

Radioactive soil contamination from the coastal towns of Walvis
Bay and Swakopmund in the vicinity of uranium mine sites of
Erongo region, Namibia

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DECLARATION

I declare that this dissertation is my own work. It is being submitted for the degree of Masters of Science in Applied Radiation Science in the North-West University, Mafikeng Campus. It has not been submitted before for any degree or examination in any other University.

Signature:

A handwritten signature in black ink, appearing to read 'A. Phivukwe', written in a cursive style.

Date:

28 May 2015

ABSTRACT

Radioactive soil contamination is one of the major causes of external gamma rays exposure in many places including the coastal towns of Walvis Bay and Swakopmund in the Erongo region of Namibia. Gamma radiation emitted from natural occurring radioisotopes such as ^{238}U , ^{232}Th and ^{40}K series and their decay products are present in all ground formations and these represent the main external source of irradiation to the human body. The levels of radioactive soil contamination due to naturally occurring radioactive materials (NORM) were investigated, followed by assessment of the radiological hazard to both the public and the environment using a well calibrated HPGe gamma ray-spectrometry setup. The activity concentration in the two coastal towns was found to vary from 14.94 ± 3.81 to 289 ± 76 Bq kg⁻¹ for ^{238}U , 17.68 ± 4.62 to 131.58 ± 19.26 Bq kg⁻¹ for ^{232}Th and 162 ± 54 to 581 ± 194 Bq kg⁻¹ for ^{40}K . The mean values of radium equivalent index (Ra_{eq}), radiation hazard index (H_{ex}) and total annual external gamma radiation dose were found to be 205 Bq kg⁻¹, 0.56 and 0.11 mSvyr⁻¹ respectively. Since the estimated radiological impact assessment factors are lower than the world permissible United Nations Scientific Committee on the Effect of Atomic Radiation (UNSCEAR), there seems to be no potential radiological health hazard associated with the soils of Walvis Bay and Swakopmund towns. This shows that the risk due to radiation contamination in Erongo region is low.

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LIST OF ABBREVIATIONS

ADC - Analog –to- digital converter

AEDE- Annual Effective Dose Equivalent

ALARA – As low as reasonably achievable

BEIR – Biological effect of ionising radiation

CARST- Centre for Applied Radiation Science and Technology

GPS- Global Positioning System

HPGe detector – High Purity germanium detector

ICP –MS – Inductively Coupled Plasma Spectrometry Mass Spectrometer

LET- Linear Energy Transfer

LHU –Langer Heinrich Uranium Mine

MCA – Multichannel pulse-height analyser

MME – Ministry of Mines and Energy

MoHSS – Ministry of Health and Social Services

NEB – Namibia Equity Brokers

NIM – Nuclear Instruments Modules

NORM - Naturally Occurring Radioactive Material

NPC – National Planning Commission

NRE - Natural radiation environment

PPE – Personal Protective equipment

RBE – Relative Biological Effectiveness

RUM - Rössing Uranium mine

SEA- Strategic Environmental Assessment

TENORM – Technologically enhanced naturally occurring radioactive material

UICM – Uranium Institute of Chambers Mines

UNSCEAR - Report to the General Assembly with Scientific Annexes

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1. INTRODUCTION

1.1 Theoretical background

Naturally occurring radioactive materials (NORM) are invariably present in small but measurable amounts in all materials found in the earth's crust, which are the main sources of human's exposure to ionizing radiation (Tzortzis and Tsertos, 2004). The principal radioactive materials in NORM are due to the presence of long lived radionuclides; these are ^{238}U , ^{235}U , ^{232}Th , ^{40}K and ^{87}Rb which are the main contributors to external exposure from gamma-radiation (Bashir et al, 2013). These radionuclides vary in spatial and in temporal. For example, cosmic rays are more intense at higher altitudes than at lower altitudes. Similarly, the concentration of uranium and thorium in soils is higher in some areas due to human activities such as mining and extraction of minerals. High levels of uranium and its decay products in rock and soil are the main sources of high natural background radiations that have been identified in several areas of the world (UNSCEAR, 2000).

In nature, mineral ores contains radionuclides which exists in a more or less state of secular equilibrium. However, this equilibrium may become disturbed through human activities such as mining and mineral processing, resulting in either enrichment or depletion of some radionuclides concentrations compared to their original matrix. This disequilibrium is partly due to the differences in the chemical properties of the radionuclides in the series, geochemical migration process and differences in radionuclides half-lives (Sato, 2003).

The exposure of human being to external radiation is through inhalation of radionuclides as aerosols, ingestion of radionuclides in food, soil and water. Once inside the human body, these radionuclides can accumulate in various organs and deliver radiation doses which results in alteration in the structure of chromosomes leading to the development of different types of diseases including cancers. The potential harmfulness of radionuclides is based on their half-lives (^{232}Th : 1.4×10^{10} year, ^{238}U : 4.47×10^9 year and ^{40}K : 1.28×10^9 year) and chemical behaviour. ^{232}Th is radiotoxic while ^{238}U and ^{40}K are both radiotoxic as well as chemically toxic while ^{40}K is a nutritionally important element (Bresson et al, 2011). Environmental problems associated with NORM in solid mineral ores and processing occur in the process of drilling, leaching, handling, storage, transportation of mineral ores and the use of contaminated equipment. These usually lead to the spread of NORM contaminating the environment, resulting in potential radiation exposure to members of the public. The International Basic Safety Standards (BSS) for protection against ionizing radiation and the safety of radiation sources specify the basic requirements for the protection of health and the environment from the ionizing radiation and

these are based on the recommendation of the International Commission on Radiological Protection (ICRP) (Lozano et al., 2011).

The isotopic abundance of primordial radionuclides in many materials is low that the risk is generally regarded as negligible. However, to minimise radiological risk to the general public from the effects of naturally occurring radioactive materials, measurement of radioactivity in the environments need to be considered to assess the biological effect on human. These studies are useful for both the assessment of public dose rate and the performance of epidemiological studies.

In the recent past, there has been growing interest in studies related to measurement of natural radioactivity in soil throughout the world mostly for the assessment of the dose(Clouvas et al, 2001).It is well documented that most natural radioactive elements present in soil are primordial radionuclides from the uranium series, thorium series and potassium 40. For example, Chowdhury et al in 1999, investigated the concentration and distribution of radioactive nuclides in river sediments and coastal soils of Bangladesh and showed that the activity concentrations of ^{238}U , ^{232}Th and ^{40}K were higher than the world values which were 35, 30 and 400Bq.kg⁻¹. In 2004, Matiulah et al, reported the mean activity of ^{226}Ra , ^{232}Th , ^{40}K and ^{137}Cs in soils of Bahawalpur, Pakistan being 32.9, 53.6, 647.4 1.5Bq.kg⁻¹. Hamarneh and Awadallah determined the radioactivity levels in various geological formations of soils in the Northern Highlands of Jordan in 2009. The average radioactivity concentrations were 42.5, 49.9, 26.7 and 291.1 Bq.kg⁻¹ of ^{226}Ra , ^{238}U , ^{232}Th and ^{40}K respectively.

Since radiation is neither visible nor can it be felt by the human sense organs, it is important that the total amount of radiation emitting-NORM in the area is accurately known and kept to a level as low as reasonably achievable in order to safeguard the lives of people and ensure radiation pollution free environment. High levels of radioactivity in some places can be due to deliberate or unintentional release of radioactive substances in the environment. Thus, it is important to consider all the significant sources of radioactive nuclides present in the environment.

The Erongo region of Namibia has been reported to experience high level of background radiation due to vast deposit of uranium bearing ores(von Oertzen, 2008). This study investigated the possible uranium soil contamination due to ^{238}U , ^{232}Th and ^{40}K , and their contribution to the annual effective dose to the general population living within the region.

1.1.1 Problem statement

There is inadequate knowledge about levels of naturally occurring radionuclides resulting from uranium mining in Erongo region of Namibia. The risks of human exposure and the impact of ionizing radiation from NORM due to uranium mining and extraction have been documented in many studies (Hamid et al, 2002). These studies were mainly concentrating on radioactivity within the mine premises and the radiation doses received by the mine workers and little attention was given to areas within the vicinity of mine sites and the dose received by members of the public. Similarly, data on natural radionuclides is still very scanty in Namibia and the previous studies were limited to specific regions or geographical locations (Oyedele, 2006).

Uranium is a suspected human carcinogen and a known kidney toxin (Miller and McClain, 2007). The chemical toxicity of uranium as a heavy metal has raised public health concerns, especially in areas where contamination of local soils from radioactive material has taken place (Choy et al, 2006). This is prevalent in places in and around uranium mine sites. The problem is further compounded when mining authorities are unaware of the problems associated with enhanced levels on NORM in raw material, products, mine tailings and the environments and no protective measures are put in place so that the doses to workers and the public do not exceed the prescribed dose limits. The relevant route of exposure of the public is internal, via inhalation of dust and aerosols and ingestion of food and water.

In Namibia, uranium mines are located in the Southern region of Erongo. Most of the mines undertake only surface mining and this produces large volumes of tailings and waste that may contain enhanced levels of naturally radionuclides. Some of the radionuclides may disperse into the environment resulting in contamination of soil and water bodies. Radionuclide's such as ^{226}Ra and ^{228}Ra are known to have high mobility in the environment due to their high comparative solubility in water. The ultimate substrate of these radionuclides is the human body which is the main concern of this study.

Adequate data is important for regulatory and advisory purposes with the aim of protecting the general public from unnecessary exposure to radiation. Thus, it is imperative to evaluate radioactivity in soil of the two coastal towns of Erongo region in the vicinity of uranium mine sites to assess the radiological risk to the public and the environment. The results of this study were compared with international and national recommended values.

1.2 Research Aims and Objectives

1.2.1 Overall aims

The general aim of the study is to determine the levels and distribution of naturally occurring radionuclides of U/Th decay series and ^{40}K in the two coastal towns of Walvis Bay and Swakopmund in the vicinity of uranium mine sites of Erongo region. The activity concentration of these radionuclides were used to evaluate the elemental concentrations and the radiological risk to members of the public living in the Erongo region.

1.2.2 Specific objectives

- Determine the activity of ^{238}U , ^{232}Th and ^{40}K in the surface soils collected.
- Determine the elemental concentrations of ^{238}U , ^{232}Th and ^{40}K in soils collected.
- Assess the potential impact of the radioactivity on human health and the environment through dose measurements and calculations.

1.3 Significance of the study

In many developing countries including Namibia, data on radionuclides concentrations in raw materials, residues, fallout from uranium mining and processes and public exposure is still very scanty. As a result of this, there is generally lack of awareness and lack of knowledge of the radiological hazard exposure by members of the public. The information generated by this study will assist policy makers, planners and regulators to design regulatory framework that can be used by the mining community to minimize potential hazard from NORM and thus protecting human health and the environment. The study will add to the knowledge base and compliment a few other radiological studies done in other regions of Namibia.

2. BASICS OF NUCLEAR DECAY

2.1 Radioactivity and radioactive decay

Radioactivity is a random process whereby an unstable nuclide or 'parent' is transformed into a more stable nuclide called the 'daughter'. If the daughter is also radioactive, the process continues in a decay chain until a stable product is reached (Lilley, 2001). All naturally occurring and artificially produced radioactive nuclei are either alpha (α) active, beta (β) active or (occasionally) both and emit α , β and γ radiation.

The strength or intensity of the radioactivity is called activity and is defined as the number of atoms that decay per unit time (Turner, 2007). The probability per unit of time for the decay of a given nucleus is a constant which can be commonly named as the disintegration or decay constant (λ). Similarly, the radioactive decay which is related to the activity can be expressed by the fundamental law of radioactive decay (L'Annunziata, 2007).

$$A = -\frac{dN}{dt} = \lambda N \quad (\text{Eq. 2.1})$$

As shown in equation 2.1, A is the activity of an isotopically pure source, which is equal to the number, dN , of radioactive nuclei disintegrating in a given time, dt , and is proportional to the number, N, of a radioactive nuclei present at time, t (L'Annunziata, 2007). λ is the decay constant and the negative sign indicates that the number of radioactive nuclei decreases when the time increases (Turner, 2007). Until quite recently, the unit of measurement of activity was the Curie (Ci) which is based on the activity of 1 gram of Radium (^{226}Ra), which is equivalent to 3.7×10^{10} disintegration per second (Curie *et al.*, 1931). Currently, the standard unit of radioactivity is Becquerel (Bq) and is defined as one disintegration per second (Cember and Johnson, 2009). The Curie is related to Becquerel by the expression $3.7 \times 10^{10}\text{Bq}$.

In terms of number of nuclides, equation 2.1 can be expressed as;

$$N_t = N_0 e^{-\lambda t} \quad (\text{Eq. 2.2})$$

Where N_0 is the original number of nuclei present at time $t = 0$. The number of nuclei decreases exponentially with time. Figure 2.1 illustrates the number of nuclei decaying as function of time.

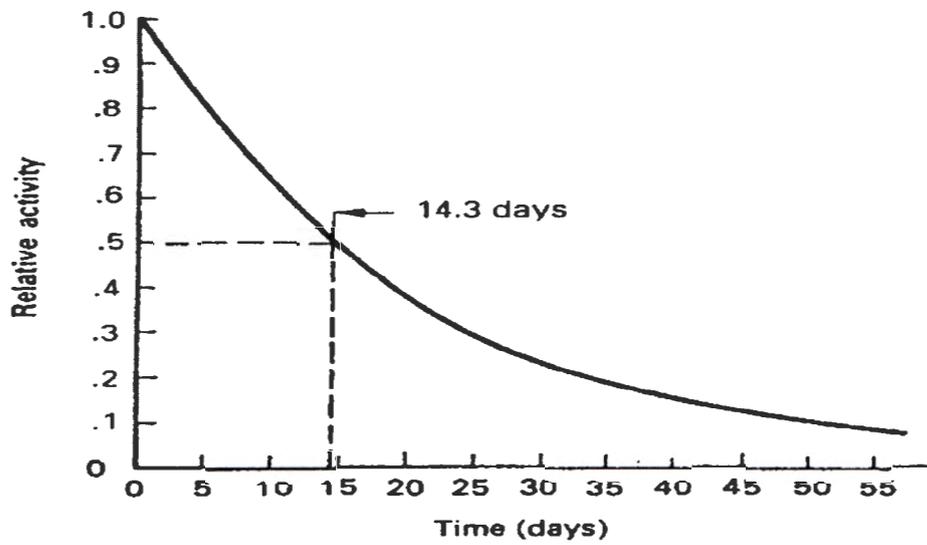


Figure 2.1: The exponential radioactive decay of ^{32}P with half-life, $T_{1/2}$ of 14.3 days (Cember and Johnson, 2009)

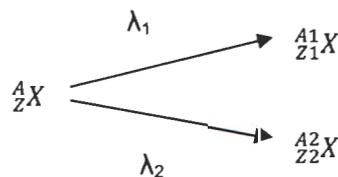
The half-life ($T_{1/2}$) is the time required for one half of a certain number of active nuclei to disintegrate as shown in Figure 2.1. The decay constant is related to half-life by the expression;

$$\lambda = \frac{0.693}{T_{1/2}} \quad (\text{Eq.2.3})$$

The half-life for radionuclides can range from fractions of a second to billions of years (Choppin et al, 2002).

2.1.1 Branching decay

In nature radionuclides exist that show two different modes of decay as shown in schematic diagram below.



The relative probability of the competing modes can be specified by the branching ratios (Krane, 1988). Each decay mode has its specific decay constant or half-life. The total observed decay constant is equal to the sum of the partial decay constant (Baisden and Choppin, 2007) as;

$$\lambda_{\text{tot}} = \lambda_1 + \lambda_2 + \dots \quad (\text{Eq. 2.4})$$

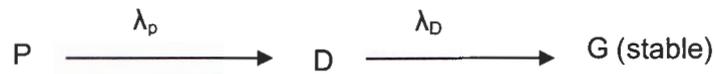
Similarly, the partial half-lives for each decay mode correlate with the partial decay constants for the n-th mode of decay

$$T_{1/2}(n) = \frac{0.693}{\lambda_n} \quad (\text{Eq. 2.5})$$

2.1.2 Radioactive decay series

If a radioactive nuclide situated in the chart of nuclide is far from the zone of stability (for the light elements at $Z=N$), the daughter nuclide after radioactive decay may be radioactive as well. This occurs with heavy nuclides such as ^{238}U and ^{232}Th decay series.

