PUBLIC EXPOSURE TO HAZARDS ASSOCIATED WITH NATURALLY OCCURRING RADIOACTIVE MATERIALS IN MINING: DOSE ASSESSMENT AND CANCER RISK ESTIMATION

BY

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A THESIS SUBMITTED TO THE SCHOOL OF GRADUATE STUDIES IN PARTIAL FULFILMENT OF THE REQUIREMENT FOR THE AWARD OF DOCTOR OF PHILOSOPHY IN ENVIRONMENTAL SCIENCE

INSTITUTE OF ENVIONMENTAL AND SANITATION STUDIES

JUNE 2015

DECLARATION

I declare that this thesis is the result of research work undertaken by Awudu, Abdel Razak of the Environmental Science Programme, University of Ghana, under the supervision of Prof. E. O. Darko and Prof. G. Emi-Reynolds of Radiation Protection Institute and School of Nuclear and Allied Sciences, Ghana Atomic Energy Commission.

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Dedications

This work is dedicated to the Almighty Allah who has seen me through a successful completion of my studies. I also dedicate it with love and sincere gratitude to my dear wife, Hajia Hamidu Labujata, my entire family, friends and well-wishers for their support, encouragement, love and care.

ACKNOWLEDGEMENTS

The successful completion of this work could have not been achieved without the assistance of the following people, your time and efforts are highly appreciated.

- Prof. E. O. Darko and Prof. G. Emi-Reynolds for their supervision and guidance.
- Radiation Protection Institute of Ghana Atomic Energy Commission for giving me the opportunity to do my study at the Radiation and Waste Safety Department and for the financial assistance.
- Mr. George Owusu Ansah, Mr. Eric Bandoh, the staff of the Environmental section and management of Adamus Gold Mine**.**
- The Director and staff of the Radiation Protection Institute for their assistance and encouragement.
- My family and friends for their moral support and motivation.
- The almighty God for giving me everything I needed to complete my study, strength, courage, hope, perseverance, understanding and love.

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ABSTRACT

Human beings are always exposed to background radiation that comes both from natural and man-made sources. Natural radioactivity is widespread in the earth environment and it exists in various geological formations such as earth's crust, rocks, soils, plants, water and air. Mining has been identified as one of the potential sources of exposure to naturally occurring radioactive materials (NORM). However, mining activities are not being monitored and regulated for NORM in Ghana. Most of the NORM industries such as mining and mineral processing are located in developing countries such as Ghana. Adamus Goldmine is one of the gold mining companies in Ghana and has been in operation since 2007 with no data on radioactivity levels. The mine currently undertakes only surface mining and the process produces large volumes of tailings and waste that may contain NORM. Some of the NORM are soluble in water and can leach into water bodies and farm lands. Mining activities lead to pre-concentration and accumulation of NORM in stockpiles, waste piles (tailings), water bodies and finally leading to exposure of humans through the food chain, as a result it is important to investigate and provide scientific data for the assessment of exposure of the general public. The objective of this study was to evaluate the contents of radionuclides ^{238}U, ^{232}Th and ^{40}K and outdoor terrestrial gamma dose rates in the Adamus Gold mine. To assess the radiological hazard of soils, the radiological hazard indices such as absorbed dose rate, annual effective dose equivalent (outdoor), hazard indices (H_{ex} and H_{in}), activity utilization index ($I_{ɣ}$) and estimated excess life time cancer risk (ELCR) are calculated. Gamma spectrometry technique was used to analyse for Uranium, Thorium and K-40 in soil, water and food crop samples from the mining environment. A total of one hundred and twelve (112) samples from thirty-one (31) sites were analysed. The absorbed dose rates, the corresponding annual effective dose and radiological hazard due to ^{226}Ra, ^{232}Th and ^{40}K in the soil materials that might possibly be used as building materials were carried out. The mean ambient gamma dose rate from direct

measurement and calculation were 55.90 nGy h^{-1} and 46.75 nGy h^{-1}, respectively. The mean activity concentrations measured for ^{238}U, ^{232}Th and ^{40}K in the soil samples were 8.09 Bq kg^{-1}, 45.16 Bq kg^{-1} and 397.77 Bq kg^{-1}, respectively. The mean activity concentrations measured for ^{238}U, ^{232}Th and ^{40}K in the mine tailings were 7.41 Bq kg^{-1}, 33.35 Bq kg^{-1} and 370.97 Bq kg^{-1}, respectively. For the water samples (ground and surface), the mean activity concentrations were 0.15 Bq l^{-1}, 0.75 Bq l^{-1} and 0.71 Bq l^{-1} for ^{238}U, ^{232}Th and ^{40}K, respectively. For the mine processed water samples, the mean activity concentrations were 0.17 Bq l^{-1}, 1.45 Bq l^{-1} and 0.86 Bq l^{-1} for ^{238}U, ^{232}Th and ^{40}K, respectively. The mean activity concentrations of ^{226}Ra, ^{228}Th and ^{40}K in the food crop samples were 5.90 Bq kg^{-1}, 12.20 Bq kg^{-1} and 279.58 Bq kg^{-1}, respectively. The food crop samples have higher estimated lifetime fatality cancer risk (ELCR) value. These values are comparable to concentrations reported for other countries. The total annual effective dose to the public was estimated to be 0.46 mSv. The results in this study compared well with typical world average values. The results indicate an insignificant exposure of the public to technologically enhanced NORMS from the activities of the Goldmine. The radiological hazard indices obtained in this study for the materials considered for use as construction materials for dwellings by the inhabitants are below the internationally recommended values. The results also show that the background radiation levels are within the natural limits and compared well with similar studies for other countries.

CHAPTER ONE

1.0 INTRODUCTION

Human beings are exposed to radiation from various sources that depend upon their activities and surroundings. Not all of the population in the world is exposed to all sources of radiation exposure. For example, workers who work in the nuclear industry or the patients cured by medical irradiation will receive radiation exposure rather than members of public. However, one of the most obvious sources which everybody is continuing and inescapably exposed is natural ionising radiation coming from the natural radionuclides in the environment, and this is also the major source of radiation exposure to humans (UNSCEAR, 2000). This exposure is in most cases of little or no concern to society, but in certain situations the introduction of health protection measures needs to be considered. Natural radiation in the environment arises mainly from Naturally Occurring Radioactive Material (NORM) which can be found both at the earth's surface and in the subsurface.

1.1 Naturally Occurring Radioactive Materials (NORM)

Naturally occurring radioactive materials (NORM) contribute about 70% of the global population exposure to ionizing radiations (UNSCEAR, 1988). NORM have been prominent features of the human environment since the creation of the earth and may be present in elevated levels in some regions or areas giving rise to potential exposure situations (Kathren, 1998; Jacob and Paretzke, 1981; Gessell and Pritchard, 1975). High levels of exposure to radioactive materials are known to cause clinical damage to the tissues of the human body. Potential pathways of exposure from these nuclides include external irradiation to gamma rays, internal exposure due to inhaled radon, its decay products and inhalation of contaminated dust and ingestion of contaminated food and/or water.

According to United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) report, natural radioactivity constitutes the largest source of population dose (UNSCEAR, 2000). The major sources of external gamma radiation exposure are ^{238}U, ^{232}Th and their decay products and ^{40}K. Also, ^{238}U and its daughters rather than ^{226}Ra and its daughter products are responsible for the major fraction of the internal dose received by humans from naturally occurring radionuclides (de Oliveira *et al*, 2001). Even though the concentrations of these radionuclides are widely distributed in nature, they have been found to depend on the geology of the local area and as a result vary from place to place (Xinwei *et al*, 2006). The specific levels are related to the type of rock from which the soil originates. Higher radioactivity levels are associated with igneous rocks such as granite and lower levels with sedimentary rocks. There are exceptions, however, as some shale and phosphate rocks have relatively high content of radionuclides (Uosif, 2007). Higher concentrations of radionuclides may also arise as a result of human activities such as mining and mineral processing (UNSCEAR, 2000; IAEA, 2005).

Natural radiation in the environment arises mainly from NORM, originating mainly from the Uranium-Thorium series and Potassium. The most important radionuclides in terms of radiation exposure are uranium-238 (^{238}U) and Thorium-232 (^{232}Th) and their decay products as well as Potassium-40 (^{40}K). NORM can be found both on the earth's surface and in the subsurface. NORM exists everywhere in the environment not only in soils, water, and the atmosphere but also in humans, and animals and from what they consume (Wilson, 1994). The origins of natural radioactivity can be classified into two main categories, the first coming from radioactive elements in the earth's crust (e.g., ^{238}U and ^{232}Th series) and the other caused by radiation from extraterrestrial sources (e.g., ^{3}H ^{14}C). A number of naturally occurring radionuclides associated with the formation of the earth are primordial radionuclides which are components of uranium and thorium radioactive series and potassium non-series radionuclides. The other natural radioactive source is cosmic radiation that originates from outer space. The primary particles of cosmic radiation have sufficiently high energies that can generate secondary particles in the upper atmosphere and then penetrate the earth's atmosphere to ground level. Additionally, some radionuclides are continuously produced by interaction of incoming cosmic rays with stable nuclides in the atmosphere, or in the earth (NCRP, 1975).

Besides naturally occurring radionuclides, the exposure of humans to radiation in the environment has resulted from other possible sources such as nuclear weapons testing in the atmosphere (e.g., ^{137}Cs), disposal of radioactive wastes from industrial and nuclear sites into the environment (e.g., ^{241}Am), or from major accidents of nuclear installations (e.g., Chernobyl). These sources remain at a very low level of exposures to members of the public (Watson *et al*, 2005).

Naturally occurring radioactive isotopes are found in rocks and soils that can be taken up by plants, and enter the food chain which can give rise to an internal radiation exposure in human beings, when man consumes these radionuclides in the foods or inhale from the air. The presence of radionuclides in rocks, soils, and building material derived from them can also produce an external radiation field to which all human beings are exposed (Watson *et al*, 2005).

Mining of all kinds affects the environment negatively. The existence of natural radioactivity in ores and in their processed wastes (liquid or solid) has long been recognised. Radiological aspects of the mining industry have been addressed in many countries and a good part of the results have been gathered into reports by international organizations such as UNSCEAR (UNSCEAR, 1982; UNSCEAR, 1993). The radiation environment in mines is complex and variable. In the area of mining, the specific concerns are occupational radiation exposure of miners and populations living in the vicinity of the mining area. Miners are exposed to airborne radon (^{222}Rn), thoron (^{220}Rn) and their short-lived decay products, long-lived radionuclides in ore dust, and external gamma and beta radiation (ICRP, 1986). All of these types of exposure warrant consideration in formulating control procedures. The radiological hazards are significant, particularly with gamma dose rate and air borne alpha-emitters. The processed solid wastes are usually disposed of on open land, where they undergo the natural processes of weathering. The impact of such disposal methods has been examined to evaluate their significance from the point of view of radiation exposure. The radiation exposure of the population living in the neighbourhood of mining activities may result from: (i) leached activity directly ingested through drinking water or indirectly through the food chain by uptake through vegetation, fish, milk and meat, (ii) an enhanced external radiation background in the area, and (iii) higher radon levels due to ground emanation. For decades, many countries have put a great deal of effort into quantifying the radiological impacts of technologically enhanced radioactivity on man and the environment (Baxter, 1996; Hedvall and Erlandsson, 1996). Some countries such as Denmark, Finland, Iceland, Norway and Sweden have taken steps to apply similar radiation procedures and control practices to non-nuclear industries as already applied in the nuclear industry (EC, 1997).

Ghana is endowed with abundant mineral resources typified by the extensive gold deposits especially in the Ashanti and Western regions, where as a result, mining activities are wide spread. There are over two thousand known minerals spread all over the country, but only about one hundred of them are common (Kesse, 1985). The most important minerals in Ghana include gold, diamond, bauxite and manganese. Some anomalies of uranium, limestone, zirconium and phosphate, among others, have also been identified. Gold contributes more than 45% of Ghana's export earnings.

In Ghana there is a general lack of information regarding doses from the non-nuclear industries. The purpose of this study is, therefore, to: (i) assess exposure to hazards associated with naturally occurring radioactive materials in mining as a result of gold mining activities of Adamus Gold Limited in Ellembele near Tarkwa in the Western Region of Ghana and surrounding communities, (ii) assess the doses and estimate the cancer risks to members of the public who are exposed to enhanced levels of naturally occurring radionuclides and (iii) contribute towards identifying naturally occurring radioactive materials (NORM) in the country.

1.2 Statement of the problem

Enhanced levels of naturally occurring radionuclides may be associated with certain natural materials, minerals and other resources. Exploitation of these resources and production of consumer items may lead to further enhancement of the radioactivity in the products, by-products, residues or waste arising from the industrial process. A potential outcome is an increase in occupational and public exposures to radiation (UNSCEAR, 2000; de Oliveira *et al*, 2001; Xinwei *et al*, 2006).

Mining has been identified as one of the potential sources of exposure to NORM. The majority of NORM issues centre on waste from industrial processes. Most of the wastes are produced in very large volumes, but are of low activity. While some wastes are disposed of, others are put to commercial uses (USEPA, 1993). The improper disposal, re-use, and recycling of NORM has led to circumstances resulting in contamination events and unnecessary public exposures. Mining activities lead to pre-concentration and accumulation of NORM in stockpiles, waste piles (tailings), pipes, water bodies and finally leading to exposure of humans through the food chain, As a result it is important to investigate and provide sound scientific data for the assessment of exposure of individual workers and the general public. Mining and milling of ores with significant amounts of uranium and thorium minerals may lead to radiological environmental impact of nearby water bodies, soils and aquifers (USEPA, 1993).

Disposal in piles or stacks can also lead to groundwater contamination and to airborne releases of radioactive particulates and radon (USEPA, 1993). Improper use and/or disposal of waste, such as for soil conditioning or fill around homes, can lead to build-up of radon gas in homes, direct exposure to individuals, and contamination of soil and of the crops growing in the soil. Re-use of NORM-contaminated materials, such as in concrete aggregate, can lead to increased radiation risks to members of the public in a variety of ways (USEPA, 1993).

These impacts may be observed during the operational phase of a mine as well as after a mine closes down, if proper remediation is not carried out. During the life of a mining operation and after a mine closes down, the tailings and waste dumps that have been formed are consequently subjected to weathering. The rate of leaching of pollutants from solid materials will change with time and be dependent on several factors that will regulate the rate of pollutants release from these sources (UNSCEAR, 2000).

In Ghana, as at 2008, there were over two hundred (200) registered companies operating small, medium and large scale mining and are wide spread over the entire country (Aryee and Aboagye, 2008). It is a fact that these industries, despite the potential radiological impacts that they may cause, are not licensed as practices, nor controlled by the Regulatory Authority. However, it has been reported in many publications (Fernandes *et al*, 2002) that undesirable NORM exposures of members of the public and workers may result from these operations. In addition to this, an overall understanding of the amount of radionuclides disposed of with the wastes, uranium, thorium and potassium concentrations in by-products that could be used in a wide variety of situations remain unknown.

Estimation of cancer risks to the public associated with NORM resulting from mining and mineral processing has not been evaluated in most mines in Ghana. Data on radionuclide concentrations in raw materials, mine tailings, scales in pipes, waste streams and public exposures in Ghana are very scanty (Darko *et al*, 2005; Darko and Faanu, 2007; Andam and Dodoo-Amoo, 1994) and the assessment of cancer risk to workers and members of the public that are exposed to these enhanced NORM is non-existent. Consequently, there is general lack of awareness and knowledge of environmental radiation pollution and cancer risk associated with enhanced NORM by operators, regulators and legislators in Ghana.

1.3 Motivation of study

The motivations of the research in the environmental radiation pollution are to assess the radiation risks to humans. This was done using a gamma spectrometry system in the following sequence:

- Radioactivity in soil, water and food crops.
- Radioactivity in mine wastes.
- Radioactivity in humans (the Public).
- Assessment of effective doses.
- Cancer risks assessment.

The study is on humans, who are constantly exposed to radioactive environment. The mining and milling of ores with significant amounts of uranium and thorium associated with the main ore have the potential to pose undue health risks to members of the public and workers. The health risks associated with ionizing radiation is due to the fact that it is very harmful as it can change the chemical make-up of many things, including the delicate chemistry of the human body and other living organisms (UNSCEAR, 2000) (UNSCEAR, 2000). This study is to assess exposure to hazards associated with naturally occurring radioactive materials in mining, assess the doses and estimate the cancer risk to the public using Nkroful Gold Mine operated by Adamus Resources Limited in Ellembele District near Tarkwa as a case study.

1.4 Objective of study

The primary objectives of the study are to assess the dose and estimate the cancer risks associated with public exposure to hazards from NORM in gold mining.

The specific objectives of this study are:

- To determine the activity concentrations of the radionuclides U/Th series and ^{40}K.
- To determine the radiation doses from these activity concentrations and compare with internationally recommended dose limits.
- To assess the hazard and estimate cancer risk to the members of the public associated with the radiation dose values.
- Recommend a suitable radiation protection programme to the mine if necessary.
- Recommend the development of a regulatory guidelines based on the established scientific data where appropriate.

1.5 Importance of the research

The research will:

- Contribute to data collection on radionuclide concentrations in soils, mine wastes, water and food crops as well as data on public exposure.
- Contribute to knowledge and awareness of environmental radiation pollution from NORM.
- Assess the doses and estimate radiation hazard and cancer risks to the public.
- Help develop radiation protection programme for public exposure associated with mining and mineral processing if necessary.

1.6 Structure of the thesis

In this thesis, there are five chapters. The first chapter describes the introduction of naturally occurring radioactive materials (NORM). The aims and objective of the work have also been described in this chapter. Chapter 2 includes the literature review on geology of NORM, radioactivity in soil, rocks, water and food crops. Details of radioactivity, sources of radiation, effect of radiation, radioactivity in cells, types of radiation effects, cancer epidemiology and cancer risk have also been covered in this chapter. Chapter 3 describes the materials and methods. It includes the details about the study area regarding location, geology, climate and vegetation. Details of the sampling and sample preparations, qualitative analysis of radioactivity in soil, water and food crops have also been given in this chapter.

Chapter 4 gives details of the natural radioactivity results in soil, water and food crops and their comparison with world averages. The impact of radioactivity has also been given in this chapter, including calculations of absorbed doses in air, effective doses and hazard indices. At the end of this chapter, cancer risk estimation has been done for the members of the public living near the study area. The last chapter (Chapter 5) constitutes the conclusions of the work done and the recommendations.

CHAPTER TWO

2.0 LITERATURE REVIEW

2.1 Radioactivity

Radioactivity is the term used to describe those spontaneous, particle and energy emitting, atomic transitions that involves changes in the state of nuclei of atoms. The energy released in such transformation is emitted in the form of electromagnetic radiations and particles (Turner, 1986). The radiations interact, in varying degrees, with matter, as they traverse through it. All radiations represent a transfer of energy from the emitting source to the surrounding media where they may interact to an extent determined by the characteristics of the radiations and the nature of the structure of the intervening medium. If the interaction between the radiation and the target medium is strong, energy is transferred at a rapid rate and radiation is strongly attenuated. If radiation attenuation is weak, the radiation may have an appreciable range and may be detected at great distance from the source.

2.2 Sources of Radiation

The world population as a whole is exposed to radiation in two ways – internal or external exposure. Whether the source of radiation is naturally occurring: originated from the earth, interstellar space, human body itself; or from man-made sources: medical, industrial applications, consumer products and occupational exposures from nuclear facilities.

2.2.1 Natural Sources

Sources of natural radiation responsible for contributing radiation hazard to mankind are from cosmic rays, terrestrial radiations and radioactivity in the body.

1. Cosmic rays impinge on the earth from outer space. These are responsible for

producing various radionuclides in the environment such as ^{7}Be, ^{14}C, ^{3}H. Doses from cosmic rays are latitude and altitude dependent. The average total dose received from external sources by a person residing at sea level is approximately 0.91 mSv per year. However, a dose twice this size may be received by a person residing at a higher elevation, such as Denver (USA), where cosmic rays are more intense, while at ground level average annual effective dose is 0.4 mSv (UNSCEAR, 2000). The intensity of cosmic rays at various altitudes is shown in Table 2.1

Table 2.1: Radiation Levels at various Altitudes (UNSCEAR, 2000).

Altitude (km)	**Exposure Rate (μSv)**
Sea level	0.03
2	0.01
6.7	1.0
10	5.0
15	10.0

2. Terrestrial radiations are released into the environment by the disintegration of primordial radionuclides potassium (^{40}K), naturally occurring radionuclides of uranium (^{238}U) and Thorium (^{232}Th) series and their progenies in the earth's crust. A person residing in a geographic region, where the radium content of the soil is relatively high, can receive a dose as high as 100 mSv per year due to inhalation of radon and its disintegration products, while annual dose limit set by ICRP for general population is 1 mSv (UNSCEAR, 2000).

3. Radioactivity in the body comes from contaminated environment through inhalation of air, ingestion of food and water. The major contributions to internal exposure are from cosmogenic nuclides (^{3}H, ^{14}C and ^{22}Na), naturally occurring radioactive nuclides (^{40}K, ^{238}U and ^{232}Th) series and fission or activation products (^{131}I, ^{134}Cs, ^{137}Cs, ^{90}Sr and ^{60}Co). Evidence show that ordinary food (e.g. cereals, milk, fruit and vegetables) also contains some radioactivity (UNSCEAR, 2000). Such activities are deposited in various critical organs of the human the body and are responsible for internal radiation hazards. The estimated activity responsible for producing internal exposure to a 70 kg adult, based on International Commission on Radiological Protection, ICRP-30 data (ICRP, 1979) due to inhalation or ingestion of the above mentioned sources is given in Table 2.

Table 2.2: Estimated activity levels of naturally occurring radionuclides in ICRP Reference Man (ICRP, 2007)

Nuclide	**Mass of Nuclide**	**Activity**	**Daily Intake**
Uranium	90 µg	1.1 Bq	1.9 µg
Thorium	30 µg	0.11 Bq	3 µg
Potassium	17 mg	4.4×10^{3} Bq	0.39 mg
Radium	31 pg	1.1 Bq	2.3 pg
Carbon-14	95 µg	15×10^{3} Bq	1.8 µg
Tritium	0.06 pg	23 Bq	0.003 pg
Polonium	0.2 pg	37 Bq	0.6 µg

2.2.2 Artificial Sources

In addition to natural background exposure, people are exposed to radiation from various man-made sources of radioactivity. Some common examples in medical, industrial and nuclear applications are listed below (UNSCEAR, 2000):

1. Medicine: Application of X-rays, radioisotopes and other modern techniques in medical diagnosis is considered to be second largest followed by natural background radiation. Although the doses delivered in different types of X-ray examinations vary from a small fraction of a mGy to tens of mGy, the trend is expected to increase in future due to widespread use of radioisotopes in medicine for diagnostic or therapeutic purposes.
2. Industrial and consumer products: Less significant sources of radiation include radioactive materials in crushed rock, building materials, phosphate fertilizers, computers/television sets, smoke detectors and other consumer products.
3. Nuclear applications: Most of the radioactivity produced in nuclear power reactors is safely contained; however, a small percentage escapes as stack gas or liquid effluent and eventually may contaminate the atmosphere and water supply. There are similar releases from nuclear-fuel reprocessing plants, which contribute to the worldwide background radiation level. There are exposures which originate from nuclear weapons testing and nuclear accidents even though these are seldom these days.

Average radiation exposures from the above mentioned natural and artificial sources are estimated to be ~2.4 mSv per year (UNSCEAR, 2000). However, this value varies depending on the geographical locations by several hundred percent. Distribution of doses from various natural and artificial sources of radiation is shown in figure 1

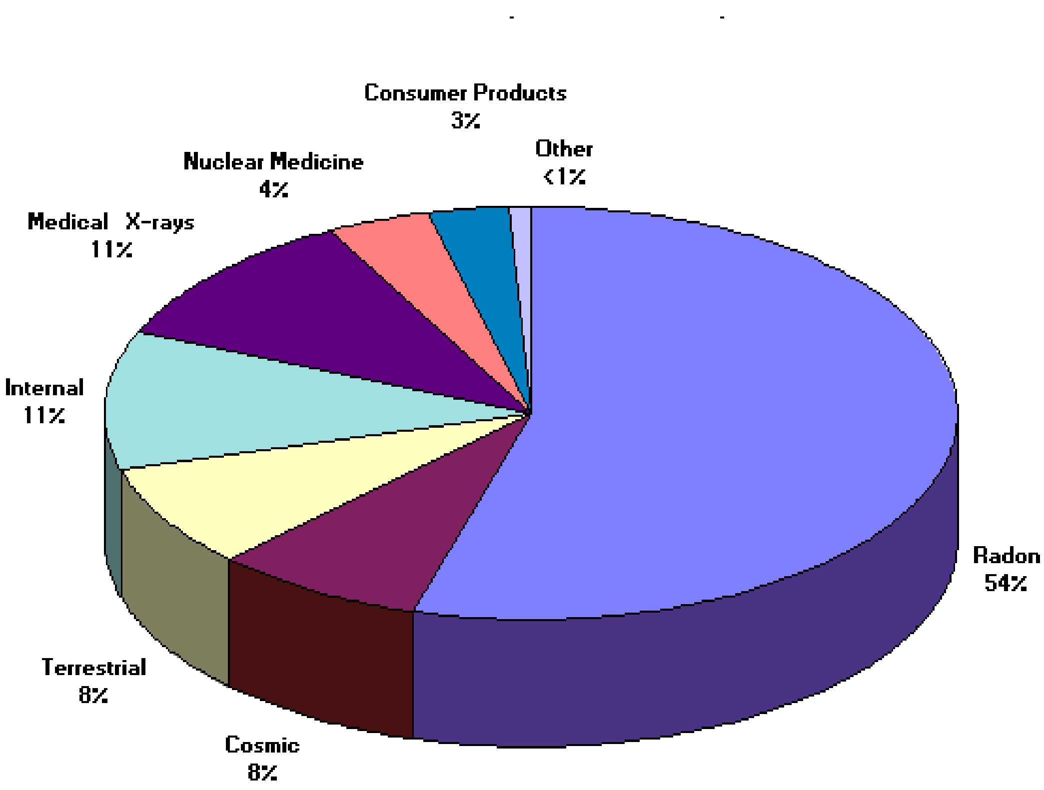


Figure 1: Contributions of radiation sources to annual effective dose (UNSCEAR, 2000).

The pie chart shows that 82% of the total average annual effective dose is from natural sources of radiation, and it is mostly from radon. Majority of the rest of 18% belongs to medical diagnosis and treatments, with less than 1% from nuclear power and fallout (UNSCEAR, 2000).

2.3 Effects of Radiation

Radiation interactions with human organisms produce ionization, which triggers morphological and functional changes, consequently damaging normal behaviour of cells, tissues and organs leading to development of cancer, and increase in mortality rate. Initiation and development of such radiation injury starts from sub-cellular mechanism such as molecules of proteins, carbohydrates, fats, inorganic salts, membrane systems. The breakage of the sub-cellular structure and its constituents provokes a chain of damages, finally leading to manifestation of morphological and functional changes. The damage developed in the cell and tissue leads to possible malfunctioning of the organ or system and ultimately the organ as a whole (UNSCEAR, 2000).

2.3.1 Mechanism of Biological Actions

Ionizing radiation penetrates living matter and gives up energy through random interactions with atoms and molecules in its path, leading to the formation of reactive ions and free radicals, depending on relative biological effectiveness (RBE) value of the impinging radiations. Charged particles have a high RBE and generally cause greater injury for a given total dose to the cell than do X-rays or γ-rays. At the same time, charged particles have less penetrating power in tissue, and pose relatively little hazard to tissues within the body, unless they are emitted by a radionuclide, or radioactive isotope, that has been deposited internally (Khan, 1984).

The molecular alterations resulting from this ionization and, in turn, the resultant biochemical changes give rise to various types of injury. The probability of injury depends on the concentration of molecular damage produced at a critical site, or target in the cell, e.g. a gene or a chromosome.

Most of the human body is made up of water. Therefore, water molecules are a likely target for being hit by photons or charged particles. The reaction, which occurs when this happens, is an ionization to form a positive ion H_2O^+ and electron (Aziz and Mateen, 1994).

Following this ionization (production of H_2O^+ and e^-) a number of other reactions may also take place. Firstly, the ion pair may recombine to stable water molecule. Secondly, if these ions do not recombine then negative (-ve) electrons may be attached to water molecule producing a third type of ion through the following reaction:

$$e^- + H_2O^+ \rightarrow H_2O^- \qquad (2.1)$$

The H_2O^+ and H_2O^- are relatively unstable and dissociate into smaller molecules as follows (Aziz and Mateen, 1994):

$$H_2O^+ \rightarrow H^+ + OH^- \qquad (2.2)$$

$$H_2O^- \rightarrow H^* + OH^- \qquad (2.3)$$

In this process, two ions (H^+ and OH^-) and two free radicals (H^* and OH^-) are produced. These ions can recombine and no biological damage would occur. The free radicals behave in a different manner. A free radical is an uncharged molecule containing a single unpaired electron in the valence or outermost shell. This free radical is very reactive. The OH^* free radicals can join together to form hydrogen peroxide by the following equation, which is poisonous and act as toxic agent:

$$OH^* + OH^* \rightarrow H_2O_2 \qquad (2.4)$$

H^* free radical can interact with molecular oxygen if it is present to form hydroperoxyl radical:

$$H^* + O_2 \rightarrow H_2O^* \qquad (2.5)$$

The hydroperoxyl radical along with hydrogen peroxide is considered to be the principal damaging product following the radiolysis of water. These free radicals can combine with each other and give a variety of potent oxidizing agents such as hydrogen peroxide, superoxide, molecular oxygen and hydroperoxyl radicals (Aziz and Mateen, 1994). Free radicals are highly reactive chemically and can alter important molecules in the cell.

One important molecule is deoxyribonucleic acid (DNA) found mainly in the nucleus of the cell. It is a large molecule and its structure is visible through microscope. DNA controls genetic information, function of the cell and its replication. Structure of a DNA molecule is shown in figure 2 (Genetics Home Reference, 2013).

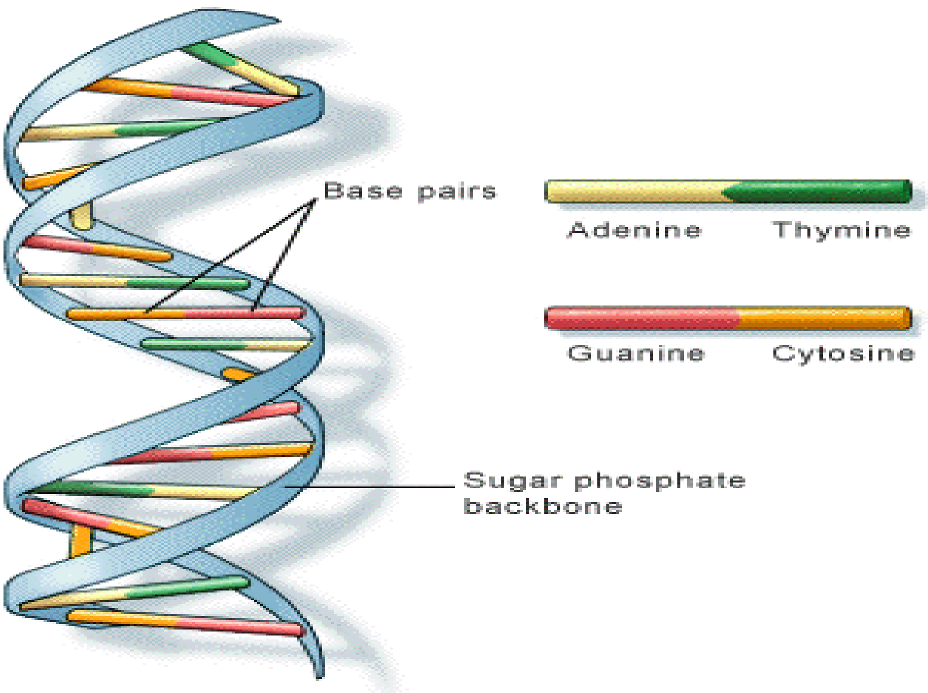


Figure 2: Structure of a DNA molecule (Genetics Home Reference, 2013).

Radiation produces damage in a cell in two ways. It may ionize a deoxyribonucleic acid (DNA) molecule directly through a chemical change or indirectly with the interaction of free hydroxyl radicals. In either case the chemical change can cause a harmful biological effects leading to development of cancers or inherited genetic defects.

2.3.2 Direct Effect

Radiation may cause ionization through a direct hit to DNA or other biological molecules. The extent of this direct effect occurring depends on the number of a particular type of molecule in the cells and its size. The larger a molecule the better target it makes. Since DNA is the largest molecule in the cell as well as the site of all the genetic information, its response has a central role in the mediation of radiation effects (UNSCEAR, 2000). Depending on how it is damaged, different results will occur. If the damage results in a strand break in its backbone (breaking the molecule in half), subsequent mitoses may fail resulting in cellular death. If the break is in one of its side groups (bases), it will then transmit different genetic data during subsequent division resulting in some kind of a mutation. Probability of radiation hit to DNA is shown in figure 3 (UNSCEAR, 2000).

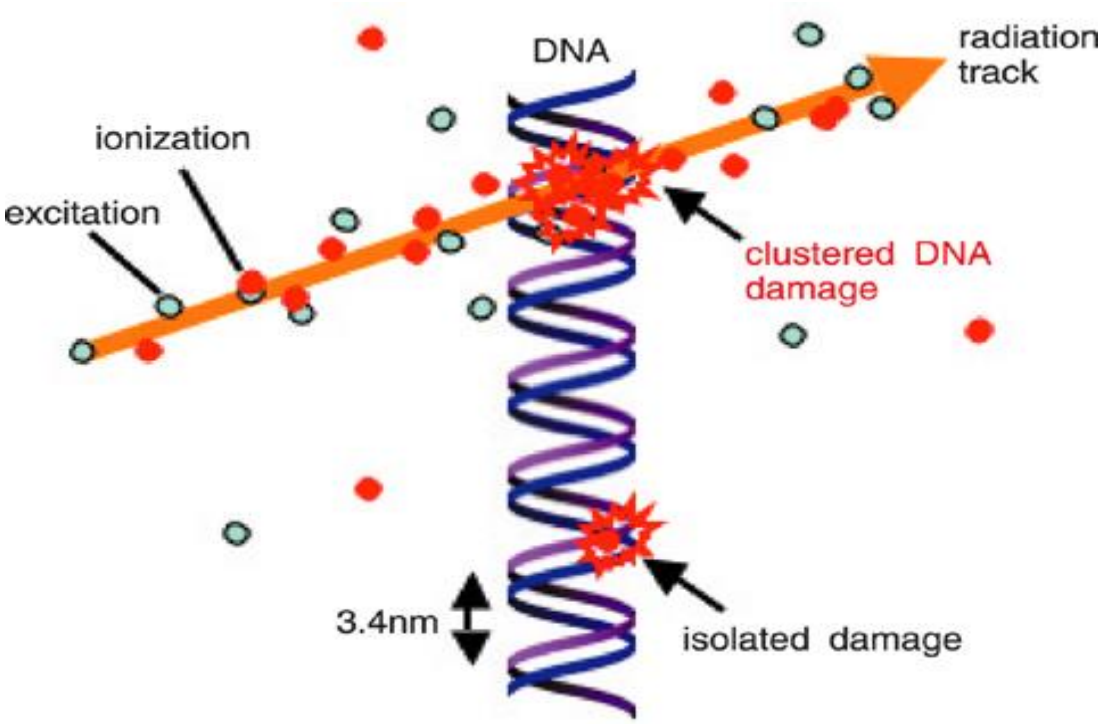


Figure 3: Probability of radiation hit to DNA (UNSCEAR, 2000).

