MEASUREMENTS OF NATURAL RADIOACTIVITY LEVELS IN THE SOIL, COFFEE AND ZUWAYE LAKE WATER SAMPLES COLLECTED FROM THE SELECTED AREA, IN ETHIOPIA, BY USING GAMMA RAY SPECTROSCOPY.

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A DISSERTATION SUBMITTED TO ADDIS ABABA UNIVERSITY COLLEGE OF NATURAL AND COMPUTATIONAL SCIENCE DEPARTMENT OF PHYSICS, IN PARTIAL FULFILLMENT FOR THE REQUIREMENT OF THE DEGREE OF DOCTOR OF PHILOSOPHY IN NUCLEAR PHYSICS.

JUNE, 2020

ADDIS ABABA UNIVERSITY

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Department: Physics

Degree: PhD Year: 2020

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Prof. A.K.Chaubey

(Supervisor)

Acknowledgements

I thank my advisor Prof. A.K.Chaubey, for he provided me insightful professional assistance in the completion of this dissertation next to Almighty God (Allah).

I would like to present my great thanks to my husband Muhammedziyad Geleto for all his continuous and endless support and patience during my study and in the whole our live. I dedicate this work to you, our sons (Muharuk and Ehan Muhammedziyad), my father and my mother.

I would like to thank Wolkite University and the department of physics at Wolkite University for awarding me sponsorship to pursue PhD study.
I am also thankful the staff of department of physics at Addis Ababa University and all my friends for their support and comments.

I would like to thank Ethiopian Radiation Protection Authority Director and their Research group staff for all their laboratory technical assistance.

Finally, I would like to thank my entire supporter who I did not raise your name here because I alone could not complete this work without your contribution all the time in need.

Rukiya Aliyi

List of Previous Publications

1. Rukiya Aliyi and Chaubey, A. K.Determination of radioactivity levels and Radiation hazards in Soil samples Used for farming purpose. International Journals of Current Research, Vol. 11, Issue, 08, pp.6454-6458, August, 2019.
2. Rukiya Aliyi and Chaubey, A. K.Measurements of levels of natural radionuclides in the soil in Zuwaye, and Shashamene area, Ethiopia, using gamma ray spectromUsed for farming.International Journals of Current Research, Vol. 11, Issue, 07, pp.5721-5725, July, 2019.
3. Rukiya Aliyi.Assessment of radiation levels around x-ray room at four Hospitals near Wolkite town, Ethiopia.International Journals of Current Research, Vol. 11, Issue, 06, pp.xxxxxxxxxx, June, 2019.

Contents

Abstract

People are exposed to radioactivity in soil, plant and even in other environment that is raised from uranium, thorium and potassium-40 radionuclides. Environmental radioactivity and the associated external exposure due to gamma radiations depend primarily on the geological, geographical and human activities. In this study the activity concentration of ^{238}U, ^{232}Th and ^{40}K radionuclides were determined in 12 soil samples, Four coffee samples and Four Zuwaye lake water samples collected from some parts West-Arsi Zone, Shashemane and Zuwaye, Ethiopia using High Pure Germanium Detector (HPGe). The average activity concentration of ^{238}U, ^{232}Th and^{40}K in the soil samples was 45.83, 100.525, 845.65Bq/Kg, respectively. These values were higher than the average world recommended values for ^{238}U , ^{232}Th , ^{40}K , respectively which are 35, 30 and 400 Bq/Kg for ^{238}U , ^{232}Th , ^{40}K respectively [UNSCEAR, 2000].

During our experimental activities our detector could identify only potassium-40 in the coffee samples. All coffee samples had 591.185 Bq/Kg average values of ^{40}K concentrations that were higher than the acceptable value (412 Bq/kg) [UNSCEAR, 2010].

Activity concentration of ^{238}U in Lake Zuwaye Water was vary from 2.94±0.25 Bq/L to 3.02±0.27 Bq/L with an average value of 2.99±0.267 Bq/L, ^{232}Th was vary from 6.9±0.73 Bq/L to 8.39±0.82 Bq/L also with an average value of 7.5 ±0.76 Bq/L and the activity concentration of ^{40}K varies from 98.12 ± 4.16154 to 101.99 ± 4.91556 with an average value 99.77±4.80Bq/L.

The activity concentration from the studied areas were found to be below WHO guidelines of 10 Bq/L for ^{238}U and higher than 1 Bq/L for ^{232}Th. The World Health Organization recommended level of ^{40}K in water meant for drinking is unavailable.

The values of radium equivalent for different soil samples in area under investigated were ranged from 124.96 to 330.7 Bq/kg with average value 248.75Bq/kg which is lower than the recommended maximum value 370 Bq/kg [Gilmore, G.R.,2008].

The calculated values of Dr were found to vary from 96.64 to 191.02nGy h^{-1}, with an average value of 149.5117nGy h^{-1}. The measured average absorbed dose rate in the air and the measured

average value of the representative level index are higher than the recommended international levels of 55 nGy h^{-1}. Such locations are not safe for human residency and agriculture.

The calculated values for Dout, Din, and Dtot respectively 0.185, 1.035, and 1.22 mSv/year to global measured values, these values were all higher than the assigned worldwide values of 0.08, 0.42, and 0.5 $year^{-1}$, respectively [UNSCEAR,2000]. The locations from which the soil samples collected were all not safe according to the Radiation Protection, and such locations cannot be classified as hazard free.

The external hazard index (Hex) and internal hazard index (Hin) due to the emitted γ -rays of the soil samples were calculated according to the following criterion: The value of Hex must be lower than unity in order to keep the radiation hazard insignificant. The calculated external hazard index values were found to vary between 0.43 and 0.78 with average value of 0.668 these values are less than unity, which is 33.2 % less than world recommended value. The calculated internal hazard index values were found to vary between 0.5 and 0.93 with average value of 0.79, which is 11.0% less than world recommended value [Dabayneh K et al,.2008].

Radioactivity level index vary from 1.19 to 2.395 Bq/Kg with average values is 1.917 Bq/Kg. The average value of Radioactivity level index is higher than the recommended safe limit ≤ 1 [Thabayneh K, Jazzar M.2012]. Therefore, the soils have radiation hazard and are harmful to society living there.

The values of radium equivalent for different coffee samples in study area were vary from 42.5 to 54.92 Bq/Kg with average value 47.44Bq/Kg which is lower than the world recommended maximum value 370 Bq/Kg [Gilmore, 2008].

The determined values of absorbed dose rate (Dr) were found to vary from 23.73 to 30.67nGy h^{-1}, with an average value of 26.49nGy h^{-1}. The measured average absorbed dose rate in the air was lower than the recommended international levels of 55 nGy h^{-1}[Turner J et al., 2007] and the coffee was safe for use.

The determined average values for Dout, Din, and Dtot respectively 0.0325, 0.181, and 0.107 mSv /year to global measured values, these values were all lower than the worldwide values of 0.08, 0.42, and 0.50 mSvyear-1, respectively [UNSCEAR, 2000].

The external (Hex) and internal (Hin) hazard index due to the emitted γ -rays of the coffee samples were calculated and examined according to the following criterion: The value of Hex must be lower than unity in order to keep the radiation hazard insignificant. This is the radiation exposure due to the radioactivity from a construction material, limited to 1.5 $mGy \cdot y^{-1}$. The maximum values of Hex equal to unity correspond to the upper limit of Raeq (370 Bq/Kg) [48]. The calculated external hazard index values were found to vary between 0.115 and 0.148 with average value of 0.128 these values are less than unity, which is 87.2 % less than world recommended value. The calculated internal hazard index values were found to vary between 0.115 and 0.148 with average value of 0.128, which is 87.2% less than world recommended value [Knoll G.F, 2000].

Radioactivity level index to vary between 0.36 to 0.475 Bq/Kg with average value is 0.4 Bq/Kg. The average value of Radioactivity level index is lower than one the recommended safe limit ≤ 1 [Van Rooyen T.J., 2002]. Therefore, the coffee had no health radiation hazard.

Hence it can be concluded that the radionuclide concentration of measured coffee samples poses no radiological health hazard to the community.

The annual effective dose calculated for the Zuwaye lake Water was varied from 0.081 to 0.09 mSv/y with an average of 0.086 mSv/y. This value, when compared with the ICPR standard value of 1mSvyr-1 is very low, hence, it can be concluded that the radionuclide concentration of Zuwaye Lake water is negligible and poses no radiological health hazard to the community.

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Chapter One

1. Introduction

1.1Background of the study

Human beings are exposed to radioactivity everyday from the ground, building materials, air, food, the universe and even elements in their own body. In addition to natural background radiation people are also exposed to, low and high-linear energy transfer radiation from the manmade sources such as X-rays and radioactive materials used in medicine, research and industry [UNSCEAR, 2000].

Natural radioactivity is found in rocks, soil, beach sand, sediment and riverbed soil, in rivers and oceans, and even in our building materials and homes. Radionuclides concentration in soil is an indicator of radioactive accumulation in the environment, which affects humans, plants and animals. Naturally occurring radioactive materials generally contain primordial radionuclides, left over since the creation of the earth [UNSCEAR, 1982].

The natural radioactivity in soil is derived mainly from the ^{238}U and ^{232}Th parent series and natural ^{40}K. Natural environmental radioactivity and the associated external exposure due to gamma radiation depend primarily on the geological and geographical conditions. 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 [UNSCEAR, 2000].

Study of soil radioactivity can provide reference data in observing possible future anthropomorphic impact and associated radiological risk to human health. The activity concentrations of radionuclides in the ^{238}U and ^{232}Th decay chains and from 40K were determined through gamma-rays spectrometry in a low background configuration [A. El-Gamal, et al.; 2007].

Ionizing radiation occur naturally either as terrestrial or extraterrestrial origin in form of high energy cosmic ray particles whose sources are galactic and extra –galactic. Terrestrial origin are

due to the presence of naturally occurring radionuclides, mainly Potassium (^{40}K) Uranium (^{238}U) and Thorium (^{232}Th) and they basically occur in geological materials making the rocks and soil [UNSCEAR, 2000]. Exposure to high level of radioactivity can lead to health problems like abnormal cell growth. It is therefore, important to measure the concentration of the radioactivity in the soil and asses the possible health radiological hazard to human and to develop standards and guidelines for the use and management of these materials [Turhan, et al, 2008].

Ionizing radiation is a flux of subatomic particles like photons, neutrons, and nuclei etc that cause ionization of atoms of the medium through which the particles pass. In order for ionization to occur a certain amount of energy must be transferred to the atom. By the law of conservation of energy, this amount of energy is equal to the decrease of kinetic energy of the particle that causes ionization. Ionization is only possible if the energy of the incident particles exceeds the ionization of the atom which is usually of the order of 10 electron volts.

In passing through matter, charged particles lose some kinetic energy by excitation of bound electrons and by ionization. The energy loss of charged particles passing through matter depends on the particle velocity, its charge and the properties of the transverse material. To ionize atoms in human tissue an energy of approximately 30 electron volts and above is required .Radiation with frequencies below 10^6 Hertz corresponding to 30 electron volts are termed as non-ionizing. Biological processes in the human body are influenced by non-ionizing radiation. Low frequency radiation affects humans via its electric and magnetic fields.

High doses of radioactivity can be harmful or even fatal. The damage caused by exposure to a radioacivity is determined by the type of radiation, the duration of exposure and the part of the body that is exposed. The interaction of ionizing radiation with the human body arises from either external sources or internal contamination which can lead to biological effects [UNSCEAR, 2000]. Radiation effects can lead to death of a cell, impairment in the natural functioning of the cell leading to somatic effects such as cancer and a permanent alteration of the cell which is transmitted to later generation i.e. genetic effect. Biological effects can also be considered in terms of stochastic effect and non-stochastic effects. Stochastic effects increases with increase in dose rate [ICRP, 2005]. While non-stochastic effect has a threshold below which

there is no effect. Human exposure to radiation also reduces the immunity of the person exposed [UNSCEAR, 2006].

Environmental radioactivity of natural origin and the exposure associated with it externally as a result of gamma radiation hinges on the geographical and geological conditions of the area and are prominent at different concentrations in the World [UNSCEAR, 2000a]. The rate of gamma dose distribution in the natural terrestrial environment is a major donor to the mean dose accepted by the populace [Tso, M. Y. W., & Leung, J. K. C., 2000]. Estimating the doses of radiation circulating in the environment is the key in examining the health exposure to a populace and provides a locus for recording variations in environmental radioactivity as a result of anthropogenic works [Obed, R. I., Farai, I. P., &Jibiri, N. N., 2005]. Human beings become exposed to radionuclides through pollution of the food chain which results from the direct deposit of radionuclides on sediment or water, leaves of plants, and from drinking of contaminated water and eating of contaminated fish [Avwiri, G. O., et al ., 2007].

Ingested radionuclides are absorbed into the bloodstream and accumulate in specific tissues such as the kidneys, bones and flesh from where they exert both chemical- and radio- toxicities [Avwiri, G. O., etal ., 2007; Bonotto, et al 2009]. Even small concentrations of a radioactive substance have the ability to produce a damaging biological effect [El-Mageed, et al, 2011].

The radioactivity to which the human population is exposed comes from many diverse sources. Some of these sources are natural; others are the result of human made. The radiation from natural sources include cosmic radiation, external radiation from radionuclides in earth's crust and internal radiation from radionuclides inhaled or ingested and retained in the body. The magnitude of these natural exposures depends on geographical, geological location and on some human activities. Height above sea level affects the dose rate from cosmic radiation; radiation from the ground depends on the local geology; and the dose from radon, which seeps from the ground into houses [Gür,et al.,2001].

Exposure to ionizing radiation arises from sources such as medical diagnostic and therapeutic procedures; nuclear weapon testing; radon and other natural background radiation; nuclear

electricity generation; accidents such as the one at Chernobyl in 1986; and occupations that increase exposure to artificial or natural sources of radiation [UNSCEAR, 2010]. Ionizing radiation in our environment can occur either naturally or can be produced artificially, through human activity. The effects of artificial or naturally occurring radiation are the same [UNSCEAR, 2010]. Naturally occurring radionuclide materials (NORM) existed since the creation of the earth. Radionuclides such as uranium, thorium and potassium are relatively abundant in rocks and soils. The gamma radiation emitted from these radionuclides gives to human beings a radiation dose.

Radioactive materials that are found in nature can be placed in three general categories, namely cosmogenic, primordial and anthropogenic. The high energy primary cosmic radiation induces various complex interactions with the earth's atmosphere, and results to cosmogenic radionuclides and hence produces secondary radiation [IAEA, 2003]. However, only a part of these radiations reach the earth's surface because the earth atmosphere acts as a filter and absorbs much of the energy of the cosmic rays. Primordial radionuclides which are left on earth since its creation, dominate in the wider environment such as in all types of soil, rocks, water, air, building materials, food and the human body [Wilson, W.F., 1994]. On the point of contribution to radiation dose received by mankind, the most important primordial radionuclides are uranium series, thorium series and ^{40}K [UNSCEAR, 1993]. Of our daily life concern, the concentration of NORM (Naturally Occurring Radioactive Materials) which is characterized by cosmogenic and primordial can be increased above the natural background levels through specific industrial activities such as waste products of sludge etc. [Bernard, Z. 1995], and higher utilization of computed tomography (CT) and nuclear medicine [NCRP, 2006].

Therefore, without these artificial sources our annual background doses do not change significantly. However, radioactive materials are subjected to ionizing radiation, which has sufficient energy to break some chemical bonds. Any living tissue in the human body can be damaged by ionizing radiation in a unique manner. The body attempts to repair the damage, but sometimes it is too widespread that the damage cannot be repaired and causes cancerous cells.

Natural sources contribute almost 80% of the collective radiation exposure of the world's population [Kaleel Mohammed Thabayneh, Mohanad Mohammed Jazzar, 2012]. Therefore,

great interest has been expressed worldwide for the study of naturally occurring radiation sources as well as environmental radioactivity has led to the interest in extensive surveys in many countries and in Ethiopia as well.

The nucleus and its collection of electrons find a lower energy state (need stability) and the excess energy is carried out by a photon (decay transition). In the nucleus, a multi particle collection of protons and neutrons held together by the strong interaction (nuclear force), whirling about themselves in bound configurations with stationary wave functions and quantized energy.

Both gravitational and Electromagnetic forces are infinite in range and their interaction strengths diminish with the square of the distance of separation. Clearly, nuclear force cannot follow the same radial dependence; else nucleons in one atom would have felt the attraction of those in nearby atoms. In fact nuclear force has a very short range, not much beyond the confine of the nucleus itself, in marked contrast to the fundamental forces that were familiar at the time.

In 1935, Yukawa proposed that the force between nucleons arises from meson exchange. This was the start of the concept of field quantum as the mediator of fundamental forces. The reason that nuclear force has a finite range comes from the nonzero rest mass of the mesons exchanged. In contrast, the field quantum for electromagnetic force is the mass less photon and for gravitational force, the graviton. The fundamental force responsible for nuclear properties is the strong interaction between quarks. Most of this interaction is restricted to between the quarks inside a nucleon with gluons as field quanta. However small ‘residue’ goes outside and gives us the interaction between nucleons.

The experimental information, both in Nuclear and Particle physics is derived from scattering measurement similar in principle to those that Ernest Rutherford and his collaborators performed in discovering the nucleus. In such experiments, beam of energetic particles area directed into a fixed target, or alternatively two beams of energetic particles are made to collide. In either case, the results of collisions in such scattering experiments provide invaluable, and often the only attainable information about sub-atomic systems.

Later, in 1920**,** the radii of a few heavy nuclei were measured by Chadwick and were found to be much smaller than the order of atomic radii. The experiments involve scattering α-particles, obtained from radioactive elements, off such heavy elements & copper, silver, and gold, and the measured cross sections were found to be different from values expected of the Rutherford formula for Coulomb scattering off point charges.

Nuclei are usually found in their individual ground states, by virtue of the fact that these are the lowest ones in energy. However, in the laboratory, and in the interior of stars, energy can be injected into nuclei to promote them to excited states. Besides energy, other properties for many of these states, such as electromagnetic moments and transition rates, can also be observed. In addition, β-decay, nucleon transfer, fission and fusion transform one nuclear species to another. The study of these quantities supplies us with information on the structure of nuclei.

In addition to its intrinsic values, nuclear structure can also provide us with the data on the nature of nuclei and the force acting on the system.

1.2Nuclear Decay Process

Radioactivity is a phenomenon related to unstable atomic nuclei with excess of energy and/or mass, which spontaneously decompose emitting ionizing radiation in the form of electromagnetic waves (gamma rays) or streams of subatomic (alpha, beta particles) [UNSCEAR., 2010]. The activity of a particular radioactive substance is characterized by the constant decay rate and the half- life ($t_{1/2}$-time taken for the activity of a given quantity of a radioactive substance to decay to half of its initial value), and it is a general rule of thumb that ten half- lives are required for each radioisotope to be eliminated [Murray R. U., 1989]. Since the half- lives of various nuclei vary from seconds to billions of years [ATSDR, 1999], the time required for their total decay significantly differ as well.

1.3 Naturally Occurring Radioactive Materials (NORMS)

NORM stands for Naturally Occurring Radioactive Material that can be found in nature. Since the earth was created, NORMs were formed in supernovae and the primary particles from our region of universe continually bombard the forming earth's crust.

NORM is an expression generally used to refer to terrestrial radionuclides like ^{40}K, ^{232}Th, ^{238}U and ^{235}U and their progeny decay products. These radionuclides are different from artificial radionuclides which do not exist in nature. This type of radionuclide called anthropogenic radionuclides or human made radionuclide. Also the term, TENORM refers to Technologically Enhanced Natural Occurring of Radioactive Material. The term is used to classify any process done by human contact causing more concentration of NORM on the earth's surface (e.g. Extraction of oil and gas) [IAEA, 2006].

NORM occurs everywhere in our life, it encircles us in the environment, for example in food, water, rocks, soil, sand, oil and gas industry, mining etc. Activities of human beings such as: using rocks, sands and cement in building materials or oil and gas extraction or phosphate industry, cause a distribution of NORM on the earth surface and lead to increases in the local activity concentration of natural radioactivity which can be significantly greater than the average background level. NORM radiation can be hazardous to human beings because of the direct emission of natural radionuclide that means all creatures are exposed to radiation during their daily lives [Wilson, W.F., 1994].

Naturally radionuclides come from the atmosphere as a result of radiation from outer space, earth's crust such as rocks mineral ores and soil, its emitted from both natural and human-made radionuclides and surrounds us at every time[HPS, 2015].
The natural radionuclides materials that have very long half-lives include ^{238}U, ^{235}U and ^{232}Th chains (terrestrial radionuclides) distribute widely on earth and ocean, they were already present when the earth was born about 4.5 billion years ago and each of these nuclides terminates in stable isotopes of (Pb) nuclide[Cember H and Johnson T E , 2009]. The other naturally radionuclides such as ^{40}K, ^{87}Rb, and ^{113}Cd are individual and the most important (^{40}K) a radioactive isotope with a long half-life (1.28×10^9 years) [Arthur R J, Miley H S and Lindsay C

BE-7, 2008]. It is widely distributed on earth and found in measurable quantities in many building materials [Cooper M B, 2005].

There are also natural radionuclides originated from interactions between the cosmic ray and the outer atmosphere such as ^{3}H, ^{14}C, ^{14}N, ^{81}Kr, ^{22}Na[Cember H and Johnson T E, 2009]. Man-made radionuclides, which are present in environment, have been created by human activities and added to the inventory of natural radionuclides for example ^{3}H, ^{131}I, ^{129}I, ^{137}Cs, ^{90}Sr and ^{239}Pu, in spite of the amount added is little compared to natural quantities. In 1996, IAEA estimated that 20% of doses contributions in the environment come from cosmic ray and man-made processes while 80% is from the natural radionuclides. The presence of the radionuclides depends on the geological and geographical conditions; therefore we can find different levels of radionuclides in the soil samples in different region in the world [UNSCEAR, 2000].

Natural radioactivity is composed of the cosmogenic and primordial radionuclides. Cosmogenic radionuclides, such as ^{3}H, ^{7}Be, ^{14}C, and ^{22}Na, are produced by the interaction of cosmic-ray particles (mainly high-energetic protons) in the earth's atmosphere. Primordial radionuclides (also called terrestrial background radiation) are formed by the process of nucleosynthesis in stars. Only those radionuclides with half-lives comparable to the age of the earth, and their decay products, can still be found today on earth, e.g. ^{40}K, and the radionuclides from the ^{238}U and ^{232}Th series. Gamma radiation from these radionuclides represents the main external source of irradiation of the human body.

The external radiation exposure arises mainly from cosmic rays and from terrestrial radionuclides occurring at trace levels in all soils. While absorbed dose rate in air from cosmic radiation outdoors at sea level is about 30 nGy h^{-1} for the southern hemisphere (UNSCEAR Report, 2000), the specific levels due to terrestrial background radiation are related to the types of rock from which the soils originate. Therefore, the natural environmental radiation mainly depends on geological and geographical conditions [Florou, H., Kritidis, P., 1992]. Higher radiation levels are associated with igneous rocks, such as granite, and lower levels with sedimentary rocks. There are exceptions, however, as some shales and phosphate rocks have relatively high content of radionuclides [UNSCEAR, 1993].

