Radiometric analysis of the gold tailings using a high-resolution LEGe γ-detector

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Abstract

Soil samples were collected from abandoned mines located west of Johannesburg. An assessment of the radioactivity con- centration was conducted. The comprehensive radiological indices were then evaluated. In this study, a coaxial LEGe HPGe γ-spectroscopy was used to measure the γ-signals emanating from the soil samples. For radioactivity concentration, the results showed that the mean activity concentrations in some tailings exceeded the global average for certain radionuclides, and the con- centrations for 226Ra, 232Th, and 40K were 338.44\$$\pm 3.48, 10.06 \pm 0.68, and 126.15 \pm 10.90 Bq/kg, respectively. Theradiologicalindicesthatsurpasstheirgloba$
$RaEq > ADR > ELCR > AEDE > Ia > I$ \$. The findings are tabulated, plotted, discussed and compared with controlling body limits. These indices exceeded the recommended values; the gold tailings pose a significant health risk to the local population, and the soil should under no circumstances be used for building purposes.

Full Text

Preamble

Radiometric analysis of gold tailings was conducted using a high-resolution LEGe γ-detector. Soil samples were collected from abandoned mines located west of Johannesburg to assess radioactivity concentrations and evaluate comprehensive radiological indices. In this study, coaxial LEGe HPGe γ-spectroscopy was used to measure γ-signals emanating from the soil samples. Results showed that mean activity concentrations in some tailings exceeded global averages for certain radionuclides, with concentrations for ^{226}Ra, ^{232}Th, and ^{40}K of 338.44\$$\pm 3.48, 10.06 \pm 0.68, and 126.15 \pm 10.90 Bq/kg, respectively. Theradiologicalindicessurpassedgloballyre$
$AGDE > RaEq > ADR > ELCR > AEDE > I$ \$ > Iγ. These findings are tabulated, plotted, discussed, and compared with regulatory limits. Since the indices exceeded recommended values, the gold tailings pose significant health

risks to the local population, and the soil should under no circumstances be used for building purposes.

Keywords: Absorbed Dose, Equivalent Dose, Excess Lifetime Cancer Risk, Radioactivity, gold tailings, Soweto.

Introduction

Radiation is a natural phenomenon, and life on Earth involves continuous exposure to background radiation from geogenic, cosmic, and anthropogenic sources [39]. Naturally occurring radioactive materials (NORM) are present in soil, water, and particulate dust, while technologically enhanced background radiation is referred to as TENORM [51]. TENORM results from anthropogenic activities such as mining, mineral extraction, fossil fuel combustion, and fertilizer production, which contribute significantly to environmental radioactive materials [2, 5, 32, 48, 51].

South Africa has long been a leader in gold production, though output has since declined. The country is also a leading producer of chromium, platinum, and rhodium, ranking as the second-largest producer of zirconium, the sixth-largest exporter of coal, and the third-largest supplier of iron ore by value [36]. This mining activity has created jobs and export earnings, contributing significantly to GDP and making South Africa one of the continent’ s most developed nations [37]. Gold mining historically overlooked uranium, typically treating it as waste. Both metals occur in the same geological settings, with uranium being significantly more abundant; specifically, the Witwatersrand gold fields yield 100 grams of uranium for every 10 grams of gold extracted [52]. Today, this uranium waste remains in tailings dams, where it can be blown as dust into nearby residential areas during windy seasons [3, 30, 53].

Assessments of radioactivity in disrupted environments are necessary for human safety and environmental health, as these environments are often contaminated with radionuclides such as ^{238}U and ^{232}Th, and non-series ^{40}K [22, 29, 33, 51]. Radionuclide concentrations vary geographically depending on geological and geophysical conditions [16, 21, 45]. Gold tailings in the Witwatersrand Basin have average uranium concentrations higher than those found in certain U-mine tailings dams in Namibia [52], which range from 45.9\$$\pm 3.0 to 1752.1 \pm$\$17.5 Bq/kg for ^{238}U [53]. Radiation affects the human body externally through γ-radiation and internally through α-radiation from inhaled radioactive ^{222}Rn gas that decays inside the body [25, 46]. Inhaling dust containing ^{222}Rn and its progeny greatly increases cancer risk [49], and exposure to gold tailings dust may cause various respiratory problems already experienced in Soweto neighborhoods [10, 38].

