Addis Ababa University College of Natural and Computational Science Department of Physics

Measurement of Radioactivity Levels and Health Risk in Maize and Sorghum Grown in the Bale zone Oromia Region, Ethiopia, by Using Gamma Ray Spectrometer (HPGe).

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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 Msc. in nuclear physics.

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Approval Sheet

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Declaration

I undersigned declare that, this research paper entitled ”Estimation of Radioactivity Levels and Health Risk in Maize and Sorghum Samples Grown in the Bale zone Oromia Region, Ethiopia, by Using Gamma Ray Spectrometer (HPGe)” is my own original work and it has not been submitted for the award of any academic degree or the like in any other institution or university, and that all the sources I have used or quoted have been indicated and acknowledged.
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Acronyms and Abbreviation

IAEA : International Atomic Energy Agency.
TECDOC : Technical Committee Document.
UNSCER : United Nations Scientific Committee on Effect of Atomic Radiation.
HPGe : High Purity Germanium.
NORM : Naturally Occurring Materials.
UNEPA : United State Environmental Protection Agency.
DNA : Deoxyribonucleic Acid.
RDANRC : Radiation Dose Assessment National Research Council.
TF : Transfer Factor.
NaI(TI): Sodium Iodized Activated by Thallium.
BDL : Below Detectable Limit.
GM : Geometric Mean
USEPA : United State Environmental Protection Agency.
GRS : Gamma Ray Spectrometry.
MCA : Multi-Channel Analyzer.
DSA : Digital Signal Analyzer.
FWHM : Full Width at Half Maxima.
FWTM : Full Width at Tenth Maxima
ICRP : International Committee for Radiation Protection.
CFSVA : Comprehensive Food Security and Vulnerability Analysis.
FAOSTAT: Food Agriculture Organization Corporate Statistical Data Base.

Contents

Acronyms and Abbreviation i
List of Table ii
List of Figure iii
Abstract iv
Acknowledgment vi

1 Introduction 1
1.1 Background of the study 1
1.2 Statements of the problem 3
1.3 Objectives of the study 5
1.3.1 General objective 5
1.3.2 Specific objectives 5
1.4 Significance of the study 6
1.5 Limitation of the Study 6

2 The Studies of Environment Natural Radioactivity 7
2.1 Radioactivity in the Natural Environment 8
2.1.1 Sources of Radiation in the Environment 9
2.2 Ingestion Exposure 9
2.3 Individual Dose Assessment 11
2.4 Radioactive Decay 11
2.4.1 Uranium decay series 12
2.4.2 Thorium 232 decay series 14
2.4.3 Potassium decay series 15
2.5 Equilibrium in radioactive decay chains 15
2.6 Gamma ray interactions in the detector 16
2.6.1 Photoelectric effect 17
2.6.2 Compton scattering 17
2.6.3 Pair production 17
2.7 Importance of the Photo-Electric Effect, the Compton Effect and Pair Production in the Absorption of γ Rays 18
2.8 Gamma Ray Spectroscopy 18

2.9 Semiconductor Detectors 19

3 Methodology of the study **21**

3.1 HPGe Spectroscopy 21

3.2 Principles of Gamma Spectrometry 22

3.2.1 HPGe Experimental Setup and Measurements 24

3.3 System Calibration 26

3.3.1 Energy Calibration 26

3.3.2 Efficiency Calibration 27

3.4 Efficiefncy and Energy Resolution 28

3.5 Background Radiation 31

3.6 Sample Area 32

3.7 Sampling and Sample preparation 33

3.8 Activity concentration Level Equation 34

3.9 Radiological Hazard Indices Equations 35

3.9.1 Radium Equivalent Equation (Raeq) 35

3.9.2 Internal Hazard Index (Hin) 35

3.9.3 Ingestion Effective Dose 36

4 Result and Discussion **37**

4.1 measurement of Radioactivity level and Health Hazard index in maize samples using HPGe gamma spectrometer 37

4.1.1 Introduction 37

4.1.2 Result and Discussion 37

4.2 measurement of Radioactivity level and Health Hazard index in sorghum samples using HPGe gamma spectrometer 41

4.2.1 Introduction 41

4.2.2 Result and Discussion 42

5 Conclusions and Recommendations **45**

5.1 Conclusions 45

5.2 Recommendations 45

References 47

Appendix 53

List of Table

4.1.Activity concentration of NORM in maize sample (2021).........................40
4.2.Radiological hazard indices..40
4.3.Activity concentration of NORM in sorghum samples 2021......................42
4.4.Activity concentration of NORM in sorghum samples...............................43

List of Figure

2.1.Ingestion exposure rate......10
2.2.Uranium decay series......14
2.3.Thorium decay series......15
2.4.Potassium decay series......16
2.5.Creation of depletion region in semi conductor crystals......20
3.1.Block diagram of basic gamma spectrometer system with MCA......22
3.2.HPGE detector and its shielding (Pb+Cd+Cu)......25
3.3.Gamma spectrometry mounted with HPGE detectors and DSA (ERPA)......26
3.4.Figure calibration curve by standard check sounds the region of interested......27
3.5.Efficiency curve by standard check sources in the region of interest......28
3.6.Counts Vs Pulse height......30
3.7.Energy resolution......31
3.8.Back ground radiation......33
3.9.Sample area in madda walabu wereda,2021......34
3.11.Sample Preparation......35
4.1.Spectrum of maize sample collected from Bale......39
4.2 Histogram expression of Activity concentration of radionuclide in maize samples......40
4.3.Spectrum of sorghum sample collected from Bale 2021......43
4.4.Histogram expression of activity concentration in sorghum samples,2021......43

Abstract

Naturally occurring radioactive materials (NORMs) are ubiquitous in our environment. ^{238}U, ^{232}Th and ^{40}K are the predominant NORMs in the soil that are found to have been existing for billions of years. Naturally occurring radioactive material (NORM) is a commonly used term to describe material containing primordial radionuclide's. Radionuclide's enter the human body through complex mechanism including ingested foodstuffs via the food chain from natural sources.The objective of the measurement is aimed at estimating the activity concentrations and the conscience of these in some areas of Bale Zone in Warre and Wadduma Kebele . The activity concentrations of five (5) maize samples and four (4) sorghum samples from the areas were determined using High purity Germanium (HPGe) detector. The activity concentration for ^{238}U, ^{232}Th and ^{40}K for the maize samples ranged from $(3.326 \pm 0.00) Bq/Kg$ to $(4.034 \pm 0.00) Bq/Kg$, $(6.260 \pm 0.00) Bq/Kg$ to $(14.257 \pm 0.00) Bq/Kg$ and $(80.935 \pm 4.004) Bq/Kg$ to $(131.092 \pm 6.038) Bq/Kg$ respectively. Hence the mean concentration obtained for each radionuclide's were $3.73 Bq/Kg$ for ^{238}U, $10.838 Bq/Kg$ for ^{232}Th and $109.370 \pm 4.29 Bq/Kg$ for ^{40}K. This showed that the values for ^{238}U and ^{232}Th were higher than activity concentrations of world reference value $0.02 Bq/kg$ and $0.003 Bq/kg$, for ^{238}U and ^{232}Th, of these radionuclides [1].The activity concentrations of radionuclides in ^{238}U,^{232}Th and ^{40}K measured for sorghum were ranged between 2.151 to $6.753 Bq/kg$, 3.349 to $20.218 Bq/kg$ and 104.184 ± 4.758 to $122.434 \pm 2.413 Bq/kg$, respectively. The average activity concentrations of ^{238}U, ^{232}Th, and ^{40}K in sorghum were $4.389 Bq/kg$, $11.295 Bq/kg$, and $112.636 \pm 4.088 Bq/kg$, respectively. The activity concentration of ^{238}U and ^{232}U implies that, higher activity concentrations than the world reference value $0.02 Bq/kg$ and $0.003 Bq/kg$, for ^{238}U and ^{232}Th, of these radionuclides [1].The ingestion effective dose in mSv/y for adult to specific activity of ^{238}U, ^{232}Th and ^{40}K in maize samples were varied from $0.24 mSv/y$ to $0.38 mSv/y$ with the average value of $0.32 mSv/y$ which is slightly greater than world reference value 0.3 mSv/y[2].

And the ingestion effective dose in mSv/y *for specific activity of* ^{238}U, ^{232}Th *and* ^{40}K *in sorghum samples were varied from* $0.2 mSv/y$ *to* $0.79 mSv/y$ *with the average value of* $0.422 mSv/y$ *which is higher than world reference value* $0.3 mSv/y$ *for ingestion[2].*

Keywords : *Radioactivity, Activity concentration level,health risk, Germanium Detector /HPGe spectrometer.*

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Chapter 1

Introduction

1.1 Background of the study

AS the studies present radioactivity is a natural part of our environment. Present-day Earth contains all the stable chemical elements from the lowest mass (H) to the highest (Pb and Bi). Every element with higher Z than Bi is radioactive. The earth also contains several primordial long-lived radioisotopes that have survived to the present in significant amounts. Some radioactive isotopes, for example 14C and 7Be, are produced continuously through reactions of cosmic rays (high-energy charged particles from outside Earth) with molecules in the upper atmosphere. As per Nuclear Science Guide in radioactive processes, particles or electromagnetic radiation are emitted from the nucleus[3]. The most common forms of radiation emitted have been traditionally classified as alpha (α), beta (β), and gamma (γ) radiation. Nuclear radiation occurs in other forms, including the emission of protons or neutrons or spontaneous fission of a massive nucleus. Of the nuclei found on Earth, the most majority was stable. This is so because almost all short-lived radioactive nuclei have decayed during the history of the Earth. The earth and the other planets of our solar system formed 4.5 billion years ago out of material rich in iron,carbon,oxygen,silicon and other medium and heavy elements.A few of the radioactive elements have half-lives that are long compared with the age of the Earth,and so we can still observe their radioactivity.This radioactivity forms the major portion of our natural radioactive environment,and also probably responsible for the inner heating of the terrestrial planets. Radioactive decay was change one nucleus to another if the product nucleus has a greater nuclear binding energy than the initial decaying nucleus. The difference in binding energy (comparing the before and after states) determines which decays are energetically possible and which are not. The excess binding energy appears as kinetic energy or rest mass energy of the decay products. Food crops from contaminated environment may accumulate radioactivity that could form a direct route of exposure to human population when consumed[4].

Therefore, the knowledge of radioactivity levels in grown food crops is very important in order to establish the doses received by populations [4]. People are exposed to radioactivity in soil, plant and even in other environment that is raised from uranium, thorium and potassium 40 radionuclide's. Environmental radioactivity and the associated external exposure due to gamma radiations depend primarily on the geological, geographical and human activities [5]. In this study the activity concentration of 238_U, 232_{Th} and 40_K radionuclide's were estimated in nine samples, five maize samples and four sorghum samples collected from some parts of Bale Zone, Madda Walabu woreda, Warre and Wadduma kebele of Ethiopia using High Purity Germanium Detector (HPGe). Naturally occurring radioactive material (NORM) is a commonly used term to describe material containing primordial radionuclide's. Of these, the radioisotopes in the uranium, actinium and thorium decay series and potassium are important for radiation protection considerations. Radiation exposure due to NORM has the potential to cause radiological harm to humans and the environment. Scientific studies have confirmed the association between inhalation of short-lived radioactive progeny of radon gas and lung cancer. This thesis investigates such measurements, with an emphasis on the regulation of naturally occurring radionuclide materials (NORM) are present in the environments; earth crust, soil, water, air, plants.These natural radionuclide's originating in uranium , thorium series and potassium.The human body digests inhales or absorbs radiations and sometimes certain radioactive elements with minerals the body is lacking [6]. The excessive uses of nuclear energy in all branches of life increase the problem. Activities related to the extraction and processing of ores can also lead to increased levels of NORM. These activities include uranium mining and milling; metal mining and melting; phosphate production; coal mining and power generation from coal burning; oil and gas drilling; rare earth and titanium oxide industries; zirconium and ceramic industries; and applications using naturally occurring radionuclides (typically isotopes of radium and thorium) [7]. The activity of radionuclide is cumulative and its effects cause damage, both in the short and long-term. This is the cause of allergic response and unexplained illnesses. Agricultural plants absorb radiations from the soil, which in turn transferred to each living organism [10]. The majority of people around the world depend on cereals grains, legumes and foodstuffs in their nutrition.

It is necessary to estimate the natural radioactivity concentrations in our daily food. However, a study of natural radioactivity concentration in food stuff have been performed in several countries [9]. In addition, human bodies are also internally exposed because of the existence of certain radionuclides in the body such as 40_K . From internal exposure, inhalation and ingestion the average values are 1.2 and 0.3 mSv respectively [8]. This study was investigate the natural radioactivity concentration of Uranium, Thorium series and Potassium in cereals grains and also the effective ingestion dose and health risk to the consumers were estimated. This provided a base line to investigate food security and suitability to human beings give confidence to research areas populations.

1.2 Statements of the problem

Natural radioactive decay like ^{238}U and ^{232}Th series in addition to radionuclide that occurs singly such as ^{40}K is found in the atmosphere and the earth in varied levels. The radioactivity in the agricultural land may transfer to the plants around. The radionuclide available in the environment is transferred to plants by two ways, first of which is the indirect method uptake from soil through roots. When food plants are grown in a polluted soil, the activity is switched to the roots from the soil and then in shoots. At the end, it transfers to the human diet [11]. This radionuclide can get into the plants during mineral uptake along with the nutrients and accrue in different areas and even it could reach the edible parts [12]. Second, the direct method absorption; it occurs through plant organs. A variety of workers have reviewed the presence of radioactivity in the plant organs [13].
Since the discovery of radiation, more than a century of radiation research has yielded extensive information on the biological mechanisms by which radiation can affect health. It is known that radiation can produce effects at the level of cells, causing their death or modification usually because of direct damage to deoxyribonucleic acid (DNA) strands in a chromosome. If the number of damaged or killed cells is large enough, it may result in organ dysfunction and even death. Also, other damage to DNA may occur that does not kill the cell. Such damage is usually repaired completely but if not, the resulting modification known as cell mutation was be reflected in subsequent cell divisions and may ultimately lead to cancer[14].

If the cells modified are those transmitting hereditary information to descendants, genetic disorders may arise. Information on biological mechanisms and on heritable effects is often gained from laboratory experiments. On the basis of the observation of their occurrence, health effects following radiation exposures are defined here as either early or delayed health effects. Generally, early health effects are evident through diagnosis of clinical syndromes in individuals, and delayed health effects such as cancer through epidemiological studies by observation of increased occurrence of a pathology in a population[14].

Maize originated in Central America and was introduced to West Africa in the early 1500s by the Portuguese traders [15].It was introduced to Ethiopia during the 1600s to 1700s [16].Maize is the popular source of food in the world and also in Ethiopia. In Ethiopia, annual maize production is 7.8 million tones with an average yield of 3.6 tonne per hectare (tha^{-1}) in 2016 the highest of any cereal in the country (Food and Agriculture Organization Corporate Statistical Database [17]. Currently, 66% of cereal farming households in Ethiopia cultivate maize on 2.1 million hectares (ha), making it the second most widely cultivated cereal in the country after teff. It is estimated that each household owns around 1 hectar of crop land, of which at least half is allocated for maize cultivation in major maize-growing areas. Subsistence maize farming accounts for more than 95% of the total maize area and production, with 75% of all maize produced being consumed by the farming household [18]. So the knowledge of the concentrations and distributions of natural radionuclide's, such as ^{40}K, ^{238}U, and ^{232}Th and their decay products and radiological associated risks in maize is essential for such studies and can be useful to estimate the degree of human risk associated with the ingestion of radionuclide's in maize through the food chain and to establish a baseline database of radionuclide's concentration, in order to monitor the possible variations in the agricultural environment radioactivity due to human activity and others. Sorghum (Sorghum bicolor L. Moench) is the sixth most planted crop in the world, and it is one of the most important cereals used as a staple food for those primarily living in arid and semiarid areas [19]. In Africa, it is the second most important cereal in which around 300 million people depend on their daily consumptions. It is consumed mostly in northern China, India, and southern Russia, where about 85% of the crops are consumed directly as human food [20].

