Banzi et al., Expert Opin Environ Biol 2016, 5:4
DOI: 10.4172/2325-9655.1000140

Expert Opinion on Environmental Biology

Research Article

A SCITECHNOL JOURNAL

Assessment of Natural Radioactivity in Soil and its Contribution to Population Exposure in the Vicinity of Mkuju River Uranium Project in Tanzania

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Abstract

The level of ^{226}Ra, ^{232}Th and ^{40}K was established in soil collected in an area about 1,300 km^2 in the vicinity of Mkuju uranium project in Tanzania and selected points in the concession using HPGe spectrometer and associated exposure risks to population were estimated. The radioactivity concentration in the vicinity range ($Bqkg^{-1}$) for ^{226}Ra (24.94 to 53.50), ^{232}Th (20.86 to 47.14), and ^{40}K (344.50 to 697.54). Whereas, radioactivity in the concession range for ^{226}Ra (2430.00 to 4200.00), ^{232}Th (130.00 to 220.00), and ^{40}K (1293.30 to 1466.10). On average the radioactivity in the concession were about 89, 5 and 3 times high than those in the vicinity. Based on the measurements, the hazards attributable to use of soil from the vicinity for different purposes is marginal. However, the hazards indices indicate that soils from the concession have relatively a significantly risk by range of factors about 27 to 29 and 40 to 42 times high than in the vicinity for the external and internal hazards, respectively. Also the radium equivalent radioactivity, dose rate and effective dose tend to be significantly high in the concession than in the vicinity by range of factors from 27 to 29, 23 to 25 and 23 to 25, respectively. These findings reveals that soils in the vicinity of the project could pose less radiological risk to human health when compared to soil in the concession.

Keywords

Baseline; Radioactivity; Mkuju river; Uranium mining; Tanzania

Introduction

In recognition of the historic of absence of good environmental practices in uranium mining and processing and the associated impacts, new supplies of uranium for the nuclear renaissance will require preparation of an Environmental Impact Assessment (EIA) prior to project approval. This requirement is necessary since regardless of techniques used for extraction, uranium mining operations generate substantial waste in the form of rock, airborne dust, liquid effluence and radon gas all of which have negative environmental impact [1-9]. In contrast with underground mining, it is generally predicted that open pit which is going to be the method of uranium extraction at Mkuju River Project (MRP) could result to large quantities of mine wastes with the potential for adverse impacts to aquatic and terrestrial life in the vicinity of the project. According to USEPA [10], the extent of environmental effects associated with open pit method depend less on the grade of ore and more on depth of the uranium ore in the host rock. Since waste rock is obtained from areas rich in uranium ore, the waste rock usually contains high concentrations of radionuclides from the uranium-238 series; thorium-232 series and potassium-40 compared to normal rock. Therefore the EIA must assess the potential radiological impacts to the public for external exposure of gamma rays due to contaminated soils or internal exposure via the contaminated food chain. For these reasons, different countries including Tanzania have formulated national regulations pertinent to uranium exploration, mining, milling and the associated waste management to ensure that uranium mining activities result in minimal environmental pollution as well as to manage risks of public exposure to radioactivity [11-16]. Consistent with these regulations, it is recognized that effective monitoring of compliance requires knowledge of site characterization of radioactivity levels before the commencement of uranium mining as well as during the operation of the mine [17].

The radiological effect associated with uranium mining is not local since radioactive dust particles are released into air, depending on source emission rate and source characteristics, meteorological (wind speed and direction, turbulence, temperature, pressure and humidity), topographical (surface roughness), particle (size, form and density) parameters [18-21]. As a result of dilution, the dust particle flux from an open pit mine decreases from the edge of the source to a distance downwind. Large particles deposit within the mine perimeter while health effects in the neighbourhood from airborne particles would be associated with the flux of smaller particles. These particulate releases are often controlled by use of large amounts of water to suppress dust emission during mining. However, from past experience, this approach was not free of environmental radiological impact since the water used for this purpose discharged onto rivers and creeks [22-25]. In some cases, it becomes necessary to pump out water that collects in the mine that hinders operations in the mining. This water has potential for environmental pollution when it is released into surface water that can be a pathway to plants, animals and the human via food chain if contaminated water is used for agriculture. In other cases, if contaminated process is released into ground water it can cause similar environmental pollution and radiological effects.

The milling as a process of recovery (extracting) of a small percentage of yellowcake from the massive amount of uranium ore could be a more detrimental source of environmental pollution and human exposure to radioactivity than mining. In this process, the uranium ore is crushed and ground to produce fine powder for extraction of uranium ore using solvent or ion exchange techniques and the remaining large volume in this process is waste called tailings [1]. As a result of ventilation systems to minimize the exposure of workers to ionizing radiation inside the mill, radon and airborne dust carrying radioactivity were released into the atmosphere through milling stacks [26]. For this reason uranium milling could cause more environmental pollution and exposure to humans from radioactivity

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Received: November 09, 2016 **Accepted:** December 05, 2016 **Published:** December 09, 2016

SciTechnol
International Publisher of Science, Technology and Medicine

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than mining for three reasons. First, since the pollutants are discharged at elevated stack height, pollution at distant locations from the mine becomes more pronounced than pollution from ground level releases. Second, the discharge rates of pollutants from the stack of the uranium mill are higher than the emission rate from the uranium extraction. Third, the tailings produced from milling are more highly concentrated than tailings from mining and therefore have higher potential for environmental pollution. As discussed earlier, drift distance and the health effects associated with the airborne dust released from the mill depend on the source term; environmental parameters and topographical characteristics [18,20,21]. From the above and this description, effective assessment of compliance with the national mining regulations of radioactive ore in the MRP requires establishment of baseline environmental radioactivity data in areas with potential to experience environmental pollution and human exposure to radioactivity before uranium extraction at MRP commences. Thus, the aim of this work is to identify the area that would be affected and establish the required baseline data for compliance with the national regulations [13,14].

