

Evaluation of primordial radionuclides and health impacts in soil, groundwater and food crops from the lower Swakop river, Namibia

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Declaration

I, Gideon Amakali (Student number: 31388582), hereby declare that this thesis represents my own research work carried out at the North-West University, Mafikeng Campus, South Africa, during the period January 2018 to December 2020. The data presented is original, except where indicated as such by means of reference. The work has not previously submitted to any other institution or awarding body for any degree.

Signed:



Gideon Amakali

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Abstract

Primordial radionuclides; ^{238}U , ^{232}Th and ^{40}K are found ubiquitously in varying concentrations in environmental media. The exploration and mining related activities have the potential to redistribute and concentrate primordial radionuclides to locations accessible to members of the public. These radionuclides are the significant contributor to the radiation doses received by humankind through various exposure pathways. The Swakop river community and environmental activists expressed concern over possible contamination of alluvial sand used in the construction industry and agricultural croplands in the lower Swakop river due to uranium mining related activities in the surrounding areas. This has motivated for the study to evaluate the primordial radionuclides in groundwater, soil and food crops in the lower Swakop river and possible associated health impacts. The study has also considered the portion of the Khan river that traversing between the uranium mining activities before confluent with the Swakop river. A total of 63 soil samples were subjected to high purity germanium (HPGe) detector gamma spectroscopic technique for the identification and quantification of primordial radionuclides present. Inductive Coupled Plasma Mass Spectrometry (ICP-MS) technique was used for the identification and quantification of total elements of U, Th and K in water and food crops samples. The average activity concentrations of ^{238}U in soil samples across the study area were found to vary from 69.19 ± 4.48 to 532.09 ± 11.90 Bq.kg⁻¹. The average activity concentrations of ^{232}Th in soil samples varied from 59.07 ± 1.11 to 211.56 ± 1.85 Bq.kg⁻¹, while for ^{40}K were from 568.73 ± 12.66 to 982.74 ± 24.56 Bq.kg⁻¹. The activity concentrations of ^{40}K and ^{238}U in groundwater along the Swakop river were varied from 737.66 to 2475.98 mBq.l⁻¹ and 0.161 to 1116.47 mBq.l⁻¹ with average values of 1449.22 ± 181.85 and 313.43 ± 127.62 mBq.l⁻¹, respectively, while ^{232}Th was below the detection limit of the technique used. The average activity concentrations for ^{238}U , ^{232}Th and ^{40}K in the groundwater samples collected from the Khan river were 1282.55 ± 230.04 mBq.l⁻¹, 0.167 ± 0.09 mBq.l⁻¹ and 1505.02 ± 153.16 mBq.l⁻¹, respectively. The minimum and maximum average activity concentration of 567.79 and 1603.56 ± 134.36 Bq.kg⁻¹ for ^{40}K were observed in spinach and beetroot, respectively. The minimum and maximum average activity concentrations of ^{238}U were 0.15 Bq.kg⁻¹ and 1.01 Bq.kg⁻¹ observed in eggplant and spinach, respectively, while for ^{232}Th were 0.09 Bq.kg⁻¹ and 0.84 Bq.kg⁻¹ observed in eggplant and spinach, respectively. The average radioactivity concentration ratios or transfer factor (TF) for ^{40}K in food crops to that in soil

were 2.77, 1.76 and 0.98 for spinach, eggplant and beetroot, respectively. The TF values for both ^{238}U and ^{232}Th were 0.014, 0.0035, 0.004 and 0.02, 0.003, 0.01 for spinach, eggplant and beetroot, respectively, relatively lower than that observed for ^{40}K .

The estimated average absorbed gamma ray dose rates at 1 meter above the ground for the study areas A and B, were $88.65 \pm 0.47 \text{ nGy.h}^{-1}$ and $100.94 \pm 0.44 \text{ nGy.h}^{-1}$, respectively, relatively higher than the global average of 60 nGy.h^{-1} . The average absorbed gamma dose rates at 1 meter above the ground for the study areas C, D and E were 135.85 ± 0.75 , 378.73 ± 1.28 and $258.17 \pm 1.37 \text{ nGy.h}^{-1}$, respectively, relatively higher than those observed in areas A and B. The maximum annual absorbed dose at 1 meter above the ground was estimated to be 6.30 mGy, based on the maximum dose rate of $717.97 \text{ nGy.h}^{-1}$ observed at the tailings storage facility. The average annual effective doses due to external exposure for the study areas A and B were 0.69 ± 0.004 and $0.80 \pm 0.002 \text{ mSv.y}^{-1}$ for the age category representative of infants, relatively higher than that of the age category representative of children, which were 0.61 ± 0.003 and $0.71 \pm 0.002 \text{ mSv.y}^{-1}$, respectively. The age category representative of adults receives the least annual doses with average values of 0.54 ± 0.002 and $0.62 \pm 0.002 \text{ mSv.y}^{-1}$, estimated for study areas A and B, respectively. The average effective dose due to external exposure for study areas A and B were greater than 0.48 mSv.y^{-1} , the global average reported by UNSCEAR. The average annual effective dose estimated for the age group representative of an adult for study areas C, D and E were 0.84 ± 0.005 , 2.32 ± 0.01 and $1.58 \pm 0.01 \text{ mSv.y}^{-1}$, respectively, significantly less than the prescribed annual limit value of 20 mSv prescribed for designated radiation workers. The annual committed effective doses by the consumption of all food crops by representative of the age categories 5 years, 15 years and adults, were estimated at 1.95 ± 0.022 , 1.32 ± 0.014 and $1.55 \pm 0.020 \text{ mSv}$, respectively. The estimated total annual effective doses for the age categories 5 years, 15 years and adults for the study area A were 2.69 ± 0.04 , 1.98 ± 0.03 and $2.14 \pm 0.04 \text{ mSv}$, respectively. The R_{eq} values for the study area A were varied from 97.37 ± 6.38 to $313.07 \pm 5.50 \text{ Bq.kg}^{-1}$ with an average value of $190.81 \pm 1.02 \text{ Bq.kg}^{-1}$. The R_{eq} values for the study area B were found to vary from 95.58 ± 4.83 to 420.44 ± 3.17 with an average value of $217.80 \pm 0.95 \text{ Bq.kg}^{-1}$. The R_{eq} for the study area C were varied from 175.42 ± 9.11 to $394.05 \pm 4.26 \text{ Bq.kg}^{-1}$ with an average value of $294.22 \pm 1.63 \text{ Bq.kg}^{-1}$. All R_{eq} values for individual samples from the study area D were in exceedance of the permissible value of 370 Bq.kg^{-1} , varied from 559.82 ± 5.44 to $1599.45 \pm 45 \text{ Bq.kg}^{-1}$ with an average value of 825.38 ± 2.78 .

Bq.kg⁻¹. The Ra_{eq} for the study area E varied from 249.78±5.97 to 1230.21±10.29 Bq.kg⁻¹ with an average value of 576.21±3.00 Bq.kg⁻¹. The predicted total excess cancer risks, due to external exposure, for the croplands and tailing's storage facility for the duration of about 1000 years were found to range from 3.8x10⁻⁴ to 4.32x10⁻⁴ and from 3.38x10⁻³ to 4.937x10⁻³, respectively.

Key Words

Primordial radionuclides, activity concentration, transfer factor, absorbed dose, effective dose, cancer morbidity risk, radium equivalent activity, NORMALYSA, Resrad

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List of Abbreviations

BEIR:	Committee on the Biological Effects of Ionising Radiation
CARST:	Centre for Applied Radiation Science and Technology
CSIR:	The Council for Scientific and Industrial Research
DNA:	Deoxyribonucleic acid
EPA:	Environmental Protection Agency of the United States of America
GPS:	Geographical positioning system
IAEA:	International Atomic Energy Agency
ICP- MS:	Inductively coupled plasma mass spectrometry
ICRP:	International Commission on Radiation Protection
ISO:	International Organisation for Standardisation
LHU:	Langer Heinrich Uranium
MET:	Ministry of Environment and Tourism
MHSS:	Ministry of Health and Social Services
MME:	Ministry of Mines and Energy
NRC:	National Research Council of the United States of America
NWU:	North-West University
RUL:	Rössing Uranium Limited
RUM:	Rössing Uranium Mine
UNSCEAR:	United Nations Scientific Committee on the Effects of Atomic Radiation
WHO:	World Health Organisation

Chapter 1: Introduction and problem statement

1.1 Terrestrial primordial radionuclides

Life on earth is inadvertently exposed to natural sources of ionizing radiation from the high-energy cosmic ray particles and terrestrial primordial radionuclides. The terrestrial primordial radionuclides are naturally occurring radionuclides that have been in existence in the earth crust, from the creation of the earth about 4.5 billion years ago (UNSCEAR, 2010). The inventory of primordial radionuclides includes ^{238}U , ^{237}Np , ^{235}U , ^{232}Th , ^{87}Rb , ^{138}La , ^{147}Sm , ^{176}Lu and ^{40}K . Presently, only the ^{238}U , ^{232}Th and ^{40}K occur in measurable quantities in environmental media such as soil, igneous and sedimentary rocks, water, foodstuff, etc. due to their respective long half-lives comparable to the age of the earth (Friedlander *et al.*, 1981; International Atomic Energy Agency [IAEA], 2003). The ^{238}U and ^{232}Th and their decay products together with ^{40}K emit either the alpha and/or beta particles, which may be accompanied by gamma ray's radiation. These radionuclides are the significant contributor to the earth's background radiation and to radiation doses received by humankind through various exposure pathways (Aswood *et al.*, 2013). Other primordial radionuclides exist in nature at extremely low levels that their contributions to the dose in humans are insignificant (UNSCEAR, 2000). Radiation has the potential to impair the functioning of tissues, thus causes adverse health effects.

Natural processes such as weathering of earth crust, igneous and sedimentary rocks release primordial radionuclides into the environment. Some activities such as mining and processing of mineral rich in primordial radionuclides, storage of tailings and combustion of coal redistributes primordial radionuclides in the environmental compartments such soil, water and air (IAEA, 2003; UNSCEAR, 1982).

Some parts of Namibia, specifically the Erongo region has noticeable relatively high level of ambient gamma dose rates, with an estimated average annual radiation dose of 7.30 mSv, presumably associated with thorium, uranium and potassium bearing minerals in the soil (Ministry of Mines and Energy [MME], 2010). The region is nicknamed "Uranium province" and hosts two active open pit uranium mines, namely the Rössing Uranium (RUM), and Swakop Uranium (SU), while Areva – Trekkopje Mine and Langer Heinrich (LHU) are under care and maintenance. There are

numerous prospecting and exploration activities for mineral deposits scattered throughout the central Erongo region. Most of the activities are taking place in proximity of the lower Swakop river, an ephemeral river that usually is dry and flows only during the heavy falls received in the high ground inland. The community of the Swakop river comprise of all age group category from infants to adults and grows food crops on the bank of the river for own consumptions and local market.

1.2 Problem statement

Namibia experienced an unprecedented rush for uranium exploration and development projects after it attained its independence in 1990 (MME, 2010). Most of these activities were concentrated in the catchment areas and basins of the Khan and Swakop rivers, the ephemeral rivers that run through the Namib desert to the Atlantic Ocean. The exploration, mining, minerals extraction and processing activities generate radioactive dust, radioactive waste rocks and fine sand of milling residues, tailings. The tailings contain some quantities of uranium residuals, other naturally occurring radionuclides and their decay products as well as chemicals used in the extraction and recovery processes (Agency for Toxic Substances & Disease Registry of the Public Health Service of the United States [ATSDR], 2019; NRC, 2012). The tailings and other radioactive waste products from mining and exploration activities are disposed of in geomorphologic depression sites such as valleys in the vicinity of the respective sites or mines. Some of the disposal sites have not been bottom lined to prevent leachates from infiltrating the groundwater table.

The communities in the surroundings of the Swakop river grow agricultural food crops, such as asparagus, olives, peppers, tomatoes, lettuce, spinach, beetroots and cucumbers, along the riverbank for own consumption and local market. The community uses groundwater extracted from the river aquifers for irrigation (de Villiers & de Beer, 2011). Some areas with proven quantities of uranium ore deposits have been identified along the lower Swakop river. The Swakop river community expressed concern on the improper disposal and management of radioactive waste at the mines, particularly the disposal of the waste rock materials, pulps, sludge and tailings. There have been speculation and suspicion that mining activities in the area contaminates the surrounding environment and agricultural crop fields on the bank of the river. The location of uranium mines, prospects sites and crop fields are illustrated in Figure 1.1.



Figure 1. 1: The location of uranium mine, prospects sites in relation to the crop fields in the Swakop and Khan rivers (Marenica Energy, 2020).

There has been concern for potential seepage of contaminants, including radionuclides from the tailing storage facilities into the surface water flow and groundwater flow in fractured rocks, reaching the nearby ephemeral Khan and Swakop rivers (Barnard, 2005; Kohrs, 2008). From the radiation protection point of view, an interest would be on the investigation of activity concentration of primordial radionuclides in soil and agricultural crops for the determination of potential effective doses to members of the public.

International and local environmental activists launched campaigns petitioning against the alluvial sand excavation as well as the exploration and mining activities in the Swakop river basin (Kohrs, 2008). Chareyron and Bourbon (2014) evaluated the activity concentration ratio of ^{226}Ra to ^{238}U for soil samples taken from the Khan river about 2 km from RUM's tailings facilities and the ratio lies between 2.3 to 5. The ratio suggests the disruption in secular equilibrium between the ^{238}U and its daughter ^{226}Ra . The disequilibrium observed could possibly be attributable to the contamination of the surface soil with processed material, specifically tailings material where ^{238}U has been extracted from the ore hence the ^{238}U residual activity in the waste is lower than that of ^{226}Ra activity.

The alluvial sand excavated from the Swakop river form the main ingredients for building materials. There are no documented studies on the level of primordial radionuclides in the alluvial sand of the lower Swakop river, hence on the building material derivatives. Judging from the estimated average effective dose for the Erongo region at 7.3 mSv.a^{-1} , an investigation on the potential exposure to radiation from the

sand used for building is justified. Enhanced levels of primordial radionuclides and their respective progenies have the potential to increase the radiation exposure level to individuals occupying dwellings or offices built from the Swakop river derived alluvial sand.

Chareyron and Bourbon (2014) expressed concern on irrigation with water high in uranium contents that might have the potential for causing radionuclides build up overtime within irrigated soil and this may increase the radionuclide uptake from soil to edible parts of crops. It is assumed that the radionuclide concentration in plants or plant parts is linearly related to its concentration in soil within the rooting zone (Shayeb *et al.*, 2017).

In recent times, the radiation protection organisations expressed concern on the possibility of long-term effects from prolonged exposure to low levels of radiation, particularly from naturally occurring radionuclides (IAEA, 2014a). It is the responsibility of any government to ensure that situations of exposure to natural background radiation are identified and evaluated so that effective protective actions are designed for optimised protection and safety (IAEA, 2018).

Although there has not been observable evidence of immediate health effects from exposure to low level of radiation (an annual dose of less than 100 mSv) encountered in the environment, the linear no threshold (LNT) hypothesis assumes that the harmful effects of radiation are linearly related to the dose received and there is no safe dose threshold value. The LNT implies that any dose however small, will increase the risk of excess cancer and/or heritable disease in a simple proportionate manner (ICRP, 2007; Hall & Giaccia, 2012).

The prospecting, exploration and uranium mining related activities within the catchment and basin of the Swakop river have the potential to redistribute or concentrate the radionuclides and lead to local accumulation and highly polluted areas accessible by members of the public. There has not been any study undertaken to characterize the activity concentrations of naturally occurring radionuclides in the agricultural land and agricultural products on the lower Swakop river. It is, therefore, imperative to evaluate the levels of activity concentration of the primordial radionuclides in the lower Swakop river's environmental media and assess the

potential effective dose to members of the public. The results would inform the regulatory bodies on the nature and extent of radiological risks and allow for appropriate intervention, where and if necessary.

1.3 Research aim and objectives

The aim of the study was to evaluate the level of primordial radionuclides and potential health impacts to members of the public due to the use of groundwater, soil and food crops in the lower Swakop river.

The objectives of this study were to:

- determine the activity concentrations of ^{40}K , ^{238}U and ^{232}Th in soil, groundwater and agricultural crops from the study area,
- evaluate the activity concentration ratios (transfer factors) for agricultural food crops and soil from the cropland in the study area,
- investigate the potential radiological hazards associated with soil and food crops in the study area
- estimate the time dependent residual activity concentrations of ^{40}K , ^{238}U and ^{232}Th in the root's zone and the resultant effective dose due to ingestion of food crops using the NORM And LegacY Site Assessment (NORMALYSA) mathematical model
- estimate the cancer morbidity risk for a worker at the tailings storage facility due to external exposure using the RESRAD-ON SITE (RESidual RADioactivity) computer code

Chapter 2: Literature review

2.1 Some of the previous studies conducted elsewhere relevant to the current study

Numerous studies conducted worldwide on the distribution of uranium, thorium and potassium in environmental media culminated in the compilation of a wide variation of activity concentrations in soil, water and agricultural crops. The concentration of a primordial radionuclide in soil is dependent on host rocks from which the soil was derived as well as other geological factors (Nguelem *et al.*, 2016). The revised global weighted average activity concentrations of ^{226}Ra , ^{238}U , ^{232}Th and ^{40}K in soil are 32, 33, 45 and 420 Bq.kg^{-1} with median values range from 17 to 60, 16 to 110, 11 to 64 and 140 to 850 Bq.kg^{-1} , respectively (UNSCEAR, 2000).

There are some areas of the world characterised by high absorbed dose rates in air due to the presence of naturally rich thorium-bearing and uranium-bearing minerals in the soil. These areas have been classified as high background radiation areas (UNSCEAR, 2010). An example is a region in Kerala, India where above global average activity concentration in the ranges of up to 3081 Bq.kg^{-1} , 11976 Bq.kg^{-1} and 216 Bq.kg^{-1} for ^{238}U , ^{232}Th and ^{40}K , respectively, have been recorded (Christa *et al.*, 2012). An investigation on the exposure to primordial radionuclides in Kerala, India by Nair *et al.*, (2009) resulted in the derived annual effective dose of over 70 mSv, while in some areas of Ramsar, Iran, an annual effective dose of up to 260 mSv has been recorded (Ghiassi-Nejad *et al.*, 2002).

A study by Kringel *et al.*, (2010) on groundwater samples collected along the length of the Khan and Swakop rivers found the uranium concentration ranges between 2 $\mu\text{g.l}^{-1}$ and 528 $\mu\text{g.l}^{-1}$ in the alluvial groundwater, with a mean of 39 $\mu\text{g.l}^{-1}$. Chareyron & Bourbon (2014) observed that the uranium content in ground water taken from the Khan river upstream and downstream of the RUM were 0.2 $\mu\text{g.l}^{-1}$ and 430 $\mu\text{g.l}^{-1}$, respectively, raising suspicion about the origin of the downstream uranium, suggesting the potential influence by mining activities. The average uranium concentration detected in the two rivers exceeded the World Health Organization (WHO) set provisional guideline value for drinking-water quality of 30 $\mu\text{g.l}^{-1}$ for chemical toxicity (WHO, 2011). Further, WHO (2001) has recommended for the general public ingestion

of soluble and insoluble uranium compounds not to exceed the tolerable daily intake (TDI) of $0.5 \mu\text{g.kg}^{-1}$ and $5 \mu\text{g.kg}^{-1}$ of body weight, respectively. The groundwater in the Khan and Swakop rivers are brackish and unsuitable for human consumption, and solely used by the community for irrigation of food crops (CSIR, 1997). Despite the water being unsuitable for human consumption, the public could be exposed to radiation through secondary transfer via the ingestion of food crops grown on contaminated land and irrigated with water high in uranium content (de Villiers & de Beer, 2011). It is documented that long term exposure to the ^{238}U and ^{232}Th decay series nuclides causes chronic lung diseases, acute leucopenia, anaemia, bone and, kidney cancers (Choppin *et al.*, 2002; NRC, 1990; Ramasamy *et al.*, 2009).

2.2 Radioactive decay

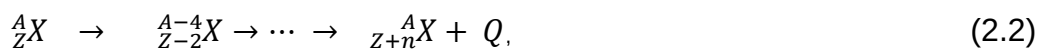
A radioactive atom (radionuclide) has an unstable configuration of protons and neutrons in the nucleus either as a result of excess neutrons or protons. In quest for nuclear stability, a radioactive atom undergoes a random, spontaneous disintegration or transformation to form new elements, a process called nuclear radioactive decay.

A parent radionuclide may be transformed directly into a stable nuclide (daughter product), through a single decay, however, there are instances where a daughter nuclide and the subsequent granddaughters are also radioactive atoms, therefore a series of decays takes place, termed radioactive decay chain, before reaching a stable nuclide. The single and serial radioactive decays can be expressed by equations 2.1 and 2.2, respectively.

Single decay



Serial decay chain



where, A_ZX , ${}^A_{Z+n}X$, ${}^{A-4}_{Z-2}X$ and Q denote the parent nuclide, stable daughter product, radioactive daughter product and decay energy, respectively (Cember & Johnson, 2009). The decay energy represents the total energy release in the decay process, and it is equivalent to the difference in mass between the parent nucleus, product

nucleus and particle (Khan & Gibbons, 2014). ^{238}U , ^{232}Th and ^{235}U are some of the naturally occurring radionuclides that undergo the serial decay process, while ^{40}K undergoes a single decay to a stable daughter product (Magill & Galy, 2005).

The mode of decay, the energy emitted during nuclear transformation and average lifetime of the nucleus before the decay are radionuclide specific, hence serve as useful tool in radionuclide identification. Radiation energy is expressed in electron volts (eV), ($1 \text{ eV} = 1.6 \times 10^{-9} \text{ J}$) (Cherry et al., 2012).

Radioactive decay is a natural, spontaneous process and is independent of any physical or chemical characteristics of an atom but dependent on the neutron to proton ratio and on the mass-energy relationship of the parent, daughter, and emitted particle(s) (Magill & Galy, 2005). The rate at which the number of atoms disintegrating, dN in a given time, dt , is proportional to the number of radioactive atoms, N present, and is mathematically expressed by equation 2.3,

$$\frac{dN}{dt} = -\lambda N \quad (2.3)$$

where λ , with a unit s^{-1} is a proportionality decay constant that is radionuclide specific, represents the probability per unit time for one atom to decay. The minus sign indicates reduction in the number of radioactive nuclei with time (Khan & Gibbons, 2014). Integrating equation 2.3 with respect to time leads to the exponential law of radioactive decay that can be expressed by equation 2.4,

$$N_t = N_0 e^{-\lambda t} \quad (2.4)$$

where, N_0 and N_t denote the number of atoms initially present at $t = 0$, and at any time $t = t$, respectively (Magill & Galy, 2005).

In the radioactive decay process, the number of radioactive atoms disintegrates within a prescribed time and is called the activity (A), measured in Becquerel (Bq). 1 Bq is therefore, equivalent to 1 atom that disintegrates per second (dps) (Cember & Johnson, 2009).

Activity can thus be expressed mathematically by equation 2.5 as,

$$A_t = \lambda N_t \quad (2.5)$$

Since $N_t = N_0 e^{-\lambda t}$,

$$\text{then } A_t = \lambda N_0 e^{-\lambda t} \quad (2.6)$$

Equation 2.5 shows that the activity A_t of a radioactive material at any given time, t , is directly proportional to the number of radionuclide atoms N_t present in the material.

Equation 2.5 then becomes,

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

where, A_0 is the initial quantity of activity and e is the base of the system of natural logarithms (Cember & Johnson, 2009).

It is convenient to express the rate of decay of a radioactive substance in terms of the half-life ($t_{1/2}$), the time it takes for either the activity or the number of radioactive atoms to decay to half the initial value. The relationship between the decay rate constant and half-life is expressed as equation 2.8,

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

where, $\ln(2)$ is the natural logarithm of 2 having an approximate value of 0.693. The activity concentration defines the relationship between the activity of a radionuclide and mass of the radioactive material, which is the number of Becquerel per unit mass or volume, Bq.kg^{-1} or Bq.l^{-1} (Cember & Johnson, 2009).

The radioactive decay equations 2.3 – 2.7 are however only applicable to a situation where a parent radionuclide decay directly into a stable atom and ignored the formation and decay of the subsequent radioactive daughters. For a radioactive decay chain, the relationship between the parent radionuclide 1 with a decay constant λ_1 and the formation of the radioactive daughter nuclide 2 with a decay constant λ_2 can be illustrated by equation 2.9,



The decay rate of the parent nuclide is $\lambda_1 N_1$ as given in equation 2.3. Therefore, the rate of growth of the radioactive daughter 2 is dependent on the rate of its production

from the decay of the parent nuclide and the rate of its own decay to nuclide 3, and can be mathematically expressed by equation 2.10,

$$\frac{dN_2}{dt} = \lambda_1 N_1 - \lambda_2 N_2 \quad (2.10)$$

Substituting equation 2.4 into equation 2.10 and rearrange yields equation 2.11,

$$dN_2 + \lambda_2 N_2 dt = \lambda_1 N_0 e^{-\lambda_1 t} dt \quad (2.11)$$

where N_0 denotes the initial number of atoms of the parent radionuclide at $t = 0$. By multiplying equation 2.11 with a factor $e^{\lambda_2 t}$ and integrating from $t = 0$ to $t = t$ yields equation 2.12, the general expression for the number of N_2 due to the growth from decay of the parent nuclide,

$$N_2(t) = N_0 \frac{\lambda_1}{\lambda_2 - \lambda_1} (e^{-\lambda_1 t} - e^{-\lambda_2 t}) \quad (2.12)$$

With reference to equation 2.5 the activity of the daughter radionuclide 2 is given by equation 2.13,

$$A_2 = \lambda_2 N_2 \quad (2.13)$$

Substituting equation 2.12 into the equation 2.13, yields the general expression 2.14, for the activity of the daughter radionuclide at time (t) due to the growth from the decay of the parent radionuclide,

$$A_2(t) = N_0 \frac{\lambda_1 \lambda_2}{\lambda_2 - \lambda_1} (e^{-\lambda_1 t} - e^{-\lambda_2 t}) \quad (2.14)$$

Equations 2.13 and 2.14 can then be generalised to cater for the determination of the activity of a daughter radionuclide n in a decay series of n chains, therefore expressed by equation 2.15 (Cetnar, 2006),

$$A_n = \lambda_n N_n = N_0 \sum_{i=1}^n c_i e^{-\lambda_i t} \quad (2.15)$$

$$\text{where } c_i = \frac{\lambda_1 \lambda_2 \dots \lambda_{n-1}}{(\lambda_1 - \lambda_i)(\lambda_2 - \lambda_i) \dots (\lambda_{n-1} - \lambda_i)}$$

With reference to the Table 2.2, the decay series for ^{238}U comprises of fourteen radionuclides with half-lives ranges from fractions of a microsecond to billions of years. The ^{238}U decay series can be divided into subseries headed by radionuclides with long half-lives, such as ^{238}U , ^{234}U , ^{230}Th , ^{226}Ra and ^{210}Pb . It is worth noting that the decay rate of the daughter nuclide is governed by the half-life of the parent and that radionuclides with shorter half-lives, decays rapidly than those with longer half-lives.

In a closed system, there will be a certain time, whereby a radioactive equilibrium will be reached, a condition where the decay and formation rates of the daughter radionuclides become equal, that is, there a fixed ratio between the activity of the parent and the daughters (Khan & Gibbons, 2014).

The radioactive equilibrium of importance to the study of ^{238}U and ^{232}Th decay series is termed secular equilibrium, a state in which the activities of all the daughter radionuclides in the decay series are almost equal with that of the parent (Cember & Johnson, 2009). Secular equilibrium can only be guaranteed in a radioactive decay chain when the half-lives of daughter radionuclides are much shorter than that of the parent, for example ^{238}U ($t_{1/2} = 4.5 \times 10^9$ years) and its daughter product ^{234}Th ($t_{1/2} = 24$ days). According to ISO (2007b), it takes about seven half-lives for the activity of the daughter to build-up reaching that of the parent.

The concept of secular equilibrium is useful in the application of gamma spectroscopic study of ^{238}U and ^{232}Th decay series. As ^{238}U and ^{232}Th do not produce any gamma energy with appreciable intensities, they cannot be measured directly by gamma spectrometry. The spectral peak interference of ^{235}U on ^{226}Ra at about 186 keV makes direct measurement by gamma spectrometry technique unreliable. Thus, the activities of ^{238}U and ^{232}Th have been generally inferred through the measurements of daughter radionuclides, with assumption that the secular equilibrium grow-up conditions, seven half-lives principle, have been complied with. The delays caused by seven half-lives hold up period is, however, considered a drawback of the approach.

2.3 Processes of radioactive decay

There are two major radioactive decay processes observed in primordial radionuclides and their respective daughters, namely, alpha decay and beta decay, which involve the emission of alpha (α) particles and beta (β) particles, respectively, each of which may or may not be accompanied by the emission of a high frequency electromagnetic gamma radiation (γ). The exact mode of nuclear transformation is dependent on the particular type of nuclear instability, whether the neutron-to-proton ratio is too high or too low for the particular nuclide under consideration and on the mass–energy relationship among the parent nucleus, daughter nucleus and emitted particle. The energy associated with radioactive decay modes varies in its penetrating power into matter (Martin, 2006).

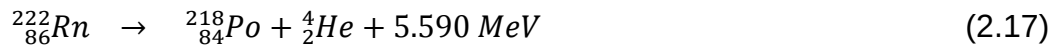
2.3.1 Alpha decay

An alpha decay is characterised by the emission of a positively charged alpha particle (α – particle), an equivalent to the nucleus of helium (${}^4_2\text{He}$). Alpha decay is more prevalent in some proton rich, heavy unstable nuclei with atomic number, $Z > 82$, some of which are naturally occurring such as ${}^{238}\text{U}$, ${}^{232}\text{Th}$, ${}^{226}\text{Ra}$ and ${}^{222}\text{Rn}$ (L'Annunziata, 2012).

In the alpha decay process, the parent radionuclide transforms into a daughter nuclide that contains two protons and two neutrons less than the parent, (the atomic mass number is reduced by 4 and atomic number reduced by 2) (Magill & Galy, 2005). An alpha decay process is illustrated in equation 2.16,



where, ${}^{A-4}_{Z-2}X$ and ${}^4_2\text{He}$ denote the daughter product and helium nucleus (alpha particle), respectively. A typical example for ${}^{222}\text{Rn}$ decay by the emission of alpha particle is illustrated by equation 2.17,



The radionuclides of natural origin emit alpha particles with a range of discrete energies, mostly lying between 4 and 8 MeV (Friedlander *et al.*, 1981).

2.3.2 Beta decay mode

In the beta decay mode, a beta particle (β) is emitted. A beta particle is a charged particle that is indistinguishable from an ordinary electron. The beta decay mode is pronounced in nuclides with excess of neutrons (neutron rich) relative to protons, or protons relative to neutron (proton rich). The proton rich radionuclides may either decay by emission of a positron (β^+) or by the atomic electron capture ($\mathcal{E}$). During a β^+ decay process, a proton converts into neutron, positron and neutrino, with subsequent emission of a β^+ and neutrino from the nucleus. The β^+ decay process is illustrated in equation 2.18,



where, ${}^1_1\text{H}$, ${}^1_0\text{n}$, ${}^0_{+1}\text{e}$ and ν denote, a proton, neutron, positron and neutrino, respectively.

The positron (β^+) decay mode results in the daughter nuclide with one atomic number (Z) less than that of the parent, while the number of neutrons (N) increases by a unit (Cember & Johnson, 2009).

The β^+ decay process and typical example are illustrated in expressions 2.19 and 2.20, respectively,



Typical example:



In an electron capture process, the parent radionuclide nucleus absorbs an atomic electron from the inner shell of the atomic orbit and unites with a proton to form a neutron according to equation 2.21,



The neutrons rich radionuclides decay by the emission of negatron (β^-) particles from the nuclei. In β^- decay mode, a neutron is converted into a proton and an electron accompanied by emission of anti-neutrino ($\bar{\nu}$). A β^- decay process is illustrated by equation 2.22,



A typical example of a β^- decay mode is given in equation 2.23,



Unlike alpha particles, beta particles are emitted with a continuous range of energies ranging from zero to the maximum energy characteristics of the particular nuclide, as shown in Figure 2.1.

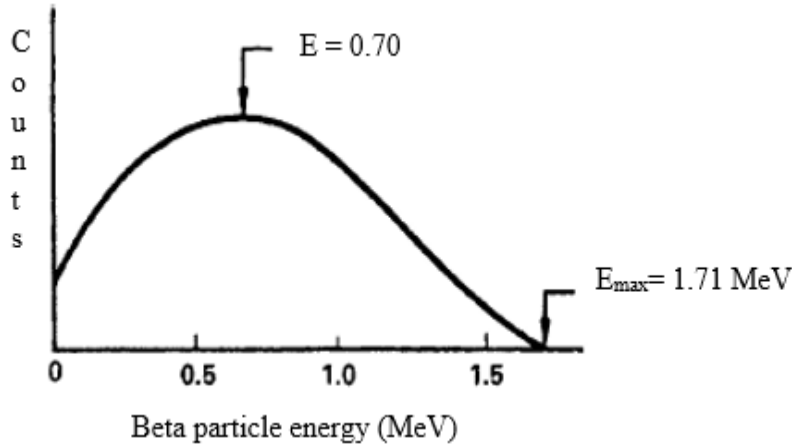


Figure 2. 1: Continuous spectrum of beta particle emission from ^{32}P (Cember & Johnson, 2009).

2.3.3 Gamma ray emission

Gamma ray emission (γ) is a phenomenon that occurs following the alpha or beta decay process (Friedlander *et al.*, 1981). The alpha or beta decay processes may leave the daughter nucleus with excess energy, in a partially excited (metastable) state. Upon de-excitation, the nucleus can either falls directly to the ground state or descends in steps to lower energy states through the emission of discrete energies of gamma radiation corresponding to the energy-state transitions of the nucleus (L'Annunziata, 2012; Magill & Galy, 2005). Unlike α -particle and β -particle, the gamma rays consist of high energy photons of up to 10 MeV, with high degree of penetration into matter (Khan & Gibbons, 2014; Plant *et al.*, 2012).

A Gamma ray emission process can be expressed by equation 2.24,



where, ${}^A_ZD^m$ represents the daughter product in a metastable excited state (Magill & Galy, 2005; Giancoli, 2014).

The emitted γ - energy (E_γ) can be expressed by equation 2.25,

$$E_\gamma = E_i - E_j \quad (2.25)$$

where, E_i and E_j represent the energy level of the nuclear transitions. This is illustrated in Figure 2.2 for the decay scheme process of ^{226}Ra , showing the decays with significant emission intensity (94.4 and 5.55%).

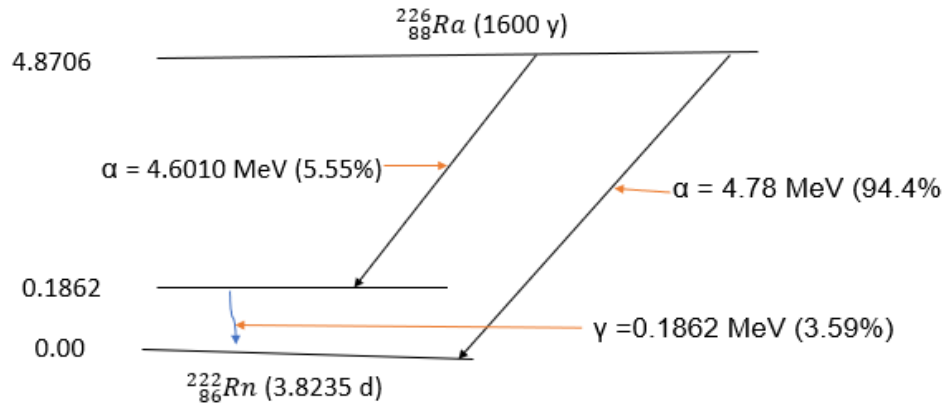


Figure 2. 2: An illustration of gamma emission from the decay scheme of ^{226}Ra (Martin, 2006).

Figure 2.2. shows that the ^{226}Ra decays by α – emission to ^{222}Rn with discrete energies. The energy difference between the major two nuclear transition, at 4.78 MeV and 4.60 MeV, is about 0.186 MeV gamma radiation (Martin, 2006).

Numerous literatures have considered the gamma ray emission as a decay process, however, there is no record of any single radionuclide that has been known to decay primarily by gamma emission. During the γ -ray emission, the atomic number or atomic mass numbers remain unchanged. Thus Martin (2006) suggests that the gamma ray emission should not be referred to as a gamma decay because gamma rays are produced only to relieve excitation energy. The properties of radiation emitted during the nuclei transformations are presented in Table 2.1.

Table 2. 1: Typical particles / radiation emitted during nuclear decays (Plant et al., 2012).

Radiation emitted	Mass (atomic mass unit)	Electrical charge	Potential to cause ionisation	Penetrating power
Alpha (α)	4	+2	Moderately high	Very low
Beta (β^+ , β^-)	0	1 or -1	Moderate	Low
Gamma (γ)	0	0	Low	High

2.4 Branching decay ratio

Some radionuclides may decay by one mode to reach a stable configuration. However, several nuclides decay by a combination of competing decay processes involving α -decay, β^- decay, β^+ decay, γ decay or electron capture to reach numerous stable atoms. The branching decay ratio, therefore, indicates relative intensities of a particular decay mode with respect to the total decay modes (Krane, 1987). ^{40}K is a typical nuclide that undergoes such a competing decay mode, which disintegrates by a β^- decay with about 89.25% of events to the ground state of ^{40}Ca , by ϵ with about 10.55% of events to a 1460.82 keV level of ^{40}Ar , by ϵ with about 0.2% and by β^+ with about 0.001% of events to the ground state of ^{40}Ar (Nucleide, 2009). The decay scheme and branching decay ratio for ^{40}K are schematically illustrated in Figure 2.3.

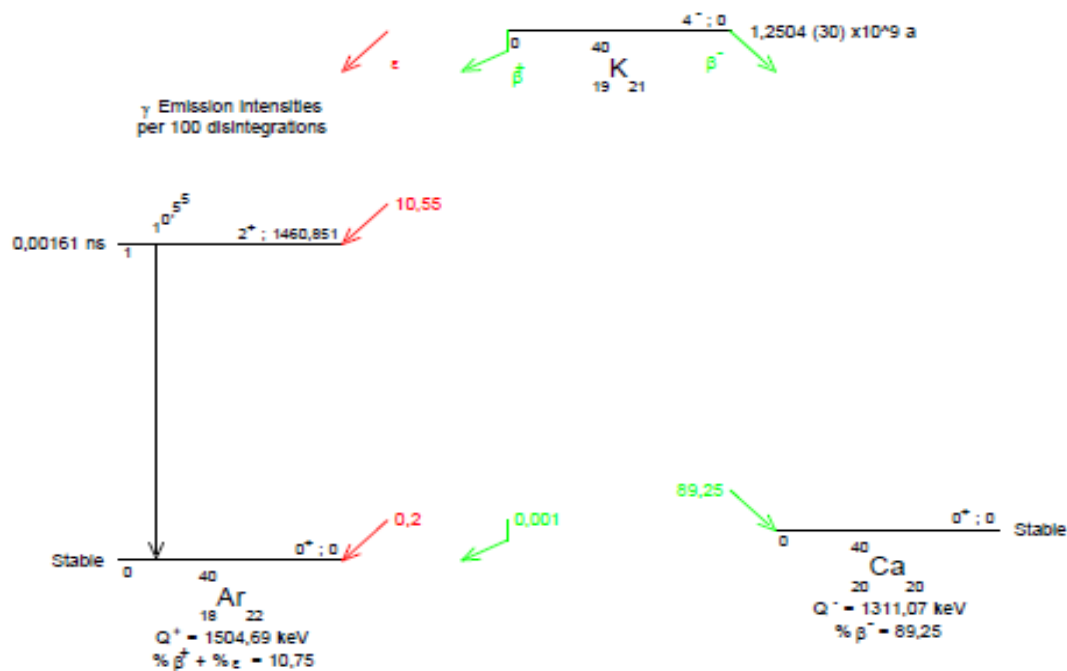


Figure 2. 3: The decay scheme and branching ratios for ^{40}K (Nucleide, 2009).

2.5 Interaction of radiation with matter

Whenever radiation interacts with matter along its path, some or all of its energy may be deposited or absorbed to the particles in the matter. The amount of energy deposited, referred to as absorbed dose, is expressed in terms of Joule (J) per unit of

mass of a matter (kg), given a special name of a “Gray” (Gy) and can be related by equation 2.26 (Cember & Johnson, 2009),

$$1\text{J. kg}^{-1} = 1\text{ Gy} \quad (2.26)$$

It is the energy deposited into matter that causes the chemical, physical, and biological changes that allows for the observation of radiation effects, and to determine the nature of the transition inside the nucleus (Loveland *et al.*, 2006).