2.3.3 Indirect Effect

Both the initial radicals and these products can migrate to biologically important molecules (like DNA – the structural material of genes) and cause bond breakage and/or oxidation of attached groups. In this way, energy of the radiation is transferred to biologically significant molecules, changing their structure. This mode of energy-transfer is known as the indirect effect and can account for an appreciable fraction of damage (UNSCEAR, 2000).

Both direct and indirect effects contribute to the overall number of such damaging events to the DNA and vary for individual cell types. Direct and Indirect actions of radiation are shown in figure 4.

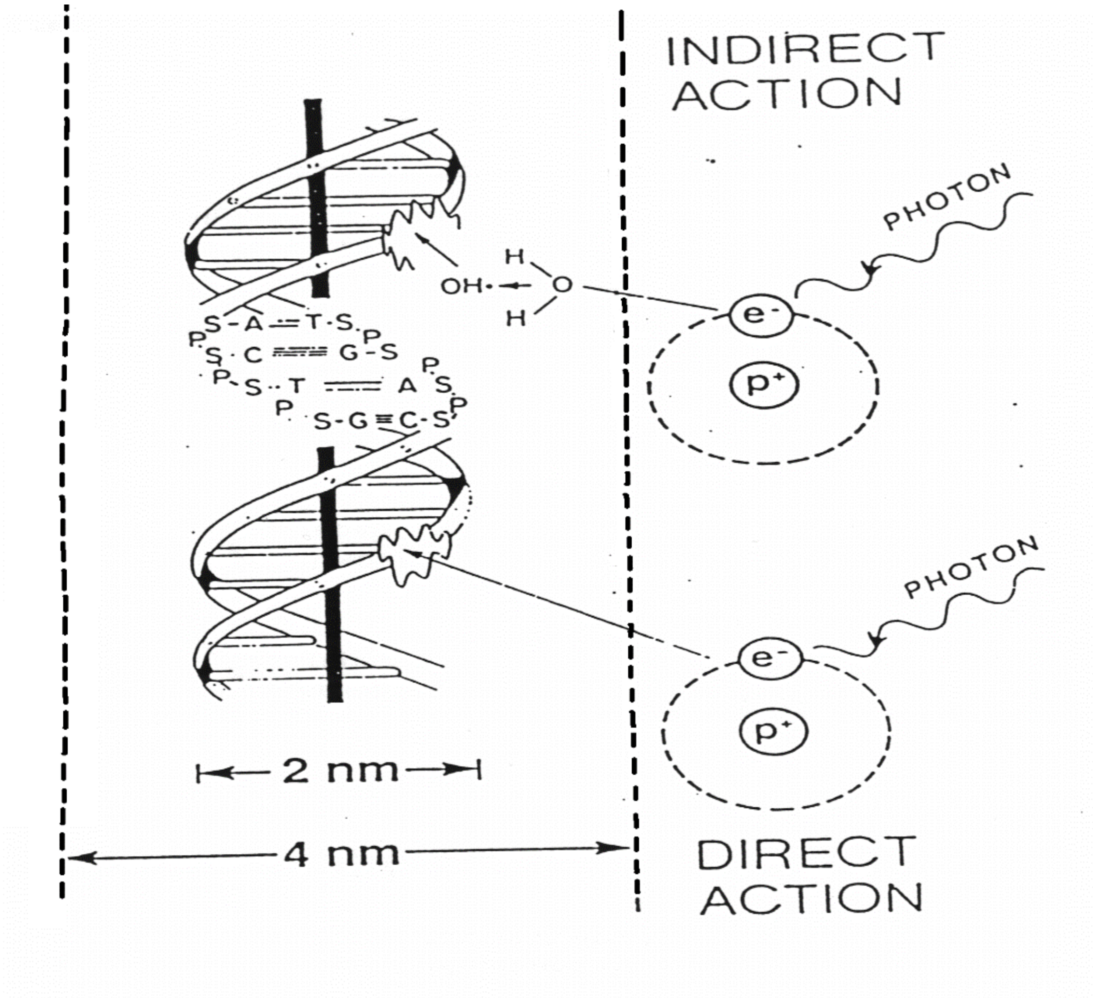


Figure 4: Direct and Indirect actions of radiation (UNSCEAR, 2000).

2.4 Radiosensitivity in Cells

The radiosensitivity of a particular cell is directly proportional to its reproductive activity and inversely proportional to the degree of differentiation. Tissues consisting of rapidly dividing stem cells (like blood or sperm cell precursors) are quite sensitive to radiation whereas cells that do not divide or only rarely divide (like nerve or muscles cells) are considerably more resistant. Other factors involved include metabolic rate, state of nourishment, oxygen level and presence of particular enzymes within the cell. The latter are most likely involved in the repair of some of the radiation damage (UNSCEAR, 2000).

A great diversity exists among the different kinds of cells found in the body. Many have a brief lifespan, undergoing division (a process called mitosis) in a period of hours, while others (such as nerve cells) do not divide at all after birth. Any alteration of the genetic information associated with mitosis can result in either a permanent change in the nature of the cell (mutation), or in the cell's death.

When a cellular component is damaged by agents (chemicals, radiation, excessive heat) a multitude of measurable effects can result. The changes may initially be restricted to a single or a few types of cells. In time, whole organs or organ systems may be affected due to the absence of a required function that upsets the equilibrium or control of the whole interrelated system. Gross physiological or morphological changes may result from an initial damage to a sufficient number of many kinds of cells. The type of cell damage depends on the specific agent the cell is exposed to and the amount of damage is related to how much of the agent reaches that particular kind of cell (UNSCEAR, 2000).

It has been observed from experiments that divided doses of gamma radiation or X-radiation let the body to get itself repaired about 60 percent within several hours from single brief exposure. Therefore, the body is able to tolerate a larger total dose when the dose is accumulated slowly or when part of it is absorbed at a later time.

The cell can repair certain levels of cell damage. At low radiation doses or dose rates (chronic doses) that are received every day from background radiation, cellular damage is rapidly repaired. Whereas at extremely high doses or acute doses cell death results. Cells cannot be replaced quickly enough and tissues fail to function. Acute radiation dose usually refers to a large dose of radiation received in a short period of time. Chronic dose refers to the sum of small doses received repeatedly over long time periods, for example, 0.2 mSv per week every week for several years (Martin and Harbison, 1996).

2.5 Types of Radiation Effects

Radiation interactions with human organisms produce ionization, which triggers morphological and functional changes, consequently damaging normal behaviour of cells, organs and tissues. The damage may result in cells death or modifications. Most organs and tissues of the body are not affected by the loss of even considerable numbers of cells. However, if the number of affected cells becomes large, there will be observable harm to the organ or tissue and therefore, to the individual. These effects usually occur from acute or single large doses of radiation, known as “deterministic effects” (UNSCEAR, 2000).

If the cell is not killed but only modified, the damage in the viable cell is usually repaired. If the repair is not perfect, the modification will be transmitted to daughter cells and may eventually lead to cancer in the tissue or organ of the exposed individual. If the cells are concerned with transmitting genetic information to the descendants of the exposed individual, hereditary disorders may arise. Such effects in the individuals or in their descendants are called “stochastic”, meaning of a random nature (UNSCEAR, 2000).

In short, deterministic (acute) effects will occur only if the radiation dose is substantial, such as in accidents. Stochastic effects (cancer and hereditary effects) may be caused by damage in a single cell. As the dose to the tissue increases from low level, more and more cells are damaged and the probability of stochastic effects occurring increases. It indicates that misrepaired radiation damage gives the potential for progression to cancer induction or hereditary disease and contributes to the multistage development of cancer. These effects are described in subsequent paragraphs (UNSCEAR, 2000).

2.5.1 Deterministic Effects

These effects occur within a few weeks after the receipt of a large or acute dose in a relatively short period of time. The effects are due to major depletion of cell populations in a number of body organs due to cell-killing and the prevention or delay of cell division. The main effects are attributable to bone marrow, gastrointestinal or neuromuscular damage, depending on the dose received (UNSCEAR, 2000).

Acute absorbed doses of 1 Gy or above give rise to nausea and vomiting. This is known as radiation sickness and it occurs a few hours after exposure as a result of damage to cells lining of the intestine. Absorbed doses above 2 Gy can lead to death, probably 10-15 days after exposures (UNSCEAR, 2000).

There is no well-defined threshold dose below which there is no risk of death due to acute doses, although for doses below 1.5 Gy, the risk of early death would be very low. Similarly, there is no well-defined point above which death is certain, but the chances of surviving an acute dose of about 8 Gy would be very low (UNSCEAR, 2000).

A reasonable estimate can be made of the dose which would be lethal for 50% of the exposed subjects within 60 days of exposure. This is called LD_{50}^{60} and is thought to have a value between 3 and 5 Gy for man. For doses up to 10 Gy, death is usually due to secondary infections because of depletion of the white blood cells which normally provide protection against infection. The range of doses from 3 to 10 Gy is often called the region of infection death. In this range the chances of survival can be increased by special medical treatments, which include isolating the subject in a sterile (infection-free) environment and giving bone marrow transfusion to stimulate white blood cell production (UNSCEAR, 2000).

For doses above 10 Gy, survival time drops abruptly to between 3 and 5 days. It remains at about this figure until much higher doses are reached. In this region, the radiation dose causes severe depletion of the cells lining of the intestine. Gross damage occurs in the lining of the intestine, followed by severe bacterial invasion. This is called the region of gastrointestinal death. At much higher doses, survival times become progressively shorter. There are very few human data in this region but from animal experiments, the symptoms indicate some damage to the central nervous system death. However, it is found that death is not instantaneous even in animals irradiated with high doses in excess of 500 Gy (UNSCEAR, 2000).

Another effect, which shows up soon after an acute over-exposure to radiation, is erythema, which is, reddening of the skin. In many situations the skin is subject to more radiation exposure than most other tissues. This is especially true for beta rays and low energy X-rays. A dose of about 3 Gy of low energy X-rays will result in erythema and larger exposures may lead to other symptoms such as changes in pigmentation, blistering and ulceration (UNSCEAR, 2000).

The levels of exposure both of workers and of the public arising from the nuclear energy industry or from industrial and medical applications of radiation are far below the levels, which would induced early effects. Such high doses could only be received in the unlikely event of a major nuclear accident. However, the low doses received in normal operations may cause harmful effects in the long term (UNSCEAR, 2000).

Another radiation effect which is deterministic in nature but which may not occur for many years is damage to the lens of the eye. This takes the form of observable opacities in the lens or in extreme cases, visual impairment as the result of a cataract. Again, there is a threshold dose and so requires setting a dose limit for the lens of the eye. The occurrence of these effects can be prevented (UNSCEAR, 2000).

There is some evidence from animal experiments that exposure to radiation may slightly reduce the life expectancy of individuals who do not exhibit any specific radiation-induced symptoms. Observations of human populations exposed at relatively high levels indicate that if life shortening occurs at all, it is very slight, almost certainly less than 1 year per Sievert (Martin and Harbison, 1996).

2.5.2 Stochastic Effects

These are delayed effects which occur years of receipt of a small or chronic dose in a relatively long period of time. The effects are of two types which are identified through hereditary changes and cancer induction (UNSCEAR, 2000).

2.5.3 Hereditary Changes

The hereditary effects of radiation result from damage to the reproductive cells. This damage takes the form of alterations, known as genetic mutations, in the hereditary material of the cell. Radiation can induce gene mutations, which are indistinguishable from naturally occurring mutations. Since ionizing radiation can cause an increase in the mutation rate, its use will increase the number of genetically abnormal people present in future generations. Clearly, the consequences of excessive genetic damage would be very serious indeed and strict control must be exercised over the radiation exposure of the general population (UNSCEAR, 2000).

2.5.4 Induction of Cancer

Development of cancer is a complex, multistage process that usually takes many years. Radiation appears to act principally at the initiation stage of normal cells in tissues by introducing certain mutation in the DNA. These mutations allow a cell to enter a pathway of abnormal growth that may lead to development of malignancy (UNSCEAR, 2000).

It became apparent in the early part of the 20th century that groups of people such as radiologists and their patients, who were exposed to relatively high levels of radiation, showed a higher incidence of certain types of cancer than groups not exposed to radiation. More recently, detailed studies of the populations exposed to radiation from atomic bombs, of patients exposed to radiation therapy, and of groups exposed occupationally, particularly uranium miners, have confirmed the ability of radiation to induce cancer (UNSCEAR, 2000).

Radiobiologists have studied the relationship between large doses of radiation and cancer. These studies indicate that damage or change to genes in the cell nucleus is the main cause of radiation induced cancer. This damage may occur directly through the interaction of the ionizing radiation in the cell or indirectly through the actions of chemical products produced by radiation interactions within cells. Cells are able to repair most damage within hours, however, some cells may not be repaired properly. Such misrepaired damage is thought to be the origin of cancer, but misrepair does not always cause cancer. Some cell changes are benign or the cell may die, these changes do not lead to cancer (UNSCEAR, 2000).

Many factors such as age, general health, inherited traits, sex, as well as exposure to other cancer causing agents such as cigarette smoke can affect susceptibility to the cancer-causing effects of radiation. Many diseases are caused by interaction of several factors, and these interactions appear to increase the susceptibility to cancer (UNSCEAR, 2000).

2.6 Cancer Epidemiology

Cancer resulting from radiation exposure cannot be distinguished from those cases resulting from other causes. Radiation-associated cancer in humans is studied in population groups that have been exposed to radiation doses in excess of the normal background (UNSCEAR, 2000).

Estimates of risk have been derived from populations for whom individual doses can be reasonably estimated. Those populations include survivors of the atomic bombings, medically irradiated patients, those occupationally exposed, individuals exposed to radionuclides released into the environment and people exposed to elevated levels of natural background radiation (UNSCEAR, 2000).

It is now well known that radiation can cause cancer in almost any tissue or organ in the body, although some sites are much more prone than others. The life span study cancer incidence and mortality data are broadly similar, demonstrating statistically significant effects of radiation for all solid tumours as a group, as well as for cancers of the stomach, colon, liver, lung, breast, ovary and bladder (UNSCEAR, 2000).

One radiation-associated cancer of particular importance is cancer of the thyroid gland in children. There is strong evidence that the risk of thyroid cancer decreases with increasing age, so the risk in children under 15 years of age is substantially larger than in adults. Among children, those aged 0-5 years are five times more sensitive than those aged 10-14 years. In view of that sensitivity, it is not surprising that large increases in thyroid cancer incidence have been observed in children in Belarus, the Russian Federation and Ukraine following the Chernobyl accident in 1986. The incidence rate of thyroid cancer in children from regions of those countries was ten times higher in 1991-1994 than in the preceding five years. About 1,800 cases of childhood thyroid cancer had occurred in 1998 (UNSCEAR, 2000).

The incidence data also provide evidence of excess radiation risks for thyroid cancer and non-melanoma skin cancers. Statistically significant risks were not seen in either the incidence or the mortality data for cancers of the rectum, gall bladder, pancreas, larynx, uterine cervix, uterine corpus, prostate gland and kidney or renal pelvis. An association with radiation exposure is noted for most types of leukaemia, but not for lymphoma or multiple myeloma (UNSCEAR, 2000).

Studies of populations exposed to medical, occupational or environmental radiation provide information on issues that cannot be addressed by the atomic bomb survivors' data, such as the effects of chronic low doses, alpha doses to the lung from radon, highly fractionated doses and variability among populations. For some cancer sites, including leukaemia, breast, thyroid gland, bone and liver, very useful results come from investigations other than the life span study. Risk estimates derived from those studies generally agree well with those from the life span study (UNSCEAR, 2000).

Data from various epidemiological study shows that all cancers are not fatal (UNSCEAR, 2000). These are age and gender dependant. Average mortality from radiation induced thyroid cancer is about 10%, breast cancer is about 50% and from skin cancer is about 1%. Overall the total risk of inducing cancer by uniformly irradiating the whole body is about half as high as the risk of fatal cancer. In radiological protection, the risk of fatal cancer has great significance. The use of fatal cancer risk makes easier to compare them with other fatal risks encountered in life (UNSCEAR, 2000).

2.7 Cancer Risk

The risk of cancer is usually based on epidemiological study from the statistics of the incidence of specific disorders in specific population groups. The incidence of cancer risk can be calculated, if we know the number of people in irradiated groups and the doses they received. By comparing these numbers with un-irradiated groups, the expected occurrence of cancer can be observed (IAEA, 2004).

Most of the Japanese atomic bomb survivors and other exposed groups received high doses over a short period of time. Observations of cancer incidence in these groups indicate that for high doses and dose rates, there is a linear relationship between dose and risk (doubling the dose received would double the risk) (UNSCEAR, 2000).

However, most radiation exposure involves low doses delivered over long periods. At these low levels of exposure, studies of cancer incidence in the exposed population do not provide any direct evidence about the relationship between dose and risk involved because the number of extra cancers that might be expected to result from the radiation exposure is too small (compared to the total number of cancer cases in the population) to detect. It is, therefore, necessary to consider other scientific information about the effects of radiation on cells and organisms and to form a judgement (UNSCEAR, 2000).

For many years, the internationally accepted solution has been to assume that all the doses have detrimental effect and the relationship is linear for high and low doses, all the way down to zero (known as the linear-no threshold or LNT hypothesis) as shown in figure 5 (UNSCEAR, 2000). However, some radiobiological experiments have interpreted that low doses of radiation have no detrimental effect, because the body can successfully repair all of the damage caused by the radiation, or even that low doses of radiation may stimulate the repair mechanisms in cells to such an extent that they actually help to prevent cancer. Other experiments have been used as the basis for theories that low doses of radiation are more harmful (per unit of dose) than high doses, or that the hereditary effects of radiations could get worse from generation to generation (UNSCEAR, 2000).

After a major review of biological effects at low doses of ionizing radiation, UNSCEAR concluded in 2000 that "an increase in the risk of cancer proportionate to radiation dose is consistent with developing knowledge and it remains, accordingly, the most scientifically defensible approximation of low dose response". However, UNSCEAR also accepted that there are uncertainties and stated, "a strictly linear dose-response relationship should not be expected in all circumstances" (UNSCEAR, 2000).

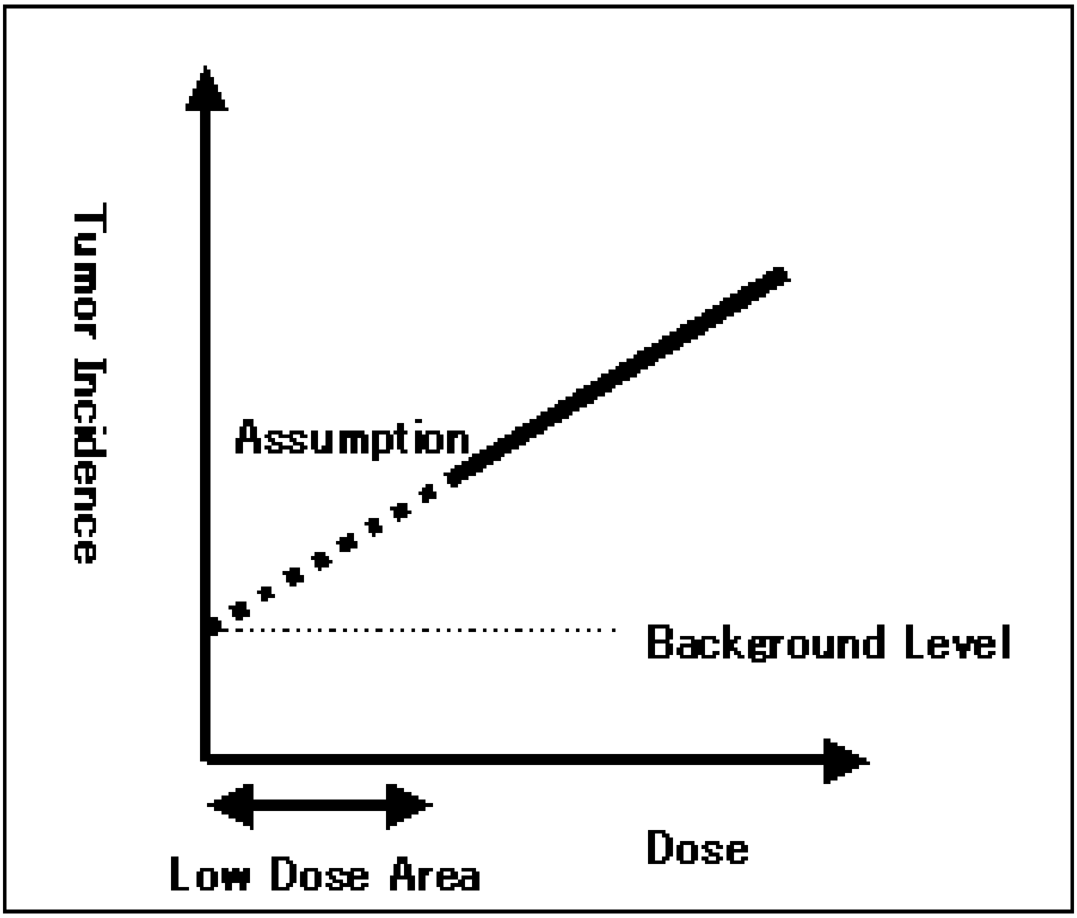


Figure 5: Dose-risk hypothesis (ICRP, 2007)

For some types of strongly ionizing radiation, such as alpha particles, the risk factor is the same at low doses as at high doses. However, for weakly ionizing radiation, such as gamma rays there is considerable radiobiological evidence that a linear relationship is a good approximation of dose response for both the low dose and high dose regions, but the risk per unit dose (the slope of the linear relationship) is less at low doses and dose rates than at high doses and dose rates. ICRP (1991) has estimated the risk factors for fatal cancers from low doses and dose rates using a judicious reduction factor of two.

ICRP has estimated that averaged over a typical population of all ages, a dose of 1 Sv to each individual would result in a radiation induced fatal cancer in about 10% of the persons exposed. This is the same as saying that average risk to an individual from a dose of 1 Sv is about one in ten or 0.1. The extrapolation of this estimate to the much lower doses and dose rates normally encountered as a result of operations in the nuclear industry and elsewhere introduces further uncertainty (ICRP, 1991).

A very conservative approach for low doses is to make a linear extrapolation from high to low dose. Since a dose of 1 Sv carries a risk of fatal cancer of 0.1, the risk from a dose of 1 mSv would be 1000 times lower, or 0.0001. The additional risk of fatal cancer imposed on an average individual by exposure to radiation at low doses and dose rates can be estimated using a dose-risk factor of 0.05 Sv^{-1} or 5×10^{-2} Sv^{-1} by using the ICRP dose and dose rate effectiveness factor (DDREF) of two (ICRP, 1991).

In reality, the risk to an actual person from a given dose depends on person's age at the time of the exposure and on his gender. For example, if a person receives a dose late in life, a radiation-induced cancer may not have time to appear before the person dies of another cause (ICRP, 1991).

A clearer understanding of physiological modifying factors, such as sex and age, has developed over the last few years, although, the absolute risk of tumour induction with sex are not large and vary with site. For most solid cancers the absolute risk is higher in women than in men. People who are young at the time of radiation exposure have higher relative and absolute risks than older people, but again this varies by site. The risk of breast cancer is virtually zero for men and twice for women (0.4×10^{-2} or 1 in 250 per Sv) (ICRP, 1991). ICRP (1991) population risk factors for fatal cancers in different organs are listed in Table 2.3 (IAEA, 2004).

Table 2.3: ICRP Population Risk Factors for Fatal Cancers (IAEA, 2004).

Tissue or Organs	**Risk Factor ($\times 10^{-2}$ Sv^{-1})**
Bladder	0.30
Bone marrow (red)	0.50
Bone surfaces	0.05
Breast	0.20
Colon	0.85
Liver	0.15
Lung	0.85
Oesophagus	0.30
Ovary	0.10
Skin	0.02
Stomach	1.10
Thyroid	0.08
Remainder	0.50
Total (rounded)	5.00

Risk factors are also different for different populations because different populations have different distributions of ages. For example, since the average age of a population of workers is generally higher than that of the population as a whole, the risk factor for the former is somewhat lower than that of the latter (ICRP, 2007).

The ICRP (1991) risk factor for workers is 4 x 10^{-2} or 1 in 250 per Sv. Different risk factors can also result from differences in the prevailing incidence of cancers (or even particular types of cancer) from all causes, because the risk from radiation is assumed to be related to this prevailing incidence. For example, the risk factor for countries with a relatively high level of cancer mortality (e.g. developed countries) would be higher than for those where cancer is less common (e.g. developing countries). However, such differences are fairly small compared to the uncertainly in the ICRP (1991) risk factors. These values can reasonably be used internationally (IAEA, 2004).

The risks of hereditary effects due to exposure of the gonads are very uncertain. ICRP (1991) estimates the risk of serious hereditary health effects in all generations following the irradiation of either parent at low doses and dose rates to be about 3 x 10^{-2} Sv^{-1}, averaged over the whole population. Clearly, only that exposure which occurs up to the time of conception can affect the genetic characteristics of the offspring and, since the mean age of childbearing is about 30 years, only a proportion of the dose received by a typical population will be genetically harmful. The total genetic risk in all generations averaged over both sexes and all ages is therefore about 1.3 x 10^{-2} Sv^{-1}. In a population of working age, because of the different age distribution, the risk is about 0.8 x 10^{-2}

Sv^{-1} (Martin and Harbison, 1996). In addition to fatal cancers, exposure to radiation also gives rise to cancers which are non-fatal or curable. These need to be taken into account but they would not have the same weight as fatal cancers.

2.8 Recommended Dose Limits

To limit consequences of radiation exposure estimates are made for deterministic and stochastic effects. ICRP recommends annual effective dose for uniform irradiation of the whole body of 1 mSv for general public and 20 mSv for radiation workers. However, in special circumstances higher values of effective doses could be allowed in any single year provided the average over 5 years does not exceed 1 mSv for general public and 20 mSv for radiation workers.

For occupational exposures, it is permissible to exceed from 20 mSv in any one year but it should not exceed 50 mSv in any single year. For non-uniform irradiation weighting factors have been assigned to various individual organs relative to whole body as 1, reflecting the harm attributable to irradiation of each organ.

The use of annual effective dose limit of 20 mSv implies that if the conditions of exposure are such that only a single tissue is exposed, the limiting annual equivalent dose for that tissue is:

$$\text{Dose limit}_T = 20/W_T \text{ mSv} \qquad (2.6)$$

For example, in the case of lung, annual dose limit is 170 mSv using its tissue weighting factor, W_T, of 0.12. Similarly, annual equivalent dose limit for thyroid is 400 mSv. The recommended dose limits from external exposure for some organs are listed in Table 2.4 (ICRP, 1991).

Table 2.4: Recommended Dose Limit for Radiation Workers and General Public (ICRP, 1991).

Effective Dose Limit	**Occupational (mSv)**	**Public (mSv)**
Whole body	20	1
Lens of eye	150	15
Skin	500	50
Hands and feet	500	-

2.9 Annual Limit on Intakes

In the cases of internal exposure, Annual Limit on Intakes (ALI) of radionuclides is based on a committed effective dose of 20 mSv y^{-1} (ICRP, 1991). ALI is a secondary limit recommended by ICRP for occupational exposure, which limits the amount of radioactive material taken into the body by an adult worker through inhalation or ingestion in a year. The ALI (Bq) for any one radionuclide can thus be obtained by dividing the annual average effective limit (0.02 Sv) by the dose coefficient, e(50) in Sv Bq^{-1}:

$$ALI = \frac{0.02}{e(50)} \tag{2.7}$$

For example, consider a radionuclide which when taken into the body, irradiates organs X, Y and Z. Suppose that, for an intake of 1 Bq of the radionuclide, the committed equivalent dose to each of these organs is H_X, H_Y and H_Z respectively. If the tissue weighting factors for organs X, Y and Z are w_x, w_y and w_z then the effective dose from an intake of 1 Bq is:

$$H_T = w_x H_X + w_y H_Y + w_z H_Z \tag{2.8}$$

The ALI is the quantity such that

$$H_E = \sum_T w_T H_T = 20\, mSv \tag{2.9}$$

The ALI is then given by

$$ALI = \frac{20}{w_T H_X + w_y H_Y + w_z H_Z} (Bq) \tag{2.10}$$

The dose resulting from an intake may be very different depending on whether the intake is by inhalation or ingestion and so separate ALIs are needed for the two routes of intake. In assessing the total dose, which a person receives in a year, both the external and internal doses are considered to ensure that the recommended dose is not exceeded.

For most of the organs and tissues of the body, the stochastic equivalent dose limits are lower than the threshold doses at which deterministic effects start to occur (generally 0.5 Sv although a few tissues show higher radiosensitivities). The exceptions are the skin, hands and feet for which an equivalent dose limit of 0.5 Sv in one year is recommended and the lens of the eye for which the limit is 0.15 Sv^{-1}.

Thus, the restrictions on effective dose are sufficient to ensure the avoidance of deterministic effects in almost all tissues and organs.

2.10 Standard Models

The human body is a complex system and radiation effects of radionuclide(s) taken into the body depend on many factors such as routes of intakes (inhalation or ingestion) of radionuclide(s), their physical and chemical form, anatomic distribution in the body, duration of retention (biological half-life) and rate of radioactive decay (physical half-life) as well as on the energies and types of the emitted radiations (α, β, γ rays).

Estimation of internal radiation doses due to inhalation or ingestion of radioactive materials is often based on tedious enumeration of assumption and calculations. To make it simple and convenient different models have been developed by the International Committee on Radiological Protection (ICRP) that describe the paths of the radionuclide(s) in the body and provide equations to calculate their transfer rates within and out of the body. The models describe human body as composite of various compartments. Each organ or tissue of deposition is assumed to consist of one or more compartments, and from each of these compartments the radionuclide is translocated at an appropriate rate to the excretion pathways.

2.11 Transport of Radionuclides in the Environment

NORM released to the environment can give rise to human radiation doses. External irradiation is exposure from environment to human directly. Internal irradiation means uptake of human via a variety of pathways such as inhalation of contaminated dust, ingestion of dirt and dust, inhalation of radon diffusing from the material and skin contamination (figures 6a and 6b).

Radioactive materials can be released into air or directly into water or soil. When released in the air, they can travel some distance, depending upon such factors as wind speed and direction and altitude of the release. The products of airborne releases can be transported to humans by a variety of paths. First, direct ingestion by inhalation is possible. Secondly, the materials will eventually deposit themselves on the ground, where they will find their way into plant and animal life and thereby, into the food chain. Third, deposition of airborne contaminants into water can reach humans either by direct ingestion or via the food chain. Similarly, direct soil and water depositions find their way into the food chain via both plant and animal life. Rain water runoff can carry soil into rivers and streams, thereby transporting any soil contamination to water. Additionally, radioactive materials can leach into porous soils and ground water (Canberra Company, 2006).