A radioactive decay chain begins with a radioactive parent nucleus, P decaying with a decay constant λ_p into a daughter nucleus D, which in turn is radioactive and subsequently decays with a decay constant λ_D into a stable grand-daughter nucleus, G, as described below (Cember and Johnson, 2009)



The decay equation of the radioactive series is given by the three equations (Krane, 1988).

$$\frac{dN_p}{dt} = -\lambda_p N_p \quad (\text{Eq. 2.6})$$

$$\frac{dN_D}{dt} = \lambda_p N_p - \lambda_D N_D \quad (\text{Eq. 2.7})$$

$$\frac{dN_G}{dt} = \lambda_D N_D \quad (\text{Eq. 2.8})$$

From the above equations, it can be deduced that the rate of depletion of the number of radioactive parent nuclei dN_p follows equation 2.1 (Santawamaitre et al., 2014). Similarly, the rate of change in the number of daughter nuclei, dN_D , equals to the difference in the supply of new daughter nuclei through the decay of the parent nuclei and the loss of the daughter nuclei from the decay of itself to a stable product. The concentration of the stable end product dN_G increases at the same rate as the decay rate of the daughter nuclear.

According to equation 2.2, the number of parent nuclei and the equation of activity can be written as

$$N_p(t) = N_p(0)e^{-\lambda t} \quad (\text{Eq. 2.9})$$

Where $N_p(0)$ is the initial number of parent nuclei at time $t = 0$. The ratio between λ_1 and λ_2 is the rate limiting factor that determines the source of daughter activity in time.

2.2 Radioactive equilibrium

Radioactive equilibrium is the term used to explain a steady state when members of the radioactive series decay at the same rate as they are produced i.e. the activity of the parent nuclide is equal to the activity of the daughter nuclide (Prince, 1979). There are three predominant state of radioactive equilibrium namely; secular equilibrium, transient equilibrium and no equilibrium.

2.2.1 Secular Equilibrium

This type of relation between parent and daughter occurs when the half-life of the parent is infinitely larger than that of daughter nuclide. Under those conditions, the activity concentration of daughter nuclide becomes equal to that of its parent with time (Lamarsh and Bell, 2009).

Considering two nuclides P and D that are in secular equilibrium with each other can be expressed as;

$$\lambda_p N_p = \lambda_D N_D \quad (\text{Eq. 2.10})$$

where λ_p is the decay constant for nuclide P, λ_D is the decay constant of nuclide D, N_p is the number of atoms in the nuclide P and N_D is the number of atoms in the nuclide D. In a decay chain, the numbers of nuclei of the various daughters present at equilibrium are inversely proportional to their decay constant. Similarly, the ingrowth of the progeny is equal to the decay rate of the parent nuclide.

For example, the secular equilibrium build-up of very short lived daughter (^{222}Rn with half-life 3.842d) from a long lived parent (^{226}Ra with half-life 1600 y). For practical reasons, equilibrium is assumed to have been attained after seven half-lives as shown on Figure 2.2.

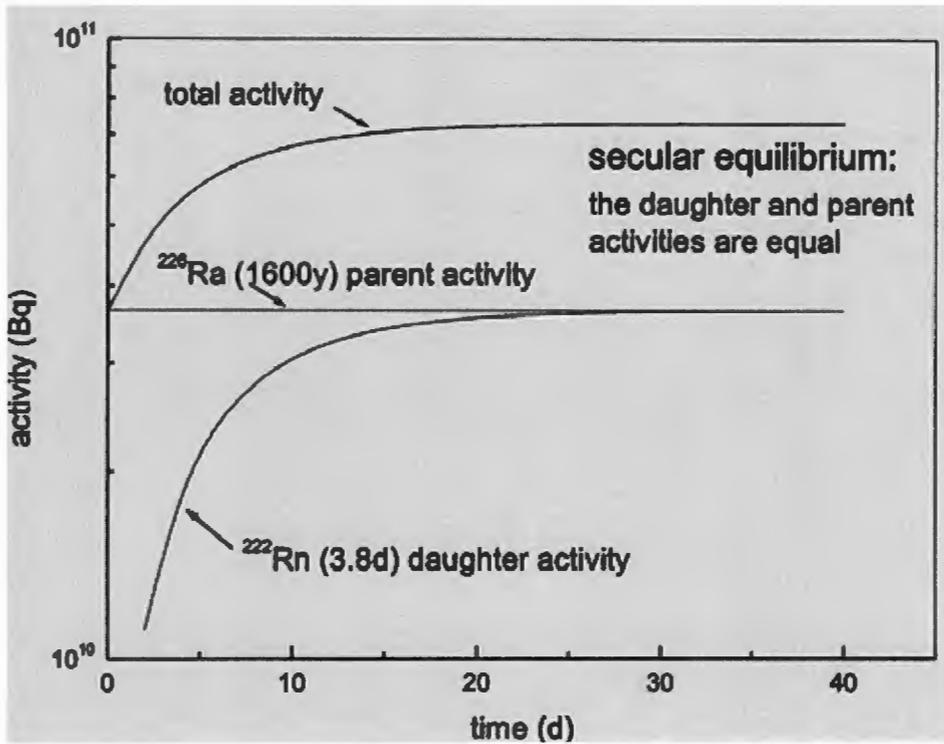


Figure 2.2: The growth of short lived ^{222}Ra until a secular equilibrium is established (Magill and Galy, 2004)

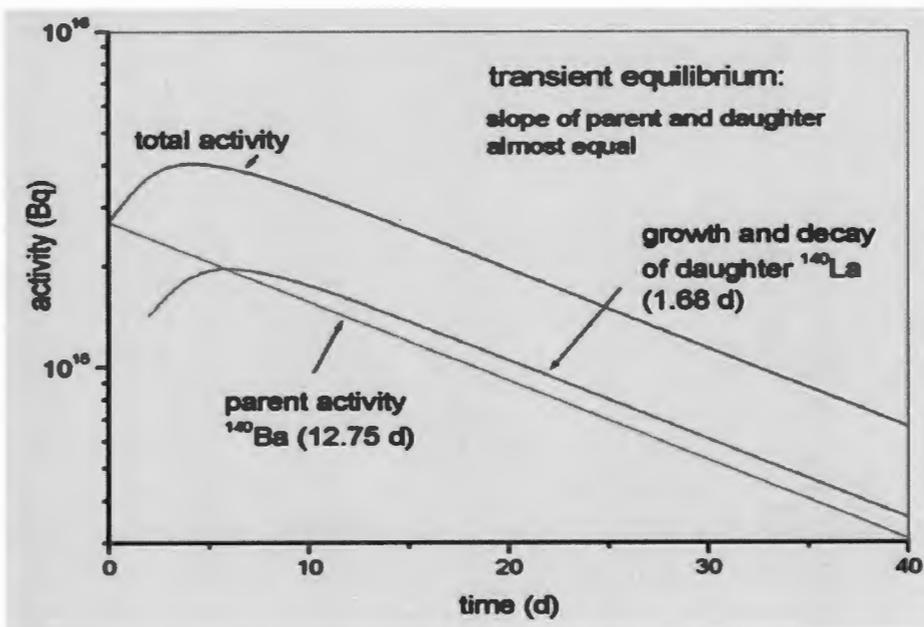


Figure 2.3: Typical transient equilibrium between ^{140}Ba and ^{140}La (Magill and Galy, 2004)

2.2.2 Transient Equilibrium

In transient equilibrium the half-life of the daughter is of the same order but smaller than that of the parent i.e. where $\lambda_p < \lambda_D$ (Magill & Galy, 2004). Under this circumstance, in the decay equation, the exponential term becomes so smaller and the activity of A_p/A_D approaches the limiting constant value $\lambda_D/(\lambda_D - \lambda_p)$. This can also be expressed as;

$$N_D / N_p = \lambda_p / \lambda_D - \lambda_p \quad (\text{Eq. 2.11})$$

A good example of radionuclide decaying by transient equilibrium is the decay of ^{140}Ba ($T_{1/2}$ 12.75d) which decays to ^{140}La ($T_{1/2}$ 1.68 d) as shown in Figure 2.3.

The daughter and parent activities are approximately equal but change with time. From equation 2.11, after a state of equilibrium is established, the ratio of number of nuclide tend to be constant value (Kaplan and Gugelot, 1955) and the parent and the daughter nuclides will decay at the same rate, related to the decay of parent

2.2.3 No equilibrium

When the parent nuclide has a shorter half-life than that of a daughter product, the state of equilibrium will not be reached. The parent nuclide which have a short half-life will decay according to its decay equation while the number of daughter nuclei will build up to maximum and then start to decrease eventually with its own characteristic half-life as can be shown in Figure 2.4.

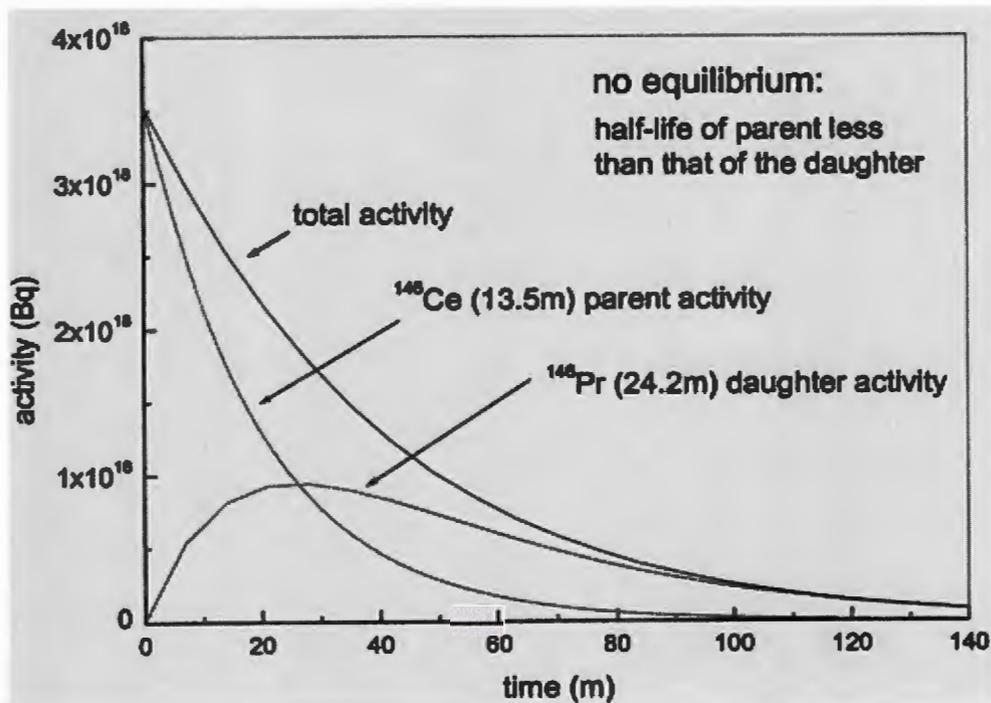


Figure 2.4: Growth and decay of long lived daughter ^{146}Pr from a short lived parent ^{146}Ce in case of no equilibrium (Magill and Galy, 2004)

2.3 Types of decay

Radioactive decay is a spontaneous nuclear transformation which results in the formation of new stable elements. The process is most often accompanied by emission of gamma radiation. If the daughter nuclide is also unstable, the radioactive decay process continues further in a decay chain until a stable nuclide is reached (Magill and Galy, 2004). Radioactive nuclides decay spontaneously by the following processes.

2.3.1 Alpha (α) decay

An alpha particle is a highly energetic helium nucleus that is emitted from the nucleus of an unstable atom when the neutron-to-proton ratio is too low i.e. proton rich nuclide (Cember and Johnson, 2009). It is positively charged, macroscopic particle consisting of two protons and two neutrons. Since the atomic numbers and mass numbers are conserved during alpha decay, it follows that the result of alpha emission is a daughter whose atomic number is less than that of the parent and whose mass number is four less than that of parent. This can be seen in the following schematic decay process of ^{238}U ;



The characteristic energy liberated in the radioactive decay (the Q value), is equal to the difference in mass-energy between the initial nucleus and the final products. The disintegration energy (which is equivalent to Q value) can be calculated by accounting for the total mass/energy change in the decay process using the following expression.

$$Q = (m_p - m_D - m_\alpha) c^2 \quad (\text{Eq. 2.13})$$

where m_p , m_D and m_α are the nuclear masses of the parent, the daughter and the emitted α particle, respectively. The reaction is energetically feasible if Q value is positive. The alpha decay is a characteristic of proton-rich, high Z number nuclides due to their greater electrostatic repulsive forces compared to their cohesive nuclear force. In addition, the emitted particle must have sufficient energy to overcome the potential barrier in the nucleus as shown in the Figure 2.5 (Cember and Johnson, 2009).

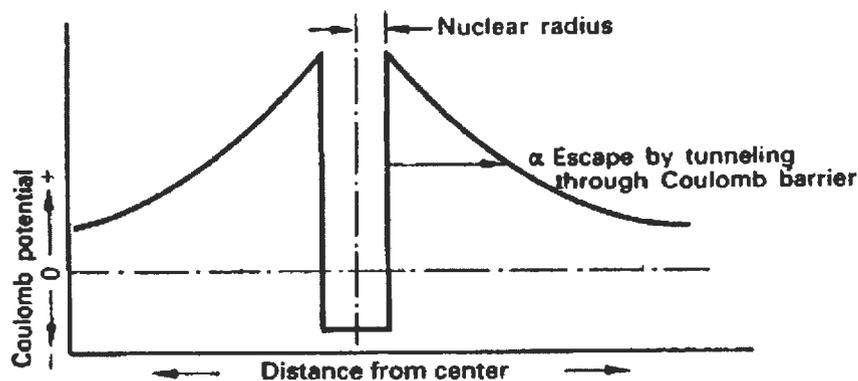


Figure 2.5: Potential inside and in the vicinity of a nucleus (Cember & Johnson, 2009).

The height of the potential barrier in Figure 2.5 is 25 MeV. However, alpha particles can escape this barrier by the process of quantum tunnelling. In the interaction with matter, the macroscopic charged alpha particles interact strongly with matter, transferring all its energy to the surrounding medium and become a neutral helium atom. Their range in solids and liquids is very short- typically of the order of micrometers and therefore they do not constitute a health hazard to the human externally. Internally, they are very dangerous owing to high linear energy transfer (LET). The quantity of LET radiation imparted into medium will increase rapidly with mass and charge of the particle (Lamarsh and Bell, 2009). Therefore, alpha particle decay will impart the highest quantities of LET radiation. Particles that decay as high LET radiation have the potential to produce a greater biological effect on absorbing medium.

2.3.2 Beta (β) decay

A beta particle is an electronically charged particle with same mass as the ordinary electron and it is ejected from an unstable radioactive nuclide whose neutron-to-proton ration is too high (Cember, 2009). In beta decay, the process occurs by emission of negative or positive beta particle or by competing process of orbital electron capture (Magill and Galy, 2004). The process changes the atomic number (Z) by one unit, while the atomic mass (A) remains constant (Krane, 1988). Unlike alpha particles, beta particles are emitted with continuous energy distribution ranging from zero to thousands electron volts.

The imbalance of neutron to proton give rise the spontaneous transformation of a neutron into proton with the emission of an electron as illustrated;



where n is the number of neutrons, p is the number of protons, β^- is beta particle, $\bar{\nu}$ is an antineutrino. Negatron has the size and mass of an electron, as such they can interact less strongly with matter as compared to alpha particles and penetrate deeper.

A second form of beta decay is positron decay which results from an excessive high proton-to-neutron ratio. Positron emission can occur only when the transformation energy is greater than 1.02 MeV (Gilmore and Hemingway, 2008). As a result, a positive electron is ejected from the nucleus where the negative electron combine with a proton converting it to a neutron within the nucleus to attain more nuclear stability as illustrated;



where p is the number of protons, n is the number of neutrons, β^+ beta particle (positron) , ν is an antineutrino. In case there is insufficient transformation energy to form an electron pair i.e. energy less than 1.02 MeV, the nuclide can undergo, electron capture (EC) (Lilley, 2001).

This process is a mechanism in which an electron, usually in a K shell, is captured by a proton rich unstable nucleus (Das and Ferbel, 2003).



