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In Situ Determination of Radioactivity Levels and Radiological Doses in and around the Gold Mine Tailing Dams, Gauteng Province, South Africa

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Abstract: The mining and processing of naturally occurring radioactive materials (NORMs) could result in elevated levels of natural radionuclides in the environment. The gold mining in the goldfields of the Witwatersrand Basin of South Africa has resulted in numerous tailing dams that have high concentrations of NORM bearing residue. The aim of this study was to evaluate the radioactivity levels in tailing dams, soils and rocks, and the consequential radiological exposure to the public in the gold mining areas of Gauteng Province, South Africa. The activity concentrations of ^{238}U, ^{232}Th, and ^{40}K were assessed using a gamma ray spectrometer (RS-230), and the activity concentrations ranges in the mine tailings were 209.95–2578.68 Bq/kg, 19.49–108.00 Bq/kg, and 31.30–626.00 Bq/kg, respectively. The radionuclides show significant spatial variability in soils, with high activities recorded in soils located in close proximity to tailings although regionally, the soil radioactivity levels mainly depend on the chemistry of the underlying rocks. The estimated annual effective doses were higher than the recommended regulatory limit of 0.25 mSv/y in particular tailing dams and soil impacted by tailings. Therefore, to ensure the protection of people and the environment, further monitoring and regulatory control measures targeting these areas are required.

Keywords: activity concentration; gold mine tailings; in situ gamma ray spectrometry; radiological exposures; South Africa

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1. Introduction

Naturally occurring radioactive materials (NORM) constitute part of the Earth and include prehistoric elements within the Earth's crust, radioactive decay products, and radionuclides that are formed in the atmosphere by cosmic ray bombardment, which eventually ends up in the water supply system [1]. Natural radionuclides such as potassium−40 (^{40}K), uranium−238 (^{238}U), and thorium−232 (^{232}Th) and their associated daughter isotopes are found in various quantities in all environmental materials and contribute a major fraction of the external radiation dose exposure to humans [2]. The occurrence of these radionuclides in the environment is commonly associated with the geological conditions of the area [3]. Rocks such as granite, shale, and phosphate bearing minerals typically contain higher concentrations of naturally occurring radionuclides [4]. Moreover, human activities such as mining have the potential to elevate radioactivity levels in the Earth's crust by introducing underground mineralised ores with natural radionuclides to the Earth's surface. These are then chemically released into the environment through numerous processes such as oxidation, as in the case of uranium-bearing lithologies. Most commonly, the mining of gold, uranium, copper, tin, and niobium results in increased levels of uranium and thorium series radionuclides in the environment [5]. In South Africa, the historic mining and processing of gold and uranium produced a large volume of tailing dams, which commonly contain

elevated concentrations of natural radionuclides and toxic residue metals associated with gold [6]. The radioactive elements contained in the tailing dams could be mobilised into the environment through weathering, infiltration of water, and passage of air through the residue material and subsequently pose radiological risks to members of the public [5]. The pathways through which residents can be exposed to radiation associated with mining operations include external gamma radiation exposure and internal exposure through inhalation and ingestion [4,7,8].

In recent years, a few studies have been conducted, particularly in the Witwatersrand gold mining region, to assess the radiological hazards due to gamma radioactivity in mine tailings and soils impacted by tailing materials. In the goldfield of the lower central part of the Wonderfonteinspruit catchment area, the average activity concentrations for ^{238}U, ^{232}Th, and ^{40}K from the mine tailings were found to be 785.3 ± 13.7 Bq/kg, 43.9 ± 1.0 Bq/kg, and 427.0 ± 13.1 Bq/kg, respectively [9]. The levels were found to pose health risks to the local population. In the Tudor Shaft, which is impacted by gold mine tailings, the activity concentrations in soils were found to be 271.96 ± 3.59 Bq/kg, 47.65 ± 3.69 Bq/kg, and 87.17 ± 5.19 Bq/kg for ^{238}U, ^{232}Th, and ^{40}K, respectively [10]. These recorded data confirm that the annual effective doses and the lifetime cancer risks were considerably higher than the world's average values reported by UNSCEAR [4], aside from providing important indications about the abandoned gold mine tailings that contain high radioactivity levels with significant radiological concern. However, there is still not sufficient data to provide a comprehensive understanding of the current radioactivity levels and the extent of surface contamination in affected mining regions in Gauteng Province, South Africa. Therefore, there is a need to assess the radioactivity contributions from natural and anthropogenic activities at a regional scale to better evaluate the impact of the abandoned mine tailings on the environmental radioactivity levels and exposure.

Hence, this study employed in situ gamma spectrometry measurements to assess the activity concentrations of ^{238}U, ^{232}Th, and ^{40}K in mine tailings, soils, and outcropping rocks in the Witwatersrand Basin in Gauteng Province. In addition, radiological doses and the risks to the human inhabitants in the area were determined. The outcome of this study provides a regional perspective of radioactivity levels in gold mining areas of Gauteng Province and the associated sources of elevated concentrations of NORMs. The findings of this work will inform the National Nuclear Regulator (NNR) during the decision-making process on the regulatory control of NORM residues associated with gold and uranium mining in Gauteng Province.

Study Area

The current study focused on the gold and uranium mining region of the Witwatersrand Basin within the Gauteng Province of South Africa, particularly areas that fall within the extent of the gold deposits of the Witwatersrand Supergroup metasedimentary rocks. This includes the West Rand, East Rand, and Central Rand areas of Gauteng Province (Figure 1). Although Gauteng Province has a surface area of 18,176 km^2, the portion of the study region was smaller due to the focused nature of the study. The focus was given to residential areas proximal to the mining activities including active and abandoned mines as well as tailing dams. The study area has more than a century long history of gold and uranium mining activities that led to the establishment of prominent residential areas such as Randfontein, Westonaria, Carletonville, Krugersdorp, Kagiso, Fochville, Wedela, Soweto, Boksburg, Brakpan, Daveyton, and Tsakane (Figure 1). Currently, it is predominately used as a residential area, with scattered mining and agricultural activities. There are numerous mines and abandoned tailing dams that are surrounded by residential households. The topographical profile of the study area varies from high ground formed by elongated quartzite ridges of the Witwatersrand and Transvaal Supergroups to plain low-lying areas [11]. The area generally experiences warm summer with an average temperature of 26 °C and cold winter with an average temperature of about 2 °C. Based on the investiga-

tion performed for the current work, the average minimum and maximum annual rainfall recorded in the area fall between 260 mm and 1078 mm, respectively.

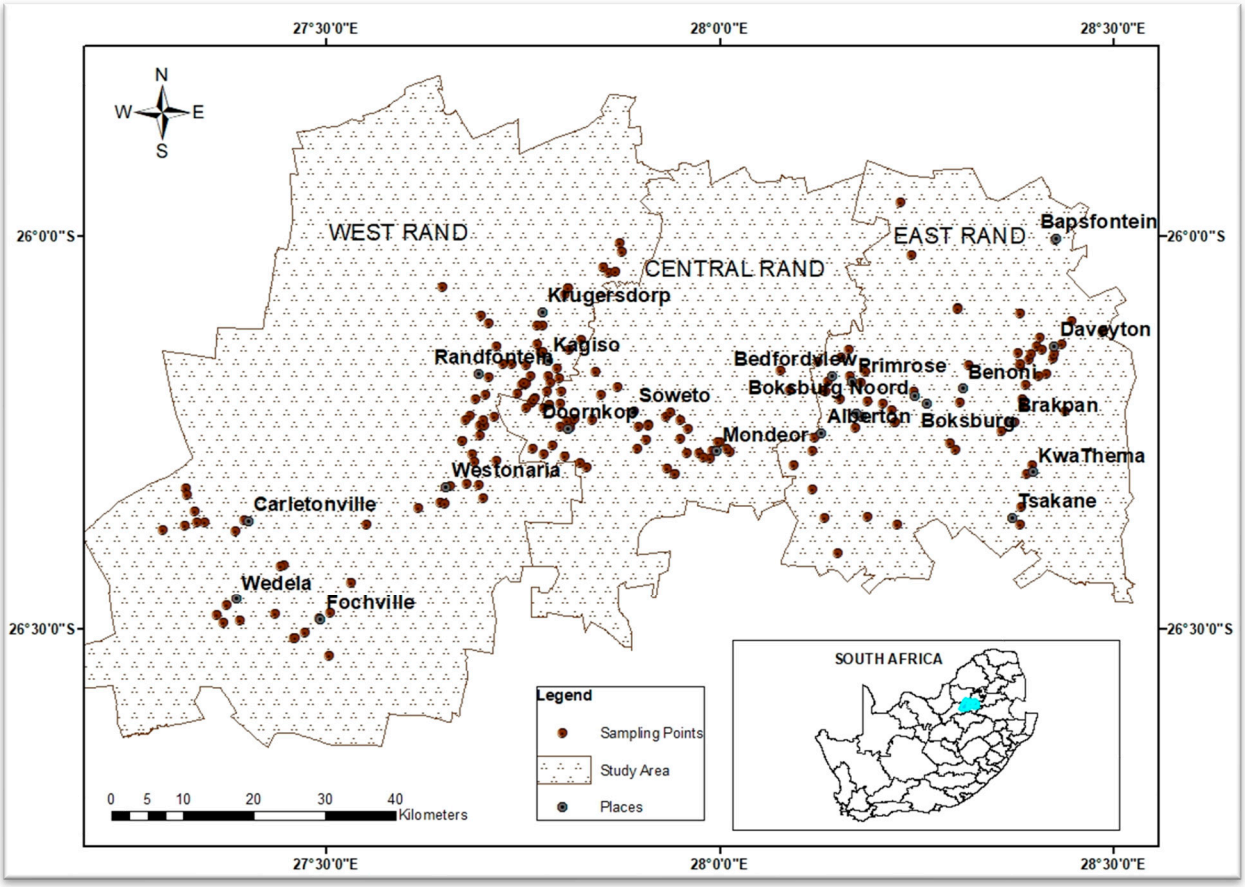


Figure 1. Study area map showing the studied areas and points where measurements were taken.

