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Measurements of Radiometric Concentrations for ^{222}Ra, ^{238}U, ^{232}Th and ^{40}K in Soil Samples from Two Uranium Mines' Stockpiles, Erongo Region, Namibia

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ABSTRACT

The continuous exposures from radiations to human population has led to research studies on evaluating the radioactivity concentrations in most parts of the world. Naturally occurring radioactive materials (NORM) have been greatly regarded as the chief sources of both terrestrial and cosmological radiations. In this study a high-purity germanium well detector was used to measure the activity concentrations of samples from mines 1 and 2 considered in the study. The measured average activity concentration for ^{226}Ra, ^{238}U, ^{232}Th and ^{40}K in mine 1 were estimated at 2546±10, 557±13, 215±2 and 1079±26 $Bq.kg^{-1}$, respectively. For mine 2, the estimated average values of the radionuclides of ^{226}Ra, ^{238}U, ^{232}Th and ^{40}K were calculated as 4414±27, 842±29, 436±5 and 2225±52 $Bq.kg^{-1}$, respectively. The high concentration of radium in samples contributed to high rate of radon emanation into the atmosphere, ground and surface water as well and, if no remedial measures are put in place, this may contribute to plants, animals and eventually human exposures in the region. The study concludes that mine tailings contribute to an increase in background radiation in the environment due to the fact that they are uncovered and deposited in the open environment, resulting in wind-blown ^{226}Ra carrying dust entering nearby residential dwellings. The exposures rates could also be increased by other possible transfer pathways, such as ingestions, inhalations and external gamma radiations. Mining companies are urged to decrease windblown atmospheric exposure by using water tanks mounted on mining equipment to sprinkle on top of the soil or covering the tailings with canvas sheet.

Keywords : Terrestrial, NORM, Radionuclides, Human Population, Exposure, Mining, Radioactivity Concentrations, Hpge Detector

I. INTRODUCTION

The continuously increasing radiations exposures to living organisms, including human beings, from the environments has been questionable worldwide. These exposures arises from ubiquitously and unevenly distribution of natural radionuclides, such as potassium (K), uranium (U) and thorium (Th) in the earth's crust (UNSCEAR, 2017). Their decay products like radon and radium are hazardous and therefore contribute to terrestrial radiations exposures. Assessments of radioactive elements in the natural environments has become one of the crucial research studies that has been and still continues to be of interest. Most researchers have evaluated radiological hazards that are accessible to a human population (Thabayneh, 2012; Samreh et al., 2015; Shimboyo and Oyedele, 2015). Exposures of living organisms is dependent to natural radionuclides present in the area (Alzubaidi et al., 2016; Isinkaye and Emelue, 2015).

Terrestrial and cosmic radiations are regarded as chief sources of a continuous external exposures. Uranium, thorium and potassium are the most important terrestrial elements that are part of the naturally occurring radioactive materials (NORM) (Mathuthu et al., 2021). Uranium is an important element in a human lifetime due to its use in production and supplying electrical energy. Radium and radon are uranium progeny that are mostly found in high concentrations in soils from uranium mines' stockpiles and dwellings, respectively.

The aim of this study was to estimate the radioactivity concentrations of NORM from stockpiles in the two mines of Erongo region. Prolonged human exposures arise from radioactivity concentrations in mining environments which may lead to health complications in the body (Issa et al., 2013). The results of this study will provide radioactivity levels that could guide the mine's leadership in implementing the effective radiation management plan and therefore conducive environment for workers and inhabitants of the area.

II. METHODOLOGY

2.1 Sampling

Random sampling strategy was employed to select the sampling sites in the two operational uranium mines closer to the town of Arandis; coded as mine A and mine B for confidentiality reasons. For each sample collected the global positioning system (GPS) of the respective point of collection was recorded. The GPS values and names of the mines under study could not be published because of the confidentiality agreements signed with the mines. Figure 1 shows some of the samples packed in their containers during sampling, transported and prepared at the analytical laboratory at Centre of Applied Radiation Science and Technology (CARST), North-West University (NWU), South Africa.