2.5.1 Alpha radiation

The mechanism for interaction of alpha particles with matter is primarily through strong coulomb forces between their positive charge and the negative charge of the orbital electrons within the absorber atoms. Upon traversing through matter, alpha particle may exert coulombic forces and sufficient amount of energy on atomic electrons. The process leads to the stripping of outer shell electrons from the atoms of the interacting materials, thereby converting the atoms into ions (ionisation) and/or excitation, raising the electrons to a higher-lying shell within the absorber atom (Knoll, 2010).

The electrical charge (+2) and large mass associated with alpha radiation create a strong interaction with electrons within the absorbing material, thus rendering alpha particles a very high rate of energy loss or transfer into matter and traverse a short range within a matter at relatively slow speed (Leo, 1994). Alpha particles are less penetrative, can easily be stopped by the dead layer of skin or piece of paper or by few centimetres of air (Etim *et al.*, 2014).

An alpha particle is, however, hazardous when ingested or inhaled or interacts with soft tissues such cornea or lining tissues of the internal organs such as bronchioles and lungs (Stoker, 2013). It transfers large amounts of energy per unit path length it travels (high linear energy transfer) that causes dense ionisation and excitation of atoms that could lead to breakage of molecular bonds, destroying or alter the normal biological functions of cells and tissues (Magill & Galy, 2005).

2.5.2 Beta radiation

A beta particle can either be positively (β^+) or negatively (β^-) charged, which in all other respect, is an ordinary electron. Therefore, both beta particle and electrons have the

same mass. When beta particle interacts with electrons of the absorbing matter, the two particles may annihilate, giving rise to two gamma-rays, whose energies are equal to the mass equivalent to the positron and electron (Cember & Johnson, 2009). The annihilation of a beta particle and an electron is illustrated in Figure 2.4.

Beta particle being a charged particle, its electric field interacts with the orbital electron of the absorbing medium to cause electronic excitation and ionization. Beta particles can also produce highly penetrating X-rays called bremsstrahlung when they interact with high-atomic-numbered absorbing materials (Khan & Gibbons, 2014).

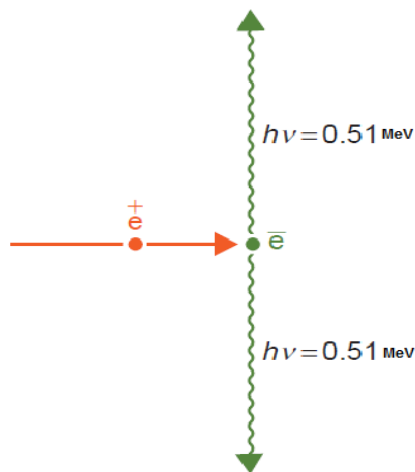


Figure 2. 4: Annihilation of electrons (Khan & Gibbons, 2014).

2.5.3 Gamma radiation

Gamma rays have no rest mass or electric charge, therefore, do not cause ionisation directly but interact with atoms of an absorber material, to liberate charged particles. The charged particles would then cause ionisation, as a secondary process (Khan & Gibbons, 2014). Whenever a beam of photons of sufficient energy passes through a material, there are three major interaction mechanisms for gamma radiation with atoms: photoelectric absorption, Compton scattering and pair production (Ahmed, 2007). Figure 2.5 illustrates the regions, where different gamma ray interaction mechanisms are dominant.

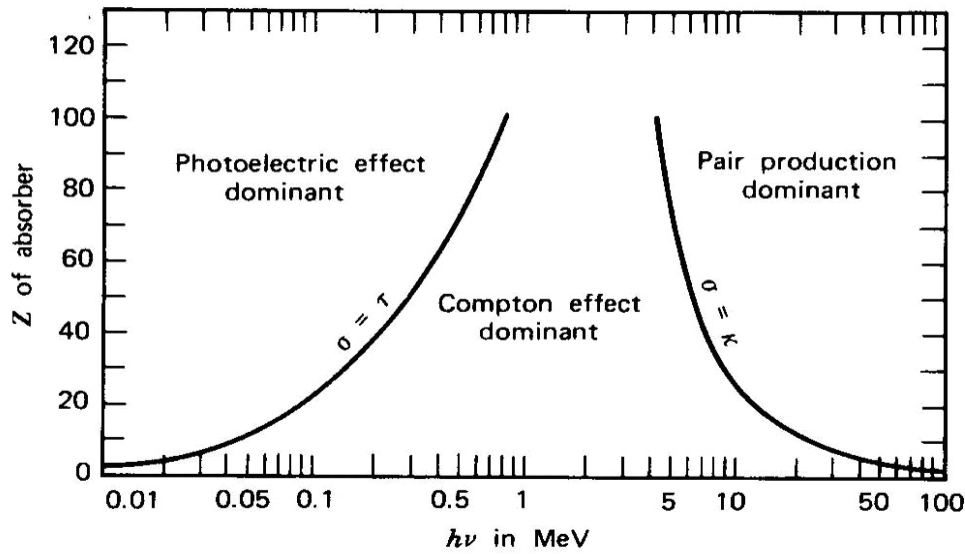


Figure 2. 5: Regions where different gamma ray interaction mechanisms are dominant (Knoll, 2010).

2.5.3.1 Photoelectric effect

The photoelectric effect is a process in which an incident photon transfers all its energy to an electron in one of the atomic shells. The orbital electron (photoelectron) is subsequently ejected with the kinetic energy given by equation 2.27,

$$E_{e-} = h\nu - E_b \quad (2.27)$$

where, E_{e-} , $h\nu$, and E_b represent the kinetic energy of the photoelectron, entire energy of the photon and binding energy of the photoelectron in its original shell, respectively. The photoelectron that leaves the atom in an excited state, subsequently emits characteristic X-ray (Cember & Johnson, 2009; Khan & Gibbons, 2014; Knoll, 2010).

The probability of photoelectric interaction, symbolised τ , is a complex process, however, depends strongly on the atomic number, Z , of the absorbing material and the energy of the incident photon ($h\nu$), therefore can be related by expression (2.28),

$$\tau \propto \frac{Z^n}{(h\nu)^3} \quad (2.28)$$

where n varies between 3 and 4 over the gamma-ray energy region of interest (Khan & Gibbons, 2014). The strong dependence of photoelectric absorption probability on the atomic number Z of the absorber material makes high Z material such as lead a

good material for shielding against X-rays and gamma radiation (Cember & Johnson, 2009).

2.5.3.2 Compton scattering

In a Compton scattering process, part of the energy of the incident photon is transferred to a free or loosely bound electron, which is at rest. The gamma-ray photon with reduced energy is deflected through an angle $\emptyset$ with respect to its original direction, with a subsequent emission of a Compton electron recoiling at an angle Θ , as illustrated in Figure 2.6. The photon can be scattered in any direction, therefore, the energy transferred to the Compton electron can vary from zero to a large fraction of the gamma-ray energy (Ahmed, 2007).

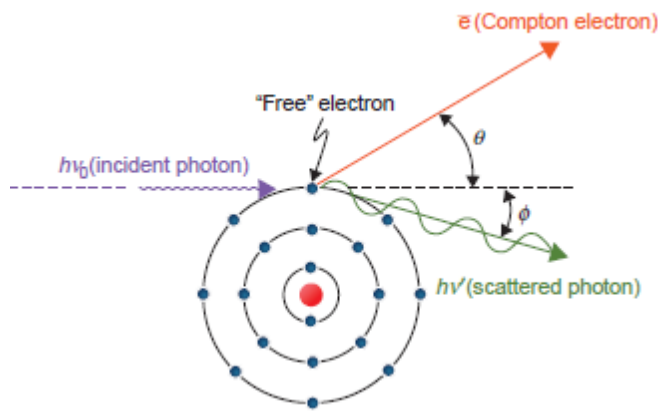


Figure 2. 6: An illustration of the Compton scatter (Khan & Gibbons, 2014).

The energy of the scattered photon $h\nu'$ can be determined by equation 2.29,

$$h\nu' = \frac{h\nu}{1 + \frac{h\nu}{m_0c^2}(1 - \cos\emptyset)} \quad (2.29)$$

where, m_0c^2 is the rest-mass energy of the electron (0.511 MeV) and $\emptyset$ is the angle between the direction of the scattered photon and its original direction (Khan & Gibbons, 2014; Knoll, 2010).

The Compton scattering process is dominant at the intermediate gamma energy region as shown in Figure 2.5. The probability of scattering effects is independent of Z , however directly proportional to the number of electrons per unit mass of the interacting material and inversely on the energy of the incoming photon (Khan & Gibbons, 2014).

2.5.3.3 Pair production

The pair production is more pronounced at photon energies above 1.02 MeV, the energy equivalent to twice the rest-mass of an electron. In the pair production process, the gamma ray is absorbed, leads to the formation of an electron (e^-) and positron (e^+) pair that possess the total kinetic energy given by $(h\nu - 1.02)$ MeV (Kahn & Gibbons, 2014; Knoll, 2010).

The positron annihilates with an electron after slowing down in the absorbing medium, to produce two 0.511 MeV annihilation photons as secondary products of the interaction. The two photons may interact with the absorbing matter either through photoelectric effect or Compton scattering (Khan & Gibbons, 2014; Knoll, 2010). The probability of pair production process, symbolized κ , increases with increasing photon energy and related to the atomic number of the material as approximately proportional to Z^2 (Khan & Gibbons, 2014).

2. 6 Properties and behavior of uranium, thorium and potassium in environment

The behaviour and hence uptake by plants of radionuclides in environment is influenced by several factors, some of which are the physico-chemical properties of soil, water and radionuclides, pH, type of vegetation, soil type etc. (Chen *et al.*, 2005; Forkapic *et al.*, 2017; Harb *et al.*, 2014; Islamović *et al.*, 2012; Rodríguez *et al.*, 2017).

It is, therefore, essential to understand the key processes and parameters affecting radionuclides mobility, bioavailability and uptake by vegetation in order to assess successfully the associated potential environmental risks. This section looked at the properties of uranium, thorium and potassium as well as on the factors influencing their mobility, bioavailability and uptake by vegetation in soil-water environment.

2.6.1 Properties and behaviour of uranium decay series in the environment

Uranium is a naturally occurring weakly radioactive element, universally found in varying small amounts in the earth crust, rocks, water, air, vegetation, humans and animals (ATSDR, 2013). The uranium average abundance in the earth's crust is about 2 parts of uranium per millions of parts of soil (ppm) with typical ranges from 0.1 to 20 ppm (WHO, 2001).

Elemental uranium is composed of three major isotopes, ^{238}U , ^{235}U and ^{234}U , their respective isotopic natural abundances, by weight, of roughly 99.2745%, 0.7200% and 0.0055%. All three isotopes of uranium have the same chemical properties but different radioactive material with different radiological properties. In terms of uranium radioactivity, ^{234}U contributes about 49%, whereas ^{238}U and ^{235}U contribute 48.7% and 2.3%, respectively (ATSDR, 2013).

^{238}U and ^{235}U are the parent nuclides of the uranium and actinium decay series, respectively, whilst ^{234}U is one of the intermediate progeny of ^{238}U decay series (Cember & Johnson, 2009). In natural environment, ^{235}U and its decay products are not considered to pose any significant radiological health hazard due to their insignificant natural abundance (UNSCEAR, 1982), hence not considered in this work. The uranium decay series is headed by ^{238}U , an alpha emitter, decaying through the fourteen-member series of radionuclides having mass number divisible by 4 with a remainder of 2, therefore referred to as a $4n + 2$ series, where n is an integer (Choppin *et al.*, 2002). The ^{238}U decay series is presented in Table 2.2.

Table 2. 2: The ^{238}U decay chain (NRC, 1999).

Isotope symbol	Half-life	Major radiation
^{238}U	4.5 billion years	α
^{234}Th	24 days	β
$^{234\text{m}}\text{Pa}$	1.17 m	β , < 1% γ
^{234}U	245 000 years	α , < 1% γ
^{230}Th	75 000 years	α
^{226}Ra	1 600 years	α , γ
^{222}Rn	3.8 days	α , < 1% γ
^{218}Po	3.1 m	α , < 1% γ
^{214}Pb	27 m	β , γ
^{214}Bi	20 m	β , γ
^{214}Po	160 μs	α , < 1% γ
^{210}Pb	22 years	β , γ
^{210}Bi	5.0 days	β
^{210}Po	138 days	α , < 1% γ
^{206}Pb	Stable	

In an undisturbed natural environment, it is assumed that ^{238}U and its radioactive decay products are approaching the state of secular equilibrium in which the

radioactivity of all nuclides are almost equal (Cember & Johnson, 2009). However, the differences in physical and chemical properties of radionuclides and geochemical processes could disrupt the radioactive secular equilibrium (Deschamps *et al.*, 2004; Mitchell *et al.*, 2013; UNSCEAR, 2000). The geochemical processes involve the dissolution and precipitation of more geochemically soluble radionuclides of a radioactive series, the diffusion of nuclides of radon and the effects of α -particle recoil, that allows dissociation of the decay radionuclides from the source material and transferred further in the environment (Papadopoulos *et al.*, 2013). It had been noted that ^{234}U is generally deficient relative to ^{238}U in soils and enhanced in aqueous media. Similarly, the concentration of ^{226}Ra could be found slightly different from that of ^{238}U , depending on the surrounding environment (UNSCEAR, 2000).

The pH of soil solution and the redox potential directly and indirectly influence all chemical processes and consequently, determine the behaviour of trace elements (Tack, 2010). Uranium reacts with oxygen to form uranium oxides, such as UO_2 and U_3O_8 (del Carmen Lamas, 2005). Uranium can mostly be found in a range of oxidation states, however, only the tetravalent U(IV) and hexavalent U(VI) are relatively stable in the environment. Uranyl ion (UO_2^{2+}) is predominant in the oxidizing environment in which the uranium is present as U(VI), which is very soluble and hence mobile in soil solution (ATSDR, 2013).

The uranyl ion forms complexes of hydroxides, carbonates, sulphates and phosphates, which are very soluble in geological fluids, hence enhance the mobility of uranium (NRC, 2012; Favas *et al.*, 2016; Mitchell *et al.*, 2013). However, U(VI) can be reduced to U(IV) under reducing conditions by microbial activity and chemical reductants (EPA, 1999; Favas *et al.*, 2016; Mitchell *et al.*, 2013) hence reduces its solubility and mobility.

U(IV) species dominate in reducing environments and tend to hydrolyse, forming strong hydrolytic complexes. U(IV) also tends to form sparingly soluble precipitates and strong complexes with naturally occurring organic materials, hydrated fluorides, and phosphates of low solubility. In areas where there are high concentrations of dissolved organic materials, organic complexes may increase U(IV) solubility (ATSDR, 2013; NRC, 2012; EPA, 1999; Mitchell *et al.*, 2013). According to EPA (1999), the hydrolysis of uranyl ion occurs at approximately pH 7, which in non-complexing media

leads to the formation of several hydrolysed species; UO_2OH^+ , $\text{UO}_2(\text{OH})_2$ and $(\text{UO})_2(\text{OH})_2^{2+}$, which limit solubility, hence minimising uranium availability for plants uptake.

A study by Ebbs *et al.*, (1998) observed the greatest uptake of uranium by plants under acidic conditions, where UO_2^{2+} is the dominant ionic species in solution. On the contrary, ATSDR (2013) suggested that uranium in the soil is not taken up by plants but adsorbed onto plant roots such as potatoes. Numerous studies had found the high concentration of uranium in roots than in other parts of plants such as leaves (Ebbs *et al.*, 1998; Shtangeeva, 2010).

2.6.2 Properties and behaviour of thorium in the environment

Thorium is a naturally occurring radioactive metal, found in small amounts in rocks, soil, water, plants, and animals. The global average of thorium in the earth crust is about $6 \mu\text{g.g}^{-1}$ with typical ranges of near zero to $25 \mu\text{g.g}^{-1}$. Elemental natural thorium exists as six isotopes; ^{227}Th , ^{228}Th , ^{230}Th , ^{231}Th , ^{232}Th and ^{234}Th . ^{227}Th and ^{231}Th belong to the ^{235}U decay series, whereas ^{230}Th and ^{234}Th belong to the ^{238}U decay series while ^{228}Th and ^{232}Th belong to the ^{232}Th decay series, whereby ^{232}Th is the head of the decay series (ATSDR, 2019).

Natural thorium is mostly occurring as ^{232}Th with a natural abundance of 100%. ^{232}Th with a half-life of 1.4×10^{10} years is a pure alpha emitter, decaying through the twelve-member series of radionuclides with characteristic mass numbers divisible by 4 with no remainder, therefore referred to as a 4n series, where n is an integer (Choppin *et al.*, 2002). The ^{232}Th decays by several alpha – beta decays sequences, some of which are accompanied by gamma ray emission until reaching a stable ^{208}Pb .

In nature, ^{232}Th is found in association with minerals such as monazites, granites and gneiss. ^{232}Th is found stable in tetravalent oxidation state (Th^{+4}) in reducing environment, sparingly soluble and less mobile in aqueous solution with exceptions in the presence of organic matter (ATSDR, 2019; EPA, 1999).

2.6.3 Properties and behaviour of potassium in the environment

Potassium belongs to group 1 elements of the periodic table. There are three naturally occurring isotopes of potassium; ^{39}K , ^{40}K and ^{41}K with their respective natural

abundance of 93.9%, 0.0177% and 6.70%. Potassium is an essential element for animal and plant cellular formations (Radović-Rajević & Zovko, 2012). Most potassium minerals are sparingly soluble. Potassium is mostly found in a stable +1 oxidation state. Of the three isotopes, only ^{40}K is a radioactive atom with a half-life of 1.251×10^9 years. ^{40}K disintegrates by β^- decay with about 89.25% of events to the ground state of ^{40}Ca , by ϵ with about 10.55% of events to a 1460.82 keV level of ^{40}Ar , by ϵ with about 0.2% and by β^+ with about 0.001% of events to the ground state of ^{40}Ar (Nucleide, 2009).

2.7 Soil to plant transfer factor

Soil is the main source of naturally occurring elements, some of which are essential nutrients for plant and animals' biochemical processes. Trace elements including radionuclides that are not needed for development and growth are taken up together with essential elements, from soil through roots and accumulate in different portions of plants (Eisenbud & Gesell, 1997). A relative measure of the transfer of the radionuclide from the soil to the plant, referred to as Transfer Factor (TF), can be expressed by equation 2.30.

$$TF = \frac{\text{Activity concentration of a radionuclide in dry crops (Bq.kg}^{-1}\text{)}}{\text{Activity concentration of a radionuclide in dry soil (Bq.kg}^{-1}\text{)}} \quad (2.30)$$

and is defined as the ratio of the activity concentration of radionuclide in the dry mass of the plant (Bq.kg^{-1}) to that in dry mass of soil (Bq.kg^{-1}) (Gaffar *et al.*, 2014; IAEA, 2010; Tack, 2010). The dry weight for soil and crop products in equation (2.30) is preferable over the “per unit fresh weight” because the amount of radioactivity per kilogram dry weight is much less variable than the latter, thus reduces uncertainties (IAEA, 2010). The soil to plant transfer factor reflects the relative mobility of radionuclides in the soil-plant system (Tack, 2010).

The transfer factor is an important parameter that is widely used in the radiological safety assessment model. The TF allows for the estimation of the amount of radioactivity that could be present in the agricultural crops and in the calculation of radiological human doses via the ingestion pathway (Chakraborty *et al.*, 2013; Mheemeed *et al.*, 2014).

IAEA (2009a and 2010) published transfer factors for several elements in various food crops in different ecosystems. These factors serve as the reference values for use by modellers in radiological assessment. It has however, been observed that the TF in the natural environment is not constant in time and dependent on numerous site-specific factors, such as soil type, physicochemical characteristic of soil and radionuclide, radionuclide concentration on the rooting zone, plant species, organic content, interfering elements (UNSCEAR, 2000). IAEA (2010) therefore, discourages the use of generic reference values and emphasized for preferential use of site-specific data or values that are in close relation to the most important physical and/or chemical variables of the site under investigation.

Shtangeeva (2010) observed that the transfer of primordial radionuclides along the terrestrial food chains had not been studied extensively in natural environments as that of artificial radionuclides. Most of the studies on the transfer of primordial radionuclides along soil-vegetation systems were conducted under controlled experimental conditions and contaminated land (El-Taher & Al-Turki, 2014; Mubarak *et al.*, 2017; Neves *et al.*, 2008; Neves & Abreu, 2009; Skoko *et al.*, 2017; Vera Tomé *et al.*, 2002). These controlled conditions are extremely different from the actual natural environmental conditions as the ones encountered in the current study area.

In recent times, numerous studies have been undertaken worldwide to assess the transfer of primordial radionuclides from soil to food crops, plants and grass (Mheemeed *et al.*, 2014; Thabayneh & Jazzar, 2012, 2014).

2.8 Exposure to primordial radionuclides

From the radiation protection point of view, IAEA (2014a) distinguishes three types of radiation exposure situations: planned exposure, emergency exposure and existing exposure. A planned exposure situation arises from the planned activities, such as handling of radioactive material, so the exposure is normally anticipated to occur or simply, there is a potential for exposure to occur. The minerals exploration, mining and processing are some of the planned activities with potential for redistribution of the primordial radionuclides from inaccessible locations to locations, where humans are present and thus an enhancement the concentration of primordial radionuclides to

above levels encountered in natural state. Such alterations of the natural environment can increase radiation exposures of the public (NRC, 1999).

An emergency exposure situation may arise as a result of an accident or any other unexpected event that requires prompt action in order to avoid or to reduce adverse consequences that has the potential of releasing radionuclides into the environment. In Namibia, any planned activity that involves handling of radioactive material requires comprehensive studies and documentation on the potential impacts associated with the activities. The documentation should include management plans, preventive measures and response actions required in order to minimise adverse consequences (Ministry of Health and Social Services [MHSS], 2005; Ministry of Environment and Tourism [MET], 2007).

The existing exposure situation is defined as a situation of exposure that already exists when a decision on the need for control needs to be taken (IAEA, 2014a). This is applicable to the prolonged or chronic exposure to inherently enhanced levels of natural background radiation in areas due to the presence of primordial radionuclides in soil, water, air and vegetation.

Numerous IAEA member states have developed and implemented programs for the identification, quantification, monitoring and managing the radiological risks associated with high background radiation areas (IAEA, 2011). In order to evaluate the dose and estimate radiological risks associated with primordial radionuclides, the potential exposure pathways should be identified and characterised. The exposure pathways are routes that the radiation takes to reach individuals. The exposure pathways are therefore, categorised as external and internal exposure pathways (IAEA, 2005; IAEA, 2014a). It is estimated that the worldwide average annual exposure to natural sources of radiation is 2.40 mSv (NRC, 2006) of which 0.31 mSv is attributable to ingestion of foodstuff and water containing ^{238}U and ^{232}Th decay series as well as ^{40}K , whereas 1.20 mSv is estimated to arise from internal exposure to radon and its progeny (UNSCEAR, 2010).

2.8.1 External exposure

An external exposure pathway exists when an individual is exposed to a penetrating radiation from radionuclides outside the body (IAEA, 2014a). As indicated earlier, the

primordial radionuclides and their respective decay products may emit a combination of alpha, beta and gamma radiation during the decay process.

Alpha particles have exceptionally low penetration power in matter and can be wholly absorbed by the dead outer layer of the skin. Beta radiation on the other hand, dependent on the energy, has low to medium capability to penetrate tissues to varying depth. Therefore, exposure to alpha emitting radionuclides from outside the body does not constitute an external radiation hazard, whereas beta radiation maybe an external radiation hazard. The gamma radiation has a high penetration power in matter and therefore, penetrates definite depth of body tissues (Cember & Johnson, 2009). Numerous studies around the world demonstrated that gamma radiation emitted from ^{40}K and the radionuclides of the ^{232}Th and ^{238}U decay series, which exist in all ground formations, represents the main external source of irradiation to the human body (Han & Park, 2018; Mubarak & Hafez, 2016; NRC, 1999; UNSCEAR, 2000; UNSCEAR, 2010).

In the assessment of external exposure to gamma radiation, two external exposure scenarios have been defined; that is, exposure in outdoor and indoor environments. In an outdoor environment, external exposure arises as a direct gamma radiation from the primordial radionuclides in soil. Sources of external exposure in an indoor environment is the direct gamma radiation from soil and from the building material. It is established that the level of radiation in soil is related to the parent rocks from which the soil originates, and generally building materials derived from or associated with igneous rocks such as granite had been known to host enhanced level of primordial radionuclides (IAEA, 2003; IAEA, 2005; IAEA, 2011; Murray, 1989). Considering the time spent indoors (about 80% of the time) and the dwellings surrounding configuration, the indoors external exposure to gamma radiation may be up to 40% greater than that from an outdoor exposure (UNSCEAR, 2000).

In Serbia, the state promulgated a law that sets limit values for primordial radionuclides contents in the building materials (Todorovic *et al.*, 2015). The same law provides for testing all building material for primordial radionuclides contents against the limit values (Bq.kg^{-1}) for internal and external design, given in Table 2.3.

Table 2. 3: Limit values for interior and external design.

Radionuclide	Limit values for internal design (Bq.kg ⁻¹)	Limit values for external design (Bq.kg ⁻¹)
²²⁶ Ra	310	410
²³² Th	210	310
⁴⁰ K	3100	5100

This intervention is aimed to control usage of materials containing elevated levels of natural radionuclides, therefore, eliminating possible cases of unnecessary exposure.

2.8.2 Internal exposure

Internal exposure refers to the radiation exposure of an individual from radionuclides incorporated in the body (IAEA, 2014a). Primordial radionuclides enter the body mainly through inhalation and ingestion. Anthropogenic activities such mineral exploration, mining and processing, sand excavation and movements of vehicles, generate voluminous dust. The primordial radionuclides may be suspended in the air and attach to dust particles, transferred distances and subsequently inhaled (UNSCEAR, 2010). The inhaled radionuclides are incorporated into body tissues and irradiate the body over the time period dependent on its physical half-life and biological retention within the body (ICRP, 2007). It is estimated that the global average effective dose from inhalation and ingestion of terrestrial radionuclides, excluding the contribution from radon and its progeny, is 310 µSv, of which 170 µSv is from ⁴⁰K and 140 µSv from the long-lived radionuclides in the uranium and thorium series (UNSCEAR, 2000).

Internal exposure to radon is of more concern, especially in enclosed spaces and habitats with inadequate ventilation systems, as radon is readily built up, reaching hazardous concentration levels. Radon gas emanates from rocks, soil and building material and dispersed in air. Once decays, its solid radioactive decay products, particularly ²¹⁰Pb, ²¹⁰Po, ²¹⁴Bi and ²¹⁴Pb, attach to aerosols and particles in the air, transported and deposited as fallouts onto soil, vegetation and water. Radon enters buildings either by emanation from the building material or diffusing through the concrete or through cracks that may occur in the concrete and walls (ATSDR, 2012).

Inhalation of radon decay products results in their deposition and being retained in the lungs for long periods, hence irradiate organs at close range. The linings of lungs and other internal organs are made of soft tissues that render no protection against alpha, beta and gamma irradiation. The alpha particles cause more ionisation because of its high linear energy transfer. However, radon is of less health concern in outdoor environments as it is readily diluted with atmospheric air (ATSDR, 2012).

Humans can also be exposed to primordial radionuclides through ingestion of food crops and water containing radionuclides. Contamination of food crops arises from the root's uptake of radionuclides in soil solution or through foliar uptake following aerial deposition (IAEA, 1990). The radionuclides are then translocated and distributed within the different edible parts of the plants. This process constitutes the crucial step of radionuclides input into the food chain (Aswood *et al.*, 2013; Černe *et al.*, 2010). Ingestion of such food crops incorporates radionuclides into body tissues (El-Taher & Al-Turki, 2014). It is estimated that the average annual dietary intake of ^{238}U , ^{232}Th and ^{226}Ra is 5 Bq, 1.7 Bq and 22 Bq, respectively, in areas of normal radioactivity (UNSCEAR, 2000). Once in the body, the radiation irradiates the exposed cells, tissues and host organs at close range (Khandaker *et al.*, 2012).

2.9 Biological effects of radiation

All living organisms are made of atoms joined together by covalent bonds to form molecules, which are further bonded together to form cells (NRC, 1990). Similar cells bind to one another forming tissues, which are subsequently grouped together as per their functionality to form organs. The cells contain organic and inorganic compounds such as proteins, carbohydrates, minerals and nucleic acids, playing their respective biological functions (Ahmed, 2007). The most important part of the cell is the chromosome, which carry the deoxyribonucleic acid (DNA). The DNA consists of two long strands of nucleotides wrapped about one another in a “double helix” as schematically illustrated in Figure 2.7. It plays a crucial role in the coding of the genetic information necessary for reproduction, growth, development and biological functioning of all known living organisms (Giancoli, 2014; Hall & Giaccia, 2012).



Figure 2. 7: Illustration of the DNA double helix (Giancoli, 2014).

It follows that the biological effects of ionising radiation begin with the interaction of radiation with atoms as described in section 2.5. The excitation or ionisations of atoms breaks bonds holding molecules together, thereby producing several chemical changes and biological damage. The fact that ionising radiation causes biological damage came to light following the manifestation of skin erythema and ulceration experienced by Becquerel after had handled the radium material in the 1896 (Hall & Giaccia, 2012). Studies from laboratory experiments on animals and survivors of radiation incidents/accidents demonstrated that the DNA is the principal target for the biological effects of radiation (Hall & Giaccia, 2012).

The radiation damage to the DNA can arise from either direct or indirect action. The direct action refers to the process when the radiation interacts with the DNA molecules, causing the excitation or ionisation of the atoms in the DNA molecules (Hall & Giaccia, 2012; NRC, 1990). This process eventually leads to the dissociation and breaking down of the chemical bonds of the DNA molecules (Leo, 1994).

In the indirect action, noting that most of the body is water, radiation interacts with and ionises water molecules to generate highly reactive free radicals, as per the chemical reaction expressed by equation 2.31,



where, H_2O^+ is an ion radical.

The ion radicals may react with other water molecules to form other highly reactive hydroxyl radicals ($\text{OH}^\cdot$), whose chemical reaction is expressed by equation 2.32,



Water molecules may also be excited and break up into the $\text{H}^\cdot$ and $\text{OH}^\cdot$ radicals as per the reaction expressed by equation 2.33,



These radicals are highly reactive and can diffuse a short distance to attack DNA in the cell, leading to the biological damage (Hall & Giaccia, 2012; NRC, 1999). The mechanism for direct and indirect actions of radiation on the DNA is illustrated in Figure 2.8.

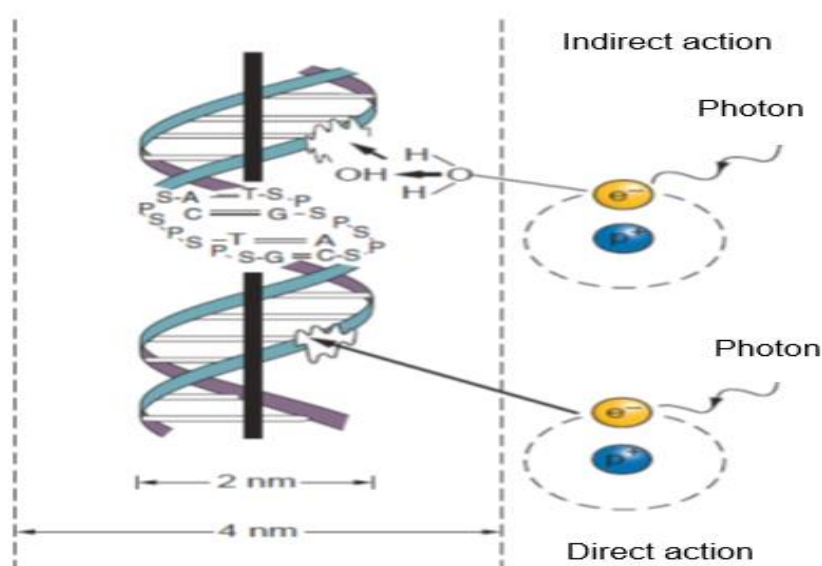


Figure 2. 8: Process for interaction of radiation with the DNA molecule (Hall & Giaccia, 2012).

The tissues or organs consist of cells, differing in their susceptibility to certain type of radiation at a given dose (Cember & Johnson, 2009). Therefore, the biological effects of radiation do not only dependent on absorbed dose but also on the type of radiation received, the duration of exposure, the organ or tissue exposed, the age and general health of the exposed individual (UNSCEAR, 1982; WHO, 2011).

The adverse health effects of radiation have been categorised into deterministic effects (tissue reactions) and stochastic effects (Cember & Johnson, 2009; Hall & Giaccia, 2012). The deterministic effects are those health effects or damage in populations of cells that can be clinically experienced few days or weeks after an exposure to a radiation dose in excess of the threshold dose of that specific group of cells or tissues. The signs and symptoms of these effects are predictable and reproducible, and the severity of the effect increases with dose (ICRP, 2007) and occurs in all individuals, who receive a dose in excess of the threshold (UNSCEAR, 2000). The threshold dose could be understood as a minimum amount of energy that results in an observable tissue reaction.

It follows then that an exposure to an above threshold dose causes a large number of cells to die, causing the organ containing such cells to malfunction. Some of the documented deterministic effects include nausea, permanent sterilisation of ovaries and testes, reddening of the skin, cataracts and bone marrow failure (Cember & Johnson, 2009).

The onset of the stochastic effects is believed to emanate when the radiation damage to the chromosomes molecules disrupts the replication and transmission of genetic information contained in the DNA to the subsequent cells. If the radiation damage occurs in the information stored in the chromosomes of the germ cells, the resultant effects do not affect the exposed person, but may be transferable to the offspring of persons exposed to radiation (NRC, 1990). UNSCEAR (2000) noted however, that the radiation induced hereditary effects are yet to be pronounced in human populations, although they have been observed in other species. Table 2.4 highlights some of the potential tissue effects in an adult human corresponding to an estimated total dose (threshold dose) received in a brief single exposure.

Table 2. 4: Threshold dose and corresponding tissue effects in the adult human indicated organs in a brief single exposure (ICRP, 2007).

Organ	Threshold absorbed dose (Gy)	Effects
Testes	0.15	Temporary sterility
	3.50 – 6.00	Permanent sterility
Ovaries	2.50 – 6.00	Sterility
Lens	0.50 – 2.00	Detectable opacities
	5.00	Visual impairment
Bone marrow	0.50	Depression of haematopoiesis

Radiation damage to the information stored in the chromosomes of a somatic cell results in the uncontrollable reproduction and development of cells of the exposed individual, leading to malignant disease such as cancer, however, may not be inheritable by their offspring (Cember & Johnson, 2009). The stochastic effects may appear late in the order of years or decade (Ahmed, 2007), and do not necessarily affect all exposed individual, but the probability of an effect occurring, not its severity, is regarded as a function of dose without threshold (ICRP, 2012a). In order to prevent the deterministic and stochastic health effects, MHSS (2005) has prescribed annual dose limits of 1 and 20 mSv for a member of the public and occupationally exposed worker, respectively, based on international recommendations. However, the dose limits are only applicable to exposure as a result of authorised activities and practices.

2.10 Assessment of radiological health risks

Epidemiological studies and data from the Japanese atomic bomb survivors, radiation workers and patients, who had been subjected to radiation for diagnostic and treatments, radiation accidents as well as from the nuclear industry and experimental studies on animals have indicated a correlation on significant increase of cancer risk at absorbed doses above 100 mGy (ICRP, 2007). Radiation risk estimates to low level of exposures have been extrapolated from studies of large population groups exposed to acute radiation doses, using mathematical models (Dauer *et al.*, 2010; NRC, 1990). However, some researchers expressed reservations on the validity of such extrapolation (Vaiserman *et al.*, 2018). The review by Kreuzer *et al.*, (2015) and Vaiserman *et al.*, (2018) on epidemiological studies on the health effects associated with low doses of radiation could not find evidence to support linear no threshold

models at low levels of radiation. Vaiserman *et al.*, (2018) noted about the scientific uncertainty and inconclusive evidence on the biological effects of low-level dose or dose rates. Some researchers recognised the challenge on the clinically and pathologically distinguishing cancer caused by low level of radiation from those caused by other cancer-causing agents (Agrawal, 2014; Mozdarani, 2012) while some opined that exposure at low doses may induce the body adaptive and repair capability for self-protection against cancer (Cember & Johnson, 2009; Luckey, 1999; Vaiserman *et al.*, 2018).

The available information suggests that the deterministic effects should not be considered as a health risk at absorbed doses of less than 100 mGy. The risk of tissue reactions, specifically lens of the eyes and circulatory system, becomes more pronounced after an acute dose or accumulated dose of about 500 mGy (ICRP, 2012a). This type of exposure may only be encountered during radiation incidents and accidents or during medical exposure, and may not be anticipated from normal occupational exposure or public situations (ICRP, 2012a). The American Health Physics Society (HPS, 2019) advises against estimating health risks to people from exposures to low level of ionizing radiation because of high statistical uncertainties at these low levels.

An absorbed dose measures the intensity of energy deposited into a tissue/organ. Knowing that different tissues react differently to types of radiation and the subsequent biological effects vary with the type of radiation and the radio-sensitivity of tissues exposed, absorbed dose is not a best estimate for stochastic effects (UNSCEAR, 2000).

The dosimetric quantities; equivalent dose and effective dose have been derived from the absorbed dose and designed to consider the differences between the different types of radiation, tissue and organ radio-sensitivity (Choppin *et al.*, 2002; Fisher & Fahey, 2017; WHO, 2011). Equivalent dose is calculated for each tissue/organ based on the absorbed dose and the weighting factor specific to the type of radiation to take into account the biological effectiveness of radiation. The equivalent dose to an organ/tissue $H_{T,R}$ is expressed mathematically by equation 2.34,

$$H_{T,R} = w_R \cdot D_{T,R} \quad (2.34)$$

where w_R and D_{T_R} represent the radiation weighting factor and the average absorbed dose from radiation R in tissue T, respectively.

The effective dose, which represents the whole-body dose is expressed mathematically by equation 2.35,

$$E = \sum_T w_T \cdot H_{T,R} \quad (2.35)$$

and defined as a summation of the organ equivalent doses, each multiplied by the appropriate tissue weighting factor, w_T , (ICRP, 2012b; IAEA, 2014a). The ICRP (1991) provides the radiation and tissues weighting factors normally used in the derivation of an effective dose based on a given absorbed dose. The effective dose serves as a primary dosimetric quantity for radiation protection upon which direct comparisons of the associated biological impacts and the regulatory dose limits have been based (ICRP, 2012b). Both equivalent and effective doses are measured in Sievert (Sv).

Radium equivalent activity (Ra_{eq}) has been one of the parameters widely applied in the assessment of radiological hazards of building materials containing ^{226}Ra (^{238}U), ^{232}Th and ^{40}K (Asaduzzaman *et al.*, 2015; Lu *et al.*, 2014; Michael *et al.*, 2010). It was formulated to express radiation exposure output from the non-uniformly distributed of ^{226}Ra , ^{232}Th and ^{40}K in material as a single index (Amanjeet *et al.*, 2017; Michael *et al.*, 2010). UNSCEAR (2000) recommended for the maximum permissible value of the Ra_{eq} to be less than or equal to 370 Bq.kg^{-1} , for the effective dose to the public to be restricted within a 1 mSv.y^{-1} . Numerous studies investigated the lifetime cancer risk (LCR), the probability of radiation-induced cancer in populations as a result of lifetime exposure to primordial radionuclides (Kekelidze *et al.*, 2017; Nguemem *et al.*, 2016; Njinga & Tshivhase, 2018; Ononugbo & Bubu, 2017; Ramasamy *et al.*, 2009).

2.11 Instrumentation for measurements of terrestrial primordial radionuclides in environmental media

There are numerous types of instruments for detection and measurement of radioactivity in the environmental media. However, the basic requirement of any such instrument is that its detector interacts with the radiation in such a manner that the

magnitude of the instrument's response is proportional to the radiation effect or radiation property being measured (Cember & Johnson, 2009).

The choice of instrument and measurement methods are determined by factors such as the purpose of measurement, physical characteristics of the sample media (matrix), types of information needed (measurement quantity), sensitivity, detection efficiency and resolution of the instrument, type and energy of radiation emitted, quantity of sample available, expertise availability as well as costs and time associated with the analysis (Knoll, 2010; Sami & Akber, 2017).

