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ESTIMATION OF RADIOLOGICAL HAZARDS DUE TO NATURAL RADIONUCLIDES FROM THE ROSTERMAN GOLD MINE TAILINGS, LURAMBI, KAKAMEGA, KENYA

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Radiological hazards associated with naturally occurring radionuclides in materials from Rosterman gold mine were assessed by analysis of 30 samples. The gamma-ray spectrometric analysis of tailing samples reported an average activity concentration of 263 ± 13, 123 ± 6 and 84 ± 4 Bq kg^{-1} for ^{40}K, ^{232}Th and ^{226}Ra, respectively. The average absorbed dose rate was 124 ± 6 nGy h^{-1}, while the annual effective dose of 0.4 ± 0.02 mSv y^{-1} for indoor and 0.3 ± 0.01 mSv y^{-1} for outdoor were reported. The mean and range of radiological parameters (external and radium equivalent) calculated from the tailing samples were within the permissible limits and hence mining of gold at Rosterman has no significant radiological health implication on the miners and the surrounding population.

INTRODUCTION

At present, mainly radionuclides whose half-life is comparable to the age of the earth exists in varying levels depending on region, meteorological, geological and other physical factors[1]. The omnipresence of naturally occurring radioactive element means that the human race has been exposed to ionising radiations from natural sources[2]. Major contributions to radiation exposure result from terrestrial materials like rocks, sand, sediments, and water, soil and mining wastes[3]. Low background radiation doses from terrestrial sources have not been reported as generating significant clinical effects on human life. However, a concern arises when the levels of radiation surpass recommended thresholds by radiation regulatory bodies.

Human activities like mining and natural calamities such as earthquakes have been known to enhance levels of natural radioactivity by exposing the underground matter, which is usually enriched with radionuclides which have been illuviated owing to the solubility of most radionuclides[4].

Mining results in extraction and disposal of large quantities of wastes that mostly contain radionuclides belonging to ^{232}Th, ^{238}U decay progressions and ^{40}K transition. The miners are at risk of radiation exposure when they inhale radon and thoron gases or their short-lived decay products encapsulated in air-bone dust[5]. There is also a potential risk of exposure of miners to radiation doses during excavation, extraction, transportation and processing of minerals such as gold since some miners spend most of their time in the mining tunnels underground more so for the artisanal mining. The general public may be exposed externally or internally to radiation doses through interaction with mine wastes like water and rubbles collected and used for construction[6,7]. Areas whose total annual effective dose exceeds a dose limit of 100 mSv y^{-1} have reported biological anomalies such as myeloid Leukemia, a form of blood cancer, stomach cancer, lung cancer and chromosomal aberrations as a result of human interaction with radiations of high energy. Further, the evidence of stochastic effects due to radiation exposure was revealed by survivors of the Hiroshima atomic bomb, who were exposed to radiation doses greater than 100 mSv[8].

Studies to assess the radiological impact due to technologically enhanced radioactivity on human and the environment have been done extensively. In Kenya, for example, previous radiological studies in rocks, sand, water and mining sediments report both low and enhanced levels of environmental radioactivity[3,6]. Most workers at the artisan gold mining sections and the general public are not aware of the potential harm as a result of radiation exposure despite a strong correlation between NORM and mining.

This radiological survey was meant to establish the current level of natural radionuclides and evaluate the corresponding radiological hazards in Rosterman gold mine. The study was done using calibrated NaI (TI) gamma-ray spectrometer. The relevant

radiological parameters, the possible health implications to the concerned and the immediate population as a result of mining in this quarry have been evaluated.