2.2 Samples preparation

All soil samples were separately spread on well-coded cartons and left on the open to dry for seven (7) days at room temperature prior to packaging, to remove all possible moistures and organic matter. They were then crushed for homogeneity, filtered through a 2.0 mm sieve, and about 500 g of weighed samples were individually packed and sealed in well coded 500 ml cylindrical plastic containers, geometrically similar to the one shown in Figure 2. The masses (m_s) of individual samples were measured and recorded for use during calculation of radioactivity concentrations. Each sample was sealed in a container for about 30 days, to allow radon and its progeny to reach radioactive secular equilibrium.

Figure 1: Packaged plastic containers and cartons used for spreading samples for drying out moistures.

2.3 Calibration of high-purity germanium (HPGe) detector

Calibrations for both energy and efficiency of the Canberra HPGe detector well-type detector were performed using IAEA radiometric reference materials; RGU-1, RGTh-1 and RGK-1, as shown in Figure 2. IAEA-RGU-1 standard sample source was prepared by dilution with silica sand of Canadian Certified Reference Materials Programme (CCRMP) Uranium Ore BL-5 (7.09% U) and IAEA-RGTh-1 standard source sample was prepared by dilution with silica sand of OKA-2 (2.89% Th, 219 µg U/g) (IAEA, 1987).

To calculate efficiency of the gamma spectrometry instrument, the following equation was used (Tedjani et al., 2016):

$$\varepsilon = \frac{C_{n,p}}{A\,(Bq) \times I_{\gamma} \times T(s)} \qquad (1)$$

where, $C_{n,p}$ is the total number of counts per peak area under calibration, I_{γ} is the gamma probability emission and T is total time of acquisition in seconds.

Efficiency calibration curve of the detector as a function of energy is a smooth curve which falls exponentially with increase in energy on the spectrum window, shown in Figure 3. These calibrations were performed well prior to counting of samples, with known activities of radioactive nuclides. This is the most important quality control and quality assurance procedures which have to be performed on every analytical instruments for accuracy and performance. Calibrations of analytical instruments were done in accordance with recommended procedures by international standard agencies, and in this case, the IAEA. The linear energy against channel graph is shown in Figure 4 for energy calibration.

Figure 2 : The IAEA radiometric reference materials; RGU-1, RGTh-1 and RGK-1 for calibration of both energy and efficiency for HPGe detector at CARST.

Calibration for efficiency was recorded and saved for use during analyses of samples. The efficiency smooth curve is shown in Figure 3.

