Comparative Analysis of Radionuclides in Farm Soil from Bilajig village North Gondar Ethiopia

By

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This Work is Dedicated to
My Wife
Genet Tekile Mariam

Table of Contents

List of Tables

List of Figures

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Symbols Used

τ_d	dead time
τ_R	recovery time
m	number of measured events per unit time
N	disintegration rate
ΔV	voltage drop on register
R	resistance
V_0	potential at anode
S	pleatue slope
V_1	threshold voltage
V_2	knee voltage
N_i	count rates
ϵ	dielectric constance
e	electric charge
$\frac{p}{c}$	peak compton ratio
L_c	critical level
P_r	probability
N_D	minimum detectable value
rsd_Q	relative standard deviation
σ_Q	standard deviation of estimated valueat quantification limit
N_Q	minimum quantifiable value
N_T	number of count with unknown sample
N_β	background count
σ	standard deviation
E	energy
$\varepsilon(E)$	energy dependent peak efficiency
f	emission probability
A	radioactivity in B_q

Abstract

Soil samples were collected, in February 2009, from a farm land in Bilajig village near Gondar town North West Ethiopia. The soil samples were taken from a farm land that uses chemical fertilizers for nearly three decades and from farm plot that has never used chemical fertilizers. Samples were also taken from border segments on which grass only is grown. The soil samples were placed for two months to settle into equilibrium. Gross beta counting using GM counter and gamma spectrometry were proposed methods to detect radioactivity in the soil samples.

Attempts to obtain detectable activity using gross beta counting method did not bear result as the activity in the samples was below the critical level which implies that the farm soils radioactivity was below the detection limit of our GM system.

Using high resolution HPGe detection system, however, the presences of radionuclides originating from ^{238}U (such as ^{226}Ra; ^{214}Pb and ^{214}Bi) and Thorium series members (such as ^{228}AC and ^{212}Pb) were detected. Further ^{40}K and ^{137}Cs were also detected.

Quantitative analysis of the samples were done using the concentration in the farm soil that has never used chemical fertilizers as unit. Thus the relative concentrations of ^{40}K and ^{137}CS is nearly the same for all farm soil samples.

When it come to the relative concentration of the natural series members significant presence of ^{214}Pb is observed, ranging from 14 to 20 times, on farm plots using chemical fertilizer.

Key words: radionuclides, gamma spectrometry, farm soils, GM counter, gross beta, counting rate

Introduction

Radiation is believed to be a constituent part of the environment since the days of the “big bang”. The natural radiation environment is composed of radiations of cosmic and terrestrial origin.

Radiations may have both harmful and beneficial aspects for living things. The availability and spatial composition of the sources of these radiation are altered due to human activities from time to time. In addition, artificially produced radio elements can get into the natural environment due to human activities ranging from power generation to detonations.

For example the concentration of Thorium in coals is redistributed due to the use of huge amount of coals in coal fired power plants. Humans add fertilizers in the soils used for agriculture to increase the yield of the farm land. The continuous and unchecked use of fertilizers in farm soils will substantially alter the natural distribution of radioisotopes in the soil.

Enhanced concentration of radioisotopes, may find their way to transfer from the environment to the living web complex. Most human foods in one way or another derived from soil, that anchors living organisms.

In a country like Ethiopia where the livelihood of the people largely depend on the agriculture economic activity the fertility and contamination level of the arable soil should be known and checked for changes that may happen due to human activities. The use of fertilizers in agriculture is belived to be started in large mechanized farms duirng the reign of Emperor Haile Selassie I. Since then the use has grown with time and these days small plots of lands owned by individual farmers are dependent on the use of artificial chemical fertilizers in order to get high yield in agricultural plot lands.

Although the natural radioactivity may vary considerably from one type of soil to another. The sources of radioactivity in soils other than those of natural origin are mainly due to, extensive use of fertilizers rich in phosphates for agricultural purposes and routine authorized low level radioactive effluent discharges or accidental release into the environment from nuclear fuel cycle installations, mineral extraction industries, industries working with mineral materials enriched in naturally active elements (Abbady et al., 2000).

The knowledge about the radioelement in soil used for many purposes, such as it enables to classify and identify soil type, possible to understand the rate of contamination of the soil by different radioelement. There are some works in the study of natural radioactivity of soil and rocks in certain specified areas of southern Ethiopia (Solomon Bekele, 1995, Investigation of natural radioactivity of soil and rock samples from Kenticha tantalite processing area of Sidamo region: Thesis, Addis Ababa University).

There are also scientific studies that showed the large difference in the composition of the radioactive elements in virgin land and land used extensive chemical fertilizers (Becegato et al., 2008).

In this work the comparative analysis of radioactive elements from different farm land soil are investigated and findings are reported.

Objectives of the study

General Objective

Compare radionuclide concentrations in different soil samples and look for possible variations due to the use of artificial fertilizers in the farm soils.

Specific Objectives

- Identify radionuclides in the soil samples.
- Compare activity concentrations in different soil types. radionuclides

Significance of the study

The results of this study will contribute to avail a baseline data on the radionuclide concentration of farm soils and possible enhancement due to human activities.

Chapter 1

Radiation

Radiation is a form of energy that is transmitted by electro magnetic waves or by strong sub atomic particles. The variation in frequency of electro magnetic waves produce different energy levels, ranging from low frequency (radio wave) to high frequency (gamma rays). Low to mid frequencies is electromagnetic waves produce non ionizing radiations. When electromagnetic waves frequency is lower than approximately 10^{17} cycle per second it can absorbed in biological materials. In this case, electrons are excited but not removed and the energy is expend as heat. Non ionizing radiation is not believed to pose a significant danger to human, except at a very exposure (Science and Technology (WASI, 1985). Radio waves, radar waves, micro waves, infrared rays, visible light, and ultra violet rays are all forms of non-ionizing radiations.

Ionizing radiation, as stated above, consists of electromagnetic wave and material particle that have sufficient energy to ionize atoms of molecules, thereby radically modify their chemical behavior. In other words, ionization radiation consists of very energetic fast moving particles, such as, beta, alpha and neutron, and gamma rays. Such radiations can penetrate and interact with matter by removing electrons from the surrounding nuclei thus producing ions. The penetration of high energy radiation into matter has two complementary aspects: The effect of the interaction upon the radiation and the effects upon the matter. These two effects may become clear in the topics discussed in other chapters.

The source of ionization radiations are unstable nuclides that a matter which emit radiation is called radioactive. In understanding radioactive decay, the radiation is actually emitted from the nucleus of radioactive atom. Furthermore, in radioactive process there is a parent and daughter sequence. The parent atom emits the radiation and in so doing the parent transformed automatically to the daughter atom. The parent and the daughter, usually are different chemical substances. However, when gamma ray emitted from a parent radioactive element both the parent and the daughter would become the same chemical substance.

The rate of emitted radiation depends on the nature of corresponding radioactive material, that is, a material which can produce intense radiation has short half life while a material that emits very weak radiation would have long half life. Since half life is one of the characteristics of radioactive substances, it is independent of any physical or chemical state of the source. For example, if one had fuel oil containing radio carbon and radio hydrogen, and burned the fuel oil, the smoke would carry the same radioactivity to the same extent as was previously present in fuel oil. Then, radioactive process cannot be speed up, no slow down, but depends only the nature of radioactive substance involved. Fundamentally this is due to the fact that the atom, and all the chemical process like burning, melting, etc., disturb only outermost electrons of the atom and affects nucleus hardly at all.

The rate of decay,or activity of a radionuclides can be expressed as number disintegrations the nuclide undergoes per second. Marie Curie was first person that quantify radioactive decay during her study of radium-226. A gram of radium-226 undergoes $3.7X10^{10}$ disintegrations per second, a value known as the curie, abbreviated Ci. The modern unit radionuclide activity is becquerel (Bq), which is equal to one disintegration per second.

1.1 Penetrating Power of Radiation

Different kinds of radiations can penetrate different distances through different materials. However, for radiation consisting of a given kind of particle, the depth of penetration in a given kind of particle, the depth of penetration depends only on the energy of that particle (radiation). For alpha, beta and gamma rays, the depth of penetration is less in more dense substances. This penetrating power difference of these different radiations is listed below.

Alpha particles emitted by naturally occurring radioactive substances move at about one-twentieth the speed of light and will penetrate up to a distance five centimeter of air before being brought to rest. They will penetrate about 0.002 to 0.005 centimeter in tissue, and since the epidermal protective layer of dead skin on the body has maximum thickness of 0.007 centimeter, alpha particles are harmless when the source emitting is outside the body.

The beta particles consists of fast moving electron essentially moving with speed of light. They can penetrate distances of a few meters in air or 0.5 cm into water or tissue before being brought to rest (Botkin & Keller, 2005).

The gamma rays, are really the same kinds of things as light or radio waves, but the energy of a single gamma ray is very much greater, amounting to something like 1Mevs. A gamma ray does not penetrate definite distance into matter, but has a certain probability of being absorbed. If a large number gamma rays are fired into matter, after some distance only half of them will remain, and at double the distance only one fourth of them being remain, and so forth. gamma rays of 1Mev energy are reduced to half intensity by one centimeter of lead, or by 10 to 20 cm of water or 100 to 2oo meters in air (Mark, 1957, p.42).

1.2 The Effect of Radiation on Matter

The materials that we used in our every day lives are made group of atoms called molecules when molecule is bombarded by radiation, it may broken up into fragments consisting of one atom or small group of atoms. These atoms may then remain permanently broken up (for example, by irradiating water one can change of it into hydrogen or oxygen, or fragment of the original molecules may recombine to form a new molecule. Most of the adverse effects of radiation of new, unstable, molecules with the irradiated material. For these events, one can take the effect of ionization radiation on life and health resulting permanent change produced in genetic materials, the chromosomes or the contained genes, as an example. The same kind of radiation always had the same effect that is if a tissue is bombarded by a beta ray from different radio nuclieds, the effect on the tissue is the same.

Some of the ionizing radiation are beta particles, alpha particles, proton particles and gamma radiation. The particles beta, alpha and protons are charged particles and can be classified them into light charged particles (electron and positron) and heavy charge particles (proton and alpha). The other form of radiation is gamma radiation as the form of electromagnetic wave.

This ionization radiation can pass through matter some of them for considerable distance and with little degradation of their energy. Another (not possessed by all of them) is their electrical charge, which creates intense disturbance in the balance of the electrical forces that hold the constituents of atoms and molecules together. The term ionizing radiation giving to the type of radiation base on their ability to inject planetary electrons from atoms or molecules. The product of this reaction ions the negatively charged electrons and the electron deficient and thus positive charged atom our molecules.

The interaction of each type of radiation with matter depend upon the mass and the charge, if any, of the radiation as well as upon its energy. Because of the energy neutron, for example, has no electrical charge, the interact feebly with atoms and therefore have great penetrating power. Eventually, they give up their kinetic energy to the atomic nuclei that they happen to collide directly. In recolling from this collisions the atoms lose one or several of their orbital electrons and themselves become charged particles.

When a swiftly moving electron, alpha,or proton penetrates matter, the molecules near which it passes are subjected to an intense, transit electrical force. Since the binding of electrons in molecules is purely electrical in character, the new force is devastating (Platzman, 1959, p.75).

The organization of electrons within each molecule is disturbed and many molecules ionized after the particle passed. These two types of primary effect are very different in nature. In an excited molecules the acquired energy alters the motion of the planetary electrons, whereas one of these electrons is actually released and wanders away from ionized molecule. More energy is required to ionize a molecule than excite it. ’

On the average, some ten to twenty electron volt of energy are transferred in each primary event, and the penetrating particle is thereby continuously decelerated. Although the lost of kinetic energy occurs in discrete amounts, the changes are so minute compared to the total kinetic energy that slowing down can be considered, for most practical purposes to be continuous process. This forms of radiations differ in degree rather than in kind in their effects on matter (Platzman, 1959, p.76).

The effect of ionizing radiation with matter, such as, absorption of the energy of ionizing radiation give us many of our most important methods for detecting and measuring the radiation itself.

The behavior of every ionizing radiation detector depends on the interaction of ionizing radiation with sensitive materials of the detector. It is therefore, important to understand, the different processes which ionizing radiation interact with matter. In describing these processes it is better to use only the above classification of ionizing radiations emitted from the natural radioactive nuclide, that is, light charged particles and heavy charged particles. This way of classification is relevant because electron interact with matter is different from proton or alpha particles. Thus, the mechanism by which beta particles interact with matter and lose energy and change in direction are similar only in principle to those experienced by heavy charged particles and are very different in reality.

The chief reason of this difference is that electron had a very small mass. Thus, electrons by virtue their mass, undergo large angle of scattering and radiative collision.

The predominant mechanism for transferring energy from fast charge particles to matter is by inelastic collisions. In the case of heavy charged particles, the impact with an electron is insufficient to deflect the incident particle appreciably, and the later will follow an approximately straight path. This is not the case for an impact between two electrons whose masses are equal.

Scattering of heavy charged particles is due to mostly elastic collusion with nuclei. Electrons are also scattered by elastic collisions with nuclei but with much greater probability than heavy charged particles, so the electron can be deflected through the large angles by an electric field which is quite distant from the molecules. So that a nuclear spectroscopy, alpha particles or protons are usually mono-energetic, while electrons have continuous of distribution of energies. Therefore, it is not necessary for the electron to pass close to the nucleus as would be required in case of heavy charged particles with the same velocity.

1.3 Radionuclides in Soil

Soil is the result of the action of weather and humans activities on the crust rocks of the earth, that is, as the rock absorbs solar energy it become heated and expand although due to the fall of its temperatures it can be contracted. In addition to these effects the frost, rain, raise and fall of temperature facilitates the possible chemical interaction of the elements of the rocks, which causes for the fragmentation of the rocks into small size until it gets the texture of soil. Then we can say that the elements of the soil are the same as that of the crust rocks of the earth.

This formation of soil tell us something about the origin of radionuclides. Understanding of origins, distributions, and behavior of natural nuclides, those emit radiation spontaneously, developed in the first twentieth century, it becomes apparent that their inherent decay provides a means of determining the rate of natural processes and type of radionuclides present in the soil as natural radionuclides (terrestrial and cosmic ray produced radionuclides).

1.3.1 Terrestrial Radionuclides

Terrestrial sources of radiation are the very long lived radionuclides that have existed within the earth since its formation several billion years ago and have not substantially decayed. The most important of these so called primordial radionuclides are $^{40}_{19}\mathrm{K}$ (half life = 1.28X10^9, $^{87}_{43}\mathrm{Rb}$ (half life = 4.7X10^{10}) $^{238}_{92}\mathrm{U}$ (half life = 4.47X10^9years) and $^{232}\mathrm{Th}$ (half life = 1.41X10^{10} years), $^{238}\mathrm{U}$ and $^{232}\mathrm{Th}$ series elements that contribute to the doses from terrestrial sources see 1.1. Other radionuclides, such as those present in the $^{235}\mathrm{U}$ decay series, have been neglected,as they contribute little to the total dose from natural background.

Primordial radionuclides are widely distributed through the crust of the earth, occurring highly concentrated in certain minerals and in very dilute form particularly every where. Then soils are naturally radioactive, because of their mineral content. In old rock minerals and ores that have not been exposed to any action, there is a radioactive equilibrium, at ratio of the radioisotopes of various radionuclides obeys the law of radioactive equilibrium (Mesmayanov, 1954, p.285). This type of radioactive equilibrium is called secular equilibrium which is the condition in which the initial member of the decay series has longer half life than any subsequent member of series.

As a result of the breakdown of rocks (a step to transform rocks into soil particles) the constituent radionuclides are migrating and the radioactive equilibrium is disturbed. Radionuclide broken down away from the parent radionuclide uranium or thorium gradually disintegrate short lived elements rapidly disappear and only such elements as $^{230}\mathrm{Th}$, $^{231}\mathrm{Pa}$ and $^{226}\mathrm{Ra}$ survive.

As discussed in the previous paragraphs the definition of primordial radionuclides indicates that they are long lived species, which have been present on the earth since its formation some 4.5X10^9 years ago. These radionuclides $^{238}\mathrm{U}$, $^{235}\mathrm{U}$ and $^{232}\mathrm{Th}$ are parent member of the three natural radioactive decay series. A mass change of four is only change permitted by ordinary radioactive decay (alpha), each member of the series differs in mass number from the others by multiple of four. It has been suggested that the mode of series in the figure given by 4n+2, 4n+3 and 4n respectively, reflecting their mass number of various components (Bleuler & Goldsmith, 1959). From these radioactive series once can understand that all other natural radioactive elements occur in nature as the product of the radioactivity decay of uranium of thorium.

