Radiation contamination in gold mine tailings soil samples using HPGe spectrometry

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Abstract. An assessment of the radiation concentration in abandoned mines located near settlements in the west of Johannesburg was carried out. In this study, a gamma spectrometre was used to measure the activity concentrations of radionuclides in the soil samples. The activity concentrations for ^{226}Ra, ^{232}Th, and ^{40}K were 338.44±3.48, 10.06±0.68, and 126.15±10.90 Bq/kg, respectively. The radiological indices that surpass their global recommended values. The mean of radiological indices in descending order, AGDE > Ra_{Eq} > ADR > ELCR > AEDE > I_a with values 1129.54 ± 46.63 μSv/y > 363.20 ± 7.81 Bq/kq > 500.56 ± 15.90 nGy/h > 5.62 ± 0.20×10^{-3} > 1.57±0.06 mSv/y > 1.45±0.30. Therefore, from the perspective of radiation exposure, using this soil for building is not safe.

1. Introduction

South Africa is endowed with numerous mineral resources, especially metals and coal. Gold has been mined in the west of Johannesburg since the late 1800s [1]. Gold mining began in 1886 [2], leaving behind an area of approximately 400 km^2 of mine waste called tailings. The 270 dams and 380 mine tailings are reported to contain approximately 6 billion tonnes of pyrite [3] and between 430,000 and 600,000 tonnes of low-grade U_3O_8 [3, 4]. On average, the tailings in the Witwatersrand Basin have uranium concentrations up to 100 mg/kg U_3O_8. This uranium amount is comparable to or even higher than that from certain uranium mines in Namibia or Germany [4].

The mining industry has had positive and negative effects on the South African economy and GDP [5]. Uranium is a radioactive and chemotoxic metal. This amount of uranium held in tailings can have a detrimental effect on the health of the residents living in the vicinity of the mining tailings during the windy season. Clouds of dust laden with ^{238}U, ^{232}Th, ^{40}K particles [6] reach settlements, and residents get exposure to ionizing radiation through inhalation of dust. The South African tailings are not only contaminated with radioactivity but also heavy hazardous metals such as arsenic, chromium and nickel [1]. The heavy metals and radioactive dust particles have a detrimental synergy when they settle in the lungs. They can cause lung issues such as silicosis, tuberculosis, chronic obstructive pulmonary disease (COPD), pneumoconiosis, asthma, emphysema, asbestosis, lung cancer, mesothelioma, and haematological damage [5]. Radiation affects the human body in two ways: externally through γ-radiation and internally through ^{222}Rn gas. Ionizing radiation causes cellular changes as it can

damage DNA; however, the body naturally recovers from radiation. Chromosomal abnormalities are among the mutations that can result from improper repair of DNA damage [7]. The nuclear decays inside the body emit α-, β-, and γ- radiations [8, 9]. Natural background radiation exposure is ubiquitous in the terrestrial environment. The radiation exposure can be due to primordial or anthropogenic radiation contamination. The exposure to radioactive tailings dust is geogenic radiation exposure [10] and anthropogenic radiation exposure, such as medical applications [11], and fallout from nuclear weapons tests and power plant failures [12, 13]. According to the International Commission on Radiological Protection (ICRP), the uranium content in the soil should not exceed 100-200 Bq/kg, according to the US Environmental Protection Agency (USEPA) 190 Bq/kg, according to the National Nuclear Regulatory of South Africa (NNR) 500 Bq/kg and according to the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) 35 Bq/kg.

Communities in Riverlea and Carletonville, south of Johannesburg and the far west of Johannesburg, respectively, complain about the dust pollution from the gold tailings in their areas. The study seeks to find out whether the tailings have radiation or heavy metal contamination in the dust. This study will analyze the concentration of radioactivity and heavy metals in the tailings. The radioactivity concentration and radiological hazard indices will be presented in this report.

