TECHNICAL ARTICLE



The Impact of Mine Tailings on the Witwatersrand and the Surrounding Water Bodies in Gauteng Province, South Africa

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Received: 4 March 2016 / Accepted: 5 June 2017 / Published online: 24 June 2017 © Springer-Verlag GmbH Germany 2017

Abstract The gross alpha and beta activity in treated water from one of the numerous gold mines in Gauteng province averaged 1.15 ± 0.13 and 0.87 ± 0.11 Bq L⁻¹, respectively. The average readings of the fissure water from the same mine were 0.56 ± 0.10 and 0.52 ± 0.11 Bq L⁻¹, respectively. Water samples collected at the foot of the Princess gold mine tailings dump in Gauteng province and from downstream, before the drainage joins other water sources, also had high average gross alpha and beta activity and high average concentrations of radionuclides. The hazard quotient value evaluated, based on 238 U of the water sample types in these vicinities, was far above one, indicating that it may pose serious health risks to the inhabitants.

Introduction

There are five mining basins in South Africa, which collectively can be considered the largest gold and uranium mining basin in the world (Naiker et al. 2003). Among the basins, three of them are currently at risk of flooding

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(Tutu et al. 2008). The West Rand basin (the smallest of the basins) has been fully flooded with acid mine water for 10 years and acid mine water has flowed uncontrolled and untreated during this period into the receiving environment (Naiker et al. 2003). The acid water in the basin contains uranium, manganese, aluminium, and copper, potentially toxic and radioactive metals that flow uncontrolled into the river systems, the receptor dams, the soil, and the environment (Blowes et al. 2003; Naiker et al. 2003; Tutu et al. 2008). In 2002, as an emergency measure, the mines and the South African government pumped the acidic mine water into Robinson Lake. The uranium concentration levels rose to 16 mg L⁻¹, 40,000 times than the permissible uranium levels for fresh water. As a consequence, the lake was declared a radiation area (Jones et al. 1989; Winde 2004).

Radionuclides release energy as gamma rays and energetic particles (alpha and beta particles) when they decompose to a more stable elements (Meindinyo and Agbalagba 2012). This ionization can damage living cells directly, by breaking the chemical bonds of important biological molecules (e.g. DNA), or indirectly, by creating chemical radicals from water molecules in the cells that can then chemically affect the biological molecules (UNSCEAR 1993). To some extent, these molecules are repaired by natural biological processes; however, the effectiveness of this repair depends on the extent of the damage (Radespiel-Troeger and Meyer 2013). The cells may die, or their natural functioning may be impaired, leading to somatic effects (such as cancer) or a mutation that could affect later generations (ATSDR 1999).

Naturally-occurring radionuclides are present in a wide range of concentrations in all rocks, soil, and water. The occurrence and distribution in ground water is controlled primarily by the local geology and geochemistry

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as well as anthropogenic activity (USEPA 1990, 2010). Mining activities has greatly altered the water quality in the crystalline rocks of Johannesburg and its environs (Durand 2012). ²³⁵U and ²³²Th 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 the uranium or thorium, which in turn are harmful to the human health when ingested (Benedik and Vasile 2008; Mariette 2014).

When a radionuclide gets into the human body through ingestion, inhalation, or absorption through the skin, it continues to decay, emitting radiation such as α , β , or γ particles so that the organ or tissue is continuously irradiated. Some radionuclides are chemically similar to other elements and mimic those elements in the organs (Arnold et al. 1992). For instance ²²⁶Ra, ³⁸Sr, and ⁵⁶Ba, are chemically similar to ²⁰Ca in the bone and so can be deposited in the bone marrow, causing bone cancer (Arnold et al. 1992).

