# LIBYAN ACADEMY

## TRIPOLI –LIBYA




**School of Basic Science**
**Department of Environmental Sciences and Engineering**

# Measurements of Natural and Man-Made Radionuclides Concentrations in Cereals Consumed in Libya Using HPGe-Detectors

***A Thesis Submitted to the school of science at the Libyan Academy in Partial Fulfillment of the Requirements of Master Degree in Environmental Sciences and Engineering***

**By:**

**Mabruka M. El khaboly**

**Under the Supervision of:**

**Dr. Nouri A. Droughi**

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# الشكر والتقدير

لابد لنا ونحن نخطو خطواتنا الأخيرة في الحياة الأكاديمية من وقفة نعود إلى أعوام قضيناها في رحاب الأكاديمية الليبية مع أساتذتنا الكرام الذين قدموا لنا الكثير .

وقبل أن نمضي تقدم أسمى آيات الشكر والامتنان والتقدير والمحبة إلى الذين حملوا أقدس رسالة في الحياة ...

إلى الذين مهدوا لنا طريق العلم والمعرفة ...

إلى جميع دكاترتنا الأفاضل.....

"كن عالما .. فإن لم تستطع فكن متعلما ، فإن لم تستطع فأحب العلماء ،فإن لم تستطع فلا تبغضهم"

وأخص بالتقدير والشكر:

**الدكتور: نورى الدروقي**

وكذلك نشكر كل من ساعد على إتمام هذا البحث وقدم لنا العون ومد لنا يد المساعدة وزودنا بالمعلومات اللازمة لإتمام هذا البحث ونخص بالذكر:

المهندس : يوسف البيري..... من مركز البحوث النووية

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الذين كانوا عونا لنا في بحثنا هذا ونورا يضيء الظلمة التي كانت تقف أحيانا في طريقنا.

إلى من زرعوا التفاؤل في دربنا وقدموا لنا المساعدات والتسهيلات والأفكار والمعلومات، ربما دون أن يشعروا بدورهم بذلك فلهم منا كل الشكر، وأخص منهم:

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**يا من أحمل اسمك بكل فخر**

**يا من أفتقدك منذ سنين**

**يا من يرتعش قلبي لذكرك**

**يا من أودعتني لله أهديك هذا البحث أبـــــي**

**"إلى روح والدي الغالي "**

**إلى حكمتي .....وعلمي**

**إلى أدبي .....وحلمي**

**إلى طريقي .... المستقيم**

**إلى طريق..... الهداية**

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# Abstract

Twenty four samples of different types of imported and locally produced cereals and grains consumed in Libya were collected and analyzed using gamma-ray spectroscopy system with a high purity germanium detector (HPGe). The concentrations of the natural radionuclides such as $^{238}U$, $^{232}Th$ and $^{40}K$ were present in the most studied samples were measured, and the radium equivalent activities (*Raeq*), were calculated, as well as the hazard index( HI). The average activity values of $^{238}U$, $^{232}Th$ and $^{40}K$ were in the range of 3.83±0.57, 1± 0.15 and 142.8 ± 21.4 $Bq.kg^{-1}$ respectively. The values of *Raeq* ranged between 17.70 – 245.64 $Bq.kg^{-1}$. The HI were ranged between 0.05–0.66, which is less than one (the higher limit of HI). The obtained results were compared with the standard accepted Nationally and International values, and was found to be within the acceptable limits.

# ملخص باللغة العربية

من خلال هذا البحث والذي تم فيه تجميع أربع وعشرون عينة مختلفة من الحبوب المستخدمة كغذاء والمستوردة من مختلف دول العالم وكذلك بعضها المنتجة محليا والتي يتم استهلكها من قبل أفراد الشعب الليبي. تم تحليل جميع هذه العينات تحت نفس الظروف وذلك باستخدام جهاز الجرمانيوم عالي النقاوة. وقد قيست تراكيز العناصر المشعة طبيعيا مثل نويدات اليورانيوم والثوريوم والبوتاسيوم و كذلك المصنعة من قبل الإنسان مثل عنصر السيزيوم. وكان متوسط نسب تركيز نويدات هذه العناصر المشعة لليورانيوم والثوريوم والبوتاسيوم على التوالي 3.83±0.57 , 1±0.15 و 21.4 ± 142.8 بيكيرل/كيلوجرام . بالإضافة لذلك فقد تم حساب مكافئ الراديوم وكان 245.64- 17.7 بيكيرل/كيلوجرام ومؤشر الخطر الإشعاعي لاستهلاك هذه المواد 0.66- 0.0 وكان اقل من الحد المسموح . نتائج هذا البحث ستساعد الجهات القانونية في وضع المعايير والحدود المسموح بها للاستهلاك المواد الغذائية في ليبيا.

# CHAPTER ONE

# INTRODUCTION

## 1. Introduction

**S**oon after the great discovery of radioactivity in 1896 by A. H. Becquerel, numerous studies have been reported on radioactivity types, properties and radiation effects. Many types of radionuclides were discovered and characterized to be found all around us in the natural environment, in the atmosphere, beneath the earth and even within ourselves. Sources of radionuclides include those that exist before the creation of the earth (primordial sources), those that result from cosmic ray interactions (cosmogenic sources) and those that were produced as result of human activities and were called man made radionuclides [1]

Natural Environmental radioactivity is composed of the cosmogenic and terrestrial radionuclides. United Nations Scientific Committee on the Effects of Atomic Radiation [69] reported that estimation of 80% of doses contribution in the environment are derived from the natural radionuclides while the remaining 20% is from cosmic ray and processes Cosmogenic radionuclides, such as $^{3}H$, $^{7}B$, $^{14}C$ and $^{22}Na$ , which are produced by the interaction of cosmic-ray particles in the earth's atmosphere. Terrestrial radionuclides ($^{40}K$, $^{238}U$ and $^{232}Th$ series) 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.Terrestrial gamma radiation is one of the main constituents of background radiation and is strongly dependent on the composition of the soil and the rocks and the radionuclides contained within them.

The dose rate of terrestrial gamma radiation is mainly attributed to differences in their geological nature of the place, gamma radiation from these radionuclides represents the main external source of irradiation of the human body [25] ,[3].

There are two prominent sources of radioactivities found in agriculture food products taken by the roots of the harvest products, namely the natural

radioactivity within the soil and the absorbed fallouts of the Man-made radionuclides . It has been found that about 85% of the measured activities attributed to the natural radioactivity present in the soil. This implies that the largest Collective Dose received by the world population attributed to the internal ingestion of food products. The exposure to ionizing radiations from natural sources occurs because of the naturally occurring radioactive materials in the soil and rocks, cosmic rays entering the earth's atmosphere from outer space and the internal exposure from radioactive elements through food, water and air. Natural radioactivity is widespread in the earth environment and it exists in various geological formations such as earth crust, rocks, soils, plants, water and air [40], [4]. So the knowledge of the distribution pattern of the both anthropogenic and natural radionuclides in soil plays an important role in radiation protection and their measurements.

Most of the radioactivity in the terrestrial environment whether it is natural or man-made, is bound to the components of the soil. Transportation of this radioactivity from soil is possible to vegetation via dust deposition or root uptake, water sources by flood wash-down, and forward to humans through inhalation, breathing and soil ingestion. Therefore, all pathways of exposure that originate from soil are potentially important for the purpose of radiation risk assessment [50].

For health and environmental safety purposes there is need to measure and evaluate the level of radioactivity in the environment regularly and proper estimation of the radiation dose distribution is vital in assessing the health risk to a population and serves as a reference for documenting changes in environmental radioactivity due to anthropogenic activities [18].

Assess and monitoring radioactivity are therefore of primary importance for humans, organisms and for environmental protection, but rapid and accurate methods for the assay of radioactivity is essential and initiating radiation map of the countries [51].

## 1.1 General statement of the problem:

Since its existence in the environment that we live in, every living creature is exposed to ionizing radiation. This radiation is a part of the earth and comes from cosmos and affects all the goods, food and even the air that we breathe and makes them partly radioactive. So, human beings are exposed to natural background radiation every day from the ground, building materials, air, food, the universe, and even elements in their own bodies. It is because of some reasons that human and its environment are affected by radioactivity, such as, natural radionuclides which are present in soil and the atmosphere, nuclear weapon testing, radioactive wastes and reactor accidents [6]. Radionuclides which give high doses rates, have serious long term effects on human, flora and fauna, including cancer. There is however, considerable controversy regarding the health effects of low-level radiation at or below typical natural background levels. Exposure of the population occurs through three main pathways: inhalation of airborne material, external irradiation from material deposited on the ground, and ingestion of contaminated food-stuff. Doses from external radiation and ingestion will dominate the overall dose. In areas of low deposition, all three pathways may be of similar importance. In Libya most of the agricultural and food-stuff are imported from other countries, some types especially cereals such as wheat imported or produced by private Libyan farmers. Since no studies have been carried out on the content of most Libyan soils and no radionuclide maps on Libyan territory has been established it is vital that the harvested farmers products to be screened for their content of radioactivity. The results of the measurements will be compared between the locally produced to that imported and the measurement and estimation of the dose is crucial for both commodities.

## 1.2 Radioactivity sources and it's pathways to human:

Radioactivity is one of the wide range pollutants contaminate human foods [23]. Radionuclides can migrate from the soil to human's body through the main pathway of plant roots, via food chain. Consumption of foods containing radionuclides make particular hazards [30]. Geographical and geological factors affect the environmental distribution of natural radionuclide concentrations [13].

The natural ($^{232}$Th, $^{226}$Ra and $^{40}$K) and man-made ($^{137}$Cs) radionuclides are known to exist in food and drinks. After ingestion, they contribute to an effective internal dose. It has been believed that a large portion, at least one-eighth, of the mean annual effective dose due to natural radioactive sources is caused by the intake of foods [32]. Natural radioactive decay series such as $^{238}$U and $^{232}$Th as well as singly occurring radionuclides such as $^{40}$K exist in the earth and atmosphere in varied levels [34].

The radioactivity present on air or in the agricultural land and in soil may transfer to the crops grown on it. It happens, however, that an amount of some radioactive elements find their way into human bodies [38].

Through inhalation and ingestion, radionuclides can enter our body. The ingested radionuclides could be accumulated in certain parts of the body. For example, $^{238}$U concentrated in human lungs and kidney, $^{232}$Th in lungs, liver and skeleton tissues and $^{40}$K in muscles [7].

The potassium in soils can be considerably altered by agricultural activities. by means of the soil and chemical fertilizers such as phosphate, radioactive elements find their way to man via the food chain [39]. Humans and other organisms in their environment are under continuous radiation of natural radioactive material found in earth's crust and cosmic rays. In addition, they receive the radiation of medical and industrial sources. The most important natural radionuclides are $^{238}$U, $^{232}$Th , $^{40}$K which with the different physical and chemical properties enter into the physical and biological environments.

$^{226}$Ra with half-life 1620 (y) belong to $^{238}$U chain is one of the main pollutants in the natural radiation environment and there is widely in different ecosystems. Higher solubility of this element than Uranium causes this element to be washed by underground water and brought to the surface. This element is chemically similar to calcium and absorbed by plants through the soil and then through the food chain enters human's body. Almost 70% of $^{226}$Ra is accumulated in the bones and the rest spread to soft tissues of the body. This radionuclide is a bone-oriented element and due to its long half-life remains in bones it may cause bone cancer. However because of alpha radiation serious dangers such as bone marrow cancer can threat human health. Average annual absorption of $^{226}$Ra through the food and drink is about 19Bq in global level that causes effective dose equivalent approximately 3.8 μSv in a year [40].

The main stages of pathways for radioactivity entering the human food chains are:

1-To be up-taken radionuclide by plants through leaves or roots and transferring it to fruit or in cereal and legumes to grains.

2- Radionuclide transport from plants, fodder animals and animal products.

3- Finally human in biological cycle, both through the polluted plants and animal products can be affected.

Therefore, it is necessary to pay attention to radioactivity pollution and their mechanism of absorption. Considering that the main objective of the study of radioactive contamination in plant sources is the impact on humans, makes it necessary to carry out this type of studies [43]. Also, the use of phosphate fertilizers in agricultural land makes multiplier radionuclide levels in soil and eventually plant contamination. Many worldwide studies have shown that food can absorb radioactive materials [40]. Wheat and corn are of important grains which are grown in different regions of the world. This radionuclide could be produced as the results of nuclear tests or nuclear

accidents, normal function of nuclear power plants or reactor accidents that are important ways to do soil contaminations. As a matter of fact $^{137}Cs$ could be found in superficial layers of soil and be absorbed by plant roots constituting one of the main root to the human food chain.

Depositions of large quantities of these radionuclides in particular organs will affect the health condition of the human such as weakening the immune system, induce various types of diseases, and finally increase in mortality rate. Grain and its products, which is the main component of daily Libyan diets serving such as breads, rice, and pasta, are considered as staple food. Since Naturally Occurring Radioactive Materials (NORM) are present in all food commodities, the levels in some grains consumed in LIBYA need to be established in order to forecast any possible radiological risk associated with the consumption of these grains. Thus, the main objectives of this research project is to quantify the presence of natural and Man-made radionuclides in some important grains consumed in LIBYA, and to estimate the radium equivalent index (*Raeq*) and hazard index (*HI*).

## 1.3 The aim of this study:

1- Measuring concentration of natural and man-made radionuclide activities in some types of Cereals consumed in Libya.

2- Determine the Hazard Index for this radionuclide and estimate the radium equivalent index (*Raeq*).

3- Comparing of the radioactivy content of the locally produced to that imported products, and the investigated levels should be compared with imported Cereals, a standard levels should be set for the regulation for any imported Cereal and Grains.

This study should be considered as a part of national programme for monitoring the main part of the food stuff radioactivity and it's dose

contribution to the Libyan population. One of the broad objectives of this study is to estimate the radioactivity levels in the cereal and other food products consumed by the Libyan people in order to estimate their radiation doses and their health hazards impact. Other aim in this study was to measure $^{137}Cs$ as an artificial gamma-ray emitter element with half-life of 30 years.

# CHAPTER TWO

# Previous Studies

## 2. Preface

Radioactivity is defined as the spontaneous break down of a nucleus. The unstable, or radioactive, ones decompose by emitting alpha (α), beta (β), and gamma (γ) radiations. The biological effects related to passage of these radiations through the cells are due to changes in their chemistry caused by ionization, excitation, dissociation, and atomic displacement [12]. Human beings are exposed to background radiation that stems both from natural and man-made sources. Therefore, knowledge of radionuclide distribution and radiation levels in the environment is important for assessing the effects of radiation exposure due to both terrestrial and cosmogenic sources. The planet Earth is a radioactive planet and natural radiations are present in every human environment; Earth's material, water, air, foods, and even human body contains naturally occurring radioactive materials. Natural background radiation, which is equivalent to 2.4 mSv per person, makes up approximately 80% of the total radiation dose a person is exposed in a year [35], [36].

Natural radioactivity has played an indispensable role in the development of earth sciences [29], usually the natural radionuclide's are very low activity concentrations. The primordial radionuclides $^{238}U$ and $^{232}Th$ exist in soil, in varying concentrations. These can be attributed to the nature of the parent rock during soil genesis. Studying transfer of natural radionuclides, like $^{238}U$ and $^{232}Th$ along with their daughter products through the biosphere is important because their ubiquitous presence and persistence in the environment. Soil–vegetables are recognized as one of the major pathways for the transfer of radionuclides to human beings Transmission of uranium and thorium along with nutrients through the absorption of minerals, and accumulate in various green parts or even up to the edible parts of the vegetation. [24]. The natural or artificial radioactive nuclides that are present in the environment are main sources of radiation exposure for human

beings and constitute the background radiation level. Evaluation of the distribution of these radionuclides is very necessary for assessing the effects of radiation exposure. The terrestrial component of the background is due to various radioactive nuclides that are present in air, soil, water and building materials whose abundances vary significantly depending on the geological and geographical features of a region. The cosmic component, on the other hand, originates from outer space as cosmic rays whose contribution to the background changes mainly with elevation and latitude. In addition to the natural sources, the level of background radiation in a region is considerably affected from man-made sources such as those from nuclear activities and accidents [68]. Therefore, many researchers throughout the world are continuously interested in measurements of natural radioactivity in air, soil and drinking water samples[6].

The distribution of natural and artificial radionuclides in environmental settings and the contributions of these radionuclides to the radiation dose to which people are exposed should be defined separately . Since the doses from these pathways are strongly related to the amount of radionuclides present, an important objective from the point of view of the radio ecological protection of the population is the accurate evaluation of the amounts received in dietary intake [15].

## 2.1 Environmental radioactivity:

Ionizing radiations enter into our lives in a variety of ways. Natural sources of radioactivity include cosmic rays, gamma rays from the Earth, radon decay products in the air, and various radionuclides in edibles and potables [2].

Human beings are exposed to ionizing radiation from natural sources throughout their lifetime. Therefore, knowledge of radionuclide distribution and radiation levels in the environment is important for assessing the effects of radiation exposure due to both terrestrial and cosmogenic sources. The planet Earth is a radioactive planet and natural radiations are present in every human

environment; Earth's material, water, air, foods, and even human body contains naturally occurring radioactive material, International Atomic Energy Agency [63], Radionuclides present in the environment are normally found in low concentration. These may be naturally occurring (e.g. uranium-238, thorium-232, and their daughter products and potassium-40 or those produced by cosmic rays interaction in the biosphere) or man- made sources (e.g. originating from nuclear fallouts or emanating from nuclear facilities) [36].

NORM has sources of natural radiation and radioactivity that are classified as extraterrestrial and terrestrial sources. In term of population radiation dose, the sources of natural radiation are the most significant and the main contributor to the population collective doses [67].

Primordial radionuclides are long-lived species, which have been present on earth since its formation about $4.5x10^9$ years ago. They are classified into:

(i) Series radionuclides ; groups of radionuclides that are headed by parent radionuclides that decay in sequence to other radionuclides with different half-lives and decay modes, and finally end as stable isotopes, National Council on Radiation Protection and Measurements (NCRP,1992). There are three natural series headed by uranium-238, uranium-235 and thorium-232, the main members of which are shown in figures 2.1.

(ii) Non-series radionuclides; they decay directly to stable nuclide. The most important radionuclides are the isotopes of potassium-40, vanadium-50, rubidium-87, cadmium-113 and indium-115. In term of population dose, the most significant radionuclides are potassium-40 and rubidium-87.

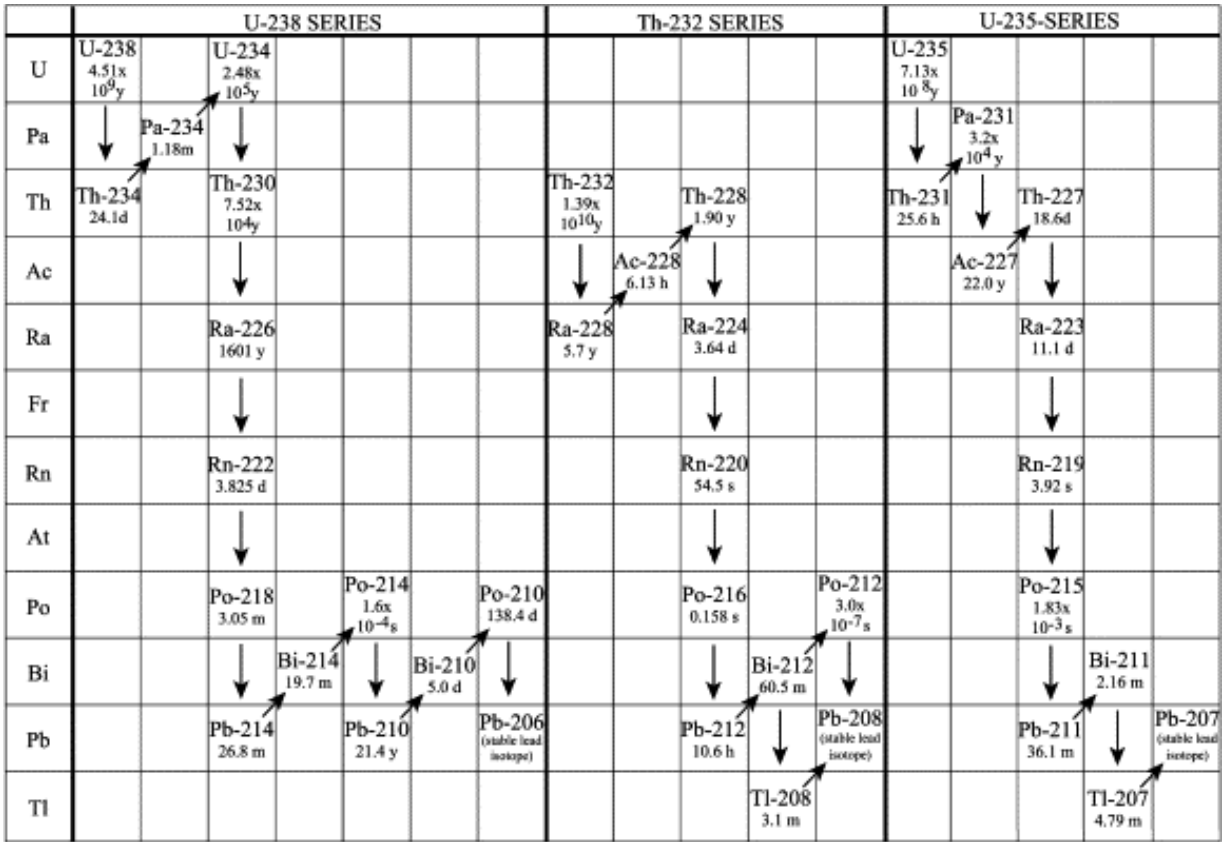



**Figure 2.1. The Uranium ($^{238}$U), Thorium ($^{232}$Th), and Uranium ($^{235}$U) decay series.**

## 2.1.1 Radioactivity level in Rocks:

Radioactivity of rocks is usually caused by one of three natural sources of gamma-radiation: potassium, uranium and thorium. Each of those elements in natural conditions contains a fraction of radionuclide that can be detected in either direct or indirect way [63].

