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Evaluation, Characterization and management of Naturally Occurring Radioactive
Materials and disused radioactive waste in Water and Soil Samples.

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LIST OF ABBREVIATIONS

ε	Porosity of dry earth material
β	Beta
γ	Gamma
β^-	Negatron
$\Delta\varepsilon$	Errors in absolute-efficiency of the detector
β^+	Positron
ΔN	Error in the net count
ΔP_γ	Error in gamma emission probability and
ρ_s	Density of the tantalum ore grains
ΔW	error in the net weight of the sample
ε_f	Full-energy peak efficiency
$\mu\text{Ci/mL}$	micro Curie per milliliter
$\mu\text{Sv/y}$	micro Sievert per year
ADC	Analog-to-Digital Converter
AED	Annual effective does
A°_{std}	initial activity of the standard at the time of manufacture
A_{Ra}	Activity concentration of ^{226}Ra
A_{Th}	Activity concentration of ^{232}Th
A_{K}	Activity concentration of ^{40}K
$^{222, 220, 219}\text{Rn}$	Randon-222, radon-220 and radon-219
$^{226, 224, 223}\text{Ra}$	Radium-226, radium-224 and radium-223
ARPANSA	Australian Radiation Protection and Nuclear Safety Agency
A_{std}	present activity of the standard
A_{u}	Activity concentrations of ^{238}U
A_{Th}	Activity concentrations of ^{232}Th
A_{K}	Activity concentrations of ^{40}K
BEIR	Biological Effects of Ionizing Radiation
BGS	Britain Geological Survey

Bq	Becquerel
Bq/kg	Becquerel per kilogram
Bq/L	Becquerel per liter
Bq/m ² s	Becquerel per square meter per second
Bq/m ³	Becquerel per cubic meter
Bq/mL	Becquerel per milliliter
CNRC	cancer nominal risk coefficient,
C _{Ra}	Activity concentration of ²²⁶ Ra
CSAE	Central Statistic Agency of Ethiopia
DCF _U	dose conversion factors for ²³⁸ U
DCF _{Th}	Dose conversion factors for ²³² Th
DCF _K	Dose conversion factors for ⁴⁰ K
D _{γ,ext}	average external gamma dose rate in μGy/h
e _A	Error in activity concentration (Bq/kg)
EC	Electron capture/radon emanation coefficient
ALARA	as low as reasonably achievable
BSS	Basic safety standard
DT	Destructive test
DU of ²³⁵ U	Depleted uranium (<0.7% ²³⁵ U) less than the natural abundance
ERPA	Ethiopian radiation protection
HEU	High enrichment uranium (~93% ²³⁵ U)
HLW	High Level Waste
HTGR	High Temperature Gas Reactor
IAEA	International Atomic Energy Agency
IAEA	International atomic energy agency
ICRP	International Commission on Radiological Protection
ICRU	Commission on Radiation Units and Measurements

IEU	Intermediate enrichment uranium (~40% ²³⁵ U)
ILW	Intermediate Level Waste
LCA	life cycle assessment
LLW	Low Level Waste
MDA	Minimum Detectable Activity
NDE	Non-Destructive Examination
NDG	Nuclear density gauge.
NDT	Non-destructive test
NID	Nuclide Identification
NORM	Naturally occurring radioactive material
SAF	Self-Absorption Factor
SGS	Segmented Gamma-Ray Scanner
SNM	Special Nuclear Material

ABSTRACT

In this study radiological hazards to members of the public and workers from exposure to natural radioactivity as a result of mining activities from Kenticha Tantalum Mines in Ethiopia, have been studied through several exposure pathways using direct gamma spectrometry to determine ^{238}U , ^{232}Th , ^{40}K , ^{226}Ra and ^{222}Rn in tantalum ore, soil, waste, waste tailing and water samples. Additionally, cancer risk assessment associated with NORM was estimated. The average activity concentrations of ^{238}U , ^{232}Th , ^{40}K , ^{226}Ra and ^{222}Rn in tantalum ore were $78.653\pm 1.431\text{Bq/kg}$, $24.945\pm 0.492\text{ Bq/kg}$, $603.170\pm 55.013\text{ Bq/kg}$, $69.478\pm 31.0\text{ Bq/kg}$ and $112.554\pm 50.249\text{ kBq/m}^3$, respectively. In soil the activity concentrations were $69.354\pm 1.081\text{ Bq/kg}$, $15.479\pm 0.231\text{ Bq/kg}$, $718.880\pm 65.531\text{Bq/kg}$, $68.923\pm 1.7\text{Bq/kg}$ and $111.655\pm 2.681\text{ kBq/m}^3$, respectively and in solid waste samples $110.496\pm 1.907\text{ Bq/kg}$, $15.009\pm 0.274\text{ Bq/kg}$, $607.269\pm 55.375\text{ Bq/kg}$, $98.300\pm 38.6\text{ Bq/kg}$ and $159.246\pm 62.607\text{ kBq/m}^3$ respectively. The values were generally above the worldwide average activity concentrations in all samples, except thorium-232. This might be due to the high contents of ^{238}U decay families and ^{40}K in the granite – pegmatite rocks of Kenticha area. The corresponding average external dose rate at 1m above the ground in air for tantalum ore, soil and solid waste samples were 76.407 nGy/h , 71.337 nGy/h , 85.408 nGy/h , respectively which were above worldwide average value of 60 nGy/h . The annual equivalent doses were also estimated as $0.021\pm 0.003\text{ mSv}$, $0.020\pm 0.001\text{ mSv}$ and $0.023\pm 0.004\text{ mSv}$ for ore, soil and solid waste samples, respectively and were found to be lower than the worldwide average of 2.42 mSv/y . Likewise, the radon emanation coefficient which is the fraction of radon generated within the grains of materials and escaped to the pore space, varied from $82\pm 2\%$ to $85\pm 2\%$ for ores, from $82\pm 2\%$ to $84\pm 2\%$ for soil, and from $53\pm 15\%$ to $83\pm 15\%$ for solid waste samples. Also, the radon exhalation rate varied in the range of 41.7 ± 25.7 to $87.3\pm 25.7\text{ mBq/m}^2\cdot\text{s}$ for ores, from 56.2 ± 1.4 to $58.1\pm 1.4\text{ mBq/m}^2\cdot\text{s}$ for soil and from 53.3 ± 32.1 to $120.2\pm 32.1\text{ mBq/m}^2\cdot\text{s}$ for solid waste, respectively. The radium equivalent activity, external hazard index, internal hazard index and representative gamma index, for all samples were estimated and generally found to be within recommended international values. Annual effective dose was evaluated from only external gamma dose rate. However, the relatively high values in all the other parameters measured implies that the Kenticha tantalum mine and its environments shown the significance of naturally occurring

radioactive material. Therefore, the results from this study will ignite in decision-making for future set-up of further research for the management of NORM wastes in Kenticha tantalum mine and for the emerging mining industry in Ethiopia. Radioactive material containing Cs-137, Ra-226, Co-60, Sr-90 and Am-241 were treated using the developed method. In the method different type of materials (cement, sands, concrete, etc.) were tested for absorption of radiation emitted from the source as well as suitability of the material for storage for long time. It was found that the best materials to be used in radioactive waste treatment is Concrete, where the surface dose reduced from 150 to 3 μ sav/h. In addition, design of storage container was proposed (with specification). Finally, based on indications of the present research findings and scientific background, recommendations were forwarded to help on amending nuclear waste management problems or indicate for other possible alternatives such as radiation map of Ethiopia.

CHAPTER ONE

INTRODUCTION

1.1.BACKGROUND OF THE STUDY.

Radiation sources are used throughout the world for a wide variety of beneficial purposes, in industry, medicine, research and education. The risks posed by these sources vary widely, depending on the amount of activities, the characteristics and chemical form of radionuclide, etc. If sealed sources are not damaged or leaking, sealed sources present a risk from only external radiation exposure. However, damaged or leaking sealed sources as well as unsealed radioactive materials may lead to contamination of the environment and internal exposure. The Goiania accident in 1987 could be listed as one of the biggest accident. After the Goiania accident, such kind of accidents occurred incessantly. These accidents caused 266 individual exposed, 39 fatalities and serious economic consequences. Recently, the International Atomic Energy Agency (IAEA) has been a growing awareness of these kinds of problems associated with radiation sources and has begun several activities. The activity of the source depends upon the duty required and varies from less than 1 micro curie to powerful sources containing thousands of curies. The higher activity sources are usually double encapsulated in a corrosion resistance metal such as stainless steel. Nuclear Waste is defined as: “any material contaminated by or incorporating radioactivity above thresholds defined in legislation is known as radioactive waste. It originates from a number of different sources such as power stations, nuclear weapons processing. On this planet, the application and study of nuclear science has been extensively increasing since the miraculous discovery of natural radioactivity by A. H. Becquerel in 1896 (Allisy, 1996). Natural and artificial radionuclides are the two main categories of radionuclides found in the environment. The sources of natural radioactivity can be divided into terrestrial and extraterrestrial (cosmic radiation and galactic cosmic origin). There are over 60 radionuclides found in nature, which can be placed in three general classes namely primordial, cosmic and anthropogenic radionuclides found in soil of the earth's crust, rocks that make up the planet, in water and ocean, in food and air, in building materials, and even within the tissues of living beings. This implies there is nowhere on Earth that natural radioactivity cannot be found (EPA, 2006). Therefore, all living organisms are exposed to low levels of natural

nuclear radiation during their daily lives. According to a review on ionizing radiation of the UK population report, natural radiation accounts for up to 85% of annual dose received by the world population expressed from building materials (18%), cosmic (14%), radon (42%) and food and drinking water (11%), as explained in Figure 1-1 (Watson et al, 2005). According to IAEA reports, exposure from natural radiation is, in most cases, of little or no concern to the public, except those who are working with mineral ores and naturally occurring radioactive material (NORM) (IAEA, 2005). However, the World Nuclear Association (WNA) states that any dose of radiation involves a possible risk to human health (WNA, 2011), even though the level of individual exposure to naturally occurring radioactive elements is usually statistically insignificant on an individual basis, from a health physics point of view.

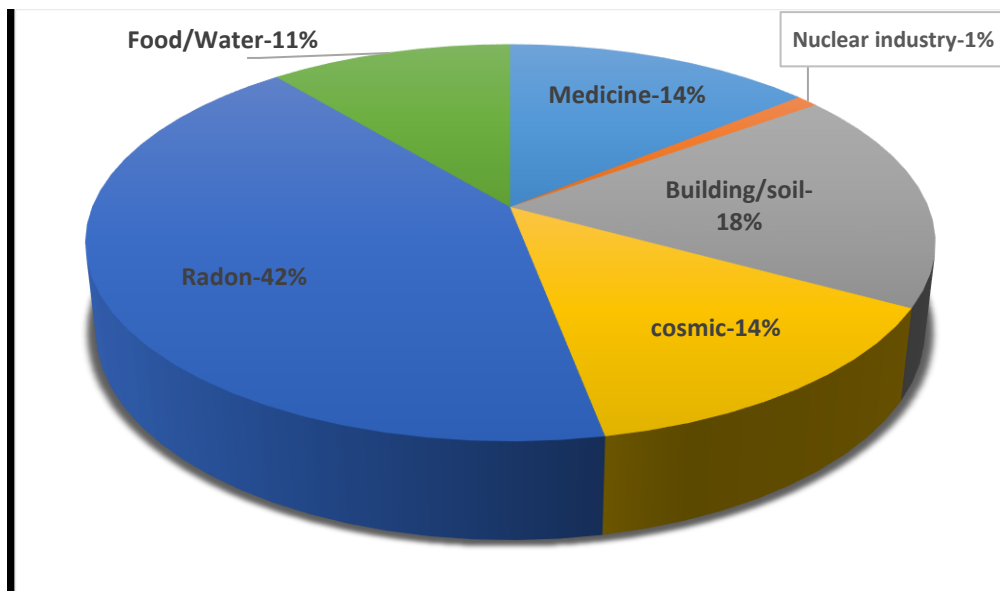


Figure 1.1-1: The world average annual dose from various sources (Todsadol, 2012).