2.3.3 Gamma (γ) Radiation

Gamma radiation are electromagnetic radiation that travels in the form of waves and do not carry any electrical charge (Gilmore and Hemingway, 2008). Gamma radiation is differentiated from other forms of electromagnetic radiation on the basis of its wavelength, frequency and origin (Krane, 1988). Following alpha or beta decay, the nuclide will be in an excited state and has to remove off excess energy. This is usually accompanied by gamma emission (Friedlander, 1981). Assuming that the nucleus is at rest while in an initial excited state E_i which decays to a final state E_f , the law of conservation of energy can be used to calculate the energy emitted by gamma-ray transition as follows;

$$E_i = E_f + E_\gamma + E_R \quad (\text{Eq. 2.14})$$

which can be rearranged as
$$\Delta E = E_i - E_f = E_\gamma + E_R \quad (\text{Eq. 2.15})$$

The recoil energy E_R of the final product can be considered to be negligible (Debertina *et al.*, 1988). Thus, the gamma-ray energy E_γ , is almost equal to the energy of the de-excitation ΔE , which is the energy difference between initial and final states. Typically, the energy ranges of gamma rays ranges from 0.1 up to 10 MeV (Lilley, 2001).

2.4 Environmental sources of radioactivity

Radioactivity is characterized by the spontaneous transformation of unstable atomic nuclei which deliver energy in form the of ionizing alpha, beta and gamma radiation, which proceeds directly from the atomic nucleus. The transition to a stable nuclide can either be direct or indirect, thus taking the form of many transformations resulting in several unstable intermediate stages. The exact mode of transformation depends on energy available for decay which in turn is closely linked to a particular type of instability i.e. whether the neutron-to-proton ratio is too high or too low for the particular nuclide under consideration and the mass and energy relationship among parent nucleus, daughter nucleus and emitted particle (Cember and Johnson, 2009).

Exposure to ionizing radiation originates from two major sources; namely; natural and man-made sources. Naturally occurring radiation which arises from the terrestrial radionuclides widely distributed in the earth's crust (Lilley, 2001). Man-made sources are mainly from medicines or medical applications such as X-rays and radiotherapy. Natural radiation is the largest contributor to the collective radiation dose to the world population. The major contribution of dose from natural radiation in normal background regions is due to ingestion of

radionuclides through the food chain, the ingestion of contaminated soil or water and inhalation of radon and its progeny and to a certain extent, due to thoron and its progeny (Awwiri et al, 2007).

2.4.1 Natural sources of Radiation

The naturally occurring radioactive materials (NORM) have the greatest contribution to human's exposure to ionizing radiation and are composed of isotopes which occur individually or are components of three main radioactive decay series (Fahmi et al., 2011). These radionuclides are categorized into two main types in relation to their origin, which are; primordial and cosmogenic.

2.4.1.1 Primordial Radionuclides

Naturally occurring radionuclides of terrestrial origin are also termed primordial radionuclides and these have been on earth since its formation. Examples of the radionuclides are ^{40}K , ^{238}U and ^{232}Th and these are characterized by very long decay half-lives which is comparable to the age of the earth (Tzortzis and Tsertos, 2004). Virtually, most minerals on earth contain minute and measurable concentrations of naturally occurring radioactive (NORM). Human activities associated with mining and mineral processing may result in increased concentrations of ^{238}U and ^{232}Th (Turcotte et al, 2001). The primordial radionuclides are further subdivided into the series and non-series (Eisenbud and Gesell, 1997). The non-series radionuclides group decays directly to stable nuclides.

Non-Series Radionuclides

These decay directly to stable nuclides. Most of these have very long half-lives and very low natural isotopic abundance resulting in negligibly low activities and therefore are usually not considered in terms of their total contribution to radiation exposure to humans. However, two such isotopes, ^{40}K (natural isotopic abundance of 0.0117%) and ^{87}Rb (natural isotopic abundance of 27.8%) are significant sources of natural radiation (Santawamaitre, 2007). ^{40}K undergoes beta decay with accompanying gamma-ray emission while ^{87}Rb is purely a beta emitter (NCRP, 1975).

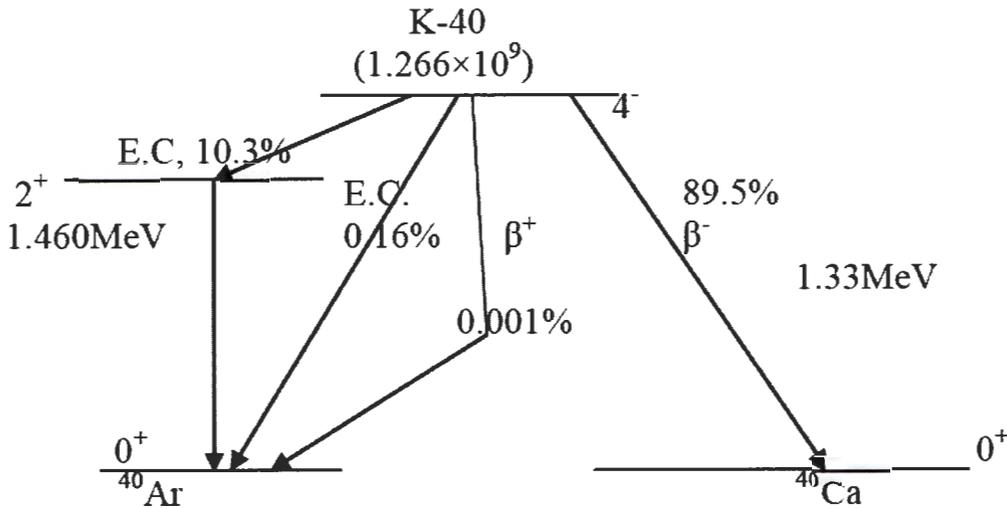
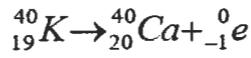
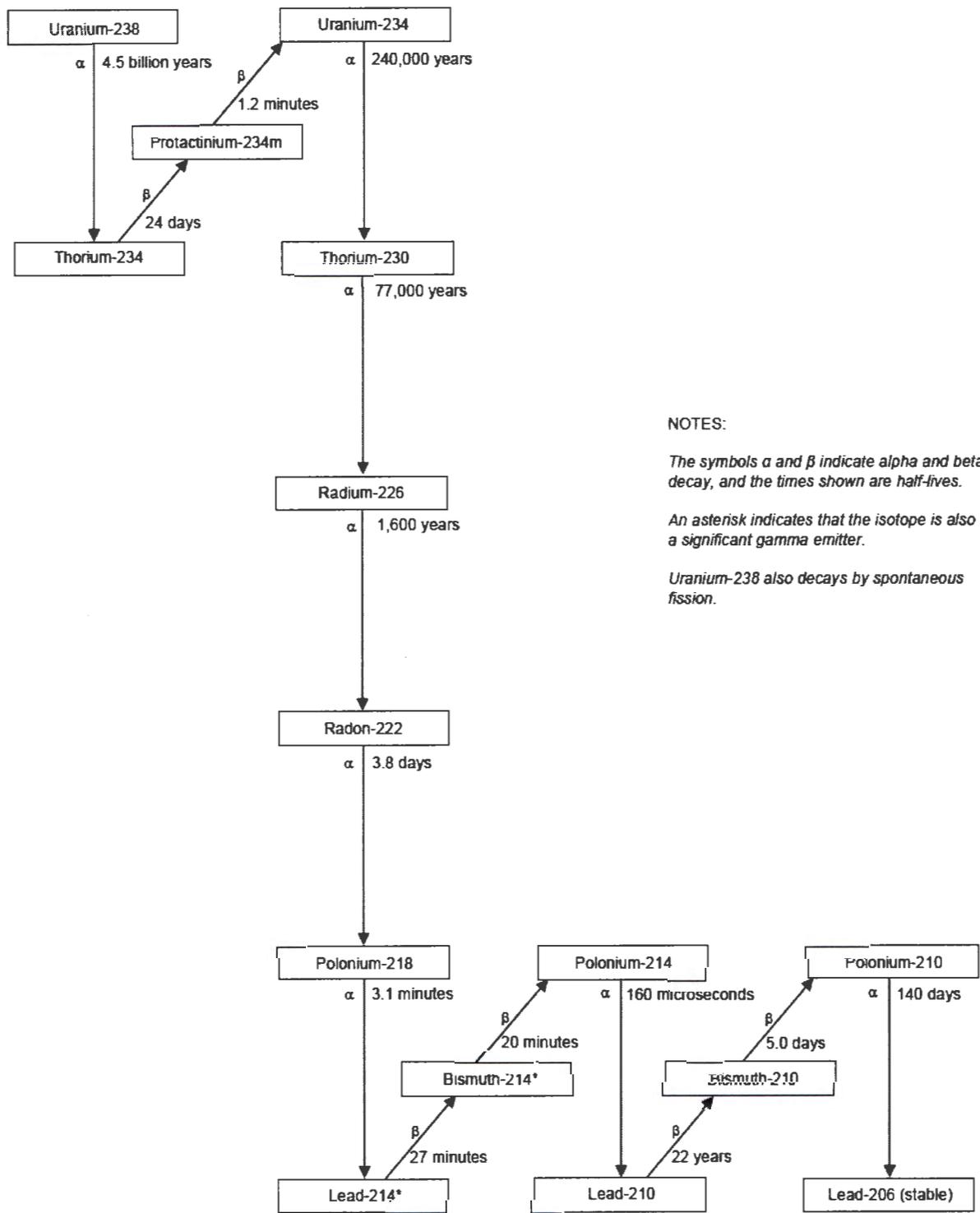


Figure 2.6: Decay scheme of ${}^{40}\text{K}$ (Faanu, 2011)

As depicted in figure 2.6, ${}^{40}\text{K}$ decays to a stable ${}^{40}\text{Ar}$ (11.2%) by electron capture and by positron emission and also decays to stable ${}^{40}\text{Ca}$ (88.8%) by beta emission. The beta emission yields 1.33 MeV of energy and the electron capture (EC) yield of 1.46 MeV.

Series Radionuclides

There are four radioactive decay series in the primordial series radionuclides and these are; Uranium, Thorium, Actinium and Neptunium series. These decay chains are headed by isotope of ${}^{238}\text{U}$, ${}^{232}\text{Th}$, ${}^{235}\text{U}$ and ${}^{237}\text{Np}$ respectively. ${}^{235}\text{U}$ and ${}^{237}\text{Np}$ may be ignored in radiation protection measurements because they do not have significant contributions; the contribution of ${}^{235}\text{U}$ to radiation exposure is less than 1% and ${}^{237}\text{Np}$ does not exist on earth anymore. The other three primordial nuclides series are headed by ${}^{238}\text{U}$, ${}^{232}\text{Th}$ and ${}^{235}\text{U}$, and they decay to yield their daughter nuclides until they reach stable nuclides such as lead. The main decay schemes of ${}^{238}\text{U}$, ${}^{232}\text{Th}$ and ${}^{235}\text{U}$ are displayed and presented in Figure 2.7; Figure 2.8 and Figure 2.9.



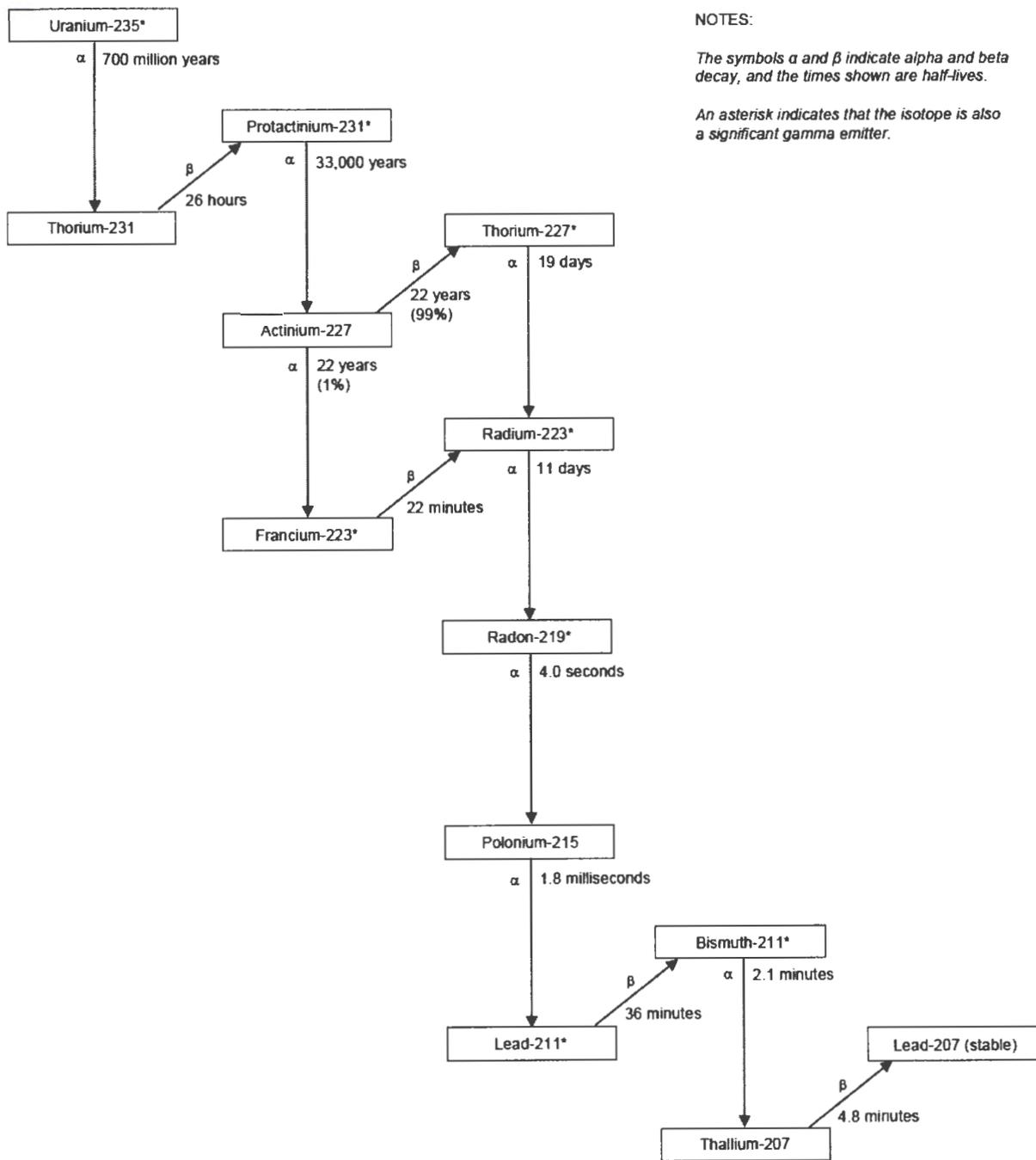
NOTES:

The symbols α and β indicate alpha and beta decay, and the times shown are half-lives.

An asterisk indicates that the isotope is also a significant gamma emitter.

Uranium-238 also decays by spontaneous fission.

Figure 2.7: Natural decay series: ^{238}U (Human Health Fact sheet, 2005)



NOTES:
 The symbols α and β indicate alpha and beta decay, and the times shown are half-lives.
 An asterisk indicates that the isotope is also a significant gamma emitter.

