

Article

Evaluation of Radionuclide Levels in Drinking Water from Communities near Active and Abandoned Gold Mines and Tailings in the West Rand Region, Gauteng, South Africa

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Abstract: The history of gold mining in the Witwatersrand Basin has led to exponential growth in the economy, residential development, and the abundance of radionuclides in the environment, including the water system. This study aimed to evaluate the radionuclide levels in drinking water (municipal water and groundwater) and the health risks associated with the ingestion of the water in the communities of the West Rand region of Gauteng Province. The activity concentrations of uranium, radium, and thorium radioisotopes were established through alpha spectrometry and the activities were subsequently used to assess the health impacts. The results indicated that the groundwaters contain elevated activities of most radionuclides owing to prolonged periods of water–rock interactions. Similarly, the highest annual effective doses were recorded in groundwaters with a range of 0.0237–0.3106 mSv/yr, with most samples exceeding the WHO- and EU-prescribed limits of 0.1 mSv/yr. Cancer morbidity and mortality risks were higher in females than in males due to the higher life expectancy of females. Nonetheless, all morbidity and mortality risks were well below the USEPA radiological risk limit of 0.001. Based on the findings of this study, continuous monitoring is paramount to ensure that the activities remain below recommended regulatory limits.

Keywords: activity concentration; annual effective dose; cancer risk; gold mining; West Rand; South Africa



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1. Introduction

The Witwatersrand Basin has a long history of gold mining dating back to the first discovery of gold in 1886 [1]. This subsequently led to the development of many cities and towns, including Johannesburg, Krugersdorp, Randfontein, Roodepoort, Carletonville, Brakpan, and Germiston. Furthermore, the rapid residential development transformed the Witwatersrand Basin into the most densely populated region in South Africa [1]. This has carried on to date, with more residential areas being developed with the continued mining and processing of gold throughout the basin. However, the mining and processing of gold in the Witwatersrand Basin has had negative impacts on the environment, the most notable being acid mine drainage (AMD) [2]. Moreover, gold mining has resulted in an increase in radionuclides released into the environment, largely through tailings dams that are ubiquitous in the Witwatersrand Basin. These commonly contain elevated concentrations of radionuclides, such as ²³⁸U, ²³²Th, ²²⁶Ra and ⁴⁰K and toxic metals [3,4]. The inevitable interaction between water and tailings continues to produce AMD even in the absence of any active mining, which may ultimately contaminate both surface water and groundwater resources [5]. Advances in effective mining practices still fail to prevent the release of toxic metals and radionuclides into the water system, resulting in adverse impacts on the environment and human health in the case of drinking water [4]. Radionuclides are introduced into the human body through ingestion or inhalation and may have adverse

health effects, such as cancer, if high concentrations are accumulated [4–6]. Communities located proximally to mining activities, such as those of the West Rand, are especially at a higher risk of radiological hazards.

Several studies have documented the adverse impacts of mining on the environment and water alike at international and local scales with an emphasis on AMD [2,7–9]. In consideration of the exacerbating water pollution associated with gold mining, the focus of most research in the basin has shifted to evaluating the pollution of freshwater resources and the associated health impacts on humans. The chemical toxicity of surface-based drinking water sources was impacted by natural uranium pollutants around a gold mine in Roodepoort [5]. The study reported high ^{238}U activity concentrations and maximum values associated with an annual effective dose of 2.1 mSv/yr, which are well above the prescribed limit of 0.1 mSv/yr. Similarly, another study in the same area focused on gamma-emitting radionuclides found in soils and drinking water [10]. The annual effective dose related to the ingestion of raw water was found to be exceptionally higher than the South African National Nuclear Regulator (NNR) and World Health Organisation (WHO) limits of 0.25 and 0.10 mSv/yr, respectively [10,11]. The radiological health risk of drinking water and soil dust from the Gauteng and North West Provinces has also been assessed [4]. The specific activity of gross alpha and beta in purified water was lower than the WHO limit. The study concluded that drinking water is radiologically and toxicologically safe for human consumption. Nonetheless, little to no research has been dedicated to assessing the radionuclide levels and radiological health risks in drinking water obtained from groundwater and municipal supplies in the West Rand areas. Although the municipal drinking water was investigated, the water was collected in the North West Province, whilst this study was limited to the Gauteng Province [4]. In light of the well-documented pollution and radiological risks associated with drinking water from gold mining areas, it is worth investigating the radionuclide concentrations and associated health effects from the ingestion of drinking water to ensure the protection of the public against radiological harm. Although the vast majority of communities receive water from Rand Water, a government-owned water board, groundwater is used to augment the water supply shortfall, which is far surpassed by water demand.

This paper focused on the radionuclide levels in drinking water and the health risk associated with the ingestion of the water in communities of the West Rand region. The results can be used by regulatory bodies to enforce compliance and contribute to the development of new policies surrounding radiological health risks associated with drinking water in South Africa.

Study Site

This study was undertaken in residential communities proximal to active and abandoned mining activities in the West Rand region of the Gauteng Province, which has an aerial extent of about 4087 km² (Figure 1). This includes numerous historical mining towns such as Krugersdorp, Fochville, Carletonville, Wedela, Kagiso, Westonaria, and Randfontein (Figure 1). Topographically, the area is characterized by a variable profile of elongated quartzite ridges of the Witwatersrand and Transvaal Supergroups that form high grounds, while shale and dolomite form low-lying plain areas [9]. The climate is marked by warm summers with maximum temperatures of 34 °C, and cold winters with temperatures falling to −1 °C. The average annual rainfall received in the study area ranges between 260 mm to 1078 mm, with most rainfall experienced during the summer months. The study area is inundated with numerous land-use activities since Johannesburg is regarded as the economic hub of Africa. However, residential development, agriculture (crop and stock farming), and gold and uranium mining are the predominant land use activities. Both active and abandoned mines, including the associated tailings dams, are scattered throughout the study area, particularly in areas where the gold-bearing Central Rand Group of the Witwatersrand Supergroup dominates. Interestingly, the residential developments mostly occur adjacent to the mines and are the most densely populated [12].