Norms contains mostly uranium, radium, thorium, potassium, and their radioactive decay products, that are undisturbed by human activities (EPA, 2008). The concentration of NORM in most natural substances is so low that the risk is generally regarded as negligible, higher concentrations may arise as a result of intervention of man. Some industrial operations, such as mining, milling, processing, melting, etc., may liberate NORM present in the Earth's crust. For example, mining can release radon from the soil. Other industrial processes may act to concentrate NORM – uranium, thorium, potassium-40, lead, radium, for example, can be concentrated (SHA, 2002). Naturally occurring radioactive materials that have been concentrated or exposed to the accessible environment as a result of human activities is called

Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM). Technologically enhanced means that the radiological, physical, and chemical properties of the radioactive material have been altered by having been processed, or beneficiated, or disturbed in a way that increases the potential for human and/or environmental exposures (EPA, 2008).

Therefore, human technical activity can increase radiation exposure, not only to the person directly involved in these activities, but also to the local or even whole population and the environment (Bou-Rabee et al., 2009).

The presence of NORM in rare earth minerals in varying concentrations is quite often significant enough to result in occupational and environmental radiation exposures during their mining, milling and chemical processing for the extraction of the rare earth elements and compounds (IAEA, 2008). Knowledge of NORMs is helpful in establishing protective controls for mining activities and use of these materials. There is, however, no regulatory radiation dose limit for natural background radiation exposures. Nevertheless, NORM may be regulated if technological activities such as mining and milling etc. and result in the material concentration of NORM above natural background levels (W. F. Wilson, 1994). In order to protect the health effect of the general public against radiation risk originating from naturally occurring radiation, the measurement of radioactivity in the environment have become the focus of greater attention by the IAEA in recent years (IAEA, 2005). For example mining processes elevate uranium, thorium, radium, potassium 40 and the radon concentrations by factors of several hundred (SHA, 2002). Therefore, studying NORM is very important to assess its influence on the natural environment and to prevent the possible health effects on humans.

1.1 STATEMENT OF THE PROBLEM.

The purpose is strictly academic and has the intention to find potential improvements in the waste management operations regarding NORM and spent radioactive waste. Two research questions will lead to finding potential improvements. The first question has the intention to reveal practical operational processes to be implemented for the improvement of the waste management process of NORM in Ethiopia. Radioactive

waste is a source of ionizing radiation and therefore it is necessary to protect both human being and the environment from the harmful effects of this radiation.

This research proposed to evaluate NORM at different mining area of , Ethiopia. NORMs such as Uranium and thorium decay families and potassium – 40 are associated with rare earth element-bearing materials. Ethiopia has the largest tantalum resources mine at Kenticha. Mining activity enhances the levels of NORM in the raw materials, mine tailings, products and waste effluent. According to ERPA report, the radium²²⁶Ra and its daughter, ²³⁸U and daughter and ²³²Th and its daughter radionuclides in the tantalum ore, solid iron waste, tailing dam samples were to have found high activity concentration and radiation doses higher than that of the background radiation measured at Kenticha tantalum mine(ERPA, 2013). This result agreed with the report of Ministry of Mining of Ethiopia, which says most parts of the Kenticha Colton, the uranium content is above the critical level of 0.5% which is the European union recommended limit(MME, 2014). In IAEA radioactive transport regulation, tantalum minerals are classed as radioactive during transport (dangerous goods) due to the small but measurable levels of naturally occurring thorium and uranium decay products present in those raw materials(TIC, 2013). Hence, the needs to determine the activity concentration of NORMs at Kenticha tantalum mine. Although, the Kenticha tantalum mine has been in operation since 1990, there is an inadequate knowledge on the level of NORM and their associated health risks. There is very little study carried out on NORM due to tantalum mining, the impact on ambient environment and radiological health risks to workers and the general public in that area, even in the world.). Radiation map of Ethiopia and Natural radioactivity level in cement raw materials is not well known. In Ethiopia there is no radiation management plan; The major issues of radiation management to be implemented include:

- ✓ Protection of workers from radiation within the factory and its premises
- ✓ Safe containment of radioactive solid waste
- ✓ Establishing radiation monitoring program at the work place and in the environment

The wastes in question are very varied and may be in the form of discrete sealed or unsealed radiation sources or may be in the form of process or consumable materials. They arise in many activities including diagnostic, therapeutic and research

applications in medicine; process control and measurement in industry; and many uses in research, teaching, agriculture, geological exploration, construction and other fields of human endeavor. . A broader overview of waste arising from this range of applications is the critical problem of radioactive waste management code of contact and code of practice. The variable range of waste types encountered and the possibility for changes to take place in the circumstances of their generation and management requires that particular attention should be given to the safety aspects of their management and regulatory control. Both management and regulatory control regimes need to be sensitive and responsive to these factors. Good operating practice can significantly reduce the amount of waste generated but in general, waste cannot be fully eliminated. The waste may contain sufficient quantities of radionuclides such that if not properly managed it has the potential to present serious risks to human health and the environment. Experience has shown this to be the case and particularly in respect of disused or spent sealed radiation sources where poor practices in the past have resulted in radiation exposure of both operating personnel and members of the public and have caused extensive contamination of the environment. Instances have arisen where such lack of control has resulted in a number of deaths and/or considerable economic loss. In addition, the EIA of radioactive release that the containment of radioactive waste in safe manner is new to Ethiopia, and therefore, further investigations will be required to precisely define the design parameters of radioactive waste (NORM) and spent sources.

1.2 RESEARCH OBJECTIVES

GENERAL OBJECTIVE

The aim of this research was to study the environmental and health related impacts of naturally occurring radioactive materials (NORMs) resulting from the operational activities of Kenticha tantalum mine.

SPECIFIC OBJECTIVE

The specific objective of this study was:

1. To assess the NORM levels at kenticha tantalum mines and Derban cement raw material mining site and environmental impacts of radioactive release
 - By identifying, quantifying and determining the activity concentration of ^{238}U , ^{232}Th and ^{40}K of NORMs at Kenticha by gamma spectrometry.
2. To evaluate radiological hazards and risks due the NORMs.
3. To justify the dose rate of the radiation by comparing to the international standard code of practice.
4. To evaluate radioactive waste management code of conduct and code of practice of Ethiopia.

1.3 RELEVANCE AND JUSTIFICATION

With the dramatic expansion of electronic industry, tantalum is the most widely used metal in the industrialized world. This leads to enormous mining activities to meet the increasing demand for electronic capacitors and others. However, tantalum-mining activities is harmful on the environment and the population because the materials, out of which tantalum ore is made, are of soil and rock origin that contains naturally occurring radioactive materials that contribute approximately 80% of the total radiation dose a person receives in a year. Moreover, as said in section (1.2), the uranium content of tantalum concentrate from Kenticha mine is above the critical level of 0.5% (MME, 2014) and also according to IAEA radioactive transport regulation, tantalum minerals are classified as radioactive due to the small but measurable levels of naturally occurring thorium and uranium decay products present in those raw materials (TIC, 2013).

However, in many African countries including Ethiopia, naturally occurring radioactive materials from mining area are not under satisfactory regulatory control. This is because; there is a general lack of awareness and knowledge by mining companies and public about the radiological hazards associated with NORM. In addition, not much research has been done on NORM (TENORM) in Ethiopia. Therefore, this research will provide familiarity on NORMs and assess the dose of the public from enhanced levels of NORMs. The work proposed in this study is therefore justified because it is useful to identify and determine concentration of specific radioactive elements in NORM, since it will contribute to creating awareness on the potential impact of naturally occurring radioactivity on the communities as a result of mining activities, the determination whether the TENORMs from this mine activity is normal to the background or contaminating the area, to reduce its influence on the environment, to emplace NORM management mechanism, to improve the health and safety practices, to recommend the mining management body and to determine (from the study) whether area monitoring at the mining activity would be necessary or not. Moreover, it may have to stakeholders giving more attention to the environmental impacts of NORMs and motivate further research into controlling NORM radiation levels in the country. Therefore, this study also allows us to analyze how much natural radionuclide the Kenticha tantalum mining releases into the environment. The results of this study will benefit the local community, regulatory authorities, health sector, water treatment agencies, mining industry, environmental protection agency, geological survey sectors, etc.

1.4 SCOPE OF THE STUDY

This thesis is concerned with radioactive sources that may be used in industry, research, teaching, agriculture, medical practice military applications and NORMS. It is not concerned with the nuclear fuel cycle as such. All efforts were being made to carry out the present study in a very systematic and organized manner, well supported by the actual field data, laboratory tests and the secondary data obtained from various sources. However, these efforts were made under the limitations of time, resources and the financial constraints This study brings to focus environmental issues related to mainly NORM since it is a key issue of radiological protection in mining areas. The scope of the research was limited principally on identifying and quantifying the

NORM activity concentrations of primordial radionuclides (^{238}U , ^{232}Th decay series and ^{40}K) of tantalum ore, soil, waste, waste tailings and water samples. For the radiological risk estimation to which an individual is exposed, the study will cover terrestrial gamma radiation (absorbed dose rate in air) measured 1m above the ground surface; the external annual effective dose; radiological cancer risks assessment, radium equivalent activity, representative gamma index, external and internal hazard indices for all tantalum ore, soil, solid waste and water samples. All computed values were compared with available reported data from other countries and with the world average value for the analyzed samples. This was achieved by collecting tantalum ore, soil, sample, water samples from Kenticha, soil sample and water sample from Derban limestone mining area by deciding on independent sample location to have representative sample.

1.5 STRUCTURE OF THIS RESEARCH

This research paper consists of six major chapters with brief description of the contents. Chapter one gives an introduction of the study that comprises background information statement of the problem, objectives of the work, relevance and justification and scope of the study. Chapter two gives an insight on what has so far been done on NORMs from past and related work. It also shows the gaps in knowledge that need to be addressed. Available theoretical approaches are relevant to the dose assessment of natural radioactivity from tantalum mine. Chapter three discusses the materials and methods used in this study as well as the relevant calculations for the work. Chapter four gives the data interpretation from this study in a clear and logical manner, aided by the use of tables and figures as required with discussion of results from the study. Finally, chapter six concludes the study and gives an overall summary of the research, recommendation and lessons learned.

CHAPTER TWO

LITRATURE REVIEW

This chapter gives the review of NORMs, information on tantalum as well as its uses, sources and reserves, its nature of radioactivity, the mineral processing of tantalum mine and the NORMs associated with tantalum. It also focuses on natural radioactivity and its sources as well as occupational and public exposure to these sources. It also focuses on radon and its sources of exposure. The detector resolution, detector efficiency of gamma spectrometry system, radiation exposure pathways and instrumentation used for measuring natural radioactivity are included in this section.