Sorghum has greater drought tolerance, soil toxicities, and temperature variation than other cereals and requires minimal fertilizers for cultivation, thus playing a critical role for food security in some semiarid areas of Asia, Africa, and Latin America [21]. It is considered one of the potential crops to alleviate the challenges of recurrent drought in Africa. In Ethiopia, sorghum is the third (3rd) most important staple cereal crop after teff and maize [22]. Sorghum crop is grown in almost all regions of Ethiopia and used as a staple food crop on which the lives of millions of poor Ethiopians depend on it. Sorghum is used in various ways in our country; the grain is used for human foods such as injera, bread, porridge, Nifro, infant food, syrup, and local beverage known as Tella and Arekie. And also, the leaf and stalk are used for animal feed, and further, the stalks are used for the construction of houses and fences and as fuel wood [23].

1.3 Objectives of the study

1.3.1 General objective

The main objective of this study was to estimate the concentration level and health risk of radioactivity in food grain (cereals),grown at Meda Welabu woreda,Bale Zone,Oromia,Ethiopia,using a Gamma Ray-spectrometer($HPGe$) .

1.3.2 Specific objectives

This study has the following specific objectives:

- To measure the activities level of ^{238}U, ^{232}Th and ^{40}K in the maize and sorghum samples.
- To calculate the health risk associated with the radioactivity in samples.
- To forward recommendation for the concerned body based on the finding of the results.

1.4 Significance of the study

In different countries there are several documents accessed on the natural radioactivity that gives information for their community.Such documents were not familiar for our country except some works that are performed by radiation protection agency, oromia science technology and communication authority and some graduated students working in this area. The exposure to radioactivity has health hazardous upon human being.Population around the study area benefit from this study. This study determined the concentration of radionuclide found in the study area and the radiological effects on the human being.

1.5 Limitation of the Study

This study was faced some limitations, due to financial and time constraints, it was not feasible to cover the entire large number area for the study. During this study it was difficult to collect large no of samples due to the time is not cultivation time and the samples were collected from household from the area, this may cause not to get enough samples from the area. Therefore, the strategies were used to success the study, the researcher was minimize the limitation of the study by assuring the farmers that the sample they provided were completely for the purpose of research and used only for academic purpose and manage its time and budget as well.

Chapter 2

The Studies of Environment Natural Radioactivity

Naturally occurring radioactive materials are found everywhere in our environment. ^{238}U, ^{232}Th and ^{40}K are the predominant NORMs in the soil that are found to have been existing for billions of years, in the sense that ^{238}U found to have a half-life of 4.5 billion years, 14 billion years for ^{232}Th and 1.3 billion years for ^{40}K, high concentration of them in foodstuffs are considered to be hazardous to the public health. Radionuclide's can be transferred to plants together with the water, minerals and other nutrients while plants uptake them by the root system for their growth and development [24]. However, there are some other factors such as type of clay, crop type, physiochemical properties of radionuclide's, pH of the soil, climate condition and types of fertilizer that influence the uptake of radionuclide's by the roots system to the plant [25]. Therefore, soil plantfoodstuff is the main corridor for radionuclide's to be transmitted to living organisms [24], while inhalation; injection and skin absorption also form other pathways for radionuclide's to get an access into human body. Thus, assessment of radioactivity in foodstuffs is considered to be one of the standard means for calculating radiological dose to humans. Maize which also known as Corn is one of the famous types of food that were said to have being consumed by a large number of populations in the globe. Furthermore, about 208 million people in sub-Saharan Africa were estimated to have dependent upon maize as their staple food stuff [26]. It is one of the most abundant food crops in Ethiopia which makes it to be available, cheap and affordable to most of the compared to teff and wheat. That it can promote food security, social amenity and socioeconomic wellbeing to the local farmers within the country. Considering the aforementioned facts, assessment of radionuclide's contents in maize is very important for Ethiopia and African population, as such, maize consumption is one of the ingestion pathway through which radionuclide's can be transferred, relocated and accommodate in the human body.

The present study focuses on the measurement of 238_U, 232_{Th} and 40_K radionuclide's concentrations in Ethiopian maize by using HPGe gamma-ray spectrometry. Annual effective dose and health risk via maize consumption was also estimated. The knowledge of natural radioactivity in man and his environment is very important since naturally occurring radionuclide's are the major sources of radiation exposure to man [27]. Radionuclide's enter the human body through complex mechanism including ingested foodstuffs through the food chain from natural sources. Ingestion of radionuclides through food intake accounts for a substantial part of average radiation doses to various organs of the body and also represents one of the important pathways for long term health considerations. Radionuclide have always been present in food at various levels depending on factors such as radioactivity contents in soil and the transfer characteristics from the environment medium to food for other regions the dose values are lower stuff and hence to man.The status of the soil on which food crops are grown determines, to a significant extent, the quality of food crops produced, the season of the year also determines to a great extent the magnitude of contamination of different foods [28].

2.1 Radioactivity in the Natural Environment

The radioactive nuclei, or radionuclides, found naturally on Earth can be grouped into three series headed by ${}^{238}U$,${}^{235}U$ and${}^{232}Th$ plus several isolated beta-particle emitting nuclei, most prominently ${}^{40}K$. The most interesting of the series is the 238_U series which decays via a chain containing 8 alpha decays and 6 beta decays to lead-206. This chain includes the longest-lived isotopes of radium and radon,radium-226 and radon-222, respectively. In each of the three chains the parent nucleus has a much greater life time than does any of the progeny. Therefore, a steady state is established in which, for a given sample of material, each member of the series has the same activity aside from deviations due to differences in chemical properties, which cause different elements to be transferred at different rates into or out of a given sample of material. Including all the succeeding decays, the total activity in the ${}^{232}Th$ and ${}^{238}U$ series is, very roughly, ten times the activity indicated for ${}^{232}Th$ and ${}^{238}U$ alone [29].

2.1.1 Sources of Radiation in the Environment

There are two main sources of radiation in the environment:

a. Naturally occurring (background radiation):Which is naturally present in our environment since the birth of the planet, includes:

1. Cosmic radiation: Radiation from space (sun and stars) and bombarding earth, and all living things on it.

2. Terrestrial radiation: uranium and the decay products of uranium, such as thorium, radium.

3. Radon gas: the largest natural source of radiation exposure to humans, it exists in air, water and soil.

4. Natural internal radiation in human body: the radiation comes from radioactive materials occur naturally in the human body. Potassium and Carbon are the primary sources of internal radiation exposures. The Potassium ^{40}K isotope enters the human body through the food chain. Carbon ^{14}C (represent 0.23 weight of the human body) enters the body both through the food chain and breathing.

b. Artificial sources (man made radiation): Artificial radiation sources are identical to the natural radiation in their nature and effect. The most important sources are:

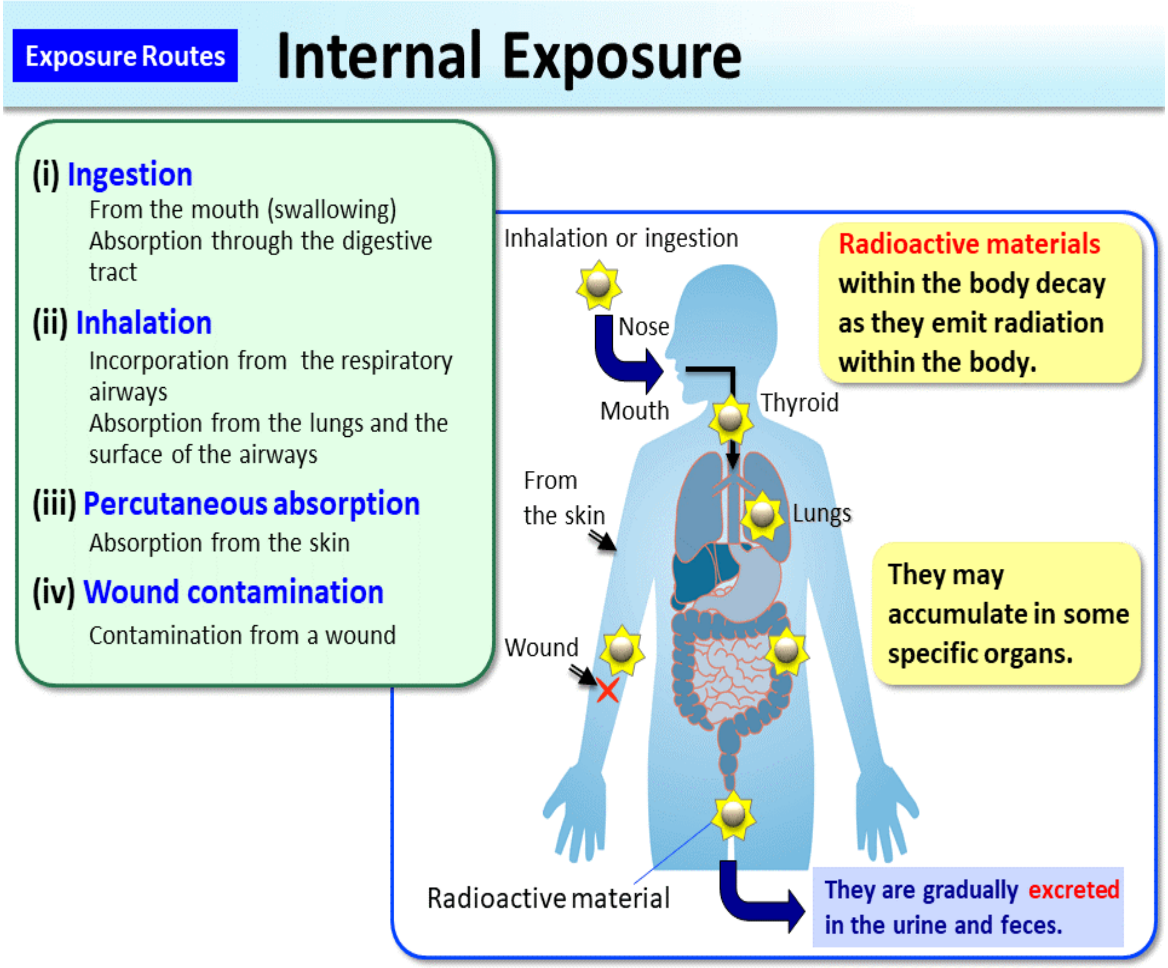
1. Medical procedures: diagnostic X-rays, nuclear medicine, and radiation therapy.

2. Consumer products: tobacco (polonium 210), building materials, fuels (gas, coal, etc.).

3. Concern isotopes: Cobalt (60Co),Cesium(137Cs),Americium(241Am), and others[30].

2.2 Ingestion Exposure

Ingestion exposure occurs when radionuclide's in the environment enter food chains. This component and that of external exposure are usually the significant and continuing sources of exposure following releases of radionuclide's to the environment. Radionuclide's released to the atmosphere may deposit onto both terrestrial and aquatic surfaces, for which different calculation methods are required. In the general case, doses from the ingestion of natural radionuclide's in foods and drinking water have been estimated from measured concentrations of the radionuclide's in body tissues or organs. For 40_K, metabolic balance maintains body levels irrespective of intake amounts. For uranium and thorium series radionuclide's, however, this is not the case, and the concentrations in foods, water and total diet have been useful for determining geographic variations in the body burdens [27].



(i) Ingestion
From the mouth (swallowing)
Absorption through the digestive tract

(ii) Inhalation
Incorporation from the respiratory airways
Absorption from the lungs and the surface of the airways

(iii) Percutaneous absorption
Absorption from the skin

(iv) Wound contamination
Contamination from a wound

Radioactive materials within the body decay as they emit radiation within the body.

They may accumulate in some specific organs.

They are gradually excreted in the urine and feces.

Figure2.1 Ingestion exposure route[14].

The procedure for calculating radiation dose from ingested radionuclide's is similar to that for calculating the dose from inhalation. Ingestion is defined as the swallowing of radionuclide's in water (or other liquids and in food). Absorption of radionuclide's from the gastrointestinal tract to blood is affected by such factors as chemical form and interactions with other food in the gastrointestinal tract. Given such variable absorption, the most practical means of estimating the dose is to use a model with average absorption kinetics and to calculate the dose to organs based on normal physiological processes. Inhaled particles brought out of the lung by ciliary action are also ingested[27].

From that point, the radiation dose is determined as if the radionuclide were ingested, not inhaled. The radiation dose to the gastrointestinal tract is largely due to the radionuclide activity within the contents of the bowel. The largest dose was be to organs that accumulate and retain the radionuclide. However, the variability in absorption of the ingested radionuclide in the gastrointestinal tract is responsible for the greatest uncertainty in the potential dose. Because radiation guidelines are usually conservative, it is likely that the commonly used absorption factors over estimate the amount of the radionuclide's that is absorbed and hence the organ dose [31].

2.3 Individual Dose Assessment

An individual dose assessment can be used in epidemiological studies, and it was needed for persons in the potentially exposed population who are interested in their own exposures. The preferred dose estimate from an individual risk assessment is the annual absorbed organ dose. The absorbed dose to an exposed individual is influenced by the exposure conditions for that person. As with the comprehensive assessment, environmental factors were influence the time-integrated activity that characterizes exposures, and an individual's lifestyle can influence the extent of contact with radionuclide's in the environment. The dependence of the absorbed dose on other factors such as gender, lifestyle, and diet must be considered, and the details applied was commensurate with the level of detail in the source term and the pathway analysis [32]. For example, the amount of time spent outdoors could affect the degree of exposure through inhalation or contact with radioactive materials deposited on soil or vegetation; the amount of milk consumed could affect the intake of radioactive doses.

2.4 Radioactive Decay

Radioactive decay is the emission of energy in the form of ionizing radiation. The ionization radiation that is emitted can include alpha, beta particles and/or gamma rays. Radioactive decay occurs in unbalanced atoms called radionuclides. When decay process takes place, a radionuclide transforms into a different atom or a decay products. The atoms keep transforming to new decay products until they reach a stable state and are no longer radioactive at the end. The most of radionuclides only decay once before becoming stable state. Those that decay in more than one step are called series radionuclides. The series of decay products under go to reach this balance is called the decay chain. Each series has its own unique decay chain. The decay products within the chain are always radioactive. Only the final, stable atom in the chain is not radioactive. Some decay products are different chemical elements. Every radionuclide has a specific decay rate which is measured in terms of half-life. Half-life is the time required for half of the radioactive atoms present to decay or transform [35].

2.4.1 Uranium decay series

The average content of uranium in the Earths crust in reference of igneous and sedimentary rocks, is approximately 2 to $4ppm$; and for thorium, 7 to $13ppm$. Its concentration depend on geochemical differences in environment during volcanic magma differentiation at the time of crystallization. Most of the uranium however, moves from the rock in hydro thermal solutions. It can easily oxidize and taken into solution after primary crystallization of volcanic magma. Therefore, uranium can be carried over long distances and redeposited in other rocks, in many cases, at higher concentration than the original occurrences. Uranium and thorium has no known biological function. They are chemo toxic, radio toxic and a carcinogen. Long term exposure to such nuclide increases the chances of getting health problems [36,37].