The high energy primary cosmic radiation induces various complex interactions with the earth's atmosphere, and results to cosmogenic radionuclides and hence produces secondary radiation [IAEA, 2003].

However, only a part of these radiations reach the earth's surface because the earth atmosphere acts as a filter and absorbs much of the energy of the cosmic rays. Primordial radionuclides which are left on earth since its creation, dominate in the wider environment such as in all types of soil, rocks, water, air, building materials, food and the human body [Wilson, W.F., 1994].

However, humankind can be exposed to radiation originating from artificially radioactive sources such as cesium-137 (^{137} Cs) present in the earth's environment as a result of nuclear weapon testing or nuclear fallout from nuclear technology [Ramasamy V, et al, 2009].

High terrestrial background radiations zones were generally attributed to local geology, location, altitude, and geochemical effects [UNSCEAR, 1993, Ramasamy V, et al, 2009]. Thus, the activity concentrations of radionuclides in granite locations were found to be higher than those of clay, sand stones, and limestone soils [Rafique M , et al ,2011, Khan H, et al., 2012]. Normally, the presence of NORMs in soil generally originates from the disintegrating rocks that are carried to soil by rain and flows [Taskin H, et al.,2009].

Naturally abundant radionuclides (^{226}Ra, ^{232}Th and ^{40}K) in the environment, and releases from fertilizers, agrochemicals, research and medical facilities form the bulk of radionuclides in ground and surface water, [Wisser, S., 2005]. Therefore presence of radioactivity in contaminated environment can be attributed to naturally occurring and artificially induced sources. Naturally occurring radioactivity are due to bedrock formations which are weathered, resulting in mineral leaching that leads to contamination, [Martin S, et al., 1995]. Artificial radioactivity is due to human activities, mainly as a result of agriculture, medicine, research as well as other activities like mining and milling of mineral ore which exposes the earth surface. All this contamination may have health effect; that poses great danger to human and other living organism in the biosphere. Study of natural radioactivity is usually done in order to gain information about the present levels of harmful pollutants that are discharged to the environment itself or in the living creatures [Samer J. Al-Kharouf, et al., 2008]. Radionuclides lead to production radiations, whereas radiation is known to trigger or induce cancer in living tissue.

In general, the main path origins of NORM are the primordial, cosmic rays and human made.

Human bodies are exposed externally to gamma rays as a result of radionuclide decay from naturally occurring radionuclides (such as: ^{238}U, ^{232}Th and ^{235}U) and their products and ^{40}K. These radionuclides occur in soil and building materials with different concentrations. The worldwide effective dose rate of public exposure due to radionuclides which occur in:

a) Soil (with weighted mean activity concentrations of 33, 45, and 420 Bq/kg) and;
b) For building material (50, 50, 500 Bq/kg) for ^{238}U, ^{232}Th, and ^{40}K respectively are given in (UNSCEAR 1999-2000).

In addition, human bodies are also internally exposed because of the existence of certain radionuclides in the body such as ^{40}K or through inhalation of ^{222}Rn. The worldwide average annual effective doses (mSv) from external sources (such as cosmic rays and terrestrial gamma rays) are 0.4 and 0.5 mSv respectively, with the typical range of 0.3-1 and 0.3-0.6 mSv respectively. From internal exposure, inhalation and ingestion the average values are 1.2 and 0.3 mSv respectively, with typical ranges of 0.2-10, 0.2-0.8 mSv respectively [UNSCEAR,2000].
The IAEA publication mentioned that if the activity concentration is less than 1 Bq/g there is no need for regulatory control and it is under exemption from regulatory requirement if the occupational exposure is less than 1 mSv/y [IAEA, 2004].

NORM as a phenomenon has been studied by different organizations, especially the International Atomic Energy Agency (IAEA) which organizes different conferences and technical meetings to study the effect of NORM on human beings to discuss the different methods to cover the environmental impact and the biological effects. NORM has been considered an important environmental issue that contributes to issuing various regulations in many countries in order to discuss the all consequences which arise from dealing with activities from included NORM, and it is disposal.

Natural radionuclides occur in different types of rocks such sedimentary, metamorphic and igneous rocks in the earth for example, ^{235}U, ^{238}U, and ^{232}Th and their radionuclide daughters which are called terrestrial radionuclides source. Such radionuclides have long half-lives about billions of years. Besides, their unstable daughter decay leads to a stable nuclide,^{206}Pb.

Radioactive decay series can be defined as group of isotopes representing various stage of radioactive decay in which heavier members of the group are transformed into successively lighter ones, the lighter being stable [Dictionary, 2008].

Most naturally occurring radioactive material obeys a chained series of transformation rather than decaying in a single step. During the decay the nuclei in the chain emit various types of radiation (usually either beta or alpha particle), until ending in a radioactively stable nuclide. Usually the concentration of parent isotopes decreases as the decay progresses. In the meantime, the concentration of their daughter products increases [European Nuclear Society Web add.].
In the Earth's crust, ^{238} U constitutes 99.27% of natural uranium by mass, in comparison, ^{235} U makes up only about 0.72% [Gilmore, 2008].

Terrestrial radionuclides are classified in three main nuclear chains;
The first is the ^{238}U chain, which represents 99% of uranium in nature and contains 14 radionuclides ^{238}U decays to ^{234}Th through alpha decay until it reaches the stable ^{206}Pb. All ^{238}U daughters have half-lives shorter than that of ^{238}U which means the daughter and the parent are in secular equilibrium.
The second nuclear chain is Actinium series starts with ^{235}U decay chain, which represent 0.72 % of Uranium in nature. The chain is shorter compared to the ^{238}U chain as it contains 12 radionuclides.
The last nuclear chains are from ^{232}Th. It represents 100% of thorium in nature. This series contains 10 radionuclides. ^{232}Th is a naturally occurring, radioactive element discovered in 1828 by the Swedish chemist Jons Jakob Berzelius. It is found in small amounts in most rocks and soils, where it is about three times more abundant than uranium. Soil commonly contains an average of around 6 parts per million (ppm) of this nuclide. The main pathways of exposure are ingestion and inhalation. It was found to be present in the highest concentrations in the pulmonary lymph nodes and lungs, indicating that the principal source of human exposure is inhalation of suspended soil particles.

Naturally occurring radionuclides of Th and U are significant contributors of ingestion dose and are present in the biotic system of plants, animals, soil, water and air. Distribution of these radionuclides in different parts of the plant depends on the chemical characteristics and several parameters of the plant and soil. Presence of radioactivity in plant organs has been reviewed by various workers [Markose, P. M.,1990, Jibiri,et al., 2007]

Finally, it is important to know that not all radionuclides in the chains emit characteristic gamma rays, only those radionuclides under lined shown in figure (1) are measurable by gamma spectrometry.

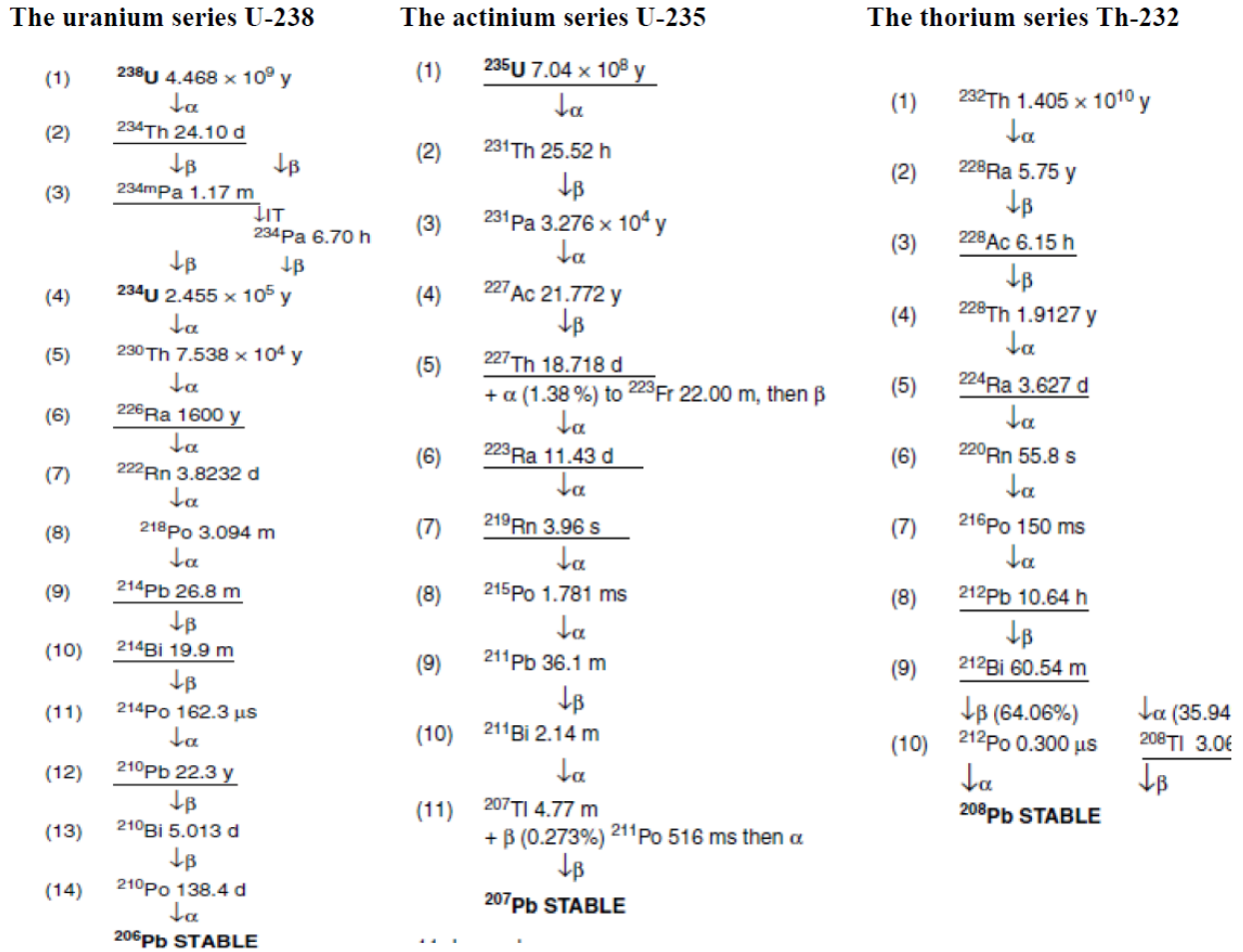


Figure 1: The decay scheme of three nuclear chains [Turner J, 2007**]**

As we see in the above figure, all series start with long half-life radionuclide and the radon member represents in each chains in a gas state. Finally all three chains end with stable lead isotope.

Non-series radionuclides such as ^{40}K also occur in the nature. ^{40}K (half-life $1.28x10^9$ years) is a natural radionuclide which exists in the environment and human body. It is widely distributed in the earth with low volume concentration ranging from 0.1% up to 3.5% .It occurs everywhere in plants, animal, even in the human body. The bones of an average age of a man and woman contain concentrations of approximately 20 mrems to tissues and 25 mrem to bones, the average annual radiation (the bones make up a concentration of almost 17mg of potassium which corresponds to activity 4.4kBq of ^{40}K) [Wilson, W.F.,1994].

1.4 Equilibrium in radioactive decay chains

The radioactive decay chain and the decay of parent radionuclide will lead to the production of progeny products. Accordingly, radiation is emitted. Hence, the progeny itself produces radiation during its decay. This happens continuously until it reaches to the stable state resulting in total collective activity. The contributions of the activity from the parent versus the progeny depend on both the parent and progeny half-life. It can be stated that equilibrium occurs when the quantity of the activity being produced is the same quantity of decay.

In a radioactive decay chain the parent nucleus (A) decays with a certain decay constant (λ_A), and while the parent decays, the daughter nucleus (B) concentration starts to growth to some point and then decay away with another decay constant (λ_B). The process continues until it reaches a final stable nuclide. Radioactive equilibrium takes place when each radioactive nuclide decays at the same rate of the parent nuclide. Thus,

$$A \xrightarrow{\lambda_A} B \xrightarrow{\lambda_B} C \quad \text{.....1}$$

From activity equation $A = \lambda N$, the activity of each nuclide is given by

$$A_A = \lambda_A N_A \quad \text{And} \quad A_B = \lambda_B N_B \quad \text{.....2}$$

There are different types of equilibrium that depend on the comparison between the parent and progeny's half-lives as follows:

For the sake of simplicity, assuming that the decay chain is only two steps and the daughter (B) is also radioactive, and the half-live of parent and daughter in equilibrium can be classified as:
When the half-life of the parent (A) is much larger than the half-life of the daughter product.
The daughter nuclide produces more radiation. After about 7 half-lives of the daughter (B), the mother and daughter activities become equal. This kind of equilibrium is called secular equilibrium. Therefore assuming that there are several succeeding generations of radioactive nuclear decay, we can generalize that:

$$\lambda_A N_A = \lambda_B N_B = ... = \lambda_n N_n \ldots\ldots\ldots\ldots\ldots\ldots\ldots 3$$

The equation is usually quoted as the ‘’ Bataeman equation in secular equilibrium’’.

When the half-life of the daughter product is longer or the same as the parent (A). As a result of the combined decay of both parent the (A) and daughter (B), the total activity increases and finally equilibrium is obtained. The total activity then decays at about the same rate of parent nuclide. This is called transient equilibrium [Radiation Protection, 2008**].**

1.5 Sources of Radioactive materials

Terrestrial Radiation

Radioactive material is found throughout nature. It occurs naturally in the soil, water, and vegetation. The major isotopes of concern for terrestrial radiation are uranium and the decay products of uranium, such as thorium, radium, and radon. Low levels of uranium, thorium, and their decay products are found everywhere. Some of these materials are ingested with food and water, while others, such as radon, are inhaled. The dose from terrestrial sources varies in different parts of the world. Locations with higher concentrations of uranium and thorium in their soil have higher dose levels [Pöschl,et al., 2007].

Cosmic Radiation

The high energy particles that flow into our solar system coming from far distance in the galaxy are another source of natural radionuclides which are called Cosmic rays. Cosmic rays are atomic particles coming from outer space. They are capable of penetrating the atmosphere due to their high energies. 90% of them are proton (hydrogen atom nucleus), 9% alpha particles and 1% electrons. When the cosmic rays reach the earth's atmosphere, they are called primary rays, and they are called secondary when they interact with it. Cosmic rays are one of the main natural radionuclides sources because it creates the radionuclide isotopic such as: H-3 (half-life 13.2 years), Be-7 (half-life 53.3 day), Be-10 (half-life $5.73*10^3$ years) Al-26 (half-life $7.1*10^5$ years), Cl-36 ((half-life $3.1*10^5$ years). As mentioned above, cosmic rays contain high energy particles such as alpha particles and neutron. Thus, during their interaction in the upper layer of the earth atmosphere with nitrogen and oxygen atoms and molecules, cosmic rays produce ^{14}C, ^{7}Be and other different radioisotopes. [Merril E., Thomas G., 1997].

The Earth, and all living things on it, is constantly bombarded by radiation from space, similar to a steady drizzle of rain. Charged particles from the sun and stars interact with the Earth’s atmosphere and magnetic field to produce a shower of radiation, typically beta and gamma radiation. The dose from cosmic radiation varies in different parts of the world due to differences in elevation and the effects of the Earth’s magnetic field [Pöschl,et al., 2007].

Anthropogenic

Anthropogenic radionuclides or human made radionuclide are produced by human being for different purposes. Also the term, TENORM refers to Technologically Enhanced Natural Occurring of Radioactive Material. The term is used to classify any process done by human contact causing more concentration of NORM on the earth's surface (e.g. Extraction of oil and gas) [IAEA,2006].

Finally, we have to talk about the man-made sources as another source of radionuclides in the earth crust. During the period 1945 to 1980, tests performed on nuclear weapons in the atmosphere added a lot of radionuclide to the environment. In addition, nuclear accidents such as Chernobyl contributed to distributing different radionuclides with both short and long half-life radionuclides. Consequently; human beings were irradiated either by inhalation or ingestion [UNSCEAR, 2000].

Artificial radionuclides such as ^{137}Cs, ^{90}Sr are classified as human made sources. Some activities of human beings lead to radiation exposure as they daily use radionuclides for different applications. In fact, this type of sources does not exist in nature, but it is produced due to different activities, mainly in nuclear power plant, nuclear weapons test or nuclear accidents as mentioned above. Artificial radionuclides occur in different concentration in the environment.
Moreover, they can be specified in environmental samples such as soil or food and so on [Pöschl,et al., 2007]. ^{137}Cs artificially occurs in nuclear sites when the fission products arise due to the use of nuclear weapons.
Two distinct groups are exposed to man-made radiation sources members of the public and occupationally exposed individuals.

Man-made radiation sources that result in an exposure to members of the public are: Tobacco, Televisions, Medical x-rays, Smoke detectors, Nuclear medicine and Building materials.

By far, the most significant source of man-made radiation exposure to the public is from medical procedures, such as diagnostic x-rays, nuclear medicine, and radiation therapy. Some of the major isotopes are I-131, Tc-99, Co-60, Ir-192, and Cs-137.In addition, members of the public are exposed to radiation from consumer products, such as tobacco (polonium-210), building

materials, combustible fuels (gas, coal, etc.), ophthalmic glass, televisions, luminous watches and dials (tritium), airport x-ray systems, smoke detectors (americium), road construction materials, electron tubes, fluorescent lamp starters, lantern mantles (thorium), etc.

Of lesser magnitude, members of the public are exposed to radiation from the nuclear fuel cycle, which includes the entire sequence from mining and milling of uranium to the disposal of the used (spent) fuel. These are uranium and its daughter products. The final sources of exposure to the public are shipment of radioactive materials and residual fallout from nuclear weapons testing and accidents, such as Chernobyl.

Occupationally exposed individuals are exposed according to their jobs and to the sources with which they work. The exposure of these individuals to radiation is carefully monitored with the use of tiny instruments called dosimeters. Some of the isotopes of concern are cobalt-60, cesium-137, americium-241, and others [Interaction of Radiation with matter, web add].

1.6 Statement of the Problem

Everyone on the planet is exposed to some background level of ionizing radiation that are either left over from the creation of the world (like uranium and radium) or made by interactions with cosmic radiation (like ^{14}C and Tritium). The major source of radioactivity in the upper atmosphere below the altitude of 60 km is cosmic rays, whereas the lower atmosphere near the ground (i.e. below 4 km) is dominated by terrestrial sources (Bazilevskaya et al.,2000).

Soil is the foundation of natural resource on which the life supporting system and socio-economic development depends. It provides food from the farming land, fodder and fuel for meeting the basic human and animal needs. Use of chemical fertilizers, especially phosphorus fertilizer, may lead to increased contamination of agricultural crops by enriching the soil with radioactive elements as well as chemical toxins.

So the knowledge of the concentrations and distributions of natural radionuclides, such as ^{40}K, ^{238}U, and ^{232}Th and their decay products and radiological associated risks in soils, plants, sediments, and so forth is useful for monitoring environmental radioactive contamination.

Coffee is one of the most popular and widely consumed beverages in the world and also in Ethiopia. Its consumption is increasing. The coffee trade has played a crucial role in their economic development. Coffee is important to the economy of Ethiopia; around 60% of foreign income comes from coffee, with an estimated 15 million of the population relying on some aspect of coffee production for their livelihood. Some naturally occurring radioisotopes and other elements present in soil are drawn into the roots of plants via ion channels or specific transporters. Their distribution throughout the plant tissues depends on their chemical characteristics and several parameters of soil and the plants themselves. The concentrations and distributions of natural radionuclides, such as ^{40}K, ^{238}U, and ^{232}Th and their decay products and radiological associated risks in soils and plants or coffee are useful for monitoring environmental radioactive contamination.

Lake Zuwaye serve many useful purposes by providing food (fish) for them, water for irrigation purposes, for transportation and for recreation to holiday makers. In the wake of all these uses, there is the need to regularly monitor the concentration of radionuclide present in this lake so as to know whether it poses health risk to society of produce from these areas or not, as high concentrations are injurious to humans, identify the pollution source and use the results obtained to predict levels of contamination in similar areas.

Such studies can be useful to estimate the degree of human risk associated with the ingestion of radionuclides in biota through the food chain and to establish a baseline database of radionuclides concentration, in order to monitor the possible variations in the marine environmental radioactivity due to nuclear industry and other human activities.

1.7 Objectives of the study

General Objective:

The aim of this study is to determine the level of radioactivity in soil, lake water and coffee in from selected parts of Ethiopia.

Specific objectives of the study:

a) To determine the activities of ^{238}U, ^{232}Th and ^{40}K in soil, Zuwaye lake water and four different Coffees using a Gamma Ray- spectrometer.
b) To calculate the health risk associated with the Zuwaye lake water, Coffee and soil from the selected area in Ethiopia.

1.8 Significant Of the Study

In developed countries several documents published on the natural radioactivity is giving awareness for their community. Such documents were not familiar for our country except some works that are performed by radiation protection agency and graduated students working in this area. The radioactivity has hazardous health effects upon human exposure. This study determined the concentration of radionuclide found in the study area and the radiological effects for the environment.

Chapter Two

2. Review of Related Literature

The total dose contribution in the form of natural sources comes from terrestrial gamma radionuclides (UNSCEAR, 2000). Only nuclides with half-lives comparable with the age of the earth or their corresponding decay products, existing in terrestrial materials, such as ^{40}K, ^{238}U and ^{232}Th radionuclides are of great interest. Abnormal occurrences of uranium and its decay products in rock and soil are the main sources of high natural background.

Observations on primordial natural radiation have received particular attention worldwide and led to extensive surveys in many counties [UNSCEAR, 2000]. They mainly serve as baseline data of natural radioactivity such that man made possible contaminations can be detected and quantitatively determined. They can be used to assess community dose rates and to perform epidemiological studies. The results obtained in each country can be exploited to enrich the world's radiation data map, which is highly needed for evaluating worldwide average values of radiometric and dosimetric quantities [Al-Jundi, 2002].

2.1 Radioactivity in the Environment

2.1.1 Natural radioactivity in the soil

Soil consists of mineral and organic matter, water and air arranged in a complicated physiochemical system that provides the mechanical footing for plants in addition to supplying their nutritive requirements [IAEA, 2006]. The inorganic portion of the surface soils may fall into a number of textural classes, depending on the percentage of sand, silt and clay. Sand consists largely of primary minerals such as quartz and has particle size ranging from 60 μm to about 2 mm. Silt consists of particles in the range of 2 to 60 μm, while clay particles are smaller than 2 μm in diameter [Van Wijngaarden L.B., et al, 2002].

Substantial amounts of radionuclides are found in the earth's crust. In fact the natural radioactivity is largely responsible for the fact that the interior of the earth is hot and molten [Van Rooyen T.J.,2002].
The term radiometric fingerprinting describes the identification of mineral species based on the difference in radionuclide concentrations [De Meijer R.J., et al., 1997].

Natural radioactive material in rocks and soil account for about 28 millirem or 8% of radiation dose a person receives in a year from all sources including medical exposure [M. I. Chowdhury, ,et al,. 2004].
The earth's crust contains small amount of Uranium, Thorium, and Radium as well as radioactive isotopes of several elements including Potassium. The radiation dose comes from the rocks, soil, and some building materials (such as bricks and concrete) [M. I. Chowdhury, ,et al,. 2004].

