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Radio-epidemiological evaluation and remediation in water sources from two mines in South Africa

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► ***Original article***

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ABSTRACT

Background: In this study, the health risk associated with three types of drinking waters (fissure, underground treated and surrounding waters) from two mines in South Africa were assessed. ***Materials and Methods***: The measurement of the radionuclides concentration was carried out by liquid scintillation counter and alpha spectrometer. The estimated radiological risk pose to the people consuming the water types were evaluated based on the calculated radionuclide concentrations. ***Results:*** The value of the gross alpha was 1.08 ± 0.02 Bq/L and gross beta activity was 0.65 ± 0.04 Bq/L in the treated underground water. These values were several fold above the limit value of 1.0 Bq/L for gross alpha and 0.1 Bq/L for gross beta. For the fissure water, the gross alpha activity value was 0.59 ± 0.07 Bq/L which is less than the limit of 1.0 Bq/L and the gross beta activity was high with a value of 0.54 ± 0.05 Bq/L compared to the limit value of 0.1 Bq/L. The results around the Princess gold mine showed very high gross alpha and beta activity in the collected water samples studied. In general, the concentrations of the natural radionuclides were high especially ^{238}U, ^{226}Ra, ^{230}Th, ^{235}U, ^{234}U and ^{210}Po in all the water samples. ***Conclusion:*** The calculated chemical toxicity, cancer mortality/morbidity and hazards quotient with respect to ^{238}U were very high. Hence, the waters within these vicinities are polluted with radionuclides and may posed serious health effect to the inhabitant.

Keywords: *Gross alpha and beta, cancer, water, radio-epidemiology, Gauteng, Princess Gold mine.*

INTRODUCTION

In South Africa, there are five gold mining basins and which collectively have resulted in the largest gold and uranium mining basin in the world. Radionuclides are radioactive isotopes or unstable forms of elements. They release energy in the form of alpha, beta and gamma radiation during the unstable state to decompose to the stable elements [5]. Gamma rays, alpha particles, and beta particles, which are given off by radioactive decay, have very different properties but are all ionizing radiation—each is energetic enough to break chemical bonds, thereby possessing the ability to damage or destroy living cells [6]. This ionization can damage living cells directly, by breaking the chemical bonds of important biological molecules viz., DNA, or indirectly, by creating chemical radicals from water molecules in the cells, which can then attack the biological molecules chemically [7]. At some extent these molecules are repaired by natural biological processes, however, the effectiveness of this repair depends on extent of damage [8]. Obviously, if the repair is faulty or not made at all then the cells may die, or an impairment in the natural functioning of the cell may occur leading to somatic effects (physical effects suffered by the irradiated individual only such as cancer) or a permanent alteration of the cell may result which is transmitted to a later generations leading to genetic effect [9].

Naturally occurring radionuclides are present in a wide range of concentrations in all rocks,

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soil, and water. The occurrence and distribution in ground water is controlled primarily by the local geology and geochemistry of rock and water as well as the anthropogenic activity [10, 11]. The radioactive elements, uranium-238 and thorium-232, decay slowly and produce other radionuclides (daughter elements), such as radium and radon, which in turn undergo further radioactive decay at a rate faster than that of uranium or thorium. These radioactive daughter elements have different chemical properties, decay at different rates, and emit different levels of radiation energy than either uranium or thorium which are harmful to our health when ingested [12, 13].

When a radionuclide gets into the human body through ingestion, inhalation or absorption through the skin, it continues to decay by emitting radiation such as α, β or γ radiations such that the organ or tissue is continuously irradiated. The damage is greatest with α, and least with γ radiation [14]. Some radio nuclides are chemically similar to some minerals in the human body and so when they are taken into the body, they mimic those minerals in the organs [15]. For instance ^{226}Ra, ^{38}Sr and ^{56}Ba, are found to have chemical similarity with ^{20}Ca in the bone [15]. When these radionuclides get into the body, they are deposited in the bone marrow causing damage to the bone cells resulting to bone cancer [6, 15].