Geologically, the area is composed of basement crystalline rocks, metasedimentary, and meta-volcanic rocks (Figure 2). The basement rocks are represented by the Archean (3.34 Ga) greenstone rocks that make up the Johannesburg Dome with prominent intrusions of tonalities, granodiorites, granites, gabbro, and migmatites [12–14]. The metasedimentary rocks that include the shale and quartzites of the Witwatersrand Supergroup were deposited between 3074 and 2714 Ma [15]. These rocks belong to the West Rand Group and the Central Rand Group of the Witwatersrand Supergroup. There are also quartz-pebbles conglomerates, which host the majority of the gold mineralization in the area [16]. The area is also underlain by the andesites of the Klipriviersberg Group of the Ventersdorp Supergroup, which were deposited between 2714 and 2665 Ma [17]. The rocks of the Ventersdorp Supergroup are overlain by the Transvaal Supergroup, which is Neo-Archaean to Palaeo-Proterozoic in age [18]. The Transvaal Supergroup rocks are mostly exposed in the south-eastern portion of the study area (Figure 2) and the dominant rock types include the quartzites of the Black Reef Formation, dolomites and iron formations of Chuniespoort Group as well as the Pretoria Group shale alternating with quartzitic sandstones [11,18,19]. The Karoo Supergroup, which overlies the Transvaal Supergroup, is mostly exposed in the eastern part of the area (Figure 2) and the dominant rock types include the mudrocks and sandstone of the Ecca Group [20].

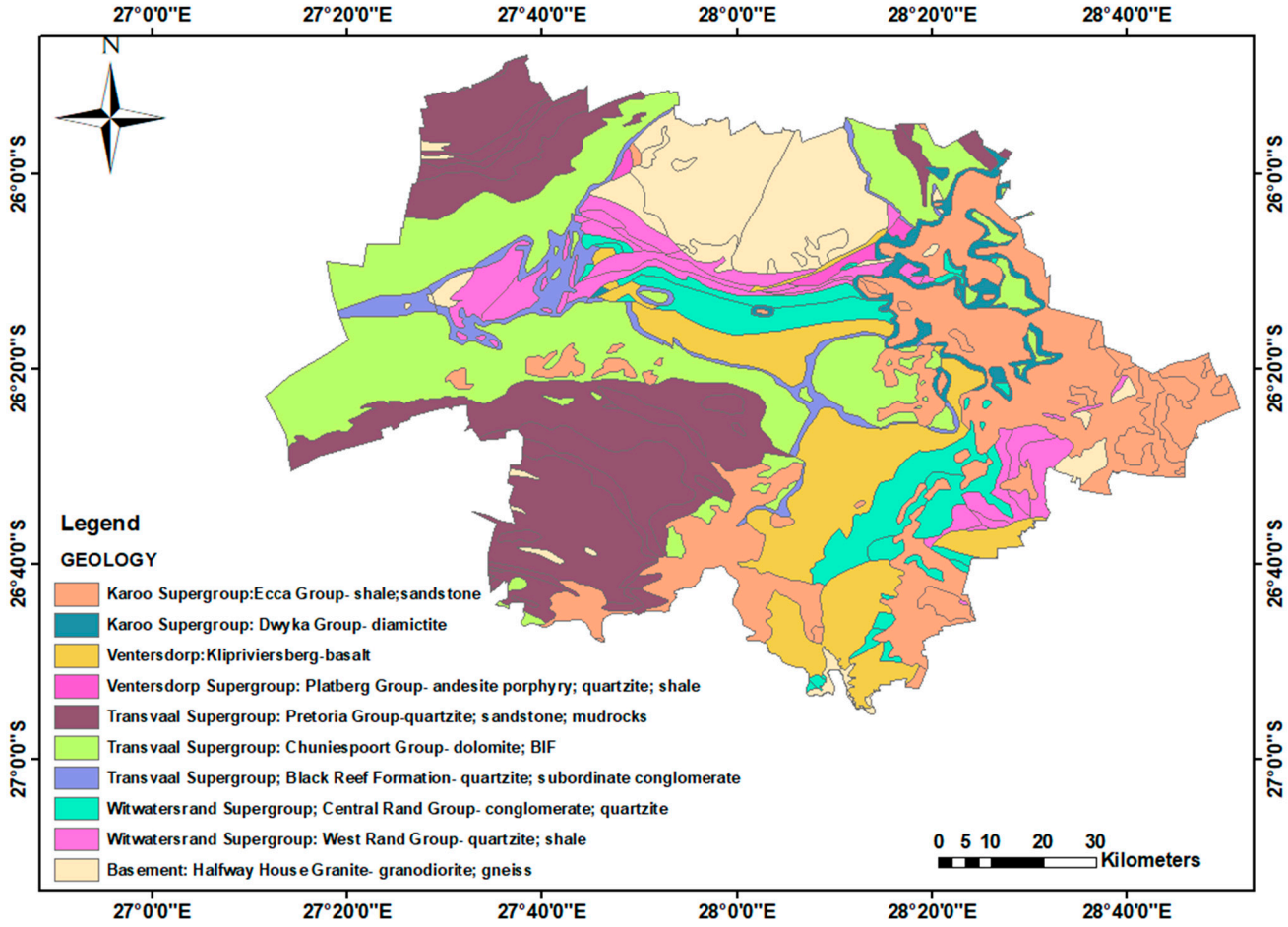


Figure 2. The geology map of the study area.

2. Materials and Methods

2.1. In Situ Gamma Survey

Portable gamma ray spectrometry is a common technique used for measuring the surface concentrations of radionuclides for regional and detailed area surveys. It has been utilized for environmental studies, uranium exploration, and geological mapping [21]. In this study, in situ measurements of activity concentrations of uranium, thorium, and potassium in soils, mine tailings, and underlying rocks were undertaken using a portable RS-230 spectrometer, with a 6.3 cubic inch Bismuth Germanate Oxide (BGO) detector. It is manufactured by Radiation Solutions Inc in Mississauga, Canada. This instrument measures the concentration of potassium in percentage (%), uranium, and thorium in parts per million (ppm) and dose rate (nSv/h) for radiation intensity. It has the energy range of 30 keV–3000 keV with a higher sensitivity that is three times greater than the sodium-iodide (Nal) detector of the same volume. The sensitivity of the detector in counts per second per unit concentration (cps/unit concentration) is 3.29 for potassium, 0.30 for uranium, and 0.13 for thorium.

The device has an automatic stabilization system that allows the unit to maintain a stable operational mode. To ensure the accuracy of the results, the measurements were taken only when the spectrometer was stabilized. The instrument was calibrated to ensure the reliability of results and the IAEA specifications with energy ranges of 1370–1570 keV for potassium, 1660–1860 keV for uranium, and 2410–2810 keV for thorium were used. The concrete test pads with known concentrations of U, Th, and K were used to calibrate the RS-230 spectrometer. The measurements were taken on each of the potassium, uranium, and thorium pads and the sample time was set at one hundred and twenty seconds (two minutes) for each measurement. The measured values were found to fall within the

acceptable concentration range, therefore ascertaining the reliability of the instrument. The uncertainty of measurements was estimated using Equation (1):

$$\text{Error } (\%) = \frac{\text{Actual value} - \text{Measured value}}{\text{Actual value}} \times 100 \quad (1)$$

The percentage errors obtained were 5.43% for U, 1.38% for Th, and 6.25% for K. Overall, the measurement error was ±5%.

In this study, the measurements were taken from the mine tailings, rocks, and soil. For the tailing dams, measurements were taken at different locations at the bottom, middle, and upper part of the tailings. The locations of the surveyed mine tailings are shown in Figure 3. The measurement points for the soils were selected randomly within the study area to capture the spatial distribution of radionuclides, while for rocks, measurements were taken where the outcrops were found. The radioelement concentrations were measured by positioning the gamma ray spectrometer detector close to the surface of the soil, mine tailings, and rocks. The counting time for every measurement was set at one hundred and twenty seconds (two minutes) at each site as this sampling time yielded sufficient statistical data [22]. A total of 188 measurement points were recorded during the survey in mining areas. The locations of the areas surveyed are shown in Figure 1.