The advancement in nuclear spectrometry instrumentations not only enables for the detection of the presence of radiation field, but for the analysis of energy distribution of the radionuclides. For decades, the solid-state semiconductor detector systems, specifically alpha spectrometry and high purity germanium (HPGe) detectors gamma (γ) – spectroscopy have gained popularity amongst others and have been primarily widely used for the qualitative and quantitative determination of primordial radionuclides in environmental media (Chakraborty *et al.*, 2013; Gaffar *et al.*, 2014; Saïdou *et al.*, 2008). The inductively coupled plasma mass spectrometry (ICP-MS) is another, emerging though complementary technique, that has gained its recognition as a fast and reliable technique for the determination of long-lived radionuclides in numerous environmental media (Aswood *et al.*, 2013; Han & Park, 2018; IAEA, 2006).

2.11.1 Overview of solid-state semiconductor detector

A semiconductor is material with electrical conductivity properties that lie between that of a conductor and an insulator. The most widely used semiconductor materials in the fabrication of solid-state detectors are silicon and germanium (Knoll, 2010). Silicon and germanium belongs to group IV of the periodic table, each having four (4) valence electrons that are available for participation in covalent bonds to form silicon or germanium crystalline lattice. In the lattice, each germanium or silicon atom forms four covalent bonds with four neighbouring germanium or silicon atoms (Cember & Johnson, 2009; Tavenier, 2010). Figure 2.9 shows a schematic crystalline lattice of germanium.

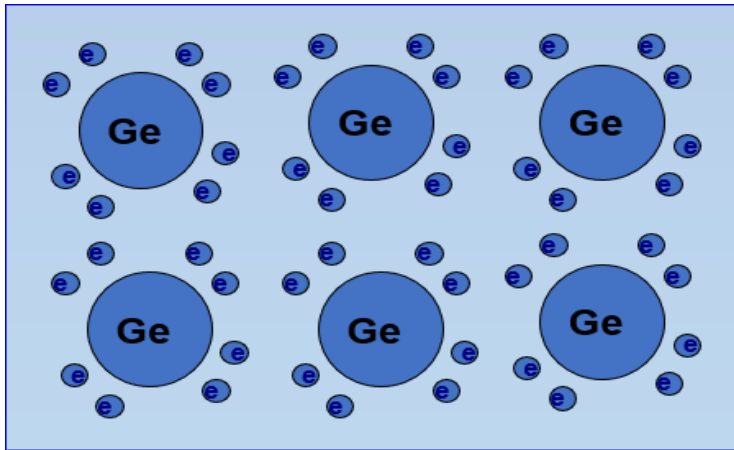


Figure 2. 9: The crystalline lattice of germanium (IAEA, 1995).

In the crystal lattice, the discrete energy levels of the atoms are broadened into valence and conduction bands, which can contain a fixed number of electrons (Ahmed, 2007; Spieler, 2005). As shown in Figure 2.10, the valence and conduction bands are separated by ranges of forbidden energies or band gaps, the size of which determines the electrical conductivity properties of the material. The size of the band gap is approximately 5 or more eV and 1 eV for an insulator and semiconductor, respectively, whereas there is no distinct separation between the valence and conduction bands in a conductor material (Tavenier, 2010).

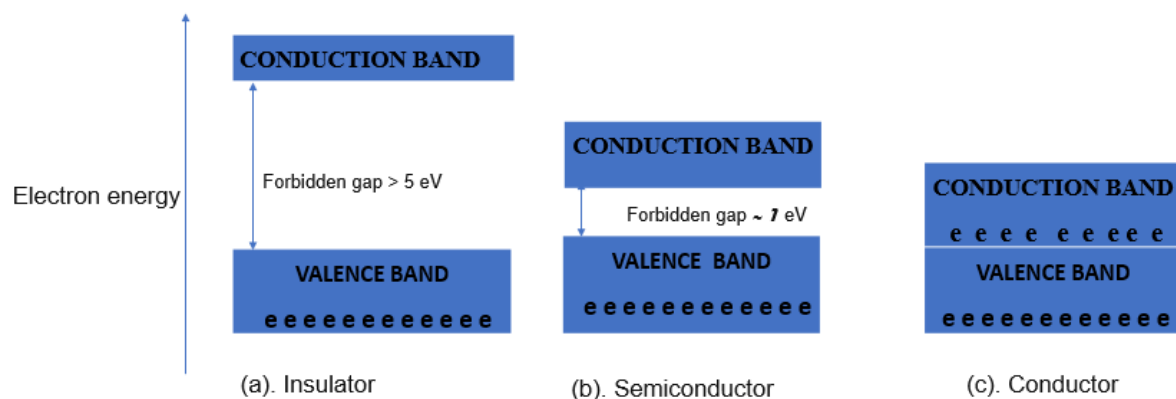


Figure 2. 10: Electronic band structures for an (a) insulator, (b) semiconductor and (c) conductor material (Tavenier, 2010).

At an absolute zero temperature (0 K), the valence band of the semiconductor is completely filled with electrons, bounded at specific sites while the conduction band is vacant as electrons do not possess energy to go through a range of energy levels to

the conduction band, therefore, no electricity conduction possible. As it can be seen in Figure 2.10 (b), the width of band gap in the semiconductor material is very small, of the order of 1eV. This hence implies that, any temperature, however small, above absolute zero, has the potential to impart thermal energy sufficient to break the covalent bonds, excite and set electrons into motion across the bandgap into the conduction band (Spieler, 2005). The moving electrons leave behind vacancies/holes in the position they formerly occupied in the valence band. These holes can be filled in by other adjacent valence electrons and so forth. If the semiconductor is connected to a closed electric circuit, (the positive and negative electrodes on opposite sides), the electrons are attracted to the anodes whereas the holes are attracted towards the cathode, hence a current flow is created in the semiconductor (Cember & Johnson, 2009). For a semiconductor material to be of use in the gamma ray detection, it must be kept at cryogenic temperature to ensure reduction in the number of electrons in the conduction band, which subsequently reduces the background current, thereby improving the detection of excitation due to the gamma-ray interactions (Gilmore, 2008).

It follows then that the fabrication and operation of a semiconductor detector is dependent on the semiconductor material having either excess of electrons or excess of holes (Cember & Johnson, 2009). The excess electrons or excess holes may be achieved by the controlled addition of impurity atoms, the process referred to as doping, to the crystal lattice. The impurity atoms, the dopants, mostly used in the fabrication process are atoms found in group III and group V of the periodic table, because they have one electron less or more in their valence band, respectively (Krane, 1987).

Substitution of Ge in the lattice with group III elements, for example Boron (B), forms covalent bonds with three neighbouring Ge atoms, one free electron and a vacant hole to a Ge atom, as illustrated on Figure 2.11.

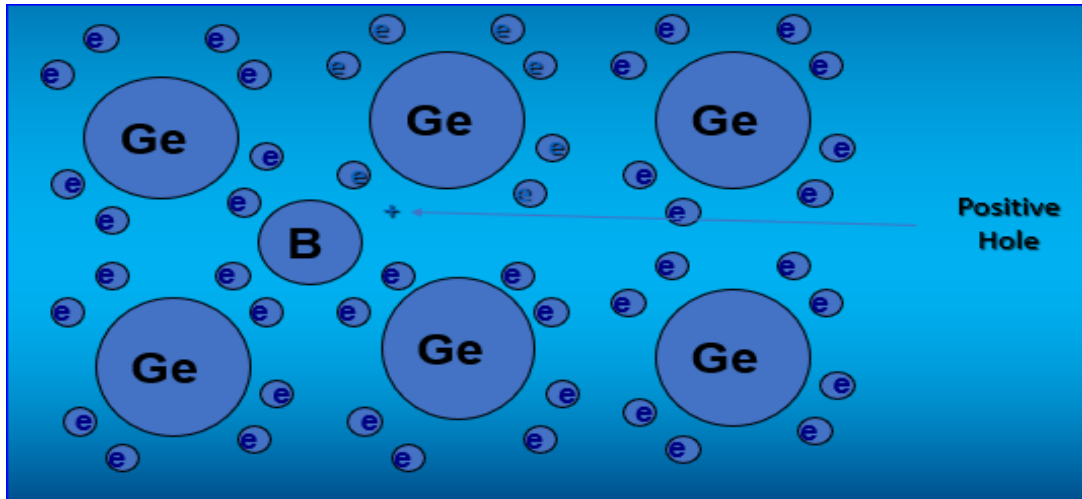


Figure 2. 11: Illustration for a Ge lattice with group III impurity (IAEA, 1995).

The vacant hole (+) can be filled by an electron from the valence band, however, the covalent bond will not be identical to the rest of the crystal, resulting in the formation of an electron site, an acceptor level, in the forbidden band gap near the bottom of the forbidden or band gap as shown in Figure 2.12.

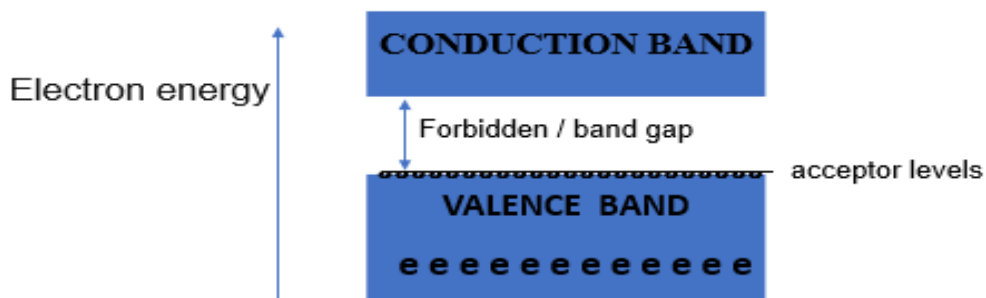


Figure 2. 12: Illustration of an acceptor levels created in the Ge bandgap (Krane, 1987).

The group III elements are referred to as electron acceptors because they can take on an electron from the valence band whereas a semiconductor material with the holes (+) in majority is called a p-type (Krane, 1987). Substituting Ge in the lattice with group V elements, for example phosphorus, results in the formation of covalent bonds with the four neighbouring Ge atoms, leaving behind an excess electron that is loosely attached to the phosphorus, as illustrated in Figure 2.13.

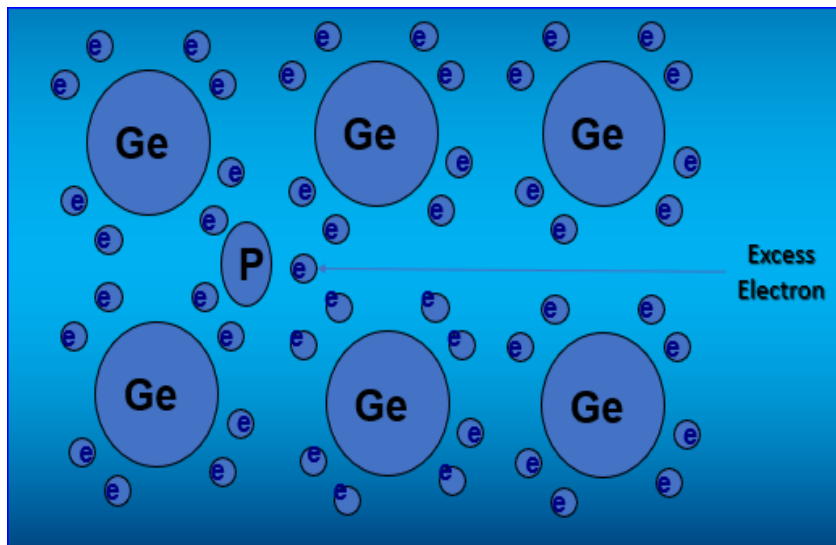


Figure 2. 13: Illustration for Ge lattice with group V impurity (IAEA, 1995).

This arrangement brings the electron energy levels close to the top of the band gap, creates an electron donor levels where electrons can easily be excited to the conduction band as depicted in Figure 2.14,

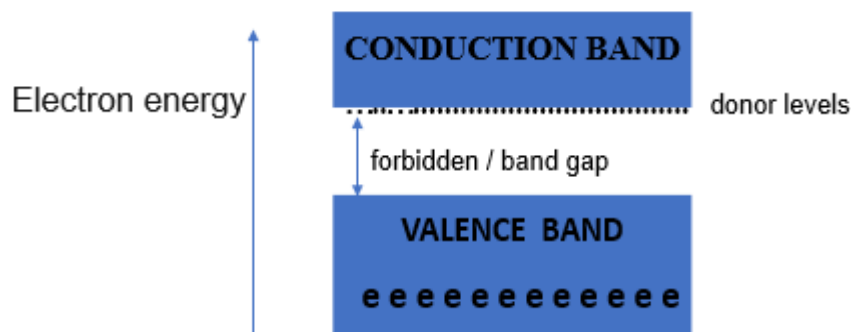


Figure 2. 14: An illustration of a donor levels created in the Ge bandgap (Knoll, 2010).

The group V elements are considered as donors because there is a high probability that an excess electron may be raised into the conduction band. A semiconductor material with excess of electrons (negative charges) is called an n-type (Gilmore,2008). Bringing together the n-type and p-type semiconductor material, forms a p-n junction with the electrical properties relatively different from those of individual types. The p-n junction technology has evolved to become backbone in the building of the semiconductor radiation detection system. At the interphase of the p-n

junction, some electrons migrate from the n-type region through and fill the holes on the p-type region, whereas some holes migrate from the p-type region through and capture the electrons on the n-type region. This recombination results in an area almost devoid of free electrons and holes, the depletion region. The depletion region is the core of the semiconductor radiation detectors (Krane,1987).

When the radiation photon or particle interact with the depletion region, the electrons–holes pairs are created, collected to the electrodes with the aid of an applied voltage. The collected electrons are amplified into electric pulses, whose amplitude is proportional to the incident photon or particle into the detector (Krane,1987). The semiconductor detectors operate at high reverse bias voltage in order to increase the volume of the depletion region, thereby enhancing detection sensitivity and increasing the electric effect required for efficient collection of charges (Krane,1987; Knoll,2010).

The semiconductor detectors made of silicon crystal are preferentially used for the charged particles and low energy gamma emitters detectors (Cember & Johnson, 2009), while germanium (Ge) with relatively high atomic number of 32 is efficient in stopping and detecting high gamma energy, hence mainly used for γ -spectrometry, specifically HPGe (Cherry *et al.*, 2012). The p-type detector is recommended for use on samples with potentially high energy photons (above 200 keV), whereas the n-type detector has been recommended for both the low and high energy gamma rays' measurements (International Organisation for Standardisation [ISO], 2007b).

2.11.2 HPGe gamma spectroscopy

Gamma spectroscopy is an analytical, non-destructive technique for the identification and quantification of gamma emitting radionuclides in samples. Identification of any radionuclide is achieved through the analysis of its specific characteristic photon energies while quantification is achieved through the analysis of the peak areas of the full - energy peaks of gamma lines. Unlike alpha spectroscopy and other radio-analytical techniques, gamma spectroscopy technique has the ability to measure simultaneously gamma emitting nuclides directly without chemical separation or with minimal sample preparation involved (IAEA, 1989).

2.11.2.1 Hyper purity germanium detection system

The HPGe detection system comprises of an integrated set of interrelated hardware, software and electronic devices working in tandem, as illustrated in Figure 2.15. The system generally consists of a semiconductor detector, made of ultrapure germanium crystals. Since germanium has low band gap, the detector is generally housed in vacuum-tight cryostat dipped in the liquid nitrogen to reduce the thermal generation of charge carriers to an acceptable level (Rittersdorf, 2007). The detector is enclosed in a thick heavy lead shield or other material to minimise effects from the ambient background.

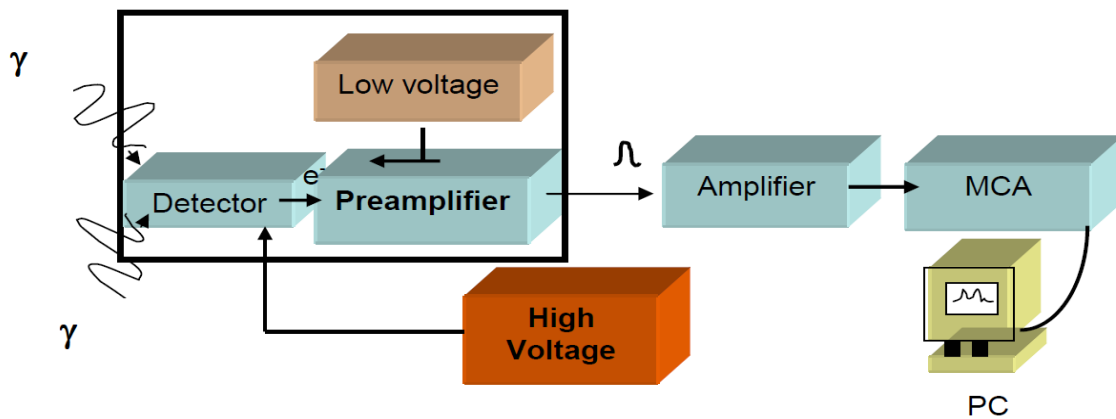


Figure 2. 15: Basic arrangement of a typical gamma spectrometry system (Reguigui, 2006).

High-voltage power / bias supply provides the electric field to the detector and the rest of the system. When radiation interacts with the detector material, electrons-holes pairs are formed. The preamplifier, which serves as an interface between the detector and amplifier, then collects the charges and convert them into pulse voltage (Gilmore, 2008).

The amplifier serves to shape the pulses and increases their signal, minimises the electronic noise. The analogue to digital convertor converts the pulse into numerical data that are fed into the multi-channel amplitude analyser (MCA). The MCA sorts the pulses by pulse height and counts the number of pulses within individual pulse height intervals (Gilmore, 2008). The personal computer (PC) processes and displays the measurement spectra. There are numerous types of commercial software packages

available in the market designed for spectral acquisition, analysis and evaluation with features for peak searching and fitting, area determination, correction for coincidences summing, decay corrections, background correction and nuclide's identification and quantification.

2.11.2.2 Calibration of the hyper purity germanium detection system

The gamma spectroscopy detection system should be calibrated both for energy response, peak energy resolution and detection efficiency for the accurate identification and quantification to be realised. Energy calibration is the process of determining the function that describes the (linear) relationship between the channels of the analyser and the energies of the known radionuclide. Generally, energy calibration is achieved by counting a multi radionuclides standard source with several gamma energy lines preferably covering the whole energy range of interest. An energy calibration curve is then derived from plotting the analyser channel numbers against the corresponding known gamma energy lines (Darwish *et al.*, 2015; Knoll, 2010). The germanium detector is characterised by its relatively high energy resolution, the ability of separation numerous closely spaced gamma-ray energies (Knoll, 2010). The energy resolution (R) is derived from the peak full width at one-half of the maximum height (FWHM). The energy resolution is mathematically expressed by equation 2.36,

$$R = \frac{FWHM}{E_0} \times 100 \quad (2.36)$$

where, E_0 , is the specified energy. For analysis, where the accuracy is of concern, ISO (2007b) recommends the use of detectors whose energy resolution is less than 2.20 keV (for the ^{60}Co peak at 1332 keV).

The factors that govern the energy resolution of a germanium detector may include the inherent statistical spread in the number of charge carriers, variations in the charge collection efficiency, and contributions of electronic noise. The dominance of whichever the factor depends on the energy of the radiation and the size and inherent quality of the detector in use (Knoll, 2010).

In order to interpret the spectrum in quantitative terms, the detection system has to be calibrated for efficiency. Detection efficiency is one of the crucial parameters in the assessment of the detector performance. There are several definitions encountered in

the field of gamma spectroscopy, however, of interest is the absolute full energy peak efficiency, which specifies the percentage of photons detected in the full-energy peak relative to the number of photons emitted (Gilmore, 2008). Efficiency calibration allows for the establishment of the detection efficiency of the detector as a function of the energy of the radiation (IAEA, 2007). Traditionally, the efficiency calibration is achieved by using a certified radionuclide standard source, having different emission lines or a mixed-radionuclides source covering energy ranges of interest.

Generally, the data needed for efficiency calibration are obtainable from the certificate supplied by the manufacturer of the certified standard source, and includes amongst others, the radionuclide, the gamma ray energies and their probability of emission, half-life of the nuclide, the radioactivity and reference date, associated uncertainty of measurements and a statement of metrological traceability. It should, however, be noted that data such as the gamma ray energies and their probability of emission or half-life of the nuclide are being reviewed and updated regularly, therefore, analysts should strive to keep abreast with new developments (Gilmore, 2008). It is, therefore, advisable for analysts to update their respective libraries supplied by the manufacturer of their detection systems, accordingly.

The detection efficiency (ϵ) at the gamma ray energy (E) is mathematically expressed by equation 2.37,

$$\epsilon_E = \frac{N_c}{A m T P_E} \quad (2.37)$$

where, N_c , A , m , T and P_E is the net-peak area (s^{-1}) of a γ -spectrum at the energy E , activity concentration of each radionuclide in the calibration source ($Bq.kg^{-1}$), mass of the source (kg), the counting time (s) and probability of the emission of gamma radiation with energy, E , for each radionuclide, per decay, respectively.

It should be noted that the detection efficiency is dependent on the size of the radioactive source, geometrical arrangement of source and detector (Gilmore, 2008) as well as on the physical and chemical properties of the source matrix and gamma radiation energy. Ideally, the sample measurement conditions should replicate the conditions for efficiency calibration, this includes similarity in the settings of electronic gains and voltage as well as between the standard used for efficiency calibration and

the sample being analysed. Satisfying these conditions, minimises the effects of summing and self-absorption of gamma radiation otherwise, correction factors should be determined and applied (ISO, 2007b).

Since primordial radionuclides are ubiquitously in all environmental media; water, soil and sediment, vegetation, etc., it has proved to be a challenge to acquire the matrix-matched standards for efficiency calibration. The situation is further exacerbated by the lack of technical capabilities at national or regional level for manufacturing efficiency calibration standards in required geometries. There are, however, several international organisations with certified and accredited management systems, specialised in manufacturing calibration sources traceable to international measurement standards (IAEA, 1989). This calls for respective laboratories to provide technical specifications such as the detector diameter and types of environmental samples anticipated, for the manufacturer to supply tailor made efficiency calibration standards. Several studies on natural radioactivity in soil and sediments relied on efficiency calibration derived from the IAEA-RGU-1, IAEA-RGTh-1 and IAEA-RGK-1 reference materials supplied by the IAEA (Alshahri, 2017; Mubarak & Hafez, 2016; Njinga & Tshivhase, 2018; Papadopoulos *et.al.*, 2013).

In addition to conventional method of detection efficiency calibration, there are several tested methods for efficiency calibration. These include the computer codes and spreadsheets-based software's designed to simulate the detector's response to radiation energy interaction (Krneta Nikolić *et.al.*, 2018). Vidmar *et al.*, (2010) evaluated the equivalence amongst the Monte Carlo codes (GEANT3, PENELOPE, MCNP and EGS4) and dedicated packages for efficiency determination in gamma-ray spectrometry (ANGLE, DETEFF, GESPECOR, ETNA and EFFTRAN) by applying them to the calculation of efficiency transfer (ET) factors for a set of well-defined sample parameters, detector parameters and energies typically encountered in environmental radioactivity measurements and there were no significant differences observed between the results of the different codes.

The dimensions of the germanium detectors are not necessarily standardized to any degree regardless of their similarities in terms of size, make and model. Therefore, the replication of efficiency calibration data set amongst germanium detectors should be discouraged since the random variability in the detector active volume and window

thickness coupled with the long-term changes in charge collection efficiency may lead to drifts in the detector efficiency over time (Knoll, 2010).

2.11.2.3 Determination of ambient background radiation for the HPGe detector

Although the detector is enclosed in the radiation shield, it is essential to determine the baseline background radiation associated with detection system. The background radiation is observed as the pulses registered by the detection system in the absence of a radiation source. The pulses could arise as a result of the interaction of the detector material and the cosmic rays, decays of primordial radionuclides and their progeny in the environment, as well as radioactive impurities in the detector and shielding material (Gilmore, 2008; Martin, 2006).

Ideally, the background measurements should be carried out using a blank standard with similar matrix as for the efficiency calibration standards and samples to be measured and under similar geometric conditions, but without any measurable amount of the radionuclide of interest present (Cember & Johnson, 2009). The background measurement is critical in determination of the minimum detectable activity (MDA). The MDA is defined as the smallest quantity of radioactivity that could be distinguished from the blank under specified conditions and is dependent on the limit of detection (L_D) and on the counting efficiency of a counting system (Cember & Johnson, 2009).

In some radioactivity measurements in sediments and soil samples studies, the background measurements were performed by using inactive deionised water as blank standard (Al-Sulaiti *et al.*, 2012; Santawamaitre *et al.*, 2011), whereas Michael *et al.*, (2010) performed measurements with an empty Marinelli beaker to determine the ambient background in the laboratory. The spectrum of the background is then used for correction of the background effect on the actual sample.

2.11.3 Inductively coupled plasma-mass spectroscopy

Since its commercialisation in the 1983s, the inductive coupled plasma-mass spectrometry (ICP-MS) has evolved rapidly and became one of the reliable techniques for the determination of a wide range of elements in a wide variety of samples. The most striking features of the ICP-MS lies on its capability to achieve extremely low detection limits, high precision and speed of analysis. It offers an advantage of

simultaneous measurement of trace elements and isotopes of an element (Barron, 2015; Beauchemin, 2017; Taylor, 2001; Thomas, 2013).

2.11.3.1 Overview of an ICP-MS system

There are several makes and designs of the ICP-MS system in the market, however, both share a similar set of integrated software and hardware components, as schematically shown in the block diagram in Figure 2.16.

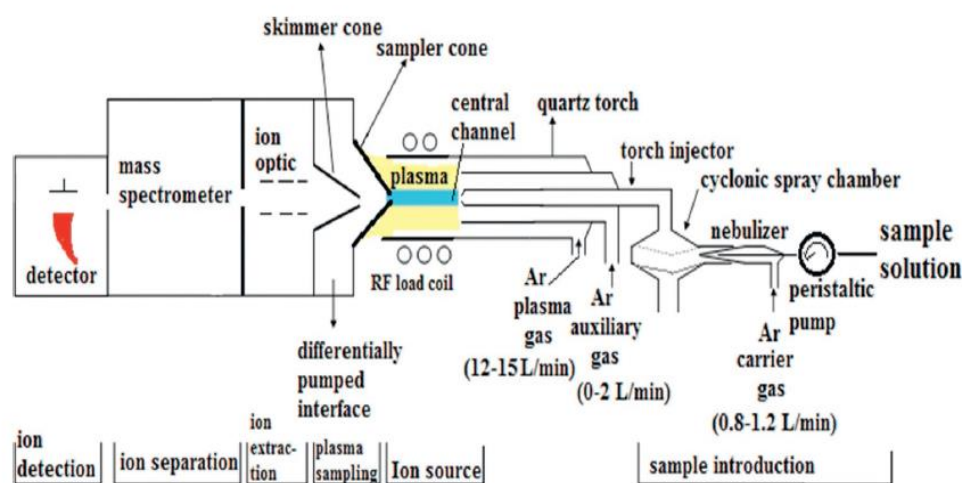


Figure 2. 16: Illustration of an ICP-MS system (Beauchemin, 2017).

As it can be seen in Figure 2.16, the main basic components of the ICP-MS comprise of the sample introduction system, plasma ion source, an ion transfer interface, a mass spectrometer and detector. The ICP-MS was primarily developed to analyse liquid samples, however, the technique has evolved and undergone adaptation to handle solids and slurries samples (Thomas, 2013). In this work, the application of ICP-MS was restricted to the analysis of samples in aqueous form.

2.11.3.2 Sample preparation

Prior to analysis, the samples are prepared to be transformed into suitable form compatible with the ICP-MS system. Sample preparation is critical for the accuracy and integrity of the generated analytical data. Hence the laboratory should maintain a high level of cleanness to avoid sample contamination, ultra-pure chemicals should be considered at all times and laboratory glassware with potential for leachable contaminants (glass and metals) should be avoided (Barron, 2015). For solid samples,

the preparation could include the grinding, sieving, weighing, liquefaction through dissolution by means of acid digestion or fusion, filtration and dilution.

2.11.3.3 Sample introduction

The aqueous sample is delivered, with a peristaltic pump into the nebuliser and spray chamber. The nebuliser converts the sample into droplets of aerosol in the stream of argon gas. The spray chamber then separates and injects the fine droplets of aerosol into the plasma torch via an injector while draining out the larger droplets of aerosol.

2.11.3.4 Plasma formation

A plasma is an electrically neutral gas, (preferably argon) high - temperature (6000 – 10 000 °K) made up of positive ions and free electrons, that can virtually atomize, ionize, and excite all elements in the periodic table. It is generated in the plasma torch from the interaction of strong magnetic field with argon gas flowing through a quartz glass torch. Applying a high voltage spark to the argon gas, causes the electrons to be stripped from their argon atoms. The electrons are accelerated in the magnetic field and collide and ionise other argon atoms and create a chain reaction that sustain the plasma (Thomas, 2013).

2.11.3.5 Ionisation

The fine droplets of aerosol from the spray chamber in the stream of argon gas is then injected into the plasma, where they can be dried, dissociated, atomized and ionized. The ions formed by the ICP discharge are typically positive ions, M^+ or M^{2+} .

2.11.3.6 Mass analysis

The ions produced in the plasma are extracted and transferred into a high vacuum mass analyser, where they are sorted on the basis of their charge to mass ratio (m/z) (Beauchemin, 2017; Taylor, 2001; Thomas, 2013).

2.11.3.7 Ions detection

The ions, typically singly charged ions (M^+), are transmitted to the detection system for conversion into an electrical signal proportional to the elemental concentration present in the sample (Barron, 2015).

2.11.3.8 Quantitative elemental analysis

The instrument is calibrated using traceable certified calibration standards of all elements of interest at a range of concentrations likely to be encountered in the unknown samples. The calibration curve of the measured intensity versus concentration for each element in the standard solutions is created, where the concentration ($\text{g}\cdot\text{unit}^{-1}$) of the elements of interest in the sample can then be derived from (Thomas, 2013).

2.12 Applications of modelling tools in health risk assessment

The fact that primordial radionuclides are long lived, dictates for a risk assessment that covers a long-term in the future. The behaviour of radionuclides in environment is influenced by competing processes and factors, therefore, poses challenges to researchers in predicting the reliable dosimetric quantities for estimation of radiological risk over time. Thus, numerous robust simulation models have been developed to aid in conducting radiological health risk assessment for complex dynamic systems. Modelling tools of interest to this study were NORMALYSA and RESRAD.

NORMALYSA is one of the recognised tools for assessment of radiological risks associated with material containing naturally occurring radionuclides and radioactively contaminated sites. The tool was developed with the support of the International Atomic Energy Agency and is freely available for its member states use. NORMALYSA comprises of a simulator program engine integrated with a set of interlinked compartmental models for simulation of environmental transport and distribution of radionuclides from the source term such as contaminated site, through exposure pathways to the receptor environment and estimation of doses to reference individuals (Avila *et al.*, 2018). The NORMALYSA environmental transport and exposure assessment models comprise of possible predefined scenarios that may require minimal adjustment to suit an individual's conditions as well as default reliable values for most parameters that are needed for running models, however, the model allows for users' site-specific parameters, where available (Avila *et al.*, 2018). The performance of NORMALYSA has been benchmarked with other models such as RESRAD-OFFSITE and was found to produce similar results (Koliabina *et al.*, 2018).

In this study, NORMALYSA was used to predict the temporal distribution of primordial radionuclides in soil and estimate the internal exposure from ingestion of food crops grown in cropland irrigated with water drawn from a borehole in cropland.

RESRAD is a computer code developed by Argonne National Laboratory under the United States' Department of Energy (U.S DoE) to serve as a multifunctional guiding tool in regulatory decisions regarding the residual radioactive material of remediated nuclear sites. Since its issuance in 1989, the code gained popularity and has been widely used by various departments of the government of the United States in the derivation of limits for radionuclides in soil at remediated nuclear sites as well as in the evaluation of associated radiation dose and health risks (Yu *et al.*, 2001). The code allows modellers to specify the site-specific soil characteristics and meteorological conditions, however, there are default values matching possible soil characteristics and environmental conditions (Yu *et al.*, 2001). The success of modelling is achieved through the formulation of an exposure scenario and the execution of a logical sequence of processes. In the current study, the code was used to estimate the cancer morbidity risks due to external exposure to radiation from the ^{238}U decay series, ^{232}Th decay series and ^{40}K for a hypothetical member's representative of a public resident of Swakop river and a maintenance worker or guard at the uranium mine's tailing facility.

CHAPTER 3: Study methods

3.1 Description of the study area

The study area is presented in Figure 3.1 with sampling points indicated as SWR, RU, KHN, RTU and SS along the Swakop river and its tributary Khan river. Swakop river and its tributaries drain a catchment area covering about 31000 km², extending from the semi-arid inland central high ground at an elevation of about 2000 m, flowing across the western Namib desert into the Atlantic Ocean, south of Swakopmund town (Greenbaum *et al.*, 2014; MET, 2002).

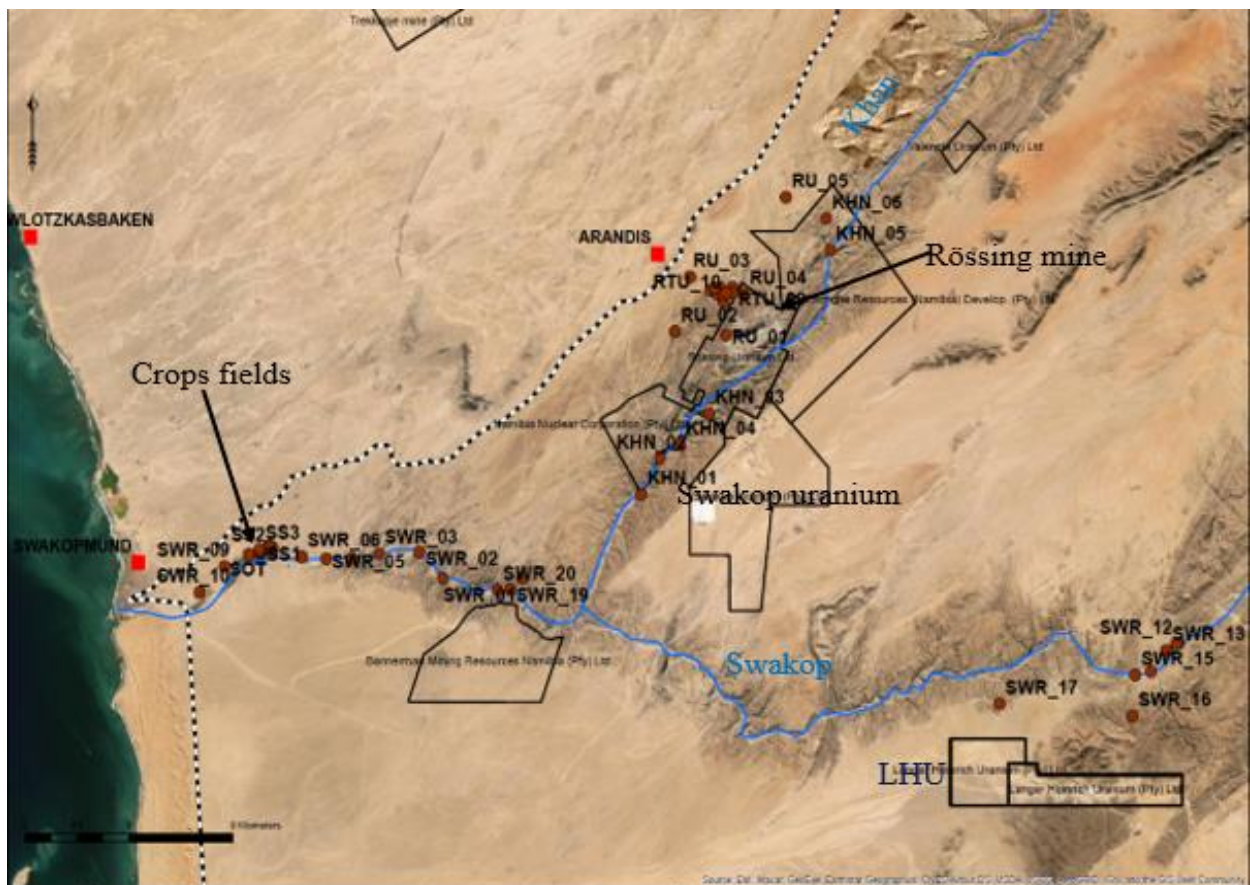


Figure 3. 1: Study area showing the sampling points.

Exploration and mining activities in the catchment area dates back as far as 1800s, however, uranium mining commenced in 1976 (Bulley, 1986). There are two operational uranium mines in the catchment of the Swakop river; Rössing uranium (RUM) and Swakop Uranium (SU) mines while Areva Trekkopje and Langer Heinrich uranium (LHU) have been placed under care & maintenance since 2013 and 2018, respectively. All uranium deposits in Namibia are mined by shallow open pit mines.

There are also numerous spots with proven uranium deposits, while uranium exploration and development activities are ongoing in the catchment area of the river.

The study area falls within the hyper arid sector of the Namib desert, with an average annual precipitation of about 50 mm (Bulley, 1986; MME, 2010), high humidity, an average of 140 days of thick fog per year and an average annual temperature of 20°C – 22°C (MET, 2002). The wind direction is predominated by Southerly and South-westerly winds during the summer, and North-easterly winds in the winter that sometimes could reach gale forces capable of moving desert surface, including tailings (MME, 2019).

The geology of the area is characterised by feldspathic quartzites, gneisses, amphibolites, meta-sediments, meta-arkoses arenite, marbles, schists, calc-silicates, alkaline volcanic and evaporites, carbonates, shales basal chuos and quartzites. The area is rich in uranium mineralization that is associated with the granitic and calcrete bodies (Brandt, 1985; Greenbaum *et al.*, 2014; MET, 2002). The alluvial aquifers of the Swakop and Khan rivers are characterised by non-homogenous compartments created by outcropping bedrock or narrowing of the river. It has been reported that these aquifers are only recharged by vertical flow rather than lateral flow, therefore, not replenished on a continual basis from upstream, but rather from occasional flood events when heavy rainfall is received in the catchment areas (MME, 2010). The latest surface water flows along the river were observed during the 2009–2011 rain seasons.

The study area lies between 22.40° S - 22 90° S and 14.40° E – 15.50° E, stretching about 80 km along the lower portion of Swakop river and about 27 km along the Khan river in the neighborhood of the RUM and SU operations. The portion of the Khan river was considered in the study due to its locality in relation to the uranium mining activities, the potential sources of environmental radioactivity contaminants. The study area is characterized by complex terrain, with alluvial sand, rocky mountainous, thick shrubs, hardly accessible, with dangerous wildlife such as cheetahs, leopards and poisonous snakes, which served as the limiting factors in the collection of samples in some parts of the study area.

The Swakop river alluvial aquifer's ground water are saline, not fit for human consumption and mainly used for irrigation purposes (MME, 2010), while potable water is supplied from other water sources by local authority and the national water utility

(Kringel *et al.*, 2010). The agricultural crops are planted in rows of soil raised beds on the bank of the river (Figure 3.2). No artificial fertilizers have been used, but guano manures (seabirds).



Figure 3. 2: Spinach and other crops on soil raised beds.

The alluvial sand mining activities and coarse aggregates for construction have been sourced from the Swakop river as early as 1890 during the formation of the Swakopmund town (Bulley, 1986).

3.2 Sample collections

The emphasis in any analytical laboratory is to produce valid and accurate analytical results. Since it is impossible to analyse infinite material of interest, the concept of sampling was introduced. Sampling is aimed at achieving a mass or volume reduction by collection of a small but representative portion (sample) of material selected from a larger quantity of material, collected for analysis while preserving complete similarity of the crucial parameters of the sample and its original material (IAEA, 2004a; Thomas, 2013).

In the case of radiometric and radiochemical analysis of environmental samples, the sample must maintain the same activity concentration of radionuclides, and the same relative abundance of their physical and chemical forms as the population from which it was taken (Macášek, 2008). This study adopted a judgemental random sampling strategy, where sampling locations were determined based on professional judgment. For this reason, the study area was subdivided into 5 segments (A-E), based on the land utilisation and/or industrial activities in the neighbourhood.

The study area A comprised of agricultural cropland on the bank of the Swakop river downstream the mining activities. The study area A is indicated by sampling points designated by SS_i and SOT in Figure 3.1 and amplified in Figure 3.3. It comprises of settlement for the Swakop river community, about 10 km from the town of Swakopmund.