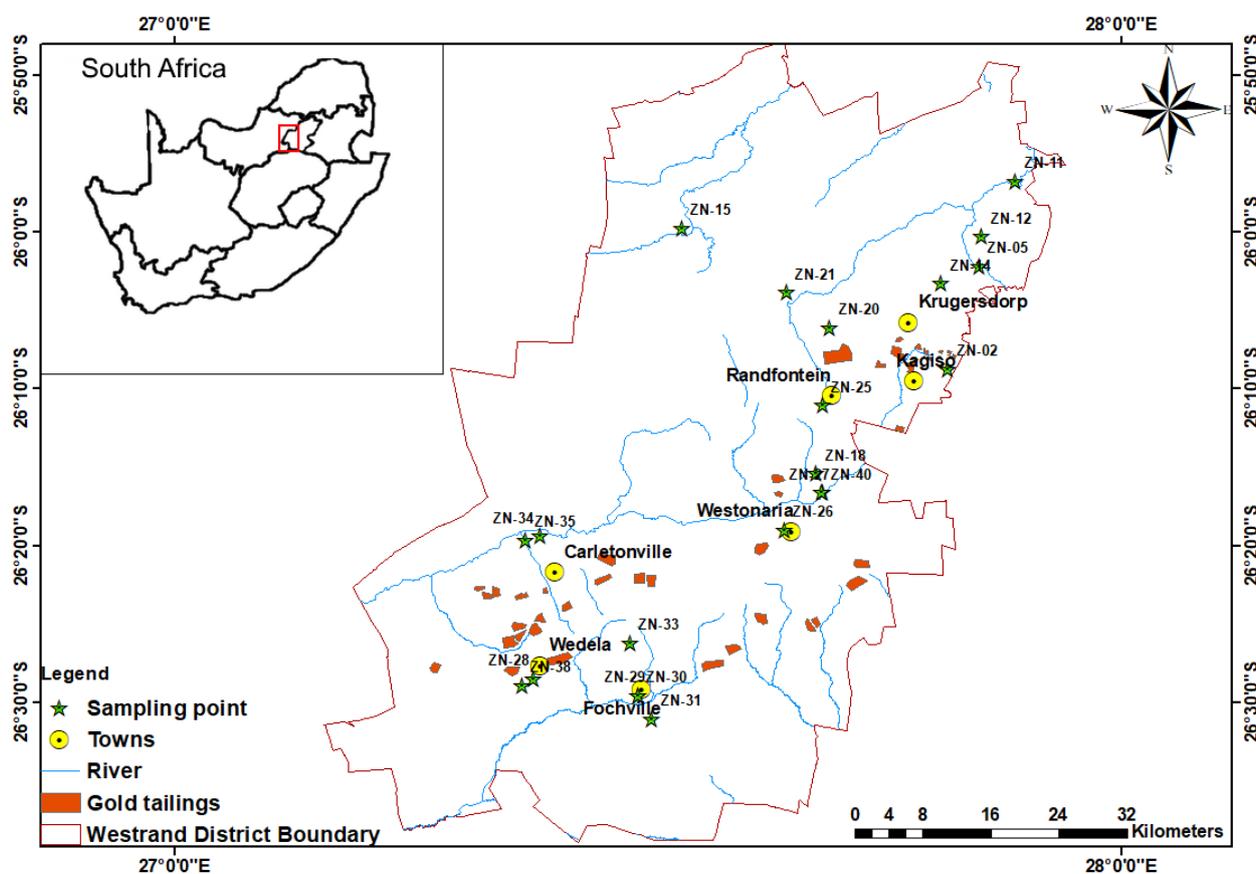


Figure 1. Locality map of the West Rand with sampling points and gold tailing dams.

The study area is characterised by complex geology composed of basement crystalline rocks, meta-sedimentary and meta-volcanic rocks (Figure 2). The basement geology represents the oldest rocks in the area and is composed of Archean (3.34 Ga) greenstone remnants that have a characteristic circular outline commonly known as the Johannesburg dome [13,14]. Furthermore, the dome is intruded by granodiorite, granite, tonalite, and migmatites. The majority of the granitoids in the dome are I-type magmatic rocks due to the high mafic mineral content (pyroxenes, olivine and minor feldspars) [14]. The basement geology is unconformably overlain by the Witwatersrand Supergroup, which comprises a thick succession of argillaceous and arenaceous meta-sedimentary rocks that are divided into the lower West Rand Group and upper Central Rand Group [15,16]. The West Rand Group is made up of quartzites and shales whilst the Central Rand Group is composed of conglomerates and quartzites [15,16]. Geochemically, the West Rand Group shales are predominantly iron-rich, with eight magnetite-bearing iron-formations. In addition, the quartzites are siliceous with a high content of feldspars and very limited pyrite mineralisation [16]. Conversely, the Central Rand quartzites are feldspar-poor. The Central Rand rocks are highly mineralised with gold, pyrite, and uraninite, and constitute the world-class gold-bearing reefs of the Witwatersrand Basin [15,16]. The mineralisation of the Witwatersrand Basin is attributed to the erosion of provenance areas (hydrothermally-altered granites and greenstones) and footwall sequences, and also the hydrothermal deposition of gold, pyrite, galena, and chalcopyrite [15,16]. The Transvaal Supergroup of the Proterozoic era overlies the Witwatersrand Supergroup and comprises of volcanic (basaltic and andesitic extrusions) and sedimentary rocks (dolomites, banded iron formations, diamictite, conglomerate, and quartzitic sandstones) [17,18]. The Transvaal Supergroup is geochemically diverse, containing silicates, carbonates (dolomite and calcium carbonate), iron, sporadic gold, pyrite, base metals (lead, zinc, fluorite), manganese, asbestos and andalusite [16–18].

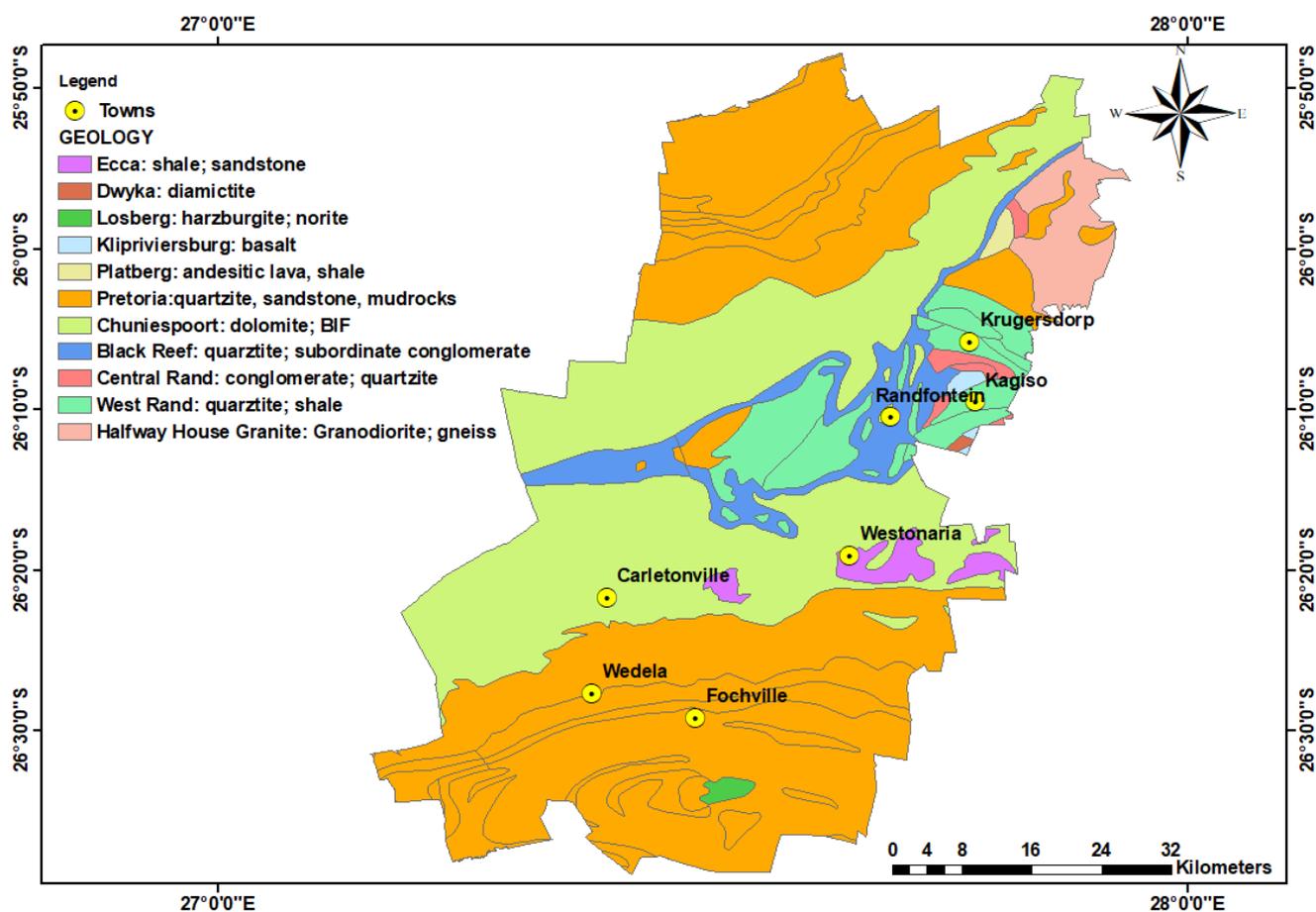


Figure 2. Geological map of the study area.