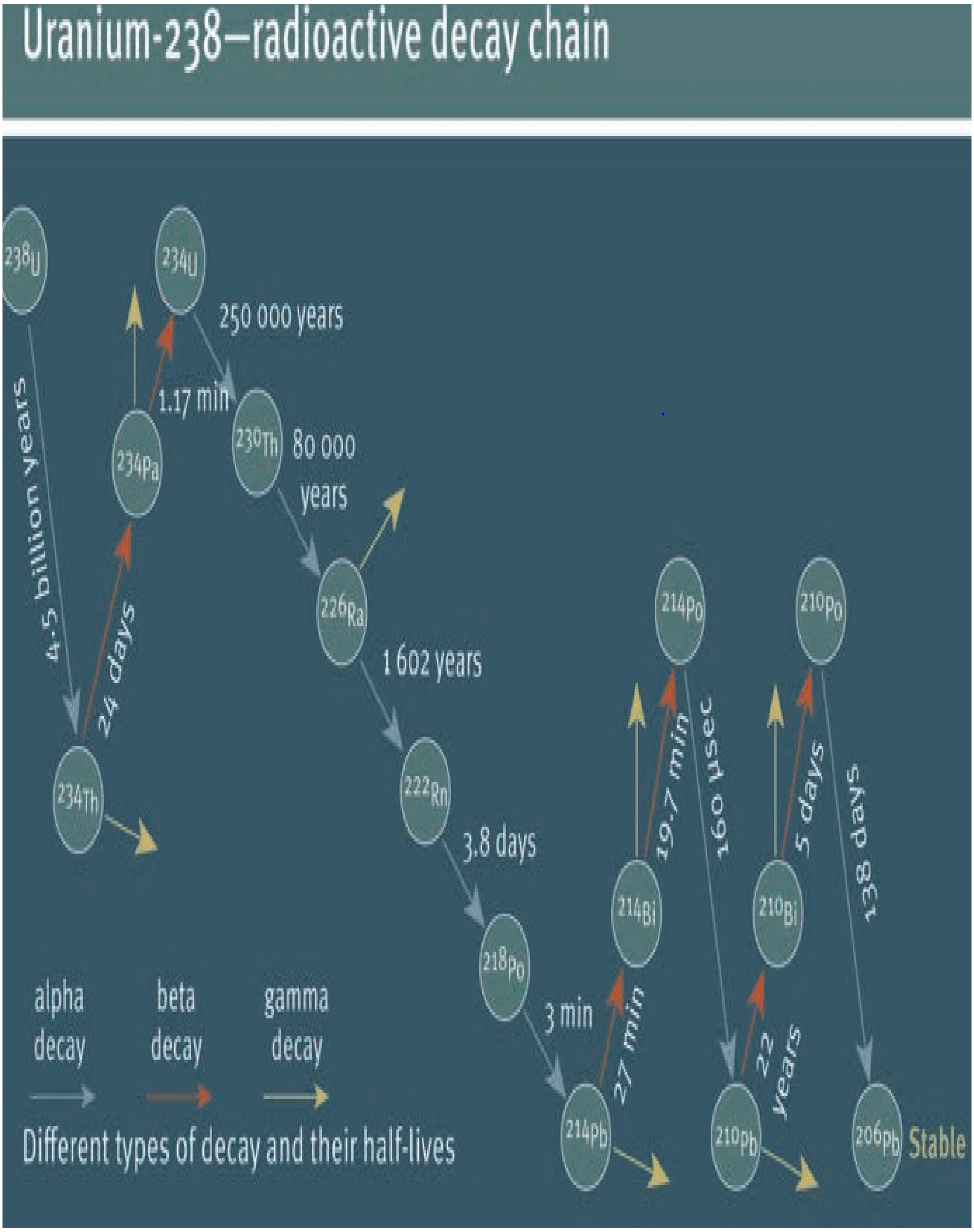


Figure 2.2.Uranium decay series[14].

2.4.2 Thorium 232 decay series

232_{Th}, is one of the most abundant heavy elements, that present in many minerals and contributes to the general background radioactivity. The minerals in which thorium mainly originated are igneous rocks, as disseminated and discrete minerals. It is not as soluble as uranium and is thus not as mobile in chemically solvent environments. Thorium is about three times more abundant than uranium in nature. The more important minerals that contain thorium are monazite, thorite, thorianite, uranothorite, and thorogummite [38]. Thorium and its radioactive progenies decay through the emission of alpha, beta and gamma until its stable end product, 208Pb [39,12].

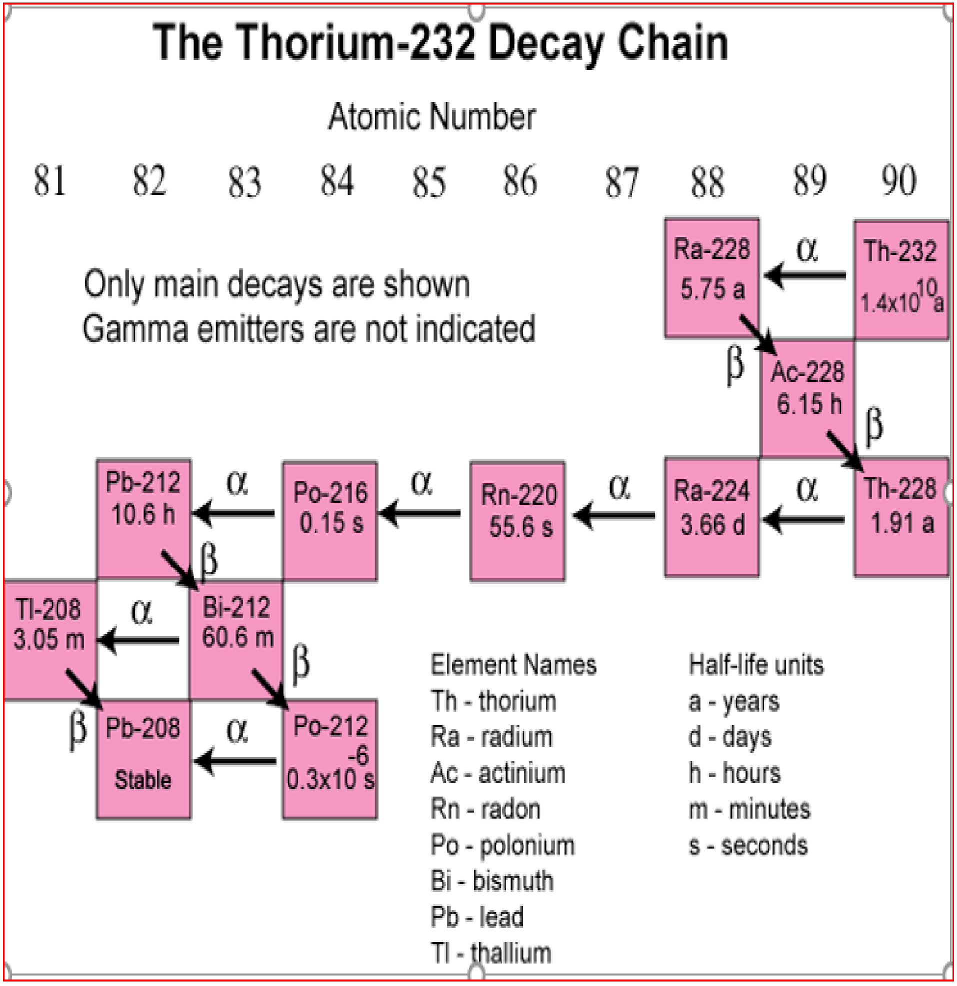


Figure2.3 Thorium Decay Series[58].

2.4.3 Potassium decay series

Potassium is a major constituent in many igneous rocks, and their petrographic classification where potassium40, (40_K) represents about 0.0119% of the total potassium inventory on Earth. It is one of the major radiation dose contributors from natural sources, with both electron and photon emissions as figure 4 below.

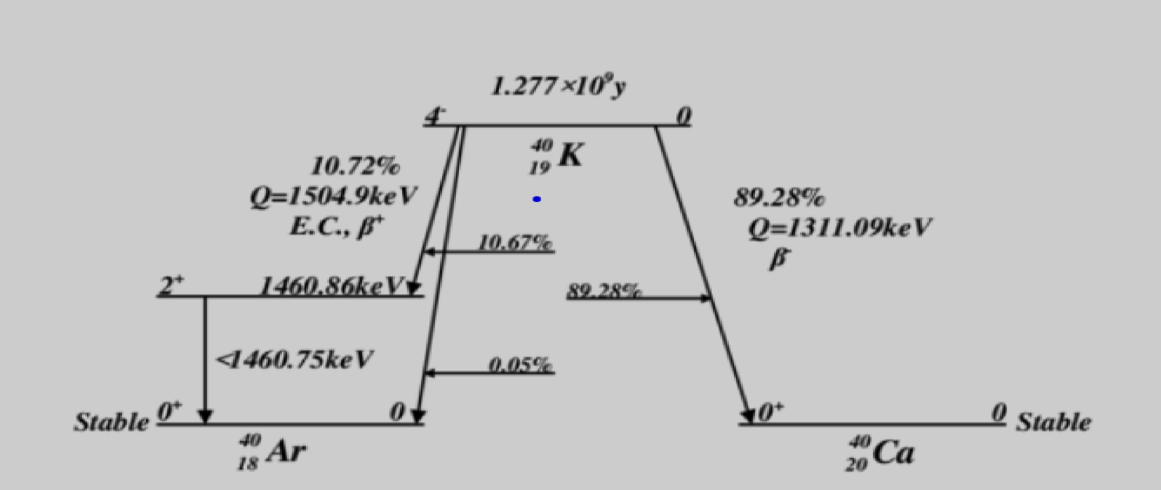


Figure2.4 Potassium decay series

Since potassium is a basic nutrient required for life, 40_K also cycles and accumulated freely in the biosphere. The combined dose from internal and external exposures of 40_K is estimated to be about 8% of the total radiation dose from all sources [39].

2.5 Equilibrium in radioactive decay chains

The radioactive decay chain and the decay of parent radionuclide was lead to the production of progeny products,and 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,

$$A \xrightarrow{\lambda_A} B \xrightarrow{\lambda_B} C \tag{2.1}$$

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

$$A_A = \lambda_A N_A \tag{2.2}$$

$$A_B = \lambda_B N_B \tag{2.3}$$

There are different types of equilibrium that depend on the comparison between the parent and progenys half-lives as follows (2.4) .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 parent (A) is much larger than the half-life of the daughter product. The daughter nuclide produces more radiation. 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:

$$\lambda_A N_A = \lambda_B N_B = ... = \lambda_n N_n \tag{2.4}$$

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 [40].

2.6 Gamma ray interactions in the detector

The process of detection of radiation is initiated when an incident gamma ray enters the crystal. The energy is totally absorbed under ideal conditions.

This is accomplished by three main interactions between gamma ray and the crystal. These interactions are photoelectric effect ,Compton scattering, and pair production[41].

2.6.1 Photoelectric effect

Photoelectric effect is an interaction between a low energy incident photon and an inner shell orbital electron. The energy of the incident photon is totally absorbed and used to eject the orbital electron from its orbital shell. This electron is known as photo electron. This happens if the incident photon exceeds the binding energy of the electron. The ejected electron leaves a vacancy in the inner orbital shell causing the emission of characteristic x-rays or auger electron when an outer shell electron full fills this vacancy [42,43].

2.6.2 Compton scattering

The interaction process of Compton scattering takes place between the incident gamma ray photon and an electron in the detector material [44]. The incoming gamma ray photon is deflected through an angle with respect to its original direction and hence the photon transfers part of its energy and momentum to a free electron, creating a recoil electron. Since all angles of scattering are possible, the energy transferred to the electron can vary from zero to a large fraction of the gamma ray energy [42].

2.6.3 Pair production

This interaction mechanism occurs in the intense electric field near the protons of the nuclei in the absorbing medium (detector) and corresponds to the creation of an electron-positron pair at the point of complete annihilation of the incident gamma-ray photon.

For this process to take place, the gamma ray energy should exceed twice the rest mass energy of the electron (1.022MeV). Both positron and electron disappear and are replaced by two annihilation photons of energy mc^2 (0.511MeV) each emitted back to back [43].

2.7 Importance of the Photo-Electric Effect, the Compton Effect and Pair Production in the Absorption of γ Rays

It is obvious from the above that all three processes can occur at the same time, if the γ ray energy is greater than 1.02 MeV, but that their relative importance varies widely with energy and the value for Z of the absorber. For γ rays below 60 keV in Aluminum and 600 keV in Lead, the photo-electric effect is predominant. The Compton effect then becomes predominant up to about 15 MeV for Aluminum and 5 MeV for Lead. At higher energies pair production predominates.

2.8 Gamma Ray Spectroscopy

Gamma ray spectrometry (GRS) provides a fast, multi elemental and non- destructive method of radioactivity measurement. Qualitative and quantitative analysis of samples can be performed by gamma-ray spectrometry systems in which a qualitative measurement identifies the radionuclides of interest from the energies and intensities of the peaks present in the spectrum under investigation. In a quantitative measurement, the activities of 3 radionuclides of interests are determined. Therefore, the accurate determination of the detector efficiency is arguably the most important parameter when gamma-ray spectrometry is used for radionuclide measurement. Gamma ray spectroscopy is an extremely important method in environmental radioactivity [45] gamma-ray photon or an x-ray is uncharged and does not create direct ionization or excitation of material through which it passes. The detection of gamma-rays is therefore critically dependent on causing the gamma-ray photon to undergo an interaction with detector material transferring all or part of the energy to an electron in the absorbing material.

Therefore, in order for a detector to serve as a gamma-ray spectrometer, it must carry out two distinct functions. First, it must act as a conversion medium in which incident gamma rays have a reasonable probability of interacting to yield one or more secondary electrons. Secondly, it must function as a conventional detector for these secondary electrons [46].

2.9 Semiconductor Detectors

A semiconductor detector is fabricated from either elemental or compound crystal materials having conduction band gap in the range of approximately 1 to 5 eV. The band gap is created by a pn junction at one electrode and no current passes through when there is no ionizing radiation in the region. This creates a region called depletion layer.The figure 2.5 shows creation of depletion layer.

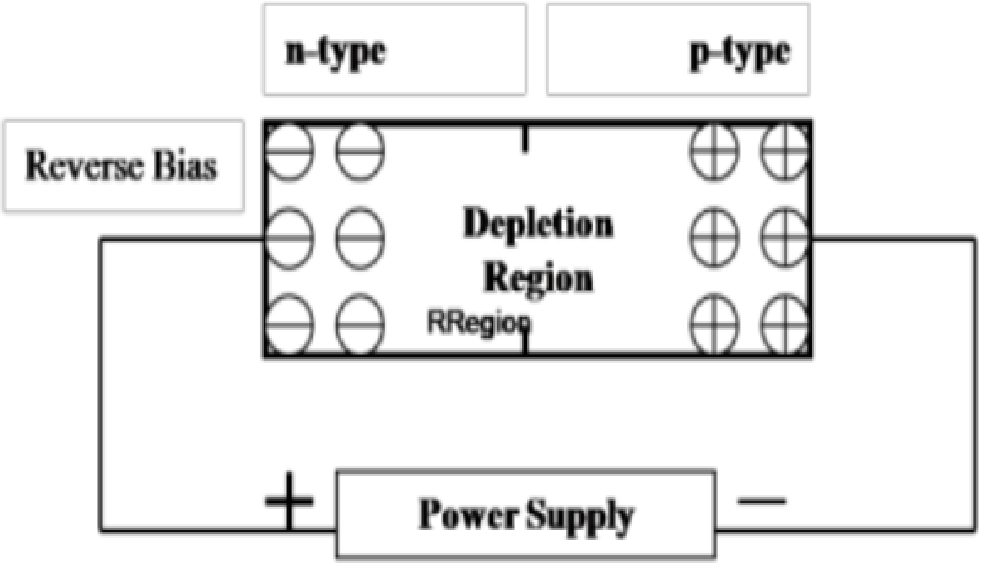


Figure 2.5 Creation of depletion region in semiconductor crystal[41].