Materials and Methods

Location of MRP and study area

The MRP shown in Figure 1 is a large scale, uranium development project located in the Namtumbo district in the Ruvuma region between latitudes 9° 59′ 50″ to 10° 07′ 15″ S, and longitudes 36° 30′ 00″ to 36° 37′55″ E. This area hosts a viable uranium deposit of sandstone type about 25,200 tU, with an estimated production of 1,600 tU in a year at its maximum capacity over a minimum of 12 years [27]. Since the uranium ore occurs at shallow depths, open-pit methodology employing heavy mining machinery such as drills, shovels and trucks will be used. With this method, it was estimated that large amount of waste rock piles could be produced in the concession area during open pit mining. Through wind and water erosions contents of waste piles could be potential to access the neighbourhood. Thus the study area with high probability to be affected by by-products of project activities was estimated using AERMOD dispersion model developed by US EPA as described previously [28] an area about 1300 km^2 around the project's boundary

The study area estimated above includes populated villages of Mandela, Mtonya, Mfuate and Likuyu-Sekamaganga located in Namtumbo district, and Selous wildlife World heritage. Permanent residents of the populated villages approximately 54 km from the MRP at present was estimated to number about 10,000; however, the population may increase seasonally to provide a workforce for various activities undertaken on the project. The area is characterized by rainy seasons which commence in January and end in April with an average annual rainfall of approximately 70 mm with temperatures ranging from 11 to 29°C, and a dry season which begins in May and ends in December with temperatures ranging from 14 to 37°C. The average wind is 6 ms^{-1} with maximum (NE and N) and minimum (SW and S) speeds used to demarcate the study area were about 13 ms^{-1} and 2 ms^{-1}, respectively [28].

Dose rate survey and sample collection

In order to assure a reasonable coverage of the vast sampling area two approaches were used to collect representative soil samples. In the first, walkover gamma radiation survey was conducted using hand-held instruments to establish background levels. This survey was conducted on the entire area in accordance with dose rates measurements as described elsewhere [29]. A high performance calibrated survey meter, Canberra, inspector 1000™, coupled with a NaI probe capable of measuring the dose rate equivalent H*(10) down to 10 nSvh^{-1} was held horizontally one metre above the ground surface. A dose rate was recorded after readings on the detector attained a stable dose rate. Based on the results of dose rate measurements we clustered together locations with equivalent records of dose-rates to form a sampling block in order to optimize the number of representative samples and time of collection. This, approach created a total of 42 sampling blocks in area of 1300 km^2 that was estimated around the MRP.

In a sample collection, randomly selected locations in each block were marked with GPS coordinate and used as sampling points. Soil samples were removed from ground with a 25 mm diameter stainless steel auger. Since exposure to natural terrestrial radiation arises mainly from radionuclides within the top 30 cm depth of soil [30-32], at each sampling block, five samples of approximately 500 g each were collected in an area of 10 km x 10 km with sample depth ranges of 0 to 30 cm from the surface. At the time of sampling, grass, rocks, sticks and foreign objects were removed from the soil samples. Before collecting samples in next block, the sampling tools (auger and stainless steel rod) were cleaned thoroughly with brush to minimize cross contamination of the samples. In order to obtain a representative sample, subsamples collected randomly from similar depth layer in the block were mixed to form a composite and then a sample represented a block was drawn for laboratory tests. In addition, ten locations in the concession area were selected and samples collected to produce data for making reference during rehabilitation. The locations of blocks were recorded using a global positioning system and presented in Table 1 for future reproducibility and for monitoring of radiological elevation related to future project activities. The samples represented the sample blocks were packed in a separate cleaned polyethylene bag, sealed and labelled by writing the block number, sample date and depth on each bag before packed it for subsequent transportation to the gamma spectrometry laboratory.

Sample preparation

Composite soil samples from 52 locations collected onsite and offsite MRP were prepared consistent with the IAEA standard methods [33]. The samples were dried in oven at 100°C for overnight to minimize its moisture content until dry weight was achieved. The dried sample was ground using an agate mortar and pestle into fine powder then sieved by a 2 mm stainless steel sieve to obtain homogenous sample of similar matrix with reference material. In order to achieve a secular equilibrium and constancy between radium-226, thorium-232 and its gamma emitting progeny, after the in-growth of radon as described elsewhere [34,35], the fine powdered dry-weight that ranged from 250 to 400 g were placed in stainless steel canisters that were sealed air tight with silicone and electrical tape and stored for a period more than three weeks before measuring their radioactivity.

Determination of radioactivity

The radioactivity concentrations of radionuclides investigated in soil samples was measured according to standard procedures [33-36], by using a coaxial high-resolution HPGe spectrometer. The HPGe used had a relative efficiency of about 51%, and a resolution of 2.1 keV for the 1332-keV gamma ray line of the ^{60}Co source. The HPGe detection system was shielded in a cylindrical lead shield with 100 mm thick. This thickness was considered enough to reduce the influence of background counts on the sample. In addition, the lead shield was

Table 1: Coordinates of 42 sampling locations for soil samples in the vicinity and 10 locations in concession area.

Locations of sampling points in the vicinity								Locations of sampling points in the concession area			
Locations	**Samples**	**Eastings**	**Northings**	**Locations**	**Samples**	**Eastings**	**Northings**	**Locations**	**Samples**	**Eastings**	**Northings**
BH2B	5	37L0232200	8890109	MRP8	5	37L0239677	8887855	S1	5	37L0237542	8888890
BH4A	5	37L0230319	8882798	MRP13	5	37L0232250	8890145	S2	4	37L0235679	8887826
BH5A	5	37L0233126	8885087	MRP14	5	37L0233182	8885255	S3	4	37L0234895	8884073
BH5B	5	37L0233140	8885367	MRP15	5	37L0236080	8881586	S4	5	37L0237226	8885415
BH3	5	37L0235989	8881609	MRP16	5	37L0230489	8882699	S6	5	37L0237217	8887638
BH30	5	37L0239874	8887885	MRP23	5	37L0228199	8889639	S7	5	37L0235680	8887826
HDL2	5	37L0239796	8887938	MRP24	5	37L0240409	8891490	S8	5	37L0237219	8887637
HDL8	5	37L0227608	8887464	SW1U	5	37L0232976	8880136	S9	5	37L0237227	8885415
HDL9	5	37L0228956	8888266	SW1D	5	37L0232976	8880139	S10	5	37L0234896	8884073
HDL15	5	37L0239012	8892875	SW2U	5	37L0229538	8877601				
MB2	5	37L0234000	8801002	SW2D	5	37L0229512	8877624				
MBL1	5	37L0234153	8889570	SW3U	5	37L0227941	8874697				
MBL2	5	37L0240380	8888047	SW3D	5	37L0227913	8875007				
MBL3	5	37L0235004	8888447	SW4U	5	37L0223240	8867535				
MBL4	5	37L0237288	8892284	SW4D	5	37L0213216	8867527				
MBL5	5	37L0240397	8891490	SW5U	5	37L0209629	8861303				
MBL6	5	37L0239012	8892878	SW5D	5	37L0209618	8861304				
MBL7	5	37L0236143	8891776	SW6U	5	37L0206213	8860975				
MBL8	5	37L0233602	8884984	SW6D	5	37L0209213	8860986				
GWS1	5	37L0228061	8889395	SW7U	5	37L0207694	8860501				
GWS2	5	37L0228139	8889592	SW7D	5	37L0207700	8860559				

