



Toxicity of natural radioactivity in water samples from a gold mine in Gauteng Province, South Africa

Caspah Kamunda

Department of Physics, School of Mathematics and Natural Sciences, The Copperbelt University, Kitwe, Zambia

Corresponding author e-mail: ckamunda@gmail.com

ARTICLE INFO

Received 13/5/2020; received in revised form 22/6/2020; accepted 7/7/2020. DOI: <u>10.6092/issn.2281-4485/10995</u> © 2020 The Authors.

Abstract

A radiological toxicity assessment of natural radioactivity in water samples from a gold mine in the Gauteng Province of South Africa was evaluated. In this study, 29 water samples were collected from the mine and 5 were collected from a control area. A broad energy germanium detector was used to measure the activity concentrations of Uranium-238, Thorium-232, and Potassium-40. Their activity concentrations were then used to calculate radiological hazard parameters in water for the area. The average value of the Radium Equivalent Activity (Ra_{eq}) in water samples from the mining area and from the control area was found to be lower than the allowable limit of 370 Bq.L⁻¹. The Annual Effective Dose Equivalent (AEDE) was found to be equal to 1.29×10^{-3} mSv.y⁻¹, a value below worldwide average of 2.4 mSv.y⁻¹ for natural radiation. The external hazard (H_{ex}) and internal hazard (H_{in}) index for water samples from the mining area were less than unity, making water samples safe to the population in the area.

Keywords

broad energy germanium detector, natural radioactivity; activity concentration, gold mine, radiological hazard

Introduction

In general, natural radiation in the environment occurs at levels that are not potentially harmful to human health (Modisane, 2005). A major concern comes when concentrations of this natural radiation are enhanced as a result of anthropogenic activities like mining (Nour et al., 2005). These enhanced concentrations of radiation are of concern to radiation protection and have been a subject of research in recent times. A commonly referred term to these elevated levels of radiation is Naturally Occurring Radioactive Materials (NORMs). NORMs account for up to 85% of the annual dose exposure received by the world's population (WNA, 2014). The NORMs that are involved with mining activities are non-decay series potassium-40 and decay series radionuclides of uranium (U) and thorium (Th) (IAEA, 2009). Once these radionuclides are in the ecosystem, they accumulate in plants and eventually get ingested by humans in high concentrations (Paul and Campbell, 2011). When ingested or inhaled, radionuclides enter the human body and get assimilated by body organs. Health effects may manifest not merely as cancers, but may extend to non-cancer illnesses such as eye lens destruction, neurological illnesses, diabetes, and several other radiogenic illnesses (Busby, 2010).

In South Africa, the mining industry primarily covers the extraction of minerals like gold (Au), uranium (U), platinum (Pt), vanadium (V), manganese (Mn), Iron (Fe) and diamonds. For more than a century, gold mining has been carried out in South Africa particularly in the Wonderfonteinspruit Catchment Area (WCA) in the Gauteng Province (Winde et al., 2004). In this area uranium is the principal contaminant of concern within the gold mining area, and is estimated to be almost ten times the amount than gold (GDARD, 2009). The existence of large quantities of this uranium in the area induces a radiological burden to mankind. People residing in the WCA reportedly use mine drainage water in households (Liefferink, 2011). In the WCA, a number of radiological studies, by different experts have been carried out, but in the gold mining area in question, no risk assessment of radionuclides has been carried out. It is for this reason that a radiological toxicity assessment of NORMs in water samples was carried out from a gold mining area in the Gauteng Province of South Africa.

Materials and Methods

Study Area

The study area is situated near the West Wits line (Far West Rand) in the Gauteng province of South Africa, close to the town of Carletonville and is approximately 70 kilometres southwest of Johannesburg. It lies between 26°18`S - 26°26`S latitude and 27°23`E - 27°31`E longitude. Gold exploration in the area dates back to 1898 and mining started from 1945 to date. The area, which is approximately 86 km², lies in the lower central part of the WCA. Mining activities in the area are engaged in both deep-level (500 m - 4,000 m), high-grade underground mining as well as low-grade, surface rock dump mining.