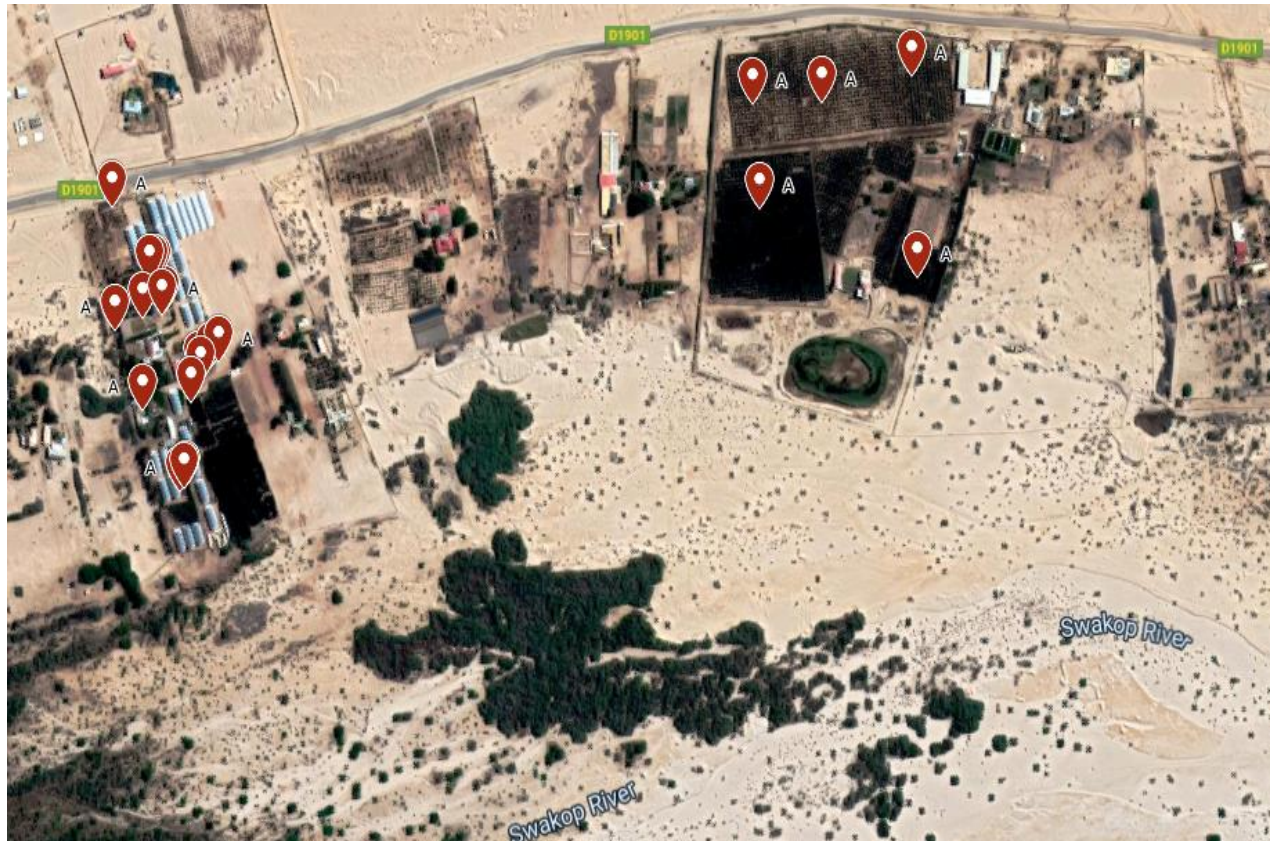


Figure 3. 3: Study area A (crop fields) in relation to the Swakop river.

With reference to Figure 3.1, the study area B stretches from the cropland, covering a length of about 80 km along the Swakop river riverbed to the vicinity of the LHU mining activities. The study area B is indicated by sampling points designated by SWR_01-SWR_20. The study area C is indicated by sampling points designated by KHN_01-KHN_06 and comprised of a portion of the Khan river riverbed, traversing between the RUM and SU mining operations, about 40 km away from the cropland. The study area D, sampling points designated by RTU_01 -RTU_10 comprised of the tailing's storage facility of the RUM mine, while area E, sampling points designated by RU_01-RU_05, covering areas within the premises of the RUM operations and the nearby surroundings, about 60 km away from the cropland.

3.2.1 Soil samples

Each sampling position was cleared of extraneous material and about 2 kg of soil sample was collected to the depth of up to 20 cm using a hand trowel. To avoid cross contamination, the trowel was wiped clean between subsequent samples (IAEA, 1989; IAEA, 2004a; ISO, 2007a).

In the study area A, a total of twenty-two samples were randomly collected from the cultivated area in the surroundings of the roots of the food crops. Additional nine soil samples were randomly collected from uncultivated open space within the cropland, where no irrigation had taken place or manure been applied. A total of twenty soil samples coded SWR_01 to SWR_20 were collected from the study area B, the dry riverbed along the Swakop river.

For the study area C, six samples (KHN_01 – KHN_06) were randomly collected along a portion of the Khan river, while ten samples were randomly collected from the tailing's storage facility (area D) and five samples were randomly collected in the surrounding of the RUM operations (area E). The samples were placed in polyethylene plastic bags, clearly marked with a marker of their identities and bags closed tight with cable ties to avoid cross contamination. All sampling positions were recorded by global positioning system (GPS) and are presented in Tables A1-A2 of the Annexure A. Samples were transported to the laboratory for further processing. Figure 3.4 shows the hand trowel and plastic bag used for soil sample collection.



Figure 3. 4: Hand trowel and polyethylene plastic bag for soil sample collection.

3.2.2 Agricultural food crops

At the time of sampling, only food crops that were ready for harvesting were sampled for evaluation and indicated in Table 3.1.

Table 3. 1: Agricultural food crops collected from the study area A.

Item No.	Species Name	Scientific name	Parts sampled	Fresh weight (kg)	No. of samples
1	Beetroot	<i>Beta vulgaris</i>	taproots	1.31	1 composite
2	Egg plant	<i>Solanum melongena L</i>	fruit	0.61 – 0.62	3
3	Spinach	<i>Spinacia oleracea L</i>	leaves	1.000 -1.01	9

About 1.31 kg (fresh weight) of beetroots were randomly collected from the raised beds and combined to constitute a representative composite sample. For the eggplant and spinach, edible parts were randomly collected from the selected raised beds with their corresponding soil samples collected from the surrounding of their respective roots. The common centre of the sampled raised bed was recorded with the GPS (Table A3 of the Annexure A). The vegetable/fruit samples were rinsed with tap water and placed in polyethylene plastic bags, clearly marked of their identities and bags closed tight to avoid cross contamination before being transported to the laboratory for further processing.

3.2.3 Water samples

In total, twenty water samples were collected directly from the boreholes in the cropland, Swakop and Khan rivers and water from the RUM tailing's storage facility in the study area. Water samples from boreholes were preceded by determination of the borehole depth and water level by an electronic dipper. Water samples were then collected at midway of the fill level by lowering a bailer on a scaled rope into the borehole. The bailer and the depth meter (dipper) were rinsed with distilled water after each borehole sampled to minimise chances of cross contamination. The water samples were collected into marked 2l polyethylene plastic containers that were rinsed with distilled water. Figure 3.5 shows some of the tools used for the collection of water samples.



Figure 3. 5: Some of the tools used for the collection of water samples.

The sampling locations (Figure 3.6) were recorded by GPS and presented in Table A4 of the Annexure A.

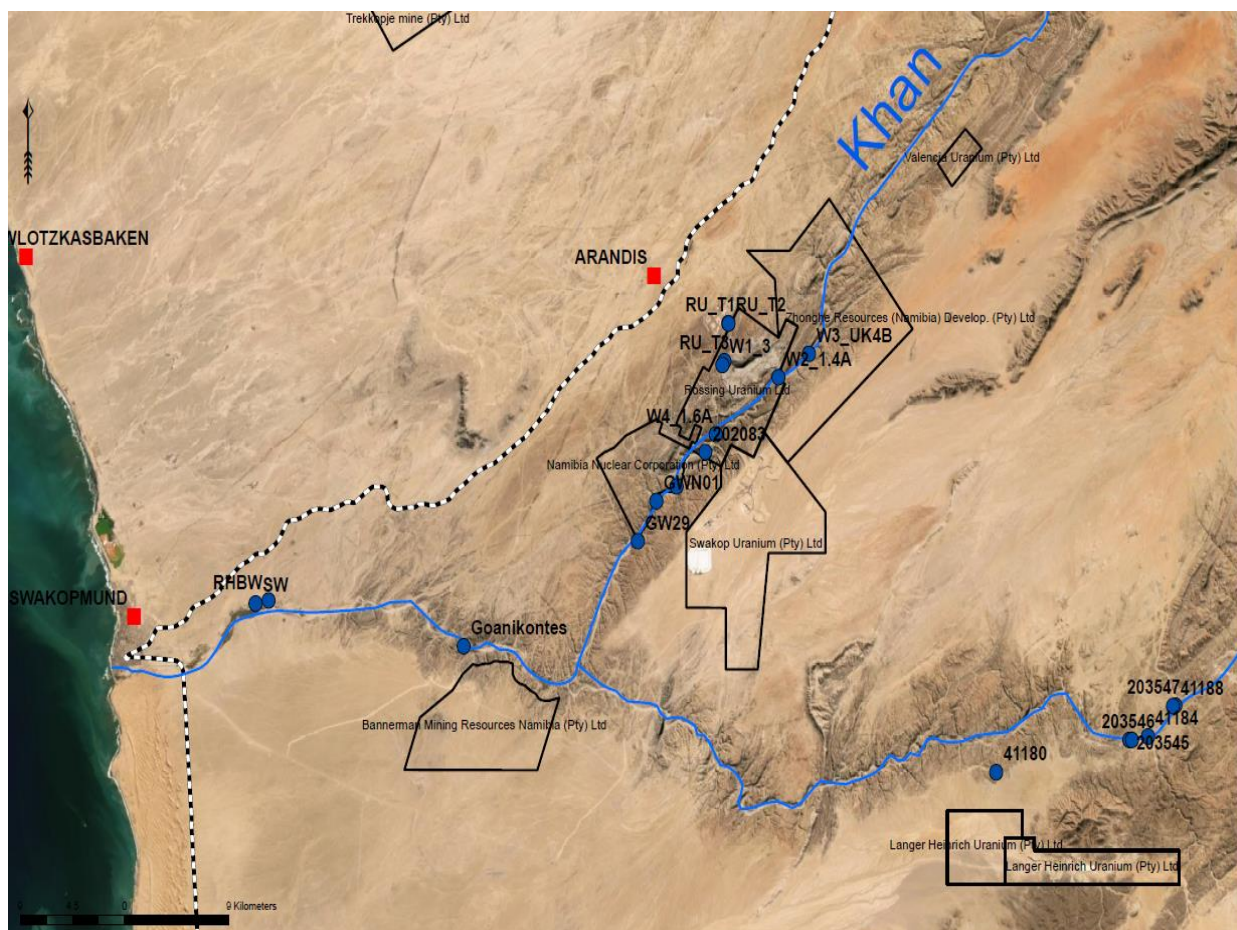


Figure 3. 6: Water sampling points along the Swakop and Khan rivers.

3.3 Samples preparation

Prior to analysis, samples have to undergo physical processing for transformation into suitable form compatible with the analytical techniques. Sample pre-treatment and preparation involves the physical processing of samples that may include drying, crushing, sieving and sealing into appropriate containers. All these steps together with the sampling planning and its execution play a critical role in the validity and accuracy of analytical results and should it not be done properly, may lead to losses of analytes of interest and introduce contamination. In our case, the laboratory glassware used in the sample preparation process were subjected to thorough cleaning by soaking in nitric acid, scrubbed and rinsed with distilled water to prevent cross contamination.

3.3.1 Soil samples

Samples were screened with radiation survey meter to assess the dose rate level. Samples were segregated according to dose rates, spread on trays lined with aluminium foils, cleared of debris and allowed to dry at ambient temperature for about

five days. The samples were then left in the low-temperature drying cabinet at 50 °C for 24 hours to ensure total removal of moisture without loss of radionuclides. Samples from cropland were then split into two sub-samples of at least 500 g. Samples earmarked for gamma spectrometry were pulverised by means of a pestle and mortar, sieved through a 2 mm mesh for homogeneity. A portion of at least 500 g was packed and sealed in a 500 ml cylindrical plastic container for at least 30 days prior to gamma counting. The holding period of at least 30 days is essential to establish a secular equilibrium between radium, radon and their progenies (IAEA, 1989). The non-pulverised or non-sieved samples were used in the determination of the physicochemical characteristics of cropland soil. Figure 3.7 presents some of the apparatus used in the sample preparation process.



Figure 3. 7: Some of the apparatus used in sample preparation (a) survey meter, (b) 2 mm mesh for sieving, mortar and pestle for pulverising of samples (c) soil sample being pulverised, (d) soil sample sealed in a 500 ml container on a weighing scale.

3.3.2 Vegetables samples

Vegetables and fruits were prepared as for cooking or consumption, whereby only the edible parts were considered for analysis. The edible parts were chopped into slices

and air dried on trays lined with aluminium foils for several days and progressively ash dried in the laboratory oven to a maximum temperature of 450 °C for 12 hours. The samples were then pulverised by means of a pestle and mortar. About 300 mg aliquots were sent to the Eco-Analytica Laboratory, North-West University, Potchefstroom for ICP-MS elemental analysis.

3.3.3 Water samples

The water samples were sent for preparation and analysis to the Eco-Analytica Laboratory, North-West University, Potchefstroom for ICP-MS elemental analysis.

3.4 Analytical methods and measurement

Noting that ^{238}U , ^{232}Th and most of their respective progenies disintegrate by alpha emissions with high emission probabilities, the alpha solid-state spectrometry would have been the primary radio-analytical technique of choice supplemented by the HPGe spectrometry. Therefore, the determination of the activity concentration in soil samples was achieved by using the HPGe detector.

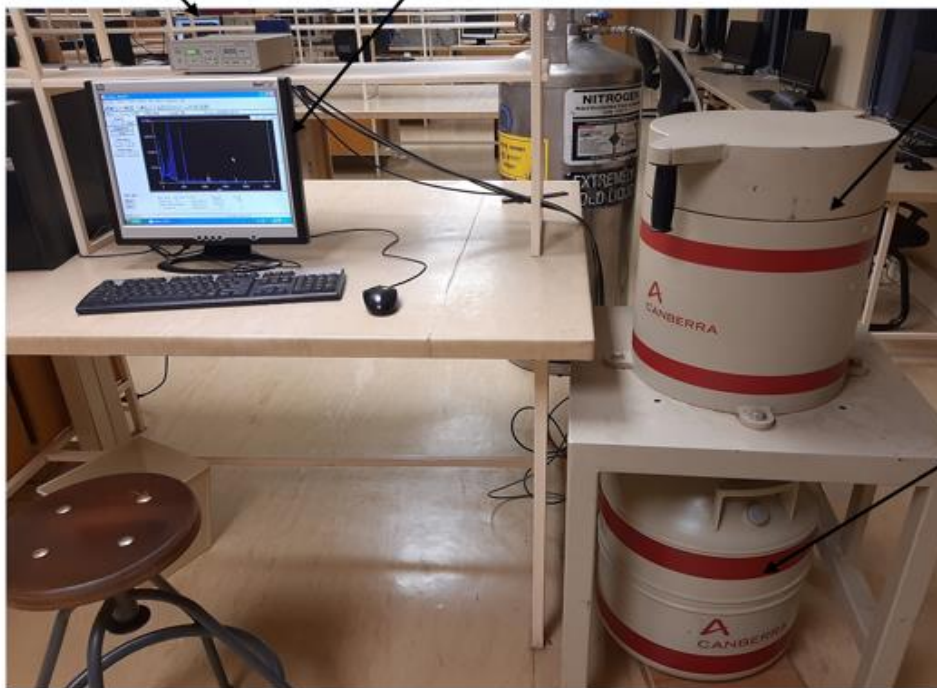
3.4.1 Measurements by HPGe

The measurement of the activity concentrations for the primordial radionuclides was achieved using the co-axial HPGe detector, Canberra Industries, model GCW 2021 (Figure 3.8) with a relative efficiency of 36.4% and an energy resolution of 1.842 keV at 1332 keV gamma ray emission of ^{60}Co . The HPGe system is coupled to a computer based digital multi-channel analyser (MCA), with Genie 2000 software program for qualitative and quantitative spectral analysis. The software performs search for statistically significant peaks, evaluation of peak position and estimation of peak area in the spectrum and assignment of peaks to nuclides based on the best match in the nuclide library. The genie 2000 has the capabilities for determination of the nuclide MDA per sample measurement, performs background analysis and subtraction from corresponding peaks of the sample spectrum and determination of activity concentration with its associated measurement uncertainty in appropriate units (Canberra Industries, 2006). The germanium detector was kept cold with a liquid nitrogen to reduce the thermal generation of charge carriers to an acceptable level. The detector and the sample to be counted were enclosed in the thick cylindrical low-level background lead lined to reduce the amount of both cosmic and terrestrial background radiation reaching the detector. On the inner surface, the lead shield is

graded with a copper liner to reduce the effects of lead X-ray fluorescence and backscatter photons from the interaction of gamma radiation with the shield wall (Canberra Industries, 2006). Before sample measurements, the HPGe detector was calibrated for energy and efficiency.

Digital Spectrum Analyser
integrated MCA

Display monitor



Lead shield

Dewar for liquid
Nitrogen

Figure 3. 8: HPGe system used in the current study, showing the MCA, Dewar for liquid nitrogen, lead shield and data display.

3.4.1.1 Energy calibration

The energy calibration was accomplished by using an ^{152}Eu standard source counted long enough to obtain appreciable counts that produced well defined photo peaks. ^{125}Eu emits numerous gamma ray energies ranging from 121.78 keV to 1769.09 keV. About 8 prominent energy peaks were identified based on their relative intensity and expected position in the spectrum and were assigned to their respective channel numbers as indicated in Table 3.2. Therefore, the first highest peak at lower energy on the spectrum was expected to correspond to energy 121.78 keV, whereas the last highest peak on the spectrum to correspond to energy 1408.01 keV.

Table 3. 2: Relationship between channel numbers and the MCA of the HPGe detector.

Channel No	E (keV)	Intensity (%)
369	121.78	28.41
742	244.69	7.55
1043	344.27	26.59
1246	411.11	2.23
2358	778.9	12.97
2919	964.07	14.50
3288	1085	10.21
3367	1112.26	13.64
4264	1408.01	20.85

The energy calibration resulted in the best fit straight-line curve (Figure 3.9) demonstrating a linearly relationship between the energy and channel number of the MCA. The linear relationship is mathematically expressed by the equation (3.1):

$$E(keV) = 0.3304Ch - 0.1466 \quad (3.1)$$

where, Ch represents the channel number.

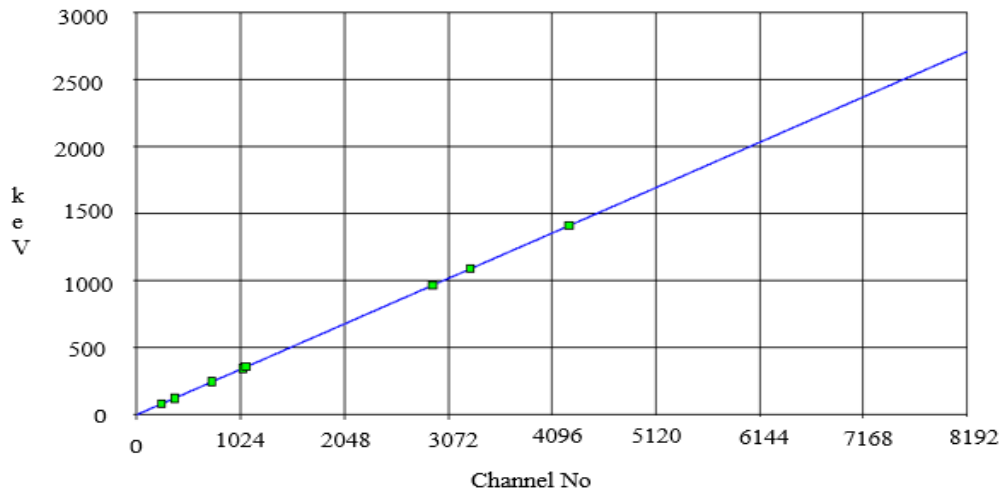


Figure 3. 9: The energy calibration curve.

3.4.1.2 Detection efficiency calibration

The detection efficiency was achieved using the certified IAEA uranium and thorium ores reference material (IAEA-RGTU-1 and IAEA-RGTh-1) in the 500 ml cylindrical

volumetric plastic container. The counting detection efficiency at the gamma ray of the energy concern was then calculated using equation 3.2,

$$\varepsilon_E = \frac{N_c}{A m T P_E} \quad (3.2)$$

where, N_c , A , m , T and P_E is the net-peak area (s^{-1}) of a γ -spectrum at the energy E , activity concentration of each radionuclide in the calibration source ($Bq.kg^{-1}$), mass of the source (kg), the counting time (s) and probability of the emission of gamma radiation with energy, E , for each radionuclide, per decay respectively (ISO, 2007b).

The calibration for detection efficiency resulted in a fourth-degree polynomial curve as illustrated in Figure 3.10 and mathematically expressed by 3.3,

$$\begin{aligned} &Ln(Eff) \\ &= -0.05511 \ln(E)^4 + 0.1442 \ln(E)^2 \\ &+ 61.82 \ln(E) - 100.90 \end{aligned} \quad (3.3)$$

where, Eff is the absolute detection efficiency and E is the gamma ray energy.

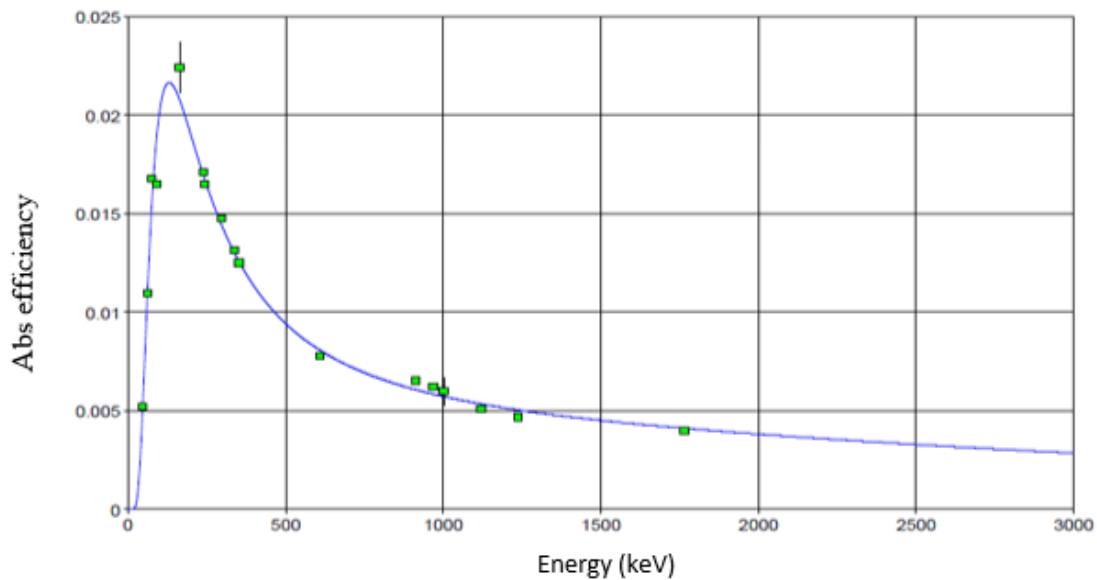


Figure 3. 10: Illustration of the absolute efficiency curve as a function of the gamma ray energy.

3.4.1.3 Background spectrum

The background level of the detection system was determined based on an empty detector and counted for 43200 seconds with no radiation source in the surrounding of the detector. The background spectrum is shown in Figure 3.11, indicating the radionuclides made up the background. The background spectrum was used to determine the MDA by using equation 3.4 proposed by Currie in 1968,

$$MDA = \frac{4.65\sigma_b}{T\varepsilon_E P_E} \quad (3.4)$$

where, σ_b , T , ε_E and P_E are the standard deviation of the background counts under the peak of interest, background counting time, detection efficiency and probability of the emission of gamma radiation at the gamma ray energy of interest, respectively (Canberra Industries, 2006). The MDA values are presented in Table 3.3.

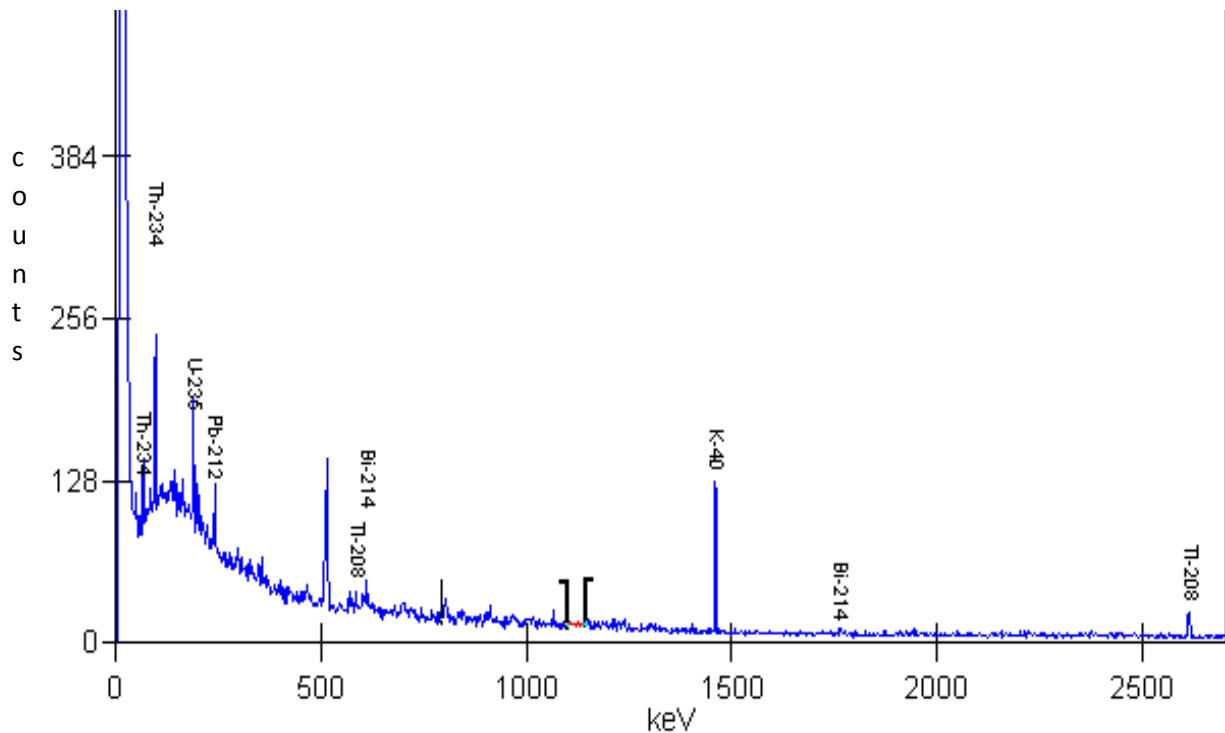


Figure 3. 11: Background spectrum measured for 43200 seconds.

Table 3. 3: Calculated values of the MDA.

Radionuclide	Measured daughter	Energy (keV)	Background counts (counts)	Detection limit (counts)	MDA (Bq)
^{238}U	^{234}Th	63.29	354	87.48	4.30
		92.59	497	103.67	2.60
	^{228}Ac	911.20	97.68	38.29	0.53
	^{212}Pb	238.72	144	55.80	0.18
^{40}K		1460.82	709	123.81	5.88
^{226}Ra	^{214}Pb	295.22	36.20	27.98	0.24
	^{214}Bi	609.32	129	52.81	0.34
		1764.54	59.9	35.99	1.34
^{210}Pb		46.54	30.00	25.49	2.67

3.4.1.4 Measurements and determination of activity concentration for soil samples

The soil sample measurements were carried out under similar conditions as that for the counting efficiency calibration and background determination. The generated respective soil spectra were evaluated based on the range of gamma ray energies characteristics of individual radionuclides of interest as shown in Table 3.3. The gamma ray energies characteristic of ^{235}U decay were ignored. Figure 3.12. is a typical spectrum showing identified peaks and their respective radionuclides present in the soil sample SWR_20. In some samples, several radionuclides could not be detected at some energies (^{210}Pb at 46,54 keV, ^{234}Th at 63,20 and 92,60 keV, $^{234\text{m}}\text{Pa}$ at 1001,03 keV, ^{212}Bi at 1620,74 keV) therefore, denoted as being below the detectable limit (BDL) in the activity concentrations tables.

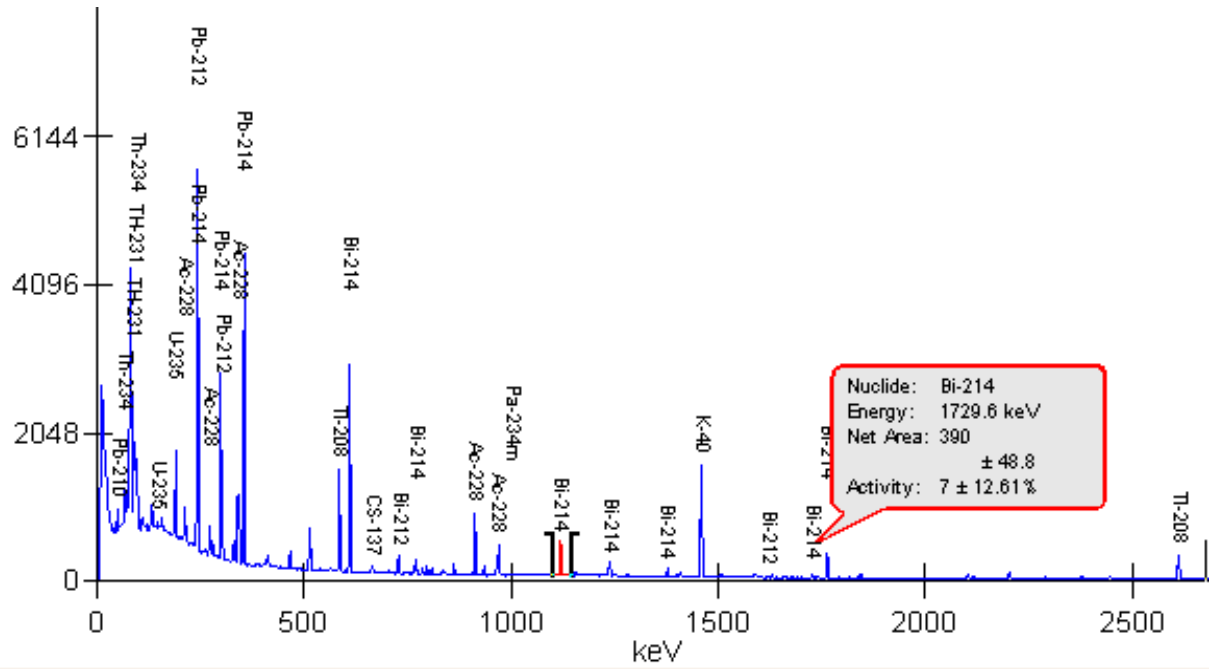


Figure 3. 12: Spectrum analysis for sample SWR_20.

The activity concentration for individual radionuclide at a specified energy of interest was determined using the equation 3.5,

$$A = \frac{N_c}{P_E \epsilon_E m T} \quad (3.5)$$

where, N_c , is given by equation 3.6 and corresponding to the net peak area or counts corrected for background effects.

$$N_c = N_s - N_b \quad (3.6)$$

N_s = gross counts under the peak area of interest in the sample spectrum (cps)

N_b = corresponding background counts under the peak area of interest in the (cps)

P_E = the gamma ray emission probability corresponding to the peak energy of interest

ϵ_E = the counting efficiency at the photo peak of interest

m = mass (or volume) of the measured sample (kg) or (l)

T = live time of counting in seconds

The activity concentration of ^{238}U , ^{232}Th and ^{226}Ra were determined as the weighted averages based on the normalized activity of individual peaks of their respective daughter products using equation 3.7. The activity concentration of ^{238}U was

determined as the weighted average of activity concentrations of ^{234}Th using gamma ray energies 63.280 keV (4.10%), 92.600 keV (4.81%) and of $^{234\text{m}}\text{Pa}$ 1001.03 (0.839%). ^{226}Ra decays to ^{222}Rn by emitting alpha particles accompanied by gamma ray emission at energy 186.1 keV (3.6%), which overlaps with gamma radiation at energy 185.72 keV from ^{235}U with a relative high emission probability (57.2%). Assuming the radioactive equilibrium has been achieved between ^{226}Ra and its daughters, the activity concentration of ^{226}Ra was determined as the weighted average of activity concentrations of ^{214}Pb using gamma ray energy at 295.22 keV (18.15%) and at energy 351.95 keV (35.10%) and of ^{214}Bi at 609.31 keV (44.60%), 1120.29 keV (14.70%) and 1764.49 keV (15.10%). Similarly, the activity concentration of ^{232}Th was calculated as the weighted average of the activity concentrations of; ^{228}Ac at energies 338.32 (11.27%) and 911.20 (25.80%), ^{212}Pb at 238.63 (43.30%) ^{212}Bi at 727.33 (30.40%) and 1620.74 (1.51%). The activity concentration of ^{210}Pb and ^{40}K were determined directly from their respective γ -ray energy transition at 46.5 keV and 1460.82 keV.

$$A_{Av} = \frac{\sum_{i=1}^N \frac{A_i}{\sigma_{A_i}^2}}{\sum_{i=1}^N \frac{1}{\sigma_{A_i}^2}} \quad (3.7)$$

where, N , A_i and σ_{A_i} are number of nuclide energies included in the weighted average calculations, the activity of the i^{th} nuclide at specified energy and the uncertainty of the individual peak measurements A_i , respectively (Canberra Industries, 2006).

3.4.1.5 Estimation of uncertainty for the activity concentration

The uncertainty of the weighted average activity (σ_{A_i}) from multiple selected γ -lines was calculated using equation 3.8,

$$\sigma_{A_i} = \sqrt{\frac{1}{\sum_{i=1}^N \frac{1}{\sigma_{A_i}^2}}} \quad (3.8)$$

where, σ_{A_i} is the uncertainty activity concentration A_i (Canberra Industries, 2006).

3.4.1.6 Determination of activity concentration ratios for primordial radionuclides for soil samples taken from the study areas

In a natural undisturbed isotopic system, the activity concentration of ^{226}Ra should be approaching or is in secular equilibrium with ^{238}U since they belong to the same decay series (Eisenbud & Gesell, 1997), while the average activity concentration ratio of

$^{232}\text{Th}/^{238}\text{U}$ in the continental earth crust lies close to 1,20 (IAEA, 2014b). The ^{232}Th decay series is known to exist in secular equilibrium in nature due to the relatively short half-lives of its decay products (IAEA, 2003). In order to ascertain the state of equilibrium between ^{238}U and ^{226}Ra , the activity concentration ratios of $^{238}\text{U}/^{226}\text{Ra}$, for all soil and tailings samples were determined.

3.4.2 Measurements by inductive coupled plasma-mass spectrometry

The water and vegetables samples were analysed by ICP-MS at the Eco-Analytica Laboratory, North-West University, Potchefstroom. The ICP-MS was operated on the collision reaction cell mode to eliminate the possibility of polyatomic interferences, especially with the quantification of elemental potassium.

3.4.2.1 Derivation of activity concentration for ^{238}U , ^{232}Th and ^{40}K in water and food crops

The activity concentration (Bq.kg^{-1} or Bq.l^{-1}) for ^{238}U , ^{232}Th and ^{40}K in samples were derived from the relation of atoms associated with the elemental mass concentration of the isotope (mg.kg^{-1} or mg.l^{-1}), taking into account the half-life of an isotope and its isotopic natural abundance. The formula for deriving activity concentration is given by equation 3.9,

$$A_i = \frac{\ln(2)}{T_{\frac{1}{2}}} \frac{\rho_i m_e}{M_i} N \quad (3.9)$$

where, A_i , $T_{\frac{1}{2}}$, m_e , ρ_i , M_i , and N , are the radioactivity concentration (Bq.kg^{-1} or Bq.l^{-1}) half-life time (s) of an isotope i , isotopic natural abundance of an isotope i , mass concentration of element e corresponding to isotope i (mg.kg^{-1} , mg.l^{-1}), atomic molar mass of an elemental isotope (g.mol^{-1}), and Avogadro's number (6.023×10^{23} atoms mol^{-1}) (Han& Park, 2018; Lariviere, *et al.*, 2006). Table 3.3 presents the isotopic nuclides parameters used in the determination of activity concentration.

Table 3. 4: Isotopic nuclides parameters used in the determination of activity concentration.

Isotope	Atomic molar mass number (g.mol^{-1})	Half-life (seconds)	Isotopic natural abundance %
^{238}U	238	1.41E+17	99.27
^{232}Th	232	4.43E+17	99.99
^{40}K	40	3.95E+16	0.012

3.4.3 Determination of physicochemical characteristics of the agricultural soil

The samples were sent to the Eco-Analytica Laboratory, North-West University, Potchefstroom for the determination of soil texture, pH, cations exchange capacity (CEC) and the anions PO_4^{3-} , SO_4^{2-} , NO_3^- , Cl^- , and HCO_3^- .

3.5 Determination of transfer factors of ^{238}U , ^{232}Th and ^{40}K from soil to food crops

The TF of a primordial radionuclide was calculated as the ratio of the activity concentration of a primordial radionuclide in the edible parts of food crops (in Bq.kg^{-1} dry weight) to the activity concentration of the same primordial radionuclide in the corresponding soil collected from the surrounding of the root zone of the crop (in Bq.kg^{-1} dry weight) according to the equation 3.10 (Tack, 2010):

$$TF = \frac{\text{Activity concentration of a radionuclide in dry crops (Bq.kg}^{-1}\text{)}}{\text{Activity concentration of a radionuclide in dry soil (Bq.kg}^{-1}\text{)}} \quad (3.10)$$

3.6 Radiological health risks assessment

The exposure scenarios considered were the external exposure by gamma rays associated with the activity concentration of ^{238}U series, ^{232}Th series and ^{40}K in soil and internal exposure due to ingestion of agricultural food crops containing primordial radionuclides. Due to limited resources the assessment has excluded the exposure to radon concentration in air, inhalation of radioactive dust and ingestion of soil. The activity concentrations data were subjected to software models to predict the trends of ^{238}U series, ^{232}Th series and ^{40}K , and evaluating radiation dose and cancer morbidity risks over time.

3.6.1 Absorbed gamma ray dose rate in air

The absorbed gamma dose rate at 1 meter above the ground was related to the activity concentrations of ^{238}U , ^{232}Th and ^{40}K in soil by the standardised equation 3.11 (Al-Sulaiti *et al.*, 2012; UNSCEAR, 2000),

$$D (\text{nGy.h}^{-1}) = 0.462A_U + 0.604A_{Th} + 0.0417A_K \quad (3.11)$$

where, A_U , A_{Th} , and A_K are the average activity concentrations (Bq.kg^{-1}) of ^{238}U , ^{232}Th and ^{40}K in a sample, while 0.462, 0.604 and 0.0417 are dose coefficient estimates (nGy.h^{-1} per Bq.kg^{-1}) as republished in the UNSCEAR (2010), for conversion of activity concentration to gamma dose rate.

3.6.2 Estimation of the annual effective dose due to external gamma ray exposure

The annual effective dose (AED) (mSv.y⁻¹) due to external gamma ray exposure was estimated as a sum of the effective dose from the outdoor and indoor exposure, using the equation 3.12.

$$AED \text{ (mSv.y}^{-1}\text{)} = \sum D \text{ (nGy.h}^{-1}\text{)} \times 8760h \times F \times DCF \text{ (Sv.Gy}^{-1}\text{)} \times 10^{-6} \quad (3.12)$$

where, $8760h$, F and DCF are the time in hours for one-year, fractional time spent outdoor and indoor (occupancy factor) and dose conversion coefficients for the corresponding age category, respectively (UNSCEAR, 2000). The standardised occupancy factor of 0.2 for outdoor and 0.8 for indoor as well as age-dependent dose conversion coefficients of 0.9, 0.8 and 0.7 Sv.Gy⁻¹, for the age categories of 0-5 years (infant), 6-15 years (child) and ≥ 16 years of age (adult), respectively as published in the UNSCEAR (2000) were used.