2.1.1 Overview of Nuclear Waste.

Radiation sources are used throughout the world for a wide variety of beneficial purposes, in industry, medicine, research and education. The risks posed by these sources vary widely, depending on the amount of activities, the characteristics and chemical form of radionuclide, etc. If sealed sources are not damaged or leaking, sealed sources present a risk from only external radiation exposure. However, damaged or leaking sealed sources as well as unsealed radioactive materials may lead to contamination of the environment and internal exposure. The Goiania accident in 1987 could be listed as one of the biggest accident. After the Goiania accident, such kind of accidents occurred incessantly. These accidents caused 266 individual exposed, 39 fatalities and serious economic consequences. Recently, the International Atomic Energy Agency (IAEA) has been a growing awareness of these kinds of problems associated with radiation sources and has begun several activities. The activity of the source depends upon the duty required and varies from less than 1 micro curie to powerful sources containing thousands of curies. The higher activity sources are usually double encapsulated in a corrosion resistance metal such as stainless steel. Nuclear Waste is defined as: “any material contaminated by or incorporating radioactivity above thresholds defined in legislation is known as radioactive waste. It originates from a number of different sources such as power stations, nuclear weapons processing.). The control of radioactive waste is achieved by an effective waste management program. The program, shown schematically in Figure 2.1 prescribes processes, which may be required to ensure that waste can be disposed of safely.

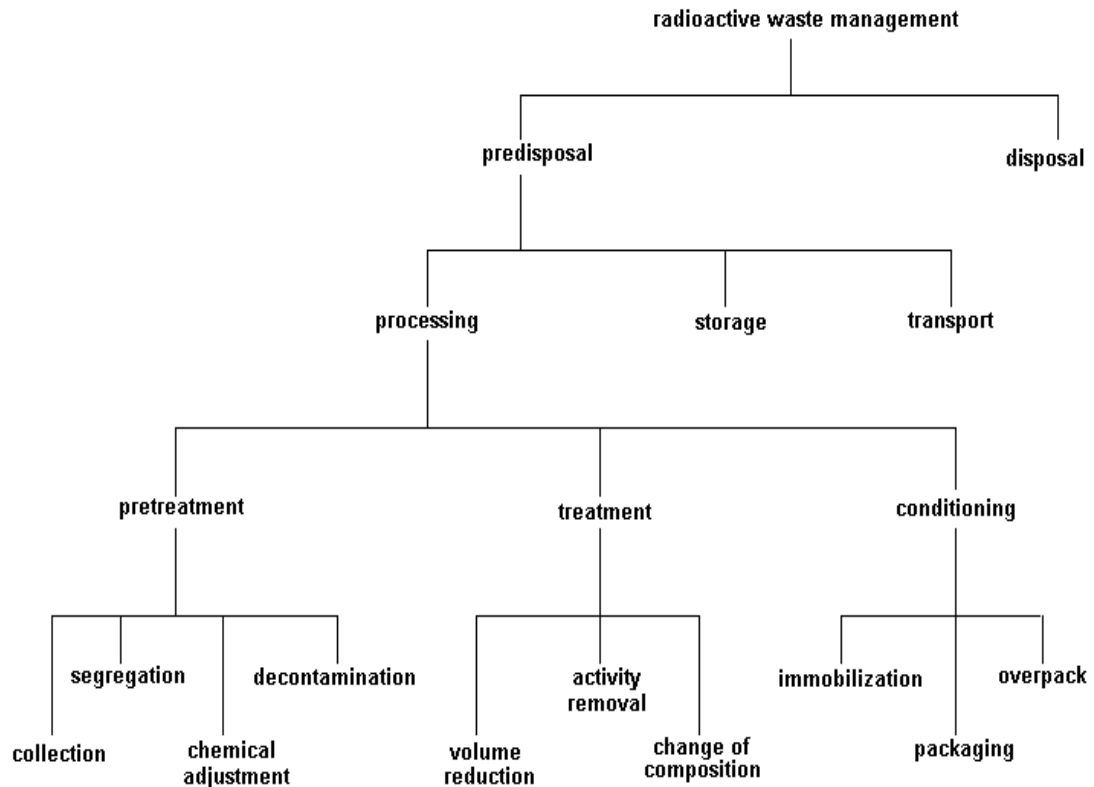


Figure 2.1 radioactive waste management code of practice

2.1.2. Half-Life

Different radioactive atoms are transformed at different rates and each radionuclide has its own characteristic transformation rate. For example, when the activity of ^{32}P is measured daily, over a period of about 3 months and the percentage of the initial activity is plotted as a function of time; the curve shown in Figure 4-10 is obtained. The data show that one-half of the ^{32}P is gone in 14.3 days, half of the remainder in another 14.3 days, half of what is left during the following 14.3 days, and so on. The time required for any given radionuclide to decrease to one-half of its original quantity is a measure of the speed with which it undergoes radioactive transformation. This period is called the half-life and is characteristic of the particular radionuclide. Each radionuclide has its own unique rate of transformation, and no operation, either chemical or physical, is known that will change the transformation rate; the decay rate of a radionuclide is an unalterable property of that nuclide. Half-lives of radionuclide range from microseconds to billions of years. From the definition of the half-life, from the definition of the half-life, it follows that the fraction of a radionuclide remaining after n half-lives is given by the relationship (Mohammed elberedi (2012)).

$$\text{half life} = \frac{0.693}{\lambda} T_{1/2}$$

2.1.3. Radioactive Equilibrium

Radioactive equilibrium is the term usually used to explain the state when the members of the radioactive series decay at the same rate as they are produced. The three predominant cases of the state of equilibrium are listed as follow.

Secular Equilibrium; ‘Secular equilibrium’ is a steady-state condition that in which the half-life of the parent is very much greater than that of the daughter

Transient Equilibrium; A different state of equilibrium, called ‘transient equilibrium’ can occur in which the parent half-life is longer-lived than that of the daughter but not significantly long where.

No Equilibrium; In case the half-life of the parent nuclide has shorter lived than that of the daughter product, the state of equilibrium will not be attained

2.1.4. Waste management considerations with respect to NORM

Various solid and liquid wastes containing NORM are produced (generated/enhanced) in large volumes by the oil and gas facilities during production. Other NORM waste (mostly solid) is produced when decontamination operations are held and during decommission and rehabilitation of an oil and gas production facility, waste management facility and/ or treatment facilities. Depending on the radioactive level of these wastes, they could have radiological effects on the workers associated to the waste, as well as on other personnel or members of the public who may be exposed to the radiation if the wastes are not managed correctly and these radionuclides end up spread in the environment. Besides the radioactive hazards of NORM, these wastes might possess other chemical characteristics adverse to human health or the environment (IAEA 2003). 2.4.7. The major naturally occurring radioactive materials (NORM) ^{238}U , ^{232}Th , and ^{40}K give rise to external, γ -radiations in the environment (UNSCEAR, 1993). Radon (^{222}Rn) and Thoron (^{220}Rn), which are rare gases formed in the decay chain of ^{238}U and ^{232}Th series escape through the pores of the soil and diffuse into the atmosphere in quantities depending on the radioactive content and types of the soil

and the prevailing atmospheric conditions and give rise to radiations to the respiratory system due to inhalation (ICRP Publication 50; ICRP Publication 65). Most NORM contains radionuclides from the so-called primordial decay chains resulting from the decay of uranium-238 (^{238}U), uranium-235 (^{235}U) and thorium-232 (^{232}Th). In the majority of situations, the naturally occurring radionuclide concentrations are not high enough to pose problems for the environment or human health, but in those situations where the radionuclide concentrations are significantly higher than the average level, human health may be affected. For the two most important primordial decay series, the average worldwide background concentrations are: ^{238}U : 30-50 Bq kg⁻¹ ^{232}Th : 40-60 Bq kg⁻¹. People have always been subjected to natural background radiation. Typical background rocks and soil contain in the order of 1 to 10 ppm each of uranium and thorium although much higher concentrations, especially in areas of mineralization, are not uncommon (UNSCEAR 2000, U.S. NCRP 1987). People are also exposed to sources of radiation that we ourselves create. X-rays and other kinds of radiation used for medical purposes, fall-out from past nuclear weapons tests and the small quantities of radioactive materials that are allowed to escape to the environment in the course of normal operation of nuclear installations are some examples. Natural background radiation typically results in a dose rate of about 2 to 3 mSv per year. (UNSCEAR 2000), although some places in the world experience much higher exposure rates, i.e. 10 mSv per year and higher. The amount of radionuclide, $N(t)$ at any time t is given by the following equation. Another quantity of interest is what is referred to as the half-life $t_{1/2}$ of a nuclide. It is the time it takes a given number of nuclide to decrease by half in number via radioactive decay. The half-life of a nuclide is related to the decay constant of the nuclide. If $t_{1/2}$ is the half-life of a nuclide, then equation (2) implies: The unit of activity is the Curie (Ci). $1 \text{ Ci} = 3.7 \times 10^{10}$ disintegration per second (dps). The SI unit for activity is Becquerel (Bq).

2.1.5. Stochastic Effects

In contrast to the deterministic effects, arising at high dose situation very little is known about the occurrence of cancer and genetic effects from exposure to low-level radiation. For cancer, this is due in part to the long delay between irradiation and the

appearance of the effect, and in part, to the difficulty of isolating radiation from other possible causes such as drugs, cigarettes, chemicals, etc. In the case of genetic effects, laboratory experiments on mice and other animals have shown such injuries (Leo, 1994). Present knowledge of the genetic effects of radiation on man is based entirely upon extrapolation from these experiments. Nevertheless, it is generally accepted that stochastic effects do not have a threshold dose and they are probabilistic in nature. The probability of incidence of such effect increases with accumulated dose rather than its dose rate. As there is a certain non-zero probability depending on other effects too, these are referred to as stochastic effects.

2.1.6. International radiation protection regulations and guidelines

The International Commission on Radiological Protection (ICRP) specified that any exposure to ionizing radiation may be potentially harmful to health, and advocates three fundamental principles for managing radiation exposures:

a) Justification

No activity involving ionizing radiation for any purpose can be justified unless it is possible to demonstrate that it will lead to a positive net benefit.

b) Optimization

The magnitude of individual exposures, the number of people exposed and the likelihood of incurring exposures shall be kept as low as reasonably achievable, economic and social factors being taken into account (the ALARA principle).

c) Limitation

No individual is to be exposed to a radiation risk that is judged unacceptable in any normal circumstances. The ICRP recognizes that everyone is subject to a significant background radiation exposure. However, even smaller-than-background doses from occupational practices are unjustifiable if there is no associated benefit, or they can be readily avoided. The situation with Naturally Occurring Radioactive Materials (NORM) is complicated by the fact that NORM is a major contributor to background radiation, and natural background levels are highly variable. In the United Kingdom, the current regulatory controls, the Ionizing Radiation Regulations 1999 (UKIRR,

1999), do not specifically address NORM IN order to determine the optimum regulatory approach

2.1.7. Exclusion is the deliberate exclusion of a particular category of exposure from the scope of an instrument of regulatory control because it is not considered amenable to control through the regulatory instrument in question.

2.1.8. Exemption is the determination by a regulatory body that a source or practice need not be subject to some or all aspects of regulatory control on the basis that the exposure (including potential exposure) due to the source or practice is too small to warrant the application of those aspects or that this is the optimum option for protection irrespective of the actual level of the doses or risks.

2.1.9. Clearance is the removal of radioactive material or radioactive objects within authorized practices from any further regulatory control by the regulatory body. Clearance level is a value, established by a regulatory body and expressed in terms of activity concentration and/or total activity, at or below which a source of radiation may be released from regulatory control.