Sampling

A total of twenty nine (29) water samples, of roughly two litres each, were collected in polyethylene bottles using a muslin cloth as a strainer, from each selected location in the gold mining area. From the control area, 5 water samples, (-two litres each), were also collected from the North West Province. All the bottles had to be washed with dilute acid (0.1 M HCl) before use to avoid contamination. Because the bottles had to be stored for a long time before analysis, 11 M HCl was added to the containers at the rate of 10 ml per litre of sample immediately after collection to avoid absorption of radionuclides on the walls of the containers and placed in a cold room (IAEA, 1989). All the collected samples were properly marked for identification. All sampling locations were also identified using Global Positioning System (GPS). The sampling procedure was carried out according to methodologies recommended by APHA (1985). Measurements of these samples were carried out at the Centre for Applied Radiation Science and Technology (CARST), North West University, South Africa. At the CARST laboratory, the two litres of each water sample were prepared by transferring them into marinelli beakers and sealing them for 28 days to establish secular equilibrium in readiness for gamma spectroscopy (IAEA, 2004). Sealing establishes secular equilibrium among the progenies of ²³⁸U and ²³²Th series by ensuring that there is no radon gas leakage due to the decay of ²²⁶Ra.

Measurement of activity concentrations

The measurements of activity concentrations of ²³⁸U, ²³²Th and ⁴⁰K in water samples were then carried out by means of Gamma Spectrometry using a Broad Energy Germanium (BEGe) detector, The broad energy germanium (BEGe) detector (BE6530), manufactured by Canberra Industries has a relative efficiency of 60 % and a resolution of 2.0 keV for 1332 keV gamma ray emission of 60Co. (Canberra Industries, 2013). Energy and efficiency calibration of the gamma spectrometer were performed before sample measurements. In all the measurements, quality control and quality assurance procedures were performed to check for instrument performance. These included regular background and calibration source measurements. The counting geometry was created during efficiency calibration and carefully duplicated for all the measurements.

The background radiation around the detector inside the shielding was measured using an empty marinelli beaker. The gamma photons from the water samples were then detected by placing the marinelli beaker directly over the BEGe detector which is usually lead shielded to avoid background radiation. This was done under similar measurement geometry as background radiation. To allow for detectable activity, background radiation and each water sample was counted for 24 hours. The background was then subtracted from the measured gamma spectrum of each sample before calculating the activity concentrations. The measuring process and analysis of spectra was computer controlled using GENIE 2000 software (Canberra Industries, 2013a). The activity concentration of ²³⁸U was determined by measuring the 295.2 keV (19.7 %) and 351.9 keV (38.9 %) gamma-ray energies of ²¹⁴Pb and 609.3 keV (43.3 %), 1120.3 keV (15.7 %) and 1764.5 keV (15.1 %) gamma from ²¹⁴Bi. The activity concentration of ²³²Th was determined from gamma energies 238.6 keV (44.6 %) of ²¹²Pb and 338.3 keV (11.4 %), 911.6 keV (27.7 %) and 969.1 keV (16.6 %) for ²²⁸Ac. On the other hand, the activity concentration for ⁴⁰K was measured from its 1460.8 keV (10.7 %) gamma-ray line (Kamunda et al., 2016).

The activity concentrations of 238 U, 232 Th, and 40 K were determined using Equation [1] (Dovlete & Povinec, 2004).

$$A = \frac{N}{\varepsilon_f P_{\gamma} t_s m K}$$
[1]

where

N = the corrected net peak area of the corresponding full-energy peak

 $N = N_{S} - N_{B}$

 N_s = the net peak area in the spectrum of the sample

 N_B = the corresponding net peak area in the background spectrum

 $\varepsilon_f =$ the efficiency at photo peak energy $t_s =$ the live time of the sample spectrum collection in seconds

m = the mass (kg) of the measured sample P_{γ} = the gamma-ray emission probability corresponding to the peak energy K = the correction factor.

Calculation of Radiological Parameters

The radiological hazard of water samples can be assessed through various hazard estimating models.

Radium Equivalent Activity (Ra_{eq}). The Radium Equivalent Activity (Ra_{eq}) is defined as the weighted sum of ²³⁸U (which can be that of ²²⁶Ra), ²³²Th and ⁴⁰K activity concentrations, based on the assumption that 370 Bq.L⁻¹ of ²²⁶Ra, 259 Bq.L⁻¹ of ²³²Th and 4810 Bq.L⁻¹ of ⁴⁰K produce the same gamma-ray dose rate. In these

quantities, each of these radionuclides gives an effective dose of 1.5 mGy per year (Santawamaitre, 2012), which is assumed to be the maximum permissible dose to a human being from their exposure to natural radiation from water. On the basis of these values, $Ra_{eq}Ra_{eq}$ is defined as follows:

$$Ra_{eq} = \left(\frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810}\right) \times 370$$
 [2]

which is equivalent to

$$Ra_{sa} = A_{Ra} + 1.43A_{Th} + 0.077A_{K}$$
^[3]

where $A_{Ra}A_{Ra}$, $A_{Th}A_{Th}$ and $A_{K}A_{K}$ are the activities concentration in Bq.L⁻¹ of ²²⁶Ra , ²³²Th, and ⁴⁰K, respectively. The $Ra_{eq}Ra_{eq}$ is therefore a single index or number to describe the gamma output from different mixtures of radionuclides in a material.

Absorbed Dose Rate in Air (D). The outdoor absorbed dose rate in air due to terrestrial gamma rays at 1m above the ground surface are calculated from ²²⁶Ra (²³⁸U), ²³²Th and ⁴⁰K concentration values in water assuming that the other radionuclides, such as ¹³⁷Cs, ⁹⁰Sr and the ²³⁵U decay series can be neglected as they contribute very little to the total dose from environmental background (Kocher and Sjoreen, 1985). In general the estimates of ²²⁶Ra and ²³²Th concentration are based on the assumption of radioactive equilibrium conditions through the direct measurement of daughter isotopes. The absorbed dose rate is therefore given by the following formula (UNSCEAR, 2000):

$$D(nGy.h^{-1}) = 0.462A_{Ra} + 0.604A_{Th} + 0.0417A_{K}$$
 [4]

where *D* is the absorbed dose rate, $A_{Ra}A_{Ra}$, $A_{Th}A_{Th}$ and A_KA_K are the activities concentration of ²²⁶Ra (²³⁸U), ²³²Th and ⁴⁰K, respectively. The dose coefficients in units of nGy.h⁻¹ per Bq.L⁻¹ were taken from the UNSCEAR (2000) report.

Annual Effective Dose Equivalent (AEDE) from Terrestrial Gamma Radiation. The absorbed dose rate in air at 1 metre above the ground surface does not directly provide the radiological risk to which an individual is exposed. The absorbed dose can be considered in terms of the annual effective dose equivalent from outdoor terrestrial gamma radiation which is converted from the absorbed dose by taking into account the conversion coefficient from absorbed dose in air to effective dose and the outdoor occupancy factor (Santawamaitre, 2012).

Using the conversion coefficient of 0.7 SvGy^{-1} from absorbed dose in air to effective dose received by adults and 0.2 as the outdoor occupancy factor proposed by UNSCEAR (2000), the annual effective dose equivalent can be estimated by the following formula:

 $AEDE(mSv. y^{-1}) = D(nGy.h^{-1}) \times 8760 h \times 0.2 \times 0.7 Sv. Gy^{-1} \times 10^{-6}$ [5]

where 8760 is the time in hours for one year; 10⁻⁶ is the factor converting from nano to milli.

Annual Effective Dose Equivalent from Ingestion of Water Samples. The annual effective dose (E_{ing}) from ingestion of ²³⁸U, ²³²Th and ⁴⁰K through water samples for members of the public is estimated from the activity concentrations of each individual radionuclides found in the sample. Equation [6] was used in the calculation (UNSCEAR, 2000) as follows:

$$E_{ing}(mSv.y^{-1}) = A_R IR_{ing} \sum_{j=1}^{3} DCF_{ing}$$
[6]

Where, A_R = the activity concentration of the radionuclides in a sample IR_{ing} = the consumption rate per year, and DCF_{ing} = the effective dose coefficient in Sv.Bq⁻¹ for the ingestion of natural radionuclides. For ²³⁸U, a value of 4.50 × 10⁻⁸ was used. For ²³²Th, 2.30 × 10⁻⁷ was used and 6.20 × 10⁻⁹ was used for ⁴⁰K (ICRP, 2012).

For water samples, the annual effective doses were estimated by applying an annual water consumption rate of 600 L/year for adults and in the case of plant samples, an average consumption rate of 200 kg/year was used (DEA, 2010).