Figure 2.8: Natural decay series: ^{235}U (Human Health Fact sheet, 2005)

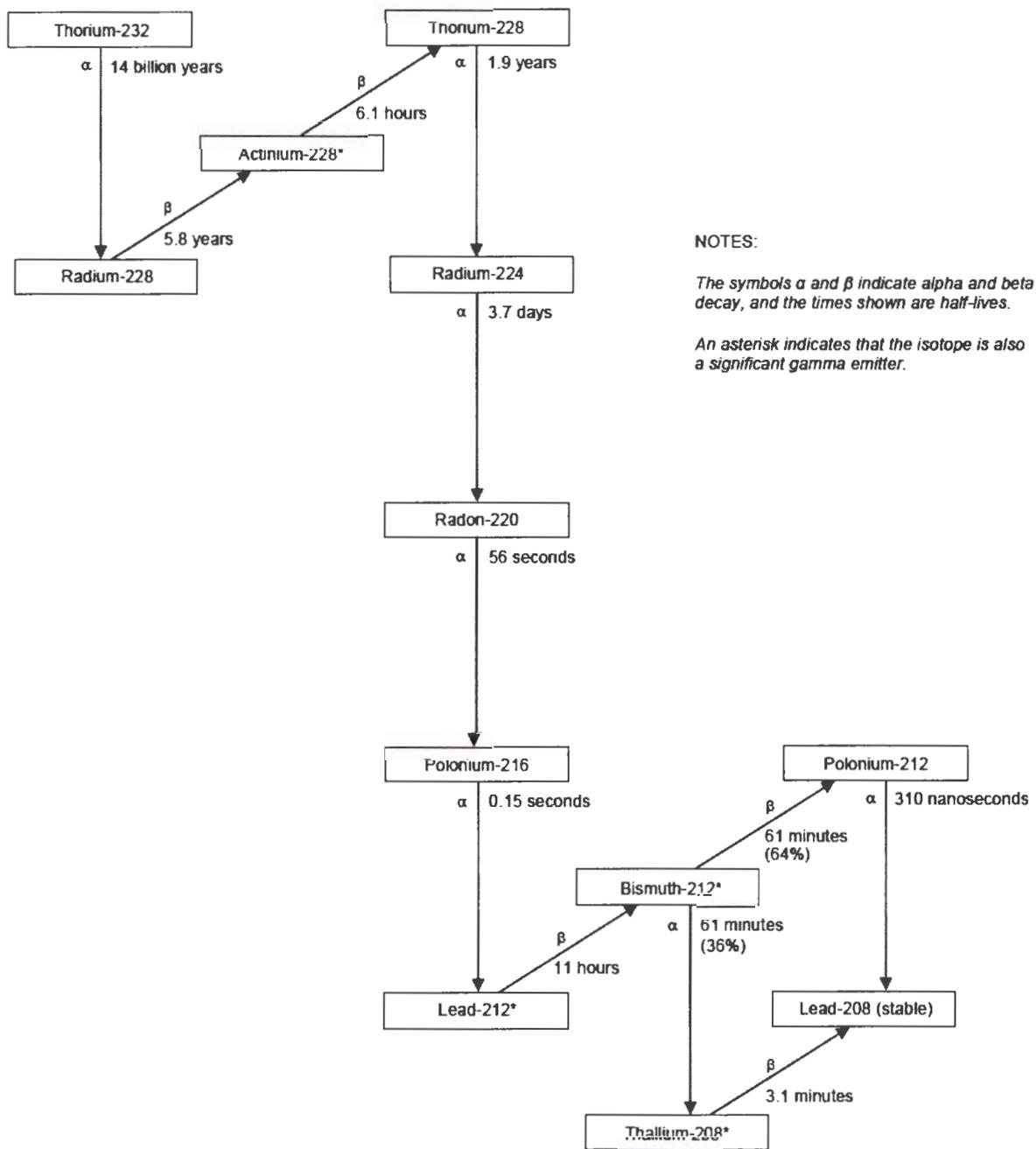


Figure 2.9: Natural decay series: ^{232}Th (Human Health Fact sheet, 2005)

2.4.1.2 Cosmic Radiation

These are radionuclides that are produced when high energy protons interact with constituents of the earth's atmosphere. Consequently, secondary showers of particles from such interaction, principally neutrons, yield cosmogenic radionuclides at relatively uniform rates (Watson et al., 2005).

2.4.1.3 Cosmogenic Radionuclides

Cosmogenic radionuclides are produced continuously by the interaction of nucleons released from cosmic radiation with the target nuclei in the earth's atmosphere, mostly the upper atmosphere (Cember and Johnson, 2009). These interaction leads to a variety of spallation or neutron capture reactions (UNSCEAR, 2000).

Spallation and charge exchange results in lighter or same mass radionuclides being created compared with the target atom. On the other hand, the radionuclides products from neutron capture reactions have one mass unit heavier than the original stable 'target' nuclide. The major target atoms in the atmosphere are oxygen, argon and nitrogen gases.

Several cosmogenic radionuclides are produced with half-lives ranging from minutes to millions of years; the majority of them are of no importance to radiation protection (Martin et al, 2012). These are; ^{10}Be , ^{26}Al , ^{36}Cl , ^{80}Kr , ^{32}Si , ^{39}Ar , ^{37}Ar , ^{35}S , ^{32}P , ^{24}Na , ^{18}F , $^{34\text{m}}\text{Cl}$, which are produced in small quantities. However, only four of them are important in radiation protection and these are; tritium (^3H), Beryllium-7 (^7Be), carbon-14 (^{14}C) and sodium-22 (^{22}Na). The production mechanism of important cosmogenic radionuclides and their typical concentration in the biosphere are shown in table.

Table 2.1: Typical cosmogenic radionuclides

Nuclide	Source	Typical natural activity
^{14}C	$^{14}\text{N}(n, p)^{14}\text{C}$	220 Bq.kg ⁻¹
^3H	Cosmic ray interaction with nitrogen, Oxygen and Lithium	1.2 Bq.kg ⁻¹
^7Be	Cosmic ray interactions with nitrogen and oxygen	10 Bq.kg ⁻¹

2.4.2 Anthropogenic sources

Human activities such as mining and extraction of ores can lead to release of radionuclides in the environment. There are several pathways by which radionuclides can reach humans. These are dependents on the pathway involved and can categorize as on-site and off-site. On-site exposure situation affects mainly the workers and people who are within the mining premises NORM could present in raw materials, waste piles, storage tanks, build-up of surface contamination on equipment and in pipes. The exposure tends to be direct from external gamma radiation or internal exposure resulting from inhalation of radioactive dust or radon progeny. Underground mining activities can lead to enhanced levels of radioactive dust and radon isotopes (O'Brien and Cooper, 1998). While off-site exposures are those exposures that occur to people who are living within or near the mining sites. This form of exposure is indirect and more complex and members of the public become the target of exposure. External exposure to members of the public can result from exposure to gamma radiation from the passage of sky shine cloud, reconstituted radioactive dust, the use of industrial waste containing NORM or through the food chain via ingestion of contaminated food, soil and water (Dahlgard, 1996). Analysis of the pathways by which these materials can move through the environment is necessary to ensure that the impact of mining and mineral processing on the environment and the public is minimized (O'Brien and Cooper, 1998).

Agricultural activities such as application of fertilizers can also lead to the discharge of large quantities of radioactive material originating from phosphate rock. Phosphate rock, which is the main ingredient in the manufacture of fertiliser and gypsum, contains high amount of uranium and its radioactive progeny (Paschoa and Godoy, 2002). In about 90% of the cases, the ore is treated with sulphuric acid. The fertilizer become somewhat enriched in uranium (up to 150% relative to the ore), while 80% of the ^{226}Ra , 30% of the ^{232}Th and 5% of the uranium are left in the phosphogypsum. The use of phosphate fertilizers in agriculture and gypsum in building material is a source of possible exposure to background radiation to the public. Elevated radon exposure can be further expected in sites being developed for housing (Vandenhove et al., 2006).

3. INTERACTION OF GAMMA RADIATION WITH MATTER

3.1 Interaction of gamma rays with matter

The measurement of gamma ray radiation is based on the interaction of photons with matter; it is empirical to consider the various mechanisms that are significance in gamma spectroscopy. The method of photon interaction was first explained by Einstein. He indicated that the type of interaction depends to a large extent on the energy of the photon. The major photon interactions mechanisms will be discussed in this section.

3.1.1 Photoelectric absorption

In this interaction, the incident gamma ray interacts with the entire atom, the gamma ray disappears and one of the atomic nucleus is ejected from the atom. The atom recoils in this process but carries with it very little kinetic energy (Lamarsh and Baratta, 2001).The energy of the photoelectron released by the interaction is the difference between the incident photon ($h\nu$) and the binding energy of electron in its original shell (E_b) (Kantele, 1995).

$$E_{e^-} = h\nu - E_b \quad (\text{Eq.3.1})$$

The photoelectron is most likely to emerge from the K shell, for which the typical binding energies ranges from few keV for low Z materials to tens of keV for material with high-Z atomic number (Knoll, 2000).

If a gamma ray succeeds in ejecting an inner atomic electron, this creates a vacancy in the shell which leaves the atom in an excited state (Lamarsh and Baratta, 2001).This vacancy is later filled through rearrangement of electrons from the shells of the atom as shown in figure 3.1.

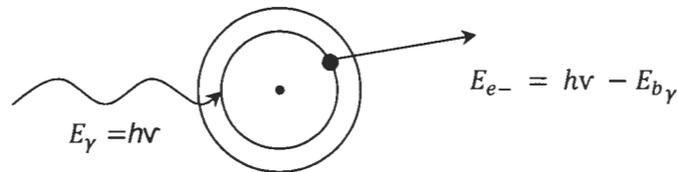


Figure 3.1: Schematic of the photoelectric absorption of process(Gilmore and Hemingway, 2008)

This transition is accompanied by emission of X-ray characteristic of the atom or by ejection of Auger electrons as shown in figure 3.2.

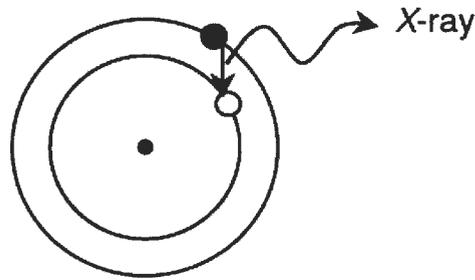


Figure 3.2: Schematic of the photoelectric absorption of process(Gilmore and Hemingway, 2008)

The interaction cross section or the probability (τ) of the photoelectric process depends on the photon energy, the binding energy and the atomic number of the absorbing material. The probability is given by the approximation (Knoll, 2000).

$$\tau = \alpha \cdot \frac{Z^n}{E_\gamma^m} \quad (\text{Eq.3.2})$$

where α is a constant, Z is the atomic number of the atom and n and m are numbers over the gamma ray energy of interest. The strong Z dependence indicates that material with high Z is very effective in the absorption of photons.

3.1.2 Compton scattering

The process involves a collision between the incident gamma ray photon and a loosely bound or free electron in the absorbing material. As a result of this collision, some of the energy is transferred to the electron. At the same time, the incoming gamma-ray photon is degraded in energy and is deflected from its original direction and an electron known as a recoil electron is formed. The magnitude of scattering depends on the angle between the directions before and after the interaction as shown in figure 3.3.

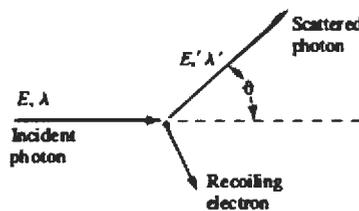


Figure 3.3: The electron energy and energy of scattered photon(Lamarsh and Baratta, 2001).

$$E_\gamma = E_{\gamma 1} + E_g \quad (\text{Eq.3.3})$$

By considering the momentum conservation law in the plane of interaction, equation 3.3 can be expressed as (Knoll, 2000):

$$E_{\gamma} = E_{\gamma 1} \cos \theta + E_g \cos \phi \quad (\text{Eq. 3.4})$$

The energy of the recoil electron can vary from ($\theta = 0$) up to a maximum value ($\theta = \pi$) depending upon the angle of scatter. The maximum energy of recoil electron is given by (Knoll, 2001);

$$E_{\gamma} = \frac{E_{\gamma}}{1 + \frac{m_0 c^2 (1 - \cos \theta)}{E_{\gamma}}} \quad (\text{Eq. 3.5})$$

where E_{γ} is the energy of the incident photon, $m_0 c^2 = 0.511$ MeV, is the rest mass energy of the electron. Compton scattering is the dominant interaction process for gamma ray energies ranging from 0.1 to 10 MeV (Das and Ferbel, 2003). At higher energy, another interaction mechanism, known as pair production becomes more significant.

3.1.3 Pair Production

In this process photons interacts within the coulomb field of the nucleus in which the gamma ray photon is absorbed into the hole and is converted into an electron positron pair. The process is only possible if the gamma ray which interacts has energy greater than 1.022 MeV. i. e. energy greater than twice the rest mass of an electron. Since an initial photon energy of at least $2 m_0 c^2$ is required for the creation of an electron-positron pair, any excess energy ($E_{\gamma} - 2 m_0 c^2$) carried in by the photon above 1.022 MeV is imparted to and shared equally by the positron and electron as kinetic energy as shown by the equation below (Knoll, 2000).

$$E_{e-} + E_{e+} = h\nu - 2 m_0 c^2 \quad (\text{Eq. 3.6})$$

The electron and positron pair transfers their energy to the surrounding medium through ionization, excitation and/or bremsstrahlung i.e. they can transfer their energy to the detector by secondary collisions with electron in the material. The two photons can be detected as single or double peaks depending on the detector. For example, using HPGe detector for environmental samples it possible to observe three ^{208}Tl peaks with energy 2614 keV, 2103 keV and 1592 keV. The positron can slow down and due to its positive charge it can combine with an electron and subsequently annihilate to form two photons with each energy of about 0.5 MeV and they are emitted at 90° and opposite in their direction.

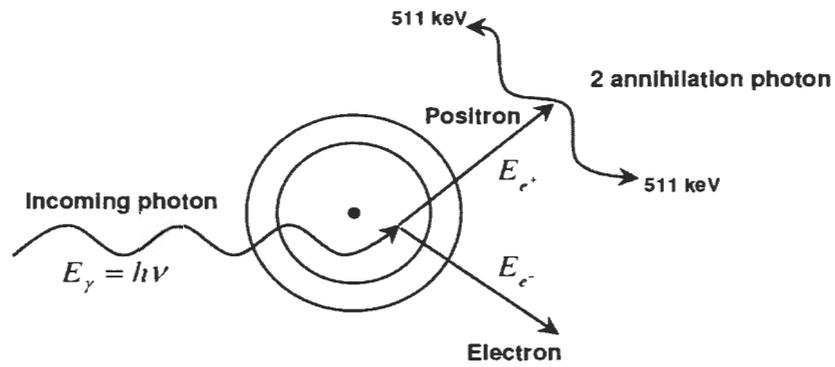


Figure 3.4: Schematic of the pair production process and annihilation(Gilmore and Hemingway, 2008)

The interaction cross section for pair production is related to the atomic number of the material as proportional to Z^2 (Turner, 2007). The chance of an electron undergoing annihilation depends the gamma ray energy above the threshold of 1.022 MeV, pair production becomes the dominant interaction process for gamma energy greater than 10 MeV (Das and Ferbel, 2003).

The relative importance of the three interaction process as a function of gamma ray energy and the value of Z absorber can be shown in the figure 3.5.

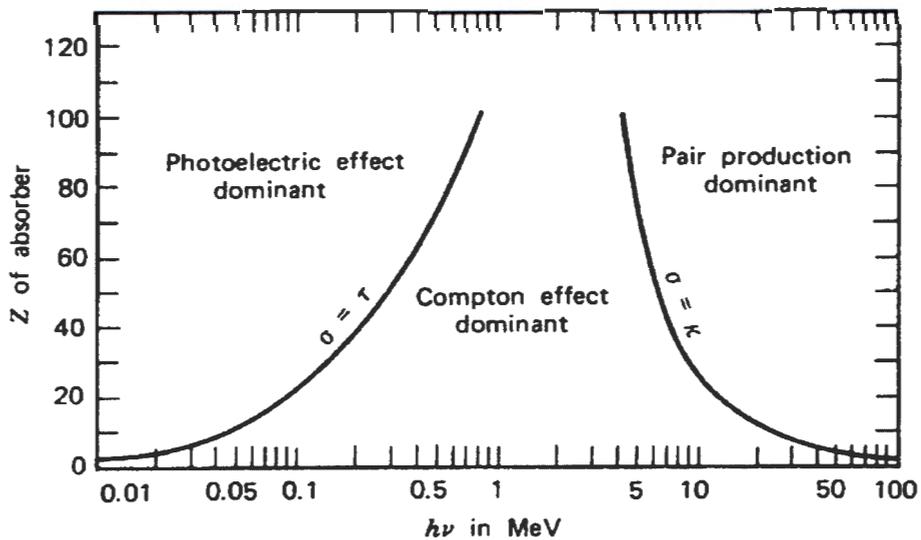


Figure 3.5: Interaction of three gamma ray photons and their region of dominance(Knoll, 2000)

3.2 Determination of radioactivity

The interaction between radiation and matter is accompanied by a number of effects such as the emission of photons, charged particles and liberation of heat. All of these can be used to detect radiation, measure particle flux density, intensity or the radiation spectrum. The difference of each detector depends on its detecting medium which can be a gas, liquid or solid materials. The three main classifications of devices used in the determination of ionizing radiation are:

- (i) Gas filled detector (ionizing chambers, proportional counter and Geiger-Muller Counter)
- (ii) Scintillation counters (organic phosphors, inorganic phosphors and the various types of scintillation counters)
- (iii) Solid state detectors (semiconductor detectors)

All types of radiation detection instruments however, involve generating an electrical signal which results from the interaction of radiation with the detector material. The detection mechanisms are basically based on the process of excitation or ionization of atoms in the detector by the passage of a charged particle.