Radionuclides are present in rocks in varying amounts, and they are easily mobilized into the environment. Radioactivity in soil results from the rock from which they were derived. The distributions of naturally occurring radionuclides depend on the distribution of rocks from which they originate and the processes which result to their removal from the soil and migrate them. Therefore, the natural environmental radioactivity mainly depends on geological and geophysical conditions [17]. According to the [68]. report, higher radiation levels are associated with igneous rocks such as granite and

the levels are lower in sedimentary rocks. Also phosphate rocks are known to contain relatively high content of radionuclides. There are exceptions, however, as some shale's and phosphate rocks have relatively high content of radionuclides, are widely used as building materials [25]. It is therefore important to measure the concentration of radionuclides in rocks that are used and those that have the potential of being used as building materials for assessing the radiological risks to human health.

Extra-terrestrial cosmic radiation in the soil and rock give us an average dose of up to 60 μ rads $y^{-1}$ [66]. the activity levels in soils are related to the types of rocks from which the soils originate and hence it will effect the radionuclide uptake by the their roots.

The radionuclides existing in phosphate rocks can enter the human environment through several ways such as fertilization of agricultural lands, usage of phosphogypsum in agriculture and building materials. Large deposits of rock phosphate, and other phosphorous compounds, are found in many geographical localities. These deposits are of great economic value for the manufacture and agricultural fertilizers (superphosphates), which will in turn effects radioactivity level uptake by the harvest crops [11].

**Table2.1. Typical activity concentration of $^{40}K$ , $^{238}U$ and $^{232}Th$ in common rocks and estimated absorbed dose in air 1 m above the surface (UNSCEAR, 1977).**

| Type of rock | Typical activity concentration (pCi $g^{-1}$) | | | Absorbed dose rate in air (μ rad $h^{-1}$) |
|---|---|---|---|---|
| | **$^{40}K$** | **$^{238}U$** | **$^{232}Th$** | |
| **Igneous** | | | | |
| Acidic(e.g. granite) | 27 | 1.6 | 2.2 | 12 |
| Intermediate(e.g.diorite) | 19 | 0.62 | 0.88 | 6.2 |
| Mafic(e.g.basalt) | 6.5 | 0.31 | 0.30 | 2.3 |
| Ultrabasic (e.g.durite) | 4.0 | 0.01 | 0.66 | - |
| **Sedimentary** | | | | |
| Limestone | 2.4 | 0.75 | 0.19 | 2.0 |
| Carbonate | - | 0.75 | 0.21 | 1.7 |
| Sandstone | 10 | 0.5 | 0.3 | 3.2 |
| Shale | 19 | 1.2 | 1.2 | 7.9 |

## 2.1.2 Radioactivity level in soil:

Natural radioactivity in soil constitute one of the most component of the background radiation exposure to population. Human beings always exposure to ionizing radiation owing to primordial radionuclides such as $^{238}U$ decay series ,$^{232}Th$ series and $^{40}K$ that are found in the earth's crust, and to cosmic ray second arias, always each of us is exposed to naturally occurring quantities of radiation sources of ionizing radiation found in soil result from the rock from which the soil is derived. Levels of ionizing radiation will vary as a result of diminution by leaching of water, dilution by

increased porosity, and by augmentation from water and organic matter by sorption and precipitation of radionuclides [16]. Average activity concentrations of $^{40}K$ , $^{238}U$ and $^{232}Th$ in various types of soil and estimated absorbed dose in air 1 m above the surface is shown in Table 2.2. It should be noted, however, that the main factor influencing the concentration of the natural radionuclides in soil is not the soil forming process but the corresponding concentration in the soil forming rocks (UNSCEAR, 1977).

**Table 2.2. Average activity concentrations of $^{40}K$ , $^{238}U$ and $^{232}Th$ in various types of soil and estimated absorbed dose in air 1 m above the surface.**

| Type of Soil | Average Activity Concentration(pCi $g^{-1}$) | | | Absorbed dose rate in air (μrad $h^{-1}$) |
|---|---|---|---|---|
| | $^{40}K$ | $^{238}U$ | $^{232}Th$ | |
| Serozem | 18 | 0.85 | 1.3 | 7.4 |
| Gray-brown | 19 | 0.75 | 1.1 | 6.9 |
| Chestnut | 15 | 0.72 | 1.0 | 6.0 |
| Chernozem | 11 | 0.58 | 0.97 | 5.1 |
| Gray forest | 10 | 0.48 | 0.72 | 4.1 |
| Sodpozolic | 8.1 | 0.41 | 0.60 | 3.4 |
| Podzlic | 4.0 | 0.24 | 0.33 | 1.8 |
| Boggle | 2.4 | 0.17 | 0.17 | 1.1 |
| World average | 10 | 0.7 | 0.7 | 4.6 |
| Typical range | 3-20 | 0.3-1.4 | 0.2-1.3 | 1.4-9 |

### 2.1.3 Radioactivity levels in staple food and vegetation

Food can be contaminated with a wide range of pollutants including radioactivity, ingestion of foodstuffs constitutes an important pathway by which can be transferred to humans. There are many human activities which can enhance the level of naturally occurring radioactivity levels in the environment. As is commonly known a large contribution to the radiation dose received by humans comes from natural occurring uranium series radionuclides accumulated in the body.

Investigation of natural radioactive isotopes present in foodstuffs and assess the dose are extremely important for controlling radiation levels to which mankind is direct or indirectly exposed. Another important fact is that, importation of food that might contains some levels of radioactivities "contamination" from any region that suffered a nuclear accident can be indirectly affect people health around the world [31].

Consumption of food is usually the most important route by which natural radionuclides can enter the human body [64], 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. The status of the soil on which food crops are grown will determine, to a significant extent, the quality of foodstuffs produced. [28].

The naturally occurring radionuclides in food come mainly from natural isotopes of potassium, uranium and thorium and their daughter products. The majority of radionuclides in the environment are present as daughter products of $^{232}$Thm and $^{238}$U, distributed by natural geological and geochemical processes, in addition to the unrelated naturally occurring $^{14}$C and $^{40}$K. There are many human activities which can enhance the level of naturally occurring radioactivity levels in the environment. [27]. Radionuclides together with some other heavy metals are the primary toxic

pollutants from the manufacture of phosphate fertilizer. Artificial fertilizing has steadily replaced the natural fertilizing in agriculture; therefore, fruits and vegetables may contain radionuclides found in phosphate fertilizers. This fact is of serious concern, because the daily intake of radionuclides with food may be considered as chronic ingestion. An investigation of the radionuclides pathway from fertilizers to plants and to humans is, therefore, very important from the viewpoint of radiological protection within the general population .

Plants organisms assimilate and deposit in their-various natural radioactive isotopes present in the soil irrespective of biological necessity. Radium enters the above-ground plant organs show no tendency to form chemical compounds of low mobility in plant tissues. Under normal environmental conditions about 90% of $^{226}Ra$ enters the human body through food. Ingestion is therefore the most important mode of exposure to be considered when assessing the dose to man. To support that work, the magnitude of soil/plant transport of naturally occurring radionuclides such as $^{226}Ra$ has to be evaluated in different regions of the world. This is necessary both for dose assessment and for predictive modeling along the terrestrial food chain [47].

In Sudan the most important kind of food are grains and roots. The maize constitutes a large percentage of the total diet, while Tomato, Okra and the leafy vegetables like Portulaca and (molokhia) come at the second level in combination with (maize) sorghum bread (ksra).

## 2. 2 Literature review

The nationwide average concentrations of $^{226}Ra$, $^{238}U$, $^{232}Th$, $^{40}K$ and $^{137}Cs$ determined were 31.62, 20.11, 19.10, 280.29 and 4.12 $Bq.kg^{-1}$, respectively. This shows that there is little contamination due to fallout radioactivity at survey sites.

In central Sudan, [48], in uranium mineralization areas at Jebel Uro, Jebel Kurun and Jebel Mun in Western Sudan, absorbed dose rate received from natural external irradiation was evaluated from the measured activity concentrations of $^{238}U$, $^{232}Th$ and $^{40}K$ in rock samples. Uranium was the principal producer of the surface radioactivity at Uro and Kurun as it contributed 99.6% and 95% of the total absorbed dose whereas, in Jebel. Measurements were performed using a high-resolution gamma spectrometer. Independent of location, the data showed that the activity concentration for $^{226}Ra$ and $^{232}Th$ increased in the order: soil>spoil heap>ore. This order basically confirms that the activity levels in the heap were low relative to natural background (i.e. soil samples), and thus the expected contribution in enhancement of natural background radiation was correspondingly low.

The concept of concentration ratio (CR) has been applied to quantify the degree of accumulation of radionuclides in the solid waste (heap). The calculated CR values revealed that $^{232}Th$ was most accumulated while there was no accumulation of $^{40}K$.

The principal contributors to the total dose rate were $^{40}K$, $^{214}Bi$ and $^{228}Ac$ which generate almost 90%.

In Western Sudan has carried out an environmental monitoring study at Miri Lake which is identified as high level background natural radiation areas in Nuba Mountains. They used lake and well water for drinking and irrigation. Food, water and soil samples were collected and measured for their radioactivity using high resolution gamma-spectrometry, laser flourimetry, X-ray fluorescence, gross alpha-beta measurement and alpha spectrometry.

Exposure of population to the external and internal radiation was estimated. A sodium iodide scintillation detector was used for measurement of the external exposure. Environmental samples were collected, processed and measured. Samples from places of normal background radioactivity were measured to serve as reference level for comparison. From these results the author concluded that Miri lake area was an elevated natural radioactivity level. The level of elevation is in the range of 2.5-11 times greater than in places with normal background natural radioactivity. The upper limit of the average annual effective dose to the population was estimated at 39.5 mSv due to the external and ingested food and due to radon inhalation in the area. In Northern Sudan, [16] has carried out assessment and GIS mapping of radioactivity in the Northern State. Activity concentration of $^{238}U$, $^{232}Th$ , $^{40}K$ and $^{137}Cs$ in soil samples collected from different locations have been measured using high resolution γ-spectrometry. $^{137}Cs$ was found to be of very little contribution to the total exposure. Absorbed dose rate in air at a height of 1m from the ground was calculated using six sets of dose rate conversion factors and the corresponding annual effective dose was estimated, study area was lie within the worldwide range for normal background radiation areas. Additionally, [58]. have been carried out Investigation for naturally occurring radionuclides in surface soils in selected regions of the Northern State of Sudan ,reported activity concentrations of $^{226}Ra$, $^{232}Th$ and $^{40}K$ in different regions of the Northern State was found to be 82.6 ± 6.8, 26.0 ± 2.9 and 726.4 ±72.0 $Bqkg^{-1}$ [37] , comprehensive studied was carried out for the determination of different radionuclides activities in foodstuff consumed and evaluation of dose levels in different food stuffs were collected from eight states in Sudan (cereals, vegetables, meat, fruits, milk and fermented milk, baby milk, cans, spices, additives, others). The concentrations of different radionuclides in the food samples were determined by gamma - spectrometry using an HPGe detector.

Radionuclides observed include: $^{212}$Bi, $^{214}$Bi, $^{134}$Cs, $^{137}$Cs, $^{40}$ K, $^{212}$ Pb, $^{214}$Pb, $^{224}$Ra, $^{226}$Ra, $^{228}$Th, $^{228}$Ac, $^{208}$Tl, $^{232}$Th and $^{238}$U. The activity concentration of these radionuclides were found in the following ranges: 0.51 - 19.42 Bq.kg-1, 0.47 - 12.13 Bq.kg-1, 0.5 - 1.29 Bq.kg-1, 0.001 - 3.41 Bq.kg-1, 19.25 – 2521.82 Bq.kg-1, 0.08 - 6.84 Bq.kg-1, 0.02 - 6.87 Bq.kg-1, 6.08 - 32.02 Bq.kg-1 , 0.03 - 21.53 Bq.kg-1, 0.92 - 26.77 Bq.kg-1, 0.91- 12.00 Bq.kg-1, 0.14 - 2.58 Bq.kg-1, 0.03 - 9.65 Bq.kg-1 and 0.82 - 5.27 Bq.kg-1 respectively. High concentrations were typically found in Portulaca; the lowest concentrations were found in barley and bread additives. The annual effective dose due to the different foodstuff estimated was found to be 2.78 ±0.44 mSvy$^{-1}$ and 1.18 ± 0.18 mSvy$^{-1}$ for age categories 7-12 y and > 17y respectively.

In [66], [67]. a value of 2.4 mSv was quoted for the population living at Places of normal background radioactivity. The maximum annual committed effective dose equivalent of 5 mSv was recommended by the ICRP. This recommendation was justified by comparison with normal natural background radiation which has a value of 2 mSv. The value presently recommended by the ICRP for the principal limit for members of the public is 1μSv.

Various studies concerning radioactivity bound to soil were carried out in Worldwide, in Saudi Arabia a study was Measurement Radioactivity and Radiation Dose assessments in Soil from Albaha Region, Saudi Arabia [42]. That reported activity concentration and the gamma absorbed dose rates of the terrestrial naturally occurring radionuclides $^{226}$Ra, $^{232}$Th, and $^{40}$K were determined in soil samples collected from twenty different locations of Albaha region in Saudi Arabia, were performed using a NaI(Tl) gamma-ray spectrometer. The typical concentrations of $^{226}$Ra , $^{232}$Th and $^{40}$K were found in surface soil samples ranged from 30.3±1.6 (sample18) to 45.3±1.9 (sample3) Bq.kg-1 and from 26.0±1.8 (sample 17) to 37.5±1.8 (sample 18)

Bq.kg-1 and 263.2±6.4 (sample 4) to 434.9±5.4 Bq.kg-1 sample 13) with overall mean values of 37 Bq.kg-1,32 Bq.kg-1 and 343 Bq.kg-1, respectively. The mean radium equivalent (*Raeq*) and outdoor radiation hazard index (*Hex*) for the area under study were determined as 116 $Bqkg^{-1}$ and 0.29 respectively. The absorbed dose rate due to three primordial radionuclides lies in the range from 43to 60.3 $nGyh^{-1}$ with a mean of 50 $nGyh^{-1}$, which yields the annual effective dose of 63 $\mu Svy^{-1}$ which is well below the permissible limit . The measured values ware comparable with other global radioactivity measurements and are found to be save for public and environment . The baseline data of this type will almost certainly be of importance in making estimations of populations exposure.

In Pakistan was reported Measurement of naturally occurring/fallout radioactive elements and assessment of annual effective dose in soil samples collected from four districts of the Punjab Province, Pakistan using a P-type coaxial high purity germanium (HPGe) c-ray spectrometer[52]. The specific activities of $^{226}Ra$ , $^{232}Th$ and $^{40}K$ of the soil samples ranged from 26.02 ± 7.11 to 93.54 ± 8.13 Bq.kg-1, 29.34 ± 2.58 to 114.41 ± 2.80 Bq.kg-1 and 348.15 ± 3.20 to 752.98 ± 4.20 Bq.kg-1, respectively. Activity due to $^{137}Cs$ was observed in some locations which ranged from 0.4 ± 0.2 to 7.8 ± 0.3 Bq.kg-1. From the measured activity concentrations, radium equivalent activity concentrations were determined followed by calculations of mean absorbed dose rate and mean annual effective dose for the inhabitants of the studied area. The mean radium equivalent activity, internal and external hazard indices values came out to be 179.26 ± 11.93 Bq.kg-1, 0.64 ± 0.05 and 0.48 ± 0.03, respectively. Indoors and outdoors average annual effective dose values were found to be 0.42 ± 0.03 and 0.10 ± 0.01 mSv, respectively. Present data have been compared with the published data for other parts of the world and found to be safe for public and environment.

In Spain a programmer of studies and surveys of natural radiation and radioactivity was organized to determine the concentrations of natural radionuclides in soil [70]. The results showed that the averages and ranges of concentrations of $^{226}$Ra, $^{232}$Th, and $^{40}$K in soil in most autonomous regions and nationally were higher than the world figures reported by [67], (world average concentrations of 30, 25, and 370 Bq.kg-1 for $^{226}$Ra, $^{232}$Th, and $^{40}$K respectively and typical ranges were 15-70 Bq.kg-1 For $^{226}$Ra, 7-50 Bq.kg-1 for $^{232}$Th, and 100-700 Bq.kg-1 for $^{40}$K ). Only the regions of Andalucia, Baleares, Murcia, Navarra, and PaisValenciano showed results comparable to the typical UNSCEAR values for the entire world, except for the range of the $^{40}$K concentrations measured in the region of Andalucia, whose upper limit was more than double that given by UNSCEAR. In relation to the geological structure of the country, the region of lower average concentrations of $^{226}$Ra, $^{232}$Th, and $^{40}$K in soil match those areas that essentially consist of recent marine sediments (mainly limestone , marbles, and clays) ranging in age from the Late Oligocene to the present.

In Bangladesh the studied have been estimated and Assessment of Natural Radioactivity and Radiation Hazard in Soil samples collected from different Upazila of Rajbari district of Bangladesh were analyzed by gamma-ray spectrometry [46]. The average natural radioactivities of $^{238}$U, $^{232}$Th and $^{40}$K have been measured as 29.03±5.67 Bq.kg$^{-1}$, 50.91±10.17 Bq.kg$^{-1}$ and 535.32±89.19 Bq.kg$^{-1}$ respectively. To estimate health effect due to the activity of those radionuclides, the radium equivalent activity (Ra absorbed dose rate (D) and effective dose have been calculated and compared to the world average values. The results are also compared with the literature values reported for other regions of the world and found that the soil of the study area are not hazards by the radiation and does not pose any harmful effect to the environment. The outcome of this study may provide valuable

information about radiation hazard and also may take part in the monitoring of environmental radioactivity.

In the Central and Eastern region of the Brazilian State of Rio Grande do Norte, 51 soil samples were collected to a depth of 30 cm (Malancoet al., 1993). Concentrations of background radionuclides in soil samples were determined by gamma ray spectrometry with an HPGe detector. The average concentrations of $^{226}$Ra, $^{232}$Th and $^{40}$K in the surveyed soils were 29.0 ±19.4 Bq.kg-1 46.6 ± 36.2 Bq.kg-1 , and 677.8 ± 434.9 Bq.kg-1 respectively. The bedrock of Sanata do Matos (Rio Grando do Norte) showed fairly high radioactivity (90 Bq.kg-1 of $^{226}$Ra, 285.6 Bq.kg-1 of $^{232}$Th and 1.414 Bq.kg-1 of $^{40}$K).

In South Africa the aim of this study was to investigate the potential radiological impact of foodstuffs grown Ina catchment area influenced by mining and mineral reprocessing industries in South Africa. Natural radionuclides were determined in a number of foodstuffs harvested from and/or grown in the area. The radionuclides were measured by non-destructive techniques such as Instrumental Neutron Activation Analyses (INAA) and low background gamma spectrometry. The estimated dose for the adult age group, resulting from consumption of these foodstuffs, was evaluated [64]. Sensitive measurement of three major radionuclides (in addition to $^{238}$U, $^{234}$U, $^{232}$Th, $^{226}$Ra, $^{224}$Ra and $^{223}$Ra) is necessary to calculate the estimated annual dose with a high degree of certainty i.e. $^{230}$Th, $^{210}$Pb and $^{210}$Po, while $^{231}$Pa, $^{227}$Ac and $^{228}$Ra also require improved sensitivity. In Orderto evaluate the yearly dose due to an individual source at a screening level of 25 μSv/a, one is faced with a required lower limit of determination (LLD) of 0.1 to 0.5 Bq/kg for certain foodstuffs. The assessment of natural radionuclides in foods allowed us to evaluate the items that present the highest risk to the population, and compare this to the limits established by the National Nuclear Regulator (NNR).From the public's point of view it is important to ensure the population that the

contaminant levels in specific food as a result of mining activities, do not exceed the permissible limits. For the majority of the analyses foodstuffs, the estimated dose was less than 250 μSv/a. Overestimation of dose, due to poor measurement detection limits, was clearly indicated for some of the samples. For most of the analyses nuclides, suitable data for evaluation of the yearly dose at the screening level of 25μSvy$^{-1}$ was obtained by broad energy gamma analysis of ached samples. However, nuclides such as $^{230}$Th, $^{210}$Po and $^{231}$Pa has to be analyses by radiochemical separation through acid destruction of dried foodstuffs followed by individual element separations to provide suitable data with a low enough LLD not to result in an overestimation of the calculated dose.

In Tanzanian was measured Radioactivity levels of staple foodstuffs products (maize and rice) from various localities of Tanzania and dose estimates [27]. The average activity concentrations of $^{40}$K, $^{232}$Th and $^{238}$U in maize were 48.79 ± 0.11, 4.08 ± 0.01and 13.23±0.10 Bq.kg$^{-1}$, respectively. In rice the concentrations of $^{40}$K, $^{232}$Th and $^{238}$U were 24.67±0.03, 3.82±0.02 and 5.02±0.02 Bq.kg$^{-1}$, respectively. $^{137}$Cs was detected in only one sample collected in Zanzibar. The sample, with activity concentration of 5.57 ± 0.01Bq.kg$^{-1}$, had been imported from Thailand. The relatively high average concentrations of the radionuclides in maize compared to rice may be attributed to the extensive use of phosphate fertilizers in maize production in Tanzania. Total annual committed effective doses due to total $^{232}$Th and $^{238}$U intakes as a result of consumption of staple foodstuffs for infants, children and adults were 0.16, 0.29 and 0.36 μSvy$^{-1}$, respectively, which are lower than the annual dose guideline for the general public.