Exposure to natural sources of radiation has become an important issue in terms of radiological protection. Much of this comes from ^{222}Rn and its solids, short lived daughter products, ^{218}Po, ^{214}Pb, and ^{214}Bi. The National Radiological Protection Board, NRPB, (1992), estimates that Radon accounts for approximately 50 % of person's annual dose of radiation from all sources in most of the world.

Radon is a naturally occurring radioactive gas that may be emitted from any rock that contains Uranium and Thorium. The radionuclides and radon concentration for a given region is likely to be the result of combination of properties of the rocks and the soil, such as distribution of Uranium and Radium, porosity, permeability, and moisture content, as well as meteorological and seasonal variation. Naturally occurring radionuclides of terrestrial origin (also called primordial radionuclides) are present in various degrees in the environment, including the human body itself [NRPB, 1992].

Only those radionuclides with half-lives comparable to the age of the earth, and their decay products, exist in significant quantities in these materials. Irradiation of the human body from external sources is mainly by Gamma Radiation from radionuclides in the ^{238}U and ^{232}Th series and from ^{40}K. These radionuclides are also present in the body and irradiate the various organs

with Alpha and Beta particles**,** as well as Gamma Rays. Some other terrestrial radionuclides, including those of the ^{235}U series, ^{87}Rb, ^{138}La, ^{147}Sm, and ^{176}Lu, exist in nature but at such low levels that their contributions to the dose in humans are small. External exposures outdoors arise from terrestrial radionuclides present at trace levels in all soils [A. B. Tanner, 1980].

The specific levels are related to the types of rock from which the soils originate. Higher radiation levels are associated with igneous rocks, such as granite, and lower levels with sedimentary rocks. There are exceptions, however, as some shales and phosphate rocks have relatively high content of radionuclides. There have been many surveys to determine the background levels of radionuclides in soils, which can in turn be related to the absorbed dose rates in air. The latter can easily be measured directly, and these results provide an even more extensive evaluation of the background exposure levels in different countries. All of these spectrometric measurements indicate that the three components (^{238}U, ^{232}Th and ^{40}K) of the external radiation field, namely from the gamma-emitting radionuclides in the ^{238}U and ^{232}Th series and ^{40}K, make approximately equal contributions to the externally incident gamma radiation dose to individuals in typical situations both outdoors and indoors [A. B. Tanner, 1980].

The radionuclides in the Uranium and Thorium decay chains cannot be assumed to be in radioactive equilibrium. The isotopes ^{238}U and ^{234}U are in approximate equilibrium, as they are separated by two much shorter-lived nuclides, ^{234}Th and ^{234}Pa. The decay process itself may, however, allow some dissociation of the decay radionuclide from the source material, facilitating subsequent environmental transfer. Thus, ^{234}U may be somewhat deficient relative to ^{238}U in soils and enhanced in rivers and the sea [A. B. Tanner, 1980].

The radionuclide ^{226}Ra in this chain may have slightly different concentrations than ^{238}U, because separation may occur between its parent ^{230}Th and uranium and because radium has greater mobility in the environment. The decay products of ^{226}Ra include the gaseous element radon, which diffuses out of the soil, reducing the exposure rate from the ^{238}U series. The radon radionuclide in this series, ^{222}Rn has a half-life of only a few days, but it has two longer-lived decay products, ^{210}Pb and ^{210}Po, which are important in dose evaluations. For the ^{232}Th series,

similar considerations apply. The radionuclide ^{228}Ra has a sufficiently long half-life that may allow some separation from its parent [A. B. Tanner, 1980].

The gaseous element of the chain, ^{220}Rn, has a very short half-life and no long-lived decay products. The activity concentration of ^{40}K in soil is an order of magnitude higher than that of ^{238}U or ^{232}Th. In its first assessment of representative concentrations of these radionuclides in soil, in the UNSCEAR 1982 Report, the Committee suggested the values of 370, 25, and 25 Bq/kg for ^{40}K, ^{238}Uand ^{232}Th, respectively.

On the basis of the higher levels reported for China and the United Bq/kg in the UNSCEAR 1993 Report [Knoll, G. F,2000].

In gamma-ray spectroscopic analysis of collected soil samples, standard procedures often call to remove the soil moisture by oven drying. After drying and processing, the soil sample is transferred and sealed in a beaker or other container and sent to the gamma-ray counting laboratory to determine the radionuclide specific activities (Bq/kg), defined here as the radionuclide activity per unit mass of soil. When quantifying members of the natural uranium (^{238}U) decay series, the emanation of ^{222}Rn from the soil must be considered.

To avoid the effects of ^{222}Rn loss, most laboratory procedures require storage time of 21±3 d (day) for sealed samples before gamma-ray counting. Radon emanates naturally from soil. There are two mechanisms for ^{222}Rn emanation from soil described as recoil emanation and diffusion emanation. From the alpha decay of ^{226}Ra, the ^{222}Rn nucleus acquires 0.086 MeV (Mega Electron Volt) in recoil energy [D. A. Bossus, 1988].

The recoil emanation consists of those ^{222}Rn atoms which terminate their recoil paths in soil pores. The diffusion emanation accounts for those Rn atoms embedded in the soil particles which subsequently diffuse into soil pores. In addition to soil type, the ^{222}Rn transport and diffusion coefficient depend on several soil parameters moisture content. The emanation of ^{222}Rn is strongly dependent on temperature and moisture [M. E. Kitto, 1992].

Temperature has been documented to have a smaller effect on radon emanation than moisture content [E. Stranden, 1983]. In general, as moisture content increases, the radon emanation rate increases until maximum is reached near a soil moisture content of 25±5%.

2.1.2 Radioactivity in the Water

Most drinking water sources have very low levels of radioactive contaminants (radionuclides), levels low enough not to be considered a public health concern. Of the radionuclides that have been observed to occur in drinking water sources, most are naturally occurring.

However, contamination of drinking water sources by anthropogenic (human-made) nuclear materials also occurs. Naturally occurring radionuclides are found in the Earth's crust and are created in the upper atmosphere. For example, trace amounts of long-lived isotopes (e.g., uranium-238, which has a half-life of almost five billion years) have been present in earth's crust since the crust first formed. As these long-lived trace radionuclides decay, shorter-lived (more radioactive) daughter products are formed. Of particular concern are naturally occurring uranium and the naturally occurring radium isotopes, radium-226 and radium-228, which have been observed to accumulate to levels of concern in drinking water sources. Most of the naturally occurring radionuclides are alpha particle emitters (e.g., the uranium isotopes and radium-226), but naturally occurring beta particle emitters do occur (e.g., radium-228 and potassium-40) [BEIR V, 2000].

Certain rock types contain trace amounts of the radioactive isotopes of uranium, thorium, and/or actinium. As these parent rocks weather, the resulting clays and other aquifer-forming materials may become a source of naturally-occurring radionuclides to drinking water sources. Other naturally occurring radionuclides include tritium, a beta particle emitter, which forms in the upper atmosphere interactions between cosmic rays (nuclear particles coming from outer space) and the gases comprising the atmosphere. Tritium can be deposited from the atmosphere onto surface waters via rain or snow and can accumulate in ground water via seepage. Tritium is also formed from human activities, as described below.

Natural tritium tends not to occur at levels of concern, but contamination from human activities can result in relatively high levels.

The man-made radionuclides, which are primarily beta and photon emitters, are produced by any of number activities that involve the use of concentrated radioactive materials. These radioactive materials are used in various ways in the production of electricity, nuclear weapons, nuclear

medicines used in therapy and diagnosis, and various commercial products (such as televisions or smoke detectors), as well as in various through academic and government research activities. Release of man-made radionuclides to the environment, which may include drinking water sources, are primarily the result of improper waste storage, leaks or transportation accidents [BEIR V, 2000].

The source of naturally occurring radionuclides is the earth's crust. Among these radionuclides there exists three principal decay series originating from ^{238}U, ^{235}U and ^{232}Th. Being one of the most abundant sources of naturally occurring radioactivity, the ^{238}U series has been widely investigated. A schematic diagram of the uranium-238 decay series is given in figure-1, showing the members of this series, their half-lives and their decay modes. The levels of uranium in igneous rocks are typically in the range 0.01-10 PPM (Pulse per Minute) and often increase with Si content, and can in extreme cases exceed thousands of PPM [M. Ivanovich, & R. S. Harmon,,1982].

U-series nuclides are expected to be in secular equilibrium (i.e. having the same activity concentrations) in closed bedrock systems. Groundwater systems significant disequilibrium is observed, which indicates that in open systems, rock interactions induce significant elemental and isotopic fractionations [J. K. Osmond and J. B. Cowart, 1982].
High natural levels of alpha and Radon in Natural Waters radioactivity in groundwater are often associated with areas of granite bedrock [O. Xavier, V. Isabel & S. Isabel,1996].

Although the physical and chemical properties of these elements are well known, it is difficult to predict the behavior and mobilization in more complex natural system. The transfer or disposition of an atom created in this series into groundwater systems depends on many factors involving the physical and chemical properties of the element. In addition to that groundwater systems vary from region to region.

In general the rock porosity and permeability control the behavior of groundwater. For example igneous rocks, such as granite, have high permeability, resulting from fractures in the rocks that permit faster flow relative to, for example shale. The general properties of uranium, thorium, and

polonium are metallic while radium is an alkaline earth and radon is a gaseous element. Soluble uranium complexes, e.g. uranyl, are readily produced in oxidizing environments, whereas in reducing groundwater environments uranium, and especially the uranyl ion, is readily precipitated or adsorbed on organic compounds [B. De Vivo, et al., 1984].
Release of radium from rock to water is mainly due to alpha recoil [S. Hongbing & M. S. Thomas, 1998].

Radium dissolves readily in waters of low pH (e.g. newly formed groundwater) and its concentration is often related to concentrations of total dissolved solids, barium and sulphate. Radon concentration in groundwater is mainly controlled by the lithology [M. A. Misdaq & A. Elharti, 1997] of the aquifer. Radon enters the water phase to form a metastable clathrate-hydrate with water, (Rn.6H2O) Clathrates are chemical compounds consisting of lattice molecules and inclusions of smaller molecules within the crystal lattice, that traps molecules in its crystal structure without chemical bonding [Lawrence Stein, 1983].

As ^{222}Rn is the direct daughter of ^{226}Ra, a high level of ^{226}Ra in bedrock is expected to generate high levels of ^{222}Rn in water, which then migrates away from the source. Groundwater can carry ^{222}Rn over large distances depending on flow rate, e.g. at flow rates between 0.5-1.0 meter per day Rn could be transported about 20-40 m before it is completely decayed. High concentrations of ^{222}Rn could lead to detectable levels of both ^{210}Pb and ^{210}Po. In aquifers that have elevated ^{210}Po concentrations, normally ^{222}Rn levels are also elevated. However, due to absorption/ desorption processes, both the ^{210}Pb and ^{210}Po levels show large variations [Lawrence Stein, 1983].
In groundwater, as mentioned earlier in this chapter, uranium is not usually in equilibrium with its progeny. The reasons for this behavior are mainly due to the differences in the mobilization in groundwater and physical-chemical processes such as physical half-lives of these nuclides, their transport between rock and water systems and their water chemistry (e.g. absorption/desorption processes; ion exchange capacities) Therefore, in order to draw a complete picture about groundwater radioactivity that comes from uranium, it is necessary to investigate each nuclide involved [Lawrence Stein, 1983].

2.1.3 Radioactivity in the Air

Small amount of radon, a radioactive gas which comes from radioactive decay of uranium, seeps into the atmosphere from the soil. On average, inhalation of the radon in homes and other buildings accounts for 200 millirem per year. This about 55% of total radiation dose an individual receives in a year from all sources, including medical x-rays, cosmic rays, building materials, the earth's crust, and ingested radioactivity materials [B. A. Al-Bataina, et al., 1997].

Chapter Three

3. Methodology of the study

Since the discovery of the phenomenon of radioactivity about a century ago and particularly during the last half century when nuclear sciences and instrumentation were developed, nuclear analytical methods have provided an increasing potential for applications in various disciplines, not the least of which is in the life sciences.

Nuclear analytical methods from now on abbreviated NAMs-are based on properties of the nucleus of the atom, in contrast to nonnuclear methods, which are based on properties of the atom or molecule, where actually the electrons play the decisive role. In NAMs, the nuclei of isotopes (belonging to a given element) are distinguishable, and thus, nuclear methods are determining isotopes, rather than elements as a combination of isotopes.

Nuclear parameters that may serve as a base for NAMs are: spin and magnetic moment, mass, excited states and associated parameters, probability for a given nuclear reaction, and-only for radioactive nuclei-half-life, types and energies of the emitted radiation[J. J. M. de Goeij, 1994].

In 1895, the German physicist Wilhelm Roentgen identified penetrating radiation, which produced fluorescence, and which he named X-rays. In 1896 two months later, Henri Becquerel discovered that penetrating radiation, later classified as α, β and γ rays, were given off in the radioactive decay of uranium and thus opened a new field of study of radioactive substances and radiations they emit [Shapiro J., 1972].

Rutherford and Soddy were the first to suggest that radioactive atoms disintegrate into lighter structures as they emit radiation. This suggestion received powerful support with the discovery that the α-particle was just an ionized atom of the element helium. Many of the newly discovered elements were found in various fractions of uranium ores, and this, the heaviest naturally occurring element, was soon suspected to be the parent substance [Ralph E. Lapp & Howard L. Andrews,. 1972]. It is presently known that uranium consists naturally of a mixture of ^{238}U (99.27%), ^{235}U (0.72%), and ^{234}U (0.006%); thorium and potassium were also identified as the radioactive parents with some decay products.

Gamma ray is electromagnetic radiation produced by nuclear interactions. It is generally characterized as high energy radiation and short wavelengths within the electromagnetic spectrum. This high energy can cause serious damage when absorbed by living cells. Because of its deep penetration property, shielding of gamma ray requires large amounts of mass. Usually materials with a high atomic number and high density are used for better absorption [Knoll, 1989; Gilmore and Hemingway, 1995].

Gamma-ray spectrometry can be performed using different types of radiation. Ge-detectors combine high resolution with low background to an extent not achievable with thallium activated sodium iodide (NaI) detectors, despite the latter being widely employed for gamma-ray spectrometry. Calculating of the amount of radionuclide present requires knowledge of the efficiency of the detector in the counting geometry. Several methods of determining the efficiency in these unusual geometries have been developed over the years [Hult, 2007; Metzger et al., 2002].

Gamma ray spectrometry is an analytical method that allows the identification and quantification of gamma emitting isotopes in a variety of matrices. In one single measurement and with little sample preparation, gamma ray spectrometry allows you to detect several gamma emitting radio-nuclei in the sample. The measurement gives a spectrum of lines, the amplitude of which is proportional to the activity of the radionuclide and its position on the horizontal axis gives an idea on its energy.

A gamma-ray spectroscopy system used to determine the energy and intensity of the nuclear radiation. This system consists of a series of equipment, including a high resolution HpGe coaxial detector, high voltage power supply, preamplifier, and amplifier, multichannel analyzer (MCA) and a computer with specific software (PCA3). The process of detecting the nuclear radiation is that the electronic signal from the detector directly goes to preamplifier which changes particle charge pulses to voltage pulses. Next the voltage signals pass through an amplifier which provides the voltage gain to bring the millivolt preamplifier output to the range of a few volts. The amplifier has a linear response, so that the amount of energy deposited is proportional to the pulse height. The pulse height can then be displayed on a MCA. After that the

spectra of either a point source or environmental sample will ready to analyze by means of software associated with it (PCA3).

There are three methods for collecting radiation data while performing a survey. A direct measurement is obtained by placing the detector near or against the surface or in the media being surveyed and reading the radioactivity level directly. Scanning is an evaluation technique performed by moving a portable radiation detection instrument at a constant speed and distance above the surface to semi-quantitatively detect elevated areas of radiation. Sampling is the process of collecting a portion of an environmental medium as representative of the locally remaining medium. The collected portion of the medium is then analyzed to determine the radionuclide concentration [MARSSIM, August 2000]. From the method that rose above during our work we used the sampling method for the radiation measurements.

Sample collection and analysis is typically less representative of true radionuclide concentrations at a specific measurement location than performing a direct measurement. This is caused by the additional steps required in collecting and analyzing samples, such as sample collection, field sample preparation, laboratory sample preparation, and radiochemical analysis. However, direct measurement techniques with acceptable detection limits are not always available. When sampling is required as part of a survey design, it is critical that the sample collection procedures consider representativeness.

Samples should be collected and analyzed by qualified individuals using the appropriate equipment and procedures.

3.1 Soil Sample Collection and Preparation

Sample collection procedures are concerned mainly with ensuring that a sample is representative of the sample media, is large enough to provide sufficient material to achieve the desired detection limit, and is consistent with assumptions used to develop the conceptual site model.

The volume of soil collected should be specified in the sample collection procedure. In general, large volumes of soil are more representative than small volumes of soil. In addition, large

samples provide sufficient sample to ensure that required detection limits can be achieved and that sample reanalysis can be done if there is a problem. However, large samples may cause problems with shipping, storage, and disposal. All of these issues should be discussed with the sample collection team and the analytical laboratory during development of sample collection procedures. In general, surface soil samples range in size from 100 g up to several kilograms.

The sample collection procedure should also make clear if it is more important to meet the volume requirement of the survey design or the surface area the sample represents. Constant volume is related to comparability of the results while surface area is more closely related to the representativeness of the results. Maintaining a constant surface area and depth for samples collected for a particular survey can eliminate problems associated with different depth profiles. The actual surface area included as part of the sample may be important for estimating the probability of locating areas of elevated concentration.

The material present in the field at the sample location may or may not provide a representative sample. Vegetative cover, soil particle size distribution, inaccessibility, or lacks of sample material are examples of problems that may be identified during sample collection. All deviations from the survey design as documented in the Standard Operating Procedures (SOPs) should be recorded as part of the field sample documentation.

Sample content is generally defined by the assumptions used to develop the conceptual site model. A typical agricultural scenario assumes that the top few centimeters of soil are available for resuspension in air, that the top 15 cm (6 in.) are homogenized by agricultural activities (*e.g.*, plowing), that roots can extend down several meters to obtain water and nutrients depending on the plant, and that external exposure is based on an assumed thickness of contaminated soil (usually at the surface). Depending on the dominant exposure pathways for each radionuclide, this can result in a complicated set of instructions for collecting representative samples. This situation can be further complicated by the fact that the site is not currently being used for agricultural purposes. For this situation it is necessary to look at the analytical results from the preliminary surveys (*i.e.*, scoping, characterization, remedial action support) to determine the expected depth of contamination.

In most situations the vegetative cover is not considered part of the surface soil sample and is removed in the field. For agricultural scenarios where external exposure is not the primary concern, soil particles greater than 2 mm (0.08 in.) are generally not considered as part of the sample (EPA 1990). Foreign material (*e.g.*, plant roots, glass, metal, or concrete) is also generally not considered part of the sample, but should be reviewed on a site-specific basis. It is important that the sample collection procedure clearly indicate what is and what is not considered part of the sample[MARSSIM, August 2000].

A total of 12 soil samples near Ajje, Asasa, shashemene, Zuwaye, Toga and Bishan Guracha were collected randomly from sites for the measurement of the specific radioactivity of ^{226}Ra, ^{232}Th and ^{40}K. Unwanted material (such as plant leave, roots and rocks) were isolatedfrom the collected samples and the samples dried at a temperature of 105° C for 10 h, crushed and sieved through a 200 mesh. The dried samples were transferred to polyethylene Marinelli beakers. Each deposit sample was left for at least one month to reach secular equilibrium between radium and thorium, and their progenies [Dabayneh K, 2008].

Figure 2: Sample preparation

3.2 Coffee Sample Collection and Preparation

Four raw coffee samples were purchased at markets in Shashamene, Ethiopia. Each was roasted and grinded and weighing. The samples were then transferred to polyethylene Marinelli type beakers of known weight were hermetically sealed with an insulating tape to impede contact with air moisture say labeled, and packed into radon-impermeable plastic containers to prevent radon gas escape as much as possible. The samples were stored and kept for a period of 1 month to attain secular radioactive equilibrium among ^{226}Ra, ^{232}Th, and their respective short-lived decay products ^{226}Ra and its decay products in the uranium series and ^{228}Ra and its decay products in the thorium series [Kurnaz A,2007, Samad MA,2012]. Finally, each Marinelli container was analyzed using a HPGe detector.

3.2.1 Study area

Lake Ziway (Oromo: Lake Dambal) is one of the freshwater Rift Valley lakes of Ethiopia. It is located about 100 miles south of Addis Ababa[Leslau, Wolf, 1999], on the border between the Regions of Oromia Region and of the Southern Nations, Nationalities, and Peoples' Region; the woredas holding the lake's shoreline are Adami Tullu and Jido Kombolcha, Dugda, and Ziway Dugda. The town of Batu lies on the lake's western shore. The lake is fed primarily by two rivers, the Meki from the west and the Katar from the east, and is drained by the Bulbar which empties into Lake Abijatta. The lake's catchment has an area of 7025 square kilometers.[Robert Mepham , 1992]

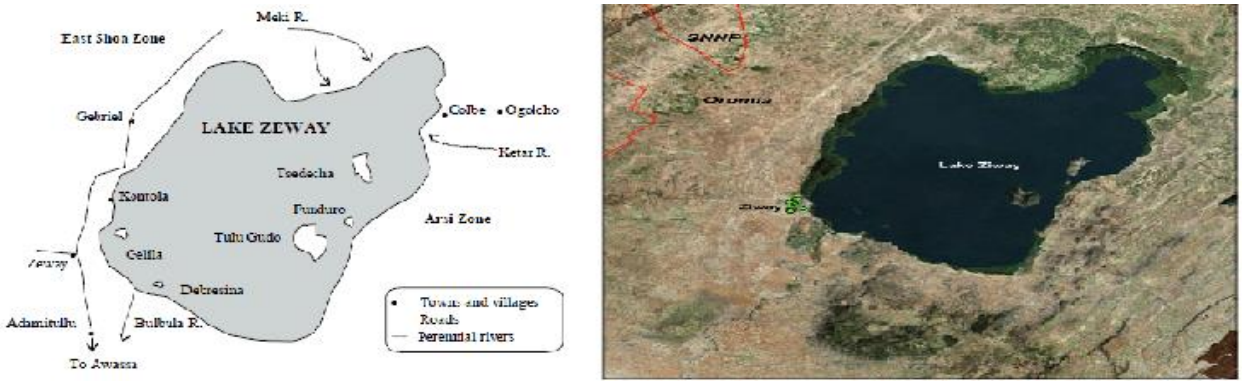


Figure 3: Lake Zuwaye

Lake Ziway is 31 kilometers long and 20 km wide, with a surface area of 440 square kilometers. It has a maximum depth of 9 meters and is at an elevation of 1,636 meters.[Water Resources and Irrigation Development in Ethiopia – IWM, 2011][Google Earth] According to the Statistical Abstract of Ethiopia for 1967/68, Lake Ziway is 25 kilometers long and 20 km wide, with a surface area of 434 square kilometers. It has a maximum depth of 4 meters and is at an elevation of 1,846 meters[Climate, National Statistics,2008]. It contains five islands, including Debre Sina, Galila, Bird Island and Tulu Gudo, which is home to a monastery said to have housed the Ark of the Covenant around the ninth century.