Gas filled detectors depend on the interaction of the electric field of moving particles with detector material (gas) to produce ionization which is converted to an electrical pulse.

Scintillation counters require the use of scintillators, organic or inorganic crystals with special properties. The radiation from the source must be absorbed in the scintillator leading to re-emission of light photons (Choppin et al., 2002). In gamma spectroscopy, thallium-activated sodium iodide scintillator (NaI(Tl)) is the most frequently used because it has higher efficiency for detecting gamma radiation and does not require cooling. However, scintillation detectors do not provide the energy selectivity to deal with a complex gamma-ray spectrum, due to their relatively poor energy resolution (Cember and Johnson, 2009). To achieve good energy resolution, semiconductor detectors such as Si or Ge are used. Upon interaction with radiation material, electron-hole pairs are created. The pairs created are separated from recombination with the help of an externally applied electrical field. The electrons drift to the anode and holes to the cathode. The signal collected in the preamplifier then goes to the shaping amplifier. It should be stated that high purity germanium (HPGe) detector has become the solid state detector of choice for many samples from the environment because of its superior energy resolution due to its lower potential and compact size.

3.2.1 The HPGe detector

The HPGe detector is used for both quantitative and qualitative determination of gamma ray photons in environmental samples. This is a non-destructive technique which has great advantage of not going through the process of sample preparation. The principle underlying gamma-ray spectrometry is that each gamma-ray has a discrete energy, which is a characteristic of the source. Therefore, by measuring the energy of gamma ray photons of any material, the source of radiation can be determined with great accuracy. The most spectacular aspect of gamma ray is its superior energy resolution.

3.2.2 Detector assembly

Germanium detectors of any type cannot operate at room temperature due to small band gap (0.7 eV) which stems from large thermally-induced leakage current at this temperature (Gilmore and Hemingway, 2008). To circumvent this problem, germanium detectors are cooled to the point where this thermal leakage no longer spoils the excellent energy detections and this temperature occurs at -196.15°C and is achieved through the use of liquid nitrogen to cool the detector as shown in Figure 3.6.

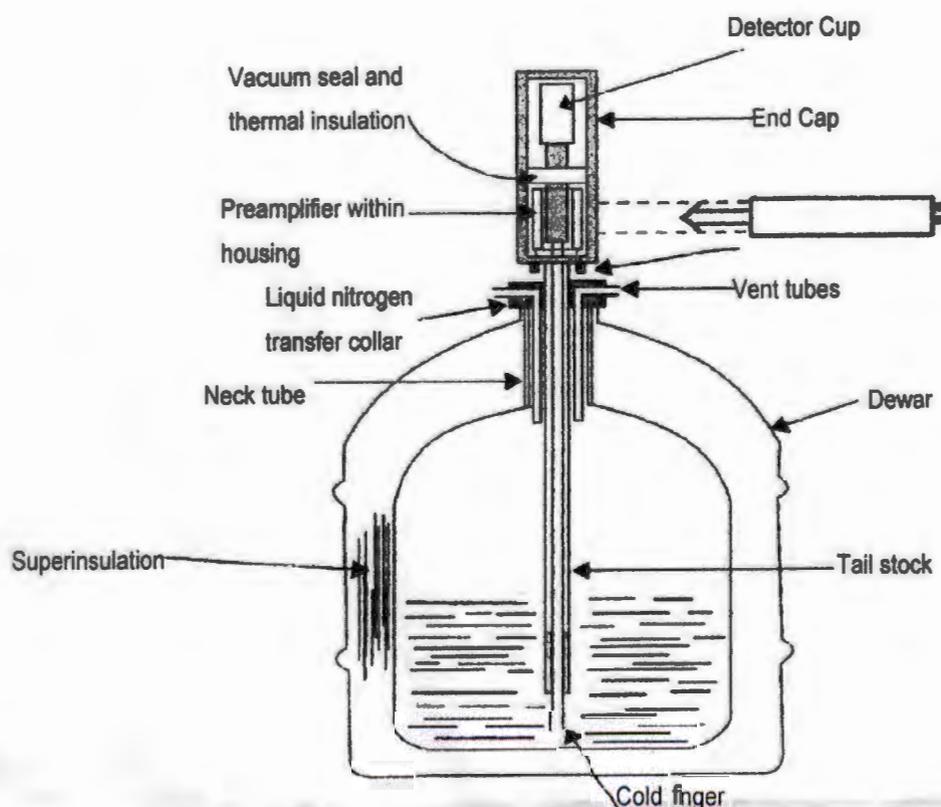


Figure 3.6: A typical germanium detector cooled by reservoir of liquid nitrogen (Gilmore and Hemingway, 2008)

3.2.3 Characteristic of gamma spectrometry

Gamma ray spectrometer was used for the estimation of the activity concentration of ^{238}U , ^{232}Th and ^{40}K in soil samples. Components of the gamma spectrometer consisted of the following;

- A Canberra hyper pure germanium coaxial detector with 15% relative efficiency of vertical configuration that is mounted on 10 liter liquid nitrogen Dewar for germanium crystal temperature control. The preamplifier is coupled to the detector and connected to the germanium crystal, so that an input component is kept at liquid nitrogen.
- A high voltage power supply with positive and negative polarity, with very low noise and very voltage stability
- Bin and power supply
- Spectroscopy amplifier which is an integral part of the low noise system to obtain an optimum signal to noise ratio.
- An analog-to-digital converter (ADC) which is intended to provide ultimate resolution, stability and linearity
- Multichannel analyzer with 9192 channels and with counting capability of 228 counts per channel.

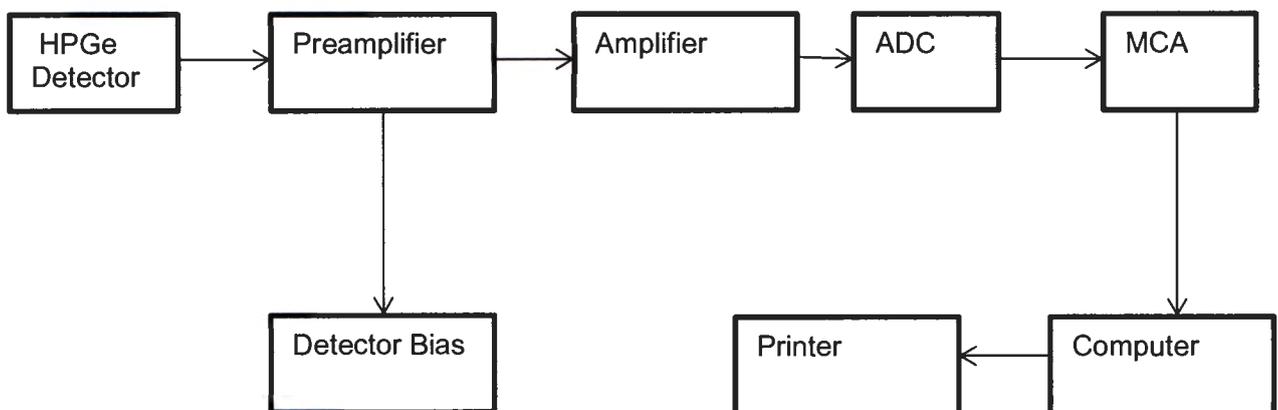


Figure 3.7: Block diagram of gamma-ray Spectrometer set up

3.2.3.1 Energy calibration of the detector

Energy calibration of the detector is carried out to identify the photo peaks present in the spectrum (IAEA, 1989). The detector system was calibrated using a mixed in-house reference standard of ^{152}Eu (half-life, 13.48 y) and ^{133}Ba (half-life, 10.53 y). The reference standard of ^{133}Ba and ^{152}Eu have well defined gamma energies within the region of interest from 79.61 keV to 1408 keV. The standard was weighed in 1.0 L Marinelli beaker and measured for 86400 seconds to acquire spectra data. The channel number that corresponds to the centroid of each full energy event on MCA was recorded and plotted to obtain a linear curve with second order polynomial. The linear curve obtained from the data points is an indication that the system is operating properly (IAEA, 1989).

3.2.3.2 Efficiency of the detector

The efficiency of the detector refers to the ratio of actual events registered by the detector to the total number of events emitted d by the source of radiation. An accurate efficiency calibration of the system is necessary to quantify the radionuclide present in the sample.

In general, the efficiency of the detector decreases logarithmically as a function of energy and it is geometric dependent. In this study a three point calibration point was used to determine the efficiency of the detector. The following expression was used to determine the efficiency (Darko and Faanu, 2007).

$$\eta(E) = \frac{N_T - N_B}{P_E A_{STD} T_{STD}} \text{ (Eq. 3.7)}$$

where;

P_E is the gamma emission probability for energy (E),

$\eta(E)$ is the efficiency of the detector,

N_T is the total counts under photo peak,

N_B is the background count,

A_{STD} is the activity in (Bq) of the radionuclide in the calibration standard at the time of calibration,

T_{STD} is the counting time of the standard.

4. BIOLOGICAL EFFECT OF IONIZING RADIATION

4.1 Biological effect of ionizing radiation

When the human body is exposed to any form of radiation, either from external or internal sources, ionization and excitation of atoms and molecules can be produced. This results in the alteration and eventual damage or death of cells that will affect the function of these organs or tissues. The damage to the deoxyribonucleic acid (DNA) in the nucleus is the main initiating event by which radiation causes long-term harm to organs and tissues in the body (UNSCEAR, 2000). It has been reported that double strand breaks in DNA are responsible for causing critical damage. Disrepair and radiation damage could also lead to potential for progression to cancer induction or hereditary disease (UNSCEAR, 2000). The mechanism of the biological effect arising from exposure to ionizing radiation is a result of direct and indirect ionization as figure 4.1.

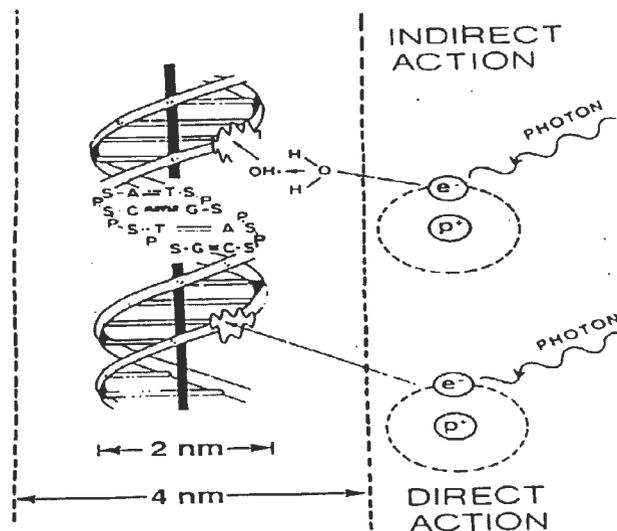


Figure 4.1: Mechanism of direct ionization and indirect ionization of DNA molecule (Faanu, 2011)

4.1.1 Direct ionization

When ionization radiation interacts with matter in the human body, a series of long and complex events are initiated through ionization and excitation of relatively few molecules. Every interaction involves the transfer of a small amount of energy from the radiation medium, usually in packets of about 100 electron volts (eV) or less. The transfer occurs in a very short time and are spatially distributed in a haphazard fashion throughout the medium along the tracks of the

charged particle. The dissociation, due to ionization or excitation of an atom on the DNA molecule leads to alteration in the genetic code. Consequently, the information in the DNA will be distorted and mutation will occur. If such point mutation occurs in gametic cells this information will be passed on to the next generation. Since these point mutation are eventually passed to successive generation, it is clear that the dose is cumulative, every little dose may result in a change in genetic makeup, which is continuously transmitted (UNSCEAR, 2001).

4.1.2 Indirect ionization

In indirect ionization, the positive ions and free electrons react with neutral molecules forming two kinds of extremely dangerous products free radicals and peroxides. These materials are extremely reactive i.e. they attack any molecule in their vicinity. This free radical can move some distance from the site of original event and cause further chemical changes such as causing the molecule to break up. The molecule attacked by a free radical can become a free radical itself; in this way a cascading of chemical reactions is set in motion. Essential molecules are thus destroyed and function less or even poisonous molecules which are foreign to the function of the cell are produced. In this way, the essential life –functions of the cell may be suppressed and genetic material may be damaged.

The variation of the biological effects of radiation depends on the type of radiation and its energy which is transferred to the irradiated parts of the tissues and organs during the exposure time (Noz et al, 2008). The quantification of the amount of ionization which occurred and the energy absorbed by particular cells are associated with biological effectiveness which can be considered in terms of radiation dosimetry.

4.2 Radiation dosimetry

4.2.1 Basic radiation Quantities and units

Exposure

The amount of ionization that X-ray and gamma rays produced in the air is known as exposure and is measured in units called Roentgen, R. This unit accounts for the production of 1esu of electrical charge of either sign in 1cm³ of air at standard temperature and pressure. The standard unit of exposure can be expressed as 2.58×10^{-4} C/kg. The exposure is designated only for limited energy range X-ray and gamma rays interacting with air (Noz et al., 2008).

4.2.1.1 Absorbed dose, D

The major weakness with exposure is that it does not reflect the biological significance of the radiation. Another unit known as the absorbed dose was introduced which take into account the quantity of energy absorption by any kind of ionizing radiation. The absorbed dose is the amount of energy imparted by ionizing radiation in a volume element of a specified material divided by the volume element.

$$D = \frac{\Delta\varepsilon}{\Delta m} \quad (\text{Eq. 4.1})$$

where D is the absorbed dose and $\Delta\varepsilon$ is the mean energy imparted into matter of mass Δm . The energy imparted is the incident energy minus the energy leaving the mass; minus the energy released in nuclear transformations. The international unit (SI) of measure for absorbed dose is the gray (Gy), which is defined as 1 joule of energy deposited per kilogram of medium. As the mass of a sample decreases in general the energy per unit mass will become more random (stochastic). For this reason, the term relative biological effectiveness (RBE) was introduced as a dimensionless quantity of the amount of absorbed dose of ionizing radiation relative to X-rays and gamma rays of particular energy to provide some biological response. (Cember and Johnson, 2009). Because of the difficulty in applying such complicated functions of energy, the RBE has been normalized to a factor known, as the radiation weighting factor (W_r) by ICRP (Noz et al., 2008).

4.2.1.2 Equivalent and Effective dose

The equivalent dose is used to assess the extent of biological damage from different types of radiation. This takes into consideration the tissue weighting factor. The equivalent dose, H_T in an organ or tissue is equal to the sum of absorbed dose $D_{T,R}$ in an organ or tissue caused by different types of radiation with so called weighting factors, W_R (Lilley, 2001);

$$H_T = \sum_R W_R \times D_{T,R} \quad (\text{Eq.4.2})$$

The standard unit of equivalent dose is called the Sievert (Sv). The old unit was the radiation equivalent man "rem" and 1 Sv equals to 100 rem. The weighing factor is introduced to measure the absorbed dose for biological effectiveness of the particles. The variation of radiation sensitivity of each organ is taken into account in the contribution of the equivalent dose in all tissues and organs of the body.

The new terms effective dose and the tissue weighting factor (W_T) are introduced. Thus, the equivalent dose is defined as the sum of equivalent doses weighted by tissue weighting factors for each tissue as given by the following expression.

$$E(Sv) = \sum_T W_T \times H_T \quad (\text{Eq.4.3})$$

The effective dose takes into account the following; the absorbed dose to all organs; the relative harm of the radiation and the sensitivity of each organ to radiation. Some tissue weighting factors for tissues and organs of the human body are shown in table 4.1

Table 4.1: Tissue weighting factors (ICRP, 1991)

Tissue or organ	Tissue weighting factors, W_T
Gonads	0.20
Colon	0.12
Lung	0.12
Red bone marrow	0.12
Stomach	0.12
Bladder	0.12
Breast	0.05
Oesophagus	0.05
Liver	0.05
Thyroid	0.05
Skin	0.01
Bone surfaces	0.01
Remainder	0.05

4.3 Radiation Protection and dose limits

Much of the knowledge of radiation effects on humans have been obtained from a group of people who survived from the atomic bombs in Hiroshima and Nagasaki and those individuals who received radiation exposure from routine work or nuclear accidents (Noz et al., 2008). The relationship between biological effect and radiation exposure was studied by Biological Effect of Ionizing Radiation (BEIR) in agreement with United Scientific Committee on Effect of Atomic Radiation (UNSCEAR) and the commission on Radiological Protection

ICRP).The effect of radiation exposure can be classified into deterministic effects and stochastic effects (Martin et al., 2012).