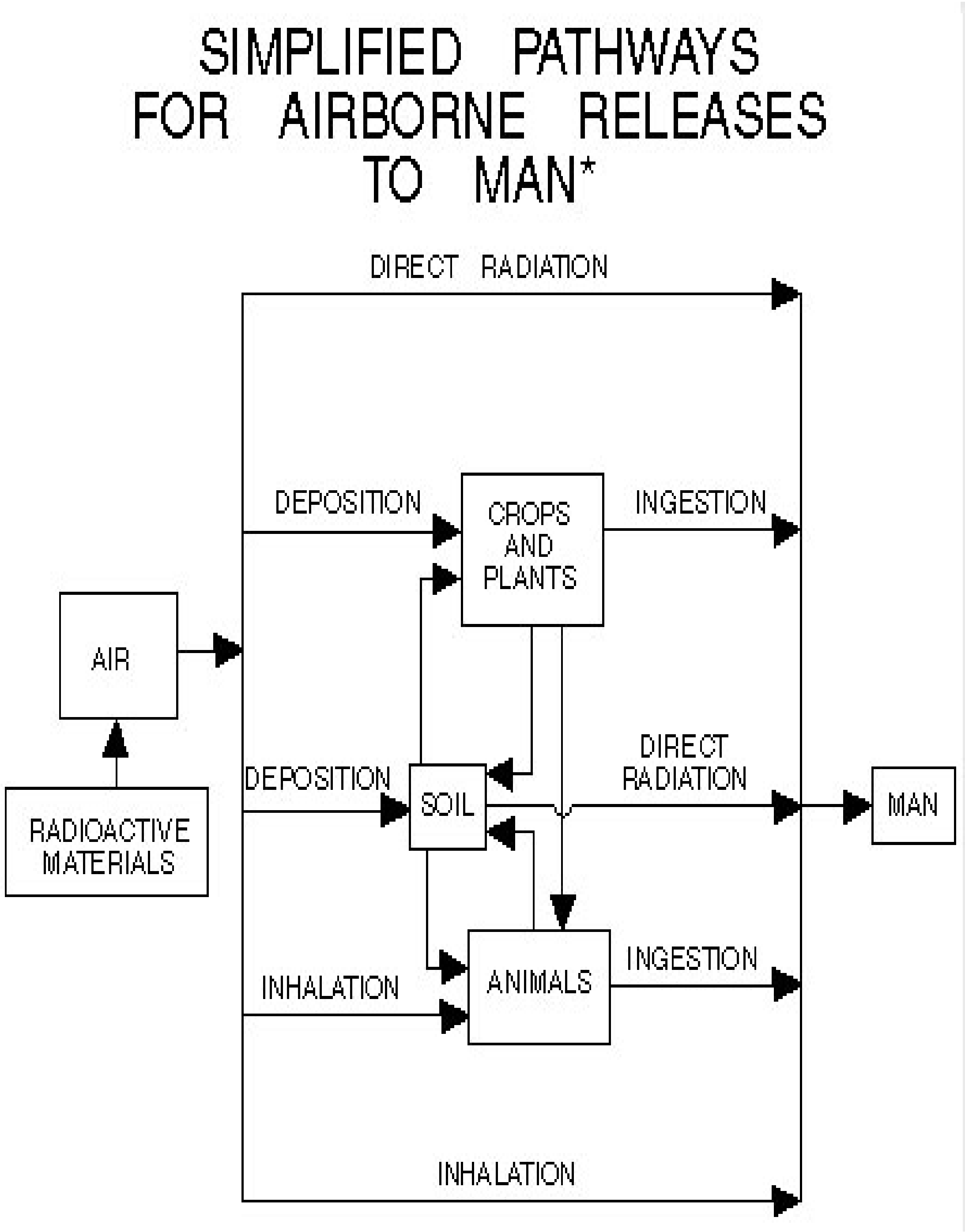


Figure 6a: Simplified possible pathways for airborne releases to humans (Canberra Company, 2006)

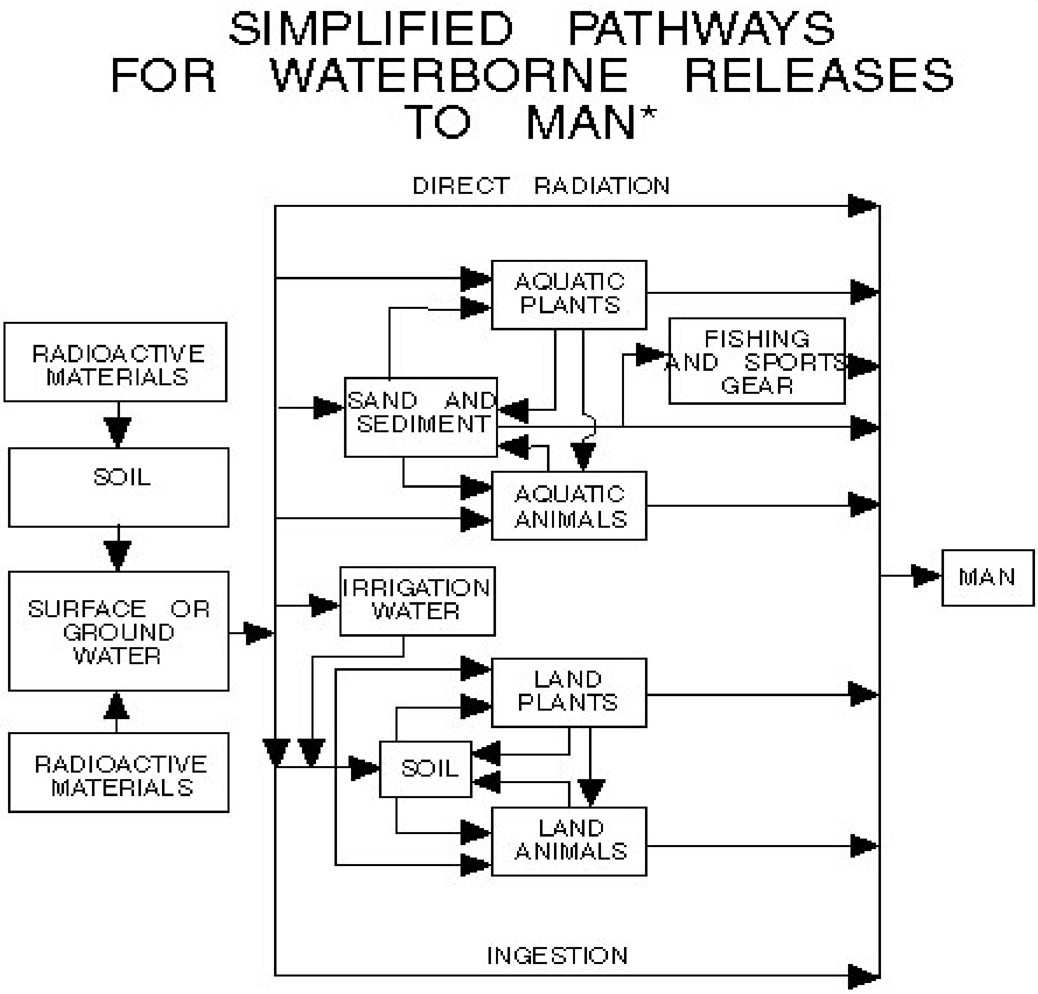


Figure 6b: Simplified possible pathways for waterborne releases to humans (Canberra Company, 2006)

2.12 Committed Dose Equivalent (H_{50})

The committed dose equivalent (H_{50}) in a particular organ or tissue is the total dose equivalent to which that organ or tissue would be committed during the 50 years after intake of a radionuclide. Estimates of H_{50} per unit intake of a radionuclide are made on the characteristics of reference Man.

For purposes of planning in radiological protection, it is assumed that risk of a given biological effect is linearly related to dose equivalent. In these circumstances, risk of an effect is determined by the total dose equivalent averaged throughout the organ or tissue at risk, independent of the time over which that dose equivalent is delivered.

The ICRP recommends that the appropriate period for integration of dose equivalent is a working lifetime of 50 years. The total dose equivalent averaged throughout any tissue over the 50 years intake of a radionuclide into the body is termed the committed dose equivalent, H_{50}, which is therefore given by

$$H_{50} = \sum \frac{\int M D_{50,i} W_{Ri} N_i dm}{\int M dm} \qquad (2.11)$$

where, M is the mass of the specified organ or tissue, $D_{50,i}$ is the total absorbed dose during a period of 50 years intake of the radionuclide into the body in the element of mass dm of the specific organ or tissue, W_{Ri} radiation weighting factor and N_i is the product of all other modifying factors such as dose rate, fractionation, etc.

Therefore, using the values of W_R listed in Table 2.11, which are constant for any type of radiation i, the expression for H_{50} shown in Eqn. (2.11) simplifies to:

$$H_{50} = \sum_i W_{Ri} D_{50,i} \qquad (2.12)$$

where $D_{50,i}$ is the total absorbed dose during the 50 years intake of the radionuclides into the body averaged throughout the specified organ (Table 2.12) or tissue for each radiation of type i.

2.13 Dose Coefficients

The biokinetic models play a central role in the dosimetry of internally deposited radionuclides. The structure of these models makes it possible to incorporate a broad range of physiological information and to adjust specific parameters as a function of age and calculation of dose coefficients.

Now these models are being used extensively in radiation protection to assess radiation doses and calculation of their health impact making life simple for health physicist with the help of the dose coefficients. The values are calculated by using different age-dependent models for respiratory tract, gastrointestinal tract and for the systemic biokinetic behaviour of radionuclides.

Age-dependent dose coefficients are now available from ICRP for various mode of intake of radionuclides (inhalation and ingestion) and of different age groups, e.g. 3 months (from 0 to 1 year), 1 year (from 1 year to 2 years), 5 years (>2 years to 7 years), 10 years (>7 years to 12 years), 15 years (>12 years to 17 years) and adult. For most purposes, the Committee consider the age categories of infants, children and adults and used the available dose coefficients corresponding to 1-2 years, 8-12 years and >17 years respectively. The fractional distribution of infants, children and adults was assumed to be 0.05, 0.3 and 0.65 respectively (UNSCEAR, 2000).

Details of some dose coefficients, e.g. Committed Effective Dose per unit intake e(g) (Sv Bq^{-1}) for members of the public for some radionuclides of interest via ingestion and inhalation (ICRP, 1996) are listed in Table 2.5 to 2.7.

Table 2.5: Ingestion Dose Coefficients e(g) for Thorium

Radionuclides	**Physical Half-life**	**Dose Coefficients (Sv Bq^{-1})**					
		<1a	**1-2a**	**2-7a**	**7-12a**	**12-17a**	**>17a**
Th-228	1.91a	3.7x10^{-6}	3.7x10^{-7}	2.2x10^{-7}	1.5x10^{-7}	9.4x10^{-8}	7.2x10^{-8}
Th-230	7.70 x 10^{4}a	4.1x10^{-6}	4.1x10^{-7}	3.1x10^{-7}	2.4x10^{-7}	2.2x10^{-7}	2.1x10^{-7}
Th-232	1.40 x 10^{10}a	4.6x10^{-6}	4.5x10^{-7}	3.5x10^{-7}	2.9x10^{-7}	2.5x10^{-7}	2.310^{-7}
Th-234	24.1d	4.0x10^{-6}	2.5x10^{-8}	1.3x10^{-8}	7.4x10^{-9}	4.2x10^{-9}	3.4x10^{-9}

Table 2.6: Ingestion Dose Coefficients e(g) for Uranium

Radionuclides	**Physical Half-life**	**Dose Coefficients (Sv Bq^{-1})**					
		<1a	**1-2a**	**2-7a**	**7-12a**	**12-17a**	**>17a**
U-230	20.8d	7.9x10^{-7}	3.0x10^{-7}	1.5x10^{-7}	1.0x10^{-7}	6.6x10^{-8}	5.69x10^{-8}
U-231	4.20d	3.1x10^{-9}	2.0x10^{-9}	1.0x10^{-9}	6.1x10^{-10}	3.5x10^{-10}	2.8x10^{-10}
U-232	72.0a	2.5x10^{-8}	8.2x10^{-7}	5.8x10^{-7}	5.7x10^{-7}	6.4x10^{-7}	3.3x10^{-7}
U-233	1.58x10^{5}a	3.8x10^{-7}	1.4x10^{-7}	9.2x10^{-8}	7.8x10^{-8}	7.8x10^{-8}	5.1x10^{-8}
U-234	2.44x10^{5}a	3.7x10^{-7}	1.3x10^{-7}	8.8x10^{-8}	7.4x10^{-8}	7.4x10^{-8}	4.9x10^{-8}
U-235	7.0x108^{8}a	3.5x10^{-7}	1.3110^{-7}	8.5x10^{-8}	7.1x10^{-8}	7.0x10^{-8}	4.7x10^{-8}
U-236	2.34x10^{7}a	3.5x10^{-7}	1.3x10^{-7}	8.4x10^{-8}	7.0x10^{-8}	7.0x10^{-8}	4.7x10^{-8}
U-237	6.75d	8.3x10^{-9}	5.4x10^{-9}	2.8x10^{-9}	1.6x10^{-9}	9.5x10^{-10}	7.6x10^{-10}
U-238	4.47x10^{9}a	3.4x10^{-7}	1.2x10^{-7}	8.0x10^{-8}	6.8x10^{-8}	6.7x10^{-8}	4.5x10^{-8}
U-239	0.392h	3.4x10^{-10}	1.9x10^{-10}	9.3x10^{-11}	5.4x10^{-11}	3.5x10^{-11}	2.7x10^{-11}
U-240	14.1h	1.3x10^{-8}	8.1x10^{-9}	4.1x10^{-9}	2.4x10^{-9}	1.4x10^{-9}	1.1x10^{-9}

Table 2.7: Ingestion Dose Coefficients e(g) for Potassium

Radio-nuclides	**Physical Half-life**	**Dose Coefficients (Sv Bq^{-1})**					
		<1a	**1-2a**	**2-7a**	**7-12a**	**12-17a**	**>17a**
K-40	1.28x10^{9}a	6.2x10^{-8}	4.2x10^{-8}	2.1x10^{-8}	1.3x10^{-8}	7.6x10^{-9}	6.2x10^{-9}
K-42	12.4h	5.1x10^{-9}	3.0x10^{-9}	1.5x10^{-9}	8.6x10^{-10}	5.4x10^{-10}	4.3x10^{-10}
K43	22.6h	2.3x10^{-9}	1.4x10^{-9}	7.6x10^{-10}	4.7x10^{-10}	3.0x10^{-10}	2.5x10^{-10}
K-44	0.369h	1.0x10^{-9}	5.5x10^{-10}	2.7x10^{-10}	1.6x10^{-10}	1.1x10^{-10}	8.4x10^{-11}
K-45	0.333h	6.2x10^{-10}	3.5x10^{-10}	1.7x10^{-10}	9.9x10^{-11}	6.8x10^{-11}	5.4x10^{-11}

Annual effective doses and committed effective doses are calculated by simply multiplying these dose coefficients with total intake of any radionuclide or mixture of radionuclides and radiological impact is assessed without going into tedious mathematical calculations for different population groups (ICRP, 1996).

History of assessment of impact of radiological important elements on the human being started with the invention of radioactivity, to meet the needs of radiation protection. The early efforts in this context were made by Cook (Cook, 1948) followed by the meticulous undertaking of a Task Group of ICRP. A Reference human model was developed and documented in ICRP-2 and later on in ICRP-23 (ICRP, 1975). The model is based on numerous anatomical, physiological, metabolic data and compositional information for 51 elements. This information served as a valuable data source to meet the health physics requirements for estimation of radiation dose of a typical radiation worker.

With the passage of time and refinements of analytical techniques, a number of authors documented data on trace elements in biological media. Since 1980, well- planned investigations of extensive trace elements and analysis of tissue, body fluid and human diets were carried out on international scale and reliable data have been generated. A few sources are cited here as examples (Iyengar, 1982; Friberg and Vahter, 1983; Grandjean, 1983; Vanoeteren *et al*, 1985; Woittiez and Iyenger, 1988; Minoia *et al*, 1990; Parr *et al*, 1991; Abdulla *et al*, 1992; Wise *et al*, 1993; Christensen, 1995; WHO, 1996; IAEA, 1992). Thus, the database has steadily expanded to include elements not previously reported in ICRP-23.

2.14 Geology of NORM

Gamma radiation detected by exploration geologist looking for uranium was discovered in 1828 by the Swedish Chemist Jons Jakob Berzelius. The great interest expressed worldwide for the study of naturally occurring radiation and environmental radioactivity has led to the performance of extensive surveys in many countries of the world (UNSCEAR, 1993). Such investigations can be useful for the assessment of public exposure dose rates, and the performance of epidemiological studies, in order to ascertain possible changes in environmental radioactivity due to nuclear, industrial, and other human activities, as well as to keep reference data records.

Rock phosphate is a terrestrial source of NORM whose concentration varies from one place to another on the Earth. Efforts are being undertaken to determine radioactivity associated with rock phosphate throughout the world (Makweba and Holm, 1993; Brady, 1990; Tufail *et al*, 2006; Guimond, 1990).

Rocks are of two types, sedimentary and igneous in origin. Sedimentary rocks deposits supply about 80-85% of the rock phosphate as a raw material for manufacturing phosphate fertilizers throughout the world. Igneous type rock phosphate deposits supply the remaining 15-20% of rock phosphate to the fertilizer industry.

Rocks of Morocco, USSR (Russia and the former states of the Soviet Union) and the United States of America (USA) have been analyzed for radioactivity concentration by Guimond (Guimond, 1990), who found the uranium contents to be 1700 Bq kg^{-1} and 1500 Bq kg^{-1} respectively. Rock phosphate of the sedimentary origin contain on average 0.015% of U_3O_8. Oguneleye *et al*. (Oguneleye *et al*, 2002) analyzed the ^{226}Ra component of the Nigerian phosphate rock from Sokoto and recorded up to 967 Bq kg^{-1}. Ashraf *et al.* (Ashraf *et al*, 2001) estimated the exposure to mine workers in Egypt, to be greater than the maximum permissible level in ICRP-60 report (http://www.tenorm.com/). Habshi (Habshi, 1994) determined the uranium component up to 200 ppm in the rock phosphate in Egypt. Khan *et al* (Khan *et al*, 1998) determined the natural radioactivity in Pakistani phosphate rock and have reported very high ^{226}Ra content in phosphate rock and in local/imported fertilizers which varied from 307.7 Bq kg^{-1} to 617.5 Bq kg^{-1}.

2.14.1 Igneous Rocks

The original sources of uranium-series, thorium-series, actinium-series, potassium and rubidium radioactivity in the terrestrial environment are the earth's crust and mantle (Table 2.8). As molten magma cools, silicate minerals are formed (i.e. magmatic differentiation). In the early stages of the cooling, the silicates tend to be mafic (which are predominately iron and magnesium), and deficient in aluminium, silicon, sodium, and potassium. The mafic rocks are dark in colour. As cooling and differentiation progress, the balance tends to reverse, the silica, containing mostly silicon-aluminium igneous rocks is formed. They are generally lighter in colour or speckled. Figure 7 shows a generalization of the process, known as the Bowen reaction series (Montgomery, 1990). Neither uranium nor thorium is compatible with the crystal structure of the major silicates. In addition, they are present in such small quantities as to have little tendency to form mineral in which they would be essential components. The result of this relationship is that the remainder of the magma cools to form miscellaneous and varied minor minerals which contain the uranium, thorium and other minor and trace elements. The last major silicates to crystallize are also those which contain most of the potassium and rubidium.

Table 2.8: Crustal concentrations of terrestrial radionuclides (NCRP, 1994).

Rock Type	**Uranium**	**Thorium**	**Potassium**	**Rubidium**
	ppm	**ppm**	**%**	**ppm**
Mafic (Dark Coloured)	0.5 – 1	3 – 4	0.8	40
Salic (Light Coloured)	3	17	4	170 - 200

2.14.2 Weathering of Igneous Rocks

Mechanical (physical) and chemical processes break rock down into soil. Weathering plays a key role in this process. Where mechanical processes dominate the breakdown, the separation usually occurs along mineral boundaries that lead to a separation of the major silicates from the minor ones containing the thorium and uranium. These minor minerals include zircon and monazite. They are stable and resistant to chemical decay and are often found as small individual grains.

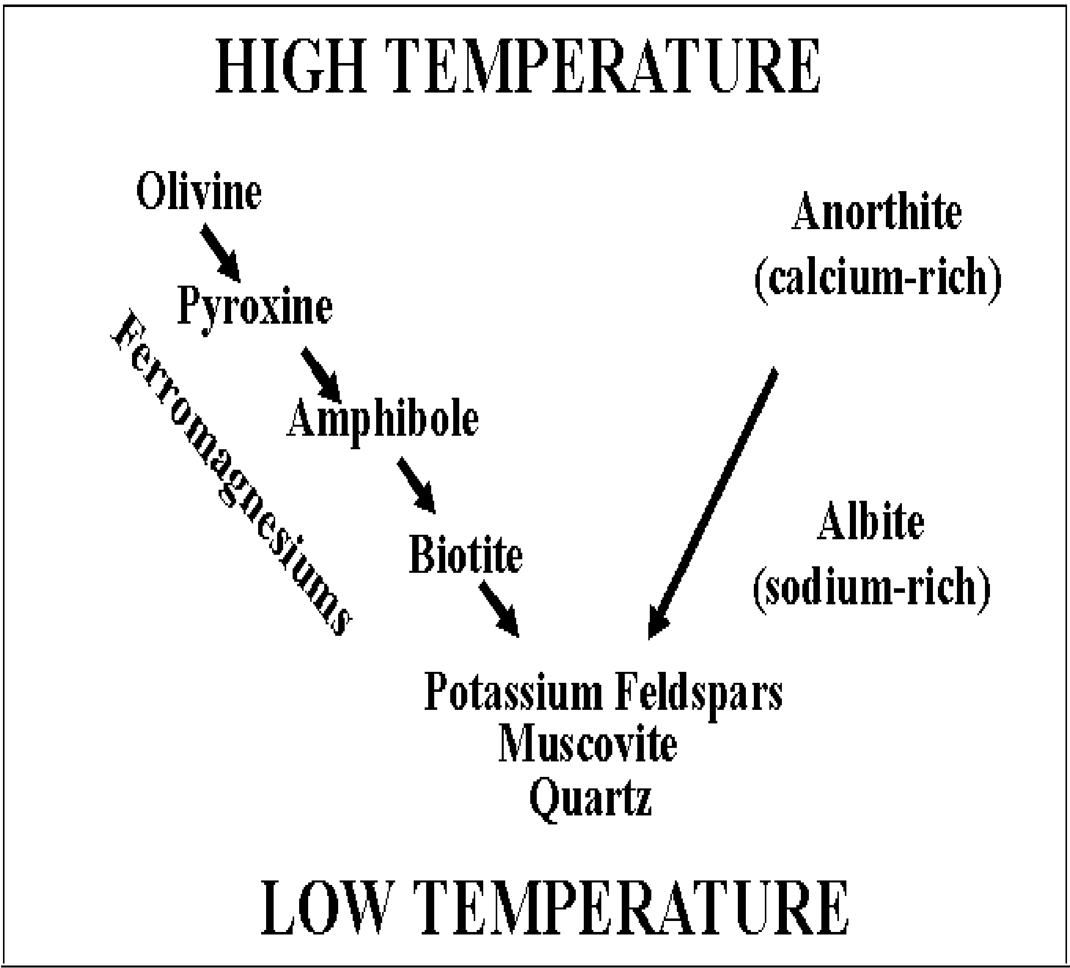


Figure 7: Bowen reaction series (NRC, 1994)

Where chemical (or biological) action predominates, the thorium - and uranium-bearing minor minerals can give up their radionuclides to layers of cations in clay minerals. When the host rocks erode, the clay minerals containing the adsorbed series radionuclides tend to be separated from the major minerals. Therefore, if igneous rock is broken down to individual grains, the products end up as:

- sands of the major mineral, depleted of the radionuclides;
- fine-grained clay minerals, slightly enriched in the radionuclides;
- relatively small quantities of resistant, dense grains of the minor minerals containing most of the series radionuclides; and
- the potassium and rubidium are removed in solution.

2.14.3 Sedimentary Rocks

Although they are only a small part of the earth's crust, sedimentary rocks cover about 85% of the land area of U.S.A. Therefore, much of the surface soil is derived from sedimentary rock. Sedimentation processes naturally sort the products of weathering and develop several major sedimentary rock types of significantly differing radionuclide concentrations. The major types are:

- shales;
- sandstones; and
- carbonate rocks.

As with the igneous rocks, thorium and uranium tend to be minor or disseminated. The radionuclides may become mobile or be deposited by migration of water or oil. Some organic complexes, notable humic acids, create mobile complexes of uranium. Uranium and other minor and trace elements have an affinity for crude oil. They are probably residues of consolidated organic and marine deposits. Petroleum is often assumed to have migrated to a position of minimum hydraulic potential in a reservoir rock, which may be derived from the same source deposits as the petroleum.

2.14.4 Shales

Shales normally contain at least 35% clay minerals, and a significant fraction contains potassium as an essential constituent. Shales can adsorb the series radionuclides. The radionuclides may also be present bound to organic matter in minor minerals or as precipitates or co-precipitates in the cementing material that binds the rock.

2.14.5 Sandstones

Sandstones are usually made of grains that are primarily quartz but may contain some potassium-containing feldspar. Those sandstones that contain more than 25% feldspar are called arkoses, and the chief feldspars are those containing potassium. On the whole, sandstones are low in both the series and non-series radionuclides. However, many deposits of uranium are found at the boundary of different layers of sandstones.

2.14.6 Carbonate Rocks

Carbonate rocks are limestones or dolomites derived by chemical precipitation from water or by the build-up of shells, bones, and teeth of organisms. Although the carbonate minerals themselves are relatively free of radionuclides, the intergranular spaces may contain elements found in the sea water from which they were deposited. Potassium is very soluble and does not stay in the deposited matter. Thorium is depleted in sea water and is not metabolized by marine organisms. Therefore, potassium and thorium are usually of low concentrations in carbonate rocks, but uranium may be present because it may be fixed by reducing conditions in decaying organic matter where the rocks are deposited. Uranium can replace calcium or be adsorbed in the principal phosphate minerals.

2.15 Radioactivity in Soil

Radioactivity in soil results from the rock from which it is derived. It is:

- diminished by leaching of water,
- diluted by increased porosity and by added water and organic matter, and
- augmented by sorption and precipitation of radionuclides from incoming water.

It is the top 0.25 m of soil that contributes significantly to background dose. Table 2.9 is a summary of concentrations of major radionuclides in major rock types and soil. Background concentrations of radionuclides in soil vary because of many factors. Soil may have been produced from the weathered top layer of still-intact bedrock below or transported laterally from the same rock unit or type some distance away. Some methods of transport are:

1. Natural phenomena such as earthquakes, volcanoes, glaciers and changes in soil composition from flooding.
2. Water is the dominant transporting medium. Glacier-derived deposits are common in the Great Lakes area, New England, and Alaska. Outwash erosion products from mountains may produce a soil surface that is more radioactive than the underlying bedrock.
3. Wind can be a significant factor.

Primordial radionuclides have very long half-lives (~ billions of years). Terrestrial radioactivity, and the associated external exposure due to gamma radiation, depend primarily on the geological and geographical conditions, and appear at different levels in the soils of each region in the world. Naturally occurring radionuclides (uranium and thorium series) are the largest contributors of radiation doses received by human beings (UNSCEAR, 1998).

Uranium is found in all rocks and soils. In the common rock types, the uranium concentrations range from 0.5 to 4.7 ppm, corresponding to activity concentrations for ^{238}U of 7 to 60 Bq kg^{-1}. Some ores mined and processed for nonradioactive materials can produce residues with elevated concentrations of radionuclides. A well-known example is phosphorus ore, which contains uranium up to 120 ppm.

Many scientists of the world worked in this field of natural radioactivity mostly by using gamma ray spectroscopy to determine ^{232}Th, ^{226}Ra, ^{40}K and ^{137}Cs. Hamid *et al*. (Hamid *et al*, 2002) determined the distribution patterns of both anthropogenic and natural radionuclides in the soils of Dhaka and northern districts of Bangladesh.

Table 2.9: Summary of concentrations of major radionuclides in major rock types and soil (Phillip *et al*, 1978)

Material	**Potassium-40**	**Thorium-232**	**Uranium-238**
	Bq kg^{-1}	**Bq kg^{-1}**	**Bq kg^{-1}**
Igneous rocks			
Basalt (crustal ave)	300	10-15	7-10
Mafic	70-400	7	7
Salic	1100-1500	60	50
Granite (crustal ave)	>1000	70	40
Sedimentary rocks			
Shale sandstone	800	50	40
Clean quartz	<300	<8	<10
Dirty quartz	400	10-25	40
Arkose	600-900	<8	10-25
Beach sand	<300	25	40
Carbonate rocks	70	8	25
All rocks (range)	700-1500	7-80	7-60
Continental crust (ave)	850	44	36
Soil (ave)	400	37	22

Ebaid *et al*. (Ebaid *et al*, 2000) and El-Shershaby (El-Shershaby, 2002) studied the Egyptian soils; in the northeastern area of the river Nile. Northeastern desert in Egypt has been monitored by gamma ray spectrometry. In this desert, uranium prospect is located near the Red Sea coast. Nnavas *et al*. (Nnavas *et al*, 2002) studied natural radioactivity in soil profiles in central Spain. Pyrenees soil profiles developed on

Tertiary sedimentary mountains landscape, in Spain Amrani and Tahtat (Amrani and Tahtat, 2001) carried study of natural radioactivity in Algerian soils.

Slvasekarapandian *et al*. (Slvasekarapandian *et al*, 2000) studied the distribution of radioactivity in Indian soils having high background radiation on the west coast of India. Singh Baldev and Singh Ajay (Singh and Singh, 2004; Singh and Singh, 2003) determined the activity variation levels in Himachal Predesh, India due to the presence of enhanced concentration of natural radioactive materials in the soil. Bikit *et al*. (Bikit *et al*, 2001) found out the radiation levels in the Vojvondina, agricultural soils in Yugoslavia were contaminated with depleted uranium. Sitaram and Umawaiti (Sitaram and Umawaiti, 2002) determined the activity levels in the Fiji, in the Pacific oceanic region having volcanic eruptions. They found that gamma dose was doubled in that region than that of the coralline area. El-Alvarado *et a*l. (El-Alvarado *et a*l, 2002) found out the natural radioactivity levels in a soil of a nuclear facility site located in a coniferous forest in central Mexico and the results indicate that the specific activities of ^{137}Cs, ^{226}Ra and ^{235}U were in general below the recommended screening limits in NCRP (NCRP, 1999) for surface soils from open fields and forested sites. Anastas (Anastas, 2000) determined the level of activity in the geological and mineralogical as well as their physico-mechanical properties and reserves in Albanian clays and the results obtained show that the concentration of radioelements in clays and soil are within the values accepted as "normal" (the value of 0.37 Bq per gram, as recommended by Smirnov (Smirnov, 1984). Akhtar *et al*. (Akhtar *et al*, 2005; Akhtar *et al*, 2003) determined the outdoor gamma radiation level in the Lahore and Faisalabad districts saline soils. The subject of radioactive monazite was studied by Brazilian Academy of sciences (Brazilian Academy of Sciences, 1997). Zahid *et al*. (Zahid *et al*, 2001) and

Amanat *et al*. (Amanat *et al*, 2002) assessed the radioactivity in the Peshawar plain and the mountain area of Pakistan including salt range. Baggoura *et al*. (Baggoura *et al*, 1998), Karunkara *et al*. (Karunkara *et al*, 2001) and Narayana *et al*. (Narayana *et al*, 2001) determined the natural and artificial radioactivity in Algeria, Kaiga and Karnataka with its long, south west coast and south coast of India, by means of gamma spectroscopy. These parts of India are covered by industry, nuclear power plants and fertilizer factories. Miah *et al*. (Miah *et al*, 1997)] studied the activity distribution in Decca city, Bangladesh. Tufail *et al*. (Tufail *et al*, 1992; Tufail *et al*, 2006) studied the natural radioactivity from saline soil of Faisalabad and the building materials used for dwellings in Islamabad and Rawalpindi, Pakistan. Mu-Ming *et al*. (Mu-Ming *et al*, 1987) measured the terrestrial gamma radiation in Taiwan, Republic of China, which is a part of geosynclines, an orogenic arc of islands, marking an active fault line of the Earth's tectonic plates. They found that radioactivity in igneous rocks was greater than sedimentary rocks. Osavaldo *et al*. (Osavaldo *et al*, 2001) determined the distribution of natural radioactivity in soils of Cuba. Chiozzi *et al*. (Chiozzi *et al*, 2003) determined radiation levels in the volcanic islands of Southern Tyrrhenian Sea.

Calculations of individual effective dose per annum have been made from the activity concentrations of dominant gamma-emitting radionuclides from uranium and thorium decay chains in ore, solid waste (spoil heap) and soil samples collected from Adassedakh and Hadal Auatib gold mines in Ariab (Eastern Sudan). The study basically confirmed that the activity levels in the spoil heap are low relative to natural background (i.e. soil samples), and thus the expected contribution in enhancement of natural background radiation is correspondingly low. The corresponding annual effective doses fall within the range: 0.01-0.20 mSv y^{-1}. Although, these estimates did not include the internal radiation burden due to inhalation of radon and its decay products, the values obtained are insignificant relative to the dose limit specified for occupational exposure of any worker (Sam and Awad Al-Geed, 2000).

2.16 Radioactivity in Water

Minute traces of radioactivity are normally found in all types of drinking water. The concentration and composition of these radioactive constituents vary from place to place, depending principally on the radiochemical composition of the soil and rock strata through which the raw water may have passed.

Many natural and artificial radionuclides have been found in water, but most of the radioactivity is due to a relatively small number of nuclides and their decay products. Among these are the following emitters of radiation of low linear energy transfer (LET): potassium-40 (^{40}K), tritium (^{3}H), carbon-14 (^{14}C), and rubidium-87 (^{87}Rb). In addition, high-LET, alpha-emitting radionuclides, such as radium-226 (^{226}Ra), the daughters of radium-228 (^{228}Ra), polonium-210 (^{210}Po), uranium (U), thorium (Th), radon-220 (^{220}Rn), and radon-222 (^{222}Rn), may also be present in varying amounts. Radionuclides that are responsible for the natural radioactivity in drinking water come from radioactive elements, and their decay products, that were incorporated in the earth at its formation, and others are produced continuously by cosmic ray bombardment (Jacobs, 1968). Of all the natural radionuclides that occur in water and emit low-LET radiation, potassium-40 is likely to be the most significant. This primordial radionuclide occurs as a constant percentage (0.0118%) of total potassium.

Radionuclides that are produced by the decay of uranium-238 and thorium-232 are widely distributed throughout the earth's crust. The majority of them are alpha-emitters and includes isotopes of polonium, radon, and radium (UNSCEAR, 1972). Concentrations of uranium in drinking water are extremely variable, apparently ranging from 0.02 to 200 $\mu g\ l^{-1}$ in fresh waters. The thorium content of drinking water has not been extensively measured, but its concentration in the human skeleton is about 37 $\mu Bq\ g^{-1}$ of ash; the corresponding abundance of uranium in the skeleton is about 10 times greater.

The primordial uranium found ubiquitously in nature consists of two isotopes with mass numbers 235 and 238. In the earth's crust, ^{238}U constitutes 99.27% of the uranium by mass, and ^{235}U, the parent isotope of the actinium chain, 0.72%. ^{234}U, a shorter-lived member of the ^{238}U chain, is usually in radioactive equilibrium or near-equilibrium with the parent isotope. Oxidation-reduction processes play a major role in the occurrence and behaviour of uranium in aqueous environments. Uranium transport generally occurs in oxidizing surface water and groundwater as the uranyl ion, UO_2^{2+}, or as uranyl fluoride, phosphate, or carbonate complexes. UO_2^{2+} and uranyl fluoride complexes dominate in oxidizing, acidic waters, whereas the phosphate and carbonate complexes dominate in near-neutral and alkaline oxidizing waters, respectively. Hydroxyl, silicate, organic and sulphate complexes might also be important, the sulphate complex being important especially in mining and milling operations that use sulphuric acid as a leaching agent.

Uranium also occurs in air, water and food and so is present in human tissues. It is introduced into water supplies as a result of leaching from natural sources, from mill tailings, from emissions from the nuclear industry, from the combustion of coal and other fuels, and from phosphate fertilizers. The average annual intake of uranium from all dietary sources is about 13 Bq (NCRP, 1987). In the United States, the typical concentration of uranium in skeleton (wet weight) is about 8 mBq kg^{-1}. Lung, kidney and bone receive the highest annual doses of radiation from uranium, estimated at 11, 9.2 and 6.4 µSv respectively for US residents (NCRP, 1987).