2. Materials and Method

2.1. The soil sample collection and preparation

The soil samples were collected from the gold mines tailings in Crown Mines (CMO), Main Reef Gold (CMRG), Roodepoort (RDP), and Kloof (KLF) in the west of Johannesburg, and then dried in an oven at 105 ^{o}C, crushed, homogenized, and pulverized into powder. The samples were then taken to iThemba LABS for analysis. Empty Marinelli beakers were weighed, and the differences in masses were used to determine the masses of the soil samples. The samples were stored in sealed Marinelli beakers for 42 days to establish secular radioactive equilibrium between radon and its daughters before γ-spectroscopy analysis, as naturally occurring radioactive materials (NORM) contain radon gas that can escape from the samples, introducing disequilibrium. The background radiation of the γ-spectrometre was negligible, as the detector was covered with a 10 cm thick lead layer.

2.2. The analysis of soil samples

The investigation used a coaxial high-purity germanium (HPGe) γ-ray spectrometre to determine the activity concentrations of ^{238}U, ^{232}Th, and ^{40}K in soil. The detector, linked to Genie-2000 analysis software, had an energy resolution (FWHM) of 1.9 keV at the 1332 keV γ-ray line of the ^{60}Co source. Since the analysis was done after the samples had established secular equilibrium, the γ-ray lines of 186.20 keV (^{226}Ra), 351.9 keV (^{214}Pb), and 1765 keV (^{214}Bi) were used to estimate ^{238}U concentration. For ^{232}Th, the γ-lines 583.1 keV (^{208}Tl) and 911.20 keV (^{228}Ac) were used, and a 1460.80 keV γ-line was to determine ^{40}K concentration in samples.

3. Numerical Calculations

3.1. The efficiency of the detector and energy calibration curves

The gamma detector used in this analysis is the Lower Energy Germanium (LEGe) detector. The LEGe is a high-resolution germanium (HPGe) detector design optimized for detecting gamma rays at low energies. For efficiency calculation, the energy sources 45.539 keV for ^{210}Pb, 59.5 keV for ^{241}Am, 88 keV for ^{109}Cd, 122.1 keV for ^{57}Co, 165.9 keV for ^{139}Ce, 279.2 keV for ^{203}Hg, 391.7 keV for ^{113}Sn, 514 keV for ^{85}Sr, 661.65 keV for ^{137}Cs, 898 keV for ^{88}Y, 1173.2 keV for ^{60}Co, 1332.4 keV for ^{60}Co, and 1836.1 keV for ^{88}Y were used. The emission probabilities of

different radionuclides were taken from various sources in the literature [13]. Figure 1(a) and Figure 1(b) show the efficiency and energy calibration curves, respectively.