Research work on radionuclides contents of underground fissure and treated water in relation to mines in South Africa are scanty. In this study, three categories of water were selected:-

– The fissure water referred to as the underground waters that were leaching out through the cracks of the rocks, soils, walls inside the mine in Gauteng.
– The treated underground water referred to as the samples collected from the mine in Gauteng which are treated (by the mine) before being dispatched to the general public and
– The surface water from the surroundings environs of the Princess Gold mine, Roodepoort. This water constantly pollute the surrounding boreholes and wells which are used in drinking and cooking.

The aim of this study was to determine the gross alpha/beta activity and radionuclides concentration in these three categories of water and to assess the radiological health risks to the people living around these vicinities.

MATERIALS AND METHODS

From the Princess Gold mine, Roodepoort, the water sampling was done during the rainy season because during this period large volume of water get to the surrounding streams, river and boreholes. The water samples from Princess Gold mine, Roodepoort, were collected at the foot of the tailings and along the water flow path before it joins other water sources. Figure 1 shows the map of the abandoned Princess Gold mine tailings. The underground treated and fissure water samples were collected from a mine in Gauteng.

The three categories of water samples collected were large enough (2 L) for adequate aliquots to be taken in order to obtain the required sensitivity. The samples were collected in plastic containers and transported immediately to the laboratory. The samples were filtered (X-121) using Whatman™ 1005-070 grade 5 filter paper with pore size of 2.5 µm to remove coarse materials, suspended particles and colloidal. Thereafter, the water samples were preserved with Nitric acid (HNO_3) to ensure that the radionuclides are not adsorbed on container walls.

The preparation and analysis of these samples were performed in Nuclear Energy Corporation of South Africa (NECSA). An outlined in the South Africa National Accredited System (SANAS). The accredited laboratory (Testing laboratory T0111) is based on International Organizational Standard (ISO/IEC Standard 17025). The seven analytical methods developed are shown in table 1.

Water volume of 50 ml each were evaporated to dryness and counted for 300 minutes on a gas proportional counter to screen for gross alpha\beta activity (X-161).

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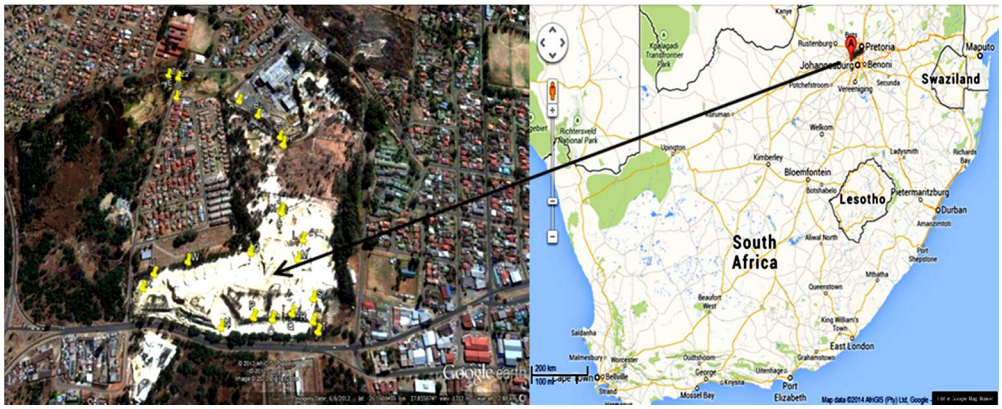


Figure 1. A map of the abandoned Princess Gold mine tailings (right) of the study area marked 'A' (left).