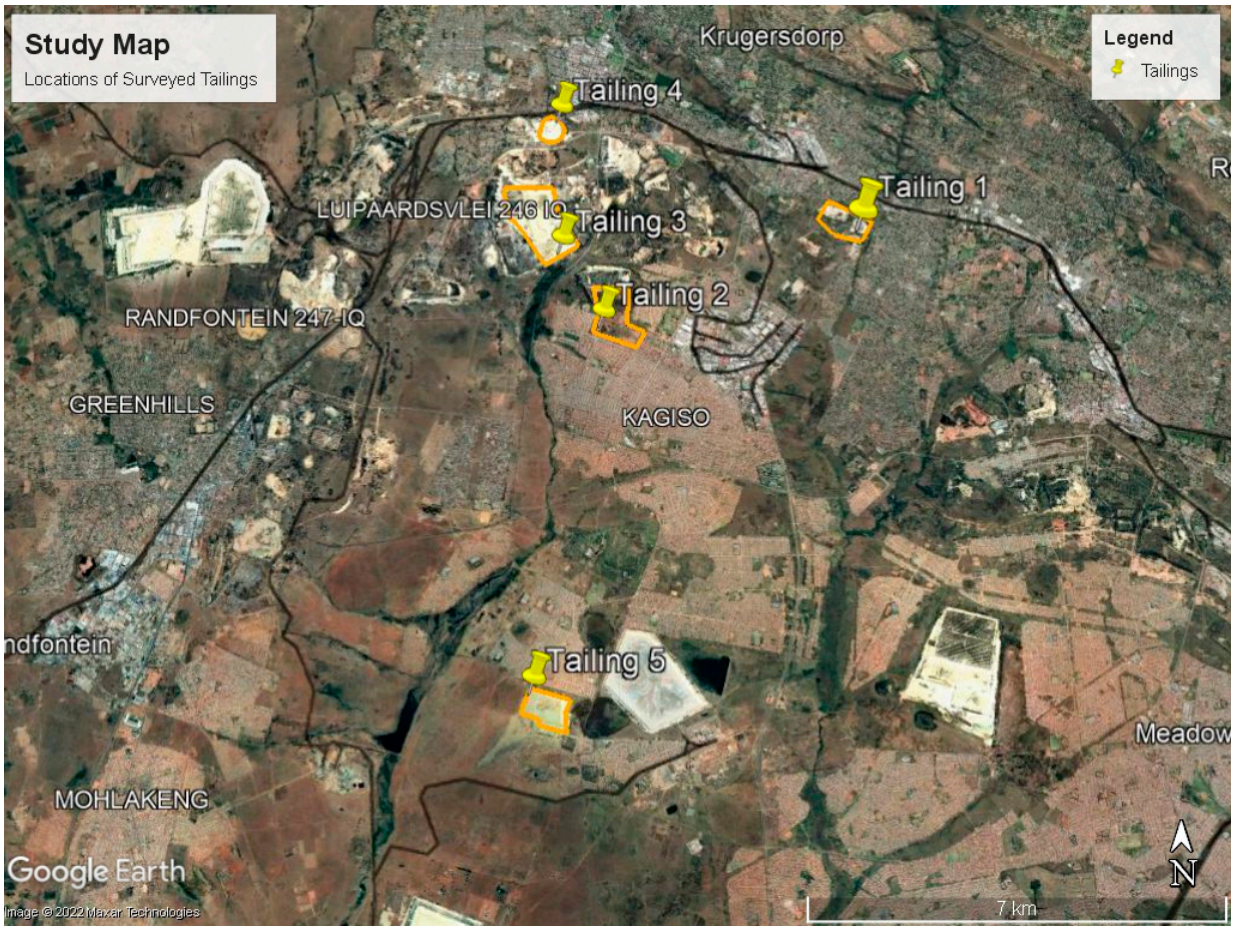


Figure 3. The locations of all the surveyed mine tailings in this study.

The concentrations of all of the measured radioelements were converted to activity concentration in Bq/kg using the following conversion factors [21]:

1 ppm = 12.35 Bq/kg for ^{238}U, 1 ppm = 4.06 Bq/kg for ^{232}Th, 1 % = 313 Bq/kg for ^{40}K. The dose rate measurements were converted from nSv/h to nGy/h.

2.2. Calculation of Radiological Doses

In this study, the outdoor absorbed dose rate and effective dose were calculated to determine the external gamma radiation exposure due to NORMs. The absorbed dose rate for mine tailings, soils, and rocks was calculated from the activities of ^{40}K, ^{238}U, and ^{232}Th using Equation (2):

$$D(nGyh^{-1}) = DCF_K . A_K + DCF_U . A_U + DCF_{Th} . A_{Th} \quad (2)$$

where D is the absorbed dose and A_K, A_U, and A_{Th} are the activity concentrations for ^{40}K, ^{238}U, and ^{232}Th, respectively. DCF_K, DCF_U, and DCF_{Th} are the absorbed dose rate conversion factors for ^{40}K, ^{238}U, and ^{232}Th in nGy/h/Bqkg^{-1}. The conversion factors used are given below [4]:

DCF_K = 0.0417 nGy/h/Bqkg^{-1}, DCF_U = 0.462 nGy/h/Bqkg^{-1}, and DCF_{Th} = 0.604 nGy/h/Bqkg^{-1}.

The average annual effective dose (E) was estimated from the absorbed dose rate (D) by using the dose conversion factor of 0.7 SvGy^{-1}, an outdoor occupancy factor of 0.2, and exposure duration per year (8760 h), as represented in Equation (3) [4]. The effective dose represents the sum of the tissue-weighted equivalent doses in all specified organs and tissues of the human body. It is mainly used for regulatory purposes to demonstrate compliance with the exposure dose limits [23].

$$E\ (mSvy^{-1}) = D(Gyh^{-1}) \times 0.2 \times 8760\ (h) \times 0.7\ (SvGy^{-1}) \times 10^{-6} \quad (3)$$

3. Results

3.1. Activity Concentration of Natural Radionuclides in the Tailing Dams

The results in Table 1 present the statistical summary of naturally occurring radionuclides found in the studied gold mine tailings in the West Rand region, which are shown in Figure 3. The activity concentrations for ^{238}U, ^{232}Th, and ^{40}K range from 209.95 to 2578.68 Bq/kg, 19.49 to 108.00 Bq/kg, and 31.30 to 626.00 Bq/kg, respectively. The levels of ^{238}U in the mine tailings were generally higher than the concentrations of ^{232}Th and ^{40}K. Elevated levels of ^{238}U were found in mine tailings two (2) and three (3), with average values of 1830.68 Bq/kg and 1525.64 Bq/kg, respectively. In comparison to the other studied mine tailings, it was found that the overall average activity concentrations obtained in this study were comparable with the levels found in the mine tailings in the Wonderfonteinspruit Catchment Area in the lower part of the West Rand District [9], as shown in Figure 4. Therefore, this indicates that the abandoned mine tailings found in the upper and lower part of the West Rand District have similar radioactivity characteristics.

Table 1. In situ activity concentration of ^{238}U, ^{232}Th, and ^{40}K in the gold mine tailings of the West Rand area and the corresponding absorbed dose and effective dose.

Source (No. of Measurements)	Statistical Parameters	Activity Concentration (Bq/kg)			Absorbed Dose	Annual Effective Dose
		^{238}U	^{232}Th	^{40}K		
Mine tailing 1 (n = 8)	Min	260.59	31.67	31.30	173.70	0.21
	Max	381.62	47.50	219.10		
	Median	321.72	37.35	140.85		
	Average	314.00	38.21	133.03		
	SD	38.67	4.63	70.49		
Mine tailing 2 (n = 3)	Min	1638.85	36.13	344.30	891.76	1.09
	Max	1963.65	55.62	626.00		
	Median	1889.55	45.88	344.30		
	Average	1830.68	45.88	438.20		
	SD	170.22	9.74	162.64		

Table 1. *Cont.*

Source (No. of Measurements)	Statistical Parameters	Activity Concentration (Bq/kg)			Absorbed Dose	Annual Effective Dose
		^{238}U	^{232}Th	^{40}K		
Mine tailing 3 (n = 3)	Min	958.36	33.29	125.20	753.46	0.92
	Max	2578.68	108.00	281.70		
	Median	1039.87	54.81	250.40		
	Average	1525.64	65.37	219.10		
	SD	912.87	38.45	82.81		
Mine tailing 4 (n = 2)	Min	224.77	19.49	31.30	135.39	0.17
	Max	293.93	25.58	62.60		
	Median	259.35	22.53	46.95		
	Average	259.35	22.53	46.95		
	SD	48.90	4.31	22.13		
Mine tailing 5 (n = 1)	Average	209.95	20.30	93.90	113.17	0.14
Average for all tailings	Min	209.95	19.49	31.30	113.17	0.14
	Max	2578.68	108.00	626.00	891.76	1.09
	Average	827.92	38.46	186.24	413.50	0.51
Wonderfonteinspruit Catchment Area Mine tailings [9]	Min	87.2	20.5	226.5		
	Max	2668.9	89.7	781.0		
	Average	785.3	43.9	427.0		

