Assessment and Investigation of Natural Radioactivity Levels in Leaves of Khat and Agricultural Soil Samples Collected from Haramaya, East Hararghe, Ethiopia, Using Gamma Spectrometry

A thesis Submitted to the Department of Physics, Addis Ababa University

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Declaration

With the approval of my supervisor, I submitted this thesis to the Department of Physics at Addis Ababa University as my original work.

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Abstract

The radiation released from natural radioactive materials (commonly, ^{40}K,^{238}U, and ^{232}Th series) performing natural radioactivity, which is out of control, absorbed by the human's body, arising from a natural source such as cosmic ray, terrestrial, and exposure from inhalation or intake radiation sources. Those natural radionuclides are typically present in rock and soil in different amounts and levels of activity, with subsequent distribution to the environment and transfer to humans through inhalation and food ingestion (soil-plant-humans) causes health risks.

This study aims to assess and investigate radioactivity levels, radiological hazard indices in six samples of leaves of Khat plants, and six of its soil (0-30 cm deep) samples collected from Haramaya University area. The measurement had been carried out using a high-efficiency gamma-ray spectrometer with Digital Signal Analyser, (DSA) coupled with a High Purify Germanium (HPGe), p-type detector. The gamma-ray spectrometers are controlled by GENIE(2K)-high channel number performance, computer software, and output result are displayed on personal computer. The investigations identify naturally occurring radionuclide's from ^{238}U, and ^{232}Th series, and ^{40}K, in addition to those, ^{137}Cs in agricultural soil and ^{54}Mn in leaves of Khat samples. The activity concentrations of ^{238}U, ^{232}Th, and ^{40}K in Khat farm soils samples range from 21.43 ± 0.98 to $30.77 \pm 0.62, Bqkg^{-1}$, with a mean value of $23.72 \pm 0.85 Bqkg^{-1}$, 69.27 ± 3.17 to $168.78 \pm 7.55, Bqkg^{-1}$, with an average value of $111.50 \pm 4.21 Bqkg^{-1}$, and 542.15 ± 22.73 to $1045.96 \pm 43.4, 4Bqkg^{-1}$, with an average value of $794 \pm 26.44 Bqkg^{-1}$, respectively. The activity concentrations of ^{40}K and ^{232}Th obtained in all samples of the soils are higher than the recommended reference levels. The radium equivalent activity range from 221.61 ± 9.83 to $331.57 \pm 13.81, BqKg^{-1}$, with an average value 244.31 $BqKg^{-1}$, which is relatively higher. Gamma-ray radiation hazards caused by primordial radionuclides of ^{40}K, ^{238}U and ^{232}Th, evaluated were, the absorbed dose rate range from 76.52 ± 3.40 to $152.57 \pm 6.7, nGyh^{-1}$, with a mean value of $113.74 \pm 4.12 nGyh^{-1}$, which is higher than the global mean value, annual effective dose range from 0.09 ± 0.00 to $0.19 \pm 0.01, mSvy^{-1}$, with an average value of $0.14 \pm 0.01 mSvy^{-1}$, which is above the world average. The calculated values of the external and internal index for agricultural soil samples range from 0.44 to 0.72, with a mean value of 0.63, and 0.51 to 0.96, with a mean value of 0.72, respectively, which are relatively high but less than the world permissible value. Also the result of radioactivity level index (I_γ) in soils sample is ranging from 0.61 ± 0.03 to 1.21 ± 0.05, with an average values of 0.90 ± 0.03 , where estimated values of I_γ in some soil samples are higher than one and an average relative

higher.

The average concentrations of ^{238}U, ^{232}Th, and ^{40}K in leaves of Khat samples were 1.32 ± 0.24, 10.36 ± 0.96, and 273.39 ± 7.39, $Bqkg^{-1}$, respectively. The radium equivalent activity, and internal hazard indices calculated to this plant were varied from 25.20 ± 2.471 to 39.50 ± 2.137, $BqKg^{-1}$ with average 34.44 ± 2.186 $BqKg^{-1}$ and from 0.05 to 0.11, with average 0.09, respectively. The average ingestion effective dose due to intake this plant samples ^{238}U, ^{232}Th, and $40K$ calculated for adult is, 0.2±0.04, 1.31 ±0.12, and 0.81±0.03, mSv/y,respectively, with the total summation an average, 2.32±0.18 $mSvy^{-1}$. These values are higher than the permissible limits of the world's total average ingestion effective dose. The obtained results were compared with the international recommended values.

Key word: Natural Radioactivity, Khat Plants, Soil, Gamma ray spectrometry, HPGe detector, Activity Concentration, and Radiological Hazard.

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1 Introduction

Radionuclides are unstable nuclei that arise naturally in the environment constitute the majority of natural background radiation [11]. Those natural radionuclides include the primordial radioactive elements, their radioactive decay products, and radionuclides produced by cosmic-radiation interactions are present in soils at various levels of activity, with subsequent distribution to the environment and transfer to humans via inhalation and food consumption (soil-plant-humans), resulting in human health risk. Only a few cosmogenic radionuclides, such as ^{6}C and ^{22}Na, are formed in sufficient numbers and have long enough lives to provide significant radiation to the environment. The primordial radionuclides U, Th, and ^{40}K are present in various amounts in the soil, depending on the parents rock's composition during soil formation [6, 12]. Natural events (such as volcanic eruptions) and man-made acts (such as poor farming practices) expose previously unknown radioactive materials in the earth's crust and increase the number of these natural radionuclides that poisoned the environment. As a result, studying radioactive components in soils is an important aspect of understanding radioactivity's behavior in the ecosystem, because of radionuclides concentration in soil is an indicator of radioactive accumulation in the environment, which has an impact on humans and animals [13].

Humans are exposed to radioactivity in a variety of ways, the most common of which is through external and internal exposure to background radiation in soil, which can contribute to environmental radioactivity. The external radiation from the natural background is cosmic rays and natural radionuclides of ^{40}K, and ^{238}U, ^{232}Th , and their progenies. However,the internal radiation exposure caused by γ-radiation is originating from radioactive material enter the body through inhaled of ^{222}Rn and through ingestion food mainly from ^{40}K, and ^{238}U, ^{232}Th and their daughter products. Those long-lived naturally occurring radionuclide may be transferred to plants together with the nutrients during mineral uptake, grow in other parts, and even reach the edible parts. In addition to uptake, direct deposition on leaf surfaces may occur, and the contaminants may be absorbed metabolically by the plants or transferred directly to animals and humans who ingest the contaminated foliage [13–15].

Gamma Spectrometry System can be used to measure the qualitative and quantitative properties of a radionuclide that emits gamma radiation. The accuracy of the measurement is highly dependent on the performance of the various parts of detectors system, and one of the most well-known types of detector is HPGe detector, which is designed to detect low levels of natural and artificial environmental radiation.

In this sudy, the analyzed samples are agricultural soils and leaves of Khat plants collected from Haramaya univeristy area, east Hararghe, Ethiopia using Canberra p-type high purity germanium (HPGe) coaxial detector with 1.9 keV relevant resolutions at 1332.5 keV, a relative efficiency of 70% and multichannel analyzer of 8192 channel performance.

Khat(Catha-edullis) is psychoactive shrubs, or is a plant whose leaves are chewed for its stimulant effect. The origin of these plants, many believe Ethiopian, others state that Khat originated in Yemen before spreading to Ethiopia and the nearby countries Arabia, Kenya, Somalia, Uganda, Tanzania, Malawi, Congo, Zambia, Zimbabwe, and South Africa; it has also been found in Afghanistan and Turkestan [16, 17]. It is widely cultivated in eastern Ethiopia, highlands of Oromia region, Hararghe, where the center place of the production in the world and the center of exportation, from the district place called Aweday to Dire Dewa to Djibouti, Uk (London), Somalia, and Arab countries [18]. It is largely marketed and consumed with in-country out of production. The majority of people in the study area are seeking the drug stimulant-effect obtained by chewing fresh leaves, while simultaneously smoking a cigarette and/or tobacco in shisha, and do so on a daily basis for at least three hours without safe themself. This is common among both males and females, from school students to higher education. Also Khat producer (farmers) are work in chat fields while also consuming these plants for long periods of time. Majority of them had wound of the mouth, dental caries, lung disease, some of Khat users are act as mental disorder,and carelessness behavior. Not only in study area as Ethiopia country, Khat chewing has become a popular spending time for many young people and consumers were found in the highest number among drivers and farmers followed by student and shopkeeper. As data of Ethiopia Public Health Associate (EPHA) shown, the magnitude of common mental disorders is higher among Khat chewers than non-Khat chewers. Common mental disorder were 17.6%(39/222) among Khat chewers while only 6.1%(27/441) among non-Khat chewers [19].

The leaves intake has many negative effects, depends on the amount of consumption, can lead to effect health such as Oral and lung cancer [20]. The central and peripheral nerve systems, as well as the gastrointestinal system, are also all affected by Khat chewing [17, 21]. The subject effect, also known as thinking, is characterized by a rush of ideas but a lack of concentration. However, the user may experience depressive mood, irritability, anorexia, and difficulties sleeping after a khat session [21]. One of the causes of oral and lung cancer is exposure to radioactive elements. As reported by Al-Jalali and Shaltout(2014) many radioactive elements and the most hazardous

heavy metals were detected in Khat, including As, Cr, Zn, Fe, Cd, Cu, Pb, Cr, Zn, Hg, ^{226}Ra, ^{214}Bi, ^{238}U series, ^{228}Ac, ^{228}Ra, ^{232}Th series, ^{40}K, ^{137}Cs, and ^{210}Po [22, 23]. The extensive use of Khat (Catha edulis) is suspected to raise the concentration of these extremely harmful substances inside the human body, increasing the risk of a variety of diseases, including oral and lung cancer. Furthermore, due to the presence of heavy minerals and different radioisotopes in Catha edulis (Khat) plants, the distribution of radioactivity in Catha edulis (Khat) plants is higher than the natural radioactivity distribution. According to the Hill(1965) [24], by conversationed with Khat chewers in the study area revealed that they believe a minimum of 500 grams to three kilos of Chat can be chewed in one day with no harmful effects. This is one of the signs that this plant is widely used in the research domain. To my knowledge, there is no information in the literature study about the natural radioactivity distribution of foodstuff, and Khat plants in Ethiopia. Due to the hazards of radioactive material to humans and the environment, it is necessary to measure the natural environmental radiation levels.

In this work, the natural radioactivity distribution in leaves of Khat plants and its soil samples collected from study area, has been measured and evaluated. The results are used to calculate radiological hazard indices, evaluate epidemiological study performance, and maintain reference information to determine changes in environmental radioactivity caused by natural and human activities.

1.1 Statement of the Problem

For the safety of the publication due to exposure, environmental monitoring and assessment are essential in regulatory and advisory policy-making. Natural radionuclides materials under certain conditions can reach hazardous radiological levels. Some studies have been carried out to assess the levels of radiation in Ethiopia. However, most of the studies are focus only on areas of known background radiation and no information on radionuclides levels in food. Khat plants and soil are containing naturally radioactive material and heavy metal, which can be hazardous to humans if the radiation exposure from such elements exceeds the maximum allowed. There was no data on the natural radioactivity levels and hazard indices from such materials and related samples before to this investigation in the study area. The activity concentration of primordial radionuclides in the Khat plant and soil in the study area was not yet studied. Most people's in this area spending more time chewing Khat and majority of them had wound of the mouth, dental caries, lung disease, some Khat users have symptoms of mental disorder,and carelessness behavior. As a consequence, the findings of this study provided the necessary information on radionuclide concentration levels and their

contribution to external as well as internal exposure in this area. Furthermore, investigating natural radioactivity levels in environments especially materials direct contact with human beings daily basis used to assess the radiation dose received by a certain group of people for the population to understand the health hazards.

1.2 Objective of the Study

The overall goal of this thesis is to assess and investigate natural radioactivity levels in leaves of Khat and its' farms soil of selected samples.

Particularly;

i. To measure the activity concentration levels of naturally occurring radionuclides in leaves of Khat plants and agricultural soil samples collected from selected area.

ii. To calculate the radiological hazard parameters(like radium equivalent dose, hazard indices, and absorbed dose rate) of human exposure to gamma rays due to Khat leaves and soils samples.

1.3 Significance of the Study

The main source of health risk is radiation from naturally occurring radioactive materials (NORM) and technologically enhanced NORM. These natural radioactive materials are naturally present in the soil at different activity levels, distribution to the environment, and transfers to humans mainly through food consumption and inhalation. Gamma radiation emitted from radioactive materials has several health effects, such as the death of the cells, damage in the natural functioning of the cell leading to somatic effects like cancer, and genetic effect- permanent alteration of the cell which is transmitted to later generation. The Khat plant is chewed regularly in the study area for stimulating effects, and the majority of the people chewed this plant. With little information regarding radionuclide activity concentrations, the data obtained will provide understanding into the amount of radiation in soil and Khat plants, as well as its effect on public health. Organizations such as the Ethiopian Radiation Protection Authority (ERPA), and the Ethiopian Public Health Association (EPHA) will benefit from the research findings in terms of the environment monitoring and radiation effect control. The obtained data uses as a baseline for future changes in environmental radioactivity caused by natural and human activities.

1.4 Limitation of the Study

The research financial resources and methodology were the study's limitations.

The collected samples in this study plan have a sampling area limit. The samples were gathered from farmland, and because farmers are not allowed to sell khat samples on farmland, they are made very expensive. As a result, we did not cover the sampling area as planned.

The constraints in methodology come from the experimental laboratory's limited access to gamma spectrometry. The measurements were conducted at the Ethiopian Radiation Protection Authority (ERPA), the country's only single laboratory. There was no way to reduce statistical and technical error by repeating the measurements, thus they were only done once. To overcame, we carried out measurements by correctly and efficiently in laboratory.

2 Review of Related Literature

2.1 Introduction

This chapter discusses the behavior of selected natural radionuclides in the environment, as well as the sources and variability of natural radiation, human doses, and radiation's health effects. Its goal is to study and disseminate information about natural radioactive materials in the environment, and also detection methods.

The majority of the overall radiation dosage received by people and other living things releases from natural radioactive elements (commonly, ^{40}K, and ^{238}U, and ^{232}Th series) performing radioactivity decay, which is out of control, arising from a natural source such as cosmic ray, terrestrial, and exposure from inhalation or intake radiation sources. Other contributing factors include the results of human activity. As a result, the amount and levels of these natural exposures depends on geographical and geochemical, as well as some human activities [11]. Natural radionuclides are typically present in rock and soil in different amounts and levels of activity, with subsequent distribution to the environment and transfer to humans through inhalation and food ingestion causes health risks [14, 25]. High efficiency and large size gamma spectrometry are required to detect low levels of environmental radiation from such sources, and HPGe detectors are commonly used for gamma-ray measurement.

2.2 The Study of Radiation in the Environment

All matter is made up of atoms. Some atoms are naturally stable, others are unstable. Of the 118 elements , there are(1 is hydrogen and 92 is uranium) 92 elements occur naturally. The remainder have been produced artificially, including technetium, americium and plutonium [26]. Atoms that have different mass number called isotope. Radioisotopes or radionuclides are radioactive materials that are unstable isotopes of an atom [27]. Radiation in the environmental comes from different radioactive materials performing radio-activity, and accelerated particle(s) from nucleon synthesis [6]. The released radiation may take the form of particles (including electrons, neutrons and alpha particles) or of electromagnetic gamma radiation or X-rays, all with different amounts of energy. As a result, everyone is constantly bombarded by high-energy electro-magnetic radiation from space, as well as long-lived radionuclides present on the earth. These different forms ionizing radiation differ in their capacity to do damage and their ability to penetrate materials. The ionizing radiation or particles and/or elementary particle emissions are caused by radioactivity(radioactive decay) of atoms

that disintegrated spontaneously and transformed into different atoms. The new atom created by decaying of radionuclides, stable or unstable. By interacting with the human body, ionizing radiation released by radioactive decay has a variety of effects on human health. When ionizing radiation (such as *gamma*-rays, X-rays, and particles) travels through anything, including living tissue, it deposits energy in the substance, causing ionization and excitation. The absorbed dose is calculated by dividing the amount of energy deposited by the mass of tissue exposed, and it is commonly expressed in milligrays [27]. Radiation's biological effects are dependent on the amount of energy deposited. However, the fact that different types of radiation have various biological effects for the same amount of energy deposited and that tissues react differently is taken into account when estimating the possible biological effect. In radiation protection, the effective dose is a weighted quantity that is the most often used indicator of the potential biological effects of ionizing radiation exposure in humans. The effective dose ("dose") is usually expressed in millisieverts (mSv). The total exposure of a group of people to radiation is called the collective dose and is expressed in man-sieverts (man Sv). The annual worldwide average dosage from natural background radiation is 2.4 mSv$y-1$, whereas the comparable annual collective dose to the global population from natural background radiation is approximately 16 million man-years Sv [6, 12, 15, 27].

Generally, environmental radiation sources are categorized into two.