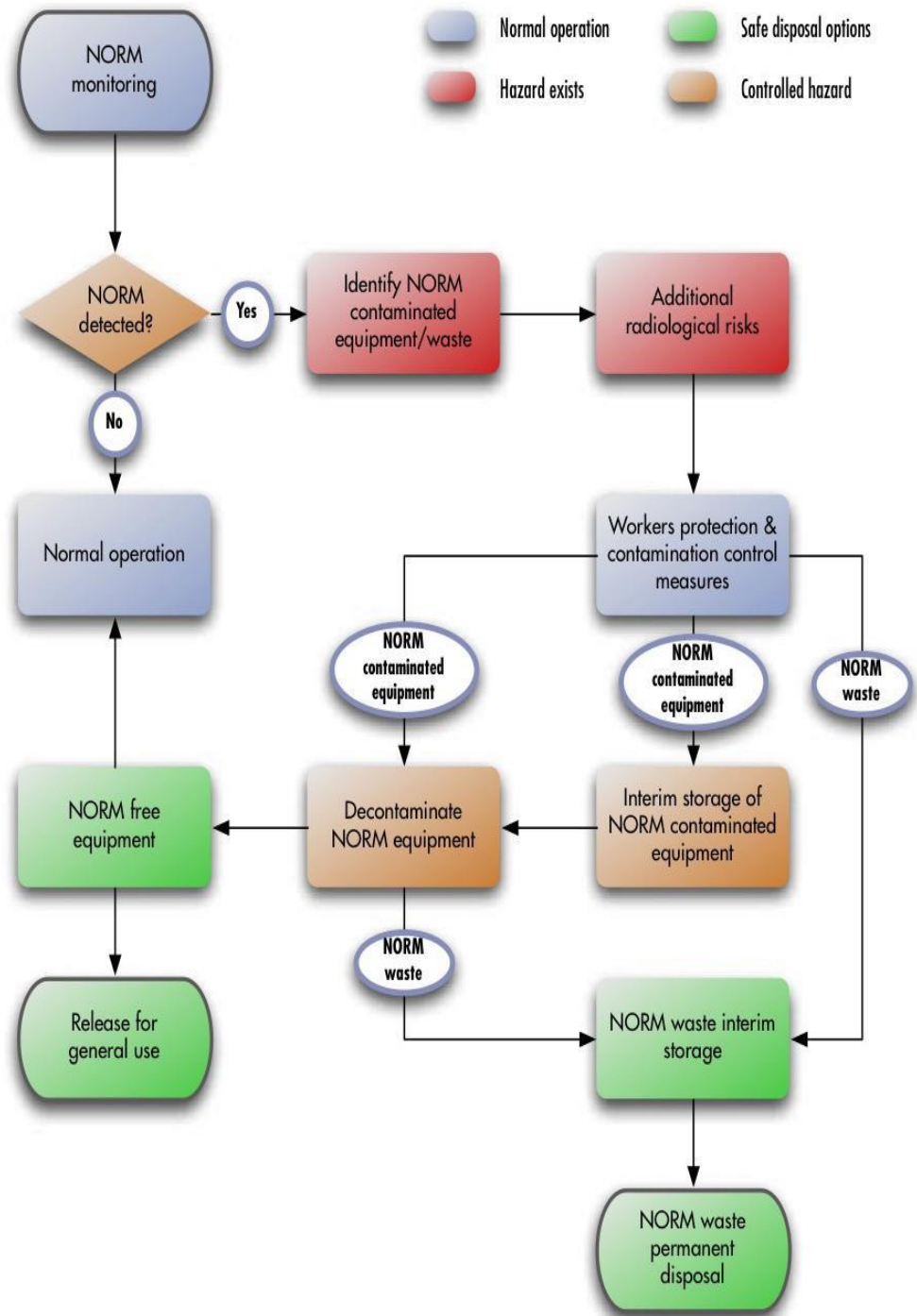


Fig 2.1.2: Process cycle of NORM Management

The worldwide average activity concentrations of ^{238}U , ^{232}Th and ^{40}K are 33, 45 and 420 Bq/kg, respectively (Saher et al, 2013; UNSCEAR, 2008a). However, the natural levels of uranium in soils in the UK can be up to 40Bq/kg. Granites and black-shales are occasionally rich in these elements, with typical concentrations of up to 200Bq/kg while Monazite can contain uranium and thorium at up to 200kBq/kg(Sobanski, 2004). The world nuclear association report said, the Australian mineral concentrate contains

thorium and uranium with activity concentration of 0.3 - 3.0Bq/kg and 0-0.8 Bq/kg, respectively. Also it said that the procsstedtantalum minerals is reported with the concentrate and its total activity concentration of NORMs sometimes exceed the Transport Code threshold of 10 kBq/kg, and some reaches 75 kBq/kg(WNA, 2009). However, the activity levels of NORMs in the niobium industry may be high, with pyrochlore containing 10,000–80,000 Bq/kg of ²³²Th. In one niobium facility in Brazil, activity levels in waste ranged up to 200,000 Bq/kg of ²²⁸Ra (in barium sulphate) and 117,000 Bq/kg of ²³²Th (in the slag)(UNSCEAR, 2008a). Whereas, phosphate ores typically contain about1,500 Bq/kg of uranium and radium; although some phosphate rocks contain upto 20,000 Bq/kg of U₃O₈. In general, phosphate ores of sedimentary origin have higher concentrations of nuclides of the uranium family(UNSCEAR, 2008a).

According to the reports of technical university of Denmark, the radiation protection issues in relation to NORM is a growing consensus that natural radioactivity actually leads to significant doses to workers in a variety of industries (DTU, 2015). Table 2.1-1 presents a rough overview of average doses to workers in a variety of industries. The table shows that doses associated with NORM work are of equal magnitude - or even higher - than doses in industries.

Table 0-1: Worldwide average doses to workers in a variety of industries (DTU, 2015)

Industrial activities	Worldwide average annual dose (mSv/y)
Medical	0.5
Industrial	1
Nuclear	1.5
Air crew	2
NORM	0.5 - 5.5

The wide range of doses to NORM workers is reflecting the fact that NORM industries are very diverse in terms of NORM quantities, activity concentration, isotopes, and primary exposure pathway (Table 2.1-1). Within the NORM field exposure to radon in mines is by far the most significant dose contributor. Generally, the worldwide average dose received by all human being from background radiation is 2.4 mSv/y, that varies depending on the geology and altitude where people live and it ranges

between 1 to 10 mSv/y, but can be more than 50 mSv/y (UNSCEAR, 1993, 2000b). The city of Ramrasin Iran hosts some of the highest natural radiation levels on earth, and over 2000 people are exposed to radiation doses ranging from 0.01 to 0.26 Sv/y. Surprisingly, inhabitants of this region seem to have no greater incidence of cancer than those in neighboring areas of normal background radiation levels (Kerim, 2002). Moreover, the highest identified background level of radiation level affecting a substantial population is in Kerala and Madras states in India where some 140,000 people received doses, which averaged over 15 mSv/y from gamma radiation, in addition to a similar dose from radon. Comparable levels occur in Brazil and Sudan, with average exposures up to about 40 mSv/y to many people. Also another highest level of natural background radiation recorded is on Brazilian beach: 800 mSv/y, although people do not live there (WNA, n.d.). Table (2.1-2) shows the range of radioactivity levels measured in some local rocks and soil samples including cosmic and terrestrial radiation of the various contents.

Table 0-2: Average and maximum annual doses to some areas of world (Kerim, 2002; UNSCEAR, 2000c)

Country	Area	Approximate population	Absorbed Dose rate in air (nGy/h)
Brazil	Guarapari	73 000	90-170 (street), 90-90 000 (beaches)
Iran	Ramsar	2 000	70-17 000
India	Kerala	100 000	200-4 000
China	Yangjiang	80 000	370 (average)

Many places in Iran, India and Europe are noted for the natural background radiation that gives an annual dose of more than 100 mSv to people and up to 260 mSv (at Ramsar in Iran, where some 200,000 people are exposed to more than 10 mSv/y). Lifetime doses from natural radiation ranges up to several thousand mSv. Nevertheless, there is no evidence of increased cancers or other health risks arising from these high natural levels (WNA, n.d.).

At Kenticha tantalum mine in Ethiopia, there is very little study on NORMs that describes the concentration of uranium and thorium families and ⁴⁰K. The only of

this study done by Zerihun et al. 1995 described only the qualitative detection limit (QDL) (i.e. 0.009 ppm) of uranium in the rock samples collected from Kenticha area (Zerihun et al, 1995). Another report by Journal of African Earth science revealed that granite rocks from Shakiso area contains 7.84 ppm and 1.54 ppm of thorium and uranium, respectively. The latest EMDSC environmental and social impacts assessment 2013 report, the tantalum concentrate concentration of uranium and thorium is above the allowable limit of the international trade which needs to be lowered to acceptable average percentage concentration of 0.81 and 0.07 for uranium and thorium, respectively. Therefore, this study is expected to provide data in terms of activity concentration (in Bq/kg and/or Bq/l) of ^{238}U , ^{226}Ra , ^{232}Th and ^{40}K for the mineral samples collected from Kenticha tantalum mine in Ethiopia. In addition, the radon concentration and emanation coefficients from dry samples will be carried out and compared with the UNSCREAR 2000 reports of 78kBq/m^3 (UNSCEAR, 2000b).

2.2. INFORMATION ON TANTALUM

Tantalum is a rare, hard, lustrous transition metal, which is located in-group five, period six of the periodic table. It's high melting and boiling points confer significant heat resistance. It is highly resistant to corrosion and immune to chemical attack at temperatures below 150°C . Tantalum is twice as dense as steel and highly durable. It is also highly ductile and surpasses most other refractory metals in workability and weld ability. Other properties are superconductivity and a high co-efficient of capacitance, which means that it can store and release an electrical charge (Hayes and Burge, 2003; Theron, 2010). Tantalum metal is highly unreactive towards nearly all of the mineral acids except HF. The reason for this extreme corrosion resistance is the formation of a thin layer of tantalum oxide (Ta_2O_5). Ta_2O_5 , unlike most other metal oxides, does not degrade to the metal itself, it adheres very well to the metal surface, which therefore adds to the protection of the metal, much like aluminum and its oxide.

2.1.1 NATURALLY OCCURRING RADIOACTIVE MATERIALS (NORMs)

The term NORM is used to describe situations where human activities have increased the potential for exposure to these radionuclides in comparison to the naturally occurring situation. Naturally occurring radioactive materials has existed in the Earth's crust since creation of the Universe. The natural radiation environment consists of cosmic radiation (producing cosmogenic radioactivity) and terrestrial

Table 2.1.1-1: Long-lived cosmogenic radionuclides in atmosphere (Choppin et al, 2002)

Nuclides	Half-life(years)	Decay mode	Particle energy (MeV)	Production rate (atoms/m²s)
³ H	12.32	β-	0.0186	2500
¹⁰ Be	1.52x10 ⁶	β-	0.555	300
¹⁴ C	5730	β-	0.1565	17000-25000
²² Na	2.605	β+	0.545	0.5
²⁶ Al	7.1x10 ⁵	β+	1.16	1.2
³² Si	160	β-	0.213	1.6
³⁵ S	0.239	β-	0.167	14
³⁶ Cl	3.01x10 ⁵	β-	0.709	60
³⁶ Ar	268	β-	0.565	56
⁵³ Mn	3.7x10 ⁶	EC	0.596	
⁸¹ Kr	2.2x10 ⁵	EC	0.28	

2.1.2 TERRESTRIAL RADIOACTIVITY

The natural occurring radionuclides of terrestrial origin also called primordial radionuclides have been in earth since the formation of the earth. Although some have long disappeared to levels that are not detectable anymore, some radioisotopes take a long time to decay (on the order of hundreds of millions of years), they are still present today. Some of the original primordial nuclides, whose half-lives are about as long as the earth's age, are still present, with half-lives often on the order of hundreds of millions of years(Read and Hayder, 2011; Watson et al, 2005). Radionuclides that exist for more than 30 half-lives are not measurable (NAS, 1999). The naturally occurring radionuclides are participants of one of the long chains, i.e. uranium (4n+2), Thorium (4n), actinium (4n+3) and neptunium (4n+1) series, the most radiologically important being those of ²³⁸U, ²³²Th and ⁴⁰K.