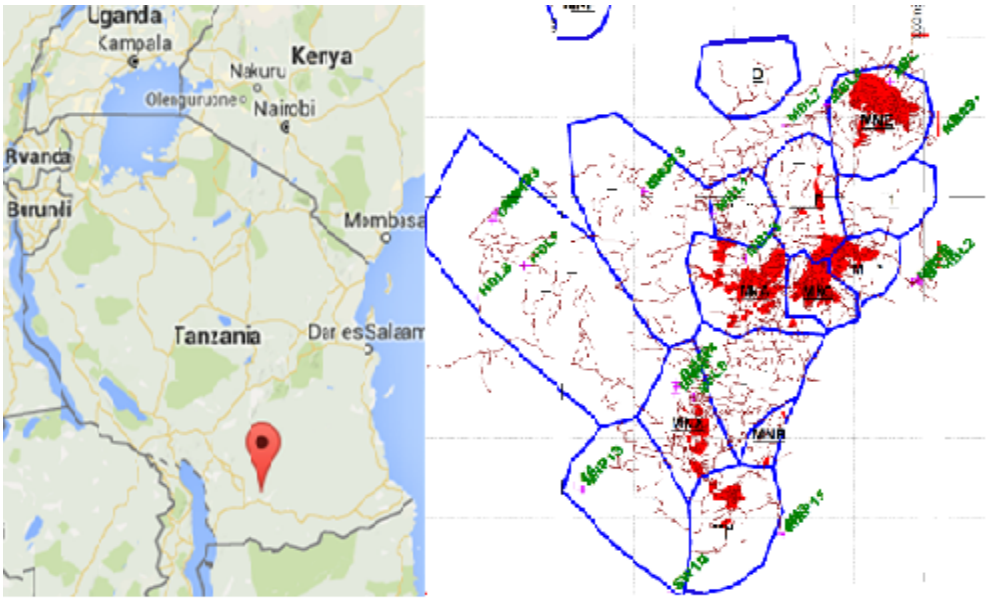


Figure 1: A map of Tanzania showing a location of MRP (left) and a sketch of 42 sampling points for soil samples in the vicinity of the mine and 10 sampling locations within the concession (right). The uranium ore deposits are shaded in red, and the sampling points are indicated by green markers.

lined with absorbers consisted of cadmium and copper metals with 3 and 30 mm thick, respectively to block the X-rays emitted from the lead shielding. To ensure the energy-channel's linear relationship, the detector was calibrated in the energy range between 60 and 2614 keV, using standard radiation sources before measurement. Sample spectra were acquired using a Multi-Channel Analyser (MCA) with 819 channels coupled to a computer for data processing to determine activity concentration of radionuclides investigated. Spectral analysis was performed with the Genie 2000™ and In Situ Object Counting System (ISOCS)™ software [37].

Due to low count rates, the soil samples was placed directly on top of the detector and counted for more than 12 hours to achieve reasonable counting statistics. To get the net count rates, R_N generated by the soil samples, it was necessary to subtract the background contribution, R_b, from the total count rates, R_t, of the soil and the background measured inside the detector chamber. The spectra of background contribution were collected by counting an empty canister overnight in the same procedure and with the same container as the soil samples. Using net count rates, R_N, the specific activity C_s ($Bqkg^{-1}$) of the corresponding radionuclides in the soil sample was obtained as follows [38,39]:

$$SA_s = \frac{R_s - R_b}{\eta\gamma W} = \frac{R_N}{\eta\gamma W} \quad (1)$$

Where: η is the gamma peak efficiency obtained from a calibration curve based on software (In Situ Object Counting-system) incorporated in the Genie 2000 software. The probability of γ is emissions was obtained from the literature [40] while W was the sample's dry weight (kg). Since a radionuclide can emit more than one kind of photon energies because of different emission probabilities, the SAs calculated for different peaks according to Equation 1 would be comparable. Thus, the desired specific activity of a given radionuclide in a sample obtained according to Equation 1 was obtained by averaging the counts of peaks showing similar activity [41]. The specific radioactivity of ^{226}Ra was determined using count rates, R_s of the gamma ray energies (keV) of ^{214}Bi (241.98, 295.21, 351.92, 609.31, 1120.29, 1238.11, 1377.82, 1729.60, 1764.49, 2204.22), and ^{214}Pb (241.98, 295.21 and 351.92). The radioactivity of ^{232}Th was evaluated using the peak energies (keV) of ^{212}Pb (238.63 and 300.09), ^{212}Bi (727.17) and ^{228}Ac (338.40, 583.2, 911.07, 969.11, 965 and 795) and ^{208}Tl (583.19, 860.56 and 2614.53). The radioactivity concentrations of ^{40}K were calculated directly using single gamma-ray energy (1460.81). The mean minimum detectable activity for counting time of 12 hours were estimated at 95% of confidence level for ^{226}Ra, ^{232}Th and ^{40}K were 0.75, 0.98, and 2.82, respectively.