The studied gold tailings dams are located north of Soweto and potentially affect areas including Meadowlands East Zone 1 in the southwest, Orlando in the south, Fleurhof in the west, and Diepkloof in the far east. Diepkloof is also close to another tailings dam, Mooifontein (Crown Mines Tailings). Residents of Riverlea, Dobsonville, Diepkloof, and Meadowlands are particularly affected [24]. Phytoremediation attempts keep failing because bacteria and microbes necessary for life cannot survive in the contaminated areas due to acid in the tailings soils [24]; therefore, the tailings remain barren and will forever be epicenters of dust laden with hazardous elements such as arsenic, mercury, lead, and uranium [22]. With changing climate, uncertainty looms, leaving citizens caught between historically irresponsible uranium waste handling and future storms.

Based on numerous complaints about yellow dust pollution in Soweto [28, 30], this project aims to provide important baseline data on radiation concentrations in gold tailings, which have received limited attention to date, and enable probabilistic assessments of health hazard indices.

Materials and Methods

Sample Collection and Processing

A total of 20 soil samples were collected from gold mine tailings at the Consolidated Main Reef (CMR), west of Johannesburg and north of Soweto. Samples were stored in transparent plastic bags, labeled with sample IDs, dried in an oven at 105°C, crushed, homogenized, and pulverized into powder. Samples were then taken to NRF iThemba LABS for further preparation, processing, and analysis. At iThemba LABS, empty Marinelli beakers were weighed and filled with samples, and mass differences were used to determine sample masses. An attempt was made to keep sample masses as uniform as possible to minimize errors. Samples were stored in sealed Marinelli beakers for 42 days to establish radioactive secular equilibrium between radon and its daughters before γ-spectrometric analysis, as NORM contains radon gas that can escape from samples, thus introducing disequilibrium.

Analysis of Soil Samples

Activity concentrations of NORM were measured using Lower Energy Germanium (LEGe) high-resolution γ-ray spectrometry with 10 cm thick lead shielding on all sides and inner Cu and Sn lining to reduce background activity by about 95%. Gamma spectrometers require shielding against interfering background radiation, which for LEGe comes from three sources: (a) cosmic rays, (b) environmental radioactivity, and (c) internal radioactivity in construction materials and detectors themselves [19, 20]. Background radiation is measured using an empty Marinelli beaker with the same geometry as the samples once all setups are complete [13, 34]. The detector' s energy resolution (FWHM) was found to be 1.9 keV at the 1332 keV γ-ray line of a ^{60}Co source, following procedures from previous studies [11, 14].

The detector was connected to a data acquisition system applying Genie-2000 analysis software, version 2.0, for both γ-ray energy and radionuclide identification. Since analysis was performed after samples had established secular equilibrium, the γ-ray lines of 186.20 keV, 295.27 keV, 351.9 keV, 609.02 keV, 1120.16 keV, and 1765 keV were used to estimate ^{238}U (^{226}Ra) concentration. For ^{232}Th concentration, the γ-lines 583.1 keV and 911.20 keV were used, and a γ-line of 1460.80 keV was used for ^{40}K concentration in soil samples [1, 14, 51].

Numerical Calculations

Detector Efficiency and Energy Calibration Curves

This study used a Lower Energy Germanium (LEGe) high-purity germanium (HPGe) detector from Canberra, interfaced with a multichannel analyzer (MCA) via an electronic data acquisition system (Canberra DSA-1000 digital signal processing (DSP) system). The LEGe detector can detect gamma rays with energies ranging from 0.0 to 2.0 MeV. An IAEA multi-isotope source was used to calibrate the detector system' s energy resolution and efficiency by emitting known gamma-ray energies ranging from 45.54 keV for ^{210}Pb to 1836.10 keV for ^{88}Y at the same geometric position as the experimental samples. This energy range is equivalent to the range covered by the LEGe detector, from low energies to 2 MeV. The emission probabilities of these radionuclides were taken from various literature sources [50]. Fig. 1 [Figure 1: see original paper] shows the efficiency and energy calibration curves.