2.2.3. URANIUM SERIES (4n +2 SERIES)

The uranium atom consists of three different isotopes: ²³⁸U (99.3%), ²³⁵U (0.72%) and ²³⁴U (0.005%). The ²³⁸U and ²³⁴U belong to one family called the uranium series

(4n+2) (Cember, 2009). This series commences with the ^{238}U isotope, due to its very long half-life, which produces 14 radioactive isotopes in 14 decay steps (by emitting 8 α -particles and 6 β -particles; which is accompanied by gamma radiation) before ending with a stable isotope of ^{206}Pb (see Figure 2.8-1).

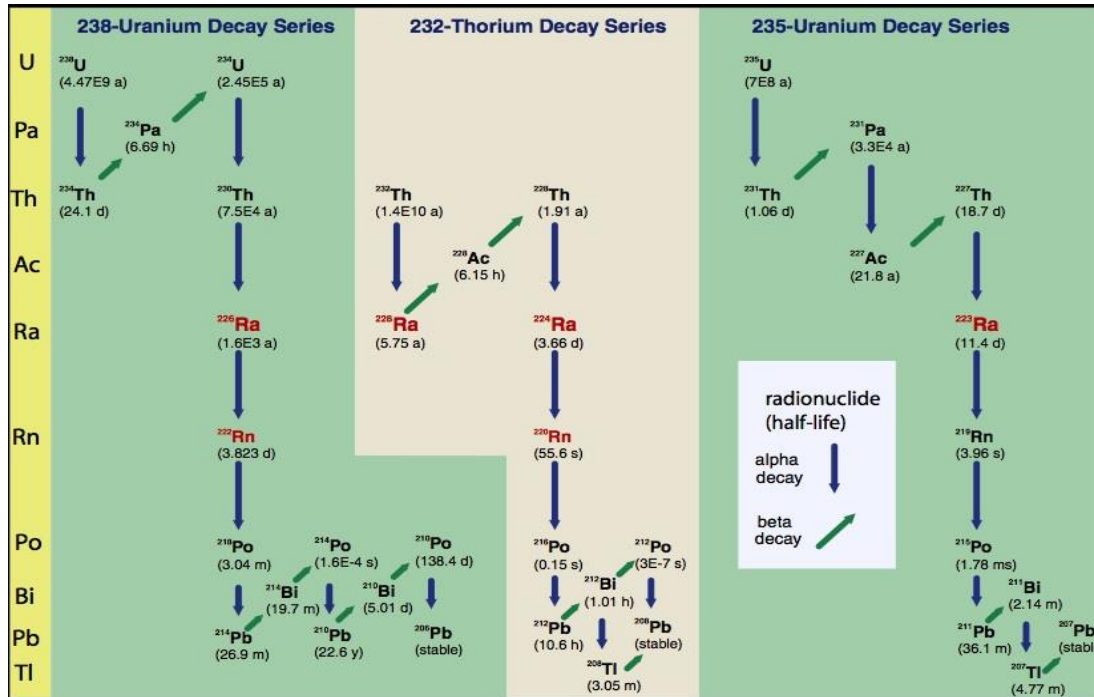


Figure 2.1.1-1: Decay series of the natural ^{238}U , ^{235}U and ^{232}Th

Pb-214 and ^{214}Bi can be measured easily by gamma spectrometry. The decay products of greatest importance is ^{226}Ra , ^{222}Rn , and radon daughters, principally ^{210}Pb and ^{210}Po (Jason, 1998; Richard and Martin, 1997). This series is said to be in secular equilibrium because all its daughters following ^{238}U have shorter half-life than the parent nuclide ^{238}U (Richard and Martin, 1997). Under normal conditions, in a natural material, the $^{235}\text{U}/^{238}\text{U}$ ratio will be constant and all nuclides in each of the series will be in equilibrium (Read and Hayder, 2011).

2.2.4. ACTINIUM SERIES (4n +3 SERIES)

The uranium-235 isotope belongs to a series called the actinium series or 4n+3 series. It starts with ^{235}U and ends up in a stable lead ^{207}Pb . The decay series involves 12 nuclides in 11 decay stages with the emission of 7 α -particles (ignoring a number of minor decay branches) as shown in Figure 2.8-1. Within this series, only ^{235}U itself can readily be measured, although ^{227}Th , ^{223}Ra and ^{219}Rn can be measured with more

difficulty. Even though, the uncertainties may be high, measurement of the daughter nuclides can provide useful support information confirming the direct ^{235}U measurement or giving insight into the disruption of the decay series. Although only a small proportion of the element (^{235}U), its shorter half-life means that in terms of radiations emitted, its spectrometric significance is comparable to ^{238}U .

2.2.5. THORIUM SERIES (4n SERIES)

Of the 26 known isotopes of thorium, only 12 have half-lives greater than one second, and of these only 3 (^{232}Th , ^{230}Th and ^{229}Th) have half-lives sufficiently long to warrant an environmental concern (ANLESD, 2007). Thorium-232 is the most abundant (about 100%) of the naturally occurring radioisotopes which is the first member of long series called the thorium series (4n) as shown in Figure 2.8-1. During 10 decay stages, 6 α -particles are emitted. Four nuclides can be measured easily by gamma spectrometry (^{228}Ac , ^{212}Pb , ^{212}Bi and ^{208}Tl). The decay of ^{212}Bi is branched – only 35.94% of decays produce ^{208}Tl by α -decay. The β -decay branch produces ^{212}Po (64%) that cannot be measured by gamma spectrometry. If ^{208}Tl measurement is to be used to estimate the thorium activity, it must be divided by 0.3594 to correct for the branching (Harb et al, 2008; Read and Hayder, 2011).

2.2.6. NEPTUNIUM SERIES (4n+1 SERIES)

The name of the series comes from the longest lived radionuclide ^{237}Np which is considered as the parent species, it has a half-life of 2.14×10^6 yr. The end product of the Np-series is ^{209}Bi , which is the only stable isotope of bismuth. Eight (8) α and 5 β -decays are required in the sequence from the parent ^{237}Np to ^{209}Bi . An important nuclide in the neptunium decay series is the ^{233}U . It is fissionable by slow neutrons (Nadia, 2014).

2.2.7. POTASSIUM RADIONUCLIDE

The average concentration of potassium in crustal rocks is about 27 g/kg and in the ocean is about 380 mg/L and plants and animals, including humans is about 1.7 g/kg (Camber, 2009). Its average activity concentration in the earth's crust is 850 Bq/kg and 65 Bq/kg in human body (bones). In healthy animals and people, ^{40}K represents the largest source of radioactivity, greater even than ^{14}C . A 70 kg person has 4400 Bq of ^{40}K and 3000 Bq of ^{14}C . It is eliminated from the body with biological half-life of 30 days; which is under strict homeostatic control (in which the amount (dose) retained

is actively regulated by the body to achieve the normal range required for system functions), and it is not influenced by environmental variations (ANLESD, 2007). Potassium-40 with half-life of 1.3 billion years decays by three general modes (β emission, K-electron capture and Positron emission). In the first mode, ^{40}K radionuclide disintegrates directly into the ground state of ^{40}Ca by the emission of β -particle of energy 1.321 MeV (88.8%) and no γ emission is associated with this type of formation (Read and Hayder, 2011). Over the second mode, ^{40}K nuclide can be transformed into stable state (ground state) of ^{40}Ar by two ways, in the first one, ^{40}K disintegrates directly with one jump into ground state of ^{40}Ar (0.16%) by electron capture. In the second way, ^{40}K nuclide can be decayed indirectly into the ground state of ^{40}Ar by two stages. Firstly, ^{40}K decays into the excited state of ^{40}Ar . Secondly, the excited nuclide ^{40}Ar , decays into ground state, accompanied by gamma radiation of 1.460 MeV energy with a probability of 11% of the ^{40}K atoms undergoing this change. In the last one (β emission), a proton will be decayed into positron and ^{40}K changed into ^{40}Ar with a probability of 0.0011% as the decay scheme shown in Figure 2.8-2 (Read and Hayder, 2011).

2.2.8. HAZARDS AND RISKS ASSOCIATED WITH NORM

Literally, hazard is defined as the potential to cause harm; therefore, radiological hazard can be defined as the potential of radiation to cause biological effects on human cell, tissue, organ or organ system. Biological effects of ionizing radiation in humans, due to physical and chemical processes, occur following the passage of radiation through their body. These processes will involve successive changes at the molecular, cellular, tissue and whole organism levels. For acute whole-body exposures above a few gray of radiation of low linear energy transfer (LET), damage occurs principally because of cell killing. This can give rise to organ and tissue damage and, in extreme cases, death. These effects, termed early or deterministic, occur principally above a threshold dose and stochastic effects has no threshold dose where the effect may or may not occur (UNSCEAR, 2008b).

The radiological impact of radiation on human's exposure to radiation has been recognized via studies that, radiation exposure above certain threshold limits can damage living cells, causing death in some of them and modifying others (UNSCEAR, 2000c). However, in the case of low doses, studies are inconclusive as to the effect

from the exposure to low background doses. It is also important to add that much of the studies on the effect of exposure to radiation have been based on information on radiation-induced cancer available from epidemiological studies of a number of human populations. These include the survivors of the atomic bombings in Japan and groups that have been exposed to external radiation or to incorporated radionuclides, either for medical reasons or occupationally. Such studies provide quantitative information on the risk of cancer at intermediate to high doses(UNSCEAR, 2000c). Consideration of possible radiation exposures is the primary method of estimating the potential health hazard associated with NORMs. The exposure to ionizing radiation due to NORMs depends on the magnitude of absorbed dose, time, dose rate and types of organ exposed due to the inhalation and ingestion of radionuclides such as ^{238}U and ^{232}Th families specifically ^{222}Rn and ^{40}K . Adverse effects and hazards, which have their origin from radiation using total radiation doses, are well known. However, risk from exposure to environmental level radiation requires an assessment of the radiological hazard following the exposure. According to the IAEA safety glossary, risk is defined as the probability of a specified health effect occurring in a person or group as a result of exposure to radiation whereas risk assessment is an assessment of the radiological risks associated with normal operation and possible accidents involving a source or practice (IAEA, 2007). Due to the stochastic nature of the adverse effects of the radiation exposure, together with their extremely low probability of occurrence, risk assessments/estimates has always been based on studies on large population groups using mathematical models. The most current of such studies are the U.S. National Academy of Sciences Committee on the Biological Effects of Ionizing Radiation(BEIR) Committee reported on the health effects of exposure to low levels of ionizing radiation. This assessment was based on a review of new scientific epidemiological studies from the survivors of the Japanese bombings, radiation accidents, radiation workers, persons who had been treated medically with radiation, populations living in high-background areas, and relevant laboratory studies on chemistry, physics and biology of ionizing radiation, and presented its conclusions in what is known as the BEIR VII Report, Health Risks From Exposure to Low Levels of Ionizing Radiation (Cember, 2009). In estimating the risk of radiogenic cancers, the BEIR VII committee found that the data supported a dose-related increase in the relative risk of a radiogenic cancer. The committee found that for all cancers except leukemia and also for the genetic effects observed in laboratory studies, the data were

compatible with linear, zero-threshold model (LNT model) (Cember, 2009; ICRP, 1993). The committee concluded, “the current scientific evidence is consistent with the hypothesis that there is a linear, no-threshold dose–response relationship between exposure to ionizing radiation and the development of cancer in humans”. The ICRP conclude, for the purposes of radiological protection, the incidence of cancer or hereditary disorders will rise in direct proportion to an increase in equivalent dose to organs and tissues below about 100 mSv (Faanu, 2011; ICRP, 2007). [Therefore, the radiological fatality cancer risks and severe hereditary effects due to exposure to NORMs were assessed from the Kenticha tantalum mine based on the total annual effective doses equivalent received by the population].