Radium-equivalent radioactivity

In practice, a total radioactivity attributable to the detriment risk to members of public in the environment often is estimated from the knowledge of the three radionuclides (i.e. ^{226}Ra, ^{232}Th, and ^{40}K) given in terms of the radium-equivalent radioactivity Ra_{eq} expressed using Equation 2 [42]:

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_K \quad (2)$$

Where: C_{Ra} is the radioactivity concentrations of ^{226}Ra, C_{Th} is the radioactivity concentrations of ^{232}Th and C_K is the radioactivity concentrations of ^{40}K. All radioactivities measured in Bqkg^{-1} dry weight). The numbers in the equation represent weighting factors that estimates the specific radioactivity of 370 Bqkg^{-1} for ^{226}Ra, 259 Bqkg^{-1} for ^{232}Th, and 4810 Bqkg^{-1} for ^{40}K emits the same dose rate of gamma rays. In this context, if Ra_{eq} does not exceed 370 Bqkg^{-1}, then it is likely that the soil from which the sample was collected is within the acceptable safe limits [42].

Quality assurance

For the sake of verification that the measured values were accurate, the HPGe spectrometer was energy calibrated with point sources: ^{133}Ba, ^{22}Na, ^{137}Cs, ^{60}Co and ^{152}Eu, using standard procedures [33]. To assure the accuracy and reproducibility of the measured data, reference soil standards from the IAEA (IAEA-375) was prepared in a similar geometry and analysed using similar procedure as unknown sample. The specific activities of radionuclides obtained in the IAEA-375 were compared with the certified values to determine the levels of agreement. In this test, we found the radioactivity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K reproduced by the measurements were within deviations of about ± 5%, ± 6%, and ± 10%, respectively. Implicit in these results is that the performance of the measurement and the uncertainties were good.

Estimation of absorbed dose rate

The outdoor absorbed dose rates in air were estimated using radioactivity concentrations of three radionuclides: $C_{Ra,}$ C_{Th} and C_K determined from the top 30 cm layer of soil [30-32]. *The radioactivity concentrations* in soil was transformed into exposure dose rate in air at an average gonad height of 1 m above the ground's surface using conversion factors (nGy h^{-1}/Bqkg^{-1}) of 0.427, 0.662 and 0.048 for ^{226}Ra, ^{232}Th and ^{40}K, respectively as described elsewhere [39-43]. A relation presented in Equation 3 was used to calculate the exposure dose rate D:

$$D = 0.427C_{Ra} + 0.662C_{Th} + 0.048C_K \quad (3)$$

In order to interpret a risk associated with the radioactivity in soil, the exposure dose rate in air D was transformed to effective dose and then the computed annual effective dose E_D (mSvy^{-1}) was compared with the annual effective dose-limit of 1 mSvy^{-1} that is recommended for exposure of members of public [44]. We used a conversion factor of dose rate into effective dose, f_c of 0.7 SvGy^{-1} and residence time, t_r of 20% (8760) hours spent outdoors per year, to compute the annual effective dose E_D as shown in Equation 4 [43]:

$$E_D = D\, f_c\, t_r \quad (4)$$

Radiological hazard indices

Since inhabitants around MRP use soil as building material and for agricultural purposes, a risk of radiological exposure of population residing around MRP was estimated using the $C_{Ra,}$ C_{Th} and C_K determined in soil. The hazards indices were used to assess the risk were based on two exposure scenarios [42]. The first is external hazard due to handing the soils in construction. The second is the internal hazard attributable to exposure of radon and its progenies as a result of inhalation or ingestion of air or soil. Assuming a radium equivalent radioactivity limit of 370 Bgkg^{-1}, the external hazard index (H_{ex}) as a relative of radium equivalent radioactivity, can be obtained by dividing Equation 2 by this value.

$$E_D = D\, f_c\, t_r \quad (5)$$

Since the effect of radon, which is a progeny of radium, is more radiologically detrimental for internal exposure being a gas, the increased risk was taken into account by scaling the concentration of radium by a smaller value than 370, as suggested elsewhere, to obtain the internal hazard index (H_{in}) [42]:

$$H_{in} = \frac{C_{Ra}}{185} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \quad (6)$$

Results and Discussion

Ambient dose-rate

The ambient dose rates in air measured directly in the field using radiation survey detector are presented in Figure 2 and 3 for locations in the vicinity and the concession areas, respectively.. In Figure 2 It was found that the recorded mean dose rate in the vicinity ranged from 0.01 to 0.03 μSvh^{-1} with an overall mean of 0.02 ± 0.01 μSvh^{-1}. Furthermore, assuming that the outdoors occupancy is 20%, a simple calculation would show that a population dose attributable to the dose rate measured in the vicinity will range from 17.53 to 52.60 μSvy^{-1} with a mean of 43.41 ± 0.3 μSvy^{-1}. In contrast to worldwide population exposure dose attributable to terrestrial radiation sources of 2.4 mSvy^{-1}[43], the calculated exposure dose to population residing in the vicinity of MRP is low (about 2%). In addition, an overall mean dose rate recorded in the vicinity of MRP is also below the outdoors exposure dose rates, commonly reported by different countries of the world that ranges from 18 to 93 μSvy^{-1} with a mean of 55 μSvy^{-1}[43].



Figure 2: Ambient dose rate recorded at 1 m above ground for different locations in the vicinity of MRP.

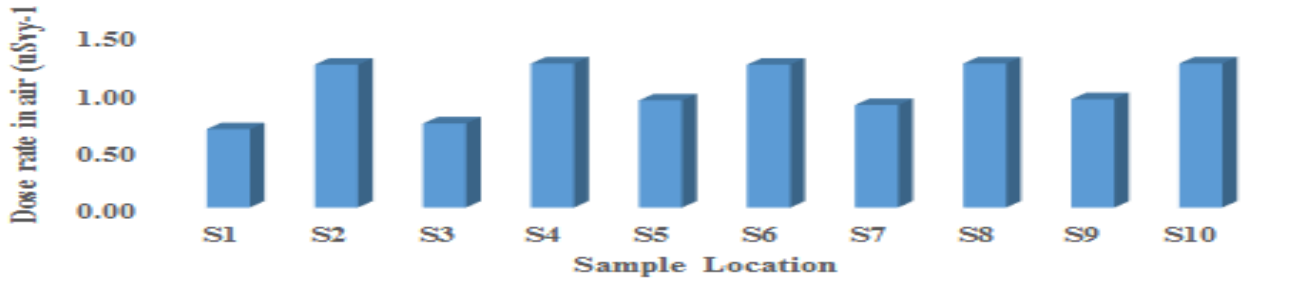


Figure 3: Ambient dose rate recorder at 1 m above ground for different locations in the concession area of MRP.