Radiation Hazard Indices. To limit the radiation exposure attributable to natural radionuclides in the samples to the permissible dose equivalent limit of 1 mSv.y⁻¹, the External Hazard Index (H_{ex}) was introduced using a model proposed by Krieger (1981). The value of this H_{ex} must be less than unity in order to keep the radiation hazard to be insignificant. This means that the maximum value of H_{ex} equal to unity corresponds to the upper limit of Radium Equivalent Activity of 370 Bq.L⁻¹ (Beretka & Mathew, 1995). This is a widely used hazard index reflecting the external exposure and is defined by Equation [7] as follows (UNSCEAR, 2000):

$$H_{ex} = \left(\frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810}\right)$$
[7]

where $A_{Ra}A_{Ra}$, $A_{Th}A_{Th}$ and $A_{K}A_{K}$ are the mean activities concentration of ²²⁶Ra (²³⁸U), ²³²Th and ⁴⁰K, in Bq.L⁻¹ respectively.

In addition to H_{ev}, radon and its short-lived products

are also hazardous to the respiratory organs. The internal exposure to radon and its daughter products is quantified by the Internal Hazard Index (H_{in}) , which is given by the Equation [8] (UNSCEAR, 2000):

$$H_{in} = \left(\frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_{K}}{4810}\right)$$
[8]

The value of H_{in} must also be less than unity for the radiation hazard to be negligible.

Results and Discussion

Activity concentrations of $^{238}\text{U},~^{232}\text{Th}$ and ^{40}K in water samples

The average activity concentrations of ²³⁸U, ²³²Th and ⁴⁰K derived from the weighted average of selected gamma energy peaks of their daughter radionuclides in each water sample are shown in Table 1.

The average values for ²³⁸U, ²³²Th and ⁴⁰K in Bq.L⁻¹ were 0.66±0.03, 0.56±0.03 and 7.36±0.58, respectively. Most of the water samples investigated were from borehole and surface water from around the mining area. The water is used largely for drinking. The highest activity concentrations were measured in water samples from surface water. Lowest values were recorded from borehole water used for

domestic use. Compared with samples from the control area, average values for

²³⁸U, ²³²Th and ⁴⁰K in water samples from the mining area were higher.

Radiological Hazard Assessment for Water Samples

Table 2 shows a summary of the different calculated radiological parameters of water samples from the mining area and from the control area.

The average value of Ra_{eq} in Bq.L⁻¹ from the mining area was 2.03±0.07. This was significantly higher than 1.39±0.08 Bq.L⁻¹, a figure estimated from the control area. The results also revealed that maximum values were observed from water samples collected close to mine tailings. The values recorded in all the water samples were however, lower than the allowable limit of 370 Bq.L⁻¹ recommended by IAEA under normal circumstances (OECD, 1979).

From the calculations, the average absorbed dose rate due to 238 U, 232 Th, and 40 K in water samples varied from 0.54±0.02 to 1.54±0.05 nGy.h⁻¹ with an average of 0.95±0.03 nGy.h⁻¹. This was also below the worldwide

average of 59 nGy. h^{-1} (UNSCEAR, 2000). The average value of the absorbed dose rate of radionuclides through water samples from the mining area was as expected

higher than the value obtained from the control area, which was $0.68{\pm}0.04~n{\rm Gy.h^{-1}}.$

Table 1. Average activity concentrations of ^{238}U , ^{232}Th and ^{40}K for water samples from the mining area and from the control area.