Hydrogeologically, the area is dominated by hard rocks that are characterized by low groundwater productivity due to their low porosity. However, ancient geological activities and processes (sedimentation, metamorphism, intrusion, brittle and ductile tectonics, weathering and erosion) have resulted in the generation of numerous geological and hydrogeological structures such as faults, fractures, joints, and weathering zones and these are crucial in controlling the hydrogeological characteristics of the local geology by regulating groundwater circulations and recharge [9,19]. As such, four types of aquifers have been identified [9,19,20].

- An intergranular aquifer in areas covered by alluvium and colluvium;
- Fractured aquifers in the Witwatersrand Supergroup that are associated with joints, fractures, and fissures;
- The karstic aquifer in the Malmani dolomites;
- The intergranular and fractured aquifers in the crystalline rocks.

Furthermore, the hydrostructures found within the geology promote groundwater recharge within these aquifers, thus resulting in highly productive aquifers that feed various springs linked to faults, formation contacts and dykes [9,21]. Interestingly, thermal water influx along fractures in the deep gold mines within the Witwatersrand basin and groundwater recharge by meteoric waters through faults and dykes have been noted thus highlighting the role of geological and hydrostructures in facilitating groundwater recharge [19].

In light of the densely populated state of the residential communities, the municipally supplied water is the main source of water, which is sourced from the Vaal Dam, located outside of the study area. However, extensive groundwater use also exists, especially in the areas located on the outskirts of the major towns and in farms. Therefore, it is important

to assess the radionuclide content of drinking water in these communities to ensure the protection of all life against radioactive harm linked to gold and uranium mining.

2. Materials and Methods

2.1. Water Sampling

Given the wide areal extent of the research site coupled with the abundance of residential communities and mining activities, a sampling strategy was developed to optimize sample numbers. Firstly, desktop studies were conducted to identify mining communities and population density to ensure adequate representation of each community in the sample collection. A total of 22 drinking water samples (11 boreholes, 1 spring and 10 municipal water samples) were retrieved from various water sources using 1 L high-density polyethylene (HDPE) bottles. As part of the sampling procedure, the polyethylene bottles and caps were rinsed twice with the sample water prior to collecting. Nitric acid was subsequently added to the samples to prevent the precipitation of metals out of the water and this is known as sealing. All samples were collected in duplicates for quality control following an identical sampling procedure. In the case of groundwater samples, boreholes were purged for 5 min before the sample was collected to allow older water present in the pipes to flow out thus ensuring groundwater directly from the aquifer is sampled.

Radionuclide activity concentrations were determined through alpha spectrometry and liquid scintillation at the National Energy Corporation of South Africa (NECSA) laboratory. The activity concentrations of uranium, thorium and radium radioisotopes, and gross alpha activity were measured through alpha spectrometry using a 7200-model alpha analyst spectrometer (Canberra Industries, Meriden, CT, USA), which is fitted with 12 Passivated Implanted Planar Silicon (PIPS) detectors. The PIPS detectors have an active surface area of 450 mm² and warranted alpha resolution of 18 keV that collectively provide excellent sensitivity, energy resolution and low detection limits. The Apex Alpha software was used for the acquisition, analysis, and storage of alpha spectra. A stainless-steel alpha standard source containing ²³⁸U, ²³⁴U, ²³⁹Pu and ²⁴¹Am was used to establish energy calibration (channel vs energy plot) and counting efficiencies for the detectors. All methods are accredited by the South African National Accreditation System (SANAS) and International Organization for Standardization (ISO 17025: 2017). The machine's minimum detection amount (MDA) were calculated according to Equation (1)

$$MDA \text{ (mBq/L)} = \frac{2.71 + 3.29 * \sqrt{B * (1 + t_s/t_b)}}{t_s * \epsilon * V * R} \quad (1)$$

where ϵ = counting efficiency of the α -spectrometer used, t_s = counting period of sample (seconds), B = background counts, normalised to counting period, t_b = counting period (nominally 24 h, or 86,400 s), V = volume of sample taken for analysis (litre), R = yield measured for uranium tracer in the blank.

Based on Equation (1), the MDA values for uranium, thorium and radium radioisotopes are 0.2, 2.5 and 0.8 mBq/L, respectively. Furthermore, the detection limit for gross alpha and beta activities are 10 and 30 mBq/L, respectively.

Additionally, six groundwater samples were collected from springs and household boreholes for isotopic analysis of tritium (³H). Environmental isotopes of water are regarded as a robust support tool for signifying different processes and characteristics of groundwater such as the residence time of groundwater in the subsurface (how long the groundwater has been in the subsurface). The tritium samples were analysed at iThemba Environmental Isotope Laboratory in Johannesburg at a ± 0.2 T.U detection limit. Results of the tritium analysis were reported in tritium units (T.U), and subsequently converted to Bq/L using a conversion factor of 1 T.U = 0.0118 Bq/L [22]. Furthermore, tritium-based groundwater residence times were determined according to Equation (2) [23]:

$$t = -17.93 \ln\left(\frac{at^3H}{a0^3H}\right) \quad (2)$$

where t is the mean residence time in years, a_t^3H is the residual activity of tritium remaining after decay overtime (tritium in the sample) and a_0^3H is the initial tritium activity.

2.2. Health Risk Assessments

All health risks were assessed based on ^{238}U activity due to its higher abundance in the environment (including water) and toxicity to humans [24]. The World Health Organization (WHO) and United States Environmental Protection Agency (USEPA) identified uranium as a nephrotoxin and thus its effects on human health should be investigated [6].

2.2.1. Annual Effective Dose

A radiological assessment of the risk associated with the ingestion of the water (drinking and cooking) in the study area was achieved through the estimation of the effective dose. Effective dose is defined as the sum of the weighted equivalent doses in all tissues and organs of the body [25]. This is an integral part of radiation protection studies since water-sourced radiation exposure begins with the ingestion of water containing radionuclides, which ultimately accumulate in different parts of the human body such as the liver, soft tissue, skeleton and kidneys [5,26]. Furthermore, the effective dose estimations play a crucial role in regulatory assessments by demonstrating compliance with the outlined dose limits [27]. As prescribed by the [28], the annual effective dose associated with the ingestion of drinking water was calculated according to Equation (3):

$$\text{AED} = \text{AC} \times \text{DC} \times \text{AWC} \times 1000 \quad (3)$$

where, AED is the annual effective dose (mSv/yr), AC is the activity concentration of ^{238}U (Bq/L), DC is the dose coefficient, AWC is the annual water consumption (L/yr).