The Witwatersrand Basin has been subjected to geological exploration, mining activities, industrial development, and associated settlement over the past century. The Gauteng and Princess gold mines brought with them not only development, employment, and wealth, but also pollution, negative health impacts, and ecological destruction. One of the most consistent and pressing problems has been mining's impact on the water bodies in and adjacent to the Witwatersrand. The dewatering and re-watering of the karstic aquifer overlying and adjacent to the Witwatersrand Supergroup and the pollution caused by acid mine drainage (AMD) are some of the profound consequences of gold mining that continue to affect the lives of many South Africans.

The major exposure pathway considered in this study is ingestion. People living around these mines are exposed directly because the contaminated water is readily available. There are three water types:

- Fissure water, which leaches out from inside the mine in Gauteng and flows into the community where it is used for drinking, farming, and cooking;
- Water that has been treated by the mine before being discharged; and
- Surface water from the area surrounding Princess gold mine in Roodepoort within Gauteng Province. The surface water is used for drinking and irrigation.

These three water types also leach into the groundwater and surrounding wells, and then are used in drinking and cooking. Hence, this study was intended to:

- Determine the gross alpha and gross beta activity in fissure water, treated groundwater, and surface water from around the abandoned Princess Gold and Gauteng mines,
- Determine the radionuclides concentration in the three water types, and
- Assess the radiological health risks affecting the people living near these mines.

The Witwatersrand and Water Distribution

Johannesburg, the capital of Gauteng province, is one of the few major cities in the world that was not founded near a lake or a river. The small rivers and groundwater along the Witwatersrand cannot meet the needs of its rapidly growing population (Schneiderhan 2008), and in recent years, the need for additional water has become more pressing. However, the karstified dolomites of the Malmani Subgroup of the Transvaal Supergroup contain more water than the Vaal Dam, which was built in the Vaal River between 1916 and 1923 to supply the Witwatersrand, and especially Johannesburg with water (Tempelhoff 2001). This dolomitic aquifer is the most reliable source of water for several towns, rural communities, and farms in the area. It extends from the North-West Province, to Gauteng, and into the Mpumalanga and Limpopo Provinces, and could supply the population and industries of the Witwatersrand if it were not polluted with sewage, industrial waste, and effluent from the gold mines of the Witwatersrand. Figure 1 is a map of the study area in Gauteng province and shows the tailings dumps, rivers, and streams.

Sample Collection

The abandoned Princess gold mine was sampled in January, April, August, and December, 2014. Water samples were collected at the foot of the dump (sample ID Z1005), along the water flow path (5 m before the drainage pipe-sample ID Z1004), and 5 m after the drainage pipe (sample ID A1004), before it joins other water sources (Table 1). Figure 2 is a map of the abandoned Princess gold mine tailings pile. Treated water (A1005) and fissure water (B1006) samples were also collected from one of the numerous gold mines in Gauteng province (Table 1) in January, April, August, and December, 2014.

The 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 (WIN-121) using WhatmanTM 1005-070 grade 5 filter paper with a pore size of 2.5 µm to remove coarse material and suspended and colloidal particles. Thereafter,



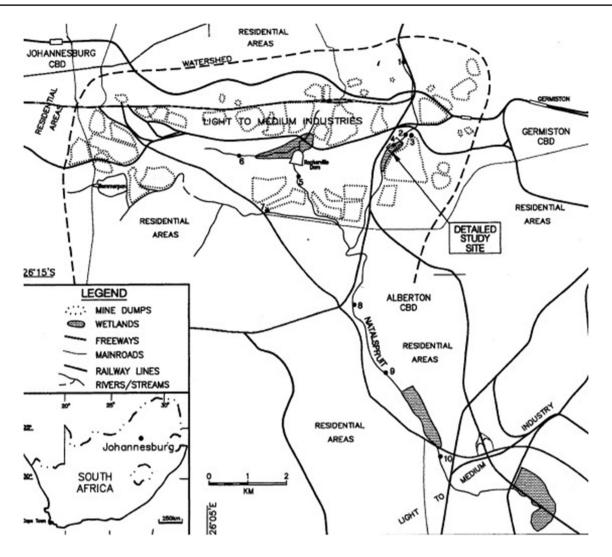


Fig. 1 A map of the study area in Gauteng province showing the tailings dumps, rivers/streams