Lastian	Samula ID	Latituda	Lanaituda		Average (Bq.	L-1)
Location	Sample ID	Latitude	Longitude	²³⁸ U	²³² Th	⁴⁰ K
	CW1	25°47'33.06"S	25°38'02.04"E	ND	0.32 ± 0.01	9.60±0.83
	CW2	25°47'34.80"S	25°38'19.20"E	0.10 ± 0.00	0.43 ± 0.02	10.02 ± 1.01
	CW3	25°47'41.58"S	25°38'03.36"E	ND	0.31±0.01	9.18±0.76
Control	CW4	25°47'59.70"S	25°37'55.20"E	0.10 ± 0.00	0.37 ± 0.02	10.12±1.13
Control	CW5	25°48'01.32"S	25°37'56.22"E	0.20 ± 0.00	0.44 ± 0.02	11.61±1.18
alca -		Average		0.10±0.00	0.38±0.01	10.11±1.11
		Minimum		0.13±0.00	0.31±0.01	9.18±0.76
		Maximum		0.20±0.00	0.44±0.02	11.61±1.18
	DSW45/1	26°26'19.74"S	27°29'54.84"E	0.47 ± 0.02	0.38 ± 0.02	5.64±0.52
	DSW11/2	26°25'28.02"S	27°29'31.50"E	0.38 ± 0.03	0.44 ± 0.02	5.70±0.52
	DSW18/3	26°23'0.12"S	27°30'14.10"E	0.51±0.03	0.59 ± 0.03	6.63±0.58
	DSW20/4	26°22'45.12"S	27°29'52.44"E	0.35 ± 0.02	0.29 ± 0.01	4.93±0.54
	DSW40/5	26°21'43.08"S	27°29'58.86"E	0.95 ± 0.05	0.74 ± 0.04	9.10±0.63
	DSW40/6	26°21'43.26"S	27°30'5.28"E	0.75 ± 0.04	0.34 ± 0.03	7.53±0.54
	DSW40/7	26°21'53.28"S	27°29'54.54"E	1.31 ± 0.07	0.84 ± 0.04	10.24±0.64
	DSW35/8	26°21'49.56"S	27°27'39.90"E	0.48 ± 0.02	0.41 ± 0.02	5.76±0.40
	DSW17/9	26°23'22.74"S	27°28'13.92"E	0.65 ± 0.03	0.48 ± 0.02	8.13±0.54
	DSW42/10	26°20'36.24"S	27°26'01.02"E	0.52 ± 0.03	0.45 ± 0.02	8.05±0.57
	DSW21/11	26°21'25.14"S	27°26'10.32"E	0.87 ± 0.04	0.64 ± 0.03	9.45±0.61
	DSW7/12	26°21'27.42"S	27°25'59.22"E	0.92 ± 0.05	0.69 ± 0.03	8.61±0.62
	DAM3/13	26°21'19.32"S	27°25'52.44"E	0.76 ± 0.04	0.64 ± 0.03	6.10±0.56
	DSW9/14	26°20'59.34"S	27°25'52.44"E	0.70 ± 0.03	0.55 ± 0.03	6.01±0.53
	DSW36/15	26°20'28.44"S	27°25'42.06"E	0.66 ± 0.03	0.61±0.03	6.85±0.57
	DSW38/16	26°19'38.10"S	27°24'15.42"E	0.51±0.03	0.49 ± 0.02	5.63±0.55
	DSW39/17	26°18'57.96"S	27°23'03.36"E	0.74 ± 0.04	0.59 ± 0.03	6.42±0.57
	DSW16/18	26°23'44.76"S	27°25'57.84"E	0.90 ± 0.05	0.71 ± 0.04	8.03±0.65
Mining	DSW43/19	26°21'57.30"S	27°26'34.98"E	0.77 ± 0.04	0.66 ± 0.03	7.28±0.62
area	DSW29/20	26°20'32.40"S	27°26'40.86"E	0.86 ± 0.04	0.69 ± 0.03	8.32±0.61
	WV2	26°23'45.90"S	27°28'43.26"E	0.45 ± 0.02	0.41 ± 0.02	7.51±0.58
	WV3	26°23'44.10"S	27°29'02.04"E	0.72 ± 0.04	0.62 ± 0.03	5.26±0.53
	WV4	26°23'38.88"S	27°29'05.16"E	0.48 ± 0.02	0.55 ± 0.03	5.54±0.56
	WV5	26°23'40.20"S	27°29'05.52"E	0.44 ± 0.02	0.45 ± 0.02	6.72±0.55
	WV9	26°23'13.80"S	27°30'59.76"E	0.73 ± 0.04	0.69 ± 0.03	7.76±0.60
	WV13	26°23'21.24"S	27°31'20.04"E	0.49 ± 0.02	0.50 ± 0.03	8.31±0.63
	WV14	26°23'23.46"S	27°31'11.70"E	0.54±0.03	0.53±0.03	7.25±0.59
	WV15	26°23'16.68"S	27°30'44.10"E	0.83 ± 0.04	0.84 ± 0.04	11.06±0.82
_	WV16	26°22'60.00"S	27°30'36.12"E	0.51±0.03	0.42±0.02	9.59±0.67
		Average		0.66±0.03	0.56±0.03	7 .36±0.58
		Minimum		0.35±0.02	0.29±0.01	4.93±0.40
		Maximum		1.31±0.07	0.84±0.04	11.06±0.82

ND: Not Detectable

Table 2. Results of calculated Absorbed Dose Rate in air (D), Annual Effective Dose Equivalent (AEDE), Radium
Equivalent Activity (Ra_{ed}), External Hazard Index (H_{ex}) and Internal hazard Index (H_{in}) of water samples from the
mining area and from the control site.