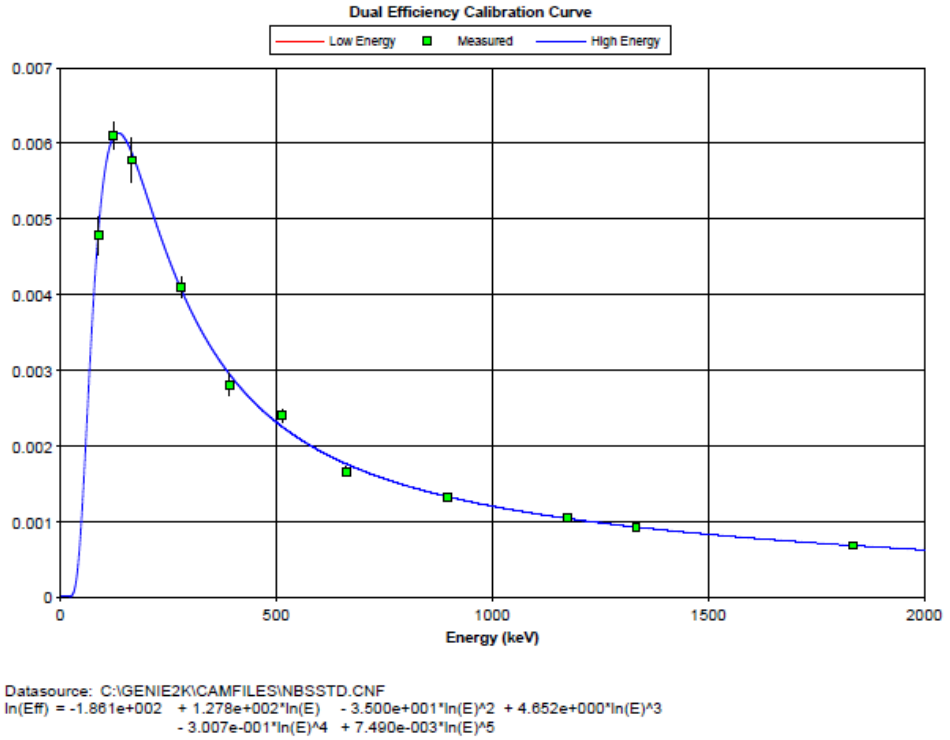

Figure 3 : Efficiency curve for the calibration of HPGe detector.

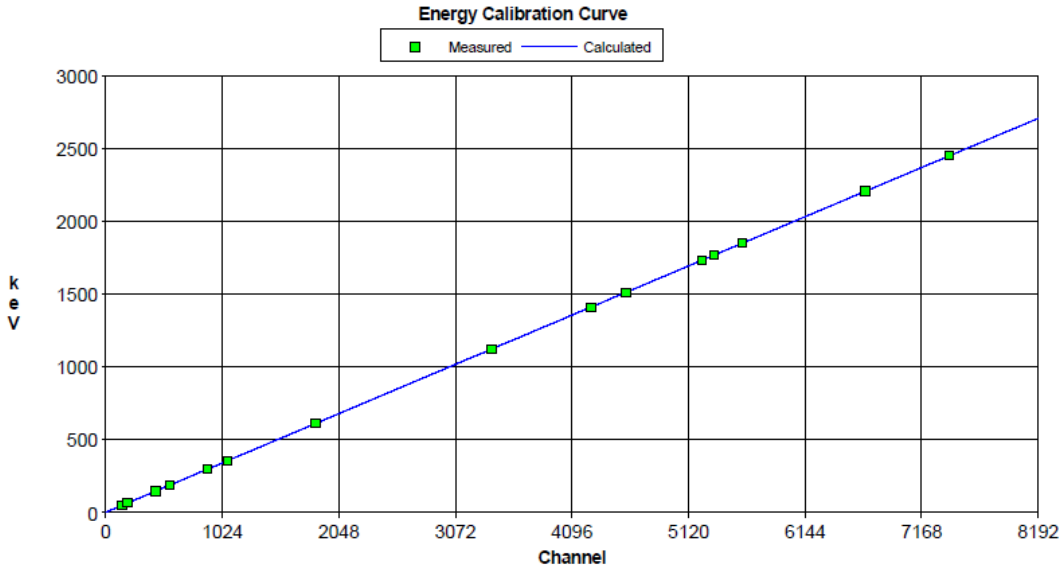

Figure 4 : Energy calibration, a linear graph, of the HPGe well-type detector.

3. Radioactivity measurements of ^{226}Ra, ^{238}U, ^{232}Th and ^{40}K

Analyses of all radionuclides of interest in this study was performed on the gamma spectroscopy detector. The detector, Figure 5, was manufactured by Canberra Industries (Meriden, CT, USA). It contains a detector model GCW2021 and relative efficiency of 36.4% and resolution of 1.842 keV at 1332 keV, γ-ray emission of ^{60}Co. The high-purity germanium system computer has Genie 2000 version 3.3 software installed for quantitative and qualitative analysis. A digital based multi-channel analyser (MCA 1000) couples the detector to the software. The

counts per peak of a specified energy were recorded and computed to obtain activity concentrations for each radionuclide under study.