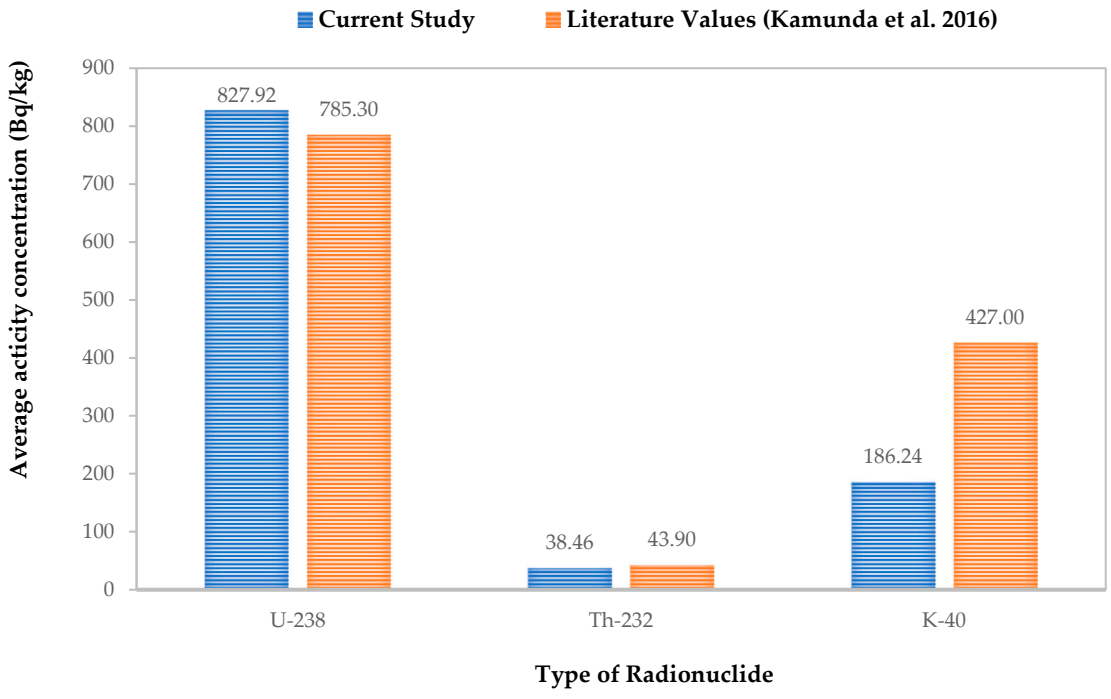
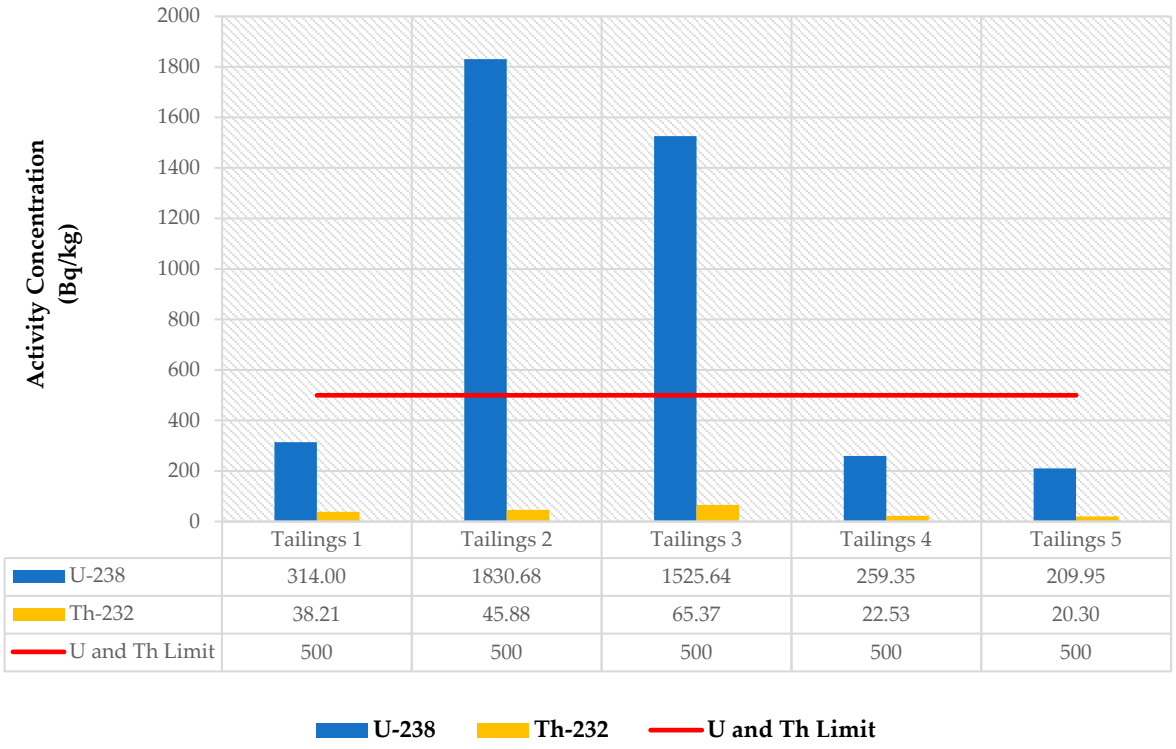


Figure 4. A comparison of the collective average activity concentrations obtained in the current study with those found in mine tailings in the Wonderfonteinspruit Catchment Area [9].

Based on the regulatory limits, materials with activity concentration levels above 500 Bq/kg for uranium−238 and thorium−232 and 50,000 Bq/kg for potassium−40 are deemed to be subjected to regulatory control [24]. In this case, the ^{238}U levels in tailings two (2) and three (3) were about three times higher than the regulatory limit of 500 Bq/kg for ^{238}U [24], as shown in Figure 5. This serves as an indication that regulatory control is required to monitor and control the exposures from these tailings. Moreover, the ^{238}U levels found in these tailings fell within the concentration range of 100 ppm (1235 Bq/kg)

to 300 ppm (3705 Bq/kg) which were found in the gold reefs of South Africa [25]. The high concentrations in these tailings, which are comparable with typical levels in gold reefs, could imply that uranium was not extracted from the gold-bearing reefs and during gold refining processes, it was disposed of in tailing dams.



	Tailings 1	Tailings 2	Tailings 3	Tailings 4	Tailings 5
U-238	314.00	1830.68	1525.64	259.35	209.95
Th-232	38.21	45.88	65.37	22.53	20.30
U and Th Limit	500	500	500	500	500

Figure 5. Comparison of the activity concentrations of ^{238}U and ^{232}Th in mine tailings with the recommended regulatory limits.

On one hand, tailings one (1), four (4), and five (5) contained ^{238}U concentrations that were below the regulatory limit of 500 Bq/kg (Figure 5). The low concentrations of ^{238}U in these tailings could indicate that uranium was extracted as the by-product of gold during mining processes, or that the mine tailings could have undergone further reprocessing afterward. With regard to ^{232}Th and ^{40}K, all of the surveyed mine tailings were found to have levels that were way below the respective regulatory limit of 500 Bq/kg for ^{232}Th and 50,000 Bq/kg for ^{40}K [24].

Statistical analysis was performed to determine the contribution of each radionuclide to the total activity concentration in the assayed mine tailings. The results are presented in Figure 6. It was found that there was a good correlation (R^2 = 0.9887) between ^{238}U and the total activity concentration due to natural radionuclides in the tailing dams (Figure 6a), whereas ^{232}Th and ^{40}K showed a poor correlation of R^2 = 0.011 (Figure 6b) and R^2 = 0.100 (Figure 6c), respectively. This gives an indication that ^{238}U contributes significantly to the total activity concentration of mine tailings compared to ^{232}Th and ^{40}K. The mineralogical composition and chemistry of the tailings depend on the mineralogy of the ore as well as the leaching process used for extraction [26]. The conglomerate reefs, which are the main ores mined for gold and uranium in the Witwatersrand basin, contain uranium minerals such as uraninite [16,27]. Hence, the mine tailings have higher levels of ^{238}U nuclides compared to the ^{232}Th and ^{40}K radioisotopes.