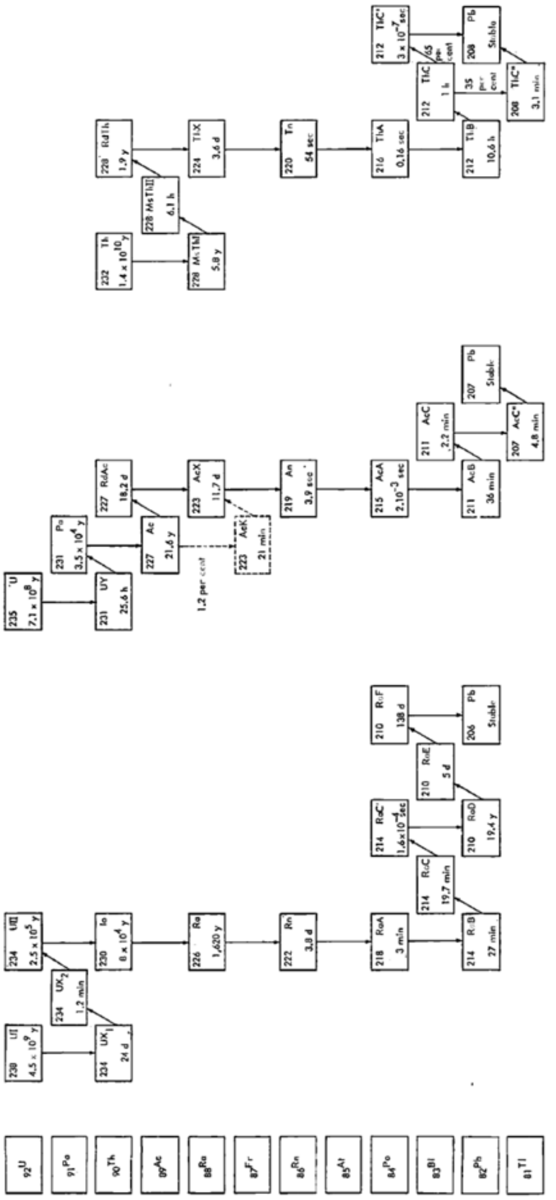

Figure 1.1: Natural Radioactive Series

Apart from uranium, thorium and their decay products, the radioactive isotopes of the elements such as K, Ca, Rb, and tin have also been found in nature (table). The average content of these elements in the earth's crust is about 0..1 percent. Therefore, with uranium, thorium, actinium and their decay products there are radioactive isotopes of other elements in the soil, for example radioisotope of potassium (^{40}K) and some of them are listed below.

1.3.2 Cosmic Ray Produced Radionuclides

The high energy radiation that enters the earth's atmosphere from other space is known as primary cosmic rays or galatic cosmic radiations. Secondary cosmic

Isotope	% isotope in natural element	Half life	Type of decay	Isotope	% isotope in natural element	Half life	Type of decay
^{40}K	0.0119	$1.3X10^{9}$	ß	^{138}La	0.089	$7X10^{10}$	α
^{48}Ca	0.179	$2X10^{16}$	ß	^{142}Ce	11.7	$5.1X10^{15}$	α
^{50}V	0.24	$> 3X10^{15}$	Ec	^{144}Nd	23.87	$5X10^{15}$	ß
^{87}Rb	27.85	$6.16X10^{10}$	ß	^{150}Nd	5.60	$5X10^{10}$	ß
^{96}Zr	2.80	$6.2X10^{16}$	ß	^{147}Sm	15.07	$6.7X10^{11}$	α
^{113}Cd	12.30	$6.2X10^{17}$	ß	^{176}Lu	2.60	$2.4X10^{10}$	ß
^{113}In	4.23	$> 10^{15}$	Ec	^{180}Ta	-----	$1X10^{12}$	ß or Ec
^{115}In	95.77	$6X10^{14}$	ß	^{180}W	6.126	$2.2X10^{17}$	α
^{124}Sn	6.11	$1.5X10^{17}$	ß	^{187}Re	62.93	$4X10^{12}$	ß
^{123}Tb	42.75	$>10^{14}$	ß	^{190}Pt	0.006	$5X10^{11}$	----
^{123}Te	0.88	$>10^{13}$	Ec	^{192}Pt	0.78	$Ca:10^{15}$	----
^{130}Te	34.11	$1.4X10^{21}$	ß	^{209}Bi	100	$27X10^{17}$	α

Figure 1.2: Natural radioactive isotope of non radioactive elements

radiation is produced when galactic cosmic radiation collides with atoms of air (oxygen, nitrogen and carbon) in upper layer of the atmosphere. The sun also emits cosmic radiation but this solar radiation has far less energy than galactic cosmic radiation. The energy of galactic cosmic radiation also much higher than the energy of any terrestrial nuclear radiation emitted by source such as uranium, radium and any other radioactive materials.

When the galactic and secondary cosmic radiation penetrates through the atmosphere, it is slowed down and the majority cosmic radiation stops in the atmosphere. The earth's atmosphere is, therefore, an effective cosmic radiation shelled. This means that the cosmic radiation intensity is much higher in upper layer of the atmosphere than at sea level (the altitude effect).

The interaction of primary cosmic rays (galactic rays with the nuclei of the atoms present in air, neutrons, protons, pions, and kaons (secondary cosmic rays) are produced, as well as a variety of reaction products (cosmogenic nuclides). The high energy secondary cosmic ray, thus formed react further with nuclei in the air to form more secondary particles (electrons and muons). Thus, cosmogenic nuclides, cosmic ray produced nuclides, are generated in the upper atomosphere in the interaction of cosmic rays with nuclei of atoms of atmospheric gases, such as, nitrogen and oxygen, The action of cosmic rays brings about splitting of atom, nuclei of atmospheric gases and, as consequence, fast neutrons appear among other things. The neutrons act on the nuclei of nitrogen atoms, as a result of which the radio isotope of carbon is formed by reaction: $^{14}N(n,p)\ ^{14}C$.

The resulting coil of atoms carbon 14 interact with oxygen, forming CO_2 which contains radioactive carbon (of half life=5730 years). And the interaction of nitrogen nuclei with neutrons created by primary cosmic radiation also gives rays radio isotope of hydrogen tritium with a half life of 12.3 years.

$$^{14}N +^{1} n \rightarrow^{12} C +^{3} H$$

In the upper layer of the atmosphere there also proceed the reaction

$$^{2}H(n,\gamma)^{3}H,^{3}He(n,p)^{3}H$$

In such a way a cosmic ray produced radionuclides are formed in the upper atmosphere and they are transported to the earth surface and their in corporation into biological and geological materials and subsequent decay provides the basis for a number of dating and tracer techniques. Cosmic ray produced radionuclides (cosmogenic radionuclides includes)
$^{3}H\ (12.3years)$, ^{10}Be (1.6X10^{5}),years
^{26}Al,(72X10^{5} years ^{32}Si,(178 years) ^{36}Cl
(3X10^{5} years) and ^{41}Ca,(1.0X10^{5} years)
^{7}Be,(53.28 days) ^{80}Kr, ^{14}C,(5730 years)
^{39}Ar, ^{22}Na, ^{35}S, ^{37}Ar, ^{33}P, ^{32}P,
^{38}Mg, ^{24}Na, ^{38}S, ^{31}Si, ^{18}F, ^{39}Cl,
^{34m}Cl (Mackenzie, 2000, p.360)

1.3.3 Man Made Radionuclides

Since 1945 there has been a large increase in the production of artificial radionuclides, originally with electrical sub atomic particle accelerators (e.g. the cyclotron), with small radium-Berilium neutron source, but, above all as a result of uranium fission and by neutron irradiation by the core of research or power- plant nuclear reactors. In the context of accidental releases there is no doubt that the fuel elements of a stationary or mobile (ship or submarine) nuclear reactor represents the greatest potential source of unintended contamination of the environment. In addition there is a possibility of an accidental release of fessile material and fission products as a result of an accident involving nuclear weaponry, which is outside the scope of this appraisal, as is the possible release of radioactive substances as a result of a war or terrorism.

Such manmade radionuclides enter the soil from varies activities, with the major sources being mentioned above and considered below.

Nuclear Weapons

First let see how to understand the term explosions. an explosion is the release of large quantity of energy in short interval of time and within a limited space. The release of this energy is accompanied by a very great increase in temperature so that the product of explosion become extremely hot gases. The explosion of air heated by nuclear detonation causes the formation of blast waves when the head of the wave (the shock front) passes a given point it results in an abrupt rise in pressure causing some of the destructive effects of the explosive.

The nuclear bomb is similar to the more conventional high explosion bomb in that a portion of it destructive action is due to the blast or shock discussed above. However, part from the fact the large TNT bomb, there are other more basic differences first, a fairly large portion of the energy from a nuclear explosion is emitted in the form of a nuclear light and heat. This emission is required to generally as "thermal radiation". It can causes fires or skin burns at considerable distances. Secondly, the explosion is accompanied by highly penetrating, but invisible, rays called "initial nuclear radiation". The substances remaining after explosion are large part of radioactive, emitting similar nuclear radiations over an extended period of time. This later radiation, arbitrarily taken as what which occurs later one minute after the bombs interaction, is commonly refer to as the "residual nuclear radioactivity".

Earlier nuclear weapons made use only the fission process in the achieving of this high energy nuclear detonation. Nuclear fission is splitting of nucleus into two parts, and it is triggered when a neutron strikes nuclei having large atomic mass and the neutron is absorbed. Plutonium is produced by fission of uranium (as shown below) when neutron strikes uranium-238, it is absorbed and uranium-239 is formed. Uranium-239 emit a beta particle and its decays. The final product is plutonium-239, which is the weapons- grade plutonium (Hanson, 2000).

$$^{238}U + n \to ^{239} U + \beta \to ^{239} Np + \beta \to ^{239} Pu$$

After having the weapons graded radionuclides, such as ^{239}Pu and ^{235}U, the process of thermal neutrons are causes of the fissionable $(^{235}U,^{239} Pu)$ materials split (fissioned) into fission products by almost instantaneous chain reaction. Such process for nuclear weapons also resulted in the release of large quantities of energy with radioactive fission products including ^{131}Cs, ^{90}Sr, ^{130}I, ^{85}Kr, ^{134}Cs, ^{135}Cs, ^{144}Ce, ^{129}I, ^{3}H, ^{241}Am, ^{99}Tc. (Chibowski, 2000, p.249),

That is fission ^{235}U or ^{239}Pu does not always produce the same fragments, as shown some of them below.

$$^{1}n +^{235} U \to^{141} Ba +^{92} Kr + 3^{1}n + Q$$

$$^{1}n +^{235} U \to^{144} Ba +^{89} Kr + 3^{1}n + Q$$

$$^{1}n +^{235} U \to^{140} Xe +^{94} Sr + 2^{1}n + Q$$

$$^{1}n +^{235} U \to^{136} Xe +^{88} Sr + 12^{1}n + Q$$

$$^{1}n +^{235} U \to^{132} Sn +^{101} Mo + 3^{1}n + Qetc$$

Neutrons and gamma rays escape from the fissioning material and bombard surrounding elements forming some radioactive isotopes. The fission products which result from the explosion constitute a very complex mixture. This mixture may consists about 170 different types of fission derbis which are isotopic forms of 35 different chemical elements. This fission derbis initially highly radioactive decay over a period of time by emittion of beta particles and gamma rays.

Later, means where found of using the fusion process to secure weapons of higher yield than where practical from the purely fission designs. A fusion process is the uniting, fusing of very light elements that great quantities of energy are given off in the process. To initiate a fusion process tremendous heat is required. The term " thermonuclear" results from the fact that such weapons use heat to maintain the nuclear reaction. For clarity let us see the action of a hydrogen bomb based on thermonuclear reaction between deuterium and tritium.

$$^{2}H + ^{3}H \rightarrow ^{4}He + ^{1}n$$

This reaction proceeds with the release of enormous energy but for this reaction to be triggered, an excessively temperature is needed as "match" to induce fusion. Such temperature can be provided by an atomic bomb. Therefore, a high hydrogen bomb, which contains a mixture of deuterium and tritium, is detonated by an atomic plutonium bomb. In the thermonuclear explosion of hydrogen bomb actually atomic bomb is exploded first, followed by thermal nuclear or fusion reaction by releasing an enormous amount of neutron, some of extremely high energies, and these can induce radioactivity in materials with which they can in contact. Naturally, two, in a thermonuclear weapons the fission portion of reaction forms a radioactive debris in the same manner as purely fission weapons. In both cases these cloud of radioactive materials produced by nuclear bomb (radioactive fallout) sweeps a path across the entire earth, contaminating water bodies and the soil.

Nuclear Power Production

Modern nuclear power plant use nuclear reactor with a sustained but controlled fission chain reaction, to generate tremendous amount of heat which is used to generate steam and produce electricity. Typical nuclear reactor has a core containing the uranium (or other fissionable elements, such as plutonium), a moderator (use to slow neutron down) and control rods of some substances that readily absorbs neutrons, such as cadmium or boron (Mckinney & Schoch, 2003, p.177).

Most releases of radionuclides from the nuclear fuel cycle occur at the uranium mining and milling and fuel processing stages, whereas in case fuel fabrications and power production stages the release of radionuclides to the environment is much small. Uranium ore is extracted using open cast and underground mining techniques as a result large quantities of tailings containing residual uranium and all of other members of decay chains have been deposited in close proximity to the mining and milling operations. Release of radionuclides in fuel production stages of fuel cycle are low, since uranium has been separated from other decay chain members before the stage of the operation. As usually, radioactive fall out due to nuclear power production contaminate the soil.

1.3.4 Chemical Fertilizers

As stated above soil is naturally radioactive due its mineral content by nature natural radioactive soil vary considerably from one type of soil to another and the radioactive elements are concentrated mostly in the surface layers of soil as their migration down ward is limited and depend on many chemical and physical conditions of the soil system. So that soil situated at high land area is in general poor as constant loss by leaching of certain elements which are essential to the plants such as nitrogen, phosphorus, potassium and calcium. Then in order to overcome these deficiencies of plot lands, the experience is replacing nutrients in soil and consequently supplying substances in order to rich high productivity by the application of chemical fertilizers, which are primarily composed of nitrogen, phosphorous and potassium whose composition formula depends on the need of soil and culture. Soil of plot land becomes contaminated radioactively due to the presence of radionuclides in phosphate fertilizers that is an extensive use of fertilizer reach in phosphate for agriculture purposes is of the source of radioactivity in the soil (Becegato et al., 2008) . Finally, extraction industries, industries working mineral materials in natural radioactivity elements such as fertilizer factories and various economic sectors in which natural or artificially radioactive elements are used provides radioactive fallout to the soil.

1.4 Pathway and Application of Radionuclides

1.4.1 The Pathway of Radionuclides in The Soil

We have seen different types of the source of radionuclides of soil, such as terrestrial natural radionuclides and radionuclide fallout from the environment to the surface of the earth. These radionuclides can transfer to the organisms of human being body through the food chain processes.

The condensed radionuclides in the atmosphere have been observed as they fall out down to the ground of the earth and distributed over the surface of the soil and on the external parts of the plants, above the surface of the earth. Those radionuclides on the external parts of plants may transfer by the action of convectional air current.

The distribution of radionuclides in the soil may not be uniform under the influence of different considerable factors, for that matter some radionuclides have restricted mobility within some soils, hence, the mobility of radionuclides the soil may depend on the properties of the soil and on the chemistry of radionuclides. That is the location of radioisotopes within the earth depends none their nuclear properties, but on their chemistry. All the three primordial radionuclides (Uranium-238, Thorium- 232 and Potassium- 40) are easily oxidized metals and they are relatively low in density and thus occur in the crust of the earth rather than in its dense mantle and metallic core (Arnold, 1959, p.85). Also their concentration difference, wetting and degree of solubility of radionuclides, temperature, pH, organic matter content, carbonate and texture of the soil are factors for the restricted mobility of the radioisotopes in the soil.

Those gaseous and dissolved radionuclides diffuse as a result of concentration gradient for the part of the soil with high concentration to the part of the soil with low concentration. In such way a gas, such as a radon diffuses out of the soil into the air. The soil moisture accelerate the rate of diffuse of radionuclides. Then, radionuclides may diffuse upward to the surface of the soil or down to the deeper part of the soil depending on the direction of concentration difference. Some radionuclides infiltrate 'leach' down in the soil. If wetting and solubility affect the mobility of radionuclides, then it is reliable to the fluctuate according to the seasonal, or even daily, variation in the rainfall and evaporation. These temporal and special change in the soil wetting can lead to significant fluctuation in oxidation-reduction potential, hence, the separation of elements which are redox-sensitive, including iodine, technetium and selenium. The activity of $^{226}\mathrm{Ra}$, $^{40}\mathrm{K}$ decreases with increased carbonate content, also an increased in oxidation state increases the mobility of U (VI), because under most oxidizing conditions U (VI) complexes are more stable than U (IV) and U (V) (Navas et al., 2002, p.635).