Silicon and Germanium are the most commonly used semiconductors with an impurity of valence 3 (acceptor) or 5 (donor). These impurities lower the energy necessary to create electron-hole pairs [47]. These systems provide much better energy resolution than scintillators. This can be attributed to the small amount of energy required to produce a charge carrier and the subsequent large output signal. Only 3eV is required to produce an electron-hole pair in Ge and 3.61eV for Si. So, low temperature of around $-2000°$C is required to avoid thermal charge carrier generation during the interactions of intrinsic (I) region by incoming photons from low level gamma energies [48,49]. The use of semiconductor detectors for gamma-ray detection has provided tremendous gains in measurement capability. The gamma-ray peaks obtained with NaI detectors are very broad by comparison, so that two peaks close to each other cannot be resolved and low- energy peaks may not be easily observed. Semiconductor detectors made of germanium or silicon compensated with lithium provide significantly better energy resolution.Germanium detectors are semiconductor diodes having a p-i-n structure (P-type contact, intrinsic layer, and N-type contact), 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. For a coaxial detector the rectifying contact that forms the semiconductor junction is typically placed at the outer surface of the crystal. Thus, the outer contact for an n-type HPGe was p+ and the inner surface was n+. The depletion layer for such a detector grows inwards as the voltage is increased. High purity germanium coaxial detectors is the first choice of detector for a general purpose high resolution gamma-ray spectrometery.These are now available with efficiencies ranging from 5% to 60% and resolution (1.33 MeV) better than 2.0 keV. Peak-to-Compton ratios in excess of 60.1 can be obtained with the larger detectors. The useful energy range of this type is from 40 keV to 10 MeV [50].

Chapter 3

Methodology of the study

There were several methods for determining the gamma-emitted radioactivity in food grains samples. We used,the most cost-effective high resolution, low-level gamma spectrometry techniques using high-purity germanium (HPGe) detectors. The radionuclides within the food grain samples emit gamma photons at known energies. These interact with the germanium in the detector, which in turn emits signals corresponding to the energies of the incoming photons. The signals from the detector crystal were routed through an amplifier and directed to a Multi-Channel Analyzer (MCA) system. Here, the signals were displayed as a spectrum in which emission counts were plotted against radionuclide energies. Software packages then converted the peak-count information to specific activities (Bq/kg) using calibration procedures.

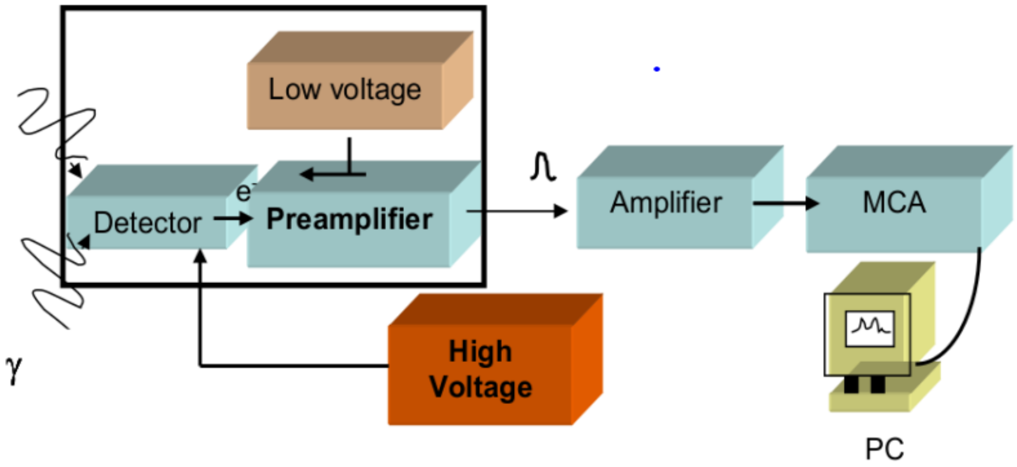


Figure 3.1. Block diagram of a basic gamma spectrometer system with MCA[55].

3.1 HPGe Spectroscopy

HPGe detectors were made by highly refining the element germanium and growing it into a crystal.

The crystal goes through a series of processing steps culminating in the attachment of positive and negative contacts which turn it into an electronic diode. The special property of this diode is that it conducts current in proportion to the energy of a photon (gamma ray) depositing energy in it. The relationship between energy and current is so precise in HPGe detectors that energies are determined to better than 1/10th of 1 percent [51]. Gamma ray spectrometry is an analytical method that allows the identification and quantification of gamma emitting isotopes in a variety of matrices. In one single measurement and with little sample preparation, gamma ray spectrometry allows us to detect several 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 [5]. High purity germanium detectors: For gamma ray detection, germanium is preferred over silicon due to its much higher atomic number. In order to obtain a sufficient sensitive thickness for detection of high energy gamma rays, earlier the detectors were made from lithium compensated p-type germanium. However, these detectors have to be kept continuously cooled at liquid N_2 temperature, due to high mobility of Li ions in germanium at room temperature. Recently, it has been possible to grow germanium crystals with very high purity such that the impurity concentrations are less than 10^{10} atoms/cc. Detectors made out of such crystals do not need any Li compensation and are called high purity germanium detectors. One advantage of this is the possibility of using n-type germanium, where a very thin window can be formed by ion implantation to extend the sensitivity to very low energy gamma rays. Presently, Ge detectors are available with large volume crystals having good photo peak efficiency for MeV gamma rays. However, in order to further reduce the Compton scattered background from the gamma ray spectra, one employs a scintillation based Compton shield around the Ge detector, which is operated in anti-coincidence mode with the Ge detector. Such Compton suppressed high purity Germanium detectors are used extensively for high resolution spectroscopic studies in nuclear physics research [52].

3.2 Principles of Gamma Spectrometry

The HPGe generates free electrons in response to absorbing energy from ionizing radioactivity (such as gamma rays from sample). The magnitude of the charge in the crystal is directly related to the energy of the incident gamma ray. The crystal behaves as a semiconductor through which a high reverse-bias voltage is passed at cryogenic temperatures. Under these conditions, the charge (electron hole pairs) produced by absorption of the gamma ray is then swept to special contacts on opposite sides of the germanium crystal by an electric field. The resulting electrical pulse is integrated by an amplifier producing an output voltage, the pulse height of which is proportional to the incident photon energy [53]. The shape, position and thickness of the contacts determine the detector configuration and its suitability for use in measuring radionuclides of a particular energy range. There are many different configurations for the shape of the germanium crystal, the ways in which voltage is applied, and how the resulting charge from the gamma rays is collected. In all cases, the detectors are suitable for measurement of which is a gamma emitter at 662 keV. Detection and quantification of emitted radioactive particles such as alpha, beta or high energy gamma and X-rays from a contaminated sample or source can easily be achieved by using different qualitative and quantitative approaches. In contrast to qualitative approach, quantitative methods of radioactivity detection are widely used because of their potential to identify and detect high as well as low radioactivity levels with increased sensitivity and counting accuracy. Qualitative and quantitative methods are used for the identification of radioactive elements and measurement of their radioactivity.

3.2.1 HPGe Experimental Setup and Measurements

These measurements were performed using a p-type high purity germanium (HPGe) coaxial detector with a relative efficiency of 70% and multichannel analyser of 8192 channel performance.

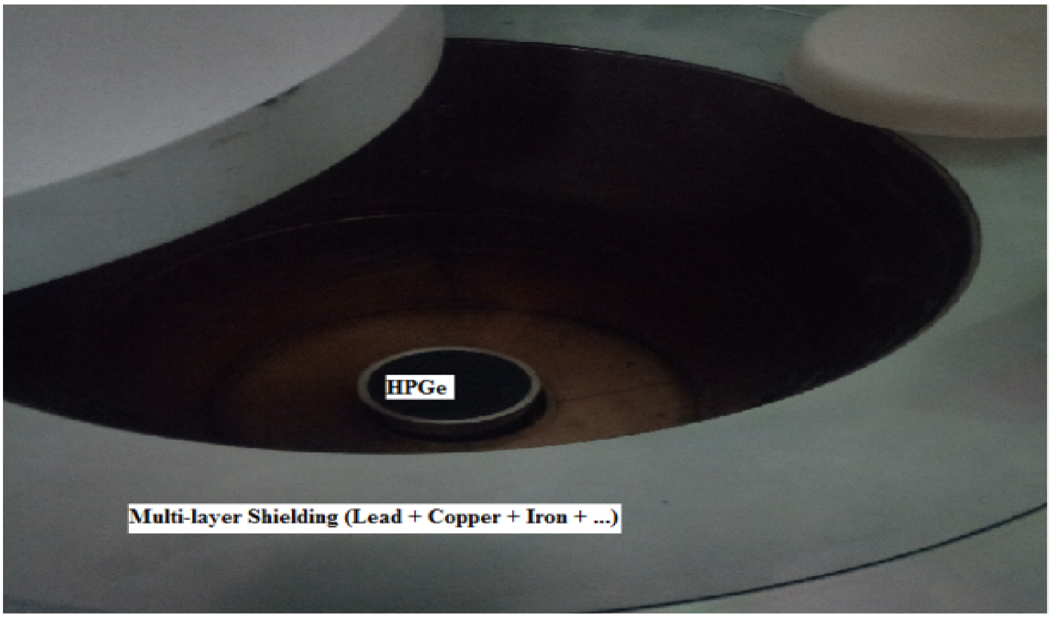


Figure3.2.HPGe-detector and its shielding (Pb+Cd+Cu)[55].

To minimize the number of photons whose sources are out of sample of study, a detector is covered in a lead of 100-mm thickness, cadmium of 2-mm thickness, and copper of 2.5-mm thickness as seen in Fig. 3.2. The 100 mm thick lead is used to reduce soft components of cosmic rays to a very low level. Copper layer suppresses X-ray emitted from the lead (specially with energy 73.9 keV), by its interaction with external radiation. The cadmium layer absorbs thermal neutrons produced by cosmic ray (Photo-neutron and alpha-neutron sources).

There is also an effect of scattered radiation from shielding materials . This can be controlled by fixing the detector at a center of shielding materials itself. After fixing all, we connected the detector to DSA and computer as in Fig. 3.3, so that we can see the amplified and resolved output spectrum on a computer screen.

Figure 3.3.Gamma spectrometry mounted with HPGe detector and DSA circuit (ERPA Lab.)

Samples were placed over the detector for 57,600 sec or 16-hours for measurements. Background radiation was also measured for an empty marinelli beaker of 0.16 kg was measured for the same counting time and an effects of background radiation were subtracted from samples spectra [54,28]. The spectra were analyzed for energies; 911 keV of 238_{Ac} for 232_{Th} radionuclide identification, 609 keV of 214_{Bi} for 238_{U} radionuclide identification, and 1460.9 keV gamma energy for 40_{K}

radionuclide identification. Levels of activity were measured for these identified radio nuclides from their photo peaks. Our set up reports more than 99% confidence for the existence of these photo peaks exactly at channel number used for calibration. Fig 3.4 [54].

3.3 System Calibration

3.3.1 Energy Calibration

An essential requirement for the measurement of gamma emitters is the exact identity of photo peaks present in a spectrum produced by the detector system. The procedure for identifying the radionuclides within a spectrum relies upon methods which match the energies of the principal gamma rays observed in the spectrum to the energies of gamma rays emitted by known radionuclides. The energy calibration of a germanium detector system (i.e. establishing the channel number of the MCA in relation to gamma ray energy) was made by measuring mixed standards sources of known radionuclides with well-defined energies within the energy range of interest. A linear curve was result in the plot of these data shows the system was operating properly[28].

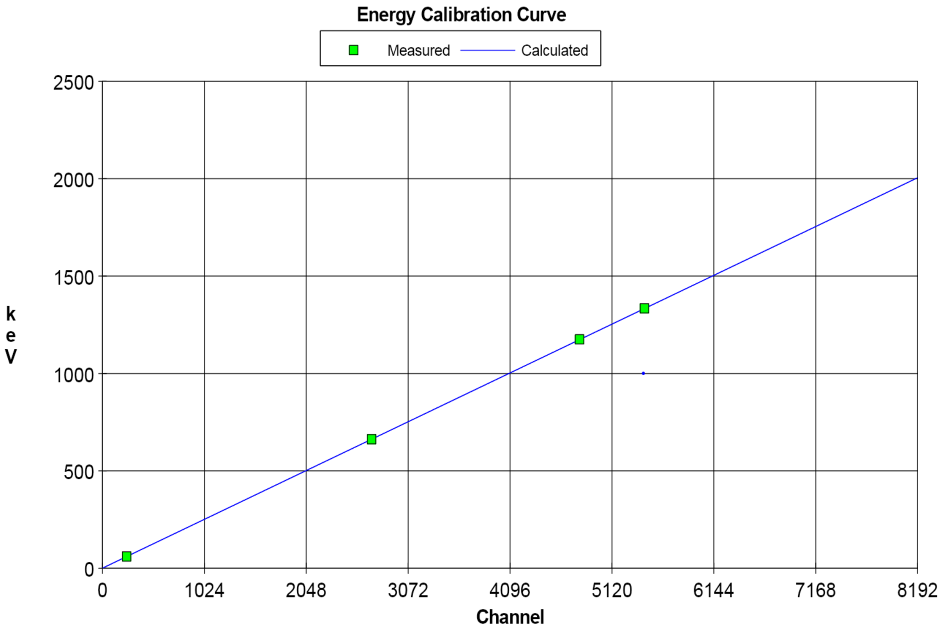


Figure 3.4. Calibration curve by standard check sources in the region of interests.

3.3.2 Efficiency Calibration

An accurate efficiency calibration of the system is necessary to quantify radio nuclides present in a sample. It is essential that this calibration be performed with great care because the accuracy of all quantitative results depend on it. It is also essential that all system settings and adjustments be made prior to determining the efficiencies and be maintained until a new calibration is undertaken. Small changes in the settings of the system components may have slight but direct effects on counting efficiency[28]. After calibration, absolute efficiency (Full Energy Peak Efficiency) of the detector was measured using the same standard sources .

$$\varepsilon_i = \frac{A_i}{C_i \times I_\gamma \times t \times M} \tag{3.1}$$

where A_i is net peak area corresponding to E_i, C_i is deduced activity by certified radionuclide, I_γ indicates probability of E_i photon emission per decay, and t is counting time, M mass of the radionuclide. The photo peak efficiencies from samples of measurement coincide with this efficiency curve and the curve is shown in Fig. 3.5 [55].

