

Human Health Risk Assessment of Environmental Radionuclides and Heavy Metals around a Gold Mining Area in Gauteng Province, South Africa

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DECLARATION

I, Caspah Kamunda (Student number: 24818852), hereby declare that the work presented in this thesis is a product of my own research. It has not been submitted in part or whole for any degree at any other university before. The data presented is original and all analyses were done by the author under the guidance of the supervisors. Any other data taken from other sources has been referred to and fully acknowledged.

Signature of the student:

Date: _____

(Caspah Kamunda)

DEDICATION

This work is most sincerely dedicated to my family, my mother and my sisters for their encouragement and loving support during my PhD studies. I can never forget my caring wife; Kudakwashe and my two special sons Tinotenda and Tadiwanashe, who tolerated my absence from home throughout the period of my studies. Without their motivation and patience, it would not have been possible to accomplish my goals. I would also like to dedicate this work to my relatives and friends who have always been there for me when things appeared to be impossible. Lastly, I would like to pay special tribute to my late father who was my main source of inspiration. May his soul rest in eternal peace.

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ABSTRACT

Naturally Occurring Radionuclide Materials (NORMs) and heavy metals are a common occurrence in the environment and have resulted in human exposure for the entire history of mankind. However, anthropogenic activities such as mining have resulted in elevated levels of these contaminants in the environment. A health risk assessment of NORMs and Heavy Metals from a gold mining area in the Gauteng Province of South Africa has been evaluated. In this study, a total of 136 soil, water and plant samples were collected from around the mine and from the control area for laboratory analyses. A broad energy germanium (BEGe) detector with a relative efficiency of 60 % and a resolution of 2.0 keV at 1332 keV gamma ray emission of ^{60}Co was used to measure the activity concentrations of NORMs. RESidual RADioactivity (RESRAD) OFFSITE modeling program (version 3.1) was then used to estimate excess cancer risk from NORMs for a hypothetical resident scenario. An Inductively Coupled Plasma Mass Spectrometer (ICP-MS) was on the other hand used to measure concentrations of heavy metals in all samples as well. The AlphaGuard Monitor was used to measure indoor radon gas. All the measurements were compared with those from the control area and reference standards.

The average activity concentrations in Bq.kg^{-1} for ^{238}U , ^{232}Th and ^{40}K in soil from the mine was found to be 574.3 ± 39.5 , 49.4 ± 8.5 and 424.7 ± 129.3 , respectively. These results showed higher levels of ^{238}U in soil from the mining area compared to the control area and the worldwide average. In plant samples, the average activity concentrations of ^{238}U , ^{232}Th and ^{40}K in Bq.kg^{-1} were 17.4 ± 3.1 , 19.7 ± 1.6 and 146.7 ± 9.2 , respectively. The values measured for ^{238}U and ^{232}Th in plant samples were higher than acceptable limits whilst that of ^{40}K was expected although it exceeded that of both ^{238}U and ^{232}Th . For water samples, the average values for ^{238}U , ^{232}Th and ^{40}K in Bq.L^{-1} were 0.66 ± 0.03 , 0.56 ± 0.03 and 7.36 ± 0.58 , respectively. Compared with samples from the control area and South African Guidelines, average values for ^{238}U , ^{232}Th and ^{40}K in water samples were higher.

Radiological hazards for soil, plant and water samples were also considered for the study area. The average values for radium equivalent activity (Ra_{eq}) in soil from the mining area was $677.7 \pm 43.8 \text{ Bq.kg}^{-1}$ compared to $86.9 \pm 1.4 \text{ Bq.kg}^{-1}$ from the control

area. This value was above the worldwide average of 370 Bq.kg^{-1} as recommended by the International Atomic Energy Agency (IAEA) under normal circumstances. For plant samples, the average value of Ra_{eq} in Bq.kg^{-1} from the mining area was 56.8 ± 4.0 compared to $44.1 \pm 2.1 \text{ Bq.kg}^{-1}$ from the control area, while in water samples it was $2.03 \pm 0.07 \text{ Bq.L}^{-1}$ compared to $1.39 \pm 0.08 \text{ Bq.L}^{-1}$. These values for plant and water samples are lower than the worldwide average.

The Annual Effective Dose Equivalent (AEDE) values in mSv y^{-1} for soil, plant and water samples from the mining area were 0.38, 0.03 and 1.17×10^{-03} , respectively. All these values are below the worldwide average of 0.48 mSv for terrestrial gamma radiation. The annual effective dose of natural radionuclides in mSv through the ingestion of water and plant samples were estimated to be 1.24×10^{-03} and 1.23×10^{-04} , respectively. Indoor radon from the mining area and the control area was also measured. The average activity concentrations of indoor radon from the mining area was 119.5 Bq/m^3 , compared to a 19.7 Bq/m^3 from the control area. This translated to an average annual effective dose of 3.01 and 0.5 mSv for the mining area and control area, respectively. According to UNSCEAR, the worldwide average annual effective dose from inhalation of radon and its decay products is 1.26 mSv. The average value from the mining area studied was higher than the worldwide average. When all the samples were put together, the total annual effective dose from the measured samples was 3.42 mSv.y^{-1} , which is higher than the worldwide average of 2.4 mSv.y^{-1} from natural radiation. This value is also higher than 1 mSv.y^{-1} , a limit recommended by ICRP for individual members of the public.

Average external hazard indices (H_{ex}) for soil, plant and water samples from the mining area were 1.8, 0.2 and 5.49×10^{-3} , respectively, while corresponding internal hazard (H_{in}) values were 3.4, 0.2 and 7.28×10^{-3} , respectively. This shows that the average H_{ex} and H_{in} values for soil samples were higher than unity, posing a potential radiological threat to members of the public in the mining area. Plant and water samples were radiologically safe to members of the public as their values were less than one.

The cancer risk for members of the public living in the mining area as a result of natural radionuclides was estimated using the RESRAD-OFFSITE Computer Code. The maximum total cancer risk for all the pathways was found to be 6.52×10^{-5} .

This was higher than the South African Individual cancer risk limit for the public of 5×10^{-6} .

Heavy metals in soil, plant, and water samples were also measured from the different locations of the gold mining area and from the control area. The average concentrations (mg.kg^{-1}) in soil decreased in the order of $\text{Cr} > \text{Ni} > \text{As} > \text{Zn} > \text{Cu} > \text{Co} > \text{Pb} > \text{Hg} > \text{Cd}$. The values were as follows: Cr (316.88); Ni (115.87); As (77.); Zn (68.01); Cu (50.79); Co (27.52); Pb (4.69); Hg (0.09); and Cd (0.05), respectively. These concentrations were higher compared to the soil from the control area. As and Cr were found to be higher than the maximum allowable limits. Average concentrations (mg.kg^{-1}) in plant samples decreased in the order of $\text{Cr} > \text{Ni} > \text{As} > \text{Zn} > \text{Cu} > \text{Co} > \text{Pb} > \text{Hg} = \text{Cd}$. Average values were as follows: Cr (6.90); Ni (2.89); As (2.50); Zn (0.44); Cu (0.32); Co (0.28); Pb (0.16); Hg and Cd (0), respectively. The results indicated that average concentrations of As and Ni in plant samples were higher than FAO/WHO and South African safe limits. Average concentrations of heavy metals in water samples (mg.L^{-1}) decreased in the order of $\text{Ni} > \text{Cu} > \text{Zn} > \text{As} > \text{Cr} > \text{Co} > \text{Pb} > \text{Hg} = \text{Cd}$. Average concentrations were as follows: Ni (0.39); Cu (0.38); Zn (0.33); As (0.19); Cr (0.14); Co (0.08); Pb (0.01); Hg and Cd (0), respectively. These average concentrations of heavy metals in water were generally higher than those from the control area. Compared with reference levels the average concentrations of As, Ni, Cr, and Zn in drinking water were higher than permissible limits.

For non-carcinogenic risk of heavy metals in all the samples, a total HQ value of 2.62 was found. This value is greater than 1, which potentially causes a health risk to the public living in the gold mining area. The total carcinogenic risk from all the samples as a result of heavy metals was found to be 1.91×10^{-4} . The sum total of the cancer risk due to natural radionuclides (6.52×10^{-5}) and that of heavy metals (1.91×10^{-4}) was found to be 2.56×10^{-4} (1 in 3906 individuals). This total value obtained was higher than the acceptable cancer risk limit. From the findings presented, it can be concluded that natural radionuclides and heavy metal pollution in the mining area are an issue of health concern.

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LIST OF ABBREVIATIONS

ADI	Average Daily Intake
AT	Average Time
BW	Body Weight
C	Concentration
HQ	Hazard Quotient
IR	Ingestion Rate
RfD	Reference Dose
nGy.h ⁻¹	nano Gray per hour
DCF	Dose conversion factor
H _{in}	Internal hazard index
H _{ex}	External hazard index
Ra _{eq}	Radium equivalent activity
USEPA	United States Environmental Protection Agency
WHO	World Health Organization
NORM	Naturally Occurring Radioactive Material
DWAF	Department of water affairs
NNR	National Nuclear Regulator
IAEA	International Atomic Energy Agency
ICRP	International Commission for Radiation Protection
GDARD	Gauteng Department of Agriculture and Rural Development
UNSCEAR	United Nations Scientific Committee on the Effects of Atomic Radiation
CARST	Center for Applied Radiation Science and Technology
ICP-MS	Inductively Coupled Plasma Mass Spectrometry
EU	European Union
mSv	millisievert (10 ⁻³ Sievert)
Bq	Becquerel
²³⁸ U	Uranium-238
²³² Th	Thorium-232
⁴⁰ K	Potassium-40

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CHAPTER 1: INTRODUCTION

1.1 Background

Mining is responsible for a series of environmental and human health disasters through the production of large volumes of waste into the environment. The global environment we live in contains small amounts of natural radionuclides that are derived from primordial and cosmogenic sources. This is as a result of the material, from which the earth was formed, about 4.5 billion years ago (Lee et al., 2004). It contained many unstable nuclides, which when they decay, continue to expose humankind to radioactivity. In most of the places on earth, levels of natural radioactivity as well as heavy metals are comparable, but in some localities it varies by large amounts (Esposito et al., 2002). This abnormally high amount is normally attributed to anthropogenic activities such as mining. Even with relatively efficient mining operations, high concentrations of natural radionuclides and of heavy metals are released into the air and water leaving a legacy of environmental contamination in nearby communities (Olawuyi and Mudashir, 2013).

1.2 Environmental Impact of Mining

Environmental impact assessments of mining projects often underestimate the potential health risks created to mankind. To dispose of large quantities of waste produced by mining activities, it has been a tremendous challenge for the mining industry. Because of their quantities and chemical characteristics, wastes produced may cause an increase in mortality rate and serious irreversible hazards to human health and the environment (WHO, 1946).

It has been established that mining activities involve long lived natural radionuclides of uranium (U), actinium (Ac) and thorium (Th) decay series (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).

Most mining operations also use toxic chemicals in order to extract valuable minerals. Such chemicals include heavy metals like mercury that is added to the ore in the process. When the precious mineral has been extracted, the crushed ore and chemicals become waste that is piled as tailings into large slime dams. In some rare cases, the tailings have often created some of the worst environmental disasters of all accidents (MINEO Consortium, 2000). When the wet tailings fail, their toxic waters can destroy aquatic life and poison drinking water supplies for many kilometres downstream (MINEO Consortium, 2000).

During mining operations, acid drainage may also be produced that may pose a long-term devastating impact on rivers, streams and aquatic life. Acid drainage occurs when sulphide-bearing minerals, such as pyrites are exposed to oxygen or water, producing sulphuric acid (Ripley, 1996). This sulphuric acid is what dissolves heavy metals, such as cadmium (Cd), arsenic (As) and lead (Pb) from the rock dumps, mine tailings and other openings. The process may take centuries because of enormous quantities of exposed rocks at some mine sites (Schmiermund and Drozd, 1997).

Mining operations also cause air pollution through particulate matter that is transported by the wind as a result of excavations, blasting, and transportation of materials. Once pollutants enter the atmosphere, they undergo either physical or chemical changes until they reach a receptor. These pollutants have the potential to cause serious effects to people's health and to the environment (USEPA, 2009).

Apart from pollution, mining operations have a disruptive effect to the environment, causing a reduction in the capacity of the land to support various post-mining activities (Mhlongo et al., 2013). Soils contaminated by chemical spills at mine sites may also pose a direct contact risk when these materials are misused (USEPA, 2009). Mining may also disturb food availability and shelter for wildlife (MacCallum, 1989).

1.3 Naturally Occurring Radioactive Materials

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).

NORMs are divided into three main categories depending on their sources of origin. These are cosmogenic, terrestrial and anthropogenic radionuclides. Cosmogenic radionuclides are formed by the interaction of cosmic rays with the earth's atmosphere (Hussain and Hussain, 2011). They interact with stable nuclides in our atmosphere to form radionuclides such as ^{14}C , ^7Be , ^{22}Na and ^3H (Tykva and Berg, 2004). Terrestrial radionuclides originate from the rocks of the earth, from our food and from our drinks. These are also referred to as primordial radionuclides. Primordial radionuclides have very long half lives in the order of a billion years, which is comparable to the age of the earth. Major examples of these primordial radionuclides are uranium-235 (^{235}U), uranium-238 (^{238}U), thorium-232 (^{232}Th), and potassium-40 (^{40}K) (Lee et al., 2004). The first three have decay chains associated with them. So they have always been present in the earth's crust and within the tissues of all living species and their decay products are referred to as daughters (Murray et al., 1987). Anthropogenic radionuclides are as a result of man-made activities (Boaretto et al., 1994).

1.4 Heavy Metals in the Environment

Human beings have also been exposed to heavy metals for an immeasurable amount of time. These heavy metals are found everywhere as a result of both natural and anthropogenic activities and have been some of the most serious problems in the vicinity of mining sites (Wang et al., 2005). By definition, heavy metals are basically toxic metals, irrespective of their atomic mass or density (Singh,

2007). Most of them have a high atomic number, high atomic weight and a specific gravity greater than 5 g/cm^3 (Singh, 2007). This classification includes some metalloids, transition metals, basic metals, lanthanides, actinides and metals of groups III to V of the Periodic Table. Examples include arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), lead (Pb), mercury (Hg), nickel (Ni), cobalt (Co), Iron (Fe), zinc (Zn), selenium (Se), aluminium (Al) and manganese (Mn) (Brandy and Weil, 1999).

While organic pollutants slowly decompose to produce carbon dioxide and water, heavy metals tend to bio-accumulate because they cannot be broken down. They persist in the environment and are transferred from one place to another. They are ingested daily by humans either through air, food, water or soil. Human symptoms and the level of toxicity depend on the type of metal, the dose absorbed, and whether or not the exposure was acute or chronic (CSIR, 2008). Some heavy metals are carcinogenic while others are detrimental to body organs. Even in very small amounts, heavy metals can be toxic to humans and animals (USEPA, 1995). Their effects on humans include increased incidence of tuberculosis, chronic bronchitis, asthma, and gastrointestinal diseases. The impacts to aquatic life may range from immediate fish killing to affecting their ability to reproduce. Heavy metals are also considered toxic to plants due to their acute and chronic effect on them. For example, high levels of Cd in soils cause a reduction in photosynthesis, nutrient, and water uptake (Wójcik and Tukiendorf, 2004).

1.5 Problem Statement

The problems of radiological hazards in mining areas have received considerable attention in recent years because of the epidemiological evidence of lung cancer among uranium miners (Mulloy et al., 2001). The hazard is not only limited to U mines, but can also extend to gold mines. Investigations have also shown that the same radiological elements that have caused lung cancer among uranium miners, also occur in other types of mines and in some instances, in sufficient concentrations to cause occupational illness (Mulloy et al., 2001). Direct access to mining areas exposes the public to radiation and heavy metals through dermal contact with soil

and water, inhalation of dust and ingestion of food and water (Coetzee, 2008). According to a report by Flowers and Zeese (2013), mining not only exposes uranium to the atmosphere, but releases other radioactive elements as well as toxic heavy metals.

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., 2004a). The WCA is the world's largest gold and uranium mining basin that has taken place from more than 120 mines, with extraction of 43 500 tonnes of gold in one century and 73 000 tonnes of uranium between 1953 and 1995 (Winde et al., 2004a). The basin covers an area of 1600 km², and has led to a legacy of some 400 km² of mine tailings and 6 billion tonnes of pyrite tailings containing low-grade uranium (Lieberink, 2009). This uranium is the principal contaminant of concern within the gold mining areas of the WCA. When pyrite tailings are oxidised with water, they produce Acid Mine Drainage (AMD) that solubilises heavy metals. These mine tailings are usually one to two kilometres in diameter and are littered everywhere, posing an environmental threat to local communities.

Although the concentrations of both gold and uranium vary widely within the ores, the gold-bearing ore is estimated to contain almost ten times the amount of uranium than gold (GDARD, 2009). The measured uranium content of many fluvial sediments in the WCA, exceeded the exclusion limit for regulation by the National Nuclear Regulator (NNR, 2007). The existence of large quantities of this uranium in the area induces a radiological burden to man and possible damage to agricultural land and water resources. People residing in the WCA reportedly use mine drainage water in households, agriculture, recreation, and fishing. Liefferink (2011a) also observed, on frequent occasions, young mothers and children eating uraniferous salt crusts.

In recent years, the IAEA has put greater attention to radiation dose measurements because of the possible health risk it may pose to humans (Santawamaitre, 2012). 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 and heavy metals has been carried out. There has not been studies to accurately measure the extent of the possible risk of these pollutants on the health of the population. This researcher has, therefore, carried out a comprehensive study on the human health risk assessment of environmental radionuclides and heavy metals on the population around the gold mining area.

1.6 Previous Studies Relevant to the Project

Environmental impact of mining projects and their potential health risks have always been of great concern to the whole world for some time now. Research in Portugal, where waste waters from uranium ore milling facilities were discharged into river basins has shown enhanced radioactivity in freshwater ecosystems (Carvalho et al., 2007). In a study carried out in uranium ore mines in Mexico to estimate effective doses from sources of natural and artificial origin in the general population and workers, it was discovered that the effective dose via the different exposure pathways in the population was two times higher than for the regional background (Gasó et al., 2005). In a related study undertaken in the Philippines, on the environmental and health impact of mercury in communities near abandoned mercury mine, results showed that certain species of fish; had exceeded the mercury levels recommended by the national guidelines (Nelia et al., 2006).

An area with elevated natural background radiation was also investigated in Iran, around Khak-Sefid, Ramsar (Ziajahromi et al., 2014). Evaluation of the health risk caused by exposure to NORMs in soils resulted in high risk values of ^{226}Ra compared to ^{232}Th and ^{40}K . This showed that ^{226}Ra was of major concern to human health in the study area (Ziajahromi et al., 2014). Assessment of the environmental impact of radionuclides and trace metals in the Taboshar and Digmai mining and tailing sites in Tajikistan, performed in 2006 and 2008, also showed enhanced levels of natural radioactivity as well as heavy metals, which presented a potential radiological and chemical impact on man and the environment (Skipperud et al., 2012).

The uranium mines in Jaduguda and nearby areas in India were also investigated for radioactivity (Tripathi et al., 2008). The gamma radiation dose rates observed at different locations 1 metre above the tailings surface varied from 0.8 to 3.3 mGy.h⁻¹. The geometric mean of the radiation dose rate indicated that it was above the local background level for the area. In a related study of the radiological situation due to exposure to gamma radiation, radon and thoron carried out at selected former uranium mining and processing areas of Kazakhstan, Kyrgyzstan, Uzbekistan and Tajikistan from Central Asian countries, it was discovered on the contrary, that doses of ionizing radiation did not present any serious hazard to the health of the resident public (Stegnar et al., 2013).

Heavy metal concentrations in rice were also investigated in Fuzhou, China. In this study, samples collected from mining settlements to analyse concentrations of Cd, Cu, Sb, Cr, Pb, Ni, and As in rice revealed high levels of Cd and As (Qing-Long et al., 2014). This showed that more attention was needed towards monitoring toxic substances in agricultural produce in order to assure food safety for consumers. The effect of mining activities on the environment was also investigated in Mongolia, Central Asia along the Boroo River (Bolormaa et al., 2005). Water samples collected for the determination of heavy metal contents revealed that metal concentrations in water samples downstream were relatively higher than those observed upstream. The findings concluded that gold mining activity in this area was influencing high metal concentrations in downstream waters. Concentrations of six heavy metals (As, Ni, Cr, Cu, Cd, and Pb) in fish and edible plants were also measured to evaluate contamination levels and health risks for Bangladeshi adults (Islam et al, 2014). The study showed that the inhabitants who consumed contaminated fish and edible plants were exposed chronically to metal pollution with carcinogenic and non-carcinogenic consequences.

In the Plateau State of Nigeria, very high concentrations of ²²⁶Ra were discovered in mine tailings from the thirty-one (31) samples collected from the mining and milling site that was analyzed for ⁴⁰K, ²²⁶Ra and ²³²Th. These high levels were of radiological concern to the public (Mangset and Sheyin, 2009). In the Zamfara State of Nigeria, Pb-containing gold ore poisoned and killed more than 400 children in

2010. Furthermore, about 180 villages were contaminated by people grinding the ore, in and around their homes (Abdulakeem and Raheem, 2013).

In Tongo, Ghana, assessments of radon and gamma radiation levels carried out in underground artisanal gold mines, on the contrary, revealed that measurements were well below the lower action level of 500 Bqm^{-3} recommended by ICRP for workplaces (Doyi et al., 2013). Investigations on the concentrations and distribution of natural radionuclides in soils and water with the aim of evaluating their radiological health hazards were also carried out from Sakwa Wagusu Area in Kenya. The values of the external and internal indices were found to be less than unity signifying safe levels. The calculated outdoor mean effective dose rate was 0.17 mSv.y^{-1} , a value less than 1 mSv.y^{-1} , the upper limit recommended for the public by ICRP (Aguko et al., 2013).

Heavy metals that were analyzed in water, fish, nails and scalp hair in children below 10 years in Migori gold mining belt of Kenya revealed that water and fish were contaminated from gold mining activities (Ngure et al., 2013). Pb and Cd concentrations in nails showed elevated levels above those reported in occupationally exposed residents. The research indicated that consumers in the study area were exposed to heavy metals through the consumption of contaminated fish in the rivers flowing through the gold mining area. This was a potential health risk to the population. Mihaljevic et al (2013) also reported that air-borne dust released from the mines and smelters during extraction and processing of copper ores in the Copperbelt of Zambia was causing a potential health risk.

Back in South Africa, Coetzee et al (2005) conducted a study in wetlands within the West and Far West Rand and broader Witwatersrand goldfields of Johannesburg, and discovered that crops that were being planted within the area contained elevated levels of radioactive and toxic heavy metals. Many of the communities make their living from subsistence farming, growing food and obtaining drinking water from the surrounding surface water sources. As a consequence of concerns reported by Coetzee et al, (2005, 2006) regarding radiological health hazards associated with the real or potential use of contaminated water, the National Nuclear Regulator (NNR) of South Africa conducted an independent assessment of the radiological impacts of

the mine water discharges on the local population (NNR, 2007). The results of the study indeed confirmed that uranium was posing a hazard to water users in the catchment area due to its chemical toxicity. It was discovered that the mean values of the heavy metals for the WCA samples not only exceeded natural background levels, but also concentrations of regulatory concern (NNR, 2007). However, potential radiation exposures caused by emissions of radon and contaminated dust from mining legacies were not investigated. In a scientific research report in March 2006, the Water Research Commission (WRC) of South Africa (Boer et al., 2006) had earlier reported that the measured uranium concentrations of many fluvial sediments in the WCA, including those of off-mine properties exceeded the limit for regulation by NNR (NNR, 2007).

In 2008, a radiometric survey of airborne nature in the West and Far West Rand goldfields, done for the Department of Water Affairs by the Council for Geoscience also revealed that many of the residential areas (e.g. Carletonville, Westonaria, Khutsong) were at high risk of radioactivity contamination (DWA, 2009). They concluded that the residents could be exposed to direct external and internal gamma radiation, which included inhalation and ingestion of radionuclides. They also concluded that the residents could be exposed to radon gas and heavy metals. The Gauteng Department of Agriculture and Rural Development (GDARD), also reported that 380 mine dumps and slimes dams in Gauteng Province were highly contaminated by radioactive materials (GDARD, 2012). The report also noted that mine tailings were causing radioactive dust fallout, soil contamination, and toxic water pollution which were presenting a potential danger to nearby informal settlements.

Hamman and Rensburg (2011) also conducted a study to detect and quantify the transfer and accumulation of Cr, Cd and U from the WCA into the surrounding environment and the cattle grazing area. The results indicated that there was evidence of Cr, Cd and U transfer and accumulating from the WCA into the surrounding environment. A related study was also conducted in the Eastern Cape Province of South Africa in a small town called Alice to investigate heavy metal pollution of edible plants cultivated in home gardens (Bvenura and Afolayan, 2012). The concentration of Mn was found to be very high in soils and spinach, whilst that of

Zn exceeded safe levels in onions, spinach and tomatoes. The high concentrations of these metals in soil was linked largely to the parent material of the soils in the area.

In the North West Province of South Africa, Kgabi et al., (2009) carried out a study of indoor and outdoor radon concentrations within the Klerksdorp gold mining areas around Midvaal Water and Botshabelo Community Health Centers and discovered that Klerksdorp area had the highest radon concentration and exposure in Southern Africa. However, this did not consider other sources of radioactivity. Kaonga and Ebenso, (2011) also carried out an evaluation of the air quality with regards to particulate matter in Kanana area of Klerksdorp gold mining town of North-West province. The study concluded that the particulate emission levels in the study area were unacceptably high.

In a summarized report by Liefferink (2011b) on the findings of the investigation conducted by NNR (2007) and other researchers, it was recommended that measurement of activity concentrations as well as radiological impact assessments as a result of mine tailings in the WCA be conducted periodically (Coetzee et al., 2006; NNR, 2007). It is also against this background that a study on the human health risk assessment of environmental radionuclides and heavy metals in the study area has been conducted.

1.7 Research Aim and Objectives

1.7.1 Research Aim

The aim of this study was to carry out a human health risk assessment of environmental radionuclides and heavy metals around a Gold Mining Area in Gauteng Province, South Africa.

1.7.2 Research Objectives

To achieve the above aim, the following were the specific objectives:

- To measure activity concentrations of different natural radionuclides in soil, water, and edible plants around the gold mining area,

- To measure the concentration of heavy metals present in soils, water and edible plants from different locations around the mine,
- To determine the indoor concentrations of radon in dwellings around the mine,
- To estimate the total effective dose from soil, water, plants and radon for individual members of the public due to natural radionuclides from the study area,
- To assess the associated radiation hazards due to natural radionuclides in the study area,
- To estimate the health risk to individual members of the public due to natural radionuclides from the gold mining area using the RESRAD-OFFSITE computer code,
- To estimate the health risk to individual members of the public from the gold mining area as a result of heavy metal contamination using theoretical models, and
- To compare the levels of health risk with those set by regulatory bodies.

1.8 Justification of the Study

The outcomes of this study would provide a baseline for Environmental Impact Assessment (EMI) for the gold mine concerned. This can be used as a reference for compliance by mine authorities. The study is also largely important in creating awareness to the population living around mining areas about the potential health risks presented by radioactivity and heavy metals. The research results are also useful in bridging knowledge gaps by providing a database of experimental data for the scientific community to quantify and improve human risk assessment. The study also assists the South African government through the National Nuclear Regulator (NNR) in developing national safety guidelines on environmental protection in mining areas.

CHAPTER 2: LITERATURE REVIEW

2.1. Radioactivity

After the discovery of radioactivity in 1896 by A. H. Becquerel (Allisy, 1996), the science of radioactivity has been extensively studied. Radioactivity is part of everyday life and is a natural and statistical process that describes the spontaneous transformation of unstable atomic nuclei called parent nuclei into a more stable configuration called daughter nuclei without the effect of physical and chemical condition. In cases where the daughter product is also not stable, the decay process carries on until a daughter nucleus reaches stability (Santawamaitre, 2012). During the transformation process, energy is released by the emission of nuclear particles or waves such as alpha particles, beta particles and gamma rays. Other possible emissions are positrons, neutrons and x-rays. These emissions are collectively called ionizing radiations. The ionizing nature of these radiations is what makes them hazardous to health. Ionizing radiation does not only arise from natural radionuclides but also from man-made sources. The most obvious radiation sources to which all individuals are exposed are radionuclides in the earth's surroundings and the interaction of cosmic rays on the earth's atmosphere (Santawamaitre, 2012).

2.1.1. Types of Radioactive Decay

As mentioned earlier, the most common emissions resulting from radioactive decay are gamma rays, alpha and beta particles. These emissions emanate from atoms (smallest particles of matter) of chemical elements. Atoms consist of negative electrons orbiting around a central massive positively charged nucleus, which contains electrically positive protons and neutral particles called neutrons.

2.1.1.1. Alpha Decay

In alpha decay, the parent atom emits an alpha particle, which is identical to a nucleus of a helium atom. This alpha particle is composed of two protons and two neutrons and is very stable. The alpha particle produced is accompanied by a daughter nucleus that contains two neutrons and two protons less than the parent. It is a process mainly found in proton rich nuclides (Peterson et al., 2007). Alpha decay only occurs in elements with high atomic numbers such as uranium, thorium and radium. The nuclei of these atoms consist of more neutrons than protons, which

make emission of alpha particles possible. These electrically charged helium atoms, can be ejected at very high speeds, approximately 30,000 km/s (speed of light) from the atom at the instant of breakdown, but slowed and stopped by about the thickness of a sheet of paper (~ 0.05 mm), or by about 30 mm of air. Alpha particles cannot penetrate the dead outer layer of the skin, but make a dense ionization trail along their stopping track, thereby producing damage to biological tissue, if emitted inside the body following ingestion or inhalation. Alpha particles are only dangerous if you either breathe or swallow the source of the alpha radiation. An example of an alpha decay involves ^{235}U as shown in Figure 2.1.

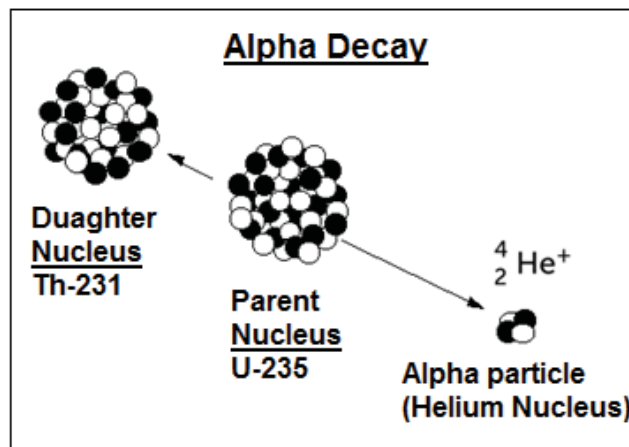


Figure 2.1: Alpha Decay (ISU, 2014)

This decay can be written in the form of a decay equation as follows:



2.1.1.2. Beta Decay

Beta decay can either produce negative particles or positive particles that have the same mass regardless of charge. Beta negative decay is a radioactive process, which emits an electron as a result of a radioactive atomic nucleus, along with an unusual particle called an antineutrino (${}^0_0\bar{\nu}$) (ISU, 2011). The electron formed is called a beta particle emitted by the conversion of a neutron into a proton. In the beta negative decay (β^-), the neutron becomes a proton, which stays in the nucleus and

an electron that escapes. Since the number of protons in the nucleus has changed, a daughter atom is formed with one less neutron but one more proton than the parent. To ensure the beta particle is not just any other, but coming from inside the nucleus, a (β^-) symbol is used. Figure 2.2 illustrates Beta negative decay.

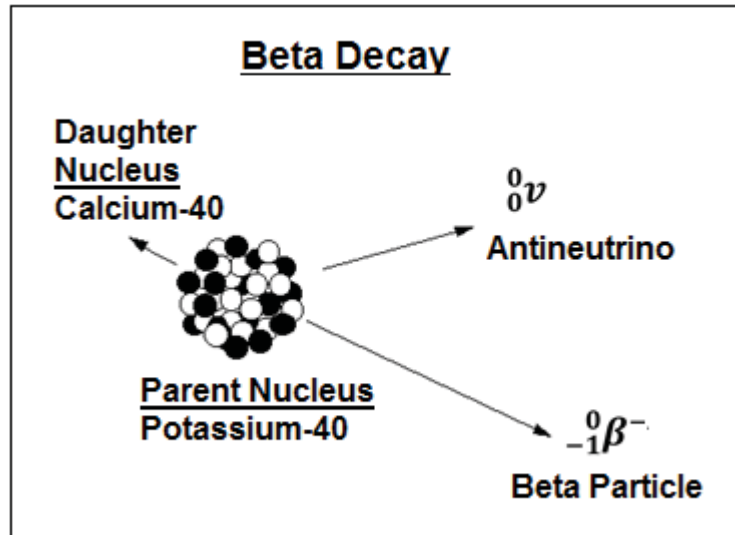


Figure 2.2: Beta negative decay (ISU, 2011)

This beta negative decay can be written in the form of a decay equation as follows:



Another way that makes a nucleus to reach stability is through the emission of a positively charged particle, called positron. A positron is equivalent to the anti-matter of the electron, which is sometimes called an antielectron. In this process, called beta-plus decay, the exact opposite of beta negative decay happens. The process involves a proton decaying into a positron and a neutron, while also emitting a neutrino (ν) for conservation of momentum (Peterson et al., 2007). Positrons have the same mass as an electron, but their charge is $+1e$. The symbol (β^+) is used to represent positrons.

An example of beta positive decay equations is as follows:



Another alternative process to positron decay is **electron capture (EC)**, which can take place when the transformation energy is insufficient to create an electron pair (Santawamaitre, 2012). This process is a mechanism in which an atomic electron, usually in K shell, is captured by a proton-rich unstable nucleus. The combination of a proton and an atomic orbital electron yields a neutron within the nucleus. The electron capture process can be shown by Equation 2.4 as follows;



Beta particles, light alpha particles, are emitted by the atom at nearly the speed of light. They can travel about 0.50 m through the air and about 10 mm into the body, and are stopped by thin layers of metal or plastic. Beta particles do not ionize as easily as alpha particles, but are a bit more of a risk. They are most dangerous if the source is ingested.

2.1.1.3. Gamma Decay

Another type of decay that leads to the formation of gamma rays is gamma decay. When alpha and beta decay occur, they form a nucleus that has more energy than usual and this surplus energy is lost by the emission of gamma rays. This radiation is characterized by a long range emission since it will be neutral. Typical energies range from a few keV to 3000 keV. Figure 2.3 illustrates gamma decay (Peterson et al., 2007).

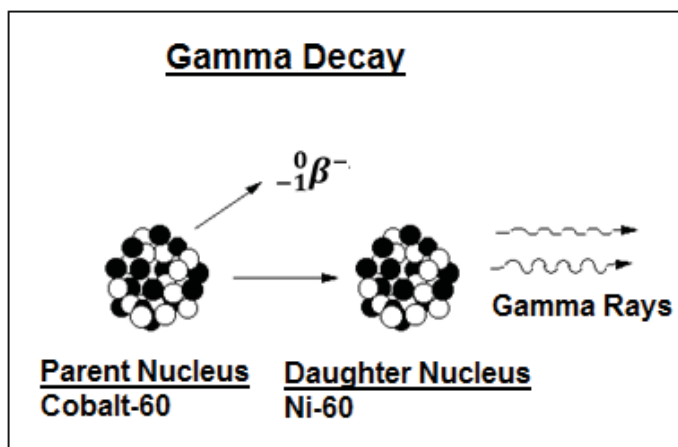


Figure 2.3: Gamma Decay (ISU, 2011)

Gamma rays, just like x rays can easily penetrate the human body, and can only be stopped by high density materials such as lead. Although they do not ionize much, they cause the most damage to a person. Even being near an unshielded source of gamma rays for a short period of time is very dangerous. Gamma and x rays are both part of the electromagnetic spectrum. While they may have almost similar ionizing properties, they differ in terms of their origin. Gamma rays originate from the nucleus while x rays originate from orbiting electrons.

2.1.2. Radioactive Decay Law and Equilibrium

The decay of radioactive material is random in nature and is impossible to estimate exactly when a particular atom is going to decay. However, if a large number of atoms are looked at as a single unit, the decay will follow a well-defined statistical pattern, known as the radioactive decay law. The decay rate is proportional to the number of radioactive nuclei of a particular type present at any time, t . The constant of proportionality (λ), termed the decay constant, is the fraction of the atoms which undergoes decay in a unit time such as a second or a year. It is related to the half-life ($T_{1/2}$) of a radionuclide which is the time required for the decay of one half of the original number of its nuclei present (Gruber, 2009).

In radiation protection, it is more important to know how much radiation is being emitted rather than how many radioactive atoms remain in the sample. Hence a quantity known as activity (A) is used to define the number of disintegrations of the radioactive material taking place in a unit time. Note that activity and atomic mass number have the same symbol (A) and number of radioactive atoms and number of neutrons have the same symbol (N). The activity of a number of radioactive atoms (N) is $N\lambda$. The rate of depletion is equal to the activity as long as there is no new supply of radioactive atoms (Equations 2.5 – 2.8) (Ivanovich and Harmon, 1982).

$$\frac{dN}{dt} = -N \cdot \lambda = A \quad (2.5)$$

$$N_t = N_0 \cdot e^{-\lambda \cdot t} \quad (2.6)$$

$$A_t = A_0 \cdot e^{-\lambda \cdot t} \quad (2.7)$$

$$\lambda = \frac{\ln 2}{T_{1/2}} \quad (2.8)$$

N_0 is the number of radioactive atoms at time $t = 0$; N_t is the number of remaining radioactive atoms at some later time t .

The unit of the activity (A) is Becquerel (Bq), named after the French physicist Henri Becquerel (1852–1908), one of the discoverers of radioactivity. One Becquerel is one transformation per second on average (1 Bq = one decay per second). The former unit was Curie (Ci), named after Marie Curie (1867–1934) and Pierre Curie (1859–1906), who discovered radium in 1898. $1 \text{ Ci} = 3.7 \times 10^{10} \text{ Bq}$, which corresponds to the activity of 1 g of ^{226}Ra . The activity concentration is related to mass and has units (Bq/kg). For liquids or gases the activity concentration is usually related to volume and has units (Bq/l or Bq/m³).

The radioactive decay law mentioned (Equations 2.5 - 2.8) takes into account only the decay of one single, separate nuclide transforming into a stable daughter nuclide. Very often the daughter product of a nuclear decay is also radioactive. This is termed radioactive decay chain, and the decay and the appearance of the daughter have to be considered together as shown in Equation 2.11.

$$\frac{dN_i}{dt} = \lambda_{i-1} \cdot N_{i-1} - \lambda_i \cdot N_i \quad (2.15)$$

N_i is radioactive nuclei of the nuclide i at time t

λ_i is the decay constant of the nuclide i

After some intermediate steps for the mother-daughter-transformation, it follows that (Stolz, 2005):

$$A_2 = \frac{\lambda_2}{\lambda_2 - \lambda_1} \cdot A_{10} \cdot (e^{-\lambda_1 \cdot t} - e^{-\lambda_2 \cdot t}) = \frac{T_1}{T_1 - T_2} \cdot A_{10} \cdot (e^{-\frac{\ln 2}{T_1} \cdot t} - e^{-\frac{\ln 2}{T_2} \cdot t}) \quad (2.16)$$

For multiple nuclides, Bateman equation can be applied (USEPA, 2009a):

$$A_n = A_{10} \cdot T_1 \cdot (K_1 \cdot e^{-\frac{\ln 2}{T_1} \cdot t} + K_2 \cdot e^{-\frac{\ln 2}{T_2} \cdot t} + \dots + K_n \cdot e^{-\frac{\ln 2}{T_n} \cdot t}) \quad (2.17)$$

$$K_n = \frac{T_n^{n-2}}{(T_n - T_1) \cdot (T_n - T_2) \cdot \dots \cdot (T_n - T_{n-1})} \quad (2.18)$$

Where

A_n = activity of nuclide n

A_{10} = activity of the mother-nuclide at time $t=0$

$\lambda_1 = \dots n$ is decay constant of nuclide 1, ... n

$T_1 = \dots n$ is half-life of nuclide 1, ... n

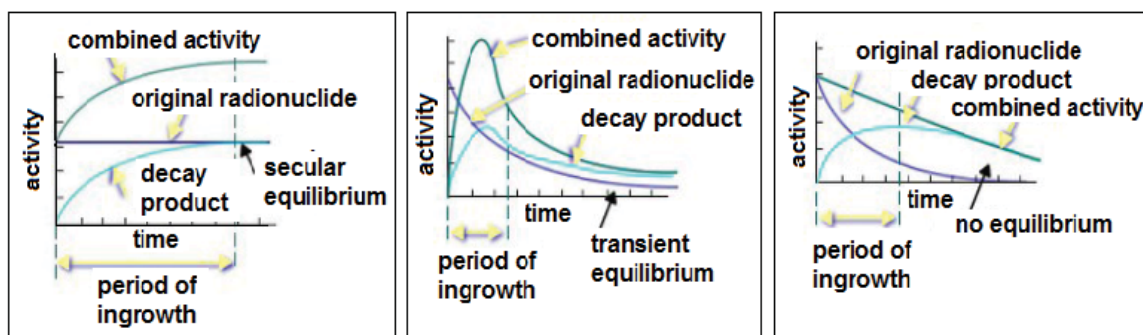
Equation 2.18 shows that the time to reach radioactive equilibrium ($A_1/A_2=\text{const.}$) depends on the half-lives of the parent and the daughter.

Three cases can be distinguished. Secular equilibrium ($T_1 \gg T_2$) arises if the half-life of the parent is much longer than the one of the daughter (progeny) i.e. the decrease of parent activity is negligible over the course of the observation period, e.g. ^{226}Ra ($T_1 = 1600$ years) and ^{222}Rn ($T_2=3.8$ days). It takes a time equal to about seven half-lives of the progeny for the activity of the progeny to be the same as the activity of the parent, i.e. to reach secular equilibrium (Gruber, 2009). Secular equilibrium has important repercussions in radiation protection in terms of the total activity of a sample. For example, when secular equilibrium is reached, the activity of the sample will double because of the two radionuclides decaying simultaneously. In some cases, secular equilibrium occurs at a number of places along the decay chain and this can greatly increase the total activity of a sample (See Figure 2.4 (a)).

Transient equilibrium ($T_1 \geq T_2$) occurs if the half-life of the daughter is of the same order but smaller than that of the parent. An example of this is in the thorium decay series. In this decay chain, ^{212}Pb ($T_{1/2} = 10.64$ h) decays to ^{212}Bi ($T_{1/2} = 1.009$ h) which decays to ^{212}Po ($T_{1/2} = 0.298$ μs) and then to stable ^{208}Pb . Obviously, there can be no progeny if the parent has not decayed. So if the parent is initially pure, the progeny activity starts from zero. As the parent decays, the progeny will start decaying at a slower rate. Hence, the activity of the progeny will build up in the sample until it reaches a maximum. After this point, the progeny seems to decay with the same half-life as the parent. Figure 2.4 (b) illustrates transient equilibrium (Gruber, 2009).

Whenever the parent nuclide has a much shorter half-life than its progeny ($T_1 < T_2$), equilibrium is not possible. After several of the parent's half-lives there is no significant parent activity remaining and the only radionuclide left is the progeny. The daughter activity grows to a maximum and then decays with its own characteristic

half-life, e.g. Cesium-146 (^{146}Cs) ($T_1 = 13.5$ months) and Praseodymium-146 (^{146}Pr) ($T_2 = 24.2$ months) (See Figure 2.4 (c)).



(a) secular equilibrium (b) transient equilibrium (c) no equilibrium

Figure 2.4: Three cases of radioactive equilibrium (USEPA, 2009a)

2.2. Important Properties of Naturally Occurring Radioactive Materials

Naturally Occurring Radioactive Materials decay to form other nuclides, which also decay to other nuclides forming a decay chain. The final atom results in a stable nuclide that is absent of radioactivity. The decay chain of ^{238}U contains several radioactive isotopes, before ending with a stable isotope of ^{206}Pb , while the decay chain of ^{235}U ends with a stable isotope of ^{207}Pb . The decay chain of ^{232}Th also contains several radioactive isotopes, before ending in another stable isotope of ^{208}Pb (Long et al., 2012). These three, ^{238}U , ^{235}U and ^{232}Th decay chains, are summarised in Tables 2.1, 2.2 and 2.3 respectively.

Natural potassium comprises three isotopes (^{39}K , ^{40}K and ^{41}K) where ^{40}K is the only radioactive isotope with a natural isotopic abundance of 0.0118 %. The beta and electron capture decay modes of ^{40}K to stable ^{40}Ca (89.28 %) and ^{40}Ar (10.72 %), respectively, is followed by the emission of 1460.8 keV gamma rays that contributes significantly to natural radioactivity (Xhixha, 2010). Potassium is soluble under most conditions and during weathering is lost into solution. It is also important to realise that the major part of exposure from primordial radionuclides is caused by radon (^{222}Rn) and to a lesser extent, thoron (^{220}Rn). Radon and thoron are gases that form part of the uranium and thorium decay chains. Inhalation of radon and its progenies is the single biggest source of radiation exposure in the environment.

Table 2.1: Properties of Radionuclides of the ^{238}U Decay Series (Wahl, 2007).