Natural radiation categories:- cosmic radiation and radiation resulting from the decay of naturally occurring radionuclides. The natural radionuclides include the primordial radioactive elements in the earth's crust, their radioactive decay products, and radionuclides produced by cosmic-radiation interactions. Natural environmental radiations mainly depend on geological and geographical conditions. Cosmic radiation, consisting mainly of high-energy particles from the sun and outer space, interacts with the earth's atmosphere and gives rise to a secondary radiation of particles and gamma rays [6, 28].

Anthropogenic sources: - these are from nuclear related technology, inorganic fertilizer and other human activities.

2.3 Naturally Occurring Radioactive Material (NORM)in the Environment

The term "naturally occurring radioactive material" refers to materials that include radionuclides of natural origin, such as primordial radionuclides (radionuclides present when the Earth was created around 4.5×10^9 years ago), ^{40}K, ^{238}U, ^{235}U, ^{232}Th, and their

radioactive decay products, as well as radionuclides produced through the interaction of cosmic rays with atoms in the atmosphere,called cosmogenic radionuclides [12, 13, 27]. Naturally occurring radionuclides are present in the environment and form most of natural background radiation [11].

Many naturally occurring elements have radioactive isotopes, only potassium (non-series) and uranium, and thorium decay series have radioisotopes that emit gamma-rays of sufficient energy and intensity to be measured by gamma-ray spectrometry. Because they are relatively abundant in the natural environment. Average crustal abundances of these elements quoted in the literature are in the range of $2 - 2.5\%$K, 2-3 ppm U and 8-12 ppm Th [27].

Most of the lighter radionuclides like ^{14}C, ^{40}K, emit only electrons or/and photons, whereas many of the heavier radionuclides like ^{232}Th, ^{238}U emit alpha particles. The decaying radionuclides create new daughter, which may be stable or may decay also [26, 27]

. The majority absorbed radiation dosage is not contributed by cosmogenic radionuclides [29]. These radionuclides' variability is related to variations in cosmic radiation fluxes, which vary with altitude and latitude, as well as temporally with changes in cosmic ray production rates. [15, 29]. Of cosmogenic radionuclides, only a few radionuclides are formed in sufficient quantities and are long-lived enough to contribute sufficient radiation in environment as some of them shown in figure 2.1.

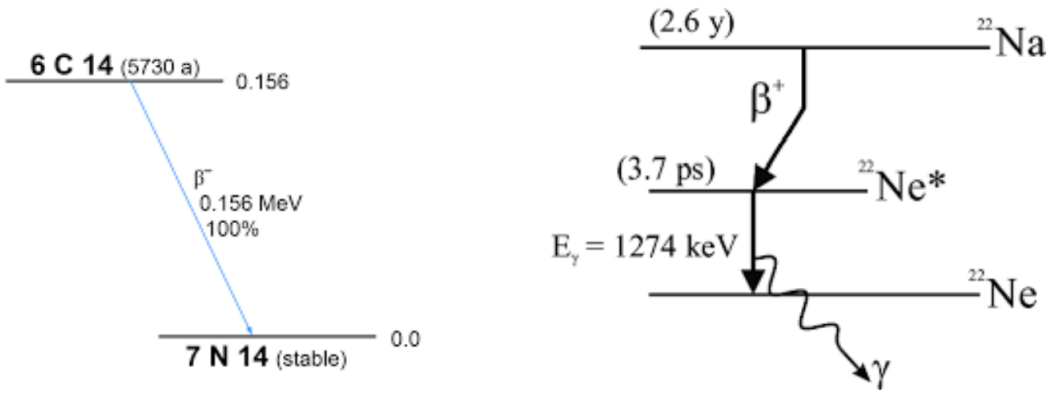


Figure 2.1: Decay scheme of Carbon-14 and Sodium-22, cosmogenically produced radio nuclides.

The naturally occurring radionuclides belong to one of the long chains, particularly the uranium (4n+2), thorium (4n), actinium (4n+3), and neptunium (4n+1) series, with the most radiologically important being $238U$, $232Th$, and $40K$. [6, 12, 29].

The levels of naturally occurring radionuclides vary widely in the environment. The

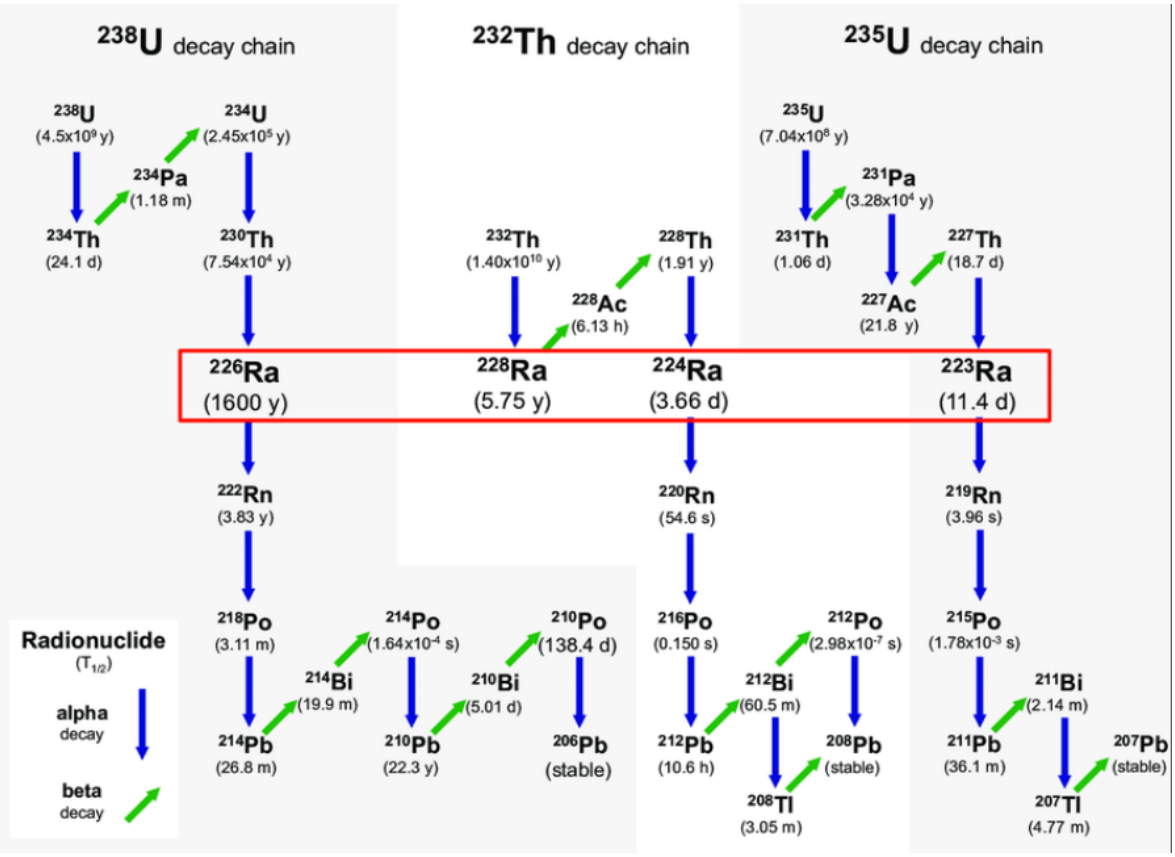


Figure 2.2: Decay series of the radionuclides of ^{238}U, ^{232}Th, and ^{235}U. Reprint [1]

distribution of the geological media from which radionuclides are formed, as well as the mechanisms that concentrate them at a specific location in certain media, determine the concentration of radionuclides in the geosphere. Typically, they are considered to be low, with global average concentrations of 35, 30 and 400 $Bqkg^{-1}$ for ^{238}U, ^{232}Th and ^{40}K , respectively [29].

The global population weighted 6 annual effective dose is 2.03 mSv. However, in specific areas in the world, such as Guarapari, Brazil; Kerala, India; Ramsar, Iran; and Yangjiang, China, these levels are elevated naturally due to the geological and geochemical characteristics of the region, resulting in a significant effective dose level, as much as 131 mSv per year [6,29]. These areas are termed High Natural Background Radiation (HNBR) areas. There is another situation where the level of naturally occurring radionuclides has been technologically altered by certain human activities [27,29]. This alteration may be deliberate, as in uranium mines [29], or unintentional, as in oil and gas industries [11].

Man-made radionuclides have gone into the environment due to human activities like for example medical purposes that use radionuclides to diagnosis and therapy the body and other man made causes are power plant reactors that use radioactive

uranium and thorium as fuel. Most of anthropogenic radionuclides are short-lived, but some have half-lives of many year and a potential risk. For example, ^{137}Cs(half life 30.2 years) one of the important,because its follows the course of potassium ecosystem and its persist in environment for long years and the others are ^{90}Sr (half-life = 28.1 years) and ^{80}Kr (half-life = 10.73 years) [15, 28].

2.4 Behaviour of NORM

The majority of radionuclide in NORM(principally radium and radon) a rise from Uranium and Thorium. The behaviour of NORM in the environment is governed not only by the physical and chemical properties of its individual radionuclides, but also by the type of human activities taking place [11, 29]. NORM contains a wide range of radionuclides with great variations in their physicochemical properties. These properties allow certain radionuclides to be released into the surrounding environment and migrate from their source of origin, causing disruption in the radioactive equilibrium state of the natural series. The radionuclides released into the environment may establish their own subseries away from the original series, this is the biogeochemical behavior of radionuclides in a given decay chain [11, 12].
For example in the ^{238}U(4n+1) decay series, several subseries, headed by ^{230}Th, ^{226}Ra, ^{222}Rn and ^{210}Pb, have the potential to be separated.

Uranium ($_{92}U$).
The uranium atom is found naturally as three different isotopes: $^{238}U(99.3\%)$,$^{235}U(0.72\%)$ and $^{234}U(0.005\%)$,a short lived member ^{238}U chain, is usually in radioactive equilibrium with parent isotope.

Uranium transport usually occurs in oxidizing surface water and groundwater as the uranyl ion, UO_2^{2+}, or as uranyl fluoride, phosphate, or carbonate complexes. The highest absorption of uranyl ions on natural materials (organic matter; iron, manganese, and titanium oxyhydroxides; zeolites, and clays) occurs at pH 5.0 − 8.5.

Uranium occurs in rock, air, water, and food and also is present in human tissues. The average annual intake of uranium of all dietary sources is about 13 Bq (350 pCi) [11, 29]. The decay products of uranium, particularly radium and its decay products, are further important than uranium itself with respect to dose to humans from both external and internal exposures.

Radium
Radium (Ra) has four natural radioactive isotopes: ^{228}Ra, ^{226}Ra, ^{224}Ra and ^{223}Ra. Ra exhibits only one oxidation state (Ra^{2+}) and is sensitive to pH changes. ^{226} Ra and

its decay products, part of the uranium chain, are responsible for a major part of the internal dose received by humans from the naturally occurring radionuclides [29].

^{226}Ra is present in all rocks and soils in variable amounts.The radium contents of soils can show considerable spatial variability, both locally and regionally. These are the result of differences in parent materials and in soil-forming factors such as climate and weathering time. Extensive studies on Ra behaviour have shown that Ra in water is soluble as chloride, sulfate, carbonate, nitrate and bromate salts, while it is insoluble as carbonate, iodate, oxalate and sulfate salts [11, 29].

Radium is chemically similar to calcium and is absorbed from the soil by plants and passed up the food chain to humans. Because the radium in food originates in soil and the radium content of soil is variable, the radium content of foods varies. In addition, it is reasonable to expect that such chemical factors as the amount of exchangeable calcium in the soil will determine the rate at which radium is absorbed by plants.

^{226}Ra is an alpha-particle emitter that decays, to $^{222}Rn(3.82d)$. The decay of ^{226}Ra(1600y) is followed by the successive disintegration of a number of short-lived alpha-particle- and beta-particle/gamma-ray-emitting progeny. After six decay steps, in which radionuclides that range in half-life from 1.6×10^{-4} to 26.8 min are produced, ^{210}Pb(half-life= 22.3y) is produced. This nuclide decays through ^{210}Bi to produce ^{210}Po, which decays by alpha-particle emission to stable ^{206}Pb. Radium itself adds little to the gamma-ray activity of the situation, but it does so indirectly through its gamma- ray-emitting decay products.The National Council on Radiation Protection and Measurements [30] estimates an average dietary intake of 0.05 Bq/d (1.3 pCi/d). Worldwide, the ^{226}Ra content of adult skeletons ranges from about 0.3 to 3.7 Bq (8 to 100 pCi), and the population-weighted average skeletal content is 0.85 Bq (23 pCi) [30], which corresponds to annual equivalent doses of 170 μ Sv (17 mrem) to cortical and trabecular bone, 90 μSv (9 mrem) to the bone lining cells, 15 μSv (1.5 mrem) to the red marrow, and 3 μ Sv (0.3 mrem) to soft tissues [29].

Thorium ^{232}Th,(4n)

Thorium-232 is the most abundant (about (100%) of the naturally occurring radioisotopes which is the first member of long series called the thorium series(4n), like uranium, it is ubiquitous in nature. Shorter-lived isotopes of thorium occur in all three of the natural decay chains [18, 29].

Th mainly occurs in the tetravalent Th(IV) state [11, 27, 29]. It is not soluble as uranium and is thus not as mobile in chemically solvent environments. In aqueous media, the level of pH can play a role in Th solubility . In near neutral pH, Th is removed from the dissolved phase onto colloids and particulates . At low pH, Th undergoes

hydrolysis and becomes more soluble and mobile . In soil, the mobility of Th may be less affected by pH changes .

Thorium and its radioactive pro-genies decay through the emission of alpha, beta and or gamma, until it's stable end products, ^{208}Pb(see fig.1). The common gamma emitters of it's pro-genies are; ^{212}Pb(10.6 hr) $(238.632kev, 47\%)$,^{212}Bi(1.01 hr) (727.330 kev 37%),^{212}Po and ^{228}Ac.

Potassium-40

Of the three naturally occurring potassium isotopes, only $^{40}K(1.3 \times 109y)$ is unstable and represent about 0.0118% of total potassium inventory on earth. It is one of the major radiation dose contributors from natural sources, with both β^-Photon emission.

It decays by three general modes(β emission, K-electron capture and Positron emission). Beta-particle emission to calcium-40 (89%) and by electron capture to argon-40(11%) and produces 1.46-MeV gamma rays after electron-capture decay. ^{40}K cyclic and accumulated freely in biosphere, since is the basic nutrient required for life.

Potassium-40 is present most by mass in natural potassium, thereby imparting a Seawater contains ^{40}K at about $11kBq/m^3$ (300 pCi/L). Because of its relative abundance and its energetic beta-particle emission (1.3 MeV), ^{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. A person who weighs 70 kg contains about 140 g of potassium, most of which is in muscle. From the specific activity of potassium, it follows that the ^{40}K content of the human body is around 4 kBq (0.1 μCi). NCRP (1987a) has estimated that this radionuclide delivers an annual dose of 0.18 mSv to the soft tissues and 0.14 mSv to bone. In health animals and people, ^{40}K represents source of radioactivitygreater even from ^{14}C.

2.5 Exposure to NORM

Exposure of humans to ionizing radiation of natural and anthropogenic origin is inevitable [31]. Exposure to ionising radiation emitted by individual radionuclides of NORM can be hazardous to humans and the environment [6, 11, 23, 29]. The radiation dose rate due to exposure to NORM is generally low in most environments. However, certain human activities can increase the concentration of NORM and/or alter exposure conditions [11]. In order to determine the radiation dose from NORM, exposure paths must be determined. There are two main types of NORM exposure pathways.

Internal Exposure

Humans are exposed to natural radiation by internal radiation, when radionuclide members of NORM enter the body through inhalation and ingestion. Various forms of ionising radiations are emitted as radionuclides decay inside the body. The radionuclides in the soil are taken up by plants through their roots, and can be passed to humans through crops harvested from such plants, causing in internal ionizing radiation exposure. Food-borne radionuclides are important sources of radiation for long-term health considerations, contributing significantly to average radiation doses to various organs and tissues in the human body [15, 27, 31].

The amount of NORM consumed and the quantity of NORM in the terrestrial and aquatic environment determine the doses from these pathway. ^{226} Ra is one of the most radiotoxic radionuclides present in food and water, as well as one of the most common sources of radiation from ^{238}U daughter [11, 15]. Due to its long life and radiological effect, it is one of the most important isotopes to be determined among the naturally occurring radionuclides in environmental samples. It decay into radon-222 by emission alpha particles. Deposition from radon-222 decay in atmosphere is often the main source of these radionuclide in foodstuff.

The other most widespread and inescapable internal exposure is caused by the beta/gamma emitting radionuclides of ^{40}K [11, 15] .

Potassium is the essential for human life and its concentration in body tissues and homeostatic control. However, it should be noted that for public exposure due to natural radiation, the ingestion of radionuclides is generally the least significant exposure pathway. From internal exposure, inhalation and ingestion the average value are 1.2 and 0.3mSv, respectively with typical ranges of 0.2 - 1 and 0.2 - 0.8 mSv [27].