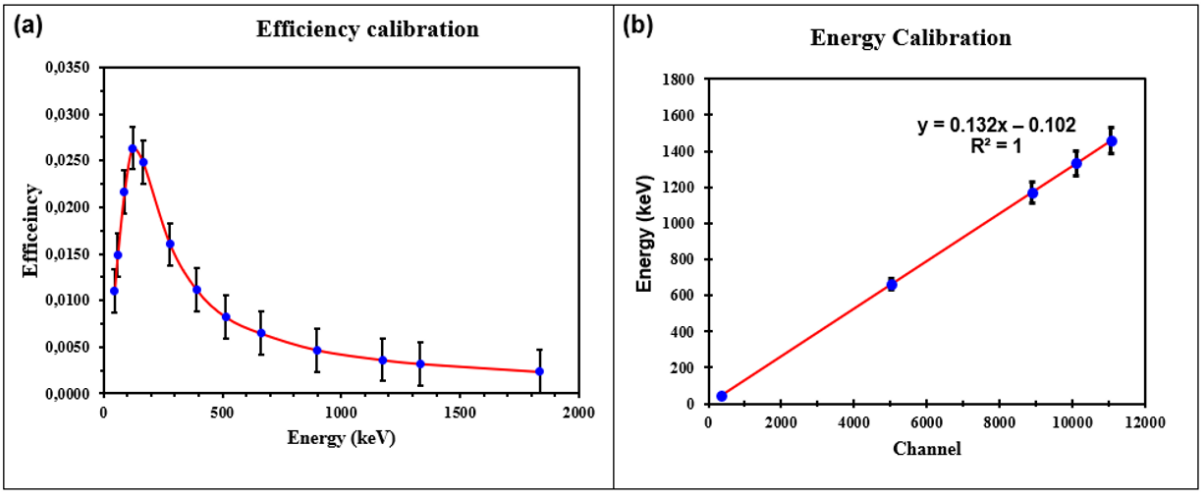

Figure 1: (a) Efficiency of detector and (b) energy calibration for characteristic γ-ray energies.

3.2. The activity concentration in soil samples

Each sample was counted for 25200 seconds to reduce statistical uncertainty. The activity concentrations in the measured samples were computed using the following relation [14, 15]:

$$A(Bq/kg) = \frac{C_s - C_b}{I_\gamma \varepsilon m} \tag{1}$$

where C_s is the activity of the sample, and C_b is the background activity, ϵ is the detector efficiency of the specific γ - radiation, ε is the absolute detector efficiency of the specific γ-ray, and m is the mass of the sample in kilograms. I_γ is the emission probability of a specific energy photopeak.

3.3. Evaluation of radiation indices

Following the calculation of the concentration of specific activity in the samples, the radiological indices of the samples were estimated from the following relations. The radium equivalent activity (Ra_{Eq}) was calculated using Equation (02) [14, 7]:

$$Ra_{Eq}(Bq/kg) = C_{Ra} + 1.43s + C_{Th} + 0.077C_K \tag{2}$$

The indoor and outdoor absorbed dose rate (ADR) due to naturally occurring radioactive materials in air 1 metre above the ground is calculated using Equation (03) and Equation (04) [7, 16]:

$$D_{In}(nGy/h) = 0.920C_{Ra} + 1.100C_{Th} + 0.0810C_K \tag{3}$$

$$D_{Out}(nGy/h) = 0.462C_{Ra} + 0.604C_{Th} + 0.0417C_K \tag{4}$$

where the coefficients of C_{Ra}, C_{Th}, and C_K are dose conversion factors in nGy/h per Bq/kg. The annual effective dose equivalent (AEDE) indoors and outdoors was estimated with Equations (05) and (06) as follows [7]:

$$AEDE_{In}(mSv/y) = D_{In}\times T\times 0.8\times F \tag{5}$$

$$AEDE_{Out}(mSv/y) = D_{In}\times T\times 0.2\times F \tag{6}$$

where T is the number of hours in a year (365× 24h = 8760 hours). F is a conversion factor from nGy/h to mSv/y with a value of 0.7×10^{-6}. The excess lifetime cancer risk (ELCR) in the indoor and outdoor environment was estimated using Equations (07) and (08) [7, 16, 17, 18]:

$$ELCR_{In} = AEDE_{In}DL\times RF \tag{7}$$

$$ELCR_{Out} = AEDE_{Out}DL\times RF \tag{8}$$

where DL is the life expectancy, which is about 70 years, and RF is the risk factor, which is given as 0.05 Sv^{-1}. The annual gonadal dose equivalent (AGDE) due to the specific activities of ^{226}Ra, ^{232}Th, and ^{40}K was calculated using Equation (09) [19]:

$$AGDE(\mu\text{Sv/y}) = 3.09C_{Ra} + 4.18C_{Th} + 0.314C_K \tag{9}$$

The alpha index (I_α), which is used to estimate α-radiation from building materials is estimated using Equation (10) [20]:

$$I_\alpha = \frac{C_{Ra}}{200} \tag{10}$$

In the above equations, ^{226}Ra, ^{232}Th, and ^{40}K represent the radionuclide concentrations.