Table 1. Sample preparation and analytical/description

Method	Description	Water around mine from Princess Gold			Treated water	Fissure water
X-121	Filtration of suspended solids	A1	B1	C1	D1	E1
X-161	Gross alpha/beta analysis	A1	B1	C1	D1	E1
X-145	Uranium by alpha spectrometry	A1	B1	C1	D1	E1
X-142	Thorium by alpha spectrometry	A1	B1	C1	D1	E1
X-124	Radium by alpha spectrometry	A1	B1	C1	D1	E1
X-129	Polonium-210 by alpha spectrometry	A1	B1	C1	D1	E1
X-129	Lead-210 by alpha spectrometry	A1	B1	C1	D1	E1

The water samples were further analysed for selected radionuclides in the uranium and thorium decay series using alpha spectrometry (X-145 to X-129). In the analyses of ^{238}U, ^{234}U, ^{235}U, ^{230}Th, ^{227}Th and ^{228}Th, the solid phase extraction was performed to first extract the radionuclides from the water samples before using the cation exchange resin. A volume of 200 mL of the water samples, each were acidified with HNO_3 and pre-concentrated to a volume of 10 mL. This volume was loaded on a pre-conditioned Truspec resin after which was rinsed with diluted HNO_3 and hydrochloric acid (HCl). The analytes were eluted with 15 mL of 0.1M HCl/0.1M HF (first eluent), 15 mL of 2M HCl (second eluent), and 15 mL 2M H_2SO_4 (third eluent). The first eluent was rinsed with 10 mL of 0.1M HCl/0.01M HF. A volume of 0.1 mL lanthanum oxide, 0.5 mL concentrated HF and titanium chloride were added to the second eluent while 0.1 mL lanthanum oxide and 0.5 mL concentrated HF were added to the third eluent to form a co-precipitation of uranium and thorium respectively with lanthanum flouride precipitate. The precipitate was then collected by filtering the solution using a filter paper. The filtrate was then dried and analysed using an alpha spectrometer equipped with alpha spec Genie software.

In the analysis of ^{226}Ra, ^{223}Ra and ^{224}Ra, the radionuclides were extracted by adding barium solution into 500 mL of the water samples each. The radium radionuclides in the water samples were extracted by forming co-precipitate with barium sulphate in an acidic solution. The precipitate was enhanced by adding 0.1M Ethylenediaminetetraacetic acid (EDTA) into the solution. The extract are analysed using the alpha spectrometer equipped with alpha spec Genie software. Finally, for polonium radionuclide (^{210}Po), a volume 200 mL of the water samples, each were acidified with HCl. Thereafter, ascorbic acid was added as a reducing agent to prevent the co-precipitation of polonium with iron. A silver disk was then dropped into the solution to promote the

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spontaneous deposition of polonium on to the surface of the silver disk. The solution was stirred for 4 hours to enhance the process [12 14]. The platted disk was then removed and dried prior to its analysis (X-129) on the alpha spectrometer.

Radionuclides analysis in SANAS accredited laboratory (Testing laboratory T0111) is based on the ISO/IEC Standard 17025 (which is -the standard for which most laboratories must hold accreditation in order to be deemed technically competent) and all the analytical methods are documented in the RadioAnalysis Quality System. Results of this analysis were obtained from one or more individual test reports produced by accredited methods.

Alpha spectra genie software

Alpha-particle spectra are critically dependent on the experimental set-up used in the measurements. Source preparation, detector size, and solid angle of the measurement, among other factors, have a strong influence on spectral parameters such as energy resolution and peak tailings.

In alpha spectrometry analysis, peak search is performed using either a library driven routine (for spectra of known contents) or by the Generalised Second Differential Method (for spectra of unknown contents). Peak areas are determined either by summation (singlets) or by non-linear least squares fitting (singlets and multiplets) using a modified Marquardt algorithm for fast convergence.

A statistical comparison based on standardized residuals (the differences between reported values and reference values divided by their own uncertainties) was performed. Using all cases where both a reported area and a reference area were available. A weighted average and its uncertainty of the ratios of reference peak areas and program output peak areas were determined.

RESULTS AND DISCUSSION

The measured values of the gross alpha and gross beta activity concentrations in the water samples are presented in figure 2.