External Exposure

Humans are exposed to natural radiation from external sources, which include radionuclides in the earth and cosmic radiation. Due to its long travel distance and great penetration power, gamma radiation provides the majority of the dosage from an external channel. In most places on the earth, natural radiation from external sources varies but in some localities, the variation is greater because of abnormally high or low soil concentrations of radioactive minerals. Cosmic radiation alone varies over the range of elevation that encompasses most of the world's population (0-2,000 m) and to a much smaller degree with latitude because of the variation in the earth's magnetic field.The population-weighted yearly effective dose from external exposure to these radionuclides in indoor and outdoor environments, respectively, is calculated to be 0.41 and 0.07 mSv. The worldwide average annual effect dose (mSv) from external sources

such as cosmic ray and terrestrial gamma ray are 0.39 and 0.48 mSv,with the typical range of 0.3 -1 and 0.3 - 0.6 mSv, respectively [27].

2.6 Hazards and Risks Associated with NORM

Hazard is the potential to cause harm; therefore, the radiological hazard can be defined as the potential of radiation to cause biological effects on human cells, tissue, organ, or organ system. Biological effects of ionizing radiation in humans, due to physical and chemical processes, occur following the passage of radiation through their bodies. These processes will involve successive changes at the molecular, cellular, tissue, and whole-organism levels.High doses of radioactivity can be harmful or even fatal. The radiological impact of radiation on human's exposure to radiation has been recognized via studies that, radiation exposure above certain threshold limits can damage living cells, causing death or delayed in some of them and modifying others rather than kill [6, 16]. However, in the case of low doses, studies are inconclusive as to the effect from the exposure to low background doses. The damage caused by exposure to a radiation is determined by types of radiation , duration of exposure and particular organ exposed and types of organ exposed due to the inhalation and ingestion of radionuclides such as ^{238}U and ^{232}Th families specifically ^{222}Rn and ^{40}K. Greater dose or dose rate,the grater the effects of the ionizing radiation.

Radiation effect can lead death of a cell, impairment in the natural functioning of the cell, leading to somatic effect such as cancer and a permanent alteration of the cell which is transmitted to later generation i.e genetic effect. Chronic exposure to ionizing radiation is continuous exposure to low levels of radiations over a long period. The effects can be observed some time after the initial exposure. Acute exposure is exposure to large, single dose of radiation or a series of doses, for a short period of time. This can cause both immediate and delayed effects. Biological effect can be considered in terms of deterministic or stochastic effect health. Because of their random nature, epidemiological detectable effects (malignancies and genetic effects) are referred to as stochastic effects [6]. It is also important to add that much of the studies on the effect of exposure to radiation have been based on information on radiation-induced cancer(carcinogenesis), available from epidemiological studies of a number of human populations. The basic way for assessing the potential health hazard associated with NORMs is to consider possible radiation exposures. It is generally established that radiation causes adverse effects and dangers when total radiation doses are used. However, risk from exposure to environmental level radiation requires an assessment

of the radiological hazard following the exposure.

2.7 Gamma Ray Spectroscopy

2.7.1 Introduction

The use of gamma-ray spectroscopy in the standardization and implementation of Activation Analysis is critical. Gamma ray spectrometry is a non-destructive, rapid, multi-element method of measuring radioactivity. Qualitative and quantitative analysis of samples can be performed by gamma-ray spectrometry systems in which a qualitative measurement identifies the radionuclides of interest from the energies and intensities of the peaks present in the spectrum under investigation. In a quantitative measurement, the activities concentration of radionuclides of interests are determined. Therefore, the accurate determination of the detector efficiency is arguably the most important parameter when gamma-ray spectrometry is used for radionuclide measurement. Gamma ray spectroscopy is an extremely important method in environmental radioactivity [4, 32].

A gamma-ray photon, also known as an x-ray, is uncharged and causes no direct ionization or excitation of the substance it travels through. As a result, causing the gamma-ray photon to interact with detecting material and transfer all or part of its energy to an electron in the absorbing material is important for gamma-ray detection [2]. Therefore, in order for a detector to serve as a gamma-ray spectrometer, it must carry out two distinct functions. First, it must act as a conversion medium in which incident gamma rays have a reasonable probability of interacting to yield one or more secondary electrons. Secondly, it must function as a conventional detector for these secondary electrons.

2.7.2 Radiation Detectors and Detection of Low Level Natural Radiations

All nuclear radiation detection is based on the interaction of the radiation with matter. Gamma ray detection depends upon the interaction with the solid which transfer the gamma ray energy to electron within the detector material. Radiations from natural environmental radio-elements are low level energy, as observed from their facts of long term effects on life. So large volume radiation detectors have become dominant over other detector types because of their inherently good resolution and linearity in detecting low energy level radiations. The sensitivity of such detectors system depends on several factors, like detector efficiency, detector resolution, background radiation, sample constituency, sample geometry and counting time.

Scintillator detectors and semiconductor detectors are commonly used low levels of environment radiation detectors in measuring photon energies emitted from environmental samples. Such photons are the main concerns of this dissertation. Virtual experiments and standardized radiation sources can be used to check the performance of such detectors. The commonly used standardized radiation sources are like ^{137}Cs,^{60}Co,^{152}Eu and others with long life-time and known strength which are also used in our measurements.

Virtual laboratories are developed by using Monte Carlo simulation in different machine languages. Geometry and Tracking4 (GEANT4) and Monte Carlo N-Particle simulation are the frequently used computer programs developed by C^{++} computer code and any other computer language, if geometry of a detector is clearly identified. GEANT4 is the common computer program used at CERN international laboratory for particle accelerating. In other sites, the computer codes are used for optimization of radiometric experiments and calibration purposes. The program uses Monte Carlo simulation following the trajectory of particles in the active region of particle detectors. It is a methodological way of doing what if analysis is true. From the result, it is possible to optimize the detectors and determine efficiency of detectors if geometry is clearly designed [7].

2.7.3 Gamma ray Semiconductor Detectors

A semiconductor detector is made from single crystal materials that are either elemental or compound and have a band gap of 1 to 5 eV [33]. The best energy resolution from radiation spectrometers and one of the modern detectors. In addition to superior energy resolution, semiconductor detectors also have another desirable features such as, compact size, relatively fast timing characteristics, and an effective thickness that can be varied to match the requirement of the application. Silicon and Germanium are the most commonly used semiconductors. Semiconductor detectors have a P-I-N diode structure in which the intrinsic (I) region is created by depletion of charge carriers when a reverse bias is applied across the diode. When photons interact within the depletion region, charge carriers (holes and electrons) are freed and swept to their respective collecting electrode by the electric field. The resultant charge is integrated by a charge sensitive preamplifier and converted to a voltage pulse with amplitude proportional to the original photon energy. The depletion depth is inversely proportional to net electrical impurity concentration, and since counting efficiency is also dependent on the purity of the material, large volumes of very pure material are needed to ensure

high counting efficiency for high energy photons [32, 33].

In any crystal there are charge trapping center $10^{17}/cm^3$ natural impurity atoms. These impurities have vacancy of electrons. As the free electrons are produced by incoming radiations, these are trapped in the vacancy of electron, electron pulse will not be formed. Much greater thicknesses are required for the detectors intended for gamma-ray spectroscopy. However,at a given applied voltage, greater depletion depths can only be achieved by lowering the value of impurity concentration through further reduction in the net impurity concentration. To accomplish this goal, there are two general approaches.The first one is to seek further refining techniques capable of reducing the impurity concentration to approximately $10^{10}atoms/m^3$. This techniques have been developed in germanium, not in silicon because of the present limit on available purity in silicon. The detector that are manufactured from this ultra pure germanium are called intrinsic germanium or High purity germanium (HPGe). The second approach is to create a compensated material in which the residual impurities are balanced by an equal concentration of do-pant atoms of the opposite type. This compensation cannot be carried out simply by adding the appropriate amount of dopant to the semiconductor before the crystal is grown. At the first the material always turn out to be n or p-type depending on which impurity shows a predominance, no matter how slight. The process of lithium ion drifting has been applied in both silicon and germanium crystal to compensate the material after the crystal has been grown. The semiconductor detectors produced by lithium drifting process are given the designation of Ge(Li) and Si(Li). These impurities lower the energy necessary to create electron-hole pairs [2, 4]. These systems provide much better energy resolution than scintillators. This can be attributed to the small amount of energy required to produce a charge carrier and the subsequent large output signal. Only 3eV is required to produce an electron-hole pair in Ge and 3.61eV for Si.

After many repetition of for production of ultra pure germanium, impurity level as low as $10^9 atoms/cm^3$ have been achieved. If the remaining low-level impurity are acceptors(such as aluminum)the electrical properties of the semiconductor crystal grown from material is p-type(designed πtype) and alternative, if donor impurities remaining high purity n-type (designed ν type) is the result [2].

For a coaxial detector (Figure 2.4) the rectifying contact that forms the semiconductor junction is typically placed at the outer surface of the crystal. Thus, the outer contact for an n-type HPGe will be p^+ and the inner surface will be n^+. The depletion layer for such a detector grows inwards as the voltage is increased.

Both Ge and Si photon detectors in thermal contact with liqiud nitrogen that cools

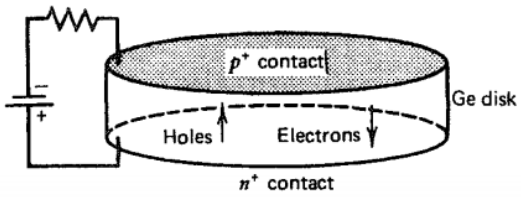


Figure 2.3: The configration planar of HPGe.The Ge semiconductors may be ν type($p^{+}contactisrectifying$), π type(n^{+} contact is rectifying)or Lithium drifting, Re-print [2]

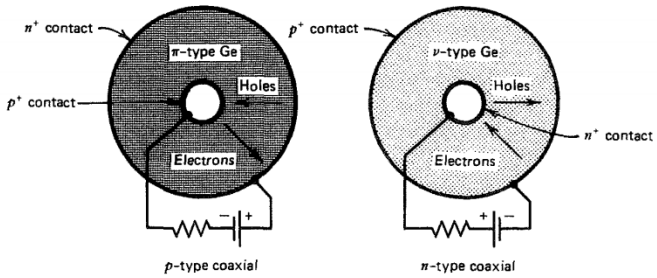


Figure 2.4: A cross section of an Schematic of p and n-type HPGe crystals coaxial detector perpendicular to the cylindrical axis . Re-print [3]

it to around $77\,°K$ or $-200\,°C$ in order to reduce the thermal charge carrier generation (noise) to an acceptable level; however, recent advances in electrical cooling systems have made electrically refrigerated cryostats a viable alternative for many detector applications. A cross-sectional view of a typical liquid nitrogen cryostat is shown in Figure 2.5 [4]

2.8 Emission of Gamma ray photons

Gamma ray is high energetic electromagnetic radiation produced by nuclear interactions. It is generally characterized as high energy radiation and short wavelengths within the electromagnetic spectrum. As a result, it has excellent penetrating and ionizing characteristics. This high energy can cause serious damage when absorbed by living cells.

In gamma decay, a nucleus goes from excited state to a state of lower energy and the energy difference between the two states is released in form of a photon [6]. An example of gamma emission for an element X of atomic number Z and mass number A;

$$ {}^{A}_{Z}X^{*} \rightarrow {}^{A}_{Z}X + \gamma \tag{2.1} $$

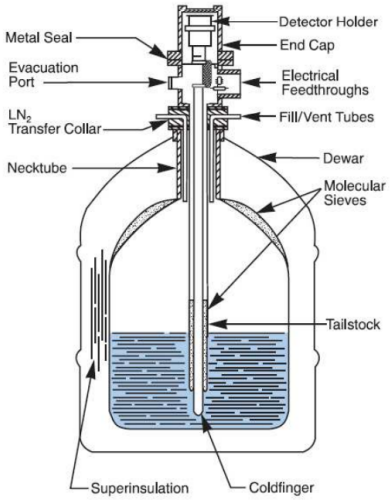


Figure 2.5: Model 7500SL Vertical Dipstick Cryostat (Canberra), Re-print [4]

The emission of gamma rays is accompanied by the change in energy state of the parent nucleus, and therefore gamma rays are always emitted with discrete energies. Most of the time gamma rays are emitted in conjunction with bets or alpha particles [34]. For example ^{40}K ($half - life = 1.3 \times 10^9 yr$), it decays with 89.3 % by emission of β^-, an electron to ^{40}Ca and 10.7% by electron capture to an excited state of ^{40}Ar. Then later decays to its ground state by emission of gamma-ray of energy 1460 keV. Fig 2.3 shows the decay scheme of ^{40}K.

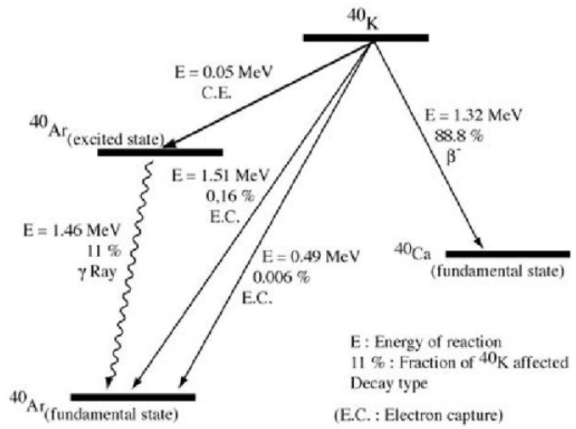


Figure 2.6: Decay scheme of Potassium-40

2.8.1 Secular Equilibrium

This type of relation between parent P and daughter D activity occurs when the half-life of the parent nuclide is infinitely larger than that of the daughter nuclide. In such

a case, the decay rate of parent P and hence the production rate of D is approximately constant.The rate of decay of any radionuclide is proportional to the number of atoms N present in the source;

$$A = \frac{-dN}{dt} = \lambda N \tag{2.2}$$

where λ is decay constant. Activity of radionuclide after time t is given by the equation

$$A = A_o \exp^{-\lambda t} \tag{2.3}$$

where A_o is the initial activity of the source. The relationship between parent and daughter activity is given by equation

$$\frac{A_D}{A_P} = \frac{\lambda_D}{\lambda_P - \lambda_D} \tag{2.4}$$

when $t_{(1/2)D} \ll t_{(1/2)P}$ then, the activity of the daughter is approximately equal to that of the parent. The state is referred to as secular equilibrium [6].

2.9 Gamma ray interaction with Detector

Gamma rays are photons that originate from the nuclei of radioactive atoms undergoing decay. They have no mass and no charge. They are quanta of electromagnetic energy that travel at the speed of light and can travel long distances in air unattenuated. When these photons interact with matter, free electrons are generated and as these electrons are slowed down by matter, they create charge pairs. The photon detectors use the charge pairs generated to determine the photon energy by measuring the quantity of charge produced by these pairs [11].

The probability of occurring of each of the three types of gamma ray interactions depends on the initial gamma ray energy and the atomic number, Z of the material the interaction takes place.

The area where each type is most significant is shown in Figure 2.7. For low Z materials, and gamma energies less than a few hundred keV, the photoelectric effect is the dominant process. Pair production becomes significant at gamma energies above 5 MeV (at high energy of ray). This leaves the Compton Effect to be most prominent at mid range energies around 1-2 MeV.
Gamma-ray photons interact with matter in three processes; namely

2.9.1 Photoelectric effect

The process by completely absorption or attenuation of the photon by atomic interaction and it gets eject an electron. A low-energy interaction and falls off rapidly with

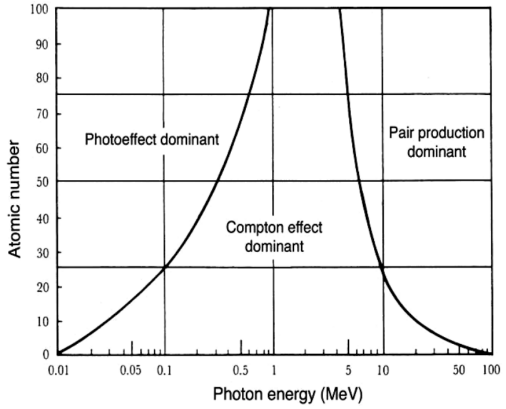


Figure 2.7: Relative predominance of photon interaction processes Re-print [5]

energy. It is not possible with a free electron, it happens with most tightly bound electrons inside an atom. Therefore, the electron absorbs an amount of energy equal to $h\nu$, where ν is the frequency of the photon and h is Planck's constant.

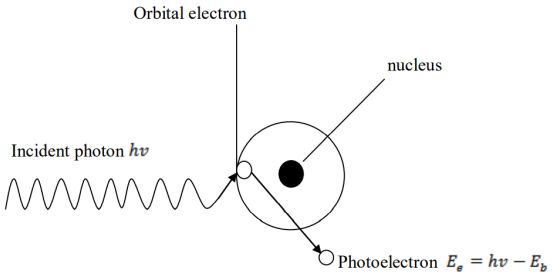


Figure 2.8: Mechanism of photoelectric effect- the incident photon interacts with an orbital electron.