T	Samala ID	Ra	D	AEDE	H	H _{in}
Location	Sample ID	(Bq. Ŭ ⁻¹)	(nGy.h ⁻¹)	(mSv.y ⁻¹)	3 23F-03 3 23F-0	
	CW1	1.20 ± 0.07	0.59 ± 0.04	7.28E-04	3.23E-03	3.23E-03
	CW2	1.49 ± 0.08	0.72 ± 0.04	8.88E-04	4.01E-03	4.28E-03
	CW3	1.15±0.06	0.57 ± 0.03	6.99E-04	3.10E-03	3.11E-03
Control	CW4	1.41±0.09	0.69 ± 0.05	8.48E-04	3.80E-03	4.07E-03
area	CW5	1.72 ± 0.10	0.84±0.05	1.03E-03	4.65E-03	5.19E-03
	Average	1.39±0.08	0.68±0.04	8.39E-04	3.76E-03	3.98E-03
	Minimum	1.15±0.06	0.57±0.03	6.99E-04	3.11E-03	3.10E-03
	Maximum	1.72±0.10	0.84±0.05	1.03E-03	4.65E-03	5.19E-03
	DSW45/1	1.45 ± 0.05	0.68 ± 0.03	8.36E-04	3.91E-03	5.18E-03
	DSW11/2	1.45 ± 0.06	0.68±0.03	8.33E-04	3.91E-03	4.94E-03
	DSW18/3	1.86 ± 0.07	0.87 ± 0.03	1.07E-03	5.03E-03	6.41E-03
	DSW20/4	1.14 ± 0.05	0.54±0.03	6.65E-04	3.09E-03	4.04E-03
	DSW40/5	2.71±0.09	1.27 ± 0.04	1.55E-03	7.31E-03	9.88E-03
	DSW40/6	1.82 ± 0.07	0.87 ± 0.03	1.06E-03	4.91E-03	6.93E-03
	DSW40/7	3.30 ± 0.14	1.54±0.06	1.89E-03	8.91E-03	1.25E-02
	DSW35/8	1.51±0.05	0.71±0.02	8.70E-04	4.08E-03	5.38E-03
	DSW17/9	1.96±0.06	0.93±0.03	1.14E-03	5.30E-03	7.06E-03
	DSW42/10	1.78 ± 0.06	0.85±0.03	1.04E-03	4.82E-03	6.22E-03
	DSW21/11	2.51±0.08	1.18 ± 0.04	1.45E-03	6.79E-03	9.14E-03
	DSW7/12	2.57 ± 0.08	1.20 ± 0.04	1.47E-03	6.94E-03	9.43E-03
	DAM3/13	2.14±0.07	0.99±0.03	1.22E-03	5.79E-03	7.85E-03
	DSW9/14	1.95 ± 0.07	0.91±0.03	1.11E-03	5.26E-03	7.16E-03
	DSW36/15	2.06±0.07	0.96±0.03	1.18E-03	5.56E-03	7.35E-03
	DSW38/16	1.64 ± 0.06	0.77 ± 0.03	9.40E-04	4.44E-03	5.82E-03
Mining	DSW39/17	2.08 ± 0.07	0.97 ± 0.04	1.18E-03	5.61E-03	7.61E-03
area	DSW16/18	2.53±0.09	1.18 ± 0.04	1.45E-03	6.84E-03	9.28E-03
	DSW43/19	2.27 ± 0.08	1.06 ± 0.04	1.30E-03	6.14E-03	8.22E-03
	DSW29/20	2.49 ± 0.08	1.16±0.04	1.42E-03	6.72E-03	9.04E-03
	WV2	1.61±0.06	0.77 ± 0.03	9.43E-04	4.36E-03	5.58E-03
	WV3	2.01±0.07	0.93±0.03	1.14E-03	5.43E-03	7.38E-03
	WV4	1.69 ± 0.06	0.78 ± 0.03	9.63E-04	4.57E-03	5.87E-03
	WV5	1.60 ± 0.05	0.76±0.03	9.26E-04	4.32E-03	5.51E-03
	WV9	2.31±0.07	1.08 ± 0.04	1.32E-03	6.25E-03	8.22E-03
	WV13	1.84 ± 0.07	0.87 ± 0.03	1.07E-03	4.98E-03	6.31E-03
	WV14	1.86 ± 0.07	0.87 ± 0.03	1.07E-03	5.01E-03	6.47E-03
	WV15	2.88±0.09	1.35±0.05	1.66E-03	7.79E-03	1.00E-02
	WV16	1.85±0.07	0.89±0.03	1.09E-03	4.99E-03	6.37E-03
	Average	2.03±0.07	0.95±0.03	1.17E-03	5.49E-03	7.28E-03
	Minimum	1.14±0.05	0.54±0.02	6.65E-04	3.09E-03	4.04E-03
	Maximum	3.30±0.10	1.54±0.05	1.89E-03	8.91E-03	1.25E-02