Thorium is a naturally-occurring, radioactive metal and the only primordial isotope of thorium is thorium-232 (more than 99%). In the environment, thorium-232 exists in various combinations with other minerals, such as silica. Like uranium, it is ubiquitous in nature. In aqueous systems, only the Th^{4+} oxidation state is known to exist. Th^{4+} undergoes hydrolysis in aqueous solutions above pH 2-3 and is subject to extensive sorption by clay minerals and humic acid at near-neutral pH. At near-neutral pH and in alkaline soils, precipitation of thorium as a highly insoluble hydrated oxide phase and co-precipitation with hydrated ferric oxides can be important mechanisms for the removal of thorium from solution with sorption reactions. Because of sorption and precipitation reactions and the low solution rate of thorium-bearing minerals, thorium concentrations in neutral waters are generally low. At low pH, such as in an acid-leach uranium mill, thorium becomes more soluble. Acid-leach milling might dissolve 30-90% of the thorium in the ore. It has been reported that acid effluents (pH 2.5) from uranium mills in the Grants Mineral Belt of New Mexico contain ^{230}Th at 5.6 to 6.3 MBq m^{-3}. The solubilised thorium can be precipitated if the acidic effluent is neutralized by contact with neutral media or by process additions of limestone to the waste solution. Similarly, under acidic conditions at some uranium mills, ^{230}Th have been shown to have migrated considerably deeper into subsoil than ^{226}Ra (USDOE, 1993).

The radium content of surface water of 4 to 19 Bq m^{-3} is lower than that of most ground waters (Hess *et al*, 1985). Radium-228 is a member of the ^{232}Th chain. NCRP (NCRP, 1987) estimates that the daily intake of ^{228}Ra is about 0.04 Bq which can be compared to its ^{226}Ra estimate of 0.05 Bq. ^{228}Ra and its products are estimated to contribute annual dose equivalents of 300 µSv to cortical bone, 84 µSv to trabecular bone, 120 µSv to the bone lining cells, 22 µSv to the red marrow and 1.5 µSv to soft tissues (NCRP, 1987).

Seawater contains ^{40}K at about 11 kBq m^{-3}. Because of its relative abundance, ^{40}K is the predominant radioactive component in common foods and human tissues. It is important to recognize that the potassium content of the body is under homeostatic control and is little influenced by environmental variations. The dose from ^{40}K in the body is therefore reasonably constant. From the specific activity of potassium, it follows that the ^{40}K content of the human body is around 4 kBq. NCRP (NCRP, 1987) has estimated that this radionuclide delivers an annual dose of 0.18 mSv to the soft tissues and 0.14 mSv to bones.

2.17 Radon

Radium-226 is one of the decay products of uranium-238, which is widespread in most rocks and soils. When this radium decays it produces radon-222, an inert gas with a half-life of almost 4 days. Radium-224 is a decay product of thorium, and it decays to radon-220, also known as thoron, with a 54-seconds half-life. Because radon is so short-lived, and alpha-decays to a number of daughter products which are solid and very short-lived, there is a high probability of its decay when breathed in, or when radon daughter products in dust are breathed in. Alpha particles in the lung are hazardous.

Radon levels in the air range from about 4 to 20 Bq m^{-3}. Indoor radon levels have attracted a lot of interest since the 1970s and in USA they average about 55 Bq m^{-3}, with a USEPA action level of 150 Bq m^{-3}. Indoor and outdoor radon concentrations, the build-up and indoor exposure, as well as meteorological parameters of Midvaal Water and Botshabelo Community Health Center within the Klerksdorp gold mining areas of the North West Province, South Africa were determined. The average indoor and outdoor concentrations of radon were measured for Botshabelo and Midvaal. The Klerksdorp area has the highest radon concentration, build-up and exposure in Southern Africa (Nnenesi *et al*, 2009). Levels in Scandinavian homes are about double the US average, and those in Australian homes average one fifth of those in USA. Levels up to 100,000 Bq m^{-3} have been measured in US homes. In caves open to the public, levels of up to 25,000 Bq m^{-3} have been measured.

Underground rock containing natural uranium permanently releases (emanates) radon to water in contact with it (groundwater). Radon-222 dissolved in potable water is another source of human exposure, mainly because the ^{222}Rn is released from solution at the tap and enters the home atmosphere. Radon-222 content of groundwater is unusually high. As radon evaporates from surface water more intensively, water from wells normally has much higher concentrations of radon than lakes and streams. The average concentration of radon in public water supplies derived from surface waters is usually less than 0.4 Bq l^{-1} and about 20 Bq l^{-1} in ground water sources. Some wells have been identified with high concentrations, up to 400 times the average and in rare cases exceeding 10 kBq l^{-1}. UNSCEAR in its Report 2000 gives reference to the National Academy of Sciences (NAS) report (NAS, 1999) regarding the air-water concentration ratio of 10^{-4} and calculates the average dose from radon in drinking water as low as 0.025 mSv y^{-1} via inhalation and 0.002 mSv $y^{-1}$from ingestion as compared to the inhalation dose from radon in air of 1.1 mSv y^{-1} (UNSCEAR, 2000).

Radon also occurs in natural gas at up to 37,000 Bq m^{-3}, but by the time it gets to consumers the radon has largely decayed. However, the solid decay products then contaminate gas processing plants, and this manifestation of NORM is an occupational health issue.

2.18 Radiation exposure through drinking water

Radiological contamination of drinking water can result from:

1. naturally occurring concentrations of radioactive species (such as radionuclides of the thorium and uranium decay series in drinking water sources);
2. technological processes involving naturally occurring radioactive materials (for example, the mining and processing of mineral sands or phosphate fertilizer production); or
3. manufactured radionuclides (produced and used in unsealed form), which might enter drinking water supplies in case of improper medical or industrial use and disposal of radioactive material.

The contribution of drinking water to the total exposure is very small and is due largely to naturally occurring radionuclides in the uranium and thorium decay series. Radionuclides from the nuclear fuel cycle and from medical and other uses of radioactive materials may, however, enter drinking water supplies. The contributions from these sources are normally limited by regulatory control of the source or practice. Contamination of drinking water is of regulatory concern for the protection of the members of the public in such water sources.

2.19 Health effects of Ionizing radiation from ingestion of drinking water

There is evidence from both human and animal studies that radiation exposure at low to moderate doses may increase the long-term incidence of cancer. There is evidence from animal studies that the rate of genetic malfunctions may be increased by radiation exposure. Acute health effects of radiation, appearing with symptoms of nausea, vomiting, diarrhoea, weakness, headache, anorexia leading to reduced blood cell counts and in very severe cases to death, occur at high doses of exposure of the whole body or large part of the body (IAEA, 1998). Therefore, acute health effects of radiation are practically not a concern for continuous monitoring of radioactivity content in central drinking water supplies. However, extreme situations of possible terrorist use of radioactive materials to contaminate drinking water supplies, theoretically, cannot be excluded.

2.20 Radioactivity in Food Crops

Plants take some fraction of the radioactivity present in the soil. The soil to plant transfer factor (TF) is broadly used as one of the parameters to estimate the intake of radionuclides through food ingestion. It depends upon many factors like soil pH, soil type, physical and chemical form of the radionuclides in the soil, oxidation-reduction potential in soil, kinds of plants etc (UNSCEAR, 1988).

Human beings are internally and externally exposed to radioactivity present in phosphate rocks and its by-products. The external exposure is from gamma rays coming from rocks. The internal effects are radon, ingestion of rock and dust, and the radioactivity transferred to food. The radiation dose taken will depend upon the intensity of consumption of food, the nature of the soil on which the particular crop has grown, the health and the age of the user. Many scientists have observed that some part of radioactivity taken by the plant from the soil (Tsukada et al, 2002; Ban-Nai et al, 1998; Pulhani et al, 2005; Uchida *et al*, 2005; Sheppard and Evenden, 1988; Morton *et al*, 2001; Chen *et al*, 2005).

2.21 Radioactivity in mining and mineral processing industry

The mining and mineral processing industry has by far the largest technologically enhanced naturally occurring radioactive materials (TENORM) solid-waste volume – an estimated US inventory of about 50 billion tons – most of it with NORM concentrations less than 10 times background. On the basis of geologic reasoning, Bliss (Bliss, 1978) has outlined the types of metallic ores whose mining and extraction might lead to NORM problems. The list includes:

1. Ores of rare-earth elements, molybdenum, gold, aluminium
2. Lead-zinc, iron, tin, vanadium, copper and other metals (commercial-scale by-product recovery of uranium has occurred in connection with the extraction of copper and gold).
3. Placer deposits of any metal (for thorium and its decay products).
4. Ores that result from intense weathering, such as bauxite.

In many areas of the world, gold and/or uranium has been extracted, either as a primary product – e.g. Rössing mine in Namibia - or as a by-product – e.g. Witwatersrand goldfields in South Africa. In such cases, large amounts of waste rock and tailings (waste material left over from various industrial processes in gold extraction) are deposited at the surface, and may migrate in the environment. Furthermore, tailings and waste rock may be used as a road fill or building material. Such practices have led to elevation of the ambient radioactivity levels in these areas, as well as contamination of water-courses, both by transported sediment as well as dissolved radionuclides which precipitate in stream sediments.

While natural radioactivity has been studied in developed countries, like USA and France, there is apparently limited work done in developing countries including Ghana in the area of natural radioactivity in the environment (soil, air, vegetation, surface and underground water (Yeboah *et al*, 1997; UNSCEAR, 1999).

The mining and processing of ores for the production of metals generate large quantities of residual bulk solid and liquid wastes. Because the minerals of value make up only a small fraction of the ore, most of this bulk material has no direct use. It is estimated that the mining and processing of ores and minerals, other than uranium and phosphate, has resulted in the production of more than 40 billion MT (44 billion short tons) of mine waste and tailings from 1910 to 1981.

The metals extraction industry typically generates about 1.5 billion MT (1.65 billion short tons) of waste per year, including about 1.0 billion MT (1.1 billion short tons) of waste rock and overburden, 0.4 billion MT (0.44 billion short tons) of ore tailings, and less than 0.1 billion MT (0.11 billion short tons) of smelter slag. Depending on the original ores and processing methods, some of these wastes contain elevated concentrations of TENORM (Table 2.10).

Table 2.10: Metal and mining industries known to involve TENORM (USEPA, 1993).

Bauxite	Lead	Thorium
Beryllium	Molybdenum	Tin
Columbium	Nickel	Uranium
Copper	Rare Earths	Titanium
Gold	Silver	Zinc
Iron	Tantalum	Zirconium

It is generally believed by geologists that the level of NORM found in ores depends more on the geologic formation or region rather than on the particular type of mineral being mined. These ores often contain many different minerals, and the radionuclide content of one type of ore or mining operation or its wastes will not be representative of other mines or waste types. For some ores, the refining process may yield a waste process that may contain higher radionuclide concentrations when compared to the original ore. It has been reported that some of the more uncommon metals have highly radioactive waste products. Also, some processes associated with metal extraction appear to concentrate certain radionuclides and enhance their environmental mobility.

Most of the metal mining waste is stored on-site or near the point of generation, in tailings ponds or used to construct dams, dikes, and embankments. About two thirds is mine waste, and one third is tailings. Metal mining processing wastes have only been reused in a limited number of applications, typically for backfilling mined out areas and for construction and road building near the mines. Some mineral processing wastes have been used to make wallboard and concrete.

Some of the mining wastes are stockpiles that are reprocessed several times to extract additional minerals. NRC staff published guidance on September 22, 1995 (NRC, 1995); allowing for certain feedstock containing uranium and thorium to be processed by licensed uranium mills. This will allow the wastes to be disposed of in the uranium mill tailings pile. There are several restrictions on the feedstock.

Studies in a number of countries have shown that mining and mineral processing activities lead to elevated levels of NORM as materials are brought up from within the earth's crust on to the surface. They are thus acknowledged as potential sources of exposure. NORM in the mines are usually concentrated and distributed in stockpiles, storage tank, waste piles (tailings dams) and build-up of surface contamination on pipes and equipment (Kathren, 1998). NORM in the mining industry has become a global key issue due to the incidence of cancer discovered in uranium and non-uranium mines (Lubin *et al*, 1990; Morrison *et al*, 1988; NRC, 1988). Regulatory authorities worldwide have become concerned with the health, safety and protection of workers and the public against the risks associated with NORM and NORM wastes in the mines (Paschoa, 1998; Heaton and Lambley, 1995; ACOP, 1985; Kolb and Wojcik, 1985; IRR85, 1985).

2.22 Pathways of Radionuclides

Radionuclides released into the environment as a result of human activities can add to exposure. These can be hazardous to living tissues when they are inside an organism where radiation released can be immediately absorbed. They may also be hazardous when they are outside of the organism but close enough for some radiation to be absorbed by the tissue. Therefore, it is important to know how much radionuclides move through the environment and into the human body. When the pathways that radionuclides follow are known, it is possible to take remedial measures to block or avoid those pathways.

Radionuclides travel through the environment along the same pathways as other materials. They travel through the air, in water (both groundwater and surface water), and through the food chain. The radionuclides may enter the human body by ingestion or inhalation, or through the skin (Fentiman *et al*, 2004). A schematic illustration of possible exposure pathways is shown in figure 8.0.

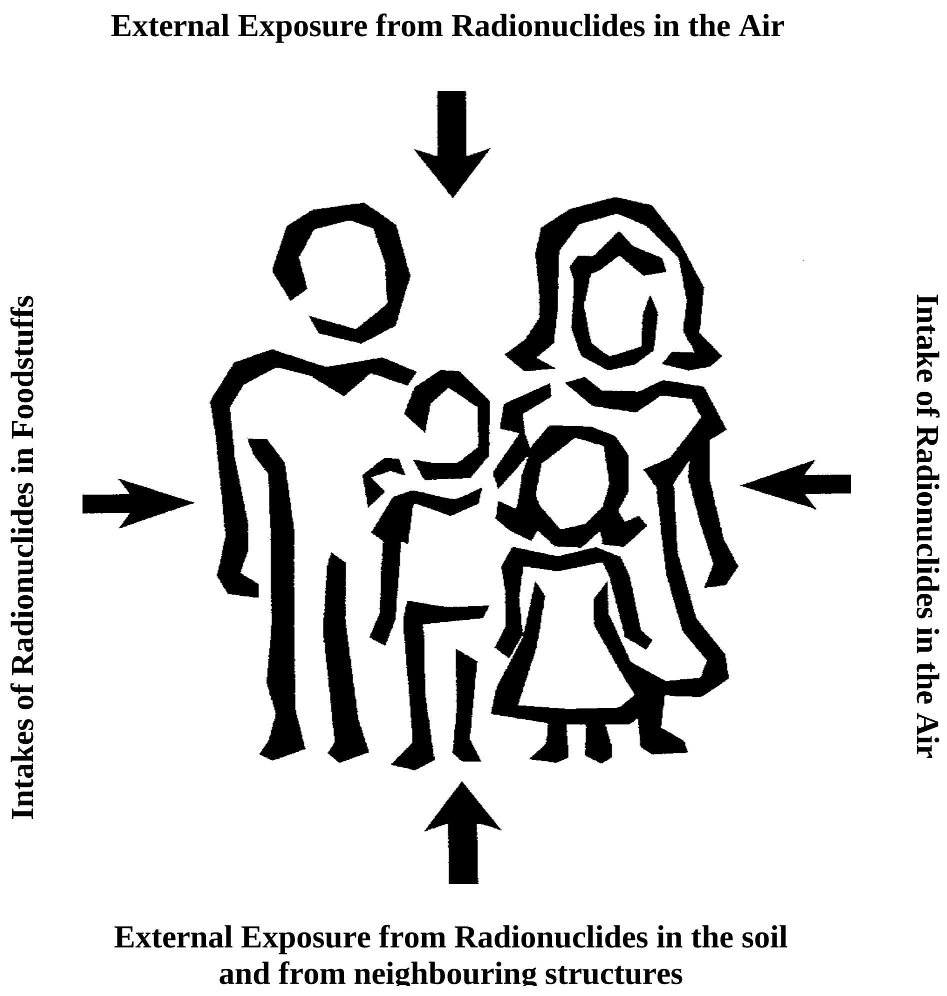


Figure 8: Potential radionuclide exposure pathways for humans.

2.22.1 Radioactivity through the Environment

The pathways of radioactive materials from the environment through atmosphere, water and food chain to the human body are described below.

2.22.2 Atmosphere

Radionuclides can be released into the air by human activities. They can also be created in the atmosphere by natural processes such as the interaction of cosmic radiation with nitrogen to produce radioactive carbon-14. Radionuclides can be removed from the air in several ways. Particles settle out of the atmosphere if air currents cannot keep them suspended. Rain or snow can also remove them.

When these particles from the atmosphere may find their way in water, on soil, or on the surfaces of living and non-living things. The particles may return to the atmosphere by re-suspension, which occurs when wind or some other natural or human activity generates clouds of dust containing radionuclides.

2.22.3 Water

Radionuclides can get into water bodies in several ways. They may be deposited from the air, as described above. They may also be released to the water from the ground through erosion, seepage, or human activities such as mining.

Some radionuclides that reach either groundwater or surface water move with the water. Others are deposited on the surrounding soils or rocks. One important factor affecting their movement is how thoroughly they dissolve in water (solubility). Another factor affecting their movement is the ability of radionuclides to adhere to the surfaces of rocks or soil through which the water flows.

2.22.4 Food Chain

Radionuclides in water or air may enter the food chain. For example, plants are capable of absorbing radionuclides from water in the same way as other minerals are absorbed. When animals drink water some of the radionuclides in the water remain in their bodies. Radionuclides from the air may settle on the surface of plants. When animals eat the plants, they ingest the radionuclides that have settled from the air or have been absorbed from the water. Plants and animals that will eventually become food for people thus provide a pathway for radionuclides from the environment to reach man.

2.22.5 Plants

Plants are the primary recipients of radioactive contamination to the food chain following atmospheric releases of radionuclides. Vegetation may be subjected to direct and indirect contamination.

The direct contamination of terrestrial vegetation refers to the deposition of radioactive materials from the atmosphere onto the plants. Indirect contamination refers to the sorption of radionuclides from the soil by the root system of plants.

2.22.6 Animals

Secondary recipients of food chain contamination are animals that consume plants or other animals. Several important pathways for the transfer of radionuclides to humans involve animal food chains, including milk and eggs from living animals and meat or flesh from animals and fish. Depending on the radionuclide and the metabolism in the organism, the concentrations may be enhanced or reduced compared with the earlier steps of the food chain. Some parts of the animal are not consumed, e.g. bones, shells, skin and feathers, and this prevents the transfer from animal products of bone seeking radionuclides such as ^{90}Sr and plutonium. Bone tissue might, however, re-enter the food chain as bone meal in various fodder products, and it might also appear in fertilizers.

The main animal pathway to humans of radiologically important radionuclides such as ^{90}Sr, ^{131}I and ^{137}Cs is milk consumption. All three radionuclides are readily transferred from animal fodder to the milk. Cesium is transferred with its chemical congener potassium to the soft tissues of animals, particularly muscle. Strontium is preferentially transferred to bone, like its congener calcium.

Fish and shellfish receive radionuclides both directly from the water and from their food. Some radionuclides that are of no concern in the terrestrial animal food chains may be concentrated in aquatic animals. This is the case, for example, for polonium, which is concentrated in crustaceans, and for polonium in fish and seafood.

2.23 Pathways into the Human body

The radionuclides present in our environment can give both internal and external doses. Internal dose is received as a result of the intake of radionuclides. The major routes of intake of radionuclides are ingestion, inhalation and absorption through the skin.

2.23.1 Ingestion

Anything that people eat or drink contains radionuclides. The common radionuclides are ^{40}K, ^{226}Ra and ^{238}U and the associated progenies. Ingestion includes the intake of the radionuclides from drinking milk and water, and consumption of food products. Some radionuclides are intentionally ingested as part of a medical therapy or diagnostic procedure. Some of the radionuclides people ingest can remain in the body for long period of time while others are quickly eliminated.

2.23.2 Inhalation

Inhalation includes the intake of radionuclides through breathing dust particles containing radioactive materials. Radionuclides suspended in the atmosphere can enter human lungs. Some radioactive particles are exhale, and some remain in the lungs where the radiation they release immediately strikes the lung tissue.

2.23.3 Through the Skin

Radionuclides may be absorbed through the skin's surface, or may enter the body through a break in the skin. Another pathway is through the injection of radionuclides as part of medical therapy.

2.24 Accumulation of Radioactivity in Critical Organs

Radionuclides enter the body by ingestion, inhalation or injection. Once taken into the body, their radiation effects depend on their anatomic distribution, duration of retention in the body, and rate of radioactive decay, as well as on the energies of the emitted radiations.

An internally deposited radioactive element may concentrate in and thus irradiate certain organs more than others. Evidence shows that iodine is chiefly accumulated in human thyroid, thorium in lungs, liver and skeleton tissue, uranium in lungs and kidneys, cesium and potassium in muscles, radium and strontium in skeletal tissues.

Different radioelements also vary in their rates of removal from the body. Radioiodine, for instance, is normally eliminated from the thyroid rapidly enough so that its concentration is halved within days. Strontium-90, on the other hand, is retained in high concentrations in the skeleton for years.

2.25 Measurement of Radiation

Human senses cannot feel radiation exposure but it can be detected through the ionization or electrical charge produced which relates to the dose received. Exposure is the total electrical charge of ions of one sign produced in per unit mass of irradiated air at normal temperature and pressure (NTP) by X- or γ-rays. The unit of exposure is coulomb per kilogram.

Ionization of radiations are also quantified in terms of absorbed dose, which is defined as the amount of energy deposited in a unit mass of material by different types of radiation (α, β, γ-rays, x-rays and neutrons). It is measured in Gray (Gy) and is equivalent to one joule per kg.

Interaction of these radiations in biological material and production of damage by same amount of absorbed energy is not the same. A dose of one Gray to tissue from alpha radiation is more harmful than one Gray of beta radiation. Therefore, measure of degree of harm or damage on equal scale from different types of radiation is expressed in terms of equivalent doses and is measured in Sievert (Sv).

2.26 Dose Assessment Methodology

The term exposure applies to ionization of air by X- or γ-rays, but the more common usage is absorbed dose (D). This is a fundamental dosimetric quantity used for radiation assessment in human. However, it is not satisfactory for radiation protection purposes because effectiveness in damaging human tissue differ for different types of ionizing radiation. Consequently, the absorbed dose averaged over a tissue or organ is multiplied by a radiation weighting factor to take account of the effectiveness of the given type of radiation in inducing health effects; the resulting quantity is termed equivalent dose, H_T.

$$H_T = \sum_R W_R \, D_{T,R} \tag{2.13}$$

where $D_{T,R}$ is the mean absorbed dose in tissue or organ T due to radiation type R and W_R is a radiation weighting factor. Values of radiation weighting factors (W_R) recommended by ICRP for the various types or energies of radiation incident on the body or radiation emitted from within the body for selected tissues or organs are listed in Table 2.11 (ICRP, 1977).

Table 2.11: Radiation Weighting Factors (W_R) for different types of Radiations

Types of Radiations	Energy Range	W_R
Photons, Electrons, Muons	All energies	1
Protons	>2 MeV	5
Neutrons	<10 keV>20 MeV 10-100 keV, 2-20 MeV >0.1-2 MeV	5 10 20
Alpha particles, Fission fragments, Heavy nuclei	All energies	20

The equivalent dose is used when individual organs or tissues are irradiated, but the likelihood of biological harm due to a given equivalent dose differs for different organs and tissues. Consequently, the equivalent dose to each organ and tissue is multiplied by a tissue weighting factor to take account of the organ's radio-sensitivity.

The sum of such weighted equivalent doses for all exposed tissues in an individual is termed as effective dose and is measured in Sv. The effective dose also takes account of energy and type of radiation and gives a broad indication of the health detriment. Moreover, it applies equally to external and internal exposure and for uniform or non-uniform irradiation.

The term effective dose is defined by the quantity, E. It is a summation of the tissue equivalent doses (H_T), each multiplied by the appropriate tissue weighting factor (W_T).

$$E = \sum_T W_T \sum_R W_R D_{T.R} \qquad (2.14)$$

To assess health detriment arising from the irradiation of various organs and tissues, ICRP recommended values of W_T for a reference population of both sexes are given in Table 2.12. These values apply to radiation workers and general public of either sex (UNSCEAR, 2000).

When radionuclides are taken into the body, the resulting dose is received throughout the period of time during which they remain in the body and this is referred to as committed dose. It represents total dose delivered during this period of time, and is calculated as specified time integral of the rate of receipt of the dose.

Table 2.12: ICRP Tissue Weighting Factors (W_T) (ICRP, 2007)

Tissue or Organ	**Weighting Factor (W_T)**
Gonads	0.20
Breast	0.05
Colon	0.12
Red bone marrow	0.12
Lungs	0.12
Stomach	0.12
Urinary bladder	0.05
Liver	0.05
Oesophagus	0.05
Thyroid	0.05
Bone surface	0.01
Skin	0.01
Remainder	0.05[a,b]

Note:

(a) The remainder is composed of the following tissue and organs: adrenals, brain, and extra thoracic region of the respiratory tract, small intestine, kidney, muscle, pancreas, spleen, thymus and uterus.

(b) The value 0.05 is applied to the average dose to the remainder tissue group. However, when the most exposed remainder tissue or organ receive the highest committed equivalent dose of all organs, a weighting factor of 0.025 is applied to that organ and a weighting factor of 0.025 is applied to the average dose in the rest of the remainder.

To compare doses delivered over different time periods, the concept of the dose commitment is introduced as the dose commitment, $H_{C.T}$ or E_C, is defined as the time integral of the average individual dose rate (per caput dose rate) delivered as a result of a specific practice:

$$H_{C.T} = \int_0^{\infty} H_T \, dt \qquad (2.15)$$

or

$$E_C = \int_0^{\infty} E(t)dt \tag{2.16}$$

The integral is taken over infinite time to account for exposures occurring during all future time and may thus involve the average individual dose rates over generations. The dose commitment from one year of a practice is numerically equal to the equilibrium dose rate, if the practice continues indefinitely at constant rate.

When prolonged exposure to a single individual from a single intake of a radionuclide is being considered, committed dose quantities are used. The time distributions of the absorbed dose rates vary with the radionuclides, their form, mode of intake, and bio kinetic behaviour. The committed equivalent dose, $H_T(\pi)$, is defined as the time integral of the equivalent dose rate, where π is the integration time in years.

$$H_i = \int_i^{i-\pi} H_T\,(t)dt \tag{2.17}$$

The value of π is taken to be 50 years for adults from time of intake and 70 years for children. The committed effective dose $E(\pi)$, is the sum of the committed equivalent doses to tissues and organs multiplied by the appropriate tissue weighting factors, W_T.

To determine aggregate quantities of doses to a population size, the ICRP has also used collective dose quantities. The collective equivalent dose, S_T, is the average equivalent dose in an exposed group of individuals multiplied by the number of individuals in each group:

$$S_T = \sum_i H_{T,i} N_i \tag{2.18}$$

where N_i is the number of individuals in a population subgroup i receiving mean organ equivalent dose $H_{T,i}$.

The collective effective dose is defined in a similar manner, i.e. the population and the time period over which the dose is determined. The collective dose commitment may become rather uncertain if applied to very long time periods in which future environmental conditions and the populations affected cannot be reasonably anticipated.

CHAPTER THREE

3.0 MATERIALS AND METHODS

The facilities and methods used in the radioactivity measurements are described in this section. Sampling and sample preparation techniques are described. Sample analysis using gamma spectroscopy are described. In general, the goal of gamma spectroscopy is to derive nuclide-specific gamma emission rates of the sample (in activity units, such as Becquerel (Bq) or decays per second) from the spectral data. A brief description of the study area is also presented.

3.1 Study Area

Ghana is Africa's second biggest producer of gold, and hosts more estimated reserves than some prolific gold producing nations. Ghana's oldest mine is in Tarkwa and operated by Goldfields Ghana Limited (Goldfields, 2008a).

The study area is the Nzema Gold Mine located in the town of Nkroful in the Nzema East Municipality of Ellembelle District of the Western Region of Ghana and operated by Adamus Resources Limited. Adamus Resources Limited is focused on advancing the Nzema gold project which consists of a contiguous block of tenements and options covering approximately 665 square kilometres. Adamus' area hosts both the Salman and Anwia deposits. The company reports that current project estimates indicate a minimum life of 10 years and average production potential of 100,000 ounces of gold per year. The study area is shown in figure 9

(Source: Ghana Exploration **http://www.endeavourmining.com/s/Ghana.asp**).

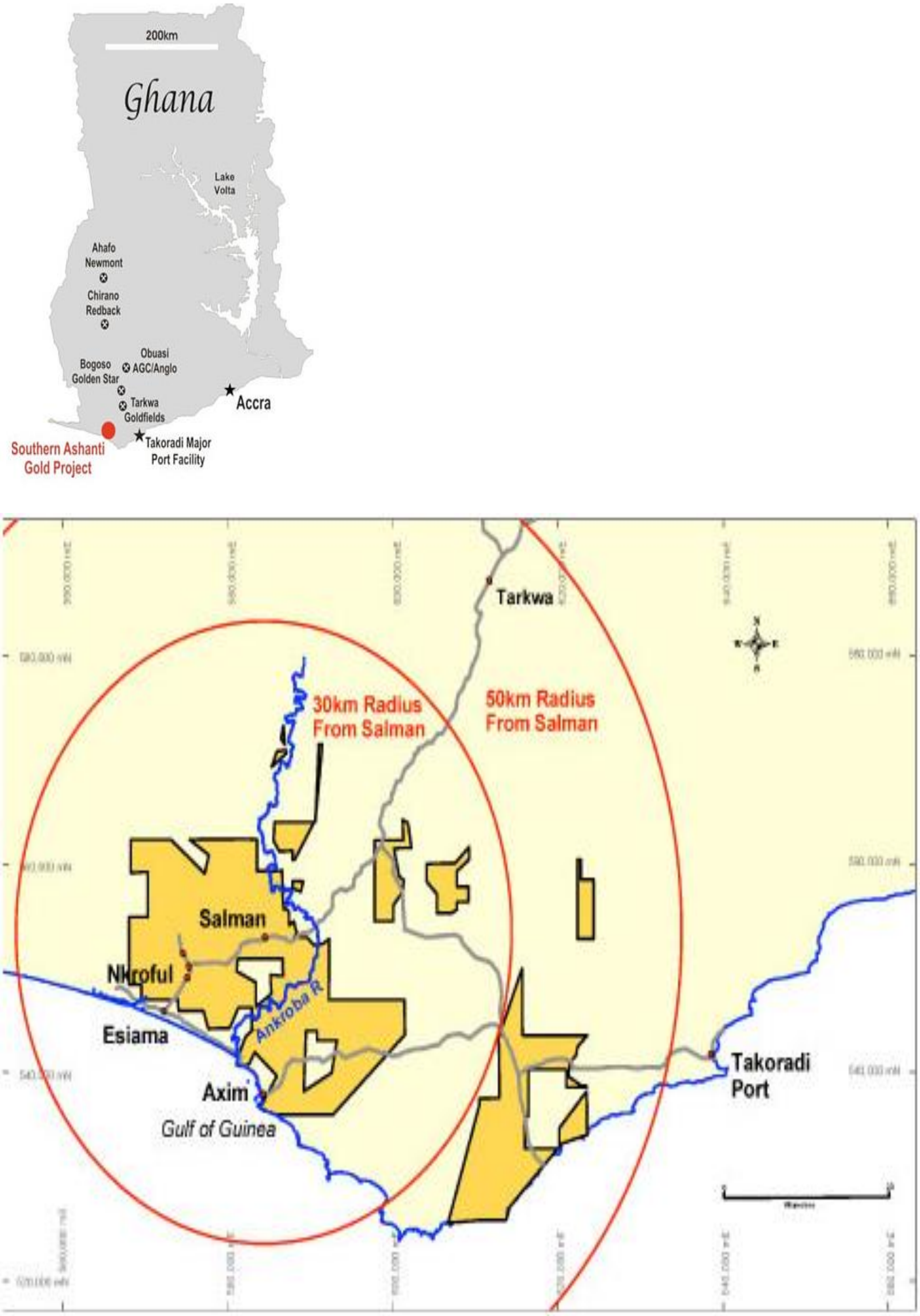


Figure 9: Map of Ghana showing the study area
(http://www.endeavourmining.com/s/Ghana.asp)

3.1.1 Location

Nkroful in the Nzema East Municipality is bordered on the west by the Jomoro District, to the north by the Wasa Amenfi West District, to the east by Wassa West and Ahanta West Districts and to the south by the Gulf of Guinea. The district capital is Axim. It is located on the shores of the Gulf of Guinea and is 81 kilometres from Takoradi, the capital of the Western Region of Ghana. The Nzema East Municipality covers an area of about 2,194 square kilometres representing 9.8 percent of the total area of the Western Region (Ghana Statistical Service, 2014).

The Nzema Gold Project is located on the southern end of the Ashanti Gold Belt in south-western Ghana, West Africa. The Nzema Gold Project is centred on latitude 5º00'N and longitude 2º14'W, approximately 280 km west of Ghana's capital city, Accra. The Nzema Gold Project consists of tenements and options held by Adamus' Ghanaian subsidiaries covering approximately 665 km^2, together with a processing facility and associated infrastructure to mine and process ore. Adamus' Nzema Gold Project includes a series of open pits located along an 8 km trend of the Ashanti Gold Belt. Significant deposits (from north to south) include Akango, Salman North, Teberu, Nugget Hill, Salman Central and Salman South (Ghana Exploration**:** **http://www.endeavourmining.com/s/Ghana.asp**).

According to the year 2010 Population census report (Ghana Statistical Service, 2014), the current population of Nzema East Municipality is 60,828 constituting 2.6% of the regions´s population. The average population density is 99.3 persons per square kilometre and a growth rate of 2.5% per annum. The rapid population growth is due to expansion in mining activities of Adamus Resources Limited and the influx of small-scale mining operations (Galamsey) in the district and subsequently migration of people especially from the rural areas. The migrants are predominantly young people looking for jobs at the mines. The Adamus Resources Limited employs approximately 275 full time workers including both expatriates and local employees, plus 661 contractors, for a total of 936 operational workers.