The photoelectric effect produces electrons whose kinetic energy,$E_e$is given by:

$$E_e = h\nu - E_b \tag{2.5}$$

where E_b is the binding energy of the liberated electron in its original shell.

The probability that a photon will undergo photoelectric absorption is expressed as cross-section τ [9, 35]. This measure of the degree of absorption and attenuation varies with the atomic number, Z, of the absorber and the gamma-ray energy, E_γ, in a complicated manner:

$$\tau(h\nu, Z) = K(constant)\frac{Z^{4.5}}{(h\nu)^3} \tag{2.6}$$

The significance of this equation is that heavier atoms absorb gamma radiation more effectively than lighter atoms, at least in terms of the photoelectric effect. It follows

that ideal detector materials would be of high Z, given that their charge collection characteristics were satisfactory.

The photoelectric attenuation coefficient, μ_{PE}, can be derived from the related cross-section in the following manner:

$$\mu_{PE} = \tau \times \rho \times N_A/A \tag{2.7}$$

where ρ is the density of the absorbing material, A its average atomic mass and N_A the Avogadro constant.

2.9.2 Compton Effect

Compton effect is the quantum theory of the scattering of electromagnetic waves by a charged particle in which a portion of the energy of the electromagnetic wave is given to the charged particle in the elastic, relativistic collision. It is the direct interaction of the gamma-ray with an essentially free and station electron,i.e it occurs for sufficient energy of photons($0.1 < compton\, energy < 10Mev$ [5, 6].

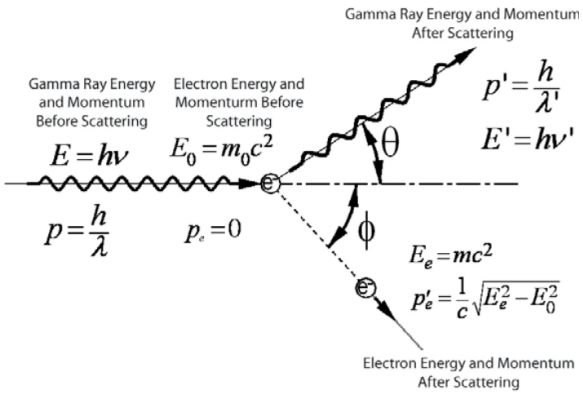


Figure 2.9: Mechanism of Compton scattering- incident photon transfer part of its energy to an electron. Re-print [5]

From theory of relativistic mechanics the energy,E_e of imparted to the recoil electron is given by;

$$E_e = h\nu - h\nu' + E_o \tag{2.8}$$

where $h\nu$ is the photon's energy, $h\nu'$ is the scattered gamma-ray energy and E_o is electron rest mass energy. The energy of scattered photon as a function of scattering angle ϕ is given by equation 2.9.

$$E' = \frac{E}{1 + \frac{E}{E_o}(1 - \cos\phi)} \tag{2.9}$$

The significance of this equation is that the scattered gamma ray energy decrease as the wavelength increases by an amount $\Delta\lambda = \frac{E}{E_o}(1 - \cos\phi)$.

The absorption cross-section, δ for Compton scattering is related to the energy $h\nu$ of the gamma ray is given by equation

$$\delta \propto K\frac{1}{h\nu} \tag{2.10}$$

Using an analogous relationship to that of Equation (2.7), we can calculate a Compton scattering coefficient, μ_{CS}. If we also take into account the fact that over a large part of the Periodic Table the ratio A/Z is reasonably constant with a value near to 2 we can show that:

$$\mu_{CS} = constant \times \delta \times \frac{1}{E_\gamma} \tag{2.11}$$

The implication being that the probability of Compton scattering at a given gamma-ray energy is almost independent of atomic number but depends strongly on the density of the material.

2.9.3 Pair production

Unlike photoelectric absorption and Compton scattering, pair production results from the interaction of the gamma-ray with the atom as a whole. The process takes place within the Coulomb field of the nucleus, resulting in the conversion of a gamma-ray into an electron–positron pair. The energy of the gamma-ray must be at least twice the rest mass of an electron which is 1.022 MeV(threshold energy), above this, the pair production increases with photon energy [5, 6, 9].

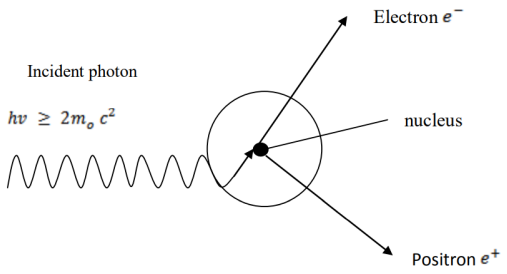


Figure 2.10: Mechanism of pair production- the process take place at the Coulomb field of nucleus. Re-print [6]

The net energy absorbed within the detector by the immediate consequences of the pair production event is (with energies expressed in keV):

$$E_e = E_\gamma - 1022 \tag{2.12}$$

The cross-section for the interaction, κ, depends upon E_γ and Z in a complicated manner which can be expressed as:

$$\kappa \propto Z^2 f(E_\gamma, Z) \tag{2.13}$$

The attenuation coefficient, μ_{PP}, is calculated in a similar manner to the photo-electric attenuation coefficient (Equation (2.7)).

2.10 Total Attenuation Coefficients

The sum of the coefficients for each of the significant interaction process:

$$\mu_T = \mu_{PE} + \mu_{CS} + \mu_{PP} + \mu_{RS} \Rightarrow (\rho \times N_A/A)(\tau \times \delta \times \kappa \times \delta_{RS}) \tag{2.14}$$

The mass-attenuation coefficient, the ratio of attenuation coefficient to the density of the material:

$$\mu/\rho = (N_A/A)(\tau \times \delta \times \kappa \times \delta_{RS}) \tag{2.15}$$

The attenuation coefficient only expresses the probability that a gamma-ray of a particular energy will interact with the material in question. It takes no account of the fact that as a result of the interaction a photon at a different energy may emerge as a consequence of that interaction. The total absorption coefficient, μ_A, must, of course, take into account those incomplete interactions:

$$\mu_A = (\rho \times N_A/A)(\tau \times f_{PE} \times \delta \times f_{CS} \times \kappa \times f_{PP} \tag{2.16}$$

In this expression, each 'f' factor is the ratio of the energy imparted to electrons by the interaction to the initial energy of the gamma-ray. Rayleigh scattering does not contribute to absorption of energy and does not appear in this equation.

By using total attenuation coefficient, we can calculate the degree of attenuation of a narrow beam of gamma radiation by using the following simple equation:

$$I = I_o e^{-\mu t} \tag{2.17}$$

where t is the thickness of the absorber in units consistent with the units of μ (i.e. cm if the attenuation coefficient, μ, is in cm^2g^{-1} and m if in m^2kg^{-1}). This equation relates the intensity of gamma-rays at a specified energy after attenuation, I, to that without attenuation at the same energy,I_o. This relationship is only valid under 'good geometry' conditions with a thin absorber and a collimated gamma-ray source. Under

the open conditions the equation(2.17) fails because of scattering from the absorber. This phenomenon is referred to as build-up and can be accounted for by a correction:

$$I = I_o e^{-\mu t} \times B \tag{2.18}$$

The build-up factor, B, is the ratio of the total photons at a point to the number arriving there without being scattered [9].

2.11 Detector Performance:

Gamma spectroscopy system are selected to take advantages of several performance characteristics. Two of most important include detector resolution and detector efficiency.

High resolution enables the spectroscopy to separate two gamma lines which are close to each other. Gamma spectroscopy system are designed and adjusted to produce symmetrical peak of the best possible resolution. In most spectra, the horizontal position of the peak is determined by the gamma ray's energy, and the area of the peak is determined by the intensity of the gamma ray and efficiency of the detector.

2.11.1 Detector resolution R:

Full width at half maximum(FWHM) used to express the detector resolution ,this is the width of the gamma ray peak at half of the highest point on the peak distribution . The resolution of a spectrometer is a measure of its ability to resolve two peaks that are fairly close together in energy. This is usually expressed as the width of the photo peak at half of its maximum height (F.W.H.M.). Resolution figures should be given with reference to specified gamma ray energies. It can be expressed as a relative value (%) or an absolute (keV) value.

$$R = (FWHM/H_o) \times 100\%$$

Where: H_o is the peak centroid These relations is then used by the system to calculate the expected FWHM.

The FWHM value increase with the energy of the photon, and its dependent on intrinsic properties of the detector, the type and setting of the electronic modules being used, and the layout of the cables. For automatic peak analysis, it is necessary to tell the computer what the peak width as a function of energy is [4].

2.11.2 Detector Efficiency

The probability that a gamma ray will interact with the detector and produce a count is the efficiency of the detector, this is because not all gamma rays that pass through a detector will produces a count in the system. High efficiency detectors produce spectrum in less time than low efficiency detectors. In general, larges detectors have higher efficiency than smaller detectors although the shielding properties of the detector material are also important factor.

Detector's efficiency is measured by taking a spectrum from a source of known activity, and comparing the count rates in each peak to the count rates expected from known intensities of each gamma ray. Efficiency, like resolution can also be expressed in absolute or relative terms. Absolute efficiency values give the probability that a gamma ray of specified energy passing through the detector will interact with the crystal and be detected.

The energy of the gamma rays being detected is an important factor in the efficiency of the detector. By plotting the efficiency at various energies, an efficiency curve can be use to determine the efficiency of the detector at energies different from those used obtain the curve.

$$Absolute \quad efficiency = \frac{Number\ of\ detected\ photon}{number\ of\ emitted\ photon} \times 100\%$$

2.12 System calibration

The calibration of an analytical system is one of the most important tasks required of any analyst. If the calibration is incorrect then all the result produced will be inaccurate. The essential requirements of a calibration are to establish an energy / efficiency / resolution relationship [9]. For gamma-ray measurement with germanium detector, the position of photo peak (full energy peak) is relevant for energy measurements. Detector calibration allows the gamma-ray spectrum to be interpreted in terms of energy, rather than channel number or voltage, and amount of radionuclide, rather than number of pulses.

There are three main calibration tasks:

2.12.1 Energy Calibration

Energy calibration is important for the qualitative analysis of the samples containing radioactive nuclei. The exact identity of photo peak present in the spectrum produced

by the detector system is a necessary requirement for the identification of gamma-emitters.

Energy calibration accomplished by measuring spectrum of a source emitting gamma-rays of precisely known energy and comparing the measured peak position with energy [9]. A lot of effort is applied to keep the linear relationship between the amount of energy transferred to the detector E (keV), and the height of the electronic pulse resulting from it; and also to the number of the channel in which this pulse is stored (K). In general:

$$E(kev) = C(kev) + G \times K(channels)$$

where: G is the linear energy scale (keV/channel) of the Multi-chanel analyser (MCA); and C is the intercept (which is usually set as close as possible to zero) and K the channel position.

An energy scale of 0,3 keV/channel is frequently used in high-energy gamma spectrometry to cover the energy region up to 2400 keV for a 8K MCA system (i.e. 8192 channels). A scale of 0,25 keV/channel would probably utilize the resolution of the detector better, but will only cover the region up to about 2000 keV. For low-energy spectrometry an energy scale of 0,5 keV/channel would often be acceptable [4]. The software usually determines the values of G and C by fitting a linear function to all the available points. An active sample containing a number of nuclides that emit photons covering the desired energy range, is counted with the detector to be calibrated. The parameters describing the Full width at Half Maximum (FWHM) and tailing of the peaks as function of photon energy are also determined as a part of this action.

2.12.2 Peak width calibration

The variation of peak width with energy i.e the width of a peak increases with energy and it is necessary, therefore, to provide the parameter of a width equation similar to the energy calibration. The peak width is the fundamental for the measurement of peak area and for peak fitting [9]. For many purposes the peak shape can be indicated to the program by supplying a single Gaussian or FWHM. The mathematical relationship between FWHM and energy(or channel number) Genie-2000 program use the equation (2.19);

$$FWHM = 2\sigma\sqrt{2ln2} \tag{2.19}$$

Where σ is standard deviation which is not a constant because it is a function of the photon energy.

2.12.3 Efficiency Calibration

Efficiency Calibration is important for the quantitative analysis of the samples containing radioactive nuclides. It is the relationship between the number of counts and the disintegration rates, and the efficiency of gamma spectrometer is a measure of how many pulses occur for a given number of gamma-rays.

To accurately determine activity concentrations, correct and reliable determination of the efficiency curve is of great importance. Detection efficiency is a complex function of the energy, spectrometer characteristics, measurements geometry, volume and density of the sample, etc [4]. The value for efficiency is dependent on the geometry of the sample – size, density, and distance from detector. For the detectors used in gamma analysis, efficiency varies significantly as a function of these parameters. In order to calibrate the spectrometer properly, calibration standards should be prepared with matrices with similar density, similar concentration for the relevant radionuclides, and in the same geometry and counting configuration as the real samples

2.13 Khat Plants and Agricultural Soil as Source of Radiation

Soil is a complex compound made up of minerals (both inorganic and organic) derived from plants and rock composition. It's a dense substance that gives plants the macro and micronutrients they need to function and grow. Naturally occurring radionuclides are ubiquitous in the environment and can be found in soil, sand, rock, minerals, air, water and biota [11, 27, 28].

Natural radionuclides in the environment, which originate from primordial radionuclides (U, and Th)series and ^{40}K present in soil, play an important role in man and biot's natural irradiation, which can be external or internal. Those long-lived radionuclides can be transferred to plants together with the nutrients during mineral uptake, grow in other parts, and even reach the edible parts [15, 29, 30] .

In Khat plants, many radioactivity material and most heavy metals and other inorganic elements present namely; Al, Ba, Bi, Br, Ca, Cl, Cu, Fe, K, Mg, Mn, Na, Ni, P, Rb, S, Si, Sr, Ti, Y, Zn and Zr ,where from those the metals such as (Na, ,Fe,Zn,K) can quickly become toxic, Whereas Ra, Bi, (^{238}U series), ^{228}Ac, ^{2}28Ra (^{232}Th series), ^{40}K, are very health risk radioactive metals [23].

The radionuclides of Khat(Catha edulis) represent the most dangerous factor, whereas this plant is chewed a daily minimum of three hours continuously. This is not restricted only to men but also women who frequently chew Khat [23]. The extensive use of Khat may increase the concentration of those elements inside the human body and

consequently the incidence of many diseases, especially oral and lung cancer,therefore these elements seem to be very dangerous and can cause a several problem on human health [23, 36]

3 Method and Methodology of the Study

3.1 Materials and Methods of the Study

3.1.1 Location of Study Area

The samples were randomly collected from different locations Haramaya woreda, east Hararghe, Ethiopia lies between Latitude 8.808028 and 8°48′29. 9008″ N and Longitude 41.601181 and 41°36′4.2516″ N using Global Position System (GPS) coordinates to measure natural radioactivity in leaves of Khat and agricultural soil samples. The samples were collected from different agricultural fields in the above mentioned study area, from three kebele, namely: Damot, Tuji Gabisa, and from Efa bate(figure 3.1).

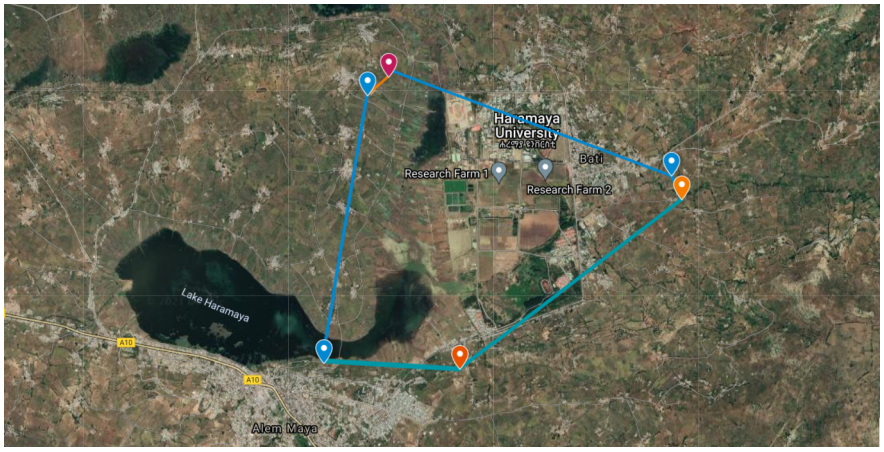


Figure 3.1: Study area

3.1.2 Sample Collection and Preparation

To evaluate levels of natural radioactivity, six samples of Khat plant (leaves) and six samples of agricultural soil (0-30 cm depth) were obtained from various Khat farmland in the above-mentioned study region. Two samples of Khat and two samples of soil were gathered from each kebele with different coordinates at different district locations (subsection kebele). For all of the samples, a single sample of Khat plant and agricultural soil was acquired at the same coordinate from the same district location (subsection kebele).

Within each kebele, soil samples were collected at similar depth and each of the sample was taken at a different depth, such as 0-5 cm, 0-15 cm, and 0-30 cm. Following collection, samples taken at various depths in a single subdivision kebele were blended, homogenized, and maintained in a plastic bag labeled with the name of the sample.