In this study, a dose coefficient of 4.5×10^{-8} [28] and AWC value of 111,325 L/yr ($305 \text{ L/capita/day} \times 365 \text{ days}$) for Gauteng residents [29] were used. The AWC value represents the total volume of water consumed by the average individual annually, and includes activities such as cooking and drinking.

2.2.2. Cancer Risk Assessment

The most prevalent effect of radiation energy on living organisms is cancer. The risks of cancer associated with water ingestion was calculated through the estimation of the lifetime cancer risk (LCR). LCR is the product of the risk coefficient (R_c) and per capita activity intake (PCAI) as outlined in Equation (4) [5]. LCR is estimated for a specific radionuclide and ^{238}U -linked LCR was calculated in this study.

$$\text{LCR} = R_c \times \text{PCAI} \quad (4)$$

The LCR was estimated for both males and females, since the two genders have different life expectancies. The applied lifetime expectancies were 59.3 and 64.6 years in males and females, respectively [30]. PCAI is a product of the average annual water intake (111,325 L), lifetime expectancy, and the activity concentration of the radionuclide of interest (^{238}U). The morbidity and mortality risks for cancer were investigated using uranium risk coefficients (R_c) of $1.71 \times 10^{-9}/\text{Bq}$ and $1.13 \times 10^{-9}/\text{Bq}$, respectively [6].

3. Results

3.1. NORMs in Water

The activities of all measured NORMs and gross activities are presented in Table 1. ^{238}U activity exhibits a narrow range in the sampled water, from 0.00473 to 0.062 Bq/L and an average activity of 0.0285 Bq/L. The highest activities were obtained in groundwater samples ZN-11 (Lanseria) and ZN-14 (Krugersdorp). Elevated ^{238}U activities were also recorded in groundwater samples ZN-29 and 31 from Wedela and Fochville, respectively (Figure 3). High activities of ^{235}U and ^{234}U activities were also noted in groundwater samples ZN-11, 14 and 3 (Table 1). Nonetheless, all sampled water contained ^{238}U activities

that are well below the WHO and European Union (EU) prescribed limits of 10 and 3 Bq/L, respectively, in drinking water albeit the relatively elevated activities [31,32]. A similar ^{238}U average activity of 0.015 in drinking water from dug wells in Finland was also reported [26]. Based on the spatial distribution map of ^{238}U presented in Figure 3, elevated activities were mostly restricted to the northernmost end of the study area (in and around Krugersdorp) with the exception of Fochville. Furthermore, ^{238}U activity progressively increased away from the centre of the study area.

Table 1. Alpha spectrometry-based activity concentrations of major radionuclides in drinking water from the West Rand and the associated recommended guidelines.

Sample Name	^{238}U (Bq/L)	^{235}U (Bq/L)	^{234}U (Bq/L)	^{232}Th (Bq/L)	^{230}Th (Bq/L)	^{228}Th (Bq/L)	^{227}Th (Bq/L)	^{226}Ra (Bq/L)	^{224}Ra (Bq/L)	^{223}Ra (Bq/L)	Gross Alpha (Bq/L)	Gross Beta (Bq/L)
ZN-02	0.0254	0.00117	0.0595	<MDA	0.031	0.00473	0.00507	−0.0021	0.011	0.0012	−0.016	0.37
ZN-05	0.0117	0.000537	0.0192	0.00454	0.0344	0.00671	0.00899	0.0019	0.004	0.0016	0.053	0.283
ZN-11 *	0.062	0.00285	0.127	0.0133	0.0479	0.0223	0.00228	0.0195	0.0136	0.002	0.341	0.502
ZN-12	0.0446	0.00205	0.112	0.00612	0.0291	0.00868	0.00673	0.0012	0.0025	0.0023	0.0081	0.15
ZN-14 *	0.0593	0.00273	0.21	0.00909	0.0726	0.0107	0.0212	0.0028	−0.0057	−0.0013	0.141	0.031
ZN-15	0.0288	0.00132	0.0459	0.00605	0.0468	0.012	0.0347	−0.0054	−0.0038	0.013	0.015	0.064
ZN-18	0.00873	0.000402	0.0245	0.0125	0.035	0.0316	0.0056	−0.0055	0.012	0.0015	0.026	−0.04
ZN-20	0.0133	0.000612	0.031	0.0036	0.044	0.0226	0.0062	−0.00097	−0.006	−0.0084	0.028	−0.47
ZN-21	0.00834	0.000384	0.104	0.0216	0.0702	0.0136	0.0097	0.0137	−0.02	−0.002	0.147	0.086
ZN-25	0.0261	0.0012	0.0473	0.00671	0.0407	0.0113	0.00777	−0.0064	0.025	0.0057	0.037	2.38
ZN-26	0.00996	0.000459	0.0316	0.00774	0.033	0.033	0.00263	−0.0044	−0.019	−0.0026	0.0092	−0.12
ZN-27 *	0.0199	0.000914	0.0199	0.00271	0.0439	0.00981	0.0102	0.0027	−0.001	−0.0071	0.02	−0.96
ZN-28	0.0368	0.00169	0.042	0.0155	0.0327	0.0121	0.0082	−0.0049	−0.0025	0.0013	−0.012	0.606
ZN-29 *	0.0403	0.00185	0.0914	0.00848	0.0795	0.0108	0.0041	0.0058	−0.011	0.0029	0.056	−1.1
ZN-30	0.0323	0.00149	0.0522	0.0235	0.028	0.024	0.0382	0.003	−0.011	−0.0031	0.132	13.5
ZN-31 *	0.0431	0.00199	0.176	0.0217	0.034	0.0154	0.00744	0.0022	−0.034	−0.0059	0.215	0.604
ZN-33	0.00473	0.000218	0.0158	0.0159	0.0789	0.0183	0.0143	0.0107	−0.0055	−0.0098	0.149	0.394
ZN-34	0.03	0.00138	0.0627	0.0089	0.018	0.0203	0.0505	0.00575	<MDA	−0.0015	0.068	0.396
ZN-35 *	0.014	0.000646	0.0281	0.00883	0.0446	0.0173	0.0377	0.0011	0.00884	0.00784	0.321	0.673
ZN-36	0.0135	0.000623	0.0153	0.00868	0.0548	0.0159	0.016	0.0022	0.0045	0.0018	0.13	0.34
ZN-38	0.0384	0.00177	0.0498	0.0065	0.0397	0.031	0.0058	0.0067	0.006	−0.00071	0.198	0.309
ZN-40 *	0.0549	0.00253	0.0723	0.0196	0.0864	0.0157	0.0101	0.00097	−0.0081	−0.0026	0.138	0.24
Minimum	0.00473	0.000218	0.0153	0.00271	0.018	0.00473	0.00228	−0.0064	−0.034	−0.0098	−0.016	−1.1
Maximum	0.062	0.00285	0.21	0.0235	0.0864	0.033	0.0505	0.0195	0.025	0.013	0.341	13.5
Mean	0.02846	0.00131	0.06534	0.01103	0.04660	0.01672	0.01425	0.00230	−0.00191	−0.00018	0.10020	0.8290

* represents groundwater.