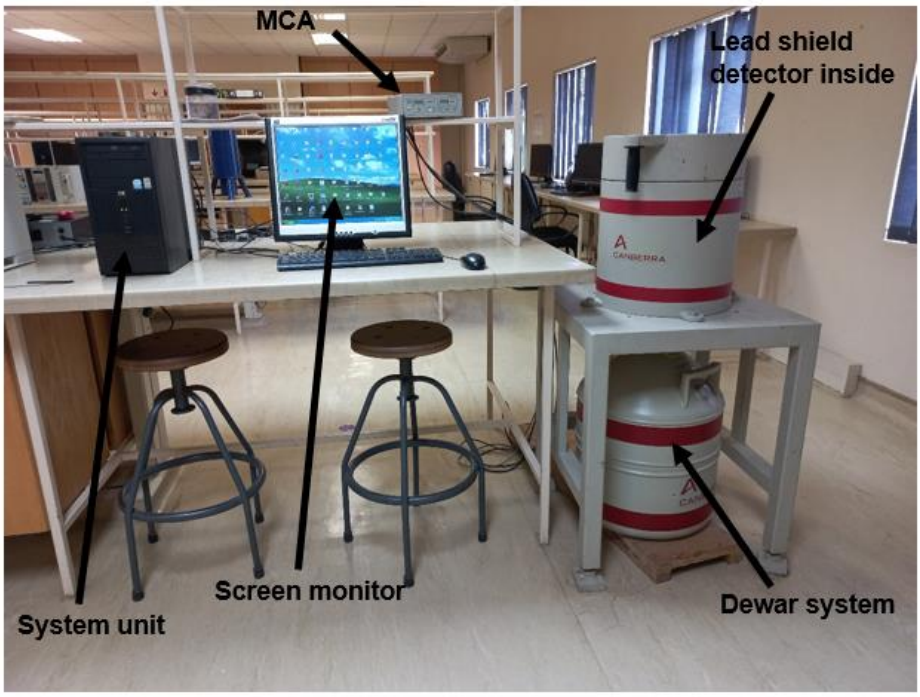

Figure 5 : The HPGe system at CARST used in this study showing the MCA, Dewar cooling system (with LN2), lead shield, computer monitor and data display.

Gamma spectrometry is a non-destructive technique that uses HPGe (or Na(Tl)I) detector for identification and quantification of all gamma emitting radionuclides present in the sample based on energies and peak area of full energy peaks in their respective gamma rays. The acquisitions of all samples data were performed on a Genie 2000 version 3.3 computerized software, under low-background conditions achieved by placing the detector inside lead shielding as in Fig 5. The gamma spectra peaks were obtained by measuring radioactivity counts for a period 12 hours (43200 s) per sample. One good and important advantage for non-destructive method of HPGe instruments is that it allows many gamma-emitting nuclides to be measured simultaneously.

The calculations of activity concentrations for radionuclides of interests were obtained from results of daughter products assuming secular equilibrium have been achieved. The energy tolerance and nuclide confidence index threshold were 3.00 keV and 0.30, respectively. Analyses from the spectrum was performed using energy lines with their respective gamma emission probability and efficiency. On HPGe detector ^{238}U, ^{226}Ra and ^{232}Th cannot be determined directly because the gamma emission probability for ^{238}U and ^{232}Th has low yield and ^{226}Ra emits gamma at energy of 186 keV (3.6%), which is interfered with that from ^{235}U at 185.9 keV of emission probability of 57.2% and hence difficult for a system to separate them. The energy lines of 63.20 keV and 92.60 keV for ^{234}Th, and 1001.03 keV for ^{234m}Pa were used to determine ^{238}U concentrations in samples. To calculate the ^{226}Ra activity concentration, both energy lines 295.22 keV and 351.93 keV for ^{214}Pb, and other three energies 609.31 keV, 1120.29 keV and 1764.49 keV for ^{214}Bi were used, provided that the samples are in radiochemical equilibrium. Gamma photons emitted with energy of 238.63 keV for ^{212}Pb, and other two energy lines 338.32 keV and 911.20 keV for ^{228}Ac were used to calculate the activity concentrations of ^{232}Th. The gamma emission with energy of 1460.63 keV was used for natural decay of ^{40}K to stable argon-40 (^{40}Ar), with photon intensity of 10.4%, by electron capture.