Parent Nuclide	Half-Life $t_{1/2}$	Decay Mode (%branch)	Decay Energy (MeV)	% Intensity	Daughter Nuclide	γ -emission Energy (keV)	% γ -emission Intensity
^{238}U	4.5×10^9 y	α (100)	4.198	79.0	^{234}Th	49.55	0.063
			4.151	20.9		113.50	0.0102
^{234}Th	24.1 d	β (100)	0.199	70.3	$^{234\text{m}}\text{Pa}$	63.28	4.1
			0.104	19.2		92.37	2.4
			0.103	7.6		92.79	2.39
$^{234\text{m}}\text{Pa}$	1.17 m	β (99.84)	2.269	98.2	^{234}U	1001.03	0.837
			1.224	1.007		766.38	0.294
		IT (0.16)		*	^{234}Pa	73.92	*
^{234}Pa	6.70 h	β (100)	0.642	19.4	^{234}U	131.30	0.029
			0.472	33.0		946.00	0.021
^{234}U	2.5×10^5 y	α (100)	4.7746	71.38	^{230}Th	53.20	0.123
			4.7224	28.42		120.90	0.0342
^{230}Th	7.5×10^4 y	α (100)	4.6870	76.3	^{226}Ra	67.672	0.343
			4.6205	23.4		143.872	0.0483
^{226}Ra	1600 y	α (100)	4.7843	94.45	^{222}Rn	186.21	3.59
			4.601	5.55		262.27	0.0050
^{222}Rn	3.82 d	α (100)	5.4894	99.92	^{218}Po	511.00	0.076
^{218}Po	3.10 m	α (99.98)	6.0024	100.0	^{214}Pb	**	
		β (0.02)		*	^{218}At		
^{218}At	1.60 s	α (100)	6.0024	100.0	^{214}Bi	*	
^{214}Pb	26.8 m	β (100)	0.671	48.9	^{214}Bi	351.93	35.1
			0.728	42.2		295.22	18.2
			1.023	6.3		241.99	7.12
^{214}Bi	19.9 m	β (99.98)	3.272	18.2	^{214}Po	609.31	44.6
			1.542	17.8		1764.50	15.1
			1.507	17.02		1120.29	14.7
		α (0.02)	5.452	53.9	^{210}Tl	1238.11	5.78
			5.516	39.2		2204.21	4.98
^{214}Po	164.3 μs	α (100)	7.6868	99.99	^{210}Pb	799.7	0.0104
^{210}Tl	1.30 m	β (100)	4.209	30.0	^{210}Pb	*	
			1.863	24.0			
^{210}Pb	22.3 y	β (100)	0.0166	84.0	^{210}Bi	46.54	4.25
			0.0631	16.0			
^{210}Bi	5.013 d	β (100)	1.1615	100.0	^{210}Po	**	
^{210}Po	138.4 d	α (100)	5.3043	99.99	^{206}Pb	803.10	0.00122
^{206}Pb	Stable end product						

* No data

** No γ rays observed

Table 2.2: Properties of Radionuclides of the ^{235}U Decay Series (Wahl, 2007).

Parent Nuclide	Half-Life $t_{1/2}$	Decay Mode (%branch)	Decay Energy (MeV)	% Intensity	Daughter Nuclide	γ -emission Energy (keV)	% γ -emission Intensity
^{235}U	$7 \times 10^8 \text{ y}$	α (100)	4.3978	55.0	^{231}Th	185.72	57.2
			4.3661	17.0		143.76	10.96
			4.2147	5.7		163.36	5.08
^{231}Th	25.52 h	β (100)	0.2881	40.0	^{231}Pa	25.65	14.5
			0.3053	33.0		84.22	6.6
			0.2060	12.8		89.94	0.94
^{231}Pa	$3.3 \times 10^4 \text{ y}$	α (100)	5.0138	25.4	^{227}Ac	27.36	10.3
			4.9513	22.8		300.07	2.47
			5.0284	20.0		302.65	2.87
^{227}Ac	21.773 y	β (98.62)	0.0448	53.0	^{227}Th	**	
			0.0355	35.0			
			0.0203	10.0			
		α (1.38)	4.9533	47.7	^{227}Fr	99.6	0.0056
			4.9407	39.6		160.49	0.0049
^{227}Th	18.72 d	α (100)	4.8727	6.3	^{223}Ra	69.28	0.0043
			6.0380	24.2		235.97	12.3
			5.9777	23.5		50.13	8.0
^{227}Fr	21.8 m	β (100)	5.7569	20.4	^{223}Ra	256.25	7.0
			1.0990	70.0		50.13	0.497
			1.0094	16.0		79.72	0.126
^{223}Ra	11.435 d	α (100)	0.9143	10.1	^{219}Rn	234.81	0.0414
			5.7162	52.6		269.46	13.7
			5.6067	25.7		154.21	5.62
^{219}Rn	3.96 s	α (100)	5.7470	9.2	^{215}Po	323.87	3.93
			6.8191	79.4		271.23	10.8
			6.5526	12.9		401.81	6.4
^{215}Po	1.781 ms	α (100)	6.4250	7.5	^{211}Pb	130.59	0.12
			7.3862	100.0		438.8	0.04
			1.379	91.3	^{211}Bi	404.85	3.78
^{211}Pb	36.1 m	β (100)	0.547	6.32		832.01	3.52
			0.974	1.54		427.09	1.76
^{211}Bi	2.14 m	α (99.72)	6.6229	83.77	^{211}Tl	351.06	12.91
			6.2782	16.23			
		β (0.28)	0.584	100.0	^{211}Po	**	
^{211}Po	0.516 s	α (100)	7.4503	98.89	^{207}Pb	897.80	0.561
^{211}Tl	4.77 m	β (100)	1.427	99.732	^{207}Pb	897.90	0.260
^{207}Pb	Stable end product						

* No data

** No γ rays observed

Table 2.3: Properties of Radionuclides of the ^{232}Th Decay Series (Wahl, 2007).

Parent Nuclide	Half-Life $t_{1/2}$	Decay Mode (%branch)	Decay Energy (MeV)	% Intensity	Daughter Nuclide	γ -emission Energy (keV)	% γ -emission Intensity
^{232}Th	1.4×10^{10} y	α (100)	4.0123	78.2	^{228}Ra	63.81	0.263
			3.9472	21.7		140.88	0.021
^{228}Ra	5.75 y	β (100)	0.0392	40.0	^{228}Ac	13.52	1.6
			0.0128	30.0		16.24	0.72
			0.0257	20.0		12.75	0.30
^{228}Ac	6.15 h	β (100)	1.158	29.9	^{228}Th	911.20	25.8
			1.731	11.66		968.97	15.8
			2.069	8.0		338.32	11.27
^{228}Th	1.9116 y	α (100)	5.4232	72.2	^{224}Ra	84.383	1.22
			5.3404	27.2		215.98	0.254
^{224}Ra	3.66 d	α (100)	5.6854	94.92	^{220}Rn	240.99	4.10
			5.4486	5.06		292.70	0.0062
^{220}Rn	55.6 s	α (100)	6.2881	99.87	^{216}Po	549.76	0.114
^{216}Po	0.145 s	α (100)	6.7783	100.0	^{212}Pb	804.9	0.0019
^{212}Pb	10.64 h	β (100)	0.335	82.5	^{212}Bi	238.63	43.3
			0.574	12.3		300.09	3.28
			0.159	5.17		115.18	0.592
^{212}Bi	60.55 m	β (64.06)	2.246	86.57	^{212}Po	727.33	6.58
			1.521	6.81		1620.50	1.49
			0.627	2.92		785.37	1.102
		α (35.94)	6.0508	69.19	^{208}Tl	39.86	1.06
			6.0899	27.12		288.20	0.337
			5.7675	1.78		452.98	0.363
^{212}Po	0.299 μs	α (100)	8.7849	100.0	^{208}Pb	**	
^{208}Tl	3.053 m	β (100)	1.796	48.7	^{208}Pb	2614.53	35.64
			1.286	24.5		583.19	30.4
			1.519	21.8		510.77	8.13
^{208}Pb	Stable end product						

* No data

** No γ rays observed

2.3. Interaction of Radiation with Matter

Matter is made up of atoms with a central nucleus that contains positively charged particles (protons) and particles that have no charge (neutrons). Atoms also contain negatively charged particles (electrons) that are in orbital motion around the nucleus. Usually, the numbers of electrons and protons inside the atom are equal. Radiation on the other hand comes in three forms, namely; charged particles (alphas and betas), neutral particles (neutrons), and photons (gammas and X-rays). When any one of these forms interacts with atoms, orbital electrons may receive extra energy that allow some of them to break free and leave the atom. When this happens, the atom becomes a positive ion while the electron which left the atom is referred to as a negative ion forming an ion pair. This process is called ionization (IAEA, 2014).

Alpha particles and all other relatively heavy and charged particles are strongly ionizing. Beta particles on the other hand cause less ionization compared to alpha particles. Gamma-rays and X-rays produce the fewest ions as they traverse through matter and are therefore considered weakly ionizing. Neutrons are a special case as they do not cause direct ionization. However, the products of their interactions with matter can range from weakly to strongly ionizing. When radiation transfers energy to the atom not enough to cause ionization, excitation is said to have occurred. As a result the inner electron of an atom may move to a higher energy level but not enough to leave the atom completely. The energy gained by the absorbing atom would eventually be released as electromagnetic radiation (IAEA, 2014).

2.3.1. Alpha Interactions

Alpha particles are relatively heavy, consisting of four nuclear particles and two positive charges. They travel at about 1/20 of the speed of light. As they move through tissue, they react strongly with matter, producing large numbers of ions. As a result, they are not very penetrating. For example, an alpha particle with an energy of 5 MeV will only travel about 3.6 cm in air and will only penetrate a piece of paper (IAEA, 2014). The same energy alpha particle would travel approximately 4 μm in tissue (IAEA, 2014). Alpha particles are the least penetrating of all radiations.

2.3.2. Beta Interactions

Beta particles are much lighter than alpha particles. They have a single negative charge and travel about the speed of light. Consequently, they interact less strongly than alpha particles for the same energy. A beta particle may produce 50 to 150 ion pairs per centimeter of air before its kinetic energy is completely dissipated. For example, a beta particle with energy 3.5 MeV will travel about 11 m in air and 17 mm in tissue (IAEA, 2014). If higher energy beta particles travel close to positively charged nuclei of absorbing atoms, they get deflected due to attractive forces they experience, thus losing energy in the form of X-rays. This form of radiation is known as bremsstrahlung (braking radiation). Interactions of beta particles with low atomic number (Z) materials produce the least bremsstrahlung whereas high Z materials produce the most (IAEA, 2014).

2.3.3. Gamma and X-ray Interactions

Alpha and beta particles continue to lose their energies until all of their energy is transferred to the absorbing medium. On the other hand, the energy for X-rays and gamma rays cannot be completely absorbed as they travel long distances. Their energies can only reduce in intensity. The interaction of gamma and x-rays with matter involves several distinct processes. These are Photoelectric effect, Compton scattering and Pair production (IAEA, 2014).

2.3.3.1. Photoelectric Effect

In the photoelectric absorption process, a gamma ray (photon) undergoes an interaction with an absorber atom and completely disappears. In its place, an energetic photoelectron is ejected from one of the bound shells of the atom causing ionization and excitation. The ejected photoelectron has kinetic energy equivalent to the initial photon minus the binding energy of the electron. Thus, the energy of the photoelectron is given by:

$$E_e = h\nu - E_b \quad (2.19)$$

where ν is the frequency of the incoming photon and E_b is the binding energy of the electron (Howley, 2003).

This process tends to dominate for gamma rays less than 100 keV. In many instances, photoelectrons originate from the innermost shells such as K shells. The vacancy created by a photoelectron is usually filled by a free electron from a higher energy shell. In this process, energy is released in the form of characteristic x-rays. In rare instances, these characteristic x-rays may interact with outer shell electrons, thereby ejecting them from the atom in the form of low energy auger electrons. Photoelectric effect is most likely to occur in high Z materials such as lead (IAEA, 2014).

2.3.3.2. Compton Scattering

Compton scattering occurs between the incident gamma photon and an outer shell electron in the absorbing material. It is the most dominant interaction in soft tissues in the energy range 0.2 MeV to 5.0 MeV. It is also common in absorbers with high Z values (IAEA, 2014).

In compton scattering, the incoming photon is deflected at an angle with respect to its incident direction. The photon transfers part of its energy to the electron (assumed to be at rest initially), which is then known as a recoil electron or a compton electron (Howley, 2003). This electron will continue to interact with the absorbing medium causing further ionization and excitation, but with reduced energy. Mathematically, if the incident photon has a frequency of ν and is scattered through an angle θ , then it can be shown using conservation of energy and momentum that the frequency of the scattered photon ν' , (Howley, 2003) is given by:

$$h\nu' = \frac{h\nu}{1 + \frac{h\nu}{m_0c^2}(1 - \cos\theta)} \quad (2.20)$$

Where m_0 is the rest mass of the electron. Thus the kinetic energy transferred to the electron is:

$$E_e = h\nu - h\nu' = h\nu \left[\frac{\frac{h\nu}{m_0c^2}(1 - \cos\theta)}{1 + \frac{h\nu}{m_0c^2}(1 - \cos\theta)} \right] \quad (2.21)$$

It can be easily shown that the maximum energy that can be transferred to an electron occurs at an angle $\theta = \pi$. This maximum energy corresponds to the

“Compton edge”. (Howley, 2003) By evaluating equation 2.21 at $\theta = \pi$, we find that this maximum energy is given by:

$$E_e|_{\theta=\pi} = hv \left[\frac{2hv/m_0c^2}{1+2hv/m_0c^2} \right] \quad (2.22)$$

And the resulting energy of the photon becomes:

$$hv'|_{\theta=\pi} = hv \left[\frac{hv}{1+2\frac{hv}{m_0c^2}} \right] \quad (2.23)$$

Since photons can be scattered up, through an angle of π , compton scattering gives rise to a continuous energy distribution known as the compton continuum.

2.3.3.3. Pair Production

If a photon enters matter with energy in excess of 1.02 MeV, it may interact by a process called pair production. When a photon passes near the nucleus of an atom, it is subjected to strong field effects from the nucleus, which may cause it to disappear and reappear in the form of electron and positron pair. The two electrons produced e^- and e^+ , are not scattered orbital electrons, but are created in the energy/mass conversion of the disappearing photon. Both the electron and the positron travel very short distances before losing their kinetic energy to the absorbing atom in a process called annihilation. A pair of two annihilation photons, of energy 0.51 MeV each, are consequently produced in this process (IAEA, 2014). These photons are emitted in opposite directions as shown in Figure 2.5.

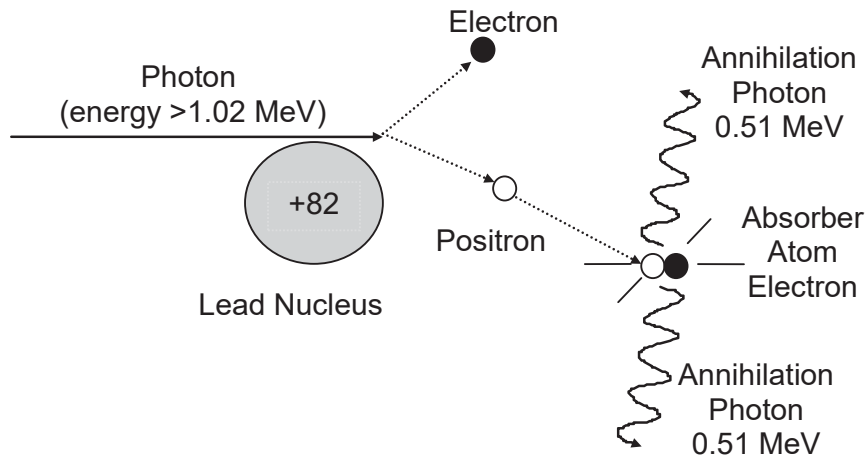


Figure 2.5: Pair production and Annihilation

The spike shown in Figure 2.5 occurs at a kinetic energy corresponding to the initial energy of the gamma ray minus the rest mass of the two particles (Howley, 2003):

$$E_{pair} = h\nu - 2m_0c^2 \quad (2.24)$$

There is a probability that pair production increases with both the atomic number (Z) of the absorber and the photon energy (Howley, 2003). The effect is slowly pronounced from 1.02 MeV to 5 MeV energies and becomes more rapid above that (IAEA, 2014). Figure 2.6 shows how the probability of each interaction depends on the energy of the incident photon and the atomic number of the absorber.

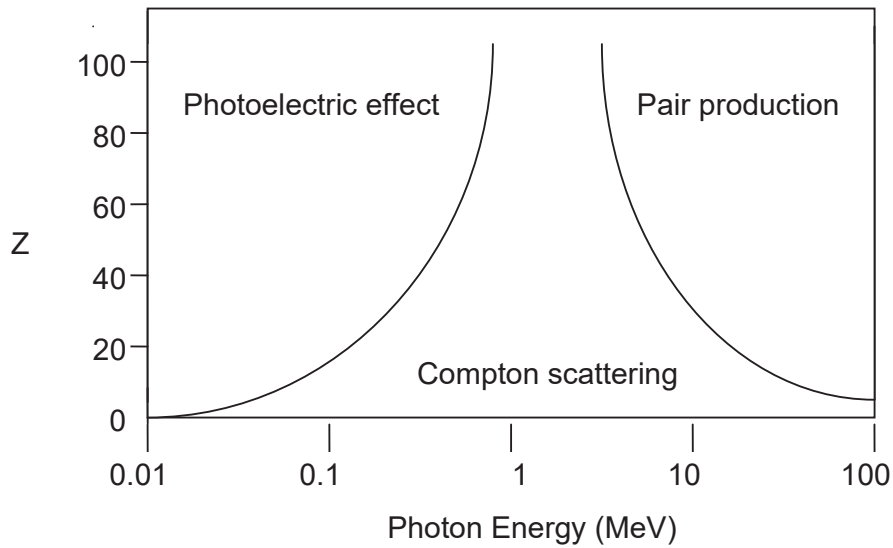


Figure 2.6: Three Major Types of Photon Interactions

2.3.4. Neutron Interactions

Neutrons are atomic particles found in the nucleus that interact with matter either by fission or nuclear reactions (Knoll, 2010). Their interactions depend on the energy of the neutron and the type of nucleus with which they collide. There are basically two forms of collisions through which neutrons interact with matter. These are through scatter and through absorption (IAEA, 2014). In scatter, energy from the neutron is transferred to the recoiling nucleus while in absorption, the neutron is captured by the nucleus. Scattering is also divided into elastic and inelastic scattering. In both processes, the neutron is slowed down as energy is lost in successive collisions. Elastic scattering is the principal mechanism of energy loss for neutrons, whilst in

inelastic scattering, the nucleus is left in an excited state and may later decay with accompanying radiation (Knoll, 2010). Neutron capture on the other hand may lead to the emission of a proton or alpha particle in boron or lithium. Neutron capture may also produce radioactive isotopes in a process called neutron activation or nuclear fission in heavy nuclei such as uranium or plutonium (IAEA, 2014).

2.4. Biological Effects of Ionizing Radiation

The biological damage caused by ionizing radiation has been a known fact for many years. Early human evidence of its harmful effects existed in the 1920s to 1930s (USEPA, 2009). Both empirical observations and epidemiological studies have consistently shown the carcinogenic properties of radiation. Early in this century, individuals who had ingested radium paint, showed increased incidences of bone sarcomas and other malignancies (USFDA, 2006). Skin cancer was noted among early practitioners; dentists and radiologists as a result of handling patients' films. Survivors of the Hiroshima atomic bomb exposed to radiation above 100 rem showed significant increases in incidence of leukaemia (USFDA, 2006). Since the 1500s, it has been discovered that underground miners in the Erzmountains of eastern Europe were susceptible to high mortality from respiratory diseases and lung cancer due to high levels of radon (^{222}Rn) and its decay products (^{218}Po , ^{214}Pb , ^{214}Bi and ^{214}Po) in these mines (USEPA, 2009)

Ionizing radiation interacts with the cell, which is the basic structure of living organisms. There is enough evidence that the DNA molecule is targeted in the cell. There is usually a time lag before the first detectable effect to the cell or tissue after the first radiation event. This is commonly referred to as the latent period. Biological effects can be divided into short term and long term effects on the basis of this. Short term could be minutes, days or weeks while long term effects could be years, decades or generations (USFDA, 2006). Following the latent period, discrete effects, such as cessation of mitosis, breaking of chromosomes, or formation of giant cells may occur. This can be permanent or temporary depending on the severity of the damage. Recovery of cells and tissues may occur particularly in the case of short term effects. However, long term effects on a large number of populations is important from a public health perspective. Long term effects thus observed so far

have been somatic damage that results in increased incidence of cancer, embryological defects, cataracts, life span shortening and genetic mutations, which may consequently affect future generations (USFDA, 2006).

Since energy deposition by ionizing radiation is a random process, even with low doses there is a certain probability in the cell resulting in cell modification or cell killing. Radiation quality, type and its energy are important factors in causing cell damage. If a cell is killed by the radiation there will be no consequences for the irradiated tissue or organism. If, however, the cell is modified, the changes may have serious consequences to the irradiated organism and for future generations that can lead into a tumor cell or mutation in a germ cell observable as hereditary effects (ICRP, 1984). Such effects are called stochastic effects (meaning random or statistical). There is no threshold of dose for stochastic effects. The probability of the cancer increases with dose. At very high doses, there are more cells destroyed and as the dose continues to increase, a whole organ or organism is destroyed. Such effects are deterministic.

The radio-sensitivity of cells and tissues also varies considerably. Eye lens, gonads and lymphocytes usually have high radio-sensitivities. Contrary to this, bones, muscles and nerve cells have low radio-sensitivity (ICRP, 1984). A single accidental exposure to a high dose of radiation for a short period is an acute exposure. When ionizing radiation is delivered to the whole body, signs and symptoms are collectively referred to as Acute Radiation Syndrome. This may produce the following biological effects: skin damage, nausea and vomiting, fatigue, loss of hair, severe diarrhoea, bone marrow damage, damage to cells lining of the small intestine, cardiovascular collapse and damage to blood vessels in the brain (USFDA, 2006). The effect of all this may vary from mild and transient to death.

2.5. Biological Effects of Heavy Metals

Certain metals, such as iron and copper, are important to human life because of their irreplaceable roles in the functioning of body organs. All living organisms require varying amounts of heavy metals, but become toxic at higher concentrations (Lane

and Morel, 2009). However, other metals have no useful role in the human physiology and most other living organisms. Examples of such elements are arsenic, lead and mercury. They may be toxic even at trace levels of exposure. Arsenic, lead and mercury are the first, second and third hazardous, respectively, on the priority list of heavy metal pollutants as designated by the United States Agency for Toxic Substances and Disease Registry (ATSDR, 2007).

Heavy metals from mining operations may contaminate agricultural soils, water supplies and the environment. Excessive accumulation of heavy metals in agricultural soils, may not only contaminate the soil, but also lead to elevated heavy metal uptake by crops (Muchuweti et al., 2006). Heavy metals may also be ingested involuntarily through food and drink. Once absorbed, they are distributed in tissues and organs. Although excretion occurs through kidneys and the digestive tract, they persist in some storage areas, such as the liver, bones, and kidneys, for years or decades causing serious health consequences (Kabata and Pendias, 1993).

Arsenic for instance, is regarded a human carcinogen from extremely low levels of exposure (ATSDR, 2007). After its absorption, it is primarily distributed in the liver, kidneys, lungs, spleen, aorta, and skin. Acute exposure to arsenic compounds can cause nausea, vomiting, abdominal pain, muscle cramps, diarrhea and burning of the mouth and throat (NRC, 1999). Chronic exposure to lower levels of arsenic results in somewhat unusual patterns of skin hyper-pigmentation, peripheral nerve damage manifesting as numbness, tingling, and weakness in the hands and feet as well as causing diabetes (UNEP, 2002).

Lead (Pb) is known to be toxic even at low concentrations and is regarded as a human mutagen and probable carcinogen (Podsiki, 2008). It induces renal tumours, reduces cognitive development, and increases blood pressure in adults. Pb toxicity also causes reduction in haemoglobin synthesis, thereby disturbing the normal functioning of the kidney, joints, reproductive and cardiovascular systems and causes chronic damage to the central and peripheral nervous systems (Ogwuegbu and Muhanga, 2005). Children and developing fetuses appear to be particularly vulnerable to the neurotoxic effects of Pb. Epidemiologic studies have also revealed

that low-level Pb exposure in children under five years of age results in deficits in intellectual development (ATSDR, 2007).

Mercury (Hg) on the other hand, occurs primarily in two forms: organic and inorganic mercury. The acute ingestion of inorganic mercury salts may cause gastrointestinal disorders such as abdominal pain, vomiting, diarrhea, and hemorrhage (UNEP, 2002). Repeated and prolonged exposure results in severe disturbances in the central nervous system as well as gastrointestinal tract, kidneys, liver and skin. It may also lead to dizziness, fatigue, headaches, hearing and memory loss, mood swings, numbness and tingling, pain in limbs, rashes, peripheral vision loss and muscle weakness in the long term (Lokeshappa et al., 2012).

Cadmium (Cd) is also known to be toxic even at low concentrations and is regarded as a probable carcinogen. Severe exposure to Cd may result in pulmonary problems such as emphysema, bronchiolitis and alveolitis (Kabata and Pendias, 1993), Cd may also induce kidney dysfunctions, and reproductive deficiencies. Renal damage may also result due to the sub chronic inhalation of Cd (Young, 2005). Ingestion of Cd-contaminated food may cause bone fracture, kidney malfunction, hypertension or even cancer (Khan et al., 2013). Arthritis, diabetes, anaemia, cardiovascular disease, cirrhosis, reduced fertility, headaches and strokes are some of its odd long term effects.

Chromium (Cr (VI)) compounds are toxic. They are also known to be both mutagenic and carcinogenic whereas Cr (III) is an essential element (Podsiki, 2008). Breathing high levels may cause irritation to the lining of the nose causing breathing problems. Long term exposure may cause damage to liver, kidney, circulatory and nerve disorders, as well as skin irritation. Nickel (Ni) on the other hand is known to be responsible for cancer, oral and intestinal. It also causes depression, heart attacks, haemorrhages and kidney problems (NRC, 1999). Additionally, excessive intake of zinc (Zn) and copper (Cu) may cause non-carcinogenic effects to humans, even though they are essential to life (Cao et al., 2010). Cu surplus had been associated with liver damage while Zn produces adverse nutrient interactions with Cu. Higher concentration of Zn may also cause impairment of growth and reproduction (Nolan, 2003).

2.6. Radiation Detection Instruments

Detection can be performed when a nuclear particle enters a device to produce either excitation or ionization. There are different types of detectors that can be used on the basis of the principles highlighted above. If a nuclear particle causes excitation followed by fluorescent de-excitation when it interacts with a special material, light is emitted that can be recognized by a Scintillation detector. Scintillation detectors are known for their high efficiency in acquisition but have poor resolution (Bode, 1998). Depending on the material from which scintillation takes place, it is possible to manufacture solid, liquid or gas scintillator detectors.

The second type of detectors operate based on ionization, where charge carriers are produced between the charged electrodes contained in the detector. Devices based on this principle can be solid semiconductor materials or gas filled, usually divided into ion chambers, proportional and Geiger-Müller tubes. Solid semiconductor detectors have a p-n junction diode structure. A depletion region is created when a reverse bias voltage is applied across the p-n diode (Gruber, 2009). When photons interact within the depleted region, charge carriers (positive holes and negative electrons) are freed and are swept to their respective collecting electrodes by a voltage supply. The collected charge is fed into a preamplifier and converted to a voltage pulse with the amplitude proportional to the original photon energy.

The most common materials for semiconductor detectors are germanium (Ge) and silicon (Si). Semiconductor detectors are most frequently used for gamma spectrometry because of certain advantages such as high energy resolution, good stability, excellent timing characteristics and simplicity of operation. There are three fundamental differences between Ge and Si detectors; the energy gap, atomic number and mobility of the major carriers. Less energy is needed to create an electron-hole pair in Ge than in Si. Thus, some Si detector may be used at room temperature while Ge detectors have to be cooled to 77 K or -196 °C to reduce the leakage current due to thermal generation of charged carriers to an acceptable level. Therefore a Ge detector has to be operated inside a vacuum chamber and cooled to liquid nitrogen temperatures (Lutz, 2001).

The sensitive detector surfaces are thus protected from moisture and other condensable contaminants. For better efficiency, a gamma detector is manufactured with a high photoelectric cross section material. The photoelectric cross section depends roughly on the fifth power of the atomic number Z , so Ge beats Si by one to two orders of magnitude. (Gruber, 2009). Gamma-spectroscopy is a very important non-destructive method for the measurement of the activity of different radionuclides in environmental samples. Many radioactive sources produce γ -rays of various energies and emission probabilities (Lutz, 2001). These gamma lines are collected in the form of a spectrum and analyzed with a gamma spectrometry system. In this study, the Broad Energy Germanium Gamma Spectrometer was used.

2.6.1. Broad Energy Germanium (BEGe) Spectrometry System

With this instrument, gamma photons are detected by the interaction properties of gamma rays with the absorbing material. Photoelectric effect, Compton scattering and pair production are the three major processes by which gamma rays interact with the detector, thereby producing charge carriers that are moved by the electric field towards the electrodes. The BEGe detector provides the best energy resolution for detection of radiation but with relatively low efficiency (Pallavicini, 2011). Depending on the impurity dopant it is possible to design different kinds of detectors: i.e. n-type: germanium crystals with donor (n-type) impurities or p-type: germanium crystals with acceptor (p-type) impurities.

Broad Energy Germanium detector configurations can be made in various shapes for particular purposes of measurement. The most common configurations of the detector are either planar or coaxial types (Santawamaitre, 2012). Planar type detectors are typically small with a diameter of a few centimetres. This size leads to the limitation of the maximum depletion region to less than 1 or 2 cm. This type of detector is suitable for the measurement of low-energy gamma radiation. However, a thicker depletion layer or a greater sensitive volume of the detector which is required for more general gamma-ray measurements can be made by constructing the detector in another type of cylindrical or coaxial geometry (Knoll, 2010).

A block diagram of a PC based Broad Energy Germanium Gamma Spectrometry System is shown in Figure 2.7. The system consists of a broad energy germanium detector that is coupled to a high vacuum cryostat dewar system. This cryostat dewar system stores liquid nitrogen and also creates a path for heat transfer from the reservoir to the detector element. The detector is always shielded with lead to reduce the background noise due to the presence of natural radioactivity in the surroundings. A High Voltage Power Supply provides the necessary high voltage to the detector and the necessary voltages to the rest of the system components. The voltage can be up to 5000 V. The system also has highly efficient electronics that include a preamplifier that produces a response proportional to the quantity of charge produced by the incident ray. The other element of the electronics is the amplifier whose main function is to amplify the signal. Pulses are properly converted into signals in a way that the ratio between the signal and background noise is maximized. This is the step in which operational decision has to be taken at the user's discretion to adjust parameters like the time unit or gain (Sundareshan, 2013). The last element is the multichannel analyzer (MCA). This is of utmost importance in the experimental measurements. It consists basically of an analogue-to-digital converter (ADC), control logic, memory and display. The MCA collects pulses in all voltage ranges at once and displays this information in real time as final results or as data for further analysis (Reguigui, 2006).

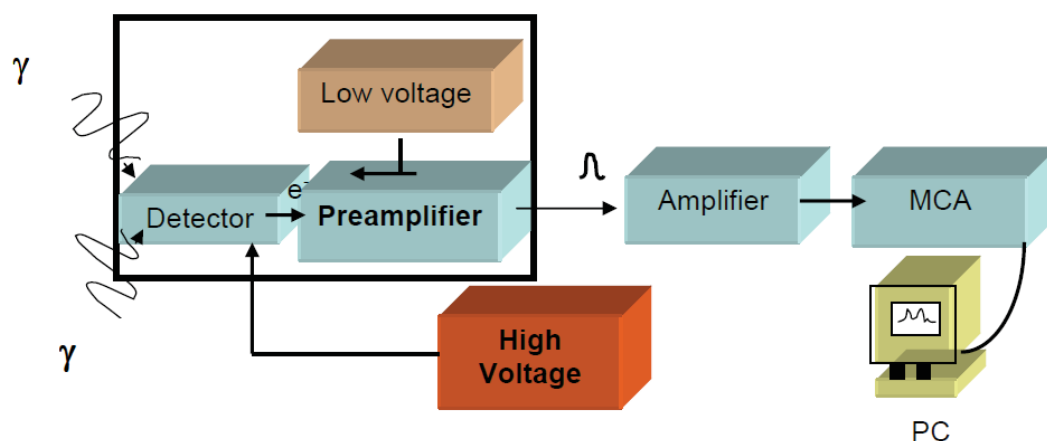


Figure 2.7: Block Diagram of High Purity Germanium Gamma Spectrometry System

2.6.2. The ALphaGuard Professional Radon Monitor

Radon-222 (^{222}Rn) is a noble gas produced by the radioactive decay of ^{226}Ra . It is abundant in uranium-rich soils and rocks and can easily escape into the surrounding water or air (Lawrence et al., 2009). The most common exposure route is through underlying soil gas into buildings. When ^{222}Rn decays, it produces a series of short-lived daughter radioisotopes (Lawrence et al., 2009). Since it is chemically inert, most of the inhaled ^{222}Rn is rapidly exhaled, whereas its daughters may deposit in the airways of the lungs. Two of its daughters, ^{218}Po and ^{214}Po , emit alpha-particles (USEPA, 2009). When this happens in the lungs, the radiation can damage the cells lining the airways, potentially leading to cancer. Nuclear decay of radon products also release energy in the form of beta particles and high energy photons, but the biological damage resulting from these emissions is regarded insignificant compared to the effects of alpha particles (USEPA, 2009).

The ALphaGuard Professional Radon Monitor is a device that measures ^{222}Rn (AlphaGuard, 2007). It is also another example of a detector that uses the ionization principle for the measurement of radon in the environment. This detector works on the principle that as radiation passes through air or gas filled space, ionization of the molecules occurs. Figure 2.8 shows the schematic diagram of a Radon Monitor. This piece of equipment is designed for instantaneous or continuous measurement of radon (^{222}Rn) gas activity. Air, water and soil exhalation measurements are performed using a large range of accessories and external probes. A Germany based company called Genitron Instruments GmbH supplied the instrument (AlphaGuard, 2007).

The device has the active volume of 0.56 dm^3 and the chamber case is held on a potential of +750 V (anode). By diffusion, the radon flows into the ionization chamber via a glass fiber filter that prevents the radon daughters from entering the ionization chamber. The alpha particles produced by radon progeny are the ones that ionize the air in the ionization chamber. When a high voltage is applied between two areas of the air filled space, the positive ions will be attracted to the detector (the cathode) and free electrons to the positive side (the anode). These charges collected by the anode and cathode will then form a very small current along the wires as it flows to

the detector (ISU, 2011). By placing a very sensitive current measuring device between the wires of the cathode and anode, the small current signal is then measured and displayed. The more the radiation that enters the chamber, the more current that can be displayed by the instrument. Data EXPERT software then performs data analysis and storage (AlphaGuard, 2007).

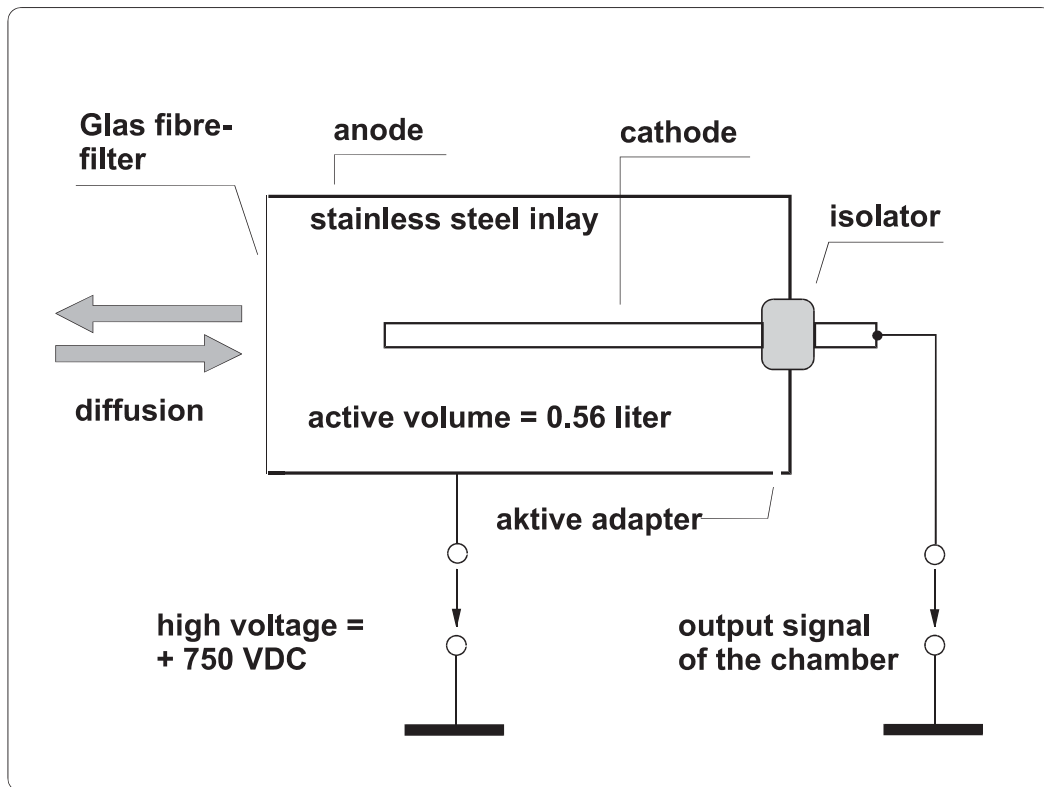


Figure 2.8: Schematic diagram of a commercial ALphaGuard Professional Radon Monitor.

2.7. Heavy Metal Detection Instruments

Heavy metals can be detected using different analytical techniques. Some examples are Neutron Activation Analysis (NAA), Proton Induced X-Ray Emission Spectroscopy (PIXE), X-Ray Fluorescence Spectroscopy (XRF), Inductively Coupled Plasma-Mass Spectrometry (ICP-MS), and others.

NAA is a non-destructive technique involving the irradiation of a sample material with neutrons in a nuclear reactor to perform quantitative analysis of major and trace elements in samples.

PIXE on the other hand is also a non-destructive technique that uses a beam of energetic ions, generated by an accelerator to produce protons in the 2-5 MeV range that creates inner electron shell vacancies. One of the inner-shell electrons such as K- and L-shell from the sample to be measured is ionized to produce an inner-shell vacancy. As these inner shell vacancies are filled by outer shell electrons through electron transition, characteristic X-rays are emitted and detected that identify with particular elements (Itoh et al., 2005).

XRF is a non-destructive technique that produces characteristic gamma rays when electrons from outer shells of an atom fill in shell electron vacancies. The vacancies are created as a result of ionization of the atom by X-rays through knocking off inner shell electrons. Analysis of the produced gamma rays from the sample by their energies, allows identification of heavy metal elements and their concentrations (Nayak et al., 2004)

In this study, ICP-MS was used to analyze for heavy metals in environmental samples. The main reasons for using this technique are the low detection limit, less interference and its ability to carry out rapid multi-elemental analysis.

2.7.1. Inductively Coupled Plasma-Mass Spectrometry (ICP-MS)

An ICP-MS basically consists of the ICP system and the MS system. The ICP system is just for the atomization of the sample in question. It uses a nebulizer, which will atomize the liquid sample into plasma. To achieve this, the liquid sample is mixed with argon that rotates when it comes in contact with the sample. The mass spectrometry (MS) system is mainly for the separation of the different atoms (Jarvis, 2006). Figure 2.9 shows the various components of the ICP-MS system.

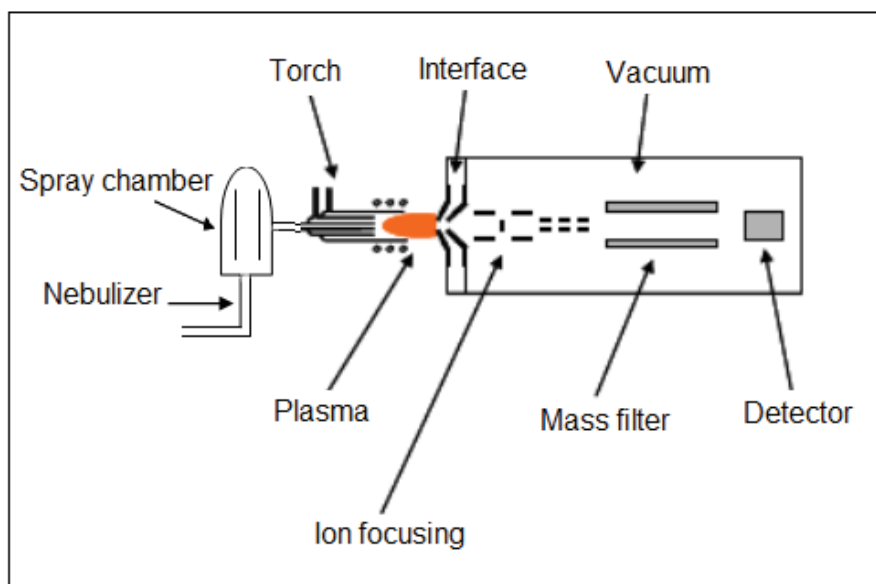


Figure 2.9: Cross section of a quadrupole-ICP-MS instrument (IAEA, 2005).

The first of ICP-MS components is the sample introduction system. This is composed of a nebulizer and spray chamber that provide the means of getting liquid samples into argon plasma as fine aerosol droplets. The high velocity stream of argon gas breaks the liquid sample into small droplets that are injected into the spray chamber by the gas stream. The ICP torch together with the RF coil, is the next component that generates the argon plasma. The plasma creates a very high temperature (6000- 10000 Kelvin) environment for atoms, ions and electrons. At these temperatures many elements make the transition from the atomic to the ionic state since there is sufficient energy to completely remove their outer shell electrons (Jarvis, 2006). Therefore, the plasma dries the aerosol, dissociates the molecules, and then removes electrons from the components, thereby forming singly-charged ions, which are directed into the mass filtering device, the mass spectrometer (PerkinElmer, 2004-2011).

The Interface then links the atmospheric pressure ICP ion source to the high vacuum mass spectrometer. The interface is the region where ions generated in the plasma are extracted and introduced to the mass spectrometer as an ion beam. The ion optics, quadrupole, and detector operate under high vacuum. Once in the high vacuum region of the mass spectrometer, the sampled ions are accelerated and

focused onto the mass filter by a set of charged plates or ion lenses. Ion optics guide the desired ions into the mass analyzer (quadrupole) while making sure that neutral species and photons are discarded from the ion beam (PerkinElmer, 2004-2011). Before the ion beam gets to the mass spectrometer, it passes through the collision/reaction cell that is used to remove interferences which can degrade the detection limits achievable.

The Mass spectrometer acts as a mass filter to sort ions by their mass-to-charge ratios. Most commercial ICP-MS systems employ a quadrupole mass spectrometer which rapidly checks the mass range. At any time, only one mass-to-charge ratio is allowed to pass through the mass spectrometer from the entrance to the exit. The MS system has a magnetic field and an electric field attached perpendicular to each other and these bend the ions as they enter the system. The radius of the arch is dependent on the mass of the ions. The ions are separated based on their mass-to-charge ratios, as they move to the detector. For example, the mass to-charge ratio is 7 for ${}^7\text{Li}^+$, 70 for ${}^{140}\text{Ce}^{++}$, 140 for ${}^{140}\text{Ce}^+$, and 238 for ${}^{238}\text{U}^+$, respectively (University of Wisconsin, 2005).

Upon exiting the mass spectrometer, ions strike the first dynode of an electron multiplier, which serves as a detector. The detector counts individual ions exiting the quadrupole. The impact of the ions produces a stream of electrons, which gets amplified until they become a measureable pulse. The last component is the data handling and system controller. This controls all aspects of instrument control and data handling to obtain final concentration results. It entails the use of software to compare the intensities of the measured pulses with standards that generate a calibration curve, to determine the concentration of the element. The measured ion intensities at different masses are proportional to the concentrations of analytes in the aqueous sample (Gross, 1999).

2.8. RESRAD-OFFSITE Simulation Model

Environmental issues are quite challenging to solve because of the complex relationships among the many variables that exist (David et al., 2004). Modelling, therefore, plays a pivotal role in this regard. One of the most frequently used modelling software is called RESRAD code. RESRAD (short for RESidual RADioactivity) was developed by Argonne National Laboratory under the U.S Department of Energy and the U.S Nuclear Regulatory Commission as a multifunctional tool to assist in developing criteria for evaluating human radiation doses and risks associated with exposure to radiological contamination (Yu et al., 2001). In this evaluation procedure, RESRAD uses U.S. Federal Guidance Report No. 12 (Eckerman and Ryman, 1993) and the ICRP-38 radionuclide database. The radionuclide database also accounts for ingrowth of daughters from initially-present parent radionuclides (Yu et al., 2001). RESRAD allows users to specify the features of their site and to predict the dose received by an individual at any time over the next 100,000 years. It models a site through the use of more than 150 variables. These parameters each have a default value assigned by the developers at Argonne, but can be changed to suit site-specific needs (Smith, 2008).