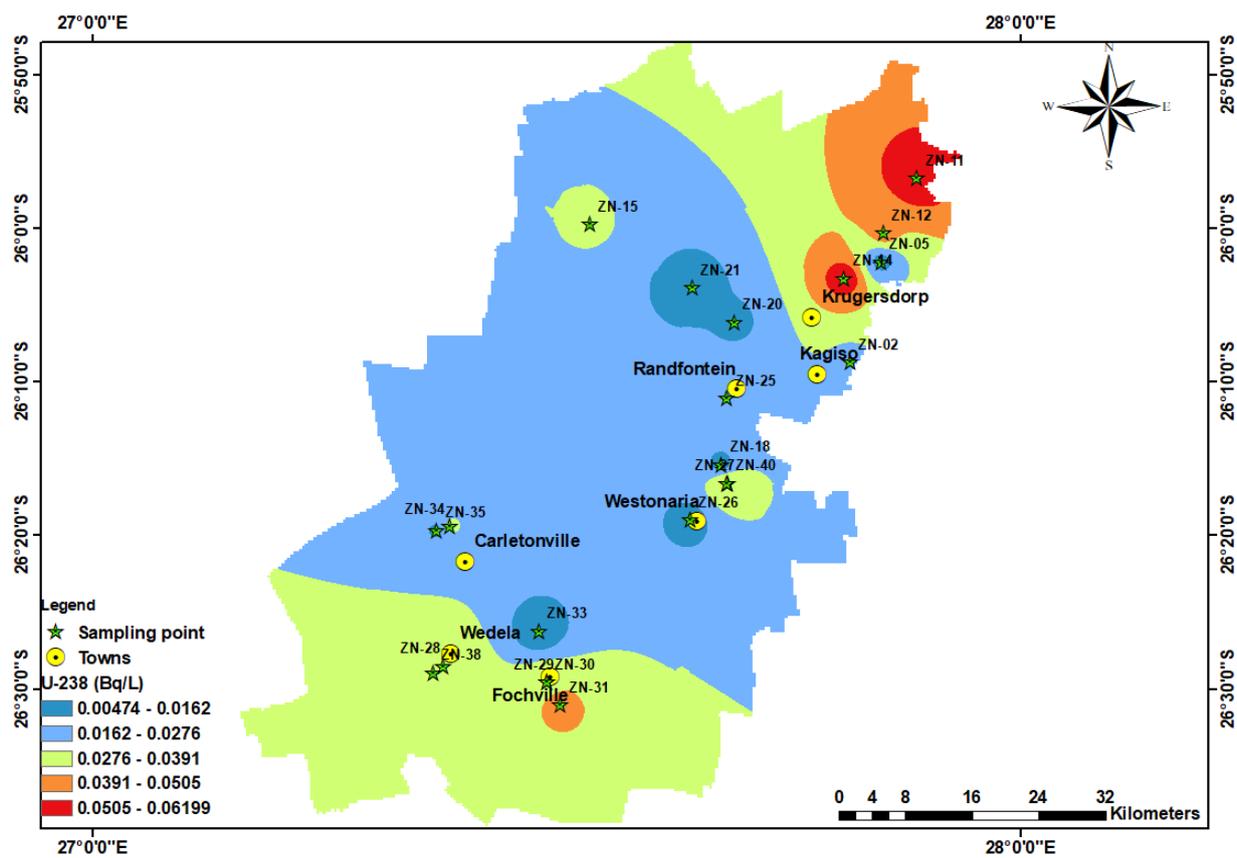


Figure 3. Spatial distribution map of ^{238}U in the West Rand region.

All thorium radioisotopes occur in very narrow ranges and have low activities (Table 1). Concerning ^{232}Th , it is largely insoluble in water, thus its occurrence and mobility in both surface water and groundwater is limited [33,34]. Furthermore, a limited occurrence of ^{232}Th in the water implies that the occurrence of most of its progenies such as ^{228}Th in the sampled water may be limited as noted in the sampled water (Table 1). Nevertheless, ^{232}Th ranged between 0.00271 and 0.0235 Bq/L, which is significantly lower than the DWAf [35] and WHO limits of 0.228 and 1 Bq/L in drinking water, respectively [11]. In contrast, ^{230}Th (a progeny of ^{238}U) activities were relatively higher in the sampled water, ranging between 0.018 and 0.0864 Bq/L. The highest activities were noted in groundwater samples ZN-14, 21, 29 and 33, as was the case with ^{238}U , and this is likely ascribed to the radioactive decay of the elevated ^{238}U (Table 1). The ^{226}Ra activities range between 0.00097 and 0.0195 Bq/L (Table 1). The negative values are due to the difference between the instrument's background activity and the sample's ^{226}Ra activity, which results in negative values that are essentially below the instrument's minimum detectable amount (MDA) for a ^{226}Ra . The highest activity was noted in groundwater sample ZN-11 from Lanseria, and this creates a cause for concern since radium is highly radiotoxic and carcinogenic and, therefore, its abundance in drinking water presents a health hazard to the public [6]. A comparison of the ^{226}Ra activities against prescribed international standards of USEPA, EU, and WHO revealed that all sampled drinking water contained ^{226}Ra activities that are well below the recommended limits of 0.185, 0.5 and 1 Bq/L, respectively. Although groundwater sample ZN-11 contained elevated activities of both ^{238}U and its progeny ^{226}Ra relative to other samples, the activities of both radionuclides were significantly different (Figure 4), since the two radionuclides occurred under different and contrasting hydrogeochemical conditions [33]. This relationship was further supported by the weak correlation ($R^2 = 0.0025$) between ^{238}U and ^{226}Ra , thus signifying the minimal influence of ^{238}U on the occurrence of ^{226}Ra in the water (Figure 4). ^{226}Ra activities from this study were

also benchmarked against reported activities in drinking water from around the world, and the results are presented in Table 2. The ^{226}Ra range obtained in this study largely coincides with ranges reported in different studies. The spatial distribution of ^{226}Ra is characterised by predominantly low activities, with sporadic elevated activities in three distinct areas (Figure 5). In the case of Lanseria (ZN-11), the elevated activity of ^{226}Ra is ascribed to the uranium and radium-rich the elevated activity of ^{226}Ra is ascribed to the uranium and radium-rich granodiorites and gneisses that collectively comprise the basement geology.