Table 1 Sample ID and descriptions

Sample ID	Description		
A1005	Treated water sample collected from one of the numerous gold mine in Gauteng province		
B1006	Fissure water sample collected from one of the numerous gold mine in Gauteng province		
A1004	Water samples from Princess gold mine, collected 5 m after the drainage pipe, before it joins other water sources		
Z1004	Water samples from Princess gold mine, collected along the water flow path (5 m before the drainage pipe)		
Z1005	Water samples from Princess gold mine, collected right at the foot of the dump (tailing)		

the water samples were preserved with nitric acid (HNO₃) to ensure that radionuclides did not adsorb on the container walls.

Experiments

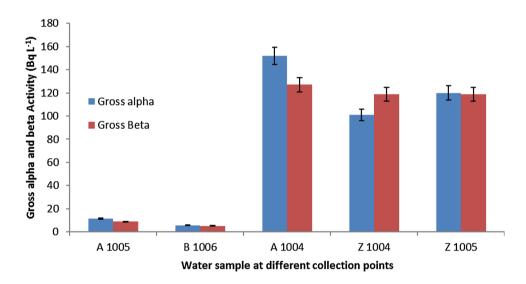
The water samples were prepared and analysed in the Nuclear Energy Corporation of South Africa (NECSA) laboratory. Water volumes of 50 mL each were evaporated to dryness and counted for 300 min on a gas proportional counter to screen for gross alpha\beta activity (WIN-161). The results were analysed using Microsoft Excel (Fig. 3). The water samples were further analysed for selected radionuclides in the uranium and thorium decay series using alpha spectrometry (WIN-145 to WIN-129).





Fig. 2 A map of the abandoned Princess gold mine tailings (right) of the study area marked 'A' (left)

Fig. 3 Gross alpha and beta distribution in the water samples



The uranium radionuclides (²³⁸U, ²³⁴U, and ²³⁵U) were first extracted from the water samples using solid phase extraction, before extraction on a cation exchange resin spectrometry. Then, 200 mL of each water sample were acidified with HNO3, pre-concentrated to a volume of 10 mL and loaded onto a pre-conditioned Truspec resin, after which were rinsed with diluted HNO3 and hydrochloric acid (HCl). The analytes were then eluted with 15 mL of 0.1 M HCl/0.1 M hydroflouric acid (HF) to make the first eluent. These were then purified by loading them on a pre-conditioned cation exchange resin and thereafter rinsed with 10 mL of 0.1 M HCl/0.01 M HF, and then eluted with 15 mL of 2 M HCl to make the second eluent. Then, 0.1 mL lanthanum oxide, and 0.5 mL of concentrated HF and titanium chloride was added to the second eluent to form a coprecipitate of uranium with lanthanum flouride. 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; the extraction yield of this method (WIN-142) was evaluated using a ²²⁹Th tracer (Benedik and Vasile 2008; Dimova et al. 2003). The thorium radionuclides (²³⁰Th, ²²⁷Th, and ²²⁸Th) were then extracted and analysed in the same manner. The results are shown in Table 2 (Fig. 4).

The radium radionuclides (²²⁶Ra, ²²³Ra and ²²⁴Ra) were extracted by adding barium solution to 500 mL of each water samples to form co-precipitates with barium sulphate in an acidic solution. The precipitate was further enhanced by adding 0.1 M ethylenediaminetetraacetic acid (EDTA) into the solution and the extract were analysed using the alpha spectrometer equipped with alpha spec Genie software. Extraction yield of this method (WIN-124) was evaluated using a ¹³³Ba tracer and the results are shown in Table 2. Finally, for the polonium radionuclide (²¹⁰Po),