3.6.3 Committed effective dose due to ingestion of agricultural food crops contain radionuclides

Committed dose measures the probability of stochastic health risks due to ingestion of radioactive material. The estimate for committed dose due to ingestion (E_{ing}) (mSv.y⁻¹) was derived as a product of the estimated activity concentration of the ²³⁸U and ²³²Th decay series and ⁴⁰K in food crops, taking into account the estimated consumption rate of food crop types and the effective dose coefficients (e_r) (UNSCEAR, 2014) using the equation (3.13) by Van *et al.*, (2019).

$$E_{ing} = \sum_i A_{r,i} * C_i * e_r \quad (3.13)$$

where, i , $A_{r,i}$, C_i and e_r denotes the food crops (spinach, beetroots and eggplant), average activity concentrations of a radionuclide r in the i food crops (Bq.kg⁻¹), age-dependent consumption rate of i food crops (kg.y⁻¹) and the age-dependent dose coefficient for effective dose by ingestion corresponding to the radionuclide r (Sv.Bq⁻¹), respectively. In the absence of formal documented data on local consumption rates of food crops, the age dependent consumption rates published by UNSCEAR (2000) were considered for the estimation of the committed effective dose (Table 3.5).

Table 3. 5: Crop types and consumption rate for the age category representative of an infant, child and adult member of the population.

Crop types	Consumption rates (kg. y ⁻¹)		
	infant	child	adult
Leafy vegetables (spinach)	20	40	60
Non-leafy vegetables (eggplant)	35	59	70
Roots (Beetroot)	60	110	170

The dose coefficient for effective dose by ingestion (Sv.Bq⁻¹) used are those published by ICRP (2012b) as presented in Table 3.6.

Table 3. 6: Effective dose coefficients (Sv.Bq⁻¹) for the age category representative of an infant, child and adult member of the population.

Radionuclide	Effective dose coefficients (Sv. Bq ⁻¹)		
	infant	child	adult
²³⁸ U	8.0 x 10 ⁻⁸	6.7 x 10 ⁻⁸	4.50 x 10 ⁻⁸
²³² Th	3.5 x 10 ⁻⁷	2.5 x 10 ⁻⁷	2.30 x 10 ⁻⁷
⁴⁰ K	2.1 x 10 ⁻⁸	7.6 x 10 ⁻⁹	6.20x10 ⁻⁹

According to ICRP (2012b), the development of the effective dose coefficients considers the factors such as distribution of radionuclides in the body, tissue retention and the rate of excretion.

3.6.4 Total annual effective dose

The total annual effective dose, E_T , (mSv.y⁻¹) to members of the public was obtained from the summation of the effective doses from external exposures due to gamma radiation arising from ²³⁸U, ²³²Th and ⁴⁰K and internal exposures due to the intake of activity concentrations of ²³⁸U, ²³²Th and ⁴⁰K in food crops (UNSCEAR, 2000), and expressed as in equation 3.14;

$$E_T = AED + E_{ing} \quad (3.14)$$

3.6.5 Radium equivalent activity

Radium equivalent activity (Ra_{eq}) was determined using equation 3.16,

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (3.16)$$

where, A_{Ra} , A_{Th} and A_K , is the activity concentration in $Bq.kg^{-1}$ for ^{226}Ra , ^{232}Th , and ^{40}K , respectively (Beretka & Mathew, 1985). UNSCEAR (2000) recommended for the maximum permissible value of the Ra_{eq} to be less than or equal to $370 Bq.kg^{-1}$, for the effective dose to the public to be restricted within $1 mSv.y^{-1}$.

3.6.6 Modelling the distribution of residual radioactivity in soil and estimation of committed dose due to ingestion of food using the NORMALYSA

Assumption was made that the observed average activity concentrations of ^{238}U , ^{232}Th and ^{40}K in root zone of a cropland represented the residual activity since the formation of the soil. The NORMALYSA model was used to estimate the temporal redistribution of primordial radionuclides in the topsoil for a period of 500 years and to estimate the resulting potential total committed dose due to food crop consumption by the community residing in the area. The modelled scenario was based on the $10000 m^2$ portion of the cropland, where the community grow agricultural crops in soil irrigated with water drawn from a borehole in the cropland. The exposure pathway considered included the direct exposure to external gamma radiation from the activity concentrations of ^{238}U , ^{232}Th and ^{40}K in the soil and internal exposure from ingestion of food crops. The crucial parameters that were input into the model are presented in Tables 3.7 to 3.9 and include the site-specific estimated average activity concentrations of ^{238}U , ^{232}Th and ^{40}K in the soil and irrigation water and activity concentration ratios (transfer factors) that were determined in section 4.2.1.1, 4.2.31 and 4.3, respectively. Other parameters were selected from the IAEA reports and default values of the model as well as literature based on the physicochemical characteristics of the soil.

Table 3. 7: Parameters of the portion for the cropland.

Parameter	Value	Reference
Area of cropland under modelling (m ²)	10000	Current study
Depth of roots zone (m)	0.2	Current study
Density of topsoil (kg.m ⁻³)	1500	NORMALYSA default value
Density of deep soil layer (kg.m ⁻³)	1700	NORMALYSA default value
Porosity of the topsoil layer	0.35	NORMALYSA default value
Porosity of the deep soil layer	0.25	NORMALYSA default value
Infiltration rate to soil (m.year ⁻¹)	0.2	NORMALYSA default value

Table 3. 8: Radionuclides specific parameters for the cropland.

Radionuclide	Average activity concentration		K _d for sand soil and pH>7, (m ³ .kg ⁻¹)	TF		
	Soil (Bq.kg ⁻¹)	Water (Bq.l ⁻¹)		Leafy crops (spinach)	Root crops (beet root)	Non-leafy crops (eggplant)
²³⁸ U	71.87	0.640	0.055	0.014	0.004	0.0035
²³² Th	52.63	BDL	60	0.02	0.01	0.003
⁴⁰ K	568.73	1.943	0.05	2.77	0.98	1.76

where the average activity concentrations of ²³⁸U, ²³²Th and ⁴⁰K in soil and water as well as the TF were determined from the current study, while K_d, the distribution coefficient values were sourced from RESRAD software applicable to a soil with pH > 7.

Table 3. 9: Habits of reference persons for the cropland.

Parameter	Unit	Adult	Child	infant	Reference
Ingestion of leafy crops	kg. year ⁻¹	60	40	20	UNSCEAR, 2000
Ingestion of roots crops	kg. year ⁻¹	170	110	60	UNSCEAR, 2000
Ingestion of non-leafy crops	kg. year ⁻¹	70	59	35	UNSCEAR, 2000

3.6.7 Estimation of radiogenic lifetime cancer morbidity risk using the RESRAD-ONSITE computer code

The RESRAD ONSITE version 7.2 was used on the estimation of cancer morbidity risks due to external exposure to ^{238}U , ^{232}Th and ^{40}K in soil for the study areas A and D, where the Swakop communities reside and RUM workers spend an appreciable amount of time, respectively. The average activity concentrations of ^{210}Pb , ^{226}Ra , ^{238}U , ^{232}Th and ^{40}K in soil obtained from the study areas A and D served as input into the model. The lifetime cancer morbidity risks were estimated based on equation (3.15),

$$\text{LCR} = A_r * ED * R_r \quad (3.15)$$

where, A_r , ED and R_r are the average time integrated activity concentration of a radionuclide (Bq.g^{-1}) in soil, exposure duration (years) and cancer risk coefficient specific to the radionuclide, the environmental medium and the mode of exposure (risk.year^{-1} per Bq.g^{-1}), respectively (Eckerman *et al.*, 1999). The radionuclide transformation was based on the ICRP 107 library, whereas the risk coefficients were based on the Department of Energy Standard 1196 -2011 (reference person), both integrated in the RESRAD code (Yu *et al.*, 2001).

Two scenarios were considered in this study as follows; i). a resident of the Swakop river (study area A), who spent about 8 hours a day outdoor in the cropland and the rest of the time indoors, and ii). a maintenance worker, who spend 8 hours a day at the uranium mine's tailings storage facility (Study area D). In both scenarios, the exposure pathway considered were restricted to the external exposure due to activity concentrations of the primordial radionuclides in soil. The site-specific characteristics and parameters, where available, have been used as input into the model, or else suitable default values from the model or literatures have been used in the estimation of cancer morbidity risks for the cropland and tailing's storage facility and are presented in Tables 3.10 and 3.11, respectively.

Table 3. 10: The characteristics and input values for the cropland.

Parameters		Site specific data	Default data	Reference
Contaminated zone:				
Area (m ²)		10 000		
Thickness (m)			1	Yu <i>et al.</i> , 2001
Density			1.55	Yu <i>et al.</i> , 2001
Erosion rate (m.y ⁻¹)			2	Yu <i>et al.</i> , 2001
Radionuclide:				
Soil activity concentration (Bq.g ⁻¹)	²³⁸ U	0.07187		
	²³² Th	0.06267		
	⁴⁰ K	0.56873		
Distribution coefficient (cm ³ .g ⁻¹)	²³⁸ U		50	Yu <i>et al.</i> , 2001
	²³² Th		60000	Yu <i>et al.</i> , 2001
	⁴⁰ K		5.5	Yu <i>et al.</i> , 2001
Exposure duration (yrs.)		30		

Table 3. 11: The characteristics and input values for the tailing's storage facility.

Parameters		Site specific data	default data	Reference
Contaminated zone:				
Area (m ²)		750 0000		RUL (2019)
Height of the tailings (m)		100		RUL (2019)
Density			1.55	Yu <i>et al.</i> , 2001
Erosion rate (m.y ⁻¹)			2	Yu <i>et al.</i> , 2001
Radionuclide:				
Soil activity concentration (Bq.g ⁻¹)	²³⁸ U	0.53		Current study
	²³² Th	0.15		Current study
	⁴⁰ K	0.98		Current study
	²¹⁰ Pb	2.95		Current study
	²²⁶ Ra	3.07		Current study
Distribution coefficient (cm ³ .g ⁻¹)	²³⁸ U		50	Yu <i>et al.</i> , 2001
	²³² Th		60000	Yu <i>et al.</i> , 2001
	⁴⁰ K		5.5	Yu <i>et al.</i> , 2001
	²¹⁰ Pb		100	Yu <i>et al.</i> , 2001
	²²⁶ Ra		70	Yu <i>et al.</i> , 2001
Precipitation		50 mm		MME,2010
Exposure duration (yr.)			30	

Chapter 4: Results and discussion

4.1 Activity concentrations of primordial radionuclides in soil, groundwater and agricultural crops from the study area

The study area A comprised of settlement for the Swakop river community about 70 km away from the uranium mining sites. The study area B stretches from the cropland, covering a length of about 80 km along the Swakop river riverbed to the vicinity of the LHU mining activities. The study area C comprised of a portion of the Khan river riverbed, traversing between the RUM and SU mining operations, about 40 km away from the cropland. The study area D comprised of the tailing's storage facility of the RUM mine, while area E covering areas within the premises of the RUM operations and the nearby surroundings, about 60 km away from the cropland.

4.1.1 Activity concentrations of ^{226}Ra , ^{238}U , ^{232}Th , ^{40}K and ^{210}Pb in soil samples

The distribution for activity concentrations of ^{226}Ra , ^{238}U , ^{232}Th , ^{40}K and ^{210}Pb for all samples from the study area A (cropland) are illustrated in Figure 4.1, while the actual results are presented in Table B1 of the Annexure B. The activity concentrations of ^{226}Ra were found to range from 24.08 ± 0.85 to 54.06 ± 0.74 Bq.kg⁻¹ with an average value of 36.18 ± 1.30 Bq.kg⁻¹. The activity concentrations of ^{238}U varied from 35.38 ± 5.96 to 127.70 ± 5.80 Bq.kg⁻¹ with an average value of 71.87 ± 4.85 Bq.kg⁻¹ while that of ^{210}Pb ranged from <2.67 to 137.20 ± 36.48 with an average value of 62.67 ± 6.74 Bq.kg⁻¹. The average activity concentration of ^{238}U was about twice the global average activity concentration in soil as documented by the UNSCEAR (2000), however, within the global average range. The activity concentrations of ^{232}Th was found to range from 28.16 ± 0.43 to 109.97 ± 1.30 Bq.kg⁻¹ with an average value of 52.64 ± 3.41 Bq.kg⁻¹. The activity concentrations of ^{40}K were relatively high than those of ^{226}Ra , ^{238}U , ^{232}Th and ^{210}Pb in all sampling points, and varied from 282.20 ± 28.44 to 723.90 ± 13.81 Bq.kg⁻¹, with an average of 568.73 ± 19.55 Bq.kg⁻¹. Fertilizer is known to be an effective source of potassium, however, no samples were collected from manure applied in cropland for radioactivity analysis (Buliga, 2010). The average activity concentrations of ^{232}Th and ^{40}K in the study area A were higher than the global average in soil reported by the UNSCEAR (2000) report. The trend observed in the study area A indicated that the activity concentrations of ^{238}U were found to be greater than those of ^{232}Th in most of the samples, so as the average activity concentration, hence in contrary to the notion

that the average activity concentration of ^{232}Th in soil is greater than that of ^{238}U as per UNSCEAR (2000). There were no significant differences observed on the activity concentrations of ^{226}Ra , ^{238}U , ^{232}Th , ^{210}Pb and ^{40}K between samples collected from the cultivated and uncultivated areas of the cropland.

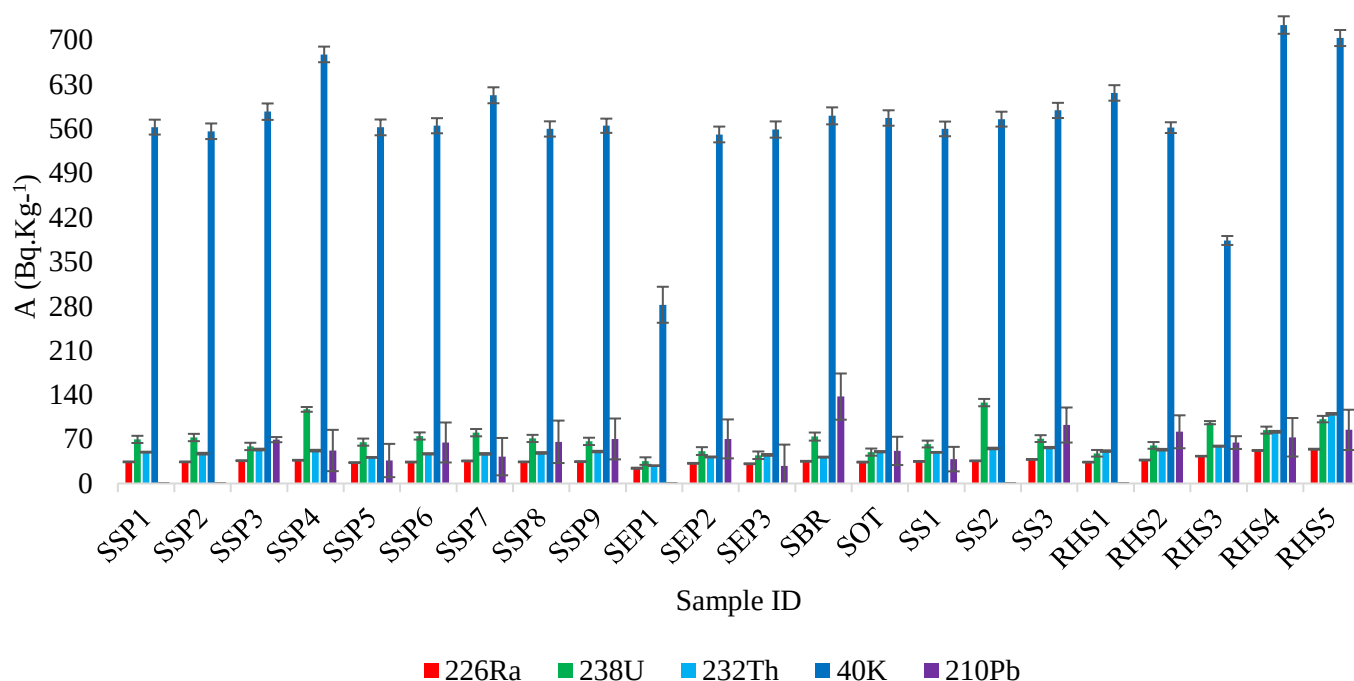


Figure 4. 1: Distribution of activity concentrations of ^{226}Ra , ^{238}U , ^{232}Th , ^{210}Pb and ^{40}K in the study area A (cropland).

The activity concentrations of ^{226}Ra , ^{238}U , ^{232}Th , ^{210}Pb and ^{40}K for the study area B are illustrated in Figures 4.2 and 4.3, respectively and detailed data in Table B2 of the Annexure B. The measured activity concentrations for ^{226}Ra were found to range from 15.50 ± 0.63 to 182.83 ± 1.48 Bq.kg⁻¹ with an average of 48.32 ± 9.25 Bq.kg⁻¹ while that of ^{238}U varied from 31.72 ± 1.61 to 168.51 ± 7.23 Bq.kg⁻¹ with an average value of 69.19 ± 8.06 Bq.kg⁻¹. The average activity concentration of ^{238}U was twice that of the global average reported in the UNSCEAR (2000) report, however, within the global average range. The activity concentrations of ^{232}Th varied from 23.45 ± 0.81 to 146.35 ± 1.41 Bq.kg⁻¹ with an average of 67.52 ± 8.77 Bq.kg⁻¹, higher than the global average, while for ^{210}Pb , they varied from <2.67 to 352.50 ± 64.88 Bq.kg⁻¹ with an average of 107.56 ± 30.80 Bq.kg⁻¹.

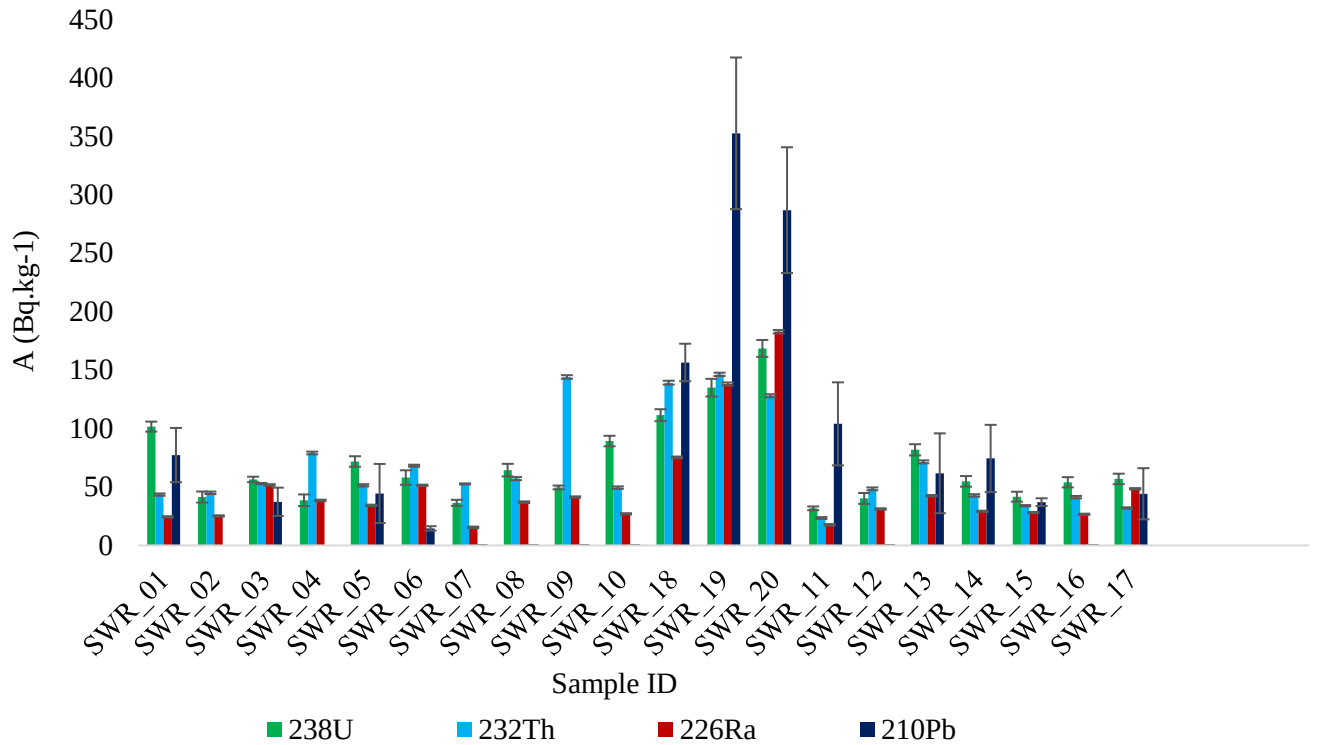


Figure 4. 2: Distribution of ²²⁶Ra, ²³⁸U, ²³²Th and ²¹⁰Pb along the lower Swakop river.

The striking feature in Figure 4.2 are the spikes observed in the activity concentrations for samples SWR_18 to SWR_20 that were up to tenfold higher than the values observed at the other sampling locations. There has been extensive mineral exploration and drilling activities in the area under the exclusive prospecting licence (EPL) 3345 (MME, 2010). The anomaly observed on the activity concentrations for samples SWR_18 to SWR_20 might be as a result of contamination of the surface soil with mineralized uranium ore brought onto the surface during the prospecting and mineral exploration activities. The activity concentrations of ⁴⁰K were found dominating in all samples, varied from 393.90 ± 8.89 to 1003.00 ± 15.82 Bq.kg⁻¹, with an average of 676.10 ± 39.41 Bq.kg⁻¹ (Figure 4.3).

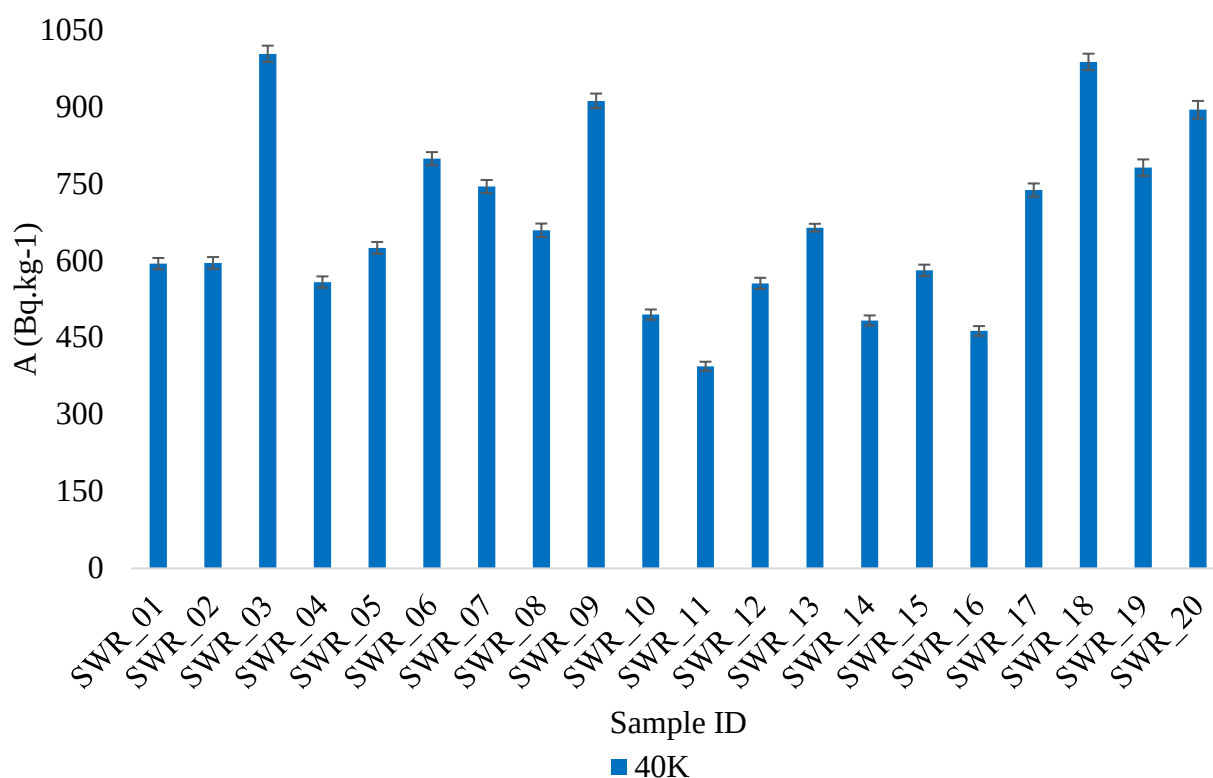


Figure 4. 3: Distribution ^{40}K along the lower Swakop river.

The distribution for the activity concentrations of ^{226}Ra , ^{238}U , ^{232}Th , ^{40}K and ^{210}Pb for samples taken from study areas C and E are illustrated in Figure 4.4, while the detailed data are presented in Tables B3 and B5 of the annexures, respectively.

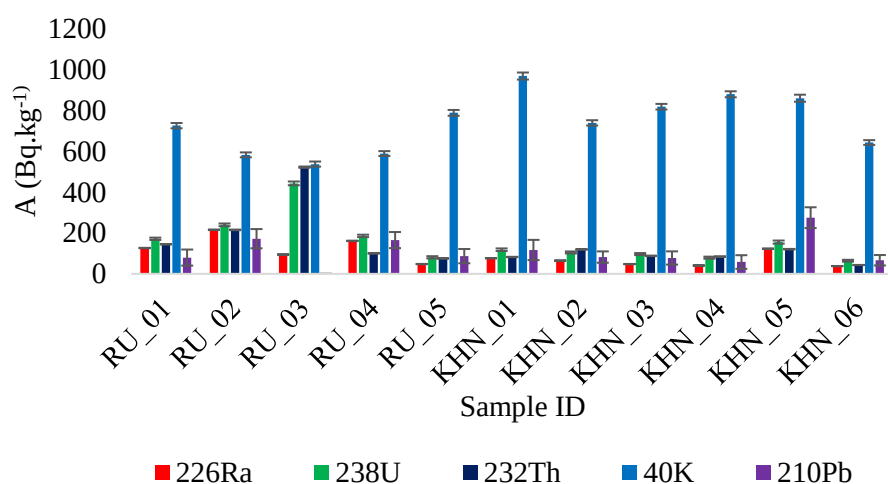


Figure 4. 4: Activity concentrations for samples taken from study areas C and E.

The distribution for the activity concentrations of ^{226}Ra , ^{238}U , ^{232}Th , ^{40}K and ^{210}Pb for samples collected from the study area C, coded KHN, are shown in Figure 4.4. The

activity concentrations of ^{226}Ra were found to range from 37.81 ± 0.60 to 123.05 ± 1.21 Bq.kg^{-1} with an average value of 65.11 ± 0.82 Bq.kg^{-1} . The activity concentrations of ^{238}U varied from 64.24 ± 4.47 to 155.30 ± 7.64 Bq.kg^{-1} with an average value of 103.01 ± 5.70 Bq.kg^{-1} while that of ^{210}Pb ranged from 57.83 ± 33.33 to 274.80 ± 50.88 Bq.kg^{-1} with an average of 112.59 ± 37.95 Bq.kg^{-1} . The activity concentrations for ^{232}Th were found to vary from 43.15 ± 0.57 to 120.69 ± 1.28 Bq.kg^{-1} with an average value of 89.70 ± 1.09 Bq.kg^{-1} while for ^{40}K , they varied from 642.50 ± 11.63 to 967.70 ± 17.24 Bq.kg^{-1} with an average value of 817.37 ± 14.57 Bq.kg^{-1} . The average activity concentrations of ^{226}Ra , ^{238}U , ^{232}Th and ^{40}K in area C were slightly higher than those of areas A and B and above the global average values. This might have been influenced by the human activities in the surrounding, probably the deposition of tailings and dust containing an enhanced level of radionuclides blown from the tailing's storage facilities and mining areas. Hamutoko *et al.*, (2014) reported cases of seasonal floods experienced during the 2009 to 2011 heavy falls, therefore, flood water might have swept radionuclides from the tailings and mining operations and deposited them along the Khan river.

The samples of the study area E are coded RU and were collected from the immediate vicinity of a uranium mine and across the boundary of the premise's operations. It was thus expected for the activity concentrations of samples taken in area E to display enhanced levels of radioactivity than for those samples from study areas A - C. The activity concentrations of ^{226}Ra varied from 47.80 ± 0.73 to 215.66 ± 1.37 Bq.kg^{-1} with an average value of 129.09 ± 1.06 Bq.kg^{-1} . The average value was about three times higher than the corresponding value observed in study area A. The activity concentrations for ^{238}U varied from 80.94 ± 5.46 to 442.61 ± 9.20 Bq.kg^{-1} with an average value of 224.10 ± 6.79 Bq.kg^{-1} , about three times higher than the activity concentrations for ^{238}U observed in the study areas A and B. The activity concentrations of ^{232}Th was found to vary from 75.66 ± 1.49 to 521.85 ± 3.15 Bq.kg^{-1} with an average value of 211.56 ± 1.85 Bq.kg^{-1} . The activity concentrations for ^{40}K varied from 537.00 ± 12.52 to 787.60 ± 14.25 Bq.kg^{-1} with an average value of 644.02 ± 12.92 Bq.kg^{-1} , although greater than the global average, it was comparable to the average values observed in study areas A to C. The activity concentrations of ^{210}Pb was found to vary from <2.67 to 172.00 ± 47.05 Bq.kg^{-1} with an average value of 125.89 ± 41.56 Bq.kg^{-1} .

The distribution of the activity concentrations for samples collected from the study area D is illustrated in Figure 4.5, while its detailed data presented in Table B4 of the Annexure B.

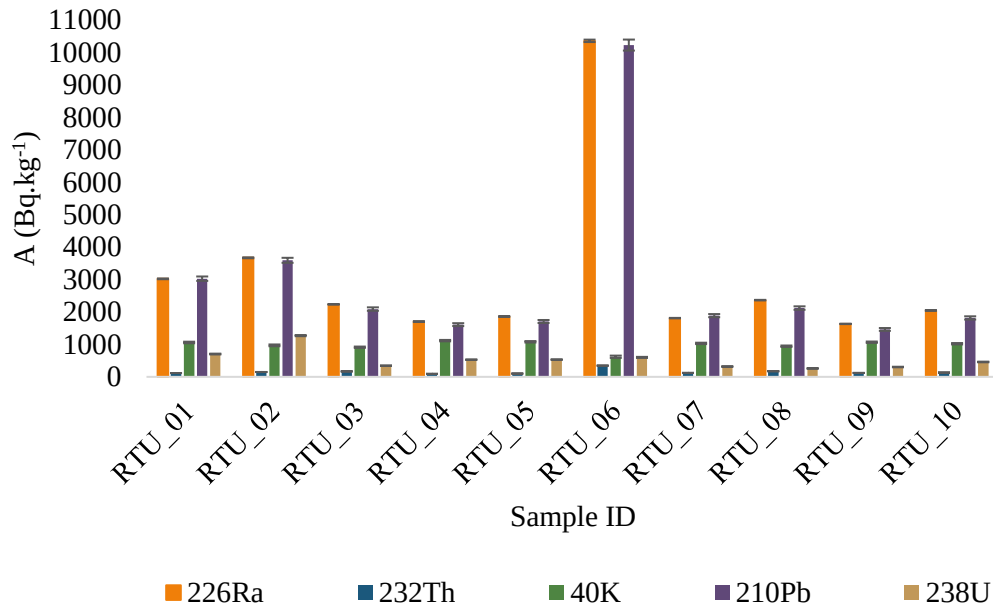


Figure 4. 5: Activity concentrations for samples taken from study area D.

Figure 4.5 shows that activity concentrations of ²²⁶Ra and ²¹⁰Pb, were relatively high, in excess of 1000 Bq.kg⁻¹ at all sampling points. The measured activity concentrations of ²²⁶Ra were found to range from 1632.98±6.70 to 10360±37.51 Bq.kg⁻¹ with an average value of 3071.01± 3.53 Bq.kg⁻¹, while that of ²¹⁰Pb varied from 1462.00±43.47 to 10230.00±170.20 Bq.kg⁻¹ with an average value of 2954.40±72.31 Bq.kg⁻¹. Notwithstanding that uranium was extracted and recovered from the ores, significant amount of activity concentrations for ²³⁸U were measured, which ranged from 259.55±9.21 to 1271.58±17.95 Bq.kg⁻¹ with an average value of 532.09±11.90 Bq.kg⁻¹. The measured values for ²³²Th suggested that the study area was not as rich in thorium as indicated by the relative low activity concentrations that ranged from 92.68±1.19 to 346.15±2.73 Bq.kg⁻¹ with an average value of 152.18±1.59 Bq.kg⁻¹. The activity concentrations of ⁴⁰K were slightly elevated compared to those observed in study areas A, B, C and E, and found to range from 619.40±35.19 to 1117.00±21.95 Bq.kg⁻¹ with an average value of 982.74±24.56 Bq.kg⁻¹, which was higher than the global average value.

Figures 4.6 and 4.7 indicate a discernible gradual increase in the average activity concentrations of ^{226}Ra , ^{238}U , ^{232}Th and ^{210}Pb across the study areas from A to E.

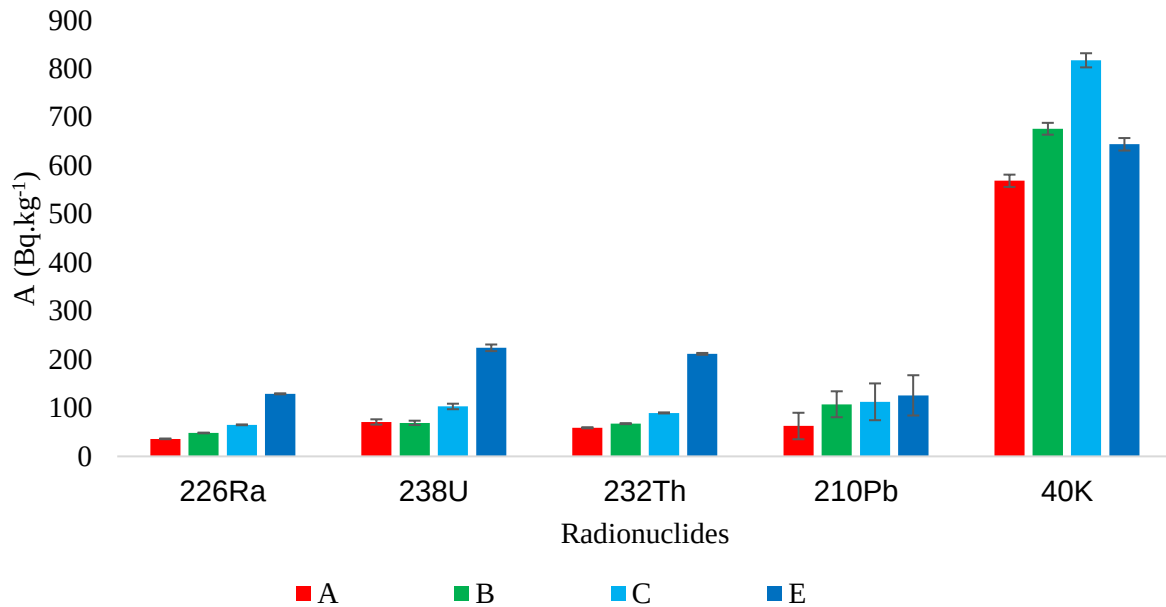


Figure 4. 6: Trends on average activity concentrations for the radionuclides in the study areas A, B, C and E.

As it can be seen in Figure 4.6 that there has been a systematic high uncertainty associated with the quantification of activity concentrations at lower energies, specifically at 46.5 keV for ^{210}Pb , probably due to effects of self-absorption and Compton continuum background.

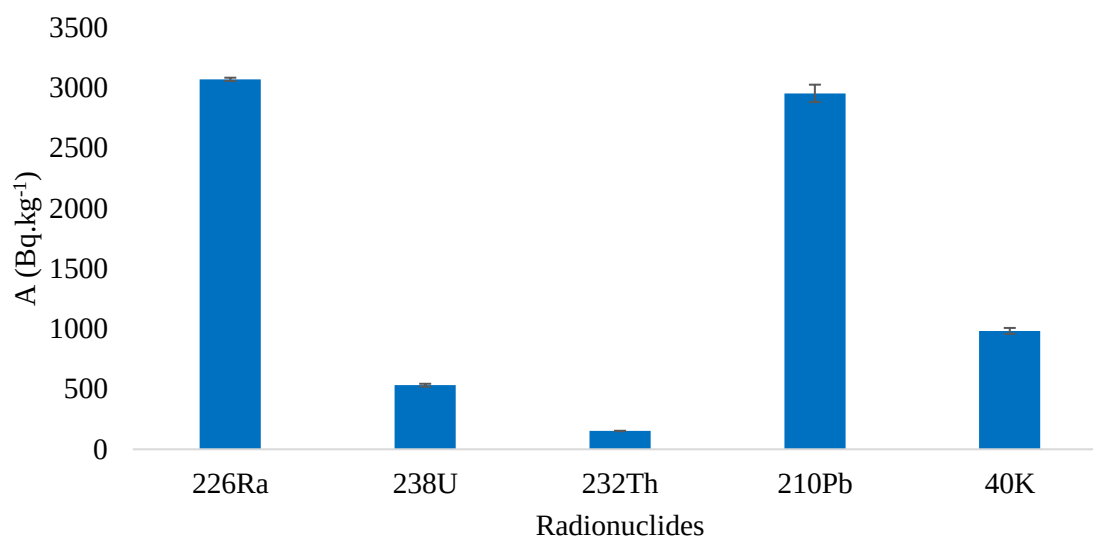


Figure 4. 7: Average activities concentrations for the radionuclides in the study areas D.

The average activity concentrations for the current study were compared to some of the published data from some studies conducted in Namibia and other countries as presented in Table 4.1.

Table 4. 1: Comparison of average activity concentrations of ²³⁸U, ²³²Th and ⁴⁰K in soil of the current study with those of the other studies.

Area / Country	Average activity concentration in soil (Bq.kg ⁻¹)			Reference
	²³⁸ U	²³² Th	⁴⁰ K	
Windhoek, Namibia	24.80 ± 5.10	35.2 ± 9.90	517.90 ± 117.40	Oyedele (2006)
Walvis Bay, Namibia	30.38 ± 11.28	32.58 ± 10.09	203.62 ± 27.00	Njinga <i>et al.</i> (2016)
Swakopmund, Namibia	99.60 ± 24.39	90.90 ± 31.99	553.07 ± 107.17	Njinga <i>et al.</i> (2016)
Algeria	30	25	370	IAEA (2000)
Egypt	37 (6 - 120)	18 (2 - 96)	320 (29 - 650)	IAEA (2000)
Panipat, India	30.24 ± 0.53	29.89 ± 0.61	291.06 ± 0.57	Amanjeet (2017)
Study area A	71.87 ± 5.44	59.07 ± 1.11	568.73 ± 12.66	Current study
Study area B	69.19 ± 8.06	67.19 ± 8.77	676.10 ± 39.41	Current study
Study area C	103.01 ± 5.70	89.70 ± 1.09	817.37 ± 14.57	Current study
Study area D	532.09 ± 11.90	152.18 ± 1.59	982.74 ± 24.56	Current study
Study area E	224.10 ± 6.79	211.56 ± 1.85	644.02 ± 12.92	Current study
Worldwide	33 (16 - 110)	45 (11 - 64)	420 (140 - 850)	IAEA (2000)

It can be seen (Table 4.1) that the average activity concentrations of ^{238}U , ^{232}Th and ^{40}K for this study were relatively higher than the global average documented by the UNSCEAR (2000) and that measured in Algeria, Egypt, India, Thailand and some parts of Namibia.

The activity concentration ratios of $^{238}\text{U}/^{226}\text{Ra}$ and $^{232}\text{Th}/^{238}\text{U}$ for soil samples taken from study areas A to E were determined and are presented in Tables B6 - B10 of the Annexure B. The activity concentration ratios of $^{226}\text{Ra}/^{210}\text{Pb}$ for soil samples taken from study area D were also determined. The distribution for the activity concentration ratios of $^{238}\text{U}/^{226}\text{Ra}$ for soil samples taken from study area A, illustrated in Figure 4.8, were found to vary from $1,42\pm0,16$ to $3,58\pm0,17$ with an average of $1,98\pm0,16$ while those of B, illustrated in Figure 4.9, varied from $0,92\pm0,04$ to $4,16\pm0,20$ with an average of $1,78\pm0,13$. The activity concentration ratios of $^{238}\text{U}/^{226}\text{Ra}$ for soil samples taken from study area C were found to vary from $1,26\pm0,063$ to $2,03\pm0,11$ with an average value of $1,68\pm0,11$ while that of study area E varied from $1,11\pm0,03$ to $4,69\pm0,11$ with an average value of $2,00\pm0,68$. The observed average activity concentration and their respective ratios for the study areas A, B, C and E indicated that the activity concentrations of ^{238}U at most of the sampling points were about twice those of ^{226}Ra , suggesting a state of secular disequilibrium. The state of disequilibrium might be attributed to conditions, where either there was a constant supply of uranium by weathering of rocks in the area or that ^{226}Ra had extensively leached out of the sampled depth and migrated into deep soil layers.