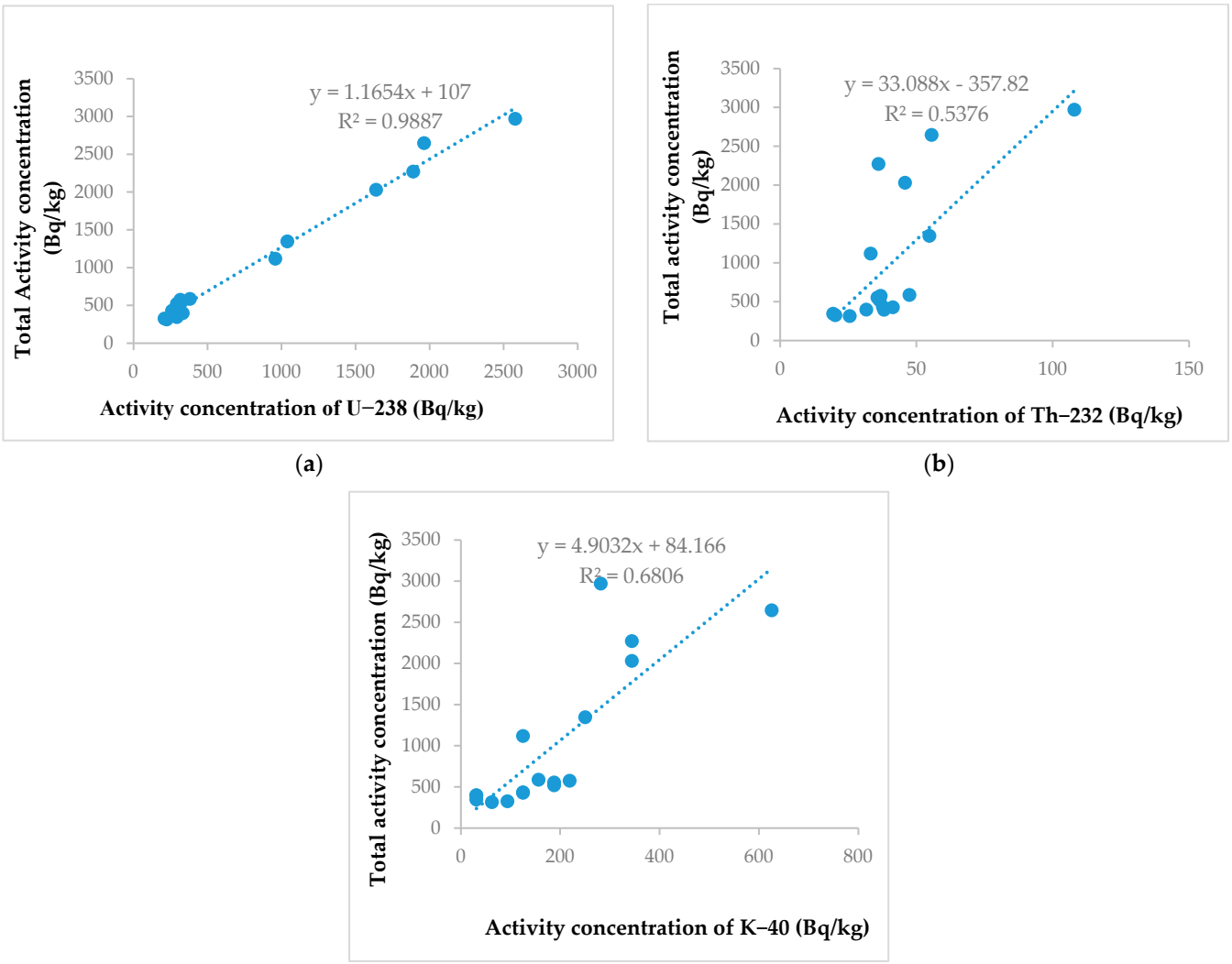


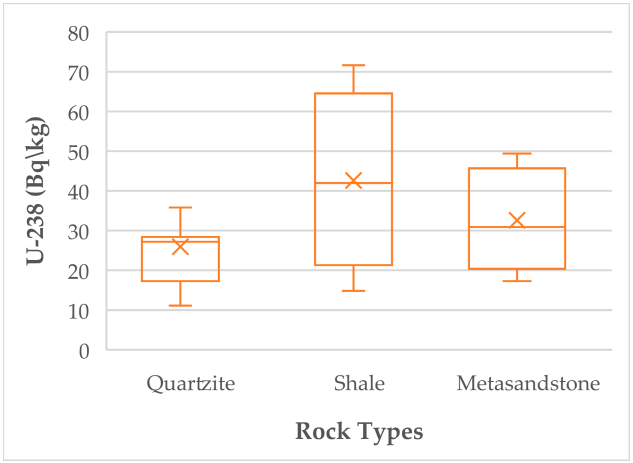
Figure 6. Contribution of each radionuclide to the total activity concentration due to ^{238}U, ^{232}Th, and ^{40}K in the tailings. (**a**–**c**) presents the contribution of ^{238}U, ^{232}Th, and ^{40}K to the total radioactivity measured, respectively.

3.2. Activity Concentrations of Natural Radionuclides in Rocks

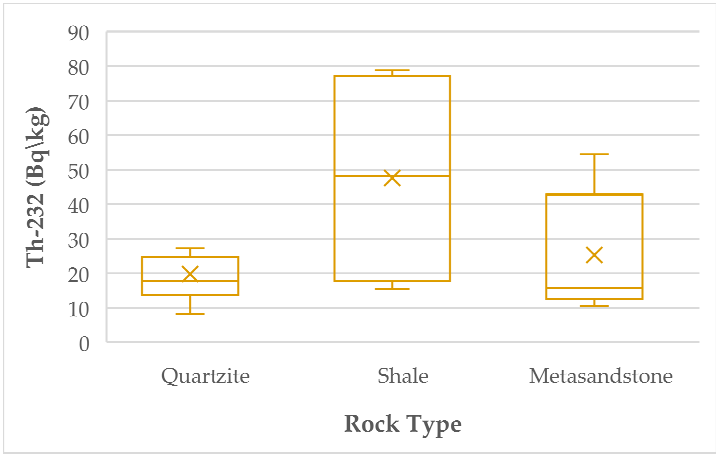
The activity concentrations of ^{238}U, ^{232}Th, and ^{40}K in rocks found in the West Rand and East Rand districts are presented in Table 2. In all of the studied rock types, the activity concentration of ^{238}U and ^{232}Th ranged from 11.12 to 71.63 Bq/kg and 8.12 to 78.76 Bq/kg, respectively. The activity concentrations of ^{40}K ranged from 31.30 to 970.30 Bq/kg. In most common rock types on Earth, the activity concentrations of ^{238}U, ^{232}Th, and ^{40}K typically range between 7 and 60 Bq/kg, 8 and 80 Bq/kg, and 90 and 1400 Bq/kg, respectively [2]. In this study, the levels of naturally occurring radionuclides present in rocks were comparable with the typical concentrations found in most of the prevalent rock types in the Earth's crust [2]. The highest radioactivity levels were observed in shales whereas quartzites generally exhibited relatively low activity concentrations (Figure 7). This may be because shales are primarily dominated by clay minerals, which have the ability to adsorb uranium and thorium more easily [28,29].

Table 2. In situ activity concentration of ^{238}U, ^{232}Th, and ^{40}K in different rock types of the West Rand and East Rand areas.

Rock Type	Radionuclides	Statistical Parameters					
		No. of Samples	Minimum	Maximum	Mean	Median	SD
Quartzite	^{238}U	11	11.12	51.87	25.94	27.17	11.35
	^{232}Th		8.12	44.66	19.82	17.86	10.10
	^{40}K		31.30	970.30	327.23	281.70	281.83
Shale	^{238}U	4	14.82	71.63	42.61	41.99	23.23
	^{232}Th		15.43	78.76	47.60	48.11	32.33
	^{40}K		31.30	939.00	618.18	751.20	401.95
Metasandstone	^{238}U	5	17.29	49.40	32.60	30.88	13.15
	^{232}Th		10.56	54.40	25.33	15.83	18.05
	^{40}K		469.50	719.90	626.00	657.30	93.90
All Rock Types	^{238}U	20	11.12	71.63	30.94	27.79	15.27
	^{232}Th		8.12	78.76	26.76	19.29	19.29
	^{40}K		31.30	970.30	460.11	453.85	303.14



(**a**)



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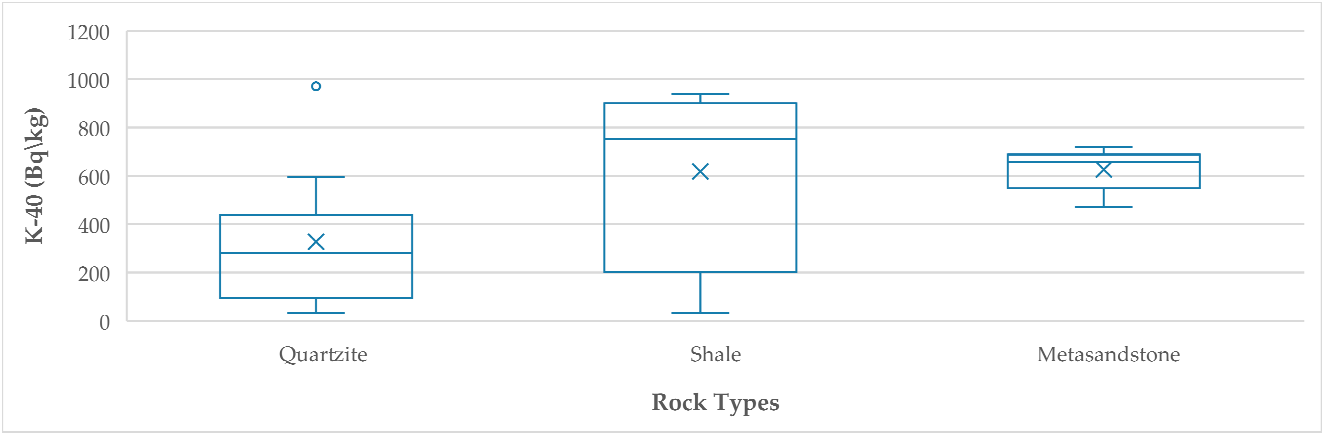

(**c**)

Figure 7. The activity concentration range of ^{238}U, ^{232}Th, and ^{40}K in the different rock types found in the West Rand and East Rand. (**a**–**c**) presents the concentration of ^{238}U, ^{232}Th, and ^{40}K in different rock types.

3.3. Activity Concentrations of Natural Radionuclides in Soil

Table 3 presents the activity concentrations of ^{238}U, ^{232}Th, and ^{40}K found in the surface soil in the study area. The activity concentration level of ^{238}U in the West Rand ranged between 12.35 and 941.07 Bq/kg, with an average value of 53.09 Bq/kg. In the Soweto area, the activity concentration of ^{238}U ranged between 14.82 and 191.43 Bq/kg, with the corresponding mean value of 47.79 Bq/kg. The highest ^{238}U levels were found in soils around the Krugersdorp area in Kagiso, with a maximum activity concentration of 941.07 Bq/kg and an average of 88.12 Bq/kg. This area is adjacent to an abandoned mine tailing (Mine tailing 2) with a high ^{238}U activity concentration averaging 1830.68 Bq/kg (Table 1). Moreover, the mixing of tailing material with soil was observed at this location. As a result, the higher concentration of ^{238}U found in soil in this area was attributed to contamination arising from the tailings dam. The ^{238}U contamination was found at a distance of approximately 80 m from the adjacent tailings dam (Mine tailing 2). However, the concentration decreased by almost 50% (1830.68 Bq/kg to 941.07 Bq/kg) within 80 m from the tailings dam. This was attributed to the mixing effect of the tailing material with uncontaminated soil. In the Soweto area, the highest ^{238}U activity of 191.43 Bq/kg was also found at a site where the surface soil was mixed with the material from a nearby tailings dam situated 100 m from the surveyed location. This suggests that the transport of materials eroded from the tailings dam was limited to short distances from the tailings dam and may not result in extensive regional contamination of surface soils.