The activity concentrations of all radionuclides of interest are computed depending on the net peak counts (C_{NP}) for a particular energy line, after correction for a background and Compton contributions, of the respective nuclides. The formula utilized for calculation of activity concentration (A_c) at each spectral gamma energy is shown in Equation 3.7 (Durusoy and Yildirim, 2017; IAEA, 1989):

$$A_c(Bq.kg^{-1}) = \frac{C_{NP}}{B_I \times \varepsilon(E_\gamma) \times m} \quad (2)$$

where, N_{CP} is the net peak counts per second under the peak area of interest corrected for background effects in Equation 3.8 as:

$$N_{CP} = N_S - N_B \quad (3)$$

N_S is the total number of counts per second (cps) under the peak area of interest in the spectrum, N_B is the gross background counts per second (cps) under the peak of interest, B_I is branching ratio intensity, ε(Eγ) is the absolute photo-peak efficiency (%) of the detector, and m is the mass of the sample in kg. Again, the uncertainty was calculated using a formula:

$$\sigma_{A_c} = 100 x A_c x \sqrt{(\frac{\sigma_S}{S})^2 + (\frac{\sigma_{\varepsilon_i}}{\varepsilon_i})^2 + (\frac{\sigma_\gamma}{\gamma})^2 + (\frac{\sigma_q}{q})^2} \quad (4)$$

where, σ_{A_c} is the combined uncertainty, ε_i is the efficiency of the i^{th} energy line considered, σ_S is the net area efficiency, σ_γ is the branching ratio efficiency, σ_{ε_i} is the i^{th} energy line efficiency, and q is the sample quantity with σ_q as the sample quantity efficiency.

The concentrations of the radionuclides were calculated from the average weighted mean activities of each of their energy lines (A_{av}) as illustrated by the formula:

$$A_{av} = \frac{\sum_{i=1}^{N} \frac{A_{c_i}}{\sigma^2 A_{c_i}}}{\sum_{i=1}^{N} \frac{1}{\sigma^2 A_{c_i}}} = \frac{\sum_{i=1}^{N} \frac{A_{c_i}}{\sigma^2 A_{c_i}}}{\sigma_{A_{av}}} \quad (5)$$

where, A_{c_i} is the activity concentration as obtained for the i^{th} energy line,

$\sigma_{A_{c_i}}$ is uncertainty in activity concentration of the i^{th} energy line, and

$\sigma_{A_{av}}$ is uncertainty of the weighted average activity concentrations of all energy lines.

From Equation 5 above, the uncertainty of weighted average activity ($\sigma_{A_{Av}}$) from multiple selected was calculated as:

$$\sigma_{A_{Av}} = \sqrt{\frac{1}{\sum_{i=1}^{N} \frac{1}{(\sigma_{A_i})^2}}} \quad (6)$$

III. RESULTS AND DISCUSSIONS

4.1 Stockpiles of mine 1

The weighted mean of activity concentrations for these samples were calculated using equation 5 above and are shown in Table 1. The soil samples were coded S01, S02, S03, …, S13 and S14.

Table 1: The activity concentrations associated with soil samples from study area B.