The pH- range of the soil indicate that the acidity, alkalinity and neutrality of the soil. Since the pH-range of the soil decrease with increase of organic matter contents of the soil and it was explained or known experimentally that, as pH of the soil increases and organic matter decrease down in the soil profile, Uranium-238 may form complexion, such as stable uranyl-carbonate and organo-oxide activity of Uranium-238 where decreased with the increase of sand content of the soil and increase with the increase of the clay content of the soil (Navas et al., 2002, p.635)., which implies that the distribution of radionuclides also depends upon the soil texture or type of the soil. Fine texture could hold more exchangeable elements (Tsidal et al., 2000, p.242).

In general, the more acid rocks, such as granites, are more radioactive than the alkaline basalt, limestones show specially low radiation levels (Arnold, 1959, p 85). These set of different factors leads to the substantial variations in the distribution of radionuclides in the soil [Shaw, 06]. At the end of the pathway, radionuclides are assimilated by plants with other necessary element for the growth of the plants, which were in the root zone. Such radionuclides absorbed by plants may be eaten by cow and some small fraction the radionuclides going to the milk and then a person drinks the milk, some small fraction of the radionuclides will go to his body. In such way neither uranium-238 nor thorium-232 enters appreciably into the metabolic process of living organisms, and neither do most of their descendants. The radiation that man receives from these elements is thus largely external. However, radium-226 and lead-210, of uranium series and radium-228, of thorium series, can be taken up by plants and travel eventually to man. In human body they concentrate in bone and thus by themselves contribute half as as much radiation to the skeleton as do all their relatives put together. Potassium also enters into biological process and distribute itself through the soft tissues. It provides the bulk of natural radiation that originate within the body.

As mentioned earlier more than 90 different radioisotopes due to nuclear detonation were identified. Among the fission process, too have required at the center of scientific and public concern: Strontium-90 and Cesium-137. Both are produced in substantial quantities by nuclear fission, both have relatively long half lives (about 25 and 30 years respectively) and both become engaged in the metabolism of human body. Neither element is a normal constituent of biological process but the chemistry of strontium resembles that of calcium. Like calcium it concentrates in the bone, where its radioactivity give rise to leukemia and bone tumors. Strontium-89, short lived fission product follows the same metabolic pathway. Cesium on the other hand, somewhat resembles potassium and its radioisotope concentrates in the soft tissues of the body with particular hazard to the genes (Arnold, 1959, p.89). To other isotopes, iodine-131 and barium-140 have attracted attention. Barium, like strontium, concentrates in the skeleton; iodine, in the thyroid gland. Because they have short half lives, radioiodine and radiobarium have been near zero levels since early this year.

The above described pathway of radionuclides of the soil until they taken up the organism of human being are generally taken (transported by atmosphere, water of the earth and biosphere and this pathway represented by the structure of a general radionuclide migration model through the soil- to plants system shown below).

The numbered processes are stated in words as follows:

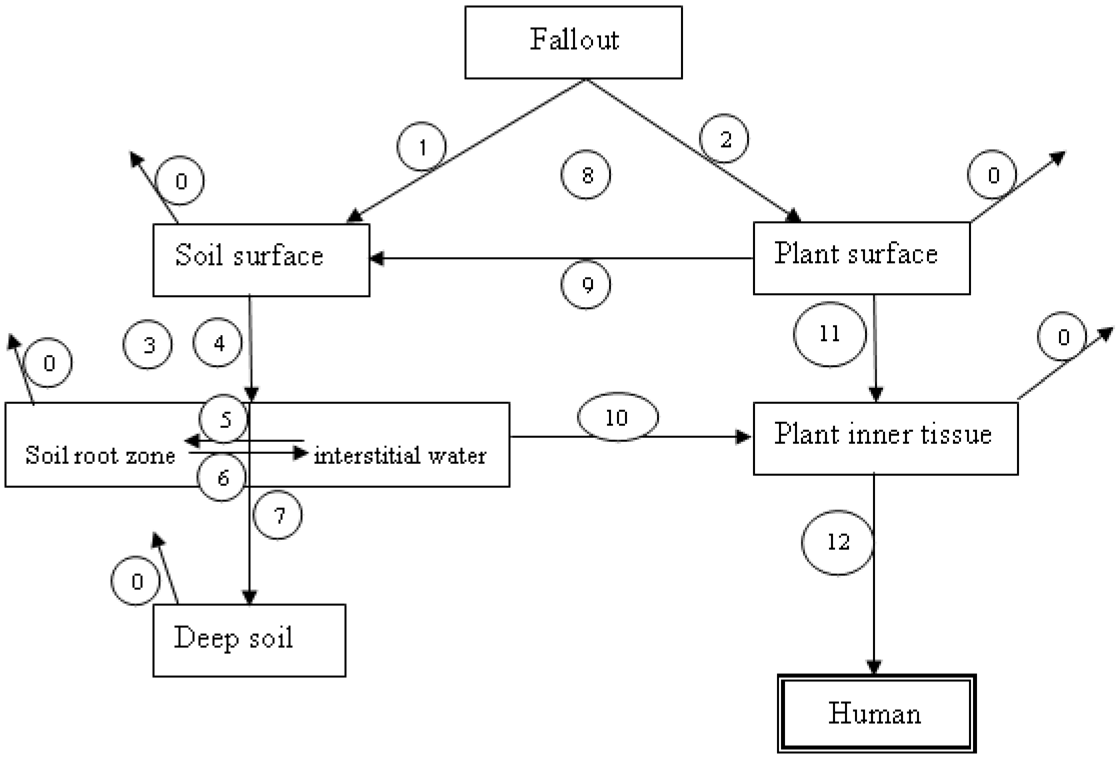

Figure 1.3: Structure of a general transfer model of radionuclide [Kabai, 04,p.201]

0- radioactive decay
1-direct deposition on soil surface
2- direct deposition on plant surface
3- advection
4- diffusion
5- absorption
6- desorption
7- leaching to deep soil
8- resuspension
9- weather process from plant surface to soil surface
10- root uptake
11- translocation to plant inner tissue
12- ingestion by human

1.4.2 Applications of Radioactive Elements

The detection of the number of radioactive atoms present in a sample of material either when they are natural origin or artificially produced has long been recognized as a powerful method of investigations. G. Hevesy in 1912 was the first to realize the minute amounts (about 10^{-17}gram) of radioactive isotopes, the *radio-traces* ,may be used as indicators to follow the path of atoms and molecules in chemical, biological and environmental systems, thus providing an extremely powerful method of investigation. Since then an enormous amount of work based on this technique has appeared on the scientific literature.

Another extremely useful field of application is that of nuclear activation analysis, which through detection of the activity of the products of irradiation of a sample of material with a beam of particles allows, in applied research, the detection of the presence of minute amounts of a given elements, and in nuclear physics research has provided more information about nuclear reaction mechanisms and nuclear structure than any other experimental technique. These studies include the identification of natural elements previously unknown such as technetium or polonium, radon and radium and of many transuranic elements up to the element with charge Z= 111.

An extremely important use of radioactivity is the estimate of the age of the earth and of the age of man made artifacts. In 1929 Rutherford realized that by an accurate measurement of the amount of a radiogenic isotope (produced in the radioactive decay of a parent) contained in a rock and of the remaining amount of a very long half-life parent decaying to this isotope, or of the relative amount of radioactive isotopes such as uranium-238 and uranium-235 one may obtain an estimate of the age of the rocks. From the relative amount of the two uranium isotopes and of lead-206 and lead-207 which are final product of the radioactive series of decays they originate, he estimated the age of samples of br ö ggerite, a mineral of uranium, to be about $4X10^9$ y, an age much younger than that of about $7X10^{12}$y estimated by Jeans for the sun, on the bases of astrophysical considerations. Jean's estimate was incorrect as we now know, while that of Rutherford although based on rather inaccurate data, is very near to the present estimate of the age of the earth, which is about $4.5X10^9$y (Hodgson, 1971, p.35).

1.4.3 Application of Soil Radionuclides

Natural radionuclides have so many practical application for the human being every day activities associated with economic sector and development of science and technology. The practical applications of radionuclides have been developed by studying the effects of interaction of ionizing radiation with matter. As stated in the previous discussion, there are two types of effect of interaction of ionizing radiation with matter, mainly effect of material body on ionizing radiation and effect of ionizing radiation on matter. Rutherford investigated that the nucleus of atom as positively charged and contain other sub particles. This was analyzed by scattering of alpha particles by the action of the nucleus of the atom. Similarly the effect of matter on radiation used for medical diagnosis, material testing, based on the evaluation of the impact of some material on ionizing radiation, that is by evaluating the radiation attenuation or scattering. While the effect of ionizing radiation on matter has application, such as, radiation therapy (medical treatment), production of mutation in plants, sterilization of foods and insects,sterilization of medical products and food preservation. Also nuclear power plant production is one of the main applications of radionuclides.

Further nuclear radiation and radioisotopes are proving very helpful in agriculture. To the world today, these tools are unlocking the secrets of many agricultural problems, which never have been possible by conventional means. Till now radioisotope and radiation are being more widely used in the field of agriculture than any other field of science and their application is leading us to the solution of agricultural problems in a shorter time and more precisely. Radioisotope and radiation gives us the opportunity to clear events that once were mysterious in the growth and nutrition of plants and evolution of new varieties by creating genetic variability. In addition to this, radioactive tracers and radiation sources have become indispensable to all the agricultural research problems. In agriculture, radiation and radioisotopes are also used in the nutritional studies of trace elements, mechanism of photosynthesis, plant protection including action of insecticides, metabolism in plant, uptake of fertilizers, ions mobility in soils and plants and food preservation. Thus, radioisotopes and radiation have contributed tremendously to fulfil the need of mankind such as food and agriculture, health and medicine, energy production, environmental protection etc.

Primarily radionuclides have provided an invaluable tool for investigating the availability of plant nutrients under field conditions. While a number of different isotopes have been used, the economic importance of phosphate fertilizer and the relative ease of handling P^{32} have resulted in particular attention being paid to phosphorus. Radioisotope of phosphorus as a tracer has a wide application in the field of plant physiology and soil chemistry. In the measurement of phosphorus uptake by plants in the soil could be determined by the difference. The recovery of phosphate fertilizer by a crop was determined by difference of the uptake of phosphorus by crops grown with and without fertilizer. The extra phosphorus in the fertilized crop was taken as the quantity coming from the fertilizer. This method assumed that fertilized and unfertilized crops take up the same amount of soil phosphorus, but tracer experiments indicate that this is often far from being the case. By using radioactive phosphorus research workers have succeeded to distinguish between soil phosphorus and the fertilizer phosphorus,taken by the plants (Alam et al., 2001)

Nuclear Power Plant

Nuclear power plant is a source of power supplied by inducing electrical energy for broad spectrum purposes. This civilian nuclear power is much safer (interms of human life lost) and less damaging of environment than other power plants, such as fossil fuel burning power plant and hydroelectric power plant (Mckinney & Schoch, 2003, p.183). Furthermore, coal-fired power plant disrupts ecosystems, and potentially change the global climate by producing carbon-base green house gases, such as carbondioxide and it spew particulates, sulfur oxide, similar harmful substances into the environment. Hydroelectric power plant although apparently clean and natural, cause untold environmental havoc by flooding up stream. The disruption the cause to natural water flow can be encourage the proliferation of disease-bearing organisms, and a large dam failure could conceivably kill several people and causes tremendous property damage. unlike the mentioned power plants, civilian nuclear power is to deliver energy from water about hundred times as great as energy available from the same amount of fuel oil. Deuterium is one of the source of material for production of nuclear power, which is heavy hydrogen found in heavy water. Deuterium occurs to the extent of 0.014% in natural (ordinary) water, and on the bases of deuterium content, the potential energy available from a gram of water is about 100 times as great as the energy available from a gram of fuel oil. Consequently, the worlds oceans represent an enormous potential energy supply [Mark, 57, P. 46]. MEDICINE: Perhaps the most important use of radioactive material is in medicine: Radiopharmaceutical, therapeutic drugs that contain radioactive materials are important in the diagnosis and treatment of many diseases. They can be injected to the body, inhaled, or taken orally as medicine or to enable imaging of internal organs and body processes. Many people around the world have benefited from diagnosis and therapeutic qualities of radioactive materials.

There are many applications of nuclear technology in medical field, ranging from diagnosis, to treatment, to diseases management. Many of this use radionuclides produce from either reactor or cyclotrons. Examples include, thyroid studies, brain studies, tumor studies, brain imaging, etc.

Diagnosis techniques

There are two distinct methods used in diagnostics. The first is to use the isotopes as an *in vivo* tracer. Here, a carefully chosen a radiopharmaceutical is administered to a patient through inhalation, injection or ingestion, to trace a specific physiological phenomenon in the living body. Detection is accomplished with special detection such as a gamma camera placed outside the body. The radiopharmaceutical can be selected to seek out only desired tissues or organs. There are hundreds of radiopharmaceuticals used in this way. The second method is to use an *in vitro* techniques for example, blood can be taken from the body, and studied, outside the living body using nuclear methods to assess exposure to infection by evaluating antibodies. It can also be used to provide detection of cancer (tumors) by studying some too dozen tumor markers.

Treatment Of Diseases

Radiation is widely used for the treatment of diseases such as cancer. The radiation is used to destroy the cancerous cells. A typical radionuclide used for this is cobalt-60 in addition to teletherapy where the radiation has no physical contact with the tumor, the radiation source may be placed immediate contact with the tumor. This form of treatment is called brachytherapy.

Disease Management

In addition to the sterilization of medical equipment, nuclear medicine is also being use to reduce pain. Radiotherapy is administrated to patients palliate the pain, thus replacing pain killing drugs, which eventually lose their effectiveness.

Industry

Industries around the world use radioactive materials in variety ways to improve productivity materials in a variety ways to improve productivity, safety, and obtain information that could not be obtained in other ways.

Radioactive materials are used in industrial radiology, civil engineering, material analysis, measuring devices, process control in factories, oil and mineral exploration, and checking oil and gas pipelines for leaks and weakness. This uses directly or indirectly influence our every day life. For example, measuring device containing radioactive materials are used in tasks ranging from testing of moisture content of the soil during road construction, to measuring the thickness of paper and plastics during manufacturing, to checking the heights of fluid when filling bottles in factories. Radioactive materials are even used in devices designed to detect explosives.

Sterilization

Sterilization is one of the most beneficial use of radiation, syringes, dressings, surgical gloves and instruments, and heating valves can be sterilized after packing by using radiation. Radiation sterilization can be where more traditional methods, such as heat treatment, cannot be used, such as in the sterilization of powders and ointments and in biological preparation like tissue grafts. For gamma rays from sources such as cobalt-60 and cesium-137 are commonly used to irradiate health care and other consumer products. The prevention of infection through this sterilization technique components the basic goal of medicine.

Food Irradiation

The use of gamma rays and electron beams in irradiation foods to control disease causing microorganisms and to extend shelf life of food products is growing through out the world.

Despite the invaluable utilization of radioactive elements for the improvement of human life discussed above, people are usually thought about the hazardous impact of radioactive elements. To give informative note I shall also discuss the disadvantages of radioactive elements below to make the thought complete.

Radionuclides were used for the production of nuclear bombs, which causes worst disaster. In addition to this well known worst effect, radionuclides have other drawback, as a result of the interaction of radiation emitted from them with matter. The significance properties of various radiation in their relative danger as they interact with living body, either inside or outside the body. For example, a substance which emits alpha particles does not constitute a hazard outside the human body. Since alpha particles are not energetic enough to penetrate the layer of dead skin and reach the living skin. A substance which emits beta particles can cause skin burns and irradiate the tissue to a depth of a few tenths of an inch. This is not likely to cause damage to internal organs. Radioactive radionuclides which emit gamma rays, however, can pass through the body and therefore can irradiate the entire body even though such gamma emitting elements are keep outside the body.

Consequently, it is convenient thing of radiation as dependent upon whether the radioactive material is outside of the body or inside of the body. In general, the danger is greater when the radionuclides gets inside the body.

Dangerous long lived radionuclides, such as cesium-137, strontium-90 and plutonium might be taken by human being through the food chain process. The ionization radiation which emit from such radionuclides changes the genetic constitution by chromosome abberations and point mutation. The chromosome abberations consists of loses and additions of whole chromosomes or chromosome parts, or alterations, called structural change in the alignment of chromosome parts. Structural changes are caused by breakage of one or more chromosomes at two or more points, followed by the junction of the fragments at their broken ends, so as form a new arrangement; that is a new linear sequence of their component hereditary particles or genes.

The changes, when occurring in genetic material of the germ cells, reach expression through reduction in the number of functional germ cells (infertility), increased mortality of zygotes of the first and subsequent generations (dominantletnals and deterimentals), and in general, the alternation of one or more herediatry characteristics, that thereafter are transmitted in their new form (mutation) [Muller, p.1065]. All this impacts of the ionizing radiation of radionuclides on living body governed by duration and penetration power of the radiations. Moreover, studies showed that the ionization of radiations on living system would curtail the life of the organism.