3.3 Lake Water Sample Collection and Sample Processing

Surface water samples of 1.5 L were taken from the middle of the lake and three various areas of the lake into clean plastic bottles washed with tap water and dried to avoid contamination or adsorption of the analyses present in the samples. A few drops of HCl were added to the samples to keep the analyses dissolved in solution and also prevent the radionuclides from adhering to the walls of the containers. The bottles were filled to the brim without any headspace to prevent trapping of gas. The bottles were covered tightly with their lids, labeled appropriately and transported to the laboratory.

In the laboratory, the water samples were filled into 1/2L Marinelli beakers with no special treatment. The samples in the Marinelli beakers were sealed hermetically to maintain a radioactive equilibrium between ^{226}Ra and its short-lived daughters, weighed and stored for a 1 month period. The radionuclides in the samples were counted using a high purity germanium detector of Ethiopian Radiation Protection Authority.

3.4 Sample Counting

The results of analysis of activity concentration of ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs radionuclides in soil samples for different locations of the study area are presented in table 1 and table 2.

The activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in the soil, coffee and Zuwaye lake water samples were determined by using gamma spectrometry coupled with aHPGe detector (Ortec) with a 70% relative efficiency and a resolution 1.9 keV for the 1332.5 keV^{60}Co gamma line and MCA with 2000 channel.

The system was calibrated for energy calibration by using sources from ^{139}Ce (166 keV), ^{113}Sn (392 keV), ^{85}Sr (514 keV), ^{137}Cs (662 keV), ^{88}Y (898 and 1836 keV) and ^{60}Co (1173 and 1332 keV) in the energy range (166 - 1850) keV [K. M. Dabayneh et al., 2008].

The background radiation and the samples were counted 36,000s. The gamma ray lines 295.21 and 351.92 keV of ^{214}Pb and 609.31, 1120.29 and 1764.49 keV of ^{214}Bi were used to determine the ^{238}U activity concentration. The activity concentration for ^{232}Th was determined using gamma lines 238.63 keV of ^{212}Pb, 911.21 and 968.97 keV of ^{228}Ac. The activity concentration for ^{40}K and ^{137}Cs were determined directly from the 1460.8 and 661.6 keV gamma lines, respectively. The net count rate photo peaks of all radio- nuclides daughter peaks were calculated by subtracting the respective count rate from the background spectrum obtained for the same counting time. Then the activity concentration of radionuclide was calculated from the background subtracted area prominent gamma ray energies [El-Shershaby A et al., 2006].

3.5 Theoretical Calculations

3.5.1 The Activity Concentration

The activity concentrations of the radionuclides in the measured samples were calculated using the following equation:

$$C = \frac{C_a}{I x \varepsilon_{ff} x M_s} \quad \text{.....4}$$

Where Ca is the net gamma counting rate (counts/second), ε_{ff} the detector efficiency of the specific γ-ray, I is the intensity of the γ-line in a radionuclide and M_s is the mass of the sample (kg).

In order to determine whether there is any radioactivity concentration in a certain sample, Curie first introduced a binary (Yes or No) decision of the presence of activity in an unknown sample. There is a certain minimum detectable activity to be measured in any radiation measurement especially in low level radioactivity in environmental sample. To perform this comparison, the net counts of a sample to the critical level (Lc) are required for a given sample.

The critical level (Lc) could be assumed at zero when there is no statistical fluctuation and other instrumental variation. Since radioactivity is the statistical process fluctuations exist to some degree in any counting measurement. Furthermore, there is a critical issue to make the decision of choosing Lc. The probability of a false positive is decreased for a high value of Lc, while by contrast, the likelihood of false negative is decreased for a low value of Lc.

In the case of no activity present, if the distribution of counting measurements obey a Gaussian distribution and only a statistical fluctuation from counting statistics are considered, the critical level is given by:

$$L_c = 2.326\sigma_{N_B} \text{.....5}$$

Where σ_{N_B} is the standard deviation of Gaussian distribution for the number of counts in the background. Therefore, any conclusions will be a false positive and the probability will be no larger than 5% [Knoll, G. F,2000].

Conversely, in the case of activity present it is important to introduce the Minimum Detectable Activity (MDA) is used to gain the actual sample count. It depends on the lower limit of detection and the counting efficiency of a counting system [Cember H. & E. Johnson, 2009]. MDA which can be found in the background spectrum and can be determined from the expression:

$$MDA = \frac{\sigma\sqrt{N_B}}{I_{(\gamma)}\varepsilon MT} \text{.....6}$$

Where σ is the statistical coverage factor equal to 1.645 with 95% confidence level and N_B is the background count under a specific photo peak energy [El Afifi E.M., 2006].

In fact five corrections factor are necessary to add to activity concentration eqn. which are characterized by the parameters K_1,K_2,K_3, K_4 and K_5.

K_1corresponds to the nuclide decay from the time that the sample was collected to the start of the measurement and can be calculated using:

$$K_1 = \exp\left(-\frac{\ln 2.\Delta t}{T_{1/2}}\right).....7$$

Where Δt is the lapsed time between the sample collection and the beginning of the measurement and $T_{1/2}$ is the radioactive nuclide half life.

K_2 is the correction factor which accounts for the decay of the nuclide during the measurement. This is given by:

$$K_2 = \frac{T_{1/2}}{\ln 2.t_r}\exp\left(1-\frac{\ln 2.t_r}{T_{1/2}}\right).....8$$

Where t_r is the elapsed real clock time during the measurement.

The correction for self-attenuation in the sample is taken in to account by the factory K_3. This is equal to the ratio of the full energy peak efficiency $\varepsilon(\mu, E)$ for a sample with the linear attenuation coefficient (μ) and the full energy peak $\varepsilon(\mu_{ref}, E)$ for a sample. The linear attenuation coefficient (μ_{ref}) is equal to unity it the matrix of both the calibration sample and measured sample are the same. However it can be investigated through the expression:

$$K_3 = \frac{\varepsilon(\mu, E)}{\varepsilon(\mu_{ref}, E)}.....9$$

In the pulse sampling cycle, when two or more photons enter the detector and are converted to signals at the same time, the sum of these two energies appear in the spectrum rather than two separate signals. Consequently, there is a net reduction in count rate or efficiency of the detector. To account for this, the correction factor K_4 is introduced which corresponds for pulse loss due to random summing phenomenon:

$$K_4 = \exp(-2R\tau).....10$$

Where R, τ are the resolution time and the mean count rate respectively. In for low count rates this factor could be taken as unity.

The energy level decay scheme for those nuclides decaying through a cascade of successive photon emissions, the sample geometry and composition and the detector parameters affect the final correction factor, K_5. If there is no cascade emission, this factor is equal to one [Dovlet, C. and Povierec, 2004].

Therefore the activity concentration becomes:

$$A = \frac{N}{I_{(\gamma)} \varepsilon M T K_1 K_2 K_3 K_4 K_5} \quad \text{.....11}$$

3.5.2 Radiological Effects

The major purpose of assessing radioactivity is to predict the possible radiation dose to be transmitted to living organisms. Exposure to radiation can be explained by various parameters [Beretka, J. and P. J Mathew, 1985]

3.5.2.1 The Radium Equivalent Activity (Raeq)

For the purpose of comparing the radiological effect or activity concentration of materials that contain ^{226}Ra, ^{232}Th and ^{40}K by a single quantity, which takes into account the radiation hazards associated with them, a common index termed the radium equivalent activity (Raeq) is used. This activity index provides a useful guideline in regulating the safety standards on radiation protection for the general public residing in the area under investigation. The Raeq index represents a weighted sum of activities of the above mentioned natural radionuclides and is based on the estimation that 1 Bq/kg of ^{226}Ra, 0.7 Bq/kg of ^{232}Th, and 13 Bq/kg of ^{40}K produces the same gamma radiation dose rates. The index is given as:

$$Ra_{eq} = C_{Ra} + 1.43 C_{Th} + 0.077 C_K \quad \text{.....12}$$

Where C_{Ra}, C_{Th} and C_K are the average activity concentration in the sample in Bq/kg of ^{226}Ra, ^{232}Th, and ^{40}K respectively [K. M. Dabayneh, et al, 2008].

3.5.2.2 Absorbed dose rate in air

The effects of gamma radioactivity originating from radioactive sources in the environment are generally expressed in terms of the total gamma radiation absorbed dose rate in air, Dr. The values of Dr in air and 1 m above the ground level are calculated from the measured activity concentrations of ^{238} U, ^{232}Th, and ^{40}K radionuclides using the following semi empirical formula [El-Shershaby A, et al., 2006].

$$D_r(nGyh^{-1}) = 0.427xAU + 0.662xATh + 0.043AK \quad \text{.....13}$$

Eq. (13) was modified to include the contributions of artificial radionuclides of cesium, (^{137} Cs) , as well as cosmic radiation via the following equation [El-Shershaby A, et al., 2006].

$$D_r(nGyh^{-1}) = 0.427xAu + 0.662xATh + 0.043xAK + 0.03XACs + 34 \quad \text{.....14}$$

Here 0.427, 0.662, and 0.043 are the dose rate conversion factors to convert the activity concentrations of ^{238} U, ^{232}Th, and ^{40}K and ^{137} Cs radionuclides into absorbed dose rates as proposed by UNSCEAR [Florou, H., Kritidis, P., 1992.]. Basically, these factors are representative of the absorbed dose rates in air per unit activity per unit of soil mass, in units of nGy h^{-1} per Bq kg^{-1}.

3.5.2.3 Annual effective dose equivalent

The annual effective dose equivalent (AEDE) received by individuals was calculated from the calculated values of Dr by applying the dose rate conversion factor of 0.7 Sv Gy^{-1} and the occupancy factors of 0.2 (5/24) and 0.8 (19/24) for outdoors and indoors, respectively. The annual effective outdoor doses, Dout; the annual effective indoor doses, Din ; and total annual effective doses, Dtot, were calculated according to the following equations [Veiga R, et al., 2006].

$$D_{out}(mSvy^{-1}) = D_r(mGyh^{-1})x24hx365.25dx0.2x0.7SvGy^{-1}x10^{-6} \quad \text{.....15}$$

$$D_{in}(mSvy^{-1}) = D_r(mGyh^{-1})x24hx365.25dx1.4x0.8x0.7SvGy^{-1}x10^{-6} \quad \text{.....16}$$

$$D_{tot}(mSvy^{-1}) = D_{out} + D_{in} \quad \text{.....17}$$

3.5.2.4 The External and Internal Hazard Index

The external (Hex) and internal (Hin) hazard index due to the emitted γ-rays of the soil samples were calculated and examined according to the following criterion:

$$H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \leq 1 \text{.....18}$$

And

$$H_{in} = \frac{C_{Ra}}{185} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \text{.....19}$$

The value of H_{ex}must be lower than unity in order to keep the radiation hazard insignificant. This is the radiation exposure due to the radioactivity from a construction material, limited to 1.5 $mGy \cdot y^{-1}$. The maximum values of Hex equal to unity correspond to the upper limit of Raeq(370 $Bq \cdot kg^{-1}$) [Veiga R, et al., 2006].

3.5.2.5 Radioactivity Level Index

The radioactivity level index, I_γ, is generally used to assess the hazardous level of radionuclide in the human body when exposed to an amount of external (indoor or outdoor) annual effective doses of γ-radiations decayed from radioactive nuclide in the soil. This index is very important for quality control of γ-radiation annual effective dose and in monitoring radiation inside human body, to ensure that such radiation does not exceed the worldwide permissible high dose values [Bernard, Z., 1995]. Values of I_γ can be calculated according to the semi empirical formula [K. M. Dabayneh, 2008, Shams I, 2013].

$$I_\gamma = \frac{C_{Ra}}{150 Bq/Kg} + \frac{C_{Th}}{100 Bq/Kg} + \frac{C_k}{1500 Bq/Kg} \text{.....20}$$

The assessed values of I_γ must be less than or equal to 1 to make sure the soil environment is generally safe or hazard free.

3.6 Experimental set up of Gamma Ray spectroscopy

3.6.1Semiconductor detectors

The detector is the center piece of the gamma spectroscopy system. The gamma photons interact with the detection material and transfer their energies to electrons or to positrons in the case of annihilation. These produced particles loose their energy within the detector, creating ionized atoms and ion pairs. These secondary entities form the basis of the detector signal. High purity Germanium is the mostly used material for gamma ray spectrometry systems.

For the past 20 years, Ge detector performance has been specified using the methods and values specified in "IEEE Standard Test Procedures for Germanium Gamma-Ray Detectors", ANSI/IEEE Std 325-1986.
These specifications are the FWHM, FW.1M, FW.02M, Peak-to-Compton ratio, and relative efficiency. These standard specifications are useful in guiding the user to an appropriate detector choice for the intended measurement.

Germanium detectors are semiconductor diodes having a P-I-N structure in which the Intrinsic (I) region is sensitive to ionizing radiation, particularly X-rays and gamma rays. Under reverse bias, an electric field extends across the intrinsic or depleted region. When photons interact with the material within the depleted volume of a detector, charge carriers (holes and electrons) are produced and are swept by the electric field to the P and N electrodes. This charge, which is in proportion to the energy deposited in the detector by the incoming photon, is converted into a voltage pulse by an integral charge sensitive preamplifier.

A radioactive source may emit gamma rays. The energy and the intensity of this radiation can be determined by using Gamma Ray Spectrometer. Gamma ray spectroscopy can be performed with either scintillation or semiconductor detectors.

Gamma ray is electromagnetic radiation produced by nuclear interactions. It is generally characterized as high energy radiation and short wavelengths within the electromagnetic spectrum. This high energy can cause serious damage when absorbed by living cells. Because of

its deep penetration property, shielding of gamma ray requires large amounts of mass. Usually materials with a high atomic number and high density are used for better absorption [Knoll, 1989; Gilmore and Hemingway, 1995].

Gamma-ray spectrometry can be performed using different types of radiation detector. Ge-detectors combine high resolution with low background to an extent not achievable with thallium activated sodium iodide (NaI) detectors, despite the latter being widely employed for gamma-ray spectrometry. Calculating of the amount of radionuclide present requires knowledge of the efficiency of the detector in the counting geometry. Several methods of determining the efficiency in these unusual geometries have been developed over the years [Hult, 2007; Metzger et al., 2002].

Semiconductor material is a material with an electronic band structure such as the band gap that provides limited conductivity [Gilmore, G.R., 2008]. It is a solid crystalline material that has an electrical conductivity between an insulator and a good conductor. It has narrow gap between the valance and conduction bands. A typical value for this band gap in germanium is about 0.7ev. This means that a small amount of energy required exciting an electron into the conduction band whereas in the valance band hole is created. The electrical conductivity of semiconductor is therefore changed when a semiconductor is exposed to radiation.

Semiconductor detectors are able to measure radiation with the aid of number of charged carriers. They are free to move in the detector, which is located between two electrodes. The ionizing emissions produce free electrons and holes. The amount of electron hole pair is proportional to the energy transmitted of the semiconductors' emission. Consequently, the amount of electrons from the valence band for conducting the strip, and an equal quantity of vacancies are created in valence band. Under the control of electric field, electrons and holes move to the electrodes where the pulse can be measured in an outer circuit; accordingly. The hole moves to the opposite direction and can be measured. It does not depend on radiant energy incident since the amount of energies required for creating an electron-hole pair is already known. The measurement of the amount of electron-hole pairs allows the radiant energy of

incident to be detected. The energy is necessary in order to produce electron-hole pair. It is very low comparing with the energy necessary to produce ion pair in the gas of the detector.

Therefore, the semiconductor detectors have many characteristic such as high resolution. Comparing with ionizing gas detector, the semiconductor detector has high density and the charged particles of the high energy can give its energies in the semiconductor of relatively small sizes [Knoll, G. F,2000].

High purity germanium (HPGe) detector is classified as a semiconductor detector. The detector was used to measure the activity concentration in the samples which were fully analyzed using a high-resolution, low background gamma-ray spectrometry system, based on a coaxial hyper-pure germanium detector (HPGe). The detector housing with a 10 cm of lead is an ideal thickness. It is usually used for the surface of the shield to reduce the surrounding radiation such as the nature radionuclide in building materials like K-40, uranium decay series and cosmic rays. Add to that, 3 mm thick layer of copper is used to cover the bulk shield of lead, which normally used to absorb the characteristic x-ray resulting from the interaction of radiation with lead. Figure (below) shows the main shield design feature in some HPGe detector.

High purity germanium detectors having high-resolution gamma-ray spectrometry has been widely used in qualitative and quantitative determination of gamma-ray emitting radionuclides in environmental samples. This is a non- destructive technique which does not require any rigorous sample preparation. It works on the principle that gamma ray photons have discrete energies which are characteristic of the emitting source. Therefore by measuring the energies of gamma ray photons in samples, the source of radiation can be accurately determined.
The great advantage to using a germanium detector is the fact that they have excellent energy resolution for gamma-ray spectroscopy. A high energy resolution means that the detector can discriminate between gamma-rays with similar energies. The more resolution a detector has, the more defined a gamma spectrum becomes.

However, often times some radiations which do not originate from the samples are detected in the gamma spectrometry. These radiations are called background radiation and are subtracted from the sample measurement during data processing. ^{40}K, ^{232}Th, ^{238}U and ^{235}U which are

radioisotopes of high-energy gamma ray of sufficient intensity are used for gamma ray mapping. High purity germanium detector is highly preferred to the other detectors because of its high resolution and sensitivity which is unmatched by any of the other detectors including NaI(Tl) detector [Hosseini, 2007].

Any HPGe detector requires a supply of liquid nitrogen to keep it at a very lowtemperature. Because germanium has a relatively low band gap, these detectorsmust be cooled in order to reduce the thermal generation of charge carriers (thusreverse leakage current) to an acceptable level. Otherwise, leakage currentinduced noise destroys the energy resolution of the detector. Liquid nitrogen,which has a temperature of 77°K or –196 °C is the common cooling medium forsuch detectors. The detector is mounted in a vacuum chamber which is attachedto or inserted into an LN2 Dewar or an electrically powered cooler. The sensitivedetector surfaces are thus protected from moisture and condensablecontaminants. The liquid nitrogen is kept in a large cryostat that can containsufficient quantity of the liquid to last at least a week between fillings (NOTE: Adetector face that is cold and wet is a sure sign that something is wrong with the vacuum system [Nafaa Reguigui , 2006].

The detector is connected to a preamplifier, a shaping amplifier and high voltage power supply. They are electronic instruments converting the event energy into a pulse height spectrum. A multi-channel analyzer (MCA) card of 8196 channels is installed in the computer for data acquisition.

A computer is required most of the times in order to visualize the spectrum and perform basic spectrum analysis using spectrum analysis software. Figure below shows a block diagram for a basic gamma spectrometry system.

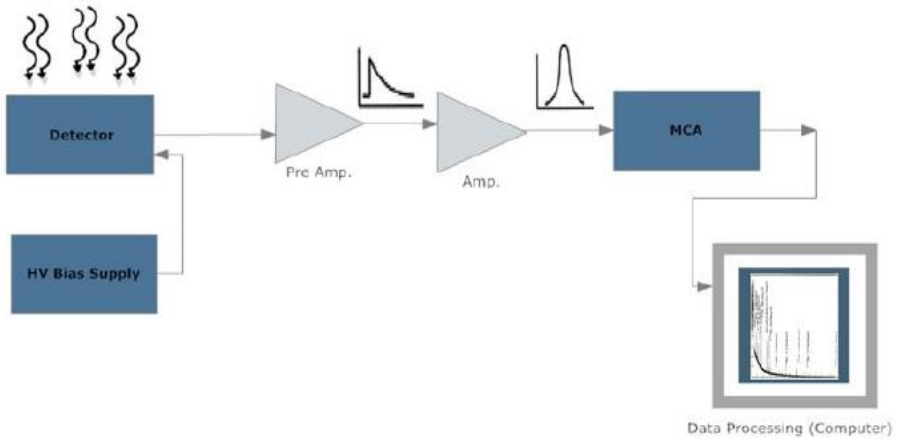


Figure 4: Block diagram of the equipments set up of high-purity germanium detector [Huda Al-Sulaiti, 2009].

3.6.2 Detector Characterization

3.6.2.1 Energy Calibration

In gamma-ray spectrometry with a HpGe detector which is operated using PCA3 software, the pulse heights are represented by channel numbers. Therefore, an energy calibration should be performed. The aim of energy calibration is to obtain a relationship between pulse heights (i.e peak position in the spectrum) and the corresponding gamma-ray energy. Energy calibrations were performed by measuring the spectrum of source emitted gamma-rays of precisely known energy and covering a wide range of energies. In other words, it was required to use a standard source such as ^{152}Eu which covers low and high energies. It is important to perform this calibration regularly during the measurement of low activity environmental sample and for long enough time to avoid any shift in the spectrum and achieve a good statistical precision for the peaks used for the calibration.

The detector calibration was performed using a certified standard reference mixed material ^{139}Ce (166 keV), ^{113}Sn (392 keV), ^{85}Sr (514 keV), ^{137}Cs (662 keV), ^{88}Y (898 and 1836 keV) and ^{60}Co (1173 and 1332 keV) in the energy range (166 - 1850) keV [De Meijer R.J.,1997]. The channel number and associated peaks were recorded. The graph between channel number and the energy was plotted.

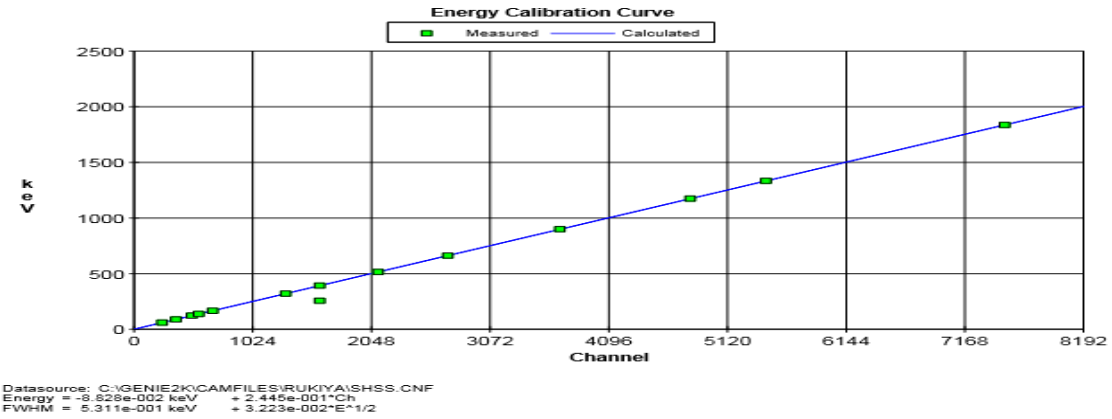


Figure 5: Energy calibration relation between channel numbers and corresponds to energy.

From the energy calibration figure the relation between channel and energy was linear.

3.6.2.2 Energy versus Efficiency Curve

One of the important of characteristic of a detector is its efficiency. In gamma-ray spectrometry the intension is to find the relation between peak area and the amount of activity present in the sample or the source. To obtain this, the absolute photo-peak efficiency should be considered. In general, considering how many quanta of radiation which come from radioactive sources compared to how many quanta reach the detector, the efficiency can be defined as the ability of the detector to measure how many pulses occur for a certain number of gamma rays emitted.