4. Results and discussion

Figure 2 shows a typical example of a γ-spectrum for soil samples analysis with the HPGe detector. The samples from four different tailings were collected and analyzed at iThemba LABS. The concentrations are dominated by ^{226}Ra and partly followed by ^{40}K in the samples, as shown in Table 1.

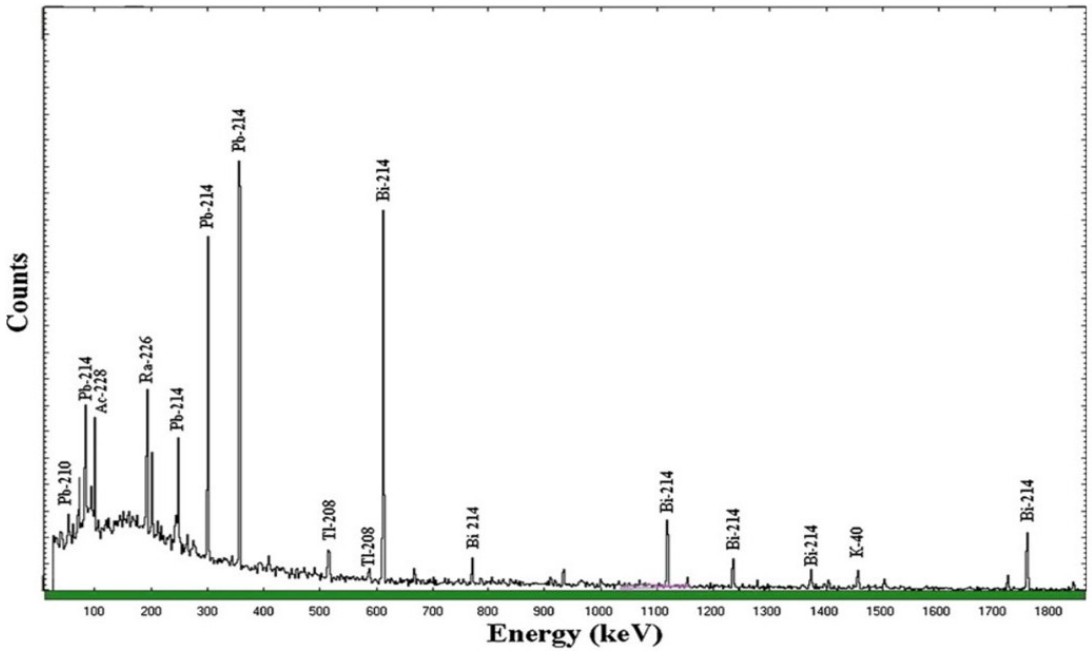

Figure 2: Typical HPGe γ-ray spectrum due to the γ-emitting NORM [21].

In the samples, the ^{226}Ra (^{238}U) ranged from 149.62±4.72 to 452.28±8.02 Bq/kg, with a mean of 338.44±3.48 Bq/kg, which is higher than the permissible limit of 35 Bq/kg [7]. The concentration of ^{40}K ranged from 113.34±20.80 Bq/kg to 145.79±22.51 Bq/kg with a mean of 126.15±10.90 Bq/kg, which is 13 times the concentration of samples but ^{40}K is below the permissible limit of 400 Bq/kg, and ^{232}Th ranged from 8.25±1.31 to 11.20±1.03 Bq/kg with a mean of 10.06±0.68 Bq/kg. The mean concentration of ^{226}Ra is 34 times that of ^{232}Th, and they follow this decreasing order: ^{226}Ra>^{40}K>^{232}Th.

Table 1: The specific activity concentrations in the tailings soil samples.