Table 3. In situ activity concentration of the ^{238}U, ^{232}Th, and ^{40}K in the soils of the West Rand, East Rand, and Soweto.

Region	Area	No. of Measurements	^{238}U (Bq/kg)		^{232}Th (Bq/kg)		^{40}K (Bq/kg)	
			Mean	Range	Mean	Range	Mean	Range
West Rand	Muldersdrift	6	45.88	12.35–77.70	52.85	30.04–83.23	1027.68	187.80–1471.10
	Krugersdorp	20	88.12	19.76–941.07	31.79	12.59–64.55	150.24	62.60–344.30
	Randfontein	15	36.80	16.06–100.04	28.91	15.83–51.97	91.81	31.30–187.80
	Westonaria	10	44.21	23.47–72.87	45.07	25.98–61.31	175.28	62.60–594.70
	Fochville	4	29.02	17.29–44.46	45.37	37.76–60.09	305.18	219.10–438.20
	Wedela	2	35.20	32.11–38.29	40.80	35.73–45.88	234.75	187.80–281.70
	Carletonville	11	35.59	12.35–61.75	39.23	24.36–62.52	167.88	62.60–313.00
	All measurements	68	53.09	12.35–941.07	37.26	12.59–83.23	232.91	31.30–1471.10
Soweto	All measurements	30	47.79	14.82–191.43	64.34	18.27–333.73	220.14	62.60–594.70
East Rand	Benoni	7	44.28	18.53–64.22	10.71	6.5–15.8	187.80	62.60–313.00
	Kwathema	7	39.70	22.23–60.52	42.80	32.07–58.46	187.80	93.90–344.30
	Brakpan	10	52.22	39.52–67.93	41.06	28.83–60.90	228.04	125.20–406.90
	Daveyton	6	42.61	23.47–60.52	43.85	23.55–81.20	203.45	93.90–250.40
	Germiston	9	39.52	13.59–144.5	30.13	14.62–54.00	222.23	125.20–438.20
	Boksburg	6	58.46	22.23–100.04	31.19	23.95–40.60	198.23	125.20–313.00
	Bapsfontein	5	35.07	25.94–50.64	37.51	22.74–54.00	306.74	93.90–469.50
	Mondeor	5	21.00	9.88–25.94	20.57	10.56–30.45	245.18	125.20–438.20
	Bedfordview	5	25.44	18.53–35.82	25.82	22.33–31.67	344.30	125.2–594.70
	Alberton	5	40.76	23.47–75.34	40.46	21.92–67.40	333.87	156.50–469.50
	All measurements	65	40.52	9.88–144.50	35.48	10.56–81.20	229.86	62.60–594.70
UNSCEAR (2000)			35		30		400	

In the East Rand region, a total of sixty-five soils were assayed from different areas and the results are presented in Table 3. The activity concentration of ^{238}U exhibited a broad range in the area, from 9.88 to 155.50 Bq/kg (average of 40.52 Bq/kg). The highest activity was measured in soil from the Germiston area (Primrose), located 5 m from a covered mine tailing. As such, the elevated activity can be attributed to the impact of tailings. Furthermore, the assayed soil is a mixture of the eroded proximal tailings and the local soil, hence the elevated activity. Similarly, high ^{238}U activity (100 Bq/kg) was also recorded in soil from the Boksburg area (Reiger Park), which is in close proximity to prominent and uncovered mine tailings. This is a common phenomenon in the study area, highlighting one of the major impacts of mine tailings to the surrounding environment. In contrast, the lowest ^{238}U activity was recorded in soil from Mondeor, an area characterised by the absence of mine tailings. Lower activities were exclusively recorded in soils found in areas with no mine tailings or mining activities, thus stressing the underpinning impact of mining on the environment. The areas of high and low concentrations of ^{238}U in the surface soil are depicted in the spatial map in Figure 8.

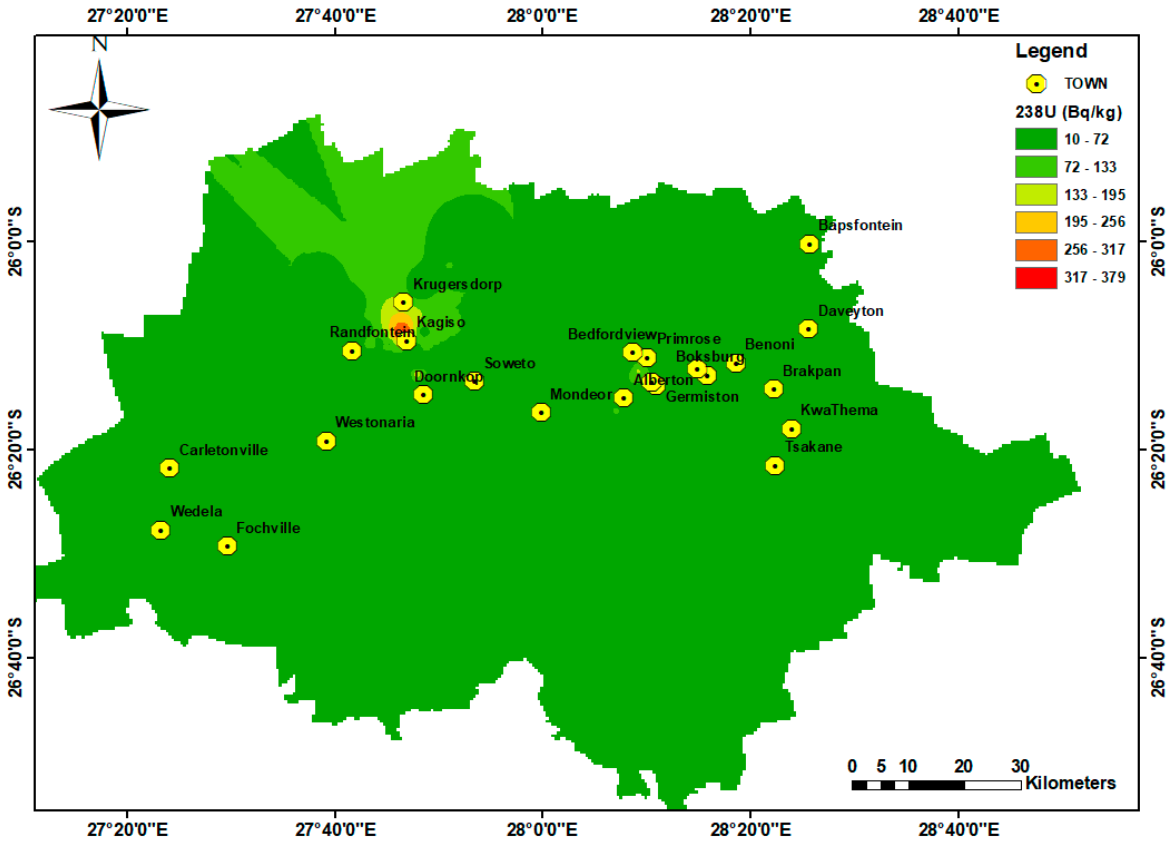

Figure 8. Spatial activity distribution map of ^{238}U (Bq/kg) in the surface soil.

With respect to ^{232}Th, the mean activity concentration in the West Rand was found to be 37.26 Bq/kg with a range of 12.59 to 83.23 Bq/kg (Table 3). In the Soweto area, the range was between 18.27 and 333.73 Bq/kg, with a mean value of 64.34 Bq/kg. The elevated activity concentration of 333.73 Bq/kg was found in Doornkop in Soweto in soil mixed with tailings material. The ^{232}Th activity showed a broad range in the East Rand, from 6.50 to 81.20 Bq/kg with an average activity of 35.48 Bq/kg (Table 3). The highest activity was more than twice the average activity of the area and was recorded in soil from Daveyton, located approximately 2.5 km from a mine tailing. The elevated ^{232}Th could be linked to the tailings since thorium commonly occurs in uranium-bearing minerals, as is the case in the Witwatersrand Basin. The spatial distribution of ^{232}Th found in the area is shown in Figure 9.