3.1.2 Geology

The geological map of Ghana (figure 10) falls on the western portion of the main shield of the West African Craton. The main shields are further divided into the Archean and the early Proterozoic units. The Archean is restricted to the Liberian Craton and constitutes highly metamorphosed gneiss older than 2.7 Ga. The early Proterozoic terrain represents large sedimentary basins and linear or acute volcanic belts corresponding to the period of accretion around 2.1 Ga (Abouchami *et al*, 1990). The main constituent of the Proterozoic rocks in Ghana are mostly underlain by Cambrian Rocks of the Birimian formation, the Tarkwaian sandstone, the Dahomeyan systems, the Togo series and the Buem formation. Several granite bodies intrude into the Birimean formation of which Nzema Gold Mine is located.

Since 2003, Adamus has focused on developing and expanding the economic potential of the Nzema Gold Project which is Adamus' principal asset. The gold deposits occur mainly in the Proterozoic Birimian formation of Ghana. The Birimian formation is characterized by NE-SW striking gold belts occurring along boundaries of metavolcanic belts and metasedimentary basins (Ghana Exploration: **http://www.endeavourmining.com/s/Ghana.asp)**. The gold ore is mainly associated with sulphide minerals in the form of arsenopyrites, pyrites and pyrrhotite and, to a minor extent, sphaderite and tetrahedrite.

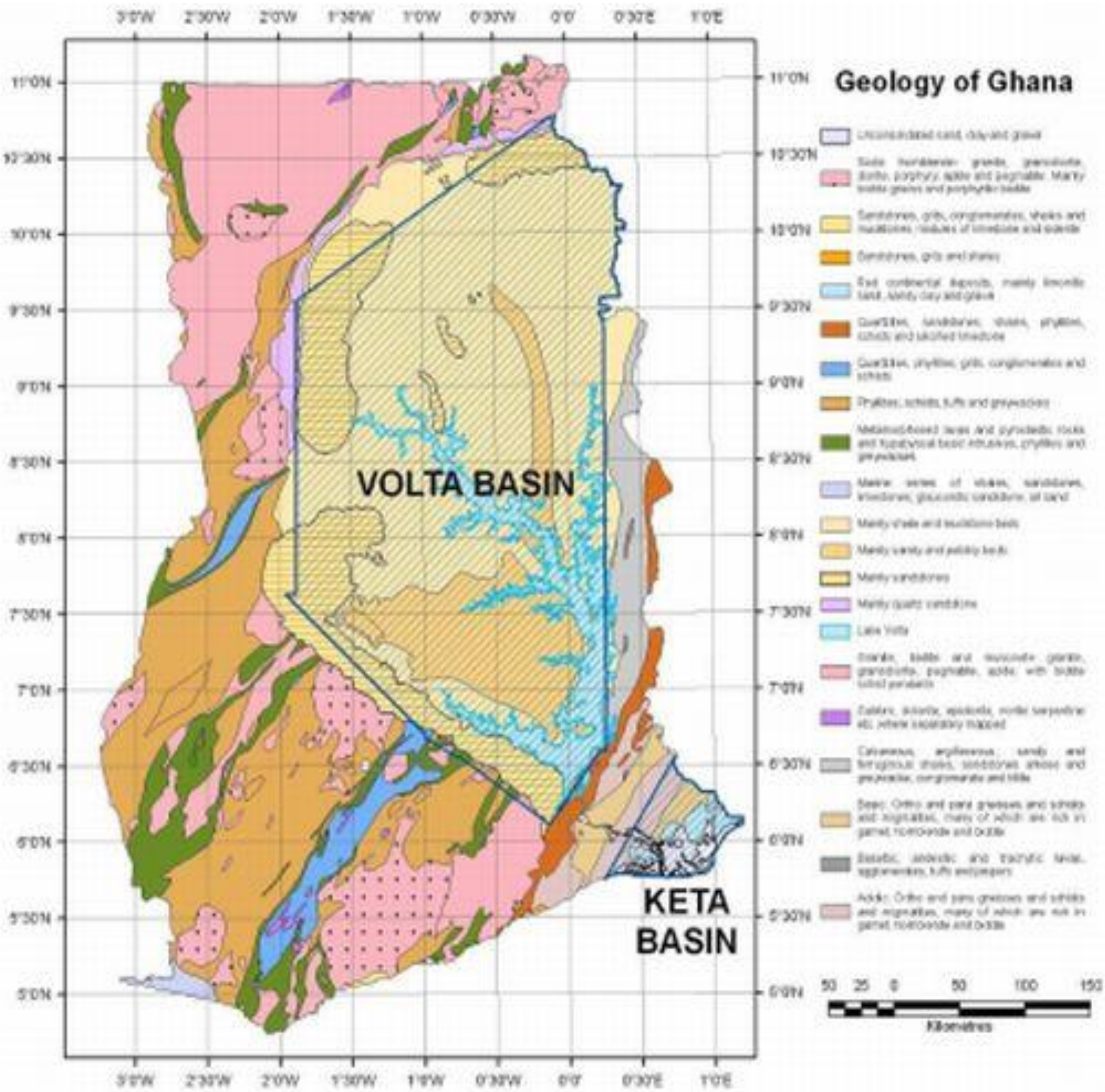


Figure 10: Geological Map of Ghana (Ghana Exploration: **http://www.endeavourmining.com/s/Ghana.asp)**

3.1.3 Climate and Vegetation

The Nzema East Municipality lies between the wet semi-equatorial climate zone of the West African Sub-region. Rainfall is experienced throughout the year with the highest monthly mean occurring around May and July each year. The average temperature in the Municipality is about 29.4 ^{o}C with temperatures ranging between 25 and 30 ^{o}C throughout the year. The vegetation of the Municipality is made up of the moist semi-deciduous rain forest mainly in the northern part, followed by secondary forest southwards mainly due to human activities like tree felling and farming and coastal savannah mainly in the south along the 30 km coastal belt (Ghana Statistical Service, 2014).

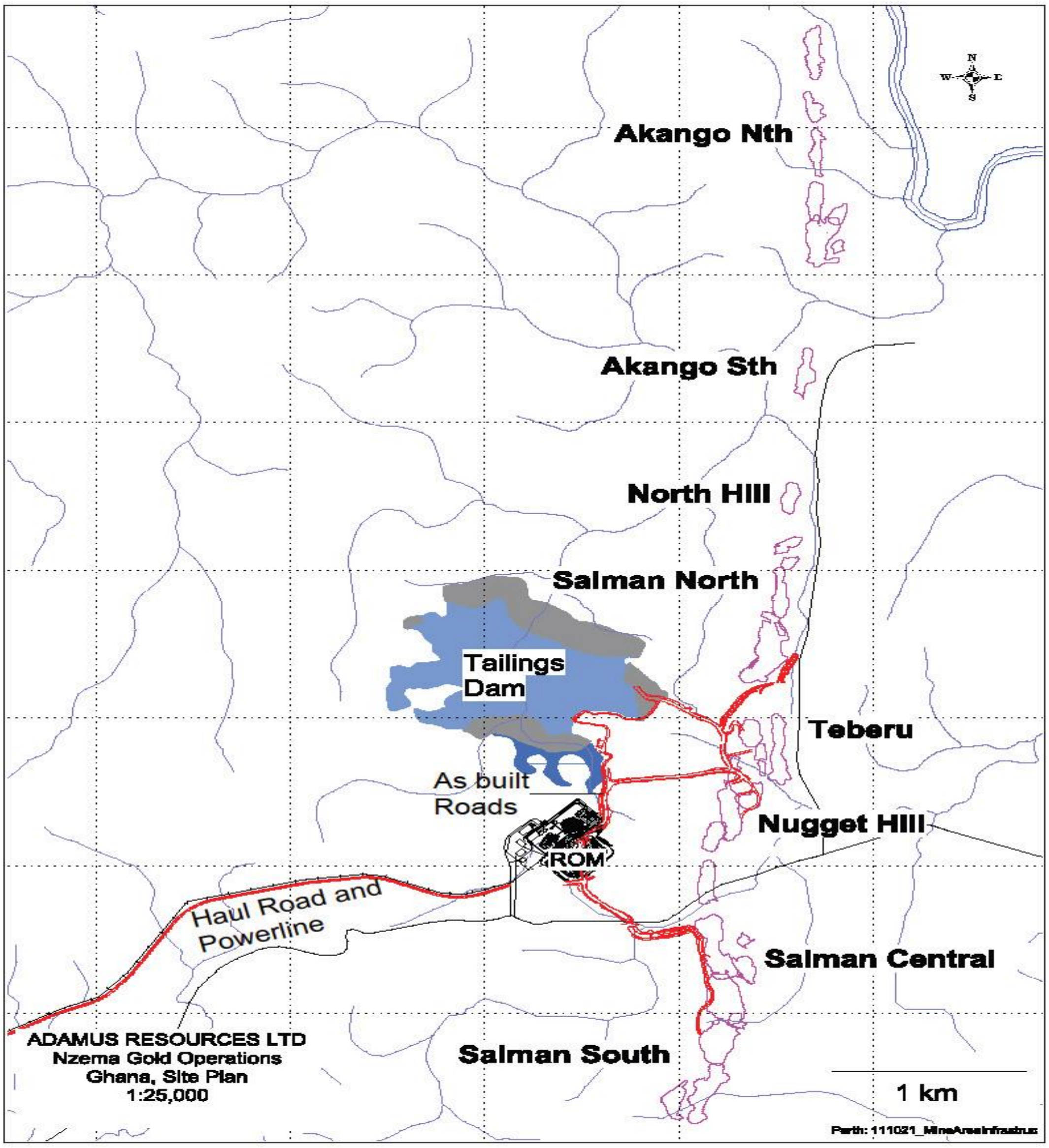


Figure 11: Map of study area showing sampling locations (Ghana Exploration **http://www.endeavourmining.com/s/Ghana.asp)**.

3.2 Sampling and Sample Preparation

3.2.1 Soil sampling

Soil samples were collected from the following locations within the mine and the surrounding communities including; Salman Town, Akango, Teberu, Aliku Town, Nkroful Bokrobo, North Hill, Nugget Hill, Angajeleh. In order to ensure representative samples were taken from the area for the analysis, initial survey was carried out in the area to determine the sampling points. The selection of the sampling locations was based on the accessibility to the public and proximity to the mine. In addition, the geological map of the area (figure 10) was used to identify the locations (figure 11) where samples will be taken. Based on these criteria, 9 locations were identified for the soil samples and 16 water sample sources. In the case of food crop samples, only food crops which were ready for harvesting was the criteria adopted. At the times of sampling, 6 farms which had cassava, cocoyam, maize and plantain crops ready for harvesting were sampled for analysis. The sampling locations were marked using a Geographical Positioning System (GPS), Geo Explorer II. Plates 1 and 2 show some of the locations within the mines where soil sampling was carried out.

PLATE 1: Plant site in the study area

PLATE 2: Mining site in the study area

The sampling strategy that was adopted for the soil samples was random (ASTM, 1983, 1986; IAEA, 2004). At each identified location samples were arbitrary collected within defined boundaries of the area of concern. Each location was divided into 50 m x 50 m grids and samples taken at different points and mixed together to give a sample. Each sampling point was selected independent of the location of all other sampling points. By this approach all locations within the area of concern had equal chance of being selected. The soil samples were taken using a coring tool to a depth of 5-10 cm. At each sampling location, samples of soil were taken from at least five different sections of the area into labelled plastic bags. One kilogram (1 kg) of each sample was collected for analysis. Sampling was carried out from 23/04/2012 to 28/4/2012 and 5/08/2013 to 10/08/2013. A total of 72 soil samples were collected from the 9 locations within the mine and the surrounding villages. The samples were transported to the laboratory of the Radiation Protection Institute (RPI) of Ghana Atomic Energy Commission (GAEC) at Kwabenya for preparation and analysis.

3.2.2 Water sampling

The water samples were taken from water sources within the Goldmine and surrounding communities. These include mine waste water, rivers, streams, boreholes and wells. The samples were collected into labelled two and half litres (2.5 L) plastic bottles. In order to maintain the homogeneity of the samples and to avoid adsorption of radionuclides on the walls of the container, all samples were acidified with 0.1N nitric acid (HNO_3). Prior to collecting groundwater samples, the wells were purged to remove any stagnant water in the well casing or gravel pack and ensure that at least 95% of the water sample originate from the aquifer formation being sampled. The sample collection occurred immediately after purging. The bottles were rinsed twice with sample water before filling with the final sample. The water samples were then transported to the Laboratory and stored in a fridge prior to preparation and analysis. A total of 16 water samples were collected. Plates 3 and 4 show some of the locations within the mines and the communities where water sampling was carried out.

PLATE 3: Pool of water within plant site in the study area

PLATE 4: Borehole in one of the communities within the study area

3.2.3 Food sampling

The average farm size is estimated to be 4 hectares. Slash and burn is the common practice of land preparation in the Municipality. Food crops such as cassava, maize, cocoyam and plantain are grown extensively both for subsistence and for sale to the public. Therefore, it is important to analyse the food crops grown in the study area by means of their radionuclide concentrations so that effective dose equivalent to humans consuming these food crops can be estimated.

In the study, four (4) different food samples including cassava, cocoyam, maize and plantain collected from the 6 farms were investigated. About 2 kg of each sample were taken from different farms and transported in polyethylene bags to the laboratory for analysis. A total of 24 food crop samples were from 6 different farms for radioactivity analysis.

3.3 Sample preparation for direct gamma spectrometry analysis.

3.3.1 Soil samples

At the laboratory, the samples were air dried in trays for 7 days and then oven dried at a temperature of 105 ^{o}C for between 3-4 hours until the samples were well dried with a constant weight (IAEA, 1989). The samples were then ground into a fine powder using a ball mill grinder and sieved through a 2 mm pore size mesh into a previously weighed one (1) litre Marinelli beaker (Plate 5). The Marinelli beakers with its content were then weighed again to determine the weight of the sample. The beakers were covered and sealed with a paper tape to prevent the escape of the gaseous radionuclides in the sample. The samples were stored for 50 days to allow for secular equilibrium between the long-lived parent radionuclide and their short-lived daughter radionuclides in the ^{238}U and ^{232}Th decay series and counted on a high purity germanium (HPGE) detector for 72000 seconds. The activity concentrations of the radionuclides of interest in the samples were reported on dry weight basis in Bq kg^{-1}

PLATE 5: Marinelli beaker used for sample counting

3.3.2 Water samples

The water samples were prepared into the one (1) Litre Marinelli beaker after filtration to remove all solid particles in the water. The samples were counted on a gamma detector (High Purity Germanium detector) for 72000 seconds. The activity concentrations of the radionuclides in the sample were reported in Bq L^{-1}.

3.3.3 Food crop samples

In the laboratory, the food crop samples were thoroughly cleaned, peeled when necessary and the edible portion chopped into smaller pieces and air dried for about a week. After that, they were oven dried at 80 °C for approximately 16 hours and prepared into powder form for analysis (Santos *et al*, 2002). The powdered samples were sieved through a 2 mm mesh and transferred to a 1 Litre Marinelli beaker and the dry weight of the sample determined (Alberto *et al*, 1995). The beakers were then sealed. The samples were counted on a HPGE detector for 72000 s and the net counts of full energy events used to determine the activity concentrations of ^{226}Ra (^{238}U), ^{228}Th and ^{40}K in Bq kg^{-1}.

3.4 Description of the Gamma Spectrometry system used for analysis

Gamma spectrometry is a rapid and convenient method for the identification and quantitative analysis of gamma emitting radionuclides (IAEA, 1989). This is a non-destructive radiometric technique usually used for the direct measurement of gamma emitting radionuclides through the discrete gamma energy lines/spectrum of various radionuclides. The instrumentation generally consists of a semiconductor detector, associated electronics and a computer-based multi-channel analyser (MCA). Usually one or more hyper pure or intrinsic germanium (HPGe) detectors based gamma-ray spectrometer (Plate 6a) operate at liquid nitrogen temperatures (-195^{o}C), by mounting the germanium crystal in a vacuum cryostat, thermally connected to a copper rod or "cold finger".

The gamma-spectrometry system used for this work consists of a coaxial HPGe detector which is useful for measurement of gamma rays with energies over the range of about 60 keV to 3.0 MeV (3000 keV), pre-amplifier, detector bias supply, linear amplifier, analog-to-digital converter (ADC), multi-channel storage of the spectrum, and data readout devices (ANSI, 1999).

To determine ^{232}Th and ^{238}U through their gamma emitting progenies, soil and food samples were packed in plastic containers, hermetically sealed and stored for 50 days prior to analysis to attain secular equilibrium between the long-lived parent and short-lived daughter products of thorium and uranium series (IAEA, 1989; Selvasekarapandian et al., 2000).

The activity of thorium and uranium was measured using a PC-based gamma spectrometry system consisting of HPGe coaxial type detector coupled to PC-based MCA acquisition board (Canberra S100). Relative efficiency of the detector was 30%, while resolution (FWHM) of the spectrum for the energy peak of ^{60}Co at 1332.5 keV was 2 keV and peak to Compton ratio was 54:1. To reduce background radiation, the system was shielded with lead blocks of 5 cm and internal cavity of 55 cm^3. The inner lining of 2 mm thick copper was followed by 2mm thick aluminium also used to absorb X-rays originating from the lead and copper (Plate 6b). The results were analysed using Windows based Aptec software (Canberra Company, 2006).

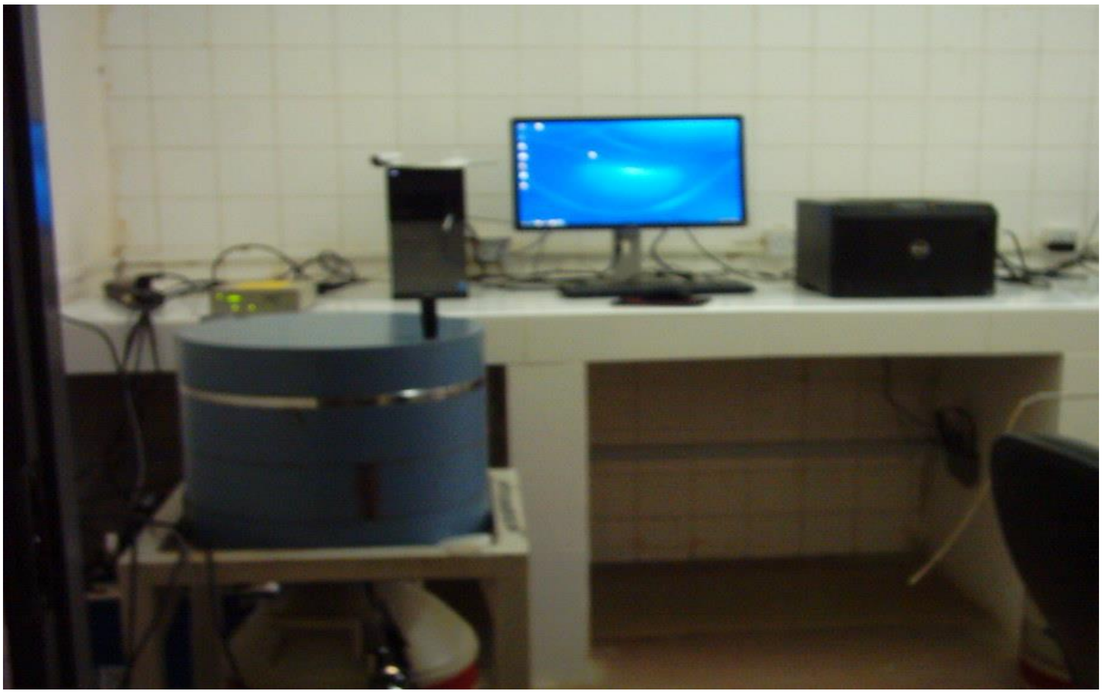


PLATE 6a: High Pure Germanium Detector System.

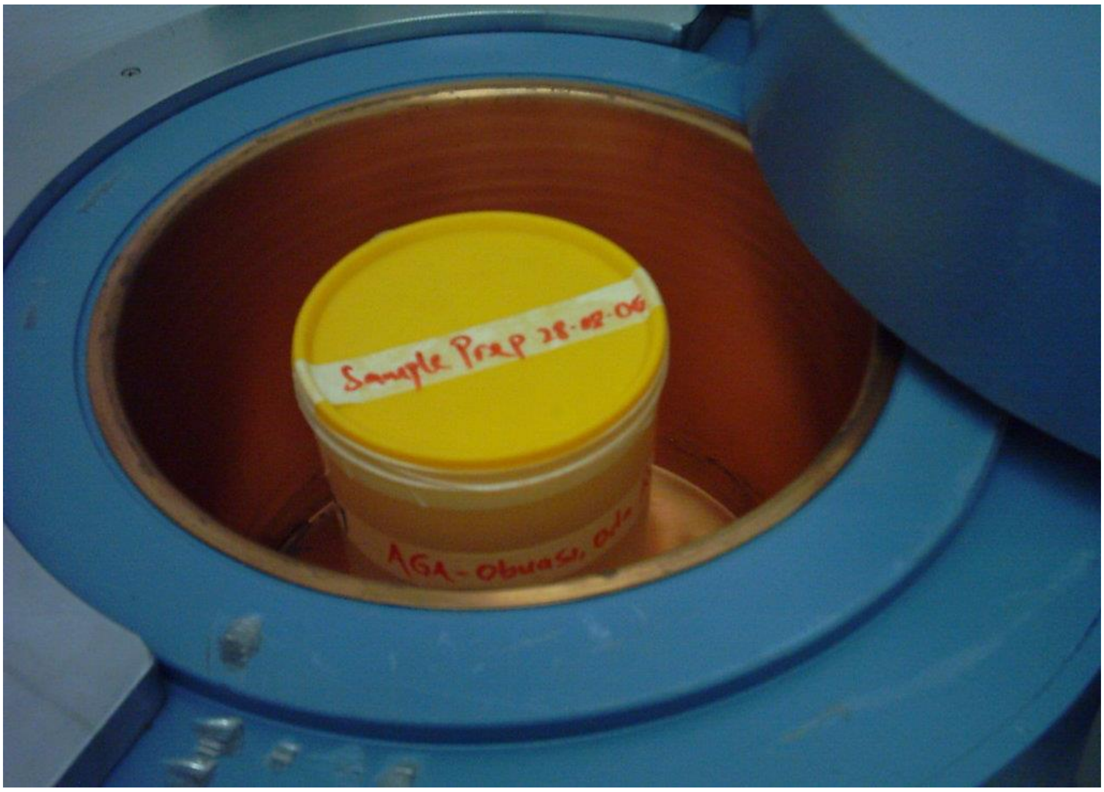


PLATE 6b: A Marinelli beaker on a detector.

3.5 Calibration of Germanium Detector System

The system was calibrated for the working range of 57-1674 keV using IAEA NW 146 gamma standard. Calibration ensures that gamma-ray spectra are accurately interpreted in terms of energy and specific activity (per unit mass or litre, e.g., Bq kg^{-1} or Bq l^{-1}).

3.5.1 Energy Calibration

Energy calibration was performed with reference to the spectrum of a standard reference source with known full-peak energies to show the relationship between channel number of the spectrometer and the energy of the radionuclides in a reference material. The standard radionuclide sources need only be counted long enough to identify the peak energies in the spectrum (Gilmore and Hemingway, 1995). The energy calibration was carried out for a 1.0 litre Marinelli beaker using a mixed twelve radionuclides gamma standard, the IAEA NW 146. The gamma standard contained the following radionuclides with the corresponding energies as shown in Table 3.1. The standard was obtained from Physikalisch Technische Bundesanstalt PTB, Germany. The standard was contained in the same type of plastic container as used for measuring the samples.

Table 3.1: Radionuclides used for efficiency calibration (IAEA NW 146)

Nuclides	Energy (keV)	Gamma rays/s	Activity Concentration (Bq)	Uncertainty (%)	Emission rate (s^{-})	Half-life time (d)
Am-241	59.54	1128	2.97×10^3	2.6	1.06×10^3	158047
Cd-109	88.03	647	1.69×10^4	6.2	6.14×10^2	462.6
Co-57	122.1	588	8.84×10^2	1.5	7.57×10^2	271.4
Ce-139	165.9	663	9.66×10^2	1.7	7.71×10^2	137.64
Hg-203	279.2	1942	2.56×10^3	1.4	2.09×10^3	46.6
Zn-113	391.7	2085	3.18×10^3	4.0	2.07×10^3	115.09
Sr-85	514	3863	3.89×10^3	2.5	3.83×10^3	64.84
Cs-137	661.6	2435	2.78×10^3	2.0	2.36×10^3	10958
Y-88	898	6231	6.62×10^3	1.8	6.22×10^3	106.6
Co-60	1173	3350	3.40×10^3	1.5	3.40×10^3	1924.9
Co-60	1333	3353	3.40×10^3	1.5	3.40×10^3	1924.9
Y-88	1836	6586	6.62×10^3	1.6	6.57×10^3	106.61

The energy calibration was performed by matching the principal gamma rays observed in the spectrum of the standard to the channel numbers. The equation relating the energy to the channel number is given by the expression (Gilmore and Hemingway, 1995).

$$E\gamma = A_o + A_1 * CN \qquad (3.1)$$

where, Eγ is the gamma energy of a given radionuclide, CN is the channel number for a given radionuclide, and A_o and A_1 are calibration constants for a given geometry. A graph of energy against channel was plotted as shown in figure 12.

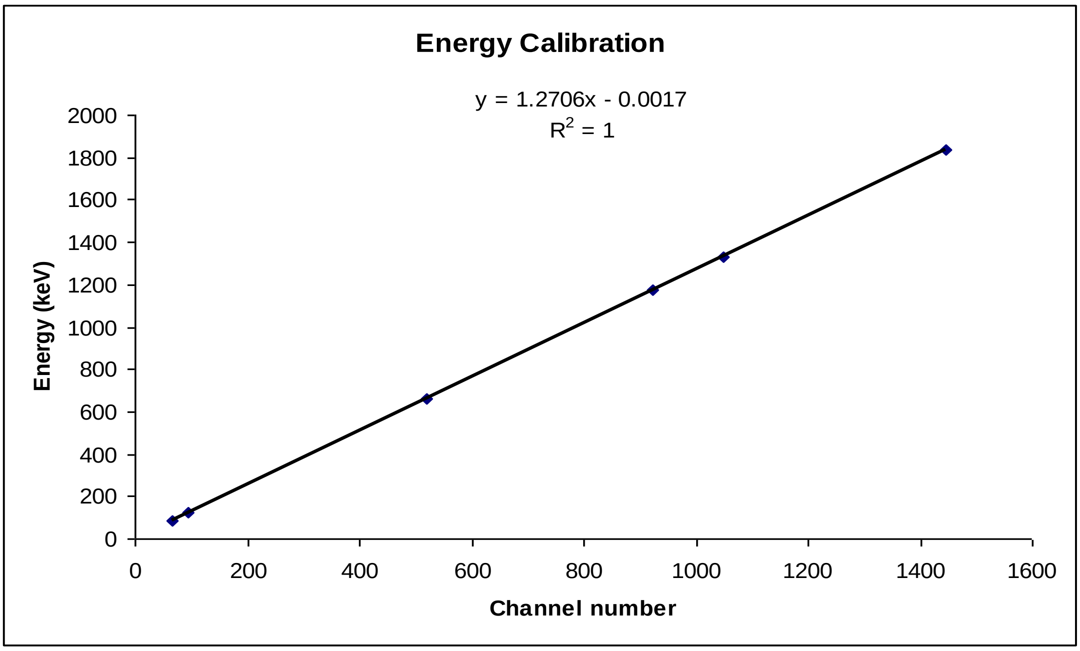


Figure 12: Energy calibration curve using mixed radionuclides standard in one litre Marinelli beaker.

3.5.2 Efficiency Calibration

Efficiency calibration is the relationship between the peak area in the spectrum and the amount of radioactivity it represents. It is the peak area in relation to the number of gamma-rays emitted by the source depending upon the geometrical arrangement of the source and the detector. It also determines the relationship between number of counts and disintegration rate. For efficiency calibration estimation, it is essential that the radionuclides gamma-ray emission probabilities are accurately known, implying that the calibration source should have known activities. Calibration sources should be prepared to have identical geometries and density as the real samples which they are to be compared with (Gilmore and Hemingway, 1995).

The efficiency calibration was performed by acquiring a spectrum of the standard until the count rate of the total absorption can be calculated with a statistical uncertainty of less than 1%. The net count rate was determined at the photo peak for all the energies to be used for the calculation of the efficiency. The efficiency was then related to the count rate and the activity of the standard by the relation (Darko et al., 2010):

$$\varepsilon(E_{\gamma}) = \frac{N}{(A * P * t)} \tag{3.2}$$

where N is the full energy peak net count corresponding to the gamma photons with energy E_{γ} and gamma emission probability P, A is the activity of the source and t is the counting time. The efficiency is related to the energy by the expression (Darko et al., 2010).

$$\ln \varepsilon(E_{\gamma}) = a_o + a_1 * (\ln E_{\gamma})^1 + a^2 * (\ln E_{\gamma})^2 \tag{3.3}$$

where a_o, a_1, a_2 are calibration constants for a given geometry and the other symbols have the usual meaning given earlier in the passage. The full energy peak efficiency as a function of gamma ray energy for a typical HPGe detector is shown in figure 13.

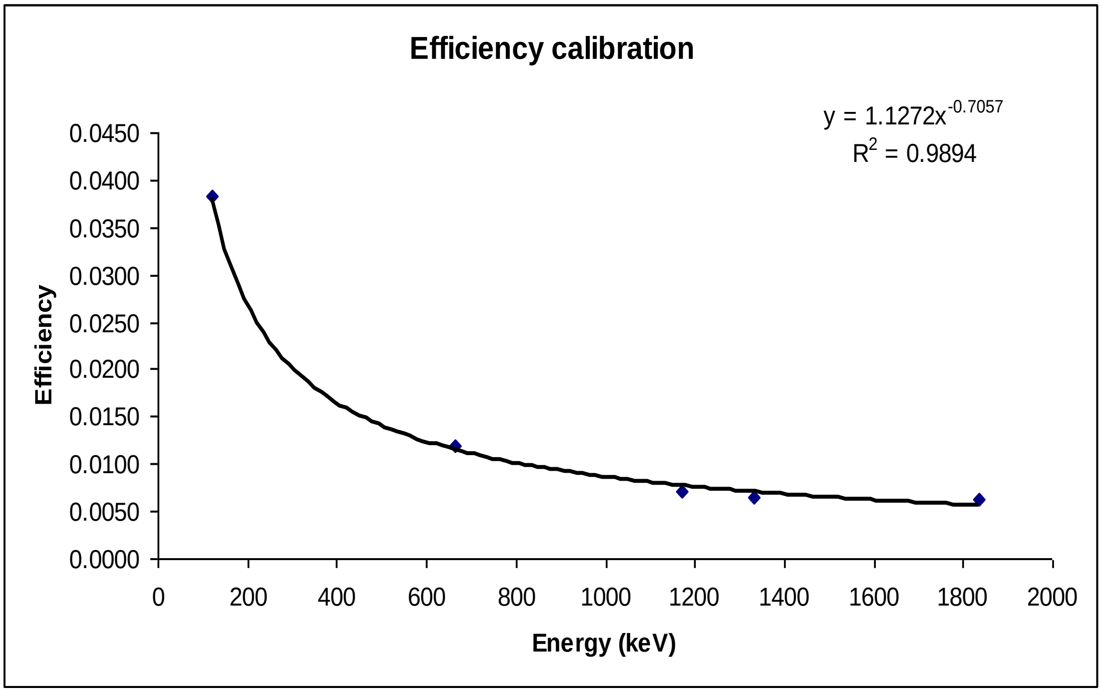


Figure 13: Efficiency calibration curve as a function of energy for mixed radionuclides standard in a one litre Marinelli beaker.

Lower limit of detection (LLD) of the system was calculated by using the following expression (IAEA, 1989):

$$LLD = \frac{4.66\sqrt{Bkg counts}}{CT\ x\ Ab\ x\ \eta} \qquad (3.4)$$

where;

CT is counting time;

Ab is the % abundance; and

η is detection efficiency of a particular radionuclide.

The calculated values of LLD of the system for some radionuclides are listed in Table 3.2.

Table 3.2: Lower Limit of Detection of some radionuclides of interest

Radionuclides	**Energy (keV)**	**Half Life (Years)**	**LLD (Bq)**
^{137}Cs	661.62	30.0	0.16
^{134}Cs	604.66	2.06	0.10
^{40}K	1460.75	1.28×10^9	5.06
^{226}Ra	609.31	1600	0.23
^{228}Ra	338.40	5.75	1.37

3.6 Radiometric Analysis

Radiometric measurements were performed for 72,000 seconds for qualitative and quantitative determination of different radionuclides present in the water, soil and food samples. Activities of ^{238}U and ^{232}Th were measured through their daughters ^{226}Ra and ^{228}Ra, respectively. The 609 keV gamma line of ^{214}Bi was used to determine ^{226}Ra, while the 911 keV gamma line of ^{228}Ac was used to detect the activity of ^{228}Ra. The analysis of ^{40}K were based upon their single peak of 1460 keV. The specific activities (A) in Bq kg^{-1} or Bq l^{-1} of various radionuclides were calculated using the formula (Darko et al., 2010):

$$A = \frac{Net\ counts}{CT\ x\ \eta\ x\ Ab\ x\ m} \qquad (3.5)$$

where;

Net counts = Sample Counts – Background Counts

CT = Counting Time

Ab = Abundance of the desired peak

η = Efficiency of the desired radionuclide

m = Mass of the samples

3.6.1 Determination of minimum detectable activity

Minimum detectable activity (MDA) is defined as the smallest quantity of radioactivity that could be measured under specified conditions. The MDA is an important concept in low level counting particularly in environmental level systems where the count rate of a sample is almost the same as the count rate of the background. Under these conditions, the background is counted with a blank, such as sample holder, and everything else that may be counted with an actual sample. In this work, 1liter Marinelli beaker filled with distilled water was counted for 36000s and the average background peaks used to determine MDA (Cember, 1996). For ^{226}Ra (^{238}U decay series), the minimum detectable activity was determined using average peak areas of the daughter gamma ray lines 295.2, 351.9 keV of ^{214}Pb and 609.31, 1764.5 keV of ^{214}Bi. The daughter gamma ray lines of 238.63 keV of ^{212}Pb, 583.2 and 2614.53 keV of ^{208}TI and 911.21 keV of ^{228}Ac keV were used to determine the MDA of ^{232}Th. The MDA of ^{40}K was determined using the gamma ray line at 1460.8 keV. The minimum detectable activities (MDA) were calculated using the following equation (Cember, 1996).

$$MDA = \frac{\sigma\sqrt{B}}{\eta.\mathrm{P.T.}W}(Bq/kg) \qquad (3.6)$$

where;

σ is the statistical coverage factor equal to 1.645 (confidence level 95%),

B is the background for the region of interest of each radionuclide,

T is the counting time in seconds,

P is the gamma emission probability (gamma yield) of each radionuclide,

W is the weight of the sample container, and

η is the detector efficiency for the measured gamma ray energy.