Activity concentrations (Bq.kg^{-1})				
Sample ID	**^{226}Ra**	**^{238}U**	**^{232}Th**	**^{40}K**
S01	87±1	141±5	107±1	821±14
S02	195±1	211±6	183±1	626±13
S03	91±1	425±8	530±3	543±12

S04	1019±11	744±38	431±9	3429±78
S05	1625±6	316±8	124±1	1025±20
S06	2722±10	337±10	206±2	1053±25
S07	1789±7	321±9	124±1	1034±21
S08	1461±53	1024±24	502±4	889±53
S09	1955±7	406±9	140±1	1006±23
S10	3484±13	1188±17	157±3	886±25
S11	31±1	60±3	40±1	135±3
S12	2791±10	653±12	107±1	968±23
S13	2248±8	347±9	170±1	895±22
S14	2745±11	1622±23	191±2	1795±37
Min	31±1	60±3	40±1	135±3
Max	3484±13	1622±23	530±3	3429±78
Ave.	1589±10	557±13	215±2	1079±26

The weighted activity concentrations for ^{226}Ra ranged from 31±1 Bq.kg^{-1} to 3484±12 Bq.kg^{-1} with an average of 1589±10 Bq.kg^{-1}. The weighted activity concentrations for ^{238}U were between 60.80±3 Bq.kg^{-1} and 1622±23 Bq.kg^{-1} with an average of 557±13 Bq.kg^{-1}. ^{232}Th had weighted activity concentrations measured between 40±1 Bq.kg^{-1} and 530±3 Bq.kg^{-1} with an average of 215±2 Bq.kg^{-1}. The non-series ^{40}K radionuclide was measured with weighted mean concentrations between 135±3 Bq.kg^{-1} and 3429±78 Bq.kg^{-1}, and an average of 1079±26 Bq.kg^{-1}.

^{226}Ra was determined with lowest activity concentration in sample S11 and highest in sample S10 weighted activity concentrations, as well with the highest average value. Except S14, the samples not collected from tailings show significantly low concentrations in comparable to the values for tailings. Only sample S14 was collected from considerable distance but close to the tailings and has a recorded activity concentrations for both ^{226}Ra and ^{238}U as: 3484±13 Bq.kg^{-1} and 1188±17 Bq.kg^{-1}, respectively. The high concentrations may be attributed to the facts that radionuclides were carried by wind-blown dusts, containing uranium and radium deposits, from tailings towards the sample locus point. Therefore, S14 collection point was considered a ^{238}U and ^{226}Ra hotspot. Another sample of interest was S11, collected from tailings but recorded low activities ranging from a minimum of 31±1 Bq.kg^{-1} for ^{226}Ra to 135±3 Bq.kg^{-1} for ^{40}K. This evidence could be attributed to leaching of radionuclides in the soil, especially the soluble ^{226}Ra nuclide resulting into radon accumulation underground. The weighted activity concentrations for these nuclides were found to be above the world's average values of 35, 33, 45 and 420 Bq.kg^{-1} for ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K, respectively.

4.2 Stockpiles of mine 2

The aim for collecting the samples from this mine's tailings, located at about 36 km away from residential town of Arandis, was to measure the possible exposure contributions from the background radiations. The samples from mine 2 were coded SU_T1, SU_T2, SU_T3, …, SU_T11 and SU_T12. The weighted of radioactivity concentrations measured were calculated as well using equation 5 above and are shown in Table 2.

Table 2 : Activity concentrations for soil samples collected from first mine (section D).

Sample ID	Activity Concentration (Bq.kg^{-1})			
	^{226}Ra	^{238}U	^{232}Th	^{40}K
SU_T1	2112±8	810±14	325±2	1301±26
SU_T2	4547±17	310±13	314±2	1300±34
SU_T3	2337±9	430±11	291±2	1282±27
SU_T4	7867±81	3583±150	1217±20	6653±15
SU_T5	2755±10	691±13	400±3	1200±27
SU_T6	10538±113	1214±66	824±16	7435±180
SU_T7	3441±13	301±12	272±2	1397±33
SU_T8	3673±14	536±13	252±2	1236±29
SU_T9	2280±9	420±11	314±2	1162±24
SU_T10	1371±5	296±9	178±1	1228±24
SU_T11	9970±36	972±22	571±4	1395±44
SU_T12	2073±8	541±12	275±2	1111±25
Min	1371±5	296±9	178±1	1111±25
Max	10538±113	3583±150	1217±20	7435±180
Ave.	4414±27	842±29	436±5	2225±52