Efficiency Calibration is the ratio between the number of counts in the full energy peak and number of radiation incident on the detector [Huda Al-Sulaiti, 2009]. Two types of efficiency can be determined

a. Absolute Photo Peak efficiency

b. Intrinsic Photo Peak Efficiency

In this study the relative efficiency was considered.

The Energy efficiency calibration curve beyond 1850 keV was constructed using different energy peaks of ^{226}R in order to cover the range from 60 up to 2000 keV [K. M. Dabayneh,et al.,2008].

Generally, the sensitivity of a HPGe system will be in direct proportion to the detector efficiency.

The graph between efficiency and the energy was plotted as shown below.

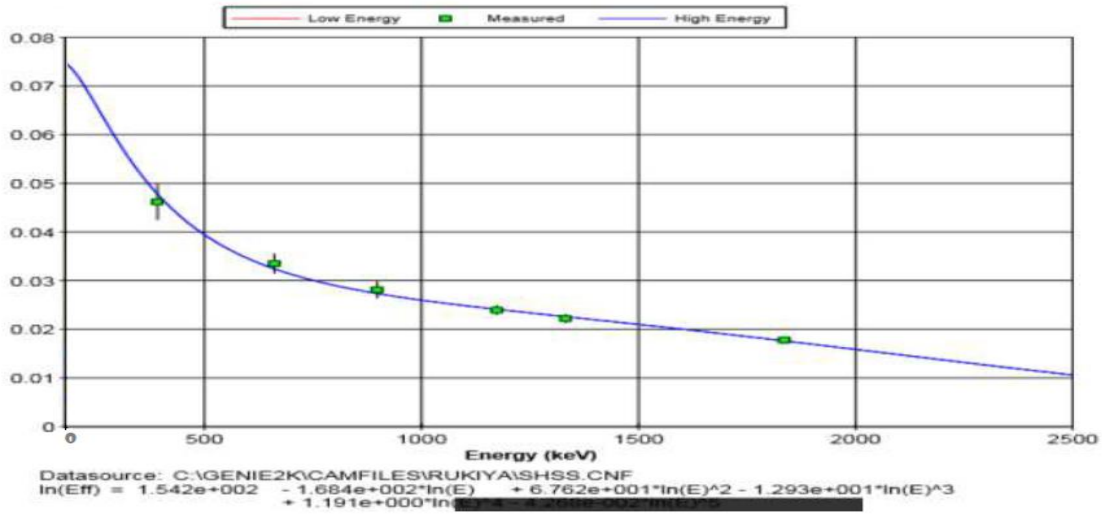


Figure 6: Detector Energy Vs Efficiency Curve

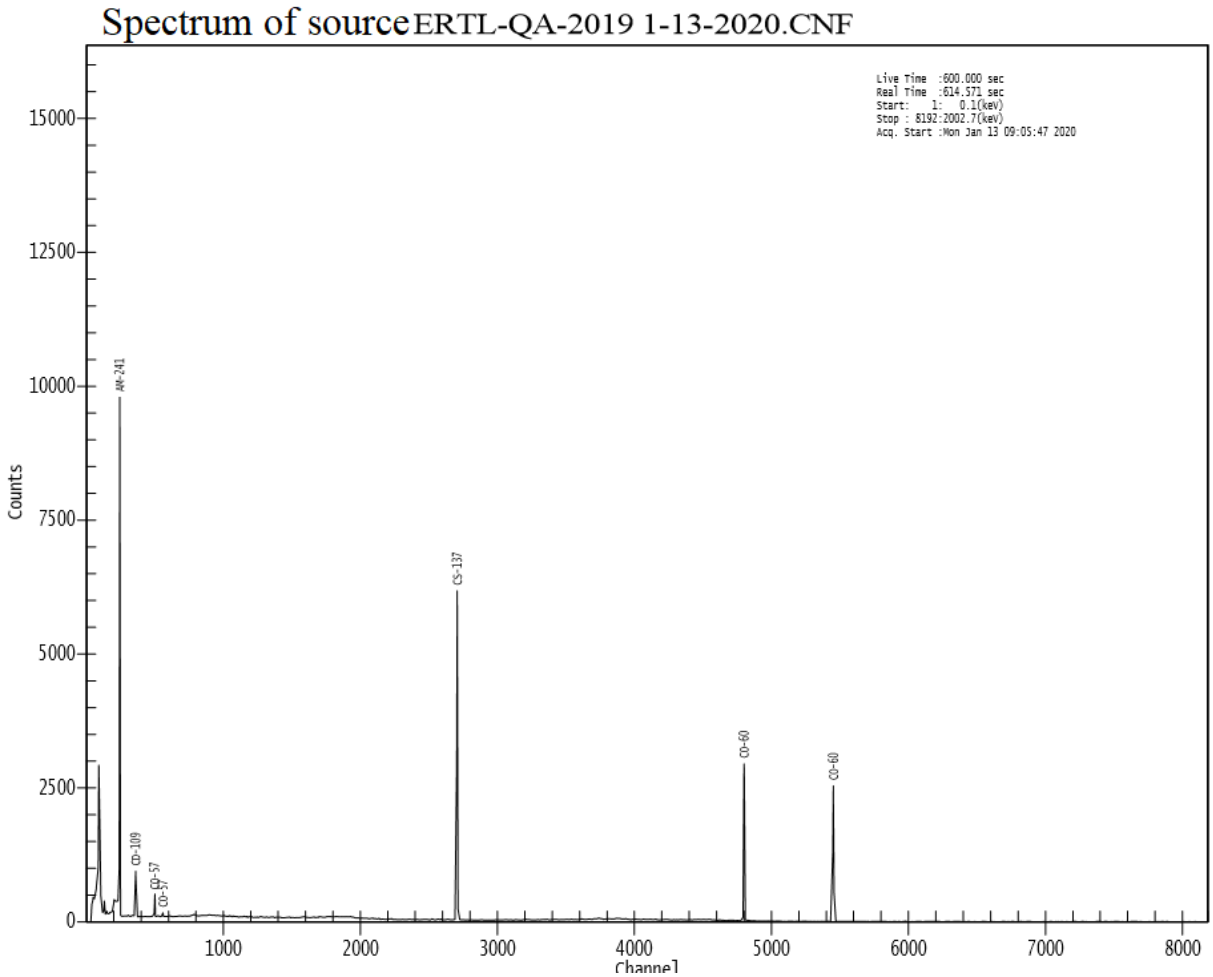


Figure 7: Spectrum for calibration source

Chapter Four

4. Measurements of Activity Concentration of Natural Radionuclides in the Soil samples collected from different sites in Ethiopia.

4.1 Introduction

Soil is the foundation natural resource on which the life supporting system and socio-economic development depends. Soils provide food, fodder and fuel for meeting the basic human and animal needs [Pulakeshiet al., 2014]. Since soil is a scarce resource with a carrying capacity that can be stretched only to a limited extent with the help of technology [Buzuayehuet al.,2002], there is an increasing demand for information on it[Fasinaet al.,2007; Nicolaescu et al., 2009].Different soil types support different land use systemsand require different management options for sustainable productivity. According to Fagbami (1990), the diverse nature of soil is a major reason behind allocation of land to wrong uses. Hence, proper understanding of its nature and properties is necessary for judicious, beneficial, and optimal use on suitable bases [Jagdishet al., 2009].

Soil-plant-man is recognized as a major pathway for the transfer of radionuclides to human beings [IAEA, 1982]. The radioactivity of environmental samples from sites and products suspected of contamination must be investigated before free access to them is given to the public [Owono ,2010]. Both routine and accidental release of nuclear waste can result in radionuclides moving into the environment and ground. The plants acquires deposited radionuclides from soil, named as soil -to-plant transfer factor (TF), which is extensively used for calculating radiological human dose via the ingestion pathway. The soil -to-plant TF is regarded as one of the most important parameters in environmental safety assessment for nuclear facilities [IAEA, 1994].

This TF is essential for environmental transfer models, which are useful in the prediction of radionuclide concentration in agricultural crops for estimating dose impact to human being [Chakraborty et al. 2013]. Radionuclides in soils are frequently transferred to different plant tissues by direct transfer via the root system, or by fallout of radionuclides and re-suspension of contaminated soil followed by deposition on plant leaves [Noordijk et al. 1992]. The uptake of radionuclides from soil to plant is characterized by the Transfer Factor (TF): the ratio of

radionuclide concentration in plant to soil per unit mass [Staven et al. 2003; Yassine et al. 2003]. The TF is usually used for assessing the impact of radionuclide releases into the environment. Due to a predicted long term transfer of radionuclides in the environment due to longevity of these radionuclides, knowledge of the geochemical and ecological cycles is also needed as they relate to the behavior of not only radionuclides but also associated elements. In general, transfer factors show a wide range of variations depending upon several factors including soil properties such as pH, clay mineral, Ca, K and organic matter content, species of plants and other environmental conditions [IAEA 1990]. Maintaining reference-data records will assist in ascertaining possible changes in environmental radioactivity due to nuclear, industrial, and other human activities.

The radioactivity due to natural radionuclides in rocks, soil and water generate a significant component of the background radiation exposure to the population. The terrestrial component of the natural background is dependent on the compositions of the rocks, soil and water in which the natural radionuclides are contained [Karahan, G. and Bayulken, 2000.].

The earth and atmosphere contain varied levels of radioactivity due to chain decays of natural radionuclides uranium-238 and thorium-228 and singly occurring radionuclides such as potassium-40 [Saleh et al. 2007]. Soil features, geological formations and human activities related to radiation and radioactivity are important factors enhancing the background levels of natural radiation [ColmeneroSujo, L. et al.,2004].

Humans are exposed to natural terrestrial radiation that originates predominantly from upper 30 cm of the soil. Humans are also exposed by contamination of the food chain which occurs as a result of direct deposition of radionuclides on plant leaves, root uptake from contaminated soil or water and from direct ingestion of contaminated water.[Santos Junior et al. 2010].

Natural radioactivity is common in the rocks and soil that make up our planet, in water and oceans, and in our building materials. We inhale and ingest radionuclides every day in our lives and radioactive material has been ubiquitous on earth since its creation. The inhalation and ingestion of theses radionuclides above the permissible level become a health hazard. Therefore, concern of the monitoring of these radionuclides in the environment is increasing at all levels, due to their harmful effects. Uranium is the ultimate source of radium and radon. Radon isotopes are the decay products of radium in uranium decay series, as an inert gas, radon can diffuse

through the soil and enter the atmosphere. Radon exposure is associated with the risk of leukemia and certain other cancers, such as melanoma and cancers of kidney and prostate.

Soil is the first link of the ecological chain soil-food-animals-human, and for this reason it has an important role in the distribution and transfer of radionuclides in fodder, which would mean that the information about the radioactive contamination of the soil and the crops are a foundation that later on all criteria and norms of radiation safety rest on. The radioactive contamination of plants is formed with dynamic continuous common action of the atmosphere, the pedosphere and the hydrosphere. From the indicated fields of life, the influence of the pedosphere is dominant, considering that the main part of the minerals which make the plant body, originate from the soil.

Soils are naturally radioactive, primarily because of their mineral content. The main radionuclides are ^{238}U, ^{232}Th and their decay products, and ^{40}K. The radioactivity varies from one soil type to the other depending on the mineral makeup and composition.

Therefore, primarily the physical-chemical characteristics of the soil are the main parameters which determine the quantity of accumulated radioactive substances in the plant organs. Some plants are capable to incorporate large amounts of radioactive substances in their tissues without visible and provable changes, however their consumption can cause serious impairments and diseases in the human organisms and the human. Of course, it may have a negative impact on the growth and development of plants if it is a matter of a stronger intensity of radioactive radiation.

The radioactive contamination of the organs and the body tissues of animal organisms basically depend on the level of contamination of the food they consume, and to a less extent it depends on drinking water and inhalation [A.G. Kudryasheva et al. 1997]. The natural radionuclides by means of migration come from the soil to the crops and contribute for total radiation burden in the population. Considering the fact that they can cause undesired effects on the human organism, it is necessary to determine the content of radionuclides in the surrounding, and on the basis of the obtained results the dose which the human receives should be calculated. However, it turned out that the understanding of the behavior of the natural radionuclides in the environment is very important, because such information can be used as the related parameter values for radiological assessments [Vera Tome, 2003].

The radioactivity concentrations in soil give information on both natural and man-made sources which is important in radiological monitoring and assessment of radiation dose for public [Vera Tome, 2003]. Measurement of natural radioactivity in soil is very important to determine the amount of change of the natural background activity with time as a result of radioactivity release. Studies of natural radioactivity are necessary not only for their radiological impact but also for their ability to act as excellent biochemical and geochemical traces in the environment. Though natural radioactivity is found in rocks and soils throughout the earth, the accession in specific areas varies relatively within narrow limits [UNSCEAR, 2000a].

Gamma radiations emitted from naturally occurring radioactive materials (NORMs) such as uranium-238 (^{238}U), thorium-232 (^{232} Th), and potassium-40 (^{40} K) are generally known as terrestrial background radiation, the main external source of irradiation of the human body [UNSCEAR, 2000.]. However, humankind can be exposed to radiation originating from artificially radioactive sources such as cesium-137 (^{137} Cs) present in the earth's environment as a result of nuclear weapon testing or nuclear fallout from nuclear technology [Ramasamy V et al.2009].

High terrestrial background radiations zones were generally attributed to local geology, location, altitude, and geochemical effects [UNSCEAR, 1993, Ramasamy V, 2009]. Thus, the activity concentrations of radionuclides in granite locations were found to be higher than those of clay, sand stones, and limestone soils [Rafique M et al., 2011, Khan H et al .2012]. Normally, the presence of NORMs in soil generally originates from the disintegrating rocks that are carried to soil by rain and flows [Taskin H et al 2009,UNSCEAR, 2000].

Knowledge of the concentrations and distributions of natural radionuclides, such as ^{40}K, ^{238}U, and ^{232}Th and their decay products, in soils, plants, sediments, and so forth is useful for monitoring environmental radioactive contamination [El-Reefy et al., 2006].
Use of chemical fertilizers, especially phosphorus fertilizer, may lead to increased contamination of agricultural crops by enriching the soil with radioactive elements as well as chemical toxins [Alharbi, 2013].

4.2 Result and Discussion

The results of the present study on the soil samples are summarized one by one as follow.

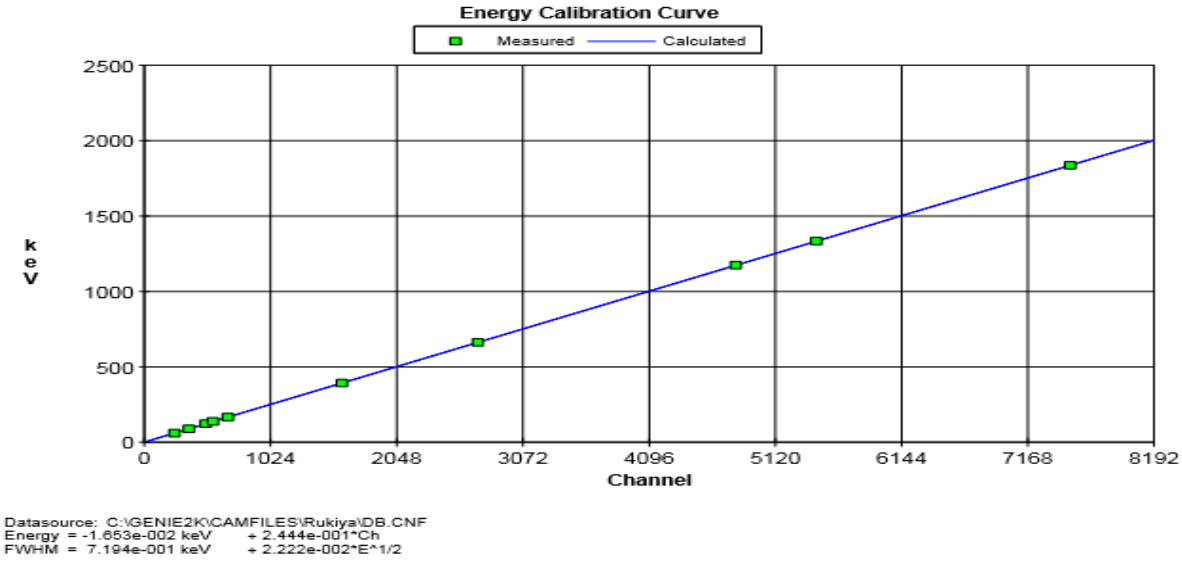


Figure8: Energy calibration for soil sample (DB) curve.

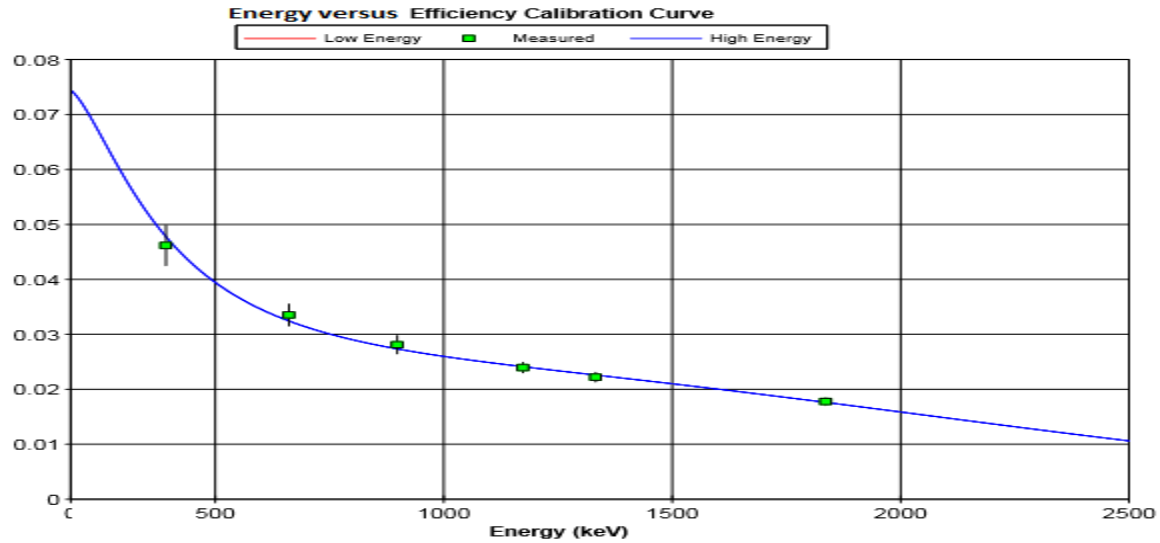


Figure9: Energy versus Efficiency curve

4.2.1 The Activity Concentration

The results of analysis of activity concentration of ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs radionuclides in soil samples for different locations of the study area are presented in table 1.

The activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in the samples were determined by standard gamma spectrometry using aHPGe detector (Ortec) with a 70% relative efficiency and a resolution 1.9 keV for the 1332.5 keV^{60}Co gamma line and MCA with 2000 channel.

The background radiation and the samples were counted for 36,000s. The 295.21 and 351.92 keV of ^{214}Pb and 609.31, 1120.29 and 1764.49 keV of ^{214}Bi gamma ray lines were used to determine the ^{238}U activity concentration. The ^{232}Th activity concentration was determined using 238.63 keV of ^{212}Pb, 911.21 and 968.97 keV of ^{228}Ac gamma lines. The activities of ^{40}K and ^{137}Cs were determined directly from the 1460.8 and 661.6 keV gamma lines, respectively. The net count rate under the most prominent photo peaks of all radio- nuclides daughter peaks were calculated by subtracting the respective count rate from the background spectrum obtained for the same counting time. Then the activity of the radionuclide is calculated from the background subtracted area prominent gamma ray energies [El-Shershaby A et al., 2006].

As we can be seen from Table 1, the value of measured activity concentration of ^{238}U , ^{232}Th , ^{40}K and ^{137} Cs were varies from 24.10 to 56.92 , 67 to 151.23 , 534.58 to 979.57 and 1.13 to 7.4Bq/Kg respectively. The average activity concentration of ^{238}U, ^{232}Th and^{40}K was 45.83, 100.525, 845.65Bq/Kg, respectively. These values were higher than the recommended world average values for ^{238}U , ^{232}Th , ^{40}K , respectively which are 35, 30 and 400 Bq/Kg for ^{238}U , ^{232}Th , ^{40}K respectively [Ramasamy V et al., 2009].

It is also observed that the measured activity concentration of ^{40}K exceeds markedly the values of both Uranium and Thorium, as it is the most abundant radioactive element under consideration. Moreover the excessive use of the Potassium containing fertilizers in the area adjacent to the sampling sites may contribute to the higher values of ^{40}K activity.

The activity concentrations of the artificial radionuclide ^{137} Cs were measured for all collected soil samples in order to assess the amount of fallout radionuclide in such locations; but the activity concentration did not detected for some samples they are given in Table 1.

The obtained activity concentration values of ^{137} Cs in all collected soil samples were found to range from 1.13 Bq kg^{-1}to 7.4 Bq kg^{-1}with an average value of 2.35Bq kg^{-1}. The minimum activity concentration value of ^{137}Cs was obtained for a soil sample collected from Zuwaye, whereas the maximum value was measured in a soil sample collected from Shashemane area. Thus, the impact of the artificial radionuclide and the corresponding additional external radiation exposure to the population were almost negligible. Consequently, the measured activities of ^{137}Cs confirmed no hazard effects due to ^{137} Cs radionuclides to the people living around the sites where soil samples were collected.

Table : 1 The measured Activity concentration level of natural radionuclides in the soil samples collected from different sites located in Ethiopia.

Sample ID	Activity Concentration			
	^{238}U	^{232}Th	^{40}K	^{137}Cs
ARDVS	30.6 ±2.109688	81.0535 ±6.4536	607.876 ±27.9023	…….
DB	56.3 ±3.90955	105.9847 ±8.734	846.001 ±40.033	0.30377 ±0.496669
SA1	48.8 ±3.3	113.93 ±9.57	881.096 ±40.1164	7.40777 ±0.0644194
SBGA-22	52.4 ±3.4934	99.4 ±6.467	908.5 ±41.4039	3.40654 ±0.237108
TSA1	52 ±3.554518	96 ±7.76264	776.3 ±36.2528	2.41325 ±0.181884
TSA2	55.94221 ±3.8107	115.4258 ±9.6338	918.545 ±42.3887	0.586469 ±0.0784295
TSA3	53.51756 ±3.7118	100.6605 ±5.77827	890.412 ±41.028	2.5377 ±0.185931
TSA4	24.1226 ±1.7042	67.90438 ±5.17216	534.982 ±23.6669	1.20966 ±0.096955
SHSS	54.02126 ± 3.423728	151.2349 ± 11.76711	784.584 ± 32.6551	……
BDSH	27.60476 ± 1.761418	80.88988 ± 6.657935	806.968 ± 39.7541	……..
BDH	37.6856 ± 81.08334	76.78563 ± 165.2413	858.251 ± 36.05	1.13734
SHKS	56.92794 ± 3.666244	117.0734 ± 9.374495	979.578 ± 40.8789	……..