Furthermore an overall mean dose rate of 43.4 ± 0.3 Svy^{-1} at MRP is extremely low about 4 % of the recommended dose limit of 1 mSvy^{-1} for a member of the public [44]. However, this is not the case for the concession area (Figure 3) where a range of terrestrial radiation dose rate varied from 0.68 to 1.25 μSvh^{-1} with an overall mean of 1.04 ± 0.23 μSvh^{-1}. Using a similar assumption of occupancy factor of 20% outdoors, the calculated population dose in the concession ranged from 1192.18 to 2191.50 μSvy^{-1} with a mean of 1801.43 ± 20 μSvy^{-1}. This is an evident that the overall mean of outdoors exposure dose to population residing in the concession area will receive approximately 42 times higher dose than those residing in the vicinity of MRP. Also an interesting point in Figure 3 is that the maximum dose rate in the concession area at locations S4, S8 and S10 (2191.5 μSvy^{-1}) are about 913 times higher than the population exposure considered to be due to the terrestrial radiation sources in the world. Also the mean dose rate obtained in the concession (1823.33 ± 20 μSvy^{-1}) is about twice the acceptable dose limit of 1 mSvy^{-1} for a member of the public [44]. The elevated dose rate recorded in the concession area could be due to the presence of radioactivity concentration of uranium ore and associated radioactive materials in the deposit. Based on the dose survey it can be concluded that radiological risks is relatively marginal in the vicinity of MRP when compared to the concession area.

Radioactivity concentration in soil

The plots of calculated radioactivity concentrations of ^{226}Ra, ^{232}Th and ^{40}K using Equation 1 for the investigated soil samples versus different sampling locations in the vicinity of the MRP are presented in Figure 4, and the corresponding radioactivity concentration for samples collected in the concession area are plotted in Figure 5. In general, (Figure 4) radioactivity concentrations of ^{226}Ra and ^{232}Th are low in all samples collected in the adjacent of the project. Figure 4 shows higher values of activity concentrations in all samples are associated with ^{40}K and lower values are associated with ^{232}Th. Although the radioactivity concentrations of ^{226}Ra and ^{232}Th do not differ much, it was noted (Figure 4) that about 69% of the locations has the radioactivity concentrations of ^{226}Ra higher than the radioactivity concentrations of ^{232}Th. However, it has been observed in Figure 4 that 13 locations in the vicinity: BH5A, BH5B, BH30, MRP14, MBL4, MBL7, MBL4, MRP13, SW2U, SW4U, SW6D, HDL15 and MRP24 presents the higher radioactivity concentrations of ^{232}Th than those of ^{226}Ra. Besides, the activity levels of ^{226}Ra, ^{232}Th and ^{40}K found in the vicinity are lower than the corresponding values found in the concession area (Figure 5). However, the values obtained in the vicinity are comparable to the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, which are reported in soil by different countries of the world (Table 2).

Figure 5 depicts outstanding higher radioactivity concentration of ^{226}Ra, ^{232}Th and ^{40}K in soil samples collected in the concession area compared to the radioactivity concentration of the same radionuclides obtained in the vicinity (Figure 4). The mean radioactivity concentrations of ^{226}Ra in the concession area are about 85 times higher than the mean radioactivity concentration of the same radionuclide recorded in the vicinity. The mean radioactivity concentration of ^{232}Th in the concession area is approximately 4 times higher than the mean obtained in the vicinity. The mean radioactivity concentration of ^{40}K is 2 times higher than the corresponding mean value of same radionuclides in the vicinity. The elevated radioactivity concentrations of ^{226}Ra and ^{232}Th in the samples collected in the concession could be associated with uranium ore in the deposit. It should be noted that the maximum mean value of ^{226}Ra in the concession area (Figure 5) was recorded in soil sample derived from location D3 and the minimum mean radioactivity concentration was found in soil sample obtained from location D4. The minimum radioactivity levels of ^{226}Ra, ^{232}Th and ^{40}K found in the concession area are significant above the radioactivity concentrations of ^{226}Ra, ^{232}Th and ^{40}K reported by different countries

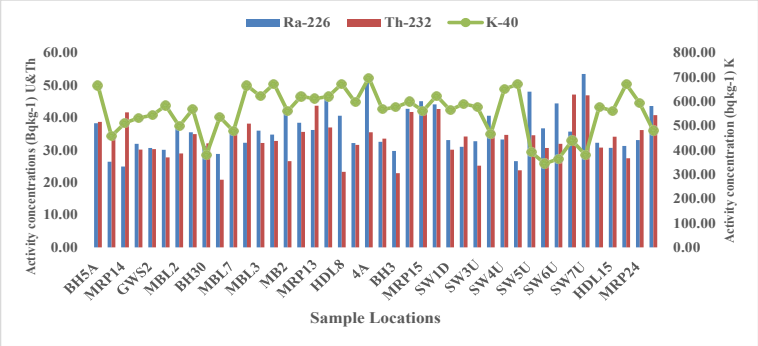


Figure 4: Radioactivity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in soil samples collected from different locations in the vicinity of the MRP .

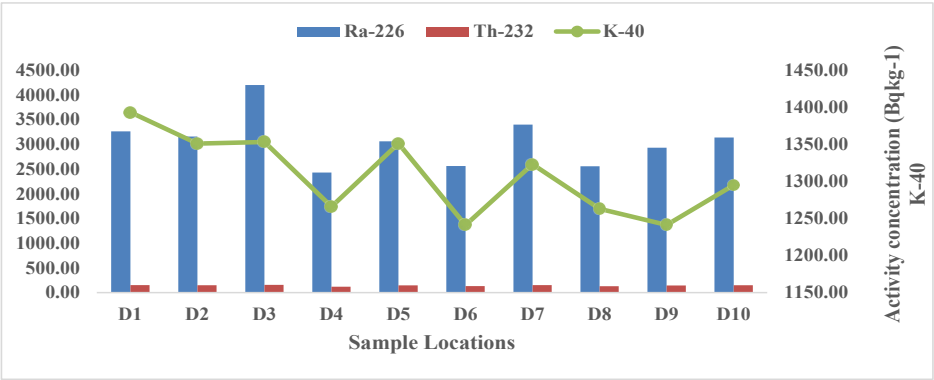


Figure 5: Radioactivity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in soil samples obtained from different locations in the concession of the MRP.

