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The Assessment of Heavy Metals and Natural Radioactivity in the Phosphate Tailings at Minjingu Mines in Tanzania

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ABSTRACT

Extraction and processing of the phosphate rocks has produced a massive amount of waste and posed a significant environmental concern. The majority of wastes generated in the fertiliser industry are overburden or waste rocks from mining, and phosphate tailings (PTs) or phosphogyp-sum from the beneficiation process. Phosphate rock mining and beneficiation expose heavy metals and radionuclides into the environment, which are harmful to living things. The purpose of this study was to determine the concentration levels of heavy metals and radionuclides activity in the phosphate tailings at Minjingu mines in northen of Tanzania. Heavy metals content and radionu-clide activity concentration were determined using energy dispersive X-ray fluoresence spectroscopy (ED-XRF) and high pure garmin energy detector (HPGe), respectively. The concentra-tion of heavy metals investigated ranges: Cu – 12.9–27.3 mg·kg^{-1}, Fe – 7944.2–19052.2) mg·kg^{-1}, Mn – 410.9–474) mg·kg^{-1}, Ni – 1.9–13.2) mg·kg^{-1}, Al – 3597–13129.2) mg·kg^{-1}, Zn – 195.2–281.7) mg·kg^{-1}, Pb – 0.7–4.5) mg·kg^{-1} and As – 2.7–11.3) mg·kg^{-1}. The result revealed that, the concentration level of heavy metals (Cu, Fe, Ni, As, and Pb) are below the permissible level while concentration level for Zn has high concentration compared to permissible level limit. However, the activity concentration of radionuclides ^{226}Ra, ^{232}Th and ^{40}K were ranging from 311 to 7,606 Bqkg^{-1}, 207 to 654 Bqkg^{-1} and 131 to 762 Bqkg^{-1}, respectively. The reported results of activity concentration of radionuclides are found to be higher compared to the recommended world value. The study results will be used as a guide for decision making in addressing problems observed in phosphate tailings, including radiation safety standards for workers and environmental systems in phosphate mines.

Keywords: phosphate rocks, heavy metals concentrations, phosphate fertilizer, Minjingu mines, radionuclides activity, phosphate tailings.

INTRODUCTION

Phosphate rock is an important source of raw materials used in fertiliser manufacturing around the world (Farid et al., 2022). Exploitation and processing of the phosphate rocks has resulted in an immense amount of waste and causes great challenge on the environment (Chiu et al., 2016). The majority of wastes generated in the fertiliser industry come from mining as overburden or waste rocks, and from the beneficiation process as phosphate tailings (PTs) or phosphogypsum (Sahu et al., 2014). The mining and beneficiation of phosphate rocks release heavy metals and radionuclides into the environment, which become toxic to living organisms (Ghose et al., 2013; Orosun et al., 2022). Phosphate rocks contain relatively high concentrations of heavy metals such as Arsenic (As), Cadmium (Cd) and lead (Pb) and naturally occurring radioactive elements (^{238}U, ^{40}K and ^{232}Th) (Atta and Zakaria, 2016; Ofomola et al., 2023). It was stated that, sedimentary rock can incorporate varying amounts of heavy metal elements such as Vanadium (V), Uranium (U), Nickel (Ni), Cromium (Cr), and Copper (Cu) based on their geological age, ore type, and origin (Boumaza et al., 2021). The phosphate manufacturing fertilizer industries are thought to be a source of natural radionuclide pollutants and expose it to the workers, the general public and environment (Sahu et al., 2014). Phosphate rock mining and beneficiation are the major processes that exposes humans to natural radionuclides such as ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K (Banzi et al., 2000). The majority of phosphate fertilisers are produced using wet process, using a reaction between sulphuric acid and phosphate rock to produce phosphoric acid (Corisco et al., 2017). It stated that mining wastes and tailings have significant concentrations of heavy metals, which can poison the food chain and infiltrate surface or subsurface waters through runoff (Chiu et al., 2016). It is reported that heavy metals like Mercury (Hg), Pb, Cd, As, Cu, Cobalt (Co), and Ni are harmful to human health and can lead to negative impacts through foods chain (Tóth et al., 2016).