The obtained activity concentration values of ^{137} Cs in all collected soil samples were found to range from 0.30Bq $kg^{-1}$to 7.4Bq $kg^{-1}$with an average value of 2.233Bq kg^{-1}. The minimum activity concentration value of ^{137}Cs was obtained for a soil sample collected from Hasasa, whereas the maximum value was measured in a soil sample collected from Aje area. Thus, the impact of the artificial radionuclide and the corresponding additional external radiation exposure to the population were almost negligible. Consequently, the measured activities of ^{137}Cs confirmed no hazard effects due to ^{137} Cs radionuclides to the people living around the sites where soil samples were collected.

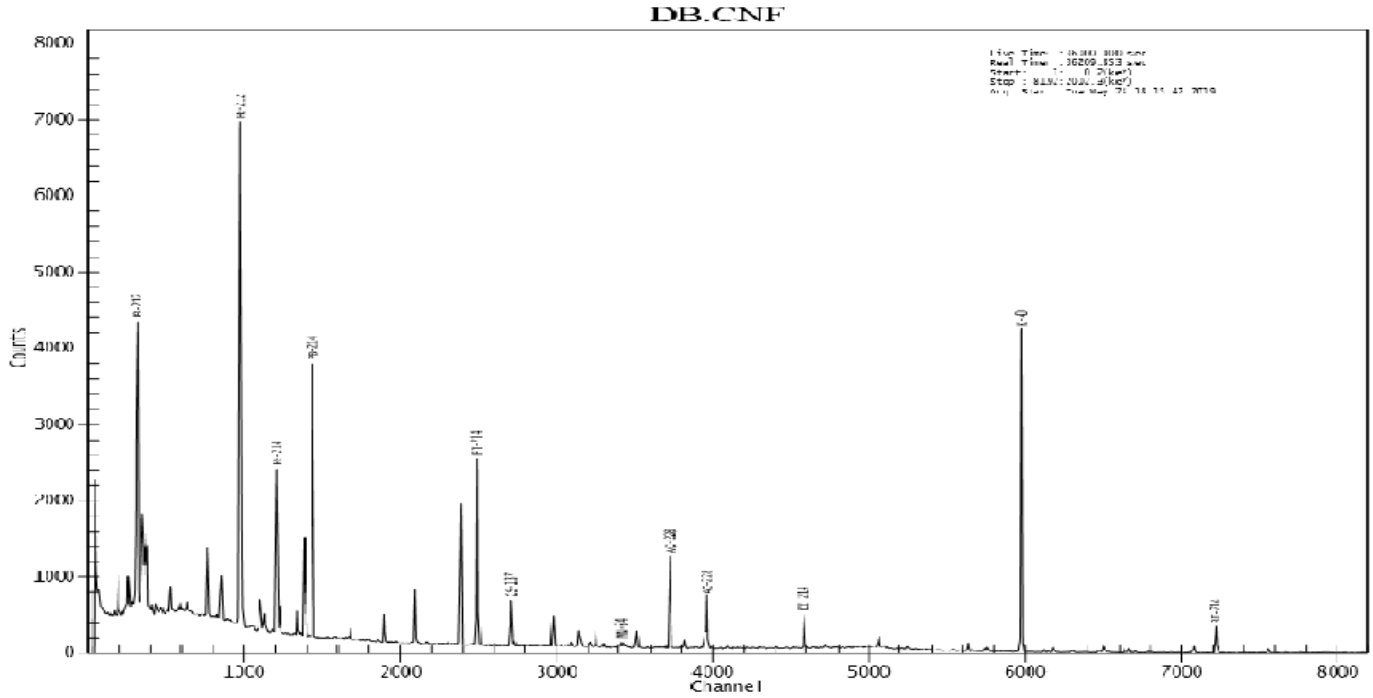
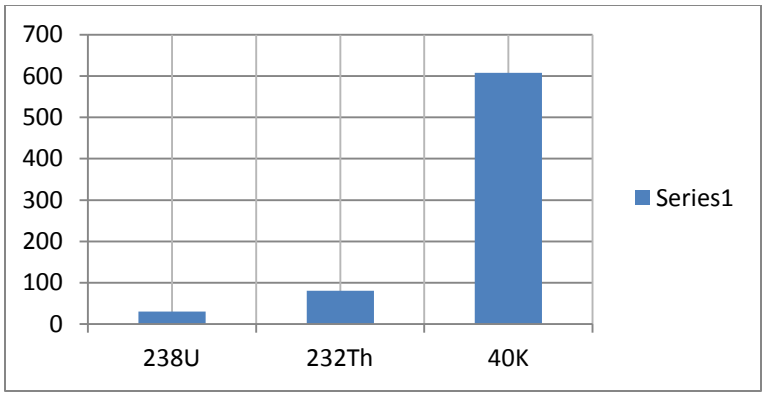
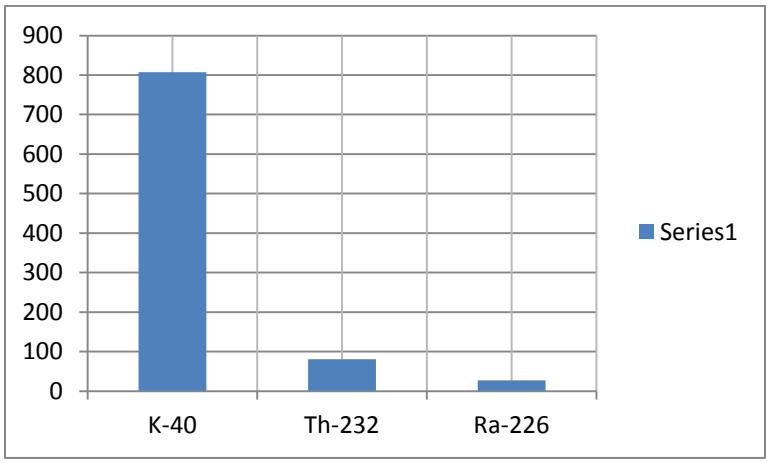


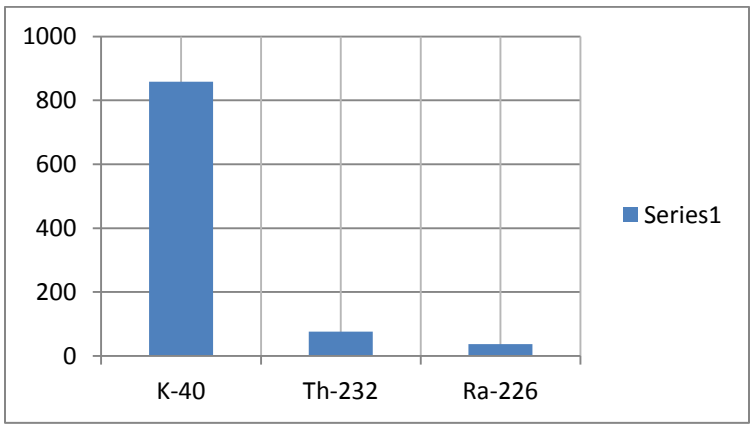
Figure 10: Spectrum of soil samples (DB) during measurement



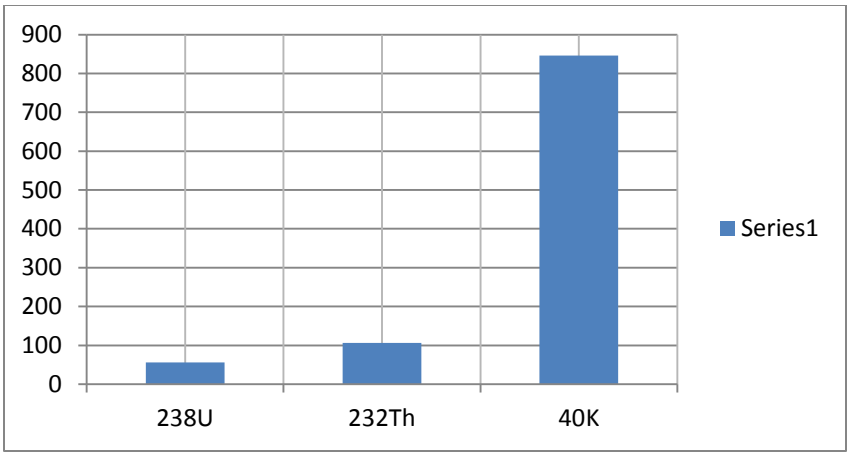
Bar graph 1: Activity of Radionuclides in the sample ARDVS



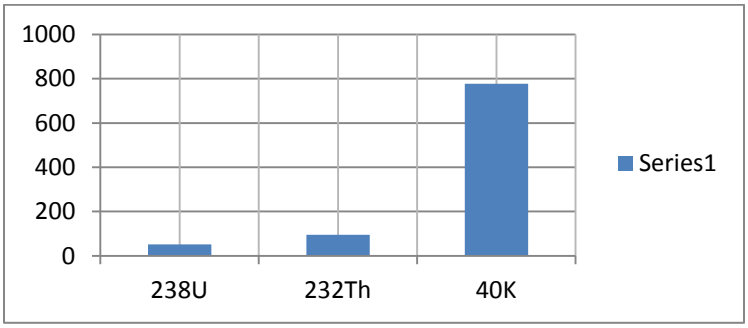
Bar graph 2: Activity of Radionuclides in the sample BDSH



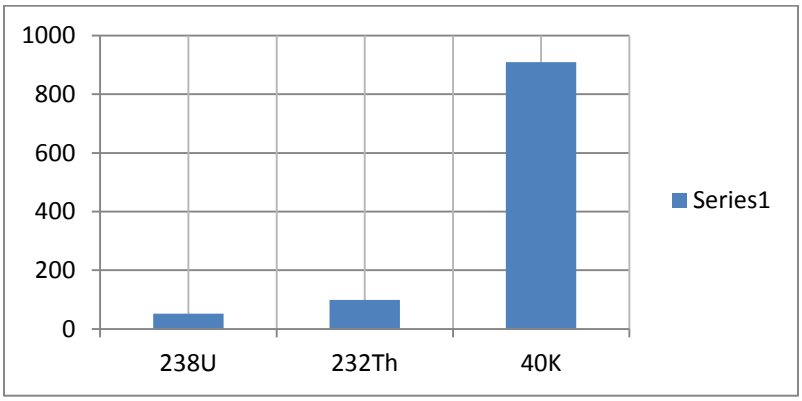
Bar graph 3: Activity of Radionuclides in the sample BDH



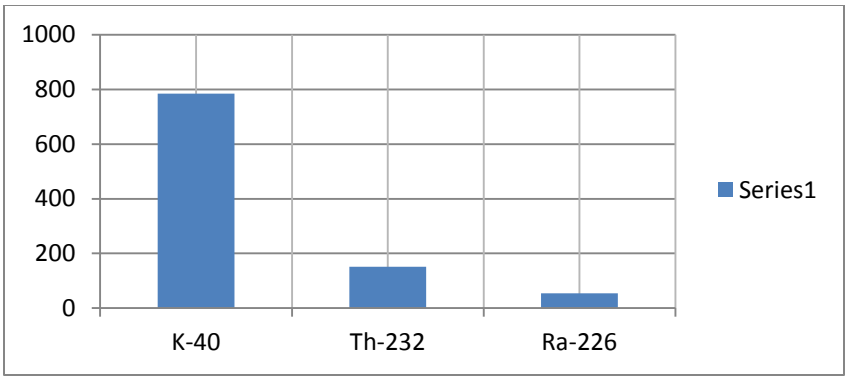
Bar graph 4: Activity of Radionuclides in the sample DB



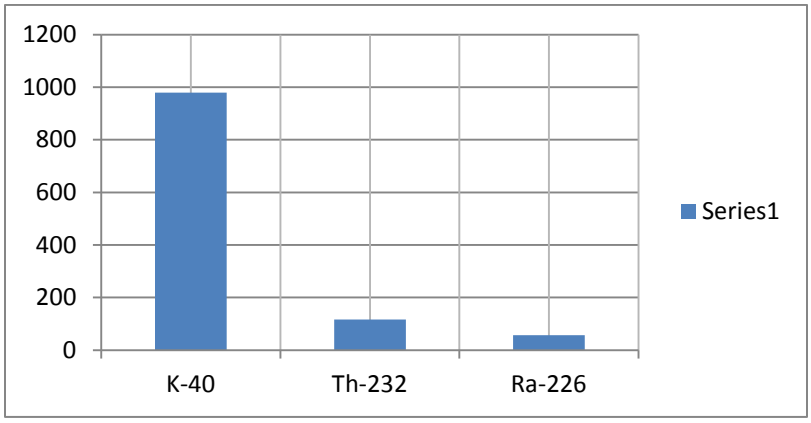
Bar graph 5: Activity of Radionuclides in the sample SA1



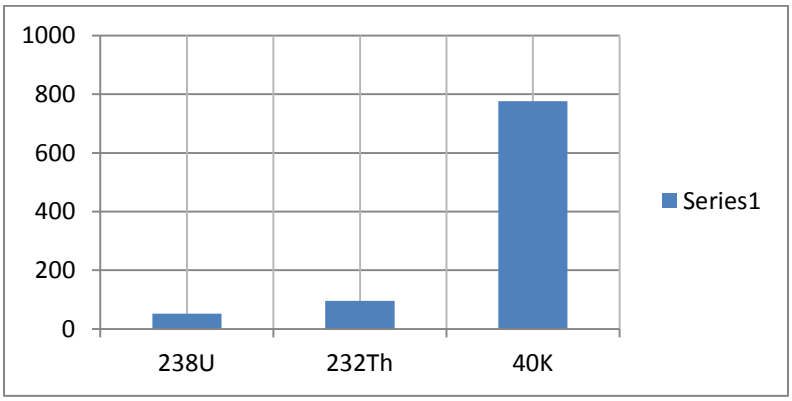
Bar graph 6: Activity of Radionuclides in the sample SBGA-22



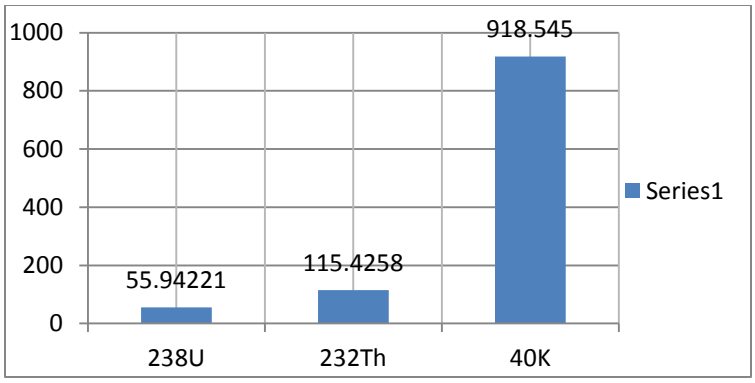
Bar graph 7: Activity of Radionuclides in the sample SHSS



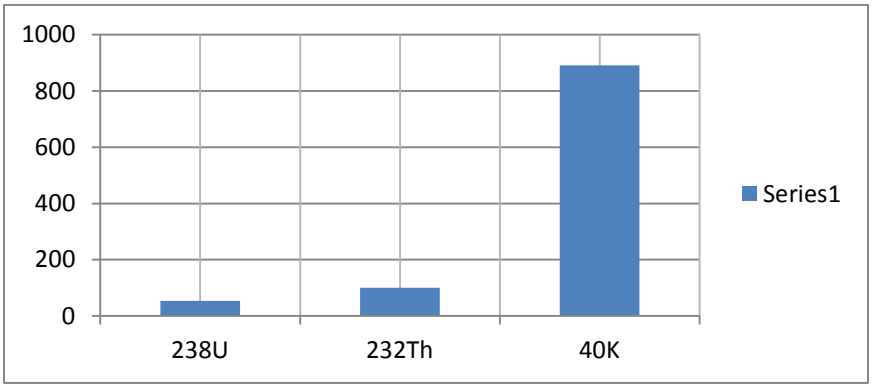
Bar graph 8: Activity of Radionuclides in the sample SHKS



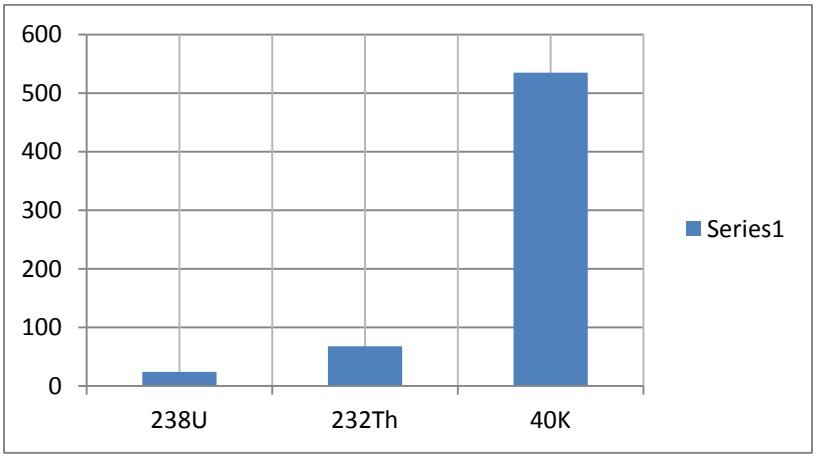
Bar graph 9: Activity of Radionuclides in the sample TSA1



Bar graph 10: Activity of Radionuclides in the sample TSA2



Bar graph 11: Activity of Radionuclides in the sample TSA3



Bar graph 12: Activity of Radionuclides in the sample TSA4

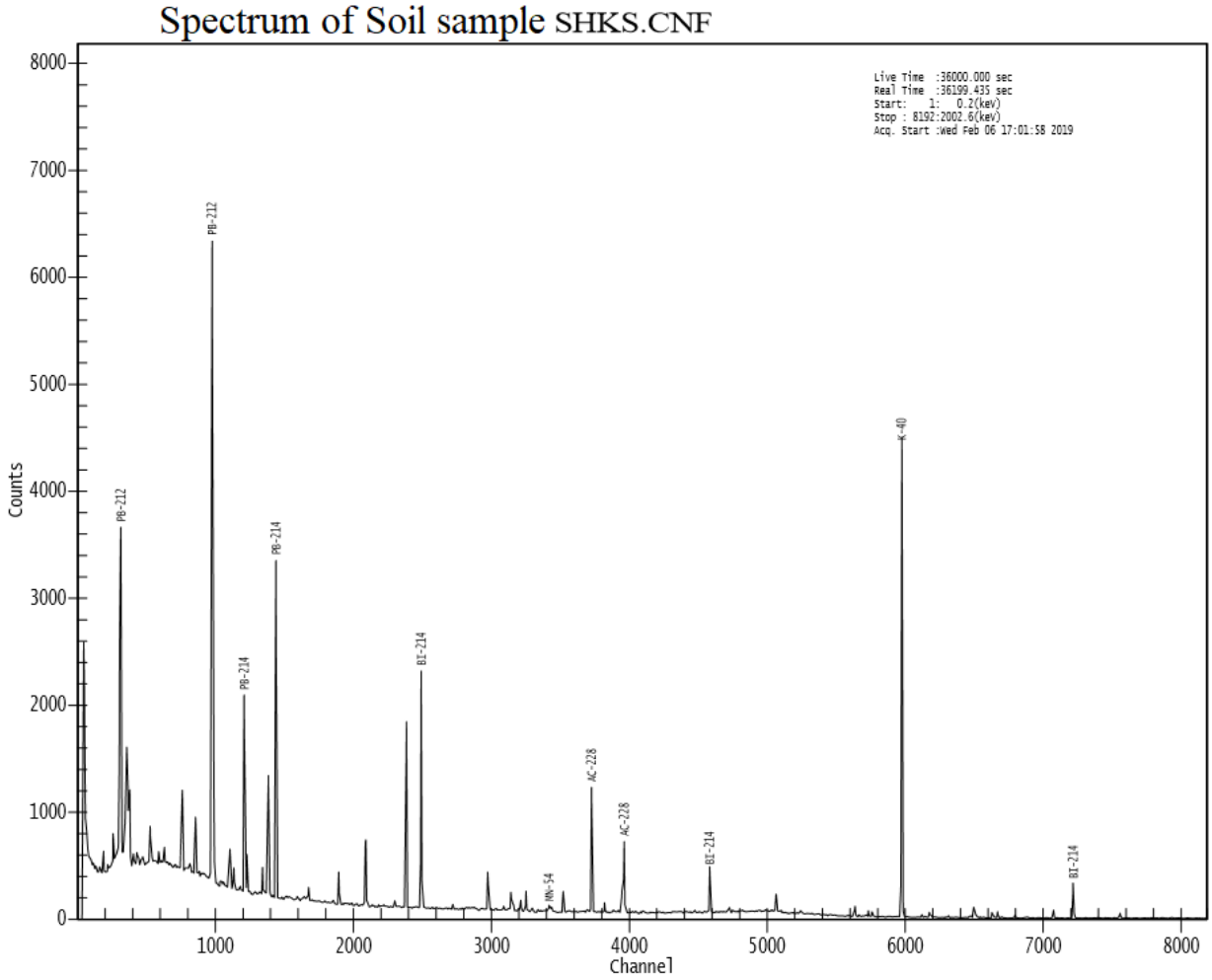


Figure 11: Spectrum of Soil sample SHKS

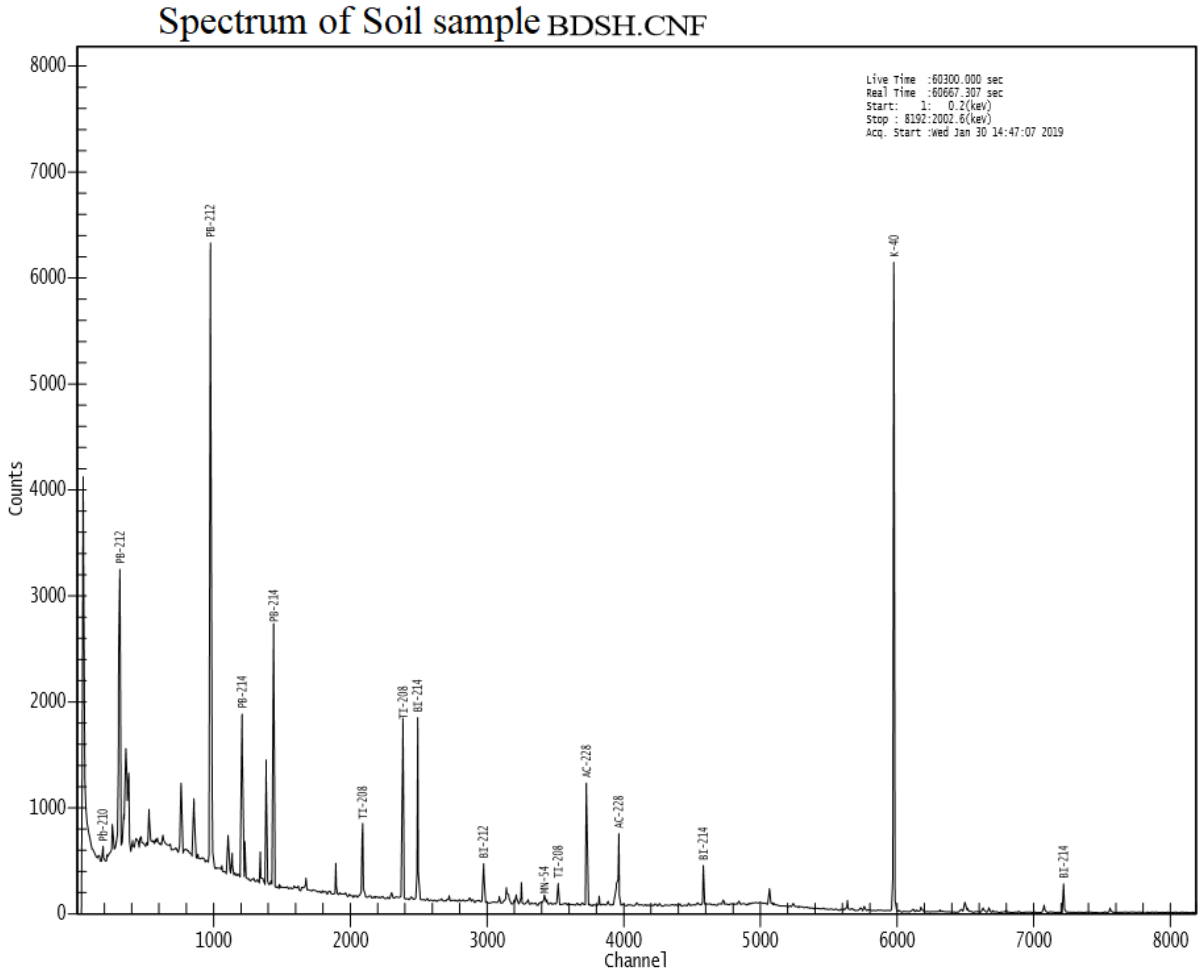


Figure 12: spectrum of Soil sample BDSH

4.2.2 Radiological Effects

The γ-radiation hazards associated with these samples were assessed according to different indices. The calculated data for Raeq, Dr, Dout, Din, Hin, Hex and I_γ of the collected soil samples are shown in table 2.