Forty samples of different types of imported and locally produced grains consumed in Jordan were analyzed using gamma-ray spectroscopy system with a high Purity germanium (HPGe) detector. The concentrations of the natural radionuclides $^{238}$U, $^{232}$Th and $^{40}$K present in

the studied samples were measured, and the radium equivalent activities $Ra_{eq}$, were calculated. In addition to that, the hazard index HI, was calculated. The average concentrations of $^{238}U$, $^{232}Th$ and $^{40}K$ were in the range of $(2.0 \pm 0.5) \times 10^{-3}$ , $(7.0 \pm 2) \times 10^{-3}$ and 49.93 ± 1.69 ppm, respectively. The values of $Ra_{eq}$ ranged between 17.70 – 245.64 $Bq.kg^{-1}$. The HI were ranged between 0.05 – 0.66, which is less than one (the higher limit of HI). The obtained results were compared with the standard accepted international values, and found to be within the acceptable limits [7].

The radioisotopes and nuclear explosions in upper layers of the atmosphere have vigorously polluted the earth. These radionuclides can be transferred to humans through the food chain. Rice is important and a major source of carbohydrates for humans. Soil contains the natural radionuclide such as Uranium, Thorium and Potassium. A study was conducted to determine the radionuclide content and their level of radioactivity in various types of rice eaten by the Malaysian. Six varieties of rice, Faiza Basmati, white glutinous, black glutinous, Siam, Kurnia and Utara were taken under consideration. Hyper Pure Germanium Detector (HPGe) was used to measure the radioactivity level. The average concentration of Uranium, Thorium and Potassium was found to be in the range of (18.33-25.10)±0.01 Bq.kg-1 , (35.49-64.97)±0.01 Bq.kg-1 and (64.802-109.929) ± 0.001Bq.kg-1 respectively. Effective dose per annum was resided in the range of 0.02 $\mu Sv.year^{-1}$ to 0.03 $\mu Sv.year^{-1}$ [45].

Radionuclides ingested in food and to a lesser extent, water, account for a substantial part of the average radiation doses received by various organs of the human body, especially the skeleton. A study of natural radionuclides in rice consumed in Saudi Arabia was performed. The study targeted the natural radionuclides $^{226}Ra$, $^{232}Th$, and $^{40}K$, in addition to heavy metals (Cd, Zn, Fe,Cr, Pb and V). It was found that rice consumption in Saudi Arabia

is radiologically safe for the presence of the investigated radionuclides. The presence of 40K in all samples in higher levels was anticipated due to its natural abundance. The levels of the toxic metals, Lead (pb), Cadmium (cd) were not detected in examined rice samples. The mean values of Iron, zinc and manganese were higher than findings reported by other studies in literature.

Saudi Arabia studied was the first at the national level to investigate the radioactivity and heavy metals of rice. The findings of that study will was to establishing a baseline of radioactivity and heavy metals for public exposure from foodstuff ingestion. the activity concentrations for $^{226}Ra$, $^{232}Th$, and $^{40}K$ in the different rice samples consumed in Saudi Arabia. $^{226}Ra$ was detected in all samples with a maximum value of 2.6 $Bq.kg^{-1}$ (a sample from India), a minimum value of 0.1 $Bq.kg^{-1}$ (a sample from USA) with average value 0.4 $Bq.kg^{-1}$. As for $^{232}Th$, it was detected in all samples with a maximum value of 2.3 $Bq.kg^{-1}$ (a sample from India), a minimum value of 0.1 $Bq.kg^{-1}$ (a sample from Thailand) with average value 0.2 $Bq.Kg^{-1}$ . $^{40}K$ was detected in all samples with a maximum value of 257.2$Bq.kg^{-1}$ (a sample from India), a minimum value of 20.9 $Bq.kg^{-1}$ (a sample from India) with average value 138 $Bq.kg^{-1}$ . The radium equivalent concept (Raeq). The calculated values are varied from 1.7 to 20.2 Bq.kg-1 with an average of 10.93 $Bq.kg^{-1}$ [5].

A study of long-lived gamma emitting radionuclides in rice consumed in Kuwait was performed. The study targeted the natural radionuclides $^{238}U$, $^{232}Th$, and $^{40}K$, in addition to the anthropogenic radionuclide $^{137}Cs$. Annual effective doses from rice consumption were estimated to be 33 and 60 µSv for the adult and child age groups respectively. These values were found to be of several orders of magnitude less than the 0.29 $mSv.year^{-1}$ world average of the ingestion exposure from natural sources reported in the

literature. Moreover, the anthropogenic radionuclide $^{137}Cs$ was detected in one sample only, out of the 21 samples measured, with an activity concentration of 0.1 $Bq.kg^{-1}$. This small value is four orders of magnitude less than the guideline limit [56].

# Chapter Three

# Materials and Experimental Methods

## 3.1 Materials and Methods

Twenty four types of grains were taken as the sample. All samples used were both local and imported grains and were obtained from local markets. The name of the samples and their number are as follows: rice (9), beans (1), peas (1), lentil (2), chickpea (1), corn (3), Bean Mdch (1), ring (1) , Barley(3) and wheat (2). This makes the total number of samples collected as twenty four shown in Table 3.4.

A total of 24 of various Cereal and Grains samples some were locally produced in Libya others were imported to be consumed by the Libyan people. The collection took place between April and June of 2015. To ensure a comprehensive and a wide-spread representation, different brands that originated from different countries were selected Table 3.4. Since cereal is locally produced in LIBYA and some samples were imported.

Because nuclear transformation cannot be sensed by God gifted human sensors, it can only be measured by especial electronic detectors. For each type of the four main emitted radiation; either in the form of electromagnetic radiation such as gamma-ray or in the particular form such as alpha, beta or neutrons especial type of detector based on it's detection theory has been appropriately made. For this research project manly concentrated on the gamma ray detection of the emitted radionuclide, the High Purity Germanium Detectors (HPGe-detector) has been used utilizing the gamma-ray spectroscopy software. The Libyan Environmental General Authority "EGA" laboratory which has the proper setting of two HPGe-detectors connected to specific electronic equipment and the needed shielding for low level germanium detector measurements. For this reason gamma ray interaction and germanium detectors will be thoroughly introduced and investigated in the next sections.

## 3.2 Gamma-ray Detectors:

For the detection and measurements of low level gamma emitting radionuclides two main types of radiation detectors are being widely utilized namely, the Scintillations detectors "NaI" and Semiconductors such as Germanium detectors. Each type of these measurements techniques has advantages and disadvantages on each other.

### 3.2.1 Scintillation detectors "NaI-detectors".

Scintillation detectors use an inorganic scintillators such as sodium iodide, which are able to be operated at room temperature. As a result, it is known as a practical and in-situ measurement systems. The electrons generated by the radiation interaction are transferred into photoelectrons. The gamma-ray photons leave their energy in the scintillators (a florescence material) leading to atoms rising to exited states [60].

The excited atoms may lose their energy through emitting visible photons and the electrons go back into the valence band. These visible photons may emit after a de-excitation that hits the photosensitive surface followed by the generation of photoelectrons. A multiplying and accelerating of the photoelectrons may lead to production of electrical pulses using photomultiplier tube. Visible photons self absorption may occur in a pure NaI crystal. Figure 3.1 shows Scintillation detector.

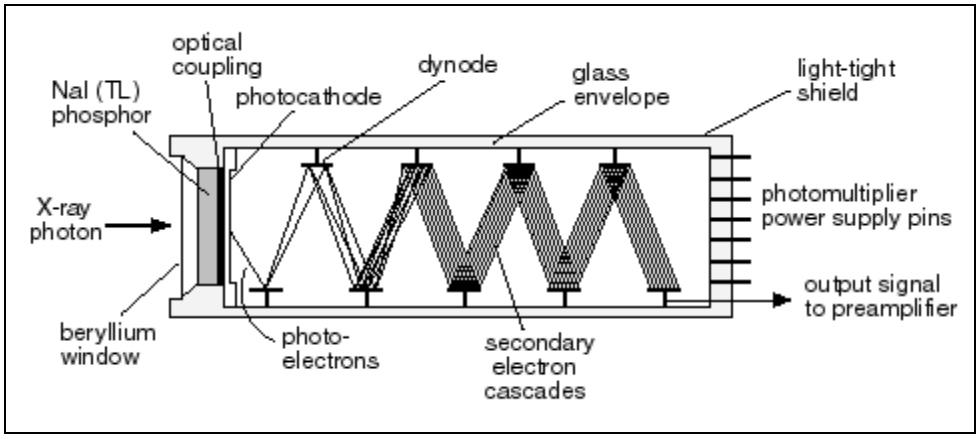



**Figure 3.1. Scintillation detectors**

To decrease this effect and increase the probability of the transmission of visible photons, an amount of impurities, called 'activators' such as the element thallium (TI), are add to the crystal [61]. The main advantage of this type of detection is having high efficiency while low resolution is one of it's disadvantages. This type of research requires high resolution detections therefore, the germanium technology has been used.

### 3.2.2 Semiconductor Detector HPGe:

These detectors HPGe are one of the major tools for the detection of photon because of its excellent energy resolution compared with scintillation detectors. HPGe detectors operates at e very low temperature which means it needs cooling system such as liquid nitrogen [30]. Concerning the efficiency an 80% efficiency detector has been utilized to gain more statistical meaningful counts for a range of gamma-ray energies in order to accurately measured radionuclides off interest as shown in the detector certificate in Appendix A. HPGe detectors convert photons into direct charge carriers [39]. They have high spectral quality, excellent peak symmetry and a low noise operation [61].

The semiconductor germanium is widely used in gamma-ray spectroscopy. It has a conductivity that range between insulators and

conductors. 1eV is the lowest amount of excitation energy needed for electrons in this semiconductor to pass across the band gap into the conduction band. The main signal information is from the electron-hole pairs. These are collected to form the corresponding energy signal [30].

P and N-type are two main materials of HPGe semiconductor that are widely used. In the semiconductor detectors electrons-holes pairs create an electrical field in a limited space known as depleted region, which is neither includes neither particles nor charges. Any charged particles or photon in this region may be influenced by the electrical field that is controlled by the bias voltage.

Germanium crystals should be kept in a low temperature by using liquid cryostat that containing nitrogen at liquid form (LN2), consequently, these charged particles that created in the depleted region would be collected and solve the current leakage problem [32].

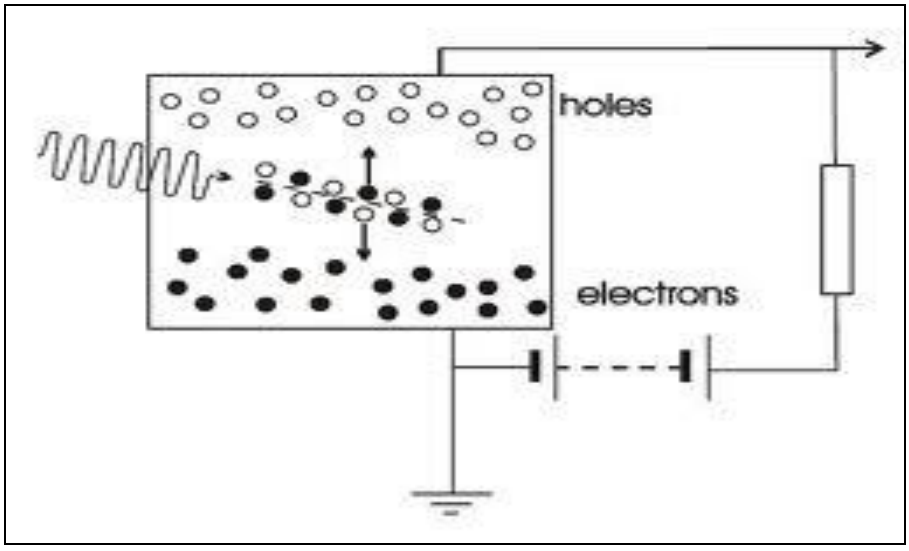



**Figure 3.2. The high-purity germanium detector system HPGe**

Compton scattering followed by photoelectric absorption of the scattered photon is the most common process, which contributes to the full-energy peak [13]. The germanium detector used for this study is a P-type

detector with an 80% relative efficiency is shown in Fig 3.4 and electronic supporting system is shown in Figure 3.5.




**Figure 3.4. Shows the Laboratory setting of the germanium detectors.**




**Figure 3.5. Shows the supporting electronic measuring system to the HPGe-detector**

### 3.2.3 Typical HPGe-detector Geometry

It is ideal for the germanium detector to maximize volume in order to absorb as many interacting gamma-rays as possible. In order to do this, the used detector is a P-type and has a coaxial geometry as shows in Figure 3.6

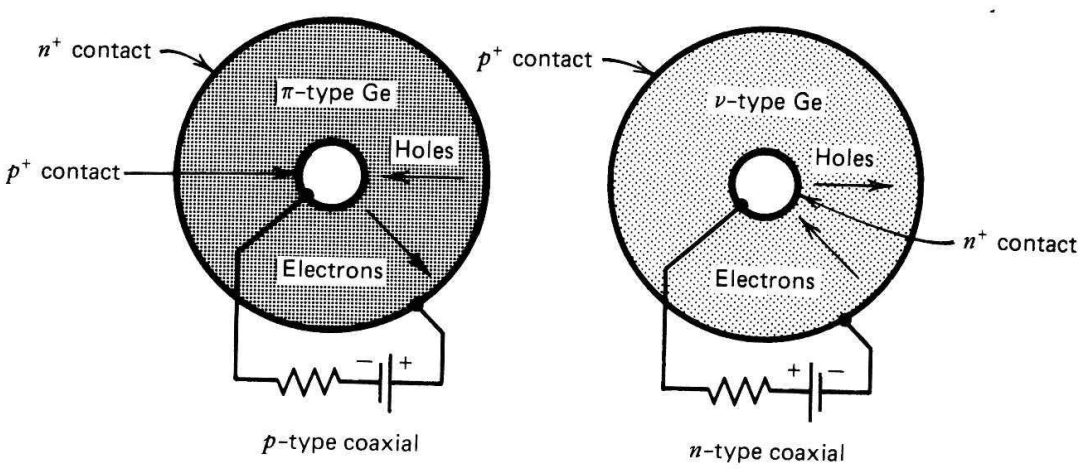



**Figure 3.6. Cross sections perpendicular to the cylindrical axis of the detector crystal**

There are electrodes connected to contact on the inside of the coaxial and the outside of the coaxial. A potential is applied across the coaxial and a potential is applied across the detector. Due to the geometry of the detector, electron-hole pairs have to travel different distances to get to the electrodes, depending on where they were created. Because this collection time is not constant, the pulse shape is also not constant as well.

## 3.3 Radionuclides and Radiations

It is a known fact that all radioactive nuclides decays in one, two or all three modes disintegration i.e. gamma-ray, beta or alpha particles. Some of the natural and the man made might decay by alpha followed by beta and gamma in order to reach stability. Even tough this project restrict it measurements on gamma-ray detection it has to be mentioned that some radionuclide are considered as either pure beta emitter or mixed mode transition as shown below:

- **Alpha particles (α) helium**

$$^{A}_{Z}\mathrm{X} \longrightarrow {}^{A-4}_{Z-2}\mathrm{Y} + {}^{4}_{2}\mathrm{He}$$

- **Beta particles (β-, β+) e- and $e^+$**

$$^{A}_{Z}X \rightarrow {}^{A}_{Z-1}Y + e^{+} + \nu_e \quad {}^{A}_{Z}X \rightarrow {}^{A}_{Z+1}Y + e^{-} + \bar{\nu}_e$$

- **Electronic Capture**

$$^{A}_{Z}X + e^{-} \rightarrow {}^{A}_{Z-1}Y + \nu_e$$

- **Gamma rays (γ ): Photons**

$^{137}Cs$, $^{210}Pb$, $^{241}Am$

## 3.3.1 Gamma-Matter Interaction:

Gamma-ray interaction processes is the main characteristic of it's principle measurements by semiconductor detectors namely germanium detectors. Gamma-ray main principle interaction mode are Photoelectric, Compton and Pair Production interaction which will be described in more details as follows:

**A- Photoelectric effect**

In this mode of interaction a photon is fully absorbed by an electron. The Momentum is conserved between the electron and its atom and a photo peak is produced which represent the energy and type of that particular incoming photon from the measured sample. It is considered the main identification of that radionuclide.

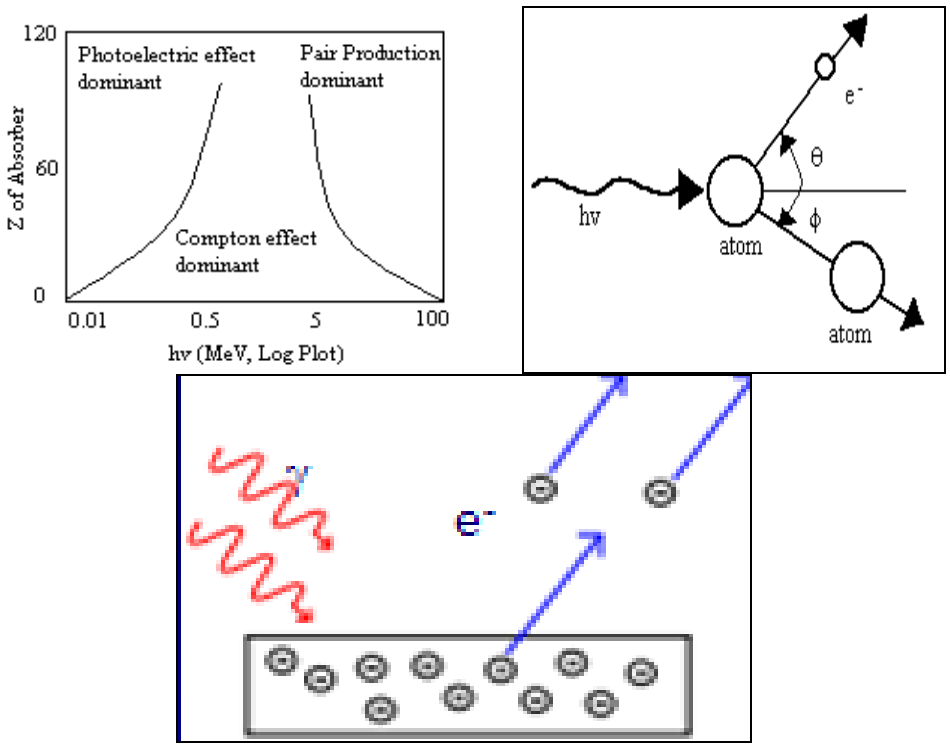



**Figure 3.7. Shows the main typical gamma-ray intraction of gamma-ray.**

The photoelectric effect is dominant in the low energy end of the spectrum**.**

$$E_e = E_\gamma - E_I$$

## B- Compton Interaction

The second mode of gamma-ray interation is the Compton Scattring which occurs in a higher energy than Photo electric. It is crucial to for this ineraction that both energy and momentum must be conserved. With this energy conservation, it is still possible for the electron to absorb all of the photon's energy. But with momentum conservation added, the amount of energy absorbed becomes a function of scattering angle. It has to be mentioned that when the interaction angle between the incoming photon and the detector surface of θ=180°, this energy is a maximum, which is written as $E_C$ $E_C(E) = E/(1+m_ec^2/2E)$ and shown in Figure 3.8

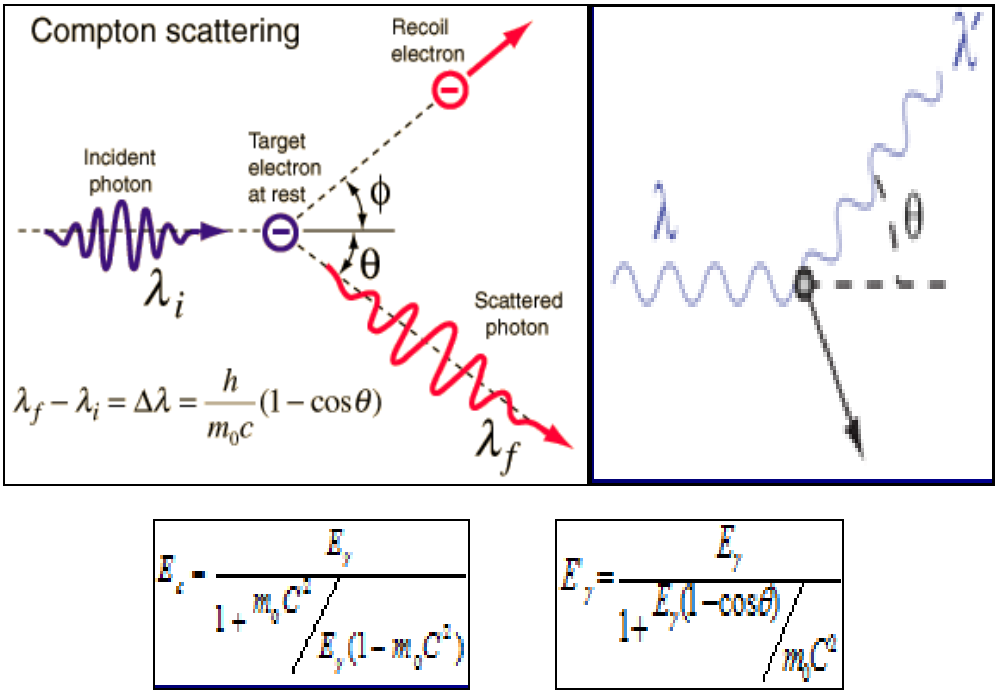
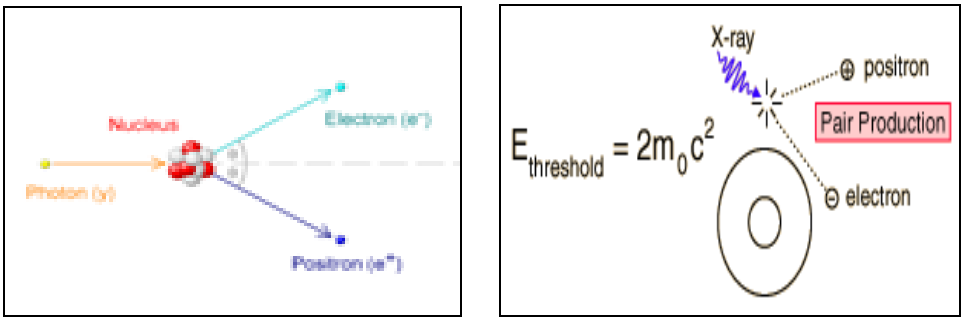



**Figure 3.8. Shows the Compton interaction process.**

### C - **Pair Production Interaction**

The third type of gamma-ray interaction is the Pair Production which only occurs when the incoming gamma photon energy exceeds 1.022 MeV because there is some frame of reference in which the relativistic theory exsist. Doppler shift reduces its frequency to bellow the frequency corresponding to the photon energy in order to creat the pair of the postron and the elecrtron. The positron that is created will only annihilate with an electron when its velocity has been reduced to nearly the velocity of the electron. Upon the annihilation procss, two gamma rays are emitted, each with energy of 511 keV, and both in the plane created by the trajectories of the positron and electron.




$$E_{e^-} + E_{e^+} = E_\gamma - 2m_0C^2$$

$2m_0C^2 = 1.02$ MeV

**Figure 3.9. Shows the pair production interaction proses.**

### 3.3.2 Gamma-ray attenuation

During the various gamma ray interactions, the incoming energy of the gamma photon will be gradually reduced until a complete absorption will occur depending on specific parameters of the initial energy and the absorbing materials that the gamma photon interacting with. A specific mathematical equations has been introduced to characterize the attenuation coefficient as shown in the Figure 3.10 .