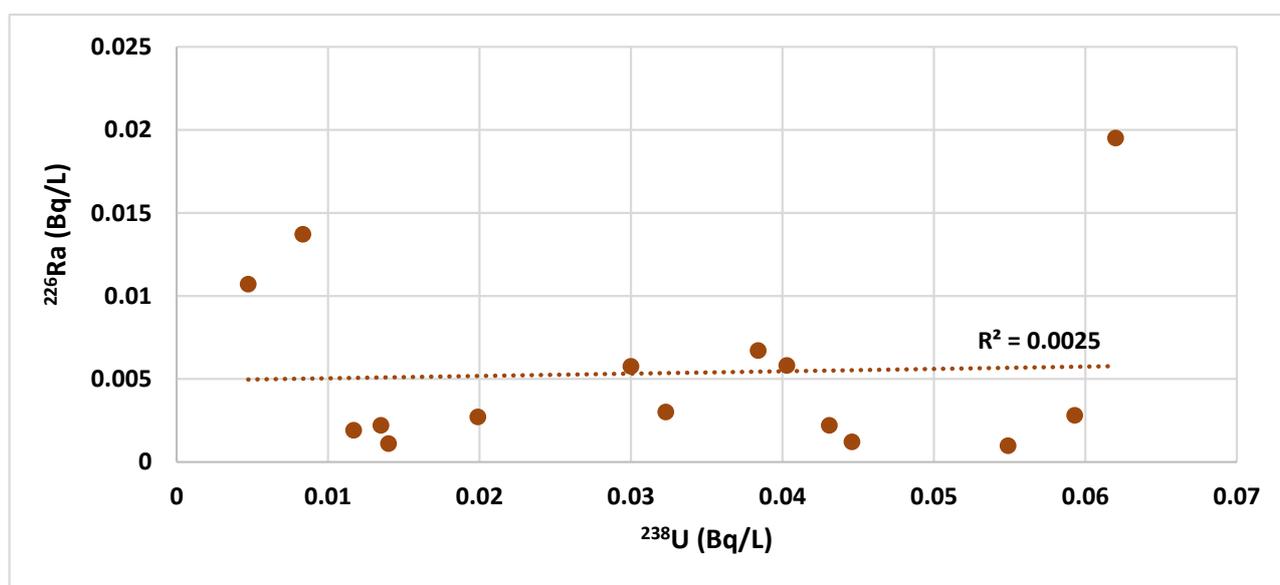


Figure 4. The ^{238}U activity concentration relative to ^{226}Ra (radiogenic product).

Table 2. ^{226}Ra activity concentrations in drinking water from around the world.

Country	^{226}Ra Activity Concentration (Bq/L)
South Africa (this study)	0.0019–0.0195
India	0.0035–0.206 [36]
Finland	0.001–1 [26]
Germany	0.00296–0.0222 [37]
United State of America	0.0185–0.644 [38]
China	0.0011–0.939 [39]
Brazil	0.002–0.492 [40]
Egypt	0.0792 [41]

All sampled water was in compliance with gross alpha regulatory limits of 0.555 and 0.5 Bq/L for domestically-used water as prescribed by EU and WHO, respectively, albeit the notable variation in activities (0.0081–0.341 Bq/L) [11]. Gross beta activity was below the WHO limit of 1 Bq/L in most samples with the exception of municipal water samples ZN-25 and 30 (Table 1) [11]. Both the gross alpha and beta activities were significantly higher than previously reported values from other studies, such as that of [4], which reported ranges of 0.0041–0.0053 Bq/L and 0.0083–0.0105 Bq/L for gross alphas and betas, respectively. Figure 5 shows the spatial distribution of gross alpha activity in the study site. Gross alpha activity has a similar spatial distribution to that of ^{238}U and ^{226}Ra , where the gross alpha activity decreases towards the centre of the study site. This pattern may be explained by the fact that both ^{238}U and ^{226}Ra are alpha emitters and therefore contribute to the total gross alpha activity.

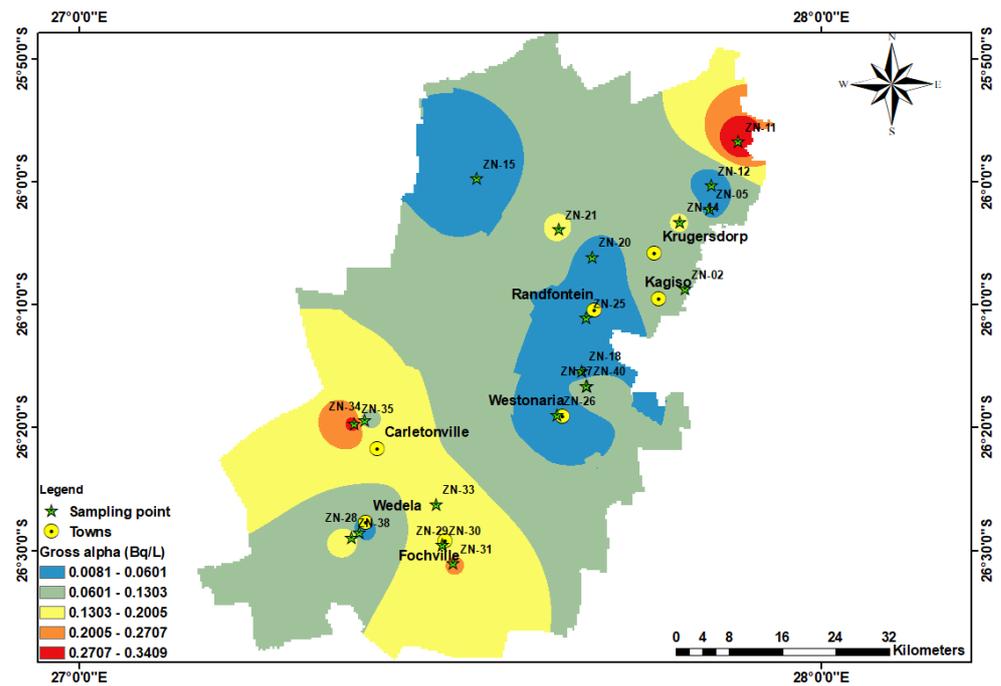


Figure 5. Spatial distribution map of gross alpha activity in the West Rand region.

3.2. Tritium

Tritium activity in the sampled groundwater ranged from 0.0094 Bq/L in sample ZN-31 to 0.02832 Bq/L in spring sample ZN-27. The high tritium activity in the spring water translates to a shorter residence time of the groundwater below the surface and more recent groundwater recharge as indicated in Figure 6. On the contrary, older groundwater is characterized by lower tritium concentration since the tritium would have undergone radioactive decay in the aquifer. Figure 7 highlights this relationship, where higher tritium activities are encountered in groundwater with shorter residence times. Given that the longer residence time of groundwater implies older recharge age of the groundwater, it is evident from Figure 7 that elevated NORM activities are largely due to water–rock interactions in the aquifers, as opposed to contributions from polluted water that may recharge the aquifers. In light of the drinking water assessment, all samples contained tritium activities that were exceptionally below the 100 Bq/L limit prescribed by the EU and WHO [11].

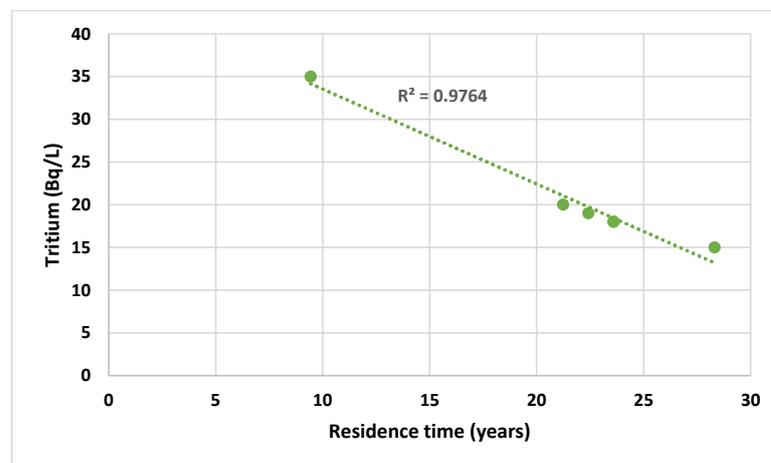


Figure 6. Activity concentration of tritium (³H) in groundwaters with variable residence time from the West Rand area (n = 6).