From figure 2, it is observed that the gross alpha and beta activity concentrations in the water samples were high compared to the National Primary Interim Drinking Water Regulations [16]. It was observed that the gross beta activity for B1 (water sample collected 5 m before the drainage pipe was higher than the gross alpha activities compared to the other samples (Figure 2). This could be attributed to

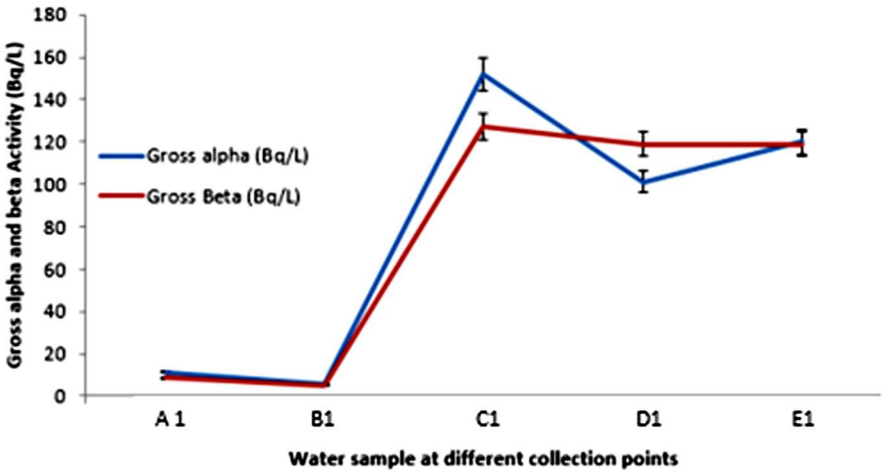


Figure 2. Gross alpha and beta distribution in the water samples

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the proximity of the slimes. The gross beta activity in water sample primarily comprised of the uranium decay products which are harmful to biological cells. The recommended levels of gross beta activity concentration by WHO is 1.0 Bq/L. If this activity doses does not exceed 1.0 Bq/L, it can be assumed that the annual total indicative dose of adults should be less than 0.1 mSv per year. The results obtained in this study showed that the measured activity concentrations of the gross beta in all the water samples are several folds above the recommended 1.0 Bq/L limit by the WHO [17].

A study by Wagner *et al*, [6] on the radionuclides contents in the public drinking water revealed the possibility of leukaemia due to exposure of 1.00 - 4.99 µg/L in adults [6, 8]. They also found that women with exposure level of >5µg/L, had a significant elevated risk of kidney cancer [6, 8]. A study by [18-20] revealed that over 70% of the water used in both rural and urban areas in South Africa is surface water drawn from rivers, streams, lakes, ponds and springs. This work as seen in figure 2 revealed the activity concentration of natural radionuclides in Bq/L.

As observed in figure 2, the measured radioactivities of the treated underground water (D1) for all the radionuclides measured were higher than the activity concentration of the fissure water (E1) except for ^{230}Th and ^{228}Th. This implies that the activity concentration of the radionuclides in the water before treatments (fissure) were lower compared to the treated water. It was also observed that ^{232}Th for water sample A1 collected 5 m after the drainage pipe had high values of 0.24 ± 0.04 and for sample Z1004 collected 5 m just before the drainage pipe had the highest value of 0.59 ± 0.06. These two sampling locations (A1 and B1) also recorded higher values of ^{238}U (figure 2). The higher values of activity concentrations of ^{238}U and ^{232}Th in the water samples collected at the foot of the dump. This suggest that the water is more acidic due to the mine dumps which may contained uranium, manganese, aluminium, copper and potentially toxic radioactive metals which flow uncontrolled into the water path (streams).

From figure 2, it was observed that activity concentration of ^{238}U was very high in all the water samples with the lowest of 4.46 ± 0.22 Bq/L for E1 and the highest of 181 ± 3.00 Bq/L for Z1004. Based on this results, the health risk assessment of ^{238}U in the water samples was evaluated in relation to lifetime cancer risk. The lifetime cancer risks LCR, associated with the intake of ^{238}U was evaluated by multiplying the applicable risk coefficient *Rc* and the per capita activity intake (PCAI) as follows:

$$LCR = PCAI \times R_c \quad (1)$$

In this evaluation, we used 62.2 years as the average life expectancy at birth in South Africa and 731 L as the annual consumption of water for an individual. From the average life expectancy at birth and the annual consumption of water for an individual, we calculated the lifetime intake of water (LTI) to 45468.2 L.