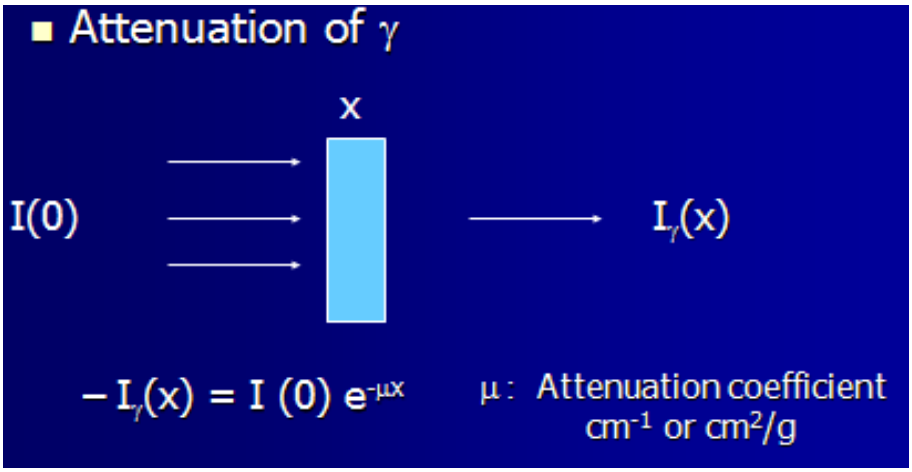



**Figure 3.10. Shows the attenuation process of gamma-ray photons.**

The total absorption process is

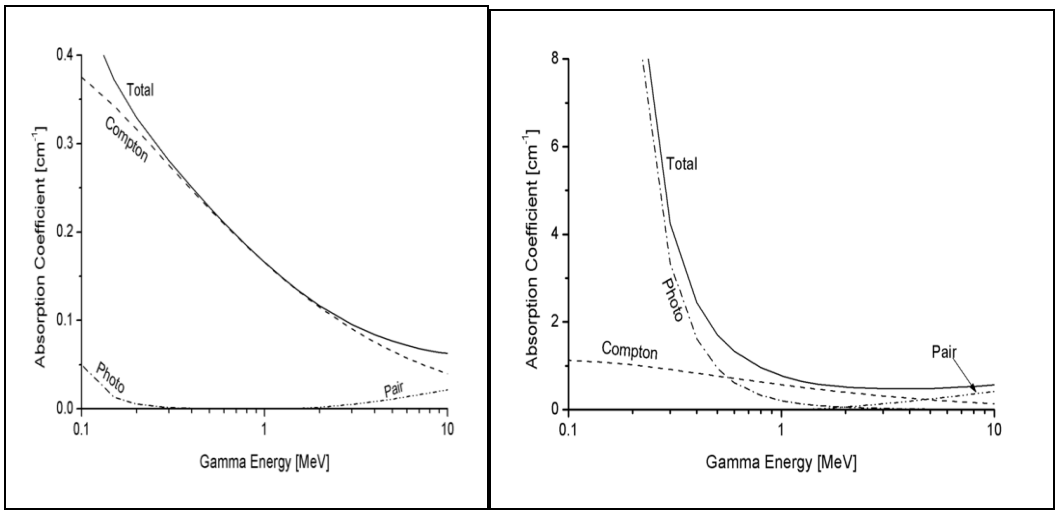



**Figure 3.11. Shows the gamma-ray main interaction modes and the total absorption process.**

## 3.4 Radioactive Equilibrium

One of the main aims of this research work is the determination and measurements of natural radioactive decay series, it is crucial to concentrate on the three main occurring radioactive series namely the uranium series headed by $^{235,238}$U and $^{232}$Th). The parent radionuclides as well as their daughter products are present in a significant amounts in earth and the plants that grows in it. The number of parent nuclei reduces with time as a result of radioactive decay to the daughter radionuclides. Concentrations of the nuclide after radioactive decay of an *n*-nuclide series that presented in linear chain may be found by applying the *Bateman* equations [14].

$$\frac{dN_1}{dt} = -\lambda_1 N_1 \qquad (3.1)$$

$$\frac{dN_i}{dt} = -\lambda_{i-1} N_{i-1} - \lambda_i N_i \quad (i = 2, n) \qquad (3.2)$$

where the stable final end product of the decay chain is *n.* The decay of the parent nuclei leads to increases in the number of atoms of the daughter nuclei but due to its own decay, this number also decreases with time. The decay constant of the initial daughter, granddaughter, great granddaughter decays are represented by $\lambda_1$ $\lambda_2$ etc, and $\lambda_i$ is the decay constant of $i^{th}$ nuclide.

$$dN_2 = \lambda_1 N_1 dt - \lambda_2 N_2 dt \qquad (3.3\ )$$

By integrating equation (3.4) and using the initial condition $N_2(0) = 0$ the following results are obtained

$$N_1(t) = N_0 e^{-\lambda t} \qquad (3.4\ )$$

$$N_2(t) = N_0 \frac{\lambda_1}{(\lambda_2 - \lambda_1)\left(e^{-\lambda_1 t} - e^{-\lambda_2 t}\right)} \qquad (3.5\ )$$

Then, the activity of daughter may be expressed as:

$$A_2(t) = \lambda_2 N_2(t) = N_0 \frac{\lambda_1 \lambda_2}{(\lambda_2 - \lambda_1)\left(e^{-\lambda_1 t} - e^{-\lambda_2 t}\right)} \qquad (3.6)$$

A radionuclide decay parent transform into other radionuclides that are known as daughters. These daughters radionuclides can also transform into grand-daughter nuclei. The activity of the daughter and grand-daughter grows gradually and simultaneously while the parent nuclides decay. After a significant period of time the daughter and grand-daughter activity may stop growing, and in turn start to decay. Moreover, after a significant period of time (t) called $t_{max}$ the daughter activity reaches its maximum value and equals to the activity of the parent [30].

The quantitative relationship of several members of series depends on the parent and daughter half-life. The activity in radioactive decay chains that is available in each stage may be estimated by understanding the "equilibrium conditions". Radioactive equilibrium can be classified into three particular cases as follows [34].

### 3.4.1 General relationship with non-equilibrium

In the case that the half-life of the parent is less than that of the daughter, there will not be equilibrium. Figure 3.12. illustrates the decay activity reaches the maximum at a finite time and followed by gradual reduction while the parent decays [34]. The ratio of parent and daughter activities can be determined from Eq. (3.4) [30].

If

$$\frac{A_2}{A_1} = \frac{\lambda_2}{\lambda_2 - \lambda_1}\left(1 - e^{-(\lambda_2 - \lambda_1)t}\right) \quad (3.7)$$

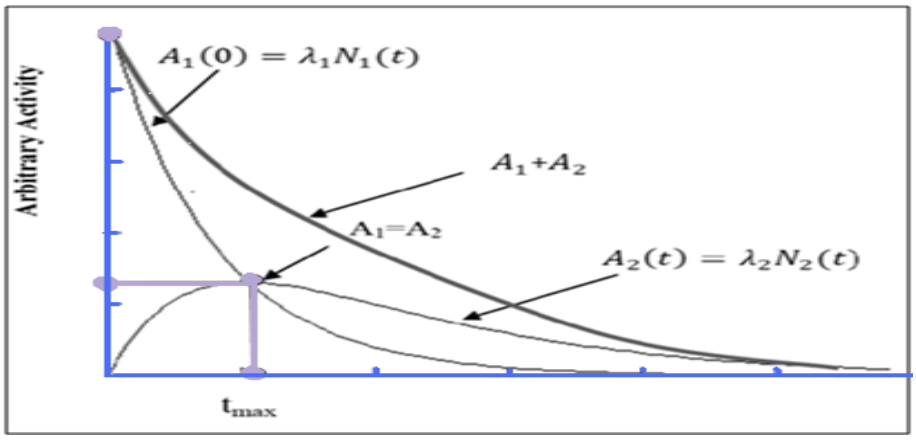



**Figure 3.12 . Shows the case of non-equilibrium condition.**

### 3.4.2 Transient Equilibrium.

In cases where the parent half-life is longer than that the daughter, the activity increases to a maximum after equilibrium [34]. The relationship

between the parent and daughter activities is estimated by Eq. 3.2 , if $\lambda_2 > \lambda_1$ [30]. Figure 3.13 shows transient equilibrium.

$$\frac{A_2}{A_1} = \frac{\lambda_2}{\lambda_2 - \lambda_1} \qquad (3.8)$$

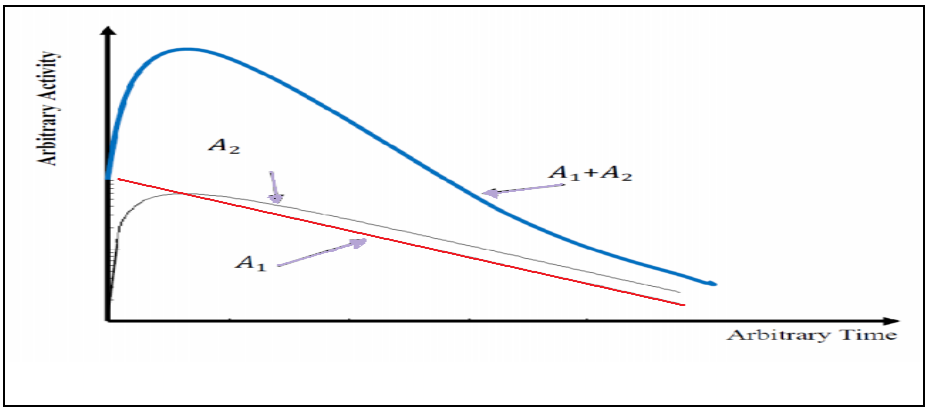



**Figure 3.13. Shows the condition of Transient equilibrium**

### 3.4.3 Secular Equilibrium

Secular equilibrium is a the case, where the initial parent radionuclides have a much longer half-life than their subsequent daughter decay, it is observed in the U and Th natural decay series. The parent radionuclides undergo a quite very slow rate of decay without a significant change in its activity during several half-lives of the production process of decay. However, its daughters grow-in and decay thereafter. The daughter nuclei are able to reach their parent activity in a closed system [38].

Consequently, it is an acceptable approximation to assume that the daughter activity is similar to the parent activity (assuming secular equilibrium occurs).

To calculate the activity in the given sample the following expression is commonly used, which depends on the number of different radionuclides species (n) that are involved in the decay series [30].

$$\lambda_n N_n = \lambda_{n-1} N_{n-1} = \ldots\ldots\ldots = \lambda_3 N_3 = \lambda_2 N_2 = \lambda_1 N_1 \qquad (3.9)$$

There are two conditions that are important to achieve this type of equilibrium:

1. The parent radionuclide must have a half-life much longer than it's progenies (e.g. $^{238}U$, $T_{1/2}$ =4.468 x$10^9$ y). [61].
2. To allow for in-growth of the decay products very long period of time must have significantly elapsed. [61].

For example, after 10,000 years $^{230}Th/^{226}Ra$ ratio could be 1 as illustrated in Figure (3.8) above, which is nearly six times the half-life of the $^{226}Ra$. However, fractionation may occur during geological processes, due to the U decay chains being composed of various elements. This is called a radioactive disequilibrium. Since Ra is more soluble than Th, the activity ratios of ($^{226}Ra/2^{30}Th$) are often >1 in water and < 1 in sediments and soils [43].

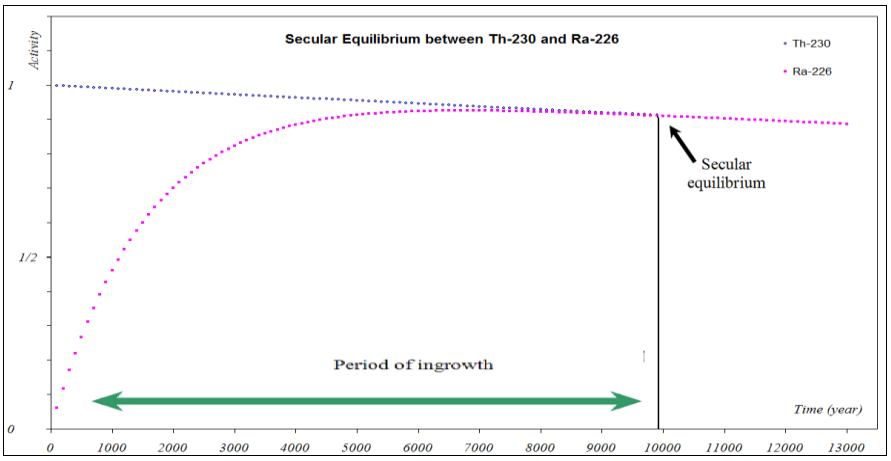



**Figure 3.14. Shows the Secular Equilibrium**

## 3.5 Practical bases for Gamma-ray spectrometry.

Gamma-ray spectroscopy has been used for decades and it has been operated by either scintillation and/or semiconductor detectors. It has been widely used to identify and measure the activity concentrations levels of the emitted radionuclides occurring from radioactive materials

decay by gamma-rays. A special dedicated electronic devices connected to a specific computer software will enable the system to operate as gamma-ray spectrometry detection system. The latest version of GammaVission computer software was used for the detailed identification and quantification at the Environmental General Authority "EGA" laboratory.

Two main functions that are needed in a workable gamma spectroscopy detector. Firstly, the detector must operate as a radiation converter medium that produces electrons at reasonable speed and the electrons must be able to travel easily through the detector. Secondly, the detector background noise must be as low as possible in order to collect the electrons that represent the detected gamma-ray photons energy emitted from the counted samples [60].

### 3.5.1 Detector Characteristic.

Using the available described P-type germanium detector with all the required accessory for the identification and quantification of the measured Cereal samples. Practically, the used computer software called GammaVission along with the detection system the main operating parameters of the detector were determined before any unknown samples to be measured. Therefore, a special standard sources are to be used in order to calibrate the detection system for energy and efficiency determination.[30]. The following subsections will describe in details the main detector components' and the two main detection parameters that characterizes the used HPGe-detector in more details.

### 3.5.2 Germanium Detector Setup

Due to the small band gap of about 0.7eV, germanium detectors are impossible to operate at room-temperature. This lack of functionality at

room-temperature stems from the large thermally-induced leakage current that results at this temperature. In order to get around this, the germanium detector is cooled to the point where this thermal leakage no longer spoils the excellent energy detections. This temperature happens to be 77 K and is achieved through the use of liquid nitrogen to cool the detector. The detector requires constant cooling from the liquid nitrogen in order to maintain the great energy resolution that it has. A special apparatus has been constructed to aid in that task. Appendix H has a sketch of such a device. The detector must be housed in a vacuum-tight cryostat to inhibit thermal conductivity between the detector crystal and the surrounding air. The cryostat is just capsule that houses the germanium crystal. A thin window is usually located near the crystal to minimize attenuation of gamma rays before they enter the germanium. As it is shown in Appendix H, the cryostat is mounted on a liquid nitrogen dewer. The dewer houses all of the liquid nitrogen and allows the germanium crystal to maintain its low temperature. The dewer must be much larger than the detector crystal itself in order to maintain liquid nitrogen for periods of time such that the dewer does not need constant filling.

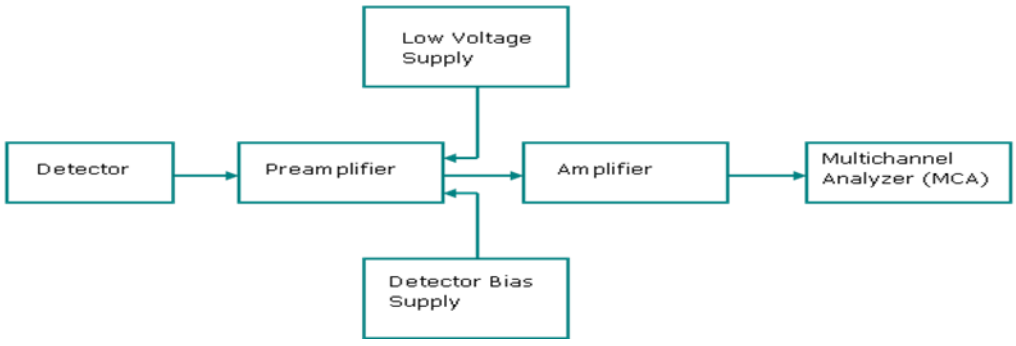



**Figure 3.15. Shows the detector setup.**

### 3.5.3 Multichannel analyzer

- A multichannel analyzer is a data acquisition instrument that counts pulses of voltage and stores them in channels corresponding to the maximum amplitude of the pulse.

- The counts are displayed as a histogram through an output to an oscilloscope or computer.

- It is important that the channel assignments are linearly related to the pulse heights so that the channel will be linear with deposited energy.

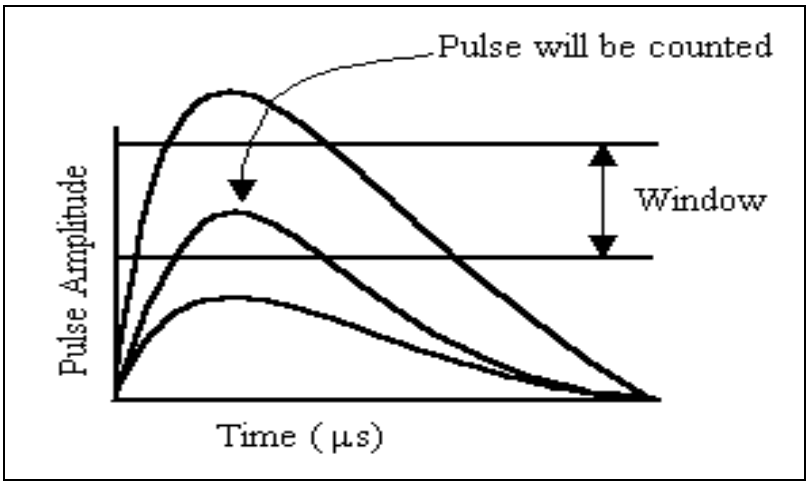



**Figure 3.16. Shows the pulse amplitude and useful counting window.**

### 3.5.4 Energy calibration.

The HPGe detector used for the measurements of the Grains and Cereal samples was calibrated for energy settings. The energy calibration evaluates the relationship between the spectrum position peak and the corresponding gamma-ray energy [61]. The energy calibration must cover the entire spectrum range of energy. Thus, the gamma-ray energies measurement are only applied to identify the spectra nuclides [61]. A specially made standard with known mixed radionuclide source was used to cary out this calibration process. The mixed radionuclide source as

described by it's source certificate contains the main know radionuclide including $Am^{241}$, $^{109}Cd$, $^{139}Ce$, $^{60}Co$, $^{57}Co$ , $^{137}Cs$ $^{203}Hg$, $^{113}Sn$, $^{85}Sr$, and $^{88}Y$ as shown in figure 3.17.

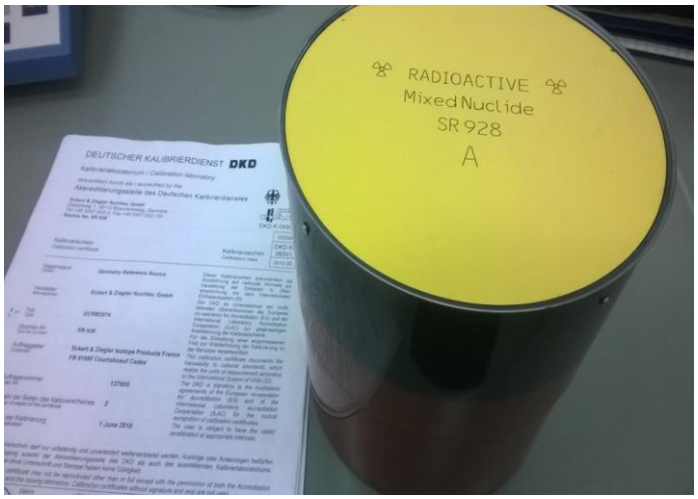



**Figure 3.17. Shows the Standard Mixed radionuclied with official Certificate.**

These radionuclides with known standard activity and energy lines and will defined activity manufactured and calibrated under secondary standard laboratory. This standard source have been selected for the calibration of energy due to the wide range of energies and a relatively long half-lives of the various radionuclides as shown in Table number 3.1.