The values of radium equivalent for different soil samples in area under investigated were calculated by using equation (12) these values presented in table2 and values ranged from 124.96 to 330.7 Bq/kg with average value 248.75Bq/kg which is lower than the recommended maximum value 370 Bq/kg [Gilmore, G.R.,2008].

The radionuclides ^{226}Ra, ^{232}Th and ^{40}K are not homogeneously distributed in soil. The inhomogeneous distribution of naturally occurring radionuclides is due to disequilibrium between ^{226}Ra and its decay products. For uniformity in exposure estimates, the radionuclide concentrations are defined in terms of 'Radium equivalent activity' (Raeq) in $Bqkg^{-1}$.

Table 2: The associated radiological hazard parameters indices Raeq, Hex,Hin, Dr and I γ of natural radionuclides collected from Ethiopia.

Sampl.ID	Raeq(Bq/Kg)	Dr(nGy/h)	Dout (mSv/y)	Din(mSv/y)	AEDE	Hex	Hin	I_γ
SHSS	330.7	191.1	0.234	1.312	1.546	0.86	1.034	2.4
BDSH	205.4	100.1	0.123	0.69	0.813	0.55	0.63	1.53
BDH	155.8	137.9	0.169	0.95	1.119	0.59	0.68	1.6
SHKS	299.8	143.9	0.177	0.989	1.166	0.81	0.963	2.20
ARDVS	150.8	92.65	0.64	0.114	0.75	0.60	0.522	1.42
DB	213.77	164.6	1.14	0.20	1.33	0.89	0.74	1.99
SA1	217.88	168.4	1.16	0.21	1.36	0.88	0.75	2.05
SBGA-2	200.9	161.35	1.11	0.2	1.3	0.86	0.71	1.95
TSA1	194.71	153.21	1.05	0.19	1.24	0.81	0.67	1.82
TSA2	227.43	173.8	1.19	0.21	1.4	0.94	0.79	2.14
TSA3	203.69	161.85	1.11	0.2	1.31	0.86	0.72	1.96
TSA4	124.97	112.3	0.77	0.14	0.91	0.51	0.44	1.19

The calculated values of Dr were found to vary from 96.64 to 191.02nGy h^{-1}, with an average value of 149.5117nGy h^{-1}. The measured average absorbed dose rate in the air and the measured average value of the representative level index are higher than the recommended international levels of 55 nGy h^{-1}[Turner J, 2007]. Such locations are not safe for human residency and agriculture.

The absorbed dose rate was converted into annual effective dose equivalent by using a conversion factor of 0.7 SvGy-1 recommended by the UNSCEAR 2000 and 0.2 for the outdoor

occupancy factors by considering that the people on the average, spent 20% of their time in outdoors. The effective dose due to natural activity in the soil samples was calculated by:

$E\ (mSvyr\text{-}1) = D \times 24 \times 365.25 \times 0.2 \times 0.7 \times 10^{-6}$

The calculated indoor, outdoor, and total AEDE values are presented in Table 2. Calculated values for Dout, Din, and Dtot respectively 0.185, 1.035, and 1.22 mSv year to global measured values, these values were all higher than the assigned worldwide values of 0.08, 0.42, and 0.5 year-1, respectively [UNSCEAR,2000]. The locations from which the soil samples collected were all not safe according to the Radiation Protection, and such locations cannot be classified as hazard free [NRPB,1992].

The external (Hex) and internal (Hin) hazard index due to the emitted γ -rays of the soil samples were calculated and examined according to the following criterion: The value of Hex must be lower than unity in order to keep the radiation hazard insignificant. This is the radiation exposure due to the radioactivity from a soil, limited to 1.5 $mGy \cdot y^{-1}$. The maximum values of Hex equal to unity correspond to the upper limit of Raeq (370 $Bq \cdot kg^{-1}$) [12]. The calculated external hazard index values were found to vary between 0.43 and 0.78 with average value of 0.668 these values are less than unity, which is 33.2 % less than recommended value. The calculated internal hazard index values were found to vary between 0.5 and 0.93 with average value of 0.79, which is 11.0% less than recommended value [Dabayneh K et al,.2008].

Radioactivity level index ranged from 1.19 to 2.395 Bq.kg-1with average values is 1.917 Bq. kg^{-1} (Table 2). The average value of Radioactivity level index is higher than the recommended safe limit ≤ 1 [Thabayneh K, Jazzar M.2012]. Therefore, the soils have radiation hazard and are harmful to society living there.

Chapter Five

5. Measurements of Activity Concentration of Natural Raionuclides in the Collected Different Coffee samples

5.1 Introduction

Coffee is one of the most popular and widely consumed beverages in the world, and its consumption is increasing [Roselli C,2013]. However, variation exists in the annual consumption between countries. In addition, coffee is grown in many countries, where the coffee trade has played a crucial role in their economic development [Roselli C et al. 2013]. Coffee comes from the plant genus Coffea, which has two primary species, C. Arabica (Arabica) and C. canephora (known as C. obusta).

Coffee is a brewed drink prepared from roasted coffee beans, the seeds of berries from certain Coffea species. The genus Coffea is native to tropical Africa (specifically having its origin in Ethiopia and Sudan) and Madagascar, the Comoros, Mauritius, and Réunion in the Indian Ocean[Maurin, O.;et al,.2007] Coffee plants are now cultivated in over 70 countries, primarily in the equatorial regions of the Americas, Southeast Asia, Indian subcontinent, and Africa. The two most commonly grown are C. arabica and C. robusta. Once ripe, coffee berries are picked, processed, and dried. Dried coffee seeds (referred to as "beans") are roasted to varying degrees, depending on the desired flavor. Roasted beans are ground and then brewed with near-boiling water to produce the beverage known as coffee.

Coffee is darkly colored, bitter, slightly acidic and has a stimulating effect in humans, primarily due to its caffeine content [Cappelletti S et al.,2015]. It is one of the most popular drinks in the world,[Oder, Tom., 2018] and it can be prepared and presented in a variety of ways (e.g., espresso, French press, caffè latte). It is usually served hot, although iced coffee is a popular alternative. Clinical studies indicate that moderate coffee consumption is benign or mildly beneficial in healthy adults, with continuing research on whether long-term consumption lowers the risk of some diseases, although those long-term studies are of generally poor quality [Poole Ret al. 2017].

While coffee is native to Ethiopia and Sudan, the earliest credible evidence of coffee-drinking as the modern beverage appears in modern-day Yemen in southern Arabia in the middle of the 15th century in Sufi shrines[Weinberg &Bealer 2001]. It was in what is now Yemen that coffee seeds were first roasted and brewed in a manner similar to how it is now prepared for drinking. But the coffee seeds had to be first exported from East Africa to Yemen, as Coffea arabica is thought to have been indigenous to the former[Wild, Antony, 2004]. The Yemenis obtained their coffee via Somali traders from Berbera (who in turn procured the beans from the Ethiopian Highlands) and began to cultivate the seed. By the 16th century, the drink had reached Persia, Turkey, and North Africa. From there, it spread to Europe and the rest of the world.

Coffee is a major export commodity, being the top legal agricultural export for numerous countries [Oder, Tom., 2018, FAO Statistics Division. 2007]. It is one of the most valuable commodities exported by developing countries. Green, unroasted coffee is one of the most traded agricultural commodities in the world [Mussatto, et al, 2011]. The way developed countries trade coffee with developing nations has been criticised, as well as the impact on the environment with regards to the clearing of land for coffee-growing and water use. Consequently, the markets for fair trade and organic coffee are expanding [Alex Nicholls; Charlotte Opal, 2005].

Coffee production in Ethiopia is a longstanding tradition which dates back to dozens of centuries. Ethiopia is where Coffea arabica, the coffee plant, originates [Thomas P. Ofcansky, David H. Shinn, 2004]. The plant is now grown in various parts of the world; Ethiopia itself accounts for around 3% of the global coffee market. Coffee is important to the economy of Ethiopia; around 60% of foreign income comes from coffee, with an estimated 15 million of the population relying on some aspect of coffee production for their livelihood [Thomas P. Ofcansky, David H. Shinn,.2004]. In 2006, coffee exports brought in $350 million,[CIA World Factbook, 2010] equivalent to 34% of that year's total exports[CIA World Factbook, 2010].

Ethiopia is the world's seventh largest producer of coffee, and Africa's top producer, with 260,000 metric tons in 2006[Food and Agricultural commodities production, 2010]. Half of the coffee is consumed by Ethiopians [Cousin, Tracey L, 1997] and the country leads the continent in domestic consumption[Major coffee producers, 2010]. The major markets for Ethiopian coffee

are the EU (about half of exports), East Asia (about a quarter) and North America[Keyzer, Merbis & Overbosch 2000]. The total area used for coffee cultivation is estimated to be about 4,000 km^2 (1,500 sq mi), the size is unknown due to the fragmented nature of the coffee farms [Belda , 2006]. The way of production has not changed much, with nearly all work, cultivating and drying, still done by hand [Cousin, Tracey L. June 1997].
The revenues from coffee exports account for 10% of the annual government revenue, because of the large share the industry is given very high priority, but there are conscious efforts by the government to reduce the coffee industry's share of the GDP by increasing the manufacturing sector [Belda, 2006].

The Tea and Coffee Authority, part of the federal government, handles anything related to coffee and tea. Such as fixing the price at which the washing stations buy coffee from the farmers. This is a legacy from a nationalization scheme set in action by the previous regime that turned over all the washing stations to farmer's cooperatives. The domestic market is heavily regulated through licenses, with the goal of avoiding market concentration [Keyzer, Merbis & Overbosch 2000].

Some naturally occurring radioisotopes and other elements present in soil are drawn into the roots of plants via ion channels or specific transporters [Sugiyama H et al.2009]. Their distribution throughout the plant tissues depends on their chemical characteristics and several parameters of soil and the plants themselves [Awudu AR et al.2012].

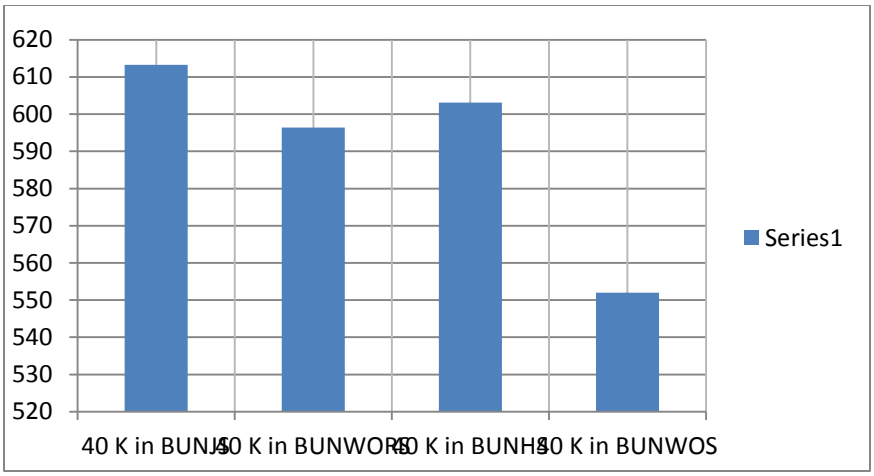
5.2 Result and Discussions

The results of the present study on the coffee samples are summarized one by one as follow. Detection of the amount of natural radioactivity for ^{226}Ra, ^{238}U, ^{232}Th, and ^{40}K in coffee powder samples was carried out with gamma spectrometry using a high-purity germanium (HPGe) detector with a 70% relative efficiency and a resolution 1.9 keV for the 1332.5 keV^{60}Co gamma line and MCA with 2000 channel.

The background radioactivity of material and the samples were counted 36,000s. The gamma ray lines 295.21 and 351.92 keV of ^{214}Pb and 609.31, 1120.29 and 1764.49 keV of ^{214}Bi were used to determine the ^{238}U activity concentration. The gamma lines 238.63 keV of ^{212}Pb, 911.21 and 968.97 keV of ^{228}Ac used to determine activity concentration ^{232}Th. The activities concentration of ^{40}K and ^{137}Cs were determined directly from the 1460.8 and 661.6 keV gamma lines, respectively. The net count rate under the well-known photo peaks of all radio- nuclides daughter peaks were calculated by subtracting the respective count rate from the background spectrum obtained for the same counting time. Then the activity of the radionuclide was calculated from the background subtracted area well-known gamma ray energies [El-Shershaby A, 2006].

Table 3: The measured Activity concentration level of natural radionuclides in the Coffee samples collected.

Sample ID	Activity concentration Bq/kg
	^{40}K
BUNJS	613.28 ± 45.995
BUNWORS	596.35 ± 33.878
BUNHS	603.12 ± 38.165
BUWONS	551.99 ± 31.0000



Bar graph 13: Activity of 40 K Radionuclides in the sample BUWOS

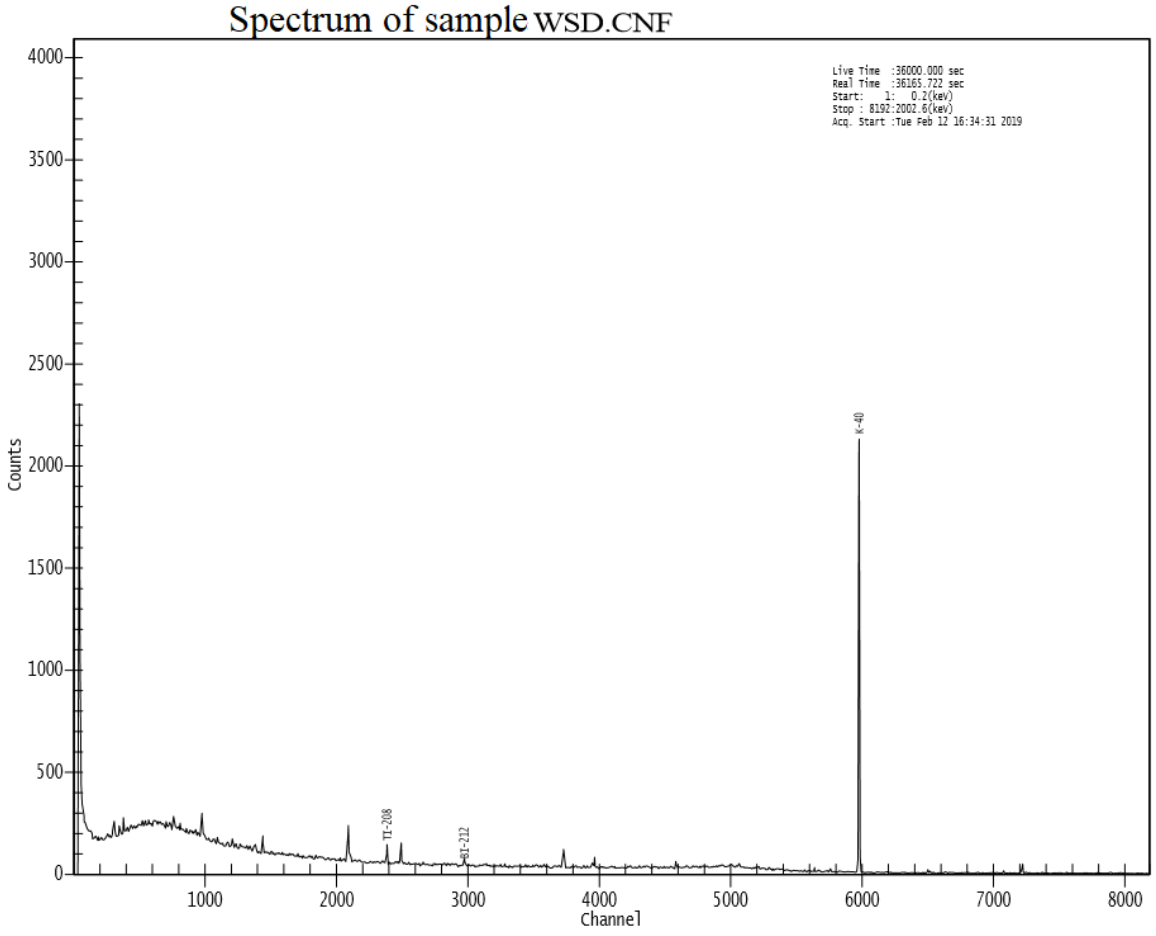


Figure 13: Spectrum of Coffee sample WSD

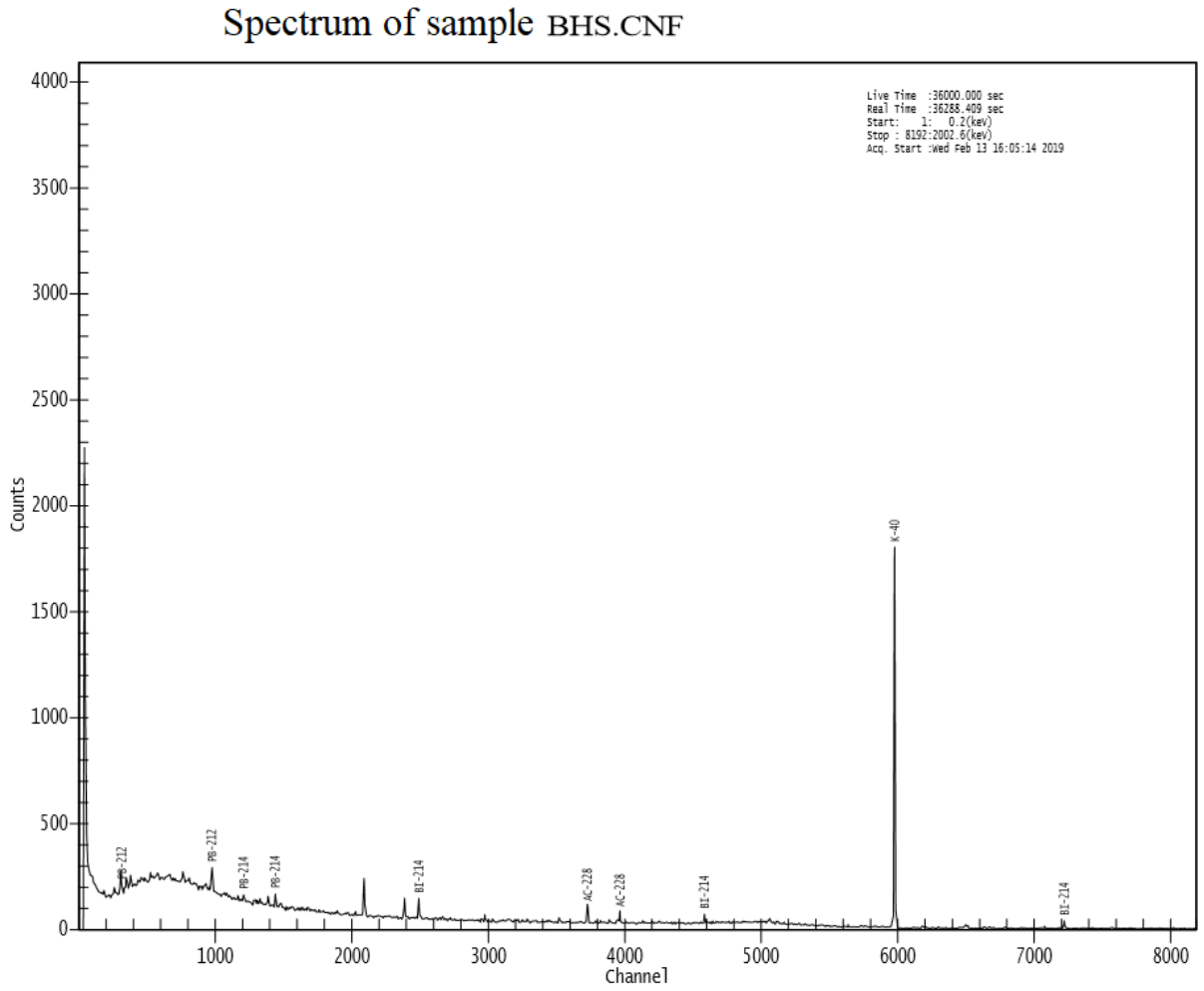


Figure 14: Spectrum of Coffee sample BHS

During our experimental activities our detector could identified only potassium-40 in the coffee samples. All coffee samples had 591.185 Bq/Kg average values of ^{40}K concentrations that were higher than the acceptable value (412 Bq/kg) [UNSCEAR, 2010].

The values of radium equivalent for different coffee samples in area under observation were calculated by using equation below these values presented in table4 and values ranged from 42.5 to 54.92 Bq/kg with average value 47.44Bq/kg which is lower than the recommended maximum value 370 Bq/kg [Gilmore, 2008].

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_{K}$$

Where C_{Ra}, C_{Th} and C_K are the average activity concentration in the sample in Bq/kg of ^{226}Ra, ^{232}Th, and ^{40}K respectively [K. M. Dabayneh, 2008]

The calculated values of absorbed dose rate (Dr) were found to vary from 23.73 to 30.67nGy h^{-1}, with an average value of 26.49nGy h^{-1}. The determined average absorbed dose rate in the air was lower than the recommended international levels of 55 nGy h^{-1}[Turner J et al.,2007] and the coffee was safe for use.

The calculated indoor, outdoor, and total AEDE values are presented in Table 4. Calculated average values for Dout, Din, and Dtot respectively 0.0325, 0.181, and 0.107 mSv /year to global measured values, these values were all lower than the assigned worldwide values of 0.08, 0.42, and 0.50 mSvyear-1, respectively [UNSCEAR, 2000].