MATERIALS AND METHODS

Survey area

The studied area is a gold mine situated in Western Kenya, Kakamega County. The area has a diversity of ancient rocks including felsic and mafic intrusive with Kavirondian conglomerates within the neighbourhood. The Rosterman gold mine site is situated in Lurambi Sub-County, Kakamega municipality and 3.4 km from Kakamega town. Lurambi Sub County is one of the elective constituencies in Kenya situated in Kakamega County. It is bordered by Ikolomani constituency to the south and globally located at 0^0 12′0″ N, 34°25′0″ E with ∼420 km^2. The population projection of Lurambi Sub County was estimated at 297 394 people according to the 2019 population census. The number of houses in Lurambi Sub County is approximated at 65 121 households[9]. Figure 1 shows the map of the studied area within Lurambi Sub County. The points marked TS represents the points where the mining by-products were collected. The artisanal miner's deposits large rock heaps after mechanical stripping of the gold vein off its parent rock. The loose irregular rocks whose size ranges from concrete to rubble masonry from underneath layers are deserted on the earth surface attracting residents who collect them and shapes them for both domestic and commercial purposes. The rocks (tailings) have been on high demand for internal and external house cladding, flooring and walling.

Sample collection and preparation

A total of 30 tailing waste samples were collected randomly up to a maximum depth of 50 cm from Rosterman gold mining site, Lurambi Sub-County, Kakamega County, Western Kenya. The raw samples were temporarily packed into 500 g plastic bottles well coded with the date of collection, sample identification number and tunnel number. The tailings were ground using electronic pulverizer and sieved through a 2 mm mesh wire to ensure homogeneity of the radionuclides present and then ovendried independently for 24 h. A mass of 0.3 kg of each sample was accurately transferred into thick standard bottles of dimension identical to those holding the standard samples to maintain counting geometry. Aluminium foil was spread to cover the mouth of each plastic bottle holding the prepared sample. A tight-fitting cork was used to seal the contents before making rounds of vinyl tape around the cork to avoid leakage of radon gas.

The prepared samples were stacked in a carton and kept for 28 days to establish secular equilibrium between ^{226}Ra and its decay progenies.

Instrument calibration

NaI (TI) gamma-ray spectrometer was used to identify the radionuclides of interest and their corresponding abundance. The detector was calibrated for energy using RGU-1, RGTh-1 and RGK-1 standard samples from the international atomic energy agency whose activity concentration in Bq kg^{-1} are 4900, 3280 and 13400, respectively[10]. Assuming ^{226}Ra and ^{222}Rn had attained secular equilibrium in each sample, two tailing samples were run per day for a live time of 30 000 s each for proper statistics. ^{226}Ra (^{228}U) was identified from the gamma lines of ^{214}Pb (351 keV) and ^{214}Bi (609 keV) and quantified from their corresponding channel contents (intensities). ^{232}Th species were identified from spectral data at energies 238.6 keV (^{212}Pb) and 911 keV (^{228}Ac). The ^{40}K was identified and quantified from its single photopeak of 1460 keV.

RESULTS AND DISCUSSION

Activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in tailing samples

The mean activity concentrations for each tailing sample were calculated using equation 3.1[11]

$$A_c = \frac{N_D}{p.\eta.m} \tag{3.1}$$

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

Figure 2 is a presentation of the activity concentration reported by this work. The varying levels of ^{40}K, ^{238}U and ^{232}Th demonstrate the non-uniform spatial distribution of natural radionuclides in the earth's crust. The crustal abundance of ^{40}K in the surveyed area was generally high compared to ^{232}Th and ^{238}U, which is a common geological occurrence in most of the crustal matter[12]. The mean activity concentrations of ^{40}K, ^{232}Th and ^{238}U for the 30 tailing samples were 264 ± 13, 123 ± 6 and 84 ± 4 Bq kg^{-1}, respectively.