However, little is known about heavy metals and radionuclides in phosphate tailings at Minjingu mines in the northern part of Tanzania. Therefore, the aim of this study was to determine the concentration of heavy metals and radionuclides present at phosphate tailings at Minjingu mines located in the northern part of Tanzania. The study results will be used as a guide for decision making in addressing problems observed in phosphate tailings, including radiation safety standards for workers and environmental systems in phosphate mines.

MATERIALS AND METHODS

The study area

The study was carried out at the Minjingu mines, where phosphate rocks are mined and processed into phosphate fertilisers using a dry technique. The Minjingu Mines is located at Minjingu Hill, near Manyara Lake and the area is between 30 42’ 21’’S 350 54’ 56’’E and 30 42’ 3’’S 350 54’ 14’’E. PTs were sampled from the tailings dumps situated near the open pit (Figure 2). Sampling was carried out to have representative samples for heavy metals and radionuclides identification and quantification.

Experimental design of the study

The experimental design chart of the study is shown in the Figure 1. The PTs samples were collected at different locations and transported to the laboratory for analysis of heavy metals and natural radionuclides. Samples were prepared and analyzed for natural radionuclides using Gamma Ray spectroscopy and heavy metal elements were analyzed by using energy dispersive X- ray fluorescence spectroscopy. The site’s environmental risk was assessed using well established models utilising the results of both analyses.

Sampling and sample preparation

The sample collection was carried out in the month of January, 2023, where a total of 10 samples were collected randomly at the points TD11-TD15 on tailing dump 1 and TD21-TD25 on tailing dump 2. The sampling points coordinates were easily recorded using Global Position System (GPS) Garmin ETREX 22 model. In every sampling point, five samples were collected randomly to form one composite sample. A 500 – 800