RESRAD-OFFSITE is an extension of the RESRAD-ONSITE computer code that was developed to estimate the radiological consequences to individuals located onsite or outside the area of primary contamination. The primary contamination is assumed to be a layer of soil and is the source of all releases that is modelled by the code. The code considers the release of radionuclides from the source to the atmosphere, to surface runoff, and to groundwater. It calculates radiation dose and excess cancer risk with the predicted radionuclide concentrations in the environment (USDOE, 2007). Nine exposure pathways are considered in RESRAD-OFFSITE: direct exposure from contamination in soil, inhalation of dust and radon, ingestion of plant foods, ingestion of meat, ingestion of milk, ingestion of aquatic foods, ingestion of water, and incidental ingestion of soil (USDOE, 2007). By selecting the relevant pathways, RESRAD-OFFSITE can be used to model various exposure scenarios such as the resident, farmer, worker or recreationist.

2.9. Environmental Exposure Pathways associated with Radionuclides and Heavy Metals in the Study Area

An exposure pathway describes how contaminants such as radionuclides and heavy metals travel through the environment from their sources to the final receptor such as humans. An exposure pathway consists of five elements (Strand and Dowdall, 2010).

The first one is the source of environmental contamination. Gold mine tailings in the study area constitute a major source of pollution to the environment. In some instances, fissure water is treated in water-purification plants and used for human consumption. Fissure water comes from underground mine workings and can be a source of contamination if it is in contact with oxidized ore. Another source of contamination is process water that is used in mining processes. It usually contains dissolved pollutants, which, when discharged into the environment through evaporation, seepage or run-off, would eventually affect humans. Other sources are rock dumps, return-water dams, storm water drainage systems, settling ponds and evaporation dams.

The second one is the environmental medium. From the source, a contaminant travels through environmental media to points where human exposure can occur. Radionuclides and heavy metals from these sources can either be leached into the underlying ground water aquifers or dissolved and drained through runoff into surface water bodies thereby contaminating water sources.

The major environmental media for humans include water, air, food, and soil (Strand and Dowdall, 2010). Water exposes humans to heavy metals and radionuclides through surface and ground water. Both surface and ground water can be used for drinking, bathing, agricultural or recreational purposes. Through the atmosphere, humans may also be exposed to indoor and outdoor air contaminated with for instance radon gas and windblown dust from mine wastes. Foods grown with contaminated water or grown where the soil is contaminated can also contribute to human exposure. The other environmental medium in the study area is soil.

Contaminated soil blown as dust in the air can be deposited on the surface or on food items that can eventually be consumed or inhaled (Health Canada, 1995).

The third element to the exposure pathway is the point of exposure. This is the location where there is contact with a contaminant. For example, people can be exposed to radionuclides and heavy metals in the home, rivers or at work. The receptor person is the fourth element to the exposure pathway. For example, swimmers may be exposed while bathing in a contaminated river or inhaling radon gas from mine tailings.

The last and fifth element is the exposure route. This describes how a contaminant enters the human body. There are generally three routes by which humans may take radionuclides and heavy metals into their bodies. These are; ingestion, inhalation and dermal contact (Strand and Dowdall, 2010). Ingestion involves swallowing something containing the contaminant. It can be food, water or soil. Inhalation involves breathing in of a substance in gaseous state. Small amounts of soil and dust can be inhaled into the lungs. Dermal contact involves absorption from water, soil or air through the skin.

The other significant exposure route, apart from the three mentioned earlier, is the contribution through direct external irradiation. (Sutton, 2008). This is exposure (from outside the body) to ionizing radiation from the ground or air. Figure 2.13 illustrates some potential exposure pathways for radionuclides and heavy metals which can be regarded as the conceptual model for the study area.

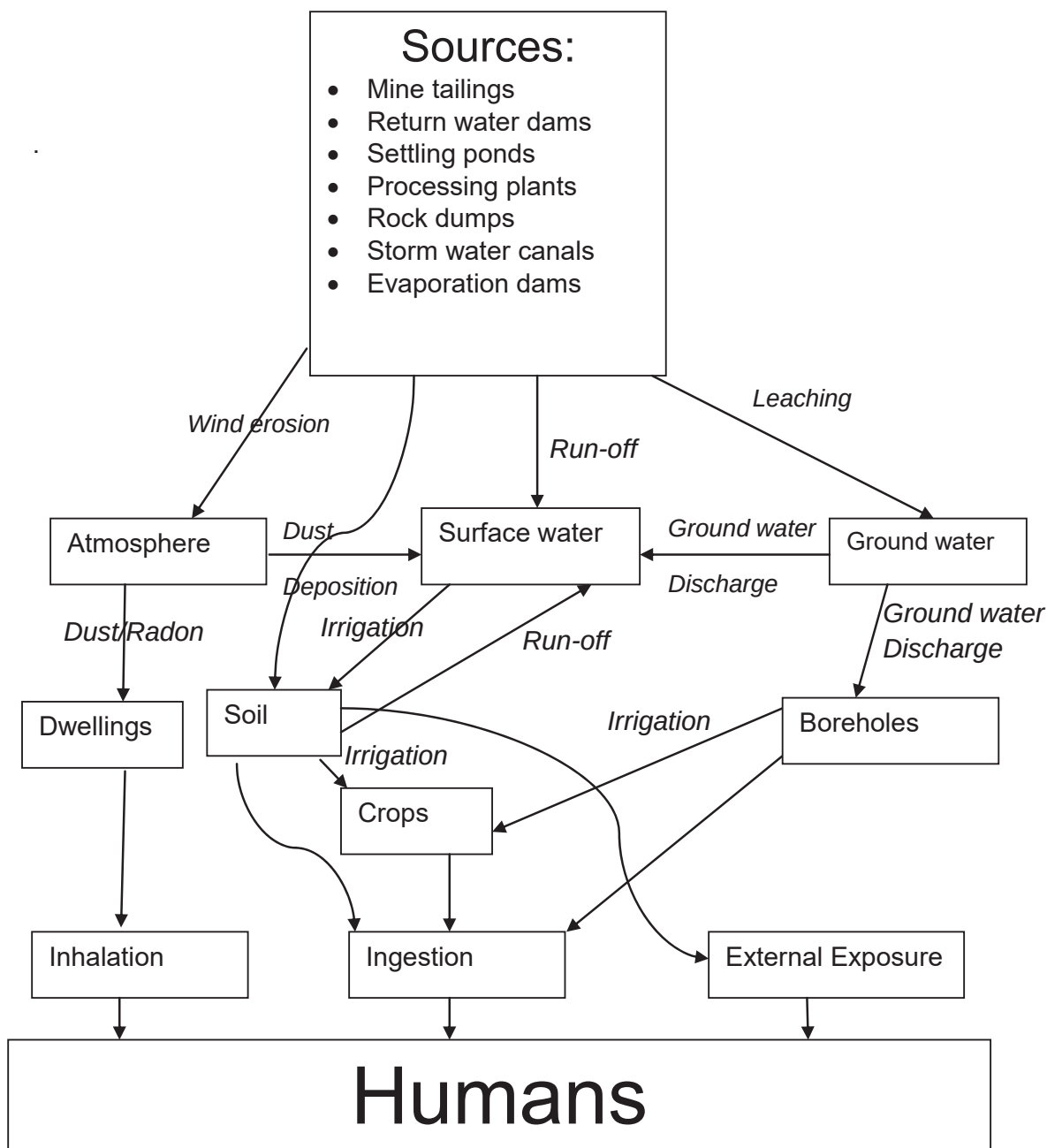


Figure 2.10: Conceptual Model for the Study Area.

CHAPTER 3: EXPERIMENTAL TECHNIQUES

3.1. Introduction

This study was carried out in the Wonderfonteinspruit Catchment Area (WCA) in the Gauteng Province, where there are potentially high levels of radionuclides and heavy metals. Another area, Moshawane Village, in the North West Province of South Africa was chosen as the control. Samples were collected from mine tailings, rock dumps, settling ponds, and from mine villages. A total of 136 samples from different media were collected for laboratory analysis. Natural radionuclides were analyzed at the Centre for Applied Radiation Science and Technology (CARST) and heavy metals were analyzed at the Department of Animal Health, both based at North West University. For heavy metals, the analysis was done using an Inductively Coupled Plasma Mass Spectrometer (ICP-MS) and natural radionuclides were measured by means of Gamma Spectrometry using a Broad Energy Germanium (BEGe) detector. Indoor radon levels from selected dwellings in mine villages were measured using the AlphaGuard Professional Radon Monitor available at CARST.

3.2. The 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 kilometers southwest of Johannesburg. Figure 3.1 shows the study area.

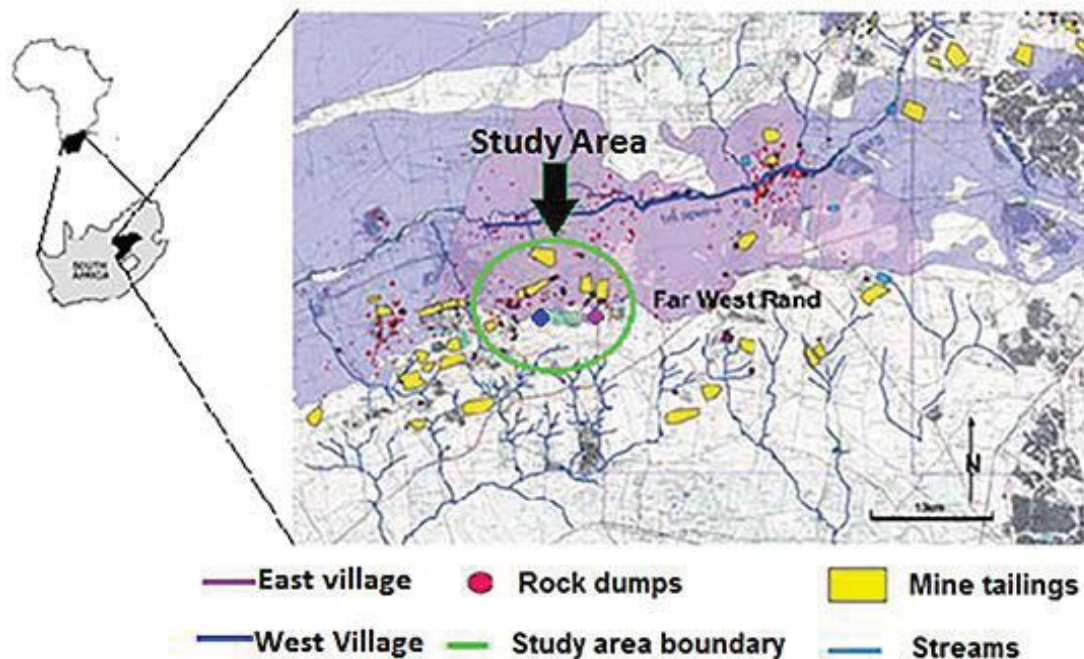


Figure 3.1: The Area of Study- Part of the Wonderfonteinpruit Catchment Area (Kamunda et al., 2016b).

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. Geologically, the area, approximately 86 km², lies in the lower central part of the Wonderfonteinpruit Catchment Area (WCA). The WCA forms part of the Witwatersrand Basin, which is the world largest gold and uranium mining basin that covers an area of 1600 km² (Winde and Stoch, 2010). The basin has led to a legacy of some 400 km² of mine tailings dams (270 tailings dams and 380 mine residue dumps) containing 6 billion tonnes of pyrite tailings and 600 000 tonnes of low-grade uranium (Winde and Stoch, 2010). 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. The area is also densely populated because of the presence of these gold mines.

The study area has several shaft systems with surface reserves represented by rock dumps and mine tailings that have been accumulated throughout the operating history of the mine. In addition, the mine owns extraction and processing facilities, which beneficiate the gold bearing ore. The topography of the area is relatively flat

and the vegetation is largely grassland. Livestock farming is also widespread in the surrounding area. The climate is temperate, with temperatures averaging 24°C in summer and 13°C in winter, occasionally dipping below the freezing point. Annual rainfall is about 750 mm (Tyson and Wilcocks, 1971). The study area has a worker population of around 14,000 (SGL, 2012). This figure is much higher if their families were to be taken into account. There are also some informal settlements residing close to the mine tailings. Figure 3.2 shows the sampling points for soil, water, rock and plant samples that were used.

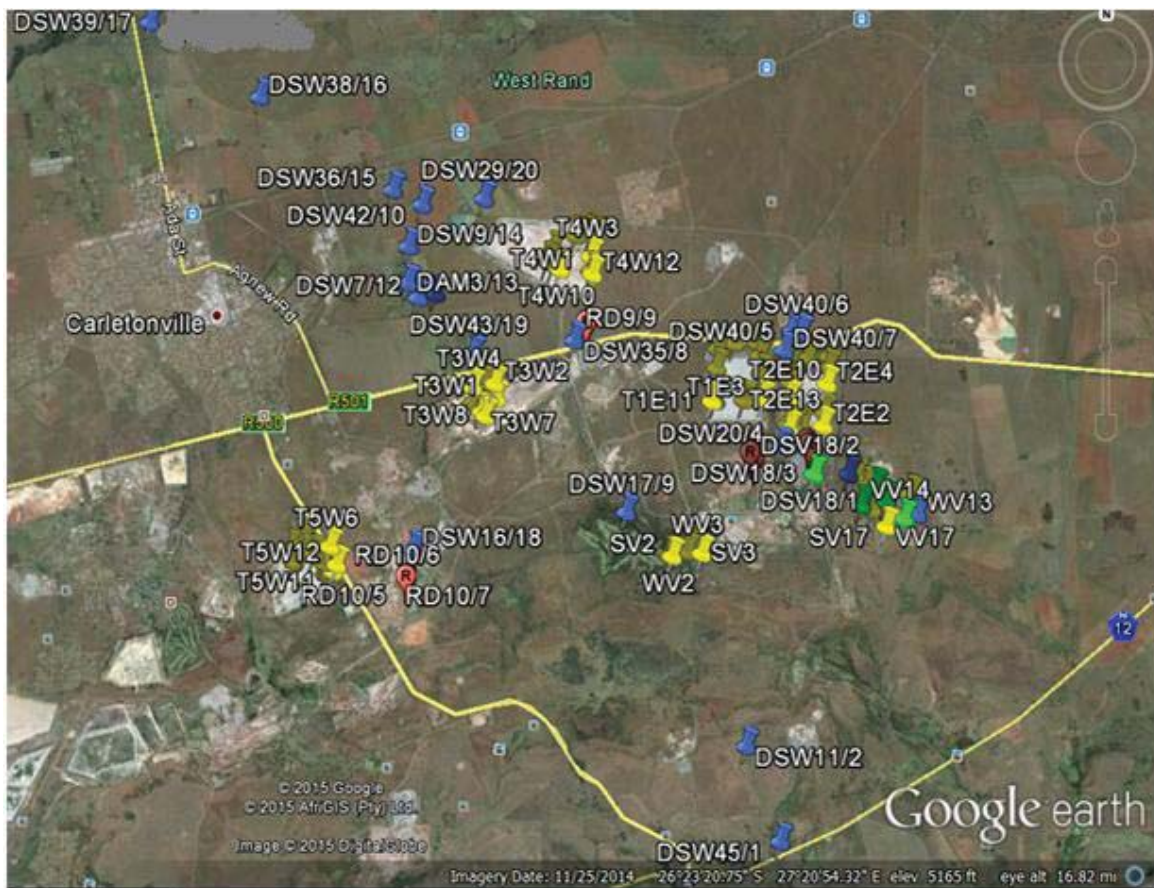


Figure 3.2: Sampling Points for Soil (Yellow), Water (Blue), Rock (Red) and Plant (Green) Samples from Mining Area.

3.3. The Control Area

Moshawane Village, in the North West Province of South Africa, was chosen as the control area as it is geologically similar to the study area. It is situated close to Mafikeng, the provincial Capital City. It is about 10 kilometers south of the Botswana Border and lies between 25°47`S - 25°48`S latitude and 25°37`E - 25°38`E longitude. The overall topography of the area is largely flat with areas of scattered trees and grassland. Figure 3.3 shows the sampling points for soil, water and plant samples from the control area.



Figure 3.3: Sampling Points for Soil (Yellow), Water (Blue) and Plant (Green) Samples from Control Area.

3.4. Sample Collection

From the study area, representative samples of soil, water, and edible plants were collected from different locations using random sampling. All the collected samples were properly marked and identified by their sampling locations using Global Positioning System (GPS) for later referencing. The collected samples were then transported to the laboratory for further processing. Figure 3.4 shows the hand corer and GPS used during soil sampling.



Figure 3.4: Hand Corer and Global Positioning System (GPS) used during Sampling

3.4.1. Soil Sample Collection

A total of 56 soil samples from five mine tailings and 17 from two mine villages representative of the the gold mining area were collected with a hand corer at a depth of 5 cm and packaged in 4 kg polyethylene bags (IAEA, 1989). Similarly 8 samples from rock dumps were also collected and packaged in 4 kg polyethylene bags. In the North West Province, 10 soil samples, of 4 kg each were collected from the control area.

3.4.2. Water Sample Collection

The sampling procedure was carried out according to methodologies recommended by APHA (1985). Twenty nine (29) drinking and waste 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 radionuclide on the walls of the bottles and placed in a cold room (IAEA, 1989).

3.4.3. Plant Sample Collection

In locations where soil samples were being collected, edible plant samples like that of vegetables (~50 g each) were also collected if available at a height 5 cm above the soil surface (IAEA, 1989). Because edible plants were not readily available, only 10 samples were collected from the mining area and one from the control area. The samples were also packaged in plastic bags carrying identification marks. The coordinates of each sampling location that corresponded to the soil samples earlier discussed were recorded for traceability.

3.5. Sample Preparation

Sample preparation is recognized as the major source of errors and if not done properly, may affect the final results. Therefore, close attention was paid to every sample to avoid cross-contamination, heavy metal and radionuclide losses amongst other precautions. The collected soil samples were first spread out on plastic sheets and allowed to air dry for 2-3 days. The soil samples were then heated in an electric oven at 110 °C for up to 24 hours to remove moisture further and thereafter placed in an electric furnace at 350 °C for 48 hours to ash the plant remains (APHA, 1985). On the other hand, the collected rock samples were crushed into small grain sizes using mortar and pestle and dried thereafter. All the samples were then passed through a sieve of mesh size 2 mm to obtain a homogenous sample matrix (IAEA, 1989). Each of the samples was weighed to 3 kg using an electronic balance. The samples, were then packed and sealed in plastic marinelli beakers for 28 days before gamma spectrometric measurements. Sealing establishes secular equilibrium among the progenies of ^{238}U and ^{232}Th series by ensuring that there is no radon gas leakage due to the decay of ^{226}Ra . A small quantity of 1 g of each sample was also packaged in clean plastic bottles for elemental analysis using ICP MS.

On the other hand, 2 L of water samples were also prepared by sealing them into marinelli beakers for 28 days to establish secular equilibrium in readiness for gamma

spectroscopy (IAEA, 2004). 5 ml of each water sample was also stored in clean containers and preserved in a cool place in readiness for elemental analysis using ICP MS technique.

The collected plant samples, were washed and dried on plastic sheets at room temperature for 1-2 weeks (Kamunda et al., 2016a). To remove moisture further, these samples were heated in an electric oven at 110 °C for up to 24 hours and then ashed at 350 °C in an electric furnace for 48 hours. The ashed samples, were then packed in 2 g plastic containers and sealed for 28 days before gamma spectroscopy measurements. About 1 g of each plant sample was also packed separately for heavy metal analysis using ICP MS technique.

3.6. Analytical Methods

The measurements of activity concentrations in soil, water and plant samples were then carried out by means of Gamma Spectrometry using a Broad Energy Germanium (BEGe) detector, whilst heavy metal concentrations of all the samples were measured using Inductively Coupled Plasma-Mass Spectrometry (ICP-MS). For indoor radon concentrations, an Instrument called AlphaGuard Professional Radon Monitor was used.

3.6.1. Broad Energy Germanium (BEGe) Gamma Spectrometry

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, 2013a). 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 include regular background and calibration source measurements. The gamma photons from the samples were detected by placing the marinelli beaker directly over the BEGe detector which is usually lead shielded to avoid background radiation. Each soil sample was counted for 12 hours to allow for measurable activity. To allow for

detectable activity each water sample was counted for 24 hours while plant samples were counted for 48 hours. The background radiation around the detector inside the shielding was also measured using an empty marinelli beaker for soil and water samples whilst a small empty container for plant samples was used. The activity concentration of each sample was later measured under similar measurement geometry as background radiation. This background was then subtracted from the measured gamma spectrum of each sample before calculating the activity concentrations.

Since detection efficiencies for gamma rays vary with energy, their determination for specific radionuclide energies was done during the calibration process. The counting geometry was created during efficiency calibration and carefully duplicated for all the measurements. The measuring process and analysis of spectra was computer controlled using GENIE 2000 software (Canberra Industries, 2013a). The activity concentration of ^{238}U was determined by measuring the 295.2 keV (19.7 %) and 351.9 keV (38.9 %) gamma-ray energies of ^{214}Pb and 609.3 keV (43.3 %), 1120.3 keV (15.7 %) and 1764.5 keV (15.1 %) gamma from ^{214}Bi . The activity concentration of ^{232}Th was determined from gamma energies 238.6 keV (44.6 %) of ^{212}Pb and 338.3 keV (11.4 %), 911.6 keV (27.7 %) and 969.1 keV (16.6 %) for ^{228}Ac . On the other hand, the activity concentration for ^{40}K was measured from its 1460.8 keV (10.7 %) gamma-ray line (Kamunda et al., 2016a). Measurements of these samples were done at the Centre for Applied Radiation Science and Technology (CARST). Table 3. 1 shows a summary of the gamma-ray energies used in the determination of activity concentrations.

Table 3. 1 Gamma Ray Energies and their Associated Intensities used in the Determination of Activity Concentrations (Wahl, 2007).

Parent Radionuclide	Daughter Radionuclide	Gamma-ray Energy (keV)	Gamma-ray Intensity (%)
^{238}U	^{214}Pb	295.2	18.2
^{238}U	^{214}Pb	351.9	35.1
^{238}U	^{214}Bi	609.3	44.6
^{238}U	^{214}Bi	1120.3	14.7
^{238}U	^{214}Bi	1764.5	15.1

^{232}Th	^{228}Ac	338.3	11.3
^{232}Th	^{228}Ac	911.2	25.8
^{232}Th	^{228}Ac	968.9	15.8
^{232}Th	^{212}Pb	238.6	43.3
^{40}K		1460.8	10.7

3.6.1.1. Energy Calibration for Gamma Spectrometry

Energy calibration for the BEGe detector was done by measuring the spectrum of ^{152}Eu point source with known full-peak energies. ^{152}Eu was chosen because it consists of multiple energy peaks that cover a large energy region. The europium point source with activity of 5 kBq as at 11 June 2008 at 12 noon was placed on the detector and ran for 1 hour. This was long enough to identify the peak energies in the spectrum. Figure 3.5 shows the energy calibration curve for the BEGe detector that was used.

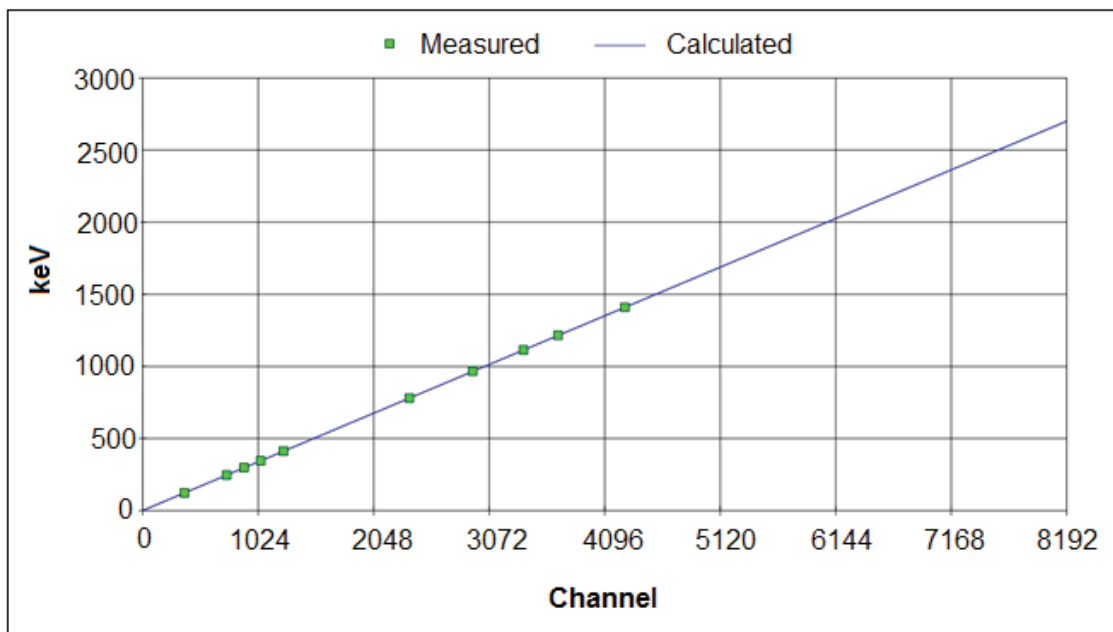


Figure 3.5: The Energy Calibration Curve for the BEGe Detector

3.6.1.2. Efficiency Calibration for Gamma Spectrometry

Efficiency calibration was done using LabSOCS (Laboratory Sourceless Calibration Software) (Canberra Industries, 2013b). This was used because of the unavailability of standard sources for the sample size that was counted. This is calibration software that eliminates the need for standard sources for efficiency calibration. A mathematical template for the detector that was characterized by CANBERRA with a combination of NIST-traceable sources was used (Canberra Industries, 2013b). This was a simplified marinelli beaker geometry. After selecting the geometry, all the relevant physical sample parameters required by the template (such as sample fill height, material, density and distance to the detector) were entered into LabSOCS Calibration Software. An efficiency calibration for the conditions specified was then generated and used for the analysis of the spectra collected, in order to yield qualitative and quantitative results. Figure 3.6 shows the dual efficiency calibration curve for the BEGe detector that was generated.

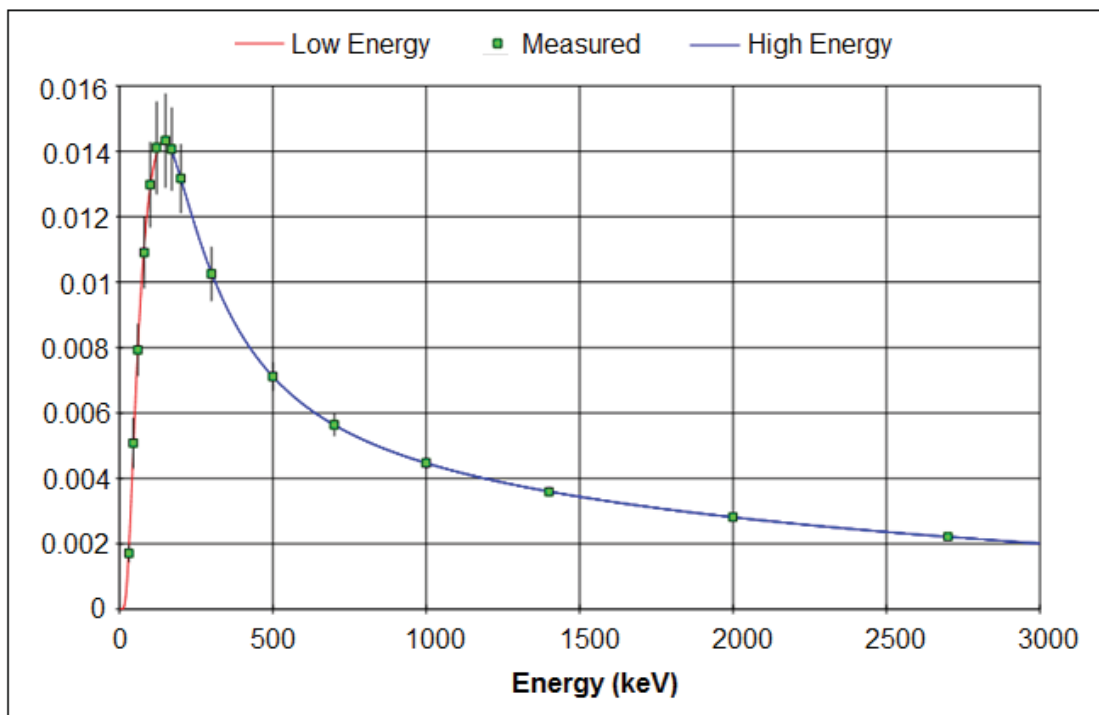


Figure 3.6: The Dual Efficiency Calibration Curve for the BEGe Detector

3.6.1.3. Calculation of Activity Concentrations

The activity concentrations of ^{238}U , ^{232}Th , and ^{40}K were determined using Equation 3.1 (Dovlete & Povinec, 2004).

$$A = \frac{N}{\varepsilon_f P_\gamma t_s m K} \quad (3.1)$$

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

ε_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 and

$$K = K_1 \cdot K_2 \cdot K_3 \cdot K_4 \cdot K_5 \quad (3.2)$$

K_1 is the correction factor for the nuclide decay from the time the sample was collected to the start of the measurement given as

$$K_1 = \exp \left[-\frac{\ln 2 \cdot \Delta t}{T_{1/2}} \right] \quad (3.3)$$

where Δt is the elapsed time from the sample that was taken from the beginning of the measurement and $T_{1/2}$ is the radionuclide half-life.

K_2 is the correction factor for the nuclide decay during counting period given as

$$K_2 = \frac{T_{1/2}}{\ln 2 \cdot t_s} \left[1 - \exp \left[-\frac{\ln 2 \cdot t_s}{T_{1/2}} \right] \right] \quad (3.4)$$

where t_s is the real time of the sample spectrum collection in seconds.

K_3 is the correction factor for a self-attenuation in the measured sample compared with the calibration sample. K_3 is defined as the ratio of the full energy peak

efficiency $E(\mu, E)$ for a sample with the linear attenuation coefficient μ to the full energy peak efficiency $\varepsilon(\mu_{ref}, E)$ for a sample with the linear attenuation μ_{ref} .

$$K_3 = \frac{\varepsilon(\mu, E)}{\varepsilon(\mu_{ref}, E)} \quad (3.5)$$

Evidently, if the matrix of both the calibration sample and the measured sample is the same, then $K_3 = 1$.

If more than one photon is absorbed by the detector during a pulse sampling cycle, the sum of the energies of two (or more) is recorded in the spectrum instead of two (or more) different signals. Any full-energy photon that is summed with another pulse is not recorded in the single photon peak and represents a loss of counts or efficiency. This loss is count rate dependent.

K_4 is the correction factor for pulses loss due to random summing:

$$K_4 = \exp(-2R\tau) \quad (3.6)$$

where τ the resolution time of the measurement system and R is the mean count rate.

For low count rates this correction factor could be taken as 1.

K_5 is the coincidence correction factor for those nuclides decaying through a range of successive photon emissions. If the nuclide has no cascade of gamma-rays then $K_5 = 1$.

As mentioned earlier, the purpose of gamma-ray spectrometry with broad energy germanium (BEGe) detectors is the determination of the activity concentration of each radionuclide species contained in the samples. Figure 3.7 therefore shows one of the spectra observed for radionuclides and their gamma-ray energies from one of the samples used in the current study.

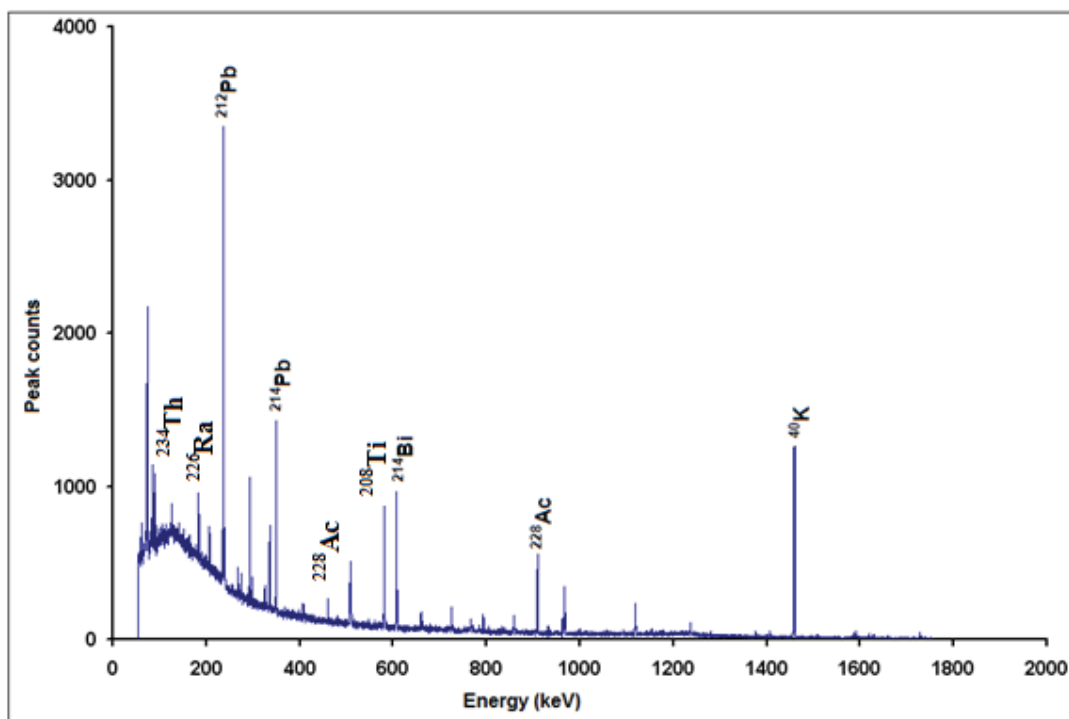


Figure 3.7: Spectrum Display for the Different Radionuclides Observed and their Gamma-ray Energies from one of the Samples Measured.

3.6.2. Inductively Coupled Plasma-Mass Spectrometry (ICP-MS)

Soil, water and plant samples were analyzed for heavy metals using ICP-MS, model PerkinElmer (NexION 300Q) (PerkinElmer, 2004-2011). ICP-MS is a powerful tool for analysing heavy metals in environmental samples. The ICP-MS system can quantitatively measure various elements and produce a value of the total amount of a specific element of interest (Bazilio & Weinrich, 2012). Before ICP-MS was done, all samples were first digested in a Multiwave 3000 microwave system as explained below.

3.6.2.1. Microwave Digestion

Digestion is done in order to extract the heavy metals from the soil, water and plant samples. For soil, 1 g dry weight of each soil sample was accurately measured and mixed with 3 ml of nitric acid and 9 ml of hydrochloric acid in a rotor vessel. About 1 ml of hydrogen peroxide was also added to the mixture. The mixture was then

digested at a temperature of 120 °C for about 45 minutes. After this, the digested samples were allowed to cool and transferred into 100 mL volumetric flasks with 2 % nitric acid. Distilled water was then used to top up to the 100 mL volume (Mathuthu et al., 2016). The digested samples were then allowed to sediment overnight and thereafter filtered with No. 40 whatman filter paper in readiness for ICP-MS.

For water samples, 5 ml of each water sample was mixed with 5 ml of nitric acid and 1 ml of hydrochloric acid was added in a rotor vessel with 1 ml of hydrogen peroxide. The mixture was again digested following the same procedure as that of the soil samples.

As for plant samples 1 g of each sample, previously ashed, was mixed with 1 ml of nitric acid and 10 ml of hydrochloric acid in a rotor vessel. 1 ml of hydrogen peroxide was also added and the same procedure for soil samples was followed to digest the plant samples.

3.6.2.2. ICP-MS Measurement

All the digested samples were then introduced into the ICP-MS. In ICP-MS, the sample components are decomposed into their atomic constituents followed by their ionization in an argon plasma at a temperature of about 6000–8000 K (PerkinElmer, 2004-2011). The positively charged ions are extracted from the plasma into a high vacuum of the mass spectrometer where they are separated by mass filters before finally being measured by an ion detector. The instrument runs each sample twice and for about 60 seconds for each complete run (USEPA, 2007). The TotalQuant method was used together with a standard solution in order to improve the analytical accuracy of the results and to be able to detect contamination and drift. The standard used is Perkin Elmer Pure Plus NexION Dual Detector Calibration Solution. TotalQuant calibration was achieved using 200 micro-grams per Liter of Al, Ba, Ce, Co, Cu, In, Li, Mg, Mn, Ni, Pb, Tb, U and Zn. For quality control, replicate samples were run together with the standard and blank solutions. The calibration process updates internal response data that correlates measured ion intensities to the concentrations of the elements in the solution. The target heavy metals were arsenic

(As), lead (Pb), mercury (Hg), cadmium (Cd), chromium (Cr), cobalt (Co), nickel (Ni), copper (Cu) and zinc (Zn).

The heavy metal concentrations obtained from the ICP-MS in mg/L were then converted into mg/kg (Temminghoff and Houba, 2004) using the following equation;

$$\frac{(a-b) \times v}{w} \text{ mg/L} \quad (3.7)$$

where a = concentration of heavy metal in the sample (mg/L);

b = concentration of the heavy metal in the blank (mg/L);

v = total volume of the digest (mL) and

w = weight of the soil sample (g).

3.6.3. Radon-222 Measurements

Radon-222 gas activity in indoor air was measured by the AlphaGuard Professional Radon Monitor. This is a portable piece of equipment designed for instantaneous and continuous measurement of radon gas activity (AlphaGuard, 2007). Indoor radon-222 concentrations in air were measured over night from the 2 mine villages. Measurements were taken 1 m above ground from 5 houses (3 east village and 2 west village) from the mine settlements as outlined by Stegnar et al. (2013). Similar radon-222 gas activity in air was also measured in 2 houses from the control area. The results were then used to estimate the annual effective dose due to inhalation of radon gas.

3.6.4. RESRAD-OFFSITE Computer Code

To estimate the cancer risk as a result of radionuclides from mining areas, RESRAD-OFFSITE computer code was used. For the study area, a number of input parameters were considered as inputs to the RESRAD-OFFSITE model. Some of these are soil density, field capacity, irrigation rate, precipitation rate, runoff coefficient, water table drop rate, and uncontaminated unsaturated zone thickness. Default parameters were also used for the input values that were not available (Yu et al., 1993).

CHAPTER 4: HEALTH RISK ASSESSMENT

4.1. Introduction

Human health risk assessment is the process of estimating the nature and probability of adverse health effects in humans who may be exposed to environmental contaminants (NRC, 1999). It is also a process used to estimate health effects that might result from exposure to carcinogenic and non-carcinogenic contaminants (USEPA, 2001). This provides information to decision-makers about the possible consequences of actions, source treatment, disposal and remediation options, as well as clean-up standards of contaminated sites. The risk assessment process is made up of four basic steps: hazard identification, exposure assessment, toxicity (dose–response) assessment, and risk characterization (USEPA, 2001). Risk is defined as the probability of suffering harm while hazard is the source of risk (USEPA, 2001). Contaminants such as radionuclides and heavy metals do not cause a risk unless exposure has occurred.

Hazard Identification basically aims to investigate what contaminants are present at a site, their concentrations, and their spatial distribution (Kamunda et al., 2016b). In the study area radionuclides and heavy metals such as As, Pb, Hg, Cd, Cr, Co, Ni, Cu and Zn were identified as possible hazards for the community.

The purpose of exposure assessment is to measure or estimate the intensity, frequency, and duration of human exposures to an environmental contaminant. In this study, exposure assessment was carried out by measuring the activity concentrations of radionuclides and the Average Daily Intake (ADI) of heavy metals through ingestion, inhalation and dermal contact by individuals from the study area.

Dose–response assessment estimates the toxicity, considering adverse effects and exposure levels of carcinogenic and non-carcinogenic contaminants. The carcinogen slope factor (CSF) and the reference dose (RfD, a non-carcinogenic threshold) are two principal toxicity indices used. RfD values are derived from animal studies using the “No observable effect level” principle. For humans, RfD values are multiplied 10-fold to account for uncertainties (Kamunda et al., 2016b).

Risk characterization predicts the potential cancerous and non-cancerous health risk of the population by integrating all the information gathered from hazard identification, exposure assessment, and toxicity assessment to arrive at quantitative estimates of cancer risk and hazard indices (USEPA, 2004).

4.2. Risk Assessment of Radionuclides

Risk assessment of radionuclides in humans is assessed through various estimating parameters as well simulation models. Some of the quantities and units used for ionizing radiation received and its biological effectiveness include Absorbed Dose, Equivalent Dose, Radium Equivalent Activity (Ra_{eq}), Absorbed Dose rate in Air (D), Annual Effective Dose Equivalent (AEDE) and Radiation Hazard Index. There are various simulation models that can be used in risk assessment, but the one that was used in the present study was the RESRAD computer model.

4.2.1. Absorbed Dose

The gray is the unit used to measure absorbed dose. This absorbed dose refers to the amount of energy actually absorbed by any material, and is also used for any radiation type. One Gray (Gy) is equal to one joule of energy deposited in one kg of a material and is named after the British Physicist Louis Harold Gray (1905–1965). The former unit is the Rad, where $1 \text{ Gy} = 1 \text{ J/kg} = 100 \text{ Rad}$. The unit gray does not describe the biological effects of the different radiations. (ISU, 2014). The absorbed dose (D) is therefore defined as the amount of the radiation energy (dE) absorbed per unit mass of material (dm). It is useful in dosimetry to define an average dose for a tissue (D_T) or organ (Equation 4.1); (ICRP, 2008)

$$D_T = \frac{dE_T}{dm_T} \quad (4.1)$$

The absorbed dose rate is the rate at which an absorbed dose is received (Gy/s). The biological effect of radiation depends not only on the energy deposited by radiation in an organism, but also on the type of radiation and the way in which the energy is deposited along the path of the radiation. Therefore, the linear energy transfer (LET) is defined as the mean energy deposited per unit path length in the absorbing material. The unit of LET is keV/μm. So for the same absorbed dose, the

biological effect of alpha particles or neutrons (high LET) is much greater than that of beta or gamma rays (low LET). For characterization of this difference in biological effects of different types of radiation, the radiation weighting factor (w_R) was established (Table 4.1) and is published in the ICRP recommendations 103 (ICRP, 2008).

Table 4.1: Radiation weighting factors for common radiations (ICRP, 2008)

Type and energy range	Radiation weighting factors, w_R
Photons, electrons and muons, all energy	1
Protons, other than recoil protons, energy > 2 MeV	5
Alpha particles, fission fragments, heavy nuclei	20

4.2.2. Equivalent Dose

Another parameter is the quantity called equivalent dose (measured in Sieverts). This relates the absorbed dose in human tissue and the effective biological damage it causes. Not all radiation has the same biological effect, even for the same amount of absorbed dose (ISU, 2014). The equivalent dose (H_T), therefore, describes the correlation between the absorbed dose and its biological effects. So the absorbed dose is multiplied by the weighting factor (w_R) of the radiation. If there are several types of radiation (R) present, the equivalent dose in the tissue (T) is the weighted sum over all contributions (Equation 4.2); (ICRP, 2008).

The unit of the equivalent dose was named after the Swedish doctor and physicist Rolf M. Sievert (1896–1966). Equivalent dose is often expressed in microsievert (μSv) or millisievert (mSv). The former unit is rem (radiation equivalent man); 1 Sv = 100 rem

$$H_T = \sum_R (w_R \cdot D_{T,R}) \quad (4.2)$$

The equivalent dose rate is the rate at which an equivalent dose is received, expressed for example in Sv/s or Sv/h. The equivalent dose is always related to a defined tissue or organ. Different tissues and organs show different sensitivities to radiation, depending on their cell cleavage frequency and their cell renewal frequency. To take these effects into account, the equivalent doses in different tissues must be weighted (Table 4.2, ICRP (2008)).

The equivalent dose (H_T) in tissue or organ T multiplied by this tissue weighting factor (w_T) is called the effective dose (E) (ICRP, 2008). In the new ICRP recommendations 103 (ICRP, 2008), the absorbed dose and the equivalent dose are calculated for women and men and for determination of the effective dose an averaged equivalent dose is used. The sum of the relative weighting factors is one, this means that the sum of the weighting risks for the organs is numerically equal to the risk for the whole body (ICRP, 2008) (Equation 4.3).

$$E = \sum_T (w_T \cdot H_T) \quad (4.3)$$

Table 4.2: Tissue weighting factors (ICRP, 2008)

Tissue weighting factors		
Tissue or Organ	w_T	$\sum w_T$
Gonads	0.2	0.2
Red marrow, colon, lung, stomach	0.12	0.48
Bladder, breast, liver, esophagus, thyroid, remainder	0.05	0.3
Skin, bone surface	0.01	0.02
	Total	1.0

4.2.3. Radium Equivalent Activity (Ra_{eq})

Due to a non-uniform distribution of natural radionuclides in samples, the actual activity level of ^{226}Ra , ^{232}Th and ^{40}K in the samples can be evaluated by means of a common radiological index named the Radium Equivalent Activity (Ra_{eq}) (Tufail, 2012). It is the weighted sum of ^{226}Ra , ^{232}Th and ^{40}K activity concentrations, based

on the assumption that 370 Bq.kg⁻¹ of ²²⁶Ra, 259 Bq.kg⁻¹ of ²³²Th and 4810 Bq.kg⁻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). On the basis of these values, Ra_{eq} is defined as follows:

$$Ra_{eq} = \left(\frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \right) \times 370 \quad (4.4)$$

which is equivalent to

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (4.5)$$

where A_{Ra}, A_{Th} and A_K are the activity concentrations in Bq kg⁻¹ of ²²⁶Ra (which can be that of ²³⁸U), ²³²Th, and ⁴⁰K, respectively. The Ra_{eq} is therefore a single index or number to describe the gamma output from different mixtures of radionuclides in a material.