The activity concentration of ^{238}U and ^{232}Th surpassed the world's average of 35 and 45 Bq kg^{-1}, respectively, while that of ^{40}K was below the world's

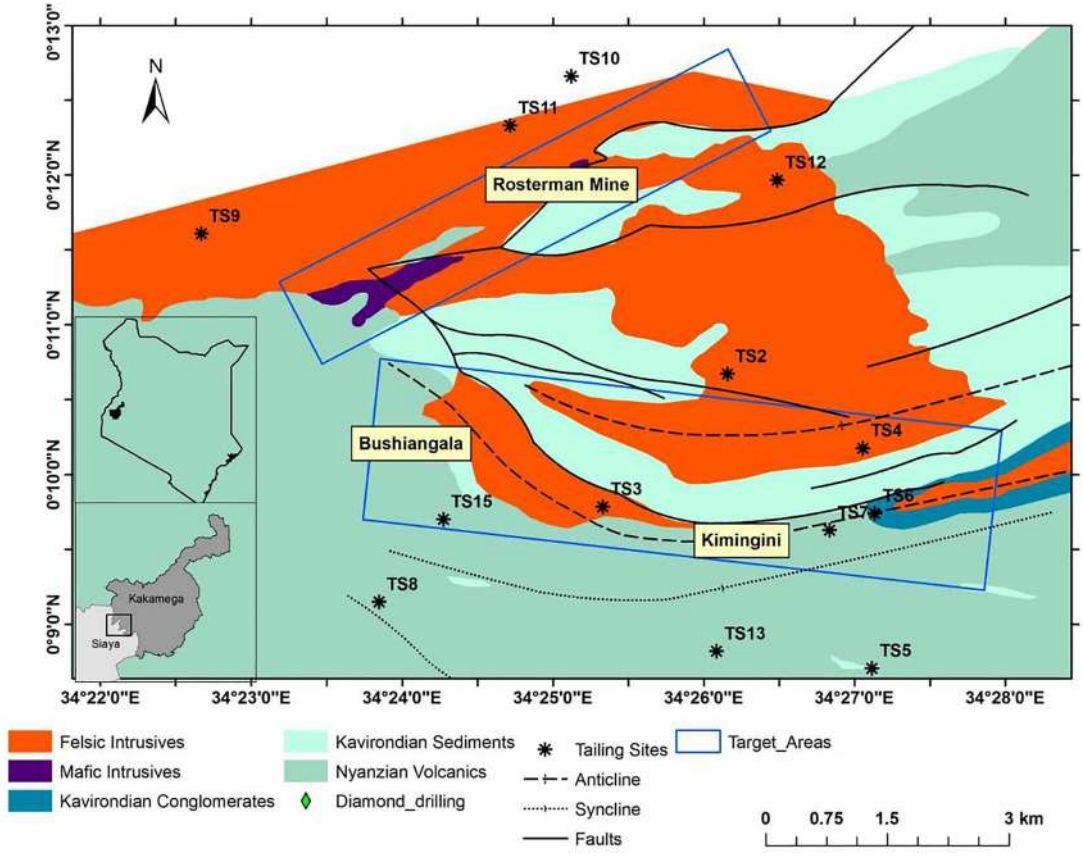

Figure 1. Map of Rosterman Gold Mine Site

mean of 400 Bq kg^{-1} [8]. The reported activity concentration averages were within the documented world's exemption levels of 1000 Bq kg^{-1}for ^{232}Th and ^{238}U[13].

Radium equivalent (Ra_{eq}) in tailing samples

The detection of gamma-ray emission from different radionuclides; ^{238}U, ^{232}Th and ^{40}K was done using NaI (TI) spectrometer. Activity concentration from tailings was converted to radium equivalent using model 3.2 below[14].

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

where C_{Ra}, C_{Th} and C_{K} are the specific activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively. The average and the range of radium equivalent in tailing samples are presented in Table 1. The radium equivalent for tailing samples averaged 280 ± 14 Bq kg^{-1}. Radium equivalent from this work was below the recommended upper level of 370 Bq kg^{-1} and thus the gold mining at Rosterman does not pose significant radiological harm to the population carrying out artisanal mining[8].

Absorbed dose rate

Absorbed dose rate (D) in the air at 1 m above the ground was calculated to provide the characteristic of external gamma radiation from terrestrial sources. The model used to estimate this dose is shown in equation 3.3[8].