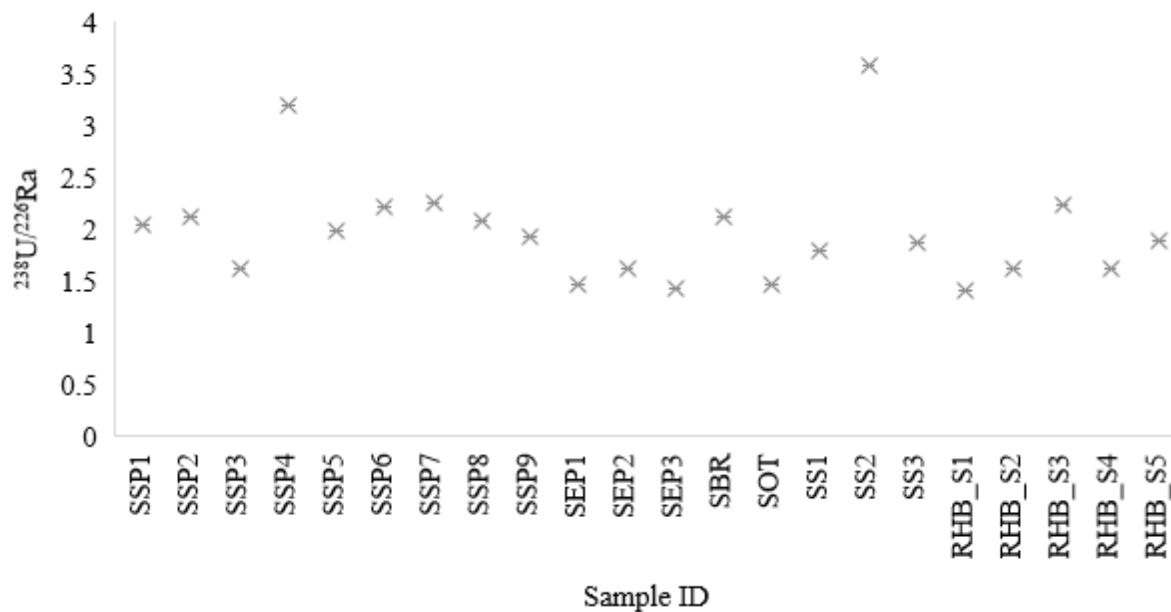


Figure 4. 8: Activity concentration ratios of $^{238}\text{U}/^{226}\text{Ra}$ for study area A.

The activity concentration ratios of $^{238}\text{U}/^{226}\text{Ra}$ for samples taken from the study area D are presented in Table B9 and were found to range from 0.06 ± 0.00 to 0.35 ± 0.01 with an average value of 0.21. All the ratio values were lower than a unity, an indication of samples depleted of ^{238}U as it had been extracted during the ore processing and elemental recovery.

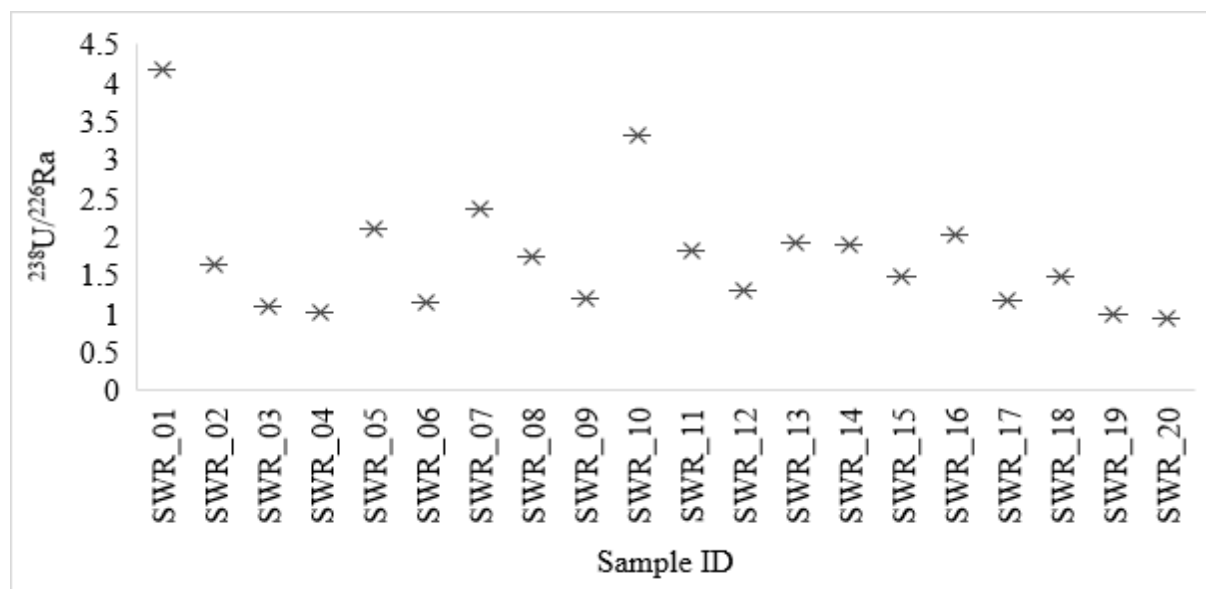


Figure 4. 9: Activity concentration ratios of $^{238}\text{U}/^{226}\text{Ra}$ for study area B.

The observed activity concentration ratios for $^{232}\text{Th}/^{238}\text{U}$ for soil samples from the study area A were found to vary from 0.43 ± 0.02 to 1.08 ± 0.06 with an average value of 0.77 ± 0.04 , while for those taken from the study area B varied from 0.43 ± 0.02 to 2.91 ± 0.10 with an average value of 1.05 ± 0.13 . The activity concentration ratios for the

study areas C ranged from 0.67 ± 0.05 to 1.14 ± 0.06 with an average of 0.88 ± 0.05 , while those from areas D and E ranged from 0.12 ± 0.00 to 0.66 ± 0.12 and 0.54 ± 0.12 to 1.18 ± 0.03 with averages of 0.34 ± 0.01 and 0.88 ± 0.03 respectively. The average activity concentration ratios for study areas A to E were both significantly lower than the theoretical value of 1.20, hence inconsistent with the expected condition for the continental crust.

An interesting observation was the maintained secular equilibrium between ^{226}Ra and ^{210}Pb for samples collected from the study area D, whose activity concentration ratios ranged from 0.96 ± 0.02 to 1.13 ± 0.03 with an average of 1.06 ± 0.02 , as presented in Table B9. The activity concentration ratios obtained in the current study will serve as useful references for environmental regulatory agencies' programmes on environmental monitoring as they will help track changes in the local radioactivity trends overtime.

4.1.2 Activity concentrations of ^{238}U , ^{232}Th and ^{40}K in water

The water sampling locations are indicated in Figure 3.6 and the results are presented in Tables 4.2, 4.3 and 4.4 for the Swakop river, Khan river and RUM processing activities, respectively.

4.1.2.1 Activity concentrations of ^{40}K , ^{238}U and ^{232}Th in groundwater samples for The Swakop river

It can be seen from Table 4.2 that there is a variation among the distribution of activity concentrations of ^{40}K and ^{238}U in groundwater along the Swakop river. The activity concentrations for ^{40}K were found to range from 737.66 to 2475.98 mBq.l^{-1} with an average value of $1449.22 \pm 181.85 \text{ mBq.l}^{-1}$, while those of ^{238}U varied from 0.161 to 1116.47 mBq.l^{-1} with the average of $313.43 \pm 127.62 \text{ mBq.l}^{-1}$. The elemental mass concentration of ^{232}Th in the Swakop river water samples was below the detection limit of the analytical technique used. This could be explained by the fact that ^{232}Th is sparingly soluble, hence limiting its concentrations in water (ATSDR, 2019; EPA, 1999).

The boreholes RHBW and SHW have been used for crops irrigation. The Goanikontes water sample recorded the highest activity concentrations of 2475.98 and 1116.47

mBq.l⁻¹ for ⁴⁰K and ²³⁸U, respectively. The activity concentrations of ⁴⁰K and ²³⁸U recorded from the cropland were 1942.54 and 639.90 mBq.l⁻¹ for SHW while 1523.59 and 308.15 mBq.l⁻¹ for RHBW, respectively, relatively lower than those observed at Goanikontes. The water sample ID: 41180 was collected at the confluence of the Gawib and Swakop rivers. The Gawib river runs directly under the non-lined tailings of LHU, therefore, expected to display elevated levels of ²³⁸U, but however, yielded the activity concentrations of 1655.71 and 528.95 mBq.l⁻¹, for ⁴⁰K and ²³⁸U respectively. Water samples IDs 203545, 203546, 203547, 41188 and 41184 were collected in the vicinity of the LHU operations against the water flow gradient and yielded varying activity concentrations of ²³⁸U and ⁴⁰K, that ranged from 0.161 to 146.91 mBq.l⁻¹ and 737.66 to 1422.10 mBq.l⁻¹, respectively, relatively lower than those of the water sample ID: 41180.

Table 4. 2: Total elements and derived activity concentrations for ⁴⁰K, ²³²Th and ²³⁸U in groundwater along the portion of Swakop river.

Sample ID / borehole ID	Total element (mg.l ⁻¹)		Activity concentration (mBq.l ⁻¹)	
	K	U	⁴⁰ K	²³⁸ U
203547	25.35	0.00116	784.38	14.30
41188	23.84	0.005307	737.66	65.57
41184	40.11	0.0119	1241.09	146.91
203545	40.72	3.92E-05	1259.96	0.4843
203546	45.96	0.000013	1422.10	0.161
41180 (Gawib river)	53.51	0.0428	1655.71	528.95
Goanikontes	80.02	0.09036	2475.98	1116.47
SHW	62.78	0.0518	1942.54	639.90
RHBW	49.24	0.0249	1523.59	308.15
Minimum	23.84	0.000013	737.66	0.161
Maximum	80.02	0.09036	2475.98	1116.47
Average	46.84	0.0254	1449.22	313.43

The activity concentrations of ⁴⁰K were relatively higher than those of ²³⁸U and ²³²Th in all sampling points along the Swakop river. It was observed that the activity concentrations of ²³⁸U in water samples collected downstream the LHU operations were relatively high than those collected elsewhere along the Swakop river.

4.1.2.2 Activity concentrations of ^{40}K , ^{238}U and ^{232}Th in groundwater samples for the Khan river

The water sample ID W214A, W3UK4B and W416A were collected from the RUM monitoring boreholes. The boreholes are in the direction of water flow from the RUM tailings' storage facility and strategically located along the frontier of the seepage plume to allow for quick determination on the impacts of mining activities on the Khan river ground water (RUL, 2018). The observed activity concentrations of ^{232}Th were extremely low, ranging from BDL to 0.630 mBq.l⁻¹ in comparison to those of ^{238}U and ^{40}K , that ranged from 353.99 to 1875.59 mBq.l⁻¹ and 923.93 to 1870.76 mBq.l⁻¹, respectively.

On its annual report, RUL (2018) recorded activity concentrations of 610 and 270 mBq.l⁻¹ for ^{238}U in the W214A and W416A boreholes, respectively, obtained through the use an alpha spectroscopic technique. The current study observed the activity concentrations of 616.30 and 353.99 mBq.l⁻¹ for ^{238}U in the W214A and W416A boreholes, respectively, as obtained by an ICP-MS and therefore, a consistent observation using a different analytical technique. The water samples collected from boreholes GW29, GWN01, 202082 and 202083 display uniform activity concentrations of the ^{40}K and ^{238}U (Table 4.3).

Table 4. 3: Total elements and derived activity concentrations for ^{40}K , ^{232}Th and ^{238}U in groundwater along a portion of the Khan river.

Sample ID	Total elemental content (mg.l ⁻¹)			Corresponding derived activity concentration (mBq.l ⁻¹)		
	K	Th	U	^{40}K	^{232}Th	^{238}U
W214A	29.86	0.000155	0.049	923.93	0.63	616.30
W3UK4B	34.3	0.0000611	0.086	1061.31	0.25	1062.46
W416A	41.46	0.00007.22	0.029	1282.86	0.29	353.99
GW29	55.68	BDL	0.128	1722.85	BDL	1580.29
GWN01	59.61	BDL	0.152	1844.46	BDL	1875.59
202083	59.11	BDL	0.140	1828.98	BDL	1724.85
202082	60.46	BDL	0.143	1870.76	BDL	1764.38
Minimum	29.86	BDL	0.029	923.93	BDL	353.99
Maximum	60.46	0.000155	0.152	1870.76	0.63	1875.59
Average	48.64	0.00004.12	0.104	1505.02±153.16	0.17±0.1	1282.55±230.04

The boreholes form a monitoring network for the Swakop Uranium (SU). The activity concentrations of ^{238}U observed in the monitoring boreholes for the SU were found to be up to 4 times higher than those of the RUM monitoring boreholes. The activity concentrations of ^{40}K were relatively high than those of the ^{238}U and ^{232}Th in all sampling points along the Khan river.

The variations in the activity concentrations of ^{238}U in both rivers were cited by MME (2010) and Hamutoko *et al.*, (2014) and attributable to the non-homogeneity, compartmentalised and discrete aquifers that limited the degree of groundwater diffuse and mixing between aquifers. Hamutoko *et al.*, (2014) further attributed the non-uniformity to the local geochemical processes and some factors such as lithology and pH that influenced the solubility of radionuclides. Looking at the distribution of the activity concentrations of ^{40}K and ^{238}U , it is likely that GW29, GWN01, 202083 and 202082 belonged to the same compartment.

A study by Kringel *et al.*, (2010) along the valleys of the Khan and Swakop rivers observed high concentrations of natural uranium in excess of the provisional WHO guideline value in 52 out of 66 (79%) water samples, hence opined that uranium is a common trace element along the rivers occurring at an elevated level. It can be seen in Tables 4.2 and 4.3 that the activity concentrations of ^{238}U were relatively higher in the Khan river than those observed in the Swakop river and the elevated level of ^{238}U downstream the confluence of the Swakop and Khan rivers up to the cropland.

4.1.2.3 Activity concentrations of ^{40}K , ^{238}U and ^{232}Th in the process and groundwater samples from RUM premises

Table 4.4 presents the activity concentrations of ^{238}U , ^{232}Th and ^{40}K observed in the process and ground water samples collected from the RUM.

Table 4. 4: Elemental concentrations and derived activity concentrations for ^{40}K , ^{232}Th and ^{238}U in the process and ground water within the RUM premises.

Sample ID	Water Source	Total elemental (mg. l ⁻¹)			Activity concentration (Bq. l ⁻¹)		
		K	Th	U	^{40}K	^{232}Th	^{238}U
RU_T1	process water	61.46	0.0011	2.023	1.902	0.0044	24.995
RU_T2	process water	23.63	11.34	54.79	0.731	46.0338	676.97
RU_T3	process water	23.26	12.51	56.66	0.72	50.7833	700.07
W1_R3	ground water	60.82	0.0017	1.794	1.882	0.00683	22.166
Minimum		23.26	0.0011	1.794	0.72	0.0044	22.166
Maximum		61.46	12.51	56.66	1.902	50.7833	700.07
Average		42.29	5.96	28.82	1.308	24.207	356.05

The process water displayed relatively higher activity concentrations for ^{232}Th and ^{238}U than those observed from other water sources. The activity concentrations for ^{238}U were found to be varying from 22.17 to 700.07 Bq.l⁻¹ with an average value of 356.05 Bq.l⁻¹. The activity concentrations for ^{232}Th were found to range from 0.0044 to 50.78 Bq.l⁻¹ with an average value of 24.21 Bq.l⁻¹. The activity concentrations of ^{40}K in the water samples collected from the tailing's storage facility ponds and seepage collection trench W1_R3, were ranged from 0.72 to 1.902 Bq.l⁻¹ with an average of 1.308 Bq.l⁻¹, hence there was no significant variations in the ^{40}K observed between the water sources.

4.1.3 Activity concentrations of ^{238}U , ^{232}Th and ^{40}K in agricultural food crops

It has been observed that the activity concentrations of ^{40}K dominated both in soil (4.1.1) and water (4.1.2) samples. A similar trend had been observed in food crops, where ^{40}K had the highest average activity concentration of 1603.56 Bq.kg⁻¹ (1401.06 - 1828.99 Bq.kg⁻¹) observed in spinach, followed by eggplant with an average of 741.27 Bq.kg⁻¹, (725.90 - 754.06 Bq.kg⁻¹), then followed by 567.79 Bq.kg⁻¹ for beetroot. It is generally believed that spinach is one of the dietary sources of potassium. The average activity concentrations of ^{40}K measured in food crops were up to two times high than the observed activity concentrations in soil samples from, which the crops were grown. Being an essential element necessary for the growth and metabolism, potassium is preferentially absorbed in large quantity, more than uranium and thorium, and in accordance with the crop's physiological and metabolic need (Eisenbud &

Gesell, 1997; IAEA, 2009a). It could be seen in Tables 4.5 to 4.7 that the activity concentrations of ^{40}K in all samples were significantly higher than those of ^{238}U and ^{232}Th in all agricultural crop types.

Table 4. 5: Elemental concentrations and derived activity concentrations for ^{40}K , ^{232}Th and ^{238}U in spinach samples.

Total elemental mass concentrations (mg.kg ⁻¹)				Derived Activity concentrations (Bq.kg ⁻¹)		
Sample ID	K	U	Th	^{40}K	^{238}U	^{232}Th
SP1	47170	0.087	0.441	1459.54	1.07	1.79
SP2	51300	0.087	0.187	1587.33	1.08	0.76
SP3	53940	0.070	0.189	1669.01	0.86	0.77
SP4	45280	0.102	0.207	1401.06	1.26	0.84
SP5	59110	0.058	0.105	1828.99	0.71	0.43
SP6	48060	0.078	0.189	1487.08	0.97	0.77
SP7	52200	0.078	0.156	1615.18	0.96	0.63
SP8	50840	0.106	0.161	1573.09	1.31	0.65
SP9	58520	0.069	0.219	1810.73	0.86	0.89
Minimum				1401.06	0.71	0.43
Maximum				1828.99	1.31	1.79
Average				1603. 56±234.36	1.01±0.20	0.84±0.41

The minimum and maximum average activity concentrations of ^{238}U were 0.15 Bq.kg⁻¹ and 1.01 Bq.kg⁻¹ observed in eggplant and spinach, respectively, while for ^{232}Th were 0.09 Bq.kg⁻¹ and 0.84 Bq.kg⁻¹ observed in eggplant and spinach, respectively. A study in Tanzania by Nyanda & Nkuba (2017) reported activity concentrations of 2520±50.5 Bq.kg⁻¹ for ^{40}K , 19.70±0.4 Bq.kg⁻¹ for ^{238}U and 30.10±0.90 Bq.kg⁻¹ for ^{232}Th in spinach, significantly higher than those of the current study area.

Table 4. 6: Elemental concentrations and derived activity concentrations for ^{40}K , ^{232}Th and ^{238}U in eggplant samples.

Total elemental mass concentrations (mg.kg ⁻¹)				Derived activity concentrations (Bq.kg ⁻¹)		
Sample ID	K	U	Th	^{40}K	^{238}U	^{232}Th
EP1	23460	0.01273	0.02812	725.90	0.16	0.11
EP2	24040	0.01177	0.02006	743.85	0.15	0.08
EP3	24370	0.01141	0.02045	754.06	0.14	0.08
Minimum				725.90	0.14	0.08
Maximum				754.06	0.16	0.11
Average				741.27±14.26	0.15±0.01	0.09±0.02

Table 4. 7: Elemental concentrations and derived activity concentrations for ^{40}K , ^{232}Th and ^{238}U in beetroot.

Sample ID	Elemental concentrations (mg.kg ⁻¹)			Derived activity concentrations (Bq.kg ⁻¹)		
	K	U	Th	^{40}K	^{238}U	^{232}Th
BR	18350	0.02277	0.05122	567.79	0.28	0.21

The roots accumulate high amount of metals and serve as natural barriers preventing them from reaching to the upper parts of plants (Shtangeeva, 2010), therefore, an expectation was of the beetroot (BR) to display high activity concentrations of radionuclides than those observed in spinach and eggplant. The current study recorded relatively high activity concentrations than the reference values of 20 mBq.kg⁻¹ and 3 mBq.kg⁻¹ for ^{238}U and 15 mBq.kg⁻¹ and 0.5 mBq.kg⁻¹ for ^{232}Th , for leafy vegetables and roots, respectively, published in UNSCEAR (2000) report. The low uptake of ^{238}U and ^{232}Th observed in the current study should be conceptualised on the view that uranium and thorium do not play any known beneficial role in biochemical processes of any organisms.

4.2 Activity concentration transfer factors of ^{238}U , ^{232}Th and ^{40}K from soil to food crops

The transfer factor of any radionuclide is viewed in the context of the physicochemical characteristics of soil such as pH, organic and inorganic contents, the concentration

of radionuclides within the root zone, soil type as well as plant species. The soil texture was found comprised of sand (92.9%), clay (4.7%) and silt (2.4%) with the pH ranged from 7.48 to 7.81 with an average of 7.67, that is, neutral to basic. The ability of soil to maintain and exchange the cations, cations exchange capacity (CEC), was found to vary from 18.68 to 20.00 cmol(+).kg⁻¹ with an average value of 19.25 cmol(+).kg⁻¹. The average values for common anions measured were 1.59 mg.l⁻¹ for PO₄³⁻, 837 mg.l⁻¹ for SO₄²⁻, 24.90 mg.l⁻¹ for NO₃⁻, 1529.45 mg.l⁻¹ for Cl⁻, and 57.96 mg.l⁻¹ for HCO₃⁻.

The activity concentration ratios of ⁴⁰K, ²³⁸U and ²³²Th in the considered food crops to soil are presented in Tables 4.8 and 4.9.

Table 4. 8: The activity concentration ratios of ⁴⁰K, ²³⁸U and ²³²Th in spinach to soil.

Ratios	Transfer factor		
	⁴⁰ K	²³⁸ U	²³² Th
SP1/SSP1	2.59	0.0154	0.0362
SP2/SSP2	2.85	0.0148	0.0161
SP3/SSP3	2.84	0.0148	0.0143
SP4/SSP4	2.07	0.0108	0.0162
SP5/SSP5	3.25	0.0109	0.0104
SP6/SSP6	2.63	0.0129	0.0164
SP7/SSP7	2.63	0.0120	0.0136
SP8/SSP8	2.81	0.0185	0.0136
SP9/SSP9	3.21	0.0129	0.0176
Minimum	2.07	0.0108	0.0104
Maximum	3.25	0.0185	0.0362
Average	2.77	0.014	0.020

The TF Values for ⁴⁰K were found to be relatively higher than those for ²³⁸U and ²³²Th in all food crop types considered in this study. The TF values for ⁴⁰K were found to vary from 2.07 to 3.25 with an average of 2.77 measured for leafy vegetable (spinach). The average value observed in the current study was relatively higher than 1.4 the reference value published by IAEA, (2010), however, lied within the range of 0.49 to 5.6. In the non-leafy vegetable category, the TF values for ⁴⁰K varied from 1.35 to 2.57 with an average of 1.76 observed for eggplant, while the least value of 0.98 was observed for beetroot. The TF values for ²³⁸U varied from 0.0108 to 0.0185 with an average of 0.014 while those of ²³²Th varied from 0.0104 to 0.0362 with average of 0.02 for spinach. The TF values of ²³⁸U observed for eggplant found to vary from 0.0028 to 0.0044 with an average of 0.0035 while for ²³²Th varied from 0.0018 to

0.0041 with an average of 0.003. The TF values for beetroot were 0.004 and 0.01 for ^{238}U and ^{232}Th , respectively.

Table 4. 9: The activity concentration ratios of ^{40}K , ^{238}U and ^{232}Th in eggplant and beetroot.

Ratios	Transfer Factor		
	^{40}K	^{238}U	^{232}Th
EP1/SEP1	2.57	0.0044	0.0041
EP2/SEP2	1.35	0.0028	0.0019
EP3/SEP3	1.35	0.0032	0.0018
Minimum	1.35	0.0028	0.0018
Maximum	2.57	0.0044	0.0041
Average	1.76	0.0035	0.0030
BR/SBR	0.98	0.004	0.01

The average TF values for ^{238}U in leafy vegetable (spinach), non-leafy vegetables (eggplant) and roots (beetroot) observed in this study were relatively low, in contrast to the reference values published by the IAEA (2010). On the other hand, the average TF values for ^{232}Th in leafy vegetables, (spinach) non-leafy vegetables (eggplant) and roots (beetroot) observed in this study were relatively higher than the reference values as published by the IAEA (2010) and presented in Table 4.10. It is generally believed that the TF for ^{238}U and ^{232}Th are relatively lower than those of ^{40}K , and those of ^{232}Th being 10-fold lower than those of ^{238}U (IAEA, 2009a).

Table 4. 10: Comparison of published TF in crop types to those observed in the current study.

Crop types									
Leafy vegetables (spinach)			Non-leafy vegetables (eggplant)			Root crops (beetroot)			Reference
²³⁸ U	²³² Th	⁴⁰ K	²³⁸ U	²³² Th	⁴⁰ K	²³⁸ U	²³² Th	⁴⁰ K	
0.17	0.0012	1.3	0.019	0.00078		0.0078	0.00084		IAEA (2010)
0.0083	0.0018	1	0.004	0.00025	0.55	0.012	0.00033	0.55	Staven <i>et al.</i> (2003)
			0.006	0.002					Aswood <i>et al.</i> (2013)
0.014	0.02	2.77	0.0035	0.0030	1.76	0.0040	0.01	0.98	Current study (average)

4.3 Potential radiological risks associated with soil and food crops samples from the study areas

The activity concentrations of ^{238}U , ^{232}Th and ^{40}K in soil were used in the derivation of potential indicators for the radiological health risks; absorbed gamma dose rates in air, effective dose, radium equivalent concentration and cancer morbidity risk. The absorbed dose rates were used in the estimation of the corresponding annual effective doses at each sampling location. The activity concentrations of ^{238}U , ^{232}Th and ^{40}K in food crops were used to estimate the committed dose due to consumption of such crops. The annual effective dose due to external exposure and committed dose were used to determine the total effective doses. The groundwater from the alluvial aquifers of the Swakop river does not fit for human consumption, hence there was no envisaged direct dose to human.

4.3.1 Absorbed gamma dose rate in air, at 1 meter above the ground

The absorbed gamma dose rates for all soil samples from the study areas A to E are presented in Tables C1 and C2 of the Annexure C, respectively. The distribution and contribution of ^{238}U , ^{232}Th and ^{40}K to the absorbed gamma dose rates at 1 meter above the ground for each sample from the Swakop river and cropland are illustrated in Figure 4.10. The estimated absorbed gamma dose rates at 1 meter above the ground for the study area A were found to vary from 45.12 ± 3.01 to 142.71 ± 2.53 nGy.h⁻¹ with an average value of 88.65 ± 0.47 nGy.h⁻¹ while for the study area B they varied from 45.24 ± 2.22 to 192.47 ± 3.50 nGy.h⁻¹ with an average value of 100.94 ± 0.44 nGy.h⁻¹. The average values for both study areas were relatively higher than the global average value of 60 nGy.h⁻¹ reported by the UNSCEAR (2000) report.

Despite the activity concentrations of ^{40}K and ^{238}U in soil samples collected from the study areas A and B being higher compared to those of ^{232}Th , it should be noted that the dose coefficient estimate of ^{232}Th is however, greater than those of ^{40}K and ^{238}U . Therefore, ^{232}Th was found to contribute the most to the absorbed gamma dose rates in most of the samples, with an average of about 37.38% followed by ^{238}U and ^{40}K with their respective averages of 34.04% and 28.61%.

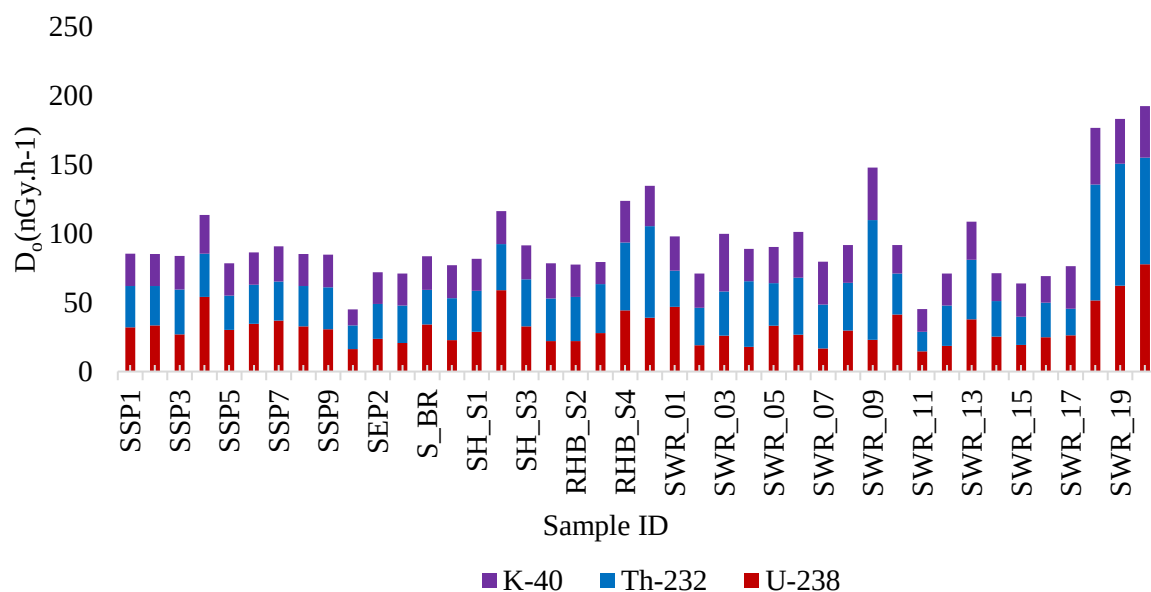


Figure 4. 10: The estimated absorbed dose rates in air due to gamma radiation arising from ^{238}U , ^{232}Th and ^{40}K for all the measured soil samples from the study areas A and B.

The distribution and contribution of ^{238}U , ^{232}Th and ^{40}K to the absorbed gamma dose rates at 1 meter above the ground for each sample from the study area C, D and E are illustrated in Figure 4.11 while the detailed data of all the samples are presented in Table C2 of the Annexure C. The absorbed dose rate for the area C were found to vary from 82.50 ± 4.21 to 180.48 ± 1.97 nGy.h^{-1} with an average value of 135.85 ± 0.75 nGy.h^{-1} . The study area D was found to record high absorbed dose rates ranging from 258.28 ± 2.51 to 717.97 ± 8.43 nGy.h^{-1} with an average value of 378.73 ± 1.28 nGy.h^{-1} followed by area E, which varied from 115.93 ± 2.75 to 542.08 ± 4.68 nGy.h^{-1} with an average value of 258.17 ± 1.37 nGy.h^{-1} . The estimated absorbed gamma dose rates for the study areas C, D and E were found to be relatively higher than those observed in areas A and B. It should be noted however, that the samples collected from study areas C – E should not be viewed as collected from undisturbed natural environment, since some of them were part of the uranium ores or might have been influenced by anthropogenic activities such as mining and processing activities in the surrounding area.

^{238}U was found to contribute the most to the absorbed gamma dose rates in most of the samples from the study areas C to E, with an average of about 49.20% followed by ^{232}Th and ^{40}K with their respective averages of 34.22% and 16.58%. Based on the overall results, the highest absorbed dose rate of 717.97 ± 8.43 nGy.h^{-1} was recorded

for sample RTU_02, which translated to an annual absorbed dose of about 6.30 mGy, based on a conservative assumption of a hypothetical individual residing at the tailings facility for uninterrupted 8760 hours a year. According to ICRP (2007), the tissue reactions (deterministic effects) manifest upon exceedance of tissue or organ threshold dose value, which is in order of 100 mGy, delivered either as a single acute dose or over a repeated low dose, therefore, there would be no potential for any manifestation of tissue reactions due to external exposure to gamma radiation arising from the ^{238}U , ^{232}Th and ^{40}K in the study area.

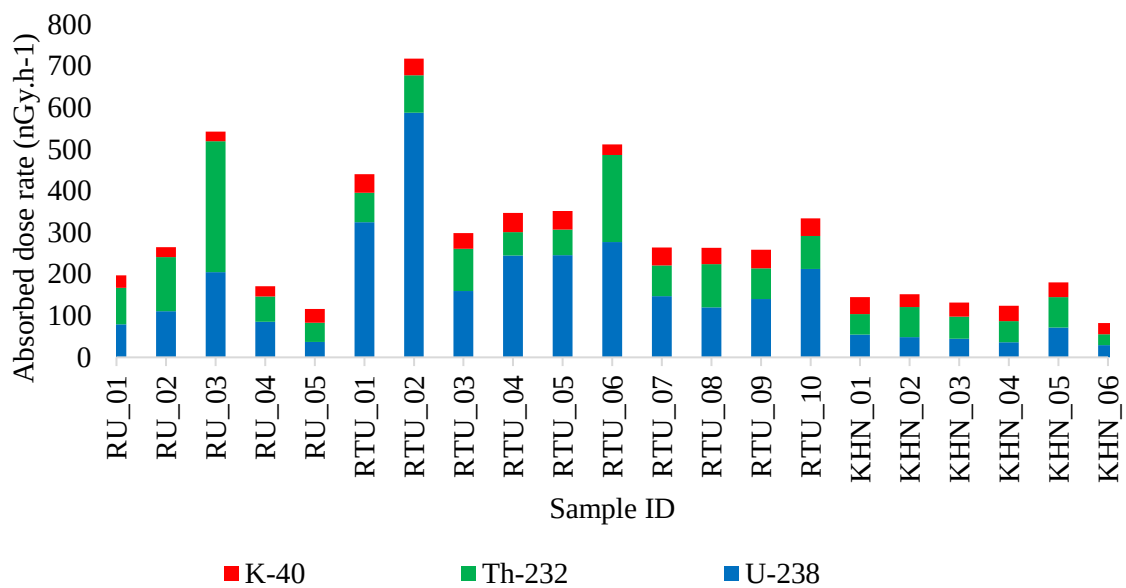


Figure 4. 11: The estimated absorbed dose rates, 1 meter above the ground, due to gamma radiation arising from ^{238}U , ^{232}Th and ^{40}K for the study areas C to E.

4.3.2 Annual effective dose due to external exposure to gamma radiation arising from ^{238}U , ^{232}Th and ^{40}K

The estimated absorbed gamma dose rates (nGy.h^{-1}) in section 4.3.1 were used in the derivation of corresponding annual effective doses due to external exposure to gamma radiation arising from the ^{238}U , ^{232}Th and ^{40}K in outdoor and indoor environments. The estimated results of the annual effective doses for external exposure in the outdoor and indoor environments assuming a hypothetical individual representative of an adult, child and infant age categories are presented in Tables C1 and C2 of Annexure C for samples collected from the study areas, whereas Tables C3 to C7 present the total effective doses to external exposure.

On soil samples collected from the study area A (Table C3 Annexure C), it was estimated that the age category representative of infants would receive high annual effective doses that varied from 0.36 ± 0.02 to 1.06 ± 0.02 mSv.y⁻¹ with an average of 0.69 ± 0.004 mSv.y⁻¹, followed by the age category representative of children with annual effective doses that ranged from 0.32 ± 0.02 to 0.94 ± 0.02 mSv.y⁻¹ with an average value of 0.61 ± 0.003 mSv.y⁻¹. The age category representative of adults receives the least annual doses that ranged from 0.28 ± 0.02 to 0.83 ± 0.01 mSv.y⁻¹ with an average value of 0.54 ± 0.002 mSv.y⁻¹. The estimated annual dose for the study area B (Table C4) were found to be slightly higher than those observed in the area A with the infants' age category receiving doses that varied from 0.36 ± 0.01 to 1.52 ± 0.02 mSv.y⁻¹ with an average of 0.80 ± 0.002 mSv.y⁻¹, while for the age category representative of children varied from 0.32 ± 0.01 to 1.35 ± 0.02 mSv.y⁻¹ with an average value of 0.71 ± 0.002 mSv.y⁻¹. The age category representative of adults would receive the least annual doses that ranged from 0.28 ± 0.01 to 1.18 ± 0.02 mSv.y⁻¹ with an average value of 0.62 ± 0.002 mSv.y⁻¹. The observed annual effective doses for both study areas and all age groups were greater than 0.48 mSv.y⁻¹, the global average reported by the UNSCEAR (2000), however, less than 1 mSv, the regulatory annual dose limit for the public.

The excavation of sand for the construction industry normally takes place at a depth high than 20 cm. Since all the soil samples for the study were collected at a depth of about 20 cm, the results might not be a true reflection of the overall activity concentrations of the primordial radionuclides at depth beyond the 20 cm. It is therefore, recommended that a comprehensive study be considered on the activity concentrations of primordial radionuclides at several soil depths beyond 20 cm.

The study areas C to E were within the mining licenced areas, where entry by the members of public is strictly prohibited. Therefore, the estimated annual effective doses due to external exposure was based on the age group representative of an adult. The estimated annual effective doses were found to be varying from 0.51 ± 0.01 to 1.11 ± 0.002 mSv.y⁻¹ with an average value of 0.84 ± 0.005 mSv.y⁻¹ for the study area C, from 1.58 ± 0.02 to 4.40 ± 0.04 mSv.y⁻¹ with an average value of 2.32 ± 0.01 mSv.y⁻¹ for the study area D, while for the study area E, the doses varied from 0.71 ± 0.01 to 3.32 ± 0.02 mSv.y⁻¹ with an average value of 1.58 ± 0.01 mSv.y⁻¹. The average annual

effective dose estimated for the study areas C to E and all age category groups are less than the prescribed annual limit value of 20 mSv for designated radiation workers.

4.3.3 Annual intake of radionuclides

The estimated annual intake of ^{238}U , ^{232}Th and ^{40}K due to the consumption of food crops are presented in Table 4.11.

Table 4. 11: Estimated annual intake of ^{238}U , ^{232}Th and ^{40}K for age categories 5 years, 15 years and adult members of the public.

Crop's type	Statistics	Radionuclides annual intake (Bq.y ⁻¹)		
		²³⁸ U	²³² Th	⁴⁰ K
Age category: 5 years				
Spinach	Range	14.23 - 26.27	8.52 - 35.77	28021.13 -36579.71
	Average	20.18±1.30	16.72±2.54	32071.10±985.07
Eggplant	Range	4.98 - 5.55	2.87- 4.03	25646.07 - 26640.87
	Average	5.22±0.17	3.28±0.38	26189.20±290.76
Beet root	n/a	16.88	12.48	34067.21
Age category: 15 years				
Spinach	Range	28.45 – 52.54	17.03 – 71.54	56042 – 73159.41
	Average	40.36±2.59	33.44±5.09	64142.21±1970.14
Egg plant	Range	8.33 – 9.29	4.81 – 6.74	42871.70 – 44534.67
	Average	8.73±0.29	5.48±0.63	43779.33±486.06
Beet root	n/a	30.95	22.87	62456.55
Age category: Adults				
Spinach	Range	42.68 – 78.80	25.55 – 107.31	86063.39 – 96213.31
	Average	60.54±3.89	50.16±7.64	96213.31±2955.21
Egg plant	Range	9.93 – 11.08	5.74 – 8.04	51139.71 – 53123.39
	Average	10.42±0.34	6.54±0.75	52222.38±579.80
Beet root	n/a	47.83	35.35	96523.76

The intakes were estimated based on the activity concentrations in food crops and the respective age dependent annual consumption rates for three age categories considered in the study. It should be noted that the consumption rate values used were for a hypothetical individual representative of the specified age category, therefore, the consumption rates used might be significantly different from that of the local community. The estimated annual intake of ^{40}K was found to be relatively high compared to that of ^{238}U and ^{232}Th within the same crops type. The minimum and maximum estimated intake values of 26189.20±290.76 and 96523.76 Bq.y⁻¹ for ^{40}K

were observed from eggplant (5 years of age category) and beet root (adult age category), respectively. The lowest average values of intake of 5.22 ± 0.17 and $3.28 \pm 0.38 \text{ Bq.y}^{-1}$ for ^{238}U and ^{232}Th , respectively, were observed from the eggplant (5 years' category). Despite the estimated high intake value for ^{40}K , it has been reported that the human body maintains the concentration of elemental potassium within the permissible limits under homeostatic control (Basu *et al.*, 2015).