Chapter 2

Instruments and Methods

2.1 Geiger Muller Counter

Geiger-Muller counter is a type of gas filled counter which is originally announced by Geiger and Muller in 1928, represented a major steps forward in detection and counting of individual radioactive events (ANSI, 1969, p.6).

There are at least three different kinds of Geiger Muller counters 'self quenched' organic, the 'self quenched halogen' and the resistor quenched halogen. The first two method have filling mixtures such that after a discharge is initially terminated, secondary discharge are presented by the action of quenching agent in the gas mixture. The self quenched counters exhibit useful plateau characteristics without use of a series anode resister. The register quenched halogen counter is self quenching just above the Geiger-Muller threshold (the voltage at which nearly all pulse are of substantially the same amplitude) but as the voltage increases a transition takes place. Above the transition voltage the counter is no longer self quenched and series anode resister or external quench circuit is needed to prevent secondary discharges.

2.1.1 Quenching

Geiger Muller counter can be build in various geometry, the most common are with cylindrical envelop which is called cylindrical GM counter. The mechanism of discharge in cylindrical GM counters; when ionizing events occur in the counter forming an electron somewhere within the volume of the counter. The cylinder is kept at negative potential with respect to the potential of central wire. The electric field lines within the volume of the counter are directed as shown in the figure 2.1.

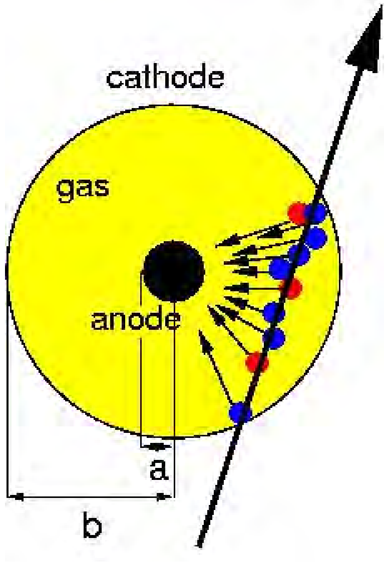

Figure 2.1: The electric field lines within the volume of the counter are directed radially

The field lines are crowded in the vicinity of the wire (anode) and thus the field strength is greatest near it. The electron formed inside by ionizing events moves towards the central wire. The field near the central wire being stronger, the electron acquires greater energy as it reaches near and near the wire. Thus, if it has energy at least equal the ionization potential of the gas, the electron ionizes the gas atom. Consequently, one more electron and a positive ion are produced; the electron so produced and previous electron repeat the same phenomena. Thus, avalanche process may begin. The avalanche is over in a very short time owing to the high mobility of electrons, which collect on the wire in a fraction of micro second. Positive ions being slow in motion form an ion sheath all around the wire.

After development of the space charge, the space charge of positive ion-sheath depresses the electric field at the anode so as to make any further multiplication by collision impossible. A second particle entering the counter at this time will not produce a pulse. When positive ion sheath reaches the cylinder, secondary electrons may be created by positive ion bombardment. The positive ion approaching the surface of the metal draws an electron out of the potential barrier at the surface. This electron then neutralizes the positive ion. The positive ion as it returns to its ground state emits characteristics recombination radiation in the form of one or more special lines. The radiation causes photoelectric effect and if 10^4 photons strike the cylinder, it is probable that one secondary electron is formed and avalanche process therefore repeats (Singh & Chauhan, Singh & Chauhan)

As mentioned above the positive ion sheath remain in the vicinity of the anode for a short period τ_d this period lasts for 10^{-4} to 10^{-3} second. During this time the counter remains insensitive to ionizing particles. The time is called dead time of the counter. In other words as the space charge moves toward the cathode the field increases again. It becomes sufficiently large so that the entering particle may produce a small pulse after a certain time τ_d, the dead time following the passage the former adjacent particle. It reaches the size of original discharge at time $\tau_d + \tau_R$, τ_R being called the recovery time.

During insensitivity (at dead time) of the GM, the GM stops counting particle entering into effective volume of the counter. Then, there is count loss in GM. However, the count loss in GM can be evaluate using two source method. Before applying this method of evaluating let us analyze using single source. Assuming that all pulse after a time τ are counted, the counting loss in the GM tube can be evaluated, if τ, for the dead time τ_d, with which it is often identified, for it is also called 'dead time'. If 'm' is the number of measured events per unit time, the total insensitivity time during this time is '$m\tau'$. If other loss are negligible, the loss of true event is

$$n - m = n(m\tau)$$

$$m = \frac{n}{1 + n\tau}$$

$$n = \frac{m}{1 - m\tau}$$

Where: n is the disintegration rate of the source

Now let us apply this knowledge from single source in the two source method by comparing the sum of measured activities of the two single source with the activity of the sum of the sources. Let n_1, n_2, n_{12} be true number of events per unit time for courses $1, 2$ and both sources respectively including background n_b, and $n_1, m_2 and m_{12}$ equal corresponding record counting rate.

$$n_1 + n_2 = n_{12} + n_b$$

$$\frac{m1}{1 - m_1\tau} + \frac{m_2}{1 - m_2\tau} = \frac{m_{12}}{1 - m_{12}\tau} + \frac{m_b}{1 - m_b\tau}$$

This leads to quadratic equation for τ. The development of the solution in the power of

$$x = m_1 + m_2 - m_{12} - m_b$$

$$\tau = \tau_1(1 + \frac{\tau_1}{2}(m_{12} - 3m_b))$$

Where

$$\tau_1 = \frac{x}{2(m_1 - m_b)(m_2 - m_b)}$$

The simplified relation is often used

$$\tau = \frac{x}{m_{12}^2 - m_1^2 - m_2^2}$$

((Bleuler & Goldsmith, 1959).)

As mentioned in mechanism of discharging of GM counter, every moving out sheath produces secondary electrons, the secondary electrons starts a new avalanches and the discharge will continue indefinitely. This discharge consists of a serious of pulses and spurts at close intervals. Hence, the quenching of discharge is essential. It may be done by having an external resistance or by filling a poly atomic gas or a halogen gas (Cl). The former is known as resistance (external) and the latter self (internal) quenching (removing of secondary electrons, pulses produced by photo electrons).

Resistance Quenching

Reducing high voltage applied to GM tube, for a fixed time after each pulse to a value that is too low support further gas multiplication then secondary avalanches cannot be formed and even free electrons are liberated as the cathode it cannot cause another GM discharge. Some electronic circuits which are called quenching circuit also use of a high resistance impulse forming circuit as shown below

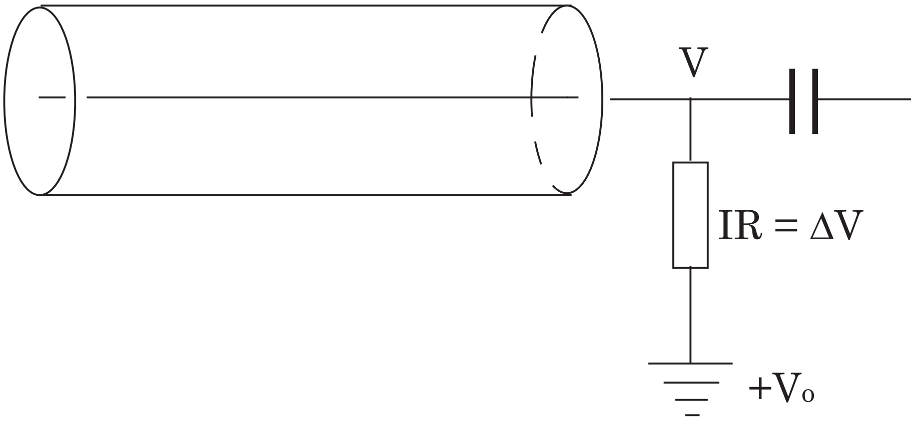

Figure 2.2: Quenching circuit

The voltage supplied to the tube from the source is

$$+V_0 = V + \Delta V$$

Where: ΔV is the voltage drop on the register and V is voltage given to the tube. Then, due to the flow of current through R the potential of central wire is decreased from V_0 to V or by $\Delta V = V_0 - V$. Earlier electric field produced in GM by V_0 at anode was strong to produce avalanche but now the voltage (potential) of anode reduced by dropping ΔV on the register R, then the corresponding field may not sufficiently strong to attract electrons produced at cathode due to excitation of argon atom (atom of gas of GM). The main criticality of external quenching is that its recovering time is too large, then during the long recovering time the counter falls to count ionizing particles which passes through it. And then external quenching counter is also called slow counter.

SELF (INTERNAL) QUENCHING

Self quenching, which is accomplished by adding a second component called quenching gas to the primary fill gas (argon). For 'self quenched' organic some polyatomic molecules added to the GM tube in addition the gas (argon) to quench the discharge themselves. This type counter have short recover of time and hence are called fast counter. Every polyatomic molecules has the same effect of quenching but they make the change in life and resolving time of counters. Generally, in counters $9cmHg$ of argon and $1cmHg$ of quenching gas (polyatomic molecules) is taken to make them self quenching. That is in counter at the time of filling some polyatomic molecules such as C_2H_5OH are also introduced at $10cmHg = 9cmHg$ of argon $+1cmHg$of C_2H_5OH total pressure. As ionize argon Ar^+ (by entering particle to active volume of the tube) moving to the cathode interacts with C_2H_5OH that is

$$Ar^+ + C_2H_5OH \rightarrow (C_2H_5 - OH)^+ + Ar$$

Instead of Ar^+ reaching to the cathode $(C_2H_5 - OH)^+$ reaches first to the cathode and neutralized with electron of the cathode

$$(C_2H_5 - OH)^+ + e^- \rightarrow (C_2H_5 - OH)$$

and in excited state, this also deexcited by emitting photons utilized in dissociation of $(C_2H_5 - OH)$ that is

$$(C_2H_5 - OH) \rightarrow C_2H_5 + OH$$

Because dissociation energy is smaller than energy needed to produce photo electrons means that the photons are absorbed by $C_2H_5 - OH$ to be dissociate and this effect knowns quenching of photon. That is photons are absorbed by quenching gas before photons arrive at the cathode. Therefore, the main function of polyatomic molecules in the avalanche is to absorb ultraviolet photons emitted by inert gas argon. When a polyatomic gas absorb ultraviolet photon and passes into excited electronic state and deexcited only by decomposition (dissociation) (Singh & Chauhan, Singh & Chauhan).

Polyatomic molecules numbers decreases in the counter after formation of each pulse, that is 'self quenched' organic counters have finite life time because at each pulse formation number of polyatomic molecules is decreasing in effective volume of the counter. Life time of counter is expressed in terms of particles (radiation) counted. Because each coming pulse consume a polyatomic molecule.

To avoid the problem of limited life time, some GM tube use halogens as quenching gas but such counter have no metal cathode because halogen families are chemically active in radiation. Due to this reason GM tube cathode should be made up of stainless-steel (which do not interact chemically). As observed in case of ionized polyatomic molecule, the ionized halogen quenching as such as $(Cl_2)^+$ is reaching the cathode neutralized by cathode electron and dissociated into $Cl +$ Cl atom that is, $Cl + Cl \rightarrow Cl_2$ which implies that quenching as Cl_2 or chlorine molecules is present always as the counter is used "self quenched" halogen counter has an infinite life time. Comparatively from the mentioned three counters “self quenched” halogen counter is preferable to perform the experiments of this thesis work. When one wants to perform an experiment using GM counter, he has to take into consideration the following facts about GM counter.

Background counting rate: Background counts means counts caused by radiation coming from sources other than to be measured. The background radiation have to be measured at a certain (specified) condition such that the measurement shall be made with counter totally enclosed in a nuclear radiation shelled. This shell shall consists of an outer layer of lead at least 50mm thick an inner liner of at least three millimeter of aluminium since lead is a substance with high atomic number, that means it has large photon absorption cross section to block the passage of the coming background photons and aluminium layer required to reflect the radiations emitted from the source placed inside the shielding. The background shall be specified as maximum of counts per unit time (ANSI, 1969, p.9). In order to determine the total pure count rate of ionizing radiation of the source required to measured.

Geiger Muller Counter Plateau

The application of GM counter requires only the operating conditions be established in which each pulse is registered by counting system (Knoll, 2000, p.208). The term GM counting plateau represents that the portion of counting-rate- versus voltage characteristics in which the counting rate is substantially independent of the applied voltage, and the minimum voltage is at which pulses are first registered by the counting is often called threshold voltage (starting voltage), where the transition between the rapid rise is of the curve and plateau is its knee.

In practice this operating point is normally chosen by recording a plateau curve from the system under conditions in which a radiation source generates at constant rate within the counter. That is one how to identify the geiger-muller regions, which the range applied voltage in which the charge collected per isolated counts independent of the charge liberated by initial ionizing event. A good GM counter has a plateau about 200 volts wide and has about one percent slope per hundred volts (Singru, 1974, p.43). Where the plateau slope is given by the next formula, for the total plateau length specified. The slope S is quoted in percent per hundred volts.

$$S = 100\left[\frac{(N_2 - N_1)}{\frac{(N_1+N_2)}{2}}\right]\left[\frac{1}{\frac{(V_2-V_1)}{100}}\right]$$

Where: V_1= Threshold voltage and V_2= the knee voltage (the voltage which corresponds to the end of the plateau) and N_1 and N_2 are counting rates at V_1 and V_2 respectively.

The continuous discharge process is potentially harmful if sustained for any length of time, and one should therefore immediately decrease the applied voltage when the end of the plateau is observed. In the interest of long operating life,on operating point is normally chosen that is only sufficiently far up the plateau to ensure that the flat region has been reached.

In real cases, the counting plateau always shows some finite slope, which indicates that response of GM counter to ionizing radiation depends on some factors, such as, on countered design, type and energy of ionizing radiation, geometric factors, temperature,high hysteresis and photosensitivity. For example, some regions near the end of the tube may have a lower than normal electric field strength and the discharge originating in this regions may be smaller than the normal.

Also, any pulse that occur during the recovery time (the minimum time from the start of a counted pulse to the instant a succeeding pulse can attain a specified percentage of the maximum amplitude of the counted pulse)will also be abnormally small. In all order to exclude this latter effect in measuring the plateau characteristic for each value of ionizing radiation flux the current and/or counting rate recorded at a stated operating voltage should be done at a count rate for which the product of dead-time and counting rate is less than 0.05[Ansi, 69, p. 8]. Or the counting rate should not exceed a few percent of the inverse of the recovery time (Knoll, 2000, p.209).

The plateau curve may also show a slight hysteresis effect, usually this is encountered when determining a count rate versus voltage characteristics. There may be a difference in the plateau obtained with an increasing voltage traverse and that with a decreasing voltage traverse. Hysteresis may also show up if a plateau is traversed and the voltage is then set at operating voltage. This effect is attributed to the charging of insulating surfaces in the counter, that is the electrical charge on the insulators are slow to equilibrate and can influence the electric field configuration inside the tube. To test for hysteresis the plateau shall be traversed in an ascending and then a descending direction with a total elapsed time or less than one hour (much shorter than the charge leakage time constant of the insulating surfaces) (ANSI, 1969, p.9).

The sources used will depend upon the applications for which the counter is designed to detect the intermediate range of gamma ray energies, one of the source shall be cobalt-60, and for, alpha, and X rays measurements any geometric factor could materially affect the evaluation of the counter response. These factors include angle of incidence of radiation, source shielding and columniation, tube shielding, air absorption and absorption due to radiation filters.

The photosensitivity of counter is indicated by a change in background counting rate on exposure of the counter to light. Since organic-quenched counters can exhibit sensitivity to light, they are usually made opaque. Halogen-quenched counters usually did not exhibit such sensitivity. Photosensitivity shall be tested by measuring the background rate first with the counter operating in the dark and then with the counter exposed to a stated illumination.

Geiger tube are often rated based on the by slope of the linear proportion of the plateau. Organic-quench tube tends to show the flattest plateau, with slopes of the ordinary 2-3 percent per hundred volt change in the applied voltage. Halogen-quenched tubes have plateau of greater typical slope but their longer useful life time can some time offset this disadvantage. Halogen tube can also be operated at lower applied voltage values (Knoll, 2000, p.210).The mechanism lead to universal low starting voltage strongly depend on the partial pressure of the halogen and $\frac{V}{P}$ value within the tube.

2.2 Semiconductor Detector

The development of semiconductor ionizing radiation detector during the last decade has completely revolutionized the field of nuclear radiation detectors. As analogy, that can be considered as ionization chambers where the gaseous medium is replaced by a semiconducting solid. In a semiconductor detector ionizing radiation produces ion pairs which are collected by electric field applied externally, and the detectors gives an electric pulse which is proportional to the energy of ionizing radiation. The semiconductor detectors have many definite advantages over the gas filled or scintillation detectors some of these advantages are:

a) smaller, compact and convenient size,
b) fast (few n seconds) rise time of output pulse,
c) linear response over wide energy range,
d) excellent energy resolutions
e) choice of sensitive depth, area and geometry

In the initial stages of development the detectors were designed for detection of heavy charged particles. Very soon development of semiconductor detectors for electrons and gamma rays followed (Singru, 1974, p.49).