Fig. 1. Efficiency (left) and energy calibration for specific γ-rays (right).

Activity Concentration in Soil Samples

All measurements were made with samples in contact with the detector housing for 25,200 s, and spectral analysis was performed with Canberra-developed Genie 2000 software. Activity concentrations in the measured samples were computed using Equation (1) [23, 50]:

$$A(\text{Bq/kg}) = \frac{N_s - N_b}{I_\gamma \epsilon T m}$$

where A (Bq/kg) is the specific activity in a sample, N_s is the activity of the sample, N_b is the background activity, I_γ is the emission probability of a specific energy photopeak, ϵ is the detector efficiency of the specific γ-radiation, T is the accumulation time (s), and m is the mass of the sample in kilograms. Following calculation of specific activity concentrations, radiological indices including Radium Equivalent (RaEq), Absorbed Dose Rate (ADR), Annual Effective Dose Equivalent (AEDE), Excess Lifetime Cancer Risk (ELCR), Internal Hazard Index (HIn), External Hazard Index (HEx), Annual Gonadal Dose Equivalent (AGDE), Alpha Indexes (Iα), and Radioactivity Index (Iγ) were estimated using corresponding models from the literature.

Radium Equivalent Activity (RaEq)

The radium equivalent quantity was developed to express the gamma yield from the mixture of radionuclides in a soil sample and represents the activity levels due to ^{238}U, ^{232}Th, and ^{40}K. The radium equivalent activity was estimated using Equation (2) [23, 51]:

$$\text{RaEq (Bq/kg)} = C_{\text{Ra}} + 1.43C_{\text{Th}} + 0.077C_{\text{K}}$$

This index assumes that 370 Bq/kg of ^{226}Ra, 259 Bq/kg of ^{232}Th, and 4810 Bq/kg of ^{40}K produce the same gamma dose rates [51]. Its maximum recommended limit is 370 Bq/kg [35], which represents a safe limit to avoid radiation hazards. It provides information about potential biological effects of gamma radiation on living organisms and helps assess radiation exposure in each area.

Absorbed Dose Rate in Air (ADR)

The absorbed gamma dose rate measures the rate at which ionizing radiation from gamma rays is deposited in a specific location, providing insight into potential biological effects. The total absorbed dose rate due to naturally occurring radioactive materials (NORM) in air 1 meter above ground is calculated using the following equations. Absorbed dose rate indoors is estimated using Equations (3) and (4) [6, 51]:

$$D_{\text{Ind}}(\text{nGy/h}) = 0.920C_{\text{Ra}} + 1.100C_{\text{Th}} + 0.0810C_{\text{K}} \quad (3)$$

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

where 0.462, 0.604, 0.0417, 0.92, 1.1, and 0.081 are dose conversion factors in nGy/h per Bq/kg, and C_{Th}, C_{Ra}, and C_{K} are radionuclide concentrations for ^{226}Ra, ^{232}Th, and ^{40}K, respectively [15, 18, 51].

Annual Effective Dose Rate (AEDE)

A value of 0.7$×10^{-6}$ Sv/y is used by the UNSCEAR [51] report for the conversion coefficient from absorbed dose in air to effective dose received by adults, with 0.2 and 0.8 for outdoor and indoor occupancy factors, respectively [17, 18]. The effective dose rate per year should be less than 1 mSv/y [31]. Indoor and outdoor annual effective dose equivalents are estimated using Equations (5) and (6) [6, 51]:

$$\text{AEDE}_{\text{Ind}}(\text{mSv/y}) = \text{ADR}_{\text{Ind}} \times T \times 0.8 \times F \quad (5)$$

$$\text{AEDE}_{\text{Out}}(\text{mSv/y}) = \text{ADR}_{\text{Out}} \times T \times 0.2 \times F \quad (6)$$

where ADR is the absorbed dose rate (nGy/h), T is hours in a year (365$×$24 h = 8760 hours), and F is the conversion factor.