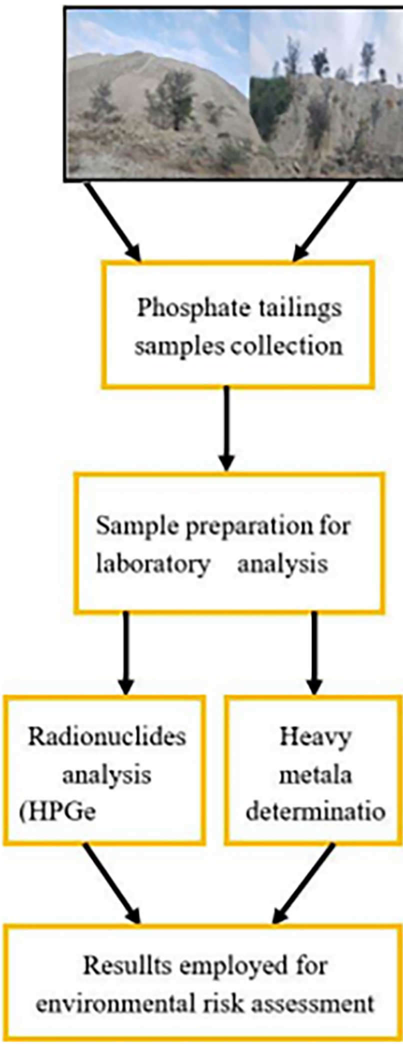

Fig. 1. Study’s experimental design

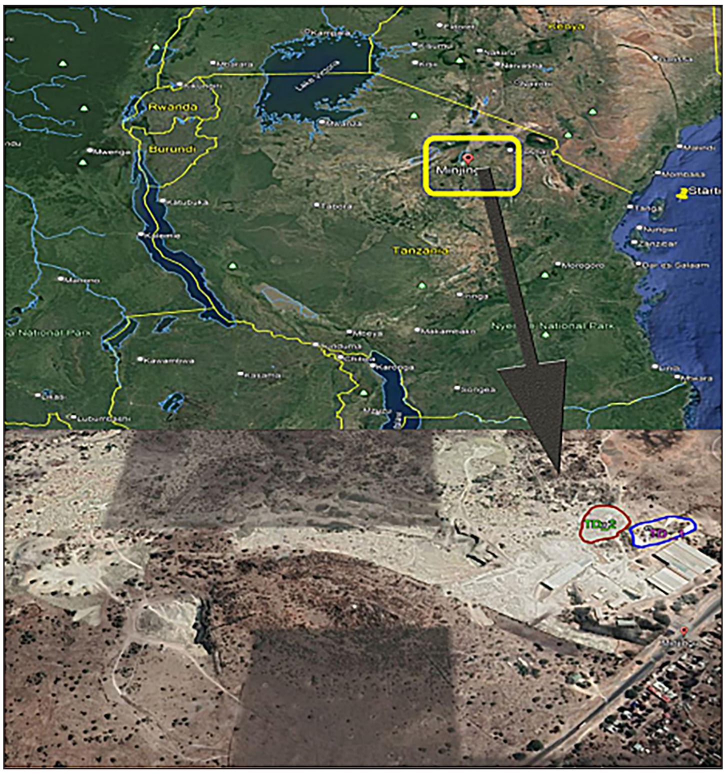

Fig. 2. The satellite image shows the geographical location of Minjingu Mines

g composite phosphate tailings sample was taken in each sampling location and put in a plastic bag and labelled. The samples were transported to the Tanzania Atomic Energy Commission (TAEC) laboratory in Arusha for analysis. The samples were dried for 24 hours at 100 °C in an oven. In order facilitate homogenization, the dry samples were crushed and put through a 500 µm sieve.

Determination of heavy metals in the phosphate tailings samples

The sample was dried, sieved, and then quartered to produce an acceptable amount of particles of each heterogeneous material component. The sample was then crushed into fine powder once more. The samples were sieved through a 500 m sieve, then the oversize was sieved once more until no grains larger than 500 µm remained. . The samples were dried using an oven at a temperature of 100 ^{o}C to reduce moisture contents below 5% before the use of XRF for identification and quantification of heavy metals. A total of 4 g weight of each sample was mixed with 0.9 g of binder and pulverized for 10 minutes at 180 rpm. The pulverized pellet was put in die of a 32 mm diameter and pressed into pellets using hydraulic pressure at a pressure of 15 bar for 1 minute to make a durable sample pellet for XRF analysis. The pulverized samples underwent analysis for Al, Mn, Ti, Fe, Zn, Cu, As, Ba, Pb, and U through the employment of a ED-XRF spectrometer, which utilized a rhodium tube and SDD detector with a detector resolution of 160 eV Mn-K and an energy range of 0–10 keV. Before conducting any analysis, it is necessary to place the spectrometer in standby mode to allow for the warming up of the X-ray tube. The commencement of analysis is contingent upon the selection of the pellet calibration technique and the input of relevant sample identification particulars. The sample compartment is interlocked during X-ray analysis. Samples of PTs were analyzed by ED-XRF to reveal their heavy metals concentrations and were studied by employing certified reference materials (CRM) IAEA-Soil-7.

Radioactivity measurements

GEM40-83-SMP, a hyper-pure germanium (HPGe) coaxial detector system, was used to measure the activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K. The analysis of samples were carried out in the laboratory of Tanzania Atomic Energy Commission (TAEC). To give ^{226}Ra and ^{232}Th time to reach secular equilibrium with their short-lived offspring, the dried and sieved sample was kept in an airtight stainless-steel canister measuring 157 cm^3 for a minimum of 21 days (El-Halim and AL-abrdi, 2021; Banzi et al., 2000). Following the radionuclides' equilibrium, each sample was placed at the top of a radiation detector and counted for 86,400 seconds. The detector's full width at half maximum (FWHM) was 0.87 keV at 122.1 keV (^{57}Co) and 1.85 keV at 1332.5 keV (^{60}Co), with a relative efficiency of 30%. A multi-nuclide standard source of type CBSS2 was used to calibrate energy and efficiency. The detector data was collected and processed using an ORTEC® DSPEC-LF digital signal processing spectrometer equipped with a 16 k-channel analyzer and Gamma Vision® software. Gamma-ray peak energies of 609.3 keV (214Bi) and 351.9 keV (214Pb), 583.1 keV (208Tl) and 911.2 keV (228Ac) and 1460.8 keV, respectively, were used to compute the activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K in a sample (Atta and Zakaria, 2016; Lolila and Mazunga, 2023). The following equation was used to determine the radionuclide activity concentrations in each sample based on the net count of each energy in the spectrum (Ofomola et al., 2023);