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

where 0.427, 0.662 and 0.043 are the activity to dose conversion factors (nGy h^{-1} per Bq kg^{-1}) for ^{226}Ra, ^{232}Th and ^{40}K, respectively. The activity to a dose

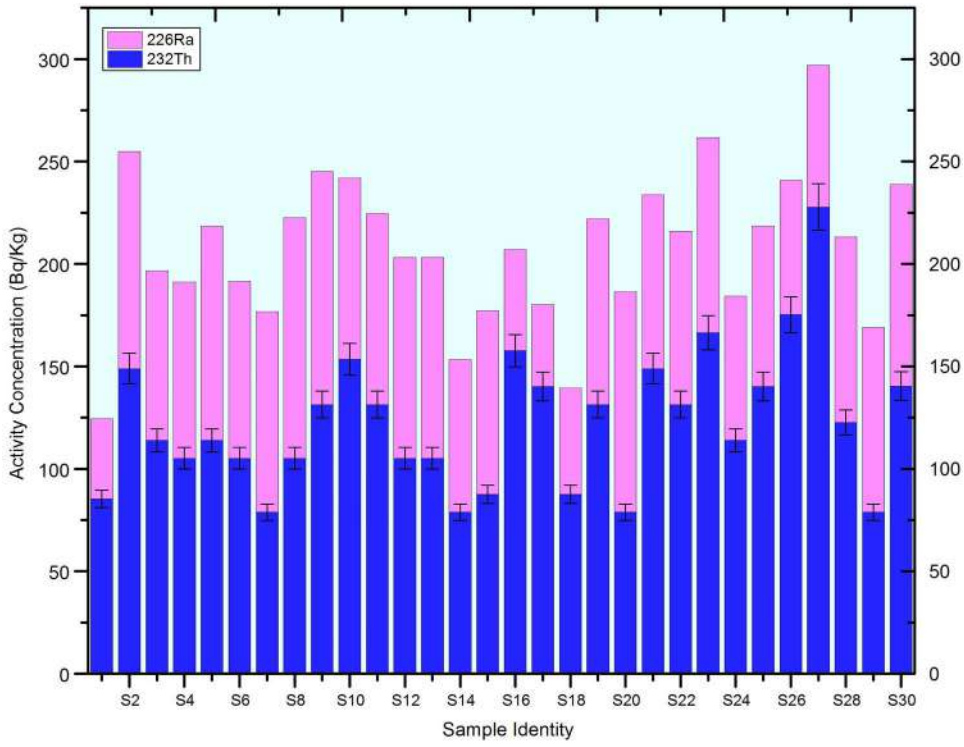

Figure 2. Activity concentration of ^{226}Ra and ^{232}Th in the tailing samples from Rosterman gold mine

Table 1. Analytical results for various parameters for tailing samples collected.

Parameter	Range	Average
Ra_{eq} (Bq kg^{-1})	168 ± 8–333 ± 16	280 ± 14
Absorbed dose rate (nGy h^{-1})	99 ± 5–183 ± 9	124 ± 6
AEDR Outdoor (mSv y^{-1})	0.10 ± 0.00–0.40 ± 0.02	0.30 ± 0.01
AEDR Indoor (mSv y^{-1})	0.20 ± 0.01–0.60 ± 0.03	0.40 ± 0.02
H_{ex}	0.50 ± 0.02–1.3 ± 0.05	0.90 ± 0.04

conversion factor of (0.427) ^{238}U is appropriate for ^{226}Ra since at secular equilibrium their activities are equal.

The data in Table 1 show that the absorbed dose from the tailing samples varied from 99 ± 5 to 183 ± 9 nGy h^{-1} with an average of 124 ± 6 nGy h^{-1}. This was twice the world's average value of 60 nGy h^{-1} [8].

Annual effective dose rate

The annual effective dose rate (E) was evaluated using equation 3.4[8]

$$E = A \times B \times C \times 8760 \times 10^{-6} \quad (3.4)$$

where A (Sv/Gy) is used in calculation of E from air kerma, B is the activity concentration in Bq/kg, C is the value of nGy/h/Bq/kg used to convert the concentration to an air kerma for a chosen geometry, 8760 converts from nGy/h to nGy/annum based on 365 days/annum and the factor 10^{-6} to converts the resulting units from nano to milli.