Excess Lifetime Cancer Risk (ELCR)

Excess lifetime cancer risk (ELCR) is used in radiation protection to estimate the potential increase in cancer risk due to exposure to ionizing radiation exceeding baseline risk. Indoor and outdoor ELCR values should average less than or equal to the global averages of 1.16$×10^{-3}$ and 0.29$×10^{-3}$, respectively. Indoor ELCR was estimated using Equation (7) [4, 6, 47, 51]:

$$\mathrm{ELCR}_{\mathrm{Ind}} = \mathrm{AEDE}_{\mathrm{Ind}} \times \mathrm{DL} \times \mathrm{RF} \quad (7)$$

Outdoor ELCR was estimated using Equation (8) [4, 47]:

$$\mathrm{ELCR}_{\mathrm{Out}} = \mathrm{AEDE}_{\mathrm{Out}} \times \mathrm{DL} \times \mathrm{RF} \quad (8)$$

where DL is life expectancy (approximately 70 years) and RF is the risk factor (0.05 Sv^{-1}).

Internal Hazard Index (HIn)

The internal hazard index assesses potential radiation exposure and hazards associated with intake of radioactive materials. It determines acceptable limits of radioactive material intake and whether additional measures such as medical follow-up or radiation protection are necessary. Inhaling radon and thoron gases can be hazardous to respiratory organs [43]. This index is calculated using Equation (9) [9, 41]:

$$H_{\mathrm{In}} = \frac{C_{\mathrm{Ra}}}{185} + \frac{C_{\mathrm{Th}}}{259} + \frac{C_{\mathrm{K}}}{4810} \quad (9)$$

For safe use of building materials in shelter construction, the index should be less than one.

External Hazard Index (HEx)

External gamma radiation dose is the amount of ionizing radiation a person receives from gamma rays from an external source, typically associated with radionuclides of concern. To limit this dose, an external hazard index (HEx) is used, given by Equation (10) [9, 26, 41]:

$$H_{\mathrm{Ex}} = \frac{C_{\mathrm{Ra}}}{370} + \frac{C_{\mathrm{Th}}}{259} + \frac{C_{\mathrm{K}}}{4810} \quad (10)$$

For individual safety outdoors, the index should be less than one.

Annual Gonadal Dose Equivalent (AGDE)

The annual gonadal dose equivalent (AGDE) estimates the potential dose to reproductive organs (gonads) from exposure to ionizing radiation over a year. AGDE due to specific activities of ^{226}Ra, ^{232}Th, and ^{40}K was calculated using Equation (11) [49]:

$$\mathrm{AGDE}(\mu\mathrm{Sv/y}) = 3.09C_{\mathrm{Ra}} + 4.18C_{\mathrm{Th}} + 0.314C_{\mathrm{K}} \quad (11)$$

AGDE considers the type of radiation, amount of exposure, and sensitivity of gonads to radiation-induced damage.

Alpha Hazard Index (Iα)

This index estimates the risk of internal exposure to alpha radiation from a mixture of α-emitting radionuclides and expresses the total hazard in a single numerical value. Excess alpha radiation from inhalation of radon from building materials is estimated using Equation (12) [7]:

$$I_\alpha = \frac{C_{\mathrm{Ra}}}{200} \quad (12)$$

The alpha index $I_\alpha \leq 1$ is equivalent to 200 Bq/kg of radium. Construction materials with ^{226}Ra exceeding 200 Bq/kg should be avoided as this may lead to 200 Bq/m^3 of radon, exposing occupants to internal radiation.