From [10] and [21], we obtained the cancer risk coefficients of ^{238}U to be 1.13 × 10^{-9} Bq^{-1} for mortality and 1.73 × 10^{-9} Bq^{-1} for morbidity. With the aid of equation (1), the cancer mortality and morbidity risks of ^{238}U over lifetime consumption of the water were calculated as (results are presented in table 3);

PCAI>Activity mBq/L =LTI = 5444 (2)

Equation (3) was used to evaluate the lifetime average daily dose (LADD) of ^{238}U through water intake to determine the effect of the carcinogenic and non-carcinogenic risks associated with ^{238}U in the water sample.

$$Ingestion\ LADD\ of\ drinking\ water = \left(\frac{EPC \times ED \times EF \times IR}{BW \times AT}\right) \mu g\, kg^{-1}\, day^{-1} \quad (3)$$

where LADD = lifetime average daily dose, EPC = exposure point concentration (µg L^{-1}), IR = water ingestion rate (L day^{-1}), EF = exposure frequency (days $year^{-1}$), ED = total exposure duration (years), AT = average time (days) and BW is the body weight (kg). From [11] and [18] we obtained the following data that were used in the evaluation of LADD and hazard quotient; IR = 2 L day^{-1}, EF = 350 days, ED = 62.2 y, AT = 22703 (obtained from 62.2 × 365) and BW = 70 kg weight of a standard adult. The following conversions were made:

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$$1Bq/L = 27.0pCi/L,\ \mu g/L = \frac{pCi/L}{0.67}$$

The hazard quotient was obtained as shown in equation 4. The results of the cancer mortality, cancer morbidity, chemical toxicity risk (LADD) and hazard quotient for ^{238}U over a lifetime consumption the waters in the study sites were estimated and presented in figure 3.

$$\text{Hazard quotient} = \frac{\text{LADD of } ^{238}\text{U through drinking water intake}}{\text{RFD of } 0.6\ \mu g\ kg^{-1}\ day^{-1}} \quad (4)$$

In figure 3, the cancer mortality risk in the study site, ranged from 2.29E+02 for E1 to 9.30E+03 for B1, while for morbidity risk ranges from 3.51E+02 for E1 to 1.40E+04 for A1. The highest cancer mortality value was found at B1 location and the highest cancer morbidity of 1.42E+04 at B1. In general, these values were between 10^{+2} and 10^{+4} higher when compared to the acceptable level of 10^{-3} for the radiological risk [10] by a factor of 10^{+7}.

The hazard quotient parameter is defined as the ratio of the potential exposure to a substance and the level at which no adverse effects are expected. This parameter, when calculated to be less than 1, it shows that no adverse health effects would be expected as a result of the exposure otherwise, the reverse is the case [11]. The hazard quotient of the measured water samples as presented in figure 4 were far above 1 in all the water samples which implies that adverse health effects would be expected as a result of the exposure.

As seen in figure 3, high values were observed in water samples for C1, B1 and A1 that were obtained around the Princess gold mine slimes.

However, ^{230}Th, ^{234}U, ^{226}Ra, ^{235}U and ^{210}Po were relatively high in A1005 and B1006 in the treated and fissure water samples respectively obtained from the mine in Gauteng. This relative high concentration of the above radionuclides (^{226}Ra, ^{230}Th, ^{235}U, ^{234}U and ^{210}Po) content in the groundwater or drinking water may result to high incidence rate of renal cell carcinoma (kidney cancer) [8].