Table 2: Comparison of values of range and mean radioactivity concentrations level in soils and effective dose measured at different locations at MRP and corresponding studies by different countries of the world.

Countries	Activity concentrations (Bqkg^{-1})						Effective Dose (μSy^{-1})		References
	^{238}U (^{226}Ra)		^{232}Th		^{40}K				
	Range	Ave	Range	Ave	Range	Ave	Range	Ave	45
Egypt	5-64	17	2-96	18	29-650	320	20-133	32	45
USA	8-160	40	4-130	35	100-700	370	14-118	47	45
China	2-440	32	1-360	41	9-1800	440	2-340	62	45
Japan	6-98	33	2-88	28	15-990	310	21-77	53	45
Malaysia	38-94	67	63-110	82	170-430	310	55-130	92	45
India	7-91	29	14-160	64	38-760	400	20-110	56	45
Iran	8-55	28	5-42	22	250-980	640	36-130	71	45
Denmark	9-29	17	8-30	19	240-610	460	35-70	52	45
Poland	5-120	26	4-77	21	110-970	410	18-97	45	45
Greece	1-240	25	1-190	21	12-1570	360	30-100	56	45
Romania	8-60	32	11-75	38	250-1100	490	21-122	59	45
Spain	6-250	32	2-210	33	25-1650	470	40-120	76	45
Luxenberg	6-52	35	7-70	50	80-1800	620	14-73	49	45
Bangladesh	28-67	48	33-70	53	345-674	481	62-100	77	45
MRP Previous study		52		36		564			47
MRP in vicinity	25-54	36	21-47	34	345-698	556	52-79	65	In
MRP in concession	2430-4200	3263	130-220	164	1293-1466	1391	1537-2385	1924	In
Worldwide (UNSCEAR 2000)	16-110	35	11-64	30	140-850	400	18-93	55	45

of the world in soil [43]. The outstanding difference of radioactivity concentration of the radionuclides between the concession area and in the vicinity of the project suggest that there is a pollution potential as a result of emissions of radioactive materials via water and/or wind pathways during the mining operations.

Data analysis

In principle, radionuclides that occurred naturally have a certain association because of their common origin. Therefore statistical analysis could indicate the distribution of the radionuclides in soil.

Activity concentration: The statistical data for radioactivity concentrations of 226Ra, 232Th and 40K determined using Equation 1 in the investigated soil samples obtained from different sampling locations on the site and in the vicinity of MRP are presented in Table 3. It was observed that the mean values of radioactivity concentrations of ^{226}Ra in soil were higher than that of ^{232}Th. The radioactivity concentrations of ^{226}Ra ranged from 24.94 to 53.50 Bqkg^{-1} with an average of 36.27 ± 6.74 Bqkg^{-1} and the activity of ^{232}Th ranged from 20.86 to 47.14 Bqkg^{-1} with an average of 33.80 ± 6.33 Bqkg^{-1} In contrast, radioactivity of ^{40}K was higher than that of ^{226}Ra and ^{232}Th in all soil samples investigated. The levels of ^{40}K ranged from 344.50 to 697.54 Bqkg^{-1} with an average of 555.87 ± 93.18 Bqkg^{-1}. The highest radioactivity concentration for ^{226}Ra, ^{232}Th and ^{40}K were found (Figure 2) in locations: SW7U, SW6D and 4A, respectively. The lowest radioactivity concentrations for ^{226}Ra, ^{232}Th and ^{40}K were recorded in locations: MRP14, MRP8 and SW5D, respectively. The possible reason for ^{40}K concentration exceeding values of both radium and thorium could be due to the variability of formations and textures of the soil in the study area. The type of soil in the study area is dominated by clay soil that resulted from runoff of rain water. The higher potassium level was probably attributed to its abundant in soil. The mean values of ^{226}Ra, ^{232}Th and ^{40}K in the investigated samples agreed with previous results of 51.7 Bqkg^{-1} for ^{238}U, 36.4 Bqkg^{-1} for ^{232}Th and 564.3 Bqkg^{-1} for ^{40}K found by Mohammed and Mazunga [45]. Both studies reported the mean values of ^{226}Ra, ^{232}Th and ^{40}K in all locations in the vicinity slightly above the corresponding mean values reported in the worldwide for :^{226}Ra (35 Bq kg^{-1}), ^{232}Th (30 Bq kg^{-1}) and ^{40}K (400 Bq kg^{-1}) [43].

Table 3 indicate that radioactivity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in soil samples collected in the concession are significantly high. The reason could be due to the uranium deposit. The radioactivity concentration of ^{226}Ra ranged from 2430.00 to 4200.00 Bqkg^{-1}) with an mean of 3263.25 ± 726.28, the radioactivity of ^{232}Th ranged from 130.00 to 220.00 Bqkg^{-1} with a mean of 164.25 ± 38.89 Bqkg^{-1} and the radioactivity of ^{40}K ranged from 1293.30 to 1466.10 Bqkg^{-1} with a mean of 1390.93 ± 82.11 Bqkg^{-1}. Moreover, the values of ^{226}Ra, ^{232}Th and ^{40}K in the soil samples were higher than the corresponding values for: ^{226}Ra (35 Bq kg^{-1}), ^{232}Th (30 Bq kg^{-1}), and ^{40}K (400 Bq kg^{-1}) (Table 2) those reported in the world [43]. In other words the soils in the concession could potentially pose public health detriment above normal limits if migrate to the vicinity

Activity concertation ratio: Since in principle soils derived from same rock have a distinctive constant ratio of radionuclides, radioactivity concentrations ratios were calculated for Th/Ra, K/Ra and K/Th in soil samples and presented in Table 3. The data in Table 3 show that ratios tend to be variable and significantly different. This difference implies that radionuclides in soil were differentially distributed may be due to secondary weather processes. If ratios in Table 3 are compared, it is clear that values for the concession tend to be less than those in the vicinity. This difference may reflect enhanced concentrations of radium and thorium in soils derived from the concession. The ratios for Th/Ra, K/Ra, and K/Th in the vicinity ranged from 0.57 to 1.67, 7.11 to 25.21 and 8.11 to 28.85, respectively. Whereas ratios in the concession range from 0.03 to 0.09, 0.32 to 0.60, and 6.66 to 10.41 for Th/Ra, K/Ra and K/Th, respectively. These ratios reflect probably a typical natural balance of radionuclides in soil, which may be distorted towards the change of radionuclides concentration in soil. Thus radioactivity concertation ratio can serve as indicator of the radiological changes on the environment.