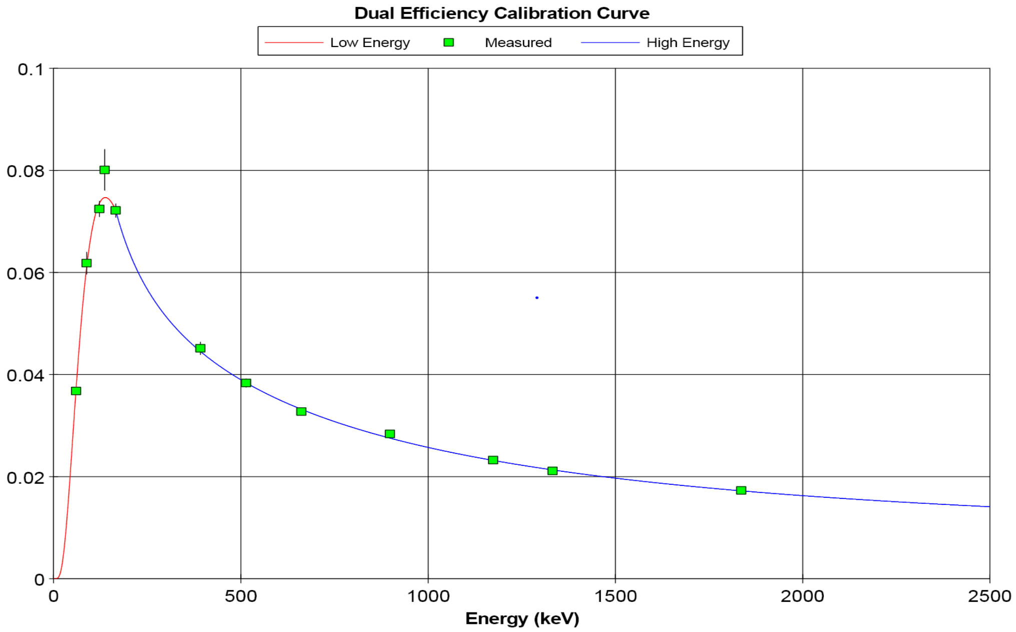


Figure 3.5. Efficiency curve by standard check sources in the region of interests.

3.4 Efficiefncy and Energy Resolution

The average value of full width at half maximum, FWHM, corresponds to the energy resolution of the high purity germanium (HPGe) detector. It describes how useful the detector is for clearly separating two adjacent energy peaks and hence, for unambiguous nuclide identification. Although, in the detector FWHM increasing with gamma-ray energy. This indicates FWHM as a function of gamma ray energies for HPGe detectors. It is shown that resolution is directly proportional to the gamma rays energies. But FWHM is much smaller in Ge detector when we compare to the scintillation type of detectors . Therefore HPGe offers very good resolution and is a good instrument for nuclide identification compared to the another detector. Efficiency is a measure of the percentage of radiation that a given detector detects from the overall yield that is emitted from the source into a solid angle of usually 4π in the photo-peak. $Eq:$ (3.1) was used to calculate the efficiency of the detector .

The HPGe detector offers less detection efficiency. It means that, HPGe is efficient in detecting nuclides with lower energy but not nuclides at higher energy. High purity germanium detector has better resolution compared to the scintillation type of detector[56].

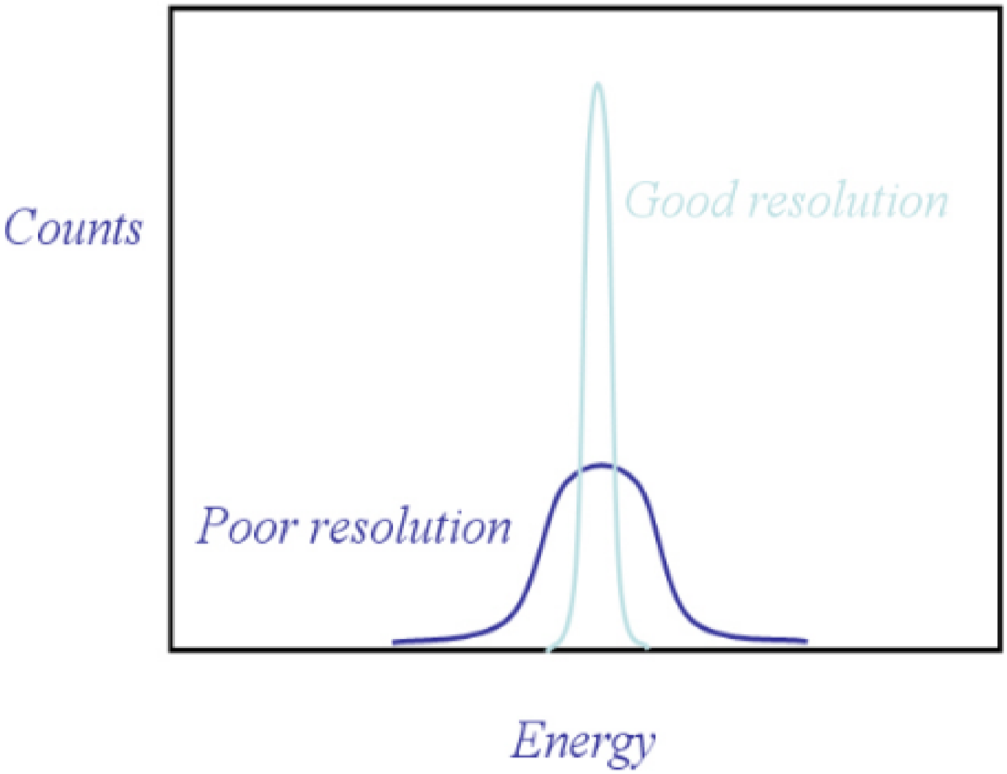


Figure 3.6. Counts Vs pulse height[55].

Pulse shape is determined by calculating ratio of Full Width at Tenth Maximum (FWTM) to Full Width at Half Maximum (FWHM) and ratio of Full Width at Fiftieth Maximum (FWFM) to FWHM. The theoretical acceptable ratio for FWTM/FWHM is 1.82 and should be less than 1.9 in practice. While for FWFM/FWHM is 2.38 for Gaussian peak and should be less than 2.5 in practice [57] From this facts, energy resolution of photo peak is calculated using Eq, 3.2.

$$ER = \frac{FWHM}{Ecentroid} \tag{3.2}$$

where FWHM is calculated using the following relation in GENIE2K compute program/code.

$$FWHM = 2\delta\sqrt{2\ln 2} \tag{3.3}$$

The FWHM of a peak in the pulse height spectrum shown in Fig: 3.7 is used as a measure of energy spread. This is a quantitative indication of the expected resolution of a detector, which is, its ability to distinguish two peaks that are close together. Energy resolution becomes good if centroids of two consecutive peaks are 3FWHM of a peak apart [58].

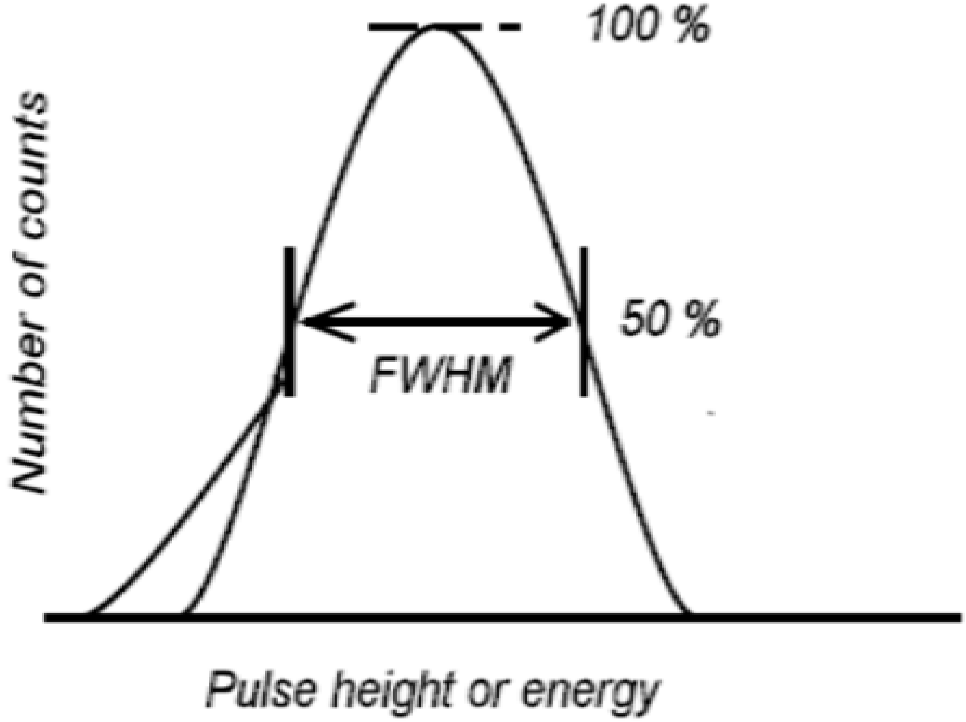


Figure 3.7.Energy Resolution.

The FWHM increase with the energy of a photon, and is dependent on intrinsic properties of the detector, the type and setting of electronic modules being used, and the layout of the cables. After checking for such equipments, a standard Marinelli beakers of 0.16kg, were used as sample holders. Range of peak locating channel were used from 180 keV to 8192 keV with nuclide energy identification tolerance of 1 keV. The dead time of cascaded DSA, as its high performance, is 0.4% in average, for 57,600 sec live time, which means approximately 144 sec difference of real time [59]. Operating parameters of the system are governed and controlled by the installed GENIE2K computer program, Canberra System. The information from prepared samples has been established in a reference of calibration measurements performed for Marinellis geometry and standard sources.

Calibration measurement is a measure of the relationship between gamma energy and channel number. Additionally, the spectrum of laboratory background has been established from prolonged measurements and subtracted from samples measurements. The resulting values allows us to perform nuclide identification and activity analysis, by referring to the information located in a nuclide library file, typically ISO-11929 MDA Report [42, 43]. So, maize and sorghum samples were analyzed using this gamma spectrometry system. The DSA and large volume HPGe detector in a system makes it more attractive in our study.

3.5 Background Radiation

Background radiation is a major factor to achieve acceptable minimum detection limits and precision in analyses. The major sources of background interference can be divided into two main types; (i) internal and (ii) external [60]. The source of the first can be in the materials used to build the detector itself, which emit interfering gamma rays at the same energies as those in environmental samples. These contribute mainly to the peak itself. For instance, [61] and [60] reported on the contribution of natural U and Th series nuclides to detector backgrounds. The second component of background noise (ii) is that external to the system. This is generally due to the inherent radiation environment to which the detector is exposed, and usually affects the whole spectral background. The quality and thickness of lead shielding around the detector can minimize the amount of interference. In this study the back ground radiation is subtracted properly to get exact radiation emitted from the sample. Figure 3.8 blew shows background radiation.

The spectrum of laboratory background has been established from prolonged measurements and subtracted from samples measurements.

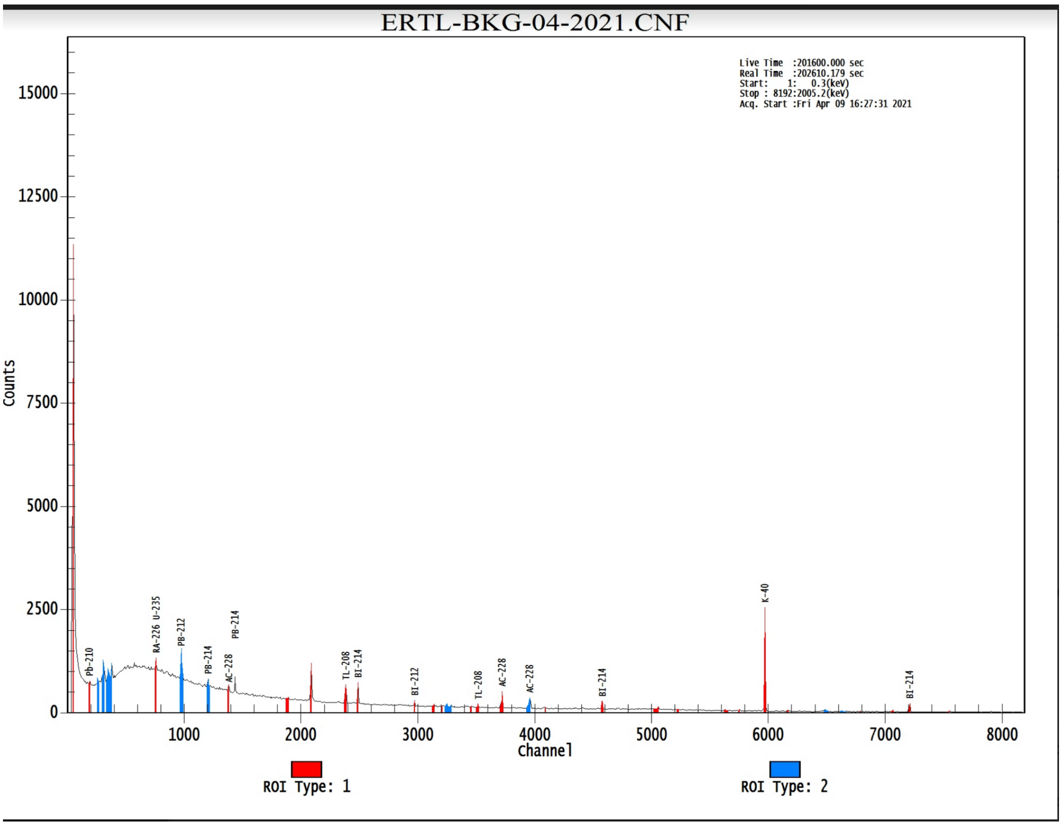


Figure 3.8.Background radiation.

3.6 Sample Area

The origin of samples of study were from Bale Zone,Madda Walabu wereda, in Ethiopia. As shown in Figure 13. So, we collected from households from warre and wadduma kebele. Bale Zone is one of the zones in the Oromia Region of Ethiopia. Bale is bordered on the south by the Ganale Dorya River which separates it from Guji Zone, on the west by the West Arsi Zone, on the north by Arsi Zone, on the northeast by the Shebelle River which separates it from West Hararghe Zone and East Hararghe Zone, and on the east by the Somali Region Bale zone is the second largest zone in Oromia National Regional State after Borena zone with a total area of 63,555km^2. It shares about 17.5 of total area of Oromia. It has 18 districts, 2 urban administrative centers, 20 urban kebeles and 351 rural kebeles. Based on the

2007 Census conducted by the CSA, this Zone has a total population of 1,402,492, an increase of 15.16 over the 1994 census, of whom 713,517 are men and 688,975 women.Bale Zone madda Walabu wereda was 623 km/386milies far from capital City Addis Ababa,Ethiopia.

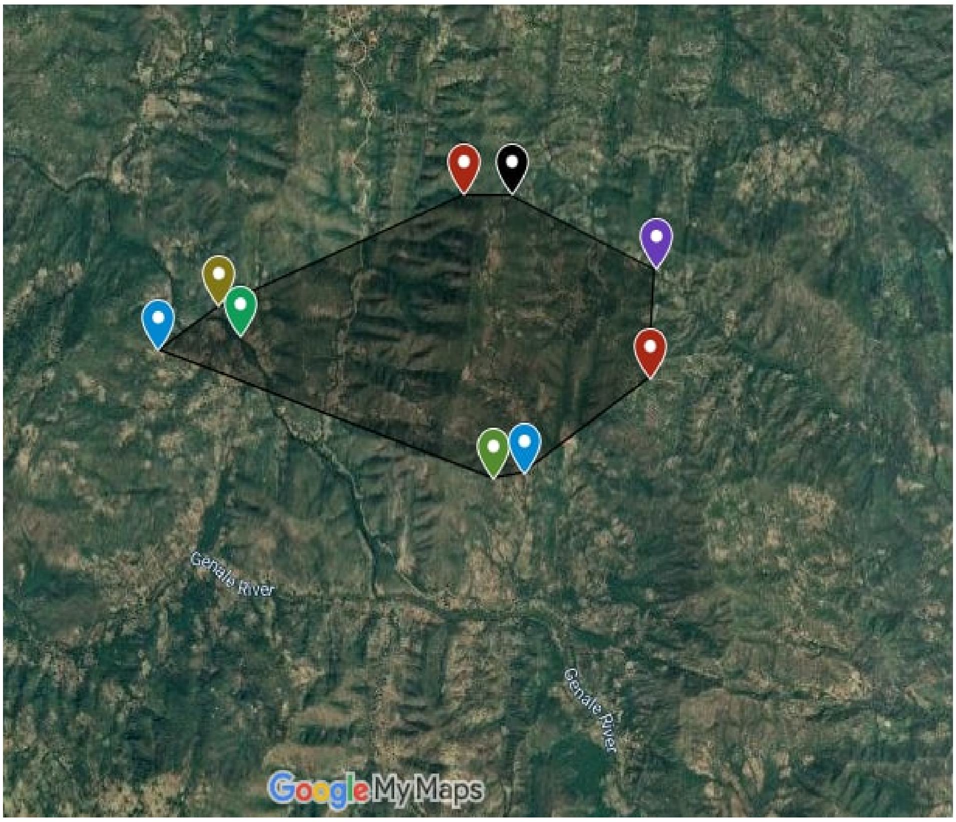


Figure 3.9. Sample Area in Madda walabu wereda,2021.