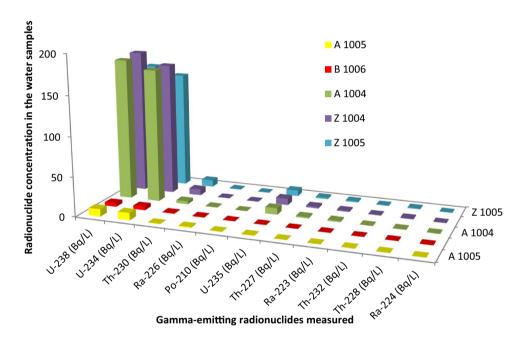


Table 2 Results of mean radionuclides activity in the water samples

Nuclides	A1005 (Bq L^{-1})	B1006 (Bq L ⁻¹)	A1004 (Bq L ⁻¹)	Z1004 (Bq L ⁻¹)
²³⁸ U	9.22 ± 0.29	4.46 ± 0.22	178.00 ± 3.00	181.00 ± 3.00
^{234}U	9.10 ± 0.29	4.54 ± 0.23	168.00 ± 3.00	167.00 ± 2.00
²³⁰ Th	0.40 ± 0.08	0.78 ± 0.13	3.87 ± 0.60	7.67 ± 1.17
²²⁶ Ra	0.99 ± 0.05	0.31 ± 0.03	0.39 ± 0.06	0.43 ± 0.04
²¹⁰ Po	0.14 ± 0.04	0.08 ± 0.13	0.05 ± 0.02	0.18 ± 0.05
^{235}U	0.43 ± 0.01	0.21 ± 0.01	8.17 ± 0.13	8.32 ± 0.12
²²⁷ Th	0.05 ± 0.03	0.05 ± 0.03	1.63 ± 0.10	2.23 ± 0.12
²²³ Ra	BDL	0.01 ± 0.02	1.47 ± 0.12	1.43 ± 0.09
²³² Th	0.07 ± 0.02	0.05 ± 0.01	0.24 ± 0.04	0.59 ± 0.06
²²⁸ Th	0.02 ± 0.01	0.07 ± 0.02	0.54 ± 0.06	0.79 ± 0.07
²²⁴ Ra	BDL	0.03 ± 0.01	0.09 ± 0.40	0.11 ± 0.03

The uncertainty is calculated mainly from counting statistic and is not the standard deviation obtained from replicate measurements

Fig. 4 Distribution of radionuclides in water samples



200 mL water samples were acidified with HCl and then 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 spontaneous deposition of polonium onto the surface of the silver disk and the solution was stirred for 4 h (Benedik and Vasile 2008; Dimova et al. 2003). The plated disk was then removed and dried prior to analysis (WIN-129) on the alpha spectrometer (Table 2).

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.

Results and Discussion

The average measured values of the gross alpha and beta activity concentrations (Fig. 3) in the water samples were high compared to the national primary interim drinking water regulations (NIPDWR 2015) and the gross beta activity for Z1004 (the sample collected 5 m before the drainage pipe) was higher than the gross alpha activities of the other samples. This could be due to the proximity of the slimes since the gross beta activity in a water sample primarily consists of uranium decay products, which were high for



Z1004 (Table 2). The WHO (2004) recommends that gross beta activity be less than 1.0 Bq L^{-1} . The measured gross beta activity of all water samples were several times 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 (DWAF 2004a, b, c). Table 2 shows the activity concentration of radionuclides from the uranium and thorium series.

The average radioactivity of the treated water (A1005) was higher than the fissure water (B1006), except for ²³⁰Th and ²²⁸Th. This implies that the water before treatment generally increased the water's radioactivity.

Water sample A1004, collected 5 m after the drainage pipe, had elevated mean 232 Th values of 0.24 ± 0.04 , but sample Z1004, collected 5 m before the drainage pipe, had the highest mean value (0.59 ± 0.06) . These two sampling locations also had higher average values of 238 U (Table 2). The 238 U and 232 Th concentrations in water samples collected at these two locations, relative to the foot of the dump, aligns with the fact that the mine dumps contribute uranium, manganese, aluminium, and copper to the flowing water (Naiker et al. 2003).