Some properties of semiconductors and their use in construction semiconductor detector are written below in a summarized form. The semiconductor materials are crystal made of silicon and germanium atoms with four balance electrons. In such materials the electron status have a band structure. The lowest band is the valance band and the electrons whose state lay in this band cannot move under an applied electric field. Above the valance band there is an energy gap between electron states. This gap, which is an insulator about 5ev, is about 1.12ev for silicon, and about 0.67ev for germanium at normal temperature (=300K) and slightly increases with decrease of temperature. Above the energy gap there is conduction band as if the electron is raised to the conduction band it may move freely under the action of external electric field. At low temperature the electrons lay in the valance band and the semiconductor act as an insulator. With increasing temperature, the electrons start to fill the conduction band living behind holes in the valance band. Both electrons in the conduction band and holes in the valance band may now move under an external electric field and the semiconductor starts to behave like a conductor (the hole migrate when they are filled by electrons of neighboring atoms.

If a pure silicon or germanium crystal is doped with a) trivalent atoms like Gallium, Thallium or Indium or, alternatively, b) pentavalent atoms like boron, arsenic, phosphorous and antimony, then in case they are insufficient electrons to fill the valance band and thus a number of holes are created in the valance band even at low temperature. The dopant also perturbs the band structure creating an additional state in the gap region, near to the valance band. The majority of the charge carriers are holes which have a positive charge and thus the semiconductor is called a p-type semiconductor. In case b the opposite will occur. There will be an excess of electrons to the conduction band the majority of the charge carries are the electrons and the semiconductors become n-type semiconductor.

At n-p junction there is transformation of holes from the p-type to the n-type semiconductor and of electrons from the n-type to the p-type semiconductor, which continues until the difference of the potential which is gradually established between the two semiconductors stops any further transformation. When this occurs, a region without free charges is established around the contact surface where, however, due to the contact potential, an electric field acts. This region is known as the depletion region of the junctions. The depletion region is similar to the active volume of a gas detector. Once a particle penetrates this region ionize its atoms, the electrons and the holes may be collected by the anode and the cathode, and the corresponding signal may be collected by the anode and the cathode, and the corresponding signal may be analyzed providing information on the energy lost by the particles (radiation). However, the depth to depletion region is very small of the order of micrometer, which is not sufficient to stop energetic particles. Its depth can be increased greatly by applying an external reverse-bias voltage (that is, a negative voltage to the p-side and positive voltage to the n-sides). The depth of depletion layer is proportional to the square root of the product of resistivity of the semiconductor and the applied voltage and, using high resistivity silicon, depletion depths of up to 5mm may be obtained. When the junction is used as a detector, its cross sectional area must be perpendicular or nearly so to the direction of the particle and there is an inactive depth of material on both sides of the junction which is very inconvenient since it may without providing any useful information (Hodgson, 1971).

This may be eliminated using semiconductor surface barrier (SSB) detectors which are obtained by making a junction between an n- or p- type semiconductor with a suitable metal in the form of a very thin surface layer. The most frequently use materials are gold with n-silicon and aluminium with p-silicon. What happen in these cases is very smaller to what we have discussed above, but there is the great advantages that there is no longer inactive surface layer which may be reduced the energy of the particle. Semiconductor surface barrier silicon detector are extensively used in low energy experiments.

The major limitation of the simple junction and surface barrier detectors is maximum depletion depths or active volume that can be created. Using silicon or germanium of normal semiconductor purity, depletion depths beyond two or three millimeter are difficult to achieve despite applying, bias voltage that are near the breaking down level. Much greater thickness are required for the detectors intended for gamma ray spectroscopy. Equation for the thickness of depletion region is given by:

$$\mathrm{d} = \sqrt{\frac{2\epsilon \mathrm{V}}{e\mathrm{N}}}$$

where: V is the reverse bias voltage and N is the net impurity concentration in the bulk semiconductor material (ϵ is the dielectric constant and small e is electric charge).

At a given applied voltage, greater depletion depths can only be achieved by lowering the value of N through further reduction in the net impurity concentration (Knoll, 2000, p.405). Based on this, there are two general approaches that can be taken to establish large depletion depths. The first is to seek further refining techniques capable of reducing the impurity concentration to approximately $10^{10}\frac{atoms}{cm^3}$. At this impurity level in germanium, the above equation predicted that a depletion depth of 10mm can be reached using a reverse bias voltage are less than 1000V. However, such a low impurity concentration corresponds to levels that are less than one part in 10^{12}, a virtually unprecedent degree of material purity. Technique have been developed to achieve this goal in germanium, but no in silicon (Knoll, 2000, p.405).

Detectors that are manufactured from this ultra pure germanium are usually called intrinsic germanium or high purity germanium (HPGe) detectors, and they have become available with depletion depths of several centimeters. The second approach to reduce a net impurity concentration is to create at compensated material in which the residual impurities are balanced by an equal concentration of dopant atoms of the opposite type. Lithium ion drifting has been applied in those silicon and germanium crystals to compensate the material after the crystal has been grown. Residual acceptor impurities are exactly balanced over a thickness of up to 2cm by the addition of interstitial lithium donor atoms. The resulting compensating material has many of the properties of intrinsic or pure material. Even if the compensation is not perfect, the residual net impurity level may be low enough so that the drifted region can easily be depleted over its entire thickness.

Germanium detectors produced by the lithium drifting process are given the designation GE(Li). The important performance characteristics such as detection efficiency and energy resolution are essential identical for Ge(Li) and HPGe detectors of the same size (Knoll, 2000, p.406).

2.2.1 Germanium Detector Operational Characteristics

A. Detector cryostat and dewar

The operation of germanium detectors at room temperature is impossible, because as a result of its small band gap (0.7eV) there will be thermal induced leakage current. These detectors must be cooled in order to reduce the thermal generation of charge carriers (thus reverse leakage current) to an acceptable level. Otherwise, leakage current induce noise destroys the energy resolution of the detector. Liquid nitrogen which has a temperature of 77^0K, is the common cooling medium for such detectors. The detector is mounted in a vacuum chamber which is attached to or inserted into liquid nitrogen dewar. The sensitive detective surfaces must protect from moisture and other contaminants.

CRYOSTAT: Cryostat consists of vacuum chamber which houses the detector element sometimes with preamplifier plus a dewar (double wall vacuum-insulated vessel) of liquid nitrogen cryogen. The detector element is held in a place by a holder, which is essentially isolated from but thermally connected to a copper finger. The cool finger transfers heat from the detector assembly to the liquid nitrogen reservoir. The detector holder as well as the outer vacuum jacket or "end-cap" are thin to avoid attenuation of low energy photons. The rest constituents of cryostat which were not described above would be shown in the total figure of cryostat below.

Gama Ray Spectroscopy with Germanium Detector

The performance of germanium detector gamma ray spectrometry are usually specified in terms of energy resolution, peak shapes, efficiency and peak-to-compton ratio. The resolution of germanium detector are specified by manufacturers commonly in terms of the FWHM at 1.33MeV. The dependence of the energy resolution on the operating temperature of germanium crystal has been studied. It is seen that, while the energy resolution is not significantly altered in the temperature range of 90^0K – 130^0K, a series degradation occurs above 150^0K (Kapoor & Ramamurthy,

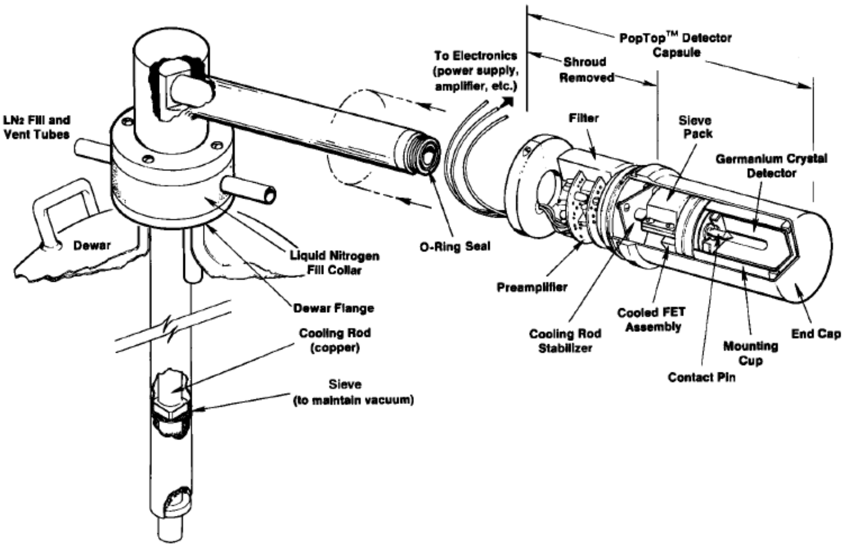

Figure 2.3: A Typical Cryostat System

1993, 133).

Energy resolution is the dominant characteristic of germanium detector. Small volume detector has a better overall energy resolution than the larger detector of the same due to the following two factors. Because the capacitance of small, detector is low, while the electronic noise of the system increases with detector capacitance and also, the effect carrier lose due to trapping are magnified in a large volume detectors with relatively large charge collection distances.

The peak shape obtainable from a detector is generally specified in terms of the $\frac{\mathrm{FWTM}}{\mathrm{FWHM}}$ ratio at 1.33Mev (here FWTM is full width at one tenth maximium) for gaussian peak, this ratio is 1.83 (Kapoor & Ramamurthy, 1993, p.133).

As stated above the small size available and lower atomic number, of Ge, results in a photoelectric cross section that is small by a factor of 10-20 compared with sodium iodide (Tl), combine to give photo peak efficiencies an order of magnitude lower in typical cases. As a direct result, the intrinsic peak efficiency of germanium detector is many factor smaller than sodium iodide (Tl) scintillator of equivalent active volume. The latter disadvantage somewhat off set by the superior energy resolution of germanium. Not only does good resolution help separate closely spaced peaks, it is also aids in the detection of weak sources of discrete energies when superimposed on the broad continuum. Detectors with equal efficiency will result in equal areas under the peak, but those with good energy resolution produce a narrow but tall peak. That may then rise above the statistical noise of the continuum (Knoll, 2000, p.427).

Compton scattering occurs when the full energy of an incident photon is not completely absorbed by the HPGe detector and thus exist the detector leaving only part of its energy to be counted. The partial energy peak appears in gamma ray spectrum as random event below the full energy peak in what is refer to as the compton continuum. The ratio of the full- energy peak to the compton continuum is called the peak-compton ($\frac{p}{c}$) ratio. In a standard HPGe detector, it is common to have a peak-to-compton ratio between 40:1 and 60:1 for the 1.33Mev peak of cobalt-60 larger detector can have a $\frac{p}{c}$ ratio nearly 100:1.

The compton continuum is a prominent part of germanium detector spectrum. Because the ratio of the compton to photoelectric cross section is much larger in Ge than NaI, a much greater fraction of all detected events lie with this continuum rather than under the photo peak.

2.3 Detection Limit

Detection limit is the smallest amount of concentration of radionuclides (or activity concentration) that can be detected in a certain arrangement of detectors. The factors contributing to the detection limit of a given arrangement are the relative position of the sample and degree of sensitivity of the detecting system.

There are some circumstances under which it is convenient to estimate the smallest signal that can be detected reliably in order to set "detection limit" for the counting system.

Some regulatory agencies required that certain minimum delectability, amount of activity be measurable while monitoring for possible presence of radioactivity contaminants.

The possible minimum detectable amount (limit of detectability) of radioactivity of radionuclides made depend on the sensitivity of the detecting system and on the statistical fluctuation to compare (distinguish) the radioactivity of the background of the environment of the source.

Therefore, it was difficult to get standard (harmonized) mechanism for the determination of the limit detectability (minimum detectable amount) of activity of radionuclides. However, several regulatory agencies collaborated to develop harmonized procedure (mechanism)to determine the limit detectability.

Some regulator agencies, such as, international organization for standardization (ISO) and international union of pure and applied chemistry (IUPAC), require that a certain minimum detection limit amount (MDA) of activity be measurable while monitoring for possible presence of radioactive contaminant. ISO and IUPAC, have been trying to develop a harmonized international chemical meteorological position on detection and quantification capabilities. After a serious effort of them the emphasis of ISO was placed on the basic meteorology, with special attention to calibration based on detection, decision and limits, and focused on radiation measurements with special attention to poission-normal distribution (Currie, 2004, p.146).

The harmonized IUPAC/ISO recommendation for detection decision and detection limits are based on the concept of hypothesis testing with default probabilities of 0.05 for false positives and false negatives and some general definitions are given that which would be realizable and mandatory that a full-defined measurement process exist, and remain in a state of statical control.

By defining relations, the basic concepts are given mathematical equations in the following three equations. In each case the parameters $\alpha and \beta$ and the probabilities of the errors where specified as first and second kinds (false positive and false negative); and rsd_Q, as the relative standard deviation at quantification limit. Detection decision (critical value), $(L_c$, α =0.05)

$$P_r[\hat{N} > L_c/N = 0] \leqslant \alpha$$

Detection limits (minimum detectable values) $(N_D, \beta = 0.05)$

$$P_r(\hat{N} \leqslant L_c/N = N_D = \beta$$

Quantification limit (minimum quantifiable value) (N_Q, rsd_Q = 0.10)

$$N_Q = K_Q\sigma_Q, K_Q = \frac{1}{rsd_Q}$$

Where $\hat{N}$ represents estimated (observed) value of the measurand N; and N_D, N_Q and σ_Q did not "true" parameter values, while σ_Q representing standard deviation of estimated value at quantification limit [Curie, 04, p. 146].

Specifically let say N_T be number of counts recorded with an unknown sample and N_β be the number of recorded counts when a blank sample is substituted to determine the background level, then the net counts resulting from the unknown

$$N_S = N_T - N_\beta$$

To make a decision whether the sample contains activity, N_s is compared with critical level (value) L_c. If $N_s < L_c$, the same sample does not contain activity and if $N_s > L_c$, then if it is assumed that some radioactivity is present. In the absence of statistical fluctuation and other instrumental variation, L_c could be set at zero, and any net positive counts could be interpreted as evidence of real activity. However, there will be many instances of positive N_s that will be observed even for sample with no activity. Therefore, one should choose the value of L_c that is high enough to minimize the likelihood of such false positives, while keeping it low to reduce the possibility of missing real activity when some is actually present (false negative). If the counts for long time ($N > 30$), then N_T and N_β follow gaussian distribution since

$$N_s = N_T - N_\beta$$

$$\sigma^2_{N_s} = \sigma^2_{N_T} + \sigma^2_{N_\beta}$$

No real activity present: If there is no actual activity in the sample, then to mean values of N_T and N_β are the same and the mean value of N_s is zero. Under this condition

$$\sigma_{N_T} = \sigma_{N_\beta}$$

$$\Rightarrow \sigma_{N_s} = \sqrt{2}\sigma N_\beta$$

Since no true activity present, then any positive indication will be false positive. This implies L_c wanted to set high enough to ensure that, under this conditions, the probability that particular measurement of N_s exceed L_c is acceptable small. From gaussian distribution having a value within the interval 1.645σ implies that the probability is 90

Since we are only concerned with positive deviation (negative $N_s \Rightarrow$ no source being present). There is a 95 percent probability that a random sample will lay below the mean pulse 1.645σ.

$$L_c = 1.645\sigma_{N_s} = 1.645\sqrt{2}\sigma_{N_\beta} \tag{2.1}$$

$$= 2.326\sigma_{N_\beta} \tag{2.2}$$

Ensures that false positive probability will be no higher than five percent. This choice has become a common bases for setting L_c (Knoll, 2000).

2.4 Soil Sampling and Preparation for counting

To detect the concentration of radionuclides or their radioactivities in the soil, there are mechanisms, such as, direct detection, scanning and sampling. A direct measurement is obtained by placing the detector near or against the surface or in the media being selected for the study and reading the radioactive level directly. Scanning is an evaluation technique performed by moving a portable radiation detection installment at a constant speed and distance above surface to semi-quantitatively detect elevated areas of radiation. Sampling is the process of collecting a portion of the farm (plot land) selected for studying purpose. These collected portion of the plot land is analyzed to determine the type of radionuclide present, the radionuclide concentration with their radioactivities. This subtopic discusses issues involved in collecting and preparing samples of soil for analyzes.

Comparing with direct measurement sample collection and analysis is less representative of true measurements of the task at a specific measurement location. This is due to the involvement of several steps of the procedure, such as, sample collection, sample preparation and radiochemical analyzes. However, direct measurement techniques with acceptable detection limits are not always available (MARSSIM, 2000, p.7-6).