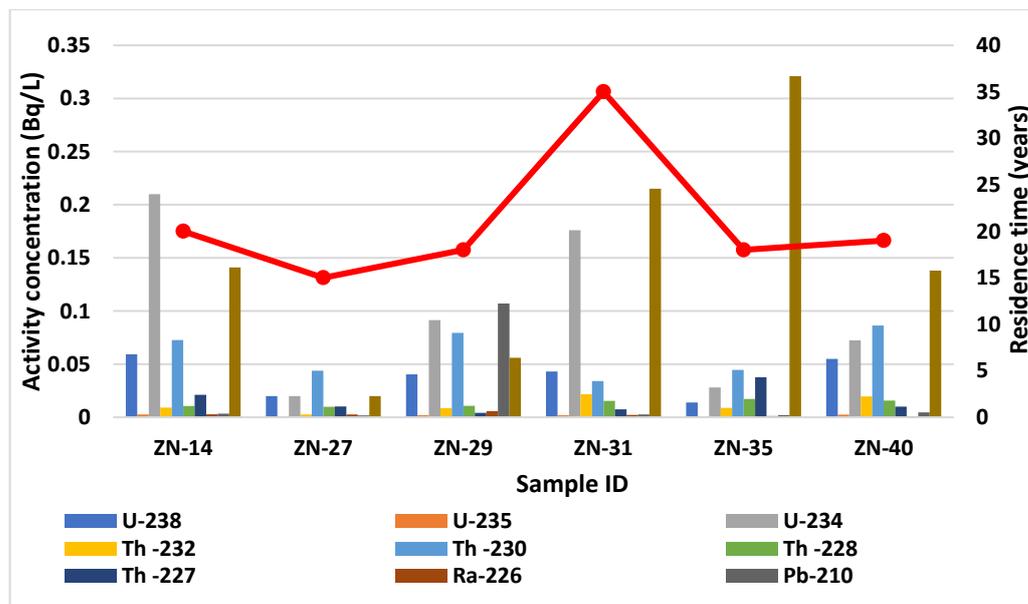


Figure 7. Radionuclide activities in groundwater and the associated residence time of groundwater.

3.3. Annual Effective Dose

The effective dose of each sample was estimated so as to quantify the risk associated with the ingestion of the water in the study area. The results of the annual effective dose estimates are presented in Figure 8. It is evident from the results that the annual effective dose in the sampled drinking water is highly variable, with ranges of 0.0237–0.3106 mSv/yr and 0.0499–0.2234 mSv/yr in groundwater and municipal water, respectively (Figure 8). Over 50% of the sampled water contained AED that exceeded the set WHO and EU limits of 0.1 mSv/yr in drinking water. In contrast, all other samples, including one groundwater sample, were well below the set limit, thus indicating good compliance with regulatory limits from the water supply body.

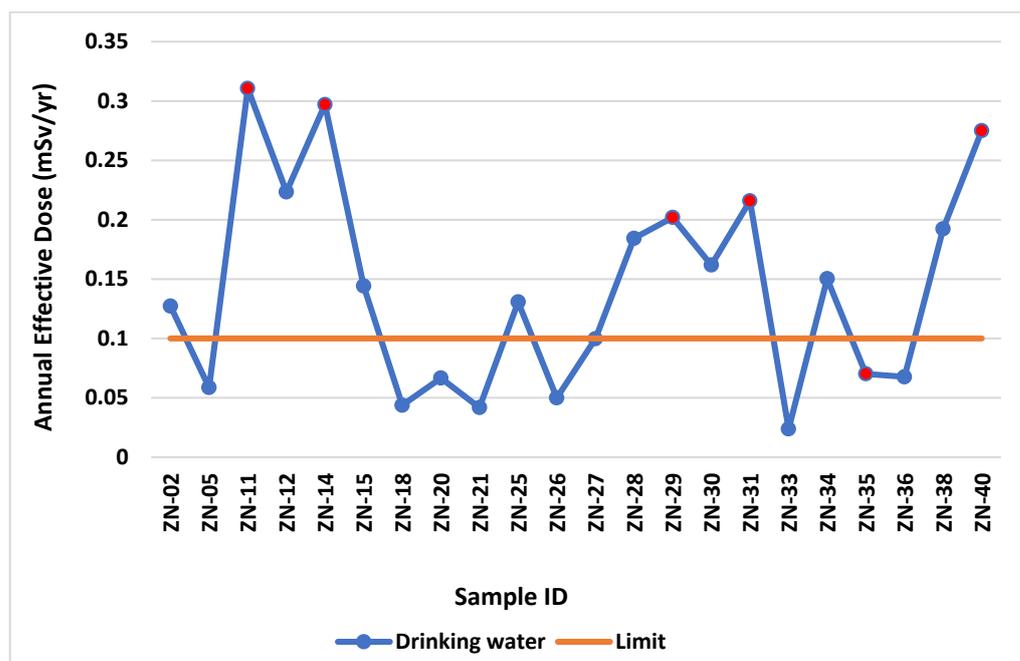


Figure 8. ²³⁸U-related Annual Effective Dose (AED) and associated regulatory limit for drinking water. Red points represent groundwater samples.

3.4. Lifetime Cancer Risks

Figure 9 presents the cancer morbidity and mortality risks associated with the ingestion of uranium through water. The morbidity cancer risk ranges are 5.340×10^{-6} – 6.999×10^{-4} and 5.817×10^{-5} – 7.625×10^{-4} in males and females, respectively. Conversely, the mortality risks are 3.528×10^{-5} – 4.625×10^{-4} and 3.844×10^{-5} – 5.038×10^{-4} in males and females, respectively. Cancer morbidity and mortality risks were higher in females than males in all sampled drinking water. Furthermore, the mortality risk was lower than the morbidity risk in both genders, which implies that the risk of developing cancer far exceeded that of dying from cancer. This phenomenon could be explained by the ability to treat (some) cancers, thus lowering the risk of cancer-related deaths, albeit the higher morbidity risks. All cancer risk values fell below the recommended radiological risk limit of 0.001 (10^{-3}) [6].