4.3.4 Committed effective dose due to ingestion of food crops

Table 4.12 presents the estimated annual committed effective doses to a reference person member of the public of age category 5 years, 15 years and adult, derived by using equation 3.12. It can be seen in Table 4.12 that the contribution of individual radionuclides to the committed dose in each age categories and from each food crops were dominated by ^{40}K , which on average contributed about 98% whereas ^{232}Th and ^{238}U contribute merely 1.47% and 0.38%, respectively. A similar observation was made by Nyanda & Nkuba (2017) on the primordial radioactivity contents of spinach in Tanzania, where an estimated annual effective committed dose of 0.036 ± 0.001 , 0.277 ± 0.008 and $0.625 \pm 0.013 \text{ mSv}$ due to ^{238}U , ^{232}Th and ^{40}K , respectively, were reported.

The absorption of thorium through the gastro-intestinal track has been known to be low, therefore, the contribution of ^{232}Th to the committed dose could be negligibly low (UNSCEAR 1982). It was also found that the annual committed effective doses arising from each of the food crops were higher than the global average of 0.3 mSv , however, within the global range of $0.2 - 0.8 \text{ mSv.y}^{-1}$ reported by the UNSCEAR (2000) report.

For individual members of the public representative of the age categories 5 years, 15 years and adults, who consumed these food crops sourced from the study area, as per the corresponding consumption rates, the estimated annual committed effective doses would be 1.95 ± 0.022 , 1.32 ± 0.014 and $1.55 \pm 0.020 \text{ mSv}$, respectively, slightly high than the annual intervention exemption level of 1 mSv recommended for prolonged exposure from dominant commodities (ICRP, 1999; IAEA, 2014a).

Table 4. 12: Estimated annual average committed effective doses to member of the public of age categories 5 years, 15 years and adult.

Age category	Food crop	radionuclide	Committed dose by individual radionuclide (mSv.y ⁻¹)		Committed effective dose (mSv.y ⁻¹)
			Range	Average	
5	Spinach	²³⁸ U	0.0011 – 0.0021	0.0016±0.0001	0.68±0.020
		²³² Th	0.0030 – 0.125	0.0059±0.0009	
		⁴⁰ K	0.59 – 0.77	0.67±0.021	
	Eggplant	²³⁸ U	0.00039 – 0.00044	0.00042	0.55±0.01
		²³² Th	0.0010 – 0.0014	0.00114	
		⁴⁰ K	0.54 – 0.56	0.55±0.01	
	Beet root	²³⁸ U	n/a	0.001	0.72
		²³² Th	n/a	0.004	
		⁴⁰ K	n/a	0.715	
15	Spinach	²³⁸ U	0.00191 – 0.00352	0.0027	0.50±0.014
		²³² Th	0.0043 – 0.0179	0.0083	
		⁴⁰ K	0.425 – 0.556	0.487	
	Eggplant	²³⁸ U	0.00056 – 0.00062	0.00059	0.34±0.003
		²³² Th	0.00120 – 0.00168	0.00137	
		⁴⁰ K	0.326 – 0.338	0.333	
	Beet root	²³⁸ U	n/a	0.002	0.48
		²³² Th	n/a	0.006	
		⁴⁰ K	n/a	0.474	
Adults	Spinach	²³⁸ U	0.00192 – 0.00355	0.00272	0.61±0.02
		²³² Th	0.0059 – 0.025	0.012	
		⁴⁰ K	0.521 – 0.681	0.597	
	Egg plant	²³⁸ U	0.00044 – 0.00049	0.00047	0.33±0.003
		²³² Th	0.0013 – 0.0019	0.0015	
		⁴⁰ K	0.317 – 0.329	0.324	
	Beet root	²³⁸ U	n/a	0.0081	0.61
		²³² Th	n/a	0.0022	
		⁴⁰ K	n/a	0.598	

n/a not applicable.

It was also observed that the estimated committed effective doses among crop types were in the order of beet root > spinach>eggplant for the 5 years of age category and spinach >beet root >eggplant for both the 15 years and adults age categories. Beverages such as spinach juice derived from fresh vegetables had been observed in the local markets hence the assumption on estimated committed dose remains. However, in most cases, the food crops are boiled before consumption. The effect of boiling on the estimated committed effective dose was not evaluated in this study. It is therefore, recommended that comprehensive studies be conducted, covering the dominant food crops produced in the area that should also consider the effects of boiling on estimated doses.

4.3.5 Total annual effective dose

The total annual effective doses for age categories representative of 5 years, 15 years and adults were only estimated based on the samples collected along the Swakop river. The total annual effective doses for the three age categories were derived as a summation of respective average annual effective doses from the external exposure to gamma radiation in outdoor and indoor environments and the average committed effective dose from the ingestion of both crop types, as per the details provided in section 3.6.4. It was estimated that the age category 5 years would be the most exposed with the total annual effective dose of 2.69 ± 0.04 mSv, followed by the age category adult with an estimated total annual effective dose of 2.13 ± 0.04 mSv. The age group representative of 15 years would be the least exposed with an estimated total annual effective dose of 1.98 ± 0.03 mSv.

The estimated total annual effective doses for the three age categories were relatively higher than the annual intervention exemption level of 1 mSv recommended for prolonged exposure (ICRP, 1999; IAEA, 2014a). It was also found that there was no significant difference on the total annual effective dose for an adult category estimated in the current study to that of the global average value of 2.4 mSv due to background radiation reported in the UNSECAR (2000), although the current study did not consider the sources of exposure such as cosmic ray, radon and water. The principal contributor to the total effective dose was mainly the committed effective dose from the consumption of food crops grown in the Swakop river.

4.3.6 Radium equivalent activity

The R_{eq} values for all samples are presented in Tables C3 – C7 of the Annexure C. The estimated R_{eq} for the study area A were varied from 97.37 ± 6.38 to 313.07 ± 5.50 $Bq.kg^{-1}$ with an average value of 190.81 ± 1.02 $Bq.kg^{-1}$ while for the study area B, varied from 95.58 ± 4.83 to 420.44 ± 3.17 with an average value of 217.80 ± 0.95 $Bq.kg^{-1}$. The R_{eq} for samples SWR_18 to SWR_20 in the study area B exceeded the permissible value of $370 Bq.kg^{-1}$.

The R_{eq} for the study area C were varied from 175.42 ± 9.11 to 394.05 ± 4.26 $Bq.kg^{-1}$ with an average value of 294.22 ± 1.63 $Bq.kg^{-1}$ while those of area D were found to be relatively higher than those observed in other study areas, varied from 559.82 ± 5.44 to 1599.45 ± 45 $Bq.kg^{-1}$ with an average value of 825.38 ± 2.78 $Bq.kg^{-1}$. The R_{eq} for the study area E varied from 249.78 ± 5.97 to 1230.21 ± 10.29 $Bq.kg^{-1}$ with an average value of 576.21 ± 3.00 $Bq.kg^{-1}$. The study found the average R_{eq} for the study areas D and E to be greater than the permissible value of $370 Bq.kg^{-1}$. Therefore, soil and earth derived materials from those areas should not be used for construction of habitable infrastructures, as this might have the potential to give rise to stochastic effects. There should be regulatory restrictions on the utilisation of tailing's material and soil from areas in the vicinity of RUM for construction of habitable structures.

4.3.7 Residual activity concentrations of ^{40}K , ^{238}U and ^{232}Th in the root's zone and the resultant committed dose due to ingestion of food crops using the NORMALYSA mathematical model

The average activity concentrations and transfer factors of ^{238}U , ^{232}Th and ^{40}K for samples from the cropland were fed into the NORMALYSA model to estimate the residual radioactivity in the root zone and subsequent committed dose for the next 500 years. The modelling predicted the progressive decrease on the residual activity concentrations of ^{238}U , ^{232}Th and ^{40}K in soil root zones to about 0.54, 13.69 and 0.10 $Bq.kg^{-1}$, respectively by the year 2520 as is illustrated in Figure 4.12. The ^{238}U , ^{232}Th and ^{40}K have half-lives of billions of years, therefore, the predicted decreases in activity concentrations would not be a result of radioactive decay but due to leaching by infiltration flow and bioturbation activities. The activity concentrations for both ^{238}U and ^{40}K were predicted to decrease faster than those of ^{232}Th because the adsorption

distribution coefficient (K_d) of the latter was relative greater than that of the formers (Avila *et al.*, 2018).

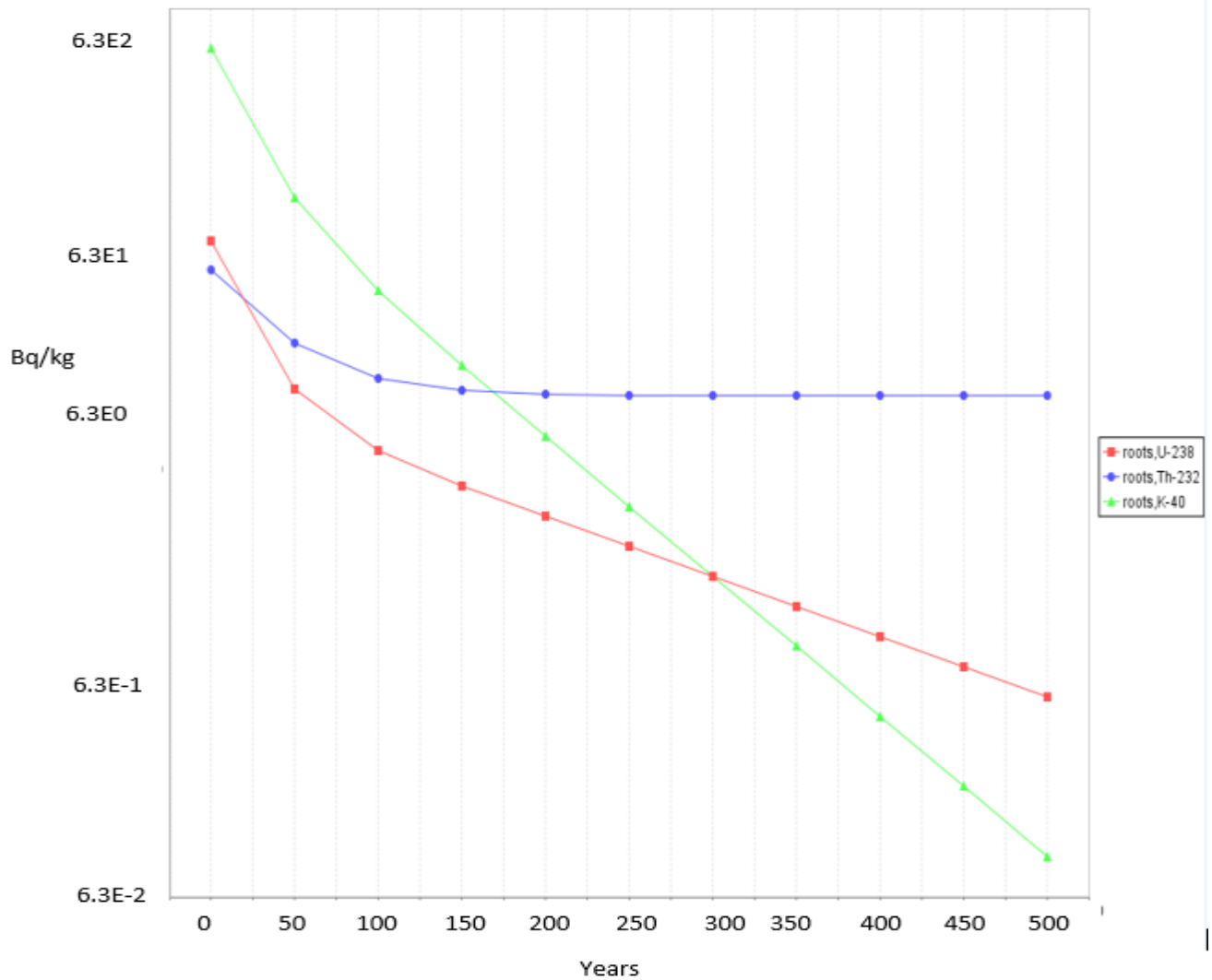


Figure 4. 12: The simulated residual activity concentrations of ^{238}U , ^{232}Th and ^{40}K in root zones up to 500 years.

The committed doses from ingestion of food crops for each radionuclide for the next 500 years are illustrated in Figure 4.13. The committed doses decreased in time in relation to the decreases of the primordial radionuclides' activity concentrations in the soil rooting zone. The NORMALYSA further illustrated that the contribution of ^{238}U and ^{232}Th to the dose would be insignificant in relation to ^{40}K .

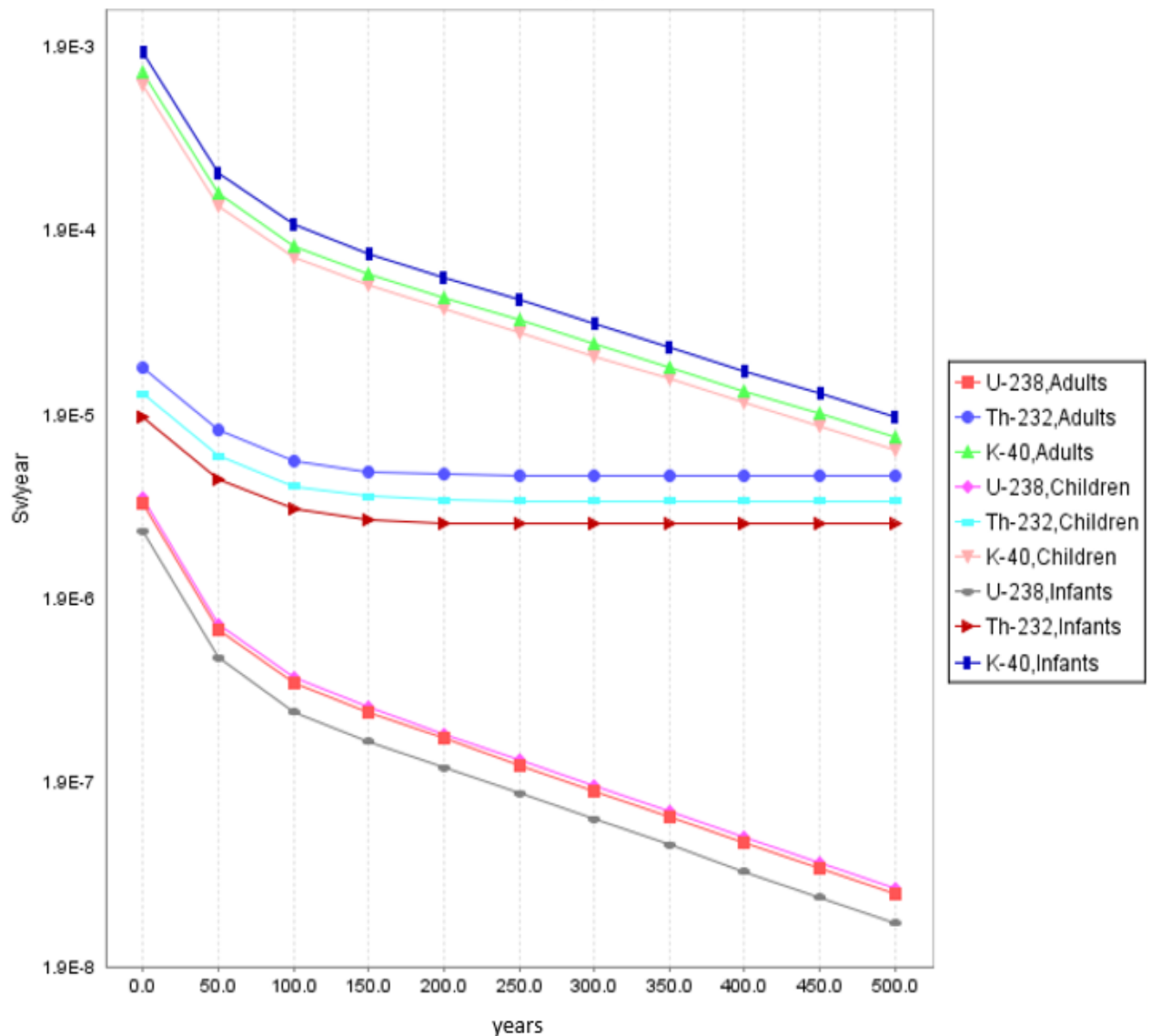


Figure 4. 13: Doses from ingestion of all food crop types for each radionuclide for the next 500 years.

Figure 4.14 is a simulation chart from the NORMALYSA model, illustrating the effective committed dose from ingestion of all food crop types summed overall radionuclides. It shows that the age category 5 years would be the most exposed among the three followed by the adult category. The age category 15 years would be the least exposed and this scenario could be attributed to the greater elimination rates from tissues and excretion of radionuclides associated with the 15 years of age category (ICRP, 2012b).

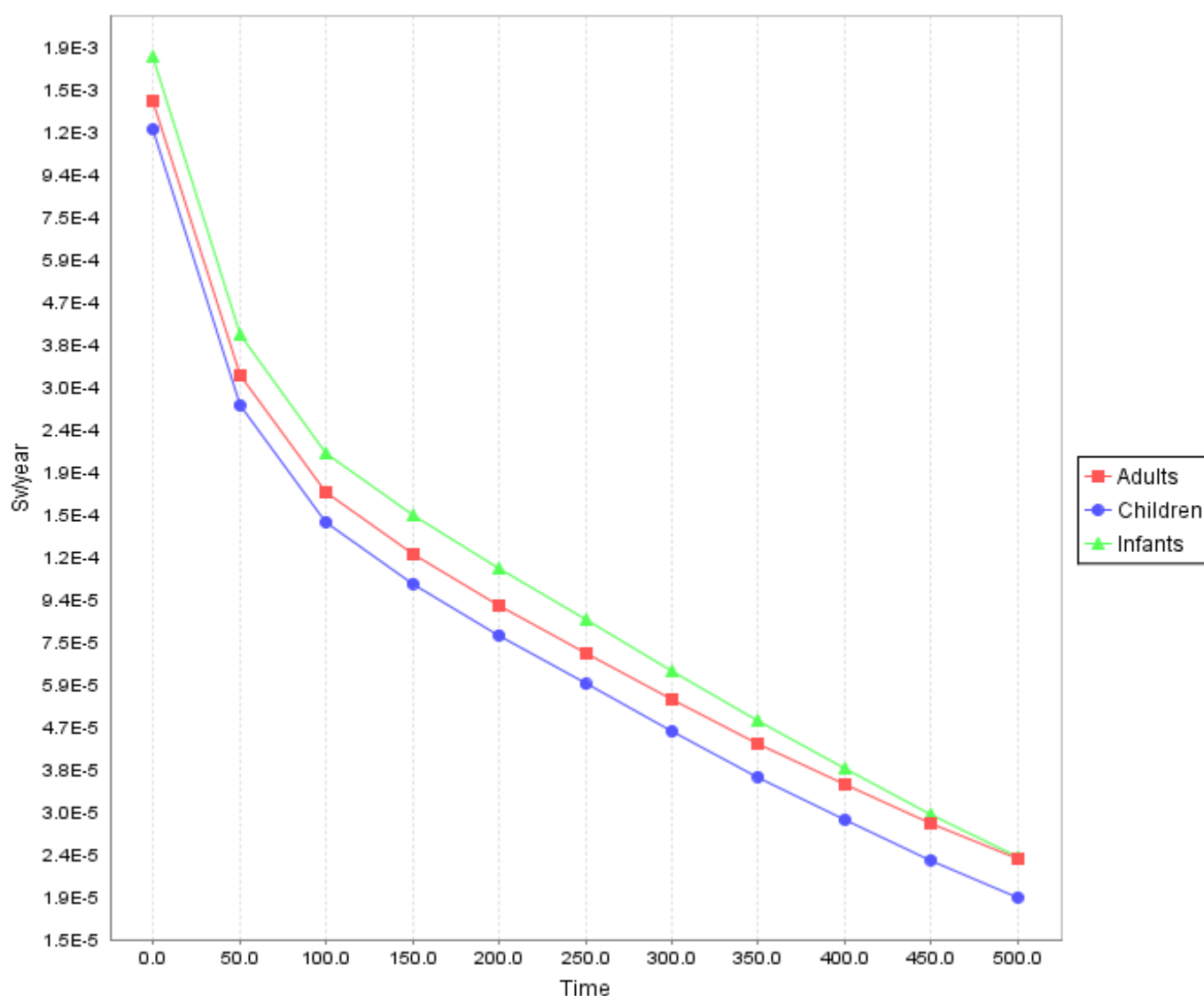


Figure 4. 14: Committed dose from ingestion of all food crop types of summed overall radionuclides for the next 500 years.

A consistency was observed between the annual average committed effective doses, presented in Table 4.12, estimated for time 0.00 year (first year) using equation 3.12 to those predicted by the NORMALYSA software model, as presented in Figures 4.13 and 4.14.

4.3.8 Cancer morbidity risk due to external exposure from ^{238}U , ^{232}Th , ^{226}Ra , ^{210}Pb and ^{40}K

The RESRAD-Onsite computer code was used to predict the cancer morbidity risks for the study areas A and D, the results are presented in Tables 4.13 and 4.14, respectively.

Table 4. 13: Cancer morbidity risk for the study area A (cropland) as a result of external exposure from ^{40}K , ^{232}Th and ^{238}U summed up for the first 800 years.

Time (Years)	Individual nuclide's cancer risk			Total
	^{40}K	^{232}Th	^{238}U	
1	1.251E-04	3.022E-04	4.778E-06	4.320E-04
5.0	9.899E-05	3.464E-04	4.653E-06	4.501E-04
6.3	9.177E-05	3.574E-04	4.613E-06	4.537E-04
8.0	8.343E-05	3.691E-04	4.563E-06	4.571E-04
10.0	7.400E-05	3.811E-04	4.501E-06	4.596E-04
13.0	6.362E-05	3.925E-04	4.424E-06	4.606E-04
16.0	5.261E-05	4.027E-04	4.330E-06	4.596E-04
20.0	4.141E-05	4.108E-04	4.213E-06	4.564E-04
25.0	3.063E-05	4.166E-04	4.071E-06	4.513E-04
31.6	2.096E-05	4.201E-04	3.899E-06	4.450E-04
40.0	1.300E-05	4.219E-04	3.693E-06	4.386E-04
50.0	7.125E-06	4.226E-04	3.449E-06	4.332E-04
63.1	3.342E-06	4.228E-04	3.164E-06	4.293E-04
79.4	1.289E-06	4.228E-04	2.839E-06	4.269E-04
100.0	3.882E-07	4.227E-04	2.477E-06	4.256E-04
158.5	1.280E-08	4.226E-04	1.680E-06	4.243E-04
200.0	1.168E-09	4.224E-04	1.279E-06	4.237E-04
316.2	1.290E-12	4.218E-04	5.894E-07	4.224E-04
400.0	1.086E-14	4.210E-04	3.422E-07	4.214E-04
500.0	2.651E-17	4.197E-04	1.727E-07	4.199E-04
631.0	1.366E-20	4.182E-04	7.352E-08	4.182E-04
795.0	9.139E-25	3.833E-04	2.395E-08	3.833E-04

The RESRAD model predicted downward trends for individual cancer risks by ^{40}K and ^{238}U for the study area A (Table 4.13), decreases from 1.251×10^{-4} and 4.778×10^{-6} in year 1 to 9.139×10^{-25} and 2.395×10^{-8} in year 795, respectively. The cancer risk by ^{232}Th was predicted to be rising during the first 63 years, increasing from 3.022×10^{-4} in year 1 to reach maximum value of 4.228×10^{-4} in year 63, thereafter, to then continue at a slightly downward trend for the next 730 years.

With reference to the study area D (Table 4.14), it was predicted that the individual cancer risks by ^{40}K and ^{238}U would be constant for the first 25 years, at 1.40×10^{-4} and 1.14×10^{-5} , thereafter, to gradually decrease to 1.25×10^{-4} and 1.12×10^{-5} by the year 1000, respectively. The individual cancer risks by ^{210}Pb and ^{226}Ra indicate downward trends, decreasing from 1.43×10^{-6} and 4.57×10^{-3} in year 1 to 4.04×10^{-20} and $2.93 \times 10^{-}$

⁰³ in year 1000, respectively. However, the cancer risk by ²³²Th was predicted to be rising during the first 50 years, increasing from 2.18×10^{-4} in year 1 to reach 3.10×10^{-4} in year 50, thereafter, to remain constant for the next 950 years. Ziajahromi *et al.*, (2015) attributed the observed initial increases in cancer risk by ²³²Th to the ingrowth of ²²⁸Ra and ²²⁸Th before attaining their circular equilibrium after about 7 to 10 half-lives.

According to the model, the cancer risk for the study area A would be dominated by ²³²Th followed by ⁴⁰K for the first 60 years, whereas for the study area D would be dominated by ²²⁶Ra, contributing to about 92% to 87% for 1000 years, followed by ²³²Th and ⁴⁰K while ²³⁸U and ²¹⁰Pb would contribute the least.

The total cancer risks estimated for the study areas A and D followed an upward trend for the first 13 and 16 years, increase from 4.320×10^{-4} and 4.937×10^{-3} on year 1 to reach total maximum cancer risks of 4.606×10^{-04} and 4.982×10^{-03} to be observed by the year 13 and 16, respectively. Then the total cancer risks for both study areas would decrease progressively to reach 3.833×10^{-4} and 3.38×10^{-3} at years 790 and 1000, respectively. The downward trends observed on cancer risks should not be construed as a result of radioactive decay but due to the vertical migration of radionuclides due to leaching, infiltration flow and bioturbation activities (Avila *et al.*, 2018).

Table 4. 14: Cancer morbidity risk for the study area D (tailings' storage facility) as a result of external exposure from ^{40}K , ^{210}Pb , ^{226}Ra , ^{232}Th and ^{238}U summed up for about 1000 years.

Years	Individual nuclide's cancer risk					Total
	^{40}K	^{210}Pb	^{226}Ra	^{232}Th	^{238}U	
1.0	1.40E-04	1.43E-06	4.57E-03	2.18E-04	1.14E-05	4.937E-03
4.0	1.40E-04	1.30E-06	4.56E-03	2.44E-04	1.14E-05	4.956E-03
5.0	1.40E-04	1.26E-06	4.56E-03	2.51E-04	1.14E-05	4.961E-03
10.0	1.40E-04	1.08E-06	4.55E-03	2.77E-04	1.14E-05	4.977E-03
13	1.40E-04	9.97E-07	4.54E-03	2.86E-04	1.14E-05	4.980E-03
16	1.40E-04	9.00E-07	4.54E-03	2.94E-04	1.14E-05	4.982E-03
20.0	1.40E-04	7.92E-07	4.53E-03	3.00E-04	1.14E-05	4.980E-03
25.0	1.40E-04	6.74E-07	4.52E-03	3.04E-04	1.14E-05	4.974E-03
50.	1.39E-04	3.09E-07	4.47E-03	3.10E-04	1.13E-05	4.929E-03
63.	1.39E-04	2.06E-07	4.44E-03	3.10E-04	1.13E-05	4.903E-03
80.	1.39E-04	1.24E-07	4.41E-03	3.10E-04	1.13E-05	4.871E-03
100.0	1.39E-04	6.50E-08	4.37E-03	3.10E-04	1.13E-05	4.831E-03
126.0	1.38E-04	2.90E-08	4.32E-03	3.10E-04	1.13E-05	4.780E-03
159.0	1.38E-04	1.05E-08	4.26E-03	3.10E-04	1.13E-05	4.718E-03
200.0	1.37E-04	2.91E-09	4.18E-03	3.10E-04	1.13E-05	4.641E-03
251.2	1.36E-04	5.79E-10	4.09E-03	3.10E-04	1.13E-05	4.545E-03
300.0	1.35E-04	1.26E-10	4.00E-03	3.10E-04	1.13E-05	4.457E-03
316.2	1.35E-04	7.59E-11	3.97E-03	3.10E-04	1.13E-05	4.428E-03
398.0	1.34E-04	5.89E-12	3.83E-03	3.10E-04	1.13E-05	4.285E-03
501.0	1.32E-04	2.35E-13	3.66E-03	3.10E-04	1.13E-05	4.113E-03
631.0	1.30E-04	4.09E-15	3.46E-03	3.10E-04	1.13E-05	3.906E-03
794.3	1.28E-04	2.49E-17	3.21E-03	3.10E-04	1.12E-05	3.663E-03
1000.0	1.25E-04	4.04E-20	2.93E-03	3.10E-04	1.12E-05	3.380E-03

WHO (2011) considers an acceptable risk that may results only in one additional case of cancer per 100 000 of the population being exposed (1×10^{-5}). The predicted total excess cancer risks for study areas A and D for the duration of about 1000 years ranged from 3.8×10^{-4} to 4.32×10^{-4} and from 3.38×10^{-3} to 4.937×10^{-3} , respectively. This implies that approximately 4 out of 10 000 and 4 to 5 out of 1000 could develop some form of cancer in any single year for the study areas A and D, respectively. The predicted cancer risks in this study, therefore, exceeded the acceptable risk recommended by WHO. It suffices to emphasise that the cancer risk assessment only considered the external exposure pathway. A comprehensive study should thus be

recommended to evaluate the contribution of other exposure pathways such as inhalation of radioactive dust or radon as well as the ingestion of food crops.

Chapter 5: Summary, conclusions and recommendations

5.1 Summary and conclusions

The community of the Swakop river and environmental activists raised concern on possible radiological contamination of the alluvial sand used in the construction industry and agricultural croplands because of the uranium mining related activities in the proximity of the lower Swakop river. This has then motivated for the study to evaluate the primordial radionuclides and radiological health impacts associated with soil, groundwater and agricultural food crops of the lower Swakop river inclusive of its main tributary, the Khan river. The study area was subdivided into 5 segments (A-E) based on the land utilisation and/or industrial activities in the neighbourhood.

To achieve the aim of the study, the soil, water and food crop samples were randomly collected from the accessible areas of the lower Swakop river and its surroundings. The activity concentrations of ^{238}U , ^{232}Th and ^{40}K in soil samples were determined using the hyper purity germanium detector. The activity concentrations of ^{238}U , ^{232}Th and ^{40}K in food crops and water samples were derived from the total mass elemental concentrations of uranium, thorium and potassium in samples that were determined by using inductively coupled plasma mass spectroscopy.

The study found a relatively great variation in the distribution of activity concentrations of ^{238}U , ^{232}Th and ^{40}K in samples amongst segments of the study areas. The average activity concentrations of ^{238}U in soil samples across the study areas varied from 69.19 ± 4.48 to 532.09 ± 11.90 Bq.kg⁻¹. The average activity concentrations of ^{232}Th in soil samples across the study areas varied from 59.07 ± 1.11 to 211.56 ± 1.85 Bq.kg⁻¹. The average activity concentrations of ^{40}K was found to be relatively higher than those of ^{238}U and ^{232}Th across the study areas, which varied from 568.73 ± 12.66 to 982.74 ± 24.56 Bq.kg⁻¹. The average activity concentrations of ^{238}U , ^{232}Th and ^{40}K in soil across all study areas were found to be higher than the world global average values reported by UNSCEAR. The study further revealed that the average activity concentrations of ^{226}Ra , ^{238}U , ^{232}Th and ^{210}Pb for the study area A (cropland) were relatively lower than those from the other segments of the study. A discernible gradual increase on the average activity concentrations of ^{226}Ra , ^{238}U , ^{232}Th and ^{210}Pb in soil was observed across the study areas as moving from the cropland (A) towards the study areas B to E.

The activity concentrations of ^{40}K in groundwater along the Swakop river were found to vary from 737.66 to 2475.98 mBq.l^{-1} with the average of $1449.22 \pm 181.85 \text{ mBq.l}^{-1}$, while for the Khan river, they varied from 923.93 to 1870.76 mBq.l^{-1} with an average of $1505.02 \pm 153.16 \text{ mBq.l}^{-1}$. The activity concentrations of ^{40}K in the water samples collected from the tailing's storage facility ponds were found to be varied from 0.72 to 1.902 Bq.l^{-1} with an average of 1.308 Bq.l^{-1} , hence there was no significant variations in the ^{40}K observed between water sources across the study areas.

The activity concentrations of ^{238}U were found to be varied from 0.161 to 1116.47 mBq.l^{-1} with an average value of $313.43 \pm 127.62 \text{ mBq.l}^{-1}$ for water samples collected along the Swakop river, and varied from 353.99 to 1875.59 mBq.l^{-1} with an average of $1282.55 \pm 230.04 \text{ mBq.l}^{-1}$ for water samples collected along the Khan river. The activity concentrations of ^{238}U observed in water samples collected from the tailings' storage facility and within mining operations were relatively higher than those observed from the other areas of study, which ranged from 22.17 to 700.07 Bq.l^{-1} with an average value of 356.05 Bq.l^{-1} .

The activity concentrations of ^{232}Th were found to range from BDL to 0.630 mBq.l^{-1} with an average value of $0.167 \pm 0.09 \text{ mBq.l}^{-1}$ for water samples along the Khan river and from 0.0044 to 50.78 Bq.l^{-1} with average value of 24.21 Bq.l^{-1} in water samples from the uranium mining operations, however, nothing was detected in water samples collected along the Swakop river.

The trends observed suggested that the distribution of activity concentrations of ^{238}U and ^{40}K in water along the Swakop and Khan rivers were not uniform. The activity concentrations of ^{238}U and ^{40}K were found to be relatively high at the agricultural cropland and the neighbourhood as well as at the confluence of the Gawib and Swakop rivers, however, lower at the upstream of the LHU activities.

In order to ascertain the potential ingestion of radionuclides through food crop consumption, the activity concentrations of ^{238}U , ^{232}Th and ^{40}K were determined. The elemental potassium is an essential element necessary for plant growth and metabolism whereas, there is no documented evidence on the beneficial roles for uranium and thorium in plant growth and biochemical processes. It turned out that food crops preferentially absorbed ^{40}K in large quantity than those of ^{238}U and ^{232}Th . This was evident in the average activity concentrations for ^{40}K of $1603.56 \pm 234.36 \text{ Bq.kg}^{-1}$

recorded for spinach, $741.27 \pm 14.26 \text{ Bq.kg}^{-1}$ for eggplant and $567.79 \text{ Bq.kg}^{-1}$ for beetroot. The average activity concentrations of ^{238}U were only found to be $1.01 \pm 0.20 \text{ Bq.kg}^{-1}$ in spinach, $0.15 \pm 0.1 \text{ Bq.kg}^{-1}$ in eggplant and 0.28 Bq.kg^{-1} in beetroot, whereas that of ^{232}Th were $0.84 \pm 0.41 \text{ Bq.kg}^{-1}$ for spinach, $0.09 \pm 0.02 \text{ Bq.kg}^{-1}$ for eggplant and 0.21 Bq.kg^{-1} for beetroot. The observed activity concentrations in the current study area were found to be higher than the reference values of 20 mBq.kg^{-1} and 3 mBq.kg^{-1} for ^{238}U and 15 mBq.kg^{-1} and 0.5 mBq.kg^{-1} for ^{232}Th , for leafy vegetables and root vegetables, respectively, published in UNSCEAR.

The TF values for primordial radionuclides in food crops to those in soil were determined. The TF values for ^{40}K were found to be varying from 2.07 to 3.25 with an average of 2.77 for spinach, from 1.35 to 2.57 with an average of 1.76 for eggplant and 0.98 for beetroot. The TF values for ^{238}U were found to be in the range of 0.0108 to 0.0185 with an average of 0.014 for spinach, 0.0035 for eggplant and 0.004 for beetroot. The TF values for ^{232}Th were found to be varying from 0.0104 to 0.0362 with an average of 0.02 for spinach, varied from 0.0018 to 0.0041 with an average of 0.003 for eggplant and 0.01 for beetroot. A TF above a unity observed in favour of ^{40}K was an indication that potassium is required in large quantities for biochemical processes in plants while low TF as observed for ^{238}U and ^{232}Th signified their non-essentialities.

The potential radiological health risks were evaluated based on the key indicators; gamma ray absorbed dose rate, external exposure to gamma radiation, internal exposure due to consumption of food crops, effective dose, excess cancer risks and radium concentration equivalent. The study found that the estimated average absorbed gamma ray dose rates at 1 meter above the ground for the study areas A and B, were $88.65 \pm 0.47 \text{ nGy.h}^{-1}$ and $100.94 \pm 0.44 \text{ nGy.h}^{-1}$, respectively, relatively higher than the global average of 60 nGy.h^{-1} . The average absorbed gamma dose rates at 1 meter above the ground for samples collected from the study areas C, D and E were 135.85 ± 0.75 , 378.73 ± 1.28 and $258.17 \pm 1.37 \text{ nGy.h}^{-1}$, respectively, relatively higher than those observed in areas A and B.

The maximum annual absorbed dose at 1 meter above the ground was estimated at 6.30 mGy, based on the maximum dose rate of $717.97 \text{ nGy.h}^{-1}$ observed at the tailings' storage facility. The maximum absorbed dose assumed of a worst-case scenario for a hypothetical individual residing at the TSF for an uninterrupted 8760 hours in a year. The maximum annual absorbed dose estimated from the current study was

significantly lower the threshold dose for tissue reaction. It could therefore be concluded that there would be no potential for any manifestation of deterministic effects due to external exposure to gamma radiation arising from the ^{238}U , ^{232}Th and ^{40}K across the study areas.

The average annual effective doses due to external exposure for the study areas A and B were 0.69 ± 0.004 and 0.80 ± 0.002 mSv.y^{-1} for the age category representative of infants, relatively higher than those of age the category representative of children with an average of 0.61 ± 0.003 and 0.71 ± 0.002 mSv.y^{-1} , respectively. The age category representative of adults would receive the least annual doses with average values of 0.54 ± 0.002 and 0.62 ± 0.002 mSv.y^{-1} , estimated for study areas A and B, respectively. The average effective dose due to external exposure for study areas A and B were greater than 0.48 mSv.y^{-1} , the global average reported by UNSCEAR, however, less than 1 mSv , a regulatory dose limit prescribed for members of the public. The average annual effective dose estimated for age group representative of an adult for the study areas C, D and E were found to be 0.84 ± 0.005 , 2.32 ± 0.01 and 1.58 ± 0.01 mSv.y^{-1} , respectively, all less than the prescribed annual limit value of 20 mSv for designated radiation workers.

The annual committed effective doses due to consumption of all food crops were estimated at 2.48 ± 0.02 , 1.66 ± 0.01 and 1.88 ± 0.02 mSv for the age categories 5 years, 15 years and adults, respectively. ^{40}K was found to contribute most to the dose at about 98%. The total annual effective dose was evaluated for the study area A, and the age category 5 years was found to be the most exposed with the total annual effective dose of 3.22 ± 0.04 mSv , followed by the age category adult, with an estimated total annual effective dose of 2.46 ± 0.04 mSv , while the age group representative of 15 years was found to be the least exposed with an estimated total annual effective dose of 2.32 ± 0.03 mSv . The total annual effective doses were relatively higher than the annual intervention exemption level of 1 mSv recommended for prolonged exposure.

The study found that, on average, the radium equivalent activities for soil samples collected from the study areas A to B, to be below the permissible limit of 370 Bq.kg^{-1} , therefore, may be used for the construction of habitable structures. The study also found the average radium equivalent activities in excess of the permissible value of 370 Bq.kg^{-1} for samples collected from study areas C to E, therefore, could increase

the chances of stochastic effects should the materials found their way into public domain. Therefore, there should be a regulatory restriction on the utilisation of tailing materials and soil from the areas in the vicinity of the RUM for construction of habitable structures.

The estimated total excess cancer risks due to external exposure, during the period of 1000 years were found to be varied from 4.32×10^{-4} down to 3.8×10^{-4} for the study area A, and from 4.937×10^{-3} down to 3.38×10^{-3} for the study area D. This implies that approximately 4 out of 10 000 and 5 out of 1000 could develop some form of cancer in any single year for the study areas A and D, respectively, an unacceptable risk by WHO, which recommended a risk value of 1×10^{-5} .

5.2 Recommendations to the regulatory body responsible for radiation protection

Noting that the soil samples considered in this study were collected at a depth of about 20 cm, it is a known fact that the excavation of sands for building and construction material is done beyond the 20 cm depth. It is therefore, recommended that a comprehensive study be considered on the activity concentrations of primordial radionuclides at several soil depths beyond 20 cm.

It is also recommended that building material fabricated from the Swakop river soil be subjected to a thorough evaluation for radioactivity contents. It is further recommended that comprehensive studies be conducted covering the dominant food crops produced in the Swakop river cropland, taking into account the actual local consumption rate and effects of boiling on estimated doses.

A comprehensive study to evaluate the cancer risk for workers at the tailing's storage facilities, considering all possible exposure pathways such as inhalation of radioactive dust, radon and external gamma rays exposure. There should be a regulatory restriction on the utilisation of tailing materials and soil from the areas in the vicinity of the RUM for construction of habitable structures.

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Annexures

Annexure A: Sampling positions in the study areas

Table A1: Soil sampling: crop fields and Swakop river (Area A and B).