$$C = C_n/C_{fk} \tag{1}$$

The conversion factors (cps/Bqkg–1) for ^{40}K, ^{226}Ra, and ^{232}Th are 0.001554, 0.002086, and 0.002119, respectively. C_n is the count rate (counts per second), defined as C_n = Net count/live time. Meanwhile, C_{fk} is the detector's calibration factor.

Phosphate tailings radiological hazard index estimation for radionuclides

Radium equivalent activity (Ra_{eq})

Based on the presumption that 370 $Bqkg^{-1}$ of ^{226}Ra, 259 $Bqkg^{-1}$ of ^{232}Th, and 4810 $Bqkg^{-1}$ of ^{40}K provide the same gamma radiation dosage, the Ra_{eq} describes the weighted uniform activities of ^{232}Th, ^{226}Ra, and ^{40}K in a sample. The Ra_{eq} was determined using the relation given in Eq (2).

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \tag{2}$$

where: A_{Ra}, A_{Th} and A_K – the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the samples respectively.

The indexes of external hazards (H_{ex}) and interior hazards (H_{in})

The H_{ex} and H_{in} were calculated using Eqs (3) and (4), respectively, to account for the radiation exposure received by the samples under examination from ^{226}Ra, ^{232}Th, and ^{40}K, both internally and externally (Al Shaaibi et al., 2021).

$$H_{ex} = A_{Ra}/370 + A_{Th}/259 + A_K/4810 \tag{3}$$

$$H_{in} = A_{Ra}/185 + A_{Th}/259 + A_K/4810 \tag{4}$$

where: the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in samples are represented by A_{Ra}, A_{Th}, and A_K, respectively. H_{ex} and H_{in} values must be less than 1, meaning that the radiation risk is considered insignificant if it falls within the International Commission on Radiological Protection's (ICRP) dose equivalent limit of 1 $mSvy^{-1}$.

Absorbed dose rate (DR)

The presence of ^{226}Ra, ^{232}Th and ^{40}K in the phosphate tailings can result to radiation dose during dumping of the tailings. Thus, conversion factors of 0.462, 0.604, and 0.0417 for ^{226}Ra, ^{232}Th, and ^{40}K, respectively, were applied to determine the radiation absorbed dose rate (DR) in air at a height of 1 metre above ground (Orosun et al., 2022) as follows;

$$DR = 0.462A_{Ra} + 0.604A_{Th} + 0.0417A_K \tag{5}$$

where: A_{Ra}, A_{Th} and A_K are the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K respectively.

Annual effective dose (AED)

The annual dose that people receive outside is known as the outdoor annual effective dose. It is calculated from the absorbed dose rate using time occupancy factors and dose conversion, which indicate that the adult outdoor time occupancy factor is 0.8 (80%) for indoor exposure and 0.2 (20%) for outdoor exposure (UNSCEAR, 2008). The following formulas were used to determine the outdoor *AED* in $mSvy^{-1}$ (Ademola et al., 2014).

$$AED = DR \times 8760\ h \times 0.2 \times 0.7 SvGy^{-1} \times 10^{-6} \tag{6}$$

The total hours in a year are 8760; the dosage conversion factor is 0.7 $SvGy^{-1}$, the factor 10^{-6} converts the nano scale to milli, and *D*R is the absorbed dose rate in air in $nGyh^{-1}$.