Mine Tailings Location	Sample Code	Specific Activity (Bq/kg)		
		^{226}Ra (^{238}U)	^{232}Th	^{40}K
Johannesburg, West	KLF	149.62±4.72	10.31±1.48	145.79±22.51
	CMO	303.02±6.60	8.25±1.31	113.34±20.80
	CMRG	448.86±8.00	10.46±1.56	117.92±21.97
	RDP	452.28±8.02	11.20±1.03	127.54±21.85
	Mean	338.44±3.48	10.06±0.68	126.15±10.90
	Minimum	149.62±4.72	8.25±1.31	113.34±20.80
	Maximum	452.28±8.02	11.20±1.03	145.79±22.51

The radiological indices were estimated, and the results are presented in Table 2. The CMO and KLF samples have the Ra_{Eq} below the permissible limit of 370 Bq/kg [7]. CMRG and RDP samples had mean Ra_{Eq} values of 475.61±1.91 and 478.10±1.76 Bq/kg, respectively. In all samples, indoor and outdoor ADR were above permissible limits of 84 and 59 nGy/h, respectively. The total AEDE is higher than the sum of 0.48 mSv/y and even higher than the global average of 1 mSv/y. The Indoor and outdoor ELCR were high in all samples. The AGDE is high in all the samples, with the maximum mean value of 1484.42±26.10 μSv/y in RDP and the minimum of 551.22±17.39 μSv/y in KLF samples. The mean I_α index in KLF samples was less than unity at 0.75±0.02, and the rest of the samples had I_α more than unity.

Table 2: The statistics of radiological indices and global recommended values (RV).

Tailing Site	Radiological Indices	Min	Max	Median	Mean	STDev	RV
CMO	Ra_{Eq}(Bq/kg)	245.14±6.12	440.06±8.98	316.78±6.42	323.50±7.04	43.90	370
	ADR_{In} (nGy/h)	222.65±5.65	399.97±8.16	287.37±5.81	293.98±5.94	39.98	84
	ADR_{Out}(nGy/h)	113.52±2.84	203.65±4.16	146.42±2.97	149.73±3.27	20.31	59
	$AEDE_{Tot}$(mSv/y)	1.23±0.01	2.21±0.14	1.59±0.10	1.63±0.02	0.22	0.48
	$ELCR_{Ind}(\times10^{-3})$	3.83±0.10	6.87±0.14	4.94±0.04	5.05±0.11	0.69	1.16
	$ELCR_{Out}(\times10^{-3})$	0.49±0.01	0.87±0.02	0.63±0.01	0.64±0.01	0.09	0.29
	$AGDE_{Out}(\mu Sv/y)$	764.13±19.27	1368.26±28.13	983.76±20.06	1006.34±22.10	136.10	300
	I_α	1.12±0.03	2.07±0.04	1.48±0.03	1.51±0.03	0.22	≤1
CMRG	Ra_{Eq}(Bq/kg)	236.77±6.25	702.52±10.23	476.93±6.08	475.61±1.91	114.36	370
	ADR_{In} (nGy/h)	217.37±5.68	644.74±3.91	437.11±5.48	436.52±1.73	104.94	84
	ADR_{Out}(nGy/h)	109.56±2.90	324.465±4.73	220.36±2.81	219.91±0.88	52.81	59
	$AEDE_{Tot}$(mSv/y)	0.67±0.05	1.99±0.12	1.35±0.06	1.35±0.02	0.32	0.48
	$ELCR_{Ind}(\times10^{-3})$	1.88±0.07	5.57±0.19	3.78±0.09	3.78±0.03	0.91	1.16
	$ELCR_{Out}(\times10^{-3})$	0.47±0.14	1.39±0.38	0.95±0.19	0.94±0.02	0.23	0.29
	$AGDE_{Out}(\mu Sv/y)$	736.11±19.61	2174.44±31.80	1479.65±18.95	1476±26.51	353.87	300
	I_α	1.11±0.03	3.40±0.05	2.24±0.03	2.26±0.03	0.55	≤1
RDP	Ra_{Eq}(Bq/kg)	142.74±4.78	1406.45±14.16	458.66±8.11	478.10±1.76	266.97	370
	ADR_{In} (nGy/h)	130.86±4.42	1292.61±13.04	420.91±7.47	438.75±7.86	245.46	84
	ADR_{Out}(nGy/h)	66.07±2.23	650.43±6.56	212.08±3.76	221.08±3.87	123.46	59
	$AEDE_{Tot}$(mSv/y)	0.72±0.02	7.14±0.06	2.23±0.04	2.42±0.04	1.36	0.48
	$ELCR_{Ind}(\times10^{-3})$	2.25±0.08	22.19±0.22	7.23±0.13	7.53±0.13	4.21	1.16
	$ELCR_{Out}(\times10^{-3})$	0.28±0.01	2.79±0.03	0.91±0.02	0.95±0.02	0.539	0.29
	$AGDE_{Out}(\mu Sv/y)$	444.81±15.16	4361.69±44.12	1423.89±25.37	1484.42±26.10	827.58	300
	I_α	0.65±0.02	6.80±0.07	2.17±0.04	2.26±0.04	1.30	≤1
KLF	Ra_{Eq}(Bq/kg)	80.03±3.68	301.23±7.50	86.94±4.00	175.57±2.18	113.66	370
	ADR_{In} (nGy/h)	73.17±3.34	276.11±6.79	79.42±3.63	160.80±4.95	104.42	84
	ADR_{Out}(nGy/h)	27.20±7.72	138.49±11.89	40.39±8.12	81.48±9.69	52.71	59
	$AEDE_{Tot}$(mSv/y)	0.40±0.02	1.53±0.04	0.44±0.04	0.89±0.03	0.58	0.48
	$ELCR_{Ind}(\times10^{-3})$	1.26±0.06	4.74±0.02	1.36±0.06	2.76±0.09	1.79	1.16
	$ELCR_{Out}(\times10^{-3})$	0.16±0.06	0.60±0.12	0.17±0.07	0.35±0.09	0.539	0.29
	$AGDE_{Out}(\mu Sv/y)$	252.791±11.85	943.72±23.70	274.39±25.37	551.22±17.39	355.07	300
	I_α	0.32±0.02	1.32±0.03	0.34±0.02	0.75±0.02	0.52	≤1