4.3.1 The deterministic effects

The deterministic effects occur when the whole body or part of the body is exposed to sufficient radiation causing cell damage or killing of the whole cell, thus hinders proper functioning of the irradiated tissue or organ. The degree of cell damage of deterministic events depends on dose rate imparted to the exposed individual. For example, when different individuals with varying susceptibilities are exposed to radiation, the threshold for deterministic effects of sufficient severity will occur at low dose rate in more sensitive people (Canu et al., 2011). However, the dose rate has been found to increase with time of exposure until the whole population show signs of deterministic effects at high dose (Nui et al., 2010). According to Cember and Johnson (2009), different types of organs show significant variation in radiosensitivity when exposed to the same time of exposure. For example, actively growing cells that are young and undifferentiated are more radiosensitive than matured and differentiated cell.

4.3.2 Stochastic effects

These effects occurs based on probability and can be found in both exposed and unexposed individuals (Cember & Johnson, 2009). Such events are thought not to have a no threshold value and thus, are not related to exposure to noxious agents or radiation. It is therefore imperative that even when standards are met, there still exist a small possibility for the occurrence of stochastic effects. It is difficulty to completely eliminate stochastic effects but their effects can be minimized (Simmons et al., 1995). Conventionally, any mutation that has occurred on the DNA can lead to cancers in the somatic cells.

To avoid unnecessary exposure causing biological effects of radiation to radiation workers and the general public, all doses must be kept as low as reasonably achievable (ALARA) as recommended by ICRP (Noz et al., 2008). The dose limits recommended by ICRP are shown in table 4.2.

Table 4.2: ICRP 600 recommended effective dose limits (Cember & Johnson, 2009).

Application	Dose limit	
	Occupational	Public
Whole body	20mSv ^a per year, averaged over a period of 5 years ^b	1 mSv ^a in a year
Annual equivalent dose in		
Lens	150mSv	15 mSv
Skin	500 mSv	50 mSv
Hand and feet	500 mSv	-

^a To find the recommended limit in rem, 1mSv = 0.1 rem

^b Maximum dose in any single year do not exceed 50 mSv

5. RESEARCH METHODOLOGY

5.1 Sample collection

The Erongo region of Namibia is located at central western part of Namibia as shown in figure 5.1. The region covers land area of 63,549 km² and has a population of about 150,400 and a population density of 2.4 km⁻² (<http://www.gov.na/documents>). Much of the region is occupied by the Namib desert which stretches parallel to the coast for about 120 to 150km inland. The two coastal towns under this study, Walvis bay and Swakopmund are 60 km and 40 km from most uranium mine sites including Rössing Uranium Mine (RUM) as depicted in figure 5.1

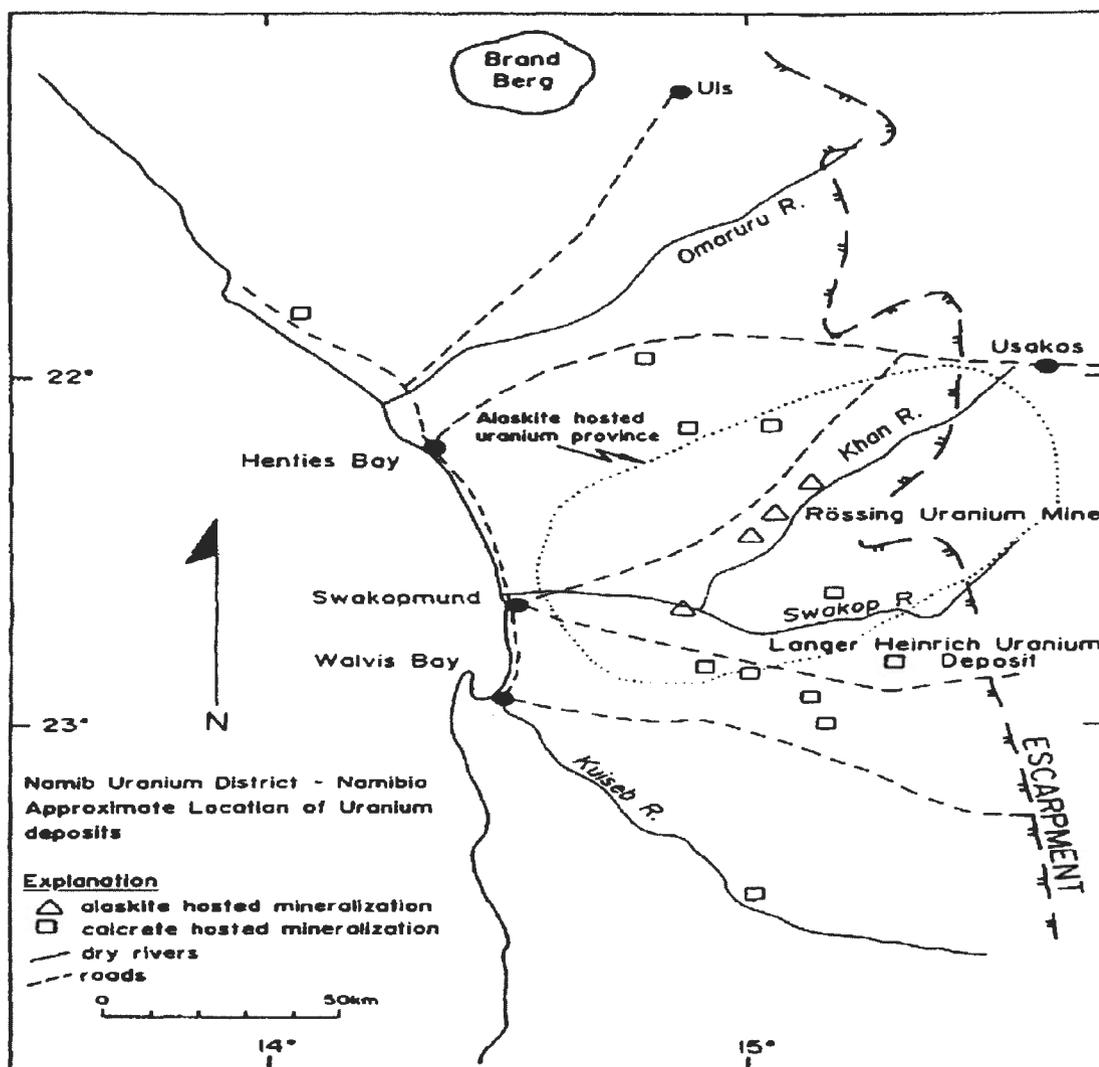


Figure 5.1: Map of Erongo region of Namibia showing the relative distance of Walvis bay and Swakopmund town from Rössing Uranium mine (Palfi, 1997).

The arid nature of the landscape means that very little of the area has agricultural potential. Only 10 km² of the Erongo Region is cleared for cultivation; this includes the area of small-scale farming in the Swakop River bed, as well as small areas at Omaruru and Okombahe. The main industrial activities in Erongo region are fishing, tourism, and mining. In Namibia, there are many mineral resources including uranium and therefore the concentration of radioactive material in the soil may be high in some locations, including the Erongo region thus making such places areas of high background radiation. High levels of natural radiation near uranium mines may make living conditions hazardous (SEA, 2010).

A total of twenty surface soil samples were collected from different geographical areas of the two towns of Walvis Bay and Swakopmund towns which are shown in Figure 5.2 and 5.3 respectively. At each geographical area for each town, ten soil samples of approximately 2.0 kg were randomly collected after marking the sampling site with a Global Positioning System (GPS). The sampling sites include open space such as playground, areas along the road, areas along the beach and those areas within the residential areas. For each sampling site, an area of about 1 m² was marked and carefully cleared of debris to a few centimetres depth. Surface soils were extracted using a soil auger to a depth of about 0.5 m and placed in plastic bags. The soil samples were mixed together thoroughly, in order to obtain a representative sample of the area. Each soil sample was labelled according to the geographical coordinates of the sampling area, and those coordinates were later used to indicate the position on the simplified map by an open circle point. All the samples were transported to the Centre for Applied Radiation and Technology (CARST), laboratory, North West University, Mafikeng Campus, South Africa, for processing.



Figure 5.2: Location of the ten geographical sampling sites where soil samples were collected in Walvis Bay



Figure 5.3: Location of the ten geographical sampling sites where soil samples were collected in Swakopmund

5.2 Sample Preparation

The samples were air dried for about four days to ensure that all the moisture was eliminated. The rocks and pebbles were removed from the sample leaving fine soil particles. Each soil sample was weighed to about 1.0 kg and placed in a pre-weighed 1 L Marinelli beaker. The beaker was hermetically sealed and stored for about four weeks prior to counting to allow radionuclides to attain secular equilibrium with their progeny. It should be noted that equilibrium is common in rocks older than 10^6 years, and the ^{232}Th series may be considered in equilibrium in most geological environments (Chiozzi et al, 2002).

5.3 Sample counting and measurements

5.3.1 Activity concentrations

Considering that secular equilibrium is established between ^{238}U and ^{232}Th and their decay products, the activity concentration of ^{238}U was calculated from the average peak energies of 1120.27 keV of ^{214}Bi and 609.31 keV of ^{214}Pb . Similarly, the activity concentration of ^{232}Th was determined from the average energies of 238.63 keV of ^{212}Pb and 911.21 keV of ^{208}Tl (Hamby and Tynybekov, 2002). The activity concentration of ^{40}K was determined from the energy of 1460.83 keV. The activity concentrations of each the radionuclides were divided by the mass to express the results in Bq kg^{-1} . An in-house reference material of $^{133}\text{Ba}/^{152}\text{Eu}$ was used. The nuclear data of the radionuclides used are presented in Table 5.1.

Table 5.1: Nuclear data of radionuclides used in the analysis.

Parent nuclide	Daughter nuclide	Gamma Energy (KeV)	Gamma Ray Abundance (%)
^{238}U	^{214}Bi	1120.27	15.0
	^{214}Pb	609.31	46.10
^{232}Th	^{212}Pb	238.63	53.65
	^{208}Tl	911.21	26.60
^{40}K	-	1460.83	10.67

The environmental gamma ray background at the laboratory site was determined using an empty Marinelli beaker under identical conditions. The background was then appropriately

subtracted from the measured gamma ray spectrum of each sample. The data for sample was acquired with WINSPEC software and it was analysed by identify software.

5.3.2 Elemental concentration of ^{238}U , ^{232}Th and ^{40}K in surface soils.

The measured activity of ^{238}U , ^{232}Th and ^{40}K (Bq.kg^{-1}) were used to calculate the elemental concentrations of uranium in ppm or mg kg^{-1} , thorium in ppm or mg kg^{-1} and potassium in (%) by using conversion factors recommended by IAEA technical document 1363 as follows;

$$1\% \text{ K} = 313 \text{ Bq.kg}^{-1} \text{ of } ^{40}\text{K}$$

$$1 \text{ ppm U} = 12.35 \text{ Bq.kg}^{-1} \text{ of } ^{238}\text{U}$$

$$1 \text{ ppm Th} = 4.06 \text{ Bq. kg}^{-1} \text{ of } ^{232}\text{Th}$$

5.3.3 Absorbed dose rate (D)

The measured activity of ^{238}U , ^{232}Th and ^{40}K were converted into doses by applying the factors 0.462, 0.604 and 0.0417 for uranium, thorium and potassium respectively (Tzortzis et al, 2003). These factors were used to calculate the external gamma dose rate (D) at 1.0 m above the ground level using the equation 5.1 (Uosif, 2011).

$$\text{Absorbed dose rate (D) (nGyh}^{-1}\text{)} = 0.0417A_{\text{K}} + 0.462A_{\text{U}} + 0.604A_{\text{Th}} \quad (\text{Eq.5.1})$$

where A_{K} , A_{U} and A_{Th} are the activities of ^{40}K , ^{238}U and ^{232}Th in Bq kg^{-1} respectively. The absorbed dose rate at each geographical location and each site where the samples were collected were calculated. The results are shown in table 6.6.

5.3.4 Annual effective dose equivalent (AEDE)

To estimate the annual effective doses, account must be taken of (a) the conversion coefficient from absorbed dose in the air to effective dose and (b) the outdoor occupancy factor. Annual estimated average effective dose equivalent received by a member is calculated using a conversion factor of 0.7SvGy^{-1} which is used to convert the absorbed rate to annual effective with outdoor occupancy of 20% (UNSCEAR, 1993). This is given by the equation in 5.2.

$$H_{\text{E}} = D \times T \times F \quad (\text{Eq.5.2})$$

where H_{E} is the annual effective dose (mSv), D is the absorbed dose rate (nGyh^{-1}), T is the outdoor occupancy time (365 days x 24h x 0.2) and F conversion factor ($0.7 \times 10^3 \text{ mSv}/10^9 \text{ nGy}$). The results obtained were used to calculate the average effective dose equivalent for each

geographical site of each town. Table 6.6 shows the annual outdoor effective dose equivalent and the range of each geographical site of each town.

5.3.5 Radium Equivalent Activity (Ra_{eq})

It is not conclusive to estimate the radiation hazards associated with soil by using effective dose only since absorbed dose is applicable to soils with uniform radioactivity. In nature the concentration of radionuclides are not uniform so it is imperative to express the radiological effects by a single term which takes into account the hazards associated with all the primordial radionuclides. This term is the “radium equivalent”. The radium equivalent is the most useful guideline for regulating safety standards on radiation protection for the general public. The radium equivalent is the weighted sum of activities of ^{226}Ra , ^{232}Th and ^{40}K and can be expressed by the mathematical equation 5.3 (Beretka and Mathew, 1985). The equation is based on the assumption that 259 Bq kg^{-1} of ^{232}Th , 370 Bq kg^{-1} of ^{226}Ra and 4810 Bq kg^{-1} of ^{40}K produce the same gamma –radiation dose rates.

$$Ra_{eq}(\text{Bq kg})^{-1} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (\text{Eq.5.3})$$

where Ra_{eq} represents the radium equivalent activity; and A_{Ra} , A_{Th} and A_K are the activities of radionuclides in Bq kg^{-1} . The permissible maximum value of radium equivalent activity is 370 Bq kg^{-1} which corresponds to the dose limit of 1.0 mSv for the general population (UNSEAR, 1988).

5.3.6 The external hazard Index (H_{ex})

To ensure that the radiation exposure due to natural radionuclides in the soil samples does not exceed the permissible dose equivalent of 1 mSv yr^{-1} , the external hazard index have been introduced using a model proposed by Krieger (1981) which is given by the mathematical equation 5.4. (Krunaz et al, 2007)

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (\text{Eq. 5.4})$$

In order to ensure that the radiation hazard is insignificant, the external hazard index must be less than unity. The unity of one represent maximum of value of H_{ex} which corresponds to the upper limit radium equivalent of 370 Bq kg^{-1} (Dragović and Onjia, 2006).

6. RESULTS AND DISCUSSIONS

6.1 Activity concentrations of ^{238}U , ^{232}Th and ^{40}K in surface soils.

The results for average activity concentrations of ^{238}U , ^{232}Th and ^{40}K in soil samples collected from Walvis Bay and Swakopmund are presented in Table 6.1 and Table 6.2 respectively.

6.1.1 Activity concentrations of ^{238}U , ^{232}Th and ^{40}K in surface soils collected in Walvis Bay

Table 6.1: Average radionuclides concentrations of the ten geographical soil samples of Walvis Bay town.