**Table 3.1. shows the main standard radionuclides and energy calibration parameters**.

| Iso.Name | Channel | En. keV | Fit ( keV ) | FWHM( keV ) | Fit( keV ) | Delta |
|---|---|---|---|---|---|---|
| Cd - 109 | 445.34 | 88.03 | 88.00 | 0.91 | 0.92 | -0.87% |
| Ce - 139 | 838.64 | 165.85 | 165.83 | 0.99 | 0.99 | -0.49% |
| Sn – 113 | 1289.81 | 255.06 | 255.11 | 1.08 | 1.08 | -0.20% |
| Hg - 203 | 1411.46 | 279.20 | 279.18 | 1.12 | 1.10 | -1.93% |
| Sn - 113 | 1980.08 | 391.69 | 391.71 | 1.22 | 1.21 | -1.04% |
| Cs - 134 | 2846.94 | 563.25 | 563.26 | 1.38 | 1.37 | -0.65% |
| Cs - 134 | 2877.75 | 569.33 | 569.36 | 1.33 | 1.37 | -3.17% |
| Cs - 134 | 3056.51 | 604.72 | 604.73 | 1.41 | 1.41 | -0.28% |
| Cs - 137 | 3344.18 | 661.66 | 661.66 | 1.47 | 1.46 | -0.48% |
| Cs - 134 | 4022.26 | 795.86 | 795.86 | 1.61 | 1.58 | -1.75% |
| Cs - 134 | 4053.01 | 801.95 | 801.94 | 1.55 | 1.58 | -1.91% |
| Mn - 54 | 4219.22 | 834.85 | 834.84 | 1.63 | 1.61 | -1.19% |
| Y - 88 | 4538.54 | 898.04 | 898.03 | 1.65 | 1.67 | -1.25% |
| Cs - 134 | 6898.76 | 1365.19 | 1365.15 | 2.07 | 2.06 | -0.37% |
| Y - 88 | 9278.11 | 1836.06 | 1836.08 | 2.42 | 2.43 | -0.08% |

The calibration of primary energy were conducted to detect the full use of the low to the maximum energy from the source record spectrum with an activity and specification of various included radionuclides in the source certificate present in the APPINDEX B. The collected spectrum using the standard sources using the range of energies 59.54 up to 81 keV, 122 keV, 661.62 keV, 1173.32 keV and 1332.52 keV 1836keV, were used for the calibration of the primary energy scale of the GammaVission software program as shown in Figure 3.5.

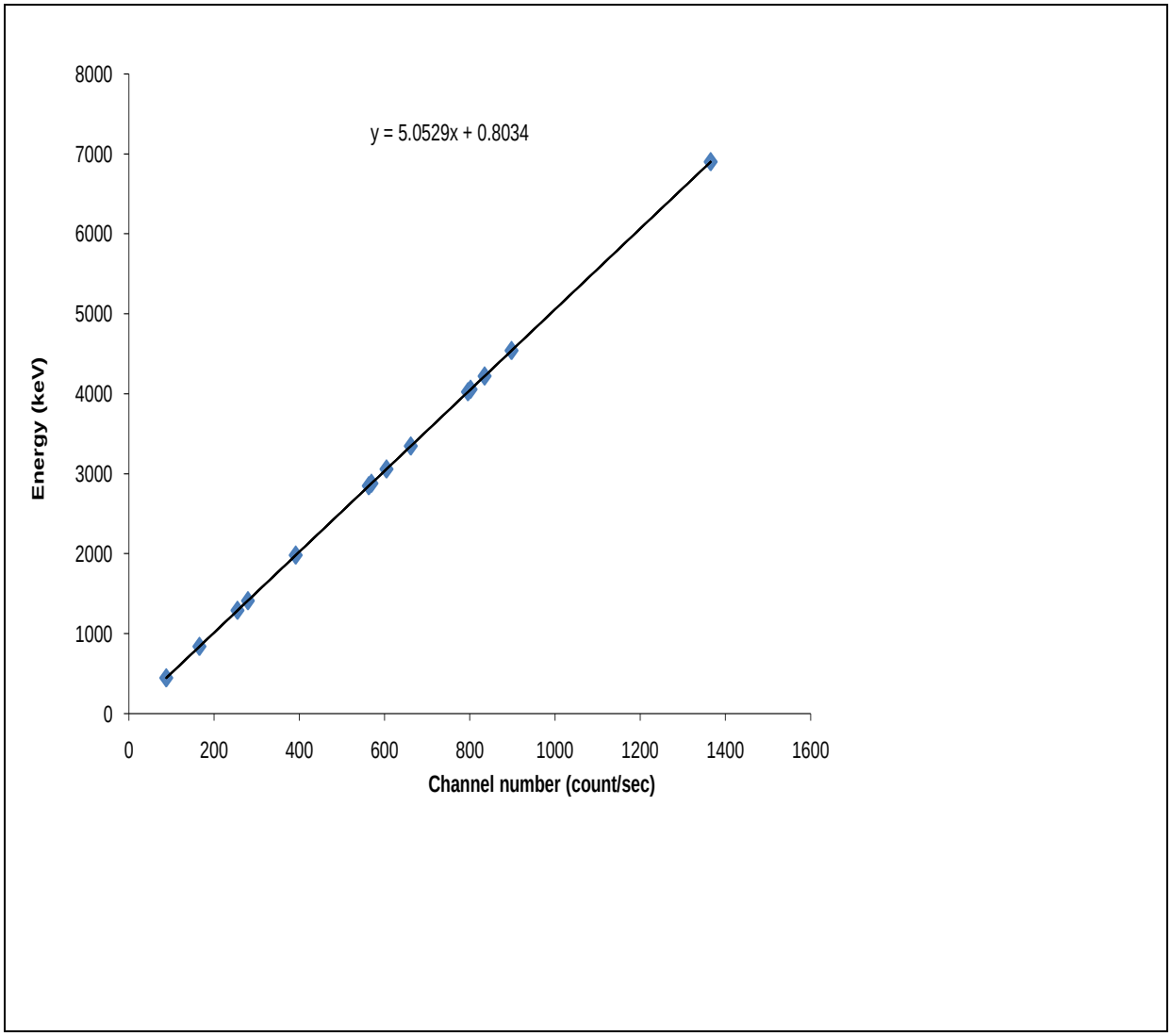



**Figure 3.18. Shows the collected energy calibrated spectrum using the mixed radionuclide source.**

## 3.5.5 Efficiency Calibration:

The absolute total efficiency of the HPGe detector is needed to calculate the activity concentration for the samples of the Cereals and the grains. The following equation illustrates the relationship between the decay rate and the absolute total efficiency. The absolute efficiency peak of the photon is the ratio between the numbers of counts in the full energy peak in the spectrum to the number of radiations incident on the detector.

$$\varepsilon_t = \frac{C_t}{D_s I_\gamma}$$

where $D_s$ is the number of disintegrations per second of the radioisotope which is the activity in Bq. $I_\gamma$ Is the fractional number of gammas emitted per disintegration.

The photo peaks from $^{226}$Ra, $^{232}$Th, and NG3 (a mixed radionuclide source containing $^{241}$Am, $^{109}$Cd, $^{139}$Ce, $^{137}$Cs, $^{60}$Co, $^{57}$Co, $^{203}$Hg, $^{113}$Sn, $^{85}$Sr, and $^{88}$Y) sources have been used to derive the efficiency calibration values. The geometries of sources was identical to the samples and it was assumed that the differences in the determination of the attenuation of gamma rays in the sources and the samples were minimal. Spectra were obtained on the calibration for 50,000 seconds. The obtained efficiency results from the calibration curves were gathered and used to calculate the actual measured activities from the Cereals and grain samples and are presented in the Table number 3.2.

TCC Polynomial coefficients:-1.203188 -2.498739 -0.656869 6.052196 0.000000 0.000000

Uncertainty = 13.8706 %

**Table 3.2. Shows the Efficiency values for each energy**

| Iso. Name | En. keV | Efficiency | Fit | Delta |
|---|---|---|---|---|
| Cd – 109 | 88.04 | 1.5063E-001 | 1.2518E-001 | -1.03 % |
| Ce – 139 | 165.85 | 2.3532E-001 | 2.4184E-001 | -2.77 % |
| Sn – 113 | 255.04 | 2.3662E-001 | 2.0189E-001 | -14.68 % |
| Hg – 203 | 279.17 | 1.8689E-001 | 1.9102E-001 | -2.21 % |
| Sn – 113 | 391.71 | 1.5404E-001 | 1.5304E-001 | -0.65 % |
| Cs – 134 | 475.35 | 7.6865E-002 | 1.3435E-001 | -74.78 % |

| | | | | |
|---|---|---|---|---|
| Cs – 134 | 563.26 | 5.9555E-002 | 1.1990E-001 | -101.33 % |
| Cs – 134 | 569.29 | 6.0626E-002 | 1.1905E-001 | -96.37 % |
| Cs – 134 | 604.66 | 7.6160E-002 | 1.1437E-001 | -50.18 % |
| Cs – 137 | 661.62 | 1.0861E-001 | 1.0775E-001 | -0.79 % |
| Cs – 134 | 795.76 | 6.3563E-002 | 9.5447E-002 | -50.16 % |
| Cs – 134 | 801.84 | 5.1629E-002 | 9.4973E-002 | -83.95 % |
| Mn – 54 | 834.81 | 9.2125E-002 | 9.2502E-002 | -0.41 % |
| Y – 88 | 898.02 | 6.6835E-002 | 8.8190E-002 | -31.95 % |
| Cs – 134 | 1167.86 | 9.4919E-002 | 7.4174E-002 | -21.86 % |
| Cs – 134 | 1365.13 | 1.1490E-001 | 6.6754E-002 | -41.90 % |
| Y – 88 | 1836.01 | 3.9533E-002 | 5.4054E-002 | -36.73 % |
| Y – 88 | 2734.03 | 6.5364E-002 | 3.9252E-002 | -93.99 % |

**Table 3.3. Shows the Calibration Certificate**

| Iso. Name | E. keV | Pct | Half life | Activity | GPS | Error |
|---|---|---|---|---|---|---|
| Cd – 109 | 88.03 | 3.61 | 4.63 E+002 | 13750.00 | 496.38 | 5.00% |
| Ce – 139 | 165.85 | 79.89 | 1.38 E+002 | 462.34 | 369.34 | 5.00% |
| Cs – 134 | 475.36 | 1.49 | 7.54 E+002 | 653.39 | 9.71 | 5.00% |
| Cs – 134 | 563.25 | 8.35 | 7.54 E+002 | 653.39 | 54.56 | 5.00% |
| Cs – 134 | 569.33 | 15.38 | 7.54 E+002 | 653.39 | 100.49 | 5.00% |
| Cs – 134 | 604.72 | 97.62 | 7.54 E+002 | 653.39 | 637.84 | 5.00% |
| Cs – 134 | 795.86 | 85.53 | 7.54 E+002 | 653.39 | 558.84 | 5.00% |
| Cs – 134 | 801.95 | 8.69 | 7.54 E+002 | 653.39 | 56.78 | 5.00% |
| Cs – 134 | 1167.97 | 1.79 | 7.54 E+002 | 653.39 | 11.69 | 5.00% |
| Cs – 137 | 661.66 | 85.10 | 1.10 E+004 | 404.50 | 344.23 | 5.00% |
| Cs – 134 | 1365.19 | 3.01 | 7.54 E+002 | 653.39 | 19.69 | 5.00% |

| | | | | | | |
|---|---|---|---|---|---|---|
| Hg – 203 | 279.20 | 81.46 | 4.66 E+001 | 936.66 | 763.00 | 5.00% |
| Mn – 54 | 834.85 | 99.98 | 3.12 E+002 | 531.79 | 531.66 | 5.00% |
| Sn – 113 | 391.69 | 64.00 | 1.15 E+002 | 828.07 | 529.96 | 5.00% |
| Sn – 113 | 255.06 | 1.82 | 1.15 E+002 | 828.07 | 15.07 | 5.00% |
| Y-88 | 898.04 | 93.70 | 1.07 E+002 | 1486.90 | 1393.23 | 5.00% |
| Y-88 | 1836.06 | 99.20 | 1.07 E+002 | 1486.90 | 1475.00 | 5.00% |
| Y-88 | 2734.00 | 0.71 | 1.07 E+002 | 1486.90 | 10.56 | 5.00% |

## 3.6 Sample Preparation

The samples were prepared for the natural radioactivity measurement. Each dried sample weighing between (600g - 1Kg) was sealed in polyethylene plastic Marinelli beakers, the plastic Marinelli beakers were sealed with a PVC tape, labeled according to its number and kept at room temperature (25 C) for at least 22 days before counting in to allow reaching the secular equilibrium of $^{232}$Th and $^{238}$U with their respective decay products, in which the activities of all radionuclide within each series are nearly equal. Each sample was given a serial number as shown in Table 3.4.

**Table 3.4 List of the collected samples and their codes**

| Sample | Name | Country | Sample | Name | Country |
|---|---|---|---|---|---|
| 1 | Red lentil | Turkey | 13 | Wheat | Gamata |
| 2 | Corn | Argentine | 14 | Barley | Gamata |
| 3 | Peas | Egypt | 15 | Corn | Gamata |
| 4 | Chickpea | India | 16 | Telda Rice | India |
| 5 | Beans | Egypt | 17 | Italian long grain Rice | Italy |
| 6 | Ring | Egypt | 18 | Aalosra Rice | Egypt |
| 7 | Bean Mdch | Egypt | 19 | Uncle Ben's Rice | England |
| 8 | Green lentil | Egypt | 20 | U.S.A Two girls Rice | U.S.A |
| 9 | Barley | Emsilata | 21 | Daawat Basmati Rice | India |
| 10 | Corn | Zliten | 22 | Zad alkher Rise | U.S.A |
| 11 | Barley | Zliten | 23 | El Nagib Rise | Egypt |
| 12 | Wheat | Argentine | 24 | Scotti round grain Rise | Italy |

Prior to the measurement, each sample underwent a pre-treatment that consisted of powdering. This step was crucial for achieving a homogeneous state for the sample. Because cereal comes dried, the samples did not undergo any drying process. Each sample was then placed in a cylindrical container with dimensions 14 cm in diameter and 13 cm in length. After being sealed, the sample filled into containers and were left for a period of at least 3 weeks to reach secular equilibrium between parent radionuclides and their daughters.

The samples were weighted and transferred to 550 ml labeled Marinelli breaks as illustrated in Figure 3.18. In order to reach radioactive secular equilibrium the samples were stored and sealed for at least one month.



**Figure 3.19. Shows the Marinelli samples beakers**

## 3.6.1 Samples analysis.

By applying the spectroscopic information for the identification of nuclei in the Cereals and grain samples of radionuclides, one can determine the individual component activities in these samples. In each sample the radionuclides could be identified by determining Centroid peaks of energies, compared with previously known gamma-ray energies from the published references.Samples were placed directly on the front face of the detector then kept for the same time as the acquisition period used to determine the spectral efficiency 50,000 seconds. The spectrum was calibrated to be associated with gamma rays ranging in energy from 40 keV up to 2.800 MeV. The background count to be used for background subtraction from the samples counts. These were analyzed independently to obtain more statistically significant overall results.

# Chapter Four

# Experimental Results and Discussion

# 4. Preface

In order to make use of the available scientific features of the Gamma-Vision software, the measured values for the main peaks of standard source were introduced to the energy and efficiency calibration wizard. This feature characterizes the main detection principles which outlines the spectrum resolution and efficiency. The resolution which is defined by the Full With Half Maximum ( FWHM), and the relative efficiency of the detector which is defined by detector manufacturer " Ortec " and quoted to be 80% . Figure 3.1. shows the actual measured FWHM values and the graph depicting the detector resolution characteristics using the standard mixed radionuclides. At the same Figure 4.1 presents the efficiency working graph for the same detector using the main peaks of the mixed radionuclides. Using these two operating characteristic of the used detector namely; the resolution and the efficiency the prepared samples were all measured each for 50,000 seconds.

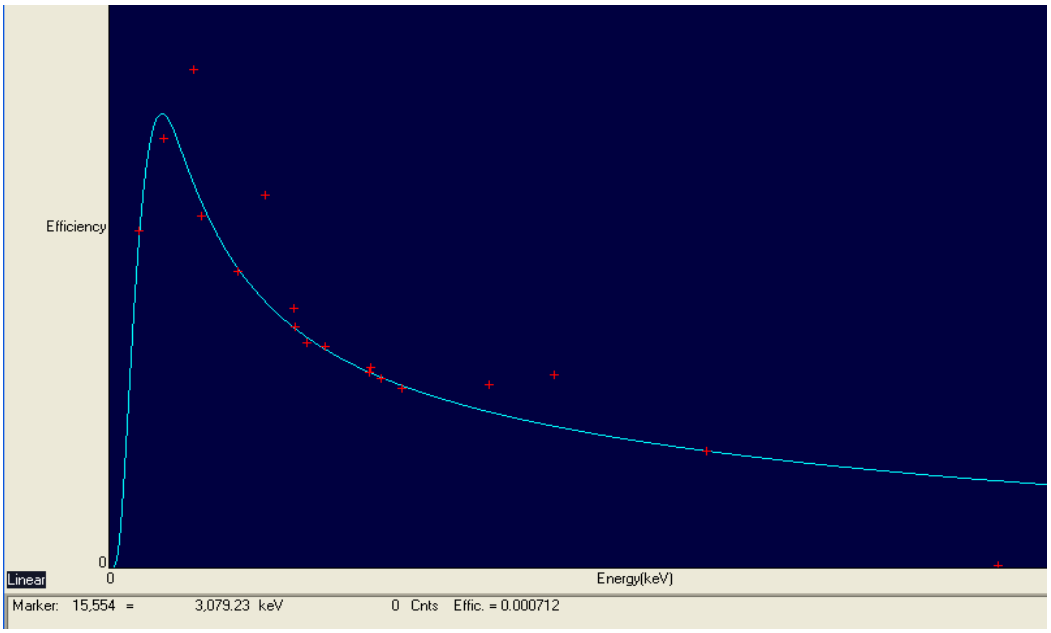



**Figure 4.1. Presents the efficiency values along with the main energies.**

## 4.1 Natural radioactivity measurement

For each Grain or Cereal Type, in total of 24 samples were prepared and sealed as mentioned in the previous Chapter for the natural radioactivity measurement. The measurement was conducted for 15 hours using the specified HPGe – Ortec type counting system. The system was calibrated with the standard mixed radionuclides calibration sources using the main known energy and emission rates of the peaks of $^{57}$Co, $^{241}$Am, $^{203}$Hg $^{133}$Ba,, $^{137}$Cs, $^{60}$Co and $^{88}$Y, which covers the full energy spectrum range from 59.54 keV from $^{241}$Am up to 1880keV from $^{88}$Y . The counting efficiency was determined previously for the same counting geometry. The measured peaks of the natural radionuclides were identified according to their individual photo peaks, which are 609.31keV ($^{214}$Bi) and 351.92keV($^{214}$Pb) for $^{238}$U , 238.63keV ($^{212}$Pb)and 583.19keV, ($^{208}$Tl) for $^{232}$Th. And 1461keV for $^{40}$K. Since the samples were sealed for more than 21 days, the activity of $^{226}$Ra during the secular equilibrium was assumed to be the same as its parent $^{238}$U. All collected spectra of all the samples were analyzed for their activities and representative spectra are presented in Figures No 2,3,4.