The results show that ^{226}Ra weighted activity concentrations was between 1371±5 Bq.kg^{-1} and 10538±113 Bq.kg^{-1}, with an average of 4414±27 Bq.kg^{-1}. ^{238}U level varied from 296±9 Bq.kg^{-1} to 3583±150 Bq.kg^{-1}, with an average of 842±29 Bq.kg^{-1}. The calculated radioactivity concentrations for both ^{232}Th and ^{40}K were 178±1 Bq.kg^{-1} to 1217±20 Bq.kg^{-1} and 1111±25 Bq.kg^{-1} to 7435±180 Bq.kg^{-1}, respectively. Their average values were 436±5 Bq.kg^{-1} for ^{232}Th and 2225±52 Bq.kg^{-1} for ^{40}K. The concentrations of ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K in the samples were higher than world's average levels 35, 33, 45, 420 Bq.kg^{-1}, respectively (UNSCEAR, 2018).

The minimum and maximum activity concentrations recorded were 178±1 Bq.kg^{-1} in SU_T10 and 10538±113 Bq.kg^{-1} in SU_T6 for ^{232}Th and ^{226}Ra, respectively. It was shown by this study that the lowest determined activity concentration in tailing's soil was higher than the world average reported by UNSCEAR. Thorium series consists of thoron product and other two soluble isotopes of radium which can be dangerous to human if inhaled or ingested. The high concentration of radium in samples contributed to high rate of radon emanation into the atmosphere, ground and surface water as well and if the necessary measures were not put in place, this may contribute to exposures in plants, animals and eventually human.

4.3 Comparisons of determined average activity concentrations of NORM for samples

The minimum and maximum values for measured radioactivity concentrations of NORM in samples are shown in Table 3 and are compared to other studies as well to world's average values.

Table 3 : The tabulated average values of activity concentrations (Bq.kg^{-1}) associated with samples from all sections.

Study area		Activity concentrations (Bq.kg^{-1})				References
		^{226}Ra	^{238}U	^{232}Th	^{40}K	
Mine 1 soil		1589±10	557±13	215±2	1079±26	Current study
Mine 2 soil		4414±27	842±29	436±5	2225±52	Current study
Gold Mine, RSA		-	785.3 ± 13.7	43.9 ± 1.0	427.0 ± 13.1	(Kamunda et al., 2016)
Tin Mine, Bangka Island	Range	2.47 – 4231.06	5.41 – 5411.27	2.91 – 7543.46	2.79 – 217.43	(Murniasih et al., 2021)
Walvis Bay, Namibia	Ave	99.59±24.39	-	90.90±31.99	553.07±107.17	(Njinga et al., 2016)
Henties Bay, Namibia	Ave	175.59±0.92	-	40.17±27.00	349.66 ± 8.00	(Onjefu et al., 2017)
bauxite ore deposit, Cameroon	Ave	125	99	157	671	(Nguelem et al., 2016)
World	Ave (Range)	33 (16 – 110)	35	45 (11 – 64)	420 (140 – 850)	(UNSCEAR, 2000; UNSCEAR, 2018)