SAMPLING: Concentration of the radionuclides may vary widely from different parts of a farm. The variation or heterogeneity , has to be reflected in the number of samples to be taken. Sampling positions have to be chosen with care. For monitoring exercises involving large areas, the area should be subdivided using grids and ideally the sampling points should be at a fixed locations with each grids or over the whole area along a zigzag (cross) path in equal and short space interval the sample have to be taken at specific (range) of the depth of the soil. Even the soil composition may vary greatly on a small area in certain depth of the soil, of course, some causes for the variation of radionuclides in soil were mentioned in the previous chapters of this paper.

Sampling is always a problem with soils as any agitation tends to fractionate mixtures according to particle size. The smaller particles tend to fall below the larger particles. The simplest way to overcome this difficulty is the cone and quartering technique. The total sample is formed into a symmetrical cone. The cone is then divided vertically into segments and alternate quarters are obtained, the remaining half being rejected. The process can be repeated successfully until the required sub-sample size is produced (Reeve & Barnes, 1994, p.155).

This is to provide and developing appropriate collection procedures of soil samples. That is sample collection procedures are concerned mainly with ensuring that a sample is representative of the sampled media, is large enough to provide sufficient soil to achieve the desired detection limit. Although the volume of soil collected should be specified in the sample collection procedure. Since, large volume of the soil are more representative than small volume of soil then large samples provide sufficient sample to ensure that required detection limits can be achieved and that sample re-analyzes can be done if there is a problem.

Tools for some sampling purpose may differ depending on different factors, such as, type of soil, sample depth, number of sample required and training of available personnel the selection of a sampling tool may also be base on the expect use of the results. For example, if a soil sample is collected to verify the depth profile used to develop the calibration for in-situ gamma spectrometry, it is important to preserve the soil core (MARSSIM, 2000, p.7-13) soil samples also recommended to be contained in polyethylene bottles with screw caps and wide mouth. Because these containers are fairly economical, provide easy access for adding or removing samples, and resist chemicals, breaking, and temperature extremes. Glass container are also acceptable but they are fragile and tend to break during trucking (displacement).

Proper sample preparation and preservation are essential of any radioactive sampling program. Precise records of sample collection and handling are necessary to ensure that data obtained from different locations or time frames are correctly compared. The appropriateness of sample preparation techniques is a function of the analysis to be performed.

Soil sample preparation includes removal of sticks, vegetation, rocks, exceeding about 0.6cm in diameter and foreign objects. If no volatile element are the any contaminants of concern the sample can be dried at approximately 110^0C for a minimum of 12 hours; volatile radionuclides (^{3}H, ^{99}Tc and iodides) must be separated from the sample drying in such a way to avoid loss of radionuclide of interest. Otherwise, the samples have to be dried out at room temperature isolating from other contaminants.

Dried samples are homogenized by mortar and pestle, jaw crusher, ball mill, parallel plate grinder, blender or a combination of this techniques, and sealed to obtain uniform sample. In addition, samples for chemical separations are also usually ashed in muffle-furnace to remove any remaining organic materials that may interfere with the procedures.

For total radionuclides concentration (activities) analysis extreme conditions have to be used to dissolve the soil such as dissolution in hydrogen fluoride, perchloric acid mixture or fusion with alkaline flux (example, sodium carbonate) and subsequent dissolution in dilute acid. For example, laboratory sample preparation in analysis of soil sample for beta emitting radionuclides all the above mentioned steps of sample preparations are necessary (cleaning, drying, grinding and dissolution). That is once the sample has been prepared, a small portion is dissolved, fused or leached to provide a clear solution containing the radionuclide of interest. The only way to ensure that the sample is solubilized completely to dissolve the sample. However, this can be an expensive and time consuming steps in analysis Gross beta measurements may be performed on material that has not been dissolved (MARSSIM, 2000, 7-21).

After dissolution, the sample is purified using a variety of chemical reactions to remove bulk chemical and radionuclides impurities. The objective is to provide chemically and radiologically pure samples for measurement. Example of purification techniques include precipitation-liquid extraction, ion-exchange, chromatography, distillation and electro deposition. Gross-beta measurements may be performed on material that has not been purified (MARSSIM, 2000, p,7-22).

Similarly, there is no special soil sample preparation required for counting samples using germanium detector or a sodium iodide detector beyond placing the sample in a known geometry for which the detector have been calibrated. After sample purified, it is prepared for counting. Beta or gamma emitting radionuclides are usually prepared for a specific type counter in specific geometry. Purification step is necessary to remove any interfering radionuclides.

Data reduction is usually the critical step in measuring photo emitting radionuclides and beta emitting radionuclides. There are often several hundred individual gamma ray energies detected within a single sample. Computer software is usually used to identify the picks, associated them with proper energy, associated the energy with one or more radionuclide, correct for the efficiency of the detector and geometry of the sample, and provide results in terms of the sample, and provides results in terms of concentrations with associated uncertainty. The data reduction for beta emitting radionuclides is less complicated than that of photon emitting radionuclides. Since the beta detectors report total beta activity, the calculation to determine the concentration for radionuclide of interest is straight forward.

The the procedure involved in the preparation of soil samples includes, cleaning, drying, grinding, sieving, resolution and purification.

Chapter 3

Apparatus and procedure of the experiment

3.1 Experimental setup

In this experiment, to count gross beta of the samples, the instruments used were:

- GM counting system with A.C. main cord
- GM detector (end window) stand or GM detector-source holder bench
- GM detector or GM stand (in PVC cylindrical encloser) with counting cable (Experiments with GM counter published by NUCEONIX SYSTEMS PRIVATE LIMITED)

The counting system was connected to the GM detector by MHV to UHF coaxial cable shown below. And the main cords from the counting system was connected to the AC power supply.

The GM detector was shielded by 40mm thick lead shielding cylindrical rings assembly with total of eight lead assembly parts. There is a hinged door in the bottom ring through which the sample can be loaded into the GM stand sample tray.

GM tube is a gas filled device which reacts to individual ionizing events, thus enabling them to be counted. Ionizing events are initiated by quanta or particles entering the tube either through the window or through the cathode and colliding with the gas molecules which yields free electrons and ions. As this set of electrons and ions collected by the corresponding electrodes induce electrical pulse height which amplified and counted by the counting system.

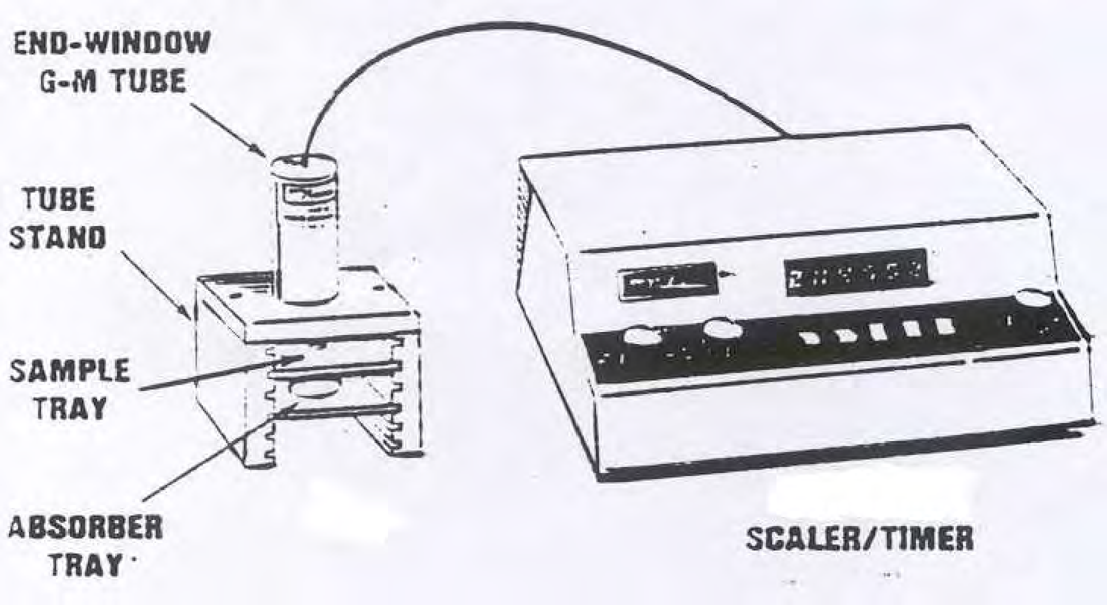

Figure 3.1: Schematic diagram of GM system

The other experimental equipment used was gamma spectrometry constituted of the following components

- Ge detector, cryostat, and preamplifier
- UPS and 4002D power supply
- amplifier model 2026
- MCA-with 8192 ADC range, 4696 memory and digital readout
- Detector bias supply-model 3106D
- computer system

[Canbera Germanium detector,2003,Cambera industries,inc. p.66 USA]

The mentioned components were connected as shown in the figure below.

The gamma spectrometry canbera system used consists of a HPGe detector (model GC 11519), and relative efficiency and resolution of the detector for 1332Kev (Co-60) are 1.9Kev, and 15 per cent respectively. The detector and preamplifier are placed inside a low-background lead shield and cooled by liquid nitrogen from the integral cryostat (model 7915-30) consists of pulse height analysis system to transform pulse, which are collected and stored by computer-based MCA. standard small Marinell beaker (8.5cm diameter) with 6cm height are used as sample containers. The signal processor contains high resolution spectroscopy amplifier with pile-up rejector and live time corrector, which allows the spectrum analysis nearly independent of a system count rate (Oczkowski, 2001, p.32).

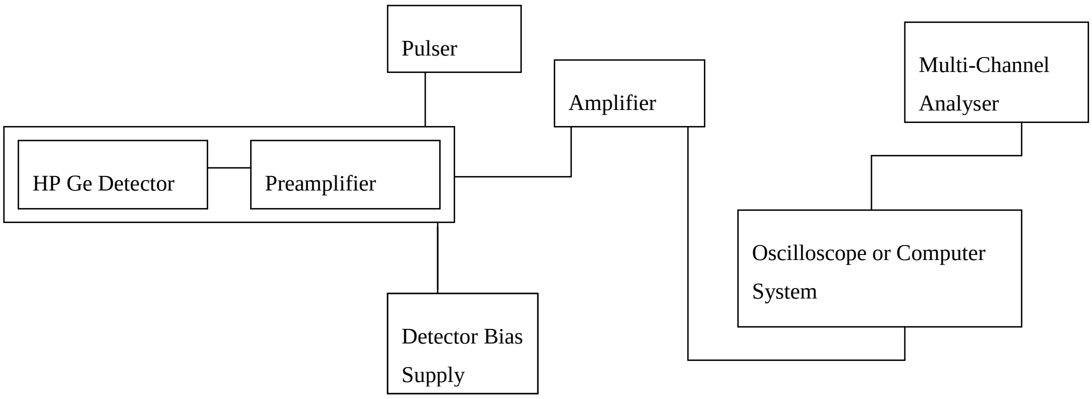

Figure 3.2: Electronic block diagram for HPGe

Data were stored in 8192 sequential channels and operating parameters of the system are governed and controlled by a computer program. The information has been established from calibration measurements performed for Marinell geometry and standard source: The relation between gamma energy and channel. Additionally, the spectrum of laboratory background activity has been established from prolonged measurements. The other necessary but which was not performed is the absolute efficiency calibration and the dependence of peak shape parameter or gamma energy.

3.2 Experimental procedure and Results

Soil samples were collected, on the month of February 2009, from three farm plots located in the surroundings of Gondar town. The first three sample were taken from Bilagig Michael Village at a locale known as Tegedicho (GBT-site). The remaining two samples were taken from the same village but from a different locale known as Shinta (GBS-site). All the sample were taken form holes bored at a distance of 50 cm along the diagonals of the farm land each hole has a depth of up to 20 cm. The soil dug out from each hole was then mixed and a quarter of it was taken to Gondar Soil Test Institute for grinding and drying. Up to 500 gm of each dried and grind soil sample was brought to the nuclear physics laboratory for analysis.

The soil samples collected both from plot lands fertilized by industrial fertilizers, such as urea and DAP; from the boarder of such fertilized form, and from farm land in which artificial fertilizers were not used to enhance the fertility of the farm. The out line that the samples were collected is tabulated in table 3.1.

The soil in farm 1(FS1) were fertilized by industrial fertilizers since 1968 E.C while farm 3(FS3) soil fertilized with artificial fertilizers begin from 1997 E.C;

Table 3.1: Soil Samples Collected from in and around farm plots

Sample #	Sample Code	Location	Farm/Grass land Area	Previous Harvest	Remark
1	FS1	GBT	Farm Soil	Teff	Fertilizer has been used since 1991
2	BS1	GBT	Border Soil	Grass	
3	FS2	GBT	Farm Soil	Teff	No Fertilizer has been used
4	FS3	GBS	Farm Soil	Teff	Fertilizer has been used since 2003
5	BS2	GBS	Border Soil	Grass	

but farm 2 has never fertilized by industrial fertilizer.

The owner of farm 2(FS2) explained that sandy plot provides great yield than other kind of farm land. She believed that the use of fertilizer would sap the natural capacity of the land to yield crop.

Concerning the topography of plot lands; farm 2 is plain land not exposed to soil erosion while farm1 is located at slightly inclined plane that exposed to soil erosion, as a result, it is traced every year. Farm 3 is located around a river with several ups and downs. In comparison with other farms, farm 3 was not protected well to prevent soil erosion.

Farmers add fertilizers on their plot land as they were instructed by specialists. Both farmer of farm 1 and farm 3 took 50 kg of urea and 50 kg of DAP. They would add the fertilizer in humid day to enhance the solubility of the fertilizer. In the first stage they would mix 50 kg of DAP with 25 kg of urea and sowed on the soil. Finally, during the time of weed removal they will sow the remaining 25 kg of urea.

Farm 1 cultivated mostly for teff crop with rotation of higher seed every tired year after the two successive plantation years of teff. However, before land reform of 1988 E.C by the incumbent government the land will remain as fallow for a year after the two successive plantation of teff. It had not experienced crop rotation with other crops such as legumes for the farmer believed that the soil was not appropriate for legume plantation. The farmer explained that the Niger seed plantation is considered as leaving the land as fallow. He is not expect a good yield of niger seed.

Farm 2, that is natural farm land, cultivated for teff and "duragha" (barley & wheat mixture species) by rotation every year.

Unlike the previous two farms, farm3 cultivated by dividing the plot land in to one larger and one smaller parts. The larger pilot planted with wheat while the smaller plot planted with Niger - seed for one season and next time the whole farm land planted with teff. The farmer describe ed that the soil fertility of the plot land show difference so he class fied and used crop rotation for different kinds of crops.

Farmers believed that yields of plot lands vary with respect to the rate of rain fall not due to the utilization of artificial fertilizers. The farmer of farm 1 got 8 "Chan" or 24 quintal of teff; 4 quintal of teff; 4 quintal of Niger. seed every crop season. The second farm yield 10 'madiga' or 3.6 quintal every year for both crops, the 3rd farm yield 17 'madiga' or 5.1 quintal to 20'madiga' or 6 quintal teff and 40.5 kg Niger seed and 7 to 10 'madiga' or 2.1 quintal to 3 quintal of wheat in every crop season.

To detect the radioactivity of the soil, the soil should pass through the process of dissolution and purification, which are expensive and time taking processes. For the process of dissolution and purification were not possible in our laboratory. Therefore, we were forced to count the gross beta in the untreated soil sample Since detection of radioactivity using untreated soil samples is difficult due to the effect of self absorption. Based on the mentioned assumption the untreated soil samples gross beta counting were made using GM tube. To perform this experiment, measuring gross beta GC602A GM counter with mica window thickness $1.75 g/cm^2$ and shielded by 4cm thick cylindrical lead. The small portion of soil samples were placed in different but identical photo camera film cases. The soils were put inside the shielder at a distance whose open end surface (sample free surface) become five millimeter below the thin mica window of the GM counter to measure the radioactivity of each sample. It was clear that the first step of the procedure was determination of the characteristics of GM tube. And it was done using strontium sample with the background, which gives the following information, such as threshold (starting) voltage of 471.28 volt, Knee (transition) voltage of 952.38 volts, $N_1 = 1055.97$ and $N_2 = 10336.67$ counts respectively with the above stated voltages, operating voltage of 710 volt and 4.8 percent per hundred volt slope of the GM range of the characteristic curve which is plotted applied voltage versus counts per 120 seconds time interval that shown below.