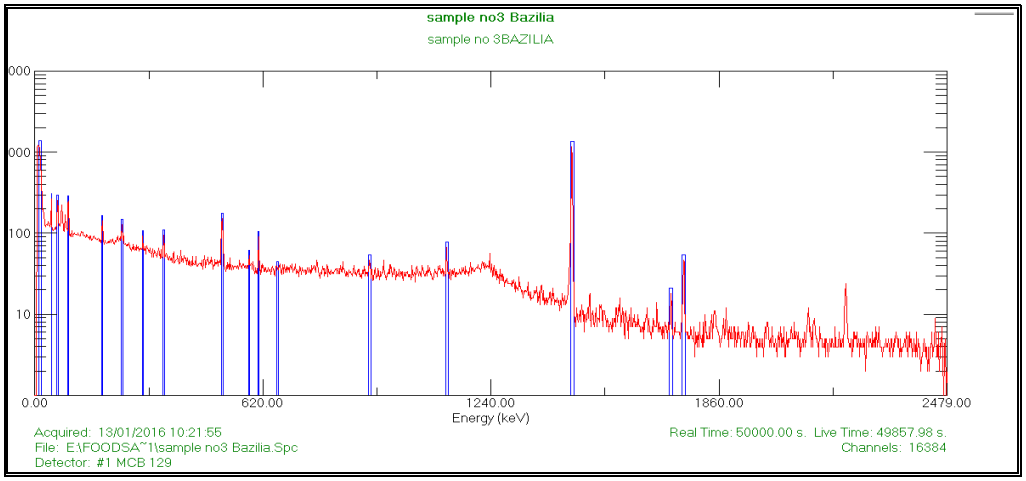



**Figure 4.1 Shows the collected spectrum without detail of photo peak identification of Peas-Egypt**

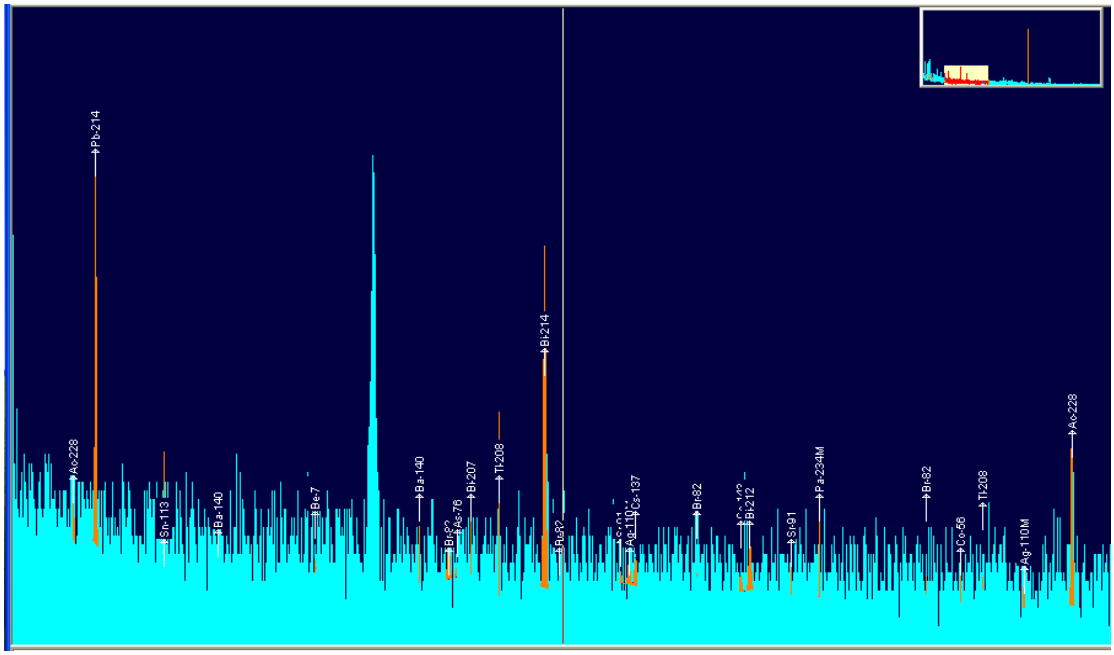



**Figure 4.2 Shows the collected spectrum with detail of photo peak identification of Wheat -Argentine**

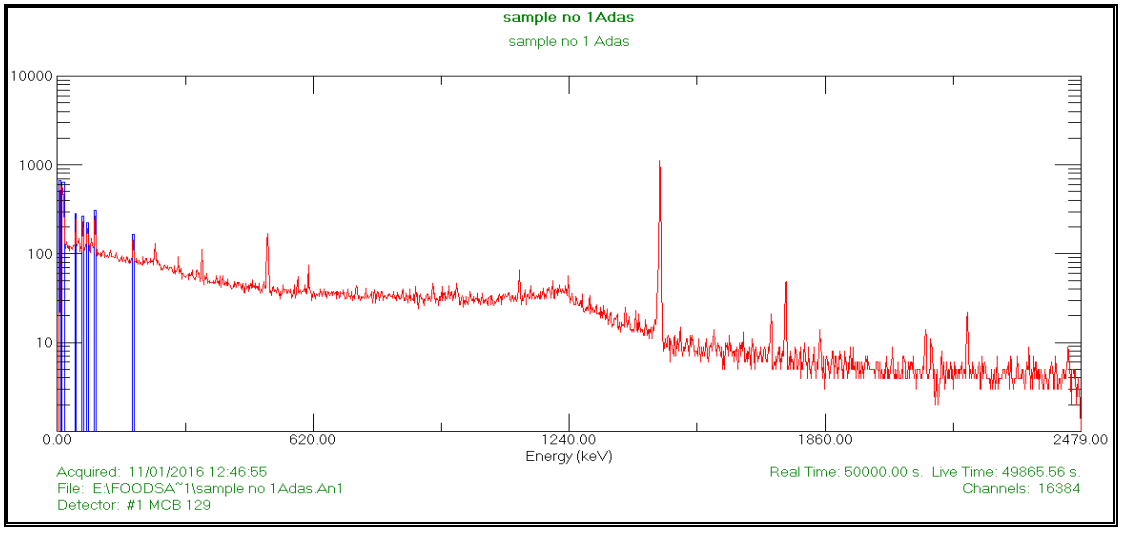



**Figure 4.3 Shows the collected spectrum of Red lentil -Turkey**

As it can be seen from the above three collected spectra for the measured samples all the main energy photo peaks are clearly delineated from which the net sample activity was calculated. From the collected of all measured

samples the Average activity of the main energy peaks representing $^{238}U$, $^{232}Th$, were calculated using Average Activity= Bq.kg-1 using two main energies of Uranium =(Pb214+Bi214) and for Thorium Th = (Pb 212+Tl 208). Also for $^{137}Cs$ and $^{40}K$ were gathered in the Tables No. 4.1,4.2,4.3 and 4.4 .

**Table 4.1 Shows the calculated Average activities for Uranium daughter nuclides in Bq.kg-1**

| samples | Ave. activity | Code Peak MDA |
|---|---|---|
| 1 | 2.66 ±0.39 | 0.69 |
| 2 | 3.02 ±0.45 | 0.46 |
| 3 | 2.17±0.33 | 0.24 |
| 4 | 3.23 ±0.48 | 0.21 |
| 5 | 4.23±0.63 | 0.27 |
| 6 | 4.39±0.66 | 0.34 |
| 7 | 3.69±0.55 | 1.02 |
| 8 | 2.06 ±0.31 | 0.18 |
| 9 | 4.14±0.62 | 0.17 |
| 10 | 2.38 ±0.36 | 0.17 |
| 11 | 9.95 ±2.91 | 0.20 |
| 12 | 4.02±0.60 | 0.26 |
| 13 | 2.60±0.39 | 0.25 |
| 14 | 6.15±0.92 | 0.27 |
| 15 | 3.29±0.49 | 0.17 |
| 16 | 2.96±0.44 | 0.16 |
| 17 | 3.21±0.48 | 0.17 |

| | | |
|---|---|---|
| 18 | 3.11±0.47 | 0.18 |
| 19 | 2.44±0.37 | 0.18 |
| 20 | 2.42±0.36 | 0.20 |
| 21 | 2.40±0.36 | 0.19 |
| 22 | 2.32±0.45 | 0.16 |
| 23 | 2.45 ±0.37 | 0.16 |
| 24 | 2.76 ±0.41 | 0.18 |

From the above Table 4.1, it can be seen most of calculated average activities are within the standard deviation of each other. However, the lowest calculated mean Average activities of 2.06±0.31Bq.kg$^{-1}$ was measured from sample No. 8 which was green lentil from Egypt. However the highest was 4.39± 0.66Bq.kg$^{-1}$ from sample No. 6 which was Ring also from Egypt. The mean Average activity of all the sample was calculated to be 3.83±0.57 Bq.kg$^{-1}$ . The Minimum Detectable Activity (MDA)is also shown for each nuclide, which is known as the lowest detection level of the detector for that particular radionuclide.

**Table 4.2 Shows the calculated Average activities for Thorium daughter nuclides**

| samples | Ave activity | Code Peak MDA |
|---|---|---|
| 1 | 0.97±0.15 | 0.09 |
| 2 | 1.01±0.15 | 0.08 |
| 3 | 1.03±0.16 | 0.10 |
| 4 | 0.86±0.13 | 0.09 |
| 5 | 1.13±0.17 | 0.13 |
| 6 | 1.29±0.19 | 0.17 |
| 7 | 0.95±0.14 | 0.09 |
| 8 | 1.25±0.19 | 0.10 |
| 9 | 1.01±0.15 | 0.09 |
| 10 | 0.96±0.14 | 0.08 |
| 11 | 1.03±0.15 | 0.12 |
| 12 | 1.20±0.18 | 0.14 |
| 13 | 1.30±0.19 | 0.13 |
| 14 | 1.25±0.19 | 0.15 |
| 15 | 1.04±0.15 | 0.08 |
| 16 | 0.90±0.13 | 0.08 |
| 17 | 0.89±0.13 | 0.09 |

| | | |
|---|---|---|
| 18 | 1.20±0.18 | 0.09 |
| 19 | 0.82±0.12 | 0.08 |
| 20 | 1.01±0.15 | 0.10 |
| 21 | 0.94±0.14 | 0.08 |
| 22 | 0.64±0.09 | 0.08 |
| 23 | 0.67±0.10 | 0.08 |
| 24 | 0.76±0.11 | 0.08 |

From the above Table 4.2, it can be seen that most of calculated Average activities are within the standard deviation of each other. However, the lowest calculated mean Average activities of 0.64±0.09 $Bq.g^{-1}$ was measured from sample No. 22 which was Zad Alkher rise from U.S.A. And the highest was 1.31± 0.19 $Bq.kg^{-1}$ from sample No. 13 which was Wheat from Gamata Libya. The mean Average activity of all the sample was calculated to be 1± 0.15 $Bq.kg^{-1}$ .

**Table 4.3 Shows the calculated Average activities for Potassium-40 nuclides in Bq.Kg-1**.

| **samples** | **Ave. activity** | **Code Peak MDA** |
|---|---|---|
| 1 | 272±40.8 | 0.59 |
| 2 | 102±15.3 | 0.38 |
| 3 | 283±42.5 | 0.64 |
| 4 | 283±42.5 | 0.65 |
| 5 | 370±55.5 | 0.65 |

| | | |
|---|---|---|
| 6 | 267±40.05 | 0.87 |
| 7 | 308±46.2 | 0.57 |
| 8 | 288±43.2 | 0.54 |
| 9 | 85.7±12. 9 | 1.35 |
| 10 | 108±16.2 | 0.41 |
| 11 | 170±25.5 | 0.57 |
| 12 | 126±18.9 | 0.79 |
| 13 | 135±20.3 | 0.71 |
| 14 | 132±19.8 | 0.67 |
| 15 | 108±16.2 | 0.53 |
| 16 | 71.1±10. 7 | 0.39 |
| 17 | 85.6±12.84 | 0.43 |
| 18 | 69.7±10. 5 | 0.42 |
| 19 | 75.8±11. 4 | 0.44 |
| 20 | 84.7±12. 7 | 0.49 |
| 21 | 75±11.3 | 0.43 |
| 22 | 71.1±10.7 | 0.37 |
| 23 | 72.9±10. 9 | 0.39 |
| 24 | 67.2±10. 08 | 0.39 |

From the above Table 4.3 for Potasium-40, it can be seen that most of calculated Average activities are within the standard deviation of each other. However, the lowest calculated Average activities of 67±10.05 $Bq.Kg^{1}$ was measured from sample No. 24 which was Scotti round grain

ricse from Italy. And the highest was 370± 55.5 Bq.kg$^{-1}$ from sample No.5 ,which was Beans from Egypt. The mean Average activity of all the sample was calculated to be 142.8 ± 21.4 Bq.kg-1 .

**Table 4.4 Shows the calculated Average activities for Cesium nuclides in Bq.kg$^{1}$.**

| samples | Ave. activity | Code Peak MDA |
|---|---|---|
| 2 | 0.12±0.018 | 0.04 |
| 3 | 0.10±0.015 | 0.04 |
| 4 | 0.11±0.017 | 0.05 |
| 5 | 0.09±0.01 | 0.07 |
| 6 | 0.15±0.02 | 0.07 |
| 7 | 0.13±0.02 | 0.05 |
| 8 | 0.07±0.01 | 0.05 |
| 9 | 0.09±0.01 | 0.04 |
| 10 | 0.11±0.02 | 0.04 |
| 11 | 0.08±0.012 | 0.06 |
| 12 | 0.11±0.017 | 0.06 |
| 13 | 0.17±0.03 | 0.05 |
| 14 | 0.11±0.017 | 0.07 |
| 15 | 0.04±0.006 | 0.03 |

| | | |
|---|---|---|
| 16 | 0.07±0.01 | 0.03 |
| 17 | 0.07±0.01 | 0.04 |
| 18 | 0.17±0.03 | 0.05 |
| 22 | 0.10±0.015 | 0.04 |
| 23 | 0.10±0.015 | 0.04 |

From the above Table 4.4 for the detection of 137Cs was detected in most of the sample except for samples No. 1, 19, 20, 21 and 24 and the detected levels were very low and within the standard deviation of each other. However, the lowest calculated average activity of 0.04±0.006 Bq.kg-1 detected in sample No 15 which was Corn from Gamat area in Libya. And the highest detected level was 0.17±0.026 Bq.kg-1 from sample No.13 which was which was Wheat from Gamata and sample No.18 which was Alosra rice from Italy. The mean Average activity of all the sample was calculated to be 0.12±0.018 Bq.kg-1.

**Table 4.5 Shows the Americium-241 detected activities from the detector contamination in Bq.Kg$^{-1}$**

| samples | Ave activity | Code Peak MDA |
|---|---|---|
| 1 | 0.33 | 0.12 |
| 2 | 0.3 | 0.12 |
| 3 | 0.0 | 0.10 |
| 4 | 0.67 | 0.15 |
| 5 | 0.52 | 0.22 |
| 6 | 1.06 | 0.20 |
| 7 | 0.37 | 0.14 |

| | | |
|---|---|---|
| 8 | 0.19 | 0.14 |
| 9 | 0.91 | 0.17 |
| 10 | 0.29 | 0.13 |
| 11 | 0.15 | 0.14 |
| 12 | 0.38 | 0.20 |
| 13 | 0.33 | 0.18 |
| 14 | 0.20 | 0.17 |
| 15 | 0.0 | 0.10 |
| 16 | 0.50 | 0.13 |
| 17 | 0.33 | 0.11 |
| 18 | 0.0 | 0.11 |
| 19 | 0.78 | 0.14 |
| 20 | 0.0 | 0.08 |
| 21 | 0.24 | 0.09 |
| 22 | 0.35 | 0.12 |
| 23 | 0.0 | 0.045 |
| 24 | 0.28 | 0.13 |

These detected levels is expected due to the wide spread of radioactive fallouts due to the Chernobyl reactor accident during the year 1986. Low levels of radioactivity especially Ceasium-137, was detected during the first two weeks after the accident in various parts of the costal cities of Libya particularly Tripoli.

## 4.2 Radium equivalent index ($Ra_{eq}$ )

The radium equivalent activity ($\mathbf{Ra_{eq}}$ ) concept allows a single index or number to describe the gamma output from different mixtures of $^{238}U$ (i.e., $^{226}Ra$) $^{232}Th$, and $^{40}K$ in a Material [ 38]. $\mathbf{Ra_{eq}}$ for each sample in Bq/kg is calculated using the following formula proposed by UNSCEAR [7] in $Bq.kg^{-1}$.

$$\mathbf{Req} = \mathbf{A}Ra + 1.43ATh + 0.077AK \quad (1)$$

Where $\mathbf{A_{Ra}}$ ,$\mathbf{A_{Th}}$ and $\mathbf{A_K}$ in $Bq.kg^{-1}$ are the activity concentration of $^{226}Ra$, $^{232}Th$, and $^{40}K$, respectively.

To reduce statistical counting error, each sample was counted for a period of 50,000 sec . An empty container was also counted under the same conditions to determine the background counts. For spectrum analysis, Gamma Vision software (Ortec, USA) was used, where the photo peaks for the sample analysis considered were 609.31keV, 351.92 keV ($^{226}Ra$), 238.63 keV, 583. 19 keV($^{232}Th$), 661.62( $^{37}Cs$) and 1461keV ($^{40}K$).

## 4.3 Hazard index

The Hazard index, is used to estimate the level of $\gamma$ -radiation hazard associated with the natural radionuclides in Grains and Cereal samples. The hazard index (HI) is calculated from the equation (2).

$$\mathbf{HI} = \frac{\mathbf{ARa}}{\mathbf{370}} + \frac{\mathbf{ATh}}{\mathbf{259}} + \frac{\mathbf{AK}}{\mathbf{4810}} \leq \mathbf{1} \qquad (2)$$

## 4.4 Activity concentration of $^{226}Ra$, $^{232}Th$ and $^{40}K$

Table 4.6 presents the activity concentrations for $^{226}Ra$, $^{232}Th$ and $^{40}K$ in the different Cereal and Grain samples consumed in **LIBYA**,

The radium equivalent activity (**$Ra_{eq}$**) and hazard index **HI.** The activity concentration of $^{238}U$, $^{232}Th$ and $^{40}K$ in $Bq.kg^{-1}$ present in the samples are given in Table 4.6, 4.7,4.8 respectively. The concentration of the parent nuclides of the natural radioactive series for each sample had been calculated in the unit of $Bq.kg^{-1}$

**Table 4.6 The Radium Equivalent activity ($Ra_{eq}$) and hazard index HI from Rice**

| No | Sample | $Ra_{eq}$ | HI Bq |
|---|---|---|---|
| 16 | TeldaRice | 8.26± 0.595 | (22.23±e( 1.6-3 |
| 17 | Italian long grain Rice | 9.32±0.620 | (25±e( 4.27-3 |
| 18 | Aalosra Rice | 8.30±0.713 | (22.3±e( 1.92-3 |
| 19 | Uncle Ben's Rice | 8.60±0.603 | (23.1±e( 1.62-3 |
| 20 | U.S.A Two girls Rice | 8.43±0.772 | (19.18±e( 2.07-3 |
| 21 | Daawat BasmatiRice | 7.54±0.678 | (20.29±e( 1.83-3 |
| 22 | Zadalkher Rise | 7.52±0.613 | (20.16±e( 1.65-3 |
| 23 | El Nagib Rise | 8.28±0.616 | (22.2±e( 1.66-3 |
| 24 | Scotti round grain Rise | 6.917±0.628 | (18.51±e( 1.69-3 |

**Table 4.7. The Radium Equivalent activity ($Ra_{eq}$) and hazard index HI from samples as indicated in the Table.**

| No | Sample | $ARa_{eq}$ | HI Bq |
|---|---|---|---|
| 9 | Barley Emsilata | 15.76± 0.68 | (41.8±e( 1.83-3 |
| 10 | Corn Zliten | 10.37±0.62 | (27.7±e( 1.64-3 |
| 11 | Barley Zliten | 14.25±0.88 | (37.6±e( 2.32-3 |
| 12 | Wheat Argentine | 11.74±1.05 | (31.6±e( 2.84-3 |
| 13 | Wheat Gamata | 12.33±0.98 | (32.3±e( 2.62-3 |
| 14 | Barley Gamata | 15.04±1.66 | (40.2±e( 2.93-3 |
| 15 | Corn Gamata | 11.45±0.34 | (30.7±e( 1.78-3 |

**Table 4.8 The Radium Equivalent activity ($Ra_{eq}$) and hazard index HI from Wheat and Barley.**

| No | Sample | AReq | HI Bq |
|---|---|---|---|
| 1 | Red lentil | 22.78± 0.65 | (1.57±e( 1.75-3 |
| 2 | Corn | 10.21±0.618 | (1.27±e( 1.67-3 |
| 3 | Peas | 23.42±0.73 | (1.63±e( 1.99-3 |
| 4 | Chickpea | 23.66±0.76 | (7.63±e( 2.07-3 |
| 5 | Beans | 29.54±1.09 | (7.79±e( 2.96-3 |

| | | | |
|---|---|---|---|
| 6 | Ring | 23.27±1.44 | (4.62±e( 3.84-3 |
| 7 | Bean Mdch | 26.10±0.77 | (69.5±e( 2.10-3 |
| 8 | Green lentil | 23.96±0.73 | (63.7±e( 1.99-3 |

As it has been seen from the above Tables 4.6-4.8, the activities from Radium Equivalent and the calculations of the respective Hazards Index in Bquerel units for all the measured samples which constitute a representative agricultural food products that consumed by Libyan people. From the presented data for all the samples, the Corn had shown the least activity of 2.07±0.53 $Bq.Kg^{-1}$ and the highest was from Rings from Egypt with 4.39±0.62 $Bq.kg^{-1}$ the rest of the samples have shown a rang of 2- 5 $Bq.Kg^{-1}$ While the radiation Hazards Index for all the samples were below the value of unity one which is the acceptable level according to the equation No.2

**Table 4.9 The $^{137}Cs$ activity ($Ra_{eq}$) and hazard index HI from various samples**

| No | Sample | $^{137}Cs$ Bq. at 661.66 KeV |
|---|---|---|
| 1 | Red lentil | BDL |
| 2 | Corn | BDL |
| 3 | Peas | BDL |
| 4 | Chickpea | 0.01±0.08 |
| 5 | Beans | 0.11± 0.10 |

| | | |
|---|---|---|
| 6 | Ring | BDL |
| 7 | Bean Mdch | 0.09± 0.08 |
| 8 | Green lentil | 0.10± 0.08 |
| 9 | Barley Emsilata | 0.04± 0.07 |
| 10 | Corn Zliten | 0.06±0.07 |
| 11 | Barley Zliten | BDL |
| 12 | Wheat Argentine | BDL |
| 13 | Wheat Gamata | BDL |
| 14 | Barley Gamata | BDL |
| 15 | Corn Gamata | BDL |
| 16 | Telda Rice | BDL |
| 17 | Italian long grain Rice | 0.10±0.07 |
| 18 | Aalosra Rice | 0.01±0.08 |
| 19 | Uncle Ben's Rice | BDL |
| 20 | U.S.A Two girls Rice | BDL |
| 21 | Daawat Basmati Rice | BDL |
| 22 | Zad alkher Rise | BDL |
| 23 | El Nagib Rise | BDL |
| 24 | Scotti round grain Rise | BDL |

From Table 4.9 for $^{137}Cs$ detected levels were very low in a number of samples in the range of 0.01- 0.10Bq while in the other samples were below detection limit(BDL) of the detection system. It should be noted that sample Barley from Emsilata and Corn from Zlitan have shown some detectable levels.

# Chapter 5

# Discussion of Results and Conclusions

## 5.1 Presentation of Results discussion

All 24 samples were measured for 15 hours using the Gamma-Ray HPGe –detector counting system. The system was calibrated using $^{57}Co$, $^{212}Pb$, $^{133}Br$, $^{238}U$, $^{137}Cs$, $^{60}Co$ and $^{40}K$, for their known energy (which covers the energy range from 122keV to 1461keV) and peak width of gamma-ray emission. The counting efficiency was determined previously for all of its counting geometry. The radionuclides were identified according to their individual photo-peak, which are 609keV ($^{214}Bi$) and 351.9keV ($^{214}Pb$) for $^{238}U$ , 238keV ($^{212}Pb$), 583.191 and 510.80keV, ($^{208}Tl$) for $^{232}Th$. And 1460keV for $^{40}K$. The activity of $^{226}Ra$ during the equilibrium was assumed to be the same as its parent, $^{238}U$. The average specific activity for each radionuclide was calculated using equation proposed by [40] , [10]

## 5.2 Results and data discussion

Concentrations of natural radionuclides in the measured samples and the estimated annual effective dose due to ingestion of type of Cereals lines are presented in Table 5.1. Tables 4.1, 4.2 for $^{238}U$ data were in the range of 2.07±0.62 to 4.54±0.45 , and 0.64±0.05 to 1.30±0.06 for $^{234}Th$, with a mean values of 2.72±0.52 , and 1.75±0.43 $Bq.kg^{-1}$ for all Grains and Cereal samples respectively. While the range for $^{40}K$ was 67 to 371 in $Bq.kg^{-1}$, The upper calculated values of $^{238}U$ was found in sample No.11 which was Barley from Zletan area is somewhat questionable and has to be investigated further with other Barley samples and soil measurements from the same area. With the exception of the sample No.11 which gave the highest detected level

was 20Bq.kg$^{-1}$, while sample No.8 which was Green Lentil from Egypt gave the lowest detected level 2.07Bq.kg$^{-1}$ for $^{238}$U daughters .