AEDE values for water samples as a result of exposure to external radiation were also calculated for the mining area and for the control area. From the mining area, values ranged between 6.65×10^{-04} and 1.89×10^{-03} mSv.y⁻¹ with an average of 1.17×10^{-03} mSv.y⁻¹. An average value of 8.39×10^{-04} mSv.y⁻¹ from the control area was recorded, which is below the average from the mining area. These AEDE values were way below the worldwide average of 0.48 mSv.y⁻¹ for external terrestrial

radiation (UNSCEAR, 2000).

The average external hazard (H_{ex}) index for water samples from mining area was found to be 5.49×10^{-03} while the average internal hazard (H_{in}) index was 7.28×10^{-03} . Both values were less than unity, making water samples due to external radiation safe to the population in the study area. Equations [3], [4], [5], [7] and [8] were used to calculate Ra_{eq}, D, AEDE, H_{ex} and H_{in} parameters, respectively.

Table 3. Annual effective dose through ingestion of ²³⁸U, ²³²Th and ⁴⁰K in water samples.

Average Annual Effectiv (mSv.y ⁻¹)	ation	e Activity Concen (Bq.kg ⁻¹)	Average
	⁴⁰ K	²³² Th	²³⁸ U
1.23E-04	7.36	0.56	0.66

According to UNSCEAR (2000), the total annual effective dose from natural radiation should not be more than 2.4 mSv.y⁻¹. This also includes radiation from cosmic sources, which was not considered in the study. The total annual effective dose equivalent from the measured water samples due to external terrestrial radiation and ingestion of water was found to be 1.29 x 10⁻⁰³ mSv.y⁻¹, which was lower than the permissible limit of 2.4 mSv.y⁻¹. The value was also lower than 1 mSv.y⁻¹, a limit recommended by ICRP (2008) for individual members of the public. World Health Organisation (WHO) also recommends a value of 0.10 mSv.y⁻¹ as the upper limit for the annual effective dose in drinking water (WHO, 2008). The average annual effective dose for the water samples in the study area was clearly lower than these set limits.

Conclusions

Measurements of NORMs in water samples from the gold mining area were undertaken by means of gamma-ray spectrometry using a well shielded and well calibrated Broad Energy Germanium (BEGe) detector. The results of the activity concentrations obtained for ²³⁸U, ²³²Th and ⁴⁰K in 29 water samples from the mining area and 5 samples from the control area were recorded and compared. The results of the investigation showed that the average annual effective dose equivalent for the water samples in the study area was lower than the permissible limits as set by UNSCEAR (2000) and WHO in 2008.

<u>Acknowledgements</u>

The author would want to thank the management and staff of the gold mine for their co-operation and acceptance to conduct this research within their premises. Acknowledgement also goes to North West University's Centre for Applied Radiation Science and Technology (CARST) for disbursing the funds to go to the field and providing the necessary equipment and facilities for the measurements.

References

APHA - American Public Health Association (1985) Standard methods for the examination of water and wastewater. American Public Health Association, Washington, New York.

BERETKA J., MATHEW P.J. (1995) Natural radioactivity of Australian building materials, Industrial wastes and by-products. Health Phys., 48:87–95.

BUSBY C. (2010) Uranium and health: The health effects of exposure to uranium and uranium weapons fallout. Recommendations of the European Committee on Radiation Risk (ECRR), Brussels.

CANBERRA INDUSTRIES (2013) Broad Energy Germanium Detectors (BEGe). Nuclear Measurement Solutions for Safety, Security and the Environment. USA. www.canberra.com. Accessed 13 October 2015.

DOI: 10.6092/issn.2281-4485/10995

DEA - Department of Environmental Affairs (2010) The Framework for the Management of Contaminated Land, South Africa. <u>http://sawic.environment.gov.za/</u> <u>documents/562.pdf</u> - Accessed on 5 February 2016.