The Minimum Detectable Activities for ^{238}U (^{226}Ra), ^{232}Th and ^{40}K are shown in Table 3.3 with estimated values of 0.12, 0.11 and 0.15 Bq kg^{-1}, respectively.

Table 3.3: The minimum detectable activities of ^{238}U, ^{232}Th and ^{40}K.

Nuclide	MDA, Bq kg^{-1}
^{238}U	0.12
^{232}Th	0.11
^{40}K	0.15

3.7 Determination of radiological effects

3.7.1 External Gamma Dose Rate

The natural uranium-thorium decay products which exist at trace levels in all ground formations represent the main external source of radiation to the human body. The outdoor terrestrial gamma dose rates were measured at 1 metre above the ground by a portable digital survey metre (RADOS 200) at all sampling sites. A total of five readings were taken at each spot and average taken. The gamma dose rate (D) in nGy h^{-1} in outdoor air at 1 metre above the ground was calculated using the conversion factors given by UNSCEAR (UNSCEAR, 2000).

$$D = 0.462 * A_{apU} + 0.604 * A_{spTh} + 0.0417 * A_{spK} \qquad (3.7)$$

where 0.462 is Dose constant for ^{238}U, 0.604 is Dose constant for ^{232}Th and 0.0417 is Dose constant for ^{40}K. A_U, A_{Th} and A_K are the activity concentrations (Bq kg^{-1}) of ^{238}U, ^{232}Th and ^{40}K respectively. Using the conversion factor of 0.70 Sv Gy^{-1} (UNSCEAR, 2000) the dose equivalents in µSv y^{-1} were computed.

3.7.2 Annual Effective Dose

The annual effective dose (AED) in µSv received by a person from external exposure was calculated from the absorbed dose rate by applying dose conversion factor of 0.7 Sv Gy^{-1} and the occupancy factor for outdoor and indoor was 0.2 (5/24) and 0.8 (19/24) respectively (Veiga *et al*, 2006). The annual effective dose to external gamma irradiation was determined using the following equation:

$$\text{AED (Outdoor) } (\mu\text{Sv}) = (\text{Absorbed dose})\ \text{nGy h}^{-1} \times 8760\ \text{h} \times 0.7\ \text{Sv Gy}^{-1} \times 0.2 \times 10^{-3} \qquad (3.8)$$

$$\text{AED (Indoor) } (\mu\text{Sv}) = (\text{Absorbed dose})\ \text{nGy h}^{-1} \times 8760\ \text{h} \times 0.7\ \text{Sv Gy}^{-1} \times 0.8 \times 10^{-3} \qquad (3.9)$$

The annual effective dose in µSv received by a member from ingestion (internal exposure) of radionuclide consumed in water was calculated on the basis of the mean activity concentrations of the radionuclides. The daily water consumption rate was considered to be 2 litres per day (730 litres per year) and the conversion factor or dose per unit intake by ingestion for naturally occurring radionuclides for adult members of the public is 4.5 x 10^{-5} mSv Bq^{-1} for ^{238}U, 2.3 x 10^{-4} mSv Bq^{-1} for ^{232}Th and 6.2 x 10^{-6} mSv Bq^{-1} for ^{40}K as reported by the International Commission on Radiological Protection (ICRP, 1996) were used. The annual effective dose owing to ingestion of U, Th and K in water was calculated as:

$$E\ (\mu Sv\ y^{-1}) = \sum_{j} I_j x 730 x D_j \qquad (3.10)$$

where, I_j is the daily intake of radionuclide j (Bq d^{-1}), and D_j is the ingestion dose coefficient (Sv Bq^{-1}).

The annual intakes (Q) of ^{226}Ra, ^{228}Ra, ^{228}Th and ^{40}K were determined in food crops using the activity concentration levels (C) in the food crops and the mean annual food consumption rates (F) by the Ghanaian population according to the Ministry of Food and Agriculture (2008) (MOFA, 2008; SRID, 2008). The calculated annual intakes and the internal dose conversion factors (IDCF) of 2.8 x 10^{-4}, 6.9 x 10^{-4}, 7.2 x 10^{-5} and 6.2 x 10^{-6} mSv Bq^{-1} of ^{226}Ra, ^{228}Ra, ^{228}Th and ^{40}K, respectively, were used to estimate the committed effective dose (H) (ICRP, 1995), using the following equations:

$$H = Q \times IDCF \qquad (3.11)$$

where $$Q = C \times F \qquad (3.12)$$

3.7.3 Radium Equivalent and Hazard Indices

The radiological risk of NORM in soils and rocks which may be used as building materials was assessed by calculating the radium equivalent activity (Ra_{eq}) and the internal hazard (H_{in}) and external hazard (H_{ex}) indices.

Radiation exposure can be defined in terms of many parameters. Radium equivalent activity (Ra_{eq}) is a widely used hazard index. It is calculated, as given by the following equation, assuming that 370 Bq kg^{-1} of ^{226}Ra, 259 Bq kg^{-1} of ^{232}Th and 4810 Bq kg^{-1} of ^{40}K produce the same gamma-ray dose rate (Beretka and Mathew, 1985):

$$Raeq\ (Bq\ kg^{-1}) = A_{Ra\text{-}226} + 1.43A_{Th\text{-}232} + 0.077A_{K\text{-}40} \qquad (3.13)$$

where $A_{Ra\text{-}226}$, $A_{Th\text{-}232}$ and $A_{K\text{-}40}$ are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1}, respectively.

Other parameters used to estimate the levels of gamma-ray radiation associated with natural radionuclides are defined by the term external hazard index (H_{ex}) and internal hazard index (H_{in}). The prime objective of these indices is to limit the radiation dose to the dose equivalent limit of 1 mSv y^{-1} (ICRP, 1990). The external hazard index (H_{ex}) is given by a model proposed by Krieger (1981) as:

$$H_{ex} = \frac{A_{Ra-226}}{370} + \frac{A_{Th-232}}{259} + \frac{A_{K-40}}{4810} < 1 \qquad (3.14)$$

H_{ex} must not exceed the limit of unity for the radiation hazard to be negligible. On the other hand, the internal hazard index (H_{in}) gives the internal exposure to carcinogenic radon and its short-lived progeny and is given by the following formula (Beretka and Mathew, 1985):

$$H_{in} = \frac{A_{Ra-226}}{185} + \frac{A_{Th-232}}{259} + \frac{A_{K-40}}{4810} < 1 \qquad (3.15)$$

The values of H_{in} must also be less than unity to have negligible hazardous effects of radon and its short-lived progeny to the respiratory organs (UNSCEAR, 2000).

3.7.4 Representative level index

In order to examine whether the samples meet these limits of dose criteria, another radiation hazard index, the representative level index, $I_{\gamma,}$ used to estimate the level of γ-radiation hazard associated with the natural radionuclides in specific investigated samples, is defined in the following Equation (NEA-OECD, 1979),

$$I_{\gamma} = \frac{A_{Ra}}{150} + \frac{A_{Th}}{100} + \frac{A_{K}}{1500} \qquad (3.16)$$

where A_{Ra}, A_{Th} and A_{K} are the concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively, in Bq kg^{-1}.

3.7.5 Annual Gonadal Dose Equivalent

In the same context, the bone marrow and the bone surface cells are considered as the organs of interest by UNSCEAR (UNSCEAR, 1988). Therefore, the annual gonadal dose equivalent (AGDE) due to the specific activities of ^{226}Ra, ^{232}Th and ^{40}K were calculated using the following formula (Mamont-Ciesla *et al*, 1982):

$$AGDE\ (\mu Sv) = 3.09A_{Ra\text{-}226} + 4.18A_{Th\text{-}232} + 0.314A_{K\text{-}40} \quad (3.17)$$

where 3.09 is dose conversion factor for ^{238}U, 4.18 is dose conversion factor for ^{232}Th and 0.314 is dose conversion factor for ^{40}K. $A_{Ra\text{-}226}$, $A_{Th\text{-}232}$ and $A_{K\text{-}40}$ are the activity concentrations (Bq kg^{-1} or Bq l^{-1}) of ^{238}U, ^{232}Th and ^{40}K, respectively.

3.7.6 Cancer risks estimation

Research and biological knowledge of molecular and cellular mechanisms confirm that cancer is a highly complex multi-step process and it is likely to be occurring at low level of external as well as internal radiation. This makes unlikely the hypothesis that low level radiation or any contributor to DNA damage enhances the probability of cancer (Cuttler, 2006). Cancer incidence is defined as the probability of contracting a fatal cancer by individuals or groups of individuals. The annual fatal cancer risk varies from group to group and from individual to individual. Estimated lifetime cancer risk (ELCR) over a period of 70 years of exposure for children and 50 years for adults was calculated by using the following equation (Kapdan et al., 2011)

$$ELCR = AED \times DL \times RF \quad (3.18)$$

where AED is annual effective dose, DL is duration of life (70) for members of the public and RF is risk factor (Sv^{-1}), fatal cancer risk per Sievert. For stochastic effects, ICRP 103 uses values of 0.057 for the public and 0.042 for adults (ICRP, 2007).

3.8 Statistical analysis of samples

Paired Sample t-test statistical technique of Statistical Package for Social Scientists (SPSS) statistical software was used to compare the Means of the radionuclides concentrations in the water and soil samples. This technique was used because sampling was carried out at two different periods for the study**;** first batch April 2012 (relatively wet period) and second batch August 2013 (relatively dry period). If the probability value P is greater than the significance level at 5 % ($P>0.05$), then it implies that the paired sample Means are insignificant or the Mean of the two paired samples are equal. On the other hand if the P-value is less than the significance level at 5 % ($P<0.05$) then there is a significant difference between the means of the two sets of data. The paired sample t-test computes the difference between two variables for each case, and tests to find out if the average difference is significantly different from zero at 95 % Confidence level. The paired sample t-test is calculated from the expression below:

$$t = \frac{\bar{d}}{\sqrt{s^2/n}} \tag{3.19}$$

Where $\bar{d}$ is the mean difference between two samples, s^2 is the sample variance, n is the sample size and t is a paired sample t-test with n-1 being the degrees of freedom.

3.9 Uncertainty estimation

Every analytical measurement is always associated with a number of uncertainties which have to be identified and quantified. These uncertainties are also referred to as the quantification of the doubts associated with the measurement namely random and systematic uncertainties. Random uncertainties vary from measurement to measurement and are equally likely to be positive or negative. Some of the factors which give rise to this type of uncertainty include fluctuation in environmental conditions, e.g. temperature, pressure, humidity, etc, due to differences in the chemical and physical composition of samples. Random uncertainties are always present in a series of measurement. Random uncertainties can be detected through repeated measurements but they cannot be eliminated.

The second type of uncertainty is referred to as systematic uncertainty. This type of uncertainty remains the same throughout a set of measurements. This may arise because the experimental set up is different from that assumed by theory or the instrument is being used outside of its calibration range. It may also arise due to an instrument being poorly calibrated or calibrated with poorly prepared standards. These types of uncertainties may be difficult to detect.

In any analytical measurement, results should be quoted accompanying a statement of the uncertainty in the measurement. Uncertainty estimation involves the following steps:

- Identifying all of the potential sources of uncertainty in the measurement,
- Estimation of the size of uncertainty from each source of uncertainty,

- Combine all of the estimated uncertainties to give an overall figure of merit for the quantity being measured.

In quoting the results of measurements, the quantity measured must be quoted with the uncertainty. In addition, a statement of the coverage factor and the confidence level should be stated.

In this study, the uncertainties associated with the determination of activity concentrations of each radionuclide were estimated from expression used in the calculation of the specific activity concentrations, viz equation (3.20).

$$A_{SP} = \frac{N.e^{\lambda.Td}}{\eta.P.M.\mathrm{T}c} \qquad (3.20)$$

Where;

A_{sp} is the specific activity in Bq kg^{-1},

N is the background corrected net peak area,

η is the absolute detection efficiency,

P is the gamma ray yield,

Tc is the counting time of the sample,

λ is the decay constant of individual radionuclides,

Td is the time between sampling and time of counting.

Some of the uncertainties identified for the quantification of the uncertainty in the determination of the specific activity concentrations include the following:

- Net peak area,

- Detection efficiency,
- Sample mass,
- Counting time.

The overall uncertainty in the determination of the activity concentrations was obtained using equation (3.21).

$$dA_{sp} = A_{sp} *[(\frac{dN}{N})^2 + (\frac{d\eta}{\eta})^2 + (\frac{dM}{M})^2]^{\frac{1}{2}} \quad (3.21)$$

dN is determined from the uncertainty in the integration of the peak area of each full energy event.

dM is the standard uncertainty on the weighing balance used to weigh the samples and the standard uncertainty was quoted to be 0.1 mg, and

dη is the uncertainty in the efficiency calibration of the counting system.

CHAPTER FOUR

4.0 RESULTS AND DISCUSSION

The summary of the dose rate based on the conversion factors given by UNSCEAR (2000) and the outdoor terrestrial gamma dose rates measured at 1 meter above the ground using the RADOS 200 portable digital survey meter and the calculated absorbed dose rate at the sampling sites and their corresponding annual effective dose rates are presented in Tables 4.1 and 4.2. Tables 4.3-4.5 show the activity concentrations of ^{238}U, ^{232}Th and ^{40}K in the soil, water and food crop samples. Table 4.6 shows the mean radioactivity concentrations and mean annual effective dose due to external gamma irradiation from ^{238}U, ^{232}Th and ^{40}K in soil samples from the mine site, mine tailings from the Adamus mine and soil samples from the surrounding villages. Table 4.7-4.8 show the mean radioactivity concentration and annual effective dose from ingestion of ^{238}U, ^{232}Th, and ^{40}K in water and the mean annual radionuclide intake and effective ingestion dose from ingestion of selected food crops from the study area. Table 4.9 shows the mean absorbed dose rate, mean annual effective dose, hazard indices, and activity utilization index and estimated cancer risks from external gamma rays within the study area. Tables 4.10-4.11 show the mean absorbed dose rate, mean annual effective dose and estimated cancer risks from ingestion of water and mean annual radionuclide intake, mean annual effective dose and estimated cancer risks from ingestion of the selected food crops from the study area.

4.1 Soil and Mine tailings

Tables 4.1-4.2 show the absorbed dose rate due to external irradiation from radionuclides in the ground and from direct measurements at 1 meter above the ground and the corresponding annual effective dose at the mine site, tailings dam site and the surrounding villages. From the tables 4.1 and 4.2, it can be observed that the measured and the observed dose rates range from 45-67 nGy h^{-1} and 25.94-73.21 nGy h^{-1}, respectively, with mean values of 55.90 nGy h^{-1} and 46.75 nGy h^{-1}, respectively. The corresponding annual effective dose rate was estimated to be in the range of 55.19-82.17 µSv y^{-1} and 31.81-89.79 µSv y^{-1}, respectively, with mean annual effective dose rate of 68.56 µSv y^{-1} and 57.33 µSv y^{-1}, respectively.

Table 4.1: Mean absorbed dose rate measured at 1 meter above the ground and mean absorbed dose rate from external gamma rays within the Adamus mine and surroundings villages

Location		**Absorbed Dose Rate (nGy h^{-1})**	
		Direct measurement of ambient gamma dose	**External irradiation from sources in the ground**
Villages	Salman Town	58	27.84
	Akango	52	25.94
	Teberu	58	30.96
	Aliku Town	51	43.08
	Nkroful Bokrobo	59	52.09
	North Hill	52	61.71
	Nugget Hill	54	73.21
	Angajeleh	55	70.06
Mine site		67	43.58
Mine Tailings		63	39.04
Range		**51-67**	**25.94-73.21**
Mean		**55.90**	**46.75**

Table 4.2: Mean annual effective dose rate measured at 1 meter above the ground and mean annual effective dose from external gamma rays within the Adamus mine and surroundings villages

<table>
<tr><th colspan="2" rowspan="2">Location</th><th colspan="2">Mean Annual Effective Dose Rate (μSv y⁻¹)</th></tr>
<tr><th>Direct measurement of ambient annual effective dose</th><th>Annual effective dose from irradiation from sources in the ground</th></tr>
<tr><td rowspan="8">Villages</td><td>Salman Town</td><td>58.87</td><td>34.14</td></tr>
<tr><td>Akango</td><td>76.04</td><td>31.81</td></tr>
<tr><td>Teberu</td><td>71.13</td><td>37.97</td></tr>
<tr><td>Aliku Town</td><td>62.55</td><td>52.83</td></tr>
<tr><td>Nkroful Bokrobo</td><td>72.36</td><td>63.88</td></tr>
<tr><td>North Hill</td><td>63.77</td><td>75.68</td></tr>
<tr><td>Nugget Hill</td><td>66.23</td><td>89.79</td></tr>
<tr><td>Angajeleh</td><td>55.19</td><td>85.92</td></tr>
<tr><td colspan="2">Mine site</td><td>82.17</td><td>53.45</td></tr>
<tr><td colspan="2">Mine Tailings</td><td>77.26</td><td>47.87</td></tr>
<tr><td colspan="2"></td><td></td><td></td></tr>
<tr><td colspan="2">Range</td><td>55.19-82.17</td><td>31.81-89.79</td></tr>
<tr><td colspan="2">Mean</td><td>68.56</td><td>57.33</td></tr>
</table>

According to UNSCEAR report, the average absorbed dose rate in air from terrestrial gamma radiation is 59 nGy h^{-1} (UNSCEAR, 2000). Comparing the average gamma absorbed dose rates in this study with UNSCEAR report, the average absorbed dose rates from direct measurements and from external irradiation in this study are lower than the world average value (59 nGy h^{-1}). The highest absorbed dose rate and from external irradiation were recorded in samples from mine site with a value of 67.0 nGy h^{-1} and from Nugget Hill with a value of 73.21 nGy h^{-1}, respectively. These values are higher than the world average value because these sites are at higher altitude relative to the other sites. It has been reported that places at higher altitude are associated with higher external gamma dose rates. It was observed that these results compared very well with similar studies carried out in other mines in Ghana (Darko *et al*., 2010).

Table 4.3 shows the mean activity concentration for ^{238}U, ^{232}Th and ^{40}K in soil samples. The mean activity concentration ranges from 6.79-15.58 Bq kg^{-1} with an average of 8.03 ± 1.38 Bq kg^{-1}, 12.71–86.48 Bq kg^{-1} with an average of 43.98 ± 1.32 Bq kg^{-1} and 284.23–507.67 Bq kg^{-1} with an average of 395.09 ± 2.82 Bq kg^{-1}, respectively.

These values show the ranges of ^{238}U and ^{232}Th activity concentrations. The activity concentration of ^{232}Th is generally higher than ^{238}U in all the soil samples. It has been reported that the total potassium content of rocks has a wide range of values from 0.3% to 4.5% for various rock types that corresponds to an activity concentration range of 90 to 1,400 Bq kg^{-1}. It has been estimated that about 110 TBq of ^{40}K is added annually to the soils in the form of fertilizer (Guimond, 1978). Because of its relative abundance, ^{40}K is the predominant radioactive component in common foods and human tissues. The activity concentration of ^{238}U, ^{232}Th and ^{40}K for all measured soil samples were below the world average value of 50 Bq kg^{-1}, 50 Bq kg^{-1} and 500 Bq kg^{-1} for ^{238}U, ^{232}Th and ^{40}K respectively (UNSCEAR, 2000). The comparison of the mean activity concentration of the samples from the various locations with the average world value is shown in figure 15.

Table 4.3: Radioactivity levels of ^{238}U, ^{232}Th and ^{40}K in soil samples and mine tailings from the Adamus mine the surrounding villages.

Sampling Sites		**Activity Concentration (Bq kg^{-1}) in soil samples and mine tailings**					
		U-238		**Th-232**		**K-40**	
		Range	**Mean**	**Range**	**Mean**	**Range**	**Mean**
Villages	Salman Town	7.13-8.11	7.47 ± 0.32	20.11-21.55	20.75 ± 0.33	201.23-342.63	284.23 ± 2.39
	Akango	7.11-8.01	7.55 ± 0.49	8.18-19.32	12.71 ± 0.54	305.01-385.55	354.22 ± 2.47
	Teberu	6.98-7.88	7.41 ± 0.74	10.99-26.25	19.24 ± 0.55	378.79-385.55	381.60 ± 2.64
	Aliku Town	6.71-7.56	7.18 ± 0.63	24.26-58.48	39.02 ± 0.76	354.77-406.82	388.26 ± 2.50
	Nkroful Bokrobo	6.69-7.24	7.07 ± 0.19	37.90-74-52	52.79 ± 0.55	385.55-430.36	406.15 ± 2.44
	North Hill	6.64-6.98	6.79 ± 0.76	58.69-89.65	68.57 ± 0.44	403.94-434.59	411.36 ± 2.71
	Nugget Hill	6.45-7.79	6.90 ± 0.82	74.52-96.64	86.48 ± 0.66	420.55-437.71	426.66 ± 2.99
	Angajeleh	6.52-7.89	6.90 ± 0.89	30.17-97.26	81.73 ± 0.34	279.10-469.71	419.80 ± 2.60
Mine site (Soil)		6.98-7.88	15.58 ± 1.80	14.99-16.94	25.19 ± 1.18	170.21-178.79	507.67 ± 3.50
Mine Tailings		26.71-27.56	7.41 ± 3.41	13.95-19.71	33.35 ± 1.34	182.68-198.08	370.97 ± 2.75
Mean			**8.03 ± 1.38**		**43.98 ± 1.32**		**395.09 ± 2.82**
Range			**6.79-15.58**		**12.71–86.48**		**284.23-507.67**

The comparison of measured and calculated absorbed dose rate in soil samples from the study area is shown in figure 14. Figures 15-17 show the comparison of the mean activity concentration of ^{238}U, ^{232}Th and ^{40}K in soil samples from the various locations with the world average values, the contribution of ^{238}U, ^{232}Th and ^{40}K radionuclides to the total activity concentration in the soil samples and mean activity concentration of radionuclides in ground, surface and mine processed water from the study area. Figures 18-19 show the comparison of mean absorbed dose rate in soil samples taken from different locations and comparison of mean absorbed dose rate in the water sample types taken from the study area.

Figure 14 shows that the measured absorbed dose rates at the sampling locations were higher than the calculated absorbed dose rate. These high absorbed dose rate from the direct measurements could be due to the contribution of cosmic rays and possible statistical uncertainties in the measurements.

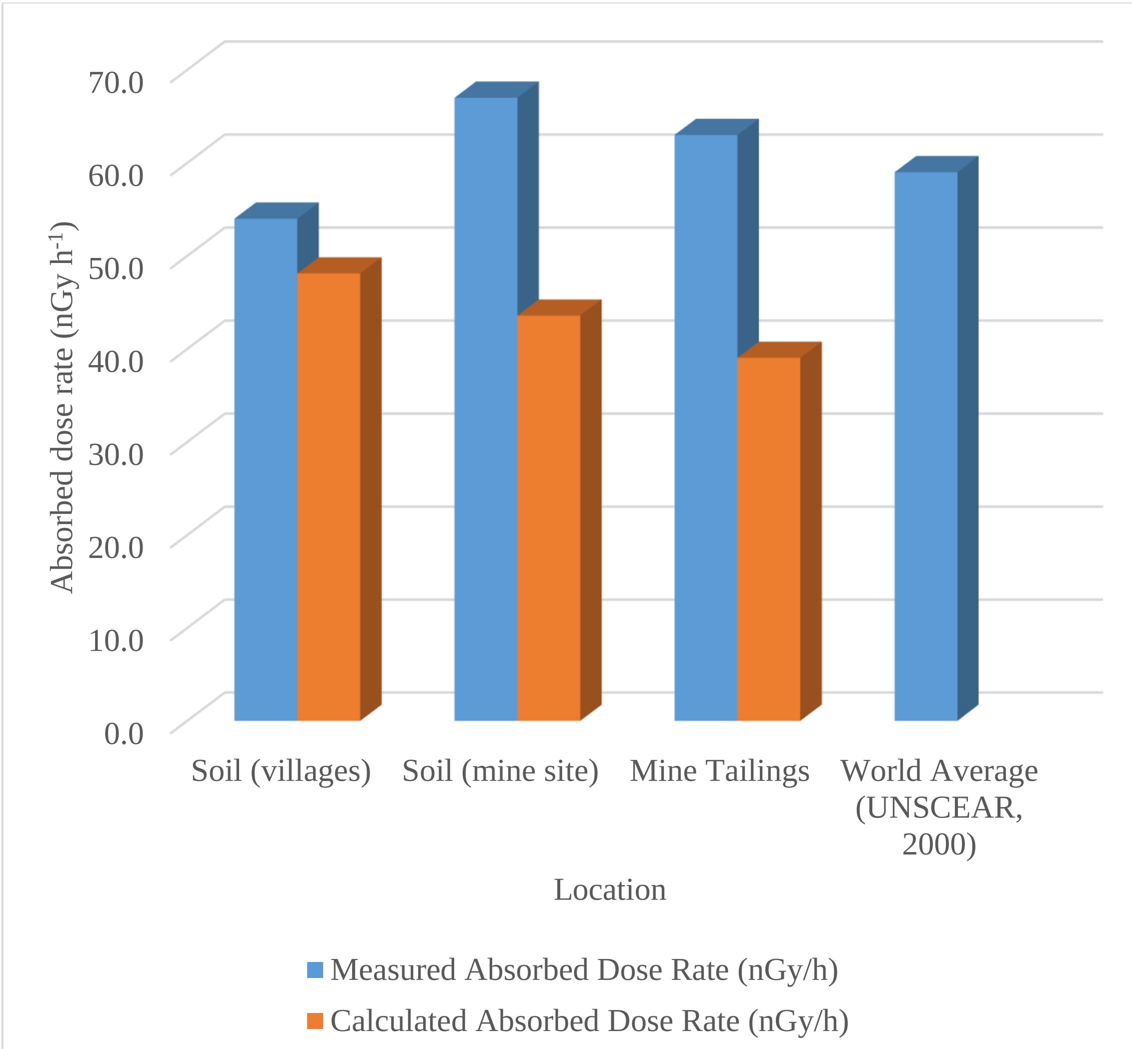


Figure 14: Comparison of measured and calculated absorbed dose rate measured at the soil in study area.

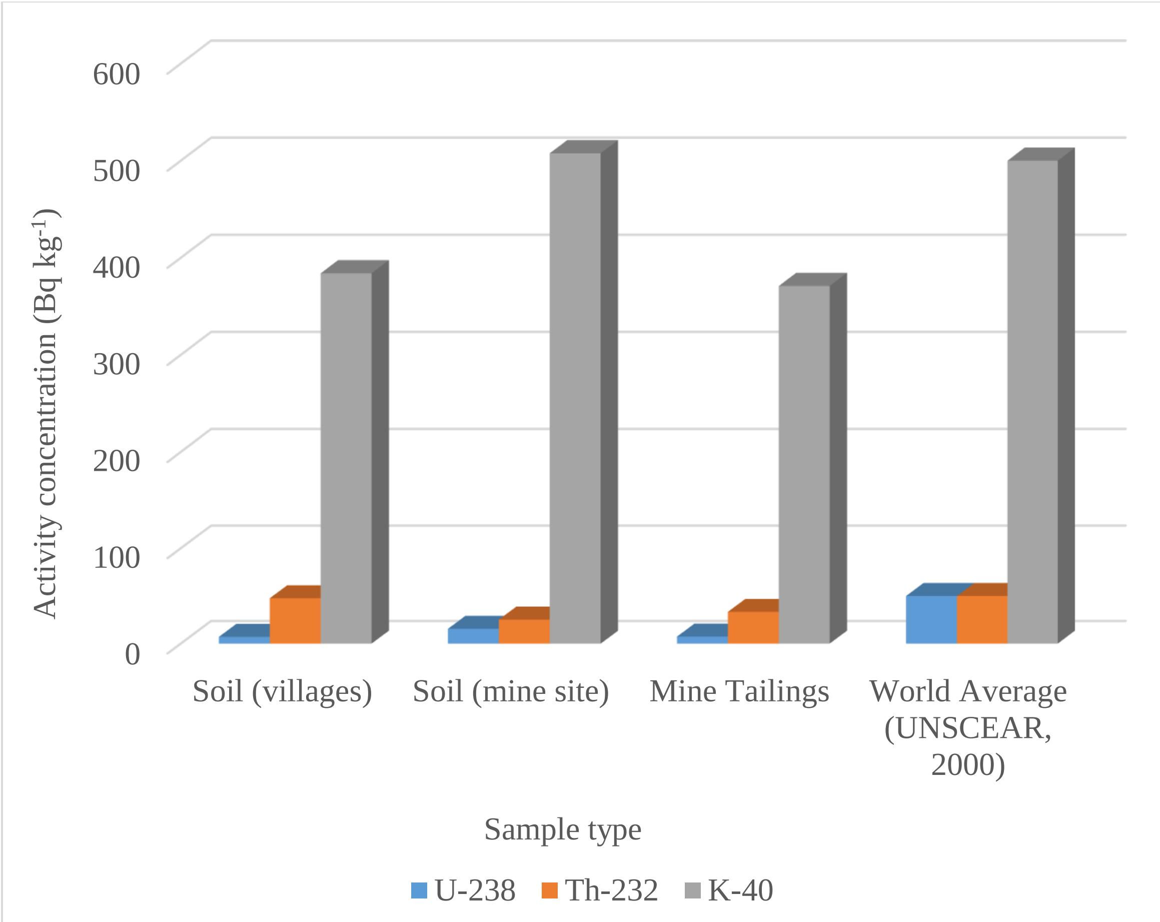


Figure 15: Comparison of the mean activity concentration of ^{238}U, ^{232}Th and ^{40}K in soil samples from the various locations with the average world values.

Figure 16 shows the contribution of ^{238}U, ^{232}Th and ^{40}K radionuclides to the total activity concentration in the soil samples. It is observed from figure 16 that ^{40}K contributes significantly to the total activity concentration in the soil samples with ^{232}Th being the second contributor. The high activity concentration in the study area may be due to the total potassium content of rocks and the use of fertilizers with significant amount of potassium in the study area.

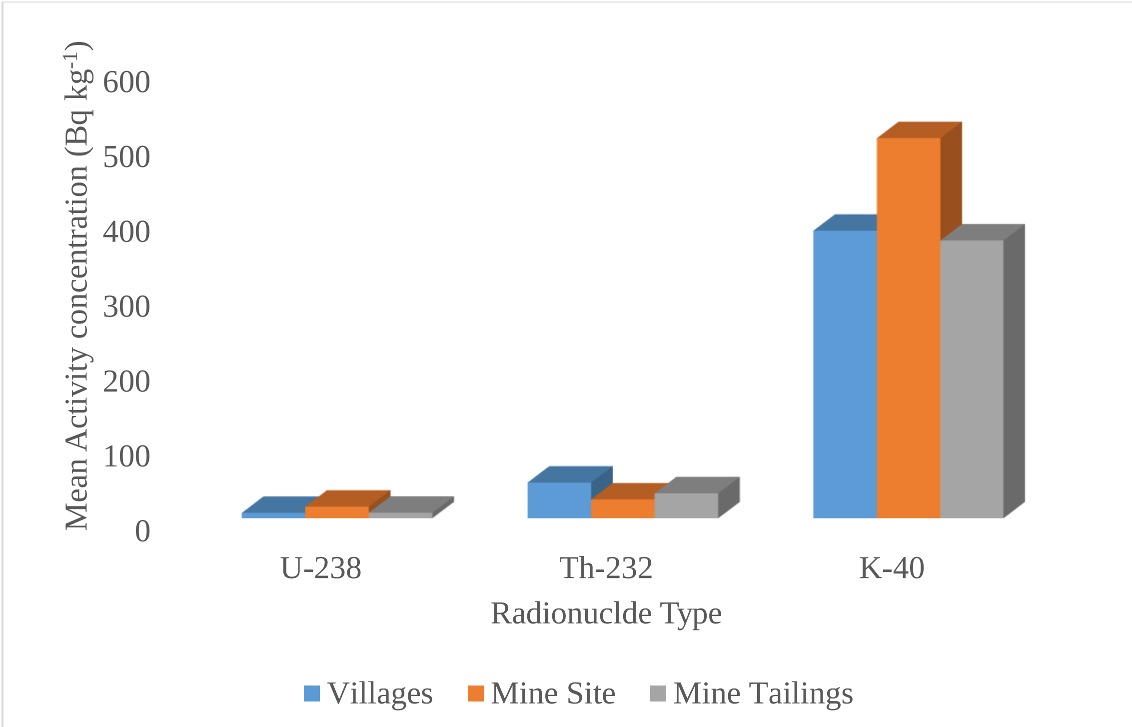


Figure 16: Contribution of ^{238}U, ^{232}Th and ^{40}K radionuclides to the total activity concentration in the soil samples.