These calculated average specific activities of Uranium daughter nuclides values means that the value of $^{226}$Ra is somewhat in accordance with the literature value in corn (grain) from Ghana [8], and the lower value is similar to the mean value obtained from phosphate region of Pernambuco located in northeastern Brazil [10]. The mean value of 2.72 Bq.kg$^{-1}$ shows a relatively lower result than the values obtained in corn flour in Brazil [53]; [55] in corn from a contaminated vegetable garden in Ontario, Canada [59], in the diet of Americans [33] and in Indian corn flour[21], The higher detected level of the Zletan area from Libya is probably due to a significantly higher activity of $^{226}$Ra similar to that reported for Nigerian maize grown in the tin mining area of Bitsichi, Jos [40] and in Turkish maize [11] than in this study. The low activity of $^{226}$Ra obtained from the other samples may be attributed to the poor migration of radium from soil to plant, or a low level of radium or uranium in the soil. Furthermore, there is no previous record of mining activities in or around the land used to grow this Barley, therefore soil samples from Barley grown areas should be investigated for uranium and thorium levels. $^{228}$Ra shows very low concentrations ranging from 0.64 to 1.31 Bq.kg$^{-1}$ with a mean of 1.75 Bq.kg$^{-1}$. This mean activity of $^{228}$Ra is higher than the references from Brazil [54]; [55], but lower than the reference from Ghana [8]. The low concentration level of $^{228}$Ra (an indicator for $^{232}$Th ) in this study may be due to a poor migration of radium from substrate to plant in the environment where the corn was grown[8] . The results obtained show that the specific activity of $^{238}$U, $^{232}$Th in all wheat flour samples appeared lower than the recommended limit of [69],[62].

This mean value of $^{226}$Ra is somewhat in accordance with the literature value in corn (grain) from Ghana [8], and the lower value is similar to the mean value obtained from phosphate region of Pernambuco located in northeastern Brazil [10]. The mean value of 2.72 Bq.kg$^{-1}$ shows a relatively lower result than the values obtained in corn flour in Brazil [53]; [55], in corn from a contaminated vegetable garden in Ontario, Canada [59], in the diet of Americans [33], and in Indian corn flour [21] There is a significantly higher activity of $^{226}$Ra reported for Nigerian maize grown in the tin mining area of Bitsichi, Jos [40], and in Turkish maize [11] than in this study. The low activity of $^{226}$Ra obtained in this study may be attributed to the poor migration of radium from soil to plant, or a low level of radium or uranium in the soil. Furthermore, there is no previous record of mining activities in or around the land used to grow this Barely. $^{228}$Ra shows very low concentrations ranging from 0.64 to 1.33 Bq kg$^{-1}$ with a mean of 0.79 Bq kg$^{-1}$. This mean activity of $^{228}$Ra is higher than the references from Brazil [54]; [55] but lower than the reference from Ghana [8]. The low concentration level of $^{228}$Ra (an indicator for $^{232}$Th ) in these sample may be due to a poor migration of radium from substrate to plant in the environment where the Cereals samples were grown [8].

The calculated specific activities for $^{40}$K was found to be in the range of 67±8 to 371±10Bq.kg$^{-1}$. In contrast with the results obtained from Brazil in which the $^{40}$K contents have actually been found to be between 30 and 54 Bq.kg$^{-1}$ in market corn flour [53]; [54]. The present value is higher than other reported studies when compared with the $^{40}$K activity in Ghanaian maize [9], and in Nigerian maize [57]. It has to be noted, that the primary source of naturally occurring radionuclides in food is the soil where it is planted; the potassium-40 content of the samples ranged from

41.84±5.88Bq.kg$^{-1}$ to 264.73±3.84Bq.kg$^{-1}$ with an average 133Bq.kg$^{-1}$, the relatively comparable concentration of $^{40}$K as obtained especially in number of sample and the lowest detected $^{40}$K was in sample number 24 from Scotti round grains from Italy. Apart from this, the relatively high activity of $^{40}$K detected in a number of the measured sample could also be traceable to the conditions under which the maize is grown, e.g. whether the maize has been treated with phosphate fertilizers. The application of fertilizers on farmlands generally increases the amount of potassium in the soil [65], its uptake and retention in Cereals food crops and hence, result in a relatively high percentage of $^{40}$K. On the other hand, the activity concentration obtained is considerably comparable with the reported activity of $^{40}$K 243.2Bq.kg$^{-1}$ in maize from Jos-Plateau, an area of high radioactivity due to mining activities [40], [41] in Turkish maize 491.62Bq.kg$^{-1}$; [11] and in Mexico 90Bq.kg$^{-1}$; [22].

The intake of $^{40}$K is the highest with an annual value of 431Bq, which does not cause any concern from the health physics point of view. This is because potassium is an essential element in the body and it is homeostatically controlled by the human metabolism, and as a result, the body content of $^{40}$K is largely determined by physiological characteristics and not intake. However, the annual intakes of $^{226}$Ra (30.2Bq) and $^{228}$Ra (6.16Bq) are very low compared to 213Bq for $^{226}$Ra and 313 Bq for $^{228}$Ra reported from Ghana [8] and well fitted within the UNSCEAR annual intake of 22Bq for $^{226}$Ra and 15Bq for $^{228}$RaBq [68].

Along with the natural radionuclides Man-made radioactivity was also measured especially $^{137}$Cs which is well known fission product and was spread during the nuclear bomb testing and various nuclear reactor accidents particularly Chernobyl in 1986. The detected levels were

low and below permissible standards. At the same time $^{241}Am$ was detected but as detector contamination and it has to be dealt with the manufacturer company.

## 5.3 Comparison of Results

As the 24 samples were gathered from the locally planted farm products and some were imported products from a number of foreign countries, it is suitable to make a compression between the natural and man made radioactivity content of both of them. As it can be seen from the Tables 4.1, 4.2, 4.3, most of the samples gave comparable results except for sample No.11 The measured samples have shown a considerable amount of the natural radionuclides and most of them were man made nuclides free such $^{137}Cs$ except for the two samples from Msalata and Zlitan that were found to contain detectable amounts of this fallout radionuclides.

Also a worldwide comparison of this project results with other published data on grains, corn and cereal samples as it can be seen from Table 5.1.

**Table 5.1 Comparison of mean activity concentrations of the radionuclides ($Bq.kg^{-1}$) and total effective dose (μSv )**

| Mean Concentration (Bq.kg−1) | | | Effective Dose (μSv y−1) | |
|---|---|---|---|---|
| $^{226}Ra$ | $^{228}Ra$ | $^{40}K$ | | Data Source |
| 2.72±0.52 | 1.75±0.43 | 10±133 | - | Present study |
| 3.88 ± 5.52 | 0.79 ± 0.93 | 52.2 ± 30.3 | 15.24 | [19] |
| 0.07 ± 0.03 | — | 30.0 ± 0.3 | — | [53] |
| — | — | 16.29 ± 3.20 | — | [57] |

| | | | | |
|---|---|---|---|---|
| 34.1 ± 14.2 | — | 243.2 ± 21.2 | 740a | [40] |
| 0.088±0.044 b | 0.474 ± 0.577 | 70.78 ± 33.7 b | — | [21] |
| — | — | 0.167 | — | [9] |
| — | — | 90 ± 30 | 58.1 | [22] |
| 25.82 ± 2.34 | — | 491.62 ± 52.61 | — | [11] |
| 0.07 | — | — | — | [59] |
| 0.0568 | — | — | — | [33] |
| 4.86 ± 2.20 | 8.43 ± 2.90 | 231.83 ± 15.23 | 370 | [8] |
| — | — | 243.2 ± 21 | 63.5 | [41] |
| 0.13 ± 0.009 | — | — | — | [10] |
| 0.016 (0.006–0.036) | 0.03(<0.007–0.065) | — | — | [55] |
| 0.2< | 0.3< | 54 ± 1 | — | [54] |

**a:** the effective dose is due to maize and other cereals.

**b:** activity concentrations are due to maize and other cereals.

### 5.4 Conclusion and Recommendations

Activity concentrations of natural radionuclides ($^{226}$Ra, $^{228}$Ra, $^{40}$K and $^{137}$Cs ) have been determined using HPGe γ-ray spectroscopy. Among the detected radionuclides, concentrations of $^{40}$K in all the samples were found in considerable amount, while $^{226}$Ra and $^{228}$Ra were present in nominal concentrations. However, the activities of all the radionuclides are somewhat insignificant compared to the recommended limits by international organizations. Also a number of samples have shown a detectable amounts of $^{137}$Cs radioactivity as shown in Table 4.9.

It can be concluded henceforth that even though consumption of these food products does not pose a significant risk, the obtained results may be helpful to address the health impacts of consumption `of these levels as well as will be the basis for setting Libyan legal limits for the Libyan population, as well as to monitor soil and environmental contamination. The following are number of recommendations need to be studied and may be applied.

- Libya imports most of the food and food products from various worldwide countries and some are locally produced. Therefore, the food consumed by the Libyan people should be measured for natural and Man-Made radioactivity and very important to be certain that food was radiation free or within an acceptable limits according to national and International standards.
- The samples of Cereals and Grains were gathered from local market particularly from Zlitan and Gamat areas and constitute a mixture good margin of the Libyan daily food intake.
- The range for average activity of the main daughter radionuclide calculated in this study was found to be 2.72±0.51 for $^{238}U$ and 1.75±0.41 for $^{232}Th$ in $Bq.kg^{-1}$. These levels are comparable with worldwide values and did not exceed the normal set values by other studies as well as the worldwide range18-93 $Bq.kg^{-1}$ as shown in Table 4.1 .
- The higher detected levels of $^{238}U$ in Gamata Barely and Zlitan is a matter need to be further investigated and it is a reason good for the need to make a thorough survey of the all areas of Libya.
- The measured radioactivity values for $^{40}K$ was found to be relatively higher than other studies but does not constitutes a medical health issue.

- Cesium-137 was found in almost of all the samples above the minimum detection limits which is normal because of the wide worldwide spread of $^{137}Cs$.
- The Radium equivalent activity was found to be in the range 10-30 $Bq.kg^{-1}$ with a mean value of 23 and Hazards Index was found to be less than one. Indicating that the worldwide mean value was not exceeded, confirming that the upper limit of radium equivalent activity did not reach 370 $Bq.kg^{-1}$ .
- The findings of this research project should be used as basis for other studies for similar food products or others to be used by regulatory authority when making the acceptable legal standard limits for food importation from other countries.
- It is recommended that other means of measurement such as Alpha spectroscopy or beta counting should be applied as these measurement criteria differs from gamma spectroscopy which only measures gamma-ray photons.

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# APPENDIXES

# APPINDEX A Detector specification Certificate

## QUALITY ASSURANCE DATA SHEET

**GMX Series HPGe (High-Purity Germanium) Coaxial Detector System**

**Model and Serial Numbers**

Detector Model No. GMX70P4-95
Cryostat Configuration CFG-PV4 — Ship Date 14-May-10
Dewar Model — Serial No. 50-TN42147A
Preamplifier Model A257N — When calling Customer Service, always
Preamplifier Serial No. 10036738 — reference the Detector Serial No.
H. V. Filter Model 138EMI — Sales Order No. 15002712
H. V. Filter Serial No. 10098161
SMART-1-N Serial No.

**Cryogenic Information**

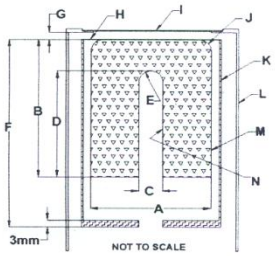
Dewar Capacity 0 — Static Holding Time
Detector Cool-Down Time

**High Voltage Bias**

Recommended Operating Bias — NEGATIVE 4900 Volts

**Performance Specifications***

| | Warranted | Measured | Amp Shape Time |
|---|---|---|---|
| Resolution (FWHM) at 1.33 MeV, $^{60}$Co | 2.3 keV | 2.3 keV | 6 µs |
| Peak-to-Compton Ratio, $^{60}$Co | 60:1 | 60:1 | 6 µs |
| Relative Efficiency at 1.33 MeV, $^{60}$Co | 70 % | 70 % | 6 µs |
| Peak Shape (FWTM/FWHM), $^{60}$Co | 2.0 | 2.0 | 6 µs |
| *Peak Shape (FWFM/FWHM), $^{60}$Co | 3.0 | 3.3 | 6 µs |
| *FWFM/FWHM is typical not warranted | | | |
| Resolution (FWHM) at 5.9 KeV, $^{55}$Fe | 1100 eV | 713 eV | 6 µs |




E: NOMINAL 5-mm RADIUS
F: 130-mm CUP LENGTH
G: 4-mm SPACE
H: 0.03-mm/0.03-mm Al/Mylar
I: 0.76-mm Be
J: NOMINAL 8-mm RADIUS
K: 0.8-mm Al
L: 1-mm Al
M: 0.3-micron Ge/B DEAD LAYER
N: 700-micron Ge/Li DEAD LAYER

A= 69 mm — Crystal Diameter
B= 74.9 mm — Crystal Length
C= 10 mm — Hole Diameter
D= 67.2 mm — Hole Depth

OTHER Capsule: NUCA # 9444
Cryo: PV4 # 13595

Data Certified By: Thomas Jennings — DATE 5-14-10

Form no. 0308BK vers. 1

# APPINDEX B Standard Source Certificate

DEUTSCHER KALIBRIERDIENST DKD

Kalibrierlaboratorium / *Calibration laboratory*

Akkreditiert durch die / *accredited by the*
Akkreditierungsstelle des Deutschen Kalibrierdienstes

**Eckert & Ziegler Nuclitec GmbH**
Gieselweg 1, 38110 Braunschweig, Germany
Tel +49 5307 932-0, Fax +49 5307 932-194
**Source No. SR 928**

DKD-K-06501

| 022588 |
|---|
| DKD-K-06501 |
| 2010-05 |

Kalibrierschein
*Calibration certificate*

Kalibrierzeichen
*Calibration mark*

| | |
|---|---|
| Gegenstand *Object* | **Geometry Reference Source** |
| Hersteller *Manufacturer* | **Eckert & Ziegler Nuclitec GmbH** |
| Typ *Type* | **QCRB2074** |
| Strahler-Nr. *Source number* | **SR 928** |
| Auftraggeber *Customer* | **Eckert & Ziegler Isotope Products France FR 91955 Courtaboeuf Cedex** |
| Auftragsnummer *Order No.* | **137805** |
| Anzahl der Seiten des Kalibrierscheines *Number of pages of the certificate* | **2** |
| Datum der Kalibrierung *Date of calibration* | **1 June 2010** |

Dieser Kalibrierschein dokumentiert die Rückführung auf nationale Normale zur Darstellung der Einheiten in Übereinstimmung mit dem Internationalen Einheitensystem (SI).
Der DKD ist Unterzeichner der multilateralen Übereinkommen der European co-operation for Accreditation (EA) und der International Laboratory Accreditation Cooperation (ILAC) zur gegenseitigen Anerkennung der Kalibrierscheine.
Für die Einhaltung einer angemessenen Frist zur Wiederholung der Kalibrierung ist der Benutzer verantwortlich.
*This calibration certificate documents the traceability to national standards, which realize the units of measurement according to the International System of Units (SI).*
*The DKD is signatory to the multilateral agreements of the European co-operation for Accreditation (EA) and of the International Laboratory Accreditation Cooperation (ILAC) for the mutual recognition of calibration certificates.*
*The user is obliged to have the object recalibrated at appropriate intervals.*

Dieser Kalibrierschein darf nur vollständig und unverändert weiterverbreitet werden. Auszüge oder Änderungen bedürfen der Genehmigung sowohl der Akkreditierungsstelle des DKD als auch des ausstellenden Kalibrierlaboratoriums. Kalibrierscheine ohne Unterschrift und Stempel haben keine Gültigkeit.
*This calibration certificate may not be reproduced other than in full except with the permission of both the Accreditation Body of the DKD and the issuing laboratory. Calibration certificates without signature and seal are not valid.*

| Stempel *Seal* | Datum *Date* | Leiter des Kalibrierlaboratoriums *Head of the calibration laboratory* | Stellvertreter *Deputy* | Bearbeiter *Person in charge* |
|---|---|---|---|---|
| Deutscher Kalibrierdienst DKD-K-06501 | 27 May 2010 | Dr. Thieme | Schott / Lehmacher | Lehmacher / Schohl / Schott / Schüler |

Eckert & Ziegler
Nuclitec

## Geometry Reference Source

| | |
|---|---|
| Source no. | SR 928 |
| Drawing | VZ-1685-001 |
| Volume | approximately 1000 ml |
| Density | approximately 1.0 g/cm³ |
| Construction | The radionuclidic mixture is homogeneously incorporated in the matrix of the source. |

| Nuclide | Gamma-ray energy [MeV] | Activity [Bq] | Emission rate [$s^{-1}$] |
|---|---|---|---|
| Americium-241 | 0.060 | 3.39E03 | 1.22E03 |
| Cadmium-109 | 0.088 | 1.69E04 | 6.14E02 |
| Cobalt-57 | 0.122 | 6.32E02 | 5.41E02 |
| Cerium-139 | 0.166 | 8.07E02 | 6.44E02 |
| Mercury-203 | 0.279 | 1.93E03 | 1.57E03 |
| Tin-113 | 0.392 | 2.94E03 | 1.91E03 |
| Strontium-85 | 0.514 | 3.79E03 | 3.73E03 |
| Caesium-137 | 0.662 | 2.67E03 | 2.27E03 |
| Yttrium-88 | 0.898 | 6.17E03 | 5.80E03 |
| Cobalt-60 | 1.173 | 3.15E03 | 3.15E03 |
| Cobalt-60 | 1.333 | 3.15E03 | 3.15E03 |
| Yttrium-88 | 1.836 | 6.17E03 | 6.13E03 |

| | |
|---|---|
| Reference date | 1 June 2010 at 12:00 UTC |
| Leakage and contamination test | Wipe test according to ISO 9978. |
| Wipe test passed on | 27 May 2010 |
| Measuring method | The activity was measured with a gamma spectrometer system consisting of a calibrated high purity germanium detector and a multi-channel analyser. |
| Traceability | Additional to the direct traceability to the PTB through the DKD this product complies with the requirements for traceability to NIST specified in the American National Standard "Traceability of Radioactive Sources to the NIST and Associated Instrument Quality Control (ANSI N42.22-1995)". As a requirement of the ANSI N42.22-1995 **Eckert & Ziegler Nuclitec GmbH** participates in the NEI/NIST Measurements Assurance Program of the Nuclear Power Industry. |
| Uncertainty | The relative uncertainty of the activity is 3 % (Cd-109: 5 %).<br>The reported uncertainty, determined according to the DKD-3 report is based on the standard uncertainty multiplied by a coverage factor of k = 2, providing a level of confidence of 95 %. (Ref. NIST Technical Note 1297/"Guide to the Expression of Uncertainty in Measurement" ISO Guide, 1995) |
| Radioactive impurities | At the time of calibration the following radioactive impurities were detected: Co-56<1 Bq; Se-75<1 Bq; Rb-84<1 Bq; Ag-110m<1 Bq; In-114m<1 Bq; Cs-134<4 Bq |
| Quality assurance system | The quality assurance system of **Eckert & Ziegler Nuclitec GmbH** was certified by Lloyd's Register Quality Assurance (LRQA) according to ISO 9001, issue 2000. Isotrak products meet the requirements of 10CFR50 Appendix B in the USA. |

# Explanations for Certificates (Page 2 of Certificates)

## Uncertainty

The reported uncertainty is based on standard uncertainty multiplied by a coverage factor k = 2, providing a level of confidence of approximately 95 %. (ISO Guide, 1995)

## Traceability

This certificate documents the traceability of measurement results to national standards, standard measuring equipment and methods for the realisation of physical units of measurement according to the International System of Units (SI).
Traceability is defined as 'the property of a result of a measurement whereby it can be related to appropriate standards, generally international or national standards, through an unbroken chain of comparisons'.

QSA Global GmbH has been accredited as DKD (Deutscher Kalibrierdienst) calibration laboratory by the Physikalisch-Technische Bundesanstalt (PTB) and is authorized to issue reference sources which are traceable to national standards held at the PTB in Germany.
Because of the European co-operation for Accreditation (EA) mutual recognition agreement the certificates are also accepted by all EA-members (e. g. UKAS).

This product complies with the requirements for traceability to NIST specified in the American National Standard "Traceability of Radioactive Sources to the NIST and Associated Instrument Quality Control (ANSI N42.22-1995)".
As a requirement for the ANSI N42.22-1995 QSA Global participates in the NEI/NIST Measurements Assurance Program of the Nuclear Power Industry.

## Leakage and contamination tests

Stringent tests for leakage are an essential feature of radioactive sources production. They are based on ISO 9978. Some standard methods used for testing radiation sources are listed below.

### Wipe test I

The source is wiped with a swab or tissue, moistened with ethanol or water, the activity removed is measured.
Limit: 200 Bq
(Limit USA: 5 nCi)

### Immersion test II

The source is immersed in a suitable liquid at 50 °C for at least 4 hours and the activity removed is measured.
Limit: 200 Bq
(Limit USA: 5 nCi)

### Bubble test III

The source is immersed in water or a suitable liquid and the pressure in the vessel reduced to 13 kPa (100 mm Hg). No bubbles must be observed.

## ISO classification

The International Organization for Standardization (ISO) has proposed a system of classification of sealed radioactive sources based on safety requirements for typical uses (see ISO 2919 and ANSI N43.6-1997). This system provides a manufacturer of sealed radioactive sources with a set of tests to evaluate the safety of his products. It also assists a user of such sealed sources to select types which suit the application he has in mind. The tests to which specimen sources are subjected are listed in the following table.