Radium equivalent activity: Of interest is not only the radioactivity concentration of individual radionuclide but the radium equivalent radioactivity of radionuclides in soil, which gives a picture of sum of radioactivity concentrations of the radionuclides in soil samples investigated. The radium equivalent radioactivity was obtained as described in Equation 2 and presented in the 10th column of Table 3. The minimum and maximum values of radium equivalent radioactivity in soil samples obtained in the concession were both approximately 29 times higher than the minimum and maximum values in soil samples collected in the vicinity. The radium equivalent radioactivity in soil samples from the concession ranged from 2857.49 to 4490.10 Bqkg^{-1} while in soil samples collected in the vicinity of MRP ranged from 99.96 to 155.50 Bqkg^{-1}. Based on the acceptable safety limit for radium equivalent radioactivity in building material of 370 Bq kg^{-1}[42], the soils obtained in the vicinity which is 42% of the recommended upper value tend to be safe. On the other hand use of soils from the concession as building material is not safe since has the minimum radium equivalent radioactivity higher about 8 times the acceptable safety limit [42]. Since there is an indication of excess radium equivalent radioactivity in the concession it is plausible that by products associated with the mining activities could potentially lead to pollution in the vicinity of MRP, if proper waste management

Table 3: The minimum, maximum, average, standard deviation skewness for radioactivity concentrations of radionuclides (Bqkg^{-1}), radioactivity concentration ratios, radium equivalent radioactivity and dose rate and hazard indices for soil samples collected from different locations onsite and offsite MRP.

Sampling Location	No of samples	Statistics	Activity concentration (Bqkg^{-1}) ^{226}Ra ^{232}Th ^{40}K			Activity concentration ratios Th/Ra K/Ra K/Th			Radium Equivalent (Bqkg^{-1})	Dose rate (nGyh^{-1})	Effective Dose (μSy^{-1})	Hazard indices H_{EX} H_{IN}	
In vicinity of MRP	42	Min	24.94	20.86	344.50	0.57	7.11	8.11	99.96	51.87	63.61	0.27	0.35
		Max	53.50	47.14	697.54	1.67	25.21	28.85	155.50	78.77	96.61	0.42	0.56
		Ave	36.27	33.80	555.87	0.95	15.88	17.14	127.41	64.55	79.16	0.34	0.44
		Std	6.74	6.33	93.18	0.20	3.92	4.84	13.95	6.78	8.31	0.04	0.05
		Skewness	0.69	0.10	-0.70	1.12	-0.34	0.53	0.25	0.16	0.16	0.25	0.44
Inside the MRP Concession	10	Min	2430.00	120.00	1241.10	0.04	0.32	8.62	2699.09	1177.82	1444.48	7.29	13.89
		Max	4200.00	157.00	1393.30	0.05	0.52	10.55	4528.71	1962.29	2406.55	12.24	23.59
		Ave	3070.50	143.50	1307.88	0.05	0.43	9.15	3376.41	1468.88	1823.33 20	9.12	17.42
		Std	514.89	12.04	53.90	0.00	0.06	0.61	532.77	28.59	0.88	1.44	2.83
		Skewness	0.97	-0.99	0.11	-1.86	-0.39	1.48	0.89	0.88		0.89	0.93

Table 4: Linear Pearson correlation coefficient for the investigated radiological variables for soil samples in the vicinity of MRP.

	^{226}Ra	^{232}Th	^{40}K	Th/Ra	K/Ra	K/Th	Raeq	D	E	HEX	HIN
^{226}Ra	1										
^{232}Th	0.43	1									
^{40}K	-0.16	-0.17	1								
Th/Ra	-0.48	0.55	-0.04	1							
K/Ra	-0.78	-0.40	0.72	0.31	1						
K/Th	-0.38	-0.79	0.69	-0.43	0.70	1					
Raeq	0.68	0.77	0.33	0.10	-0.27	-0.34	1				
D	0.59	0.69	0.49	0.11	-0.10	-0.19	0.98	1			
E	0.59	0.69	0.49	0.11	-0.10	-0.19	0.98	1.00	1		
HEX	0.68	0.77	0.33	0.10	-0.27	-0.34	1.00	0.98	0.98	1	
HIN	0.85	0.71	0.18	-0.09	-0.47	-0.38	0.97	0.92	0.92	0.97	1

is not properly done.

Absorbed dose rate: Since about 10,000 people resides in villages of Namtumbo district adjacent to MRP, therefore an assessment of radiological exposure to the general public in the early stage of the uranium mining is necessary. Equally important is an estimation of radiological risk to mine workers inside the concession area under similar exposure scenario. Radioactivity concentrations obtained by Equation 1 were used as input in Equation 3 to estimate the outdoors exposure dose rate and presented in Table 3 for each location in the vicinity and the concession area. In Table 3 outdoors dose rate in air found in the vicinity ranges from 51.87 to 78.77 nGyh^{-1} with a mean of 64.55 ± 6.78 nGyh^{-1}. This mean is slightly higher when compared with the mean of terrestrial dose rate reported by different countries of the world which is 60 nGyh^{-1}[43]. However a maximum dose rate (78.77 nGyh^{-1}) was found to be less than a maximum value reported in the world [45], suggesting that the dose rates obtained in this study could be from the natural sources. On the other hand Table 3 shows, in the concession dose rate vary from 1177.82 to 1962.29 nGyh^{-1} with a mean of 1468.88 ± 28.59 nGyh^{-1}. In comparison with the world values a mean value exceed about 25 times the world mean values which is 60 nGyh^{-1} [43]. Furthermore, the minimum and maximum values found in the concession were about 23 and 35 times higher than the corresponding values found in the vicinity. This result suggests that there is a probability of potential radiation exposure in the concession of MRP compared to the vicinity. These results compared with the results of the dose rate measurement in the field survey indicating that the using soil radioactivity and measurement of directly ambient dose rate.