Table 1. Concentration of heavy metals in the phosphate tailings at Minjingu mines

Sample ID	Heavy metals concentration (mg.kg^{-1})							
Tailing dump 1	Al	Mn	Fe	Ni	Cu	Zn	As	Pb
TD11	5352.7	424.0	11014.6	4.1	20.4	247.0	7.7	0.7
TD12	7858.7	410.9	14840.6	11.2	23.9	281.7	2.7	4.1
TD13	3597.0	422.3	7944.2	1.9	12.9	245.1	11.3	1.1
TD14	7409.7	463.8	12687.0	4.1	21.3	276.2	7.7	1.6
TD15	6637.0	433.4	11335.6	2.4	17.3	271.8	10.7	1.2
Min	3597.0	410.9	7944.2	1.9	12.9	245.1	2.7	0.7
Max	7858.7	463.8	14840.6	11.2	23.9	281.7	11.3	4.1
Mean	6171.0	430.9	11564.4	4.7	19.2	264.4	8.0	1.8
Tailing dump 2								
TD21	11600.9	430.5	17618.3	13.2	22.0	224.1	6.1	4.3
TD22	13129.2	458.9	19052.0	13.0	27.3	209.5	6.7	4.5
TD23	8381.6	462.9	11087.3	3.2	17.4	217.0	7.8	1.6
TD24	8381.1	429.9	14235.0	9.4	19.0	236.7	5.5	4.3
TD25	7571.4	474.0	12778.3	7.4	18.7	195.2	10.0	3.0
Min	7571.4	429.9	11087.3	3.2	17.4	195.2	5.5	1.6
Max	13129.2	474.0	19052.0	13.2	27.3	236.7	10.0	4.5
Mean	9812.8	451.2	14954.2	9.2	20.9	216.5	7.2	3.5
Permissible level	N.A	2000	50000	50	100	200	20	100

RESULTS AND DISCUSSION

Heavy metal concentration in the phosphate tailings

The heavy metal concentration in the phosphate tailings samples taken from the Minjingu mines is displayed in Table 1. The concentrations of the heavy metals (Pb, Mn, As, Al, Fe, Ni, Cu, and Zn) in the tailing phosphate samples from Minjingu mines were determined, and compared to heavy metals permissible level established by WHO. The following are the ranges of heavy metal concentrations (mg·kg^{-1}) in the areas under study:

- Copper (Cu): the highest concentration of copper in this study was recorded at TD22 in the tailing dump 2 (27.3 mg·kg^{-1}) and lowest concentration recorded in the sample TD13 in the tailing dump 1 (12.9 mg·kg^{-1}). All sample findings fall below the suggested level of 100 mg·kg^{-1} when compared to the World Health Organization's (WHO) maximum allowed limit for copper in soil.
- Iron (Fe): sample TD22 had the highest iron concentration (19052 mg·kg^{-1}), whereas sample TD13 had the lowest value (7944.2 mg·kg^{-1}). The iron concentration was found to be the lowest when compared to the WHO's maximum permissible limit of 50,000 mg·kg^{-1} for iron.
- Manganese (Mn): the manganese in the tailings phosphate samples has the highest and lowest concentration in the sample TD25 (474.0 mg·kg^{-1}) and TD12 (410.9 mg·kg^{-1}), respectively. When the results of sample tests for manganese are compared to the WHO maximum permitted limit of 2000 mg·kg^{-1}, it is evident that every sample is below the WHO permissible limit.
- Zinc (Zn): the concentration of zinc in the sample observed to be highest in the sample TD12 (281.7 mg·kg^{-1}) and the lowest concentration in the sample TD25 (195.2 mg·kg^{-1}). From the results of the samples (TD11, TD12, TD13, TD14, TD15, TD21, TD22, TD23 and TD24) indicated high concentration than permissible allowable limit of 200 mg·kg^{-1} except sample TD25 with concentration (195.2 mg·kg^{-1}) below the permissible limit.
- Nickel (Ni): the sample TD21 has the highest content of nickel (13.2 mg·kg^{-1}) while the sample TD23 has the lowest concentration (1.9 mg·kg^{-1}). All sample concentrations fall within the 50 mg·kg^{-1} acceptable limit when comparing the nickel result to the WHO maximum allowed level.