3.7 Sampling and Sample preparation

Nine samples of different food grains, five of maize and four of sorghum samples were collected during the year of the experiment (march, 2021) from the local kebele of Bale Zone of Ethiopia (ware and wadduma kebele). The collected samples were dried properly so that it could be grinded into powder. The powdered samples were then sieved using a fine aperture mesh screen (mesh size2μ m) in order to remove extraneous items like plants, materials and etc, and to obtain a fine grained sample that would present a uniform matrix to the detector. Then the powder of the samples was dried by oven until a constant weight achieved. Thus it ensure that

any significant moisture was removed from the samples. Upon collection, all the samples were properly packed and marked for their identification code.
The samples where transported, stored properly and processed at the sample preparation laboratory of Ethiopian Radiation protection Authority Centre, Addis Ababa. Then the samples were transferred to cylindrical plastic containers. The weights of the samples were recorded using an electrical Weighing balance. The samples were stored in the ERPA laboratory until secular equilibrium were met.

Figure 3.10.Sample preparation.

3.8 Activity concentration Level Equation

Following the spectrum analysis, for photo peaks representing decay progenies of Uranium, Thorium and photo peak of Potassium, level of activities were calculated using eq, 3.4.

The specific activity, $CE_i(Bq/kg)$ of a nuclide i for a photo peak at energy E, is after rearranging Eq,3.1 [51] is given by;

$$CE_i(Bq/kg) = \frac{A_i}{\varepsilon_{E_i} \times I_\gamma \times t \times m} \tag{3.4}$$

3.9 Radiological Hazard Indices Equations

An exposure of population to environmental radiation sources increases appreciably as industries and natural disasters increases. Therefore, it is important to assess the radiological risks of radiation sources from environments because of human activities. The widely used radiation hazard indices for assessment were radium equivalent activity, ingestion effective dose rate and internal hazard indices that give information about radiation exposure to population [55].

3.9.1 Radium Equivalent Equation (Raeq)

Gamma radiation hazards from specific radio nuclides could be evaluated using different indices.(Raeq), is one of an indicator, which is the weighted sum of activities of the three radio nuclides based on the supposition that 370Bq/kg of 238_U, 259Bq/kg of 232_{Th}, and 4810 Bq/kg of 40_K were give same gamma-ray dose rate. Therefore, Raeq can be calculated from these concept as;

$$Ra_{eq} = C_U + 1.43C_{Th} + 0.077C_k \tag{3.5}$$

where ^{238}U, ^{232}Th and ^{40}K are activity concentrations in Bq/Kg of ^{238}U, ^{232}Th and ^{40}U respectively [53].

3.9.2 Internal Hazard Index (Hin)

This hazard can be quantified by the internal hazard index [62].This given by the following expression.

$$H_{int} = \frac{A_U}{185} + \frac{A_{Th}}{259} + \frac{A_k}{4810} \tag{3.6}$$

where A_U, A_{Th} and A_k are activity concentrations of ^{238}U, ^{232}Th and ^{40}K respectively.

3.9.3 Ingestion Effective Dose

Effective dose is a measure of the energy deposited in a tissue or organ and therefore a measure of the biological harm that may be caused. Effective dose is often referred to simply as radiation dose or dose. Effective dose is measured in units called Sieverts (Sv) [63]. The ingestion effective dose due to the intake of ^{238}U ,^{232}Th , ^{40}K in foods can be evaluated using the following expression [64,65].

$$H_{T,r} = \sum i(U_i \times C_{i,r}) \times g_{t,r} \tag{3.7}$$

U=consumption rate (kg/year),$C_{i,r}$ is activity concentration (Bq/kg),$g_{t,r}$ dose conversion coefficient for ingestion of radionuclide (Sv/Bq) in tissue T. For adult members of the public ,the dose conversion coefficient $g_{t,r}$ for ^{40}K,^{226}Ra (^{238}U) and ^{232}Th ,are 6.2 $\times 10^{-9}$, 2.8 $\times 10^{-7}$ and 2.2 $\times 10^{-7}$ Sv/Bq respectively [66].

Chapter 4

Result and Discussion

4.1 measurement of Radioactivity level and Health Hazard index in maize samples using HPGe gamma spectrometer

4.1.1 Introduction

Most of grains, vegetables fruits, and foods are exposed to various radiations, both from terrestrial,atmosphere and from Cosmo genic, it is followed by a series of decay [36,67,68]. We eat and swallow in our daily consumption many of the things that we do not know the content level of radioactive materials, one of these is maize, which is a staple food for many families in Ethiopia. Knowing the concentrations and permissible dosages of radioactive materials is important to know the harmful limits for humans [19]. Some geographical and terrestrial factors affect the ranges of concentrations of radioactive materials and the activity level are strongly dependent on the origin of the ores [55].

4.1.2 Result and Discussion

Spectra of each samples were recorded by using HPGe detector cascaded with the necessary electronic devices.

From the observed spectra, energies representing daughters of long living radioactive materials and direct representing energy for ^{40}K were clearly identified.

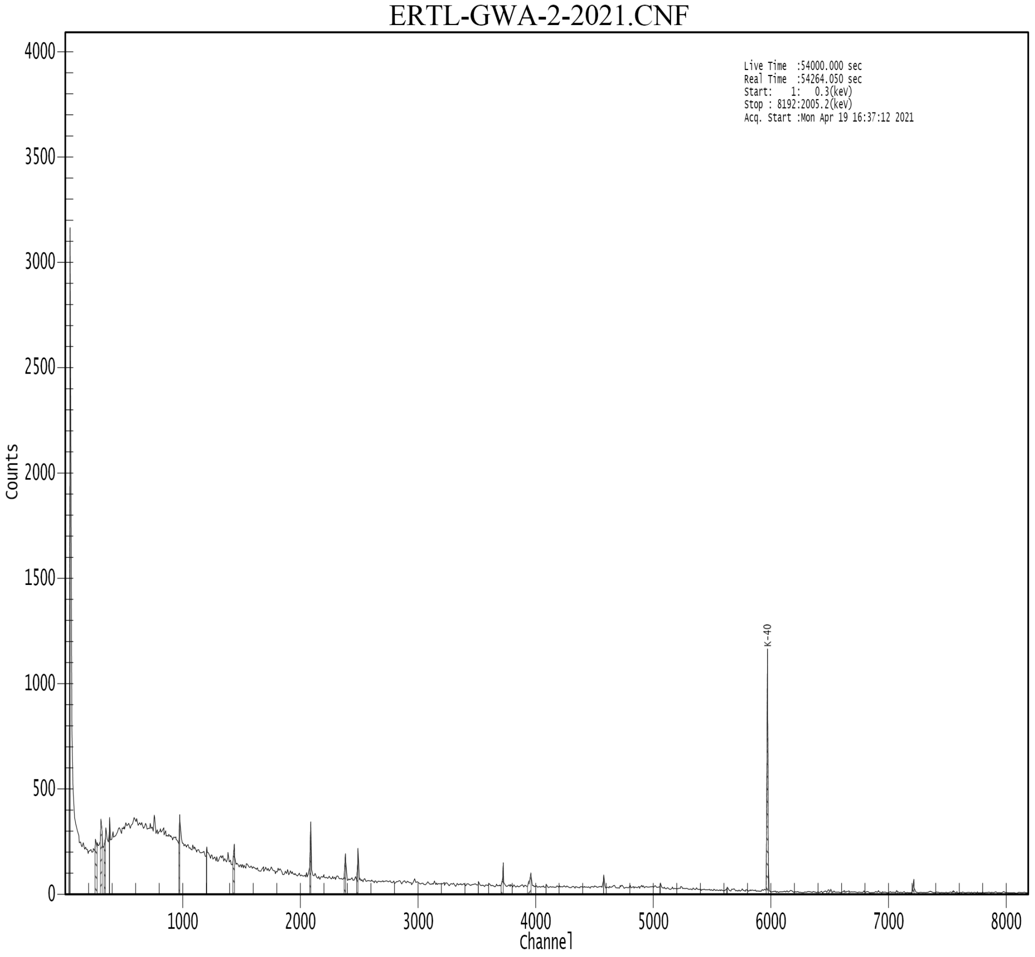
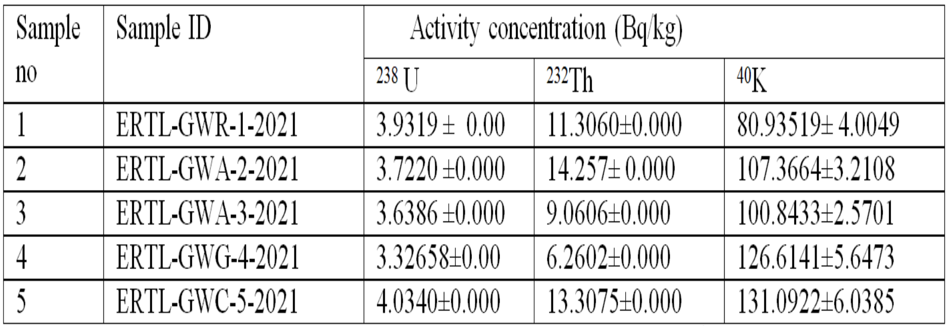


Figure 4.1. Spectrum of maize sample collected from Bale,2021.

The spectra in above figure were recorded for maize sample ERTL-GWA-2 .The activity concentrations of all maize samples for ^{238}U, ^{232}Th and ^{40}K were calculated in the table 4.1.



Sample no	Sample ID	Activity concentration (Bq/kg)		
		^{238}U	^{232}Th	^{40}K
1	ERTL-GWR-1-2021	3.9319 ± 0.00	11.3060±0.000	80.93519± 4.0049
2	ERTL-GWA-2-2021	3.7220 ±0.000	14.257± 0.000	107.3664±3.2108
3	ERTL-GWA-3-2021	3.6386 ±0.000	9.0606±0.000	100.8433±2.5701
4	ERTL-GWG-4-2021	3.32658±0.00	6.2602±0.000	126.6141±5.6473
5	ERTL-GWC-5-2021	4.0340±0.000	13.3075±0.000	131.0922±6.0385

Table 4.1. Activity concentration of NORM in maize sample (2021).

Table (4.1) presents the consequences of the activity concentration of natural radionuclides, ^{238}U ,^{232}Th and ^{40}K in maize samples collected from Bale Zone warre and wadduma Kebeles. The activity concentrations of ^{238}U, ^{232}Th and ^{40}K ranged from 3.326 to 4.0340 $Bqkg^{-1}$, 6.260 to 14.257 $Bqkg^{-1}$ and 80.935 ± 4.004 to $131.092 \pm 6.038 Bqkg^{-1}$, respectively. The average activity concentrations of ^{238}U, ^{232}Th and ^{40}K in maize were $3.73 Bqkg^{-1}$, $10.838 Bqkg^{-1}$ and $109.370 \pm 4.29 Bqkg^{-1}$ respectively. The activity concentration of ^{238}U and ^{232}Th demonstrate that they have higher activity concentrations than the world reference value $0.02 Bq/kg$ and $0.003 Bq/kg$, for ^{238}U and ^{232}Th, of these radionuclides [1].The activity concentration of the radionuclide in maize samples discussed above in table 4. 1 were also expressed below in figure 4.2 by histogram expression.

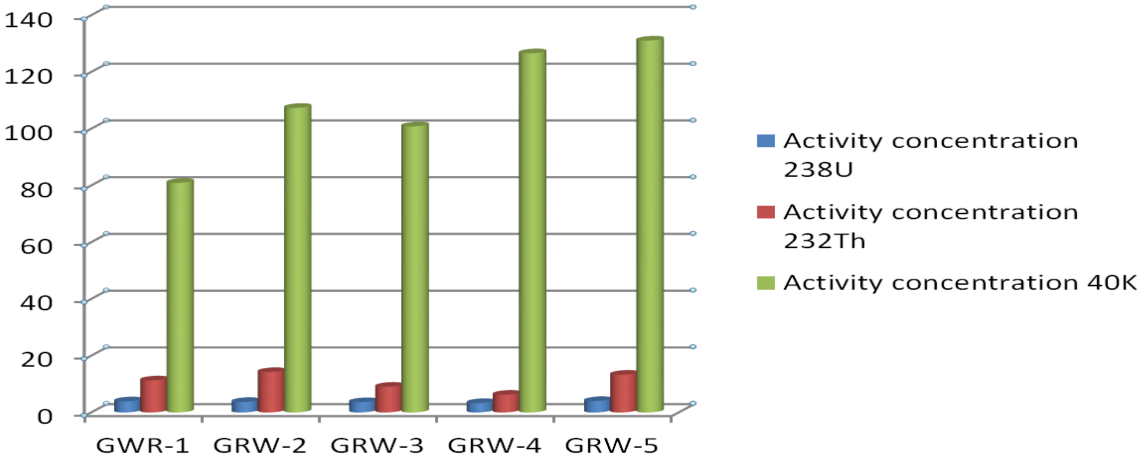
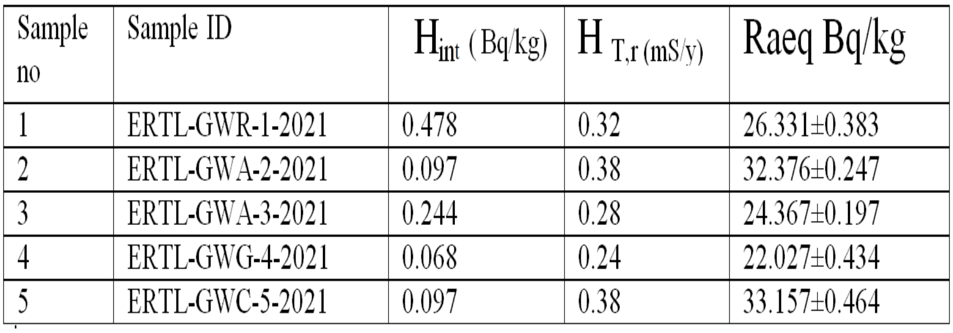


Figure 4.2.Histogram shows Activity concentration of Radionuclide in maize sam-

ple ,2021.
The internal hazard, ingestion effective dose and Radium equivalence of the maize samples were discussed in the table 4.2 blow.