A hazard index is a parameter that is represented by a single value that takes into account the measured activity concentrations of ^{238}U , ^{232}Th and ^{40}K in the sample. The various types of radiological hazard indices are the radium equivalent activity, external and internal hazard indices, representative gamma index, radioactivity level index, dose rate, annual effective dose and total absorbed dose rate (Saher et al, 2013). The worldwide average concentrations of the radionuclides of $^{238}\text{U}/^{226}\text{Ra}$, ^{232}Th and ^{40}K are 370, 259 and 4810 Bq/kg, respectively; which produces the same gamma-dose rate (Faanu, 2011; Darko et al, 2010; Keser et al, 2013; Todsadol, 2012) in the environment. The radium equivalent concept allows a single index or number, which is a widely used hazard index to assess the radiation hazards due to exposure of gamma radiation from different mixtures of uranium, thorium and potassium in the soil samples from different locations. The values of the external and internal hazard indices must be less than 1.0 for the radiation hazard to be considered negligible i.e. the radiation exposure due to the radioactivity from the construction material is limited to 1.5 mSv/y. Also, radon and its short-lived products are hazards to the respiratory organs and as a result, the internal exposure to radon and its daughter products is quantified using the internal hazard index (Faanu, 2011; Darko et al, 2010; Keser et al, 2013; Todsadol, 2012).

2.2.9. TECHNOLOGICALLY ENHANCED NORM

There are a number of circumstances in which materials containing natural radionuclides are recovered, processed, used, or brought into position such that radiation exposures result. These human activities cause extra or enhanced exposures.

The exposures generally included in the category of enhanced exposures are those arising from the mineral processing industries and from fossil fuel combustion(UNSCEAR, 2000b).In recent decades, the development of new technologies has resulted also in the production of by-products and waste with the so-called technologically enhanced naturally occurring radioactive materials (TENORM). IAEA defined TENORM as materials containing naturally occurring radionuclides (potassium and uranium and thorium decay series)whose radioactivity has been concentrated or exposed to the accessible environment as a result of human activities, such as tantalum mineral processing (IAEA, 2006). Whereas according to US Environmental Protection Agency, TENORMs are defined as the Naturally occurring radioactive materials that have been concentrated or exposed to the accessible environment as a result of human activities such as manufacturing, mineral extraction, or water processing and “technologically enhanced” means to alter the radiological, physical and chemical properties of the NORM such that there is an increase in the potential for human and environmental exposures(EPA, 2008). Therefore, human technological activity can increase radiation exposure, not only to the person directly involved in these activities, but also to the local or even whole population(IAEA, 2006):

1. The concentrations of NORM can be enhanced above its natural levels in a product, byproduct or residue.
2. The availability for release into the biosphere of the NORM in products, byproducts or residues can be enhanced through physicochemical changes or simply due to the method by which the residues are managed. Table 2.10-1 illustrates the NORM concentrations of major radionuclides in main rock types and soil (IAEA, 2003a).

TENORM often comprises industrial wastes from thorium and uranium mining/milling; niobium, tin and gold mining; water treatment; oil and gas production; phosphate fertilizer, coal fire ash and aluminum production. All these technologically enhanced materials result in exposure to individual and group, increased environmental mobility and its contamination, improper disposition and various problems in re-use and re-cycling of wastes. In many cases, relatively low level of radiation occurs in very large TENORM areas. This situation causes problem to concerned authorities over the economic burden of disposing the waste materials

thinking that the low dose radiation could not make any problem to the environment. It is also one of the reasons why a large number of TENORM waste sites are uncovered and may be found in the many of the thousands of abandoned sites(EPA, 2008).

2.3. EXTERNAL EXPOSURE

External exposures outdoors arise from terrestrial radionuclides present at trace levels including such as tantalum ores and water bodies. The specific levels are related to the types of rock from which the soils originate. Higher radiation levels are associated with igneous rocks, such as granite, and lower levels with sedimentary rocks. There are exceptions, however, as some shale and phosphate rocks have relatively high content of radionuclides. There have been many surveys to determine the background levels of radionuclides in soils, which can in turn be related to the absorbed dose rates in air(UNSCEAR, 2000b). The latter can easily be measured directly, and these results provide an even more extensive evaluation of the background exposure levels indifferent countries. Table (2.10-1) provides information regarding the abundance of radionuclides in different natural materials (IAEA, 2003a). The radionuclides in the uranium and thorium decay chains cannot be assumed to be in radioactive equilibrium. The isotopes ^{238}U and ^{234}U are in approximate equilibrium, as they are separated by two much shorter-lived nuclides, ^{234}Th and ^{234}Pa . The decay process itself may, however, allow some dissociation of the decay radionuclide from the source material, facilitating subsequent environmental transfer. Thus, ^{234}U may be somewhat deficient relative to ^{238}U in soils and enhanced in rivers (UNSCEAR, 2000b). The radionuclide ^{226}Ra in this chain may have slightly different concentrations than ^{238}U because separation may occur between its parent uranium and because radium has greater mobility in the environment. The decay products of ^{226}Ra include the gaseous element radon, which diffuses out of the soil, reducing the exposure rate from the ^{238}U series. For the ^{232}Th series, similar considerations apply. The radionuclide ^{228}Ra has a sufficiently long half-life that may allow some separation from its parent, ^{232}Th . The gaseous element of the chain, ^{220}Rn , has a very short half-life and no long-lived decay products.

Table 2.3-1: Abundance concentrations of major radionuclides in main rock and soil (IAEA, 2003a)

Rock type	⁴⁰ K		²³² Th		²³⁸ U	
	Total K (%)	Bq/kg	ppm	Bq/kg	ppm	Bq/kg
Igneous rocks						
Basalt, crustal average	0.8	300	3–4	10–15	0.5–1	7–10
mafic	0.3–1.1	70–400	1.6, 2.7	7, 10	0.5, 0.9	7, 10
Salic	4.5	1100–1500	16, 20	60, 83	3.9, 4.7	50, 60
Granite, crustal average	>4	>1000	17	70	3	40
Sedimentary rocks						
Shale, sandstones	2.7	800	12	50	3.7	40
Clean quartz	<1	<300	<2	<8	<1	<10
Dirty quartz	2?	400?	3–6?	10–25?	2–3?	40?
Arkose	2–3	600–900	2?	<8	1–2?	10–25?
Beach sands (unconsolidated)	<1	<300?	6	25	3	40
Carbonate rocks	0.3	70	2	8	2	25
Continental upper crust (ave.)	2.8	850	10.7	44	2.8	36
Soils	1.5	400	9	37	1–8	66
Note: Question marks indicate estimates in the absence of measured values						

2.3.1 GAMMA SPECTROMETRY ANALYTICAL TECHNIQUES

There are many different types of radiation detectors such as filled detectors (ionization chamber counters, proportional counters, and Geiger-Muller counters), scintillation or semiconductor detectors. All types of radiation detection instruments however basically involve generating electrical signals which result from the interaction of radiation with the detector's active volume (sensitive to ionizing radiation) (Cember, 2009; Knoll, 1999). Among many types of detectors, the

experimental work presented in this thesis project used HPGe detector for the analysis of gamma emitting naturally occurring radionuclides as described in the following sections.

CHAPTER THREE

1.1.MATERIALS AND METHODS

The findings of the current research were born through an organized and systematic methodology followed. Broadly, four major activities briefed hereunder were addressed to fulfill the preset objectives of the research:

- ✓ Literature review
- ✓ Field work
- ✓ Laboratory tests on collected samples
- ✓ Data analysis and Interpretation of results
- ✓ Conclusion and recommendation

The literature survey has encompassed both published and unpublished reports of investigations, case studies, text books and Journals that were obtained from different sources for better understanding of aspects related to the current research topic such as: building the framework concepts on radioactive waste controlling mechanism, available standard specifications, integration of various adopted methodologies on international atomic energy agency studies and collection of secondary data. The fieldwork has involved four major activities: Collection of representative samples for desired laboratory tests, insitu tests for radiation dose and surface contamination test. Laboratory tests, on collected samples, for permissible naturally occurring radioactive material in the natural environment the lab have been carried out in Ethiopian radiation protection authority. The British Standard (BS) and American Society for Testing and Material (ASTM) and BSS of IAEA standards were used for the testing procedures and analysis of radionuclides. The present research has made use of both secondary and primary data for analysis and interpretation. Existing data for the currently targeted nuclear radiation leakage that was made for use by ERPA. Besides, primary data generated from laboratory test of collected samples and from insitu, tests during the current research were used. Mostly in this research, I used both Destructive and non-destructive test and the material used during the research are: The analysis of data and interpretation of results has followed several integrated radioactive waste and source assessment techniques provided by IAEA researchers. Besides, correlation was made for results from the adopted techniques themselves and with the actual condition of the record and insitu model experimental result observations and environmental

indicators (uniformitarian's approach). Software's such as, raise, rad calculator global radiation map, google earth, and, Microsoft offices (word and excel) were used as an assisting tool in this research. The norm laboratory test (natural occurring radioactive material) has been tested by sophisticated gamma spec and gamma cell. Which can identify the radionuclide in the norm, and it is a new technology, which is capable to analysis the concentration of radionuclide, its peak energy and detectable nuclides. Based on the type of research, the case study is the most suitable strategy for my master thesis. Case studies do not possess control over the behavioral events and focuses on contemporary events (Yin 2009). Moreover, case studies rely on different sources of evidence. The aim of the research is not to describe or explain a situation or setting, but rather to discover "*How to improve...*" something. Therefore, an exploratory approach is needed. The unit of analysis is a supply chain, but the focus will be one echelon, which is the waste management company. I think that a single case study with multiple units of analysis is appropriate. The data collection can be divided into primary and secondary. The main difference between these is that primary data is being founded and collected by the researcher (data that is not available), while secondary data is available data collected by third persons (hard and soft data). Primary and secondary data can once more be divided in two categories, internal and external. The secondary data was collected throughout scientific journals, local and international documents referring to official regulations involved in waste management, radiation related topics. Moreover, this thesis makes use of several recommended guidelines from public and private institutions related to operations in the waste management process for NORM and disused radioactive wastes originated from nuclear medicine, agriculture construction and mineral processing facilities. Other sources are Master thesis, Ph.D. dissertation and industry statistics published by the regulating authorities.



Fig 3.1.1 Gamma spectrometry typical destructive test instrument.