4.2.4. Absorbed Dose Rate in Air (*D*)

For health protection purposes, the absorbed dose rate coming from the external gamma radiation can be calculated by knowing the activity concentrations in the environmental medium such as soil. This outdoor absorbed dose rate in air due to terrestrial gamma rays at 1m above the ground surface is calculated from ²²⁶Ra (²³⁸U), ²³²Th and ⁴⁰K concentration values in the medium 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 Sjoeren, 1985). In general, the estimates of ²³⁸U and ²³²Th activity concentrations are based on the assumption of radioactive equilibrium conditions through the direct measurement of daughter radionuclides.

The absorbed dose rate is therefore given by the following formula (UNSCEAR, 2000):

$$D(\text{nGy.h}^{-1}) = 0.462A_{Ra} + 0.604A_{Th} + 0.0417A_K \quad (4.6)$$

Where D is the absorbed dose rate, A_{Ra}, A_{Th} and A_K are the activities concentration of ²²⁶Ra (²³⁸U), ²³²Th and ⁴⁰K, respectively. The dose coefficient in units of nGy.h⁻¹ per Bq.kg⁻¹ was taken from the UNSCEAR (2000) report.

4.2.5. Total Annual Effective Dose

The total annual effective dose to individual members of the public is calculated using the ICRP dose formula [ICRP, 2012]. This is determined by summing all the individual components of the equivalent doses for the different exposure pathways. In this study, the following components were considered:

- Annual effective dose due to external gamma irradiation from ^{238}U , ^{232}Th and ^{40}K in the soil, plant and water samples;
- Annual effective dose from ingestion of water and plant samples containing ^{238}U , ^{232}Th and ^{40}K and
- Annual effective dose due to Inhalation of indoor radon gas from soil.

4.2.5.1. Annual Effective Dose Equivalent from Terrestrial Gamma Radiation

The absorbed dose rate in air at 1 meter 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 Sv Gy^{-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:

$$\text{AEDE}(\text{mSv.y}^{-1}) = D(\text{nGy.h}^{-1}) \times 8760 (\text{h}) \times 0.2 \times 0.7 (\text{Sv.Gy}^{-1}) \times 10^{-6} \quad (4.7)$$

Where 8760 is the time in hours for one year; 10^{-6} is the factor converting from nano to milli.

4.2.5.2. Annual Effective Dose from Ingestion of Water and Plant Samples.

The annual effective dose (E_{ing}) from ingestion of ^{238}U , ^{232}Th and ^{40}K through water and plant samples for members of the public is estimated from the activity concentrations of each individual radionuclides found in the sample. Equation 4.9 was used in the calculation (UNSCEAR, 2000) as follows:

$$E_{\text{ing}}(\text{mSv} \cdot \text{y}^{-1}) = A_{\text{R}} I_{\text{R}_{\text{ing}}} \sum_{j=1}^3 \text{DCF}_{\text{ing}} \quad (4.8)$$

Where, A_{R} = the activity concentration of the radionuclides in a sample
 $I_{\text{R}_{\text{ing}}}$ = the consumption rate per year, and
 DCF_{ing} = the effective dose coefficient in $\text{Sv} \cdot \text{Bq}^{-1}$ for the ingestion of natural radionuclides. For ^{238}U , a value of 4.50×10^{-8} was used. For ^{232}Th , 2.30×10^{-7} was used and 6.20×10^{-9} was used for ^{40}K (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).

4.2.5.3. Annual Effective Dose from Inhalation of Radon

The annual effective dose (E_{inh}) from the inhalation of radon due to indoor air is determined by an expression suggested by UNSCEAR (2000):

$$E_{\text{Rn}}(\text{mSv} \cdot \text{y}^{-1}) = \text{DCF}_{\text{Rn}} F_{\text{Rn}} A_{\text{Rn}} T_{\text{exp}} \times 10^{-6} \quad (4.9)$$

Where;

E_{Rn} = the annual effective dose of radon through inhalation,
 DCF_{Rn} = the dose conversion factor of radon via inhalation (assumed to be 9 nSv/Bqhm^{-3}) (UNSCEAR, 2000),
 F_{Rn} = the indoor equilibrium factor between radon and its progeny (assumed to be 0.4) (UNSCEAR, 2000),
 A_{Rn} = the radon activity concentration in $\text{Bq} \cdot \text{m}^{-3}$, and
 T_{exp} = the exposure time to this concentration (assumed to be 7000 hours in one year) (UNSCEAR, 2000).

4.2.6. 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 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.kg⁻¹ (Beretka & Mathew, 1995). This is a widely used hazard index reflecting the external exposure and is defined by Equation 4.10 (UNSCEAR, 2000):

$$H_{ex} = \left(\frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \right) \quad (4.10)$$

Where A_{Ra}, A_{Th} and A_K are the mean activity concentrations of ²²⁶Ra (²³⁸U), ²³²Th and ⁴⁰K, in Bq.kg⁻¹ respectively.

In addition to H_{ex}, 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 Equation 4.11 (UNSCEAR, 2000):

$$H_{in} = \left(\frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \right) \quad (4.11)$$

Where A_{Ra}, A_{Th} and A_K are as defined in Equation 4.8. The value of H_{in} must also be less than unity for the radiation hazard to be negligible.

4.2.7. Risk Assessment of ²³⁸U, ²³²Th and ⁴⁰K using RESRAD-OFFSITE Code

For the study area, a number of parameters were considered as inputs to the RESRAD-OFFSITE code (version 3.1). The resident scenario was chosen as the critical receptor in this risk assessment. The parameter values were carefully selected to achieve a more realistic estimation of the dose or risk. Where necessary, site-specific parameters replaced default parameters (Yu et al. 1993).

Input parameters to the model included the measured activity concentrations of ^{238}U , ^{232}Th , ^{40}K , and meteorological data (annual wind speed and direction). Hydrogeological parameters for the various zones were also taken into consideration. These included hydraulic conductivity and density of the soil. Some hydrological parameters, such as total porosity, field capacity, irrigation rate, erosion rate, precipitation rate, runoff coefficient, and soil-specific exponential parameters were also estimated based on the condition of the study area (Yu et al. 1993). The area and thickness of the different zones were also taken into account.

The main exposures in this scenario included direct exposure to the radionuclides in soil, inhalation of dust, and ingestion of contaminated vegetables, water, and soil. Dietary information on vegetables, water, and soil was also considered through regulatory guidelines set by the National Nuclear Regulator (NNR, 2013). Inhalation rate, soil ingestion rate, and the distribution coefficient for radionuclides in the study area were also taken into account according to guidelines (NNR, 2013). The RESRAD library of radionuclides was then used in this assessment. A summary of the most relevant data used for the risk assessment is shown in Table 4.3.

Table 4.3: Summary of Input Parameters for the RESRAD-OFFSITE code (Mathuthu et al., 2016).

Parameter	Value	Description and/or Reference
Average activity concentrations of ^{238}U , ^{232}Th , and ^{40}K in soil	Table 5.1	The values were based on measurements
Area of contaminated zone (m^2)	45,000,000	This was the average estimated over the whole area
Thickness of contaminated zone (m)	0.05	This was the average estimated over the whole area
Cover depth (m)	0	There was no cover depth for the radionuclides
Density of contaminated zone (g/m^3)	1.44	For sandy loam soil (Yu et al. 1993).
Erosion rate for contaminated zone (m/y)	0.001	RESRAD default value (Yu et al. 1993).
Total porosity for contaminated zone	0.43	For sedimentary material which is sand fine (Yu et al. 1993).
Field capacity for contaminated zone	0.2	RESRAD default value (Yu et al. 1993).
Hydraulic conductivity of contaminated zone (m/y)	1090	For sandy loam soil (Yu et al. 1993).
Soil-specific	4.9	For sandy loam soil (Yu et al. 1993).

exponential b parameter		
Average annual wind speed (m/s)	5	For the area (Hagemann 2013)
Evapotranspiration coefficient	0.5	RESRAD default value (Yu et al. 1993).
Precipitation rate (m/y)	1	The area is relatively humid. (Yu et al. 1993).
Irrigation rate	0.2	RESRAD default value (Yu et al. 1993).
Runoff coefficient	0.65	For moderately steep, residential area
Inhalation rate (m ³ /y)	8059.2	For South Africa (NNR 2013).
Exposure duration (y)	30	RESRAD default value (Yu et al., 1993).
Fraction of time spent onsite		For South Africa (NNR 2013).
Indoor	0	
Outdoor	0.1	
Fraction of time spent offsite		For South Africa (NNR 2013).
Indoor	0.8	
Outdoor	0.1	
Soil ingestion rate (g/y)	37	For South Africa (NNR 2013).
Leach rate	0	RESRAD default value (Yu et al. 1993).
Water drinking rate (L/y)	600	For South Africa (NNR 2013).
Distribution coefficient (K _d) - uranium (cm ³ /g)	200	For South Africa (NNR 2013).
Distribution coefficient (K _d) - thorium (cm ³ /g)	60000	RESRAD default value (Yu et al. 1993).
Distribution coefficient (K _d) - potassium (cm ³ /g)	5.5	RESRAD default value (Yu et al. 1993).

4.3. Risk Assessment of Heavy Metals

The potential risk from heavy metals in contaminated soils, water and plants are calculated based on recommendations by several USEPA publications (USEPA, 1989; USEPA, 1991; USEPA, 2001; USEPA, 2004; USEPA, 2007; USEPA, 2011a). The average daily intake (ADI) (mg/kg-day) for the different pathways were calculated using the following exposure Equations (4.12- 4.15) as prescribed by USEPA (1989):

Ingestion of Heavy Metals through Soil, Water and Plants:

$$ADI_{ing} = \frac{C \times IR_{ing} \times EF \times ED}{BW \times AT \times 10^6} \quad (4.12)$$

Where ADI_{ing} (mg/kg-day), is the average daily intake of heavy metals ingested from soil, C = concentration of heavy metal in mg/kg for soil and plants and in mg/L for water. IR_{ing} (mg/day) is the ingestion rate, EF (days/year) is the exposure frequency, ED (years) is the exposure duration, BW (kg) is the body weight of the exposed individual, AT (days) is the time period over which the dose is averaged and 10^6 is the conversion factor in kg/mg.

Inhalation of Heavy Metals via Soil Particulates:

$$ADI_{inh} = \frac{C_s \times IR_{inh} \times EF \times ED}{BW \times AT \times PEF} \quad (4.13)$$

Where ADI_{inh} (mg/kg-day) is the average daily intake of heavy metals inhaled from soil, C_s (mg/kg) is the concentration of heavy metal in soil, IR_{inh} (m^3 /day) is the inhalation rate, PEF (m^3 /kg) is the particulate emission factor. EF , ED , BW and AT are as defined earlier in Equation 4.12 above.

Dermal Contact with Water:

$$ADI_{demw} = \frac{C_w \times SA \times PC \times ET \times EF \times ED \times CF}{BW \times AT} \quad (4.14)$$

Where ADI_{demw} (mg/kg-day) is average daily intake of heavy metals via dermal contact from water. C_w (mg/L) is the concentration of metal in water, SA (cm^2) is the exposed skin area, PC (cm/h) is the dermal permeability coefficient, ET (hour/day) is the exposure time during bathing and shower, CF (L/cm^{-3}), the unit conversion factor. EF , ED , BW and AT are as defined earlier in Equation 4.12 above.

Dermal Contact with Soil:

$$ADI_{dems} = \frac{C_s \times SA \times FE \times AF \times ABS \times EF \times ED}{BW \times AT \times 10^6} \quad (4.15)$$

Where ADI_{dems} (mg/kg-day) is average daily intake via dermal contact from soil. C_s (mg/kg) is the concentration of metal in soil, SA (cm^2) is the exposed skin area, FE is the fraction of the dermal exposure ratio to soil, AF (mg/cm^2) is the soil adherence factor, ABS is the fraction of the applied dose absorbed across the skin, 10^6 (kg/mg) is the conversion factor. EF , ED , BW and AT are as defined earlier in Equation 4.12

above. Table 4.4 shows the exposure parameters used for the health risk assessment through the different exposure pathways.

Table 4.4: Exposure Parameters used for Risk Assessment through Different Exposure Pathways (Kamunda et al., 2016b).

Parameter	Media	Unit	Individual	References
Body weight (BW)	all	kg	70	(DEA, 2010)
Exposure frequency (EF)	all	days/year	350	(DEA, 2010)
Exposure duration (ED)	all	years	30	(DEA, 2010)
Ingestion rate (IR _{ing})	soil	mg/day	100	(DEA, 2010)
	water	L/day	2	(USEPA, 2004)
	plants	mg/day	200	
Inhalation rate (IR _{inh})	soil	m ³ /day	20	(DEA, 2010)
Skin surface area (SA)	soil & water	cm ²	5800	(DEA, 2010)
Soil adherence factor (AF)	soil	mg/cm ²	0.07	(DEA, 2010)
Dermal Absorption factor (ABS)	soil		0.1	(DEA, 2010)
Dermal exposure ratio (FE)	soil	none	0.61	(USEPA, 2004)
Particulate emission factor (PEF)	soil	m ³ /kg	1.3 x 10 ⁹	(DEA, 2010)
Average time (AT)	all	days		
For carcinogens			365 × 70	(DEA, 2010)
For non-carcinogens			365 × ED	(DEA, 2010)
Exposure time during bathing and shower (ET)	water	hours/day	0.3	(USEPA, 2004)
Dermal permeability coefficient (PC)	water	cm/h	1.0 x 10 ⁻³	(USEPA, 2004)
Unit conversion factor (CF)	water	L/cm ⁻³	1.0 x 10 ⁻³	(USEPA, 2004)

4.3.1. Non-carcinogenic Risk Assessment of Heavy Metals

Non-carcinogenic hazards are characterized by the hazard quotient (HQ). HQ is a unitless number that is expressed as the probability of an individual suffering an adverse effect. The hazard quotient is defined as the quotient of the average daily intake or dose divided by the toxicity threshold value, which is referred to as the reference dose (RfD) of a specific chemical (USEPA, 1989):

$$HQ = \frac{ADI}{RfD} \quad (4.16)$$

Where RfD is the chronic reference dose for the heavy metal (mg/kg-day). For n number of heavy metals, the non-carcinogenic effect to the population is a result of the summation of all the HQs due to individual heavy metals. This is considered to be another term called the Hazard Index (HI) as described by USEPA document of 1989. Equation 4.17 shows the mathematical representation of this parameter:

$$HI = \sum_{k=1}^n HQ_k = \sum_{k=1}^n \frac{ADI_k}{RfD_k} \quad (4.17)$$

Where HQ_k ADI_k is RfD_k are values of heavy metal k . If the HI value is less than one, the exposed population is unlikely to experience adverse health problems. If the HI value exceeds one, then there may be concern for potential non-carcinogenic effects (USEPA, 1989).

4.3.2. Carcinogenic Risk Assessment of Heavy Metals

For carcinogens, the risks are estimated as the incremental probability of an individual developing cancer over a lifetime as a result of exposure to the potential carcinogen. The basic equation for calculating the lifetime cancer risk is:

$$Risk = ADI \times CSF \quad (4.18)$$

Where "Risk" is a unitless probability of an individual developing cancer over a lifetime; ADI is the average daily intake or dose as highlighted earlier. CSF is the cancer slope factor, expressed in $(mg/kg/day)^{-1}$. It converts the estimated daily intake of the heavy metal averaged over a lifetime of exposure directly to incremental risk of an individual developing cancer (USEPA, 1989).

The total lifetime cancer risks for an individual is calculated from the average contribution of the individual heavy metals in the different media for all the pathways using the following equations:

$$Risk_{(total)} = Risk_{(ing)} + Risk_{(inh)} + Risk_{(dermal)} \quad (4.19)$$

Where $Risk_{(ing)}$, $Risk_{(inh)}$, and $Risk_{(dermal)}$ are risks contributions through different media i.e soil, water and plants. For n number of heavy metals, the risk for the given pathway is given by Equation (4.20) as follows:

$$\text{Risk}_{\text{pathway}} = \sum_{k=1}^n \text{ADI}_k \text{CSF}_k \quad (4.20)$$

Where ADI_k and CSF_k are the ADI and CSF, for the k^{th} heavy metal.

Both non-carcinogenic and carcinogenic risk assessment of heavy metals are estimated from RfD and CSF values largely derived from the Department of Environmental Affairs (DEA), South Africa and USEPA as shown in Table 4.5

Table 4.5: Reference Doses (RfD) in (mg/kg-day) and Cancer Slope Factors (CSF) for the Different Heavy Metals (Kamunda et al., 2016b).

Heavy Metal	Ingestion RfD	Dermal RfD	Inhalation RfD	Ingestion CSF	Dermal CSF	Inhalation CSF	References
As	3.00E-04	3.00E-04	3.00E-04	1.50E+00	1.50E+00	1.50E+01	(DEA, 2010) (USEPA, 2002)
Pb	3.60E-03	-	-	8.50E-03	-	4.20E-02	(DEA, 2010) (Luo et al., 2012)
Hg	3.00E-04	3.00E-04	8.60E-05	-	-	-	(DEA, 2010)
Cd	5.00E-04	5.00E-04	5.70E-05	-	-	6.30E+00	(DEA, 2010) (USEPA, 1991)
Cr (VI)	3.00E-03	-	3.00E-05	5.00E-01	-	4.10E+01	(DEA, 2010) (USEPA, 2007)
Co	2.00E-02	5.70E-06	5.70E-06	-	-	9.80E+00	(USEPA, 2011a)
Ni	2.00E-02	5.60E-3	-	-	-	-	(DEA, 2010) (IRIS, 2007)
Cu	3.7.00E-02	2.40E-2	-	-	-	-	(DEA, 2010) (USEPA, 2011a)
Zn	3.00E-01	7.50E-02	-	-	-	-	(DEA, 2010) (USEPA, 2011a)

CHAPTER 5: RESULTS AND DISCUSSIONS

5.1. Introduction

The chapter presents the results and discussions on health risk assessment of natural radionuclides (^{238}U , ^{232}Th and ^{40}K) and heavy metals from the gold mining area. The results of the activity concentrations of these natural radionuclides performed by means of high-resolution gamma-ray spectrometry from soil, plant and water samples are presented first accompanied by their radiological hazard assessment. RESRAD-OFFSITE Computer Code was then used to calculate the cancer risk. The results obtained from Inhalation of indoor radon and the corresponding annual effective dose are also presented here. Results from the ICP-MS measured concentrations of heavy metals in soil, plants and water samples are also presented and discussed. These results of heavy metals are then used to estimate carcinogenic and non-carcinogenic risks using exposure equations prescribed by USEPA (1989). The total cancer risk as a result of both natural radionuclides and heavy metals were also calculated.

5.2. Activity Concentrations of ^{238}U , ^{232}Th and ^{40}K for Soil Samples

The results presented in Table 5.1 show the average activity concentrations of NORMs for soil samples analysed from the gold mining area and from the control area. These values were derived from the weighted average of selected gamma energy peaks of their daughter radionuclides (Table 3.1) in each soil sample. For an expanded source of information, Appendix A (Table AA1 – AA4) is referred to. The average activity concentration of ^{238}U in Bq.kg^{-1} was found to be 574.3 ± 11.1 . For ^{232}Th the average activity concentration was 49.4 ± 1.9 and that of ^{40}K was $424.7 \pm 25.7 \text{ Bq.kg}^{-1}$. On the other hand, the average activity concentrations in Bq.kg^{-1} for ^{238}U , ^{232}Th , and ^{40}K from the control area were 17.0 ± 0.4 , 22.2 ± 0.5 and 496.8 ± 15.2 , respectively. The errors in the activity concentrations were calculated using propagation of uncertainty equations based on the weighted averages of the radionuclides (see Appendix F).

Table 5.1: Activity Concentrations of ^{238}U , ^{232}Th and ^{40}K for Soil Samples from the Mining Area and from the Control Area.

Location	No. of Samples	Parameter	Activity Concentrations (Bq·kg ⁻¹)		
			²³⁸ U	²³² Th	⁴⁰ K
Control area	10	Average	17.0±0.4	22.2±0.5	496.8±15.2
		Min	12.5±0.3	16.6±0.4	424.3±13.0
		Max	23.6±0.5	30.4±0.6	648.4±19.8
Tailings one	11	Average	733.4 ± 12.7	41.3 ± 0.9	339.9 ± 10.7
		Min	304.4 ± 5.4	36.5 ± 0.8	271.5 ± 9.0
		Max	1243.4 ± 21.6	54.5 ± 1.2	468.4 ± 14.6
Tailings two	13	Average	794.9 ± 13.8	44.9 ± 1.0	460.7 ±14.4
		Min	616.6 ± 10.7	38.4 ± 0.9	226.5 ±7.8
		Max	1391.8 ± 24.1	49.5 ± 1.1	681.9 ± 21.0
Tailings three	8	Average	1556.2 ± 27.0	59.0 ± 1.3	354.3 ± 11.4
		Min	390.9 ± 6.8	22.3 ± 0.6	233.8 ± 7.5
		Max	2668.9 ± 46.2	89.7 ± 1.9	497.1 ± 15.6
Tailings four	12	Average	232.0 ± 4.1	33.2 ± 0.8	489.5 ± 14.1
		Min	87.2 ± 1.6	20.5 ± 0.6	257.9 ± 8.3
		Max	618.2 ± 10.8	49.1 ± 1.1	781.0 ± 23.9
Tailings five	12	Average	609.7 ± 10.6	41.1 ± 0.9	490.7 ± 15.2
		Min	236.1 ± 4.2	25.3 ± 0.6	281.2 ± 8.8
		Max	2054.7 ± 35.6	67.1 ± 1.4	574.9 ± 17.7
West village	6	Average	47.7±0.9	50.0±1.0	231.2±7.3
		Min	34.4±0.7	42.6±0.9	160.4±5.1
		Max	61.0±1.2	55.7±1.2	362.0±11.3
East village	11	Average	50.1±1.0	55.6±1.2	219.3±7.0
		Min	37.1±0.7	41.9±0.9	133.8±4.4
		Max	75.8±1.4	76.6±1.6	280.8±8.8
Rock dumps	8	Average	570.2±18.7	70.3±8.0	811.9±125.4
		Min	52.9±5.6	21.9±7.0	527.8±82.7
		Max	3681.6±87.1	167.5±12.6	1177.0±114.1
Overall Average for Mining Area			574.3±39.5	49.4±8.5	424.7±129.3
Overall Minimum for Mining Area			34.4±0.7	20.5±0.6	133.8±4.4
Overall Maximum for Mining Area			3681.6±87.1	167.5±12.6	1177.0±114.1

The results presented in Table 5.1 are further illustrated in Figure 5.1 below. The average activity concentration of ^{238}U was highest in Tailings three followed by Tailings two, Tailings one, Tailings five, and then Rock dumps followed by East village and lastly West village. High levels of ^{238}U were found in gold mine tailings and rock dumps as these were wastes extracted from the underground mine. These results, compared to the control area indicated that mining activities contributed

significantly to the elevated levels of ^{238}U . The results also showed enhanced levels of ^{232}Th in rock dumps and in gold mine tailings compared to the control area. Soil samples from the mine villages had the least amount of ^{232}Th . The rock dumps followed by mine Tailings three had the highest average activity concentration of ^{232}Th compared to other areas. The other areas had less activity concentration because gold ore had been reprocessed to extract uranium. In soil obtained from the village there was less contamination from mining activities. The average activity concentration of ^{40}K was higher in rock dumps and comparable to the control area than in the rest of the mining areas. The activity concentration of ^{40}K from the control area would have been expected to be lower than that from the mining area, but this was not the case in many areas. These high levels of ^{40}K in the control area are believed to be as a result of the use of fertilizers for agricultural production some years back.

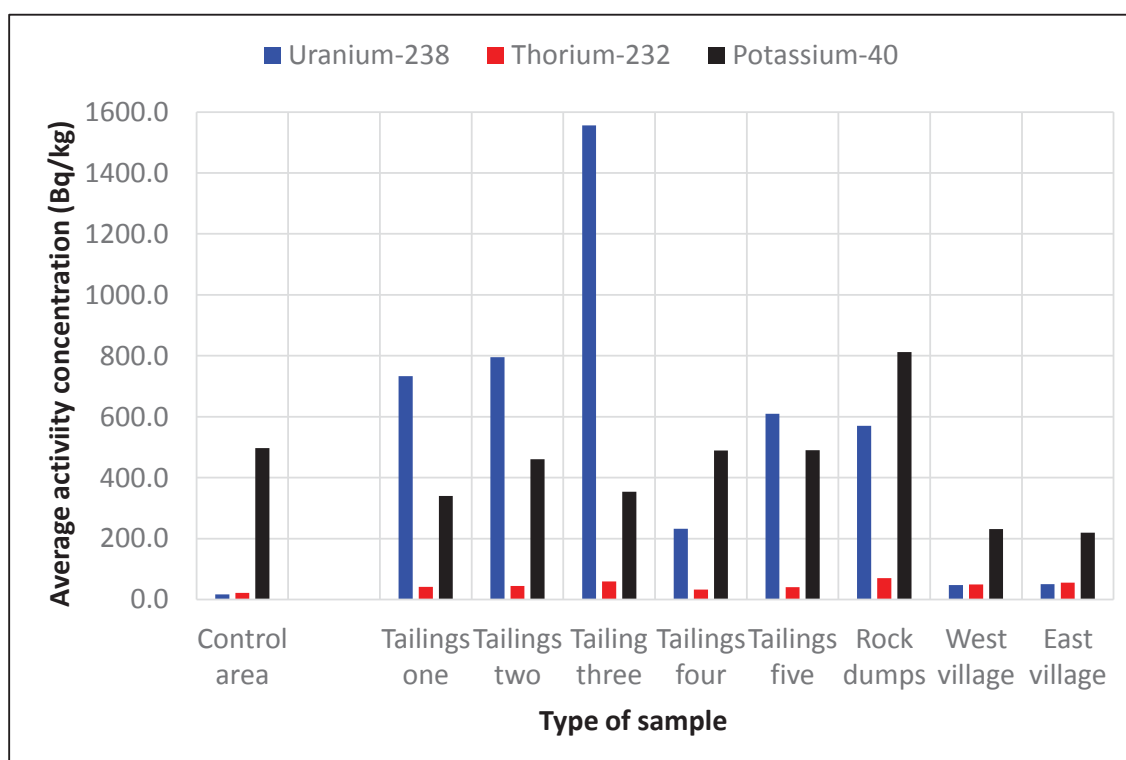


Figure 5.1: Average activity concentrations of ^{238}U , ^{232}Th and ^{40}K for soil samples from the mining area and the control site.

5.3. Comparison of Activity Concentrations of ^{238}U , ^{232}Th and ^{40}K in Soils from the Study Area with other Countries

The activity concentrations of ^{238}U , ^{232}Th and ^{40}K in soils from the mining area were also compared with results published from other countries including high uranium areas as presented in Table 5.2. The worldwide average activity concentrations of ^{238}U , ^{232}Th and ^{40}K in soil samples from various studies around the world are 33, 45 and 420 Bq.kg⁻¹, respectively (UNSCEAR, 2000).

Table 5.2: Comparison of activity concentrations of ^{238}U , ^{232}Th and ^{40}K in soils from the study area with other countries (Kamunda et al., 2016a).

Country	Activity Concentrations in Soil (Bq.kg ⁻¹)					
	^{238}U		^{232}Th		^{40}K	
	Mean	Range	Mean	Range	Mean	Range
South Africa(**)	574.3	34.4-3681.6	49.4	20.5-167.5	424.7	133.8-1177.0
Nigeria (++)	37	6-67	63	17-87	998	85-1565
Ghana (*)	29	-	25	-	582	-
Egypt (+)	37	6-120	18	2-96	320	29-650
Algeria (+)	30	2-110	25	2-140	370	66-1150
United States (+)	35	4-140	35	4-130	370	100-700
Romania (Uranium area) (+)	57	-	31	-	486	-
China (+)	33	2-690	41	1-360	440	9-1800
India (+)	29	7-81	64	14-160	400	38-760
Japan (+)	29	259	28	2-288	310	15-990
Syrian Arab Republic (+)	23	10-64	20	10-32	270	87-780
Ireland (+)	37	8-120	26	3-60	350	40-800
Switzerland (+)	40	10-150	25	4-70	370	40-1000
Hungary (+)	29	12-66	28	12-45	370	79-570
Russian Federation (+)	19	0-67	30	2-79	520	100-1400
Greece (+)	25	1-240	21	1-190	360	12-1570
Portugal (+)	49	26-82	51	22-100	840	220-1230
World average (+)	33		45		420	

Legend: (+) UNSCEAR 2000 Report, (++) Nasiru., 2013; * Darko, 2007; (**) Present work

The average activity concentration of ^{238}U from the study area was very high compared to the other countries (including uranium areas) and the world average.

This is attributed to the large volumes of uranium waste found in the gold mine, estimated to be almost ten times the amount of gold itself. Figure 5.2 illustrates this point.

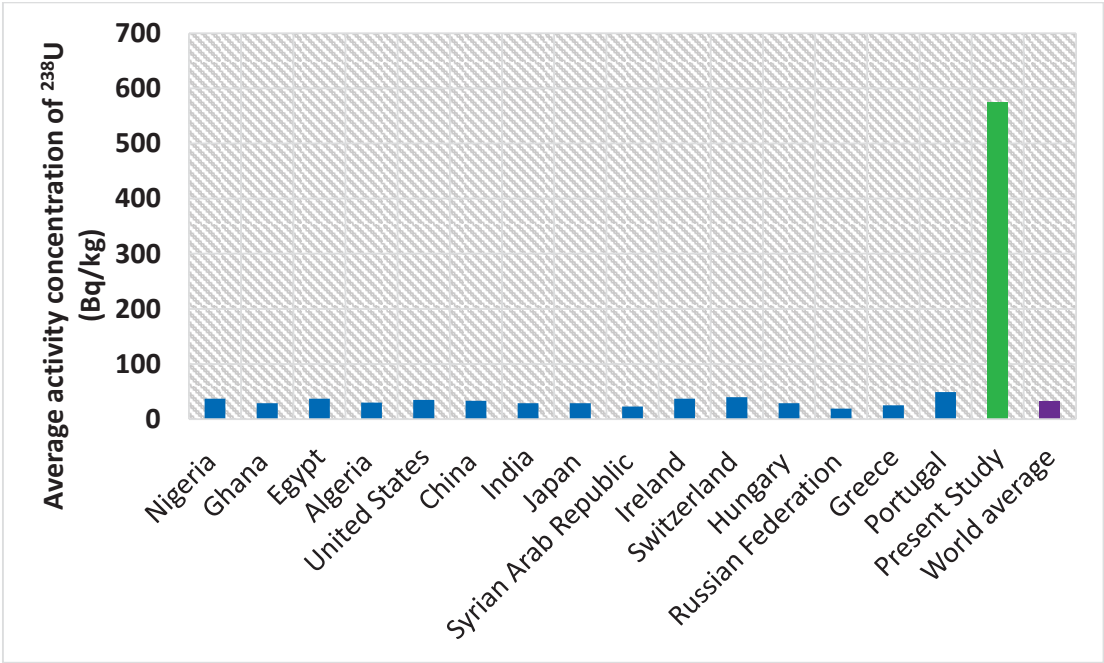


Figure 5.2: Comparison of Average Activity Concentration of ^{238}U relative to other Countries in the World

For ^{232}Th and ^{40}K , average activity concentrations from the mining area was comparable to the world average. Figures 5.3 and 5.4 show this.

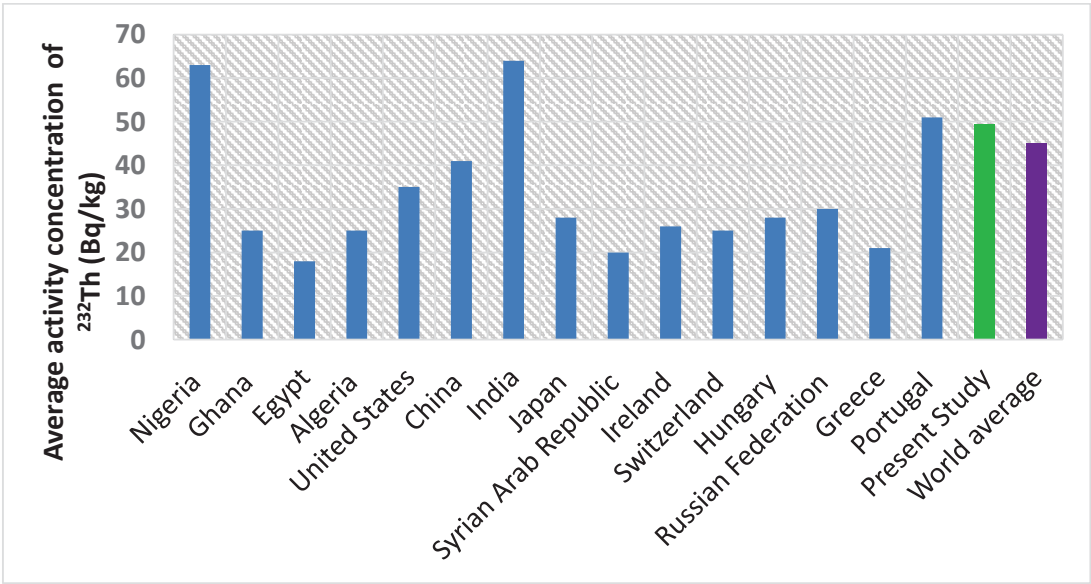


Figure 5.3: Comparison of Average Activity Concentration of ^{232}Th relative to other Countries in the World

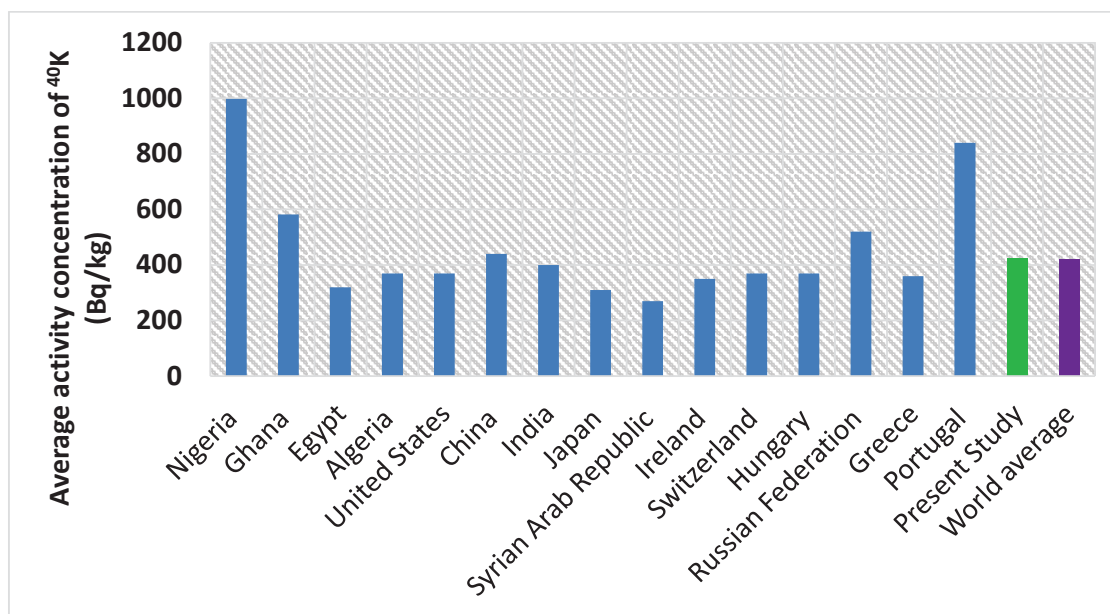


Figure 5.4: Comparison of Average Activity Concentration of ^{40}K relative to other Countries in the World

5.4. Radiological Hazard Assessment for Soil Samples

All the radiological hazard parameters for soil samples were considered for the study area and the results of the findings are summarized in Table 5.3. Detailed information about the sources of origin of these parameters is shown in Appendix A (Table AA5). Equations 4.5, 4.6, 4.7, 4.10 and 4.11 in chapter 4 were used to calculate R_{eq} , D , AEDE , H_{ex} and H_{in} parameters, respectively.

Table 5.3: Calculated Radium Equivalent Activity (Ra_{eq}), Absorbed Dose Rate in air (D), Annual Effective Dose Equivalent (AEDE), External Hazard Index (H_{ex}) and Internal hazard Index (H_{in}) of Soil Samples from the Mining area and from the Control site (Kamunda et al., 2016a).

Location	No of Samples	Parameter	Ra _{eq} (Bq.kg ⁻¹)	D (nGy.h ⁻¹)	AEDE (mSv.y ⁻¹)	H _{ex}	H _{in}
Control area	10	Average	86.9±1.4	41.9±0.7	0.05	0.2	0.3
		Min	76.4±1.4	37.5±0.7	0.05	0.2	0.2
		Max	103.8±1.5	49.1±0.8	0.06	0.3	0.3
Tailings one	11	Average	818.6±12.9	377.9±5.9	0.46	2.2	4.2
		Min	390.8±5.6	181.1±2.6	0.22	1.1	1.9
		Max	1342.2±21.7	618.7±10.0	0.76	3.6	7.0
Tailings two	13	Average	894.6±14.0	413.6±6.4	0.51	2.4	4.6
		Min	707.2±10.9	327.3±5.0	0.40	1.9	3.6
		Max	1480.0±24.2	682.3±11.2	0.84	4.0	7.8
Tailings three	8	Average	1667.9±27.1	769.4±12.5	0.94	4.5	8.7
		Min	440.9±6.9	203.9±3.2	0.25	1.2	2.2
		Max	2811.9±46.3	1297.3±21.4	1.59	7.6	14.8
Tailings four	12	Average	317.1±4.4	147.6±2.1	0.18	0.9	1.5
		Min	160.3±2.1	74.5±1.0	0.09	0.4	0.7
		Max	746.7±11.0	346.9±5.1	0.43	2.0	3.7
Tailings five	12	Average	706.4±10.8	327.0±5.0	0.40	1.9	3.6
		Min	317.8±4.4	148.0±2.1	0.18	0.9	1.5
		Max	2182.1±35.7	1007.3±16.5	1.24	5.9	11.5
East Village	11	Average	146.5±2.0	65.9±0.9	0.08	0.4	0.5
		Min	107.4±1.5	48.0±0.7	0.06	0.3	0.4
		Max	206.9±2.7	93.0±1.2	0.09	0.6	0.8
West Village	6	Average	137.0±1.9	61.9±0.8	0.08	0.4	0.5
		Min	114.2±1.6	51.3±0.7	0.06	0.3	0.4
		Max	168.5±2.2	76.9±1.0	0.11	0.5	0.6
Rock dumps	8	Average	733.3±26.1	339.8±12.2	0.42	2.0	3.5
		Min	148.2±13.8	71.0±4.8	0.09	0.4	0.5
		Max	3964.9±89.5	1825.8±41.3	2.24	10.7	20.7
Overall Average for Mining Area			677.7±43.8	312.9±20.3	0.38	1.8	3.4
Overall Min for Mining Area			107.4±1.5	48.0±0.7	0.06	0.3	0.4
Overall Max for Mining Area			3964.9±89.5	1825.8±41.3	2.24	10.7	20.7
Worldwide Average Range			370	59 18-93	0.48	<1	<1

The Ra_{eq} average values for the mining area and the control area ranged from 107.4 ± 1.5 to $3964.9.9 \pm 89.5$ Bq.kg⁻¹ with an average of 677.7 ± 43.8 Bq.kg⁻¹ and from 76.4 ± 1.4 to 103.8 ± 1.5 Bq.kg⁻¹ with an average of 86.9 ± 1.4 Bq.kg⁻¹ respectively. The minimum values were largely as a result of measurements from the East and West Villages. Maximum values were recorded from rock dumps and from tailings three. In most of the soil samples from the mining area, Ra_{eq} values were above the worldwide value of 370 Bq.kg⁻¹ as recommended by IAEA under normal circumstances (OECD, 1979) rendering the area around the mine tailings unsafe. This was attributed mostly to the high levels of ²³⁸U found in the mine tailings and rock dumps.

Table 5.3 also shows that the average outdoor absorbed dose rate in air from terrestrial gamma rays was found to be 312.9 ± 2.3 nGy.h⁻¹. This value was much higher than the worldwide average of 59 nGy.h⁻¹ (UNSCEAR, 2000). However, from the control area, a value of 41.9 ± 0.7 nGy.h⁻¹ was calculated, which is lower than the worldwide average.

The AEDE values for the mining area and the control area were also calculated. These values were found to be in the range 0.06 to 2.24 mSv.y⁻¹ with an average of 0.38 mSv.y⁻¹, 0.05 to 0.06 mSv.y⁻¹ with an average of 0.05 mSv.y⁻¹, respectively. Although some AEDE values were below the worldwide average of 0.48 mSv (UNSCEAR, 2000), it is also important to note that there were also some samples above the worldwide average. The International Commission on Radiation Protection (ICRP) recommends an AEDE limit of 1 mSv.y⁻¹ for individual members of the public and 20 mSv y⁻¹ for radiation workers (ICRP, 2008). In South Africa, the dose constraint applicable to the average member of a critical group from a single source within the exposed population is 0.25 mSv per annum (NNR, 2013). This means that the AEDE average values from the mining area were considered unsafe to the population, but those from the control area were below the regulatory limit.

Calculated values of hazard indices for soil samples from mining area ranged from 0.3 to 10.7 with an average of 1.8 and from 0.4 to 20.7 with an average of 3.4 for the external hazard (H_{ex}) and internal hazard (H_{in}) respectively. This shows that the

average values for H_{ex} and H_{in} were higher than unity, posing a significant radiological threat to the population in the area.

5.5. Indoor Radon and Annual Effective Dose from Inhalation Measured from Selected Mine Dwellings and from the Control area

Table 5.4 shows activity concentrations of Indoor radon from selected mine dwellings and from the control area. Measurements were taken over night for a period of 24 hours. Table 5.4 also presents calculated values for the corresponding annual effective dose to which members of the public are exposed. According to ICRP (2008), the international limit of indoor radon is 200 Bq/m^3 . The results in Table 5.4 indicate that the average activity concentrations of indoor radon from the mining area is 119.5 Bq/m^3 , a value much higher than the average 19.7 Bq/m^3 from the control area. This was because ^{238}U is the source of Radon-222, which had a high activity concentration in the area. Although the average value from the mining area was lower than the international limit, some values were higher in some dwellings. The activity concentrations of indoor radon from the mining area ranged from 1.0 to 472.0 Bq/m^3 , compared to a range of 0.1 to 35.0 Bq/m^3 from the control area. The annual effective doses from inhalation of radon corresponding to the activity concentrations were calculated using Equation 4.9 and the results are also indicated in Table 5.4. The annual effective dose ranged from 0.03-11.89 mSv with a mean value of 3.01 mSv. These dose values were higher than the results estimated from the control area that ranged from 0.00-0.88 mSv with an average of 0.5 mSv. The results were also significantly higher than those published for normal areas around the world. According to UNSCEAR (2000), the average annual effective dose from inhalation of radon and its decay products is 1.26 mSv.

Table 5.4: Activity concentrations of Indoor radon and the corresponding annual effective dose from Inhalation measured from selected mine dwellings and control area.

Location	Source	Parameter	Indoor radon activity concentrations (Bq/ m ³)	Average annual effective dose (mSv)
Control area	Dwelling 1	Range	2.5-22.5	0.06-0.57
		Average	12.1	0.30
	Dwelling 1	Range	0.1-35.0	0.00-0.88
		Average	27.3	0.69
Overall control area		Range Average	0.1- 35.0 19.7	0.00-0.88 0.50
Mining area	West Village Dwelling 1	Range	23.1-194.0	0.58-4.89
		Average	77.6	1.96
	West Village Dwelling 1	Range	3.3-177.9	0.08-4.48
		Average	56.6	1.43
	East Village Dwelling 1	Range	34.2-472.0	0.86-11.89
		Average	256.5	6.46
	East Village Dwelling 2	Range	1.0-91.5	0.03-2.31
		Average	28.5	0.72
	East Village Dwelling 3	Range	12.0-386.4	0.3-9.74
		Average	178.2	4.49
Overall mining area		Range Average	1.0-472.0 119.5	0.03-11.89 3.01

Results from indoor radon measurements taken from the mining area and from the control area have shown that levels reach their maximum in the early morning and their minimum at noon. This is illustrated in Figure 5.5, where fluctuations in indoor radon activity concentrations were recorded from one of the mine dwellings (East Village Dwelling 1). This was also supported by UNSCEAR (1982), who also stated that radon concentrations in the air vary daily and seasonally. Maximal concentrations occur in late summer, and minimal levels in winter. The availability of radon inside dwellings also depends on (i) the concentration of radon in the soil (ii) the rate of extraction of soil gas and (iii) the pressure difference needed to extract it (UNSCEAR, 1982). Inside dwellings, low rates of air exchange can result in a build-up of radon and its decay products to levels much higher than those typically observed outdoors.