Sample no	Sample ID	H_{int} (Bq/kg)	$H_{T,r}$ (mS/y)	Raeq Bq/kg
1	ERTL-GWR-1-2021	0.478	0.32	26.331±0.383
2	ERTL-GWA-2-2021	0.097	0.38	32.376±0.247
3	ERTL-GWA-3-2021	0.244	0.28	24.367±0.197
4	ERTL-GWG-4-2021	0.068	0.24	22.027±0.434
5	ERTL-GWC-5-2021	0.097	0.38	33.157±0.464

Table 4.2. Radiological hazard indices.
Average of $H_{int} = 0.2$,Average of $H_{Tr} = 0.32$,Average $Ra_{eq} = 27.70 \pm 0.345$
The value of the radiation hazard indices in this study (internal hazard indices and radium equivalent activity) are between $0.068 \pm 0.00 Bq/kg$ to $0.478 \pm 0.00 Bq/kg$ for H_{int} and the radium equivalence of the sample were measured between $22.027 \pm 0.434 Bq/kg$ to $33.157 \pm 0.464 Bq/kg$ with average value of $0.2 Bq/kg$ and $27.70 \pm 0.345 Bq/kg$ respectively. Table 4.2 also presents the result of the ingestion effective dose in mSv/y for adult to specific activity of^{238}U,^{232}Th and ^{40}K in maize samples which was evaluated using equation (3.7).The range of summation of the ingestion of effective dose were varied from $0.24 mSv/y$ to $0.38 mSv/y$ with the average value of $0.32 mSv/y$ which is slightly greater than world reference value $0.3 mSv/y$ for ^{238}U and ^{232}Th [2].

4.2 measurement of Radioactivity level and Health Hazard index in sorghum samples using HPGe gamma spectrometer

4.2.1 Introduction

Food is the primary wellspring of radioactive components [11]. Radionuclides consumed by means of foodstuff represent a huge segment of the average radiation dosage to different body organs and are likewise a standout amongst the most imperative courses for long term wellbeing contemplations; Radioactivity concentrations may be increase in local foodstuffs because they could be cultivated in high NORM areas [69]. Soil radionuclides can enter the food chain by express discharge on leaves or by transfer into parts of plants that are consumable for people and creatures [70] .Sorghum is one of most five common cereals, Teff, Maize, Wheat and Barley, account for 75 percent of the total grain area in Ethiopia. These cereals are consumed throughout the country as staples, with varying preference by cereal type [71].The contamination of these food by high concentration radioactive may affect the population's health potentially.

4.2.2 Result and Discussion

The spectra for all sorghum samples were recorded accordingly. The spectra listed in figure 4.3 were recorded for sorghum sample ERTL-WRS-7.

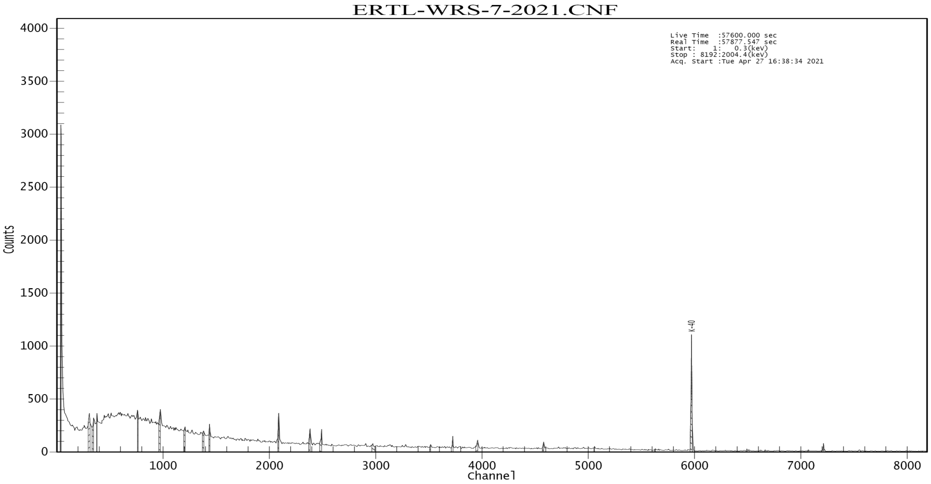
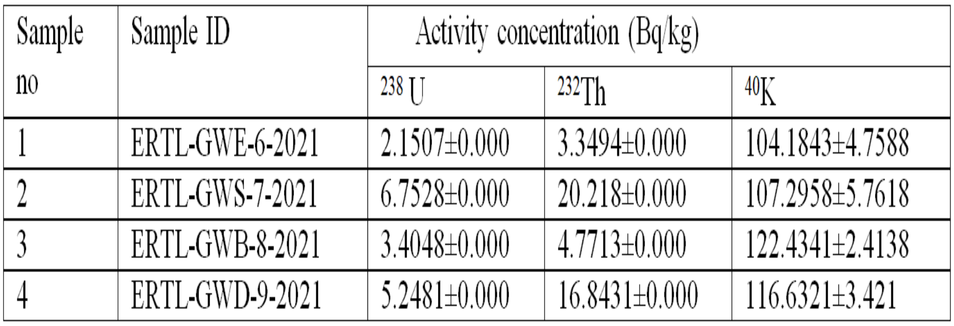


Figure 4.3. Spectrum of sorghum sample collected from Bale,2021.

The activity concentrations of all sorghum samples for ^{238}U, ^{232}Th and ^{40}K were calculated in the table 4.3 below.



Sample no	Sample ID	Activity concentration (Bq/kg)		
		^{238}U	^{232}Th	^{40}K
1	ERTL-GWE-6-2021	2.1507±0.000	3.3494±0.000	104.1843±4.7588
2	ERTL-GWS-7-2021	6.7528±0.000	20.218±0.000	107.2958±5.7618
3	ERTL-GWB-8-2021	3.4048±0.000	4.7713±0.000	122.4341±2.4138
4	ERTL-GWD-9-2021	5.2481±0.000	16.8431±0.000	116.6321±3.421

Table 4.3. Activity concentration of NORM in sorghum sample,2021. Table 3 discuss the consequences of the activity concentration of natural radionuclides, ^{238}U ,^{232}Th, and ^{40}K in sorghum samples collected from Bale Zone warre and wadduma Kebeles.

The activity concentrations of ^{238}U, ^{232}Th and ^{40}K ranged from 2.151 to 6.753$Bqkg^{-1}$, 3.349 to 20.218$Bqkg^{-1}$ and 104.184 ± 4.758 to $122.434 \pm 2.413 Bqkg^{-1}$, respectively. The average activity concentrations of ^{238}U,^{232}Th and ^{40}K in sorghum were 4.389$Bqkg^{-1}$, 11.295$Bqkg^{-1}$, and $112.636 \pm 4.088 Bqkg^{-1}$, respectively. The activity concentration of ^{238}U and ^{232}Th shows that they have higher activity concentrations than the world reference value 0.02 Bq/kg and 0.003Bq/kg , for ^{238}U and ^{232}Th of these radionuclides [1].The activity concentration of sorghum samples also presented in histogram in figure 4.4 below that used to analize the activity concentration of ^{238}U,^{232}Th and ^{40}K.

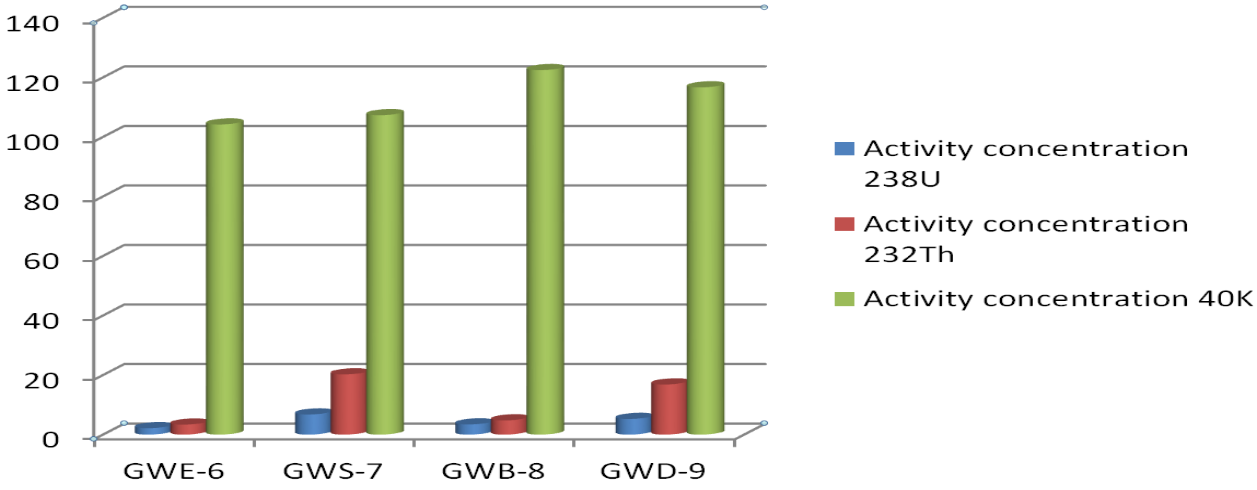
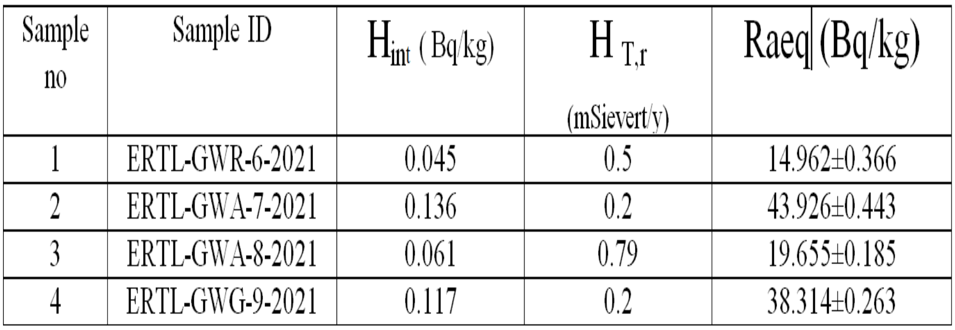


Figure 4.4. Histogram expression of activity concentration in sorghum samples 2021. The internal hazard, ingestion effective dose and Radium equivalence of the sorghum samples were discussed in the table 4.4 blow.



Sample no	Sample ID	H_{int} (Bq/kg)	$H_{T,r}$ (mSievert/y)	Raeq (Bq/kg)
1	ERTL-GWR-6-2021	0.045	0.5	14.962±0.366
2	ERTL-GWA-7-2021	0.136	0.2	43.926±0.443
3	ERTL-GWA-8-2021	0.061	0.79	19.655±0.185
4	ERTL-GWG-9-2021	0.117	0.2	38.314±0.263

Table 4.4 Radiological hazard indices in Sorghum Sample (2021).
Average $H_{int} = 0.089$,Average $H_{t,r} = 0.422$,Average $Ra_{eq} = 29.15 \pm 0.314$.

The value of internal hazard indices and radium equivalent activity are between $0.045 \pm 0.00 Bq/kg$ to $0.136 \pm 0.00 Bq/kg$ for H_{int} and the radium equivalence of the sample were measured between $14.962 \pm 0.366 Bq/kg$ to $43.926 \pm 0.443 Bq/kg$ with average value of $0.089 Bq/kg$ and $0.004 Bq/kg$ respectively. Table 4.4 also presents the result of the ingestion effective dose in mSv/y for specific activity of ^{238}U,^{232}Th and ^{40}K in sorghum samples which was evaluated using equation (3.7) for sorghum samples.The range of summation of the ingestion of effective dose were varied from $0.2 mSv/y$ to 0.79 mSv/y with the average value of $0.422 mSv/y$ which is higher than world reference value 0.3 mSv/y for ingestion [2].

Chapter 5

Conclusions and Recommendations

5.1 Conclusions

The activity concentrations of natural radio nuclides and their radiological health hazards indices in different types of maize and sorghum samples taken from Bale Zone in Ethiopia were measured using gamma ray spectroscopy coupled with HPGe detector. Grains were categorized in two groups as maize and sorghum as mentioned in the beginning. The average values of measured activity concentrations for ^{238}U, ^{232}Th and ^{40}K in maize samples were calculated. The average values of measured activity concentrations for ^{238}U and ^{232}Th are higher than the given world reference values, $0.02Bq/Kg$ for ^{238}U and $0.003Bq/Kg$ for ^{232}Th [1]. Potassium is a key element in many body functions such as digestion and heart rate, and the potassium content of the body is kept constant by metabolic processes [1]. The radiation hazard indices (ingestion effective dose) from maize sample were also slightly higher than the world permissible value,$0.3mSv/y$ for ingestion. The average activity concentration and ingestion effective dose for sorghum sample also greater than world reference value $0.02Bq/kg$ for ^{238}U and $0.003Bq/kg$ for ^{232}Th,and $0.3mSv/y$ for ingestion. From these result we observed the activity concentration and ingestion effective dose for given samples all are above world permissible values. These result shows activities of population around the area and consuming food grain cultivated around this area can cause health hazard.

5.2 Recommendations

This study, dissertation has recommendations based on the findings, for population around the area, government and for responsible sectors who can work on issues. The average radioactivity levels measured in maize and sorghum sample were above world reference values, Also the radiological hazard indices for both samples were above world permissible value.

Therefore, this study indicates that the radionuclides intake from the consumption of maize and sorghum poses threat to public health. Consuming maize and sorghum cultivated in Bale Zone warre and Wadduma kebele expose the population to high radiation dose which might be detrimental to their health. Government, population and concerned sectors must give special attention to the issue. It is recommended that further studies on radioactivity should be carried out in the area.