4.2 Water

Table 4.4 show the radioactivity levels of ^{238}U, ^{232}Th and ^{40}K in ground, surface and mine processed water samples from the Adamus mine and surrounding villages. For groundwater samples the activity concentration ranges for ^{238}U, ^{232}Th and ^{40}K are 0.10 – 0.20 Bq l^{-1} with an average of 0.16 ± 0.04 Bq l^{-1}, 0.70 – 1.00 Bq l^{-1} with an average of 0.83 ± 0.04 Bq l^{-1} and 0.61 – 0.76 Bq l^{-1} with an average of 0.70 ± 0.03 Bq l^{-1}, respectively. For surface water samples the activity concentration ranges for ^{238}U, ^{232}Th and ^{40}K are 0.10 – 0.17 Bq l^{-1} with an average of 0.13 ± 0.03 Bq l^{-1}, 0.60 – 0.74 Bq l^{-1} with an average of 0.66 ± 0.04 Bq l^{-1} and 0.64 – 0.79 Bq l^{-1} with an average of 0.72 ± 0.05 Bq l^{-1}, respectively. For mine processed water samples the activity concentration ranges for ^{238}U, ^{232}Th and ^{40}K are 0.16 – 0.18 Bq

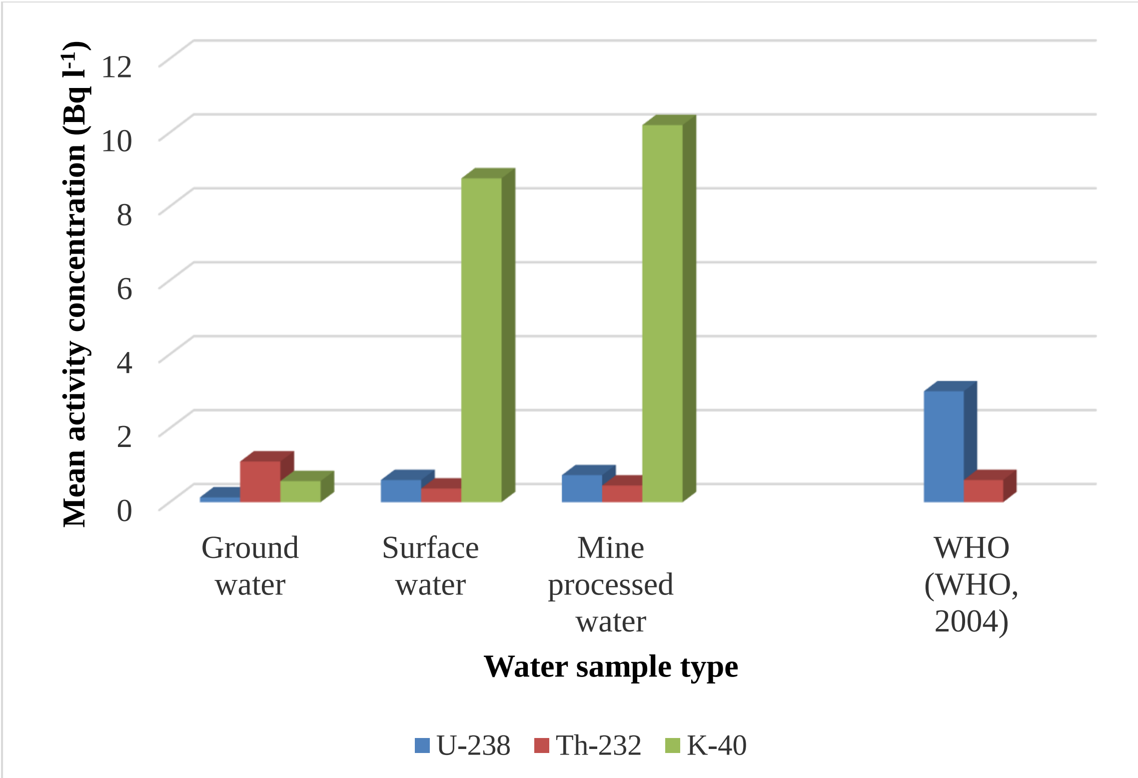
l^{-1} with an average of 0.17 ± 0.04 Bq l^{-1}, 1.40 – 1.52 Bq l^{-1} with an average of 1.45 ± 0.03 Bq l^{-1} and 0.79 – 0.93 Bq l^{-1} with an average of 0.86 ± 0.06 Bq l^{-1}, respectively.

Table 4.4: Radioactivity levels of ^{238}U, ^{232}Th and ^{40}K in ground, surface and mine processed water samples from the Adamus mine and surrounding villages.

Water Type	**Water Sample Code**	**Activity Concentration (Bq l^{-1})**		
		U-238	**Th-232**	**K-40**
Groundwater	SLHAP	0.19 ± 0.03	0.80 ± 0.05	0.76 ± 0.03
	SLACB	0.18 ± 0.04	1.00 ± 0.04	0.73 ± 0.02
	SLGCB	0.20 ± 0.06	0.70 ± 0.04	0.65 ± 0.04
	SLBCB	0.10 ± 0.03	0.84 ± 0.05	0.61 ± 0.02
	SLTWE	0.14 ± 0.02	0.81 ± 0.04	0.76 ± 0.02
	Mean	**0.16 ± 0.04**	**0.83 ± 0.04**	**0.70 ± 0.03**
Surface water	ANGST	0.10 ± 0.06	0.61 ± 0.03	0.64 ± 0.04
	BANST	0.17 ± 0.03	0.74 ± 0.02	0.65 ± 0.08
	ANBST	0.12 ± 0.05	0.66 ± 0.03	0.77 ± 0.05
	TEBST 1	0.15 ± 0.02	0.72 ± 0.04	0.79 ± 0.04
	TEBST 2	0.12 ± 0.03	0.65 ± 0.03	0.68 ± 0.04
	BROST	0.10 ± 0.02	0.60 ± 0.03	0.76 ± 0.05
	SALST	0.13 ± 0.04	0.66 ± 0.04	0.77 ± 0.08
	Mean	**0.13 ± 0.04**	**0.66 ± 0.03**	**0.72 ± 0.05**
Mine processed water	TSFSW	0.16 ± 0.03	1.47 ± 0.04	0.79 ± 0.05
	TSFST 1	0.17 ± 0.02	1.52 ± 0.03	0.86 ± 0.06
	TSFST 2	0.18 ± 0.04	1.40 ± 0.03	0.87 ± 0.05
	NKBPW	0.17 ± 0.03	1.41 ± 0.04	0.93 ± 0.08
	Mean	**0.17 ± 0.03**	**1.45 ± 0.04**	**0.86 ± 0.06**

Figure 17 shows the comparison of mean activity concentration of radionuclides in ground, surface and mine processed water from the study area. The results show small ranges of ^{238}U and ^{232}Th activity concentrations. It could be observed that the activity concentration of ^{232}Th is higher than ^{238}U in all the water samples. This is because soils in the study area are acidic and are also low in nutrients due to high leaching as a result of the high rainfall in the Municipality and because of the acidic conditions, ^{232}Th tends to be more soluble in water than ^{238}U hence the high concentration of thorium in all the water samples.

The highest activity concentration of 1.45 Bq l^{-1} for ^{232}Th was measured in water samples from the mine processed water. This may be due to the fact that the mine processed water is a collection of all processed water from the mine. Thorium is higher in groundwater compared to surface water. This may also be so because thorium is generally soluble in underground water which is slightly acidic than in surface water. Like uranium, thorium is ubiquitous in nature. In aqueous systems, thorium undergoes hydrolysis in aqueous solutions above pH 2-3 and is subject to extensive sorption by clay minerals and humic acid at near-neutral pH (USDOE, 1993). Because of sorption and precipitation reactions and the low solution rate of thorium-bearing minerals, thorium concentrations in neutral waters are generally low hence the low activity concentration in surface water. At low pH, such as in an acid-leach, thorium becomes more soluble and has been shown to migrate considerably deeper into subsoil than ^{238}U (USDOE, 1993). Small amounts of thorium are present in all rocks, soil, above-ground and underground water, plants, and animals. These small amounts of thorium contribute to the weak background radiation for such substances.



Legend: GW – Groundwater; SW – Surface water; MPW – Mine processed water

Figure 17: Mean activity concentration of radionuclides in Ground, Surface and Mine processed water from the study area.

Uranium is found in all rocks and soils. Uranium is introduced into water supplies as a result of leaching from natural sources, from mill tailings, from the combustion of coal and other fuels, and from phosphate fertilizers.

The high activity concentrations of ^{40}K compared to ^{238}U could be due to the potassium content of the surrounding rocks and the application of high volumes of potassium in the form of fertilizer on farms and during run-offs they are washed or leached into water bodies. It has been reported that the total potassium content of rocks has a wide range of values from 0.3% to 4.5% for various rock types that correspond to an activity concentration range of 90 to 1,400 Bq kg^{-1}. It has been estimated that about 110 TBq of ^{40}K is added annually to the soils in the form of fertilizer (Guimond, 1978). Because of its relative abundance, ^{40}K is the predominant radioactive component in common foods and human tissues. It is important to recognize that the potassium content of the body is under homeostatic control and is little influenced by environmental variations. The dose from ^{40}K in the body is therefore reasonably constant.

Comparing the results from the study with WHO recommended guideline activity concentration values for ^{238}U and ^{232}Th, it was observed that results from the study were lower than the guideline values except for the mine processed water as shown in figure 17.

4.3 Food Crops

The major tree crops are coconut, oil palm, rubber and cocoa with cassava maize, cocoyam and plantain being the major food crops in the Municipality. Cassava, maize, cocoyam and plantain are grown extensively both for subsistence and for sale. Radioactivity levels of ^{238}U, ^{232}Th and ^{40}K in selected food crop (cassava, maize, cocoyam and plantain) samples from the Adamus mine and surrounding villages are shown in Table 4.5. The relative concentration of ^{238}U, ^{232}Th and ^{40}K in the selected food crops analyzed range from 3.84±1.69 – 10.46±3.66, 10.49±3.42 – 14.93±2.86 and 231.83±13.23 – 368.50±18.10 Bq kg^{-1}, respectively. The highest concentrations were observed in plantain samples for ^{238}U and in cassava for ^{232}Th and ^{40}K. The lowest ^{238}U, ^{232}Th and ^{40}K concentrations were observed in cassava and in maize, respectively. Potassium-40 (^{40}K) was detected in all food samples with reasonable activity concentration levels. The levels ranged from 231.83±15.23

Bq kg^{-1} in maize to 368.50 ± 19.20 Bq kg^{-1} in cassava. This result agrees with the world range reported by Maul and O'Hara (1989) for ^{40}K concentration from 40 to 240 Bq kg^{-1}.

The concentration of potassium was found to be very high compared to uranium and this may be attributed to poor migration characteristics of uranium from the substrate to the plant in the concerned environment. It is well known that the existence of potassium in all living things with different levels leads to a wide variety of ^{40}K radioisotope levels in plants. Potassium is a micronutrient and it may be expected that the soil characteristics favour the mobilization of potassium and its subsequent migration into plant (Pietrzak *et al*, 2001).

Table 4.5: Radioactivity levels of ^{226}Ra, ^{228}Th and ^{40}K in food cassava, cocoyam, maize and plantain from the Adamus mine and surrounding villages (Bq kg^{-1} dry weight)

Food crop type	**No. of Samples**	**^{226}Ra**		**^{228}Th**		**^{40}K**	
		Range	**Mean**	**Range**	**Mean**	**Range**	**Mean**
Cassava	6	1.90-7.31	3.84±1.69	8.80-28.80	14.93±2.86	289.00-445.00	368.50±18.10
Cocoyam	6	2.74-6.65	4.43±2.20	8.61-19.90	12.62±3.15	155.00-401.00	310.50±15.61
Maize	6	1.55-10.60	4.86±2.02	5.24-19.63	10.49±3.42	134.00-372.00	231.83±13.23
Plantain	6	2.93-31.60	10.46±3.66	6.50-13.70	10.76±3.28	173.00-353.00	279.50±15.25

4.4 Comparison with published data

Table 4.6-4.9 are comparisons of the mean activity concentrations of ^{238}U, ^{232}Th and ^{40}K in soils in the study area with similar studies done in Ghana and with published reports from other countries (UNSCEAR, 2000; Darko et al., 2010). The values compared well with published data from other countries and all values were below the world average values.

Table 4.6: Comparison of activity concentrations of ^{238}U, ^{232}Th and ^{40}K in soils in the study area and published data (UNSCEAR, 2000; Darko et al., 2010)

Country	Concentration in soil, Bq kg^{-1}					
	^{238}U		^{232}Th		^{40}K	
	Range	Mean	Range	Mean	Range	Mean
Algeria+	2-110	30	2-140	25	66-1150	370
Egypt+	6-120	37	2-96	18	29-650	320
United States+	4-140	35	4-130	35	100-700	370
India+	7-81	29	14-160	64	38-760	400
Malaysia+	49-86	66	63-110	82	170-430	310
Lithuania+	3-30	50	9-46	25	350-850	600
United Kingdom+	2-330	-	1-180	-	0-3200	-
Ghana (Mine1)#	-	29	-	25	-	582
Ghana (Mine 2)#	-	35	-	21	-	682
Ghana (This work)	6.79-15.58	8.03	12.71-86.48	43.98	284.23-507.67	284.23
World average+	-	33	-	45	-	420

Legend: + UNSCEAR 2000 Report; # Darko et al., 2010 for Ghana (Mine 1 and Mine 2 values)

Table 4.7: Comparison of the Global Average Specific Activity of U, Th, K in soil and absorbed dose rate in air from UNSCEAR report (UNSCEAR, 2000) with results of this work.

Radionuclide	UNSCEAR		This Work	
	Average Specific Activity in soil (Bq kg^{-1})	**Absorbed dose rate (nGy h^{-1})**	**Average Specific Activity in soil (Bq kg^{-1})**	**Absorbed dose rate (nGy h^{-1})**
U-238	25 (10-50)	11 (4-21)	8.0(7-16)	**13.3(8-19)**
Th-232	25 (7-50)	17 (5-33)	44(13-86)	**17.1(11-30)**
K-40	370 (100-700)	16 (4-30)	395(284-508)	**32.5(27-43)**

Table 4.8: Comparison of the average values of ^{238}U, ^{232}Th and ^{40}K concentration in water samples from the present investigation with other countries.

Country	Type of water	^{238}U (Bq l^{-1})	^{232}Th (Bq l^{-1})	^{40}K (Bq l^{-1})	Reference
Nigeria (Port Harcourt)	Surface water	48.29	0.0379	0.3624	Avwiri (2005)
China	Ground water	0.392	0.011	-	Ziqiang (1998)
Finland	Ground water	0.110	-	-	Salonen (1988)
Germany (Erzgebirge)	Tap water	-	0.00052	-	Fisenne (2000)
Egypt (Qena)	Ground water	0.079	0.041	-	Ahmed (2004)
Egypt (Safaga-Quseir)	Ground water	0.113	0.051	-	Ahmed (2004)
Ghana (Obuasi)	Ground water	0.14	0.65	0.64	Awudu (2010)
Ghana (Obuasi)	Surface water	0.12	0.50	0.68	Awudu (2010)
Ghana (Adamus)	Ground water	0.16 ± 0.04	0.83 ± 0.03	0.70 ± 0.04	Present work
Ghana (Adamus)	Surface water	0.13 ± 0.04	0.66 ± 0.03	0.72 ± 0.05	Present work
Ghana (Adamus)	Mine Process water	0.17 ± 0.03	1.45 ± 0.04	0.86 ± 0.06	Present work

Table 4.9: Comparison of the average activity concentrations (Bq kg^{-1}) of ^{226}Ra, ^{228Th} and ^{40}K in food crops determined in the present study and data from different studies.

Country	Sample	^{226}Ra	^{228}Ra	^{238}U	^{232}Th	^{228}Th	^{40}K	Reference
USA	Corn	0.21	-	-	-	-	87.00	(Hosseini, 2006)
Turkey	Corn	25.82	-	-	-	-	491.62	(Bolca, 2007)
S. Brazil	Corn	30.00	-	-	-	-	-	(Scheibe, 2006)
Nigeria	Cocoyam	2.66	-	-	3.12	-	20.06	(Jwanbot, 2012)
Nigeria	Cassava	2.25	-	-	2.62	-	17.97	(Jwanbot, 2012)
Ghana	Cassava	0.60	-	-	0.74	-	2.51	(Addo, 2013)
Ghana	Maize	5.00	9.00	-	-	10.00	232.00	(Awudu, 2011)
Ghana	Cassava	3.48	-	-	-	14.93	368.50	Present study
Ghana	Cocoyam	4.43	-	-	-	12.62	310.50	Present study
Ghana	Maize	4.86	-	-	-	10.49	231.83	Present study
Ghana	Plantain	10.46	-	-	-	10.76	279.50	Present study

Tables 4.10-4.12 show the estimated absorbed dose rates and annual effective doses due to external irradiation and ingestion of ^{238}U, ^{232}Th, and ^{40}K for the different sample types collected from the study area.

Table 4.10: Mean radioactivity concentrations and mean annual effective dose due to external gamma irradiation from ^{238}U, ^{232}Th and ^{40}K in soil samples from the mine site, mine tailings from the Adamus mine and soil samples from the surrounding villages.

Types of sample	**Concentration (Bq kg^{-1})**			**Absorbed Dose Rate (nGy h^{-1})**	**Annual Effective Dose (μSv y^{-1})**
	U-238	**Th-232**	**K-40**		
Soil (villages)	7.16 ± 1.08	47.66 ± 1.34	384.04 ± 2.75	48.11 ± 1.34	59.00 ± 1.60
Soil (mine site)	15.58 ± 1.80	25.19 ± 1.18	507.67 ± 3.50	43.58 ± 1.34	53.45 ± 1.70
Mine Tailings	7.41 ± 3.41	33.35 ± 1.34	370.97 ± 2.75	39.04 ± 1.19	47.87 ± 1.32

Table 4.11: Mean radioactivity concentration and annual effective dose from ingestion of ^{238}U, ^{232}Th, and ^{40}K in water from the Adamus mine and surrounding villages.

Water Type	**Concentration, (Bq l^{-1})**			**Absorbed Dose Rate (nGy h^{-1})**	**Annual Effective Dose (μSv y^{-1})**
	U-238	Th-232	K-40		
Ground water	0.16 ± 0.04	0.83 ± 0.03	0.70 ± 0.04	0.61 ± 0.03	5.20 ± 0.04
Surface water	0.13 ± 0.04	0.66 ± 0.03	0.72 ± 0.05	0.49 ± 0.04	4.20 ± 0.04
Mine water	0.17 ± 0.03	1.45 ± 0.04	0.86 ± 0.06	0.99 ± 0.03	8.50 ± 0.04

Table 4.12: Calculated mean annual radionuclide intake and effective ingestion dose

Foodstuff	*Consumption rate (kg/yr)	Mean annual radionuclide intake (Bq)			
		^{226}Ra (^{238}U)	^{232}Th	^{40}K	Dose (mSv)
Cassava	152.90	587.0	2283.0	56344.0	0.68
Cocoyam	57.10	253.0	721.0	17730.0	0.23
Maize	43.80	213.0	459.0	10154.0	0.16
Plantain	84.80	912.0	912.0	23702.0	0.47
Total		**1965.0**	**4375.0**	**107930.0**	**1.54**

Source: MOFA, SRID (MOFA, 2008; SRID, 2008)

The estimated absorbed dose rates range from a minimum of 39.04 nGy h^{-1} measured in soil samples collected from within the mine site to a maximum of 48.11 nGy h^{-1} measured in soil samples collected from surrounding villages. The differences could be attributed to differences in geology and geochemical state of the sampling sites. Similarly, the estimated absorbed dose rates range from 0.49 nGy h^{-1} measured in ground water samples to 0.99 nGy h^{-1} measured in mine processed water samples, respectively. The estimated average absorbed dose rate for all samples are lower than the world average value of 59 nGy h^{-1} (UNSCEAR, 2000) as shown in figures 18 and 19.

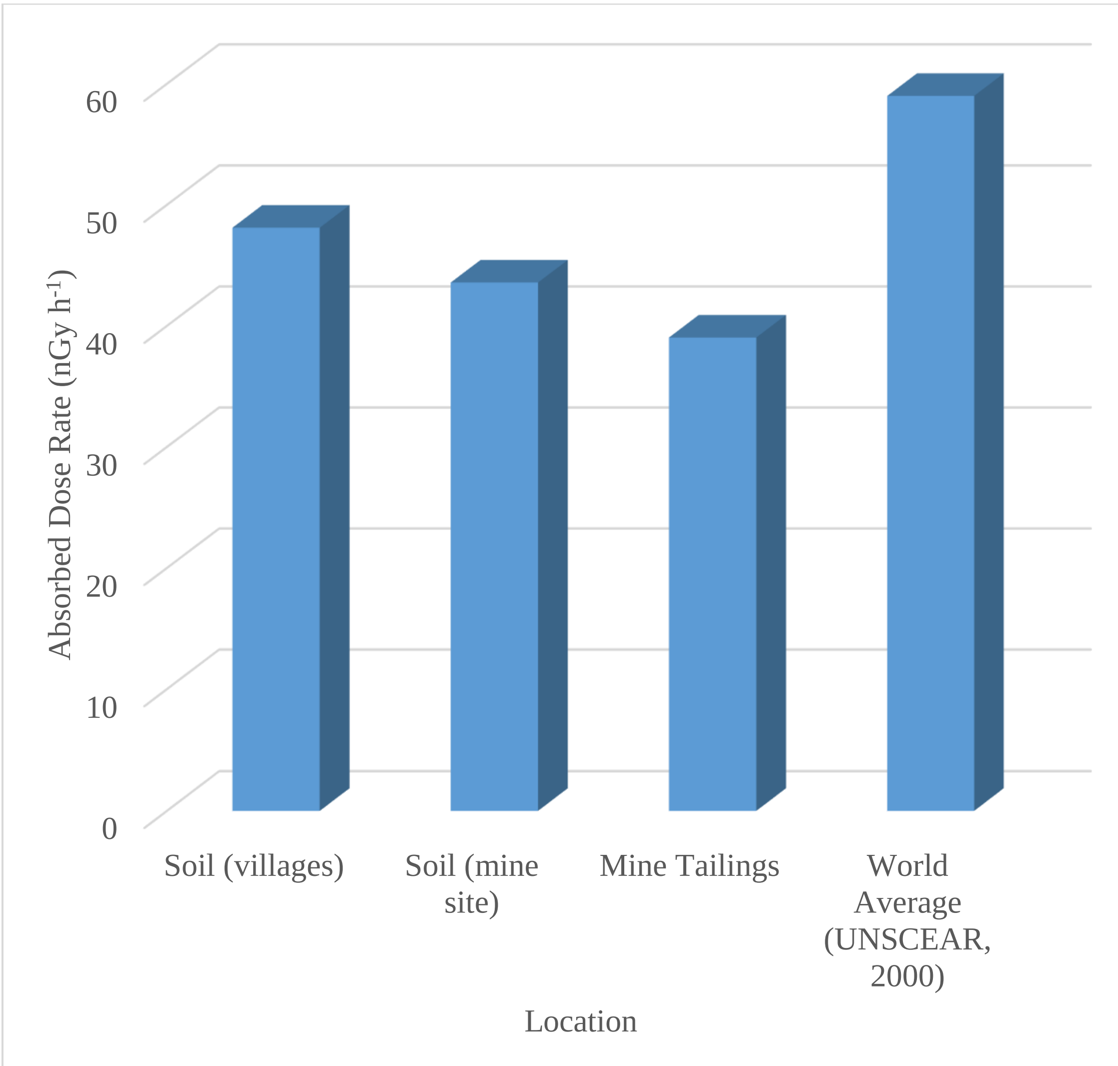
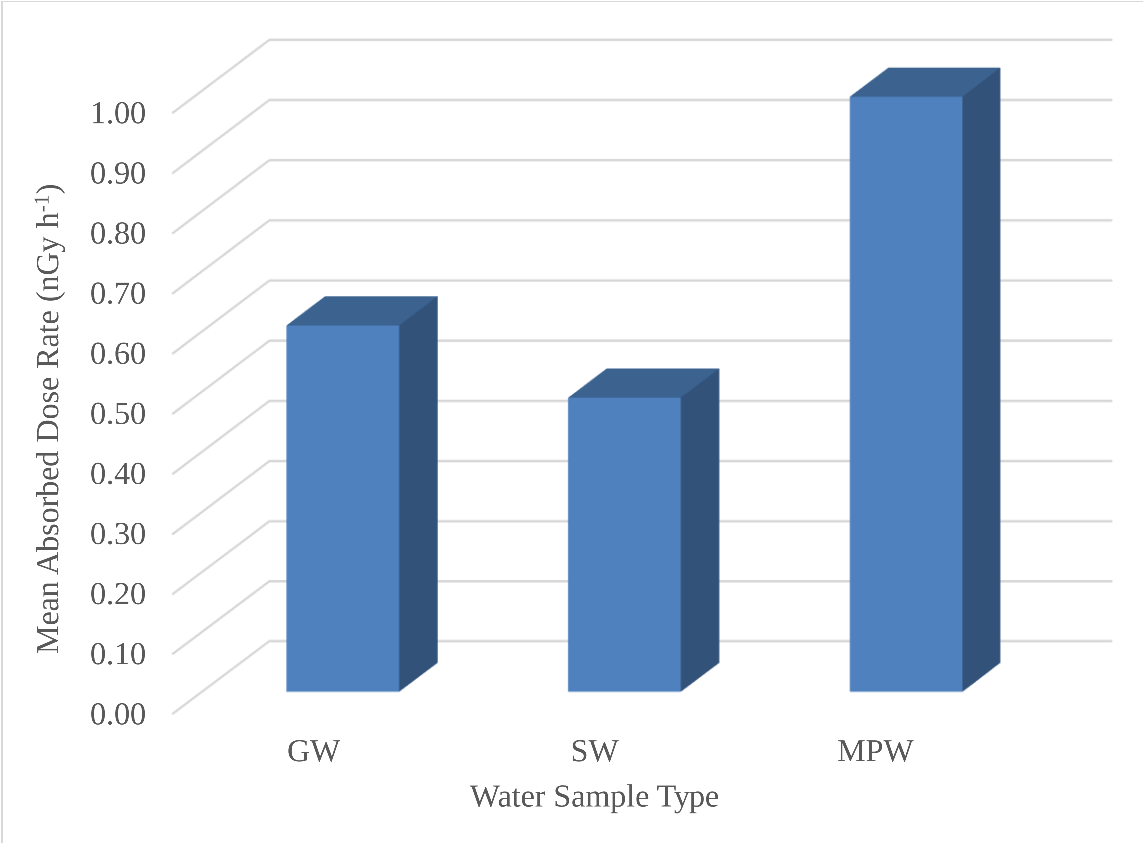


Figure 18: Comparison of mean absorbed dose rate in soil samples taken from different locations in the study area.



Legend: GW – Groundwater; SW – Surface water; MPW – Mine processed water

Figure 19: Comparison of mean absorbed dose rate in water samples taken from the study area.

The estimated annual effective dose equivalent received by members of the public due to ^{238}U, ^{232}Th and ^{40}K from the soil was calculated from the estimated average absorbed dose rate by multiplying dose conversion factor of 0.7 Sv Gy^{-1} and the occupancy factor for outdoor of 0.2 (5/24) (Veiga et al., 2006).

The intake of radionuclides with food and/or water is dependent on the concentration of radionuclides in the various food crops and water and on the food and water consumption. It is obvious that food and water consumption depends on many factors, some of which concern the individual while others are group related. Information on the range and amounts of foods consumed regularly by individuals is required. Types of food and water consumed are related, of course, to the specific geographical, as well as the cultural, economic, social and even political conditions within and amongst countries (WHO, 1988). The risk associated with an intake of radionuclides in the body is proportional to the total dose delivered by the radionuclides while staying in the various organs. In general it is assumed that stochastic effects occur linearly with dose and usually the effective dose equivalent is used to define this risk.

From ingestion of water, the estimated annual effective dose equivalent was calculated from the estimated absorbed dose rate by multiplying the daily water consumption rate of 2 litres per day and the conversion factor or dose per unit intake by ingestion of ^{238}U, ^{232}Th and ^{40}K for adult members of the public, i.e. 4.5 x 10^{-5} mSv Bq^{-1}, 2.3 x 10^{-4} mSv Bq^{-1} and 6.2 x 10^{-6} mSv Bq^{-1} for ^{238}U, ^{232}Th and ^{40}K, respectively, as reported by the International Commission on Radiological Protection (ICRP) (ICRP, 1996).

For ingestion of food crops, the estimated annual intakes and the internal dose conversion factors (IDCF) of 2.8 x 10^{-4}, 7.2 x 10^{-5} and 6.2 x 10^{-6} mSv per Bq for ^{238}U, ^{232}Th and ^{40}K, respectively were used (UNSCEAR, 2000).

The estimated annual effective dose due to external gamma irradiation ranges from 31.81 μSv in soil samples from Salman town to 89.79 μSv in soil samples from Nugget Hill with a mean annual effective dose of 57.23 μSv. This estimated mean annual dose from soil concentrations is lower than the world average value of 70 µSv y^{-1} (0.07 mSv y^{-1}) reported in UNSCEAR 2000 report (UNSCEAR, 2000).

From ingestion of water, the estimated annual effective dose ranges from 4.20 µSv in surface water to 8.50 µSv in mine processed water samples with mean annual effective dose equivalent of 5.20 µSv y^{-1} for ground water, 4.20 µSv y^{-1} for surface water and 8.50 µSv y^{-1} for mine processed water. These estimated annual effective dose equivalent values for the different water sample types are lower than the world average of 100.0 µSv y^{-1} (0.1 mSv y^{-1}) (WHO, 2004). Though the effective dose equivalents are within the acceptable values for water, the results revealed that the dose rates are highest in mine processed water even though it is not for domestic use. For domestic use, ground water samples have higher dose rate as compared with surface water due to the fact that the ground water is in close contact with the bedrocks and the high concentrations of some of the radionuclides. Generally, the doses obtained in the study are lower than the recommended reference level and from radiation protection point of view, life-long consumption of these waters may not cause any significant radiological health risk.

For food samples, the highest dose is received from the consumption of cassava and the least from maize. This high radionuclide intake was not due to high radionuclide concentration but may be due to the high consumption rate. Cassava and plantain are the staple food in the municipality. The estimated total annual dose equivalent from the consumption of the selected food crops is 1.54 mSv with a mean value of 0.39 mSv y^{-1}.

Table 4.13: Mean absorbed dose rate, mean annual effective dose, hazard indices, activity utilization index and estimated cancer risks from external gamma rays within the Adamus mine and surroundings villages

Location		Dose Rate ($nGy\ h^{-1}$)	Annual Effective Dose ($\mu Sv\ y^{-1}$)	Hazard Indices		Iγ	Estimated fatality cancer risk per year (10^{-6})	Estimated lifetime fatality cancer risk (ELCR) (10^{-3})
				H_{ex}	H_{in}			
Villages	Salman Town	27.84	34.14	0.16	0.45	0.45	1.95	0.14
	Akango	25.94	31.81	0.14	0.16	0.41	1.81	0.13
	Teberu	30.96	37.97	0.17	0.19	0.50	2.16	0.15
	Aliku Town	43.08	52.83	0.25	0.27	0.70	3.01	0.21
	Nkroful Bokrobo	52.09	63.88	0.31	0.33	0.85	3.64	0.26
	North Hill	61.71	75.68	0.37	0.39	1.01	4.31	0.30
	Nugget Hill	73.21	89.79	0.44	0.46	1.20	5.12	0.36
	Angajeleh	70.06	85.92	0.42	0.44	1.14	4.90	0.34
Mine site		43.58	53.45	0.24	0.29	0.69	3.05	0.21
Mine Tailings		39.04	47.87	0.23	0.25	0.63	2.73	0.19
Range		**25.94-73.21**	**31.81-89.79**	**0.14-0.44**	**0.16-0.46**	**0.41-1.20**	**1.81-5.12**	**0.13-0.36**
Mean		**46.75**	**57.33**	**0.27**	**0.32**	**0.76**	**3.27**	**0.23**

Table 4.14: Mean absorbed dose rate, mean annual effective dose and estimated cancer risks from ingestion of water from the Adamus mine and surroundings villages.

Water Type	Water Sample Code	Dose Rate (nGy h^{-1})	Annual Effective Dose (μSv y^{-1})	Estimated fatality cancer risk per year (10^{-7})	Estimated lifetime fatality cancer risk (ELCR) (10^{-5})
Ground water	SLHAP	0.60	5.17	2.95	2.06
	SLACB	0.72	6.16	3.5	2.46
	SLGCB	0.54	4.66	2.65	1.86
	SLBCB	0.58	4.97	2.83	1.98
	SLTWE	0.59	5.03	2.87	2.01
	Mean	**0.61**	**5.20**	**2.96**	**2.07**
Surface water	ANGST	0.44	3.79	2.16	1.51
	BANST	0.55	4.74	2.70	1.89
	ANBST	0.49	4.17	2.38	1.67
	TEBST 1	0.54	4.61	2.63	1.84
	TEBST 2	0.48	4.09	2.33	1.63
	BROST	0.44	3.78	2.15	1.51
	SALST	0.49	4.21	2.40	1.68
	Mean	**0.49**	**4.20**	**2.39**	**1.68**
Mine processed water	TSFSW	0.99	8.54	4.87	3.41
	TSFST 1	1.03	8.86	5.05	3.54
	TSFST 2	0.97	8.28	4.72	3.31
	NKBPW	0.97	8.32	4.74	3.32
	Mean	**0.99**	**8.50**	**4.85**	**3.39**

Table 4.15: Mean annual radionuclide intake, mean annual effective dose and estimated cancer risks from ingestion of the selected food crops from the Adamus mine and surroundings villages.

Food Crop Samples	Mean annual radionuclide intake (Bq)			Dose (mSv)	Estimated fatality cancer risk per year (10^{-5})	Estimated lifetime fatality cancer risk (ELCR) (10^{-3})
	^{238}U	^{232}Th	^{40}K			
Cassava	587.0	2283.0	56344.0	0.68	3.88	2.71
Cocoyam	253.0	721.0	17730.0	0.23	1.31	0.92
Maize	213.0	459.0	10154.0	0.16	0.91	0.64
Plantain	912.0	912.0	23702.0	0.47	2.68	1.88
Total	**1965.0**	**4375.0**	**107930.0**	**1.54**	**8.78**	**6.14**
Mean	**491.25**	**1093.75**	**26982.50**	**0.39**	**2.19**	**1.54**

The gamma ray radiation hazards due to the specified radionuclides in soil are assessed by calculating different indices. Even though the total activity concentration of radionuclides were calculated, it does not provide the exact indication about the total radiation hazards. The two indices are the external and internal radiation hazards. These indices were calculated (Table 4.9) by using the equation used by Oregun et al (2007). The calculated H_{ex}, H_{in} and $I\gamma$ values for the soil samples are 0.27 in the ranges of 0.14 - 0.44, 0.32 in the range of 0.16-0.46 and 0.76 in the range of 0.41-1.20, respectively. The mean values of the hazard indices (H_{ex} and H_{in}) are less than unity (permissible levels) (Orgun *et al.*, 2007). Similarly, the mean activity utilization index ($I\gamma$) is less than 2 ($I\gamma < 2$), which corresponds to an annual effective dose less than 0.3 mSv y^{-1} (El-Gamal *et al*., 2007).