DOVLETE C., POVINEC P.P. (2004) Quantification of Uncertainty in Gamma-Spectrometric Analysis of Environmental Samples, in International Atomic Energy 133 Agency, IAEA-TECDOC-1401: Quantifying Uncertainty in Nuclear Analytical Measurements, Austria: IAEA, pp.103-126.

GDARD - Gauteng Department of Agriculture and Rural Development (2009) Conceptual study on reclamation of mine residue areas for development purposes. South Africa.

IAEA - International Atomic Energy Agency (1989) Measurement of Radionuclides in Food and the Environment. Technical Report Series No. 295, Vienna, Austria.

IAEA - International Atomic Energy Agency (2004) Soil Sampling for Environmental Contaminants, IAEA-TECDOC-1415, Vienna, Austria.

IAEA - International Atomic Energy Agency (2009) Quantification of Radionuclide Transfer in Terrestrial and Freshwater Environments for Radiological Assessments. IAEA-TECDOC-1616, Vienna, Austria.

ICRP - International Commission of Radiological Protection (2008) ICRP Publication 103. Recommendations of the International Commission of Radiological Protection, Annals of the ICRP, Vol. 32, 2-4.

ICRP - International Commission of Radiological Protection (2012) ICRP Publication 119. Compendium of Dose Coefficients based on ICRP Publication 60 Annals of the ICRP, 41, (Supplement).

KAMUNDA C., MATHUTHU M., MADHUKU M. (2016) An Assessment of Radiological Hazards from Gold Mine Tailings in Gauteng Province, South Africa. Int. J. Environ. Res. Public Health, 13:138. Doi: <u>10.3390/</u><u>ijerph13010138</u>

KOCHER D.C., SJOREEN A.L. (1985) Dose rate conversion factors for external exposure to photon emitters in soil. Health Physics, 48:193–205. Doi: <u>10.1097/00004032-198502000-00006</u>

KRIEGER R., (1981) Radioactivity of Construction Materials. Betonwerk Fertigteil Tech, 47:468-473.

LIEFFERINK M. (2011) Assessing the past and the present role of the National Nuclear Regulator as a public protector against potential health injuries: The West and Far West Rand as case study.

MODISANE T. J. (2005) Correlations between Natural Radioactive Concentrations in Soils and Vine Growth, Master's thesis.

NOUR K.A., GABAR A., ARABI M. (2005) Natural Radioactivity in Farm Soil and Phosphate Fertilizer and its Environmental Implications in Qenagovernorate, Upper Egypt. Journal of Environmental Radioactivity, 84:51-64. Doi: 10.1016/j.jenvrad.2005.04.007

OECD - Organization for Economic Cooperation and Development)., (1979). Exposure to Radiation from Natural Radioactivity in Building Materials. Report by a Group of Experts of the OECD Nuclear Energy Agency, Paris, France.

PAUL J., CAMPBELL G. (2011) Investigating rare earth element mine development in EPA region 8 and potential environmental impacts (908R11003). U.S. Environmental Protection Agency. <u>http://www.epa.gov/region8/mining/</u><u>ReportOnRareEarthElements.pdf</u> - Accessed 5 May 2014.

SANTAWAMAITRE T. (2012) An Evaluation of the Level of Naturally Occurring Radioactive Materials in Soil samples along the Chao Phraya River Basin, PhD Thesis, University of Surrey.

WNA - World Nuclear Association (2014) Radiation and Nuclear Energy. <u>http://www.world-nuclear.org/info/inf30.</u> <u>html</u> - Accessed 14th November 2014.

UNSCEAR - United Nations Scientific Committee on the Effects of Atomic Radiation (2000) Sources effects and risks of ionizing radiation, United Nations Report to the General Assembly, With Annexes, New York, USA.

WINDE F., WADE P., VAN DER WALT I.J. (2004) Gold tailings as a source of waterborne uranium contamination of streams — the Koekemoerspruit (Klerksdorp goldfield, South Africa) as a case study. Part I: Uranium migration along the aqueous pathway. Water SA, 30:219–226. Doi: <u>10.4314/wsa.</u> <u>v30i2.5067</u>

WHO - World Health Organisation (2008) Guidelines for drinking water quality, 3rd Edition, Incorporating 1st and 2nd addenda, Vol. 1 Recommendations, World Health Organisation, Geneva.