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 ~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 the determination of internal and external

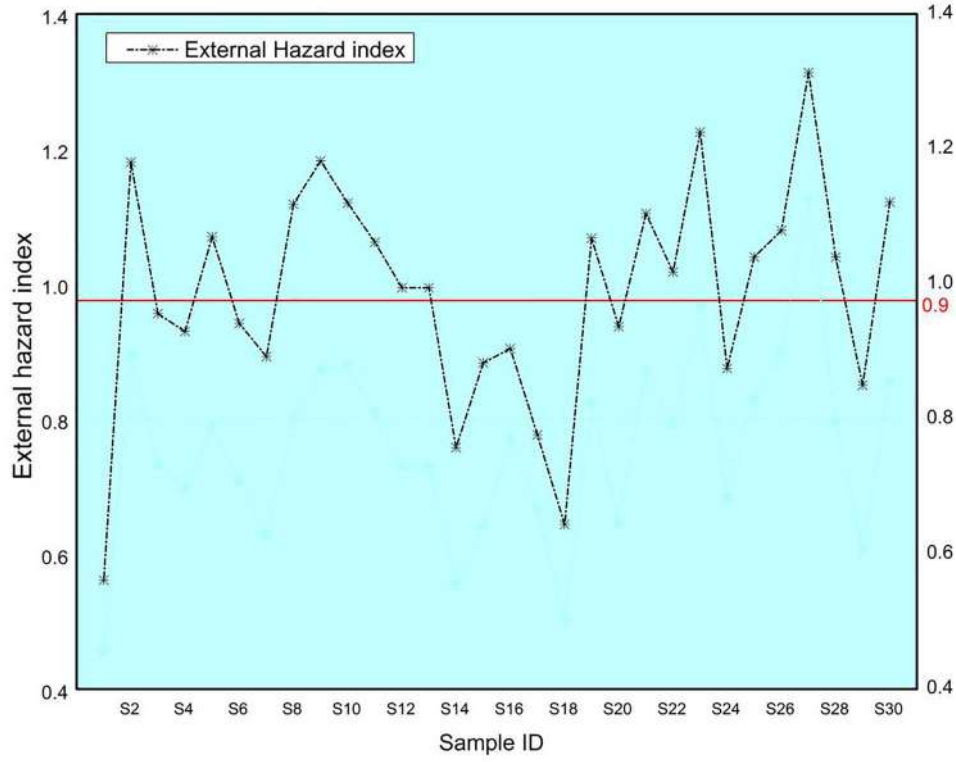

Figure 3. External hazard indices for the sampled tailings from Rosterman gold mine

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[15].

$$E_{in}\left(mSv\ y^{-1}\right) = D\left(nGy\ h^{-1}\right) \times 8760\left(h\ y^{-1}\right) \times 0.6 \times 0.7\left(Sv\ Gy^{-1}\right) \times 10^{-6} \quad (3.5)$$

$$E_{out}\left(mSv\ y^{-1}\right) = D\left(nGy\ h^{-1}\right) \times 8760\left(h\ y^{-1}\right) \times 0.4 \times 0.7\left(Sv\ Gy^{-1}\right) \times 10^{-6} \quad (3.6)$$

where E_{in} and E_{out} are the indoor and outdoor AEDR, respectively, D is the absorbed dose rate, 8760 h y^{-1} is the time expressed in hours for 1 year, 0.7 (Sv Gy^{-1}) 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[16]. The findings from this work report a mean indoor AEDR for the tailing samples of 0.40 ± 0.02 mSv y^{-1}, which is below the world's average of 0.41 mSv y^{-1} and the corresponding permissible limit of 1 mSv y^{-1}. The average outdoor AEDR was 0.3 ± 0.01 mSv y^{-1} (Table 1). The mean level of indoor AEDR is less than the recommended limit suggesting the mining of gold from Rosterman poses a negligible radiological health threat to population mining and the population residing at the mining site[17]. The mean outdoor AEDR was below the safety limit of 1 mSv y^{-1}, suggesting that the human population interacting with the tailings are safe from harmful effects due to external gamma radiation associated with elevated doses of radiations.