The mean activity concentration of ^{238}U (Table 2) was very high in all of the water samples, ranging from $4.46\pm0.22~Bq~L^{-1}$ (B1006) to $181\pm3.00~Bq~L^{-1}$ (Z1004), which may be due to the half-life of ^{238}U . However, the chemical toxicity risk or lifetime average daily dose (LADD) of ^{238}U is significant. The maximum recommended contaminant level (MCL) is 30 $\mu g/L/day$ (WRC 1997; WHO 2011). The German drinking water MCL guideline is 10 $\mu g/L/day$ (Trinkw 2001). The health risk assessment of ^{238}U in the water samples was evaluated relative to lifetime cancer risk using Eq. 1:

$$LCR = PCAI \times R_C \tag{1}$$

where LCR=lifetime cancer risks, PCAI=per capita activity intake, and R_c =risk coefficient. From the average life expectancy at birth (62.2 years; SAMRC 2015) and the annual water consumption by an individual (731 L; UNSCEAR 2000), we calculated the lifetime intake of water (LTI) to be 45468.2 L From USEPA (1996) and UNSCEAR (2000), 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 Eq. (1), the cancer mortality and morbidity risks of 238 U over lifetime consumption of the water were calculated (PCAI = Activity Bq L⁻¹ × LTI; Table 3).

Equation (2) was used to evaluate the chemical toxicity risk or LADD of ²³⁸U through water intake to determine the effect of the carcinogenic and non-carcinogenic risks associated with ²³⁸U in the water samples.

Ingestion LADD of drinking water

$$= \left(\frac{EPC \times ED \times EF \times IR}{BW \times AT}\right) \mu kg^{-1} day^{-1}$$
 (2)

where LADD=lifetime average daily dose, EPC=exposure point concentration ($\mu g \ L^{-1}$), 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 USEPA (1996) and WHO (2004) we obtained the following data for LADD and hazard quotient: IR=2 L day⁻¹, EF=350 days, ED=62.2 years, AT=22,703 (obtained from 62.2×365) and BW=70 kg as a standard value for a man. The following conversions were made:

$$1Bq/L = 27.0pCi/L, \ \mu/L = \frac{pCi/L}{0.67}$$
 (3)

The hazard quotient is defined as the ratio of the potential exposure to a substance and the level at which no adverse effects are expected. It was calculated by dividing the 238 U LADD of the drinking water intake by the reference dose of $0.6~\mu g~kg^{-1}~day^{-1}$ (USEPA 1996). The hazard quotient of the water samples were far above 1 (Table 3), which indicates that adverse health effects can be expected. The cancer mortality, cancer morbidity, LADD, and hazard quotient for 238 U over a lifetime of consuming the waters in the study sites were estimated (Table 3). The cancer mortality value was found to be 10^2 times higher and the cancer morbidity was 10^4 times the acceptable level of $1/10^3$ (USEPA 1996) at location Z1004.

High hazard quotient values were also observed in the water samples around the Princess gold mine slimes and the ²³⁴U and ²³⁸U concentrations were relatively high in A1005 and B1006 (treated and fissure water samples,

Table 3 The hazards parameters of ²³⁸U in the water samples

Location	Cancer mortality risk	Cancer morbidity risk	LADD (μg kg ⁻¹ day ⁻¹)	Hazard quotient
A1005	4.74E+02	7.25E+02	2.53E+05	4.21E+05
B1006	2.29E+02	3.51E+02	1.22E+05	2.04E+05
A1004	9.15E+03	1.40E+04	4.88E+06	8.13E+06
Z1004	9.30E+03	1.42E+04	4.96E+06	8.26E+06
Z1005	8.07E+03	1.23E+04	4.30E+06	7.17E+06



Table 4 Dose rates for different age groups

Sample ID	Annual dose rate in (μSv year ⁻¹)						
	<1 year	1–2 years	2–7 years	7–12 years	12-17 years	>17 years	
A1005	420	150	110	110	220	120	
B1006	430	150	100	95	160	97	
A1004	5100	1700	1300	1200	2100	1600	
Z1004	6300	1900	1400	1400	2300	1800	
Z1005	5700	1700	1300	1200	2100	1600	

respectively). Such high concentrations in drinking water can cause a high incidence rate of renal cell carcinoma (kidney cancer).