Radioactivity Index (Iγ)

This index examines radiation exposure risk to human settlements, particularly informal ones located near gold mine tailings dams. Residents may inadvertently excavate tailings soil for brick-making, resulting in potential gamma radiation exposure. The gamma radiation emitted by certain natural radionuclides in building materials is linked to this index, calculated with Equation (13) [4, 6, 44]:

$$I_\gamma = \frac{C_{\mathrm{Ra}}}{150} + \frac{C_{\mathrm{Th}}}{100} + \frac{C_{\mathrm{K}}}{1500} \quad (13)$$

With 1 as an upper limit, $I_\gamma \leq 1$ corresponds to 0.3 mSv/y, and $I_\gamma \leq 3$ corresponds to 1 mSv/y. For bulk building materials such as bricks, the ranges of I_γ are: $0.5 \leq I_\gamma \leq 1$.

Results and Discussion

Radioactivity Concentration in Soils

Samples from the Consolidated Main Reef (CMR) tailings were collected and analyzed at NRF iThemba LABS. Fig. 2 [Figure 2: see original paper] shows a γ-spectrum generated in this analysis. The spectrum comprised peaks due to geogenic radionuclides with no peaks emanating from anthropogenic radionuclides, proving that no artificial radionuclides are present in the gold tailings samples. The distribution of radionuclides is presented in Table 1 .

In the samples, ^{226}Ra (a decay product of the ^{238}U series) ranged from 222.61$±5.89Bq/kgto680.00±9.84Bq/kg, withameanof451.59±$8.00 Bq/kg, which is higher than the permissible limit of 35 Bq/kg [51]. According to these results, ^{226}Ra dominated in samples, followed partially by ^{40}K, while ^{232}Th was in the background in soil samples from Consolidated Main Reef gold tailings, as shown in Fig. 3 [Figure 3: see original paper]. The activity concentration of ^{232}Th ranged from 5.66$±1.13to18.41±2.21Bq/kgwithameanof10.46±$1.56 Bq/kg, and ^{40}K ranged from 67.18$±20.47Bq/kgto179.04±27.23Bq/kg, withameanof117.92±$21.97 Bq/kg. Both specific activity concentrations of ^{232}Th and ^{40}K are below their permissible limits of 30 and 400 Bq/kg, respectively, as specified in [51].

The average concentration of ^{226}Ra was significantly higher than that of the other two radionuclides, with activity decreasing in the order: ^{226}Ra > ^{40}K > ^{232}Th.

Assessment of Radiological Risk Indices

Table 2 presents radiological risk indices. RaEq had a minimum of 236.77$±6.25Bq/kgandmaximumof702.52±10.23Bq/kg, withameanof475.61±1.91Bq/kg, whichwasabovethe nGy/h, respectively, exceeding recommended values of 84 nGy/h [40] and 59 nGy/h [51].

The minimum indoor AEDE value was 0.54$±0.02mSv/y, whilethemaximumwas1.59±0.05mSv/y, withanaver 3}$ and 0.29$×10^{-3}$ [40, 51], respectively. The internal hazard index was 2.5 times higher than its recommended value, while the external hazard index was 1.3 times higher. AGDE, Iα, and Iγ were 5, 2, and 1.6 times higher than their recommended values, respectively.

The radium equivalent activity graph in Fig. 4 [Figure 4: see original paper] indicates that only 4 out of 20 samples had RaEq below the recommended limit. Samples CMR 05, 08, 10, 12, 14, and 17 displayed particularly high RaEq values.

Fig. 5 [Figure 5: see original paper] illustrates the relationship between Absorbed Dose Rate and Annual Effective Dose Equivalent in Consolidated Main Reef gold tailings samples, showing proportionality between these indices.

ELCR results show high indoor exposure risk that can lead to cancer in a lifetime across all samples, with values far above permissible limits. Outdoor exposures also exceed recommended limits. Indoor ELCR ranged from 1.88$±0.07×10^{-3}$ (minimum) to 5.57$±0.19×10^{-3}$ (maximum), with a mean of 3.78$±0.03×10{-3}, whichis3.3timeshigherthantherecommendedindoorELCRlimitof1.16×10{-3}$ [40]. Outdoor ELCR had a minimum of 0.47$±0.14×10{-3}, maximumof1.38±0.38×10{-3}, andmeanof0.94±0.03×10^{-3}$ [40]. Fig. 6 [Figure 6: see original paper] shows that both mean ELCR values are above recommended limits.