Water pollutant or contamination caused by radionuclides is a significant environmental problem particularly in densely populated developing countries where human habitats are usually in close proximity to mine sites [22]. Water draining from gold mines slimes frequently contains radionuclides at high levels which could contaminate streams and agricultural lands. This study confirmed to the

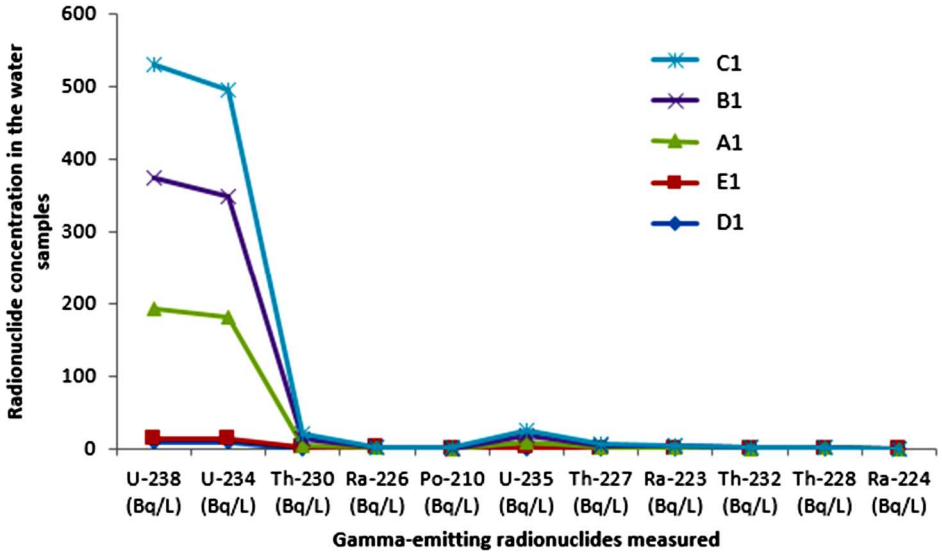


Figure 3. Distribution of radionuclides measured

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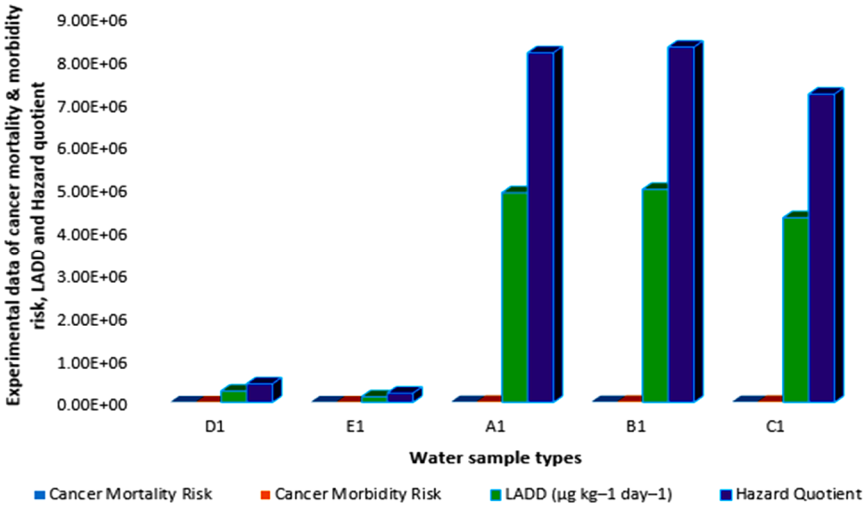


Figure 4. Evaluated health risk parameter in the water samples

high values of radionuclides pollution identified, affecting freshwater systems and resources in South Africa [22]. Mine water is a growing concern in water quality management and impacts negatively on the water environment by increasing the levels of radionuclides of the receiving water resulting to increasing cases of kidney cancer [8]. Dose rates due to the radionuclides measured in the water samples were calculated according to guidelines in the National Nuclear regulatory licensing guide LG-1032 [23]. Consumption rates for the different age groups in this guide and dose rate conversion factors published in IAEA Safety Series 115 [24] were used. Where the activity was less than the MDA, half of the values were used in the calculation. The dose rate values were reported with 2 significant digits as shown in figure 5.