- Lead (Pb): sample TD22 has the greatest content of lead (4.5 mg·kg^{-1}), whereas sample TD11 has the lowest concentration (0.7 mg·kg^{-1}). The lead (Pb) results indicate that all samples had concentration levels below the permissible limit, when compared to the WHO maximum allowable limit of 100 mg·kg^{-1}.
- Arsenic (As): the Arsenic in the samples has the highest concentration in the sample TD13 (11.3 mg·kg^{-1}) and the lowest concentration in the sample TD12 (2.7 mg·kg^{-1}). It shows that all samples has lowest concentration level below the permissible limit when compared with the maximum allowable limit for Arsenic which is 20 mg·kg^{-1}.

Concentrations of radioactive elements in the phosphate tailings

The natural radionuclide activity concentration (^{226}Ra, ^{232}Th and ^{40}K) were determined in the phosphate tailings samples by HPGe gamma spectroscopy as shown in Table 2. Based on the findings, the activity's greatest contribution originates from ^{226}Ra compared to activity from ^{232}Th and ^{40}K. The average activity concentration for ^{226}Ra, ^{40}K and ^{232}Th are 7606, 762 and 688 Bq kg^{-1} recorded in the tailing dump 2 and 3946, 683, and 654 Bq·kg^{-1} reported in tailing dump 1, respectively. Furthermore, in comparison with the world average values of 32, 45 and 420 Bq·kg^{-1} given for ^{226}Ra, ^{232}Th, and ^{40}K respectively, the average activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K recorded in both tailing dumps have higher values than the world mean value by International Atomic Energy Agency (IAEA) (Júnior et al., 2021) and United Nations scientific committee on the effect of atomic radiation, sources and effects of ionizing radiation (UNSCEAR, 2008).

The absorbed dose rate (DR) ranged between 468.30 and 2708.6 nGy·h^{-1} for tailing dump 1, 2778.0 and 4846.5 nGy·h^{-1} for tailings dump 2 with an average value of 2151.7 and 3799.1 nGy·h^{-1} respectively as shown in Table 2. The results revealed that the absorbed dose rate from the samples are higher compared to the recommended absorbed dose rate of 59 nGy·h^{-1} ((Masok et al., 2018; UNSCEAR, 2008). The comparable annual effective dosage rate (AED) was found to have an average of 2.6 and 4.7 mSv·yr^{-1} for tailing dump 1 and tailing dump 2 respectively. This value is significantly greater than the average value

Table 2. Activity concentration of natural radionuclides and radiological hazard indices in phos-phate tailings at Minjingu mines

Sample ID	Activity concentration of radionuclides (Bqkg-1)				Radiological hazard indices			
Tailing dump 1	^{232}Th	^{226}Ra	^{40}K	Ra_{eq}	H_{ex}	H_{in}	DR (nGyh^{-1})	AED (mSvy^{-1})
TD11	427.7	3780.8	486.2	4429.8	12.0	22.2	2622.7	3.2
TD12	654.1	3483.9	682.7	4471.8	12.1	21.5	2583.6	3.2
TD13	436.8	311.0	278.9	957.1	2.6	3.4	468.3	0.6
TD14	397.4	3945.8	531.2	4555.0	12.3	23.0	2708.6	3.3
TD15	636.3	3173.3	569.3	4127.0	11.2	19.7	2375.5	2.9
Max	654.1	3945.8	682.7	4555.0	12.3	23.0	2708.6	3.3
Min	397.4	311.0	278.9	957.1	2.6	3.4	468.3	0.6
Mean (study)	510.5	2939.0	509.7	3708.2	10.0	18.0	2151.7	2.6
Tailing dump 2								
TD21	687.9	3759.8	754.6	4801.6	13.0	23.1	2778.0	3.4
TD22	537.5	5408.5	761.9	6235.8	16.9	31.5	3709.7	4.5
TD23	555.6	6551.9	481.5	7383.4	20.0	37.7	4417.8	5.4
TD24	575.9	4629.6	611.6	5500.2	14.9	27.4	3243.7	4.0
TD25	207.1	7606.3	130.7	7912.6	21.4	41.9	4846.5	5.9
Max	687.9	7606.3	761.9	7912.6	21.4	41.9	4846.5	5.9
Min	207.1	3759.8	130.7	4801.6	13.0	23.1	2778.0	3.4
Mean (study)	512.8	5591.2	548.1	6366.7	17.2	32.3	3799.1	4.7
Mean (World)	45	32	420	370	<1	<1	59	0.07