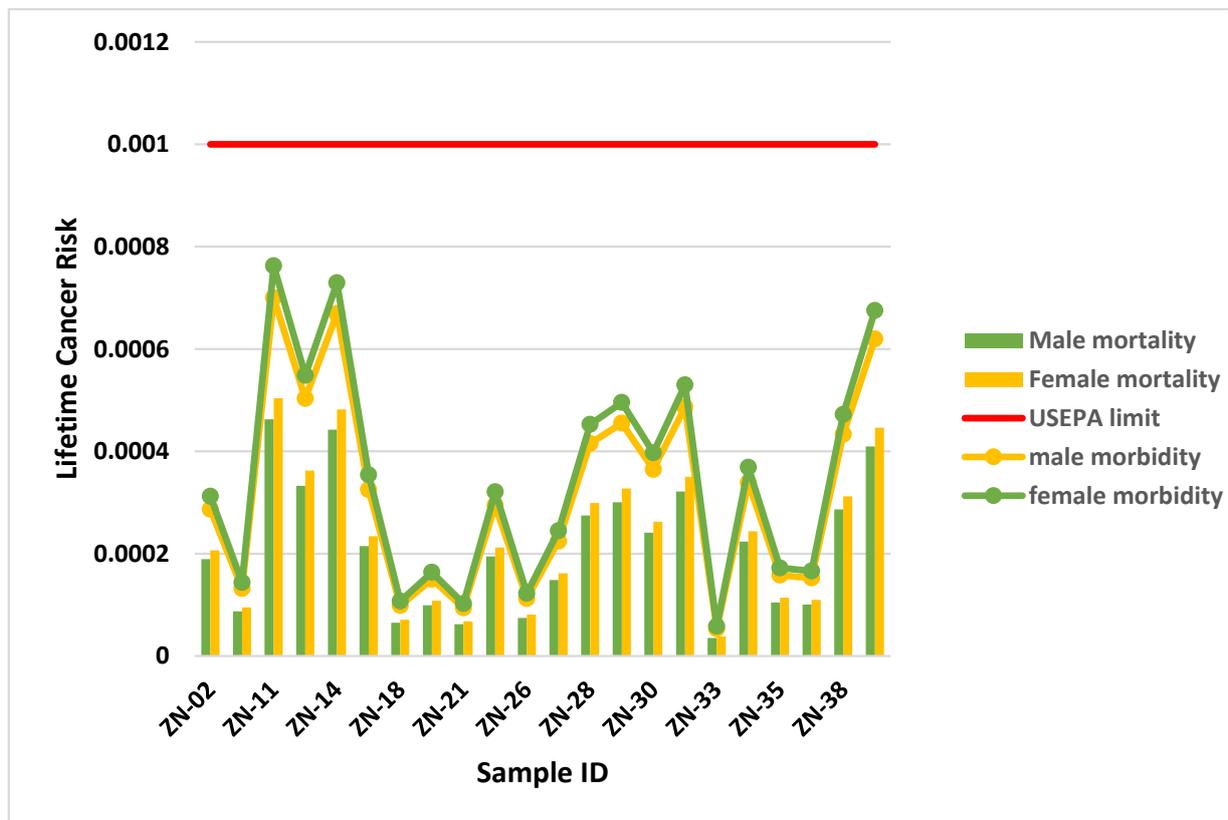


Figure 9. Mortality and morbidity risks of cancer (^{238}U related) from the ingestion of drinking water from the West Rand.

4. Discussion

Based on the findings of this study, contributions from the local geology through-water-rock interaction (mineral dissolution) are responsible for the elevated uranium activities in the sampled groundwater. In the case of groundwater samples ZN-11 and ZN-14, the Archean gneisses and granites that are known to contain elevated concentrations of uranium are responsible for the elevated activities. Similarly, elevated ^{238}U activities in groundwater samples ZN-29 and ZN-31 are attributed to the dissolution of minerals contained in the Pretoria Group (quartzites and mudrocks), which have been affected by the hydrothermal remobilisation of gold and its accessories such as uranium [18]. This is also evident from the spatial distribution map of ^{238}U , which indicates elevated activities in areas characterised by the aforementioned lithologies, therefore signifying the control of the local geology on uranium concentration in the groundwater. Likewise, there is an abundance of ^{234}U and ^{235}U in the same groundwater samples, which is expected, since the two uranium radioisotopes are progenies of the radioactive decay of ^{238}U . However, an abundance in

^{238}U does not imply an abundance in all its progenies, as is the case with ^{226}Ra (Figure 4). A similar observation was made in Finnish groundwater in which the occurrence of ^{226}Ra was independent from the occurrence of uranium [26]. The very limited occurrence of ^{226}Ra in the drinking water is a common phenomenon across the world, including in areas where the local geology favours the abundance of radium in water such as our study area [42]. A negative relationship exists between tritium activity concentrations and groundwater residence time, such that an increase in residence time leads to a decrease in tritium activity in the groundwater. This is because tritium undergoes radioactive decay, thus resulting in low activities over time [42]. Furthermore, groundwaters with higher residence times were found to commonly contain elevated activities of NORMs due to longer periods of water–rock interactions that result in higher NORM activities in the older water. The evaluation of tritium against radionuclide activities in water revealed that water–rock interactions are responsible for the elevated activities in the groundwater as opposed to contribution from polluted recharging water. The tritium-depleted groundwater sample ZN-31 is a great demonstration of this, as it contained elevated radionuclide activities albeit the highest residence time (i.e., old recharge age) in the subsurface. Moreover, this sample was located in an area inundated with tailings and an active gold mine (Figure 1). As such, the groundwater chemistry is mainly influenced by water–rock interactions as opposed to pollution emanating from local surface activities such as gold mining. Furthermore, it is apparent from the findings of this study that gold mining activities (active mining and tailing dams) have a limited influence on radionuclide levels in drinking water from the West Rand (Table 1). This is especially in the case of municipal supplied water, which undergoes treatment prior to distribution to the communities. In the case of groundwater, the radionuclides levels are exceptionally lower than values reported in other studies, regardless of most samples collected in locations that are proximal to tailing dams as shown in Figure 1 [5,10].

More than half of the sampled drinking water recorded AEDs higher than the 0.1 mSv/yr limit set by the EU and WHO owing to the elevated ^{238}U activities, which are in turn attributed to the leaching of uranium from the local geology in the case of groundwater. The AED estimates obtained in this study coincide with results from previous studies such as [10] that reported AED to vary between 0 and 2 mSv/yr in drinking water from Gauteng and the North West. Similar values were also reported in the West Rand, with a range of 0.0632 and 2.1 mSv/yr [5] and in Finnish groundwaters with a maximum dose of 3 mSv/yr [26]. However, AED estimates from this study were significantly higher than values reported in groundwater from gold mining regions in India, with a dose range of 0.00251 and 0.00576 mSv/yr [36]. Females have elevated risks of both cancer morbidity and mortality relative to males due to the relatively higher life expectancy of females, which is a crucial factor in radiological cancer risk assessments. A similar value of 3.39×10^{-4} for cancer morbidity were reported in Ogun State, Nigeria [43]. On the other hand, the cancer risk values obtained in this study are far less than the risk values obtained by [5], with ranges of 1.51×10^2 – 5.03×10^3 and 9.86×10^1 – 3.28×10^3 for morbidity and mortality, respectively. The notable difference in risks between the two studies is due to the significant difference in ^{238}U activity in the sampled water, since ^{238}U activities from this study are significantly lower than the reported values [5].

5. Conclusions

The safety of drinking water from communities in the gold-mining West Rand region of Gauteng was successfully investigated in this study. Groundwater was found to mostly contain elevated activities of radionuclides relative to the treated water supplied to most homes, and this was linked to the fact that the groundwater undergoes little to no treatment prior to consumption. Likewise, higher annual effective dose (AED) and Lifetime Cancer Risk (LCR) estimates of ^{238}U were noted in groundwater due to the elevated activities of ^{238}U . LCR assessment revealed that females are at higher risks of both cancer morbidity and mortality as opposed to males. Furthermore, all cancer risks were found to be significantly

lower than the recommended radiological risk limit, as was the case with the estimated AED. A comparison of tritium residence time (age) estimates and radionuclide activity concentrations indicated that radionuclide concentrations in the groundwater are primarily influenced by water–rock interactions with a limited effect from the polluted recharging water associated with the ubiquitous tailing dams and gold mines. The findings of this research revealed that the activities and associated health risks fall below all prescribed limits and the water is therefore radiologically safe for human consumption. In addition, the treated municipal water adheres to both local and international standards of drinking water, thus signifying good compliance from the water supply body. In light of the results of this study, the authors recommend continuous assessment and monitoring of the radiological levels in drinking water to ensure continued compliance with regulatory limits and guidelines. This is especially important for the groundwater, since it is not treated before consumption, and has elevated risks of contamination relative to the treated municipal water.

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