Figure 5, shows the exposure annual dose rates of the members of the public to the ionizing radiation emanating from these samples measured. According to National Nuclear Regulator (NNR) Regulatory Guide (23), the dose rates constraint applicable for members of the public within the exposed population is 250 µSv/yr (25-27). The annual dose rate for < 1 yr ranged from 420 µSv/yr to

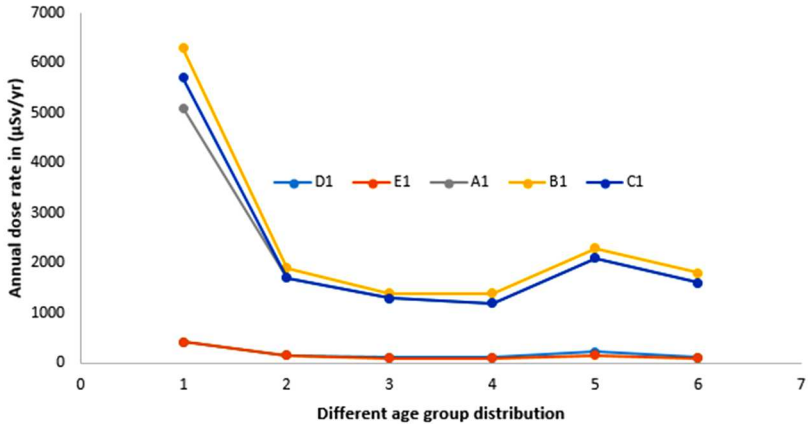


Figure 5. Evaluated annual dose rate in different age groups.

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6300 μSv/yr, therefore, these water samples are not suitable for consumption by babies. The annual dose rate for A1, B1 and C1 (water samples collected around the Princess dump mine slimes) were high and ranged from 1200 μSv/yr to 6300 μSv/yr for all the age groups measured. The annual dose rate due to radionuclides in the water samples from the treated (D1) and fissure (E1) sources were below the limit of 250 μSv/yr for 1 to > 17 yrs and vary from 95 to 220 μSv/yr.

From the foregoing analysis it is evident that there is an increase in gamma-emitting radionuclides pollutants in water samples from the mines which will end in the streams/rivers and threatens the scarce water resources of South Africa, and as a result also human health and food security in the mining areas. The water quality around the Princess mine slimes to the surrounding community and the water quality of the treated or fissure exceeds the standards set by DWAF (19-21, 25-27). From these studies, it is seen that the water is deteriorating in terms of radionuclides pollutants due to torrential rains and floods, lack of sufficient drainage, uncontrolled drainage, lack of adequate knowledge, wrong management decisions, very poor construction and rehabilitation rates of drainage systems, increase of irrigation systems without paying any attention to their adverse impacts on soil and quality of water resources, etc.

CONCLUSION

The assessed radioactivity concentration in the three types of waters (fissure, underground treated and surrounding waters) from the two mines indicate that the gross alpha and beta activity were high in the treated underground water compared to the fissure water. Also, the results around the Princess gold mine shows high gross alpha and beta activity concentration. The concentrations of the natural radionuclides (^{238}U, ^{234}U, ^{230}Th, ^{226}Ra, ^{210}Po, ^{235}U, ^{237}Th, ^{223}Ra, ^{232}Th, ^{228}Th, and ^{224}Ra) were relatively high in all the water sample types. The calculated chemical toxicity, cancer mortality/morbidity and hazards quotient with respect to ^{238}U were high. Hence, the waters within these vicinities may posed serious health effect to the inhabitant. Therefore, the fissure/treated underground water from the mine in Gauteng should undergo further and intensive treatments before usage by miners and dispatch to the public. Around the Princess Gold mine vicinity, sufficient and controlled drainage system should be emphasized.

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***Conflicts of interest*:** *Declared none.*

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