globally, which is 0.07 mSv·yr^{-1} (UNSCEAR, 2008). The findings showed that, in comparison to earlier studies, the annual effective dose rate level discovered in this one was higher. The following is the annual effective dosage rate that the researchers reported: It was reported to be between 0.2 and 0.5 mSv·y^{-1} in Tunisia (Khelifi et al., 2019), 0.02 and 0.92 mSv·y^{-1} in Morocco (Qamouche et al., 2020), 1.8 mSv·y^{-1} in Egypt (Gaafar et al., 2016), and 0.266 mSv·y^{-1} in Togo (Hazou and Patchali, 2021). Similar to Minjingu phosphate rock, these rocks are sedimentary phosphate rocks.

According to Table 2, the highest computed radium equivalent value was found in TD25 (7912.6 Bq·kg^{-1}), while the lowest value was obtained in TD13 (957.1 Bq·kg^{-1}). The average Ra_{eq} for tailing dump 1 and tailing dump 2 were 3708.2 and 6366.7 Bq·kg^{-1} respectively. These results are considered to be higher radium equivalent concentration when compared with the UN-SCEAR recommended limit of 370 Bq·kg^{-1} (Manigandan and Shekar, 2014; UNSCEAR, 2008).

The H_{ex} and H_{in} ranges were 2.6–21.4 and 3.4–41.9, respectively, with TD25 recording the highest values and TD13 recording the lowest. All samples had hazards indices that were greater than unity, both internal (H_{in}) and external (H_{ex}). There is a considerable risk of radon and its short-lived progeny on the human respiratory organ since all values are higher than the UNSCEAR recommended unity. Workers who are exposed to radiation could receive radiation doses from natural radioactive elements that affect all or part of their body and may lead to immediate or long-term effects on humans humans ((IAEA), 2014; Lema et al., 2014; Pathak, 1989). Lung cancers and laukemia (Weiss, 2016; Yiin et al., 2016) are an example of health risks associated with exposure the levels of radiation than the recommended level.

CONCLUSIONS

Heavy metal contamination (Cu, Zn, Al, Mn, Ni, Fe, Pb, and As) in phosphate tailings was assessed in comparison to the WHO permitted level limit. Heavy metal concentrations (Cu, Al, Mn, Ni, Fe, Pb, and As) at the Minjingu mines are lower than the WHO maximum permissible limit for heavy metals in soil, whereas Zn concentrations are higher than the WHO maximum allowable limit. The results showed that the mean activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K are greater than the global recommended activity threshold. Both the annual effective dose and the average outdoor dose rate were more than the globally recommended average. This suggests that employees are more susceptible to health problems such as lung cancers and laukemia due to radiation exposure, which was far greater than the comparable exposure levels documented in other parts of the world. We must put radiation regulations and standards into effect by enhancing working conditions to lower occupational radiation exposure to the allowed levels advised by IAEA safety standards. In order to minimize the workers from radiation exposure, we recommend to the management of Minjingu mines should take the following actions in accordance with IAEA and WHO guidelines (International Atomic Energy Agency (IAEA), 2013): to control workers occupancy period or shielding to minimize external exposure to the Natural Occurrence Radioactive Materials (NORM); to put barrier and warning sign to protect entrance to the areas with high activity concentrations; to provide the workers with protective respiratory equipments; to prevent resuspensions of dust; and to work together with the regulatory agency to educate employees on awareness and how to take preventative measures.

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