This study has shown that a significantly higher value for ^{226}Ra concentrations was determined at mine 2 with a concentration level of 4414±27 Bq.kg^{-1} and as well greater than all concentrations found in other studies. The concentration for ^{226}Ra in mine 1 (at 1589±10 Bq.kg^{-1}) was greater than all values except for a study conducted in a Tin Mine in Bangka Island (Murniasih *et al.*, 2021). Both mining areas 1 and 2 show high concentration of potassium-40, greater than all values in all other studies. The potassium level in the mines had averages of 1079±26 and 2225±52 Bq.kg^{-1}, greater than both world average and ranges shown by the United Nations Scientific Committee on the Effects of Atomic Radiations (2018). The ^{238}U average concentrations for both mines were greater than the world's average of 35 Bq.kg-1 and fall within the range of values in a study by Murniasih *et al.* (2021) and significantly comparable to a study conducted in a Gold Mine by Kamunda *et al.* (2016). ^{232}Th recorded values at 215±2 and 436±5 Bq.kg^{-1} for mine 1 and 2, respectively. These concentrations are greater than those from other studies conducted by researchers such as Onjefu *et al.* (2017), Njinga *et al.* (2016) in the same region. The thorium concentration was also

found to be within the range of activity concentration values in a study on a Tin Mine by Bangka Island by Murniasih *et al.* (2021). Only the concentration of thorium in mine 1 was found to be significantly comparable to a concentration found in a bauxite ore deposit, Cameroon, by Nguelem *et al.* (2016) but still higher than both the world average and range in UNSCEAR (2018)

IV. CONCLUSION

The measured weighted average concentrations for all radionuclides of interest in the study show significantly high values from all mines' tailings. This was proved by the ratio for activity concentration for ^{226}Ra (1589±10 Bq.kg^{-1}) to that of ^{238}U (557±13 Bq.kg^{-1}) approximated 3 and 5 for samples of mines 1 and 2, respectively. Hence, ^{238}U and its daughter ^{226}Ra were inequilibrium. Human activities such mining and milling, wind blowing, and rainfall and other natural factors were possible causes of the inequilibrium between these two nuclides. Hence, this was considered a disturbed soil in the mining zone and therefore the weighted activity concentration for ^{238}U could not be quantified by that of ^{226}Ra. The study had also shown very low concentrations of a soluble ^{226}Ra radionuclide in some samples, which shows that the nuclide may be absorbed into the ground water through leaching process which may result in high accumulation of radon in the soil. Radon and thoron gases may be transferred into the atmosphere and also into buildings of the nearest inhabitable residences in the surrounding area thereby increasing human exposure rate in the region.

In spite of that, the study has shown that mine tailings contribute to an increase in background radiation in the environment due to the fact that they are uncovered and deposited in the open environment, and can be blown away by wind into the atmosphere, nearby residential dwellings. Mining companies may decrease windblown atmospheric exposure by using water tanks mounted on mining equipment to sprinkle on top of the soil or cover the tailing with a canvas cloth.

Ethical Approval

The article has not been submitted to a Committee on Publication Ethics (COPE) and therefore no ethics approval received so far.

Compliance with ethical standards

The authors of this article declare that they have no known financial interests or relationship that could appear to be of influence to the research work discussed and reported in this paper. The research involves only human participants.

Consent to participate and publish

The authors have received explicit consent to submit and that they obtained consent from the responsible authorities at the institute/organization where the work has been carried out, **before** the work is submitted.

Consent to publish

All authors have consensus agreement to publish this research work.

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Competing Interests

Authors Manny Mathuthu and Vaino Indongo declare they have no competing financial interests. The authors have no relevant financial or non-financial interests to disclose. All authors certify that they have no affiliations with or involvement in any organization or entity with any financial interest or non-financial interest in the subject matter or materials discussed in this manuscript.

Author Contributions

All authors contributed to the study conception and design. Material preparation, data collection and analysis were performed by both Vaino Indongo and Manny Mathuthu. The first draft of the manuscript

was written by Vaino Indongo. Supervision was carried by Manny Mathuthu and all authors commented and approved on previous versions of the manuscript.

Availability of data and materials.

The data used in this article will be provided upon request by the journal and/or reviewers of this article.

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