### Classification of sealed source performance standard according to ISO 2919 and ANSI N43.6-1997

| | Class 1 | 2 | 3 | 4 | 5 | 6 | X |
|---|---|---|---|---|---|---|---|
| Temperature | No test | - 40 °C (20 min) + 80 °C (1 h) | - 40 °C (20 min) + 180 °C (1 h) | - 40 °C (20 min) + 400 °C (1 h) and thermal shock 400 °C to 20 °C | - 40 °C (20 min) + 600 °C (1 h) and thermal shock 600 °C to 20 °C | - 40 °C (20 min) + 800 °C (1 h) and thermal shock 800 °C to 20 °C | Special test |
| External Pressure | No test | 25 kPa absolute | 25 kPa absolute to 2 MPa absolute | 25 kPa absolute to 7 MPa absolute | 25 kPa absolute to 70 MPa absolute | 25 kPa absolute to 170 MPa absolute | |
| Impact | No test | 50 g from 1 m | 200 g from 1 m | 2 kg from 1 m | 5 kg from 1 m | 20 kg from 1 m | |
| Vibration | No test | 3 x 10 min 25 - 500 Hz at 5 g peak amplitude | 3 x 10 min 25 - 50 Hz at 5 g peak amplitude and 50 - 90 Hz at 0.635 mm amplitude peak to peak and 90 - 500 Hz at 10 g peak amplitude | 3 x 30 min 25 - 80 Hz at 1.5 mm amplitude peak to peak and 80 - 2000 Hz at 20 g peak amplitude | | | |
| Puncture | No test | 1 g from 1 m | 10 g from 1 m | 50 g from 1 m | 300 g from 1 m | 1 kg from 1 m | |

## Special applications

No test programme can cover all possible combinations of environments to which a source may be exposed.
Users should therefore consult our experts before using sources in potentially adverse environments.

## IAEA Special Form

'Special Form' is a test specification for sealed sources given in the IAEA transport regulations (IAEA TS-R-1).
It is used in determining the maximum acceptable activities for various types of transport containers.
If nothing else is stated, the reference date is identical with the date of manufacture.

## Quality assurance System

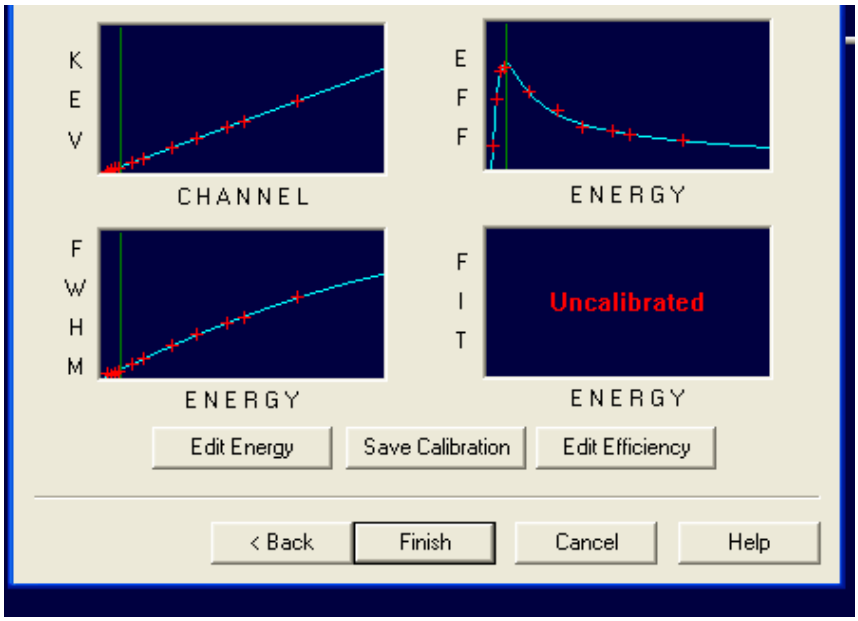
The quality assurance system of QSA Global GmbH was certified by Lloyd's Register Quality Assurance (LRQA) according to ISO 9001:2000 and according to ISO 13485:2003 for medical devices. Isotrak products meet the requirements of 10CFR50 Appendix B.

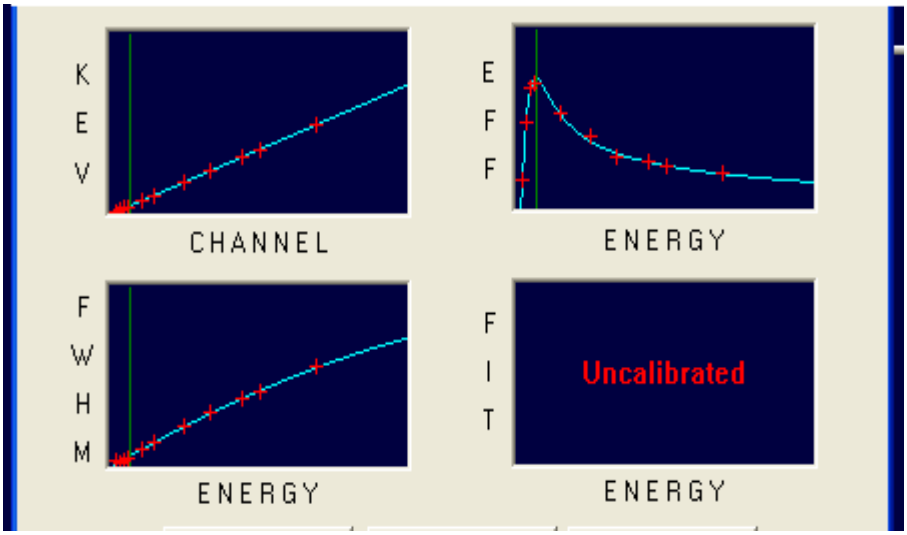
## NRC Advice

Radioactive Material - not for human use - introduction into foods, beverages, cosmetics, drugs, or medicinals, or into products manufactured for commercial distribution is prohibited - exempt quantities should not be combined

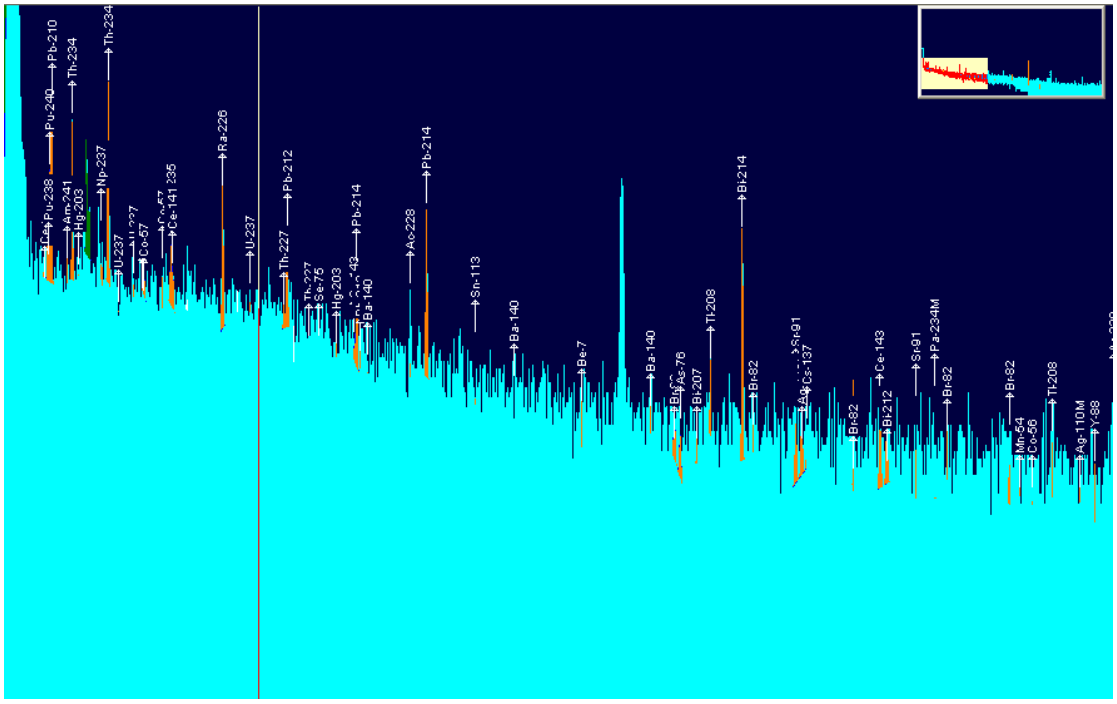
RE1, Issue 3, 2007-06-08

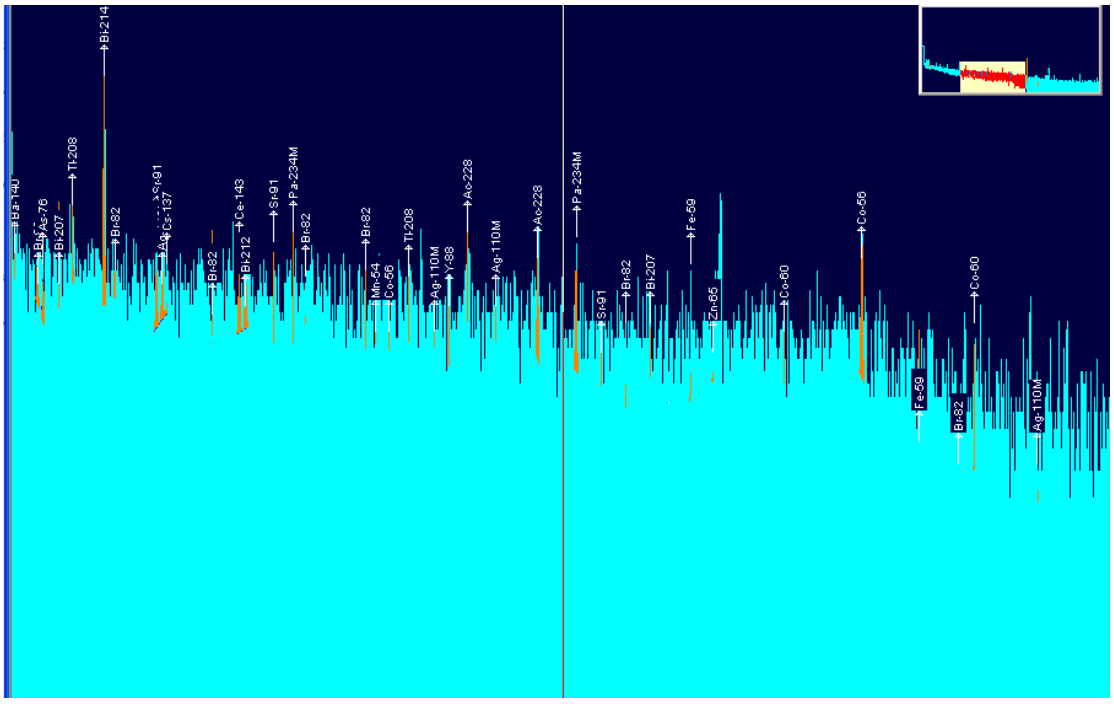
# APPINDEX C Total Channel, Energy and Efficiency calibration software wizard

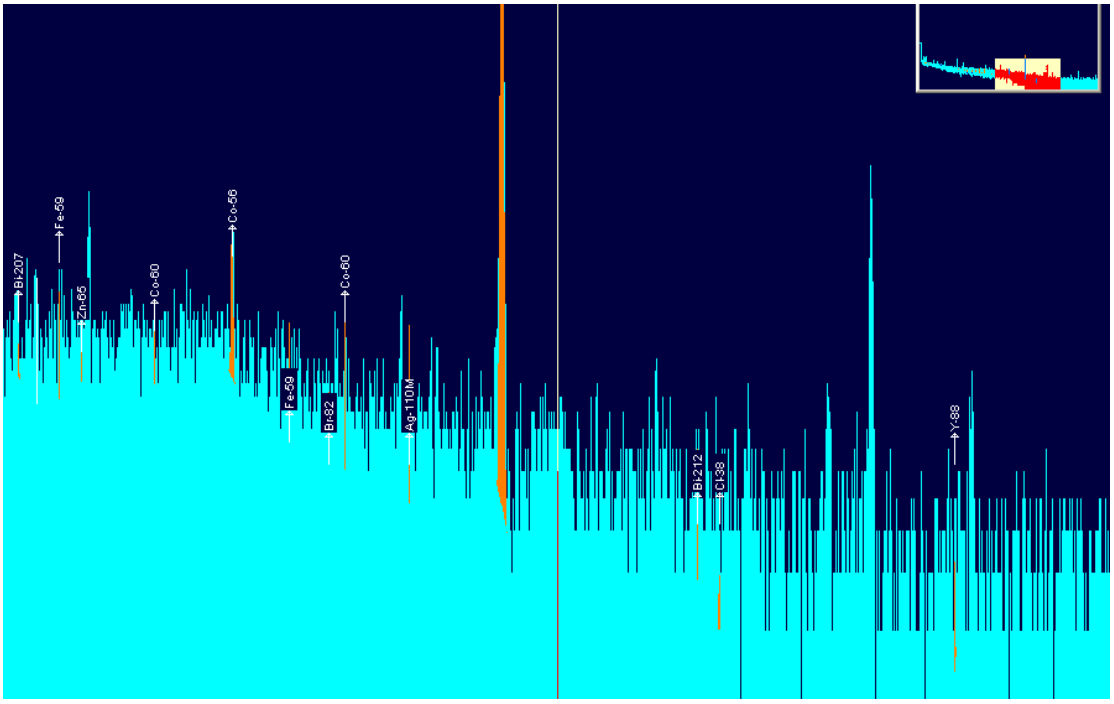


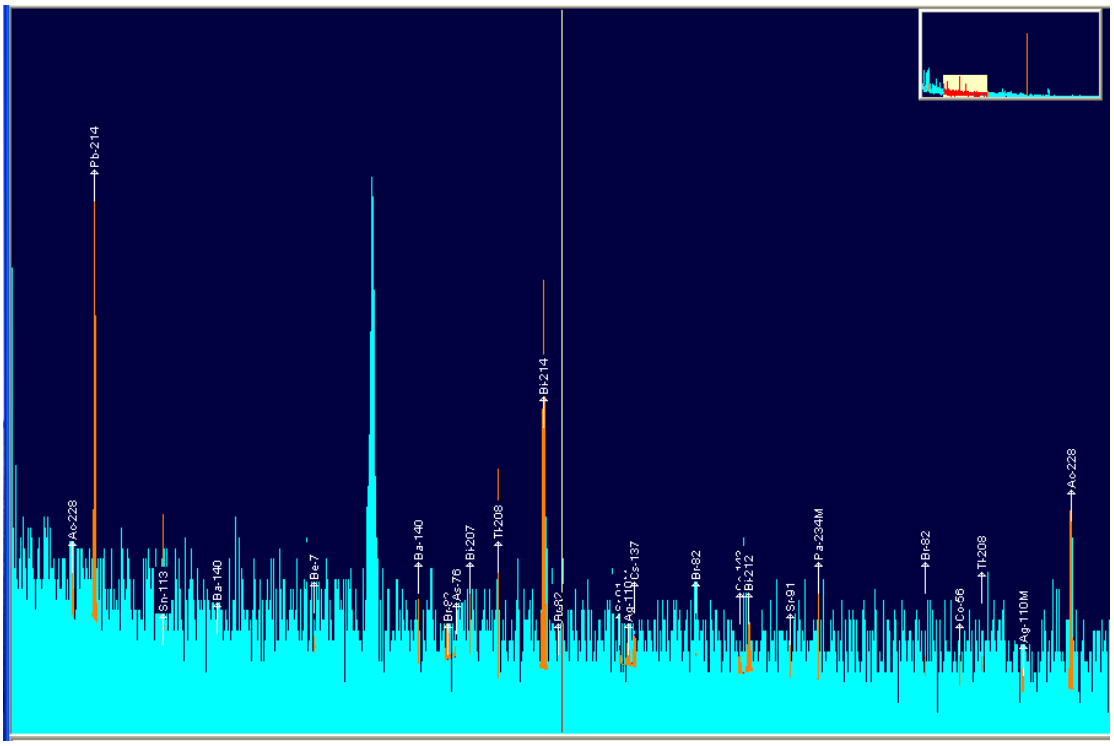
# APPINDEX D  SPECTRA OF SAMPLE N0 24

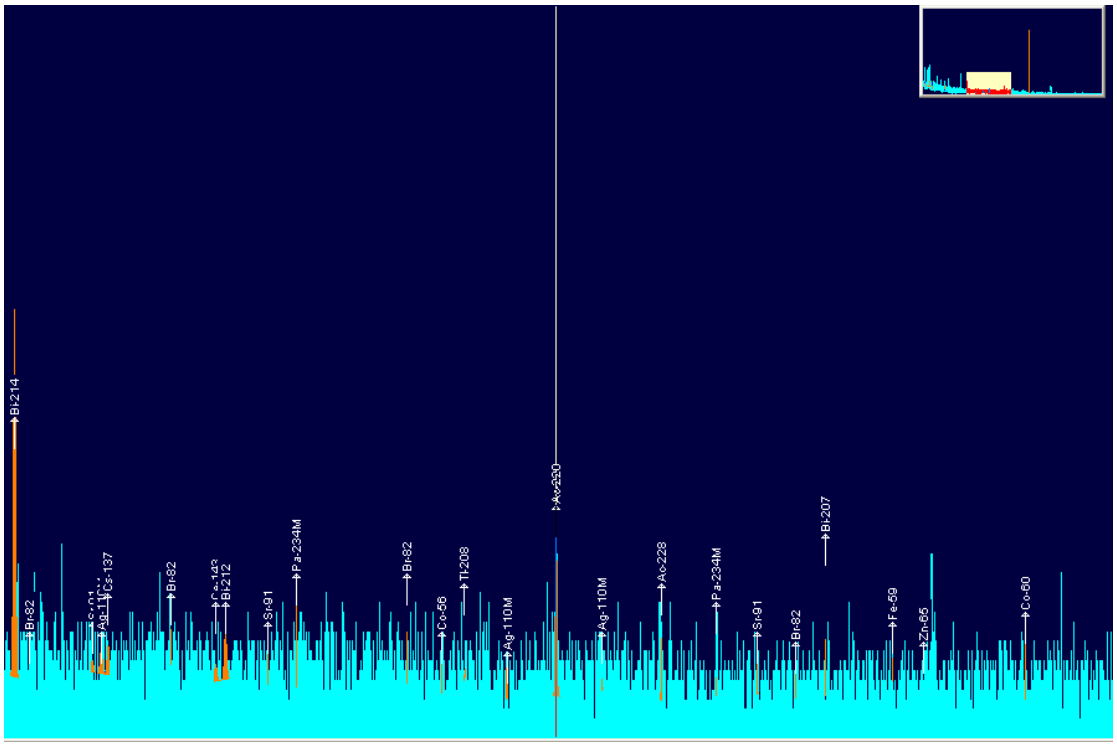


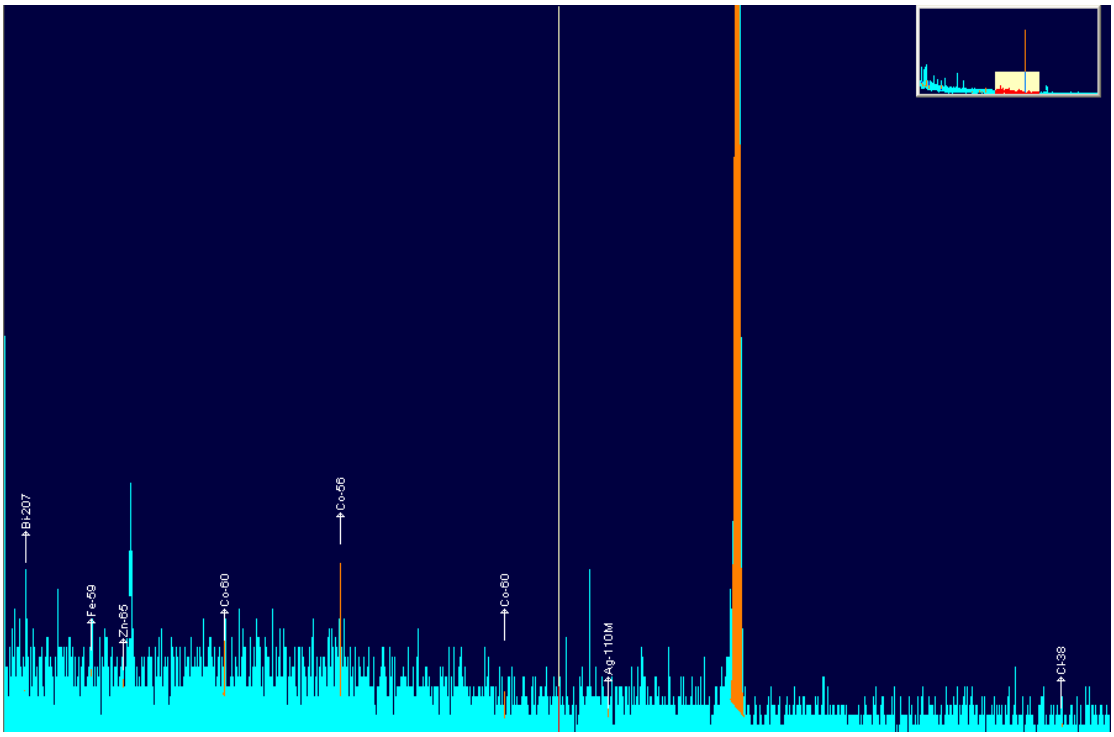




Bi-207
Fe-59
Zn-65
Co-60
Co-56
Co-60
Fe-59
Br-82
Ag-110M
Bi-212
Cl-38
Y-88

# APPINDEX E  SPECTRA OF SAMPLE N0 12




Bi-207
Fe-59
Zn-65
Co-60
Co-58
Co-60
Ag-110M
Cl-38