The external hazard index (Hex) and internal hazard index (Hin) due to the emitted γ -rays of the coffee samples were calculated and examined according to the following condition: The value of Hex must be lower than unity in order to keep the radiation hazard insignificant. This is the radiation exposure due to the radioactivity from a soil, limited to 1.5 $mGy \cdot y^{-1}$. The maximum values of Hex equal to unity correspond to the upper limit of Raeq (370 $Bq \cdot kg^{-1}$) [12]. The calculated external hazard index values were found to vary between 0.115 and 0.148 with average value of 0.128 these values are less than unity, which is 87.2 % less than recommended value. The calculated internal hazard index values were found to vary between 0.115 and 0.148 with average value of 0.128, which is 87.2% less than recommended value [Knoll G.F, 2000].

Radioactivity level index ranged from 0.36 to 0.475 Bq.kg-1with average value is 0.4 Bq. kg^{-1} (Table 4). The average value of Radioactivity level index is lower than one the recommended safe limit ≤ 1 [Van Rooyen T.J.,2002]. Therefore, the coffee had no radiation hazard.

Hence it can be concluded that the radionuclide concentration of measured coffee samples were poses no radiological health hazard to the community.

Table 4: Radium equivalent, absorbed dose rate, annual effective dose rate, external hazard index, and internal hazard index in different coffee samples.

Sample ID	Raeq Bq/Kg	Dr(nGy/h)	Dout(mSv/y)	Din(mSv/y)	AEDE	Hex	Hin	I_γ
BUNJS	54.92	30.67	0.0376	0.210	0.2476	0.148	0.148	0.475
BUNWORS	45.91	25.64	0.0315	0.176	0.215	0.124	0.124	0.397
BUNHS	46.44	25.93	0.0318	0.178	0.2098	0.125	0.125	0.4
BUNWONS	42.50	23.735	0.0291	0.163	0.1921	0.115	0.115	0.36

Chapter Six

6. Measurements of Activity Concentration of Natural Radionuclides in the Lake Water samples collected from Lake Zuwaye.

6.1 Introduction

Water is vital and, concurrently, one of the most important natural resources. About 70% of the Earth's surface is covered with water, which is estimated at a volume of approximately 1.4 billion km^3[Ashton *et al*., 2012]. However, most of it is salty, and only around 2.5% of the global water resources (about 35 million km^3) consists of freshwater (merely a fraction of the freshwater can be used for drinking water purposes). Over 30% (about 10.5 million km^3) is stored as groundwater beneath the earth's surface the most important available fresh water resource. Lakes and rivers account for far less than a half percent of the fresh water reserves.

Terrestrial radiation originates from the natural radionuclides, which emits gamma rays where individuals are in contact with it inwardly or outwardly. A large portion of these natural radionuclides which are going by uranium (^{238}U), thorium (^{232}Th) and potassium (^{40}K) are found in the earth's crust. These natural radionuclides are concentrated in rocks and soil in various types and depend on the geology of the areas and geographic conditions, which prompt to expand the effective dose from natural earthbound radiation [UNSCEAR. 2000]. The study of terrestrial radiation is noteworthy as well as marker alarm for the man who lives in an area which has a high-concentration in natural radionuclides [Ramli et al., 2005]. Terrestrial radiation in soil may be hazardous people, contingent upon methods of transmission to people, internal and external exposure to gamma rays, consequently, it is extremely imperative to evaluate the concentration of natural radionuclides in soil and water.

There are two courses in which the radionuclides can be transmitted to the human body specifically and in a roundabout way. The immediate course is the point at which the human is exposed to external gamma rays and alpha particles, which are radiated from radionuclides in soil. This external exposure would cause skin blazing and radiation disorder if the concentration

of the radionuclides surpassed the allowed universal cutoff point. Likewise, gamma rays and alpha particles are radiated from radon gas (^{222}Rn), which is the component in uranium-238 chain. Radon gas inhalation is the prime supplier of the external dosage of the total populace [UNSCEAR, 1988].

Though, the roundabout way originates from water due to contact with the soil might be subjected to temperament of radionuclides in it, particularly in lakes and rivers as suppliers for drinking water. Along these lines the human body is uncovered through adulterated water by radionuclides to the danger of bio agglomeration, which builds the concentration of these radionuclides in the human body [Malandrino, 2011].

Living organisms are being exposed continuously to naturally occurring ionizing radiation. These radiations which we are exposed to usually are cosmic rays which are sourced from outer space, the sun's surface, radionuclides which occur in the earth's crust, in building materials, water, air foods and the human body. Some of these exposures are constant and uniformly distributed for all individuals globally, for example, ingestion of ^{40}K in foods. Other exposures vary greatly depending on location [Kieran maher, 2004-2006].

Radionuclides have been released to the environment from different sources and processes. Radionuclides present in the biosphere, whether natural or artificial in origin, ultimately result in irradiation of human populations [Aheir 1995]. Naturally occurring radionuclides of terrestrial origin are present in rivers and lakes sediments as well [Krmar 2009]. A number of scientific efforts have been undertaken to characterize radionuclides in the marine environment, including biota, water and sediments [Dar and El Saharty 2013; Ibrahim and Ramzy 2013; El-Reefy et al. 2010; El Zakla et al. 2013; Darwish 2013].

Such studies can be useful to estimate the degree of human risk associated with the ingestion of radionuclides in biota through the food chain and to establish a baseline database of radionuclides concentration, in order to monitor the possible variations in the marine environmental radioactivity due to nuclear industry and other human activities [Akram 2005 and Burger 2006].

Sediment plays a role in accumulating and transporting contaminants within the geographic area and is considered the environmental host of the waste discharged by natural or artificial

processes in our world. Lakes act therefore as sinks for the materials which pass through the various aquatic chemical and biological cycles including radionuclide contaminants. They were considered as sources of radionuclides to the downstream ecosystems.

Cosmic rays, for example, are stronger at high altitudes, and concentrations of thorium and uranium in soils are elevated in localized areas. Exposures may also change as a result of human practices and activities. Mostly, building materials and design of houses and their ventilation systems strongly influence indoor concentrations of radon gas and its decay products, which contribute greatly to inhalation doses. Therefore, it is necessary to study the naturally occurring radiation levels in the different components of the environment. Such investigations could be useful for both assessment of public dose rates and the performance of epidemiological studies as well as keeping reference-data records to ascertain possible changes in the environmental radioactivity due to nuclear, industrial, and other human activities [UNSCEAR, 2000a].

The natural radioactivity is originated with radioisotopes of natural decay chains in earth, cosmic radiations and man-made artificial radiation sources. Generally radioisotope contents in water and soil are determined in environmental research. Natural waters are known to contain both alpha emitters (^{238}U) and beta emitters (^{40}K) in a wide range of concentrations. Generally, they are responsible for a small fraction of the total dose received from natural and artificial radioactivity [UNSCEAR, 2000]. Natural waters contain both α and β emitters in widely varying concentrations which are responsible for a generally small fraction of the total dose received from natural and artificial radioactivity [UNSCEAR, 1993]. Alpha activity is mostly due to uranium isotopes (^{234}U, ^{235}U and ^{238}U) and to ^{226}Ra [D.M. Bonotto et al. 2009]. Beta activity is usually due to a large extent to ^{40}K and to short-lived daughters of ^{238}U, ^{234}Th and ^{234}Pa [M. Forte et al. 2009].

Water sources from areas that are rich in naturally occurring radioactive materials exhibit high levels of radioactivity [Caroli et al., 2013]. Natural water contains several α and β emitting isotopes in wide range of concentrations [Al-Amir et al., 2012]. The predominant radionuclides found in water include ^{226}Ra, ^{232}Th, ^{137}Cs and ^{238}U and their decay products as well as ^{40}K [Khayet& Matsuura, 2013].

6.2Result and Discussion

The results of the present study on the lake water samples are summarized one by one as follow.

6.2.1 The Activity Concentration

The results of analysis of activity concentration of ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs radionuclides in lake water samples for different locations of the study area are presented in (Table 4).
The activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in the samples were determined by using gamma spectrometry coupled with HPGe detector (Ortec) with a 70% relative efficiency and a resolution 1.9 keV for the 1332.5 keV^{60}Co gamma line and MCA with 2000 channel.

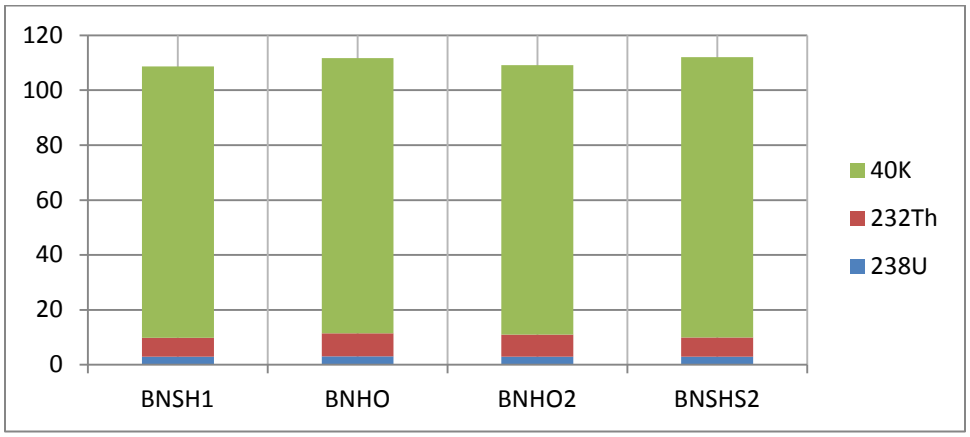
The samples packed in the Marinelli beaker and were counted for 36,000s. The activities concentration of ^{238}U in the samples was determined using the energy peak of 609.31 keV of ^{214}Bi. Similarly, the activity concentration of ^{232}Th was determined using 911.21 keV peak of ^{228}Ac gamma line. The activity concentration of ^{40}K was determined from the energy of 1460.83 keV.

The net count rate under the well-known photo peaks of all radio-nuclides daughter peaks were calculated by subtracting the respective count rate from the background spectrum obtained for the same counting time. Then the activity of the radionuclide is calculated from the background subtracted area well-known gamma ray energies [El-ShershabyA et al., 2006].

Table 4: The measured Activity concentration level of natural radionuclides in the Lake Water samples collected from different sites located in Zuwaye Lake.

Sample ID	Activity Concentration		
	^{238}U	^{232}Th	^{40}K
BNSH1	2.94998 ± 0.256548	6.9445 ± 0.73691	98.693 ± 4.89844
BNHO	3.028816 ± 0.278698	8.3902 ± 0.82405	100.3 ± 5.237678
BNHO2	3.017623 ± 0.265988	7.9978 ± 0.79643	98.12 ± 4.16154
BNSHS2	2.978876 ± 0.2665543	7.0237 ± 0.69885	101.99 ± 4.91556

The water samples from the lake Zuwaye were found to contain levels of radionuclides with average activity concentrations of 2.99±0.267, 99.77±4.80 and 7.5 ±0.76 Bq/L for ^{238}U, ^{40}K, and ^{232}Th, respectively. The annual effective dose calculated for the lake varied from 0.081 to 0.09 mSv/y with an average of 0.0858 mSv/y. This value when compared with the ICPR standard value of 1mSvyr-1 is very low, hence, it can be concluded that the radionuclide concentration of water supply in Lake Zuwaye is negligible and poses no radiological hazard to the public.



Bar graph 14: **Activity concentration level of natural radionuclides ^{40}K, ^{232}Th and ^{238}U Zuwaye Lake Water.**

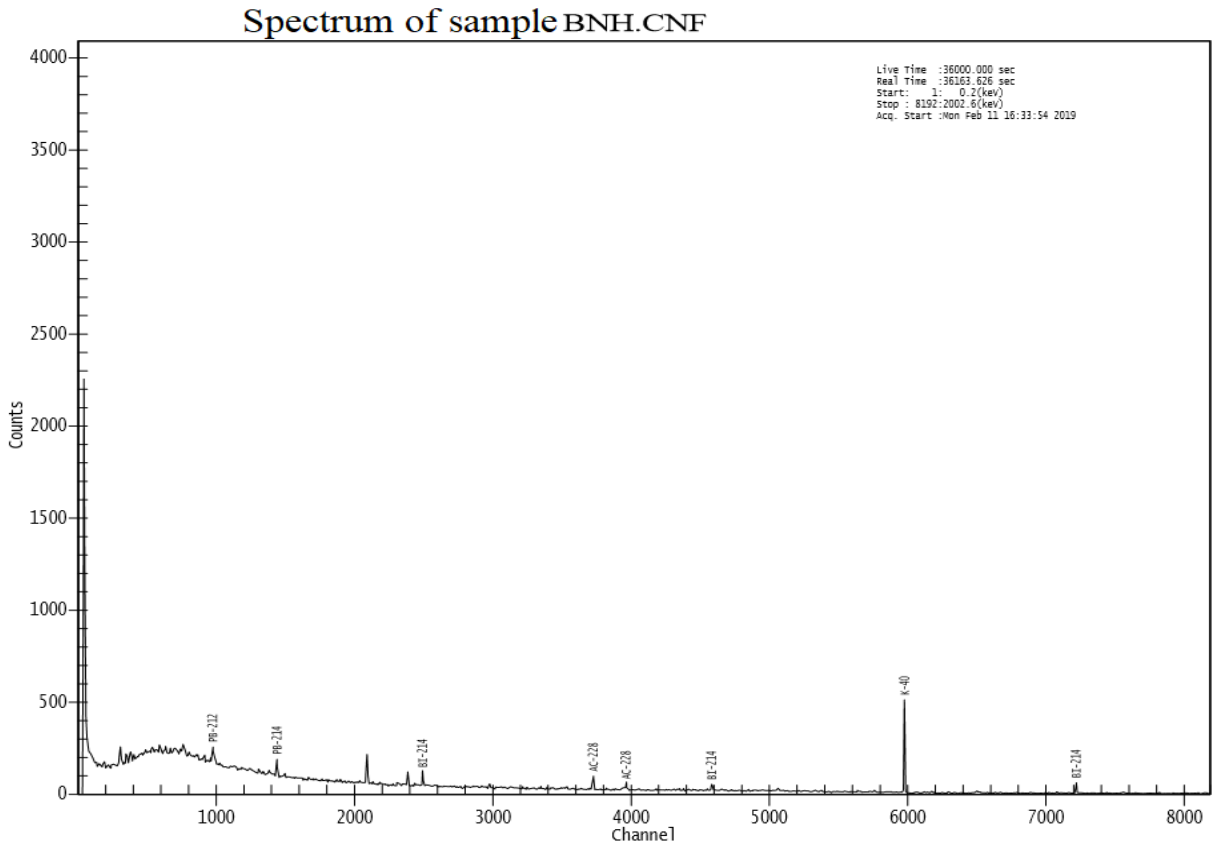


Figure 15: Spectrum of Zuwaye Lake water BNH

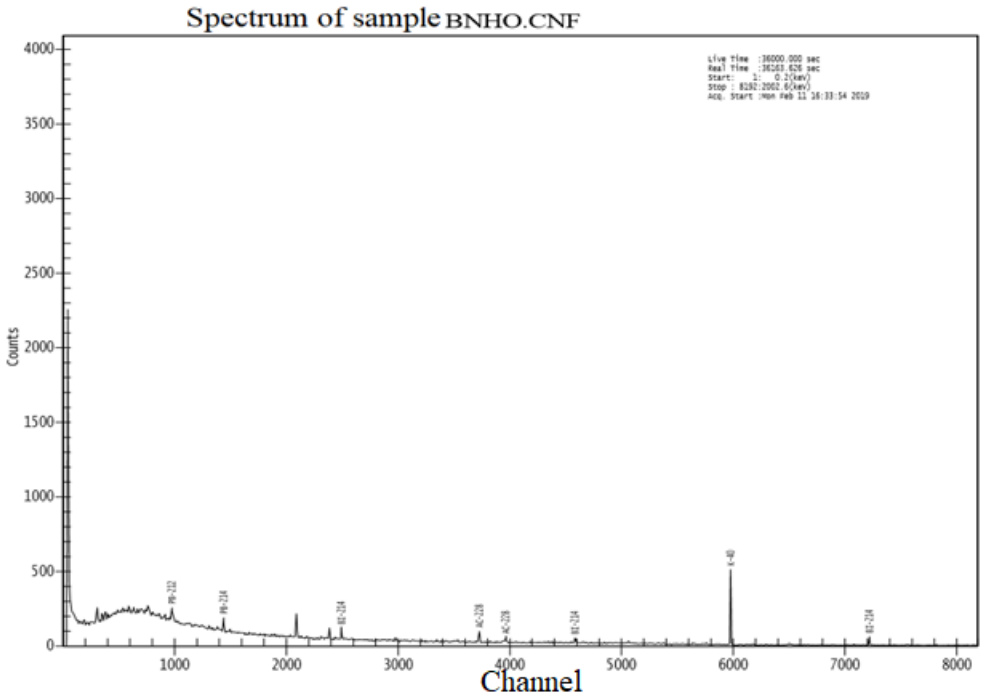


Figure 16: Spectrum of Zuwaye Lake Water sample BNHO

6.2.2 Radiological Effect

Table 5: **Radium equivalent, absorbed dose rate, annual effective dose rate, external hazard index, and internal hazard index in different Zuwaye Lake samples.**

Sample ID	Raeq Bq/L	Dr(nGy/h)	Dout(mSv/y)	Din(mSv/)	AEDE	Hex	Hin	I_γ
BNSH1	20.48	10.101	0.0124	0.069418	0.082	0.055	0.06	0.16
BNHO	22.76	11.161	0.0137	0.076701	0.092	0.0614	0.07	0.17
BNHO2	22.02	10.802	0.0133	0.074239	0.086	0.0594	0.07	0.17
BNSHS	20.876	10.307	0.0127	0.070837	0.084	0.0564	0.06	0.16

Three naturally occurring radionuclides (^{238}U, ^{232}Th and ^{40}K) were determined in the LakeZuwaye water samples listed in the table 4. Activity concentration of ^{238}U Lake Zuwaye Water ranged from 2.94±0.25 Bq/L to 3.02±0.27 Bq/L with an average value of 2.99±0.267 Bq/L, while ^{232}Th ranged from 6.9±0.73 Bq/L to 8.39±0.82 Bq/L also with an average value of 7.5 ±0.76 Bq/L and the activity concentration of ^{40}K ranged from 98.12 ± 4.16154 to 101.99 ± 4.91556 with an average value 99.77±4.80Bq/L.

The activity concentration from the studied areas were found to be below WHO guidelines of 10 Bq/L for ^{238}U and higher than 1 Bq/L for ^{232}Th. The World Health Organization recommended level of ^{40}K in water meant for drinking is unavailable. This is as a result of the maintenance at a constant amount of potassium in the human body at a fixed body mass.

Chapter Seven

7. Conclusions and Recommendations.

7.1 Conclusions

The activity concentrations of U-238, Th-232 and K-40 calculated in the soil, Lake Water and Coffee samples collected from West-Arsi Zone, Shashamene, and Zuwaye, Ethiopia respectively, was measured using high purity Germanium spectroscopy (HPGe). All results indicated that the value of measured activity concentration in the soil samples of ^{238}U, ^{232}Th, ^{40}K and^{137} Cs were varies from 24.10 to 56.92, 67 to 151.23, 534.58 to 979.57 and 1.13 to 7.4Bq/Kg respectively. The average activity concentration of ^{238}U, ^{232}Th and^{40}K was 45.83, 100.525, 845.65Bq/Kg, respectively. These values were higher than the recommended world average values for ^{238}U , ^{232}Th , ^{40}K , respectively which are 35, 30 and 400 Bq/Kg for ^{238}U , ^{232}Th , ^{40}K respectively [UNSCEAR, 2010].

It is also observed that the measured activity concentration of ^{40}K exceeds markedly the values of both Uranium and Thorium, as it is the most abundant radioactive element under consideration. Moreover the excessive use of the Potassium containing fertilizers in the area adjacent to the sampling sites may contribute to the higher values of ^{40}K activity.

During our experimental activities our detector could identified only potassium-40 in our Coffee samples. All coffee samples had ^{40}K concentrations that were higher than the acceptable value (412 Bq/kg) [UNSCEAR, 2010]. The mean activity concentrations of ^{40}K in measured coffee samples were 616.185 higher than the recommended limit.

Three naturally occurring radionuclides (^{238}U, ^{232}Th and ^{40}K) were determined in the lake Zuwaye water samples listed in the table 4. Activity concentration of ^{238}U Lake Zuwaye ranged from 2.94±0.25 Bq/L to 3.02±0.27 Bq/L with an average value of 2.99±0.267 Bq/L, while ^{232}Th ranged from 6.9±0.73 Bq/L to 8.39±0.82 Bq/L also with an average value of 7.5 ±0.76 Bq/L and the acidity concentration of ^{40}K ranged from 98.12 ± 4.16154 to 101.99 ± 4.91556 with an average value 99.77±4.80Bq/L.

The activity concentration from the studied areas were found to be below WHO guidelines of 10 Bq/L for ^{238}U and higher than 1 Bq/L for ^{232}Th. The World Health Organization recommended level of ^{40}K in water meant for drinking is unavailable. This is as a result of the maintenance at a constant amount of potassium in the human body at a fixed body mass. The annual effective dose calculated for the lake varied from 0.081 to 0.09 mSv/y with an average of 0.0858 mSv/y. This value when compared with the ICPR standard value of $1mSvyr^{-1}$is very low, hence, it can be concluded that the radionuclide concentration of water supply in Lake Zuwaye is negligible and poses no radiological hazard to the public.

Calculations of Radium equivalent activity (Raeq), dose rate (*D*), annual effective dose equivalent (AEDE), the external hazard index (*H*ex) and radioactivity level index were done for all soil, coffee and lake water samples. Dose rate (*D*), annual effective dose equivalent (AEDE), the external hazard index (*H*ex) and radioactivity level index in the measured soil values were higher than the recommended world average values and the soil was not safe for use. Only radium equivalent activity, the external hazard index (*H*ex) and internal hazard index values were lower the world average values. All values were lower than the recommended world average values and the coffee was safe for use.

It can be concluded that all the samples investigated at Lake Zuwaye posed no health risk to the public from consumption of fish, ingestion of water and use it for irrigation purposes. It is hoped that results obtained in this study would serve as a useful source of information for further study in other parts of the country to assess the doses of radioactivity that the population is exposed and also serve as a baseline data for future studies.

7.2 Recommendations

As we all know that from natural radioactivity our health condition can be disturbed depending on the energy and the accumulation of radiation in our body. Once our health condition disturbed we can face many problem economical, socially and psychologically. The peoples interest on constructing new building, road, cement factory and expanding of urbanization increase from time to time. To do this they dig earth for the purpose of finding construction material and raw materials for cement factory. While doing these if the care is not taken we can pollute our environment. So before we encounter this entire problem it is better to protect our environment. The concerned bodies have to aware the society regarding of radiation.

It is also recommended that further studies on radioactivity should be carried out to cover the remaining area of the lakes, food stuff, and soil in the different county side and radon gas in air to obtain a baseline data for future studies. Gathering and storing the obtained information would help in formulating radiological map and national guidelines for radioactivity levels in food, water and other environmental samples.

This work can be used as a baseline for future studies and the data obtained in this study may be useful for natural radioactivity mapping and also be used as a reference data for monitoring possible radioactivity pollutions in the environment for future.

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