Sample ID	Latitude	Longitude	Sample ID	Latitude	Longitude
Study Area A					
SEP1	-22.64007	14.64379	SOT	-22.64041	14.64306
SEP2	-22.63997	14.64395	SS1	-22.64034	14.64369
SEP3	-22.6399	14.64407	SS2	-22.63956	14.64269
SBR	-22.64011	14.64382	SS3	-22.63823	14.64267
SSP1	-22.64124	14.64363	RHB_S1	-22.63825	14.65122
SSP2	-22.6413	14.64356	RHB_S2	-22.63899	14.65329
SSP3	-22.64127	14.64361	RHB_S3	-22.6371	14.65204
SSP4	-22.63936	14.64336	RHB_S4	-22.63681	14.65322
SSP5	-22.63943	14.64306	RHB_S5	-22.63712	14.65112
SSP6	-22.63939	14.64332			
SSP7	-22.63901	14.64325			
SSP8	-22.63902	14.64321			
SSP9	-22.63902	14.64316			

Table A1: Soil sampling: crop fields and Swakop river (Area A and B) (Cont'd)

Study area B					
Sample ID	Latitude	Longitude	Sample ID	Latitude	Longitude
SWR_01	-22.65951	14.79696	SWR_11	-22.706799	15.41561
SWR_02	-22.64055	14.77689	SWR_12	-22.707313	15.413654
SWR_03	-22.64179	14.74308	SWR_13	-22.711967	15.40552
SWR_04	-22.645	14.72013	SWR_14	-22.727273	15.392519
SWR_05	-22.64535	14.69861	SWR_15	-22.729798	15.379261
SWR_06	-22.64366	14.67801	SWR_16	-22.759673	15.376999
SWR_07	-22.63575	14.65054	SWR_17	-22.751249	15.26516
SWR_08	-22.64197	14.63347	SWR_18	-22.659876	14.863532
SWR_09	-22.65079	14.61325	SWR_19	-22.668043	14.853303
SWR_10	-22.66954	14.59216	SWR_20	-22.668147	14.842537

Table A2: Soil and tailings for study areas C, D and E.

Sample ID	Study Area	Sample type	Latitude	Longitude
RU_01	C	Soil	-22.48256	15.03475
RU_02	C	Soil	-22.47994	14.99211
RU_03	C	Soil	-22.4401	15.00486
RU_04	C	Soil	-22.452833	15.04997
RU_05	C	Soil	-22.382087	15.08500
RTU_01	D	tailings	-22.45019	15.02205
RTU_02	D	tailings	-22.45275	15.02769
RTU_03	D	tailings	-22.44977	15.03105
RTU_04	D	tailings	-22.44807	15.02553
RTU_05	D	tailings	-22.44835	15.02553
RTU_06	D	tailings	-22.45821	15.03242
RTU_07	D	tailings	-22.45323	15.03748
RTU_08	D	tailings	-22.44760	15.04032
RTU_09	D	tailings	-22.45199	15.04023
RTU_10	D	tailings	-22.45195	15.03571
KHN_01	E	Soil	-22.59879	14.96385
KHN_02	E	Soil	-22.57239	14.97946
KHN_03	E	Soil	-22.54004	15.02074
KHN_04	E	Soil	-22.56266	14.99651
KHN_05	E	Soil	-22.42081	15.12234
KHN_06	E	Soil	-22.39752	15.11895

Table A3: Agricultural food crops.

Sample ID	Crop name	Latitude	Longitude
SEP1	Egg plant	-22.64007	14.64379
SEP2	Egg plant	-22.63997	14.64395
SEP3	Egg plant	-22.6399	14.64407
SBR	Beetroots	-22.64011	14.64382
SSP1	Spinach	-22.64124	14.64363
SSP2	Spinach	-22.6413	14.64356
SSP3	Spinach	-22.64127	14.64361
SSP4	Spinach	-22.63936	14.64336
SSP5	Spinach	-22.63943	14.64306
SSP6	Spinach	-22.63939	14.64332
SSP7	Spinach	-22.63901	14.64325
SSP8	Spinach	-22.63902	14.64321
SSP9	Spinach	-22.63902	14.64316

Table A4: Water sampling positions.

Item no.	Sample ID	Water source	Latitude	Longitude
1	SHW	groundwater	-22.63956	14.64269
2	RHBW	groundwater	-22.63762	14.65376
3	203547	groundwater	-22.706799	15.41561
4	41188	groundwater	-22.707313	15.413654
5	41184	groundwater	-22.727273	15.392519
6	203545	groundwater	-22.729673	15.376999
7	203546	groundwater	-22.729798	15.379261
8	41180	groundwater	-22.751249	15.26516
9	Goanikontes	groundwater	-22.668185	14.817576
10	W2_1.4A	groundwater	-22.49053	15.0819
11	W3_UK4B	groundwater	-22.47503	15.10767
12	W4_1.6A	groundwater	-22.52835	15.02934
13	GW29	groundwater	-22.598797	14.96386
14	GWN01	groundwater	-22.57239	14.97946
15	202083	groundwater	-22.540043	15.02074
16	202082	groundwater	-22.562663	14.99651
17	RU_T1	Process water	-22.4548	15.03989
18	RU_T2	Process water	-22.4552	15.04003
19	RU_T3	Process water	-22.4799	15.03656
20	W1_R3	ground water	-22.4826	15.03485

Annexure B: Activity concentrations and concentration ratios for food crops and soil

Table B1: Activity concentrations for soil samples of the study area A.

Sample ID	Activity concentrations (Bq.kg ⁻¹)				
	²²⁶ Ra	²³⁸ U	²³² Th	⁴⁰ K	²¹⁰ Pb
SSP1	34.14±0.63	69.61±5.68	49.45±0.74	562.70±11.75	BDL
SSP2	34.11±0.63	72.58±5.77	47.11±1.31	556.20±12.30	BDL
SSP3	36.01±0.69	58.50±5.80	53.59±1.35	587.10±12.97	69.23±4.08
SSP4	36.65±0.65	117.00±3.76	51.86±1.32	677.50±12.33	52.05±32.64
SSP5	32.94±0.67	65.31±5.61	41.14±0.69	562.30±12.47	36.23±26.28
SSP6	33.81±0.64	74.96±5.66	46.81±1.05	564.90±11.91	64.74±31.56
SSP7	35.62±0.67	80.23±5.78	46.72±1.27	613.00±12.61	42.39±29.50
SSP8	34.11±0.62	71.10±5.67	48.22±1.27	559.80±12.04	65.78±33.52
SSP9	34.53±0.70	66.49±5.77	50.39±1.20	564.90±11.23	70.32±32.26
SEP1	24.08±0.85	35.38±5.96	28.16±0.43	282.20±28.44	BDL
SEP2	31.76±0.69	51.18±6.14	41.85±0.73	551.10±12.44	70.40±30.76
SEP3	31.13±0.69	44.59±5.95	45.09±1.40	558.90±12.84	BDL
SBR	35.00±0.70	74.07±6.34	41.59±0.76	580.50±13.40	137.20±36.48
SOT	33.74±0.70	49.52±5.66	50.13±1.06	577.10±12.18	51.41±22.35
SS1	34.80±0.68	62.31±5.38	49.13±0.66	559.90±11.57	38.47±19.32
SS2	35.71±0.64	127.70±5.80	55.25±1.25	575.30±11.75	BDL
SS3	37.95±0.62	70.88±5.37	56.70±1.03	589.10±11.97	92.31±27.67
RHB_S1	33.66±0.62	47.68±5.27	51.11±1.21	616.70±12.25	BDL
RHB_S2	36.97±0.66	60.15±5.29	53.29±1.30	561.90±8.49	81.61±26.01
RHB_S3	43.05±0.56	96.03±2.50	58.86±0.84	383.70±7.05	64.58±10.08
RHB_S4	52.13±0.76	84.26±5.54	81.58±1.46	723.90±13.81	72.95±30.38
RHB_S5	54.06±0.74	101.65±5.07	109.97±1.30	703.40±12.71	84.64±31.81
Minimum	24.08±0.85	35.38±5.96	28.16±0.43	282.20±28.44	BDL
Maximum	54.06±0.74	127.70±5.80	109.97±1.30	723.90±13.81	137.20±36.48
Average	36.18±1.37	71.87±4.85	52.64±3.41	568.73±19.55	62.67±6.74

Table B2: Activity concentrations for the soil samples of the Area B.

Sample ID	Activity concentrations (Bq.kg ⁻¹) Study Area B				
	²²⁶ Ra	²³⁸ U	²³² Th	⁴⁰ K	²¹⁰ Pb
SWR_01	24.44±0.55	101.65±4.27	43.49±0.99	593.90±11.09	77.28±23.24
SWR_02	25.23±0.55	41.40±4.73	45.03±1.04	595.40±11.38	BDL
SWR_03	51.76±0.63	56.48±2.33	52.86±0.56	1003.00±15.82	37.36±12.11
SWR_04	38.52±0.64	38.65±4.98	79.07±1.18	558.00±11.15	BDL
SWR_05	34.27±0.61	71.80±4.50	51.40±0.97	624.70±11.54	44.43±25.27
SWR_06	51.41±0.59	58.08±6.19	68.17±0.87	798.60±12.73	14.55±1.90
SWR_07	15.50±0.63	36.45±2.58	52.59±0.48	744.20±12.76	BDL
SWR_08	36.99±0.68	64.41±5.39	57.15±1.32	658.90±13.22	BDL
SWR_09	41.41±0.58	49.58±1.62	144.10±1.50	911.10±14.36	BDL
SWR_10	27.02±0.55	89.29±4.51	49.44±1.07	494.50±10.09	BDL
SWR_11	17.62±0.66	31.72±1.61	23.45±0.81	393.90±8.89	104.00±35.50
SWR_12	31.15±0.60	40.20±4.66	48.42±1.21	555.50±10.87	BDL
SWR_13	42.51±0.62	81.79±4.81	71.49±1.27	664.10±7.50	61.71±34.17
SWR_14	29.18±0.53	54.82±4.52	42.78±1.05	483.00±10.00	74.37±28.79
SWR_15	28.11±0.56	41.67±4.26	33.97±0.54	580.90±11.09	37.04±3.31
SWR_16	26.65±0.52	54.03±4.36	41.28±1.03	462.70±9.51	BDL
SWR_17	48.40±0.62	56.87±4.50	32.04±0.56	737.40±12.84	44.20±21.88
SWR_18	75.25±0.80	111.41±5.10	139.23±1.65	987.20±15.98	156.50±16.06
SWR_19	138.08±1.31	134.96±7.54	146.35±1.41	781.00±16.11	352.50±64.88
SWR_20	182.83±1.48	168.51±7.23	128.04±1.26	893.90±17.20	286.80±53.79
Minimum	15.50±0.63	31.72±1.61	23.45±0.81	393.90±8.89	BDL
Maximum	182.83±1.48	168.51±7.23	146.35±1.41	1003.00±15.82	352.50±64.88
Average	48.32±9.25	69.19±8.06	67.52±8.77	676.10±9.41	107.56±30.80

Table B3: Activity concentrations for soil samples of the area C.

Sample ID	Activity concentrations (Bq.kg ⁻¹)				
	²²⁶ Ra	²³⁸ U	²³² Th	⁴⁰ K	²¹⁰ Pb
KHN_01	76.27±0.91	118.12±6.40	82.35±1.00	967.70±17.24	116.70±49.57
KHN_02	64.93±0.75	104.57±5.01	119.68±1.30	738.50±13.08	82.14±27.71
KHN_03	47.72±0.66	96.78±4.92	88.07±1.30	817.70±13.79	77.54±32.4
KHN_04	40.86±0.63	79.03±5.09	84.25±1.44	878.50±14.73	57.83±33.33
KHN_05	123.05±1.21	155.30±7.64	120.69±1.28	859.30±17.2	274.80±50.88
KHN_06	37.81±0.59	64.24±4.47	43.15±0.57	642.50±11.63	66.52±25.49
Min	37.81±0.59	64.24±4.47	43.15±0.57	642.50±11.63	57.83±33.33
Max	123.05±1.21	155.30±7.64	120.69±1.28	967.70±17.24	274.80±50.88
Average	65.11±0.82	103.01±5.70	89.70±1.09	817.37±14.57	112.59±14.57

Table B4: Activity concentrations for tailings material for the area D.

Sample ID	Activity concentrations (Bq.kg ⁻¹)				
	²²⁶ Ra	²³⁸ U	²³² Th	⁴⁰ K	²¹⁰ Pb
RTU_01	3021.83±11.74	703.09±13.10	117.25±1.34	1060.00±23.97	3027.00±67.90
RTU_02	3670.32±14.08	1271.58±17.95	149.01±1.66	971.30±27.24	3593.00±77.15
RTU_03	2235.62±8.90	345.01±9.81	167.60±1.49	912.90±21.02	2090.00±52.90
RTU_04	1704.98±6.90	530.00±10.23	92.68±1.20	1117.00±21.95	1610.00±44.47
RTU_05	1861.16±7.51	531.71±10.43	101.24±1.26	1081.00±21.98	1707.00±46.37
RTU_06	10360.68±37.51	599.69±16.49	346.15±2.73	619.40±35.19	10230.00±170.20
RTU_07	1810.93±7.35	317.67±9.21	122.54±1.23	1032.00±21.49	1887.00±48.60
RTU_08	2364.99±9.28	259.55±9.21	172.50±1.58	944.80±22.10	2121.00±52.930
RTU_09	1632.98± 6.69	303.58±8.75	121.67±1.26	1068.00±21.38	1462.00±43.47
RTU_10	2046.57±8.22	459.03±10.42	131.17±1.37	1021.00±21.21	1817.00±50.02
Min	1632.98 ±6.69	259.55±9.21	92.68±1.20	619.40±35.19	1462.00±43.47
Max	10360.68±37.51	1271.58±17.95	346.15±2.73	1117.00±21.95	10230.00±170.20
Average	3071.01 ±13.53	532.09±11.90	152.18±1.59	982.74±24.56	2954.40±72.31

Table B5: Activity concentrations for soil samples of the area E.

Sample ID	Activity concentrations (Bq.kg ⁻¹)				
	²²⁶ Ra	²³⁸ U	²³² Th	⁴⁰ K	²¹⁰ Pb
RU_01	126.28±1.03	171.50±5.66	144.82±1.12	725.00±12.97	79.69±39.34
RU_02	215.66±1.37	239.76±6.55	215.15±1.51	582.10±12.04	172.00±47.05
RU_03	94.43±1.06	442.61±9.20	521.85±3.15	537.00±12.52	BDL
RU_04	161.28±1.11	185.67±5.96	100.30±1.02	588.40±11.88	165.20±39.35
RU_05	47.80±0.73	80.94±5.46	75.66±1.49	787.60±14.25	86.65±35.02
Min	47.80±0.73	80.94±5.46	75.66±1.49	537.00±12.52	BDL
Max	215.66±1.37	442.61±9.20	521.85±3.15	787.60±14.25	172.00±47.05
Average	129.09±1.06	224.10±6.07	211.56±1.85	644.02±12.92	125.88±41.56

Table B6: Activity concentrations ratios of ²³⁸U/²²⁶Ra and ²³²Th/²³⁸U for study area A.

Sample ID	²³⁸ U/ ²²⁶ Ra	²³² Th/ ²³⁸ U
SSP1	2.04±0.17	0.71±0.06
SSP2	2.13±0.17	0.65±0.05
SSP3	1.62±0.16	0.92±0.09
SSP4	3.19±0.12	0.44±0.02
SSP5	1.98±0.18	0.63±0.06
SSP6	2.22±0.17	0.62±0.05
SSP7	2.25±0.17	0.58±0.04
SSP8	2.08±0.17	0.68±0.06
SSP9	1.93±0.17	0.76±0.07
SEP1	1.47±0.25	0.80±0.13
SEP2	1.61±0.20	0.82±0.10
SEP3	1.43±0.19	1.01±0.14
SBR	2.12±0.19	0.56±0.05
SOT	1.47±0.17	1.01±0.12
SS1	1.79±0.16	0.79±0.07
SS2	3.58±0.17	0.43±0.02
SS3	1.87±0.14	0.80±0.06
RHB_S1	1.42±0.16	1.07±0.12
RHB_S2	1.63±0.15	0.89±0.08
RHB_S3	2.23±0.06	0.61±0.02
RHB_S4	1.62±0.11	0.97±0.07
RHB_S5	1.88±0.10	1.08±0.06
Minimum	1.42±0.16	0.43±0.02
Maximum	3.58±0.17	1.08±0.06
Average	1.98±0.11	0.77±0.04

Table B7: Activity concentrations ratios of $^{238}\text{U}/^{226}\text{Ra}$ and $^{232}\text{Th}/^{238}\text{U}$ for study area B.

Sample ID	$^{238}\text{U}/^{226}\text{Ra}$	$^{232}\text{Th}/^{238}\text{U}$
SWR_01	4.16±0.20	0.43±0.02
SWR_02	1.64±0.19	1.09±0.13
SWR_03	1.09±0.05	0.94±0.04
SWR_04	1.00±0.13	2.05±0.27
SWR_05	2.09±0.14	0.72±0.05
SWR_06	1.13±0.12	1.17±0.13
SWR_07	2.35±0.19	1.44±0.10
SWR_08	1.74±0.15	0.89±0.08
SWR_09	1.20±0.04	2.91±0.10
SWR_10	3.31±0.18	0.55±0.03
SWR_11	1.80±0.11	0.74±0.05
SWR_12	1.29±0.15	1.20±0.14
SWR_13	1.92±0.12	0.87±0.05
SWR_14	1.88±0.16	0.78±0.07
SWR_15	1.48±0.15	0.82±0.08
SWR_16	2.03±0.17	0.76±0.06
SWR_17	1.18±0.09	0.56±0.05
SWR_18	1.48±0.07	1.25±0.06
SWR_19	0.98±0.06	1.08±0.06
SWR_20	0.92±0.04	0.76±0.03
Minimum	0.92±0.04	0.43±0.02
Maximum	4.16±0.20	2.91±0.10
Average	1.73±0.18	1.05±0.13

Table B8: Activity concentrations ratios of $^{238}\text{U}/^{226}\text{Ra}$ and $^{232}\text{Th}/^{238}\text{U}$ for study area C.

Sample ID	$^{238}\text{U}/^{226}\text{Ra}$	$^{232}\text{Th}/^{238}\text{U}$
KHN_01	1.55±0.09	0.70±0.04
KHN_02	1.61±0.08	1.14±0.06
KHN_03	2.03±0.11	0.91±0.05
KHN_04	1.93±0.13	1.07±0.07
KHN_05	1.26±0.06	0.78±0.04
KHN_06	1.70±0.12	0.67±0.05
Minimum	1.26±0.063	0.67±0.05
Maximum	2.03±0.11	1.14±0.06
Average	1.68±0.11	0.88±0.08

Table B9: Activity concentrations ratios of $^{238}\text{U}/^{226}\text{Ra}$, $^{232}\text{Th}/^{238}\text{U}$ and $^{226}\text{Ra}/^{210}\text{Pb}$ for study area D.

Sample ID	$^{238}\text{U}/^{226}\text{Ra}$	$^{232}\text{Th}/^{238}\text{U}$	$^{226}\text{Ra}/^{210}\text{Pb}$
RTU_01	0.23±0.00	0.17±0.00	1.00±0.02
RTU_02	0.35±0.01	0.12±0.00	1.02±0.02
RTU_03	0.15±0.00	0.49±0.01	1.07±0.03
RTU_04	0.31±0.01	0.17±0.00	1.06±0.03
RTU_05	0.29±0.01	0.19±0.00	1.09±0.03
RTU_06	0.06±0.00	0.58±0.02	1.01±0.02
RTU_07	0.18±0.01	0.39±0.01	0.96±0.02
RTU_08	0.11±0.00	0.66±0.02	1.11±0.03
RTU_09	0.19±0.01	0.40±0.01	1.12±0.03
RTU_10	0.22±0.01	0.29±0.01	1.13±0.03
Minimum	0.06±0.00	0.12±0.00	0.96±0.02
Maximum	0.35±0.01	0.66±0.02	1.13±0.03
Average	0.21±0.03	0.34±0.06	1.06±0.02

Table B10: Activity concentrations ratios of $^{238}\text{U}/^{226}\text{Ra}$ and $^{232}\text{Th}/^{238}\text{U}$ for study area E.

Sample ID	$^{238}\text{U}/^{226}\text{Ra}$	$^{232}\text{Th}/^{238}\text{U}$
RU_01	1.36±0.05	0.84±0.03
RU_02	1.11±0.03	0.90±0.03
RU_03	4.69±0.11	1.18±0.03
RU_04	1.15±0.04	0.54±0.02
RU_05	1.69±0.12	0.93±0.07
Minimum	1.11±0.03	0.54±0.02
Maximum	4.69±0.11	1.18±0.03
Average	2.00±0.68	0.88±0.10

Annexure C: Absorbed dose rate in air, annual effective dose and radium equivalent concentrations

Table C1: The estimated absorbed dose rates at 1 meter above the ground for the external exposure and the corresponding annual effective dose in an indoor and outdoor environment for age category representative of an adult, child and infant for the study areas A and B.

Sample No	D (nGy.h ⁻¹)	AED Outdoor exposure (mSv.y ⁻¹)			AED Indoor exposure (mSv.y ⁻¹)		
		Adult	Child	Infant	Adult	Child	Infant
SSP1	85.49±2.71	0.105±0.003	0.12±0.004	0.13±0.0043	0.42±0.01	0.48±0.02	0.54±0.02
SSP2	85.18±2.83	0.104±0.003	0.12±0.004	0.13±0.0045	0.42±0.01	0.48±0.02	0.54±0.02
SSP3	83.88±2.85	0.103±0.003	0.12±0.004	0.13±0.0045	0.41±0.01	0.47±0.02	0.53±0.02
SSP4	113.63±1.98	0.139±0.002	0.16±0.003	0.18±0.0031	0.56±0.01	0.64±0.01	0.72±0.01
SSP5	78.47±2.68	0.096±0.003	0.11±0.004	0.12±0.0042	0.38±0.01	0.44±0.02	0.50±0.02
SSP6	86.46±2.74	0.106±0.003	0.12±0.004	0.14±0.0043	0.42±0.01	0.48±0.02	0.55±0.02
SSP7	90.85±2.83	0.111±0.003	0.13±0.004	0.14±0.0045	0.45±0.01	0.51±0.02	0.57±0.02
SSP8	85.32±2.68	0.105±0.003	0.12±0.004	0.13±0.0042	0.42±0.01	0.48±0.02	0.54±0.02
SSP9	84.71±2.80	0.104±0.003	0.12±0.004	0.13±0.0044	0.42±0.01	0.47±0.02	0.53±0.02
SEP1	45.12±3.01	0.055±0.004	0.06±0.004	0.07±0.0047	0.22±0.01	0.25±0.02	0.28±0.02
SEP2	71.90±2.92	0.088±0.004	0.10±0.004	0.11±0.0046	0.35±0.01	0.40±0.02	0.45±0.02
SEP3	71.14±2.93	0.087±0.004	0.10±0.004	0.11±0.0046	0.35±0.01	0.40±0.02	0.45±0.02
S_BR	83.54±3.02	0.102±0.004	0.12±0.004	0.13±0.0048	0.41±0.01	0.47±0.02	0.53±0.02
S_OT	77.23±2.74	0.095±0.003	0.11±0.004	0.12±0.0043	0.38±0.01	0.43±0.02	0.49±0.02
SH_S1	81.81±2.56	0.100±0.003	0.11±0.004	0.13±0.0040	0.40±0.01	0.46±0.01	0.52±0.02
SH_S2	116.36±2.83	0.14±0.003	0.16±0.004	0.18±0.0045	0.57±0.01	0.65±0.02	0.73±0.02
SH_S3	91.56±2.61	0.112±0.003	0.13±0.004	0.14±0.0041	0.45±0.01	0.51±0.01	0.58±0.02
RHB_S1	78.62±2.59	0.096±0.003	0.11±0.004	0.12±0.0041	0.39±0.01	0.44±0.01	0.50±0.02
RHB_S2	77.65±2.59	0.095±0.003	0.11±0.004	0.12±0.0041	0.38±0.01	0.44±0.01	0.49±0.02
RHB_S3	79.34±1.30	0.097±0.002	0.11±0.002	0.13±0.0020	0.39±0.01	0.44±0.01	0.50±0.01

Table C1: (Cont'd)

RHB_S4	123.83±2.77	0.152±0.003	0.17±0.004	0.20±0.0044	0.61±0.01	0.69±0.02	0.78±0.02
RHB_S5	134.68±2.53	0.165±0.003	0.19±0.003	0.21±0.0040	0.66±0.01	0.76±0.01	0.85±0.02
SWR_01	97.99±2.11	0.120±0.003	0.14±0.003	0.15±0.0033	0.48±0.01	0.55±0.01	0.62±0.01
SWR_02	71.15±2.32	0.087±0.003	0.10±0.003	0.11±0.0037	0.35±0.01	0.40±0.01	0.45±0.01
SWR_03	99.84±1.31	0.122±0.002	0.14±0.002	0.16±0.0021	0.49±0.01	0.56±0.01	0.63±0.01
SWR_04	88.88±2.45	0.109±0.003	0.12±0.003	0.14±0.0039	0.44±0.01	0.50±0.01	0.56±0.02
SWR_05	90.27±2.21	0.111±0.003	0.13±0.003	0.14±0.0035	0.44±0.01	0.51±0.01	0.57±0.01
SWR_06	101.31±2.96	0.124±0.004	0.14±0.004	0.16±0.0047	0.50±0.01	0.57±0.02	0.64±0.02
SWR_07	79.63±1.34	0.098±0.002	0.11±0.002	0.13±0.0021	0.39±0.01	0.45±0.01	0.50±0.01
SWR_08	91.75±2.67	0.113±0.003	0.13±0.004	0.14±0.0042	0.45±0.01	0.51±0.01	0.58±0.02
SWR_09	147.94±1.32	0.181±0.002	0.21±0.002	0.23±0.0021	0.73±0.01	0.83±0.01	0.93±0.01
SWR_10	91.74±2.22	0.113±0.003	0.13±0.003	0.14±0.0035	0.45±0.01	0.51±0.01	0.58±0.01
SWR_11	45.24±0.96	0.055±0.001	0.06±0.001	0.07±0.0015	0.22±0.00	0.25±0.01	0.29±0.01
SWR_12	70.98±2.32	0.087±0.003	0.10±0.003	0.11±0.0037	0.35±0.01	0.40±0.01	0.45±0.01
SWR_13	108.66±2.37	0.133±0.003	0.15±0.003	0.17±0.0037	0.53±0.01	0.61±0.01	0.69±0.01
SWR_14	71.31±2.22	0.087±0.003	0.10±0.003	0.11±0.0035	0.35±0.01	0.40±0.01	0.45±0.01
SWR_15	63.99±2.05	0.078±0.003	0.09±0.003	0.10±0.0032	0.31±0.01	0.36±0.01	0.40±0.01
SWR_16	69.19±2.15	0.085±0.003	0.10±0.003	0.11±0.0034	0.34±0.01	0.39±0.01	0.44±0.01
SWR_17	76.38±2.17	0.094±0.003	0.11±0.003	0.12±0.0034	0.37±0.01	0.43±0.01	0.48±0.01
SWR_18	176.73±2.64	0.217±0.003	0.25±0.004	0.28±0.0042	0.87±0.01	0.99±0.01	1.12±0.02
SWR_19	183.32±3.65	0.225±0.004	0.26±0.005	0.29±0.0058	0.90±0.02	1.03±0.02	1.16±0.02
SWR_20	192.47±3.50	0.236±0.004	0.27±0.005	0.30±0.0055	0.94±0.02	1.08±0.02	1.21±0.02
Min	45.12±3.01	0.055±0.004	0.06±0.001	0.07±0.0015	0.22±0.01	0.25±0.02	0.28±0.02
Max	192.47±3.50	0.236±0.004	0.27±0.005	0.30±0.0055	0.94±0.02	1.08±0.02	1.21±0.02
Average	93.94±4.97	0.115±0.006	0.13±0.007	0.15±0.008	0.46±0.02	0.53±0.03	0.59±0.03

Table C2: The estimated absorbed dose rates for the external exposure in an indoor and outdoor environment and the corresponding annual effective dose for age category representative of an adult, child and infant for the study areas C to E.

Sample No	D (nGy.h ⁻¹)	AED Outdoor exposure (mSv.y ⁻¹)			AED Indoor exposure (mSv.y ⁻¹)		
		Adult	Child	Infant	Adult	Child	Infant
RU_01	196.94±2.75	0.24±0.003	0.28±0.004	0.31±0.004	0.97±0.01	1.10±0.02	1.27±0.02
RU_02	264.10±3.20	0.32±0.004	0.37±0.005	0.42±0.005	1.30±0.02	1.49±0.02	1.71±0.02
RU_03	542.08±4.68	0.66±0.006	0.76±0.007	0.85±0.007	2.66±0.02	3.04±0.03	3.50±0.03
RU_04	170.90±2.86	0.21±0.004	0.24±0.004	0.27±0.005	0.84±0.01	0.96±0.02	1.10±0.02
RU_05	115.93±2.74	0.14±0.003	0.16±0.004	0.18±0.004	0.57±0.01	0.65±0.02	0.75±0.02
RTU_01	439.85±6.18	0.54±0.008	0.62±0.009	0.69±0.009	2.16±0.03	2.47±0.03	2.84±0.04
RTU_02	717.97±8.43	0.88±0.010	1.01±0.012	1.13±0.013	3.52±0.04	4.03±0.05	4.63±0.05
RTU_03	298.69±4.70	0.37±0.006	0.42±0.007	0.47±0.007	1.47±0.02	1.67±0.03	1.93±0.03
RTU_04	347.42±4.87	0.43±0.006	0.49±0.007	0.55±0.008	1.70±0.02	1.95±0.03	2.24±0.03
RTU_05	351.88±4.96	0.43±0.006	0.49±0.007	0.55±0.008	1.73±0.02	1.97±0.03	2.27±0.03
RTU_06	511.96±7.93	0.63±0.009	0.72±0.011	0.81±0.013	2.51±0.04	2.87±0.04	3.30±0.05
RTU_07	263.81±4.41	0.32±0.005	0.37±0.006	0.42±0.007	1.29±0.02	1.48±0.02	1.70±0.03
RTU_08	263.50±4.46	0.32±0.005	0.37±0.006	0.42±0.007	1.29±0.02	1.48±0.02	1.70±0.03

Table C2: The estimated absorbed dose rates for the external exposure in an indoor and outdoor environment and the corresponding annual effective dose for age category representative of an adult, child and infant for the study areas C to E (cont'd).

RTU_09	258.28±4.21	0.32±0.0052	0.36±0.0059	0.41±0.0066	1.27±0.02	1.45±0.02	1.67±0.03
RTU_10	333.88±4.96	0.41±0.0061	0.47±0.0070	0.53±0.0078	1.64±0.02	1.87±0.03	2.16±0.03
KHN_01	144.67±3.10	0.18±0.0038	0.20±0.0043	0.23±0.0049	0.71±0.02	0.81±0.02	0.93±0.02
KHN_02	151.40±2.51	0.19±0.0031	0.21±0.0035	0.24±0.0040	0.74±0.01	0.85±0.01	0.98±0.02
KHN_03	132.00±2.47	0.16±0.0030	0.19±0.0035	0.21±0.0039	0.65±0.01	0.74±0.01	0.85±0.02
KHN_04	124.03±2.58	0.15±0.0032	0.17±0.0036	0.20±0.0041	0.61±0.01	0.70±0.01	0.80±0.02
KHN_05	180.48±3.68	0.22±0.0045	0.25±0.0052	0.28±0.0058	0.89±0.02	1.01±0.02	1.16±0.02
KHN_06	82.53±2.15	0.10±0.0026	0.12±0.0030	0.13±0.0034	0.40±0.01	0.46±0.01	0.53±0.01
Minimum	82.53±2.15	0.10±0.0026	0.12±0.003	0.13±0.0034	0.40±0.01	0.46±0.01	0.53±0.01
Maximum	717.97±8.43	0.88±0.0103	1.01±0.0118	1.13±0.0133	3.52±0.04	4.03±0.05	4.63±0.05
Average	280.63±35.51	0.34±0.04	0.39±0.05	0.44±0.06	1.38±0.17	1.57±0.20	1.81±0.23

Table C3: The Results of the total annual effective dose for the external exposure in an indoor and outdoor environment for the age category representative of an adult, child and infant study area A.

Sample no.	AED Adult (mSv.y⁻¹)	AED Child (mSv.y⁻¹)	AED Infant (mSv.y⁻¹)	R_{eq} (Bq.kg⁻¹)
SSP1	0.52±0.01	0.60±0.02	0.67±0.02	183.65±5.85
SSP2	0.52±0.01	0.60±0.02	0.67±0.02	182.77±6.14
SSP3	0.51±0.01	0.59±0.02	0.66±0.02	180.34±6.19
SSP4	0.70±0.01	0.80±0.01	0.90±0.01	243.33±4.31
SSP5	0.48±0.01	0.55±0.02	0.62±0.02	167.44±5.78
SSP6	0.53±0.01	0.61±0.02	0.68±0.02	185.40±5.93
SSP7	0.56±0.01	0.64±0.02	0.72±0.02	194.25±6.13
SSP8	0.52±0.01	0.60±0.02	0.67±0.02	180.31±5.82
SSP9	0.52±0.01	0.59±0.02	0.67±0.02	182.05±6.08
SEP1	0.28±0.02	0.32±0.02	0.36±0.02	97.37±6.38
SEP2	0.44±0.02	0.50±0.02	0.57±0.02	153.46±6.30
SEP3	0.44±0.02	0.50±0.02	0.56±0.02	152.11±6.36
S_BR	0.51±0.02	0.59±0.02	0.66±0.02	178.23±6.52
S_OT	0.47±0.01	0.54±0.02	0.61±0.02	165.65±5.65
SH_S1	0.50±0.01	0.57±0.02	0.65±0.02	175.68±2.29
SH_S2	0.71±0.01	0.82±0.02	0.92±0.02	251.00±3.64
SH_S3	0.56±0.01	0.64±0.02	0.72±0.02	197.33±5.06
RHB_S1	0.48±0.01	0.55±0.02	0.62±0.02	168.25±5.62
RHB_S2	0.48±0.01	0.54±0.02	0.61±0.02	179.63±5.65
RHB_S3	0.49±0.01	0.56±0.01	0.63±0.01	209.74±2.83
RHB_S4	0.76±0.01	0.87±0.02	0.98±0.02	256.65±6.02
RHB_S5	0.83±0.01	0.94±0.02	1.06±0.02	313.06±5.49
Minimum	0.28±0.02	0.32±0.02	0.36±0.02	97.37±6.38
Maximum	0.83±0.01	0.94±0.02	1.06±0.02	313.06±5.49
Average	0.54±0.002	0.61±0.003	0.69±0.004	190.81±1.02

Table C4: The Results of the total annual effective dose for the external exposure in an indoor and outdoor environment for the age category representative of an adult, child and infant study area B.

Sample no.	AED Adult (mSv.y-1)	AED Child (mSv.y-1)	AED Infant (mSv.y-1)	R_{eq} (Bq.kg-1)
SWR_01	0.60±0.01	0.69±0.01	0.77±0.01	209.57±4.58
SWR_02	0.44±0.01	0.50±0.01	0.56±0.02	151.63±5.03
SWR_03	0.61±0.01	0.70±0.01	0.79±0.01	209.30±2.75
SWR_04	0.55±0.01	0.62±0.01	0.70±0.02	194.68±5.33
SWR_05	0.55±0.01	0.63±0.01	0.71±0.01	193.40±4.80
SWR_06	0.62±0.02	0.71±0.02	0.80±0.02	217.05±6.39
SWR_07	0.49±0.01	0.56±0.01	0.63±0.01	168.95±2.84
SWR_08	0.56±0.01	0.64±0.02	0.72±0.02	196.87±5.80
SWR_09	0.91±0.01	1.04±0.01	1.17±0.01	325.80±5.04
SWR_10	0.56±0.01	0.64±0.01	0.72±0.01	198.07±5.17
SWR_11	0.28±0.01	0.32±0.01	0.36±0.01	95.58±4.83
SWR_12	0.44±0.01	0.50±0.01	0.56±0.02	152.22±4.41
SWR_13	0.67±0.01	0.76±0.01	0.86±0.02	235.15±4.66
SWR_14	0.44±0.01	0.50±0.01	0.56±0.01	153.19±4.68
SWR_15	0.39±0.01	0.45±0.01	0.51±0.01	134.98±5.75
SWR_16	0.42±0.01	0.49±0.01	0.55±0.01	148.69±7.91
SWR_17	0.47±0.01	0.54±0.01	0.60±0.01	159.47±7.56
SWR_18	1.08±0.01	1.24±0.02	1.40±0.02	386.52±3.50
SWR_19	1.12±0.02	1.29±0.02	1.45±0.02	404.39±2.89
SWR_20	1.18±0.02	1.35±0.02	1.52±0.02	420.44±3.17
Minimum	0.28±0.01	0.32±0.01	0.36±0.01	95.58±4.83
Maximum	1.18±0.02	1.35±0.02	1.52±0.02	420.44±3.17
Average	0.62±0.002	0.71±0.002	0.80±0.002	217.80±0.95

Table C5: The Results of the total annual effective dose for the external exposure for the age category representative of an adult, child and infant for study area C.

Sample no.	AED Adult (mSv.y-1)	AED Child (mSv.y-1)	AED Infant (mSv.y-1)	R _{eq} (Bq.kg-1)
KHN_01	0.89±0.02	1.01±0.02	1.16±0.02	310.40±5.60
KHN_02	0.93±0.01	1.06±0.01	1.22±0.02	332.59±7.97
KHN_03	0.81±0.01	0.93±0.01	1.06±0.02	285.68±4.63
KHN_04	0.76±0.01	0.87±0.02	1.00±0.02	267.15±2.17
KHN_05	1.11±0.02	1.26±0.02	1.45±0.02	394.05±4.26
KHN_06	0.51±0.01	0.58±0.01	0.66±0.01	175.42±9.11
Minimum	0.51±0.01	0.58±0.01	0.66±0.01	175.42±9.11
Maximum	1.11±0.02	1.26±0.02	1.45±0.02	394.05±4.26
Average	0.84±0.01	0.95±0.01	1.09±0.01	294.22±1.63

Table C6: The total annual effective dose for the external exposure for the age category representative of an adult, child and infant for study area D.

Sample no.	AED Adult (mSv.y-1)	AED Child (mSv.y-1)	AED Infant (mSv.y-1)	R _{eq} (Bq.kg-1)
RTU_01	2.70±0.03	3.08±0.04	3.53±0.04	952.37±13.36
RTU_02	4.40±0.04	5.03±0.05	5.77±0.06	1559.45±18.23
RTU_03	1.83±0.02	2.09±0.03	2.40±0.03	654.97±10.17
RTU_04	2.13±0.03	2.43±0.03	2.79±0.03	748.55±10.52
RTU_05	2.16±0.03	2.47±0.03	2.83±0.03	759.72±10.72
RTU_06	3.14±0.04	3.59±0.05	4.11±0.05	1142.38±17.16
RTU_07	1.62±0.02	1.85±0.03	2.12±0.03	572.37±9.52
RTU_08	1.62±0.02	1.85±0.03	2.12±0.03	578.98±9.64
RTU_09	1.58±0.02	1.81±0.02	2.07±0.03	559.82±5.44
RTU_10	2.05±0.03	2.34±0.03	2.68±0.03	725.23±5.37
Minimum	1.58±0.02	1.81±0.02	2.07±0.03	559.82±5.44
Maximum	4.40±0.04	5.03±0.05	5.77±0.06	1559.45±18.23
Average	2.32±0.01	2.65±0.01	3.04±0.01	825.38±2.78

Table C7: The total annual effective dose for the external exposure for the age category representative of an adult, child and infant for study area E.

Sample No.	AED Adult (mSv.y ⁻¹)	AED Child (mSv.y ⁻¹)	AED Infant (mSv.y ⁻¹)	Ra _{eq} (Bq.kg ⁻¹)
RU_01	1.21±0.01	1.38±0.02	1.58±0.02	434.42±5.97
RU_02	1.62±0.02	1.86±0.02	2.13±0.02	592.25±6.96
RU_03	3.32±0.02	3.80±0.03	4.35±0.03	1230.21±10.29
RU_04	1.05±0.01	1.20±0.02	1.37±0.02	374.41±6.20
RU_05	0.71±0.01	0.81±0.02	0.93±0.02	249.78±5.97
Minimum	0.71±0.01	0.81±0.02	0.93±0.02	249.78±5.97
Maximum	3.32±0.02	3.80±0.03	4.35±0.03	1230.21±10.29
Average	1.58 ±0.01	1.81±0.01	2.07±0.01	576.21±3.00