more than 1mSvy^{-1} is considered deleterious. The external hazard index was estimated from $R_{a_{eq}}$ equation which caps $R_{a_{eq}}$ at 370BqKg^{-1} as given in equation 3.8, 27.

$$H_{Ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \leq 1 \quad (3.8)$$

For any material to have insignificant hazardous effect due to radiation, the H_{ex} index should be less than 1mSvy^{-1} , which is equivalent to 370BqKg^{-1} 28. The average internal and external hazard indices for the tailing samples were $0.70 \pm 0.03\text{mSvy}^{-1}$ and $0.80 \pm 0.04\text{mSvy}^{-1}$ respectively (Table 3.1). Since the recorded average values for both internal and external indices were less than a unit, mining of gold at Rosterman does not predispose the miners and the general public to harmful health effects due to direct gamma radiation from ^{40}K species and inhalation of decay daughters from ^{238}U and ^{232}Th decay series.

4.0 CONCLUSION

Radiological analysis of all the collected tailing samples has gone through evaluation. The difference in the mineral present in the individual samples is the cause of the variation in the activity concentration resultant from naturally occurring radionuclides was $^{40}\text{K} > ^{232}\text{Th} > ^{226}\text{Ra}$. Average absorbed dose rate for the tailing samples was lower than the world's reported mean of 60nGy^{-1} . Indoor and outdoor AEDRs for the samples analyzed were below the world's average of 0.41mSv/y and permissible dose constraint of 1mSv/y recommended by UNSCEAR and ICRP bodies, respectively. Radium equivalent in the samples were below 370Bq/kg , the recommended limit for a material to be considered to cause negligible health hazards. The calculation of internal and external radiation hazard indices was to determine the possible risks associated with the exposure to gamma radiation through inhalation or direct external irradiation. This was found to be within the globally acceptable range. Regarding the current results, it is possible to conclude that gold mining at Rosterman has no health hazardous effect on the miners and the general public.

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CONFLICT OF INTEREST

The authors declare no conflict of interest regarding publication of this paper.

REFERENCES

1. Uosif M.A (2007). Gamma-ray spectroscopic analysis of selected samples from the Nile river sediments in Upper Egypt. *Radiat Prot Dosim.* 123(2): 215-220.
2. Kamunda, C., Mathuthu, M., and Madhuku, M. (2016). An Assessment of Radiological Hazards from Gold Mine Tailings in the Province of Gauteng in South Africa. *International Journal of Environmental Research and Public Health.* 13(1): 138.
3. Faanu A., Adukpo O.K., Tettey-Larbi, Lawluvi H., Kpeglo D.O., Darko E.O., Emi-Reynolds G., Awudu R.A., Kansaana C., Amoah, P.A., Efa A.O., Ali I.D., Agyeman B., Agyeman L., and Kpodzro R. (2016). Natural radioactivity levels in soils, rocks, and water at a mining concession of Perseus gold mine and surrounding towns in the central region of Ghana. *Springer plus* 5:98.
4. UNSCEAR, (2008). United Nations Scientific Committee on the effects of atomic radiation, sources, and effects of ionizing radiation. Report to General Assembly, with Scientific Annexes United Nations. United Nations, New York.
5. UNSCEAR (2000). United Nations Scientific Committee on the effects of atomic radiation, sources, and effects of ionizing radiation. Report to General Assembly, with Scientific Annexes United Nations. United Nations, New York.
6. Ajithra, A., and Shanthi, G. (2016). Assessment of Beach sand using Gamma Ray Spectrometer in Thiruvananthapuram District, Kerala of South India. *IRA-International Journal of Technology & Engineering* (ISSN 2455-4480), 5(3), 56-69.
7. Grasty, R.L. (1977). A General Calibration Procedure for Gamma-ray Spectrometers - Project 720084; in Report of Activities, part C; Geol. Survey of Canada, Paper 77-1C.
8. Kebwaro, M. J., Rathore, S.V., Hashim, N.O., & Mustapha, A.O. (2011). Radiometric assessment of natural radioactivity levels around Mrima Hill, Kenya. *International Journal of the physical sciences.* 6(13): 3105-3110.