Fig 3.1.2 ERPA rad-lab set up

3.1.1 KENTICHA TANTALUM MINE

The Kenticha tantalum mine in Ethiopia is considered one of the largest world-class tantalum producing assets in Eastern Africa. The mineral is currently experiencing global boom. A state-owned company, Ethiopia Mineral Development Share Company (EMDSC) since 1990 in the Adola Greenstone Belt, operates this mine. A deposit in weathered crust was delineated in 1988 with proven reserves of 25,850 tons of Colombo–tantalite ore at 0.02 to 0.03% tantalum (SEAMIC, 2006). The small-scale open pit mine has started with a pilot plant producing about 60 tons per year. The deposit is both a weathered crust ore (the top 60 meters) with proven reserve of 2400 tons of tantalum pentoxide and 2300 tons of niobium pentoxide, and primary ore with proven reserve of 2393 ton Ta₂O₅ and 2362.5 ton Nb₂O₅. At present it is producing over 190 tons of tantalite concentrate of tantalite-colombite ore per annum (MME, 2009).

Kenticha tantalum mine is selected for the study for the following reasons:

1. Kenticha Ta-Nb ore is known to host a number of elements such as rare earth elements including U, Th, K, Nd, Eu and others (like Li, Be, beryl, sulphides, etc.) (MME, 2010).
2. The bulk of the oddo shakiso population is distributed around Kenticha tantalum mine.
3. According to ERPA report, the radium-226 and its daughter, uranium-238 and its daughter and thorium-232 and its daughters in the tantalum ore, solid iron waste, tailing dam samples was found with high activity concentration and radiation doses higher than that of the background radiation measured at that site (ERPA, 2013) and this agreed with Ministry of mine of Ethiopia report which said that the uranium content of tantalum concentrate is high (MME, 2014).

Figure 3.1-2 and Figure 3.1-3 illustrates the parts of the study area; and location of Ethiopia. Kenticha tantalum mine in southern Ethiopia and the distribution of sampling points; Appendix I also described sampling co-ordinates for ore, soil, solid waste, tailings and water samples.



Figure 1.1-1: Topography of Kenticha tantalum open pit mining site (MME, 2010)

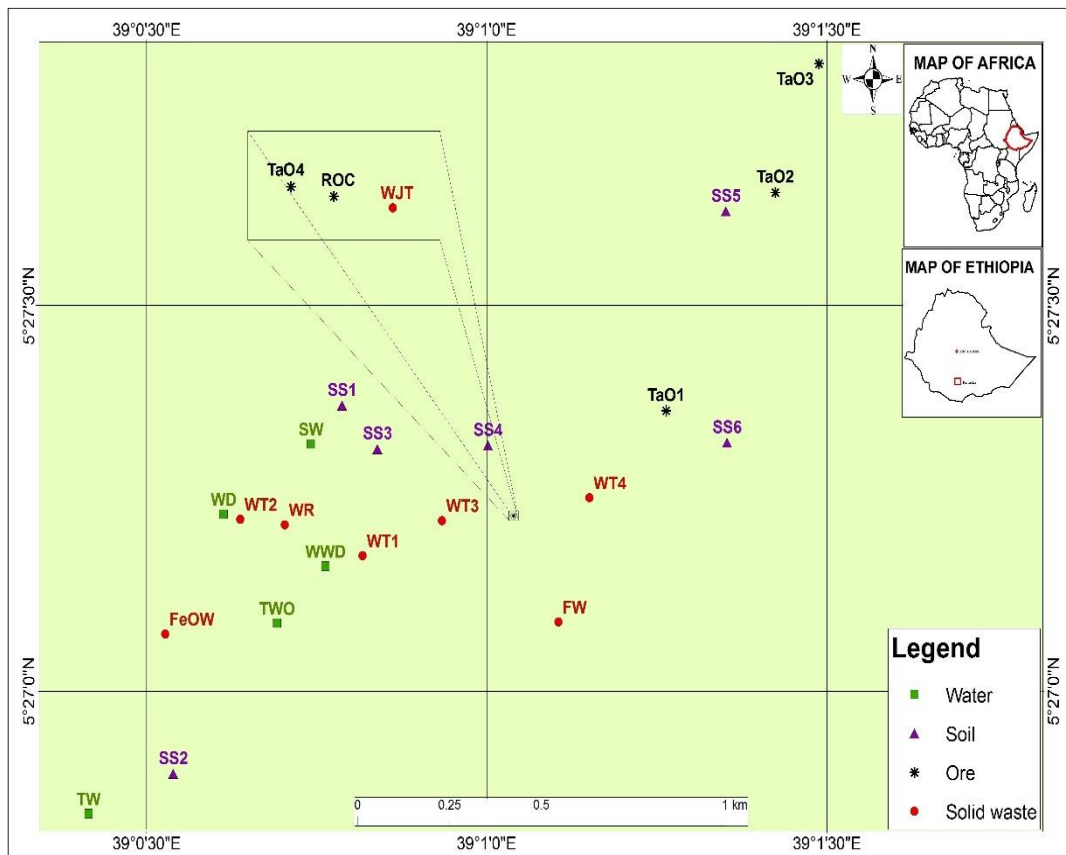


Figure 1.1-2: Outline of Kenticha tantalum mine showing sampling points

3.1.2 HYDROLOGY OF THE STUDY AREA

The climate of Kenticha area is mainly characterized by an arid and semi-arid climate (Zewdie, 2011) in which the rainy season starts from March to May and from October to November. This rainfall season is dependent on the wind that originates from the Indian Ocean. The main sources of water are borehole, cold spring, hot spring, river water and rainwater. Drainage is well developed and mainly gets water from rain. The Marmora River is the largest perennial river in the region about 8km west of the Kenticha tantalum deposit (source).

3.1.3 MATERIALS

Some materials and equipment used to effectively carry out this research are described below. Generally, the materials and equipment required in this study were plastic bags, 1.5L plastic bottles, 0.4 μm filter paper, 1M HNO_3 , lab coat, goggles, gloves, 1 liter Marinelli beakers, analytical balance, sample drying trays, sample grinder, sample drying oven and wire mesh sieve. Moreover, a gamma spectroscopy system that comprised of Genie 2000 software, a High Purity Germanium Detector (HPGe) and multichannel analyzer (MCA) was used for counting the samples. In addition, global positioning system device (GPS), statistical package for social scientists (SPSS) and Microsoft excel software were also very important for this research. A solid water standard of mixed radionuclides containing ^{241}Am , ^{109}Cd , ^{139}Ce , ^{57}Co , ^{60}Co , ^{137}Cs , ^{113}Sn and ^{88}Y in identical geometrical configurations to the sample containers were used for calibrating the HPGe detectors as well.

3.1.4. Overview of sampling and sample preparation

The experimental framework of the sample preparation for this research work to packing of the samples for secular equilibrium is summarized in Figure 3.2-1.

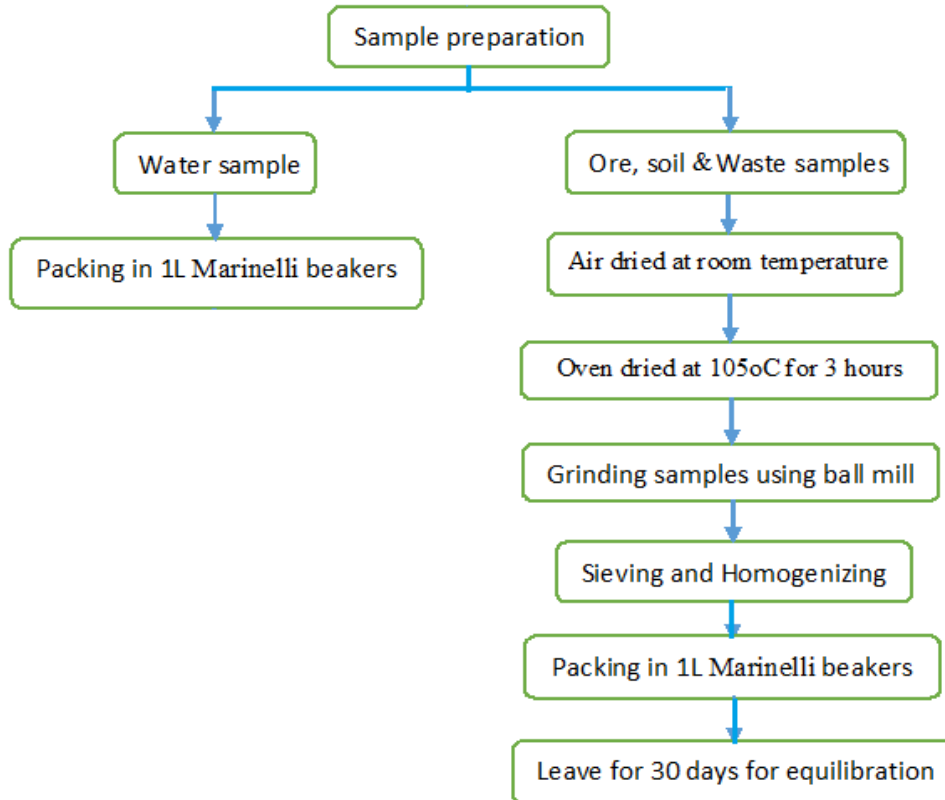


Figure 1.1-3: Flow chart for sample preparation and packing

3.1.5 SAMPLING TECHNIQUES

Initially, sampling area survey was carried out using GPS prior to sampling in the area to determine the sampling points. Sampling were considered sample representativeness using random pattern sampling provided that sampling is statistically valid (Barbara, n.d.). Tantalum ore, soil, solid waste and water samples were collected from tailing dam, waste water dumps, pure water dam, ore stockpiles, open pits, processing plant site and soil samples at its normal working condition of the mine. Sampling included outside the main mine area such as the water samples from Mormora River found 8 km away from the mine. At each sampling location, 1kg of tantalum ore, solid waste and soil samples were collected from undisturbed surface to a depth of 3m with a coring tool into clearly labelled plastic bags (Barbara, n.d.; IAEA, 1989). In general, about three cores were taken and composed to make a single sample. After each sampling all vegetation and debris were removed. Sampling were done using clean polyethylene bottle for water sampling and plastic bag for ore, soil and solid waste sample, which were packed tightly and properly to prevent exchange of

particles and contamination during transport. Samples with relatively high levels of activity were kept separated from other samples to avoid cross contamination.



Figure 3.1.5-1: Kenticha ore pit tantalum-mining site

The water samples were also collected into clean-labelled 1.5L polyethylene bottles from the Marmora Rivers, tap water and spring water. Each sample were immediately spiked (acidified) with 1M HNO₃ to prevent sorption on surface of the bottle. Then, the bottle was covered with lids and sealed. At GAEC Laboratory, all water samples were stored in a refrigerator prior to preparation and analysis.

The sample collection bottle was washed with detergent to remove grease, then with water following further washing with 10% HNO₃ and rinsing thoroughly with distilled water. When sample from the river water was taken, the sample was taken at a point where total mixing has occurred. Particulate phases were removed or separated at sampling site. Finally, all samples were transported to the GAEC radiation protection laboratory for preparation and analysis.



Figure 3.1.5-2: tailing dam of tantalum ore washing site.

3.1.6 SAMPLE PREPARATION

At GAEC laboratory, all ore, soil and solid waste samples were spread onto trays or plastic sheets, and allowed to dry at room temperature for several days and then oven dried at 105°C for 3 hours until the samples had constant weight. Composite samples of tantalum ore, soil and solid waste were prepared prior to grinding due to the required amount for HPGe Detector analysis. Then, the samples were grounded into fine powder using a ball mill and sieved through wire mesh. The weight of clean empty one liter (1L) geometric beakers were recorded using weighing balance (METTLER TOLEDO, Model: XP2001S, SNR: B044082756, mass range: 0.1g to 2100g, d = 0.1, power requirements: 12V~2.25A, Made in Switzerland) and then the homogenized grounded samples were put into the Marinelli beakers. Then, the beakers were covered and sealed with lids and a paper tape to prevent the escape of the gaseous radionuclides from the sample (i.e. radon), as shown Figure 3.4-1. Each beaker was then weighed again to determine the weight of the sample together with the beaker. After 2 hours sample preparation, each sample was counted on HPGe detector for 7200 seconds and kept for a period of about 30 days to allow radioactive equilibrium to be reached between the long-lived parent and their corresponding short-lived daughter radionuclides (Malain, 2007; Faanu, 2011; Abdullahi et al, 2014). Then after 30 days, the samples were counted on a high purity germanium (HPGe) detector for 36,000

seconds. Finally, the activity concentrations of the radionuclides in tantalum ore, soil and solid waste samples were reported on dry weight basis in Bq/kg.