The samples were then weighed, dried for 24 hours in the sun and 8 hours in an oven at 110 $^{\circ}C$ to remove moisture, and reweighed to get a constant mass. The dried soil samples were ground into a fine powder, which was then sieved at 0.5 mm. Sieved samples were weighed, and an average mass of 742 gm of various samples was put into standard Marinelli beakers. The soil samples placed within standard Marinelli beakers container with a different weighing were kept hermetically sealed for a minimum of 28 days to attain secular equilibrium between ^{226}Ra,^{232}Th,^{238}U and ^{40}K and their progeny for HPGe natural radioactivity detectors.

In addition, all leaves of Khat plants collected from each kebele subsection of the selected fields were cleaned as much as possible with water, separated from the branch, then dried in the sun for 24 hours,crushed into fine powder, weighed, and then dried in an oven at 110$^{\circ}C$ around 14 hours to avoid moisture and reweighed to find constant mass. The dried Khat plant samples were ground into a fine powder and filtered through a 0.5 mm sieve. The sieved sample was weighed and found to have an average mass of 227.5 grams, with various weights packed into standard Marinelli beakers for natural radioactivity detectors. The standard Marinelli beakers container with different weighing were kept hermetically sealed for 30 days to attain secular equilibrium between ^{226}Ra,^{232}Th,^{238}U and ^{40}K and their progeny.

Finally, six soil composite samples and six Khat plant samples were brought up for gamma-spectrometry analysis.

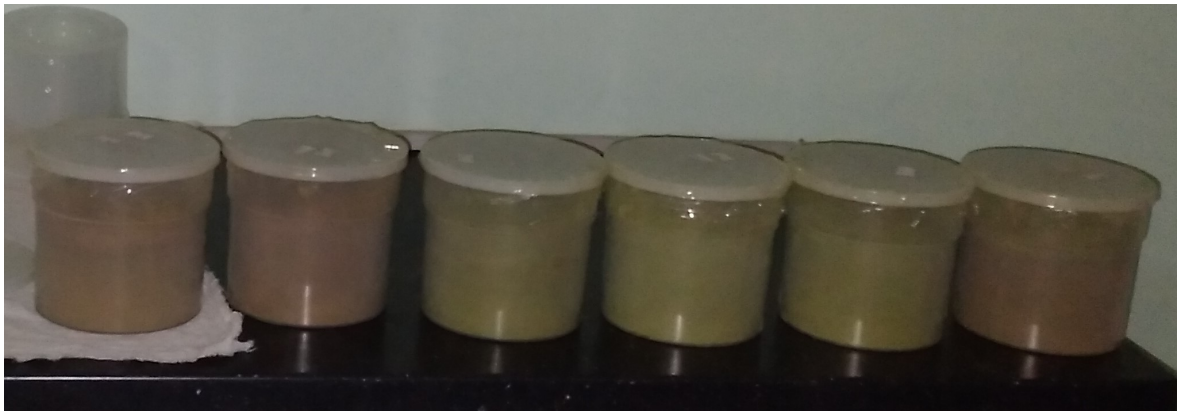


Figure 3.2: Prepared samples of soil and Chat(Khat)for HPGe detector

3.2 Gamma Spectrometry

Gamma ray spectrometry is an analytical method that allows the identification and quantification of gamma emitting isotopes in a variety of matrices. In one single measurement and with little sample preparation, gamma ray spectrometry allows us to

detect several radiations/particles emitting radionuclei in samples [33, 34].

The measurement gives a spectrum of lines , the amplitude of which is proportional to the activity of the radionuclide and its position on the horizontal axis gives an idea on its energy. Qualitative and quantitative analysis of samples can be performed by gamma - ray spectrometry system in which a qualitative measurement identifies the radionuclide of interest from the energies and intensities of the peaks present in the spectrum under investigation. In quantitative measurement, the activities of radionuclide of interests are determined.

3.2.1 Gamma Ray Spectrometer with HPGe Detector

The gamma spectrometer used for radiations detection and measurements is a Canberra system consist of Digital Signal Analyser (DSA) with High Purified Germanium detector, (HPGe) coupled with Pre-amplifier, Amplifier and Multi-Channel Analyser (MCA). This spectrometer systems is preferable due to high performance in detecting low level gamma energies of environmental radiation sources [2].

Germanium detectors are semiconductor diodes having a P - I - N structure in which the intrinsic (I) region is sensitive to Ionization radiations like, X-rays and gamma-rays. Under reverse bias, When photons interact with the material within the depleted volume of a detector, charge carriers (holes and electrons) are produced and are swept by the electric field to the opposite electrodes. This charge, which is in proportional to the energy of incoming photons deposited in the detector, is converted to a voltage pulse or signal by an integral charge sensitive pre-amplifier [3,4]. This integrated charge sensitive device is coupled to Digital Signal analyzer (DSA) consists of a pulse height analysis system to transform pulses, which are collected and stored by a computer installed GENIE2K-based software.

Pulse shape is determined by calculating ratio of Full Width at Tenth Maximum (FWHM)to Full Width at Half Maximum (FWHM) and ratio of Full Width at Fiftieth Maximum (FWFM) to FWHM. The theoretical acceptable ratio for FWTM/FWHM is 1.82 and should be less than 1.9 in practice. While for FWFM/FWHM is 2.38 for Gaussian peak and should be less than 2.5 in practice [4, 33, 37]

Energy resolution is a measure of the width (full width at half max, FWHM) of a single energy peak at a specific energy in a gamma-ray spectrum that is the smaller the width, the better the detector, the higher the resolution, either expressed in absolute keV or relative(%). When the centroids of nicely-shaped peaks are $3 times FWHM$ apart, the individual peaks are clearly separated, making peak area measurement pos-

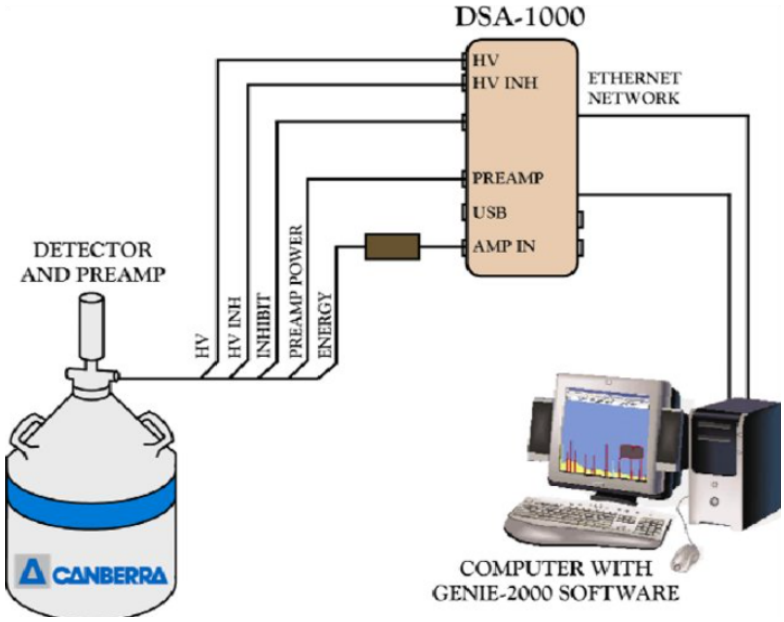


Figure 3.3: Gamma spectrometry full set up with DSA [7]

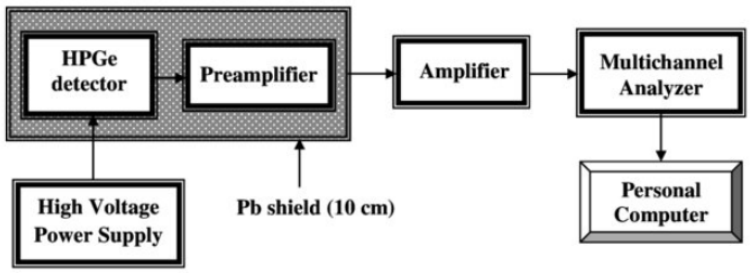


Figure 3.4: Diagram of high purity germanium detector system [8].

sible, and the detector energy resolution is good. However, if the separation were only $1 \times FWHM$, then simple measurement of the peak area is not possible [9]. Most gamma-ray spectra contain many small peaks on an uncertain background. The better the resolution, the narrower the peak, and so what few counts are in the peak will be concentrated in a fewer channels.

Energy resolution of photo peak is given by;

$$R = \frac{FWHM}{E_{centroid}} \tag{3.1}$$

where FWHM is calculated using the relation given in Equation above, in GENIE2K computer program/code.

$$FWHM = 2\sigma\sqrt{2ln2} \tag{3.2}$$

Where σ is standard deviation which is not a constant because it is a function of the photon energy. In the same way, same relation as in above equation is used for the calculation of FWTM and FWFM by putting in place of $ln2, ln10$ and $ln50$ respectively.

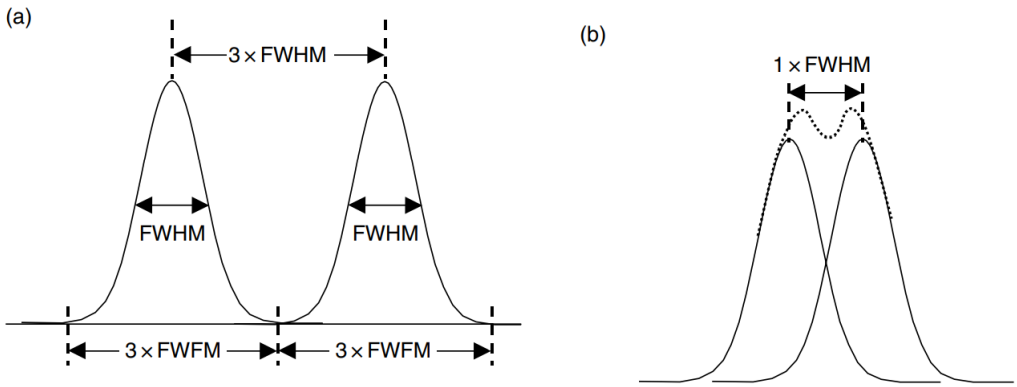


Figure 3.5: The influence of FWHM on the ease of discriminating between close energies: (a) centroids $> 3 \times FWHM$ apart should pose no problem; (b) centroids $1 \times FWHM$ apart require deconvolution programs.Reprint [9]

The FWHM increase with the energy of a photon, and is dependent on intrinsic properties of the detector, the type and setting of electronic modules being used, and the layout of the cables. After checking for such equipments, a standard Marinelli beakers of 0.16kg, mass 0.5litter, volume were used as sample holders. Range of peak locating channel were used from 180 keV to 8192 keV with nuclide energy identification tolerance of 1 keV. The dead time of cascaded DSA, as its high performance, is 0.46% in average, for 27600 sec live time, which means approximately 127.58 sec difference of real time for soil samples and 0.44% in average, for 63000 sec live time, which means approximately 239.77 sec difference of real time for Khat(Catha edulis) samples .

Operating parameters of the system are governed and controlled by the installed GENIE2K computer program, Canberra System. Additionally, the spectrum of laboratory background has been established from prolonged measurements and subtracted from sample's measurements. The resulting values allows us to perform nuclide identification and activity analysis, by referring to the information located in a nuclide library file, typically ISO-11929 MDA Report [4].

Therefore,Khat leaves and soil were analyzed using this gamma spectrometry. The DSA and large volume HPGe detector in a system makes it more interesting in our study.

The gamma photons interact with the detection material and transfer their energies to electrons or to positrons in the case of annihilation [4]. The detector is the center piece of the gamma spectroscopy system. These produced particles loose their energy within the detector, creating ionized atoms and ion pairs. These secondary entities form the basis of the detector signal [32].

3.3 Experimental Procedure

3.3.1 Gamma ray Spectrometer Coupled with HPGe Experimental Setup

Experimental Setup of gamma ray Spectrometer Coupled with HPGe used in this work is looks like as figure 3.6 to detect the activity concentration of three natural radionuclides in the samples. The measurements were performed using a p-type high purity germanium (HPGe) coaxial detector covered in a lead of 100 mm ,a cadmium depleted uranium of 2 mm , and copper of 2.5 mm thicknesses, to minimize the photon whose their source are out of the sample. Cadmium layers absorbs thermal neutrons produced by cosmic ray,as whole it absorbs radiation whose sources are environmental background. The Lead is used to reduce the soft components of cosmic rays and radiation released from Cd depleted uranium to a very low level. Copper layer prevent X-ray emitted from the lead, by its interaction with external radiation.

Figure 3.6: Gamma spectrometry mounted with HPGe detector and DSA circuit (ERPA Lab).

3.3.2 Calibration of gamma-ray spectrometer

Detector calibration allows the gamma-ray spectrum to be interpreted in terms of energy,rather than channel number or voltage, and amount of radionuclide, rather than number of pulses [9].The system was calibrated with respect to energy and efficiency for

0.16kg Marinelli beakers contain certified mixed radionuclides (type-CBSS2) supplied by IAEA. This is normally performed before measurement.

Energy Calibration

The object of energy calibration is to derive a relationship between peak position in the spectrum and the corresponding gamma-ray energy. The energy calibration was made by measuring radioactive Amersham mixed a standard source obtained from AMERSHAM a content nuclide in (Table 3.1). The HPGe detector is counted long enough for 10hrs to produce well-defined photopeaks. The calculations were carried out for all samples were estimated using the energy as a function of the number of channels, and the full width half maximum FWHM as a function of the number of channels calibrations. The energy calibration was established with 1024, 2048, 3072. 4096, 5120,6144,7168, and 8192 channels numbers(see a figure below) and well fitted with the degree function as energy;

$$Energy = 6.638 \times 10^{-2}\, Kev + 2.447 \times 10^{-1} \times Ch$$

The channel number that corresponds to the centroid of each full energy peak on the MCA recorded and plotted on linear graph paper on the X-axis versus the gamma ray energy on the Y-axis. Results obtained are shown in Figure 3.7

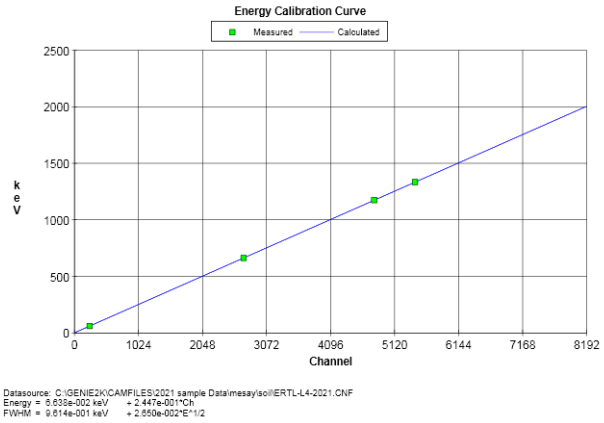


Figure 3.7: Calibration curve by standard check sources in the region of interests

The FWHM calibration performed uses the following expression:

$$FWHM = 9.614 \times 10^{-1} kev + 2.650 \times 10^{-2} E^{1/2}$$

Efficiency Calibration

Also the efficiency calibration was made using mixture standard source obtained from AMERSHAM content nuclide in (Table 3.1).The full energy peak efficiency is the

parameter the most significant in gamma ray spectrometry. Efficiency calibration is the relationship between the number of counts and the disintegration rates performed by computer software analysis [3]. The efficiency calibration was performed using different energy peaks to cover the energy range from 0 upto 2500 keV (Figure 3.7).

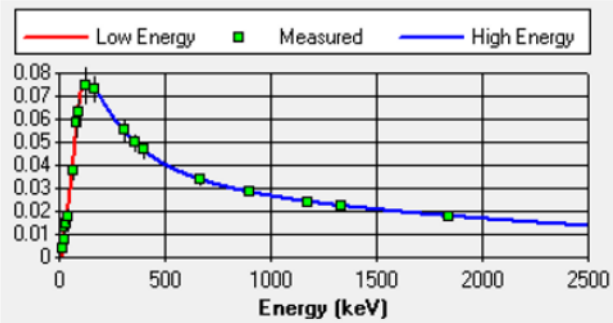


Figure 3.8: Results of efficiency (%) calibration for HPGe detector.

The results for all samples were estimated using the efficiency as a function of energy, the energy as a function of number of channels, and the full width half maxima FWHM as a function of number of channels calibrations.

The efficiency, ε of the counting for energies, E utilized in the calculations is given ;

$$\ln(\varepsilon) = -2.692 \times 10^{1} + 9.874 \times \ln(E) - 1.002 \times \ln(E)^{2}$$

Full Energy Peak Efficiency of the detector was measured using the same standard sources as in section 2.9.2 for the same geometry is given by:

$$\varepsilon_i = \frac{A_i}{C_i \times I_\gamma \times t \times M}$$

where $A_i$is net peak area corresponding to Ei , C_i is deduced activity by certified radionuclide, I_γ indicates probability of E_i photon emission per decay, and t is counting time, M mass of the radionuclide. This is the parameter of most significance in practical gamma spectrometry.

Finally, the calibrations are saved and used in the acquisition program to apply the corrections for each sample.