Sample ID	Activity concentrations, Bq.kg ⁻¹		
	^{238}U	^{232}Th	^{40}K
WB1	35.53±5.9	52.51±9.02	208.75±69.07
WB2	37.35±5.69	34.52±5.42	162.58±53.55
WB7	20.50±02.26	23.97±4.73	196.77±64.52
WB8	21.83±03.98	17.68±2.39	195.17±96.55
WB10	48.24±7.31	32.44±3.78	226.79±75.00
WB13	40.02±6.02	39.64±6.9	188.15±62.22
WB17	14.94±2.24	41.89±7.88	208.73±103.17
WB20	22.22±02.45	25.99±5.12	213.29±69.93
WB22	22.08±02.92	26.77±04.28	176.58±58.23
WB23	41.07±6.13	30.38±4.89	259.35±90.32
Range	(14.94-48.24)	(17.68-52.51)	(162.58-259.35)
Mean	30.38±04.49	32.58±5.44	203.62±74.26

The results for the average activity concentrations of ^{238}U , ^{232}Th and ^{40}K (Bq kg⁻¹) of soil samples from Walvis Bay are summarized in table 6.1, where the range and the mean are listed. The measured activity concentrations in the soil samples are in the range of 14.94±02.24 to 48.24±7.31 Bq kg⁻¹ for ^{238}U , 17.68±02.39 to 52.51±09.02 Bq kg⁻¹ for ^{232}Th , 162.48±53.55 to 259.35±90.32 Bq kg⁻¹ for ^{40}K . The highest values of ^{40}K of 259.35±90.32 Bq kg⁻¹ was measured in soil samples taken closer to the sand dunes, which is WB 23. High levels of ^{40}K may be attributed to presence of loamy and clay soils which are continuously deposited by sand dunes of the dry Namib desert (El-Gamal, Nasr, & El-Taher, 2007). The activity concentration of ^{238}U , ^{232}Th and ^{40}K obtained for the soils of Walvis Bay town are comparable to the world ranges of 11 to 64 for ^{238}U , 17 to 60 for ^{232}Th and 140 to 850 Bq kg⁻¹ for ^{40}K (UNSCEAR, 2000). The mean

activity concentrations of the radionuclides are of the order $^{238}\text{U} < ^{232}\text{Th} < ^{40}\text{K}$ and these are presented as $30.38 \pm 4.49 \text{ Bq kg}^{-1}$, $32.58 \pm 5.44 \text{ Bq kg}^{-1}$ and $203.62 \pm 74.26 \text{ Bq kg}^{-1}$, respectively. These mean activity values falls within some values obtained in some parts of Africa and the world of 30 Bq kg^{-1} for ^{238}U and 35 Bq kg^{-1} for ^{232}Th (Sahin and Cavas, 2008). While the mean activity concentration of ^{40}K is relatively higher in comparison to the ^{238}U and ^{232}Th , it is however, lower than the average world values of 500 Bq kg^{-1} (UNSCEAR, 1993). This can be attributed to the type of underlying geological rocks and a marked absence of agricultural activities which under most circumstances are the main contributors to increased levels of ^{40}K in the surface soils.

The activity ratios between parent/daughter and/or between parent/granddaughter were used to confirm the assumption of radioactive equilibrium through the decay chains. The activity of $^{226}\text{Ra}/^{214}\text{Pb}$ and $^{214}\text{Pb}/^{214}\text{Bi}$ in the ^{238}U decay series and those of $^{228}\text{Ac}/^{212}\text{Pb}$, $^{212}\text{Pb}/^{212}\text{Bi}$ from the ^{232}Th decay series was found to be approximately one. These results are indicative that the state of secular equilibrium has been achieved in all samples.

6.1.2 Activity concentrations of ^{238}U , ^{232}Th and ^{40}K in surface soils collected in Swakopmund

Table 6.2 illustrates the average radionuclides concentration of ^{238}U , ^{232}Th and ^{40}K from the soil samples collected in the ten geographical locations of Swakopmund. As can be observed from the results, the activity concentrations of ^{238}U , ^{232}Th and ^{40}K vary from 71.38 ± 11.1 to $155.80 \pm 33.71 \text{ Bq kg}^{-1}$ for ^{238}U , 41.63 ± 05.00 to 184.53 ± 28.61 for ^{232}Th and 360.82 ± 121.43 to $610.87 \pm 155.07 \text{ Bq kg}^{-1}$ for ^{40}K . SW16 has a higher concentration of ^{238}U which is $155.80 \pm 33.71 \text{ Bq kg}^{-1}$. The mean activity concentrations of ^{238}U , ^{232}Th and ^{40}K are $99.59 \pm 17.41 \text{ Bq kg}^{-1}$, $119.23 \pm 16.14 \text{ Bq kg}^{-1}$ and $553.07 \pm 155.79 \text{ Bq kg}^{-1}$ respectively. The results for the mean activity concentrations thorium and potassium are comparable with the previous studies in the same locale which has values of $91.10 \pm 39.6 \text{ Bq kg}^{-1}$ for ^{232}Th and $645.50 \pm 69.50 \text{ Bq kg}^{-1}$ for ^{40}K .

Table 6.2: Average radionuclides concentrations of the ten geographical soil samples of Swakopmund town.

Sample ID	Activity concentrations, Bq.kg ⁻¹		
	²³⁸ U	²³² Th	⁴⁰ K
SW 1	117.45±16.08	118.00±17.02	421.05±136.84
SW 4	92.19±9.54	57.51±7.18	360.82±121.43
SW 6	85.89±9.47	131.58±19.26	553.68±131.58
SW 7	79.38±14.53	56.25±08.73	574.22±187.96
SW 8	71.38±11.1	41.63±05.00	537.76±104.95
SW 9	91.40±12.60	74.06±05.97	610.87±155.07
SW 12	111.48±37.07	99.95±13.49	761.76±151.38
SW 15	87.04±15.79	127.48±15.80	551.68±182.77
SW 16	155.80±33.71	92.38±20.90	581.27±194.08
SW 20	103.93±14.10	110.20±26.45	577.63±191.79
Range	(71.38-155.80)	(41.63-131)	(360.82-761.76)
Mean	99.59±17.41	119.23±16.14	553.07±155.79

6.1.3 Comparison of activity concentrations in Walvis Bay and Swakopmund

Swakopmund town has higher mean radionuclides concentrations of 99.59±17.41 Bq kg⁻¹ for ²³⁸U, 119.23±16.14 Bq kg⁻¹ for ²³²Th and 553.07±155.79 Bq kg⁻¹ for ⁴⁰K compared to Walvis Bay with 30.38±09.98 Bq kg⁻¹ for ²³⁸U, 32.82±11.96 Bq kg⁻¹ for ²³²Th and 197.42±72.47 Bq kg⁻¹ for ⁴⁰K. This can be attributed to their geographical locations from the uranium mines. Swakopmund and Walvis Bay are located at a distance of 40 and 60 km from Rossing Uranium mine and consequently Swakopmund town receive high levels of dust emissions giving rise to ambient pollution concentrations and deposition levels derived from anthropogenic, natural and biogenic sources (Friedrich, 2009). Windblown dust from natural mineral resources is estimated to account for 89% of the global aerosol load (Satheesh and Moorthy, 2005) while mining operations and aggregate extraction sites are significant sources of fugitive dust emissions containing radionuclides such as of ²³⁸U, ²³²Th and ⁴⁰K (Neuman et al, 2009). The chemical behaviour of the radionuclides is also important in understanding their solubility and transport in water. In low pH, uranium is highly soluble in water compared to thorium and hence is highly mobile in the environment where it can be leached into soil and underground sources of water. In contrast to uranium, thorium has a low solubility in natural water and is entirely transported as

particulate matter in the environment where it will be transported adsorbed onto surface of clay minerals. These reasons account for the difference in the activity concentrations between uranium and thorium in the two towns.

6.2 Elemental concentration of ^{238}U , ^{232}Th and ^{40}K in surface soils.

The results of elemental concentration of ^{238}U (mg kg^{-1}), ^{232}Th (mg kg^{-1}) and ^{40}K (%) for the town of Walvis Bay and Swakopmund are presented in Table 6.3 and 6.4 respectively. Table 6.5 shows the comparison of mass elemental concentration uranium, thorium and potassium in this study with values from other environments.

6.2.1 Elemental concentration of ^{238}U , ^{232}Th and ^{40}K in surface soils of Walvis Bay

Table 6.3: Calculated elemental concentrations of radionuclides in the ten geographical soil samples of Walvis Bay town.

Sample ID	Elemental concentrations of ^{238}U , ^{232}Th and ^{40}K		
	^{238}U (mg kg^{-1})	^{232}Th (mg kg^{-1})	^{40}K (%)
WB1	2.88±1.05	12.93±5.03	0.67±0.22
WB2	3.02±1.10	8.50±2.89	0.52±0.17
WB7	1.66±0.44	5.90±2.37	0.63±0.21
WB8	1.77±0.63	4.35±1.14	0.62±0.31
WB10	3.91±1.42	7.99±2.07	0.72±0.24
WB13	3.24±1.18	9.76±3.87	0.60±0.20
WB17	1.21±0.31	10.32±4.30	0.67±0.33
WB20	1.80±0.48	6.40±2.57	0.68±0.22
WB22	1.79±0.66	6.59±2.29	0.56±0.19
WB23	3.33±1.20	2.64±0.83	0.83±0.29
Range	(1.21-3.91)	(4.35-12.93)	(0.52-0.83)
Mean	2.46±0.85	8.02±2.92	0.65±0.24

The range of calculated elemental concentrations in all soil samples collected from Walvis bay varied from 1.21±0.31 to 3.91±1.42 mg kg^{-1} for ^{238}U , 4.35 ±1.14 to 12.93±5.03 mg kg^{-1} for ^{232}Th and 0.56±0.19 to 0.83±0.29 % for ^{40}K . In order to observe if there is any correlation between the concentrations of natural radionuclides at all sampling sites, the values obtained for radionuclides elemental concentration were plotted between $^{238}\text{U}/^{232}\text{Th}$, $^{238}\text{U}/^{40}\text{K}$

and $^{232}\text{Th}/^{40}\text{K}$. The correlations of elemental concentrations of nuclides is poor, with coefficients of 0.103 between ^{238}U and ^{232}Th ; 0.0021 for ^{232}Th and ^{40}K ; 0.087 for ^{238}U and ^{40}K which suggest that the underlying rock may be heterogeneous.

6.2.2 Elemental concentration of ^{238}U , ^{232}Th and ^{40}K in surface soils of Swakopmund

Table 6.4: Calculated elemental concentrations of radionuclides in the ten geographical soil samples of Swakopmund town

Sample ID	Elemental concentrations of ^{238}U , ^{232}Th and ^{40}K		
	^{238}U (mg kg ⁻¹)	^{232}Th (mg kg ⁻¹)	^{40}K (%)
SW 1	9.51±1.30	29.06±4.19	1.35±0.44
SW 4	7.46±0.77	14.17±1.77	1.15±0.39
SW 6	12.70±1.58	32.41±4.74	1.77±0.42
SW 7	6.43±1.18	13.85±4.61	1.83±0.60
SW 8	9.76±0.90	10.25±1.23	1.72±0.34
SW 9	13.01±1.84	18.24±1.47	1.95±0.50
SW 12	16.26±3.00	24.62±3.32	2.43±0.48
SW 15	10.77±1.28	31.40±3.89	1.86±0.22
SW 16	1.79±0.66	22.75±2.29	1.76±0.19
SW 20	3.33±1.20	27.14±0.83	1.86±0.29
Range	(1.79-16.23)	(10.25-32.41)	(1.15-2.43)
Mean	8.06±1.57	22.39±3.69	1.77±0.50

Table 6.4 shows the calculated elemental concentrations of radionuclides in the ten geographical locations of Swakopmund which varies from 1.79 to 16.23 mg kg⁻¹ for ^{238}U , 10.25 to 32.41 mg kg⁻¹ for ^{232}Th and 1.15 to 2.43 % for ^{40}K . The mean elemental concentrations for ^{238}U , ^{232}Th and ^{40}K was found to be 8.06±1.57 mg kg⁻¹, 22.39±3.69 mg kg⁻¹ and 1.77±0.50 % respectively. The average values for elemental concentrations are comparable to the values obtained in Amman/Jordan which are 6.80 mg kg⁻¹ for ^{238}U , 20.2 mg kg⁻¹ for ^{232}Th and 1.82 % for K (Al-Jundi, Al-Bataina, Abu-Rukah, & Shehadeh, 2003). However, the values are three to four times higher than the world average values.

Table 6.5 Comparison of mass elemental concentrations of uranium, thorium and potassium in this study with values from other environments.

Country/City	Mass elemental concentrations of ^{238}U , ^{232}Th and ^{40}K			Reference
	^{238}U (mg kg ⁻¹)	^{232}Th (mg kg ⁻¹)	^{40}K (%)	
Adana/ Turkey	1.42 (0.05-6.55)	5.21 (0.15-14.0)	0.97 (0.03-2.65)	Degerlier et al. (2008)
Amman/ Jordan	6.80 (1.78-8.42)	20.2 (5.18-25.4)	1.82 (0.45-1.96)	Al-Jundi et al. (2003)
Instanbul/Turkey	1.68 (0.24-4.78)	9.14 (1.98-22.5)	1.11 (0.38-3.92)	Karahan and Bayulken (2000)
Lagos/ Nigeria	2.60 (0.97-4.05)	3.46 (BDL-7.90) ^a	0.29 (0.10-0.80)	Obed et al. (2005)
Ibadan/ Nigeria	1.64 (0.82-3.30)	7.9 (3.28-10.3)	0.72 (0.31-1.09)	Jibiri and Bankole (2006)
Lahore/ Pakistan	2.08 (2.00-2.24)	12.2 (11.2-13.0)	1.83 (1.70-1.96)	Akhtar et al. (2005)
Manisa/ Turkey	2.35 (1.78-2.84)	6.67 (4.45-8.90)	1.11 (0.68-1.53)	Erees et al. (2006)
Riyadh/ Saudi Arabia	0.82 (0.41-1.09)	1.59 (0.94-2.82)	0.36 (0.21-0.62)	Al-Kahtani et al. (2001)
Sistan, Blouchestan/ Iran	1.77 (1.67-2.00)	8.15 (7.14-9.02)	1.54 (1.29-1.88)	Hosseini (2007)
Tripoli/ Libya	0.85 (0.70-1.04)	2.34 (1.88-2.40)	0.88 (0.86-0.92)	Shenber (1997)
Windhoek/ Namibia	2.00 (1.22-3.06)	8.69 (4.32-15.3)	1.69 (0.55-2.56)	Oyedele (2006)
Belgrade/ Serbia	2.57 (0.85-4.42)	9.40 (2.73-15.7)	1.60 (0.74-2.60)	Mandic et al. (2010)
Walvis Bay/ Namibia	2.88 (1.21-3.91)	8.02 (4.35-12.93)	0.65 (0.52-0.83)	Present Study
Swakopmund/ Namibia	8.06 (1.79-16.23)	22.39 (10.25-32.41)	1.77(1.15-2.43)	Present study

6.2.3 Comparison of elemental concentration in Walvis bay, Swakopmund and worldwide values.

The mean values for the elemental concentration of radionuclides due to ^{238}U , ^{232}Th and ^{40}K in Walvis Bay fall within worldwide values. However, the mean values for Swakopmund values are four times more than the previous studies conducted world-wide as observed in Table 6.5. The high radionuclides concentrations can be attributed to the proximity of the two towns relative to the uranium mines and as well as geological underlying rock. Swakopmund town is 40 km away from uranium mines and Walvis Bay located further down, is 60 km away. Accordingly, Swakopmund is likely to share the same geological underlying rock with uranium mines than Walvis, and consequently, the concentration of radionuclides are high in those soils. The map of Erongo region is shown in figure 5.1, most of the region is covered by intrusive granite rocks which are known to have high levels of ^{238}U , ^{232}Th and ^{40}K . The concentrations of these radionuclides in rocks and soil may vary because of alteration or metamorphic process (Verdoya et al, 2001). Their concentrations ratio may provide an indication whether depletion or enrichment has occurred.

6.3 Radiological Indexes in Walvis Bay and Swakopmund

The radiological hazard due to the contribution of ^{238}U , ^{232}Th and ^{40}K were used to estimate the radiation exposure doses and assessment of biological effect on human. These are considered in terms of absorbed dose rate (D), the Annual Effective Dose Equivalent (AEDE), the radium equivalent activity (Ra_{eq}) and the external hazard index (H_{ex}). The results for radiological risk assessment for the two coastal towns of Walvis Bay and Swakopmund are depicted in Table 6.6 and Table 6.7 respectively.

6.3.1 Radiological Indexes in Walvis Bay

The contributions of natural radionuclides to radiological hazard indexes to the general population and the environment in Walvis Bay are presented in Table 6.6; where the absorbed dose, the Annual Effective Dose Equivalent, the radium equivalent activity and the external hazard index are listed.