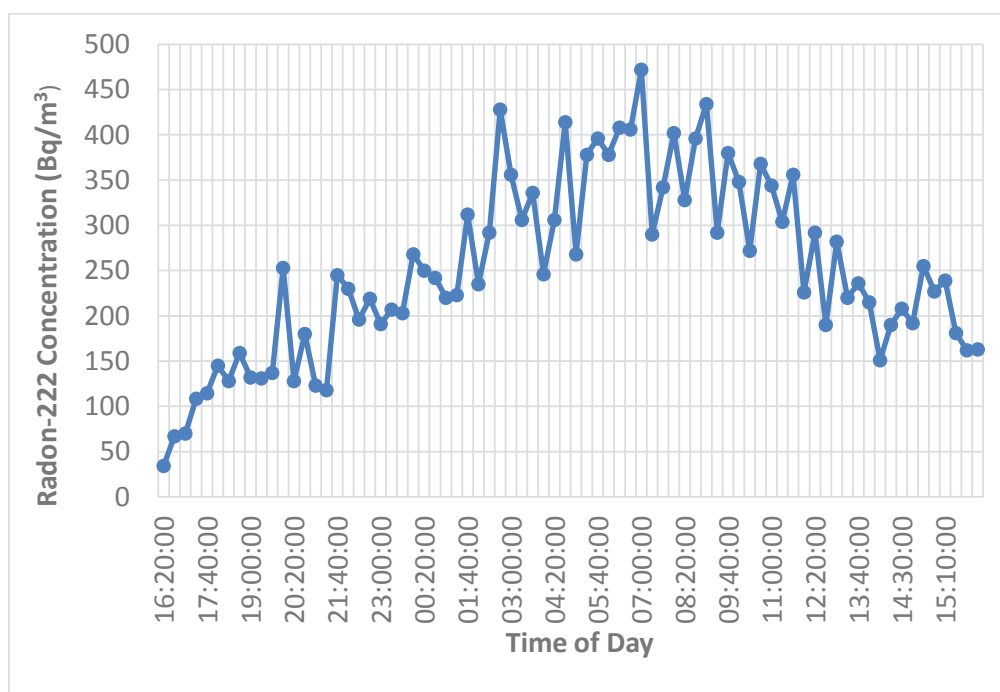


Figure 5.5: Variation of Indoor Radon Concentration in Bq/m³ with time of Day in Dwelling 1 in the East Village of the Mining Area Measured on 13th of May 2015.

5.6. Activity Concentrations of ²³⁸U, ²³²Th and ⁴⁰K for Plant Samples

The results presented in Table 5.5 show average activity concentrations of ²³⁸U, ²³²Th and ⁴⁰K for plant samples analysed from the gold mining area and from the control area. These values were derived from the weighted average of selected gamma energy peaks of their daughter radionuclides (Table 3.1) in each plant sample. For detailed description of the sample identity (ID), refer to Appendix A (Table AB). The average activity concentration of ²³⁸U in Bq.kg⁻¹ was found to be 17.4±3.1. For ²³²Th the average activity concentration was 19.7±1.6 and that of ⁴⁰K was 146.7±9.2 Bq.kg⁻¹. On the other hand, the average activity concentrations in Bq.kg⁻¹ for ²³⁸U, ²³²Th, and ⁴⁰K from the control area were 12.1±1.5, 15.6±1.0 and 125.7±4.8, respectively.

Table 5.5: Average activity concentrations of ^{238}U , ^{232}Th and ^{40}K for Plant Samples from the Mining Area and the Control Area.

Location	Sample ID	Latitude	Longitude	Average (Bq.kg^{-1})		
				^{238}U	^{232}Th	^{40}K
Control area	CV1	25°47'53.82"S	25°38'1.92"E	12.1±1.5	15.6±1.0	125.7±4.8
	VV91	26°23'13.80"S	27°30'59.76"E	37.7±6.9	32.4±2.3	248.1±9.6
Mining area	VV92	26°23'13.80"S	27°30'59.76"E	24.6±4.3	35.6±3.1	214.8±8.7
	VV101	26°23'10.38"S	27°31'15.24"E	14.9±2.4	14.9±1.2	120.5±4.9
	VV102	26°23'10.38"S	27°31'15.24"E	17.7±3.1	23.7±2.0	147.8±8.6
	VV11	26°23'6.84"S	27°30'53.04"E	14.3±2.2	17.0±1.5	150.3±6.0
	VV14	26°23'23.46"S	27°31'11.70"E	7.4±1.7	11.8±1.0	79.9±3.5
	VV15	26°23'16.68"S	27°30'44.10"E	11.6±2.0	12.5±1.2	100.7±3.8
	VV17	26°23'27.90"S	27°31'0.06"E	12.6±2.1	11.5±0.8	118.5±4.4
	DSV181	26°23'0.12"S	27°30'14.10"E	15.6±2.8	17.5±1.6	141.2±5.3
	DSV3615	26°20'28.44"S	27°25'42.06"E	17.3±3.0	19.9±1.7	145.2±5.5
	Average Mining Area			17.4±3.1	19.7±1.6	146.7±9.2
	Minimum Mining Area			7.4±1.7	11.5±0.8	79.9±3.5
	Maximum Mining Area			37.7±6.9	35.6±3.1	248.1±9.6

The results in Table 5.5 showed that activity concentration of ^{238}U was highest in leafy vegetable samples VV91 followed by VV92. This is clearly illustrated in Figure 5.5. This was expected as leafy vegetables usually have relatively higher activity concentrations compared to non-leafy and fruit vegetables. According to UNSCERAR (2000), the acceptable world reference for ^{238}U in leafy vegetables is 20 mBq.kg^{-1} . The measured values were higher than the acceptable limit. The least concentration of ^{238}U was found in fruit sample VV14, which is from a lime plant. Although the value was the lowest, it was still significantly higher than a world reference of 3 mBq.kg^{-1} for fruit samples (UNSCEAR, 2000). For ^{232}Th , all the values were higher than the world reference of 15 mBq.kg^{-1} for leafy vegetables and 0.5 mBq.kg^{-1} for fruit samples, respectively. It was also observed that the activity concentration of ^{40}K markedly exceeded that of both ^{238}U and ^{232}Th , as it is the most abundant radioactive element in normal soil and is highly soluble. High levels of ^{40}K in all plant samples were also expected since it is a macronutrient for plants. The measured values were also comparable to the ones derived from the control area.

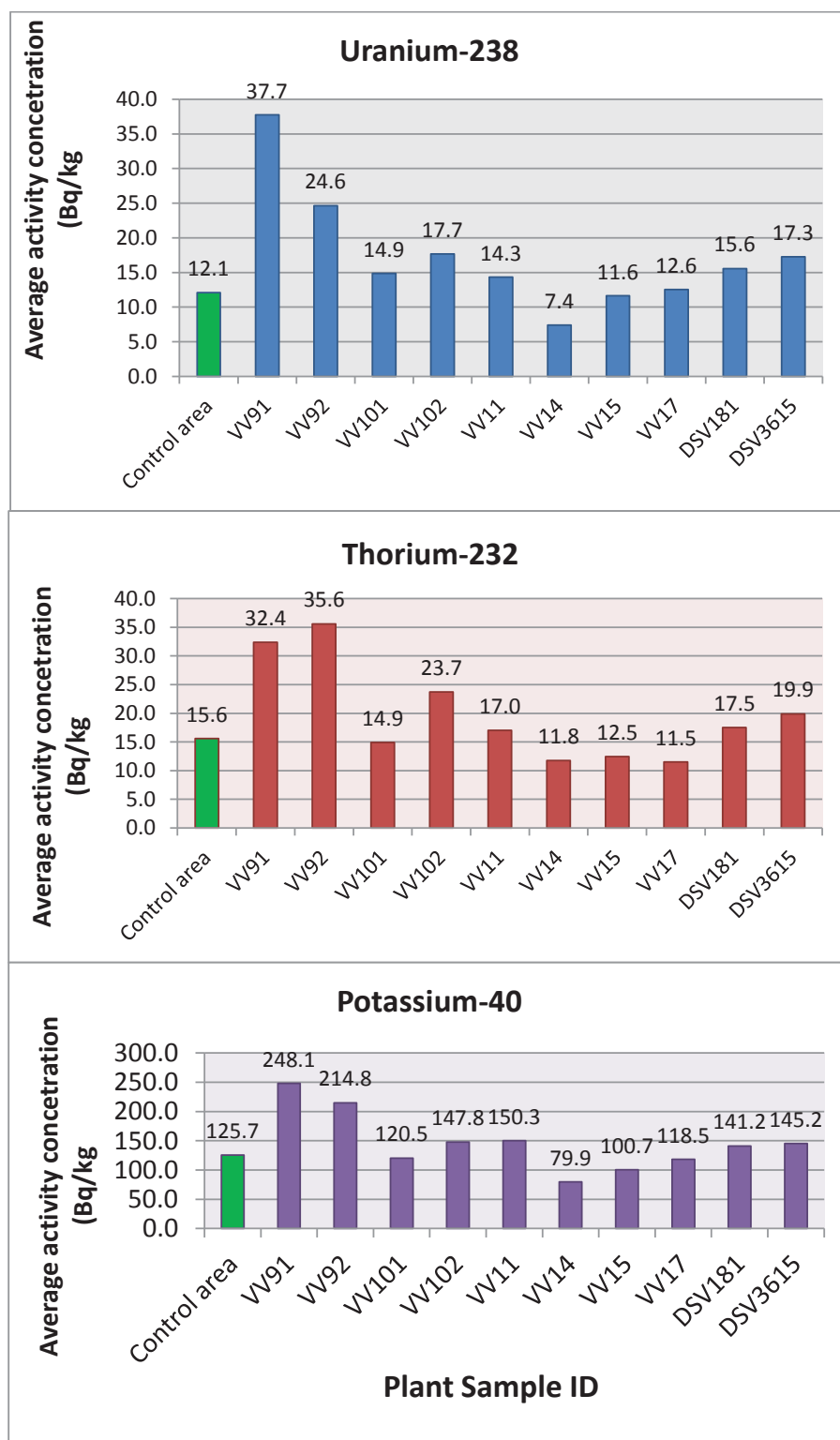


Figure 5.6: Comparison of Average Activity Concentrations of ^{238}U , ^{232}Th and ^{40}K for Plant Samples from the Mining Area and from the Control Area.

5.7. Radiological Hazard Assessment for Plant Samples

Radiological hazard assessment for the analyzed plant samples was also considered and the results are shown in Table 5.6. Equations 4.5, 4.6, 4.7, 4.10 and 4.11 in chapter 4 were used to calculate Ra_{eq} , D , AEDE, H_{ex} and H_{in} parameters, respectively.

Table 5.6: Results of calculated Absorbed Dose Rate in air (D), Annual Effective Dose Equivalent (AEDE), Radium Equivalent Activity (Ra_{eq}), External Hazard Index (H_{ex}) and Internal hazard Index (H_{in}) of Plant Samples from the Mining Area and from the Control Area.

Location	Sample ID	Ra_{eq} (Bq.kg ⁻¹)	D (nGy.h ⁻¹)	AEDE (mSv.h ⁻¹)	H_{ex}	H_{in}
Control area	CV1	44.1±2.1	20.2±0.9	0.02	0.1	0.2
Mining area	VV91	103.1±7.7	47.3±3.5	0.06	0.3	0.4
	VV92	92.0±6.2	41.8±2.8	0.05	0.2	0.3
	VV101	45.5±3.0	20.9±1.3	0.03	0.1	0.2
	VV102	63.0±4.3	28.7±1.9	0.04	0.2	0.2
	VV11	50.3±3.1	23.2±1.4	0.03	0.1	0.2
	VV14	30.4±3.5	13.9±1.8	0.02	0.1	0.1
	VV15	37.2±2.7	17.1±1.2	0.02	0.1	0.1
	VV17	38.2±2.4	17.7±1.1	0.02	0.1	0.1
	DSV181	51.5±3.6	23.7±1.6	0.03	0.1	0.2
	DSV3615	56.9±3.9	26.1±1.7	0.03	0.2	0.2
Average Mining Area		56.8±4.0	26.0±1.8	0.03	0.2	0.2
Minimum Mining Area		30.4±2.4	13.9±1.8	0.02	0.1	0.1
Maximum Mining Area		103.1±7.7	47.3±3.5	0.06	0.3	0.4

The average value of Ra_{eq} in Bq.kg⁻¹ from the mining area was 56.8±4.0 with a range of 30.4±2.4 to 103.1±7.7. Ra_{eq} from the control area was 44.1±2.1 Bq.kg⁻¹, which is a figure lower than the average value from the mining area. From the results, it can be seen that higher values of Ra_{eq} were recorded from leafy vegetables.

Table 5.6 also shows that the outdoor average absorbed dose rate in air (D) due to ²³⁸U, ²³²Th, and ⁴⁰K in plant samples varied from 13.9±1.8 to 47.3±3.5 nGy.h⁻¹ with an average of 26.0±1.8 nGy.h⁻¹. This is also below the worldwide average of 59

nGy.h⁻¹ (UNSCEAR, 2000). This D value was still higher than the average value obtained from the control area (20.2±0.9 nGy.h⁻¹).

AEDE values for the mining area and the control area were also calculated. From the mining area, these values were found to be in the range 0.02 to 0.06 mSv.y⁻¹ with an average of 0.03 mSv y⁻¹. An average value of 0.02 mSv y⁻¹ from the control area was recorded. These AEDE values were way below the worldwide average of 0.48 mSv for external terrestrial radiation (UNSCEAR, 2000), and also lower than 1 mSv.y⁻¹, a limit recommended by the International Commission on Radiation Protection (ICRP) for individual members of the public (ICRP, 2008).

The external hazard (H_{ex}) index estimated for plant samples from the mining area ranged from 0.1 to 0.3 with an average of 0.2 while the internal hazard (H_{in}) index ranged from 0.1 to 0.4 with an average of 0.2. These average values for both quantities were less than unity, making plant samples radiologically safe to the population in the area. It must also be stated that their effect may become significant with the cumulative contribution from other sources.

5.8. Activity Concentrations of ²³⁸U, ²³²Th and ⁴⁰K for 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 (Table 3.1) in each water sample are shown in Table 5.7. A detailed description of the water sampling points is shown in Appendix AC. 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 boreholes 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.

The results also showed that the average activity concentration of ^{232}Th in the water samples from the control area was higher than average activity concentration of ^{238}U for water samples from the control area. Activity concentrations of the radionuclides in water samples from the study area were compared with South African Guidelines (DWAF, 2002). The average activity concentrations of ^{238}U and ^{232}Th in all the samples were higher than the recommended values of 1.0 and 0.05 mBq.L^{-1} , respectively. ^{40}K is a very soluble and essential element for metabolism in living organisms. Its value was high as expected.

Table 5.7: Average Activity Concentrations of ^{238}U , ^{232}Th and ^{40}K for Water Samples from the Mining Area and from the Control Area.

Location	Sample ID	Latitude	Longitude	Average (Bq.L^{-1})		
				^{238}U	^{232}Th	^{40}K
Control area	CW1	25°47'33.06"S	25°38'2.04"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°38'3.36"E	ND	0.31±0.01	9.18±0.76
	CW4	25°47'59.70"S	25°37'55.20"E	0.10±0.00	0.37±0.02	10.12±1.13
	CW5	25°48'1.32"S	25°37'56.22"E	0.20±0.00	0.44±0.02	11.61±1.18
	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
Mining area	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°26'1.02"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°23'3.36"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
	DSW43/19	26°21'57.30"S	27°26'34.98"E	0.77±0.04	0.66±0.03	7.28±0.62
	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°29'2.04"E	0.72±0.04	0.62±0.03	5.26±0.53

WV4	26°23'38.88"	27°29'5.16"E	0.48±0.02	0.55±0.03	5.54±0.56
WV5	26°23'40.20"S	27°29'5.52"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

5.9. Radiological Hazard Assessment for Water Samples

Table 5.8 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 R_{eq} in $Bq.L^{-1}$ from the mining area was 2.03 ± 0.07 . This was significantly higher than $1.39 \pm 0.08 Bq.L^{-1}$, a figure estimated from the control area. The results also indicated that the maximum values were observed from water samples 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 outdoor average absorbed dose rate due to ^{238}U , ^{232}Th , and ^{40}K in water samples varied from 0.54 ± 0.02 to $1.54 \pm 0.05 nGy.h^{-1}$ with an average of $0.95 \pm 0.03 nGy.h^{-1}$. This was also below the worldwide average of $59 nGy.h^{-1}$ (UNSCEAR, 2000). The average value of the absorbed dose rate as a result of water samples was however, lower than the value obtained from the control area ($0.68 \pm 0.04 nGy.h^{-1}$).

AEDE values for water samples as a result of exposure to external radiation were also calculated for the mining area and the control area. From the mining area, values ranged from 6.65×10^{-04} to $1.89 \times 10^{-03} mSv.y^{-1}$ with an average of $1.17 \times 10^{-03} mSv.y^{-1}$. An average value of $8.39 \times 10^{-04} mSv.y^{-1}$ 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 $0.48 mSv$ 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 4.5, 4.6, 4.7, 4.10 and 4.11 in chapter 4 were used to calculate Ra_{eq} , D, AEDE, H_{ex} and H_{in} parameters, respectively.

Table 5.8: Results of calculated Absorbed Dose Rate in air (D), Annual Effective Dose Equivalent (AEDE), Radium Equivalent Activity (Ra_{eq}), External Hazard Index (H_{ex}) and Internal hazard Index (H_{in}) of water samples from the mining area and from the control site.

Location	Sample ID	Ra_{eq} (Bq.L ⁻¹)	D (nGy.h ⁻¹)	AEDE (mSv.h ⁻¹)	H_{ex}	H_{in}
Control area	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
	CW4	1.41±0.09	0.69±0.05	8.48E-04	3.80E-03	4.07E-03
	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
Mining area	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
	DSW39/17	2.08±0.07	0.97±0.04	1.18E-03	5.61E-03	7.61E-03
	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

WHO recommends a value of 0.8 mSv.y^{-1} as the upper limit for the annual effective dose in water (WHO, 2008). The average annual effective dose for the water samples in the study area was lower than the set limit. The pH is also an important water quality parameter as it affects solubility of radionuclides as well as heavy metals (WHO, 2008). Solubility of these contaminants increases with decreasing pH making their concentrations higher. South Africa requires a pH range of 6.5-8.5 for drinking water (WHO, 2008). All the water samples collected had a pH within the acceptable range.

5.10. Annual Effective Dose from Ingestion of ^{238}U , ^{232}Th and ^{40}K through Water and Plant Samples.

The annual effective dose of natural radionuclides through the ingestion of water and plant samples were also estimated using Equation 4.8 as described earlier. The average activity concentrations of ^{238}U , ^{232}Th and ^{40}K in plant and water samples from Tables 5.6 and 5.7 were used to calculate the annual effective dose as presented in Table 5.9.

Table 5.9: Annual Effective Dose through ingestion of ^{238}U , ^{232}Th and ^{40}K in Water and Plant Samples.

Sample type	Average Activity Concentration			Average Annual Effective Dose (mSv.y^{-1})
	^{238}U	^{232}Th	^{40}K	
Plant	17.4	19.7	146.7 (Bq.kg^{-1})	1.24E-03
Water	0.66	0.56	7.36 (Bq.L^{-1})	1.23E-04
Total	18.1	20.3	154.1	1.37E-03

5.11. Summary of the Annual Effective Dose from Soil, Plant, Water and Radon of each Individual Member of the Public

The total annual effective dose of an individual member of the public was determined by summing all the individual equivalent doses for the exposure pathways considered for the study area. Table 5.10 shows the breakdown from samples collected.

Table 5.10: Breakdown of Total Annual Effective Dose from Soil, Plant Samples, Water and Radon to each Individual Member of the Public.

Number	Exposure Pathway	Calculated Average Annual Effective Dose (mSv.y ⁻¹)	(UNSCEAR, 2000) Reference
1	External irradiation of ²³⁸ U, ²³² Th and ⁴⁰ K due to soil, plant and water samples	0.41	0.48
2	Ingestion of ²³⁸ U, ²³² Th and ⁴⁰ K through water and plant samples	1.37×10 ⁻³	0.29
3	Inhalation of indoor radon	3.01	1.26
Total Annual Effective Dose		3.42	2.4

According to UNSCEAR (2000), the total annual effective dose from natural radiation should be 2.4 mSv.y⁻¹. This also includes radiation from cosmic sources, which was not considered in the study. The total annual effective dose from the measured samples was 3.42, which was higher than the worldwide average of 2.4 mSv.y⁻¹ and also higher than 1 mSv.y⁻¹, a limit recommended by ICRP (2008) for individual members of the public. The highest contribution came from inhalation of radon, while ingestion through water and plant samples contributed the least. Annual effective dose via external irradiation of ²³⁸U, ²³²Th and ⁴⁰K came second with a greater contribution coming from soil samples.

5.12. Cancer Risk due to ^{238}U , ^{232}Th and ^{40}K using RESRAD-OFFSITE Computer Code.

RESRAD-OFFSITE Computer Code was used to estimate the cancer risk for individuals living in the mining area. Input parameters from Table 4.3 were used in the calculation. Table 5.11 shows cancer risks for all the pathways summed for the duration of 100 years. The maximum total cancer risk for all the pathways was 6.52×10^{-5} at year 20. This progressively decreased to 6.09×10^{-5} at year 100. ^{232}Th was driving the risk effect followed by ^{238}U and then lastly ^{40}K . This is clearly illustrated in Figure 5.7. The US Environmental Protection Agency considers acceptable for regulatory purposes a cancer risk in the range of 1×10^{-6} to 1×10^{-4} (USEPA, 1989). On the other hand, South Africa considers the individual cancer risk limit for the public to be 5×10^{-6} (Government Gazette, 2006). This means that total cancer risks from year 0 to year 100 were higher than acceptable values according to South African risk limit.

Table 5.11: Individual Cancer Risk from ^{238}U , ^{232}Th and ^{40}K from Gold Mine for all the pathways summed for a duration of 100 years.

Year	Individual Cancer Risk			Total
	Nuclides			
	⁴⁰ K	²³² Th	²³⁸ U	
0	1.84E-06	4.55E-05	7.51E-06	5.49E-05
1	9.28E-07	4.75E-05	7.37E-06	5.58E-05
5	6.23E-08	5.35E-05	6.82E-06	6.04E-05
10	2.11E-09	5.76E-05	6.20E-06	6.38E-05
15	7.44E-11	5.93E-05	5.63E-06	6.50E-05
20	2.51E-12	6.01E-05	5.12E-06	6.52E-05
25	8.51E-14	6.03E-05	4.65E-06	6.50E-05
30	2.92E-15	6.04E-05	4.22E-06	6.46E-05
35	1.11E-16	6.04E-05	3.83E-06	6.43E-05
40	7.88E-18	6.04E-05	3.48E-06	6.39E-05
45	1.57E-18	6.03E-05	3.16E-06	6.35E-05
50	4.79E-19	6.03E-05	2.87E-06	6.32E-05
55	1.55E-19	6.02E-05	2.61E-06	6.29E-05
60	5.15E-20	6.02E-05	2.37E-06	6.26E-05
65	1.68E-20	6.01E-05	2.16E-06	6.23E-05
70	5.50E-21	6.01E-05	1.96E-06	6.20E-05
75	1.80E-21	6.00E-05	1.78E-06	6.18E-05
80	5.95E-22	6.00E-05	1.62E-06	6.16E-05
85	1.95E-22	5.99E-05	1.47E-06	6.14E-05

90	6.36E-23	5.99E-05	1.33E-06	6.12E-05
95	2.08E-23	5.98E-05	1.21E-06	6.10E-05
100	6.79E-24	5.98E-05	1.10E-06	6.09E-05

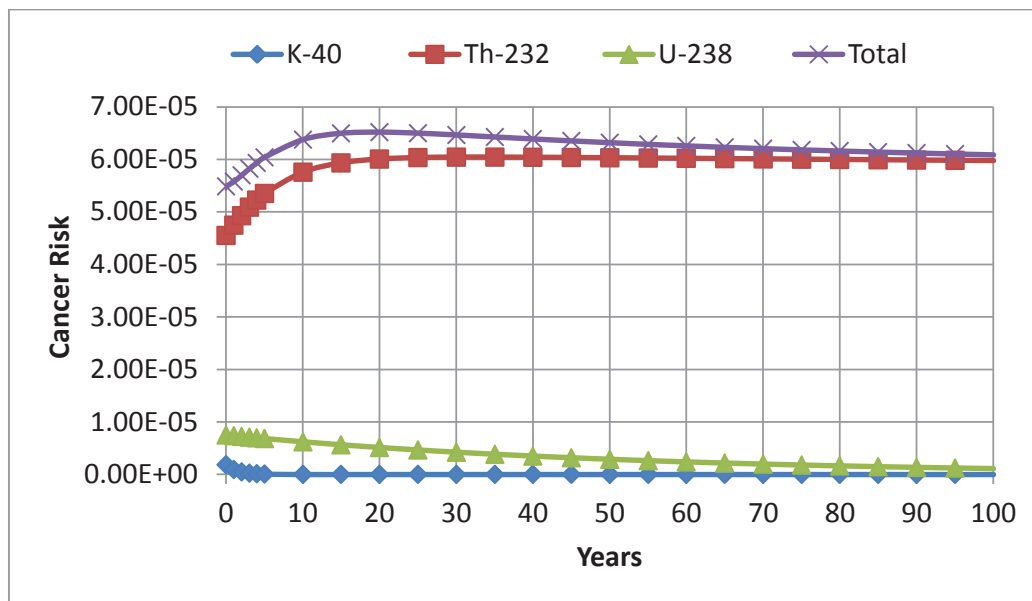


Figure 5.7: Individual Cancer Risk from ^{238}U , ^{232}Th and ^{40}K Summed over all Pathways Summed for a duration of 100 years.

From the simulation model, it was also discovered that the water independent pathway is the most significant pathway in calculating the risk. In this pathway, the cancer risk is largely caused by direct radiation from the soil followed by radon, and then inhalation followed by soil ingestion. The least component was the plant pathway. Figure 5.8 shows the cancer risk summed up based on component pathways for duration of 100 years following contamination.

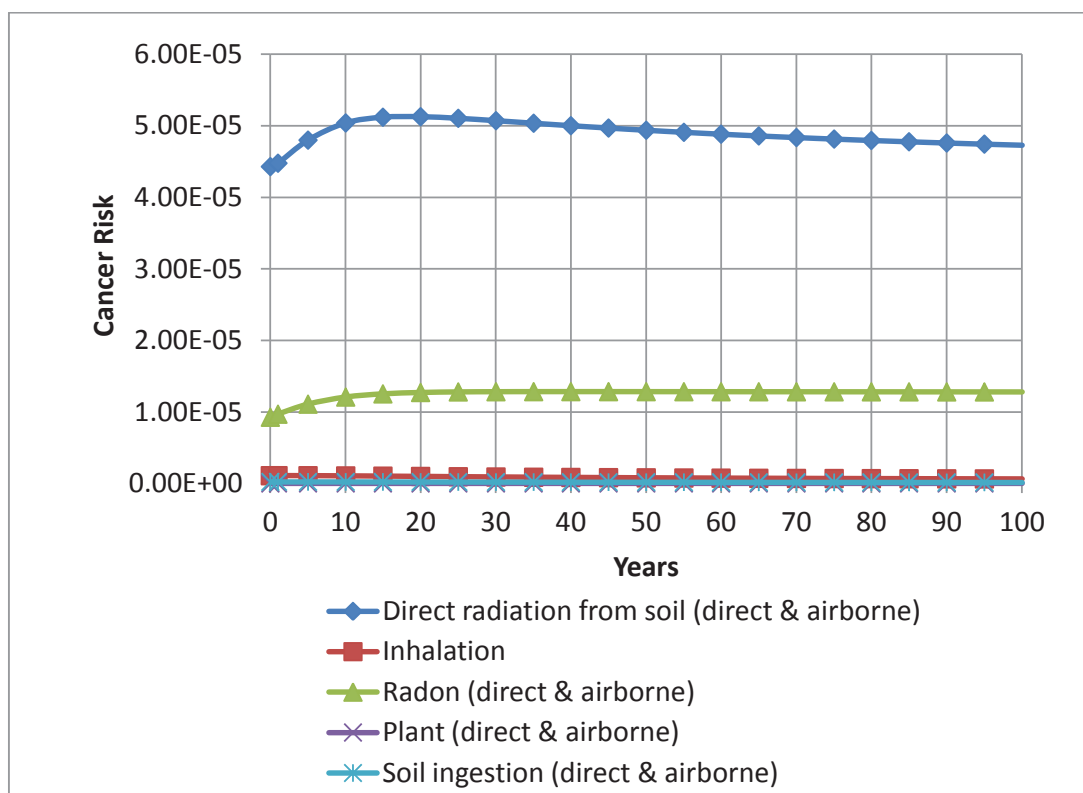


Figure 5.8: Individual Cancer Risk from ^{238}U , ^{232}Th and ^{40}K summed based on component pathways over a duration of 100 years.

5.13. Concentrations of Heavy Metals in Soil, Plant Samples, and Water from the Gold Mining Area and Control Area

The concentrations of heavy metals in mg.kg^{-1} from the different locations of the gold mining area and from the control area in soil, plant, and water samples are presented in this section.

5.13.1. Average Concentrations of Heavy Metals in Soil Samples

The average concentrations of heavy metals in mg.kg^{-1} from the different locations in soil from the mine and from the control area are presented in Table 5.12. Detailed results from the ICP-MS measurements can be found in Appendix B. These average concentrations in soil samples were used to calculate the average daily intakes for non-carcinogenic and carcinogenic risk assessment.

The results presented showed that the average concentrations of the heavy metals in soil from the gold mining area varied significantly and decreased in the order of Cr>Ni>As>Zn>Cu>Co>Pb>Hg>Cd. Figure 5.9 illustrates this observation. The average values in mg.kg^{-1} were as follows: Cr (316.88 within a range of 77.50-861.67); Ni (115.87 in a range of 68.33-152.50); As (77.25 within a range of 62.23-115.19); Zn (68.01 within a range of 21.82- 85.00); Cu (50.79 within a range of 19.09- 108.75); Co (27.52 within a range of 11.82- 41.25); Pb (4.69 within a range of 1.58-10.22); Hg (0.09 within a range of 0.06-0.13); and Cd (0.05 within a range of 0.04 to 0.08), respectively. These results also indicated that mine tailings had generally higher concentrations of heavy metals compared to the soil from the mine villages. Compared with the control area, all the values of heavy metal concentrations from the mining area were higher than those from the control area. This may have been caused by high levels of acid mine drainage that dissolves heavy metals into the ground.

Table 5.12: Average Concentrations of Heavy Metals in Soil from the Mining Area and the Control Area.

Location	No of samples	Heavy Metal Concentrations (mg.kg ⁻¹)								
		As	Pb	Hg	Cd	Cr	Cu	Zn	Co	Ni
Control area	10	2.50	2.34	0.00	0.00	44.72	8.62	14.17	5.80	0.00
Tailings one	11	94.17	8.85	0.13	0.05	441.52	46.78	46.15	33.68	131.04
Tailings two	13	115.19	10.22	0.13	0.05	270.76	45.48	51.95	31.76	115.08
Tailings three	8	71.33	2.31	0.06	0.05	77.50	46.25	82.50	30.00	152.50
Tailings four	12	73.18	2.96	0.07	0.04	104.17	55.83	60.00	21.67	99.83
Tailings five	12	67.08	3.31	0.06	0.05	97.50	47.50	48.33	21.67	125.83
Rock dumps	8	62.23	3.94	0.08	0.08	583.75	108.75	185.00	41.25	142.50
West village	6	65.17	1.58	0.10	0.05	861.67	36.67	48.33	28.33	68.33
East village	11	69.69	4.32	0.06	0.05	98.18	19.09	21.82	11.82	91.82
Average		77.25	4.69	0.09	0.05	316.88	50.79	68.01	27.52	115.87
Minimum		62.23	1.58	0.06	0.04	77.50	19.09	21.82	11.82	68.33
Maximum		115.19	10.22	0.13	0.08	861.67	108.75	185.00	41.25	152.50

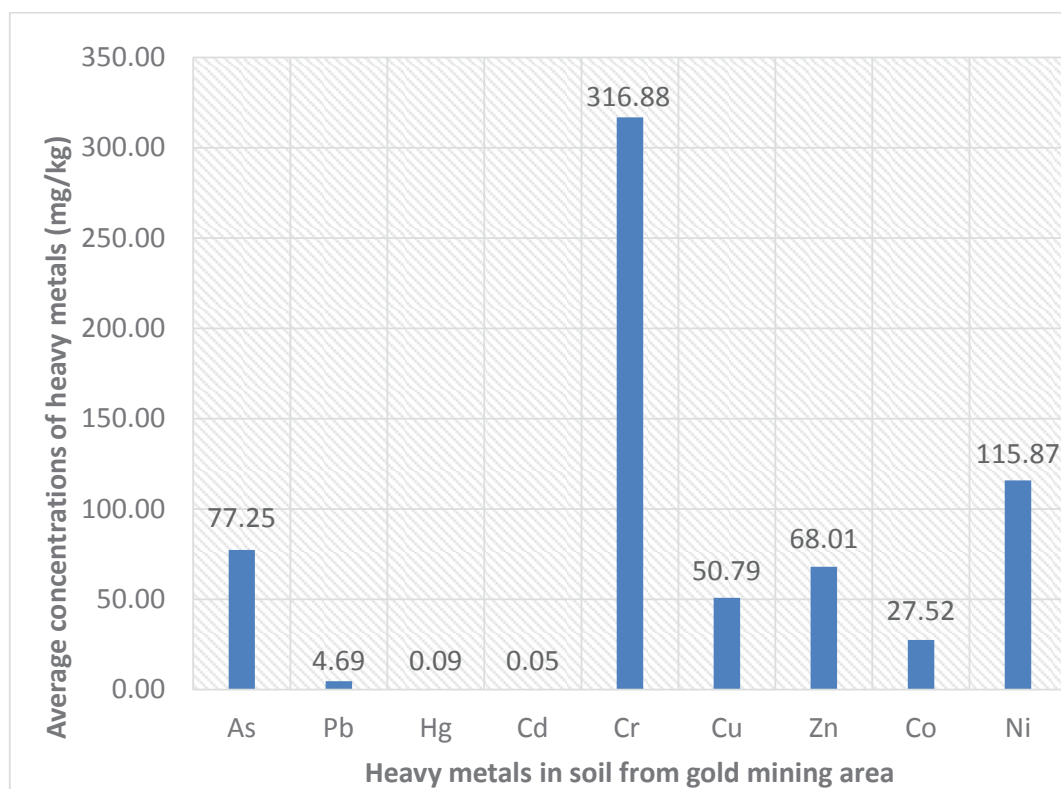


Figure 5.9: Average Concentrations of Heavy Metals in Soil from the Gold Mining Area.

The results measured were also compared with recommended maximum allowable limits for South Africa and other countries as shown in Table 5.13. As and Cr were found to be the highest in the present study. However, Pb, Cd, Zn and Co concentrations were lower than the maximum allowable limits while Hg, Cu, and Ni were comparable with other countries. For As and Cr, a graph was drawn to compare the values with those from other countries. Figure 5.10 illustrates this.

Table 5.13: Maximum Allowable Limit of Heavy Metal Concentrations in Soil for Different Countries (Kamunda et al., 2016b).

Country	Concentration Limit (mg.kg ⁻¹)									References
	As	Pb	Hg	Cd	Cr	Cu	Zn	Co	Ni	
Germany	50	70.0	0.5	1.0	60.0	40.0	150.0	n.a	50.0	(Dar-Yuan and Chia-Hsing 2011)
Poland	n.a	100	n.a	3	100	100	300	50	100	(Mtunzi et al., 2015)
UK	32	450	10	10	130	n.a	n.a	n.a	130	(CLEA, 2009)
Australia	20	300	1	3	50	100	200	n.a	60	(EPAA, 2012)
Taiwan	60	300	2	5	250	200	600	n.a	200	(Dar-Yuan and Chia-Hsing, 2011)
Bulgaria	10	26	0.03	0.4	65	34	88	20	46	(Atanasov, 2007)
Canada	20	200	0.8	3	250	150	500	n.a	100	(CME, 2009).
China	30	80	0.7	0.5	200	100	250	n.a	50	(EPMC, 2015)
Tanzania	1	200	2	1	100	200	150	n.a	100	(He et al., 2015)
FAO/WHO	20	100	n.a	3	100	100	300	50	50	(Chiroma et al., 2014)
Guidelines	20	100	n.a	3	100	100	300	50	50	
EU	n.a	300	n.a	3	150	140	300	n.a	75	(EU, 2002)
Guidelines	n.a	300	n.a	3	150	140	300	n.a	75	
South Africa	5.8	20	0.93	7.5	6.5	16	240	300	91	(DEA, 2010)

n.a: Not available

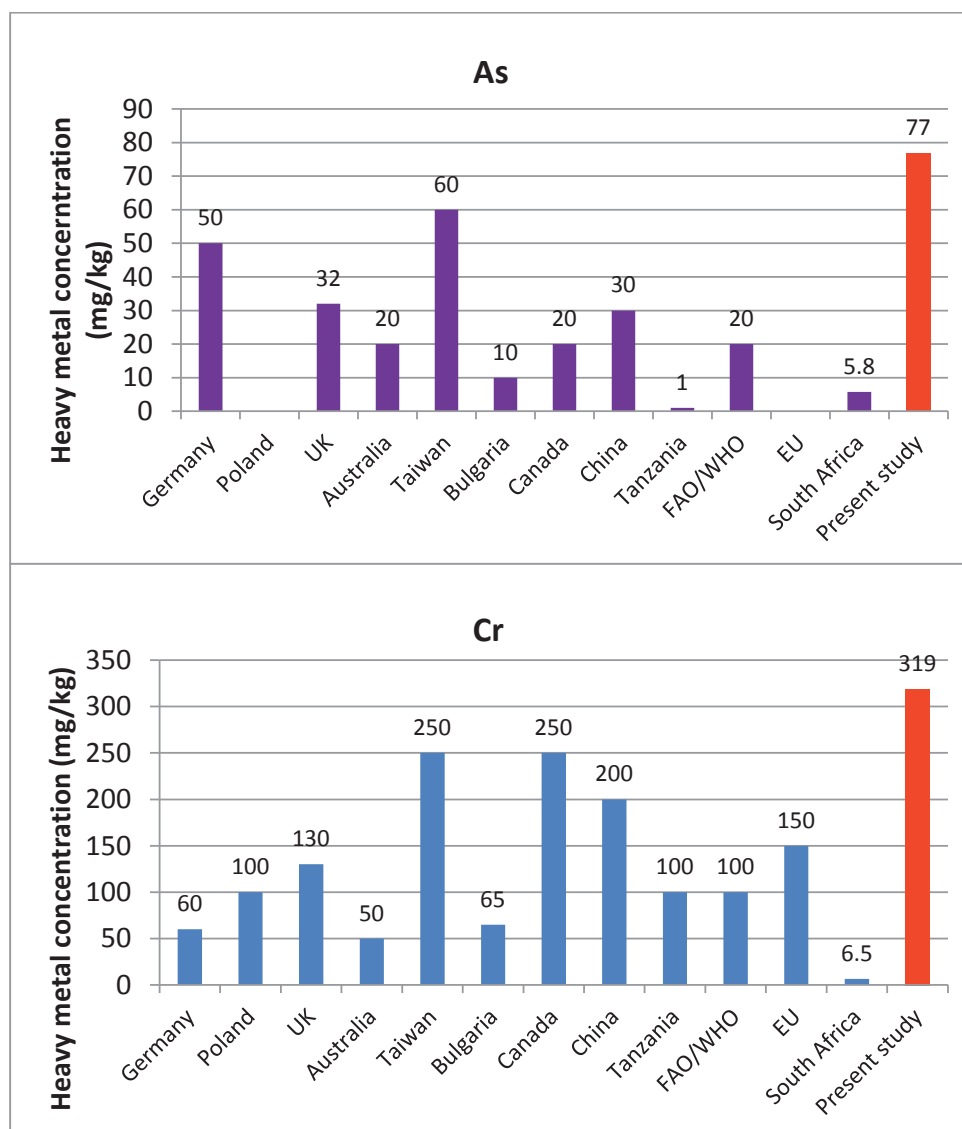


Figure 5.10: Comparison of Average Concentrations of As and Cr in Soil from the Gold Mining Area with Maximum Allowable Limits from other Countries and Regulatory bodies.

5.13.2. Average Concentrations of Heavy Metals in Plant Samples

The average concentration (mg.kg^{-1}) of heavy metals in plant samples from the different locations of the gold mining area and from the control area are presented in Table 5.14. These average concentrations were also used to calculate average daily intakes for non-carcinogenic and carcinogenic risk assessment. The results presented show that the average concentrations of the heavy metals in plant samples from the gold mining area were generally higher than those from the control area. The average concentration of chromium was the highest in the mining area with the least being Hg and Cd. The average concentrations decreased in the order of $\text{Cr} > \text{Ni} > \text{As} > \text{Zn} > \text{Cu} > \text{Co} > \text{Pb} > \text{Hg} > \text{Cd}$. The ranges in mg.kg^{-1} were as follows: Cr (4.20 to 8.30 with an average of 6.90); Ni (0.20 to 6.70 with an average of 2.89); As (2.20 to 2.70 with an average 2.50); Zn (0.20 to 0.80 with an average 0.44); Cu (0.20 to 0.60 with an average 0.32); Co (0.10 to 0.70 with an average 0.28); Pb (0.10 to 0.30 with an average 0.16), respectively. Hg and Cd were not detected in the sample.

The results of the measurements were also compared with national and international guidelines for permissible limits of heavy metals in foodstuffs as presented in Table 5.15. The results indicated that average concentrations of As and Ni in plant samples were higher than FAO/WHO and South African safe limits whilst those of Hg, Cd and Zn were below permissible levels. There were no standards for chromium and cobalt to compare with. The remaining heavy metals were within permissible limits. High levels of As and Ni were as a result of mining activities. Because Ni is an essential metal to plants, it was therefore not surprising to be high. Pb has no known biological importance in plants, hence its concentration was low.

Table 5.14: Concentrations of Heavy Metals in Plant Samples from the Mining Area and from the Control Area.

Location	Sample ID	Latitude	Longitude	Heavy metal concentration (mg.kg ⁻¹)						
				As	Pb	Cr	Cu	Zn	Co	Ni
Control Area	CV1	25°47'53.82"S	25°38'1.92"E	1.30	0.10	3.20	0.40	0.60	0.10	2.70
Mining Area	VV91	26°23'13.80"S	27°30'59.76"E	2.70	0.10	6.10	0.30	0.20	0.30	2.70
	VV92	26°23'13.80"S	27°30'59.76"E	2.50	0.20	4.20	0.20	0.40	0.10	1.10
	VV101	26°23'10.38"S	27°31'15.24"E	2.20	0.10	5.20	0.40	0.60	0.10	3.70
	VV102	26°23'10.38"S	27°31'15.24"E	2.70	0.20	8.30	0.60	0.20	0.20	0.20
	VV11	26°23'6.84"S	27°30'53.04"E	2.60	0.10	7.20	0.40	0.50	0.30	3.40
	VV14	26°23'23.46"S	27°31'11.70"E	2.70	0.20	8.10	0.20	0.30	0.20	2.10
	VV15	26°23'16.68"S	27°30'44.10"E	2.70	0.10	7.20	0.50	0.20	0.20	0.30
	VV17	26°23'27.90"S	27°31'0.06"E	2.20	0.20	8.10	0.20	0.80	0.60	4.30
	DSV181	26°23'0.12"S	27°30'14.10"E	2.50	0.30	7.20	0.20	0.40	0.70	6.70
	DSV3615	26°20'28.44"S	27°25'42.06"E	2.20	0.10	7.40	0.20	0.80	0.10	4.40
Average				2.50	0.16	6.90	0.32	0.44	0.28	2.89
Minimum				2.20	0.10	4.20	0.20	0.20	0.10	0.20
Maximum				2.70	0.30	8.30	0.60	0.80	0.70	6.70

Table 5.15: Permissible Limits of Heavy Metals in Fruit and Vegetables.

Country	Average concentrations of heavy metals fruit and vegetables (mg.kg ⁻¹)									References
	As	Pb	Hg	Cd	Cr	Cu	Zn	Co	Ni	
FAO/WHO	n.a	5.0	n.a	0.01	n.a	0.2	2.0	n.a	0.2	(FAO/WHO, 2003)
South Africa	0.2	0.1	0.5	0.05	n.a	5.0	5.0	n.a	n.a	(DOH, 2004)

n.a: Not available

5.13.3. Concentrations of Heavy Metals in Water Samples

The average concentrations of heavy metals in water samples (mg.L⁻¹) from the different locations of the gold mining area and from the control area are presented in Table 5.16. These average concentrations were used to calculate the average daily intakes for residents leading to non-carcinogenic and carcinogenic risk assessment for individual members of the public. The results presented show that the average concentration of the heavy metals in water from the gold mining area were generally higher than concentrations from the control area. The average concentrations decreased in the order of Ni>Cu> Zn>As> Cr>Co>Pb. The ranges in mg.L⁻¹ were as follows: Ni (0.08 to 2.68 with an average of 0.39); Cu(0.02 to 3.72 with an average of 0.38); Zn (0.00 to 2.12 with an average 0.33); As (0.00 to 0.99 with an average 0.19); Cr (0.00 to 0.40 with an average 0.14); Co (0.00 to 0.66 with an average 0.08); Pb (0.00 to 0.04 with an average 0.01), respectively. Although Hg and Cd were measured, their concentrations were not detectable.