HIn had a minimum of 1.24\$$\pm 0.04, maximumof 3.74 \pm 0.06, andmeanof 2.51 \pm 0.01, whichalsohasproportionalv$ indicating high gamma exposure on the skin. All indices were above recommended limits as shown in Fig. 7 [Figure 7: see original paper], indicating the extent of soil contamination in this area.

The annual gonadal dose equivalent (AGDE) is above the permissible limit of 300 µSv/y in all samples. However, samples CMR 05, 08, 09, 10, 12, 14, 17, 18, 19, and 20 have values \$ \$1500 µSv/y, which are five times the permissible limit of 300 µSv/y [27]. This high AGDE indicates that samples present elevated risk of exposure to gonads, which is dangerous due to their sensitivity to radiation [12].

Fig. 8 [Figure 8: see original paper] compares annual gonadal dose equivalent (AGDE) with radium equivalent activity (RaEq), showing a linear relationship between the indices. This is logical: greater environmental radiation exposure corresponds to higher gonadal exposure risk.

HEx, a radiological measure used to assess potential external radiation exposure from NORMs in mining environments, had a minimum of 0.64\$$\pm 0.02, maximumof 1.90 \pm 0.04, andmeanof 1.29 \pm$\$0.01, which is above the permissible limit of \$ \$1, indicating possible gamma exposure in the external environment. HIn and HEx values of unity correspond to a dosage of 1.5 mGy/y [13]. Even when values exceed unity, they pose no immediate health hazard, but the increased risk of stochastic effects becomes significant [42].

The Iα hazard index had a minimum of 1.11\$$\pm 0.03, maximumof 3.40 \pm 0.05, andmeanof 2.26 \pm 0.03, whichisabov$ had a minimum of 0.80\$$\pm 0.03, maximumof 2.35 \pm 0.05, andmeanof 1.60 \pm$\$0.03, which is above the recommended limit of unity.

All samples had high HIn values, but samples CMR 05 and CMR 12 had the highest values as shown in Fig. 7 [Figure 7: see original paper]. HIn and Iα, being indices that report alpha radiation from building materials, have proportional values with HIn leading, indicating high alpha exposure in internal channels like lungs if dust is inhaled and stomach/intestines if dust is swallowed or ingested with food. Meanwhile, HEx and Iγ, being indices that report gamma radiation exposure from NORMs, also show proportional values.

Summary and Conclusions

Samples from Consolidated Main Reef gold tailings west of Johannesburg were collected, and radioactivity concentrations were assessed through comprehensive evaluation of radiological indices. Coaxial LEGe HPGe γ-spectrometry was used to measure γ-signals in soil samples. Only geogenic radionuclides were detected, indicating no anthropogenic radionuclides. The mean activity concentration of ^{226}Ra was 13 times the UNSCEAR [51] value of 35 Bq/kg and 1.2 times the [35] value of 370 Bq/kg. Both ^{232}Th and ^{40}K had means below their acceptable values.

All radiological indices surpassed global recommended values, with mean indices in descending order: AGDE > RaEq > ADR > ELCR > Iα > HIn > Iγ > HEx > AEDE.

These high values indicate that gold tailings pose significant health risks to the local population. Although findings showed high radiation hazard indices without immediate health effects, long-term exposure for people living near gold tailings could lead to radiological problems. A voluntary study should be conducted with individuals in the area to determine if links exist between reported symptoms and documented mine tailings exposure outcomes. Authorities should apply remediation methods such as phytoremediation, covering contaminated soil with virgin soil, and restricting area access in the short term. Raising awareness through print and social media is also crucial.

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Note: Figure translations are in progress. See original paper for figures.

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