Then as mentioned above all samples fixed at that distance from the mica window of GM tube and their radioactivity (gross beta) was counted at operating voltage of 710 volt. The gross beta of each sample were counted within 100 seconds time interval continuously for 10000 seconds. However, as the obtained result evaluated statistically it become unsatisfactory as compared with the critical level (value) of the radioactivity. Then we forced to design as a mechanism of gross beta counting, such as thermal neutron activation analysing, after travelling further steps we became in doubt in the effect of neutron activation analysing method. Lastly, the only choice was to use HPGe which found in Ethiopian Radiation Protection

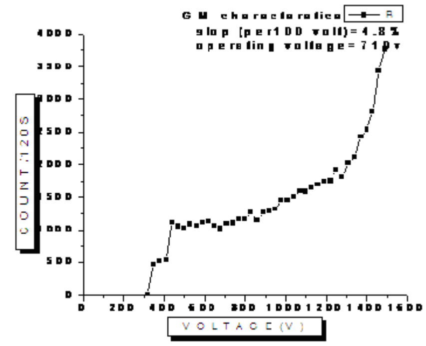

Figure 3.3: The characteristic curve of GM tube

authority.

The radioactivity of the samples were measured by low background HPGe gamma spectrometry.Then, the gamma radioactivity measurements of all soil samples of 448gm quantity contained in identical and independent beakers were done with the coaxial HPGe Canbera GC 1519 type with resolution of 1.9Kev FWHM at 1.33Mev and 44:1 peak-compton ratio at 15 percent of relative efficiency (capacity) that enables identification measurements for most natural isotopes as well as artificial radionuclides.

The crystal detector is with dimension of 59mm diameter and 32mm length. The distance between the source and the detector was set to 5mm and it is reverse biased by high voltage of 5KV (5000 V). The level of liquid nitrogen always checked by metal stick to check if it is enough for operation.

Some standard isotopes, such as ^{137}Cs and ^{60}Co, with precisely known activities were used for with calibration of energy by recording a spectrum of a calibration source with several known gamma energies, finding the corresponding peaks in the spectrum and entering the energies into multichannel analyzer. The multichannel analyzer then performs a least squares fit to the data and assign each channels as energy according to the next equation.

$$E = a + b.n$$

Where: a= offset, b= proportional factor and n= channel number. This implies that the amplification is assume to be linear, that is the channel energy are proportional to the pulse height.

And set of double standard radionuclides used for efficiency calibration in order to give specific results ($\Rightarrow$ γ- spectrometry has to be calibrated in terms of energy and efficiency). Since only some of the emitted photons reach to the detector (due to geometrical reasons and self absorption in the sample), then this fraction is dependent of the sample geometry and on the detector properties which vary with photon energy. By taking all this factors into account the detection efficiency can be calculated using

$$\varepsilon(E) = \frac{N}{t.f.A}$$

Where: $\varepsilon(E)$ is equal to energy-dependent peak efficiency, N is equal to counts in peak, f= branching ratio of observed γ-emission, A= activity in the sample and t= time taken for counting.

Using the above equation it is possible to calculate the specific activities A_s in $\frac{Bq}{Kg}$ of radionuclides by the following expression:

$$A_s = \frac{N}{t\epsilon m}[Silva; Mazzilli, 05p.124]$$

Where: m is mass of the sample in kg.

For the expression of detection limit (minimum detectable activity) L_D,is given in the next equation, where the factor 4.66 corresponds one level of confidence of 95%.

$$L_D = \frac{4.66\sqrt{N}}{\epsilon f t m}(in Bq/kg)$$

The parameter of this equation for the calculation of the L_D can be applied to the measurement of background in the photo peak of each radionuclides (1.46Mev of ^{40}K (Santos et al., 2005, p.222)and (Hamby & Zometsky, Hamby & Zometsky, p.5). However, due to lack of standard source of radioactivity (radioisotopes) efficiency calculation were not done. Even the gamma spectrometry has been calibrated in energy using only the two mentioned standard sources with extrapolating technique. Finally, the quantitative and qualitative analysis of the results performed with Genie-2000 software.

Chapter 4

Results Discussion and Conclusion

In this work an attempt was made to test whether gross beta counting can be used to detect radioactivity in the soil. Beta counting of the samples were made without any chemical treatment on the sample and all measurements using all samples in a lead castle shield have shown that the beta activity in the soil samples is below the detection limit.

4.1 Results of Gross Beta counting

The gross beta counts were obtained using GM counter at 710 operating voltage with 100 second intervals continuously for 10,000 seconds. The result of the count for the five soil samples is presented in table 4.1 below.

From the result shown in table 4.1, the average and standard deviation of the samples were calculated and tabulated in table 4.2.

To determine the presence of detectable, with in the detection limit of our system, radioactivity in the farm soils the critical level of the gross beta count for soil radioactivities were determined using equation (2.2):

$$L_c = 2.32\sigma_{BG} \qquad \text{((Knoll, 2000))}$$

The critical value the counting was, thus L_c= 24.26. However, the average gross beta counting of tested soil samples was below the critical level counting. In other words, the average gross beta counting is below 24.26. Therefore, if it is applied directly the implication that the net counts less than the critical level, it may lead to conclude that the samples have no a real radioactivity. This conclusion had the possibility to be wrong for the samples were not dissolved and purified which is worth to be performed to count gross beta in soil samples. In this study dissolution and purification of soils were not done or the farm soil samples were not passed through these processes in the experiment, therefore, it would not safe

Table 4.1: gross beta counting of soil samples

	bg	tb005	m001	MB02	EF003	T004
	71	77	72	96	69	75
	58	70	89	82	75	52
	73	82	80	87	88	67
	78	62	89	73	89	68
	64	92	68	88	76	106
	70	72	77	77	55	84
	76	70	59	66	70	68
	67	100	73	93	74	57
	81	75	84	80	60	66
	79	77	85	82	74	39
	65	91	82	80	62	84
	65	72	62	86	88	65
	74	49	103	78	76	70
	44	79	62	72	65	58
	44	81	89	86	86	85
	48	58	82	92	62	89
	61	61	53	83	82	83
	54	86	64	71	70	94
	82	79	104	102	54	83
	61	75	63	78	78	78
	44	74	94	78	86	62
	64	93	95	100	104	66
	54	80	82	116	76	86
	61	83	64	61	78	78
	51	77	60	85	72	92
	58	80	79	71	69	79
	56	70	82	82	83	83
	74	82	75	104	62	57
	64	86	95	66	73	85
	55	85	68	76	67	71
	72	79	100	66	78	60
	47	68	74	78	72	86
	52	89	81	96	99	74
	71	57	81	80	67	78
	56	94	81	99	71	88
	65	109	89	58	101	77
	63	92	104	73	73	96
	70	79	107	56	75	81
	64	82	72	102	72	86
	52	54	98	60	78	93
	40	72	93	83	83	87
	60	60	72	102	69	79
	65	81	74	76	91	50
	67	66	70	75	94	81
	66	89	93	83	64	67
	72	85	79	86	70	89
	80	78	98	59	79	72
	65	81	80	76	78	71
	43	82	75	66	60	75
	54	71	92	78	56	91
	65	79	93	78	93	60
	74	64	84	76	85	70
	65	99	75	82	73	53
	54	79	99	69	72	96
	44	82	90	67	56	67
	61	80	64	61	89	80
	46	71	76	83	107	77
	55	66	96	77	72	61
	64	90	95	92	48	86
	80	84	81	74	69	70
	68	92	85	81	82	71
	71	81	44	61	88	65
	40	84	65	89	66	70
	68	94	66	69	93	61
	48	74	102	69	67	88
	65	89	80	87	74	81
	53	77	104	110	54	92
	70	68	92	100	61	64
	67	88	71	86	90	86
	76	75	97	72	64	87
	56	101	89	77	76	85
	72	69	80	75	82	94
	76	72	80	67	92	92
	56	85	96	102	88	91
	58	66	84	82	80	76
	60	72	86	77	80	72
	48	85	80	102	98	76
	60	113	71	78	90	82
	75	76	80	94	70	86
	50	63	62	68	86	86
	54	94	80	101	77	64
	71	87	89	68	88	58
	77	103	72	69	74	52
	55	63	72	78	72	90
	43	68	69	64	87	82
	66	88	96	77	71	88
	64	68	79	80	74	90
	77	71	77	82	68	88
	66	71	75	107	57	75
	63	67	99	78	90	79
	56	88	85	108	86	93
	69	65	76	90	98	74
	70	90	72	118	53	99
	68	84	80	90	63	90
	73	80	96	74	92	82
	60	94	89	72	78	87
	49	83	59	92	90	47
	75	66	70	65	86	49
	70	102	91	92	77	83
	56	66	84	113	61	82
AVERAGE	**62.57143**	**77.85714**	**80.71429**	**80.91837**	**75.44898**	**75.91837**
STDEVA	**10.88194**	**11.89363**	**13.3713**	**13.40217**	**11.27324**	**13.25053**

to conclude that the farm soils have no real radioactivity. On the other hand, it showed the difficulty of counting the gross beta of untreated soil sample by the help of GM counter due to the bulkiness (self absorption effect) of the sample.

It was then concluded that gross beta counting, using GM counting system, can not be used to detect radioactivity in the present soil samples. Therefore gamma counting was done on the samples.

4.2 Results of gamma counting

Gamma counting of farm soils were done using HPGe. The counting gamma count obtained for the soil samples, is tabulated in tables 4.3 to 4.7 and the gamma background result is shown in table 4.8

Table 4.2: average gross beta counting of soil samples

sample	average counts	STDEVA	Ns=Nt-Nb
FS1	81.24	12.81	18.24
BS1	81.66	13.77	19.19
FS2	76.4	12.42	13.97
FS3	76.88	13.11	14.41
BS2	79.02	12.02	16.47
B.G	62.47	10.46	

4.3 Analysis of Gamma Counts

The result of tables 4.3 to 4.7 and table 4.8 were used to calculate the relative concentration of radionuclides in the soil samples studied. Those gamma rays having the highest emission probability were used in the calculation. The results of this calculation is shown in table 4.9

Table 4.3: Gamma counting observed for Sample FS1

Peak No	Possible gamma emmitting Radio nuclides	FWHM (KeV)	Energy (Kev)	Net Peak area	Peak area uncertain	Cts/sec	Error %	Peak critical level
1	Pa-231 Th-231	0.31	28.19	114	14.31	0.004	12.25	35.3
2	Pb-212 Pb-214	0.58	75.38	461	29.94	0.016	6.5	68.2
3	Pb-212 Pb-214	0.59	77.61	648	32.56	0.023	5	65.5
4	Cd - 109 Pb-212 Pb-214 Th - 227	0.77	87.72	95.7	46.72	0.0033	48.8	75.2
5	Ra-226 U-235	0.97	186.28	494	31.07	0.017	6.3	70.7
6	Ac-228 Th - 227	1.04	209.52	218	25.99	0.0076	11.9	69.2
7	Pb-212	1.11	238.89	1900	48.94	0.066	2.6	65.3
8	Pb-214 Sr-92 Xe-138(125)	1.12	241.94	299	26.27	0.01	8.8	64.8
9	Pb-214	1.25	295.36	554	30.63	0.019	5.5	60.5
10	Pb-212 Pa-231 Th-227	1.26	300.12	94	21.28	0.0033	22.6	62.1
11	Ac-228 Cs-136	1.35	338.59	374	26.78	0.013	7.2	48.1
12	Pb-214 Bi - 211	1.37	352.13	887	25.3	0.031	4	53.5
13	Sb-125 Ac-228 Cs - 136	1.59	463	87.8	19.29	0.003	22	35.8
16	Bi-214 Ru-103 Xe-135	1.84	609.58	651	31.25	0.023	4.8	48.5
17	Cs-137	1.93	661.9	523	28.19	0.018	5.4	48.5
18	Bi-212 I-132	2.02	727.72	98.6	18.66	0.0034	18.9	37.5
19	Ac-228 Cs-134	2.12	795.07	42.3	16.25	0.0015	38.5	30.9
20	Ac-228	2.28	911.52	391	25.22	0.014	6.5	45.4
21	Ac-228 Sb-124	2.35	969.31	179	20.77	0.0062	11.6	36.3
22	Sc-46 Bi-214	2.54	1120.72	172	19.64	0.006	11.4	31.9
24	Bi-214	2.67	1238.9	72.8	16.67	0.0025	22.9	28.5
25	K-40	2.91	1461.37	2540	51.87	0.088	2	31.5
27	Bi-214	3.81	1730.21	34.5	9.21	0.0012	26.7	13.4
28	Bi - 214	3.21	1765.36	186	15.62	0.0065	8.4	15.1

Table 4.4: Gamma counting observed for Sample BS1

Peak No	Possible gamma emmitting Radio nuclides	FWHM (KeV)	Energy (Kev)	Net Peak area	Peak area uncertain	Cts/sec	Error %	Peak critical level
1	Pb-212 Pb-214 Bi - 211	0.57	74.25	454	30.21	0.016	6.6	64.9
2	Pb-212 Pb-214	0.58	76.47	541	31.16	0.019	5.8	61
3	Cs - 136 Pb-212 Pb-214 Th - 227 Cd - 109	0.63	86.39	171	25.2	0.0059	14.7	77.3
4	U-235 Ra-226	0.97	185.04	394	29.84	0.014	7.6	99.7
5	Pb-212 Th-227 Nb-95	1.11	237.52	1570	45.34	0.054	2.9	66.5
6	Sr-92 Pb-214 Xe-138	1.12	240.63	329	27.1	0.011	8.2	61
7	Ba-133 Sr-91	1.2	276.48	72	21.31	0.0025	29.6	60.6
8	Pb-214	1.25	294.01	430	28.75	0.015	6.7	57.1
9	Th-227 Pa-231 Pb-214	1.26	299.04	112	21.51	0.0039	19.2	57.5
10	Ac-228	1.34	336.99	307	26.19	0.011	8.5	61
11	Bi-211 Pb-214	1.34	350.56	681	32.82	0.024	4.8	79.9
12	Zr-97	2.51	509.22	702	35.33	0.024	5	74.4
14	Sb-125	1.82	598.63	64.3	17.65	0.0022	27.4	41.6
15	Xe-135 Sb-125 Bi-214	1.84	607.61	522	29.17	0.018	5.6	57.8
16	Cs-137 Nb-97	1.92	659.79	453	27.59	0.016	6.1	56.3
17	Zr-89	2.27	909.07	369	25.06	0.013	6.8	50.3
18	Sb-124	2.35	967.07	199	21.68	0.0069	10.9	51.3
19	Kr-89	2.53	1117.73	161	20.04	0.0056	12.5	95.8
21	I-135	2.91	1457.34	2720	53.63	0.095	2	44.8
22	Ac-228	3.04	1589.29	75.1	13.23	0.0026	17.6	23.7

Table 4.5: Gamma counting observed for Sample FS2

Peak No	Possible gamma emmitting Radio nuclides	FWHM (KeV)	Energy (Kev)	Net Peak area	Peak area uncertain	Cts/sec	Error %	Peak critical level
1	Pb -212 Pb -214	0.59	77.17	321	26.76	0.013	7.4	82.1
2	Pb -212 Pb -214 Cd - 109 Cs - 136 Th - 227	0.63	87.2	132	23.2	0.0061	13.8	63
3	U - 235 Th - 231 Pb - 212 Pb - 214 Th - 227 Cd - 109	0.64	90.01	91.8	22.22	0.0038	20.8	58.3
4	Ac - 228 U - 235 Th - 227 Pa - 234	0.65	93.08	235	23.78	0.0027	11.2	60.5
5	U - 235 Ra - 226	0.97	185.58	235	25.44	0.0084	10.8	83.5
6	Ac - 228 Th - 227	1.03	208.95	110	22.15	0.003	26.3	60.2
7	Pb - 212	1.11	238.36	1310	47.76	0.045	3.2	64.2
8	Sr - 92 Pb - 214 Xe - 138	1.12	241.24	225	24.04	0.0075	11.5	63.2
9	Pb - 214	1.25	294.83	235	24.52	0.0087	10	66.8
10	Ac - 228 La - 140	1.32	327.92	64.9	18.84	0.0026	25.9	45.4
11	Ac - 228	1.34	337.95	289	24.2	0.011	7.9	63.1
12	BI - 211 Pb - 214	1.37	351.43	433	27	0.015	6.4	57.7
15	Xe - 135 Bi - 214 Ru - 103	1.84	608.7	286	23.72	0.0094	8.9	50
16	Cs - 137	1.92	660.85	551	29.21	0.018	5.6	57.7
17	Ac - 228 Zr - 89	2.28	910.29	337	23.87	0.012	7.2	50.4
18	Eu - 152 Ac - 228	2.35	963.96	65.1	16.42	0.0018	31.6	42.1
19	Sb - 124 Ac -228	2.35	968.06	170	19.85	0.0053	13.1	45.9
20	Bi - 214 Sc - 46	2.54	1119.01	939	17.36	0.0028	22	39.2
21	K - 40	2.91	1459.67	2480	51.01	0.086	2.1	40.8
23	Bi- 214	3.21	1763.35	152	14.36	0.0052	9.7	19.9