Table 5.16: Concentration of Heavy Metals in Water from the Mining Area and the Control Area.

Location	Sample ID	Latitude	Longitude	Heavy Metals Concentration (mg.L ⁻¹)						
				As	Pb	Cr	Cu	Zn	Co	Ni
Control Area	CW1	25°47'33.06"S	25°38'2.04"E	0.00	0.00	0.02	0.10	0.38	0.00	0.02
	CW2	25°47'34.80"S	25°38'19.20"E	0.00	0.00	0.00	0.12	0.16	0.00	0.00
	CW3	25°47'41.58"S	25°38'3.36"E	0.00	0.00	0.02	0.16	0.18	0.00	0.00
	CW4	25°47'59.70"S	25°37'55.20"E	0.36	0.02	0.24	0.10	0.36	0.00	0.04
	CW5	25°48'1.32"S	25°37'56.22"E	0.00	0.02	0.00	0.10	0.38	0.00	0.02
Average				0.07	0.01	0.06	0.12	0.29	0.00	0.02
Mining Area	DSW451	26°26'19.74"S	27°29'54.84"E	0.25	0.04	0.36	0.12	0.24	0.08	0.26
	DSW112	26°25'28.02"S	27°29'31.50"E	0.20	0.00	0.30	0.08	0.38	0.04	0.18
	DSW183	26°23'0.12"S	27°30'14.10"E	0.31	0.00	0.00	0.08	0.28	0.02	0.18
	DSW204	26°22'45.12"S	27°29'52.44"E	0.02	0.00	0.00	0.02	0.12	0.04	0.24
	DSW405	26°21'43.08"S	27°29'58.86"E	0.99	0.00	0.24	3.72	0.34	0.36	2.26
	DSW406	26°21'43.26"S	27°30'5.28"E	0.74	0.00	0.00	0.04	0.10	0.02	0.24
	DSW407	26°21'53.28"S	27°29'54.54"E	0.86	0.00	0.00	1.40	0.24	0.20	0.94
	DSW358	26°21'49.56"S	27°27'39.90"E	0.74	0.00	0.00	2.42	0.48	0.14	2.68
	DSW179	26°23'22.74"S	27°28'13.92"E	0.00	0.00	0.00	0.04	0.30	0.02	0.14
	DSW4210	26°20'36.24"S	27°26'1.02"E	0.00	0.00	0.00	0.04	0.00	0.02	0.12
	DSW2111	26°21'25.14"S	27°26'10.32"E	0.11	0.00	0.00	0.06	0.40	0.04	0.16
	DSW712	26°21'27.42"S	27°25'59.22"E	0.33	0.00	0.20	0.02	0.22	0.04	0.14
	DAM313	26°21'19.32"S	27°25'52.44"E	0.21	0.00	0.32	0.04	0.32	0.06	0.34
	DSW914	26°20'59.34"S	27°25'52.44"E	0.00	0.00	0.00	0.08	0.18	0.06	0.30
	DSW3615	26°20'28.44"S	27°25'42.06"E	0.00	0.00	0.00	0.04	0.12	0.04	0.26
	DSW3816	26°19'38.10"S	27°24'15.42"E	0.00	0.00	0.00	0.06	0.00	0.04	0.20
	DSW3917	26°18'57.96"S	27°23'3.36"E	0.00	0.00	0.00	0.06	0.16	0.02	0.16
	DSW1618	26°23'44.76"S	27°25'57.84"E	0.14	0.02	0.40	0.10	0.18	0.04	0.22
	DSW4319	26°21'57.30"S	27°26'34.98"E	0.06	0.04	0.24	0.22	0.26	0.08	0.26
	DSW2920	26°20'32.40"S	27°26'40.86"E	0.34	0.00	0.18	0.06	0.26	0.02	0.14
	WV2	26°23'45.90"S	27°28'43.26"E	0.12	0.02	0.28	1.84	0.28	0.66	1.04

WV3	26°23'44.10"S	27°29'2.04"E	0.00	0.02	0.30	0.08	0.48	0.02	0.14
WV4	26°23'38.88"	27°29'5.16"E	0.00	0.00	0.24	0.08	0.76	0.04	0.12
WV5	26°23'40.20"S	27°29'5.52"E	0.00	0.02	0.00	0.02	2.12	0.02	0.14
WV9	26°23'13.80"S	27°30'59.76"E	0.00	0.02	0.00	0.08	0.34	0.04	0.08
WV13	26°23'21.24"S	27°31'20.04"E	0.00	0.00	0.18	0.06	0.30	0.00	0.08
WV14	26°23'23.46"S	27°31'11.70"E	0.00	0.00	0.24	0.02	0.24	0.02	0.12
WV15	26°23'16.68"S	27°30'44.10"E	0.00	0.02	0.26	0.02	0.20	0.02	0.12
WV16	26°22'60.00"S	27°30'36.12"E	0.00	0.02	0.28	0.06	0.24	0.02	0.14
Average			0.19	0.01	0.14	0.38	0.33	0.08	0.39
Minimum			0.00	0.00	0.00	0.02	0.00	0.00	0.08
Maximum			0.99	0.04	0.40	3.72	2.12	0.66	2.68

The results of the measurements were also compared with national and international guidelines for permissible limits of heavy metals in drinking water. These results are presented in Table 5.17. The results revealed that the average concentrations of As, Ni, Cr, and Zn in drinking water were higher than permissible limits whilst those of the remaining heavy metals were within permissible limits. The high concentrations of these metal elements could be attributed to high levels of acid mine drainage reported by Winde et al. (2004) in the area, which is catalytic to the solubility and leaching of these heavy metals.

Table 5.17: Permissible Limits of Heavy Metals in Drinking Water.

Country	Heavy metals concentration limit (mg.L ⁻¹)										References
	As	Pb	Hg	Cd	Cr	Cu	Zn	Co	Ni		
USEPA	0.01	0.015	0.002	0.005	0.1	1.3	0.5	0.1	n.a	(USEPA, 2011b)	
WHO	0.01	0.01	0.006	0.003	0.05	2.0	n.a	n.a	0.07	(WHO, 2004)	
South Africa	0.01	0.01	0.001	0.003	n.a	1.0	n.a	n.a	n.a	(DOH, 2004)	

n.a: Not available

5.14. Non-Carcinogenic Risk Assessment of Heavy Metals through Soil, Plant and Water Exposure Routes

Non carcinogenic risk of heavy metals for individual members of the public were calculated based on reference doses (RfD) presented in Table 4.5 and on the results of average daily intake (ADI) values calculated for the different samples. The results were then presented in terms of the hazard quotient (HQ) and hazard index (HI) for ingestion, inhalation and dermal pathways using Equations 4.16 and 4.17.

5.14.1. Non-Carcinogenic Risk Assessment of Heavy Metals through Soil Samples

For non-carcinogenic risk calculations due to soil samples, ADI values presented in Table 5.18 were used. These ADI values were calculated using Equations 4.12, 4.13 and 4.15 described in Chapter 4.

Table 5.18: ADI Values in mg/kg/day used for Non-carcinogenic Risk Calculations in Soil Samples.

Pathway	Heavy metal ADI values (mg/kg/day)									Total
	As	Pb	Hg	Cd	Cr	Cu	Zn	Co	Ni	
Ingestion	1.06E-04	6.42E-06	1.17E-07	7.07E-08	4.34E-04	6.96E-05	9.32E-05	3.77E-05	1.59E-04	9.06E-04
Inhalation	1.63E-08	9.88E-10	1.80E-11	1.09E-11	6.68E-08	1.07E-08	1.43E-08	5.80E-09	2.44E-08	1.39E-07
Dermal	2.62E-05	1.59E-06	2.90E-08	1.75E-08	1.08E-04	1.72E-05	2.31E-05	9.34E-06	3.93E-05	2.24E-04
Total	1.32E-04	8.01E-06	1.46E-07	8.82E-08	5.42E-04	8.68E-05	1.16E-04	4.70E-05	1.98E-04	1.13E-03

HQ and HI values are important parameters used to quantify non-carcinogenic risks of heavy metals from different samples. If Both HQ and HI are less than 1, there is no obvious risk to individual members of the public, but if these values exceed one, there may be concern for potential non-carcinogenic effects (USEPA 1989; DEA., 2010). Table 5.19 shows HQ and HI Values for Heavy Metals in Soil from the Mining Area. The results presented show that the total HQ value is 2.25. This value is potentially harmful since it is greater than 1. The dermal pathway is the main driver contributing a value of 1.73 to the non-carcinogenic effect followed by the ingestion pathway with a value of 0.51. The inhalation pathway is the least of them all contributing only 0.003.

Table 5.19: Hazard Quotient (HQ) Values and Corresponding Hazard Indices (HI) for Heavy Metals in Soil from Mining Area.

Pathway	HQ Values									Total (HI)
	As	Pb	Hg	Cd	Cr	Cu	Zn	Co	Ni	
Ingestion	3.53E-01	1.78E-03	3.91E-04	1.41E-04	1.45E-01	1.88E-03	3.11E-04	1.89E-03	7.94E-03	5.12E-01
Inhalation	5.43E-05	n.a	2.10E-07	1.91E-07	2.23E-03	n.a	n.a	1.02E-03	n.a	3.30E-03
Dermal	8.74E-02	n.a	9.67E-05	3.50E-05	n.a	7.18E-04	3.08E-04	1.64E+00	7.02E-03	1.73E+00
Total	4.40E-01	1.78E-03	4.87E-04	1.77E-04	1.47E-01	2.60E-03	6.18E-04	1.64E+00	1.50E-02	2.25E+00

n.a: Indicates that reference doses were not available for calculation.

5.14.2. Non-Carcinogenic Risk Assessment of Heavy Metals through Plant Samples

For plant samples, the only pathway considered was the ingestion pathway. Table 5.20 tabulates results of ADI values used together with their corresponding HQ values. In this case, Equation 4.12 described in chapter 4 was used to calculate ADI values. Calculated values of HQ showed an HI value of 2.97×10^{-2} , which is potentially safe according to USEPA (1989) and South Africa guidelines (DEA, 2010).

Table 5.20: Average Daily Intake (ADI) and their Corresponding Hazard Quotient (HQ) Values due to Ingestion of Heavy Metals in Plant Samples from Mining Area.

Parameter	ADI and HQ Values							Total (HI)
	As	Pb	Cr	Cu	Zn	Co	Ni	
(ADI)ing	6.85E-06	4.38E-07	1.89E-05	8.77E-07	1.21E-06	7.67E-07	7.92E-06	3.70E-05
(RfD)ing	3.00E-04	3.60E-03	3.00E-03	3.70E-02	3.00E-01	2.00E-02	2.00E-02	
(HQ)ing	2.28E-02	1.22E-04	6.30E-03	2.37E-05	4.02E-06	3.84E-05	3.96E-04	2.97E-02

5.14.3. Non-Carcinogenic Risk Assessment of Heavy Metals through Water Samples

For water samples, ingestion and dermal pathways were considered in the non-carcinogenic risk assessment. Table 5.21 tabulates results of ADI values used together with their corresponding HQ values. In this situation, Equations 4.12 and 4.14 described in chapter 4 were used to calculate ADI values for water samples. Calculated values of HQ showed an HI value of 3.38×10^{-1} . This is less than 1 and potentially safe according to USEPA (1989) and South Africa guidelines (DEA, 2010). In this scenario, the dermal pathway contributed more than the ingestion pathway.

Table 5.21: Average Daily Intake (ADI) and their Corresponding Hazard Quotient (HQ) Values due to Ingestion and Dermal Contact with Water for Heavy Metals from Mining Area.

Parameter	ADI and HQ Values							Total (HI)
	As	Pb	Cr	Cu	Zn	Co	Ni	
(ADl)ing	5.25E-09	2.08E-10	3.80E-09	1.04E-08	9.01E-09	2.10E-09	1.08E-08	4.15E-08
(ADl)der	4.57E-06	1.81E-07	3.30E-06	9.01E-06	7.84E-06	1.82E-06	9.37E-06	3.61E-05
Total (ADI)	4.58E-06	1.81E-07	3.31E-06	9.02E-06	7.85E-06	1.83E-06	9.38E-06	3.61E-05
(RfD)ing	3.00E-04	3.60E-03	3.00E-03	3.70E-02	3.00E-01	2.00E-02	2.00E-02	
(HQ)ing	1.75E-05	5.77E-08	1.27E-06	2.80E-07	3.00E-08	1.05E-07	5.38E-07	1.98E-05
(RfD)der	3.00E-04	n.a	n.a	2.40E-02	7.50E-02	5.70E-06	5.60E-03	
(HQ)der	1.52E-02	n.a	n.a	3.75E-04	1.05E-04	3.20E-01	1.67E-03	3.38E-01
Total HQ	1.53E-02	5.77E-08	1.27E-06	3.76E-04	1.05E-04	3.20E-01	1.67E-03	3.38E-01

n.a: Indicates that reference doses were not available for calculation.

5.14.4. Summary of Non-Carcinogenic Risk Assessment of Heavy Metals

The results shown in Table 5.22 show a summary of the non-carcinogenic risk assessment as a result of heavy metals through the different pathways in all the samples from the mining area. The total HQ value, which is also the HI was found to be 2.62. This cumulative value is greater than 1, which potentially causes a health risk to residents of the gold mining area.

Table 5.22: Summary of HQ Values and HI in all Samples for Heavy Metals from the Mining Area.

Medium	Pathway	HQ Values								Total (HI)	
		As	Pb	Hg	Cd	Cr	Cu	Zn	Co		Ni
Soil	Ingestion	3.53E-01	1.78E-03	3.91E-04	1.41E-04	1.45E-01	1.88E-03	3.11E-04	1.89E-03	7.94E-03	5.12E-01
	Inhalation	5.43E-05	n.a	2.10E-07	1.91E-07	2.23E-03	n.a	n.a	1.02E-03	n.a	3.30E-03
	Dermal	8.74E-02	n.a	9.67E-05	3.50E-05	n.a	7.18E-04	3.08E-04	1.64E+00	7.02E-03	1.73E+00
	Sub-total	4.40E-01	1.78E-03	4.87E-04	1.77E-04	1.47E-01	2.60E-03	6.18E-04	1.64E+00	1.50E-02	2.25E+00
Water	Ingestion	1.75E-05	5.77E-08	0.00E+00	0.00E+00	1.27E-06	2.80E-07	3.00E-08	1.05E-07	5.38E-07	1.98E-05
	Dermal	1.52E-02	n.a	0.00E+00	0.00E+00	n.a	3.75E-04	1.05E-04	3.20E-01	1.67E-03	3.38E-01
	Sub-total	1.53E-02	5.77E-08	0.00E+00	0.00E+00	1.27E-06	3.76E-04	1.05E-04	3.20E-01	1.67E-03	3.38E-01
Plants	Ingestion	2.28E-02	1.22E-04	0.00E+00	0.00E+00	6.30E-03	2.37E-05	4.02E-06	3.84E-05	3.96E-04	2.97E-02
Total		4.78E-01	1.90E-03	4.87E-04	1.77E-04	1.53E-01	3.00E-03	7.27E-04	1.96E+00	1.70E-02	2.62E+00

n.a: Indicates that reference doses were not available for calculation.

An evaluation to determine the pathway that contributed the most to non-carcinogenic risk was done. Figure 5.11 shows that the dermal pathway was the greatest contributor to non-carcinogenic risk with an HQ value of 2.07 followed by the ingestion pathway with a value of 0.54. Inhalation of heavy metals only contributed 3.3×10^{-3} to the total non-cancer risk. Co contributed the most to the non-cancer risk, followed by As and then Cr. This was primarily because As is a non-essential element in plants and animals. Although Co plays a vital role in nitrogen fixing and Cr in the metabolism of fats and cholesterol, their ADI values for soil were closer to the toxicity threshold doses for the dermal pathway (Co) and ingestion pathway (Cr), thereby becoming more hazardous than the other heavy metals.

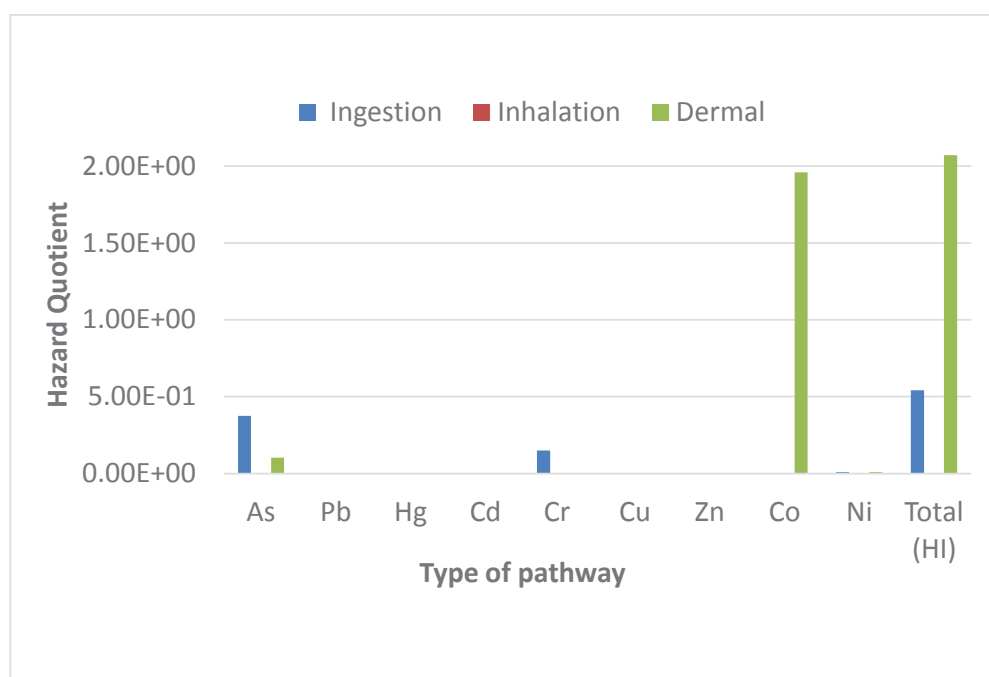


Figure 5.11: Hazard Quotient for Heavy Metals through Different Pathways in the Mining Area.

Non-cancer risks were also evaluated based on sample type. Figure 5.12 shows the results of this comparison. It was discovered that soil from the mining area had the greatest contribution with an HQ value of 2.25. This was followed by water samples with a value of 0.34. Plant samples were the least contributing only a value of 2.97×10^{-2} . Co again was realized to contribute the most to the non-cancer risk, followed by As and Cr for the reasons stated earlier.

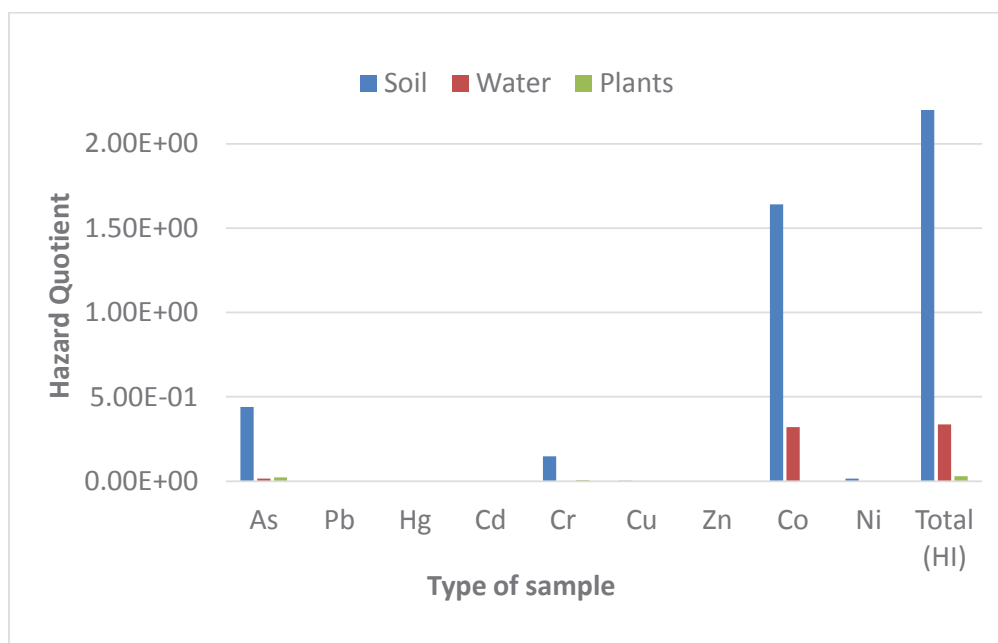


Figure 5.12: Hazard Quotient for Heavy Metals through Different Samples in the Mining Area.

For all the samples and for all pathways, HI was equal to 2.62. According to USEPA (1989) and DEA (2010), this value may pose a non-cancer health risk to residents of the gold mining area as it is greater than 1, a level regarded safe by international and national standards.

5.15. Carcinogenic Risk Assessment of Heavy Metals through Soil, Plant and Water Exposure Routes

As, Pb, Cd, Cr and Co were considered as metals that have the potential to cause carcinogenic risk. The cancer risks for individual members of the public were calculated from the average contribution of the individual heavy metals in all the environmental media for all the pathways. In order to calculate the ADI values Equations 4.12, 4.13, 4.14 and 4.15 described in chapter 4 were used. On the other hand, Equations 4.18, 4.19 and 4.20, also from chapter 4 were used to determine the cancer risk.

5.15.1. Carcinogenic Risk Assessment of Heavy Metals through Soil from the Mining Area

The carcinogenic risk values calculated for soil were based on the average daily intake (ADI) values as presented in Table 5.23.

Table 5.23: Average Daily Intake (ADI) Values in mg/kg/day used for Carcinogenic Risk Calculations in Mine Soil.

Pathway	ADI Values for Heavy Metals (mg/kg/day)					Total ADI
	As	Pb	Cd	Cr	Co	
Ingestion	4.54E-05	2.75E-06	3.03E-08	1.86E-04	1.62E-05	2.50E-04
Inhalation	6.98E-09	4.2323E-10	4.66E-12	2.86E-08	2.49E-09	3.85E-08
Dermal	1.12E-05	6.8131E-07	7.50E-09	4.61E-05	4.00166E-06	6.20E-05
Total	5.66E-05	3.43E-06	3.78E-08	2.32E-04	2.02E-05	3.12E-04

The results of the cancer risks presented in Table 5.24 showed that the total cancer risk due to soil was 1.7×10^{-4} mainly driven by the ingestion pathway with a cancer risk of 1.61×10^{-4} followed by the dermal contact with a cancer risk of 1.68×10^{-5} . The least was the inhalation pathway with a cancer risk of 1.3×10^{-6} .

Table 5.24: Cancer Risk Values of Heavy Metals for Individual Members of the Public through Soil from Mining Area.

Pathway	Cancer Risk Values					Total Risk
	As	Pb	Cd	Cr	Co	
Ingestion	6.80E-05	2.34E-08	n.a	9.30E-05	n.a	1.61E-04
Inhalation	1.05E-07	1.78E-11	2.94E-11	1.17E-06	2.44E-08	1.30E-06
Dermal	1.68E-05	n.a	n.a	n.a	n.a	1.68E-05
Total	8.50E-05	2.34E-08	2.94E-11	9.42E-05	2.44E-08	1.79E-04

n.a: Indicates that cancer slope factors were not available for risk calculation.

5.15.2. Carcinogenic Risk Assessment of Heavy Metals through Plant Samples

For plant samples, carcinogenic risk was considered as a result of ingestion of these edible plants. The results of cancer risk for these mine residents are presented in Table 5.25. The table also presents ADI values that were used for carcinogenic risk calculations. As a result of consumption of the plants (mostly vegetables), the total cancer risk was found to be 8.46×10^{-6} mainly driven by As and Cr.

Table 5.25: ADI and Cancer Risk Values through Ingestion of Heavy Metals for Individual Members of the Public for Plant Samples from Mining Area.

Parameter	ADI and Cancer Risk Values					Total Risk
	As	Pb	Cd	Cr	Co	
(ADI)ing	2.94E-06	1.88E-07	0.00E+00	8.10E-06	3.29E-07	1.58E-05
(CSF)ing	1.50E+00	8.50E-03	n.a	5.00E-01	n.a	
Risk	4.40E-06	1.60E-09	n.a	4.05E-06	n.a	8.46E-06

n.a: Indicates that cancer slope factors were not available for risk calculation.

5.15.3. Carcinogenic Risk Assessment of Heavy Metals through Water Samples

The ingestion and dermal pathways were considered for estimating the cancer risk of heavy metals in water. The results for individual cancer risk are presented in Table 5.26. The same table also presents ADI values used in the calculation of the cancer risk. For those metals without known cancer slope factors, the risk was not calculated. The results shown indicate that the total cancer risk was 2.94×10^{-6} mainly due to dermal contact.

Table 5.26: ADI values and the Cancer Risk Values through Ingestion and Dermal Contact of Heavy Metals for Individual Members of the Public for Water from Mining Area.

Parameter	ADI and Cancer Risk Values					Total Risk
	As	Pb	Cd	Cr	Co	
(ADI)ing	2.25E-09	8.91E-11	0.00E+00	1.63E-09	8.99E-10	1.78E-08
(ADI)der	1.96E-06	7.75E-08	0.00E+00	1.42E-06	7.82E-07	1.55E-05
Total (ADI)	1.96E-06	7.76E-08	0.00E+00	1.42E-06	7.83E-07	1.55E-05
(CSF)ing	1.50E+00	8.50E-03	n.a	5.00E-01	n.a	
(Risk)ing	3.38E-09	7.57E-13	n.a	8.14E-10	n.a	4.19E-09
(CSF)der	1.50E+00	n.a	n.a	n.a	n.a	
(Risk)der	2.94E-06	n.a	n.a	n.a	n.a	2.94E-06
Total Risk	2.94E-06	7.57E-13	n.a	8.14E-10	n.a	2.94E-06

n.a: Indicates that cancer slope factors were not available for risk calculation.

5.15.4. Summary of Carcinogenic Risk Assessment of Heavy Metals

The results presented in Table 5.27 show a summary of the cancer risks from all the environmental media. The total carcinogenic risk from all the samples was found to be 1.91×10^{-4} . The contribution of heavy metals to total risk is such that Cr>As>Co>Pb>Cd.

Table 5.27: Summary of Cancer Risk Values of Heavy Metals through Soil, Plant and Water Exposure Routes.

Medium	Pathway	Cancer Risk Values for Heavy Metals in the Mining Area					Total Risk
		As	Pb	Cd	Cr	Co	
Soil	Ingestion	6.80E-05	2.34E-08	n.a	9.30E-05	n.a	1.61E-04
	Inhalation	1.05E-07	1.78E-11	2.94E-11	1.17E-06	2.44E-08	1.30E-06
	Dermal	1.68E-05	n.a	n.a	n.a	n.a	1.68E-05
	Sub-total	8.50E-05	2.34E-08	2.94E-11	9.42E-05	2.44E-08	1.79E-04
Water	Ingestion	3.38E-09	7.57E-13	n.a	8.14E-10	n.a	4.19E-09
	Dermal	2.94E-06	n.a	n.a	n.a	n.a	2.94E-06
	Sub-total	2.94E-06	7.57E-13	n.a	8.14E-10	n.a	2.94E-06
Plant	Ingestion	4.40E-06	1.60E-09	n.a	4.05E-06	n.a	8.46E-06
Total		9.23E-05	2.50E-08	2.94E-11	9.82E-05	2.44E-08	1.91E-04

n.a: Indicates that cancer slope factors were not available for risk calculation.

Figure 5.13 is an extract from Table 5.27 that clearly illustrates that the ingestion pathway was the greatest contributor to the individual cancer risk. When all the ingestion components in Table 5.27 are summed up, a value of 1.7×10^{-4} is obtained. This ingestion pathway is then followed by the dermal pathway with a calculated value of 1.98×10^{-5} . The least contribution came from the inhalation pathway that recorded a risk of 1.3×10^{-6} . Figure 5.13 also reaffirms that As and Cr were the major contributors to carcinogenic risk.

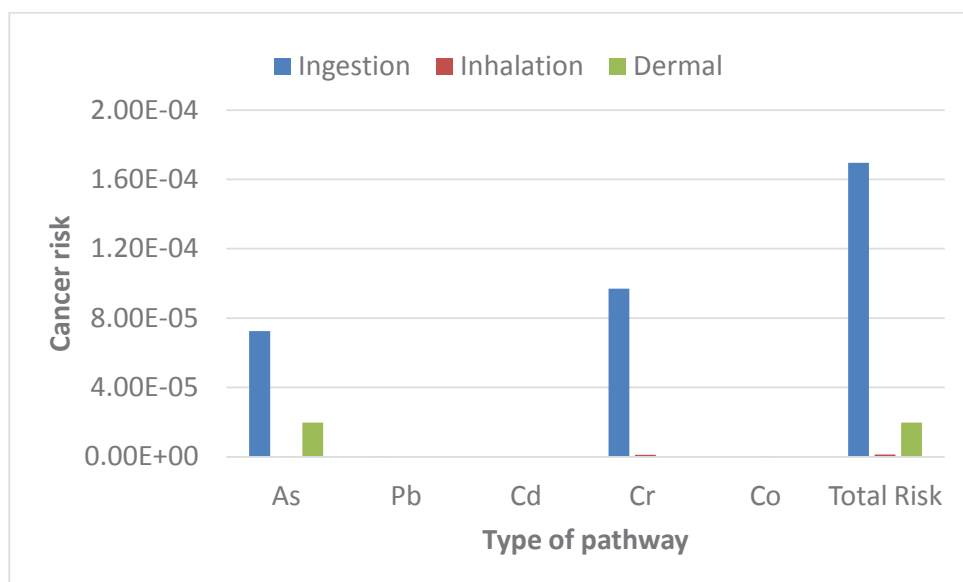


Figure 5.13: Cancer Risk Values for Heavy Metals through Different Pathways.

When the results were correlated with the type of sample, it was observed that soil samples contributed the most to the total cancer risk with a summed up value of 1.79×10^{-4} followed by plant samples with a contribution of 8.46×10^{-6} and lastly water samples with a value of 2.94×10^{-6} . The results are illustrated in Figure 5.14.

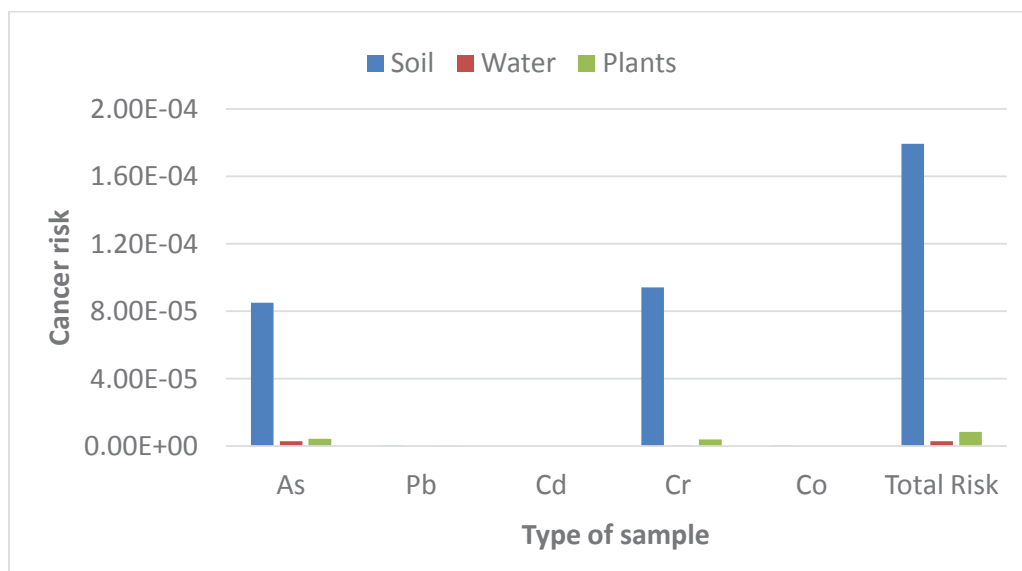


Figure 5.14: Cancer Risk Values for Heavy Metals through Different Samples.

5.16. Summary of Cancer Risk due to Natural Radionuclides and Heavy Metals

The US Environmental Protection Agency considers acceptable for regulatory purposes a cancer risk in the range of 1×10^{-6} to 1×10^{-4} (USEPA, 1989). South Africa, instead, considers a cancer risk limit for the public as 5×10^{-6} (Government Gazette, 2006). For natural radionuclides ^{238}U , ^{232}Th and ^{40}K , the maximum cancer risk with all pathways summed was found to be 6.52×10^{-5} (1 in 15337 individuals). On the other hand, total cancer risk for heavy metals was found to be 1.91×10^{-4} (1 in 5236 individuals). The sum total of cancer risk due to these radionuclides and heavy metals was found to be 2.56×10^{-4} (1 in 3906 individuals). This value is higher than acceptable values according to USEPA and South African standards. From the findings presented, it can also be stated that heavy metals contributed more to the risk compared to natural radionuclides.

CHAPTER 6: SUMMARY, CONCLUSIONS AND RECOMMENDATIONS

6.1. SUMMARY AND CONCLUSIONS

The aim of the study was to carry out a human health risk assessment of environmental radionuclides and heavy metals around a Gold Mining Area in the Gauteng Province, South Africa. The study was motivated by the fact that no previous studies had been carried out in the area to evaluate the extent of the possible risks of these contaminants to members of the public. Soil, water and plant samples were collected from representative locations for this purpose. Gamma-ray spectroscopy and ICP MS were used to measure the activity concentrations of natural radioactivity and concentrations of heavy metals, respectively, in all samples. The AlphaGuard Monitor was used to measure indoor radon gas. All the measurements were compared with those from the control area and reference standards.

The results obtained in the study showed that the average activity concentrations in Bq.kg⁻¹ for ²³⁸U, ²³²Th and ⁴⁰K in soil from the mining area varied from 34.4±0.7 to 3681.6±87.1, with an average of 574.3±39.5, 20.5±0.6 to 167.5±12.6 with an average of 49.4±8.5 and 133.8±4.4 to 1177.0±114.1 with an average of 424.7±129.3, respectively. On the other hand, the average activity concentrations in Bq.kg⁻¹ for ²³⁸U, ²³²Th, and ⁴⁰K in soil from the control area were 17.0±0.4 varying from 12.5±0.3 to 23.6±0.5, 22.2±0.5 varying from 16.6±0.4 to 30.4±0.6 and 496.8±15.2 varying from 424.3±13.0 to 648.4±19.8, respectively. The results showed that there were higher levels of ²³⁸U and ²³²Th in soil from the mining area compared to the control area. When the results were compared with published data from other countries and the worldwide average, ²³⁸U levels were very high whilst the average activity concentrations of ²³²Th and ⁴⁰K were comparable to other countries (UNSCEAR, 2000). This was attributed to the large volumes of uranium waste found in the gold mine.

In plant samples, the average activity concentration of ²³⁸U in Bq.kg⁻¹ were found to be 17.4±3.1 and ranged from 7.4±1.7 to 37.7±6.9. For ²³²Th the average activity concentration in Bq.kg⁻¹ was 19.7±1.6, ranging from 11.5±0.8 to 35.6±3.1 and that of ⁴⁰K was 146.7±9.2 Bq.kg⁻¹ in the range 79.9±3.5 to 248.1±9.6 Bq.kg⁻¹. On the other

hand, the average activity concentration in Bq.kg⁻¹ for ²³⁸U, ²³²Th, and ⁴⁰K from the control area was 12.1±1.5, 15.6±1.0 and 125.7±4.8, respectively. According to UNSCERAR (2000), the acceptable world reference for ²³⁸U in leafy vegetables is 20 mBq.kg⁻¹ and 3 mBq.kg⁻¹ for fruit samples. The values measured for ²³⁸U in plant samples were higher than these acceptable limits. For ²³²Th all the values were higher than the world reference of 15 mBq.kg⁻¹ for leafy vegetables and 0.5 mBq.kg⁻¹ for fruit samples, respectively. It was also observed that the activity concentration of ⁴⁰K markedly exceeded that of both ²³⁸U and ²³²Th, as it is the most abundant radioactive element on normal soil. These high levels were attributed to mining activities in the area.

For water samples, the average values for ²³⁸U, ²³²Th and ⁴⁰K in Bq.L⁻¹ were 0.66±0.03 (within the range 0.35±0.02 - 1.31±0.07), 0.56±0.03 (within the range 0.29±0.01 - 0.84±0.04) and 7.36±0.58 (within the range 4.93±0.40 - 11.06±0.82), respectively. Compared with samples from the control area, average values for ²³⁸U, ²³²Th and ⁴⁰K in water samples from the mining area were higher. Compared to South African guidelines (DWAF, 2002), the average activity concentrations of ²³⁸U and ²³²Th in all the samples were higher than recommended values of 1.0 and 0.05 mBq.L⁻¹, respectively. ⁴⁰K is a very soluble and an essential element for metabolism in living organisms. Its value was, therefore, high as expected.

Radiological hazards for soil, plant and water samples were considered for the study area. For soil, the Ra_{eq} average values for the mining area and the control area ranged from 107.4±1.5 to 3964.9.9±89.5 Bq.kg⁻¹ with an average of 677.7±43.8 Bq.kg⁻¹ and from 76.4±1.4 to 103.8±1.5 Bq kg⁻¹ with an average of 86.9±1.4 Bq.kg⁻¹, respectively. In most of the soil samples from the mining area, Ra_{eq} values were above the international value of 370 Bq.kg⁻¹ as recommended by IAEA under normal circumstances (OECD, 1979), mainly due to the high levels of ²³⁸U found in the mine tailings and rock dumps. Outdoor average dose rate in air from terrestrial gamma rays in soil was found to be 312.9±20.3 nGy.h⁻¹, a value much higher than the global average of 59 nGy.h⁻¹ (UNSCEAR, 2000). However, the value from the control area was found to be 41.9±0.7 nGy.h⁻¹, which is lower than the worldwide average. AEDE values for soil samples from the mining area were ranging from 0.06 to 2.24 mSv.y⁻¹ with an average of 0.38 mSv y⁻¹. For the control

area, the average AEDE value was 0.05 mSv y^{-1} ranging from 0.05 to 0.06 mSv y^{-1} . Although some AEDE values were below the worldwide average of 0.48 mSv for terrestrial gamma radiation (UNSCEAR, 2000), some samples had values above the worldwide average. Calculated hazard indices for soil samples from the mining area ranged from 0.3 to 10.7 with an average of 1.8 and from 0.4 to 20.7 with an average of 3.4 for the external hazard (H_{ex}) and internal hazard (H_{in}) respectively. This showed that the average values for H_{ex} and H_{in} were higher than unity, posing a significant radiological threat to the population in the area.

For plant samples, the average value of Ra_{eq} in Bq.kg^{-1} from the mining area was 56.8 ± 4.0 with a range of 30.4 ± 2.4 to 103.1 ± 7.7 . That of the control area was $44.1 \pm 2.1 \text{ Bq.kg}^{-1}$, a figure lower than the values from the control area. From the results, it can be seen that maximum values were recorded from leafy vegetables, but still lower than the allowable limit of 370 Bq.kg^{-1} (OECD, 1979). The outdoor average absorbed dose rate in air due to ^{238}U , ^{232}Th , and ^{40}K in plant samples from the mining area varied from 13.9 ± 1.8 to $47.3 \pm 3.5 \text{ nGy.h}^{-1}$ with an average of $26.0 \pm 1.8 \text{ nGy.h}^{-1}$. This is also below the worldwide average of 59 nGy.h^{-1} (UNSCEAR, 2000). AEDE values from plant samples for the mining area and the control area were also calculated. These values were found to be in the range 0.02 to 0.06 mSv.y^{-1} with an average of 0.03 mSv y^{-1} for the mining area and an average value of 0.02 mSv.y^{-1} from the control area. These AEDE values were also below the worldwide average of 0.48 mSv for external terrestrial radiation (UNSCEAR, 2000) and also lower than 1 mSv.y^{-1} , a limit recommended by the International Commission on Radiation Protection (ICRP) for individual members of the public (ICRP, 2007). The external hazard (H_{ex}) index estimated for plant samples from mining area ranged from 0.1 to 0.3 with an average of 0.2 while the internal hazard (H_{in}) index ranged from 0.1 to 0.4 with an average of 0.2. The average values for both quantities were less than unity, making plant samples radiologically safe to members of the public in the study area.

The average value of Ra_{eq} in water samples from the mining area was $2.03 \pm 0.07 \text{ Bq.L}^{-1}$. This was significantly higher than $1.39 \pm 0.08 \text{ Bq.L}^{-1}$, a figure estimated from the control area. The values recorded in all the water samples as a result of external exposure were however, lower than the allowable limit of 370 Bq.L^{-1} (OECD, 1979).

From the calculations the average outdoor absorbed dose rate in air 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 in air as a result of water samples was however, lower than the value obtained from the control area (0.68 ± 0.04 nGy.h^{-1}). AEDE values for water samples as a result of exposure to external radiation were also calculated for the mining area and the control area. From the mining area, values ranged from 6.65×10^{-4} to 1.89×10^{-3} mSv.y^{-1} with an average of 1.17×10^{-3} mSv.y^{-1} . An average value of 8.39×10^{-4} mSv.y^{-1} from the control area was recorded, which is below the average from the mining area. These AEDE values were also way below the worldwide average of 0.48 mSv for external terrestrial radiation (UNSCEAR, 2000). Average external hazard (H_{ex}) index for water samples from the mining area was found to be 5.49×10^{-3} while the average internal hazard (H_{in}) index was 7.28×10^{-3} . Both quantities were less than unity, making water samples due to external radiation safe to the population in the area.

Indoor radon from the mining area and the control area was also measured. The activity concentrations of indoor radon from the mining area ranged from 1.0 to 472.0 Bq/m^3 , compared to a range of 0.1 to 35.0 Bq/m^3 from the control area. The annual effective doses corresponding to the activity concentrations ranged from 0.03-11.89 mSv with a mean value of 3.01 mSv. This average value was higher compared to the results calculated from the control area. Radon is a daughter product of uranium, which was elevated in the gold mining area. The mean value of the annual effective dose of radon was also higher than that published for normal areas around the world. According to UNSCEAR (2000), the average annual effective dose from Inhalation of radon and its decay products is 1.26 mSv.

The annual effective doses of natural radionuclides in mSv through the ingestion of water and plant samples were estimated to be 1.24×10^{-3} and 1.23×10^{-4} , respectively. These values were also lower than 1 mSv.y^{-1} , a limit recommended by ICRP for individual members of the public (ICRP, 2007).

The cancer risk for members of the public living in the mining area, as a result of natural radionuclides, was estimated using RESRAD-OFFSITE Computer Code. The

maximum total cancer risk for all the pathways was found to be 6.52×10^{-5} . This was higher than the South African Individual risk limit for the public of 5×10^{-6} (Government Gazette, 2006).

Health risks as a result of heavy metals in soil, plant, and water samples collected from the different locations of the gold mining area and from the control area were also evaluated. The average concentrations of heavy metals in mg.kg^{-1} from the different locations in soil varied significantly and decreased in the order of $\text{Cr} > \text{Ni} > \text{As} > \text{Zn} > \text{Cu} > \text{Co} > \text{Pb} > \text{Hg} > \text{Cd}$. The average values in soil were as follows: Cr 316.88 (within a range of 77.50-861.67); Ni 115.87 (within a range of 68.33-152.50); As 77.25 (within a range of 62.23-115.19); Zn 68.01 (within a range of 21.82- 85.00); Cu 50.79 (within a range of 19.09- 108.75); Co 27.52 (within a range of 11.82- 41.25); Pb 4.69 (within a range of 1.58-10.22); Hg 0.09 (within a range of 0.06-0.13); and Cd 0.05 (within a range of 0.04 - 0.08), respectively. It was observed that soil from the mining area had generally higher concentrations of heavy metals compared to the soil from the control area. The results were also compared with maximum allowable limits. As and Cr were found to be the highest in the present study. However, Pb, Cd, Zn and Co concentrations were lower than the maximum allowable limits while Hg, Cu, and Ni were comparable with other countries.

The average concentrations in mg.kg^{-1} of heavy metals in plant samples from the different locations were also measured. Their values decreased in the order of $\text{Cr} > \text{Ni} > \text{As} > \text{Zn} > \text{Cu} > \text{Co} > \text{Pb}$. The ranges in mg.kg^{-1} were as follows: Cr (4.20 to 8.30 with an average of 6.90); Ni (0.20 to 6.70 with an average of 2.89); As (2.20 to 2.70 with an average 2.50); Zn (0.20 to 0.80 with an average 0.44); Cu (0.20 to 0.60 with an average 0.32); Co (0.10 to 0.70 with an average 0.28); Pb (0.10 to 0.30 with an average 0.16). The results of the measurements were also compared with national and international guidelines for permissible limits of heavy metals in foodstuffs. The results indicated that the average concentrations of As and Ni in plant samples were higher than FAO/WHO and South African safe limits whilst those of Hg, Cd and Zn were below permissible levels. There were no standards for chromium and cobalt to compare with. The remaining heavy metals were within permissible limits.