3.3.3 Background measurement

A natural background was measured for 56 hours using empty marinelli beakers 0.16 kg of similar geometry as the marinelli beakers of the samples and then subtracts the value of the natural background from the every measured value.

Table 3.1: : Gamma energies, branching ratios and the half- live of the radionuclide's used for gamma- system calibration.

Nuclide	γ-ray energy (Kev)	Half-live	Branching ratio %
^{241}Am	60	432.7 y	36.60
^{109}Cd	88	463 d	3.60
^{57}Co	122	271d	85.60
^{139}Ce	166	137.66 d	79.90
^{203}Hg	279	46.6 d	81.50
^{113}Sn	392	115 d	64.17
^{137}Cs	662	30 y	84.62
^{88}Y	898	106.01 d	94.00
^{60}Co	1173	5.27 y	99.97
^{60}Co	1333	5.27 y	99.98
^{88}Ye	1836	106.01 d	99.20

3.3.4 Analysis of samples

The activity concentrations for samples were measured using Canberra p-type high purity germanium (HPGe) coaxial detector with 1.9 keV relevant resolutions at 1332.5 keV ^{60}Co line of 5444 channel, a relative efficiency of 70% and multichannel analyzer of 8192 channel performance, including the mixed gamma sources.

At the end of the in growth period(at the end of for secular equilibrium of minimum 30 days) the samples were placed over the lead shielded HPGe detector at liquid nitrogen temperature ($77°K$) and counted for minimum 25,303 sec or 7.03-hours for the soil sample and 54,174 sec or 15.048 hours for Chat sample measurements.

The spectra were analysed for energies; 351.92 keV (^{214}Pb), and 295.21 kev (^{214}Pb),60931 keV (^{214}Bi),1120.29 kev (^{214}Bi)and 1764.94 kev(^{214}Bi) used to determine ^{226}Ra which equal to ^{238}U radionuclide. The energies 911 .21keV (^{228}Ac), and 968.97(^{228}Ac) used for ^{232}Th radionuclide identification, and 1460.9 keV gamma energy for ^{40}K radionuclide identification. Levels of activity concentration were measured for these identified radio nuclides is from their photo peak.

3.4 Activity concentration

Since the counting rate is proportional to the amount of the radioactivity in a sample, the Activity Concentration(Ac) which can be determined as a specific activity as the

follows

$$A_c = \frac{C - BG}{\varepsilon \times I_\gamma \times t \times M} \tag{3.3}$$

Where A_c is the specific activity in (Bq/kg), C is the area under the photo peaks, ε: Percentage of energy efficiency. I_γ is the percentage of gamma-emission probability of the radionuclide under consideration, t is counting time in (Sec.), M is mass of sample in (kg) and BG is background.

3.5 Calculations of Radiological Parameters

3.5.1 Radium Equivalent Activity

Radium equivalent activity (R_{aeq}) index represents a weighted sum of activities of ^{226}Ra,^{232}Th and ^{40}K in NORM based on estimation that the worldwide average concentrations of the radionuclides of ^{226}Ra,^{232}Th and ^{40}K are 370, 259 and 4810 Bq/kg, respectively will give the same gamma-ray dose rate [7].

$$R_{aeq} = A_U + 1.43A_{Th} + 0.077A_K \tag{3.4}$$

where A_U, A_{Th} and A_K are activity concentrations in Bq/Kg of ^{238}U, $^{232}Th$and ^{40}K respectively. The world average of Ra_{eq} in soil is $370Bqkg^{-1}$ [10].

3.5.2 Absorbed Dose Rate in Air (DR)

According to guidelines provided by UNSCEAR [38], the absorbed dose rate in air at a height of 1 m above ground surface was calculated from the measured activity concentrations of gamma emitters using dose-rate conversion factors that ensures a uniform distribution of the three radio nuclides for almost the same activities. If natural radioactive nuclides are uniformly distributed in the ground, dose rates D (nGy/h) at 1 m above the ground surface are calculated by the following formula;

$$D_R = 0.427A_U + 0.623A_{Th} + 0.043A_K \tag{3.5}$$

This dose rate indicates the received dose at outdoors from radiation emitted by radionuclides in environmental materials. The limit for this dose is 59nGy/h [7].

3.5.3 The Annual Effective Dose Rate(ADR)

Annual effective dose should be calculated to assess the health effects of the absorbed dose. Annual effective dose rate (A_{DR}, in $mSvy^{-1}$) received by the population can be

calculated using [6, 7]

$$A_{DR_{Indoor}} = DR(mGy/h) \times 8760h/y \times 0.8 \times 0.7Sv/Gy \times 10^{-6} \tag{3.6}$$

$$A_{DR_{outdoor}} = DR(mGy/h) \times 8760h/y \times 0.2 \times 0.7Sv/Gy \times 10^{-6} \tag{3.7}$$

Where (0.7 $SvGy^{-1}$) is a conversion coefficient used to transform absorbed dose in air to the effective dose received by humans, with an outdoor occupancy factor (0.2), which is equivalent to an outdoor occupancy of 20% and 80% for the indoors i.e the fraction of time spent indoors and outdoors is 0.8 and 0.2 respectively. This factor is suitable for determining the pattern of life in the studied area.The worldwide annual average dose of ionizing radiation by natural source of exposure is approximately 2.4mSv/y [7, 27]

3.5.4 External Hazard Index (Hex) and Internal Index (Hin)

The external hazard index for samples under investigation was estimated using the equation defined by [25, 39].

$$Hex = \frac{A_U}{370Bq/kg} + \frac{A_{TH}}{259Bq/kg} + \frac{A_K}{4810Bq/kg} \tag{3.8}$$

The external index is used to calculate the indoor radiation dose value due to the external exposure to gamma-radiation released by the natural radionuclides in the formation building substances of the lodging places.

The hazard levels from inhaled alpha particles emitted from radon short-lived radio nuclides such as 222-Rn and daughter product can be calculated by internal hazard index, Hin as [7, 40]

$$Hin = \frac{A_U}{185Bq/kg} + \frac{A_{TH}}{259Bq/kg} + \frac{A_K}{4810Bq/kg} \tag{3.9}$$

For both H_{ex} and H_{in} to be less than one, the maximum values of Ra_{eq} must be less than $370Bqkg^{-1}$ and 185 $Bqkg^{-1}$, respectively [38]

3.5.5 Radioactivity Level Index

The radioactivity level index (I_γ) is used to monitor radiation inside the human body and to compute the risk level of radionuclides in the human body when exposed to an amount of indoor or outdoor annual effective doses of radiations from radionuclides in soils. The estimated values of (I_γ) should be less than or equal to one to make sure the soil environment is hazard-free. The radioactivity level index (I_γ) proposed by the European Commission has been calculated from the activity concentrations of ^{238}U, ^{232}Th, and ^{40}K in soil samples using the following formula [39, 41].

$$I_\gamma = \frac{A_U}{300Bq/kg} + \frac{A_{Th}}{200Bq/Kg} + \frac{C_K}{3000Bq/kg} \tag{3.10}$$

The criterion for a definition of different levels of I_γ- indices are; $I_\gamma \leq 0.5$ as upper limit corresponds to $0.3 mSvy^{-1}$, $I_\gamma \leq 1$ corresponding to $1 mSvy^{-1}$ [42].

3.5.6 Ingestion Effective Dose

Dose by ingestion are mainly due to ^{40}K, and ^{238}U and ^{233}Th series radionuclides present in food. Radiation doses from the intakes of radionuclides in food is can be calculated from the amount of radionuclide deposited on foodstuff, the measurements of the activity concentration of particular radionuclide in food per unit deposition, the daily consumption rate of the food products and the dose per unit activity ingested. The consumption of foods by individuals varies widely around the world, depending on the climate, food availability and cultural dietary preferences. Food consumption data was needed for the estimation of radiation doses from ingestion.

The Ingestion effective dose due to the intake of three radionuclides in foods can be evaluated using the following expression [40]:

$$H_{T,r} = \sum (U_r)^i \times C_r^i) \times g_{T,r} \tag{3.11}$$

where, i denotes a food group, the coefficients U_r^i, denote the consumption rate (kg/y), C_r^i is activity concentration of the radionuclide r of interest (Bq/kg), and $g_{T,r}$ is the dose conversion coefficient for ingestion of radionuclide r (Sv/Bq) in tissue T. For adult members of the public, the recommended dose conversion coefficient $g_{T,r}$ for ^{40}K, ^{226}Ra(^{238}U), and ^{232}Th, are 6.2×10^{-9}, 2.8×10^{-7} and 2.3×10^{-7} μSv/Bq respectively [10]

Presently, no site specific consumption data exist in the study area and as such we have adopted on published article [24] on the study area, that 500 g to 3 kg per day without ill effect, to enable us calculate the effective dose due intake of the food stuffs using Equation (3.11). So we have been taken that the average of minimum and maximum consumption rate for this analysis.

4 Result and Discussion

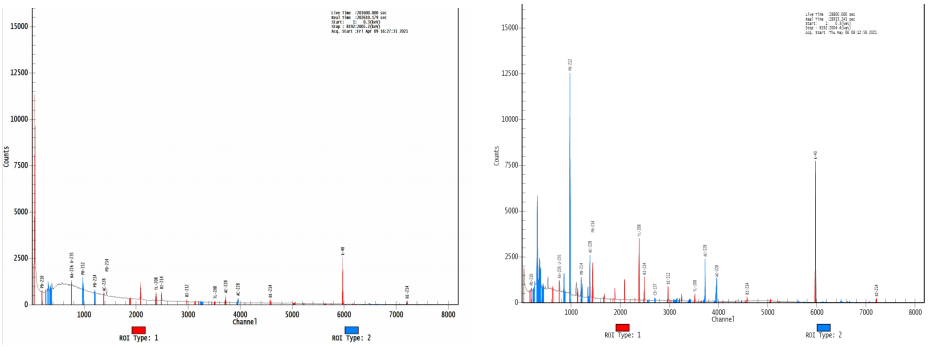
4.1 Introduction

This study was aimed at measurement of natural radioactivity level in the soil and the Khat plants in Haramaya area, eastern Ethiopia, which is the center place and the source of Khat for the exportation to neighboring countries from the district place called Aweday to Jibute, Somalia, different Arab countries, and to different countries in the world, using HPGe detector.

The soil and Khat samples were obtained from Khat farms and prepared for analysis at Ethiopia Radiation Protection Authority (ERPA)laboratory. The soil and Khat plant samples were analyzed by measuring the intensity emitted from radionuclide in samples and the calculation of activity concentration from intensity, also the calculation of radium equivalent activity, external and internal hazard indices, the radiation absorbed dose rate and annual effective dose rate was performed from activity concentration of radionuclides in the samples. The quantities have been determined and the results are presented in tables and graphs in this section and as all the results have been discussed in this chapter.

4.2 Activity Concentration of Natural Radionuclides in Soil

From experimental set up in labratory, the spectrum for each gamma emitter is one of the interest determined. The spectra were from primordial radioactivity decay progenies see in section 3.3.4. This was done for each of sample of soil and khat including back ground spectrum as shown in fig. 4.1.



(a) Background spectrum (b) Spectrum from Soil sample

Figure 4.1: HPGe background and samples output Spectrums

Radioactivity concentrations of the three primordial radionuclides ^{40}K, ^{238}U and ^{232}Th, and anthropogenic radionuclide ^{137}Cs were investigated in soil samples collected from Khat farms, Haramaya area, eastern hararghe. The sources of ^{137}Cs could be from the geological and chemical properties at these locations or might be have bee transported from elsewhere. The radioactivity concentration of these radionuclides were calculated using equation (3.3) as seen in section 3.1. The activity concentrations for three natural radioactivity in six soil samples were determined using HPGe p-type detector are presented in table 4.1.

The measured activity concentration of ^{238}U in agricultural soil samples ranged from $21.43 \pm 0.98 Bqkg^{-1}$ to $30.77 \pm 0.62 Bqkg^{-1}$, with a mean value of $23.72 \pm 0.85 Bqkg^{-1}$. The concentration of ^{232}Th ranged from $69.27 \pm 3.17 Bqkg^{-1}$ to $168.78 \pm 7.55 Bqkg^{-1}$, with an average value of $111.50 \pm 4.21 Bqkg^{-1}$. The activity of ^{40}K in agricultural soil samples ranged from $542.15 \pm 22.73 Bqkg^{-1}$ to $1045.96 \pm 43.44 Bqkg^{-1}$, with an average value of $794 \pm 26.44 Bqkg^{-1}$. Except soil sample code L5, at all sampling site the farmers using irrigation, water from underground for production of Khat and other foodstuff like vegetable etc. It is also observed that the measured activity concentration of ^{40}K exceeds markedly the values of both Uranium and Thorium, as it is the most abundant radioactive element under consideration. The average activity concentration for ^{40}K is the highest, followed by ^{232}Th and ^{238}U being the last in sampling sites.

Table 4.1: Activity concentration of the natural radionuclides ^{40}K, ^{238}U and ^{232}TH measured in the soil sample in $Bqkg^{-1}$.

Sample ID	**Sample type**	^{238}**U**	^{232}**TH**	^{40}**K**
L1	Soil	22.52±1.02	168.78±7.55	879.12 ±36.58
L2	,,	21.43±0.98	98.50±4.46	770.48±32.15
L3	,,	23.55±1.06	69.27±3.17	542.15±22.73
L4	,,	30.77±0.62	125.34±2.68	740.84 ±11.58
L5	,,	20.02±0.90	123.86±5.54	1045.96± 43.44
L6	,,	24.03±0.50	83.23±1.85	786.06±12.18
Average	-	23.72±0.85	111.50 ±4.21	794.10±26.44

The recommended reference levels of ^{238}U, ^{232}Th, and ^{40}K are 35, 30, and 400 $Bqkg^{-1}$, respectively, as listed in the world average concentrations published by UNSCEAR(2000). The activity concentrations of ^{40}K and ^{232}Th obtained in all samples of

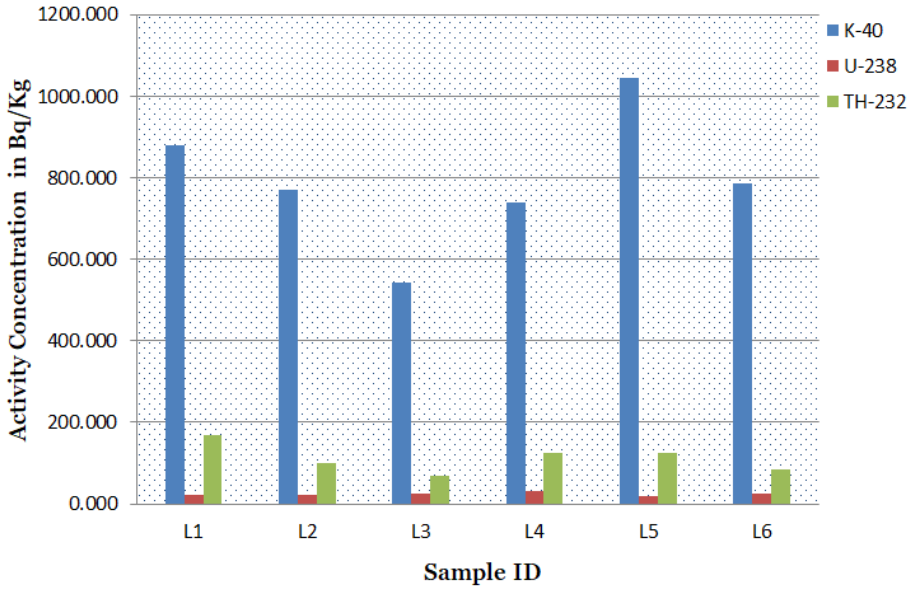


Figure 4.2: $^{238}U, 232Th$ and ^{40}K activity in sampled agricultural soil.

soil in the present study are higher than the recommended reference levels. The activity concentration of ^{238}U is below the permissible limit of world average concentration i.e 23.72Bq/kg.

4.3 Radiological Hazard Assessment

Gamma-ray radiation hazards caused by specific radionuclides of ^{40}K, ^{238}U and ^{232}Th, were evaluated using different indices.

4.3.1 Radium equivalent Activity (Ra_{eq})

The radium equivalent activity is the most widely used radiation hazard index [6, 39]. As shown in Table 4.2, the radium equivalent activity was found in soil samples to be in the range $221.61 \pm 9.83 BqKg^{-1}$ to $331.57 \pm 13.81 BqKg^{-1}$, with in average value $244.31 BqKg^{-1}$, which is higher than the world average value 89 $BqKg^{-1}$, and lower than the permissible healthy limit of 370 $BqKg^{-1}$g [6]. Radium equivalent activity in all sample was lower than the permissible limit, because of the activity concentration of the ^{238}U in all samples is lower than the permissible limit activity concentration in the soil.