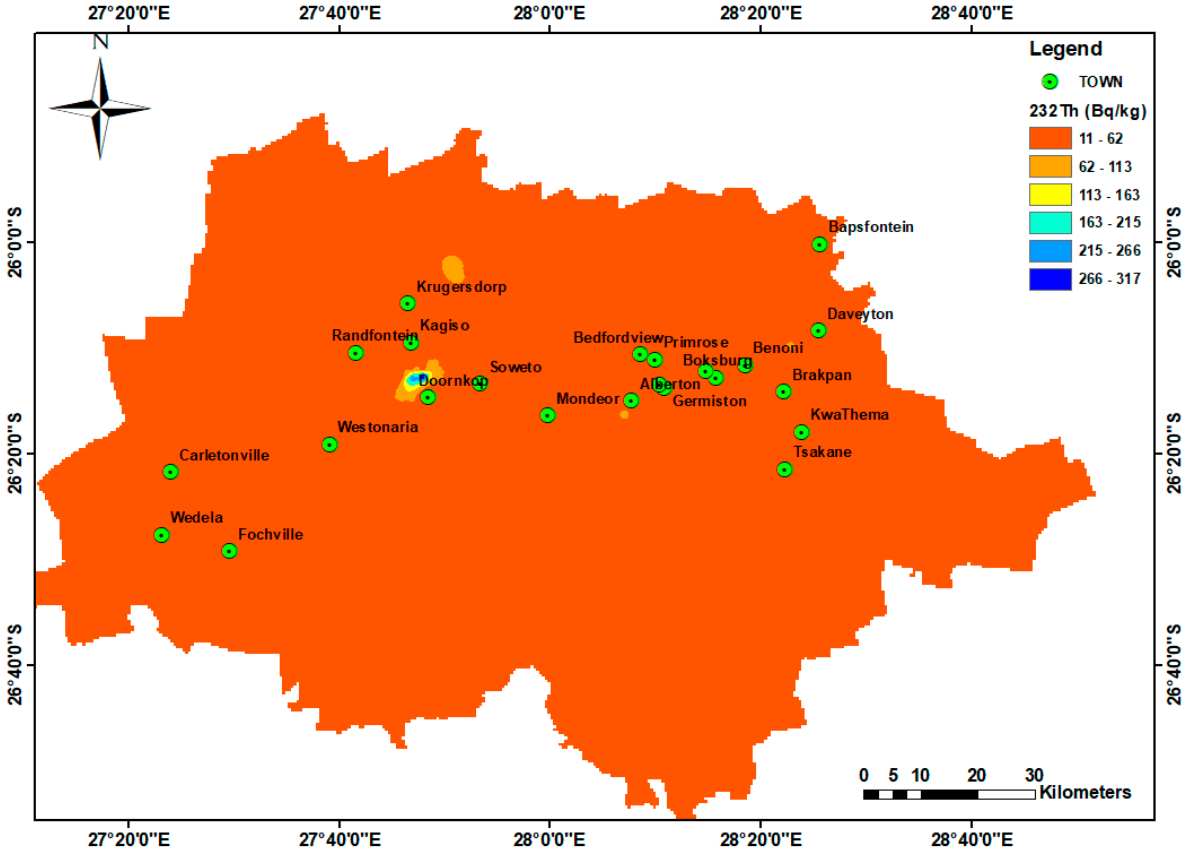

Figure 9. Spatial activity distribution map of ^{232}Th (Bq/kg).

For ^{40}K, the average activity concentration in the West Rand area was 232.91 Bq/kg, with a range of 31.30 to 1471.10 Bq/kg. In contrast, ^{40}K activity in Soweto varied between 62.60 and 594.70 Bq/kg, with a mean value of 220.14 Bq/kg. The activity concentration of ^{40}K was highly variable in the East Rand region, ranging from 62.40 to 594.70 Bq/kg with an average of 229.86 Bq/kg. Soil from the Bedfordview area contained the highest ^{40}K activity concentration, which is well above the average activity of the region and is attributed to the potassium-bearing clay minerals and feldspars found in the local geology, particularly granites found in the vicinity. However, on a regional scale, the highest activity concentration of ^{40}K was found in the northern part of the study area in the West Rand region (Figure 10). The Basement granites is the main lithology in this area (Figure 2), as such, the high activity concentration of ^{40}K is associated with potassium feldspar found in these rocks. The high ^{40}K activities are linked to the local geology as opposed to the other radionuclides whose elevated activities in soils are directly related to contributions from mining activities.

The average global activity concentration of ^{238}U, ^{232}Th, and ^{40}K in soils is reported to be 35 Bq/kg, 30 Bq/kg and 400 Bq/kg, respectively [4]. In comparison to the worldwide values, the mean activity concentration of ^{40}K in the three studied regions (West Rand, Soweto, and East Rand) was lower than the world average value of 400 Bq/kg whereas those of ^{238}U and ^{232}Th were slightly higher than the worldwide average values (Table 3). Although the average values of ^{238}U and ^{232}Th were higher than the global average concentration, all except one assayed point around the Krugersdorp area in Kagiso (941.07 Bq/kg) were still far below the NNR's regulatory limit of 500 Bq/kg for ^{238}U and ^{232}Th [24]. Similarly, the ^{40}K values measured at all sites showed levels that were below the NNR's regulatory limit of 50,000 Bq/kg.

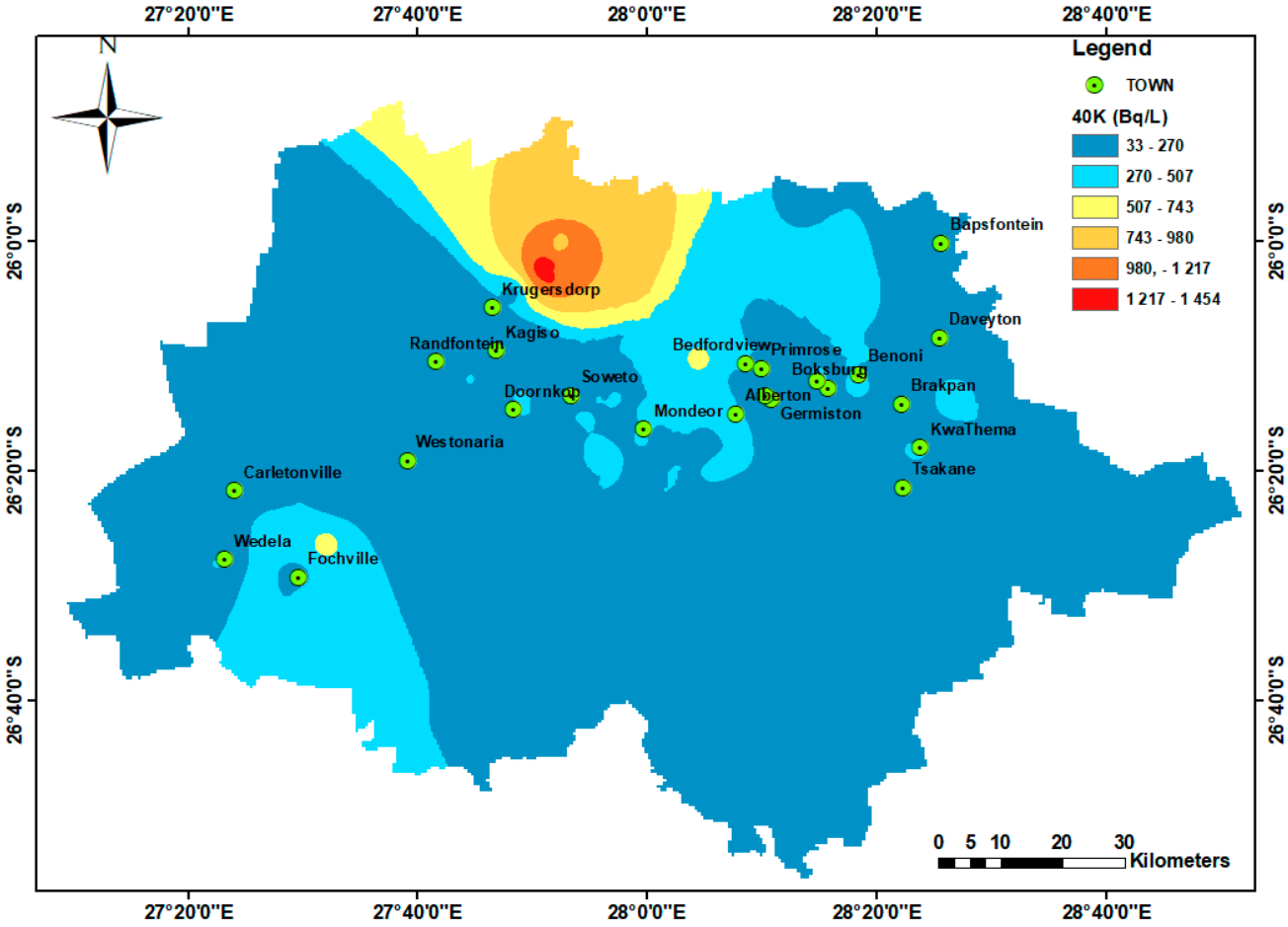

Figure 10. Spatial activity distribution map of ^{40}K (Bq/kg).

In general, the results obtained in this study indicate that the highest activity concentrations of ^{238}U and ^{232}Th were confined to areas where mine tailings were washed off and deposited on surface soils in close proximity to the tailings.

3.4. Absorbed Dose Rate in Air

The outdoor absorbed dose rate calculated from activity concentrations of ^{238}U, ^{232}Th, and ^{40}K in the mine tailings of the West Rand region ranged between 113.17 and 891.76 nGy/h (average of 413.50 nGy/h) (Tables 1 and 4). On the other hand, the absorbed dose rate from rocks in the West Rand varied between 11.35 and 119.82 nGy/h (average of 51.92 nGy/h), whereas that of rocks in the East Rand region ranged between 30.28 and 79.38 nGy/h (average of 45.30 nGy/h). The global mean absorbed dose rate in ambient air from terrestrial gamma radiation is reported as 59 nGy/h [4]. In comparison, the average absorbed dose rates from all mine tailings exceeded the global average (Tables 1 and 4) whereas the average value from rocks was slightly lower (Table 4). In the West Rand region, the absorbed dose rate from soils was highly variable with a range between 22.44 and 478.99 nGy/h (average of 56.73 nGy/h). The highest absorbed dose rate was found in an area impacted by the presence of mine tailings, however, the average value obtained compared well with the worldwide average of 59 nGy/h (Table 4). In the Soweto area, the absorbed dose rate from soils ranged from 27.50 to 293.93 nGy/h, (average of 70.12 nGy/h), which was slightly higher than the world average. The outdoor absorbed dose rate from soils in the East Rand region varied between 20.73 to 95.09 nGy/h and showed an average dose rate of 49.09 nGy/h. Interestingly, the highest absorbed dose rate was related to the soil from the Alberton area, which was attributed to the elevated activities of all of the analysed radionuclides in the soil (Table 3). Furthermore, the dose rate at this location was

above the global mean absorbed dose rate of 59 nGy/h from terrestrial gamma radiation in ambient air [4]. It is worth noting that the majority of absorbed dose rates that exceeded the global average were associated with soils located in close proximity to the mine tailings. In most instances, the soil was found within prominent residential areas of high occupancy. Therefore, this emphasizes the detrimental impacts of mining on the environment and the risks to human health.