5. Conclusion

Soil samples from gold tailings in the west of Johannesburg were analysed using a high-resolution HPGe γ-ray detector to assess radioactivity concentrations and associated health hazard indices.

The analysis showed that only geogenic radionuclides and no anthropogenic radionuclides were detected. The concentrations of ^{226}Ra, ^{232}Th, and ^{40}K were 338.44±3.48, 10.06±0.68, and 126.15±10.90 Bq/kg, respectively. The results showed that the average activity concentration in the soil was dominated by the radionuclide ^{222}Ra of the ^{238}U decay series, and ^{222}Ra was higher than the global average at all sites investigated.

The different radiological indices surpassed the recommended values (RV) as discussed. In descending order the averages are AGDE > $\mathrm{Ra_{Eq}}$ > ADR > ELCR > AEDE > I_a with values 1129.54 ± 46.63 μSv/y > 363.20 ± 7.81 Bq/kq > 500.56 ± 15.90 nGy/h > 5.62 ± 0.20×10^{-3} > 1.57±0.06 mSv/y > 1.45±0.30. The study suggests the prohibition of the use of this soil for construction purposes. Uranium, with its high content, can cause skin irritation and reactions, but conclusive conclusions cannot be drawn until data on heavy metals is available.

The uranium concentrations in the samples are acceptable according to the NNR limit value of 500 Bq/kg, but unacceptable according to UNSCEAR and ICRP limit values of 35 and 100-200 Bq/kg, indicating that South African nuclear contamination legislation needs reconsideration and harmonization to meet global standards.

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