Table 4.6: Gamma counting observed for Sample FS3

Peak No	Possible gamma emmitting Radio nuclides	FWHM (KeV)	Energy (Kev)	Net Peak area	Peak area uncertain	Cts/sec	Error %	Peak critical level
1	Pb-212 Pb-214	0.57	74.91	410	27.85	0.017	6.8	58
2	Pb-212 Pb-214	0.58	77.16	576	29.81	0.024	5.2	51.5
3	Pb-212 Pb-214 Cd-109 Cs - 136 Th - 227	0.63	87.18	229	24.38	0.0095	10.7	60.4
4	Th-231 Ac - 228 Pb-212 Pb-214 Cd-109	0.64	89.91	104	22.63	0.0043	21.8	63
5	U-235 Ra-226	0.97	185.71	343	27.08	0.014	7.9	82.8
6	Ac-228 Th-227	1.03	209.06	147	23.25	0.0061	15.8	74.4
7	Pb-212	1.11	238.47	1690	46.15	0.07	2.7	59.3
8	SR-92 Pb-214 Xe-138	1.12	241.24	319	25.44	0.013	8	56.5
9	Pb-214	1.25	295.05	343	26.17	0.0014	7.6	53.3
10	Pa -231 Pb-212 Th - 227	1.26	300.03	81.7	20.63	0.0034	25.3	54.2
11	Ac-228	1.34	338.15	336	25.38	0.014	7.5	55.9
12	Pb-214 Bi-211	1.37	351.76	532	29.38	0.022	5.5	52.3
13	Ac-228 Cs-138 Sb - 125	1.59	463.04	104	18.24	0.0043	17.6	45.8
16	Bi-214 Xe-135 Ru - 103	1.84	609.15	372	24.96	0.015	6.7	46.1
17	Cs-137	1.92	661.58	439	25.82	0.018	5.9	45.1
18	I-32,130	1.94	669.58	64.5	15.81	0.0027	24.5	38.9
19	Ac-228	2.28	911.32	412	24.66	0.017	6	50
20	Ac-228 Sb-124	2.35	969.38	186	19.97	0.0077	10.7	40.5
21	Sc-46 Bi-214	2.54	1120.42	104	16.74	0.0043	16.1	33.6
23	K-40	2.91	1460.61	1980	46.23	0.082	2.3	38.7
24	Ac-228	3.04	1587.38	30	10.44	0.0012	34.8	23.8
26	Bi - 214	3.21	1763.19	117	13.3	0.0049	11.4	16.5

Table 4.7: Gamma counting observed for Sample BS2

Peak No	Possible gamma emmitting Radio nuclides	FWHM (KeV)	Energy (Kev)	Net Peak area	Peak area uncertain	Cts/sec	Error %	Peak critical level
2	Pb-212 Pb-214	0.57	74.67	443	28.99	0.017	6.5	63.4
3	Pb-212 Pb-214	0.59	76.92	553	30.12	0.021	5.5	59.8
4	Pb-212 Pb-214 Cd-109 Cs - 136 Th - 227	0.63	87.06	197	25.55	0.0097	9.9	73
5	Ac-228 U - 238 Th-227 Pa - 234	0.65	92.73	320	27.15	0.013	8.1	92.5
6	Ac - 228	0.79	128.91	113	22.68	0.0038	22.6	69.5
7	U-235 Ra-226	0.97	185.71	397	28.37	0.015	7.1	92.3
8	Ac-228 Th-227	1.04	209.09	153	24	0.0062	14.5	72.1
9	Pb-212	1.11	238.26	1730	48.09	0.061	2.9	91.1
10	Ac-228 Rn-219	1.19	270.13	137	21.87	0.0044	18.5	75.1
11	Ba-133 Hg-203	1.21	277.22	77.2	19.61	0.003	24.3	58.2
12	Pb-214	1.25	295	342	26.32	0.012	8	58.8
13	Th-227 Pa-231 Pb-212	1.26	299.87	129	20.96	0.0048	16.4	56
14	Ac-228 La-140 Th-227	1.33	327.93	95.5	19.99	0.0034	22.3	53.6
15	Ac-228	1.35	338.11	372	26.48	0.014	7.1	74.4
16	Pb-214 Bi-211	1.38	351.67	626	31.38	0.024	4.9	54
17	Cs-136 Ac-228 Sb - 125	1.59	462.6	136	19.77	0.0053	13.8	59
20	Bi-214	1.83	609.06	473	27.2	0.018	5.7	60.5
21	Cs-137	1.93	661.42	409	25.71	0.015	6.4	45.9
22	Bi-212 I-132	2.03	727.18	105	18.68	0.0037	18.9	47.3
24	Ac-228 Zr-89	2.28	911.04	405	25.36	0.015	6.1	57.2
25	Ac-228 Sb-124	2.36	968.9	282	22.2	0.0099	8.2	50.3
26	Bi-214 Sc-46	2.54	1120.1	158	18.74	0.0058	11.9	41.4
27	K-40	2.92	1460.59	2020	48.42	0.08	2.2	40.7
29	Bi - 214	3.22	1764.11	211	16.32	0.077	7.8	18.8

Table 4.8: Gamma counting observed for Background Radiation

Peak No	Possible gamma emmitting Radio nuclides	FWHM (KeV)	Energy (Kev)	Net Peak area	Peak area uncertain	Cts/sec	Error %	Peak critical level
1	Pa-231 Th-231	0.31	28.15	48	9.43	0.0017	21.3	38.8
2	U-235 Ra-226	0.97	185.9	158	19.28	0.0055	12.5	68.7
4	Ac-228	2.28	911.61	64	15.59	0.0022	24.5	33.8
6	Bi-214 Sc-46	2.45	1120.12	52	15.21	0.0018	29.9	32.1
7	K-40	2.91	1460.95	1459	39.86	0.051	2.7	37.3
8	Bi - 214	3.21	1760.73	136	13.34	0.007	9.9	38.1

Table 4.9: Analysis of relative concentrations of radionuclides

γ E (keV)	Nuclide	Yield (%)	$T_{\frac{1}{2}}$	$T_{\frac{1}{2}}$ (in sec)	Origin	Sample FS1			Sample BS1			Sample FS2			Sample FS3			Sample BS2		
						Net Cnt rate (sec^{-1})	% Error	Conc. (nuclides/ kg soil)	Net Cnt rate (sec^{-1})	% Error	Conc. (nuclides/ kg soil)	Net Cnt rate (sec^{-1})	% Error	Conc. (nuclides/ kg soil)	Net Cnt rate (sec^{-1})	% Error	Conc. (nuclides/ kg soil)	Net Cnt rate (sec^{-1})	% Error	Conc. (nuclides/ kg soil)
186.1	Ra-226	3.28	1600Y	5.05E+10	^{238}U	0.0170	6.3	8.44E+10	0.0140	7.6	6.95E+10	0.0084	10.8	4.17E+10	0.0140	7.9	6.95E+10	0.0140	7.1	6.95E+10
351.9	Pb-214	37.1	26.8m*	1.61E+03	^{238}U	0.0310	4.0	4.33E+02	0.0240	4.8	3.36E+02	0.0015	6.4	2.10E+01	0.0220	5.5	3.08E+02	0.0230	4.6	3.22E+02
609.3	Bi-214	46.1	19.9m	1.19E+03	^{238}U	0.0230	4.8	1.92E+02	0.0180	5.6	1.50E+02	0.0099	8.9	8.27E+01	0.0160	6.7	1.34E+02	0.0180	5.7	1.50E+02
911.2	Ac-228	29	6.13H	2.21E+04	^{232}Th	0.0014	6.5	3.44E+02				0.0120	7.2	2.95E+03	0.0160	6.0	3.93E+03	0.0150	6.1	3.68E+03
238.6	Pb-212	43.6	10.6H	3.82E+04	^{232}Th	0.0066	2.6	1.86E+03	0.0550	2.9	1.55E+04	0.0450	3.2	1.27E+04	0.0680	2.7	1.92E+04	0.0630	2.9	1.78E+04
1460.8	K-40	10.67	1.28E+9Y	4.04E+16	Primordial	0.0880	2.0	1.07E+17	0.0460	2	5.62E+16	0.0860	2.1	1.05E+17	0.0810	2.3	9.89E+16	0.0800	2.2	9.77E+16
661.7	Cs-137	85.1	30.17Y	9.52E+08	Fission	0.0180	5.4	6.50E+07	0.0160	6.1	5.77E+07	0.0180	5.6	6.50E+07	0.0180	5.9	6.50E+07	0.0150	6.4	5.41E+07

Normalizing the concentration to sample FS2, the farm land in which no chemical fertilizer has been used, we obtain the relative concentrations of natural and manmade radionuclides as follows.

Table 4.10: Relative concentratively of Radio nuclide

γ E (keV)	Nuclide	Yield (%)	$T_{\frac{1}{2}}$	$T_{\frac{1}{2}}$ (in sec)	Origin	Sample FS1	Sample BS1	Sample FS2	Sample FS3	Sample BS2	Remark
						Rel. Conc.	Rel. Conc.	Rel. Conc.	Rel. Conc.	Rel. Conc.	
186.1	Ra-226	3.28	1600Y	5.05E+10	U-238	2.02	1.67	1.00	1.67	1.67	
351.9	Pb-214	37.10	26.8m*	1.61E+03	U-238	20.67	16.00	1.00	14.67	15.33	
609.3	Bi-214	46.10	19.9m	1.19E+03	U-238	2.32	1.82	1.00	1.62	1.82	
911.2	Ac-228	29.00	6.13H	2.21E+04	Th-232	0.12		1.00	1.33	1.25	
238.6	Pb-212	43.60	10.6H	3.82E+04	Th-232	0.15	1.22	1.00	1.51	1.40	
1460.8	K-40	10.67	1.28E+9Y	4.04E+16	Primordial	1.02	0.53	1.00	0.94	0.93	
661.7	Cs-137	85.10	30.17Y	9.52E+08	Fission	1.00	0.89	1.00	1.00	0.83	

As can be seen in tables 4.9 and 4.10 selected nuclides were analyzed to determine the concentration of their radionuclide. The analysis was done based on their life time and and the probability of emission of a selected gamma energy. The energy that had the greatest yield was used to determine the concentration of the corresponding radionuclide. To do so the rate of decay of the radionuclides were calculated using the following formulae.

$$\text{Count Rate} = \text{number of atoms}[\frac{0.692(Branchingratio)}{T_{1/2}]} \tag{4.1}$$

The relative concentrations were determined by the use of equation (4.1).The criteria used to select radioactive elements were the character of having long lived with high branching rates,and those coexist in equilibrium with their respective members of uranium series and thorium series specially the interest was focussed on those with high gamma emission probabilities. Here the results of the experimental data was discussed based on the elements Ra-226,Pb-214, Bi-214 from uranium series ; Ac-228, Pb-212, Bi-212 from thorium series ; Cs-137 with branching ratio 85.2 percent and potassium with branching ratio of 10.67 percent.

The counting rate of gamma emission of these radioactive nuclides in the investigated farm soils were tabulated above. The radionuclide Count rates of Radium-226, Pb-214, Bi-214 of uranium series and Ac-228, Pb-212, Bi-212 of thorium series, K-40 and Cs-137 was expressed in terms of rate of emission of gamma ray($\frac{counts}{second}$) and concentration of radionuclides in the soil.This study showed that the natural isotope Pb -214 at energy level 351.2 Kev of the uranium series is the main source of soil contamination that provides high relative concentration of radionuclides of the respective farm soil samples. Generally, as shown in the table relative concentration of nuclides,the natural radioactive nuclides from the uranium series ,Such as,Ra-226 ,Pb-214 and Bi-214 concentration is relatively high at the farm which was chemically fertilized for long time. This result is strongly associated with the studies of (Becegato et al., 2008, p.1264)that stated radioactivity concentration in various production area of chemical fertilization are mainly due to the fractioning of the chemical reaction in acidulation phase phosphoric acid and production.Products of MAP(mono-ammonia phosphate) show that radium concentration is higher due to production process of such fertilizer which use ammonia,which in turn does not react with phosphoric rock ,which is opposite of what uranium-rich phosphoric acid.

According to this principle the count rate of uranium series elements relative concentrations were slightly higher in farm soils (Fs1 and Fs3)where chemical fertilizer was added for long period of time than the farm land(FS2)that were not treated with artificial chemical fertilizers. This result has a strong correlation with the study results for the presence of some isotopes that from U-238 series in chemical fertilizers (Chibowski, 2000, P.252)

In contrast,the reverse was true about the relative concentration of radionuclides of thorium series elements.For that matter the concentration of Pb-212(238.6Kev)and Ac-228(911.2) was lowest at relatively fertilized for long time and lower at native soil farm(Fs2) than the farm found at other site area (FS3).This might be arisen from the nature of the soils of the plot lands.Nevertheless,the relative concentration of the primordial radionuclide K-40 is the same in all farm lands(FS1,Fs2 and FS3) which did not correlate with the principle that chemical fertilizers contribute for the rise of the concentration,of K-40 in soil (Chibowski, 2000, p.250) and (Becegato et al., 2008, p.1264).

However, in the case of this study there was no significant difference in gamma emission count rate of K-40 on the investigated farm soils that were fertilized by artificial chemical fertilizers and farm soils that were not treated with artificial chemical fertilizers. The result is not correlated with the results indicated in several studies (Chibowski, 2000, p.250), (Becegato et al., 2008, p.1264). This might be due to the improper addition of fertilizers by farmers on their plot lands (**??**). The other natural radioactive nuclides that were obtained from the radioactivity studies were Pb-214, Ra-226, Bi-214 from uranium series and Pb-212, Bi-212, Ac-228 from thorium series.

When the relative concentration of radionuclides compared in the margin of chemically fertilized farm(BS1)with respective main farm soils of (FS1)the relative concentration of radionuclides observed in (BS1)is lower except the thorium series element Pb-212. Similarly, the comparison of the relative concentration of radionuclides in (BS2) and (FS3) showed that the relative concentration of radionuclides observed in (BS2) is lower except the series elements of uranium such as Ra-226, Pb-240 and Bi-240, this might be due to high erosion at the heart land and sedimentation of eroded soil at the border of (FS3) farm. In the results of the border soil samples the relative concentration of thorium and uranium series elements were decreased and increase alternatively in opposite manner. In other words, while the elements in the thorium series increases the elements of uranium series will decrease and vice versa.This might arise due to high leaching of these elements in the heart of the farm land than that of the margin of the farm that eroded surface soil deposited were found.The reason for the variation of relative concentration of radionuclides in the thorium series elements and uranium series elements in the margin soil and the heart land soil might be due to the fact that radioactivities of uranium-238 increases with depth because uranium-238 is leached from the surface soil and accumulated in deeper zone (Navas et al., 2002, p.632).Further it might be due to lack of organic matter and sand soils,where uranium is usually mobile and transported as hexavalent carbonate complex or divalent uranaly ion (Rothman et al., 1979). In addition to these reasons farm one(FS1) was terraced frequently which can reduce the eroded soil deposition on the boarder of the farm.

Lastly, radionuclide Cs-137 is artificial radionuclides which distributed as the form of fall out through out the world, then as expected the gamma emission count rate of Cs-137 was the same in all examined soil of farm plots while a slight difference was observed at the border of plot lands.The similar result of K-40 in the farm soils was because they had the same chemical behavior with Cs-137.

4.4 Conclusion

The gross beta count measurement of untreated farm soils were below the critical level. However, the presence of real radioactivity in the untreated soils were confirmed using gamma spectrometry measuring instrument (HPGe). Even if the gross beta count measurement does not unfold the real radioactivity of untreated farm soils it may be used for survey study of radioactivities in farm soils.

The counting rates of radionuclides obtained using HPGe would not help to compare and discuss the relationships with other study results. This is primarily due to the absence of certified standard radionuclide that required for efficiency calibration.

Finally, the relative concentrations of radionuclides of uranium and thorium series elements were varied alternatively in opposite manner in the border soil samples as compared with their respective main farm soils. The K-40 and Cs-137 elements relative concentration in the border soil samples were lower compared with others. These variations may be due to the dependence of radionuclide concentrations on erosion, leaching, adsorption and chemical properties of the nuclides and the nature of the soil.

4.5 Recommendation

The radionuclides of farm soils should be studied in detail taking into account its importance to contribute in the development of plantation and other agricultural activities in the future Ethiopia. Secondly, the department should furnished its laboratory with equipments by buying and installing MCA for the HPGe that is not functional yet. Moreover, it would have certified standard radionuclides for calibration of energy and efficiency of study results.

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DECLARATION

I the under signed declare that the thesis is my original work, has not been presented for a degree in any other university and that all sources of material used for the thesis have been duly acknowledged.

Name: ____________________

Signature: ____________________

This Thesis has been submitted for examination with my approval as university advisor.

Name: ____________________

Signature: ____________________