4.3.2 Absorbed Dose Rate in Air (D_R).

Table 4.2 shows the absorbed dose rate calculated from the radioactivity concentrations of ^{40}K, ^{238}U and ^{232}Th,in farmland of khat soil samples. The absorbed dose rate

in khat farmland soil ranged from $76.52 \pm 3.40 nGyh^{-1}$ to $152.57 \pm 6.7 nGyh^{-1}$, with a mean value of $113.74 \pm 4.12 nGyh^{-1}$, which is higher than the global mean value of $59 nGyh^{-1}$ established by UNSCEAR [10],but below maximum allowed health limit of $1500 nGyh^{-1}$.The absorbed dose value is maximum for sample code L1 for which its ^{232}Th(168.78 Bq/kg) activity value, is six times more than the permissible limit(30 Bq/kg). The minimum absorbed dose value for sample code L5 for which its activity value ^{232}Th(76.52Bq/kg) is lower than the activity concentration in all sample determined but above the world average dose rate(59nGy/h).

Table 4.2: Radio-logical hazard indices in Soil samples (S).

S.ID	Ra_{eq} $(\frac{Bq}{Kg})$	D_R (nGy/h)	A_{DR} (mSv/y)	Hex	Hin	I_γ
L1	331.57±13.81	152.57±6.71	0.19±0.01	0.72	0.96	1.21 ±0.05
L2	221.61±9.83	103.65±4.58	0.13±0.01	0.60	0.66	0.82±0.04
L3	164.35±7.34	76.52±3.40	0.09±0.00	0.44	0.51	0.61±0.03
L4	267.06±5.35	123.09±2.43	0.15±0.00	0.72	0.80	0.98±0.02
L5	277.69±12.16	130.69±5.70	0.16±0.01	0.75	0.80	1.03±0.05
L6	203.57±16.26	95.91±1.89	0.12±0.00	0.55	0.61	0.76±0.03
Average	**244.31±10.79**	**113.74±4.12**	**0.14±0.01**	**0.63**	**0.72**	0.90± 0.03

Table 4.3: Shows the world average and maximum acceptable safety health limit of radiological parameters [6, 10].

	$Ra_{eq}(\frac{Bq}{Kg})$	$D_R(nGy/h)$	A_{DR}(mSv/y)	Hex	Hin
World average	89	59	0.07	1	1
Acceptable safety limit	370	1500	1	6	6

4.3.3 The Annual Effective Dose Rate (A_{DR})

The calculated annual effective dose in the agricultural soil samples ranged from $0.09 \pm 0.00 mSvy^{-1}$ to $0.19 \pm 0.01 mSvy^{-1}$, with an average value of $0.14 \pm 0.01 mSvy^{-1}$,which was above the world average of $0.07 mSvy^{-1}$. As shown in (Table 4.3),the worldwide average annual effective dose due to external terrestrial is approximately $0.5 mSvy^{-1}$ [10]. Indoor dose rates were not evaluated because data on average buildup of radon gas in the indoor atmosphere were not available.

4.3.4 External Hazard (H_{ex}) and Internal Hazard Index (H_{in}) .

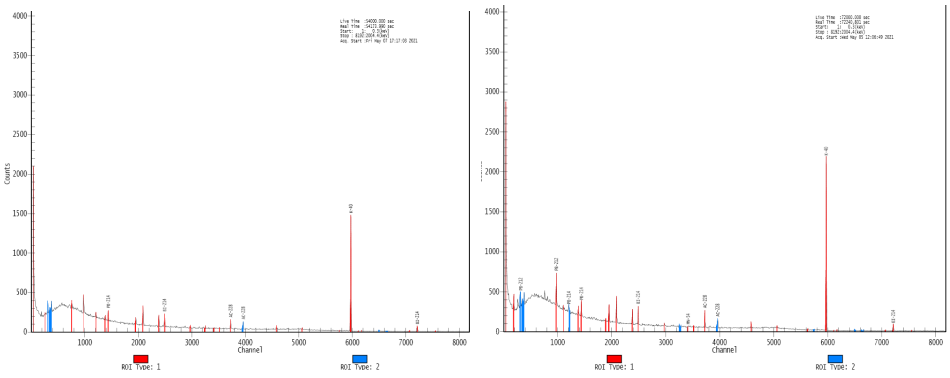
The calculated values of H_{ex} and H_{in} index for agricultural soil samples ranged from 0.44 to 0.72, and 0.51 to 0.96, with a mean value 0.63 and 0.72, respectively as illustrated in Table 4.2. The value of H_{ex} and (H_{in}) index must be less than unity to keep the radiation insignificant.The mean value of H_{ex} and (H_{in}) index calculated in this work is less than one but the value of H_{ex} and (H_{in}) index of some samples are high or relative to one.

4.3.5 Radioactivity Level Index

The calculated radioactivity level index (I_γ) in soils sample is ranging from 0.61 ± 0.03 to 1.21 ± 0.05, with an average values of 0.90 ± 0.03 (as illustrated in Table 4.2). The estimated values of (I_γ) in some soil samples are higher than one [43] and an average less than one.

4.4 Activity Concentration of Natural Radionuclides in Khat Plant

The result of gamma spectrum for each of gamma emitter graphically as shown in fig.4.2a and b.



(a) Spectrum from Khat sample L4 (b) Spectrum from Khat sample L6

Figure 4.3: Khat samples output Spectrums on a and b

The quantitative analysis of natural radioactivity of ^{238}U, ^{232}Th, and ^{40}K in Khat (Catha edulis) were determined by using HPGe p-type gamma spectrometry.

The activity concentration of natural radioactivity of ^{238}U, ^{232}Th, and ^{40} K in Catha edulis samples were determined. The confidence presence of those radionuclides

of ^{232}Th,and ^{40} K in Khat samples around 97% . Table 4.4 illustrates the radionuclide concentration in the sample under investigation. As shown from this table the average concentrations of ^{238}U, ^{232}Th, and ^{40}K in Khat were 1.32 ± 0.24, 10.36 ± 0.96, and $273.39 \pm 7.39, Bqkg^{-1}$, respectively. The concentration levels value of ^{232}Th, and ^{40}K low when compared with the average concentration of ^{232}Th, and 40K, 50.25 ± 4.88, and $687 \pm 38.18 Bqkg^{-1}$,respectively in Yemen's Khat (Catha edulis) analyzed using Wave Dispersive X-ray Fluorescence spectrometry (WDXRF) and n-type germanium detector equipped with epoxy window. The activity concentration levels of ^{238}U obtained is higher than the value of ^{238}U i.e 0.285 ± 0.05 in this paper. In this work case quantitative analysis some toxic, radioactivity, and heavy metal were determined, namely; ^{238}U series(^{214}Bi and ^{214}Pb),^{232}Th (^{228}Ac),^{40}k and ^{54}Mn.

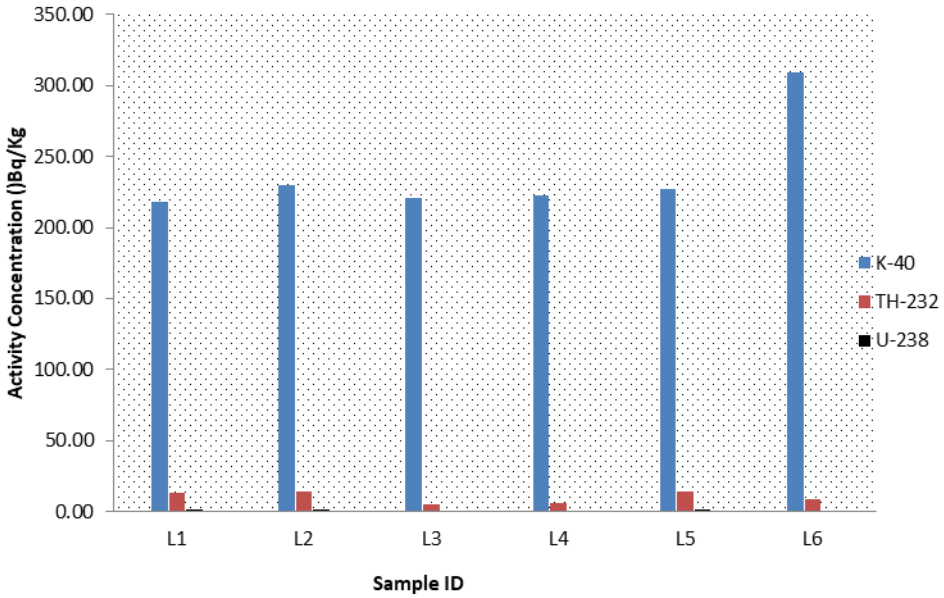


Figure 4.4: $^{238}U, 232Th$ and ^{40}K activity in leaves of Khat plant sampled.

Therefore, it is expected that the massive use of Khat (Catha edulis) may increase the concentration of these elements inside the human body and consequently increase the incidence of many diseases, especially oral and lung cancer [22].

4.5 Radiological Hazard Assessment

The radiation hazard indices (radium equivalent activity and internal hazard indices) were calculated for all samples in Khat plant as shown in Table 4.4. The radium equivalent activity, and internal hazard indices were varied from (25.20 ± 2.471) to(39.50 ± 2.137, $BqKg^{-1}$ with an average (34.44 ± 2.186, $BqKg^{-1}$ and from (0.05) to (0.11) with an average (0.09), respectively.

Table 4.4: Activity concentration, Radium equivalent activity and internal hazard index of the natural radionuclides of ^{40}K, ^{238}U and ^{232}TH measured in the Khat (Catha edulis) sample $Bqkg^{-1}$.

Sample ID	^{238}**U**	^{232}**TH**	^{40}**K**	Ra_{eq}	H_{in}
L1	1.32±0.21	13.68±0.87	218.16	37.68±1.830	0.05±4.94
L2	1.39±0.22	14.25±1.01	229.93±4.92	39.48±2.047	0.11
L3	1.14±0.29	4.95±0.97	220.58±10.20	25.20±2.471	0.07
L4	1.04±0.25	6.06±0.83	222.36 ±5.33	26.83±1.842	0.08
L5	1.97±0.28	14.03±1.01	226.78± 5.46	39.50±2.137	0.11
L6	1.07±0.23	9.16±1.07	308.91±13.45	37.95±2.789	0.1
Average	1.32±0.24	10.36±0.96	237.79 ±7.39	34.44±2.186	0.09

The values of the radiation hazard indices in this study radium equivalent activity indices is lowest value in sample (L3), 25.20±2.047 and the highest value in sample (L2, 39.48 ± 2.047 $BqKg^{-1}$ and internal indices is lowest value in all sample . This indicates that the average internal hazard index in Khat samples was lower than the permissible limits of a unity recommended by UNSCEAR, while the radium equivalent activity also were lower than the maximum permissible level of 370 Bq/kg recommended by UNSCEAR [10].

4.5.1 Annual Ingestion Effective Dose

Table 4.6 shows the results of the Ingestion effective dose in (mSv/y) for adult due to specific activity of ^{238}U, ^{232}Th, and ^{40}K in Khat(Catha edulis) samples which it is calculated using Eq. (3.10). The average ingestion effective dose due to ^{238}U, ^{232}Th, and $40K$ calculated for adult in Khat sample is, 0.2±0.04, 1.31 ±0.12, and, 0.81 ±0.03, mSv/y,respectively, with the total summation an average, 2.32±0.18$mSvy^{-1}$. The average of ingestion effective dose due to ^{232}Th is higher than due to ^{238}U and ^{40}K because of the increased the dose conversion coefficient for ingestion and activity concentration of radionuclides. The calculated values of effective dose due to intake Khat in all samples are higher than the permissible limit of the total effective dose from inhalation and ingestion of terrestrial radionuclides, which report by UNSCEAR [10], 0.31 mSv/y, of which 0.17 mSv/y is from $40K$ and 0.14 mSv/y is from the long-lived radionuclide in uranium and thorium series. Therefore, in case of this work, the total ingestion effective dose for adult in khat plant is approximately eight times higher than

permissible limit of 0.3 mSv/y [38].

Table 4.5: Ingestion effective dose for adult in Khat(Catha edulis) samples.

Sample ID	^{238}**U**$(mSvy^{-1})$	^{232}**TH**$(mSvy^{-1})$	^{40}**K**$(mSvy^{-1})$	ADR $(mSvy^{-1})$
L1	0.20±0.032	1.73±0.110	0.74±0.017	2.67 ± 0.16
L2	0.21±0.033	1.80±0.128	0.78±0.017	2.79±0.18
L3	0.17±0.045	0.62±0.1230	0.75±0.035	1.55±0.20
L4	0.16±0.038	0.76 ±0.105	0.76±0.018	1.68±0.16
L5	0.30±0.043	1.77 ± 0.127	0.77±0.019	2.84±0.19
L6	0.16±0.035	1.15±0.135	1.05±0.046	2.37±0.22
Average	**0.2±0.04**	**1.31 ±0.12**	**0.81±0.03**	**2.32±0.18**

Table 4.6: Reference of annual effective dose from natural source,reported by UNSCEA, [10].

Natural Source Exposure	**Annual average dose(worldwide)mSv/y**
External terrestrial	$0.48 \approx 0.5$
Ingestion	$0.29 \approx 0.3$
Inhalation	1.26
Cosmic Radiation	0.39
Total	2.4

5 Conclusion and Recommendation

5.1 Conclusion

The activity concentration of primordial radioactivity and their radiological hazard indices in agricultural soil and leaves of Khat plant samples collected from Haramaya University area were measured using gamma spectroscopy coupled with HPGe detector. Radioactivity of primordial radionuclides(^{238}U, ^{232}Th, ^{40}K),and ^{137}CS, was investigated in collected soil samples. Also in leaves of Khat plants, radioactivity, and heavy metal were determined, namely: ^{238}U series ^{214}Bi and ^{214}Pb, ^{232}Th series ^{228}Ac, ^{40}K and Mn. The primordial radionuclides of ^{238}U, ^{232}Th, and ^{40}K were the three most important detected in this study zone.

The average value of measured activity concentration of ^{232}Th, and ^{40}K in soil samples are higher than the given world average certified values of 30 $BqKg^{-1}$, and 400 $BqKg^{-1}$, respectively as seen in section 4.2. However, the average concentration of ^{238}U is lower than average certified values of $35BqKg^{-1}$. The calculated average absorbed dose and annual effective dose in soil samples are above world average certified values of $59nGyh^{-1}$, and $0.07mSvy^{-1}$, respectively and lower than maximum health limit. Also the calculated average values of gamma level index (I_γ) of some soil samples are slightly higher than one and on average less than one. This indicates that the radiation hazard from terrestrial natural radionuclides in soil of study area is insignificant. The obtained results of average activity concentration of ^{232}Th, and ^{40}K in leaves of Khat samples lower than similar work reported by [23], but the average concentration of ^{238}U is higher than the value reported in this paper. The annual ingestion effective dose for adult due to specific activity of primordial radionuclides of ^{238}U, ^{232}Th, and ^{40}K in leaves of Khat samples is higher than the world average of $0.30mSvy^{-1}$ and maximum permissible limit of $1mSvy^{-1}$.

The study established a map of baseline and serves as a reference for future similar works.

5.2 Recommendation

The study was conducted using soil and Khat samples from Khat plantation farms of Haramaya woreda. The study area can be classified as an area with increased background radioactive radiation, according to the findings and the abundance of three primordial radionuclides in present plant and soil samples radiation raises the intake radiation dosage in the human body. Moreover, chewing chat over a daily basis more raises the health risk, which can induce oral and lung cancer in the long run and more study need to be carried out on this area to ensure that Khat users are not exposed to radiation, radiation protection measures were required to assure the local population's radioactive safety.