Table 6.6 Radiological indexes in Walvis bay

Sample ID	Radiological indexes due to natural radionuclides in Walvis Bay			
	Absorbed Dose rate (nGyh ⁻¹)	Annual Effective Equivalent dose (mSv yr ⁻¹)	Radium(Ra _{eq}) Equivalent (Bq kg ⁻¹)	Radiation External Hazard (Hex)
WB1	56.84	0.07	126.69	0.34
WB2	44.89	0.06	99.23	0.27
WB7	32.15	0.04	69.93	0.19
WB8	28.90	0.04	62.14	0.17
WB10	51.34	0.06	112.09	0.30
WB13	50.28	0.06	111.19	0.30
WB17	40.91	0.05	90.91	0.25
WB20	34.86	0.04	75.81	0.20
WB22	33.73	0.04	73.96	0.20
WB23	48.14	0.06	104.48	0.28
Range	(28.90-56.84)	(0.04-0.07)	(62.14-126.69)	(0.17-0.34)
Mean	42.20	0.05	92.64	0.25

As observed from Table 6.6, the results for absorbed dose rate due to naturally occurring radionuclides; ²³⁸U, ²³²Th and ⁴⁰K based on their activities in the soil at 1 m above the ground varies from 28.90 to 56.84 nGyh⁻¹, with mean gamma dose rate 42.20 nGyh⁻¹. The range and the average terrestrial absorbed dose are within the values in UNSCEAR report 2000, which is 10 to 200nGyh⁻¹ and 60nGyh⁻¹ respectively (Taskin et al., 2009). The level of gamma radiation is directly associated with the activity concentrations of radionuclides in the samples and the cosmic rays.

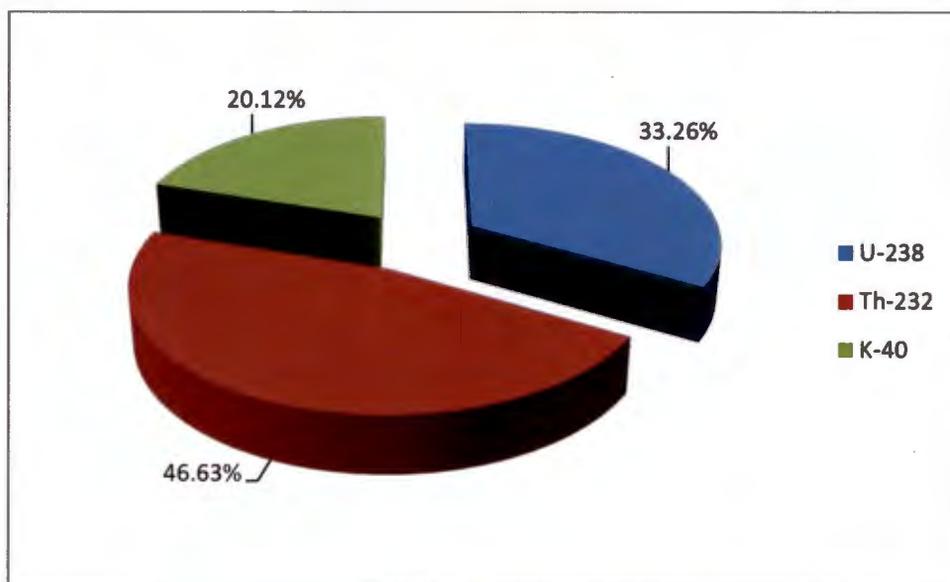


Figure 6.1: The contribution of different radionuclides to the absorbed dose rate.

Figure 6.1 shows that ^{232}Th is the main contributor to the absorbed dose rate in the measured soil samples. This can be attributed to its natural abundance in the monazite sand which is common in continental crust. The relative average contributions of ^{232}Th , ^{238}U and ^{40}K to the total absorbed dose across all the soil samples of Walvis bay are 46.63%, 33.2% and 20.12% respectively. The absorbed dose rate itself does not give a direct indication of the possible biological effects caused by radiation and hence the degree of biological risk caused by different types of radiation (Jibiri et al, 2007). Another term, known as the tissue weighing factor is introduced which is assigned as a unit for gamma radiation. The resulting quantity is the annual equivalent dose rate measured in sieverts per year (Svyr^{-1}).

Using equation 5.2, the calculated annual outdoor effective dose equivalent due to activity of natural radionuclides in all ten geographical locations of Walvis bay is displayed in Figure 6.3 and was found to vary from 0.04 mSvyr^{-1} to 0.07 mSvyr^{-1} with an average of 0.05 mSvyr^{-1} . Similarly, for indoor exposure, using the occupancy factor of 0.8 recommended by UNSCEAR 2000 report, the average annual indoor effective dose equivalent was 0.21 mSvyr^{-1} . This means that the general population of people living in Walvis Bay has the burden of total annual average effective dose equivalent from both indoor and outdoor exposure of 0.35 mSvyr^{-1} , which is less than the average external gamma dose of 0.46 mSvyr^{-1} from natural radiation sources of terrestrial origin (Yang et al., 2005). The results also shows that the mean value is below the maximum permissible dose of 1 mSvyr^{-1} recommended for the public by the International Commission on Radiological Protection, (UNSCEAR, 2000).

The radiation hazard parameters in terms of radium equivalent (Ra_{eq}) and the external hazard index (H_{ex}) were also evaluated. The radium equivalent (Ra_{eq}) was calculated from equation 5.3. The results shows that the maximum values for radium equivalent (Ra_{eq}) was $126.69 \text{ Bq kg}^{-1}$ in site WB 1 and the minimum of 62.14 Bq kg^{-1} in site WB 8. The results shows that the values reported in this work are within the permissible limit recommended by ICRP which is 370 Bq kg^{-1} (UNSCEAR, 2000). In this present study the low concentration of Ra_{eq} value may be related to the transportation of radioactive materials by weathering, sedimentation and water runoff due to desert flash rains (Murugesan et al, 2011). The external radiation hazard index (H_{ex}) was calculated from equation 5.4. The maximum external radiation hazard index is 0.34 which is lower than the maximum value of a unity. The value of a unity corresponds to the maximum radium activity of 370 Bq kg^{-1} for all terrestrial material. The external radiation hazard should be below unity (Al-Hamarneh and Awadallah, 2009). The results of the radiological indices indicate that the area does not have high background radiation and the concentrations of radionuclides are of no radiological importance. Nevertheless regular monitoring is necessary to avoid cumulative effects on the general public.

6.3.2 Radiological indexes in Swakopmund

The calculated radiological risk due to the presence of natural radionuclides in soil in the coastal town of Swakopmund are shown in table 6.7

Table 6.7 Radiological indexes in Swakopmund

Sample ID	Radiological indexes due to natural radionuclides in Swakopmund			
	Absorbed Dose rate (nGyh ⁻¹)	Annual Effective Equivalent dose (mSv yr ⁻¹)	Radium (Ra _{eq}) Equivalent (Bq kg ⁻¹)	Radiation External Hazard (Hex)
SW1	183.28	0.22	318.61	0.86
SW4	92.37	0.11	202.21	0.55
SW6	142.24	0.17	316.68	0.86
SW7	144.19	0.18	204.03	0.55
SW8	80.55	0.10	172.33	0.47
SW9	112.43	0.14	244.34	0.66
SW12	143.64	0.18	313.03	0.84
SW15	140.22	0.17	311.82	0.84
SW16	189.46	0.23	332.66	0.90
SW20	182.50	0.22	305.99	0.83
Range	(92.37-189.46)	(0.11-0.23)	(172.33-332.16)	(0.55-0.90)
Mean	141.09	0.17	272.17	0.74

As observed in Table 6.7, the absorbed dose rate due to terrestrial gamma rays at 1 m above ground were in the range 92.37 to 189.46 nGyh⁻¹ and the mean was 141 nGyh⁻¹ which is higher than the world value of 55 nGyh⁻¹ in the soil samples (Abdil et al., 2006). The outdoor annual effective dose rate equivalent was calculated from absorbed dose using the equation given 5.2. The values varies from 0.11 to 0.23 mSvyr⁻¹. The average outdoor annual dose equivalent was found to be 0.17 mSvyr⁻¹. The value is less than the average values for external gamma outdoor annual effective dose of 0.46 mSvyr⁻¹ from natural radiation sources of terrestrial origin (Yang et al., 2005). The radium equivalent activity in soil were found to vary from 172.33 to 332.16 Bq kg⁻¹ and the mean was 272.17 Bq kg⁻¹ which were lower than the average value of 370 Bq kg⁻¹ reported by OECD (UNSCEAR, 1988).

The calculated values for external hazard index in soil samples varied from 0.55 to 0.90 which is lower than the world permissible limit of 1. The value of this index must be less than a unity for the radiation hazard to be negligible. This implies, the soil in Swakopmund town can be used for construction purpose as it do not pose and any radiological risk. The basic approach to radiation protection is consistence all over the world. The ICRP recommends that any exposure above the natural background radiation should be kept as low as reasonably achievable (ALARA) for which members of the public is 1 mSvyr^{-1} . The dose limit has been established on a prudent approach by assuming that there is no threshold dose below which there will be no effect.

6.3.3 Comparison of radiological indexes in Walvis Bay and Swakopmund towns.

The radiological hazard indexes in Walvis Bay are in agreement with the world values. However, the radiological indexes for Swakopmund are three times higher than the average world values although there are within the permissible limits. The high radiological indices values in Swakopmund can be attributed to the close proximity of the town to Rossing uranium mine which share the same geological underlying rock. Accordingly, it can be assumed that Swakopmund town can receive radiation plumes through sky shine effect and the radionuclides can easily be dispersed into the air and make its way to the coastal town leading to an increase in radiological indexes.

7. CONCLUSION AND RECOMMENDATIONS

7.1 Conclusion and recommendations

The research focused on two coastal towns of Walvis bay and Swakopmund which are 60 km and 40 km from most of uranium mines respectively. The activity concentrations of ^{238}U , ^{232}Th and ^{40}K in different soil samples as well as radiological doses and risks were established. The activity concentrations for which the members of the public could be exposed were quantified through dose calculations.

7.1.1 Activity concentration of ^{238}U , ^{232}Th and ^{40}K in surface soils.

One of objective of this research project was to determine the activity concentrations of ^{238}U , ^{232}Th and ^{40}K in soil samples collected from the two coastal towns of Walvis bay and Swakopmund in the vicinity of uranium mine sites of Erongo region using gamma ray based spectrometer system. The objective was achieved and the activity concentrations of different soil samples collected across the two towns varied from 14.94 ± 03.81 to 155.80 ± 33.71 Bq kg^{-1} for ^{238}U , 17.68 ± 04.62 to 131.58 ± 19.26 Bq kg^{-1} for ^{232}Th and 162.58 ± 53.55 to 761 ± 151.38 Bq kg^{-1} for ^{40}K . The results in this study are in agreement with previous studies in the same locale and some parts of Africa and the world which are 11 to 64 for ^{238}U , 17 to 60 for ^{232}Th and 140 to 850 Bq kg^{-1} for ^{40}K (UNSCEAR, 2000). The samples satisfy the universal standards of limiting the radioactivity to within safe limits of 1000, 1000 and 4000 Bq kg^{-1} for ^{238}U , ^{232}Th and ^{40}K respectively (Gupta et al, 2010).

The samples were hermetically sealed in 1 litre Marinelli beakers for 21 days to allow secular equilibrium to be established between parent and daughter. The activity ratio between parent and daughter was calculated and found to be approximately one which affirm that a state of secular equilibrium was achieved in all samples.

7.1.2 Elemental concentration of ^{238}U , ^{232}Th and ^{40}K in collected surface soils.

The second objective was determining the elemental concentration of radionuclides in soil sample due ^{238}U , ^{232}Th and ^{40}K through calculations. The activity concentrations of the soil samples were converted to elemental concentration using factors recommended by IAEA technical document 1363 as follows;

$$1\% \text{ K} = 313 \text{ Bq kg}^{-1} \text{ of } ^{40}\text{K}$$

$$1 \text{ ppm U} = 12.35 \text{ Bq kg}^{-1} \text{ of } ^{238}\text{U}$$

$$1 \text{ ppm Th} = 4.06 \text{ Bq kg}^{-1} \text{ of } ^{232}\text{Th}$$

For clarity, the units for ppm were considered as mg kg^{-1} since ppm are equivalent to mg kg^{-1} and this facilitate easy comparison with other values obtained from other environment. It should be noted that the elemental concentration of soil samples from the town of Swakopmund was found to be $8.06 \pm 1.57 \text{ mg kg}^{-1}$, $22.39 \pm 3.69 \text{ mg kg}^{-1}$ and $1.77 \pm 0.50 \%$ for ^{238}U , ^{232}Th and ^{40}K respectively. These values are three to four times higher than the world average values and are only comparable to the values obtained in Amman/Jordan which are 6.80 mg kg^{-1} for ^{238}U , 20.2 mg kg^{-1} for ^{232}Th and 1.82% for ^{40}K (Al-Jundi et al., 2003). Determination of correlation between radionuclides in all soil samples across the two coastal towns also shows that the correlation was poor which is an indicative of a heterogeneous geological underlying rock type. The mean value for the thorium/uranium ratio in the soils analysed was 3.26, which is slightly lower than the theoretically expected for normal continental crust which ranges from 3.7 to 4.0 (Van Schmus, 1995). The close association of ratios between thorium to uranium can be used to estimate pedogenic time frame of soil development (Schumeister et al, 1993), because their equilibrium is often disturbed by physical and chemical process (Magill and Galy, 2004). The value of the ratio between potassium and uranium for the analysed soil samples was 3.78 which are extremely lower than the normal continental crust which is estimated to be 1.0 to 1.3×10^4 (Van Schumus, 1995). It should be noted that another school of thought confirmed that the ratio is highly variable and is dependent on the sources of the soil (Roger and Adams, 1969).

7.1.3 Radiological Hazard

The third objective was to estimate the contribution of ^{238}U , ^{232}Th and ^{40}K to radiological risk to the general public and the environment through dose calculations. The objective was achieved and was considered in terms of absorbed dose rate (D), the annual effective dose Equivalent (AEDE), the radium equivalent activity (R_{aeq}) and the external hazard index (H_{ex}).

The average values for absorbed dose rate (D), the annual effective dose Equivalent (AEDE), the radium equivalent activity (Ra_{eq}) and the external hazard index (H_{ex}) in Swakopmund was found to be 141.09 nGyh^{-1} , 0.17 mSvyr^{-1} , $272.17 \text{ Bq kg}^{-1}$ and 0.74 respectively. The radium equivalent was found to be less than the recommended maximum value of 370 Bq kg^{-1} , and the external indexes had value less than a unity, which falls below the equivalent maximum allowed dose for the general public of 1 mSv per year recommended by ICRP. In light of the above reasons, it can be concluded that the soils from the two coastal towns do not pose any significant radiological hazard for people working and living in Erongo region.

7.2 Summary

Soil samples collected from the two coastal towns were analysed by using HPGe detector based gamma ray spectroscopy system. Measurement of natural radioactivity in the soil is very important to determine the amount of change in natural background with time as a result of any radioactive release. Monitoring of any release radioactive contamination in the soil is important for environmental protection. The radiological concentration consequence of natural radioactivity in soil is based on the effects of gamma ray on the human body. The measurements shows the values of absorbed dose rates in the air at 1 m above the ground, the radium equivalent and the external hazard index in the investigated area are lower than the equivalent dose limit of 1 mSvyr^{-1} recommended limit by the United Nation Scientific Committee on the effect of Atomic Radiation (UNSCEAR, 2000).

The detector utilised in this investigation was appropriate for the given radionuclide and the different energies at contamination for ^{238}U , ^{232}Th and ^{40}K . The contamination can include radionuclide which emit radiation with higher energy than in house standard of ^{152}Eu / ^{133}Ba , this can happen with different radionuclides and the detector is also efficient for higher energy but not as efficient as compared to the lower energy. The detector has a good audio signal.

7.3 Recommendations

The following suggestions are provided to reduce the health effects due to radioactive soil contamination.

The research community

- Due to time constraints, the determination of activity concentrations for each soil sample was measured only once. In essence, the measurements should be repeated in order to obtain good statistical precision.
- More detailed studies on the evaluation of public exposure from soil contamination due to uranium mining and exploration activities in Erongo region of Namibia. The research can be extended to determine radioactivity in drinking water and vegetables including the transfer factor in soil to plants and vis versa.

Policy makers

The general public should be sensitised about radioactivity exposure due to uranium soil contamination and mitigation measures should also be in place to cater for accidental release of radioactive soil contamination.

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