The average concentrations of heavy metals in water samples in mg.L^{-1} from the different locations were also measured. The average concentrations from the mining area decreased in the order of $\text{Ni} > \text{Cu} > \text{Zn} > \text{As} > \text{Cr} > \text{Co} > \text{Pb}$. The ranges in mg.L^{-1} were as follows: Ni (0.08 to 2.68 with an average of 0.39); Cu (0.02 to 3.72 with an average of 0.38); Zn (0.00 to 2.12 with an average 0.33); As (0.00 to 0.99 with an average 0.19); Cr (0.00 to 0.40 with an average 0.14); Co (0.00 to 0.66 with an average 0.08); Pb (0.00 to 0.04 with an average 0.01). The results showed that the average concentrations of the heavy metals in water from the gold mining area were generally higher than concentrations from the control area. Compared with national and international guidelines for permissible limits, the average concentrations of As, Ni, Cr, and Zn in drinking water were higher than permissible limits whilst those of the remaining heavy metals were within limits.

Non carcinogenic risks of heavy metals for individual members of the public through the different pathways in all the samples were also evaluated. The total HQ was found to be 2.62, a value greater than 1, which potentially causes a health risk to the public living in the gold mining area (USEPA, 1989; DEA, 2010). The dermal pathway was the greatest contributor to non-carcinogenic risk with an HQ value of 2.07 followed by the ingestion pathway with a value of 0.54. Inhalation of heavy metals only contributed 3.3×10^{-3} to the total non-cancer risk. Co, As and Cr contributed the most to the non-cancer risk. Non-cancer risks were also evaluated based on sample type. It was discovered that soil from the mining area had the greatest contribution with an HQ value of 2.25. This was followed by water samples with a value of 0.34. Plant samples had the least, contributing a value of 2.97×10^{-2} .

The total carcinogenic risk from all the samples as a result of heavy metals was found to be 1.91×10^{-4} . The ingestion pathway was the greatest contributor to the cancer risk with a value of 1.7×10^{-4} . This was followed by the dermal pathway that recorded a value of 1.98×10^{-5} . The least contribution came from the inhalation pathway that had a risk of 1.3×10^{-6} . The results have shown that Cr and As were the major contributors to carcinogenic risk. When the results were correlated with sample type, it was discovered that soil contributed the most to the total cancer risk with a value of 1.79×10^{-4} recorded followed by plant samples which contributed a cancer risk of 8.46×10^{-6} and lastly water samples with a value of 2.94×10^{-6} .

The US Environmental Protection Agency considers a cancer risk in the range of 1×10^{-6} to 1×10^{-4} acceptable for regulatory purposes (USEPA, 1989). For natural radionuclides, ^{238}U , ^{232}Th and ^{40}K , the maximum cancer risk, with all pathways summed up, was found to be 6.52×10^{-5} (1 in 15 337 individuals). On the other hand, total cancer risk for heavy metals was estimated to be 1.91×10^{-4} (1 in 5236 individuals). The sum total due to natural radionuclides and heavy metals was a cancer risk of 2.56×10^{-4} (1 in 3906 individuals). This value is higher than acceptable values according to USEPA and South African standards. From the findings presented, it can be concluded that natural radionuclides and heavy metal pollution in the mining area are an issue of health concern.

6.2. RECOMMENDATIONS

From the findings of this study, the following recommendations should be considered for future studies;

- Dust should also be sampled as it is a potential pathway to man. It was not collected due to unavailability of equipment and time constraints. Radioactive dust carried along with heavy metals through wind erosion can be deposited on agricultural land from mine tailings or other sources impacting on the environment.
- The study was only restricted to natural radionuclides of a primordial origin and selected heavy metals, but other radionuclides and less hazardous heavy metals need to be investigated as well.
- Gross alpha and gross beta activity concentrations in drinking water sources in the study area should be considered in future work. This requires a Liquid Scintillation Counter (LSC).
- The results of the study indicated that uranium concentration is high in most of the samples. A radiological risk assessment, looking at radionuclides bound to sediments is also recommended.
- Fewer measurements on radon gas were carried out and yet it is known to contribute more than 50 % of natural radiation. More detailed studies on radon and its progeny should be performed in the gold mining area.

- A correlation of the environmental parameters such as ^{222}Rn , ^{238}U and U using the different Instruments in order to validate the measurements can also be investigated.
- Physico-chemical factors such as soil pH, temperature, electrical conductivity, type of soil also affect the migration of radionuclides and heavy metals. These could be considered in future studies.
- There are also short term and seasonal variations that affect the distribution and concentration of these contaminants. The original intention was to do sampling three times a year, but it was not possible due to logistical and time limitations. Future work should consider these factors in detail.
- The study can be expanded to cover grass/vegetation and all other food items, that are potential pathways to man.

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APPENDICES

APPENDIX A: Details of Activity Concentrations of Soil, Water and Plant Samples from the Gold Mining Area.

Table AA: Details of Activity Concentrations Soil Samples from the Gold Mining Area.

Table AA1: Calculated Activity Concentrations of ^{238}U derived from the Weighted Average of Selected Gamma-ray Transitions of ^{214}Pb and ^{214}Bi in each Soil Sample.

Sample ID	Latitude	Longitude	Average Activity concentration (Bq.kg ⁻¹)				Weighted Average	
			^{214}Pb		^{214}Bi		^{238}U	
			295.2 keV	351.9 keV	609.3 keV	1120.3 keV	1764.5 keV	
CS1	25°47'34.62"S	25°37'55.26"E	12.6±0.8	12.8±0.8	12.6±0.6	11.9±0.9	13.2±0.8	12.7±0.3
CS2	25°47'33.06"S	25°38'2.04"E	14.4±0.9	13.6±0.8	13.2±0.6	13.3±0.9	13.9±0.8	13.6±0.3
CS3	25°47'34.80"S	25°38'19.20"E	12.6±0.8	13.6±0.8	12.5±0.6	13.9±1.0	13.1±0.7	13.0±0.3
CS4	25°47'35.46"S	25°38'13.62"E	18.0±1.1	17.9±1.0	17.7±0.7	18.5±1.0	18.2±0.9	18.0±0.4
CS5	25°47'41.70"S	25°38'3.36"E	12.4±0.8	12.8±0.7	12.0±0.5	13.4±0.9	12.6±0.7	12.5±0.3
CS6	25°47'46.32"S	25°37'59.16"E	18.2±1.0	18.5±1.0	17.8±0.8	18.1±1.2	17.9±0.8	18.1±0.4
CS7	25°47'53.10"S	25°37'54.90"E	18.8±1.1	19.4±1.1	18.0±0.8	18.0±1.0	19.8±0.9	18.7±0.4
CS8	25°47'50.64"S	25°38'1.56"E	22.7±1.3	23.3±1.3	21.2±0.9	22.8±1.1	23.2±1.0	22.5±0.5
CS9	25°47'53.82"S	25°38'1.92"E	24.2±1.4	24.3±1.3	22.8±0.9	23.4±1.1	24.1±1.0	23.6±0.5
CS10	25°48'1.86"S	25°37'57.72"E	17.9±1.0	17.9±1.0	17.0±0.7	16.7±0.8	17.9±0.8	17.4±0.4
Average±Error								
T1E1	26°22'20.52"S	27°29'10.20"E	719.6±40.1	725.7±38.1	677.1±26.6	680.9±20.7	708.2±23.3	695.4±12.0
T1E2	26°22'20.40"S	27°29'28.74"E	733.5±40.9	742.5±39.0	699.3±27.5	697.8±21.3	721.8±23.9	712.3±12.4
T1E3	26°22'19.80"S	27°29'40.86"E	664.9±37.1	672.5±35.3	632.8±24.9	636.9±19.5	657.6±21.7	647.8±11.2
T1E4	26°22'5.64"S	27°29'39.66"E	785.8±43.9	797.9±41.9	753.3±29.7	765.7±23.4	795.0±26.2	776.1±13.5
T1E5	26°21'56.28"S	27°29'38.76"E	687.2±38.4	692.8±36.4	660.1±26.0	658.7±20.2	674.4±22.3	669.4±11.7
T1E6	26°21'49.02"S	27°29'37.26"E	718.3±40.1	727.5±38.2	682.8±26.9	687.9±21.2	700.8±23.2	697.2±12.1
T1E7	26°21'49.38"S	27°29'22.68"E	853.5±47.7	868.4±45.6	816.8±32.2	818.8±25.1	839.3±27.7	832.2±14.5

T1E8	26°21'52.44"S	27°29'12.24"E	734.6±41.0	744.1±39.1	698.8±27.5	696.1±21.4	728.0±24.1	713.5±12.4
T1E9	26°22'3.06"S	27°29'11.10"E	801.6±44.8	808.9±42.5	758.9±29.9	760.7±23.4	786.4±26.0	775.9±13.5
T1E10	26°22'20.94"S	27°29'10.26"E	1294.0±72.2	1307.0±68.6	1220.0±48.0	1216.0±37.1	1255.0±41.3	1243.4±21.6
T1E11	26°22'21.42"S	27°29'8.46"E	315.9±17.7	321.2±16.9	299.7±11.8	298.8±9.4	304.7±10.3	304.4±5.4
Average±Error								
T2E1	26°22'33.84"S	27°30'5.88"E	695.8±38.9	705.0±37.0	662.3±26.1	666.7±20.5	683.0±22.6	676.8±11.8
T2E2	26°22'33.72"S	27°30'19.80"E	664.9±37.1	669.1±35.1	632.8±24.9	637.5±19.6	660.3±21.8	648.4±11.3
T2E3	26°22'26.64"S	27°30'20.64"E	760.2±42.4	773.5±40.6	722.6±28.5	726.6±22.3	753.2±25.0	740.7±12.9
T2E4	26°22'10.98"S	27°30'22.08"E	730.7±40.8	744.2±39.1	699.6±27.5	704.1±21.6	737.3±24.4	718.5±12.5
T2E5	26°22'2.10"S	27°30'23.04"E	846.8±47.3	862.9±45.3	803.4±31.6	796.5±24.4	823.2±27.2	816.2±14.2
T2E6	26°21'55.86"S	27°30'23.52"E	860.4±48.0	869.3±45.6	807.5±31.8	797.9±24.5	818.7±27.1	818.0±14.2
T2E7	26°21'56.88"S	27°30'9.84"E	861.2±48.1	877.7±46.1	821.5±32.3	824.6±25.3	857.5±28.3	841.5±14.6
T2E8	26°21'59.34"S	27°29'58.72"E	890.9±49.7	900.7±47.3	847.1±33.4	845.9±25.9	872.1±28.8	862.9±15.0
T2E9	26°22'10.08"S	27°29'58.26"E	740.0±41.3	749.9±39.4	703.9±27.7	700.8±21.6	723.1±24.0	716.0±12.5
T2E10	26°22'19.20"S	27°29'57.66"E	696.3±38.9	711.6±37.4	668.0±26.3	672.4±20.7	685.2±22.7	681.2±11.9
T2E11	26°22'33.30"S	27°29'56.52"E	633.6±35.4	642.6±33.7	601.6±23.7	602.5±18.5	630.3±20.9	616.6±10.7
T2E12	26°22'34.26"S	27°30'0.78"E	1416.0±79.0	1440.0±75.5	1361.0±53.5	1368.0±41.7	1418.0±46.7	1391.8±24.1
T2E13	26°22'35.53"S	27°29'57.88"E	822.7±45.9	837.4±44.0	784.4±30.9	789.7±24.2	824.8±27.2	805.8±14.0
Average±Error								
T3W1	26°22'21.60"S	27°26'49.98"E	1416.0±79.0	1440.0±75.5	1361.0±53.5	1368.0±41.7	1418.0±46.7	1391.8±24.1
T3W2	26°22'13.62"S	27°26'48.96"E	2765.0±154.2	2805.0±147.2	2620.0±103.0	2611.0±79.4	2696.0±88.6	2668.9±46.2
T3W3	26°22'14.16"S	27°26'41.64"E	493.5±27.6	497.6±26.1	466.7±18.4	465.5±14.4	484.4±16.1	476.6±8.3
T3W4	26°22'15.66"S	27°26'34.74"E	2328.0±129.8	2356.0±123.6	2188.0±86.1	2198.0±66.9	2262.0±74.4	2240.6±38.8
T3W5	26°22'24.06"S	27°26'35.28"E	397.8±22.2	405.4±21.3	381.7±15.1	384.6±11.9	397.9±13.3	390.9±6.8
T3W6	26°22'28.20"S	27°26'39.84"E	2716.0±151.5	2751.0±144.4	2572.0±101.2	2560.0±77.9	2640.0±86.7	2617.0±45.3
T3W7	26°22'29.22"S	27°26'41.04"E	781.3±43.6	795.0±41.7	744.7±29.3	748.0±23.0	764.8±25.3	759.7±13.2
T3W8	26°22'31.38"S	27°26'43.08"E	1976.0±110.2	1997.0±104.8	1861.0±73.2	1868.0±56.9	1925.0±63.3	1904.5±33.0
Average±Error								
T4W1	26°21'11.40"S	27°27'50.82"E	155.1±8.7	158.2±8.3	148.5±5.9	151.3±4.9	153.7±5.3	152.4±2.7
T4W2	26°21'11.34"S	27°27'50.22"E	403.1±22.5	409.9±21.5	387.2±15.3	390.5±12.1	406.6±13.6	397.3±7.0
T4W3	26°21'3.06"S	27°27'50.16"E	147.3±8.3	149.0±7.8	137.5±5.4	140.0±4.6	146.0±5.0	142.7±2.6

T4W4	26°20'51.18"S	27°27'49.62"E	89.2±5.0	91.9±4.9	85.8±3.4	83.9±2.9	89.5±3.2	87.2±1.6
T4W5	26°20'51.66"S	27°27'40.26"E	185.9±10.4	188.2±9.9	179.8±7.1	179.2±5.7	185.1±6.3	182.5±3.2
T4W6	26°20'53.46"S	27°27'30.66"E	148.4±8.3	149.8±7.9	141.4±5.6	140.6±4.6	144.6±5.0	143.6±2.6
T4W7	26°21'0.18"S	27°27'25.50"E	386.7±21.6	392.0±20.6	368.8±14.5	363.0±11.4	376.6±12.7	373.0±6.6
T4W8	26°21'6.18"S	27°27'21.96"E	135.1±7.6	138.5±7.3	130.4±5.2	133.8±4.3	139.7±4.8	135.2±2.4
T4W9	26°21'7.98"S	27°27'25.62"E	194.2±10.9	197.5±10.4	187.9±7.4	189.3±6.0	202.0±6.9	193.6±3.4
T4W10	26°21'10.44"S	27°27'30.24"E	642.9±35.9	652.3±34.3	610.2±24.0	605.7±18.7	618.7±20.6	618.2±10.8
T4W11	26°21'13.61"S	27°27'38.64"E	217.3±12.2	222.6±11.7	212.0±8.4	212.2±6.7	220.4±7.5	215.9±3.8
T4W12	26°21'12.90"S	27°27'51.18"E	143.0±8.0	144.9±7.6	138.2±5.5	140.7±4.6	145.1±5.0	142.0±2.6
Average±Error			232.0±4.1					
T5W1	26°23'52.20"S	27°24'41.82"E	456.3±25.5	461.3±24.2	432.4±17.0	434.5±13.4	446.5±14.9	442.0±7.7
T5W2	26°23'45.36"S	27°24'39.96"E	245.2±13.7	246.4±13.0	230.3±9.1	230.7±7.3	240.4±8.1	236.1±4.2
T5W3	26°23'42.48"S	27°24'46.92"E	281.6±15.7	283.6±14.9	266.3±10.5	267.9±8.4	281.7±9.5	274.1±4.8
T5W4	26°23'47.82"S	27°24'48.60"E	366.1±20.5	374.8±19.7	351.7±13.9	357.3±11.1	367.6±12.3	361.5±6.3
T5W5	26°23'44.40"S	27°24'54.72"E	297.2±16.6	301.8±15.9	283.6±11.2	286.6±9.0	290.0±9.8	289.5±5.1
T5W6	26°23'43.56"S	27°25'4.86"E	608.9±34.0	616.4±32.4	578.5±22.8	576.6±17.8	592.4±19.7	588.2±10.3
T5W7	26°23'52.38"S	27°25'0.66"E	330.1±18.4	335.2±17.6	314.5±12.4	317.7±9.9	331.8±11.1	323.7±5.7
T5W8	26°23'58.62"S	27°25'1.98"E	422.4±23.6	426.0±22.4	405.5±16.0	407.9±12.6	425.4±14.2	415.3±7.3
T5W9	26°23'58.68"S	27°25'3.60"E	810.6±45.2	817.4±42.9	768.7±30.3	775.0±23.7	796.2±26.3	787.0±13.7
T5W10	26°23'57.96"S	27°25'5.76"E	2149.0±119.9	2158.0±113.3	2029.0±79.8	2013.0±61.3	2057.0±67.7	2054.7±35.6
T5W11	26°23'57.00"S	27°25'7.08"E	1303.0±72.7	1313.0±68.9	1220.0±48.0	1213.0±37.0	1257.0±41.4	1244.1±21.6
T5W12	26°23'53.40"S	27°25'9.24"E	307.2±17.2	312.5±16.4	293.9±11.6	294.4±9.2	306.5±10.3	300.6±5.3
Average±Error			609.7±10.6					
SVW1	26°23'45.41"S	27°28'41.22"E	35.4±2.0	36.0±1.9	33.3±1.4	33.7±1.3	35.0±1.4	34.4±0.7
SVW2	26°23'45.90"S	27°28'43.26"E	55.6±3.1	55.7±3.0	52.3±2.1	53.2±1.9	54.7±2.1	53.9±1.0
SVW3	26°23'44.10"S	27°29'2.04"E	46.8±2.7	47.5±2.5	45.0±1.8	46.2±1.7	45.9±1.8	46.1±0.9
SVW4	26°23'38.88"S	27°29'5.16"E	48.0±2.7	49.5±2.6	46.7±1.9	47.0±1.7	47.2±1.8	47.4±0.9
SVW5	26°23'41.94"S	27°28'57.78"E	45.1±2.6	45.6±2.4	43.2±1.7	41.8±1.6	44.3±1.7	43.6±0.9
SVW6	26°23'43.26"S	27°28'52.50"E	61.0±3.5	63.2±3.4	60.3±2.4	59.0±2.3	62.8±2.4	61.0±1.2
Average±Error			47.7±0.9					
SVE1	26°23'0.78"S	27°30'40.56"E	79.2±4.5	78.8±4.2	73.3±2.9	74.5±2.6	76.9±2.8	75.8±1.4

SVE2	26°23'6.30"S	27°30'48.30"E	43.6±2.5	44.0±2.3	42.1±1.7	43.2±1.6	42.8±1.7	43.0±0.8
SVE3	26°23'13.92"S	27°30'59.76"E	47.8±2.7	48.9±2.6	45.4±1.8	44.9±1.7	47.4±1.8	46.4±0.9
SVE4	26°23'10.38"S	27°31'15.24"E	47.3±2.7	48.3±2.6	46.7±1.9	48.1±1.8	48.4±1.9	47.7±0.9
SVE5	26°23'6.84"S	27°30'53.04"E	53.5±3.0	53.7±2.9	52.3±2.1	54.1±2.0	54.6±2.1	53.6±1.0
SVE6	26°23'18.90"S	27°30'51.36"E	53.9±3.1	54.6±2.9	51.3±2.1	52.2±1.9	52.6±2.0	52.6±1.0
SVE7	26°23'21.24"S	27°31'20.04"E	37.4±2.1	38.6±2.1	35.6±1.4	36.3±1.4	38.6±1.6	37.1±0.7
SVE8	26°23'23.46"S	27°31'11.70"E	46.8±2.7	48.1±2.6	46.4±1.9	46.5±1.6	48.7±1.9	47.2±0.9
SVE9	26°23'16.68"S	27°30'44.10"E	44.9±2.5	45.8±2.4	43.1±1.7	43.7±1.6	47.2±1.8	44.8±0.9
SVE10	26°22'60.00"S	27°30'36.12"E	47.6±2.7	47.8±2.5	45.0±1.8	45.9±1.7	46.7±1.8	46.3±0.9
SVE11	26°23'27.90"S	27°31'0.06"E	57.4±3.2	58.6±3.1	54.7±2.2	56.2±2.0	58.9±2.2	56.9±1.1
Average±Error								
			50.1±1.0					

Table AA2: Calculated Activity Concentrations of ²³⁸U derived from the Weighted Average of Selected Gamma-ray Transitions of ²¹⁴Pb and ²¹⁴Bi in each Rock Sample.

Sample ID	Latitude	Longitude	Average activity concentration of ²³⁸ U (Bq.kg ⁻¹)				Weighted Average
			²¹⁴ Pb		²¹⁴ Bi		
			295.2 keV	351.9 keV	609.3 keV	1120.3 keV	
RD91	26°21'43.86"S	27°27'50.82"E	47.6±20.5	81.0±8.6	74.8±9.9	72.9±21.8	75.3±6.0
RD92	26°21'44.40"S	27°27'50.70"E	4238.0±257.9	4350.0±260.7	3558.0±146.6	3457.0±134.3	3681.6±87.1
RD101	26°24'3.96"S	27°25'56.76"E	246.1±31.8	207.5±20.3	155.2±20.8	186.8±51.4	192.7±12.8
RD102	26°24'3.78"S	27°25'57.72"E	230.4±31.3	238.5±21.7	244.2±22.7	213.2±47.8	237.0±13.4
RD103	26°24'4.26"S	27°25'56.82"E	176.8±25.9	157.5±18.0	199.0±21.5	187.0±52.8	175.7±11.9
RD111	26°22'57.42"S	27°29'38.22"E	51.0±15.0	47.4±8.5	59.5±10.3	57.9±15.1	52.9±5.6
RD121	26°22'46.98"S	27°30'12.72"E	78.8±18.9	83.7±11.4	81.8±12.1	58.5±31.0	80.8±7.4
RD122	26°22'49.20"S	27°30'14.04"E	58.4±12.9	85.4±9.7	55.7±8.4	57.1±21.9	65.9±5.5
Average							570.2±18.7

Table AA3: Calculated Activity Concentrations of ^{232}Th derived from the Weighted Average of Selected Gamma-ray Transitions of ^{228}Ac and ^{212}Pb in each Soil sample.

Sample ID	Latitude	Longitude	Average activity concentration of ²³² Th (Bq.kg ⁻¹)				²¹² Pb		Weighted Average
			²²⁸ Ac						
			338.3 keV	911.6 keV	969.1 keV	238.6 keV			
CS1	25°47'34.62"S	25°37'55.26"E	16.0±1.1	18.5±0.8	18.9±0.8	17.0±1.1			18.0±0.4
CS2	25°47'33.06"S	25°38'2.04"E	19.1±1.3	18.2±0.7	18.4±0.8	21.1±1.3			18.7±0.5
CS3	25°47'34.80"S	25°38'19.20"E	17.5±1.2	16.0±0.7	17.2±0.7	16.2±1.0			16.6±0.4
CS4	25°47'35.46"S	25°38'13.62"E	24.3±1.5	24.8±0.9	25.4±1.0	29.5±1.9			25.3±0.6
CS5	25°47'41.70"S	25°38'3.36"E	18.4±1.1	17.9±0.7	17.8±0.8	20.5±1.3			18.2±0.4
CS6	25°47'46.32"S	25°37'59.16"E	23.1±1.3	22.3±0.8	23.1±0.8	22.7±1.4			22.8±0.5
CS7	25°47'53.10"S	25°37'54.90"E	21.7±1.3	22.8±0.8	22.9±0.9	22.2±1.4			22.6±0.5
CS8	25°47'50.64"S	25°38'1.56"E	26.1±1.5	27.1±1.0	26.5±1.0	26.2±1.6			26.6±0.6
CS9	25°47'53.82"S	25°38'1.92"E	30.2±1.7	30.9±1.0	30.1±1.1	30.2±1.9			30.4±0.6
CS10	25°48'1.86"S	25°37'57.72"E	22.4±1.3	22.1±0.8	22.6±0.8	21.8±1.4			22.3±0.5
Average±Error									
T1E1	26°22'20.52"S	27°29'10.20"E	54.6±2.9	39.9±1.3	40.6±1.4	40.8±2.5			41.5±0.8
T1E2	26°22'20.40"S	27°29'28.74"E	46.4±2.6	37.7±1.3	40.2±1.5	41.1±2.6			39.9±0.9
T1E3	26°22'19.80"S	27°29'40.86"E	41.9±2.6	34.9±1.2	37.5±1.4	37.8±2.4			36.8±0.8
T1E4	26°22'5.64"S	27°29'39.66"E	46.0±2.6	39.9±1.3	39.8±1.4	40.2±2.5			40.5±0.9
T1E5	26°21'56.28"S	27°29'38.76"E	44.6±2.6	38.7±1.4	39.8±1.5	39.3±2.5			39.8±0.9
T1E6	26°21'49.02"S	27°29'37.26"E	42.6±2.5	36.2±1.3	39.4±1.6	29.7±1.9			36.5±0.8
T1E7	26°21'49.38"S	27°29'22.68"E	52.5±3.0	42.1±1.5	41.3±1.6	43.2±2.7			43.0±1.0
T1E8	26°21'52.44"S	27°29'12.24"E	43.5±3.0	38.4±1.4	40.9±1.6	40.2±2.5			39.9±0.9
T1E9	26°22'3.06"S	27°29'11.10"E	51.0±2.9	42.9±1.5	44.7±1.7	43.6±2.7			44.5±1.0
T1E10	26°22'20.94"S	27°29'10.26"E	69.0±3.9	52.0±1.8	53.1±2.0	56.2±3.5			54.5±1.2
T1E11	26°22'21.42"S	27°29'8.46"E	38.4±2.2	36.8±1.3	36.7±1.4	38.8±2.4			37.2±0.8
Average±Error									
T2E1	26°22'33.84"S	27°30'5.88"E	50.7±2.9	42.6±1.5	43.5±1.7	44.7±2.8			44.1±1.0

T2E2	26°22'33.72"S	27°30'19.80"E	43.9±2.5	36.7±1.3	38.6±1.5	39.3±2.5	38.4±0.9
T2E3	26°22'26.64"S	27°30'20.64"E	51.4±3.5	44.3±1.8	48.3±1.8	48.7±3.1	47.2±1.1
T2E4	26°22'10.98"S	27°30'22.08"E	42.6±3.1	39.5±1.4	39.1±1.5	42.1±2.6	39.9±0.9
T2E5	26°22'2.10"S	27°30'23.04"E	53.5±3.1	44.3±1.6	46.1±1.8	46.0±2.9	46.1±1.0
T2E6	26°21'55.86"S	27°30'23.52"E	55.8±3.2	46.5±1.6	49.9±1.9	51.0±3.2	49.2±1.1
T2E7	26°21'56.88"S	27°30'9.84"E	47.7±2.8	39.8±1.4	42.3±1.6	43.6±2.7	41.9±0.9
T2E8	26°21'59.34"S	27°29'58.72"E	55.0±3.2	43.3±1.5	45.7±1.8	46.9±2.9	45.8±1.0
T2E9	26°22'10.08"S	27°29'58.26"E	55.7±3.2	46.9±1.7	51.1±1.9	48.7±3.1	49.5±1.1
T2E10	26°22'19.20"S	27°29'57.66"E	49.1±3.3	42.9±1.5	43.6±1.7	44.9±2.8	44.0±1.0
T2E11	26°22'33.30"S	27°29'56.52"E	37.6±2.8	38.3±1.4	39.9±1.5	39.4±2.5	38.9±0.9
T2E12	26°22'34.26"S	27°30'0.78"E	60.8±3.6	51.8±1.8	56.3±2.1	34.6±2.2	49.5±1.1
T2E13	26°22'35.53"S	27°29'57.88"E	59.9±3.4	47.4±1.6	48.3±1.8	47.7±3.0	49.0±1.1
Average±Error							
T3W1	26°22'21.60"S	27°26'49.98"E	115.6±6.5	78.0±2.7	83.1±3.0	45.4±2.9	71.6±1.6
T3W2	26°22'13.62"S	27°26'48.96"E	116.5±6.5	83.4±2.9	87.1±3.2	50.2±3.2	77.2±1.7
T3W3	26°22'14.16"S	27°26'41.64"E	27.3±2.0	22.3±0.9	24.7±1.1	25.4±1.6	23.9±0.6
T3W4	26°22'15.66"S	27°26'34.74"E	112.0±6.3	84.7±2.9	89.9±3.2	87.1±5.5	89.3±1.9
T3W5	26°22'24.06"S	27°26'35.28"E	22.8±1.9	22.0±0.8	22.4±1.0	23.2±1.5	22.3±0.6
T3W6	26°22'28.20"S	27°26'39.84"E	113.5±6.3	85.2±2.9	89.7±3.2	87.4±5.5	89.7±1.9
T3W7	26°22'29.22"S	27°26'41.04"E	42.3±2.5	31.7±1.2	34.9±1.4	36.3±2.3	34.3±0.8
T3W8	26°22'31.38"S	27°26'43.08"E	76.1±4.4	62.9±2.2	61.1±2.3	63.0±3.9	63.6±1.4
Average±Error							
T4W1	26°21'11.40"S	27°27'50.82"E	28.8±1.7	27.2±1.0	27.0±1.1	28.3±1.8	27.5±0.6
T4W2	26°21'11.34"S	27°27'50.22"E	23.1±2.0	19.9±1.4	19.2±0.9	22.7±1.4	20.5±0.6
T4W3	26°21'3.06"S	27°27'50.16"E	32.2±2.1	31.1±1.2	31.8±1.2	32.5±2.0	31.7±0.7
T4W4	26°20'51.18"S	27°27'49.62"E	29.9±1.8	31.2±1.1	32.3±1.2	31.3±2.0	31.4±0.7
T4W5	26°20'51.66"S	27°27'40.26"E	35.6±2.1	37.6±1.3	37.0±1.4	38.8±2.4	37.2±0.8
T4W6	26°20'53.46"S	27°27'30.66"E	27.8±1.7	28.1±1.1	28.1±1.1	28.7±1.8	28.1±0.7
T4W7	26°21'0.18"S	27°27'25.50"E	50.5±3.2	45.7±1.6	47.2±1.8	50.9±3.2	47.3±1.1
T4W8	26°21'6.18"S	27°27'21.96"E	31.5±2.0	29.3±1.1	30.6±1.2	30.0±1.9	30.1±0.7
T4W9	26°21'7.98"S	27°27'25.62"E	33.1±2.2	30.9±1.2	30.2±1.2	31.6±2.0	31.0±0.7

T4W10	26°21'10.44"S	27°27'30.24"E	52.3±3.5	47.2±1.7	49.9±1.9	50.9±3.2	49.1±1.1
T4W11	26°21'13.61"S	27°27'38.64"E	34.4±2.2	35.1±1.2	34.1±1.3	35.5±2.2	34.7±0.8
T4W12	26°21'12.90"S	27°27'51.18"E	28.5±1.9	29.8±1.1	29.7±1.2	30.7±1.9	29.7±0.7
Average±Error							33.2±0.8
T5W1	26°23'52.20"S	27°24'41.82"E	36.5±2.6	34.8±1.2	37.3±1.4	38.3±2.4	36.2±0.8
T5W2	26°23'45.36"S	27°24'39.96"E	30.7±2.1	30.8±1.1	31.1±1.2	32.7±2.1	31.1±0.7
T5W3	26°23'42.48"S	27°24'46.92"E	33.9±2.2	32.1±1.2	33.5±1.3	32.8±2.1	32.9±0.8
T5W4	26°23'47.82"S	27°24'48.60"E	39.5±2.7	39.1±1.4	40.0±1.5	40.8±2.6	39.7±0.9
T5W5	26°23'44.40"S	27°24'54.72"E	35.6±2.5	33.7±1.3	35.4±1.4	36.7±2.3	34.9±0.8
T5W6	26°23'43.56"S	27°25'4.86"E	44.8±3.0	42.8±1.5	41.7±1.6	45.1±2.8	42.9±1.0
T5W7	26°23'52.38"S	27°25'0.66"E	36.6±2.5	34.0±1.2	35.1±1.3	35.0±2.2	34.8±0.8
T5W8	26°23'58.62"S	27°25'1.98"E	36.3±2.5	35.8±1.3	35.3±1.4	38.1±2.4	35.9±0.8
T5W9	26°23'58.68"S	27°25'3.60"E	52.6±3.4	47.1±1.6	48.8±1.8	50.6±3.2	48.6±1.1
T5W10	26°23'57.96"S	27°25'5.76"E	83.0±4.8	58.8±2.1	66.3±2.5	66.0±4.1	64.3±1.4
T5W11	26°23'57.00"S	27°25'7.08"E	79.1±4.5	63.6±2.2	67.8±2.5	67.6±4.2	67.1±1.4
T5W12	26°23'53.40"S	27°25'9.24"E	25.2±1.8	25.5±1.1	25.0±1.0	25.7±1.6	25.3±0.6
Average±Error							41.1±0.9
SVW1	26°23'45.41"S	27°28'41.22"E	46.2±2.6	45.6±1.5	45.4±1.6	46.6±2.9	45.7±1.0
SVW2	26°23'45.90"S	27°28'43.26"E	53.0±3.0	54.0±1.8	55.4±1.9	54.5±3.4	54.4±1.1
SVW3	26°23'44.10"S	27°29'2.04"E	42.2±2.4	42.3±1.4	43.0±1.5	42.8±2.7	42.6±0.9
SVW4	26°23'38.88"S	27°29'5.16"E	51.1±2.8	52.5±1.7	53.4±1.8	53.7±3.4	52.7±1.1
SVW5	26°23'41.94"S	27°28'57.78"E	49.6±2.8	48.4±1.6	49.3±1.7	48.2±3.0	48.8±1.0
SVW6	26°23'43.26"S	27°28'52.50"E	53.2±3.1	56.0±1.9	56.4±2.0	55.3±3.5	55.7±1.2
Average±Error							50.0±1.0
SVE1	26°23'0.78"S	27°30'40.56"E	75.4±4.1	75.0±2.5	78.6±2.6	77.2±4.8	76.6±1.6
SVE2	26°23'6.30"S	27°30'48.30"E	56.1±3.1	55.7±1.8	56.5±1.9	56.9±3.6	56.2±1.2
SVE3	26°23'13.92"S	27°30'59.76"E	49.8±2.7	53.6±1.8	54.2±1.9	53.6±3.3	53.2±1.1
SVE4	26°23'10.38"S	27°31'15.24"E	41.0±2.4	42.4±1.5	44.9±1.6	40.1±2.5	42.7±0.9
SVE5	26°23'6.84"S	27°30'53.04"E	45.8±2.5	48.7±1.6	50.3±1.8	48.9±3.1	48.8±1.0
SVE6	26°23'18.90"S	27°30'51.36"E	63.4±3.6	64.3±2.1	65.0±2.2	65.7±4.1	64.6±1.3
SVE7	26°23'21.24"S	27°31'20.04"E	42.6±2.4	41.3±1.4	42.3±1.5	42.4±2.7	41.9±0.9

SVE8	26°23'23.46"S	27°31'11.70"E	53.7±3.0	55.7±1.9	56.1±1.9	56.8±3.5	55.7±1.2
SVE9	26°23'16.68"S	27°30'44.10"E	56.1±3.1	55.5±1.8	56.6±1.9	56.8±3.5	56.1±1.2
SVE10	26°22'60.00"S	27°30'36.12"E	55.3±3.1	53.8±1.8	55.9±1.9	55.3±3.5	54.9±1.1
SVE11	26°23'27.90"S	27°31'0.06"E	62.2±3.5	60.3±2.0	61.4±2.1	61.6±3.8	61.1±1.3
Average±Error							
							55.6±1.1

Table AA4: Average Activity Concentrations of ^{238}U , ^{232}Th and ^{40}K for Soil Samples from the Mining Area and from the Control Area.

Location	Sample ID	Latitude	Longitude	Average activity concentrations (Bq.kg ⁻¹)		
				^{238}U	^{232}Th	^{40}K
Control area	CS1	25°47'34.62"S	25°37'55.26"E	12.7±0.3	18.0±0.4	497.4±15.3
	CS2	25°47'33.06"S	25°38'2.04"E	11.3±0.3	18.7±0.5	648.4±19.8
	CS3	25°47'34.80"S	25°38'19.20"E	13.0±0.3	16.6±0.4	515.2±15.8
	CS4	25°47'35.46"S	25°38'13.62"E	18.0±0.4	25.3±0.6	493.3±15.1
	CS5	25°47'41.70"S	25°38'3.36"E	12.5±0.3	18.2±0.4	582.3±17.8
	CS6	25°47'46.32"S	25°37'59.16"E	18.1±0.4	22.8±0.5	433.7±13.2
	CS7	25°47'53.10"S	25°37'54.90"E	18.7±0.4	22.6±0.5	424.3±13.0
	CS8	25°47'50.64"S	25°38'1.56"E	22.5±0.5	26.6±0.6	452.7±13.8
	CS9	25°47'53.82"S	25°38'1.92"E	23.6±0.5	30.4±0.6	475.7±14.5
	CS10	25°48'1.86"S	25°37'57.72"E	17.4±0.4	22.3±0.5	444.6±13.5
	Average±Error			17.0±0.4	22.2±0.5	496.8±15.2
	Min±Error			12.5±0.3	16.6±0.4	424.3±13.0
	Max±Error			23.6±0.5	30.4±0.6	648.4±19.8
Tailings one	T1E1	26°22'20.52"S	27°29'10.20"E	695.4±12.0	41.5±0.8	358.7±10.9
	T1E2	26°22'20.40"S	27°29'28.74"E	712.3±12.4	39.9±0.9	324.4±10.1
	T1E3	26°22'19.80"S	27°29'40.86"E	647.8±11.2	36.8±0.8	297.1±9.2
	T1E4	26°22'5.64"S	27°29'39.66"E	776.1±13.5	40.5±0.9	290.6±9.0
	T1E5	26°21'56.28"S	27°29'38.76"E	669.4±11.7	39.8±0.9	468.4±14.6
	T1E6	26°21'49.02"S	27°29'37.26"E	697.2±12.1	36.5±0.8	327.0±10.4
	T1E7	26°21'49.38"S	27°29'22.68"E	832.2±14.5	43.0±1.0	294.9±9.5
	T1E8	26°21'52.44"S	27°29'12.24"E	713.5±12.4	39.9±0.9	296.9±9.5
	T1E9	26°22'3.06"S	27°29'11.10"E	775.9±13.5	44.5±1.0	378.5±11.9
	T1E10	26°22'20.94"S	27°29'10.26"E	1243.4±21.6	54.5±1.2	271.5±9.0
	T1E11	26°22'21.42"S	27°29'8.46"E	304.4±5.4	37.2±0.8	430.5±13.4
	Average±Error			733.4±12.7	41.3±0.9	339.9±10.7
	Min±Error			304.4±5.4	36.5±0.8	271.5±9.0
	Max±Error			1243.4±21.6	54.5±1.2	468.4±14.6
Tailings two	T2E1	26°22'33.84"S	27°30'5.88"E	676.8±11.8	44.1±1.0	496.7±15.4
	T2E2	26°22'33.72"S	27°30'19.80"E	648.4±11.3	38.4±0.9	353.6±11.1
	T2E3	26°22'26.64"S	27°30'20.64"E	740.7±12.9	47.2±1.1	591.9±18.3
	T2E4	26°22'10.98"S	27°30'22.08"E	718.5±12.5	39.9±0.9	357.4±11.2
	T2E5	26°22'2.10"S	27°30'23.04"E	816.2±14.2	46.1±1.0	543.2±16.8
	T2E6	26°21'55.86"S	27°30'23.52"E	818.0±14.2	49.2±1.1	554.7±17.2
	T2E7	26°21'56.88"S	27°30'9.84"E	841.5±14.6	41.9±0.9	359.0±11.4
	T2E8	26°21'59.34"S	27°29'58.72"E	862.9±15.0	45.8±1.0	485.9±15.1
	T2E9	26°22'10.08"S	27°29'58.26"E	716.0±12.5	49.5±1.1	681.9±21.0
	T2E10	26°22'19.20"S	27°29'57.66"E	681.2±11.9	44.0±1.0	537.1±16.6
	T2E11	26°22'33.30"S	27°29'56.52"E	616.6±10.7	38.9±0.9	453.9±14.1
	T2E12	26°22'34.26"S	27°30'0.78"E	1391.8±24.1	49.5±1.1	226.5±7.8
	T2E13	26°22'35.53"S	27°29'57.88"E	805.8±14.0	49.0±1.1	347.6±11.0
	Average±Error			794.9±13.8	44.9±1.0	460.7±14.4
	Min±Error			616.6±10.7	38.4±0.9	226.5±7.8

		Max±Error	1391.8±24.1	49.5±1.1	681.9±21.0
Tailings three	T3W1	26°22'21.60"S 27°26'49.98"E	1391.8±24.1	71.6±1.6	409.3±13.3
	T3W2	26°22'13.62"S 27°26'48.96"E	2668.9±46.2	77.2±1.7	424.4±13.8
	T3W3	26°22'14.16"S 27°26'41.64"E	476.6±8.3	23.9±0.6	240.6±7.7
	T3W4	26°22'15.66"S 27°26'34.74"E	2240.6±38.8	89.3±1.9	497.1±15.6
	T3W5	26°22'24.06"S 27°26'35.28"E	390.9±6.8	22.3±0.6	233.8±7.5
	T3W6	26°22'28.20"S 27°26'39.84"E	2617.0±45.3	89.7±1.9	363.0±12.1
	T3W7	26°22'29.22"S 27°26'41.04"E	759.7±13.2	34.3±0.8	392.1±12.3
	T3W8	26°22'31.38"S 27°26'43.08"E	1904.5±33.0	63.6±1.4	274.1±9.3
		Average±Error	1556.2±27.0	59.0±1.3	354.3±11.4
		Min±Error	390.9±6.8	22.3±0.6	233.8±7.5
		Max±Error	2668.9±46.2	89.7±1.9	497.1±15.6
Tailings four	T4W1	26°21'11.40"S 27°27'50.82"E	152.4±2.7	27.5±0.6	360.4±11.2
	T4W2	26°21'11.34"S 27°27'50.22"E	397.3±7.0	20.5±0.6	257.9±8.3
	T4W3	26°21'3.06"S 27°27'50.16"E	142.7±2.6	31.7±0.7	497.3±15.3
	T4W4	26°20'51.18"S 27°27'49.62"E	87.2±1.6	31.4±0.7	366.3±11.3
	T4W5	26°20'51.66"S 27°27'40.26"E	182.5±3.2	37.2±0.8	578.1±17.7
	T4W6	26°20'53.46"S 27°27'30.66"E	143.6±2.6	28.1±0.7	420.1±12.9
	T4W7	26°21'0.18"S 27°27'25.50"E	373.0±6.6	47.3±1.1	781.0±23.9
	T4W8	26°21'6.18"S 27°27'21.96"E	135.2±2.4	30.1±0.7	449.7±13.8
	T4W9	26°21'7.98"S 27°27'25.62"E	193.6±3.4	31.0±0.7	444.8±13.7
	T4W10	26°21'10.44"S 27°27'30.24"E	618.2±10.8	49.1±1.1	758.6±23.2
	T4W11	26°21'13.61"S 27°27'38.64"E	215.9±3.8	34.7±0.8	529.6±16.2
	T4W12	26°21'12.90"S 27°27'51.18"E	142.0±2.6	29.7±0.7	430.0±1.3
		Average±Error	232.0±4.1	33.2±0.8	489.5±14.1
		Min±Error	87.2±1.6	20.5±0.6	257.9±8.3
		Max±Error	618.2±10.8	49.1±1.1	781.0±23.9
Tailings five	T5W1	26°23'52.20"S 27°24'41.82"E	442.0±7.7	36.2±0.8	513.6±15.8
	T5W2	26°23'45.36"S 27°24'39.96"E	236.1±4.2	31.1±0.7	482.7±14.8
	T5W3	26°23'42.48"S 27°24'46.92"E	274.1±4.8	32.9±0.8	492.0±15.1
	T5W4	26°23'47.82"S 27°24'48.60"E	361.5±6.3	39.7±0.9	554.2±17.0
	T5W5	26°23'44.40"S 27°24'54.72"E	289.5±5.1	34.9±0.8	566.9±17.4
	T5W6	26°23'43.56"S 27°25'4.86"E	588.2±10.3	42.9±1.0	574.9±17.7
	T5W7	26°23'52.38"S 27°25'0.66"E	323.7±5.7	34.8±0.8	531.4±16.3
	T5W8	26°23'58.62"S 27°25'1.98"E	415.3±7.3	35.9±0.8	493.6±15.2
	T5W9	26°23'58.68"S 27°25'3.60"E	787.0±13.7	48.6±1.1	517.2±16.0
	T5W10	26°23'57.96"S 27°25'5.76"E	2054.7±35.6	64.3±1.4	460.5±14.7
	T5W11	26°23'57.00"S 27°25'7.08"E	1244.1±21.6	67.1±1.4	419.6±13.3
	T5W12	26°23'53.40"S 27°25'9.24"E	300.6±5.3	25.3±0.6	281.2±8.8
		Average±Error	609.7±10.6	41.1±0.9	490.7±15.2
		Min±Error	236.1±4.2	25.3±0.6	281.2±8.8
		Max±Error	2054.7±35.6	67.1±1.4	574.9±17.7
West village	SVW1	26°23'45.41"S 27°28'41.22"E	34.4±0.7	45.7±1.0	187.3±5.9
	SVW2	26°23'45.90"S 27°28'43.26"E	53.9±1.0	54.4±1.1	308.5±9.6
	SVW3	26°23'44.10"S 27°29'2.04"E	46.1±0.9	42.6±0.9	190.1±6.0
	SVW4	26°23'38.88"S 27°29'5.16"E	47.4±0.9	52.7±1.1	160.4±5.1
	SVW5	26°23'41.94"S 27°28'57.78"E	43.6±0.9	48.8±1.0	178.6±5.7
	SVW6	26°23'43.26"S 27°28'52.50"E	61.0±1.2	55.7±1.2	362.0±11.3

		Average±Error		47.7±0.9	50.0±1.0	231.2±7.3	
		Min±Error		34.4±0.7	42.6±0.9	160.4±5.1	
		Max±Error		61.0±1.2	55.7±1.2	362.0±11.3	
East village	SVE1	26°23'0.78"S	27°30'40.56"E	75.8±1.4	76.6±1.6	280.8±8.8	
	SVE2	26°23'6.30"S	27°30'48.30"E	43.0±0.8	56.2±1.2	183.1±5.8	
	SVE3	26°23'13.92"S	27°30'59.76"E	46.4±0.9	53.2±1.1	238.4±7.5	
	SVE4	26°23'10.38"S	27°31'15.24"E	47.7±0.9	42.7±0.9	262.1±8.3	
	SVE5	26°23'6.84"S	27°30'53.04"E	53.6±1.0	48.8±1.0	246.8±7.8	
	SVE6	26°23'18.90"S	27°30'51.36"E	52.6±1.0	64.6±1.3	261.5±8.2	
	SVE7	26°23'21.24"S	27°31'20.04"E	37.1±0.7	41.9±0.9	133.8±4.4	
	SVE8	26°23'23.46"S	27°31'11.70"E	47.2±0.9	55.7±1.2	213.1±6.8	
	SVE9	26°23'16.68"S	27°30'44.10"E	44.8±0.9	56.1±1.2	198.4±6.3	
	SVE10	26°22'60.00"S	27°30'36.12"E	46.3±0.9	54.9±1.1	167.5±5.4	
	SVE11	26°23'27.90"S	27°31'0.06"E	56.9±1.1	61.1±1.3	226.6±7.1	
			Average±Error		50.1±1.0	55.6±1.2	219.3±7.0
		Min±Error		37.1±0.7	41.9±0.9	133.8±4.4	
		Max±Error		75.8±1.4	76.6±1.6	280.8±8.8	
Rock dumps	RD91	26°21'43.86"S	27°27'50.82"E	75.3±6.0	23.5±3.3	527.8±82.9	
	RD92	26°21'44.40"S	27°27'50.70"E	3681.6±87.1	167.5±12.6	569.3±125.9	
	RD101	26°24'3.96"S	27°25'56.76"E	192.7±12.8	100.0±9.2	626.4±145.5	
	RD102	26°24'3.78"S	27°25'57.72"E	237.0±13.4	56.6±7.0	691.9±157.2	
	RD103	26°24'4.26"S	27°25'56.82"E	175.7±11.9	108.5±9.6	920.0±156.4	
	RD111	26°22'57.42"S	27°29'38.22"E	52.9±5.6	21.9±7.0	830.7±100.9	
	RD121	26°22'46.98"S	27°30'12.72"E	80.8±7.4	34.9±7.0	1152.0±120.4	
	RD122	26°22'49.20"S	27°30'14.04"E	65.9±5.5	49.5±8.4	1177.0±114.1	
			Average±Error		570.2±18.7	70.3±8.0	811.9±125.4
			Min±Error		52.9±5.6	21.9±7.0	527.8±82.7
			Max±Error		3681.6±87.1 <th>167.5±12.6<th>1177.0±114.1</th></th>	167.5±12.6 <th>1177.0±114.1</th>	1177.0±114.1
			Overall Average		574.3±39.5	49.4±8.5	424.7±129.3
		Overall Minimum		34.4±0.7	20.5±0.6	133.8±4.4	
		Overall Maximum		3681.6±87.1 <th>167.5±12.6<th>1177.0±114.1</th></th>	167.5±12.6 <th>1177.0±114.1</th>	1177.0±114.1	

Table AA5: Results of Absorbed Dose Rate in air (D), Annual Effective Dose Equivalent (AEDE), Radium Equivalent Activity (Ra_{eq}), External Hazard Index (H_{ex}) and Internal hazard Index (H_{in}) of Soil Samples from the Mining Area and from the Control Area.