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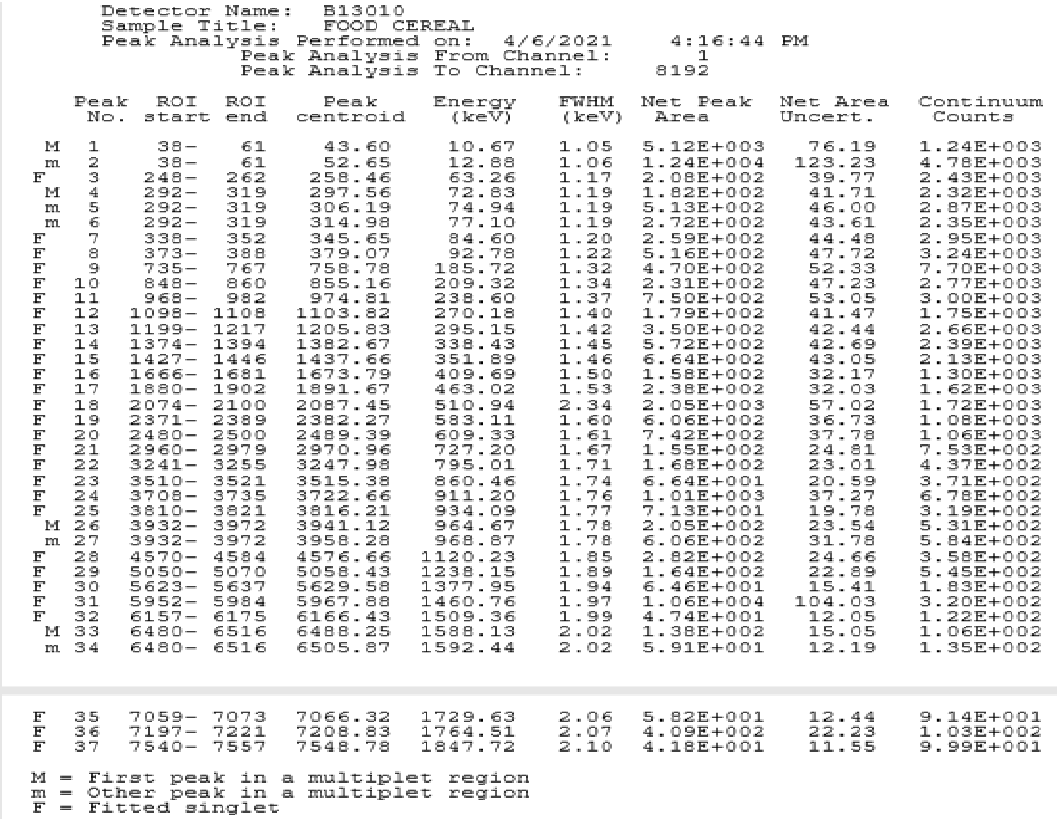
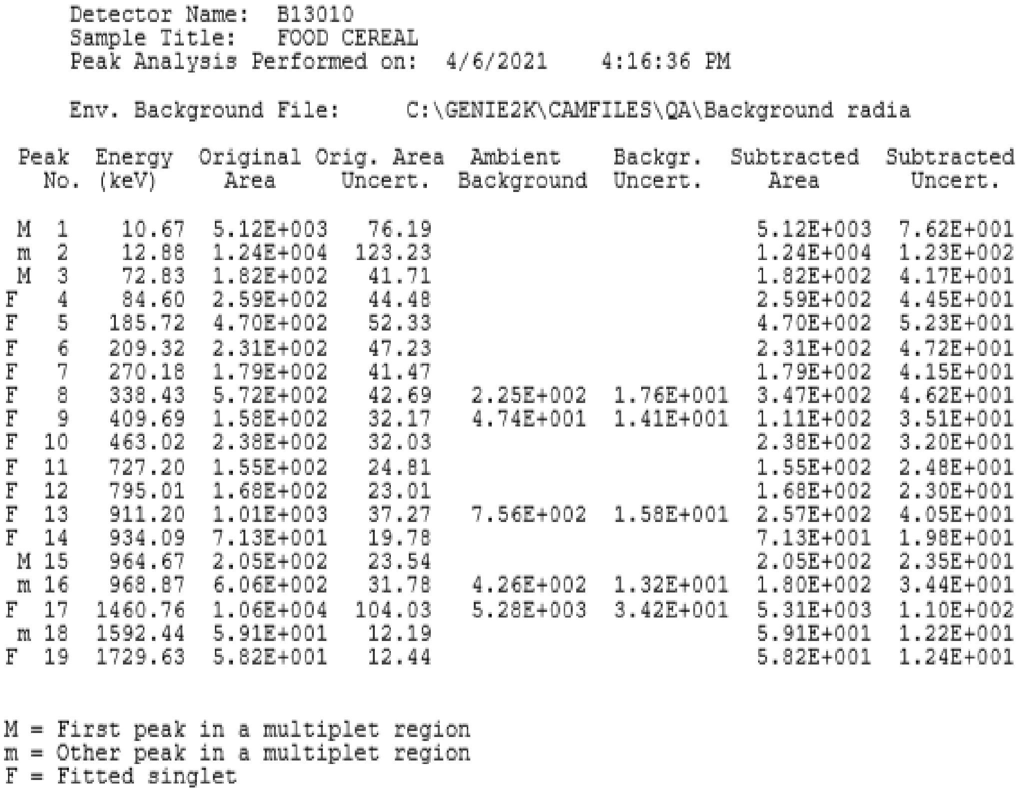
Appendix

Peak Analysis Report



```
          Detector Name:   B13010
          Sample Title:    FOOD CEREAL
          Peak Analysis Performed on:   4/6/2021     4:16:44 PM
                    Peak Analysis From Channel:       1
                    Peak Analysis To Channel:      8192

      Peak  ROI   ROI     Peak       Energy     FWHM   Net Peak    Net Area    Continuum
       No. start end    centroid     (keV)     (keV)    Area      Uncert.      Counts

   M   1     38-    61     43.60      10.67     1.05  5.12E+003    76.19     1.24E+003
   m   2     38-    61     52.65      12.88     1.06  1.24E+004   123.23     4.78E+003
  F    3    248-   262    258.46      63.26     1.17  2.08E+002    39.77     2.43E+003
   M   4    292-   319    297.56      72.83     1.19  1.82E+002    41.71     2.32E+003
   m   5    292-   319    306.19      74.94     1.19  5.13E+002    46.00     2.87E+003
   m   6    292-   319    314.98      77.10     1.19  2.72E+002    43.61     2.35E+003
  F    7    338-   352    345.65      84.60     1.20  2.59E+002    44.48     2.95E+003
  F    8    373-   388    379.07      92.78     1.22  5.16E+002    47.72     3.24E+003
  F    9    735-   767    758.78     185.72     1.32  4.70E+002    52.33     7.70E+003
  F   10    848-   860    855.16     209.32     1.34  2.31E+002    47.23     2.77E+003
  F   11    968-   982    974.81     238.60     1.37  7.50E+002    53.05     3.00E+003
  F   12   1098-  1108   1103.82     270.18     1.40  1.79E+002    41.47     1.75E+003
  F   13   1199-  1217   1205.83     295.15     1.42  3.50E+002    42.44     2.66E+003
  F   14   1374-  1394   1382.67     338.43     1.45  5.72E+002    42.69     2.39E+003
  F   15   1427-  1446   1437.66     351.89     1.46  6.64E+002    43.05     2.13E+003
  F   16   1666-  1681   1673.79     409.69     1.50  1.58E+002    32.17     1.30E+003
  F   17   1880-  1902   1891.67     463.02     1.53  2.38E+002    32.03     1.62E+003
  F   18   2074-  2100   2087.45     510.94     2.34  2.05E+003    57.02     1.72E+003
  F   19   2371-  2389   2382.27     583.11     1.60  6.06E+002    36.73     1.08E+003
  F   20   2480-  2500   2489.39     609.33     1.61  7.42E+002    37.78     1.06E+003
  F   21   2960-  2979   2970.96     727.20     1.67  1.55E+002    24.81     7.53E+002
  F   22   3241-  3255   3247.98     795.01     1.71  1.68E+002    23.01     4.37E+002
  F   23   3510-  3521   3515.38     860.46     1.74  6.64E+001    20.59     3.71E+002
  F   24   3708-  3735   3722.66     911.20     1.76  1.01E+003    37.27     6.78E+002
  F   25   3810-  3821   3816.21     934.09     1.77  7.13E+001    19.78     3.19E+002
   M  26   3932-  3972   3941.12     964.67     1.78  2.05E+002    23.54     5.31E+002
   m  27   3932-  3972   3958.28     968.87     1.78  6.06E+002    31.78     5.84E+002
  F   28   4570-  4584   4576.66    1120.23     1.85  2.82E+002    24.66     3.58E+002
  F   29   5050-  5070   5058.43    1238.15     1.89  1.64E+002    22.89     5.45E+002
  F   30   5623-  5637   5629.58    1377.95     1.94  6.46E+001    15.41     1.83E+002
  F   31   5952-  5984   5967.88    1460.76     1.97  1.06E+004   104.03     3.20E+002
  F   32   6157-  6175   6166.43    1509.36     1.99  4.74E+001    12.05     1.22E+002
   M  33   6480-  6516   6488.25    1588.13     2.02  1.38E+002    15.05     1.06E+002
   m  34   6480-  6516   6505.87    1592.44     2.02  5.91E+001    12.19     1.35E+002


  F   35   7059-  7073   7066.32    1729.63     2.06  5.82E+001    12.44     9.14E+001
  F   36   7197-  7221   7208.83    1764.51     2.07  4.09E+002    22.23     1.03E+002
  F   37   7540-  7557   7548.78    1847.72     2.10  4.18E+001    11.55     9.99E+001

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

Background Subtract Report



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     Detector Name:  B13010
     Sample Title:   FOOD CEREAL
     Peak Analysis Performed on:  4/6/2021    4:16:36 PM

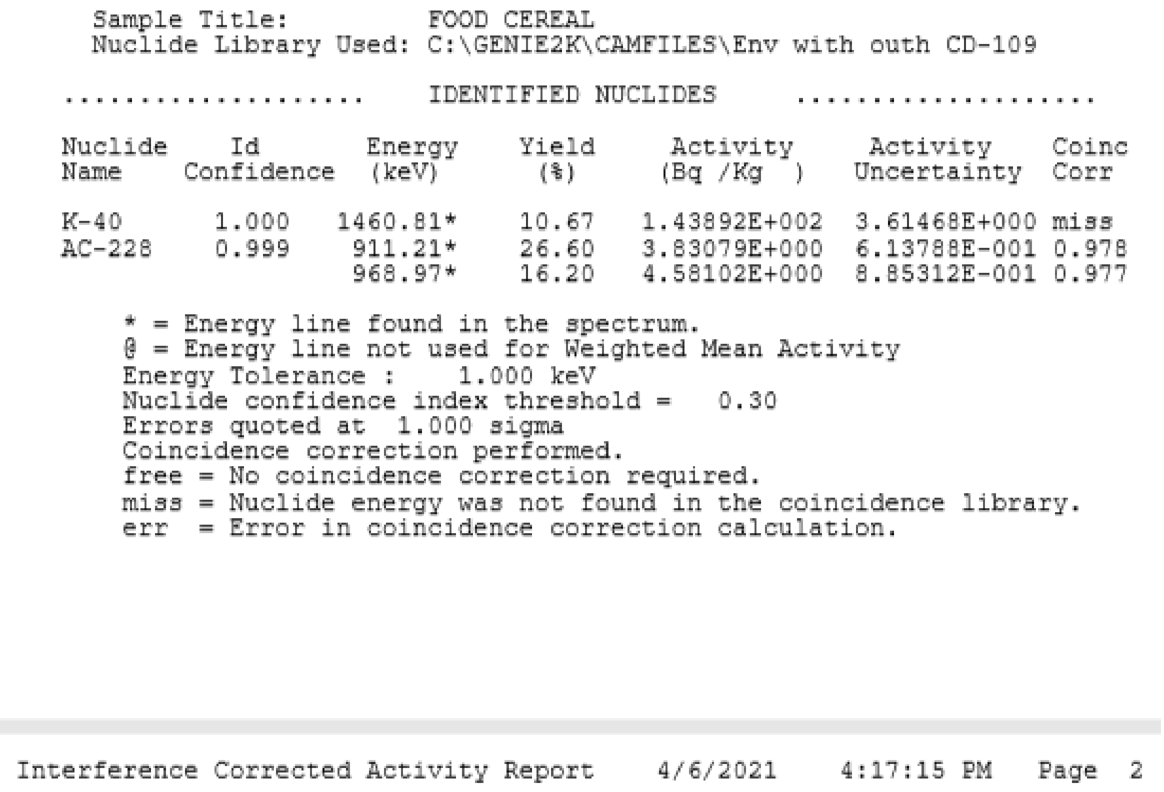
     Env. Background File:     C:\GENIE2K\CAMFILES\QA\Background radia

 Peak  Energy  Original Orig. Area  Ambient    Backgr.  Subtracted  Subtracted
   No. (keV)     Area    Uncert.  Background  Uncert.     Area        Uncert.

  M  1    10.67  5.12E+003   76.19                        5.12E+003  7.62E+001
  m  2    12.88  1.24E+004  123.23                        1.24E+004  1.23E+002
  M  3    72.83  1.82E+002   41.71                        1.82E+002  4.17E+001
 F   4    84.60  2.59E+002   44.48                        2.59E+002  4.45E+001
 F   5   185.72  4.70E+002   52.33                        4.70E+002  5.23E+001
 F   6   209.32  2.31E+002   47.23                        2.31E+002  4.72E+001
 F   7   270.18  1.79E+002   41.47                        1.79E+002  4.15E+001
 F   8   338.43  5.72E+002   42.69   2.25E+002  1.76E+001 3.47E+002  4.62E+001
 F   9   409.69  1.58E+002   32.17   4.74E+001  1.41E+001 1.11E+002  3.51E+001
 F  10   463.02  2.38E+002   32.03                        2.38E+002  3.20E+001
 F  11   727.20  1.55E+002   24.81                        1.55E+002  2.48E+001
 F  12   795.01  1.68E+002   23.01                        1.68E+002  2.30E+001
 F  13   911.20  1.01E+003   37.27   7.56E+002  1.58E+001 2.57E+002  4.05E+001
 F  14   934.09  7.13E+001   19.78                        7.13E+001  1.98E+001
  M 15   964.67  2.05E+002   23.54                        2.05E+002  2.35E+001
  m 16   968.87  6.06E+002   31.78   4.26E+002  1.32E+001 1.80E+002  3.44E+001
 F  17  1460.76  1.06E+004  104.03   5.28E+003  3.42E+001 5.31E+003  1.10E+002
  m 18  1592.44  5.91E+001   12.19                        5.91E+001  1.22E+001
 F  19  1729.63  5.82E+001   12.44                        5.82E+001  1.24E+001


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

Nuclide Identification Report



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

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

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

K-40      1.000   1460.81*    10.67   1.43892E+002  3.61468E+000 miss
AC-228    0.999    911.21*    26.60   3.83079E+000  6.13788E-001 0.978
                   968.97*    16.20   4.58102E+000  8.85312E-001 0.977

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


 Interference Corrected Activity Report    4/6/2021    4:17:15 PM   Page  2
```
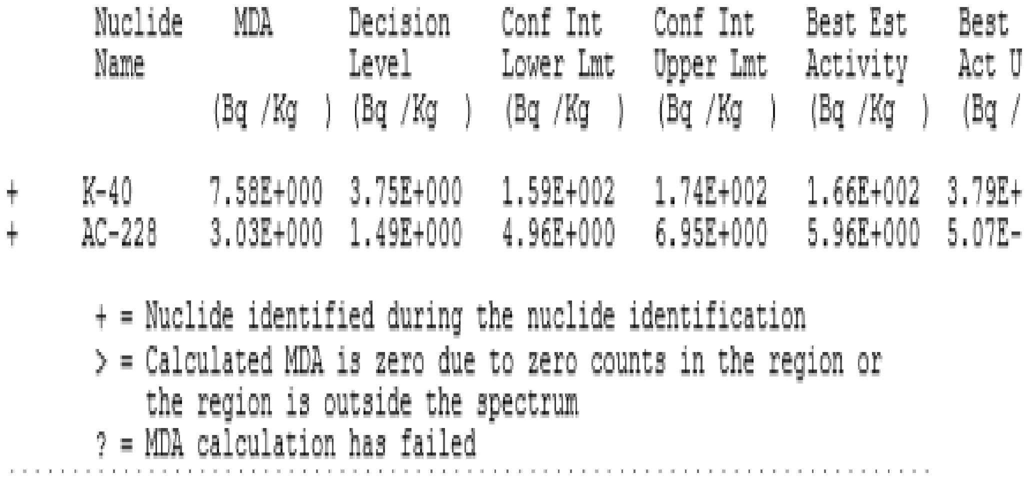

Nuclide ISO 11929 Report



```
         Nuclide    MDA       Decision    Conf Int    Conf Int    Best Est    Best
         Name                 Level       Lower Lmt   Upper Lmt   Activity    Act U
                 (Bq /Kg  ) (Bq /Kg  )  (Bq /Kg  )  (Bq /Kg  )  (Bq /Kg  )  (Bq /

 +      K-40     7.58E+000  3.75E+000   1.59E+002   1.74E+002   1.66E+002  3.79E+
 +      AC-228   3.03E+000  1.49E+000   4.96E+000   6.95E+000   5.96E+000  5.07E-

         + = Nuclide identified during the nuclide identification
         > = Calculated MDA is zero due to zero counts in the region or
             the region is outside the spectrum
         ? = MDA calculation has failed
 .........................................................................
```