Annual effective dose: The calculated annual effective dose using dose rates as described by Equation 4 for each location within and around the concession area is presented in Table 3. Based on the standards of acceptable exposure to a member of public, doses not exceeding 1 mSvy^{-1} is considered to be safe [44], In Table 3 the annual effective dose for areas adjacent to the project ranged from 52.00 to 79.11 μSvy^{-1} with a mean value of 65.12 ± 8.31 μSvy^{-1}. In contrast, these values are in same order of magnitude but lightly higher than those recorded by the radiation survey instrument from 17.53 to 52.60 μSvy^{-1} with a mean of 43.41 ± 0.3 Svy^{-1}. Lower values in the field could be due to shielding effects and temporal variability of the weather at site. However both measurements in the vicinity tend to be less than the acceptable limit of exposure of members of public [44].

However, in the concession the minimum and maximum annual effective dose were about 23 to 25 times higher than the corresponding values found in the vicinity. This indicates the association of radioactivity and effective dose. A calculated range of annual effective dose from 1444.48 to 2406.55 μSvy^{-1} with a mean 1823.33 ± 20 μSvy^{-1} in the concession is comparable ranges obtained directly by field instrument from 1192.18 to 2191.50 μSvy^{-1} with a value of 1801.43 ± 20 μSvy^{-1}. The values determined in the concession tend to be 2 times above the recommended annual dose limit for a member of the public of 1 mSvy^{-1}[44]. The results of annual effective dose suggest that may be a radiological risk related to the deposit is high.

Hazards indices: The statistical data for H_{EX} and H_{IN} of different locations obtained using the specific activities as input to Equations 5 and 6 are presented in columns 13th and 14th of Table 3 for H_{EX} and H_{IN}, respectively. For the sake of compliance with the radiological safety standards when soil is used as building material, both indices should be less than unity [46]. In Table 3, H_{EX} in the vicinity range from 0.27 to 0.42 with a mean of 0.34 ± 0.04 and in the concession H_{EX} range from 7.29 to 12.24 with a mean of 9.12 ±1.44. The mean H_{EX} in the concession is 27 times higher than a mean obtained in the vicinity. These mean values indicate that the concession has values in excess about 9 times above the unity while in the vicinity the value is below the recommended upper level of unity [46]. Similarly, H_{IN} in the vicinity range from 0.35 to 0.56 with a mean of 0.44 ± 0.05 while in the concession H_{IN} values range from 13.86 to 23.59 with a mean of 17.42 ± 2.83. In comparison a mean values of H_{IN} in the concession is about 40 times higher than that in the vicinity. This means that internal exposure from ingestion of soils in the concession is about 17 times higher than from soil in the vicinity. Besides, as expected the internal risk tends to be much higher than the external risk for both locations because of radon exposure.

Relationship of radiological parameters: In order to understand distribution of ^{226}Ra, ^{232}Th and ^{40}K in soil samples skewness of data were calculated and presented in Table 3. In Table 3 a positive skewness is an indication of asymmetric distribution of data with tail extending towards values that are more positive. Whilst a negative skewness is an indication of asymmetric distribution of data with tail extending towards values that are more negative and zero is an indication that the spatial distribution of data is normal and perfect symmetry [47]. Interesting to note in Table 3 that the skewness of ^{226}Ra and ^{40}K are positive and those of ^{232}Th are negative.

In addition, to assess a relationship and strength of association between pairs of variables, the linear Pearson correlation coefficient (r) has been used and presented in Table 4. Table 4 shows, a positive correlation (r=0.43) of radioactivity of ^{226}Ra and that of ^{232}Th. This indicate that uranium and thorium occurs together in nature as observed elsewhere [48]. In Table 3 data indicate a strong correlation between radioactivity of ^{226}Ra and Raeq (r=0.68), D (r=0.59), E

(r=0.59), H_{EX} (r=0.68) and H_{IN} (r=0.85). Similarly, data show existence of a strong correlation of ^{232}Th and Raeq (r=0.77), D (r=0.69), E (r=0.69), H_{EX} (r=0.77) and H_{IN} (r=0.71). Furthermore a strong positive correlation has been observed between Raeq and D (r=0.98), E (r=0.98), H_{EX} (r=1) and H_{IN} (r=0.97), between dose rate and E (r=1), H_{EX} (r=98) and H_{IN} (r=0.92), between effective dose and H_{EX} (r=0.98) and H_{IN} (r=0.92). A strong correlation is an indication of close relationship between the radiological parameters. This relationship could be distorted in the presence of radionuclides from foreign origin. The calculated coefficient of determination R^2 of variables also indicate a linear fit significant at p>0.05.

Conclusions

The levels of ^{226}Ra, ^{232}Th and ^{40}K and associated parameters in soil samples derived on the site and in the vicinity of MRP have been investigated to establish indicators for ready to use in verification of compliance with standards. The results of this study indicate that radioactivity and risk related to use of soil derived in the vicinity of MRP prior to mining operations is marginal. However, it become apparent that since radioactivity levels in soil from the concession was found elevated, its inadvertent access in the vicinity could lead to excess radioactivity onto the environment and risk to people above the permissible safety limit. For this reason, environmental surveillance programmes are recommended to monitor the radioactivity pathways during the mining operations. Thus, the data in the present study could serve as preliminary reference baseline data for this purpose.

Acknowledgements

The authors gratefully acknowledge the owners of MRP, the Uranium One, for permission to take samples in the area. The technical assistance and the logistical support provided by Mr Beria, Site Manager, and John Ntukula, Environmental Manager, for supplying valuable background information and transport to sampling points is highly appreciated. The authors would also like to thank Mr Mujuni Rweyemamu of TAEC for his assistance in radioactivity measurement of the samples. Thanks also go to COSTECH for financial support, and the TAEC for granting a study leave.

Conflict of Interest Statement

The authors declare that there are no potential conflicts of interest or any financial- or personal- relationships with other people or organizations that could inappropriately bias conduct and findings of this study.

Author's Contributions

All authors read and approved the final manuscript.

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