Table 4. The calculated absorbed dose rate in the air at a height of 1 m above the ground and the associated effective doses.

Region	Parameters	Statistical Parameters	Type of Material (Number of Samples)		
			Mine Tailings (*n* = 5)	**Soil (*n* = 68)**	**Rocks (*n* = 12)**
West Rand	Calculated Absorbed Dose (nGy/h)	Range	113.17–891.76	22.44–478.99	11.35–119.82
		Mean	413.50	56.73	51.92
	Calculated Effective Dose (mSv/y)	Range	0.14–1.09	0.03–0.59	–0.15
		Mean	0.51	0.07	0.06
Soweto				Soil (*n* = 30)	
	Calculated Absorbed Dose (nGy/h)	Range		27.50–293.93	
		Mean		70.12	
	Calculated Effective Dose (mSv/y)	Range		0.03–0.36	
		Mean		0.09	
East Rand				Soil (*n* = 64)	Rocks (*n* = 9)
	Calculated Absorbed Dose (nGy/h)	Range		20.73–95.09	30.28–79.38
		Mean		49.09	45.30
	Calculated Effective Dose (mSv/y)	Range		0.03–0.12	0.04–0.10
		Mean		0.06	0.06

3.5. Annual Effective Dose

The calculated annual effective dose due to natural radionuclides present in mine tailings of the West Rand was found to range between 0.14 to 1.09 mSv/y, with an average of 0.51 mSv/y (Tables 1 and 4). The effective dose related to the radionuclide content in soils from the West Rand region varied from 0.03 to 0.59 mSv/y (average of 0.07 mSv/y) while that from the rocks ranged between 0.01 and 0.15 mSv/y (average of 0.06 mSv/y) (Table 4). In contrast, the effective dose associated with soils from the East Rand region ranged between 0.03 and 0.12 mSv/y (average dose of 0.06 mSv/y) whilst that in rocks ranged between 0.04 and 0.10 mSv/y (average of 0.06 mSv/y). In the Soweto area, the calculated effective dose ranged from 0.03 to 0.36 mSv/y (average of 0.09 mSv/y) (Table 4). In relation to public exposure, the dose limit applicable to the average member of the critical group within the exposed population in South Africa is 0.25 mSv/y [24]. As such, the external gamma radiation received due to ^{238}U, ^{232}Th, and ^{40}K in the surface soil and rocks in the study area is well below the regulatory limit of 0.25 mSv/y, with the exception of two locations (soil-related): Kagiso (0.59 mSv/y) and Soweto (0.36 mSv/y). The highest annual effective doses were associated with the tailing dams. In two of the studied mine tailings (mine tailing 2 and 3), the effective doses were found to be about four (4) times higher than the recommended dose limit of 0.25 mSv/y (Figure 11). These two mine tailings and two sampling points were deemed as hotspot areas and pose a radiological risk to members of the public residing within the vicinity.

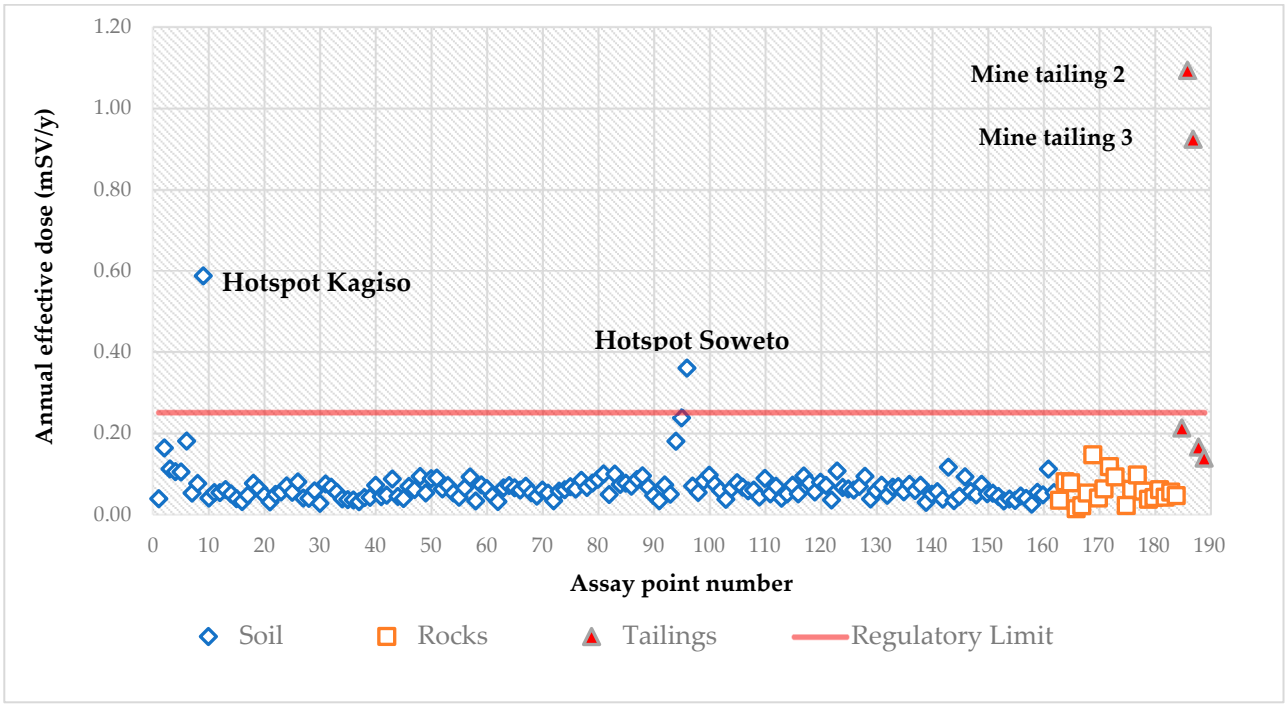

Figure 11. Annual effective dose received in the study area compared with the regulatory limit of 0.25 mSv/y.

4. Discussion

The findings of this study showed that of the three radionuclides studied, ^{238}U was found to be the most significant radioactive contaminant of radiological concern. High ^{238}U concentrations were found in the mine tailings than in the surrounding soils and rocks since the ore, which was used for gold mining, was brought to the surface through the mine shaft from deep underground. However, soils in the immediate vicinity of abandoned mine tailings were found to be contaminated with ^{238}U, which was found in elevated concentrations in the tailings. The contamination was only observed in areas where mine tailing materials are being washed off and deposited on surface soils in the vicinity. This, therefore, shows the immediate impact of tailings on the environment, particularly in soils used by local communities. However, on a regional scale, it was found that the radioactivity levels in surface soil mainly depend on the activity concentration of the underlying geology. Overall, the annual effective dose estimations indicate that the soils and rocks found in the area do not significantly contribute to the external gamma radiation received by members of the public as opposed to the mine tailings and soils impacted by tailings that were identified as significant sources of external gamma exposure.

The study revealed that mine tailings 2 and 3 and soils identified as hotspots contained high activity concentrations that resulted in higher radiological dose exposures. The use of these radioactive materials could contribute to high radiation doses and detrimental consequences to the health of members of the public in the vicinity. As such, protective actions should be implemented to ensure the safety of the public and reduce human exposure. It is therefore proposed that the nearby communities must avoid using tailing materials for construction or any other purposes. Moreover, agricultural activity is also discouraged in areas that have been identified as hotspots. In overall, the findings of this research provide the current radioactivity levels and areas of radiological concern in gold mining areas of Gauteng Province. As such, the results of this study may be used as a basis to identify areas where detailed radiological monitoring, considering all of the critical exposure pathways, should be focused in future research.

5. Conclusions

In this study, the activity concentrations of ^{238}U, ^{232}Th, and ^{40}K in abandoned gold tailing dams, soils and rocks from different parts of the West Rand, East Rand, and Soweto areas were analysed using the in situ gamma ray spectrometry technique. Additionally, radiation hazards due to the exposure of the studied radionuclides were evaluated. On the basis of the measured activity concentrations and the corresponding effective doses, the hotspot areas were found to be associated with mine tailing 2 and mine tailing 3 as well as soils in Kagiso and Soweto, which are all located in close proximity to the mine tailings. In these areas, the recommended dose limit of 0.25 mSv/y was exceeded. Therefore, further monitoring and regulatory control measures are required to ensure the protection of all residents in these locations as well as the environment.

Author Contributions: P.M.M. and S.C.M. carried out the investigation, collected field data, and drafted the original manuscript. T.A.A. supervised the work and acquired funding for this study. He also contributed to the write-up and editing of the manuscript. I.K. contributed to the write-up, review and editing of the manuscript. S.N. assisted with the acquisition of finances used for the implementation of this research and reviewed the manuscript. M.M. co-supervised the research work and reviewed the manuscript. All authors have read and agreed to the published version of the manuscript.

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Conflicts of Interest: The authors declare no conflict of interest in the findings reported in this work.

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