- 9 Otwoma, D., Patel, J.P., Bartilol, S., and Mustapha, O.A. (2013). Estimation of annual effective dose and radiation hazards due to natural radionuclides in mount Homa, southwestern Kenya. *Radiation protection dosimetry*. 155(4): 497-504.
10. Hashim, N. O., Rathore, I. V. S., Kinyua, A. M., & Mustapha, A. O. (2004). Natural and artificial radioactivity levels in sediments along the Kenyan coast. *Radiation physics and chemistry*, 3(71), 805-806.
11. Achola, S. O., Patel, J. P., Angeyo, H. K., & Mustapha, A. O. (2010). Natural radioactivity and associated radiation characteristic of the new high background radiation area of lambwe east southern Kenya
12. Tzortzis, M., Tsertos, H., Christofides, S., & Christodoulides, G. (2003). Gamma-ray measurements of naturally occurring radioactive samples from Cyprus characteristic geological rocks. *Radiation Measurements*, 37(3), 221-229.
13. IAEA. (2010). Handbook of parameter values for the prediction of radionuclide transfer in terrestrial and freshwater environments: International Atomic Energy Agency.
14. El-Taher, A., & Al-Zahrani, J. (2014). Radioactivity measurements and radiation dose assessments in soil of Al-Qassim region, Saudi Arabia. The vicinity of chirano gold mine in Ghana. *Radiant prot Dosim*. 158(1):87-99.
15. Joshua, E. O., Ademola, J. A., Apanowo, M. A. and Oloronde, D. O. *Natural radionuclides and hazard of rock samples collected from south eastern Nigeria*. *J. Radiat. Meas.* **44**, 401–404 (2009).
16. International Commission on Radiological Protection. Occupational Intakes of Radionuclides: Part 1. (Oxford: ICRP; Publication) p. 130 (2015).
- 17 IAEA. (1996). International basic safety standards for protection against ionizing radiation and for the safety of radiation sources, safety series no. 115. IAEA, Vienna.
18. Bendibbie M. M., David M. M and Jayanti P. P. (2013). Radiological analysis for suitability of Kitui south limestone for use as a building material. *International Journal of Fundamental Physical Sciences*. 3:32-35.
19. International Commission on Radiological Protection. (ICRP), Radiation Protection Dosimetry. ICRP 2007 Recommendations. Vol. **37**. (Oxford: Pentagon Press. ICRP Publication 103. Ann. ICRP) pp. 2–4 (2007).

20. ICRP (2007) 2006 recommendations of the International Commission on Radiological Protection. ICRP publication no.103, Pergamon Press, Oxford.
21. Darko EO, Faanu A, Razak A, Emi-Reynolds G, Yeboah J, Oppon OC, Akaho EHK (2010) Public exposure hazards associated with natural radioactivity in open-pit mining in Ghana. *Radiat Prot Dosim* 138(1):45–51
22. Xinwei, L., Lingqing, W., Xiaodan, J., Leipeng, Y., & Gelian, D. J. R. p. d. (2005). Specific activity and hazards of Archeozoic-Cambrian rock samples collected from the Weibei area of Shaanxi, China. *118(3)*, 352-359.
23. Faanu A., Lawluvi H., Kpeglo D.O., Darko E.O., Emi-Reynolds G., Awudu R., Adukpo O.K., Kansaana C., Ali I.D., Agyeman B., Agyeman L., and Kpodzro R. (2013). Assessment of natural and anthropogenic radioactivity levels in soils, rocks, and water in the vicinity of chirano gold mine in Ghana. *Radiant prot Dosim.* 158(1):87-99.
24. Odumo, B. O., Nanos, N., Carbonell, G., Torrijos, M., Patel, J. P., and Rodríguez Martín, J. A. (2018). Artisanal gold-mining in a rural environment: Land degradation in Kenya. *Land Degradation and Development.* 29(10): 3285–3293.
25. UNSCEAR, (2008). United Nations Scientific Committee on the effects of atomic radiation, sources, and effects of ionizing radiation. Report to General Assembly, with Scientific Annexes United Nations. United Nations, New York.
26. Faanu A., Adukpo O.K., Tettey-Larbi, Lawluvi H., Kpeglo D.O., Darko E.O., Emi-Reynolds G., Awudu R.A., Kansaana C., Amoah, P.A., Efa A.O., Ali I.D., Agyeman B., Agyeman L., and Kpodzro R. (2016). Natural radioactivity levels in soils, rocks, and water at a mining concession of Perseus gold mine and surrounding towns in the central region of Ghana. *Springer plus* 5:98.
27. OECD. Exposure to radiation from the natural radioactivity in building materials. In: Reported by a Group of Experts of the OECD. (Paris, France: Nuclear Energy Agency) (1979).
28. ICRP. (2015). Occupational Intakes of Radionuclides: Part 1. (Oxford Pentagon press: ICRP; Publication) pp. 130