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Appendix I

Peak Locate Analysis Report

Peak Locate Analysis Report 5/10/2021 4:08:15 PM Page 1

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***** P E A K L O C A T E R E P O R T *****

****

Detector Name: B13010
Sample Title: Environmental
Peak Locate Performed on: 5/10/2021 4:08:15 PM
Peak Locate From Channel: 1
Peak Locate To Channel: 8192
Peak Search Sensitivity: 3.00

Peak No.	Centroid Channel	Centroid Uncertainty	Energy (keV)	Peak Significance
1	41.58	0.0895	10.24	39.68
2	65.86	0.3476	16.18	3.09
3	162.87	0.2489	39.91	5.13
4	190.09	0.1704	46.57	11.44
5	218.19	0.2948	53.45	3.89
6	236.59	0.2743	57.95	4.17
7	258.50	0.1652	63.31	11.17
8	305.76	0.0707	74.88	65.18
9	315.34	0.0580	77.22	90.94
10	331.48	0.2893	81.17	3.67
11	344.47	0.1217	84.35	18.51
12	356.44	0.0813	87.27	40.48
13	367.74	0.0929	90.04	33.94
14	380.97	0.0925	93.28	30.44
15	406.72	0.1637	99.58	11.00
16	431.37	0.1592	105.61	10.28
17	444.13	0.2870	108.73	3.28
18	470.74	0.1944	115.24	7.52
19	527.79	0.1082	129.20	24.79
20	588.50	0.2688	144.05	4.62
21	629.54	0.1656	154.09	9.36
22	760.27	0.1023	186.08	24.58
23	855.55	0.0821	209.39	38.68
24	882.72	0.2262	216.04	4.41
25	938.87	0.3051	229.77	3.39
26	975.57	0.0390	238.75	164.86
27	986.95	0.1082	241.54	26.44
28	1032.28	0.2801	252.63	3.71
29	1104.73	0.0872	270.35	32.50
30	1134.06	0.1055	277.53	23.92
31	1177.44	0.2168	288.14	5.88
32	1206.83	0.0746	295.33	45.35
33	1226.92	0.0872	300.25	33.72
34	1340.86	0.0907	328.13	30.25
35	1358.12	0.2168	332.35	5.71
36	1383.11	0.0577	338.46	72.72
37	1438.64	0.0606	352.05	64.63
38	1657.24	0.2935	405.53	3.19
39	1674.01	0.1087	409.64	19.78
40	1851.39	0.1932	453.03	5.98
41	1892.83	0.0805	463.17	35.15

Peak No.	Centroid Channel	Centroid Uncertainty	Energy (keV)	Peak Significance
42	2087.98	0.0705	510.92	44.25
43	2234.78	0.2936	546.84	3.09
44	2247.23	0.2916	549.88	3.23
45	2299.20	0.1396	562.60	11.34
46	2384.14	0.0475	583.38	94.59
47	2490.97	0.0609	609.52	57.56
48	2554.01	0.2943	624.94	3.40
49	2576.71	0.2835	630.49	3.14
50	2704.89	0.1152	661.86	16.08
51	2720.82	0.2471	665.75	3.31
52	2973.18	0.0701	727.49	42.02
53	3035.36	0.2525	742.71	3.76
54	3087.28	0.1445	755.41	10.32
55	3120.99	0.1780	763.66	6.91
56	3141.01	0.1345	768.56	11.52
57	3156.99	0.1181	772.47	15.53
58	3197.11	0.1844	782.28	6.72
59	3211.28	0.1212	785.75	14.14
60	3249.75	0.0834	795.16	28.98
61	3296.07	0.2063	806.50	5.05
62	3394.22	0.1967	830.51	5.11
63	3416.34	0.1146	835.92	15.33
64	3435.50	0.1501	840.61	8.70
65	3517.95	0.0805	860.78	30.88
66	3652.94	0.2343	893.81	3.95
67	3696.50	0.1550	904.47	8.55
68	3725.07	0.0489	911.46	81.08
69	3818.13	0.1619	934.22	7.39
70	3920.06	0.2006	959.16	5.28
71	3943.89	0.0800	964.99	31.13
72	3961.28	0.0566	969.25	58.45
73	4093.05	0.1922	1001.49	5.40
74	4224.81	0.2402	1033.72	3.10
75	4353.82	0.2549	1065.29	3.18
76	4409.94	0.1786	1079.02	5.82
77	4471.98	0.1694	1094.20	7.28
78	4539.36	0.1999	1110.68	5.33
79	4579.90	0.0909	1120.60	23.16
80	5062.00	0.1293	1238.55	11.52
81	5238.64	0.2336	1281.77	3.25
82	5615.90	0.2602	1374.07	3.26
83	5632.37	0.1204	1378.10	11.98
84	5730.64	0.2032	1402.15	4.38
85	5755.88	0.1493	1408.32	8.03
86	5972.14	0.0332	1461.23	150.33
87	6115.56	0.1288	1496.32	10.00
88	6138.68	0.1553	1501.98	6.94
89	6169.92	0.1616	1509.62	7.38
90	6185.51	0.1909	1513.43	5.46
91	6308.65	0.2813	1543.56	3.04
92	6365.15	0.2240	1557.39	3.07
93	6460.66	0.1556	1580.76	7.25
94	6492.74	0.0880	1588.60	21.68
95	6510.70	0.1308	1593.00	10.92
96	6537.50	0.2199	1599.56	4.50
97	6625.85	0.1113	1621.17	13.29
98	6666.14	0.1042	1631.03	14.86
99	6697.92	0.1567	1638.80	6.34
100	6792.34	0.1965	1661.90	4.43
101	6813.60	0.1899	1667.11	4.80
102	7071.56	0.1178	1730.22	11.79
103	7213.82	0.0826	1765.03	22.22
104	7552.97	0.1471	1848.00	6.76

? = Adjacent peak noted

Appendix II

Peak Analysis Report

Peak Analysis Report 5/10/2021 4:08:29 PM Page 1

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***** P E A K A N A L Y S I S R E P O R T *****
****

Detector Name: B13010
Sample Title: Environmental
Peak Analysis Performed on: 5/10/2021 4:08:29 PM
Peak Analysis From Channel: 1
Peak Analysis To Channel: 8192

	Peak No.	ROI start	ROI end	Peak centroid	Energy (keV)	FWHM (keV)	Net Peak Area	Net Area Uncert.	Continuum Counts
F	1	38-	50	43.33	10.67	1.05	5.95E+003	100.27	6.87E+003
F	2	154-	169	162.90	39.92	1.13	4.94E+002	71.37	9.13E+003
	3	181-	196	190.09	46.57	0.86	5.45E+002	170.34	9.47E+003
M	4	212-	265	237.13	58.08	1.16	3.23E+002	80.38	1.16E+004
m	5	212-	265	258.47	63.31	1.17	1.52E+003	94.64	1.25E+004
M	6	296-	390	305.87	74.90	1.19	1.37E+004	152.44	1.53E+004
m	7	296-	390	315.24	77.19	1.19	1.85E+004	156.09	1.45E+004
m	8	296-	390	344.65	84.39	1.20	2.46E+003	79.87	1.31E+004
m	9	296-	390	356.46	87.28	1.21	7.81E+003	114.13	1.18E+004
m	10	296-	390	367.69	90.03	1.21	7.25E+003	139.67	1.20E+004
m	11	296-	390	380.92	93.26	1.22	6.82E+003	116.89	1.14E+004
M	12	396-	452	406.62	99.55	1.23	1.50E+003	88.42	1.11E+004
m	13	396-	452	431.11	105.54	1.23	1.62E+003	90.56	1.14E+004
m	14	396-	452	444.22	108.75	1.24	3.18E+002	83.76	1.16E+004
F	15	466-	478	470.76	115.24	1.25	8.16E+002	83.18	9.13E+003
F	16	521-	534	527.75	129.19	1.26	3.03E+003	101.67	1.07E+004
F	17	623-	640	629.78	154.15	1.29	1.17E+003	86.41	1.22E+004
F	18	749-	770	760.23	186.07	1.32	3.53E+003	97.51	1.31E+004
M	19	848-	891	855.53	209.38	1.34	5.28E+003	106.35	8.67E+003
m	20	848-	891	883.04	216.11	1.35	5.54E+002	75.59	9.31E+003
M	21	967-	998	975.62	238.76	1.37	5.32E+004	248.65	8.84E+003
m	22	967-	998	986.61	241.45	1.37	6.93E+003	110.27	7.77E+003
F	23	1097-	1112	1104.76	270.36	1.40	4.29E+003	91.18	5.75E+003
F	24	1127-	1145	1134.07	277.53	1.40	2.29E+003	78.38	6.73E+003
F	25	1172-	1183	1177.61	288.18	1.41	3.77E+002	59.85	3.90E+003
M	26	1199-	1234	1206.85	295.34	1.42	5.61E+003	97.48	5.39E+003
m	27	1199-	1234	1226.91	300.25	1.42	3.55E+003	85.41	5.34E+003
M	28	1329-	1391	1340.83	328.12	1.44	3.17E+003	81.28	5.39E+003
m	29	1329-	1391	1358.02	332.33	1.44	2.51E+002	57.58	5.21E+003
m	30	1329-	1391	1383.11	338.46	1.45	1.15E+004	126.40	5.40E+003
F	31	1429-	1446	1438.65	352.05	1.46	9.71E+003	114.68	4.35E+003
F	32	1651-	1683	1674.09	409.66	1.50	1.67E+003	63.76	6.72E+003
F	33	1844-	1863	1851.84	453.14	1.53	4.38E+002	47.08	3.22E+003
F	34	1883-	1903	1892.81	463.17	1.53	3.64E+003	76.89	3.60E+003
F	35	2075-	2100	2087.85	510.89	2.34	6.98E+003	101.96	4.07E+003
F	36	2290-	2308	2299.10	562.57	1.59	7.37E+002	47.90	2.46E+003
F	37	2371-	2397	2384.16	583.38	1.60	1.92E+004	147.06	3.43E+003
F	38	2480-	2501	2490.98	609.52	1.62	7.63E+003	98.49	2.78E+003
M	39	2549-	2583	2553.88	624.91	1.62	1.05E+002	37.02	1.71E+003
m	40	2549-	2583	2575.73	630.25	1.63	1.34E+002	38.49	2.17E+003
M	41	2696-	2727	2704.98	661.88	1.64	1.03E+003	51.37	2.49E+003
m	42	2696-	2727	2720.92	665.78	1.65	2.11E+002	40.85	2.18E+003

	Peak No.	ROI start	ROI end	Peak centroid	Energy (keV)	FWHM (keV)	Net Peak Area	Net Area Uncert.	Continuum Counts
F	43	2960	2986	2973.21	727.50	1.68	4.61E+003	79.33	2.89E+003
F	44	3030	3041	3035.43	742.73	1.68	1.38E+002	35.19	1.18E+003
M	45	3078	3168	3087.51	755.47	1.69	6.00E+002	44.38	2.03E+003
m	46	3078	3168	3121.23	763.72	1.69	3.92E+002	41.19	2.18E+003
m	47	3078	3168	3140.97	768.55	1.70	7.69E+002	46.34	2.14E+003
m	48	3078	3168	3157.16	772.51	1.70	9.46E+002	47.81	2.11E+003
M	49	3191	3222	3197.37	782.35	1.70	3.56E+002	37.79	1.44E+003
m	50	3191	3222	3211.39	785.78	1.70	9.42E+002	45.91	1.90E+003
F	51	3239	3260	3249.69	795.15	1.71	2.52E+003	61.94	1.95E+003
F	52	3290	3302	3295.88	806.45	1.71	2.15E+002	34.86	1.07E+003
M	53	3384	3445	3394.85	830.66	1.73	3.22E+002	37.43	1.82E+003
m	54	3384	3445	3416.31	835.91	1.73	9.35E+002	45.46	1.82E+003
m	55	3384	3445	3435.18	840.53	1.73	5.06E+002	40.32	1.81E+003
F	56	3504	3530	3517.96	860.78	1.74	2.62E+003	62.24	2.16E+003
F	57	3639	3662	3652.62	893.73	1.75	1.66E+002	33.63	1.77E+003
M	58	3688	3739	3696.47	904.46	1.76	3.64E+002	38.31	1.56E+003
m	59	3688	3739	3725.03	911.44	1.76	1.52E+004	129.42	1.65E+003
F	60	3809	3828	3817.97	934.18	1.77	3.66E+002	36.25	1.43E+003
M	61	3910	3972	3919.72	959.08	1.78	1.21E+002	28.39	1.60E+003
m	62	3910	3972	3944.01	965.02	1.78	2.83E+003	52.23	1.67E+003
m	63	3910	3972	3961.19	969.22	1.79	8.87E+003	83.01	1.73E+003
F	64	4087	4102	4092.87	1001.44	1.80	2.26E+002	33.29	1.12E+003
F	65	4219	4233	4225.22	1033.83	1.81	1.49E+002	31.10	9.95E+002
F	66	4338	4365	4352.95	1065.08	1.83	1.77E+002	33.59	2.03E+003
F	67	4401	4421	4410.13	1079.07	1.83	3.37E+002	35.24	1.41E+003
F	68	4460	4479	4471.70	1094.13	1.84	3.02E+002	37.28	1.55E+003
F	69	4532	4552	4539.58	1110.74	1.84	2.40E+002	35.86	1.63E+003
F	70	4567	4590	4579.89	1120.60	1.85	1.85E+003	55.71	1.93E+003
F	71	5047	5075	5061.96	1238.54	1.89	7.17E+002	46.82	2.97E+003
F	72	5230	5245	5238.52	1281.74	1.91	1.55E+002	31.93	1.04E+003
M	73	5605	5645	5616.74	1374.28	1.94	1.31E+002	23.13	6.59E+002
m	74	5605	5645	5632.26	1378.07	1.95	6.16E+002	32.25	6.39E+002
M	75	5723	5767	5730.10	1402.01	1.95	1.26E+002	22.00	4.99E+002
m	76	5723	5767	5755.70	1408.28	1.96	2.58E+002	25.39	6.21E+002
F	77	5955	5988	5972.09	1461.22	1.97	6.11E+004	247.89	7.14E+002
M	78	6105	6193	6115.58	1496.33	1.99	3.38E+002	24.24	3.33E+002
m	79	6105	6193	6138.80	1502.01	1.99	2.11E+002	21.19	3.73E+002
m	80	6105	6193	6170.39	1509.74	1.99	1.56E+002	20.15	3.84E+002
m	81	6105	6193	6185.43	1513.42	1.99	1.18E+002	19.34	3.27E+002
F	82	6353	6375	6364.76	1557.29	2.01	7.06E+001	15.26	2.50E+002
M	83	6449	6545	6461.06	1580.85	2.01	2.42E+002	20.95	2.90E+002
m	84	6449	6545	6492.75	1588.61	2.02	1.41E+003	40.87	2.81E+002
m	85	6449	6545	6510.42	1592.93	2.02	4.85E+002	25.90	2.54E+002
m	86	6449	6545	6537.28	1599.50	2.02	5.43E+001	14.76	2.16E+002
M	87	6613	6707	6625.96	1621.20	2.03	5.95E+002	28.31	2.94E+002
m	88	6613	6707	6666.15	1631.03	2.03	7.11E+002	30.03	2.64E+002
m	89	6613	6707	6697.63	1638.73	2.03	1.82E+002	18.54	2.17E+002
M	90	6785	6824	6792.30	1661.89	2.04	7.60E+001	15.80	2.25E+002
m	91	6785	6824	6813.81	1667.16	2.04	6.76E+001	15.41	2.51E+002
F	92	7060	7088	7071.81	1730.28	2.06	4.56E+002	24.93	2.56E+002
F	93	7199	7230	7213.75	1765.01	2.07	1.70E+003	43.31	2.69E+002
F	94	7539	7565	7552.81	1847.96	2.10	2.73E+002	19.94	2.02E+002

M = First peak in a multiplet region
m = Other peak in a multiplet region
F = Fitted singlet

Appendix III

Nuclide Identification And Gamma-Spectrum Report

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Interference Corrected Activity Report    5/10/2021   4:09:36 PM   Page  1

**************************************************************************
*****   N U C L I D E   I D E N T I F I C A T I O N   R E P O R T   *****
**************************************************************************

     Sample Title:         Environmental
     Nuclide Library Used: C:\GENIE2K\CAMFILES\Env with outh CD-109

   .....................    IDENTIFIED NUCLIDES     .....................

   Nuclide     Id        Energy     Yield      Activity      Activity    Coinc
   Name     Confidence   (keV)       (%)      (Bq /kg  )    Uncertainty  Corr

   K-40        0.974   1460.81*     10.67   1.04596E+003  4.34368E+001 err
   CS-137      0.992    661.65*     85.12   1.43424E+000  1.11974E-001 err
   PB-212      0.997     77.11*     17.50   7.72073E+001  7.74893E+000 err
                       238.63*     44.60   7.43401E+001  5.95762E+000 err
   BI-214      0.984    609.31*     46.30   1.95645E+001  1.27854E+000 err
                      1120.29*     15.10   2.00624E+001  1.11674E+000 err
                      1764.49*     15.80   2.00499E+001  1.00618E+000 err
   PB-214      0.997    295.21*     19.20   2.01893E+001  1.65567E+000 err
                       351.92*     37.20   2.00451E+001  1.62241E+000 err
   AC-228      0.990    911.21*     26.60   1.24468E+002  8.14926E+000 err
                       968.97*     16.20   1.23346E+002  7.54255E+000 err

       * = Energy line found in the spectrum.
       @ = Energy line not used for Weighted Mean Activity
       Energy Tolerance :    1.000 keV
       Nuclide confidence index threshold =   0.30
       Errors quoted at  1.000 sigma
       Coincidence correction performed.
       free = No coincidence correction required.
       miss = Nuclide energy was not found in the coincidence library.
       err  = Error in coincidence correction calculation.
       ISOCS/LabSOCS/Coinc. Corr. Warning/error code =        537199722

**************************************************************************
*****        G A M M A   S P E C T R U M   A N A L Y S I S         *****
**************************************************************************


Filename: C:\GENIE2K\CAMFILES\2021 sample Data\mesay\soil\ERTL-L5-2021.

Report Generated On             : 5/10/2021   4:09:51 PM

Sample Title                    : Environmental
Sample Description              : soil
Sample Identification           : ERTL-L5-2021
Sample Type                     : soil
Sample Geometry                 : 538G-E

Peak Locate Threshold           :  3.00
Peak Locate Range (in channels) :     1 -  8192
Peak Area Range (in channels)   :     1 -  8192
Identification Energy Tolerance :    1.000 keV

Sample Size                     :  8.750E-001 kg

Sample Taken On                 :
Acquisition Started             : 5/6/2021    8:12:58 AM

Live Time                       :    28800.0 seconds
Real Time                       :    28933.2 seconds

Dead Time                       :   0.46 %
```