Sample ID	Ra_{eq} (Bq.kg ⁻¹)	D (nGy.y ⁻¹)	AEDE (mSv.y ⁻¹)	H_{ex}	H_{in}
CS1	76.7±1.4	37.5±0.7	0.05	0.2	0.2
CS2	88.0±1.7	43.5±0.9	0.05	0.2	0.3
CS3	76.4±1.4	37.5±0.7	0.05	0.2	0.2
CS4	92.2±1.5	44.2±0.7	0.05	0.2	0.3
CS5	83.4±1.5	41.1±0.8	0.05	0.2	0.3
CS6	84.0±1.3	40.2±0.7	0.05	0.2	0.3
CS7	83.7±1.3	40.0±0.6	0.05	0.2	0.3
CS8	95.3±1.4	45.3±0.7	0.06	0.3	0.3
CS9	103.8±1.5	49.1±0.8	0.06	0.3	0.3
CS10	83.5±1.3	40.0±0.7	0.05	0.2	0.3
Average±Error	86.9±1.4	41.9±0.7	0.05	0.2	0.3
Min±Error	76.4±1.4	37.5±0.7	0.05	0.2	0.2
Max±Error	103.8±1.5	49.1±0.8	0.06	0.3	0.3
T1E1	782.3±12.1	361.3±5.6	0.44	2.1	4.0
T1E2	794.4±12.5	366.7±5.8	0.45	2.1	4.1
T1E3	723.3±11.3	333.9±5.2	0.41	2.0	3.7
T1E4	856.5±13.5	395.2±6.3	0.48	2.3	4.4
T1E5	762.4±11.8	352.8±5.4	0.43	2.1	3.9
T1E6	774.6±12.2	357.8±5.7	0.44	2.1	4.0
T1E7	916.4±14.6	422.7±6.7	0.52	2.5	4.7
T1E8	793.5±12.5	366.1±5.8	0.45	2.1	4.1
T1E9	868.6±13.6	401.1±6.3	0.49	2.3	4.4
T1E10	1342.2±21.7	618.7±10.0	0.76	3.6	7.0
T1E11	390.8±5.6	181.1±2.6	0.22	1.1	1.9
Average±Error	818.6±12.9	377.9±5.9	0.46	2.2	4.2
Min±Error	390.8±5.6	181.1±2.6	0.22	1.1	1.9
Max±Error	1342.2±21.7	618.7±10.0	0.76	3.6	7.0
T2E1	778.0±11.9	352.8±5.5	0.44	2.1	3.9
T2E2	730.6±11.4	261.8±5.3	0.41	2.0	3.7
T2E3	853.7±13.1	395.4±6.0	0.48	2.3	4.3
T2E4	803.1±12.6	371.0±5.8	0.45	2.2	4.1
T2E5	924.0±14.3	427.6±6.6	0.52	2.5	4.7
T2E6	931.1±14.4	430.8±6.6	0.53	2.5	4.7
T2E7	929.1±14.7	429.1±6.8	0.53	2.5	4.8
T2E8	965.7±15.1	446.5±7.0	0.55	2.6	4.9
T2E9	839.2±12.7	389.1±5.9	0.48	2.3	4.2
T2E10	785.4±12.0	363.7±5.6	0.45	2.1	4.0
T2E11	707.2±10.9	327.3±5.0	0.40	1.9	3.6
T2E12	1480.0±24.2	682.3±11.2	0.84	4.0	7.8

T2E13	902.6±14.1	416.4±6.5	0.51	2.4	4.6
Average±Error	894.6±14.0	413.6±6.4	0.51	2.4	4.6
Min±Error	707.2±10.9	327.3±5.0	0.40	1.9	3.6
Max±Error	1480.0±24.2	682.3±11.2	0.84	4.0	7.8
T3W1	1525.7±24.3	703.3±11.2	0.86	4.1	7.9
T3W2	2811.9±46.3	1297.3±21.4	1.59	7.6	14.8
T3W3	529.3±8.4	244.6±3.9	0.30	1.4	2.7
T3W4	2406.6±38.9	1109.8±18.0	1.36	6.5	12.6
T3W5	440.9±6.9	203.9±3.2	0.25	1.2	2.2
T3W6	2773.2±45.4	1278.4±21.0	1.57	7.5	14.6
T3W7	839.0±13.3	388.1±6.1	0.48	2.3	4.3
T3W8	2016.6±33.1	929.7±15.3	1.14	5.4	10.6
Average±Error	1667.9±27.1	769.4±12.5	0.94	4.5	8.7
Min±Error	440.9±6.9	203.9±3.2	0.25	1.2	2.2
Max±Error	2811.9±46.3	1297.3±21.4	1.59	7.6	14.8
T4W1	219.5±3.0	102.0±1.4	0.13	0.6	1.0
T4W2	446.4±7.1	206.6±3.3	0.25	1.2	2.3
T4W3	226.2±3.0	105.8±1.4	0.13	0.6	1.0
T4W4	160.3±2.1	74.5±1.0	0.09	0.4	0.7
T4W5	280.2±3.7	130.9±1.7	0.16	0.8	1.3
T4W6	216.1±2.9	100.8±1.4	0.12	0.6	1.0
T4W7	500.7±7.0	233.4±3.3	0.29	1.4	2.4
T4W8	212.9±2.8	99.4±1.3	0.12	0.6	0.9
T4W9	272.1±3.7	126.7±1.7	0.16	0.7	1.3
T4W10	746.7±11.0	346.9±5.1	0.43	2.0	3.7
T4W11	306.3±4.2	142.8±1.9	0.18	0.8	1.4
T4W12	217.6±2.7	101.5±1.3	0.12	0.6	1.0
Average±Error	317.1±4.4	147.6±2.1	0.18	0.9	1.5
Min±Error	160.3±2.1	74.5±1.0	0.09	0.4	0.7
Max±Error	746.7±11.0	346.9±5.1	0.43	2.0	3.7
T5W1	533.4±7.9	247.5±3.7	0.30	1.4	2.6
T5W2	317.8±4.4	148.0±2.1	0.18	0.9	1.5
T5W3	359.1±5.1	167.0±2.4	0.20	1.0	1.7
T5W4	460.9±6.6	214.1±3.1	0.26	1.2	2.2
T5W5	383.0±5.4	178.5±2.5	0.22	1.0	1.8
T5W6	693.8±10.4	321.6±4.8	0.39	1.9	3.5
T5W7	414.4±5.9	192.7±2.8	0.24	1.1	2.0
T5W8	504.7±7.4	234.1±3.5	0.29	1.4	2.5
T5W9	896.4±13.8	414.5±6.4	0.51	2.4	4.5
T5W10	2182.1±35.7	1007.3±16.5	1.24	5.9	11.5
T5W11	1372.3±21.7	632.8±10.0	0.78	3.7	7.1
T5W12	358.4±5.4	165.9±2.5	0.20	1.0	1.8
Average±Error	706.4±10.8	327.0±5.0	0.40	1.9	3.6
Min±Error	317.8±4.4	148.0±2.1	0.18	0.9	1.5
Max±Error	2182.1±35.7	1007.3±16.5	1.24	5.9	11.5
SVW1	114.2±1.6	51.3±0.7	0.06	0.3	0.4
SVW2	155.4±2.1	70.6±0.9	0.09	0.4	0.6
SVW3	121.6±1.6	54.9±0.7	0.07	0.3	0.5

SVW4	135.1±1.8	60.4±0.8	0.07	0.4	0.5
SVW5	127.2±1.8	57.1±0.8	0.07	0.3	0.5
SVW6	168.5±2.2	76.9±1.0	0.09	0.5	0.6
Average±Error	137.0±1.9	61.9±0.8	0.08	0.4	0.5
Min±Error	114.2±1.6	51.3±0.7	0.06	0.3	0.4
Max±Error	168.5±2.2	76.9±1.0	0.09	0.5	0.6
SVE1	206.9±2.7	93.0±1.2	0.11	0.6	0.8
SVE2	137.4±1.9	61.4±0.8	0.08	0.4	0.5
SVE3	140.8±1.9	63.5±0.8	0.08	0.4	0.5
SVE4	129.0±1.7	58.8±0.8	0.07	0.3	0.5
SVE5	142.4±1.9	64.5±0.8	0.08	0.4	0.5
SVE6	165.1±2.3	74.2±1.0	0.09	0.4	0.6
SVE7	107.4±1.5	48.0±0.7	0.06	0.3	0.4
SVE8	143.2±2.0	64.3±0.9	0.08	0.4	0.5
SVE9	140.3±1.9	62.9±0.8	0.08	0.4	0.5
SVE10	137.7±1.9	61.5±0.8	0.08	0.4	0.5
SVE11	161.7±2.2	72.6±1.0	0.09	0.4	0.6
Average±Error	146.5±2.0	65.9±0.9	0.08	0.4	0.5
Min±Error	107.4±1.5	48.0±0.7	0.06	0.3	0.4
Max±Error	206.9±2.7	93.0±1.2	0.11	0.6	0.8
RD91	149.6±9.9	71.0±4.8	0.09	0.4	0.6
RD92	3964.9±89.5	1825.8±41.3	2.24	10.7	20.7
RD101	383.9±21.5	175.6±10.1	0.22	1.0	1.6
RD102	371.2±20.7	172.5±10.0	0.21	1.0	1.6
RD103	401.7±21.8	185.1±10.3	0.23	1.1	1.6
RD111	148.2±13.8	72.3±6.5	0.09	0.4	0.5
RD121	219.4±15.5	106.5±7.4	0.13	0.6	0.8
RD122	227.3±15.9	109.4±7.4	0.13	0.6	0.8
Average±Error	733.3±26.1	339.8±12.2	0.42	2.0	3.5
Min±Error	148.2±13.8	71.0±4.8	0.09	0.4	0.5
Max±Error	3964.9±89.5	1825.8±41.3	2.24	10.7	20.7
Overall Average	677.7±43.8	312.9±20.3	0.38	1.8	3.4
Overall Min	107.4±1.5	48.0±0.7	0.06	0.3	0.4
Overall Max	3964.9±89.5	1825.8±41.3	2.24	10.7	20.7

Appendix AB: Description of Sampling Points for Plant Samples from the Mining Area and from the Control Area.

Location	Sample ID	Latitude	Longitude	Description
Control area	CV1	25°47'53.82"S	25°38'1.92"E	Pumpkin Leaves from the Control Area
Mining area	VV91	26°23'13.80"S	27°30'59.76"E	Spinach leafy vegetable from East Village
	VV92	26°23'13.80"S	27°30'59.76"E	Turnip leafy vegetable sample from East Village
	VV101	26°23'10.38"S	27°31'15.24"E	Tomato fruit sample from East Village
	VV102	26°23'10.38"S	27°31'15.24"E	Turnip vegetable sample from East Village.
	VV11	26°23'6.84"S	27°30'53.04"E	Mulberry fruit sample from East Village
	VV14	26°23'23.46"S	27°31'11.70"E	Lime fruit from East Village
	VV15	26°23'16.68"S	27°30'44.10"E	Mulberry fruit from East Village
	VV17	26°23'27.90"S	27°31'0.06"E	Mulberry fruit from East Village
	DSV181	26°23'0.12"S	27°30'14.10"E	Mustard green leafy vegetable grown on sewage treatment point DSW18
	DSV3615	26°20'28.44"S	27°25'42.06"E	Mustard green leafy vegetable grown on sewage treatment point DSW36
11 Plant Samples				

Appendix AC: Description of Water Sampling Points from the Mining Area and from the Control Area.

Location	Sample ID	Latitude	Longitude	Mass of Sample	pH	Description
Control area	CW1	25°47'33.06"S	25°38'2.04"E	1356.67	8.066	Borehole drinking water from control area.
	CW2	25°47'34.80"S	25°38'19.20"E	1391.49	8.101	Borehole drinking water from control area.
	CW3	25°47'41.58"S	25°38'3.36"E	1403.53	7.992	Borehole drinking water from control area.
	CW4	25°47'59.70"S	25°37'55.20"E	1650.34	7.984	Borehole drinking water from control area.
	CW5	25°48'1.32"S	25°37'56.22"E	1633.61	8.100	Borehole drinking water from control area.
Mining area	DSW451	26°26'19.74"S	27°29'54.84"E	1944.25	7.413	Stream water from mine shaft that flows into a private dam for public use.
	DSW112	26°25'28.02"S	27°29'31.50"E	1971.86	7.495	Sewage water from mine shaft that flows into a mine dam.
	DSW183	26°23'0.12"S	27°30'14.10"E	1911.11	7.679	Sewage water from mine shaft. The water is treated at this point and eventually flows to the settling point through a canal via a series of mine dams. The settling point has 16 ponds that help purify the water.
	DSW204	26°22'45.12"S	27°29'52.44"E	1977.01	7.341	Storm water from run-off around the mine that flows through a canal to the return water dam of mine tailings 1 and 2. The water from the return dam is pumped back to the process plant for reuse.
	DSW405	26°21'43.08"S	27°29'58.86"E	1954.16	7.909	Raw water of return water dam, section 1, of mine tailings 1 and 2. The water from the return dam is pumped back to the process plant for reuse
	DSW406	26°21'43.26"S	27°30'5.28"E	2176.65	7.950	Raw water of return water dam, section 2, of mine tailings 1 and 2. The water from the return dam is pumped back to process plant for reuse
	DSW407	26°21'53.28"S	27°29'54.54"E	2069.46	8.096	Raw water before return water dam of mine tailings 1 and 2. The water from the return dam is pumped back to process plant for reuse.

DSW358	26°21'49.56"S 27°27'39.90"E	2178.51	7.556	Sewage water from mine shaft, treated at this point and can be used by the public.
DSW179	26°23'22.74"S 27°28'13.92"E	2344.70	7.755	Sewage water from DSW183 originally from mine shaft. The water is used for irrigation and eventually flows to the settling point via a canal.
DSW4210	26°20'36.24"S 27°26'1.02"E	2381.38	8.150	Fissure water straight from underground used for public use
DSW2111	26°21'25.14"S 27°26'10.32"E	2322.63	7.996	Process water from the water plant for public use.
DSW712	26°21'27.42"S 27°25'59.22"E	1926.40	8.336	Water coming from the mine shaft and from process plants. The water flows to the settling point via a canal.
DAM313	26°21'19.32"S 27°25'52.44"E	1946.70	8.077	Water from the settling point. The settling point has 16 ponds that help purify the water.
DSW914	26°20'59.34"S 27°25'52.44"E	1832.41	8.020	Water from the settling point. It flows via a canal down for blending with fissure water.
DSW3615	26°20'28.44"S 27°25'42.06"E	1915.80	8.040	Blended water from DSW914 and DSW4210 used by the public. There is also an informal settlement nearby.
DSW3816	26°19'38.10"S 27°24'15.42"E	1936.42	7.765	Water from 1 m pipe discharge from another mine.
DSW3917	26°18'57.96"S 27°23'3.36"E	1932.80	8.041	Mixed water from DSW3615 and DSW3816. The water flows outside mine property for public use.
DSW1618	26°23'44.76"S 27°25'57.84"E	1943.32	8.066	The water comes from the mine shaft and from process plants.
DSW4319	26°21'57.30"S 27°26'34.98"E	1856.50	8.113	Purified water for farm irrigation and domestic use.
DSW2920	26°20'32.40"S 27°26'40.86"E	1960.40	8.210	Raw water from the return water dam of mine tailings number 4. The water is recycled back to the process plant for reuse.
WV2	26°23'45.90"S 27°28'43.26"E	2013.10	7.115	Borehole water for public use from west village.
WV3	26°23'44.10"S 27°29'2.04"E	1911.65	7.688	
WV4	26°23'38.88"S 27°29'5.16"E	1950.75	7.777	
WV5	26°23'40.20"S 27°29'5.52"E	1972.40	7.257	
WV9	26°23'13.80"S 27°30'59.76"E	2005.24	7.807	Borehole water for public use from east village.

WV13	26°23'21.24"S	27°31'20.04"E	1962.02	7.489
WV14	26°23'23.46"S	27°31'11.70"E	1905.26	7.863
WV15	26°23'16.68"S	27°30'44.10"E	1942.55	8.187
WV16	26°22'60.00"S	27°30'36.12"E	1940.92	7.860
34 Water Samples				

APPENDIX B: Concentrations of Heavy Metals in Soil from the Mining and Control Area.

Location	Sample ID	Latitude	Longitude	Concentrations of heavy metals (mg.kg ⁻¹)									
				As	Pb	Hg	Cd	Cr	Cu	Zn	Co	Ni	
Control Area	CS1	25°47'34.62"S	25°37'55.26"E	1.50	0.90	0.00	0.00	34.20	7.20	9.90	7.40	0.00	
	CS2	25°47'33.06"S	25°38'2.04"E	0.10	3.90	0.00	0.00	45.40	6.60	13.70	12.90	0.00	
	CS3	25°47'34.80"S	25°38'19.20"E	2.30	1.60	0.00	0.00	29.80	6.00	15.80	2.00	0.00	
	CS4	25°47'35.46"S	25°38'13.62"E	2.40	1.70	0.00	0.00	24.90	7.10	14.60	2.10	0.00	
	CS5	25°47'41.70"S	25°38'3.36"E	3.50	2.40	0.00	0.00	20.20	5.90	12.20	2.00	0.00	
	CS6	25°47'46.32"S	25°37'59.16"E	2.20	2.10	0.00	0.00	54.40	11.40	11.90	9.20	0.00	
	CS7	25°47'53.10"S	25°37'54.90"E	3.30	1.80	0.00	0.00	31.90	8.10	10.10	3.30	0.00	
	CS8	25°47'50.64"S	25°38'1.56"E	2.30	2.40	0.00	0.00	71.70	13.20	17.00	7.20	0.00	
	CS9	25°47'53.82"S	25°38'1.92"E	4.50	3.90	0.00	0.00	74.90	12.20	16.00	8.00	0.00	
	CS10	25°48'1.86"S	25°37'57.72"E	2.90	2.70	0.00	0.00	59.80	8.50	20.50	3.90	0.00	
Mine Tailings One				Average±Error	2.50	2.34	0.00	0.00	44.72	8.62	14.17	5.80	0.00
	T1E1	26°22'20.52"S	27°29'10.20"E	108.90	12.80	0.20	0.10	610.90	49.10	54.50	32.00	121.10	
	T1E2	26°22'20.40"S	27°29'28.74"E	84.60	7.50	0.00	0.10	58.70	48.40	27.50	28.10	91.40	
	T1E3	26°22'19.80"S	27°29'40.86"E	96.60	7.10	0.10	0.00	491.70	47.70	38.10	32.50	107.10	
	T1E4	26°22'5.64"S	27°29'39.66"E	153.80	7.50	0.10	0.00	554.10	53.00	42.40	48.50	136.90	
	T1E5	26°21'56.28"S	27°29'38.76"E	106.70	5.20	0.10	0.10	695.60	46.30	41.70	26.30	110.70	
	T1E6	26°21'49.02"S	27°29'37.26"E	69.30	8.80	0.10	0.00	50.70	33.50	27.30	23.30	78.20	
	T1E7	26°21'49.38"S	27°29'22.68"E	100.90	9.50	0.20	0.10	486.40	40.50	42.90	35.70	107.20	
	T1E8	26°21'52.44"S	27°29'12.24"E	106.10	7.00	0.10	0.00	72.00	42.40	36.40	33.60	99.00	
	T1E9	26°22'3.06"S	27°29'11.10"E	79.60	13.00	0.10	0.10	519.50	38.70	46.50	27.50	195.50	
	T1E10	26°22'20.94"S	27°29'10.26"E	86.30	10.40	0.20	0.10	434.90	51.60	35.60	41.60	176.20	
T1E11	26°22'21.42"S	27°29'8.46"E	43.10	8.50	0.20	0.00	882.20	63.40	114.70	41.40	218.10		
Mine Tailings Two				Average±Error	94.17	8.85	0.13	0.05	441.52	46.78	46.15	33.68	131.04
	T2E1	26°22'33.84"S	27°30'5.88"E	48.00	6.70	0.00	0.10	57.20	35.10	47.80	17.90	116.90	
	T2E2	26°22'33.72"S	27°30'19.80"E	65.00	7.40	0.10	0.00	62.60	37.70	56.30	20.90	68.50	
	T2E3	26°22'26.64"S	27°30'20.64"E	218.30	17.10	0.20	0.10	160.30	74.90	114.70	45.00	184.40	
	T2E4	26°22'10.98"S	27°30'22.08"E	299.50	15.80	0.20	0.10	91.90	47.40	73.50	59.00	143.10	
	T2E5	26°22'2.10"S	27°30'23.04"E	73.50	5.10	0.20	0.00	454.60	36.10	43.80	22.50	92.50	

	T2E6	26°21'55.86"S	27°30'23.52"E	72.60	4.00	0.20	0.10	79.00	38.30	46.80	25.10	93.50
	T2E7	26°21'56.88"S	27°30'9.84"E	86.70	6.80	0.10	0.00	81.00	34.20	34.80	25.20	86.30
	T2E8	26°21'59.34"S	27°29'58.72"E	133.90	8.60	0.20	0.00	899.00	48.50	56.30	33.20	147.50
	T2E9	26°22'10.08"S	27°29'58.26"E	63.50	7.60	0.10	0.00	73.20	34.10	40.20	19.80	77.50
	T2E10	26°22'19.20"S	27°29'57.66"E	95.30	9.90	0.10	0.10	685.30	45.40	55.90	31.30	116.10
	T2E11	26°22'33.30"S	27°29'56.52"E	75.40	7.40	0.10	0.00	72.40	36.40	35.00	23.70	83.20
	T2E12	26°22'34.26"S	27°30'0.78"E	207.90	28.00	0.10	0.00	75.10	71.50	26.70	48.20	97.10
	T2E13	26°22'35.53"S	27°29'57.88"E	57.90	8.50	0.10	0.10	728.30	51.60	43.60	41.10	189.40
	Average±Error 115.19 10.22 0.13 0.05 270.76 45.48 51.95 31.76 115.08											
Mine Tailings Three	T3W1	26°22'21.60"S	27°26'49.98"E	95.60	4.30	0.10	0.00	130.00	110.00	230.00	90.00	220.00
	T3W2	26°22'13.62"S	27°26'48.96"E	39.40	2.00	0.10	0.10	150.00	100.00	200.00	70.00	210.00
	T3W3	26°22'14.16"S	27°26'41.64"E	55.60	0.00	0.00	0.00	40.00	10.00	10.00	0.00	160.00
	T3W4	26°22'15.66"S	27°26'34.74"E	93.90	0.00	0.10	0.10	80.00	40.00	80.00	30.00	80.00
	T3W5	26°22'24.06"S	27°26'35.28"E	48.70	0.00	0.00	0.00	30.00	10.00	0.00	0.00	120.00
	T3W6	26°22'28.20"S	27°26'39.84"E	94.10	5.10	0.10	0.10	30.00	40.00	60.00	20.00	210.00
	T3W7	26°22'29.22"S	27°26'41.04"E	43.20	7.10	0.00	0.00	120.00	30.00	40.00	10.00	120.00
	T3W8	26°22'31.38"S	27°26'43.08"E	100.10	0.00	0.10	0.10	40.00	30.00	40.00	20.00	100.00
	Average±Error 71.33 2.31 0.06 0.05 77.50 46.25 82.50 30.00 152.50											
Mine Tailings Four	T4W1	26°21'11.40"S	27°27'50.82"E	81.20	0.00	0.10	0.10	30.00	30.00	20.00	30.00	10.00
	T4W2	26°21'11.34"S	27°27'50.22"E	54.50	1.00	0.10	0.00	150.00	60.00	70.00	20.00	80.00
	T4W3	26°21'3.06"S	27°27'50.16"E	113.10	3.30	0.00	0.10	110.00	40.00	50.00	20.00	180.00
	T4W4	26°20'51.18"S	27°27'49.62"E	62.10	4.10	0.00	0.00	90.00	30.00	50.00	10.00	60.00
	T4W5	26°20'51.66"S	27°27'40.26"E	77.10	8.10	0.20	0.00	70.00	30.00	40.00	10.00	70.00
	T4W6	26°20'53.46"S	27°27'30.66"E	64.10	1.00	0.00	0.00	150.00	80.00	60.00	30.00	20.00
	T4W7	26°21'0.18"S	27°27'25.50"E	39.70	4.20	0.10	0.10	90.00	80.00	110.00	20.00	110.00
	T4W8	26°21'6.18"S	27°27'21.96"E	76.20	4.30	0.10	0.10	90.00	100.00	100.00	30.00	138.00
	T4W9	26°21'7.98"S	27°27'25.62"E	84.60	2.20	0.10	0.10	150.00	90.00	70.00	30.00	138.00
	T4W10	26°21'10.44"S	27°27'30.24"E	92.30	5.10	0.00	0.00	150.00	60.00	80.00	40.00	167.00
	T4W11	26°21'13.61"S	27°27'38.64"E	63.30	2.20	0.10	0.00	130.00	50.00	60.00	20.00	113.00
	T4W12	26°21'12.90"S	27°27'51.18"E	69.90	0.00	0.00	0.00	40.00	20.00	10.00	0.00	112.00
	Average±Error 73.18 2.96 0.07 0.04 104.17 55.83 60.00 21.67 99.83											

Mine Tailings Five	T5W1	26°23'52.20"S	27°24'41.82"E	120.00	2.60	0.10	0.10	0.10	110.00	50.00	40.00	10.00	100.00
	T5W2	26°23'45.36"S	27°24'39.96"E	60.00	8.10	0.00	0.10	0.00	90.00	30.00	20.00	10.00	210.00
	T5W3	26°23'42.48"S	27°24'46.92"E	60.00	5.10	0.00	0.00	0.00	100.00	30.00	30.00	10.00	190.00
	T5W4	26°23'47.82"S	27°24'48.60"E	46.00	0.10	0.10	0.10	0.10	130.00	40.00	50.00	20.00	70.00
	T5W5	26°23'44.40"S	27°24'54.72"E	70.00	2.70	0.10	0.00	0.00	110.00	30.00	50.00	20.00	40.00
	T5W6	26°23'43.56"S	27°25'4.86"E	60.00	2.10	0.00	0.00	0.00	70.00	30.00	30.00	10.00	10.00
	T5W7	26°23'52.38"S	27°25'0.66"E	78.00	6.10	0.10	0.10	0.10	110.00	40.00	40.00	10.00	130.00
	T5W8	26°23'58.62"S	27°25'1.98"E	50.00	5.10	0.10	0.00	0.00	80.00	30.00	20.00	10.00	110.00
	T5W9	26°23'58.68"S	27°25'3.60"E	80.00	0.00	0.00	0.00	0.00	100.00	30.00	30.00	10.00	210.00
	T5W10	26°23'57.96"S	27°25'5.76"E	68.00	5.80	0.10	0.10	0.10	110.00	150.00	160.00	100.00	230.00
	T5W11	26°23'57.00"S	27°25'7.08"E	53.00	2.00	0.10	0.10	0.10	100.00	90.00	80.00	40.00	110.00
	T5W12	26°23'53.40"S	27°25'9.24"E	60.00	0.00	0.00	0.00	0.00	60.00	20.00	30.00	10.00	100.00
				Average±Error 67.08 3.31 0.06 0.05 97.50 47.50 48.33 21.67 125.83									
Mine Rock Samples	RD91	26°21'43.86"S	27°27'50.82"E	42.00	6.30	0.10	0.20	0.10	160.00	50.00	30.00	20.00	0.00
	RD92	26°21'44.40"S	27°27'50.70"E	45.00	4.10	0.10	0.00	0.00	40.00	80.00	70.00	120.00	190.00
	RD101	26°24'3.96"S	27°25'56.76"E	91.00	1.00	0.10	0.10	0.10	60.00	160.00	350.00	10.00	90.00
	RD102	26°24'3.78"S	27°25'57.72"E	42.00	3.70	0.10	0.10	0.10	40.00	190.00	360.00	10.00	140.00
	RD103	26°24'4.26"S	27°25'56.82"E	62.00	6.20	0.00	0.00	0.00	180.00	260.00	490.00	30.00	150.00
	RD111	26°22'57.42"S	27°29'38.22"E	61.20	1.00	0.10	0.10	0.10	1710.00	20.00	80.00	70.00	210.00
	RD121	26°22'46.98"S	27°30'12.72"E	71.30	7.10	0.00	0.00	0.00	930.00	60.00	40.00	30.00	160.00
	RD122	26°22'49.20"S	27°30'14.04"E	83.30	2.10	0.10	0.10	0.10	1550.00	50.00	60.00	40.00	200.00
				Average±Error 62.23 3.94 0.08 0.08 583.75 108.75 185.00 41.25 142.50									
Mine West Village	SVW1	26°23'45.41"S	27°28'41.22"E	82.10	2.00	0.10	0.00	0.00	1100.00	30.00	70.00	20.00	10.00
	SVW2	26°23'45.90"S	27°28'43.26"E	91.30	1.20	0.10	0.10	0.10	150.00	20.00	30.00	20.00	60.00
	SVW3	26°23'44.10"S	27°29'2.04"E	43.30	0.00	0.10	0.00	0.00	210.00	30.00	30.00	20.00	40.00
	SVW4	26°23'38.88"S	27°29'5.16"E	64.40	2.10	0.20	0.10	0.10	1080.00	40.00	20.00	30.00	30.00
	SVW5	26°23'41.94"S	27°28'57.78"E	61.20	2.20	0.10	0.10	0.10	1360.00	50.00	40.00	40.00	120.00
	SVW6	26°23'43.26"S	27°28'52.50"E	48.70	2.00	0.00	0.00	0.00	1270.00	50.00	100.00	40.00	150.00
				Average±Error 65.17 1.58 0.10 0.05 861.67 36.67 48.33 28.33 68.33									
Mine East	SVE1	26°23'0.78"S	27°30'40.56"E	71.30	1.10	0.10	0.10	0.10	190.00	30.00	20.00	20.00	10.00
	SVE2	26°23'6.30"S	27°30'48.30"E	78.20	1.00	0.00	0.00	0.00	90.00	20.00	10.00	10.00	210.00

Village	SVE3	26°23'13.92"S	27°30'59.76"E	48.50	1.20	0.10	0.10	0.10	140.00	30.00	50.00	20.00	110.00
	SVE4	26°23'10.38"S	27°31'15.24"E	91.80	20.20	0.10	0.10	0.10	90.00	20.00	50.00	10.00	120.00
	SVE5	26°23'6.84"S	27°30'53.04"E	58.20	2.10	0.00	0.00	0.00	110.00	20.00	20.00	20.00	90.00
	SVE6	26°23'18.90"S	27°30'51.36"E	67.20	0.10	0.00	0.10	0.10	90.00	20.00	10.00	10.00	110.00
	SVE7	26°23'21.24"S	27°31'20.04"E	74.30	3.10	0.10	0.00	0.00	90.00	10.00	10.00	10.00	150.00
	SVE8	26°23'23.46"S	27°31'11.70"E	84.30	1.10	0.20	0.10	0.10	80.00	10.00	30.00	10.00	60.00
	SVE9	26°23'16.68"S	27°30'44.10"E	64.60	8.10	0.10	0.00	0.00	90.00	20.00	20.00	10.00	60.00
	SVE10	26°22'60.00"S	27°30'36.12"E	56.10	4.20	0.00	0.00	0.00	60.00	10.00	10.00	0.00	50.00
	SVE11	26°23'27.90"S	27°31'0.06"E	72.10	5.30	0.00	0.00	0.00	50.00	20.00	10.00	10.00	40.00
	Average±Error 69.69 4.32 0.06 0.05 98.18 19.09 21.82 11.82 91.82												

APPENDIX C: Gamma Spectroscopy Operation Procedure.

Prior to launching the Gamma Acquisition and Analysis (GAA) application, ensure that liquid nitrogen has been transferred to the dewar and that the detector is connected to the computer. This process usually takes about 6 hours, especially when the dewar was previously empty. After filling the dewar with liquid nitrogen, allow the detector to cool down for approximately one hour. When this is done, you are now free to switch on the MCA hardware located in the back of the DSA and on the left side in readiness for operation. Once this is switched ON, a green light on the detector will go ON to show that the detector has been cooled down.

1. Starting the Genie 2000 program

- On the Start menu go to **All programs | Genie 2000 | Gamma acquisition analysis.**

2. Opening the Detector

- Having opened the program go to **File | Open Datasource | Select detector** (e.g BE6530) and click **Open.**

3. Selecting the Appropriate Acquisition Time

This is used to define how long the acquisition period is going to be.

- Go to **MCA | Acquire set up | Live time** (enter 43200 sec for example) | **OK.**

4. Setting up the high voltage power supply (HVPS)

- Go to **MCA | Adjust | HVPS** | Type in 4500V | **OK | Exit**

5. Start Acquisition

- **Acquire | Start**

6. Saving the spectrum

After acquisition of the gamma spectrum, you can save the spectrum

- **File**| save in **CAMFILES** | **Save as** | **File name**| save as type (*.CNF).

7. Analyzing the spectrum

After opening a CAM file datasource, the desired calibration parameters can be loaded and saved with the spectral data.

This can be achieved as follows:

- Go to **Calibrate** | **Load** | **Select in CALFILES** | **Load** |

When the Load Calibration dialog box is first displayed, the boxes labelled Energy/Shape and Efficiency will contain a check mark, Leave these boxes checked.

Descriptive information is then entered to uniquely identify each acquired spectrum.

This is done as follows;

- Go to **Edit** | **Sample information** | Type in the desired entries in the Edit Sample Information dialog box | **LoadCal** | **OK** | .

After this, the analysis process can be done in the GAA window menu. This is done by going through the following execution sequence:

- Go to **Analyze** | **Peak Locate** | **Unidentified 2nd Diff**. The start channel must be 1 and the Stop channel must be 8192. The Significance Threshold must be 3 and the Tolerance must be 3 keV. Then click **Execute** button.
- Next Go to **Analyze** | **Peak Area** | **Sum/Non Linear LSQ Fit**. Check for the Start and Stop channel. They must be the same as above. After this, one has to click **Execute** button.
- Next Go to **Analyze** | **Interactive Peak Fit**. Make sure that the **STDLIB** is selected and then click **Execute** button. This allows one to edit the peak areas. If you are happy with it, you can just click the **Ok** button after the pop up window.
- Next Go to **Analyze**| **Area Correction** | **Std. Bkg Subtract**. When the new window pops up, check the **Yes** button and browse for the file you used for

your background radiation. NB. This must have been analysed prior to the subtraction. After this selection, one has to click **Execute button**.

- E. Next Go to **Analyze | Efficiency Correction | Standard**. After this, one has to click **Execute button**.
- F. Next Go to **Analyze | Reporting |Standard**. In this menu, one has to select the activity units to be used in the report. (E.g Bq). After this, one has to click **Execute button**
- G. Next Go to **Analyze | Nuclide Identification |NID**. In this window, you must verify for the Start and Stop channel as well as the tolerance. The **Generate Report Button** must be checked for the report to be display in the report window. After this, one has to click **Execute button**.

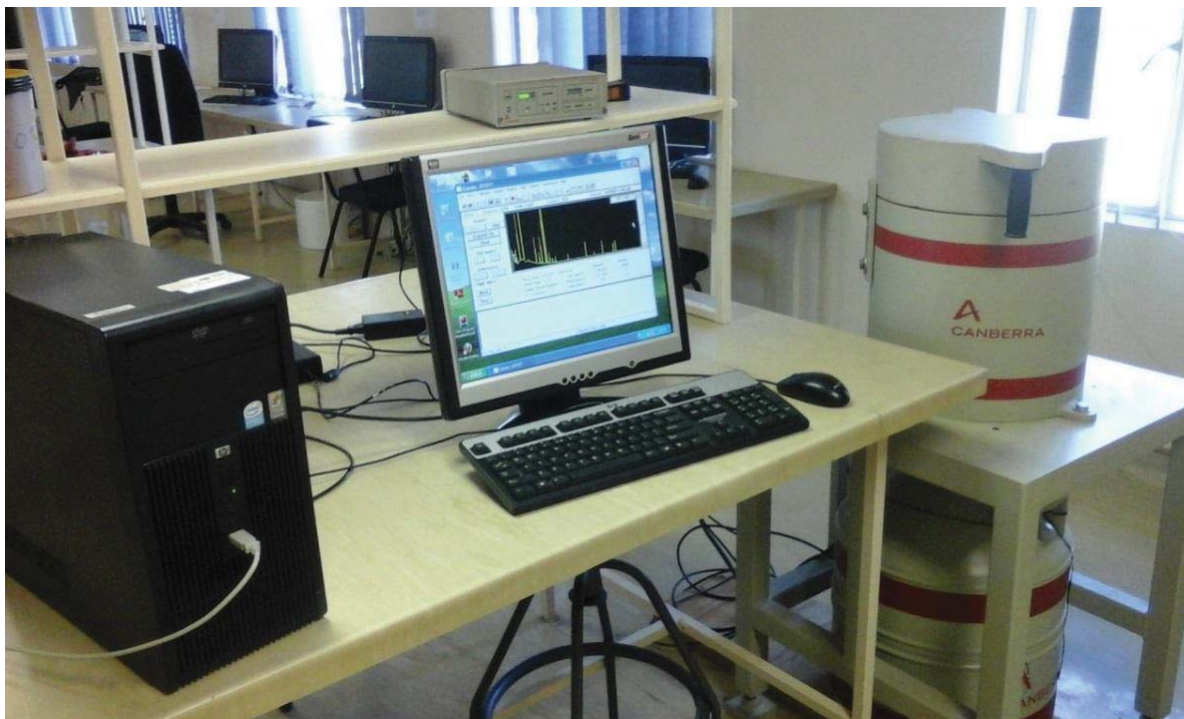
It must also be noted that the sequence of analyses can be changed to suit the requirements of the analyzer. The spectra analyzed and reports can then be saved as pdf files for further use.

8. Exiting the GAA Application

When all spectral data analyses have been completed, click on **File |Close** to exit from the GAA application.

APPENDIX D: Instrumentation for Data Analysis in Pictures.

a) Broad Energy Germanium (BEGe) Gamma Spectrometry System



b) Inductively Coupled Plasma Mass Spectrometry System



c) ALPhaGuard Professional Radon Monitor



APPENDIX E: List of Publications and Presentations.

Peer Reviewed Publications from this Work.

1. **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: [10.3390/ijerph13010138](https://doi.org/10.3390/ijerph13010138).
2. Mathuthu, M., **Kamunda, C.**, and Madhuku, M. **(2016)**. Modeling of Radiological Health Risks from Gold Mine Tailings in Wonderfonteinspruit Catchment Area, South Africa. Int. J. Environ. Res. Public Health, 13, 570; doi: [10.3390/ijerph13060570](https://doi.org/10.3390/ijerph13060570).
3. **Kamunda, C.**, Mathuthu, M., and Madhuku, M. **(2016)**. Health Risk Assessment of Heavy Metals in Soils from Witwatersrand Gold Mining Basin, South Africa. Int. J. Environ. Res. Public Health, 13, 663; doi: [10.3390/ijerph13070663](https://doi.org/10.3390/ijerph13070663).

Conference Presentations

1. **Kamunda, C.**, Mathuthu, M., and Madhuku, M. **(2016)**. Assessment of Radiological Hazards from Gold Mine Tailings in Gauteng Province, South Africa. 14th International Congress of the International Radiation Protection Association (IRPA), Cape town International Convention Centre, South Africa; 9th – 13th May 2016.
2. **Kamunda, C.**, Mathuthu, M., and Madhuku, M. **(2016)**. Modelling of Radiological Health Risks from Gold Mine Tailings in Gauteng Province, South Africa. The 61st Annual Conference of the SA Institute of Physics (SAIP2016), University of Cape Town, South Africa; 4th – 8th July 2016.

APPENDIX F: Equations for Weighted Average and Error Propagation Calculation.

Weighted Average

Given functions $b \pm \delta b$, $c \pm \delta c$ and $d \pm \delta d$, the weighted average W_m in these is:

$$W_m = \frac{\frac{b}{\delta b^2} + \frac{c}{\delta c^2} + \frac{d}{\delta d^2}}{\frac{1}{\delta b^2} + \frac{1}{\delta c^2} + \frac{1}{\delta d^2}}$$

The corresponding propagated uncertainty in this weighted average will be:

$$\Delta W_m = \frac{1}{\sqrt{\frac{1}{\delta b^2} + \frac{1}{\delta c^2} + \frac{1}{\delta d^2}}}$$

Error propagation

The simplified formulas for propagation of errors are summarized below.

Function ¹	Manipulation	Uncertainty ²
$y = a_1 x_1 - a_2 x_2$ or $y = a_1 x_1 + a_2 x_2$	Addition/subtraction	$s_y = \sqrt{a_1^2 s_{x_1}^2 + a_2^2 s_{x_2}^2}$
$y = a_1 x_1 x_2$ or $y = a_1 \left(\frac{x_1}{x_2} \right)$	Multiplication/Division	$\frac{s_y}{y} = \sqrt{\left(\frac{s_{x_1}}{x_1} \right)^2 + \left(\frac{s_{x_2}}{x_2} \right)^2}$
$y = x^m$	Power of x	$\frac{s_y}{y} = \frac{m s_x}{x}$
$y = e^{-ax}$	Power of e	$\frac{s_y}{y} = a s_x$

¹ a , a_1 , a_2 and m represent constants, x , x_1 and x_2 are independent variables and y is the dependent variable.

² s_j is the estimated standard deviation in variable x_j .