Water contamination by radionuclides is a significant environmental problem, particularly in densely populated developing countries where human habitats are usually in close proximity to mine sites (Scott 1995). Water draining from gold mines slimes frequently contains radionuclides at high levels. This study confirmed the high values of radionuclides pollution affecting freshwater systems and resources in South Africa (McCarthy 2010). Mine water is a growing concern in water quality management and negatively impacts the water environment by increasing the levels of radionuclides, resulting in increasing cases of kidney cancer (Radespiel-Troeger and Meyer 2013).

Dosage rates due to the radionuclides measured in the water samples were calculated according to guidelines in the National Nuclear regulatory licensing guide (LG-1032; NNR 2015). Consumption rates for the different age groups in this guide and dose rate conversion factors published in the IAEA (1996) Safety Series 115 were used. Where the activity was less than the MDA, half of the values were used in the calculation and the following radioactive equilibria were assumed; ²³⁸U up to ^{234m}Pa (²³⁸U), ²²⁶Ra up to ²³⁸Bi (²²⁶Ra), ²¹⁰Pb and ²¹⁰Po (²¹⁰Po), ²³¹Pa up to ²²⁷Th (²²⁷Th), ²²³Ra and ²¹¹Pb (²²³Ra), ²³⁵U and ²³¹Th (²³⁵U), ²²⁸Ra up to ²²⁸Th (²²⁸Th), and ²²⁴Ra up to ²¹²Bi (²²⁴Ra). The dose rate values were reported with 2 significant digits (Table 4).

According to National Nuclear Regulator (NNR 2015) Guide 002, the dosage rates constraint for members of the public within the exposed population is 250 μ Sv year⁻¹ (DWAF 2004d, e, f). The annual dosage rate for <1 year ranged from 420 to 6300 μ Sv year⁻¹ (Table 4). It is evident that the water quality around the Princess mine slimes threatens the surrounding community and that the water quality of the treated and untreated fissure water exceeds the DWAF standards (2004a, b, c, d, e, f).



This study has indicated that surface ground water around the abandoned Princess gold mine is contaminated due to its tailings dumps, which is not only affecting the quality of surface water, but is also contributing radioactivity to the surface aquatic environment of the Witwatersrand watershed. This study shows that around the abandoned Princess gold mine, the average values of gross alpha and beta activity in the water samples were approximately 20 times higher than the fissure water and the treated water from one of the numerous gold mines in Gauteng province. A similar trend was observed for the average naturally occurring radionuclide (238U, 234U, 230Th, 226Ra, ²¹⁰Po, ²³⁵U, ²³⁷Th, ²²³Ra, ²³²Th, ²²⁸Th, and ²²⁴Ra) activity concentrations for the three types of water samples analysed in this study. The concentrations of the natural radionuclides were relatively high in all of the water sample types and the calculated chemical toxicity, cancer mortality/morbidity, and hazards quotient with respect to ²³⁸U were high. Hence, the water within these vicinities may pose a serious health risk to the inhabitants due to ingestion exposure. This study emphasised that abandoned mines, mine closure, and especially environmental rehabilitation need to be addressed urgently. According to the Department of Mineral Resources and Brown (2007), there are over 8000 dilapidated and ownerless mines in South Africa, which would take approximately 800 years to rehabilitate at a huge cost.

Acknowledgements The author thanks the Council for Geoscience for their support of current work.

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