External hazard index

Radon and its decay daughters can be a radiolabel hazard to human health. The external hazard index was estimated from Ra_{eq} equation which caps Ra_{eq} at 370 Bq kg^{-1} as given in equation 3.7[18]. External hazard index (H_{ex}) is used to determine the potential danger due to external gamma-ray and it is useful for

Table 2. Summary statistics for the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in the tailing samples from Rosterman gold mine.

Variables	Activity concentration (Bq kg^{-1})		
	^{238}U	^{232}Th	^{40}K
Maximum	117 ± 6	227 ± 11	369 ± 19
Minimum	39 ± 2	78 ± 4	88 ± 4
Median	51	87	244
Mean	84	123	264
Standard Deviation	54	44	82

Table 3. Comparison of activity concentrations in tailings at Rosterman gold mine with other radiometric surveys.

Region	Mean activity concentration (Bq kg^{-1})			Reference
	^{238}U	^{232}Th	^{40}K	
Kenya	84 ± 4	123 ± 6	263 ± 13	Present study
Kenya	21 ± 1	49 ± 3	782 ± 39	(20)
Kenya	27 ± 2	60 ± 9	112 ± 12	(21)
South Africa	17 ± 0.4	22 ± 0.5	496 ± 15	(2)
Nigeria	69 ± 3	63 ± 3	487 ± 24	(12)
World wide	33	45	420	(8)

the characterisation of building materials.

$$H_{Ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \leq 1 \qquad (3.7)$$

where C_{Ra}, C_{Th} and C_K are the specific activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1}, respectively. For any material to have an insignificant hazardous effect due to external irradiation, the H_{ex} index should be <1, which is equivalent to the maximum permissible limit of 370 Bq kg^{-1} [19].

The mean external hazard indices for the tailing samples were 0.90 ± 0.04 (Table 1). The mean level of the external hazard index is shown by the red horizontal strike at 0.8 in Fig. 3. The figure further shows that the external index varied for each sample.

Since the reported average for external hazard index was less than a unit (Fig. 3), use of rocks from Rosterman for construction purposes does not predispose the residents and the general public to significant harmful health effects due to direct gamma radiations.

Descriptive data for the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in tailing samples

To ease the interpretation of the activity statistics, the entire data on specific activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in tailing samples were summarised and expressed in a tabular form. Table 2 shows a presentation of measures of spread and central tendency evaluated. The mean values for dose rates and the radiation hazards were in reasonable agreement with the reported values, thus mining of gold at Rosterman has insignificant radiological implication on the health of miners and the general public. Table 3 presents the mean for the current work compared with radiometric surveys by other scholars.

CONCLUSION

Radiological analysis of all the tailing samples and subsequent evaluation points out that, generally, the variation in the activity concentration of the tailing samples was attributed to the differences in the minerals present in the individual samples. The absorbed dose rate for the tailing averaged 124 ± 6 nGy h^{-1}, which was higher than the world's reported mean of 60 nGy h^{-1}. The major contribution to the absorbed dose was due to the high ^{40}K concentration in the tailing samples. Indoor and outdoor AEDRs for the samples analysed were below the world's average of 0.41 mSv y^{-1} and the maximum dose constraint of 1 mSv y^{-1} recommended by ICRP. Radium equivalent in the samples was below 370 Bq kg^{-1}, the recommended maximum limit for a material to be considered to contribute negligible

health hazards. The possible risks associated with exposure to gamma radiation through direct external irradiation were examined by calculating the external radiation hazard index, which was found to be within the global acceptable range. Based on the present results, gold mining at Rosterman on radiological context is not a threat to the health of those mining or the general public in the surrounding environment.

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CONFLICT OF INTEREST

The authors have none to declare.

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