The estimated cancer risk component for stochastic effects were evaluated from the estimated annual effective dose of the samples using ICRP methodology. The risk to the public was then estimated using the 2007 recommended risk coefficients in ICRP (103) report and assumed 70 years lifetime of continuous exposure of the public to low levels of radiation. The results of the estimated fatal cancer risk to the public per year ranged from 1.81 x 10^{-6} to 5.12 x 10^{-6} with the associated estimated lifetime fatality cancer risk of 0.13 x 10^{-3} to 0.36 x 10^{-3} for soil samples. For water samples, it ranges from 2.65 x 10^{-7} to 3.50 x 10^{-7}, from 2.15 x 10^{-7} to 2.70 x 10^{-7} and from 4.72 x 10^{-7} to 5.05 x 10^{-7} in ground, surface and mine processed water, respectively. The corresponding estimated lifetime fatality cancer risk is in the range of 1.86 x 10^{-5} to 2.46 x 10^{-5}, from 1.51 x 10^{-5} to 1.89 x 10^{-5} and from 3.31 x 10^{-5} to 3.54 x 10^{-5} for ground, surface and mine processed water, respectively.

For food crop samples, it ranges from 0.91×10^{-5} to 3.88×10^{-5} with a mean of 2.19×10^{-5} with a corresponding estimated lifetime fatality cancer risk of 0.64×10^{-3} to 2.71×10^{-3} with and a mean value of 1.54×10^{-3}.

From Tables 4.9-4.11, it is clear that the risk coefficient has a direct link with the number of Sieverts. The dose from ingestion of food crops is greater than from ingestion of water and from external irradiation from gamma sources in the soil (food crop samples > soil samples > water samples).

In terms of the estimated lifetime fatality cancer risk to the members of the public means that on the average approximately 2 out of 10,000 may suffer from some form of cancer fatality from external irradiation from gamma sources in the soil, 2 out of 100,000 may suffer from some form of cancer fatality from ingestion of only ground water, 1 out of 100,000 may suffer from some form of cancer fatality from ingestion of only surface water and 3 out of 100,000 may suffer from some form of cancer fatality from ingestion of mine processed water. For the ingestion of ground and surface water, 1 out of 100,000 may suffer from some form of cancer fatality and for ingestion of water from all the three sources, 2 out of 100,000 may suffer from some form of cancer fatality.

From ingestion of any of the selected food crops, on the average 1 out of 1,000 may suffer from some form of cancer fatality. Table 4.11 shows that the contribution of the consumption of cassava to the estimated lifetime fatality cancer risk is greater than the rest. The negligible cancer fatality risk value recommended by USEPA (1993; 2006) is in the range of 1.0×10^{-6} to 1.0×10^{-4} (i.e. 1 person out of 1 million or 10,000 suffering from some form of cancer fatality is considered trivial). Comparing the estimated results of the lifetime fatality cancer risk in the study with the acceptable risk factor, it can be concluded that, all the estimated results of the lifetime fatality cancer risk for members of the public in the Adamus mine and its surrounding villages due to external irradiation of gamma sources in the soil and ingestion of food crops and water in the study area are within the range of acceptable risk values recommended by USEPA (USEPA, 1993; 2006).

CHAPTER FIVE

5.0 CONCLUSION AND RECOMMENDATIONS

5.1 CONCLUSION

The aim of this research work was to assess the dose and estimate the cancer risks associated with public exposure NORM in the study area as a result of the mining and mineral processing activities using Adamu Gold mine in Ellembele District near Tarkwa as a case study. The three (3) exposure pathways considered for the study were; direct external gamma ray irradiation from natural sources in soil, internal exposure from ingestion of water containing naturally occurring radioactive materials and ingestion of some selected food crops (cassava, plantain, cocoyam and maize). The areas covered during this work include mine site, Salman, Akango, Aliku, Nkroful Bokorobo, North Hill, Nugget Hill, and Angajeleh.

The research interests in the environmental radiation pollution was to assess the radiation doses and radiation risks to humans. This was done using a gamma spectrometry in the following sequence: Radioactivity in soil, water and food crops, doses to humans (the Public) and estimating cancer risks. The study deals with the humans, who are constantly exposed to radioactive environment. The mining and milling of ores with significant amounts of uranium and thorium associated with the main ore which has the potential to pose undue health risks to members of the public. Ionizing radiation is especially harmful because it can change the chemical make-up of many things, including the delicate chemistry of the human body and other living organisms.

No investigations have been conducted to obtain data on the activity concentration levels of the natural radionuclides ^{238}U, ^{232}Th and ^{40}K in the area. Consequently, the radiation doses and risks associated with these radionuclides have never been investigated. High levels of these elements could pose chemical and/or radiological hazards.

In this study, the radioactivity levels of ^{238}U, ^{232}Th and ^{40}K in different types of samples, the corresponding radiation doses and cancer risks to members of the public were estimated. The mean activity concentrations of ^{238}U, ^{232}Th and ^{40}K in the soil samples were estimated to be 8.03, 43.98, 395.09 Bq kg^{-1}, respectively. For the water samples, the mean activity concentrations of ^{238}U, ^{232}Th and ^{40}K were 0.16, 0.83, 0.70 Bq l^{-1} respectively for ground water, 0.13, 0.66, 0.72 Bq l^{-1}, respectively, for surface water and 0.17, 1.45, 0.86 Bq l^{-1}, respectively, for mine processed water. The measured concentration of radioactive materials and the annual effective doses were within the range found in other countries. They were also found to be within the acceptable permissible levels for drinking water set by the WHO and therefore do not pose health problems to the populace of the host communities and do not affect the background ionization radiation of the environment

For the food samples the mean activity concentrations of ^{238}U, ^{232}Th and ^{40}K (dry weight) were 491.25, 1.93.75 and 26982.50 Bq kg^{-1}, respectively. The results in this study compared well with other studies carried out in other countries and with the worldwide average activity concentrations (UNSCEAR, 2000).

The ICRP philosophy of radiological protection aims at preventing deterministic effects and also reducing the occurrence of stochastic effects of cancer to acceptable levels. This is achieved by a system of protection that requires justification of practice to ensure it produces a net benefit, optimisation of protection to keep exposures as low as reasonably achievable (ALARA) and the protection of individuals by imposing either dose limits or controls on the risks from potential exposures. As a result, the potential exposure of the public in the study area was assessed by estimating the annual effective doses in various samples and the total annual effective dose was estimated from the mean annual effective doses from all the exposure pathways considered for purposes of comparison with recommended dose limits.

The estimated mean annual effective were 57.33 $\mu Sv\ y^{-1}$, 5.97 $\mu Sv\ y^{-1}$ and 390.00 $\mu Sv\ y^{-1}$ for soil, water and food crop samples, respectively. The estimated total annual effective dose for all the exposure pathways was 453.30 $\mu Sv\ y^{-1}$ (0.45 $mSv\ y^{-1}$). The total annual effective dose is lower than the 1 mSv per year dose limit recommended by the ICRP for public radiation exposure control.

The hazard indices of the soil samples are less than Unity (permissible level) implying that the mining activities do not pose any significant radiological hazard to the communities in this area. The radiological hazards to the public in the study area were assessed based on the calculation of hazard indices (external and internal) and activity utilization index for the soil samples. The external and internal hazard indices are less than Unity (permissible level). The calculated activity utilization index is less than two; this indicates that soils from the study area can be safely used for construction of buildings. This is an important information for the local people in terms of utilize the soils.

The cancer risks to members of the public, from exposure to naturally occurring radioactive materials (NORM) as a result of the mining and mineral processing activities of Adamus Goldmine using the ICRP risk assessment methodology for fatal cancer risk was estimated. The estimated mean lifetime cancer risk for soil samples is 0.23 x 10^{-3}, 2.46 x 10^{-5} for ground water, 1.68 x 10^{-5} for surface water and 3.39 x 10^{-5} for mine processed water respectively. For food crop samples, the estimated mean lifetime cancer risk is 1.54 x 10^{-3}.

The estimated lifetime fatality cancer risks from all the samples considered varied from 0.13 x 10^{-6} in soil to 2.71 x 10^{-3} in food crop. The total lifetime cancer risk was estimated to be 1.57 x 10^{-3}. This, in terms of the lifetime fatality cancer risk means, that approximately 2 out of 1000 may suffer from some form of cancer fatality. The negligible cancer fatality risk value recommended by USEPA is in the range of 1 x 10^{-6} to 1 x 10^{-4} (i.e. 1 person out of 1 million or 10,000 suffering from some form of cancer fatality). The estimated values of the lifetime cancer risks in this study exceeded the range of acceptable risk for all the samples put together. Also, the estimated net lifetime risk was above the world average of 0.29 x 10^{-3} given by UNSCEAR (2000). The data shows that the estimated cancer risk maximum due to the high consumption rate of cassava food crop.

The results obtained in this study show that the background radiation levels are within the natural limits and compared well with similar studies for other countries. The data from this study can be used as baseline for future investigations.

The results of the estimated doses of the other sources of exposure compare quite well with earlier studies on radioactivity in other mines in Ghana and elsewhere (Darko et al., 2010; UNSCEAR, 2000). This study considers that the ingestion of food crops (cassava and plantain) could be the most significant mode of exposure in the study area. On the basis of the results from this study, consumption of food crop and water do not pose any significant source of radiation hazard to the population. The results from this study could form part of data collection to help in the development of reference levels and the develop guidelines for the regulation of NORM in Ghana for radiation protection of workers and the public.

Therefore, the study area is still in the zone of normal radiation level, which leaves the soil and water radioactivity there less a threat to the environment as well as to human health. However, this data may provide a general background level for the area studied and may also serve as a guideline for future measurement and assessment of possible radiological risk to human health in the region.

5.2 RECOMMENDATIONS

This is a baseline study for assessing the natural radioactivity levels due to the activities of Adamus Goldmine and the possible radiological risk to human health. In order to make the study more comprehensive, the following areas are recommended for further research in future:

1. A study should done to determine the gross alpha and gross beta activity concentration in drinking water sources in the study area using a gross alpha and beta counter.
2. A study should done on the assessment of the ^{226}Ra and ^{222}Rn emanation coefficient for technologically enhanced naturally occurring radioactive material (TENORM) scales in pipes of the gold treatment plants in Ghana.
3. A study should be extended to other food crops, including vegetables.
4. A study should also be done to see the effect of heat on the radioactivity levels during the preparation of food crops into food for consumption.
5. The study should be extended to the other mines to help come out with a national guidelines on radioactivity levels and associated dose levels for radiation protection purposes.

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APPENDICES

Appendix A

Calibration

The high purity germanium detector used for the study was calibrated for energy and efficiency. Standard radionuclides, homogenously distributed in solid water matrix, in a one (1) litre Marinelli beaker geometry with density approximately 1 g cm^{-3} was used. The standard source was used to calibrate the system for the soil, water and food samples. Efficiency calibration is geometry dependent and necessary for the quantification of ^{238}U, ^{232}Th and ^{40}K radionuclides in particular geometry. The soil, water and food samples were measured in a 1 litre Marinelli beaker on a high purity germanium detector (HPGE).

The energy calibration curves of the detector using standard radionuclides in the Marinelli beaker is shown figure 12 with a correlation of R^2 =1. The energy calibration curves indicate the correlation between the energy of radionuclide and the corresponding channel number at the centroid of a full energy peak. The plot fitted a linear function which indicates that the detector system is performing well for the matrix of standard radionuclides used for the energy calibration.

The efficiency calibration curve is also shown in figure 13 with a correlation coefficients of R^2 = 0.989 for energies between 100 and 2000 keV. The equation for the efficiency curve obtained is

$$Y = 1.127X^{-0.7057} \qquad (1)$$

The generally accepted expression for efficiency calibration is given by equation (3) (IAEA, 1989):

$$\ln \varepsilon = a_1 + a_2 \ln E \tag{3}$$

where ε is absolute full energy event efficiency, a_1 and a_2 are the fit parameters, E is the nergy (keV). By applying natural log of both sides of equations (1) and (2) will result in identical equations as equation 3.

$$\ln \varepsilon = 0.1196 – 0.7057\ln E$$

The expression is suitable for determining efficiencies of gamma energies between 100 keV and 2000 keV.

The energy resolution of the detector measured at 1332 keV of a ^{60}Co source was 0.19%. The typical range of the resolution of HPGe detectors is 0.1-0.2% (Sood et al., 1981). The measured resolution of the detector used for this study is within the range of the resolution of HPGe detectors which also indicates that the detection system was performing well and suitable for use in this study.

The results of the minimum detectable activities of ^{238}U, ^{232}Th and ^{40}K which were determined by measuring a 1 L Marinelli beaker filled with distilled water on the detector are shown in table 5.1. The minimum detectable activities of ^{238}U, ^{232}Th and ^{40}K were estimated to be 0.12, 0.11 and 0.15 Bq kg^{-1} respectively. The minimum detectable activities of these radionuclides depends on a number of factors including the background radiation in the area, adequacy of shielding from environmental background radiation and the inherent activity concentrations of these radionuclides in the sample containers.

Energy calibration results for 1.0 Litre Marinelli beaker Geometry

Nuclide	Gamma ray energy (keV)	Channel Number
Cadmium-109	85.13	67
Cobalt-57	121.98	96
Caesium-137	662	521
Cobalt-60	1174.07	924
Cobalt-60	1334.17	1050
Yttrium-88	1838.61	1447

Radionuclides used for efficiency calibration

Energy (keV)	Efficiency
122	0.0383
662	0.0120
1173	0.0072
1332	0.0064
1836	0.0062

The mixed standard source that was used for the energy and efficiency calibrations of the gamma detector has the following specifications as shown in table 3.1.

Geometry of Reference Source

Source number: NW146

Volume: Approximately 1000 ml

Density: Approximately 1.0 g/m^3

Reference date: 1st February, 2006 at 12:00 GMT.

Appendix B

Sampling points of soil samples and sample code

Type of water sample: Groundwater samples

Name of Location	Sample Code	Mass of Sample (kg)	Date of Collection	Mass of Sample (kg)	Date of Collection
Salman Hand Pump	SLHAP	0.93	24/04/2012	0.94	6/08/2013
Salman Ash Quarters Borehole	SLACB	0.92	24/04/2012	0.93	6/08/2013
Salman Green Quarters Borehole	SLGCB	0.92	24/04/2012	0.91	6/08/2013
Salman Blue Quarters Borehole	SLBCB	0.93	24/04/2012	0.92	6/08/2013
Salman Well	SLTWE	0.93	24/04/2012	0.92	6/08/2013

Type of water sample: Surface water samples

Name of Location	Sample Code	Mass of Sample (kg)	Date of Collection	Mass of Sample (kg)	Date of Collection
Anganjele Stream	ANGST	0.93	25/04/2012	0.92	7/08/2013
Bangara Stream	BANST	0.93	25/04/2012	0.94	7/08/2013
Angajeleh + Bangara Stream	ANBST	0.93	25/04/2012	0.94	7/08/2013
Teberu Stream 1	TEBST 1	0.94	25/04/2012	0.93	7/08/2013
Teberu Stream 2	TEBST 2	0.94	25/04/2012	0.94	7/08/2013
Broma stream	BROST	0.93	25/04/2012	0.92	7/08/2013
Salman Stream	SALST	0.94	25/04/2012	0.94	7/08/2013

Type of water sample: Mine processed water samples

Name of Location	Sample Code	Mass of Sample (kg)	Date of Collection	Mass of Sample (kg)	Date of Collection
TSF Seepage Water	TSFSW	0.92	26/04/2012	0.93	8/08/2013
TSF Silt Trap 1	TSFST 1	0.91	26/04/2012	0.90	8/08/2013
TSF Silt Trap 2	TSFST 2	0.93	26/04/2012	0.93	8/08/2013
Nkroful Bokrobo Pit Water	NKBPW	0.92	26/04/2012	0.93	8/08/2013

Appendix C:

Absorbed dose rates in air and estimated annual effective doses at 1 meter above the ground at the soil sampling points.

Sample code	Measured dose rate (nGy h^{-1})		Calculated Effective Dose (μSv y^{-1})	Location
	Range	Average		
SS_1	58-60	59	72.36	Salman Town
SS_2	57-59	58	71.13	
SS_3	56-58	57	69.90	
SS_4	57-59	58	71.13	
SS_5	57-59	58	71.13	
SS_6	50-52	51	62.55	Akango
SS_7	51-53	52	63.77	
SS_8	52-54	53	65.00	
SS_9	50-52	51	62.55	
SS_{10}	52-54	53	65.00	
SS_{11}	58-60	59	72.36	Teberu
SS_{12}	57-59	58	71.13	
SS_{13}	56-58	57	69.90	
SS_{14}	57-59	58	71.13	
SS_{15}	57-59	58	71.13	
SS_{16}	49-51	50	61.32	Aliku Town
SS_{17}	50-52	51	62.55	
SS_{18}	50-52	51	62.55	
SS_{19}	51-53	52	63.77	
SS_{20}	50-52	51	62.55	
SS_{21}	58-60	59	72.36	Nkroful Bokrobo
SS_{22}	59-61	60	73.58	
SS_{23}	57-59	58	71.13	
SS_{24}	59-61	60	73.58	
SS_{25}	57-59	58	71.13	
SS_{26}	50-52	51	62.55	North Hill
SS_{27}	51-53	52	63.77	
SS_{28}	52-54	53	65.00	
SS_{29}	50-52	51	62.55	
SS_{30}	52-54	53	65.00	
SS_{31}	53-55	54	66.23	Nugget Hill
SS_{32}	52-54	53	65.00	
SS_{33}	52-54	53	65.00	
SS_{34}	54-56	55	67.45	
SS_{35}	54-56	55	67.45	
SS_{36}	61-63	62	76.04	Angajeleh
SS_{37}	63-65	64	78.49	
SS_{38}	61-63	62	76.04	
SS_{39}	62-64	63	77.26	
SS_{40}	63-65	64	78.49	

SS_{41} SS_{42} SS_{43} SS_{44} SS_{45}	61-63 63-65 61-63 62-64 63-65	62 64 62 63 64	76.04 78.49 76.04 77.26 78.49	Plant site (within the plant)
Average	**49– 65**	**56**	**68.56**	

Appendix D:

Soil sampling points within the mine and its surrounding communities.

Sample code	Location	Location coordinates
SS_1	Salman Town	N 4° 57’22.02” W 2° 19’ 31.21”
SS_2		N 4° 57’55.11” W 2° 19’ 22.26”
SS_3		N 4° 57’13.42” W 2° 19’ 23.73”
SS_4		N 4° 57’59.40” W 2° 19’ 34.22”
SS_5		N 4° 57’01.28” W 2° 20’ 36.34”
SS_6	Akango	N 4° 47’59.76” W 2° 21’ 10.07”
SS_7		N 4° 48’02.74” W 2° 22’ 04.56”
SS_8		N 4° 47’49.91” W 2° 21’ 22.90”
SS_9		N 4° 49’44.03” W 2° 20’ 11.85”
SS_{10}		N 4° 49’44.03” W 2° 20’ 11.85”
SS_{11}	Teberu	N 4° 47’ 49.84” W 2° 21’ 22.82”
SS_{12}		N 4° 47’ 49.84” W 2° 21’ 22.82”
SS_{13}		N 4° 51’29.52” W 2° 22’ 54.87”
SS_{14}		N 4° 51’29.52” W 2° 22’ 54.87”
SS_{15}		N 4° 48’21.23” W 2° 21’ 04.28”
SS_{16}	Aliku Town	N 4° 49’48.63” W 2° 22’ 18.27”
SS_{17}		N 4° 49’15.26” W 2° 21’ 44.83”
SS_{18}		N 4° 48’58.62” W 2° 21’ 23.70”
SS_{19}		N 4° 49’29.66” W 2° 21’ 21.76”
SS_{20}		N 4° 49’22.34” W 1° 28’ 36.40”
SS_{21}	Nkroful Bokrobo	N 4° 48’57.84” W 1° 28’ 37.02”
SS_{22}		N 4° 48’52.03” W 1° 29’ 47.23”
SS_{23}		N 4° 48’ 47.44” W 1° 29’ 56.72”
SS_{24}		N 4° 51’54.82” W 1° 29’ 58.46”
SS_{25}		N 4° 49’18.99” W 2° 29’ 00.87”
SS_{26}	North Hill	N 4° 49’47.45” W 1° 28’ 59.53”
SS_{27}		N 4° 42’24.39” W 2° 21’ 07.49”
SS_{28}		N 4° 52’59.51” W 2° 33’ 55.51”
SS_{29}		N 4° 49’56.60” W 2° 20’ 11.36”
SS_{30}		N 4° 49’56.60” W 2° 20’ 11.36”
SS_{31}	Nugget Hill	N 4° 49’08.02” W 2° 20’ 25.21”
SS_{32}		N 4° 47’10.90” W 2° 23’ 33.46”
SS_{33}		N 4° 47’38.31” W 2° 22’ 34.25”
SS_{34}		N 4° 47’59.82” W 2° 22’ 59.89”
SS_{35}		N 4° 48’10.83” W 2° 20’ 26.59”
SS_{36}	Angajeleh	N 4° 47’53.71” W 2° 20’ 43.66”
SS_{37}		N 4° 47’53.71” W 2° 20’ 43.66”
SS_{38}		N 4° 47’53.71” W 2° 20’ 43.66”
SS_{39}		N 4° 47’53.71” W 2° 20’ 43.66”
SS_{40}		N 4° 47’53.71” W 2° 20’ 43.66”

SS_{41} SS_{42} SS_{43} SS_{44} SS_{45}	Within the Plant	N 4º 57'29.02" W 2º 29' 41.21" N 4º 57'25.11" W 2º 29' 42.26" N 4º 57'23.42" W 2º 29' 43.73" N 4º 57'29.40" W 2º 19' 44.22" N 4º 57'21.28" W 2º 29' 46.34"

Appendix E:

Water sampling points within the mine and its surrounding communities.

Sample code	Location	Location coordinates	Description of samples
WS_1	Salman	N 4º 51’22.02” W 2º 21’ 31.21”	Hand pump water
WS_2	Salman Ash Quarters	N 4º 51’55.11” W 2º 21’ 22.26”	Borehole water
WS_3	Salman Green Quarters	N 4º 51’13.42” W 2º 21’ 23.73”	Borehole water
WS_4	Salman Blue Quarters	N 4^0 50’59.40” W 2º 21’ 24.22”	Borehole water
WS_5	Salman	N 4º 51’01.28” W 2º 20’ 26.34”	Well water
WS_6	Anganjeleh	N 4º 47’59.76” W 2º 22’ 10.07”	Stream water
WS_7	Bangara	N 4º 48’02.74” W 2º 22’ 44.56”	Stream water
WS_8	Angajeleh / Bangara	N 4º 47’49.91” W 2^0 21’ 22.90”	Stream water
WS_9	Teberu	N 4º 49’44.03” W 2º 20’ 21.85”	Stream 1 water
WS_{10}	Teberu	N 4º 49’44.03” W 2º 20’ 11.85”	Stream 2 water
WS_{11}	Broma	N 4º 47’ 49.84” W 2º 21’ 22.82”	Stream water
WS_{12}	Salman	N 4º 47’ 49.84” W 2º 01’ 22.82”	Stream water
WS_{13}	Tailings Storage Facility (TSF)	N 4º 51’29.52” W 2º 22’ 54.87”	Seepage water
WS_{14}	TSF	N 4º 51’29.52” W 2º 22’ 54.87”	Silt Trap 1 water
WS_{15}	TSF	N 4º 48’21.23” W 2º 21’ 04.28”	Silt Trap 2 water
WS_{16}	Nkroful Bokrobo	N 4º 49’48.63” W 2º 22’ 18.27”	Pit water

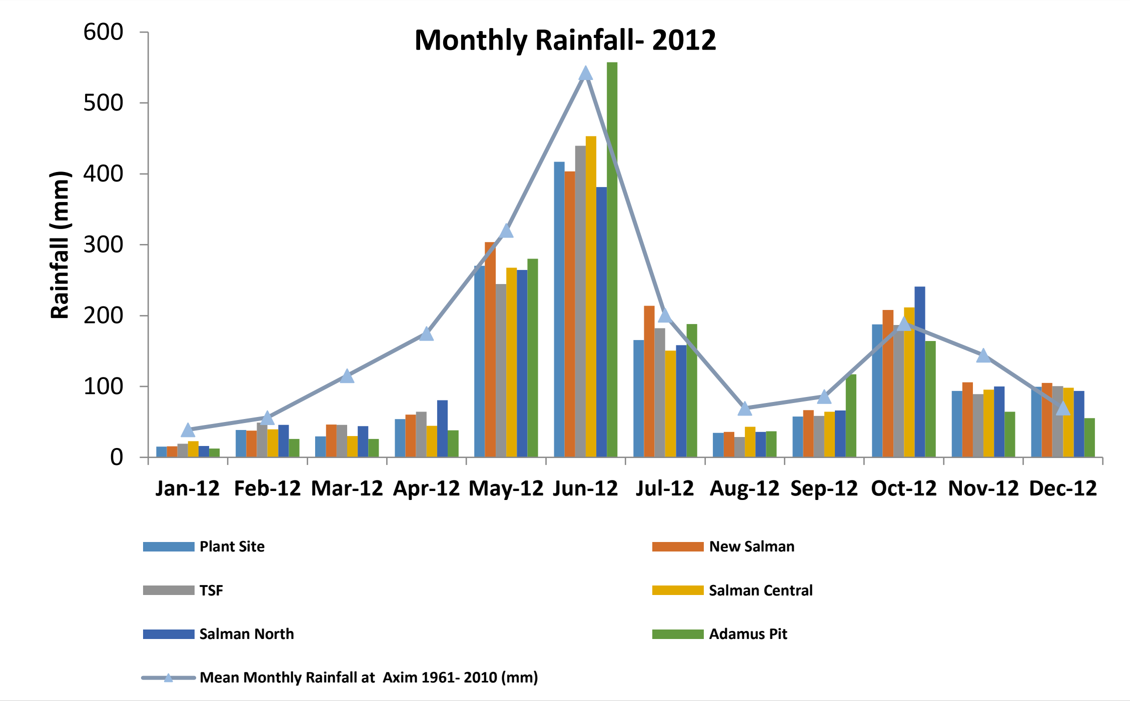
Appendix F:

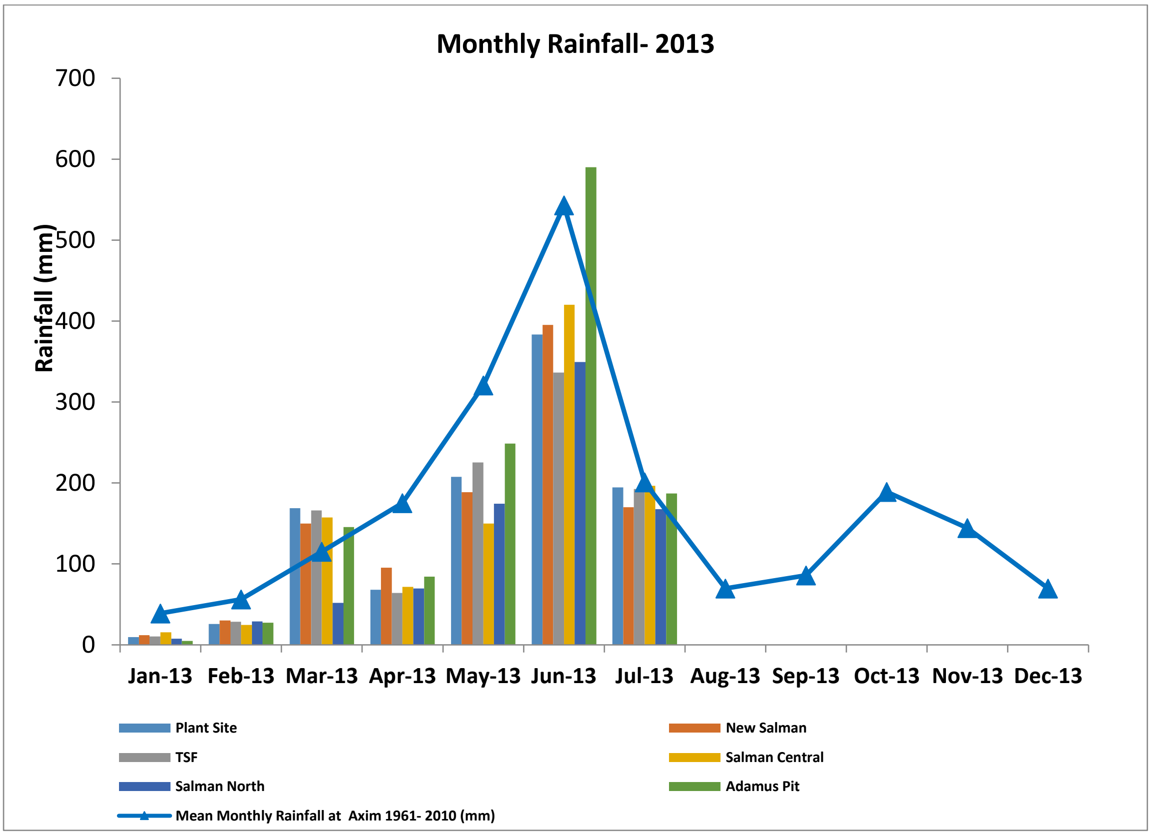
Food crop sampling points within the mine and its surrounding communities.

Sample code	Location	Location coordinates	Description of samples
FS_1	Salman	N 4^0 51'22.02" W 2^0 21' 31.21"	
FS_2	Akango	N 4^0 49'18.99" W 2^0 20' 00.87"	Cassava / Cocoyam / Maize / Plantain
FS_3	Teberu	N 4^0 49'56.60" W 2^0 20' 11.36"	
FS_4	Angajeleh	N 4^0 49'08.02" W 2^0 20' 25.21"	

Appendix G:

Monthly Rainfall Totals (mm) for 2012-2013





Appendix H:

Mean Daily Temperature (°C)

Year	Jan	Feb	March	April	May	June	July	Aug	Sept	Oct	Nov	Dec
1961	26.7	27.4	28.1	27.4	27.6	26.4	24.9	24.4	248	24.8	26.7	26.7
1962	27.3	28.1	27.6	27.4	27.1	26.5	25.3	26.2	26.2	26.1	26.5	26.8
1963	27.8	27.2	27.4	27.6	27.1	26.5	25.2	26.1	26.2	26.1	26.8	26.7
1964	26.7	28.2	27.8	26.8	25.7	26.1	25.4	25.1	25.2	26.6	26.3	26.2
1965	26.1	26.7	27.6	27.3	27.1	26.2	25.1	25.1	25.6	26.6	27.1	26.8
1966	27.8	27.5	27.9	27.4	27.5	26.2	26.4	25.3	25.3	26.1	26.9	27.2
1967	26.6	28.1	26.7	27.9	27.6	25.5	24.9	24.5	24.5	25.2	26.2	27
1968	25.7	26.8	26.8	26.9	26.4	25.4	25.3	25.1	24.8	25.6	26.1	26.4
1969	26.6	28.1	27.6	27.9	26.9	25.5	24.9	24.5	24.5	25.2	26.2	27
1970	26.7	27.7	27.8	27.7	26.8	25.6	25.1	24.5	24.6	26.1	26.1	26.6
1971	26.4	26.6	26.9	26.6	26.9	25.4	25.3	24.2	24.5	25.6	26.2	25.7
1972	26.7	27.1	27.1	26.8	26.6	25.7	25.1	23.8	24.6	26.1	26.4	26.8
1973	27.4	28.2	27.7	27.7	26.7	25.9	25.6	25.4	25.5	26.1	26.6	26.1
1974	26.1	27.3	27.3	27.1	26.5	25.5	25.3	26.5	24.6	25.5	26.2	26.1
1975	26.1	27.2	27.2	27	26.5	25.7	24.7	24.3	24.6	25.7	25.9	26.2
1976	25.8	26.4	27.2	27	26.3	25	24.5	24	24.3	25.1	26	26.4
1977	26.4	27.7	27.9	28	26.9	25.6	25.3	24.5	25.5	26.3	26.7	26.4
1978	27.3	27.6	27.6	26.5	26.7	25.5	24.7	24.7	25.2	26.8	26.5	27
1979	27.7	27.8	28	27.8	26.7	25.9	25.1	25.5	25.9	26.1	26.5	26.9
1980	27.2	27.6	27.9	27.9	26.5	26.3	25.3	25.3	25.7	26	26.4	25.6
1981	26.7	28.2	27.7	28.1	26.6	26.2	25.3	25	25.7	26.6	26.9	27.5
1982	27	27.9	28	27.9	26.4	26.5	25.6	24.4	25.1	25.9	26.9	27
1983	26	28.1	28.9	28.4	26.6	26.4	25.5	24.8	25.4	26.6	27.1	27
1984	26.9	27.7	27.8	28	26.8	26.5	25.5	25.6	25.5	25.9	26.2	26.3
1985	27	27.4	27.6	27.9	26	25.6	25.7	25.5	25.3	26.5	26.2	26.1
1986	26.4	27.1	27.1	27.9	27	26	24.7	24.5	25.3	25.9	26.3	26.2
1987	27.3	27.3	27.7	28.6	27.5	26.1	25.7	25.6	25.8	26.2	27.4	26.7
1988	26.7	28	27.5	27.2	26.9	25.5	25.3	24.6	24.8	25.7	26.3	26.4
1989	26.2	26.4	27.6	27.6	27	25.8	24.7	23.7	25.3	26.4	27.1	26.6
1990	26.9	26.5	28.8	27.9	26.8	26.1	26.1	25.2	25	26.3	27.3	27.2
1991	27.3	28.1	28	27.4	27	26.9	25.9	24.9	25.5	25.5	26.4	27
1992	26.7	28.2	28	27.9	26.8	26	24.8	24.7	25.3	26.2	26.1	26.9
1993	26.1	27.4	26.3	26.9	27.4	26.1	25.5	24.7	26.1	26.7	26.9	27
1994	26.5	27.7	27.7	27.4	26.9	26.2	25.2	24.8	25.2	25.6	26.4	26.8
1995	26.2	28.2	27.6	28	27.4	26.8	26.2	25.8	25.5	26.4	27	27.1
1996	27.4	27.9	27.8	27.3	26.9	25.6	25.4	24.7	24.8	26.3	26.7	27.1
1997	27.2	27.7	27.9	27.4	27	25.9	25.5	24.7	26	27.2	27.5	27.8
1998	27.8	29.5	29.9	29.3	28.1	26.9	26.1	25.2	26.2	26.8	27.6	27.7
1999	27.5	27.5	28.3	27.7	27.4	26.7	26.1	25.4	25	25.9	26.9	27.2
2000	27.1	27.5	28.3	28	27.5	26.4	25.6	25.2	25.4	26.1	27.4	27.2
Mean	26.8	27.6	27.7	27.6	26.9	26	25.3	25	30.8	26.1	26.6	26.7