Figure 3.1.6-1: Sample filled in a geometrical beaker and sealed

Moreover, the 1L water samples were filled into weighed geometrical beakers to its one-liter (1L) mark. Then, the beakers were tightly sealed with lids and paper tape as shown in Figure 3.4-2, then recorded the volume of water samples and left for 30 days for equilibration. Finally, the beakers with the samples were counted on HPGe detector for 36,000 seconds. Then, the activity concentrations of the radionuclides in the sample were recorded in Bq/L (Faanu, 2011; IAEA, 1989).



Figure 3.1.6-2: soil sample filled in a geometrical beaker

3.1.7 DESCRIPTION OF THE HYPER PURE GERMANIUM DETECTOR

In this research, analyses of the samples (tantalum ore, solid waste, soil and water) were performed with a computer-based gamma-spectrometry system for qualitative and quantitative determination of gamma-emitting radionuclides, NORMs (see Figure 3.6-1) for the summary of sample analysis flow chart). The High Purity Germanium (HPGe) detector is coupled to a Multi-Channel Analyzer (MCA) to measure activity concentrations of the radionuclides in the samples. A number of discrete gamma-ray energy peaks in each sample spectrum reveal the presence of several radionuclides in the samples and the peak area under each spectrum reveal the quantity of the radionuclides in that sample (see appendix II). Using Genie 2000 software associated with the detector, the gamma ray spectra were analyzed.

The detector was cooled with liquid nitrogen (cryostat) to temperature of -196.25°C to reduce noise and for good resolution. The relative efficiency of the detector was 45% with energy resolution of 0.66 keV at gamma ray energy of 136.409 keV of cobalt - 57. Detector specifications and performance data is shown in Appendix IV.

To determine background radiation around the detector, six cleaned empty 1L Marinelli beakers filled with distilled water were counted for 7,200 and 36,000 seconds in the same geometry as the samples. Then, the background average count obtained under each peak were used to correct the net peak area of gamma rays of measured isotopes and to determine the minimum detectable activities of ^{238}U , ^{232}Th , and ^{40}K .

3.1.8 ENERGY CALIBRATION

Before any actual counting of the samples, the HPGe detector was calibrated with respect to energy and efficiency. The objective of energy calibration is to identify radionuclides in the sample by deriving a relationship between spectrum peak position and the corresponding gamma-ray energy. Energy calibration is performed by measuring the spectrum of a standard source emitting γ -rays of precisely known energy and comparing the measured peak position with their energy. In this study, the energy calibration was carried out by counting standard radionuclides (a mixture of ^{241}Am , ^{109}Cd , ^{139}Ce , ^{57}Co , ^{60}Co , ^{137}Cs , ^{113}Sn and ^{88}Y) of known activities emitting gamma energy in the energy range from 60keV – 2MeV. The radionuclides and their

activities in the standard are shown in Appendix III. The standard was counted for 36000 seconds. A good multi-channel analyzer has a linear response in its full dynamic range. That is, the bins of the MCA are directly proportional to the energy absorbed in the gamma spectrometry detector. This implies that, if one plots the absorbed energy against the channel number of the MCA, the result would be linear (Ahmed, 2007) as shown in Figure 4.1-1, which is an indication that the system is operating properly (Faanu, 2011; IAEA, 1989).

3.1.9 MINIMUM DETECTABLE ACTIVITY (MDA)

The minimum detectable activity (MDA) is defined as the smallest quantity of radioactivity that could be measured under specified conditions; i.e. the minimum concentration level of radioactivity that can be determined to be statistically significant from an analytical blank, and includes sample holder and everything else that may be counted with an actual sample (Faanu, 2011; Moussa and Mounia, n.d.; Todsadol, 2012); to ensure that there is a 95% probability of the true net counts. For a low-level counting particularly in environmental level systems, MDA is significant above which counts are statistically significant of the measurement (Faanu, 2011). For this research, only Marinelli beaker filled with distilled water was counted for 10 hours (36,000 seconds) such that the average background peaks were used to determine the MDA. The MDA of ^{238}U was estimated from the average activity concentration of ^{214}Pb at gamma peaks of [351.92keV (35.6%)] and ^{214}Bi [609.31keV (45.5%) and 1764.49keV (15.3%)], while ^{232}Th was estimated from peaks of ^{212}Pb [238.63keV (43.6%)] and ^{208}Tl [583.19keV (85%)]. Moreover, the ^{40}K photo peak energy [1460.83keV (10.7%)] was used for computing its MDA. The minimum detectable activity depends on the gamma emission probability (P_γ) and absolute efficiency (E_f) of the detector. The mathematical expression of MDA is given by equation (3.5-3):

$$MDA = \frac{1.645\sqrt{N_B}}{\varepsilon_f P_\gamma T C m} \quad (3.5-3)$$

Where,

- N_B is the background counts in the region of interest for a particular radionuclide,
- ε_f is the photo peak efficiency,
- P_γ is gamma emission probability,

- T_C is the counting time and
- m is the weight of the sample container. The unit of MDA is Becquerel per kilogram (Bq/kg).



Fig 3.4.3 Weathered tantalum ore body (white) underlying talc-tremolite schists and serpentines. The quartz core is standing out



Fig 3.4.4 limestone raw material mining of Derban cement factory

CHAPTER FOUR

CHARACTERIZATION OF RADIOACTIVE WASTE

4.1. Preamble

The radioactive material assessment in the present research is more specific to evaluation of radioactive waste management code of practice and code of conduct compatibility (Ethiopian rad waste management with the international atomic energy code of practice). For the waste of both NORM and Spent disused radioactive material, characterization, disposal option, categorization has been evaluated by integrated waste management assessment techniques in conjunction with cross-validation of actual conditions. This chapter forwards details of the basic considerations, techniques, input data types and sources utilized in the evaluation of disused radioactive waste.

4.1.1 Radioactive waste;

A fundamental element at all stages of waste management, from waste arising to disposal, is the characterization and classification of waste. Classification can be used for different purposes, such as identifying the potential hazards associated with the particular types of waste, designation of waste to a particular processing, storage or disposal option and planning and designing waste management facilities. Classification enables the selection of the most appropriate waste management option.

4.1.2 Safety Culture

Efforts should be directed to ensuring that an appropriate safety culture exists in all the organizations involved with the generation and management of radioactive waste and its regulatory control. This should be aimed at creating a necessary awareness of the need for proper management of radioactive waste at all levels within the various organizations and should discourage complacency in any aspect of related operations.

4.1.3 Radiation Protection

A radiation protection program should be in place that adequately ensures radiation safety and control of the access to areas where radioactive wastes are or will be managed. Appropriate radiation monitoring of the areas should be carried out and of

material being removed from controlled areas. Radiation dosimetry should be provided to the workers who could be occupationally exposed to radiation.

4.1.4. Environmental Monitoring

Environmental monitoring will be a regulatory requirement for any large waste management facility, but may also be required for smaller facilities depending on the activity of radioactive waste handled and/or discharged. The need for monitoring should be closely linked to the possibility of significant radiation doses to the general public being received.

4.1.5. Physical Protection

Physical security arrangements at the facility should be in place to ensure that radioactive wastes are not accidentally or deliberately removed from their proper location without authorization. Particular attention should be given to materials or equipment, which may have some intrinsic value.

4.1.6 PROCESSING

Processing of radioactive waste can involve a number of operations that change the characteristics of the waste and involves pretreatment, treatment and conditioning steps. Processing may be required for safety, technical and financial reasons. From a safety perspective, processing is required to eliminate or reduce associated hazards (e.g. radiological, physical, chemical, biological). Waste should only be processed after its precise characterization. The processing method selected should ensure that the waste would meet the acceptance requirements for storage, disposal and transport, or release from regulatory control as approved by the regulatory body. The methods for processing should be selected based on the waste characteristics. Radioactive waste generated from small-scale use of radioactive materials can generally be categorized into the following main groups: solid waste, liquid waste and gaseous waste. The wastes in the groups may consist of waste with radionuclides which are differentiated by activity (alpha, beta/gamma emitters), half-life, and by their physical, chemical and biological properties. The generation of secondary radioactive waste should always be taken into account when selecting a processing method. The implications of secondary waste arising should be taken into account in the safety and environmental impact assessment. This is of particular concern with operations such as decontamination, dismantling, sawing and cutting, shredding and crushing of solid waste for volume reduction purposes. These operations are often associated with decommissioning of facilities.

4.2. Basic Consideration

The present radioactive waste assessment research has made its basis on considering the NORM concentration spent radioactive deposits and in use radioactive material. So that the basic analysis and interpretation techniques followed for the assessment are well known for radioactive substance.

Engineering behavior similarity and existing constraints for radioactivity are taken while correlating the norm in sedimentary terrain with other igneous and metamorphic terrain some of the supporting arguments on this regard are listed here under:

- The insitu ionizing radiation dose of the different rock progeny
- Most of the raw material for cement industry deposits in the study area are highly dispersive so non-destructive test method was appropriate.
- According to Pettijohn (1984), pyroclastic debris deposits such as pumice, gypsum limestone and volcanic breccia are considered as “volcano-classitic sediments” composed of that could range from Bombs ($\geq 64\text{mm}$) to fine ash ($\leq 0.25\text{mm}$), the radionuclide concentration detrimentally very high as compared to other source rocks. The number of nuclei in a radioactive sample that disintegrate during a given time interval decreases exponentially with time of exposure.

4.2.1. Evaluation Techniques

The evaluation approach followed during the present research broadly can be classified into two categories;

- Analysis of data and interpretation of results applying integrated radioactive waste management assessment techniques
- Cross-validation of findings of approach- (i) with actual condition indications; obtained from IAEA records and insitu norm measurement and insitu dose leakage measurement during the present research.

Three well-known and efficient preliminary radioactive assessment techniques are adopted for this research:

- The “activity Ratios” $R = \frac{\text{Activity}}{D \text{ Value}}$, suggested as a criteria to evaluate the waste categorization of spent and in use radioactive material/waste.
- The term ‘radioactive waste’ covers a wide variety of material, ranging from wastes that can be put safely into a dustbin, to items that need remote handling,

heavy shielding and cooling to be managed safely. Radioactive waste is considered in different categories, as follows: (Mitchell, 1981; U.S. ACE, 1984; Verfel, 1989) is adopted. Generally, radioactive material/waste is considered as high level, intermediate and low level. Low-level radioactive waste has a radioactive content not exceeding 4 giga-becquerels per/ton (GBq/tn) of alpha activity or 12GBq/tn beta-gamma activity and is suitable for disposal in near surface engineered facilities.

- High-level radioactive waste is radioactive enough for the decay heat to increase significantly its temperature and the temperature of its surroundings. This means that heat generation has to be taken into account when designing storage and disposal facilities.
- Intermediate-level radioactive waste is more radioactive than low-level radioactive waste, but does not generate enough heat to require this to be taken into account of in storage or disposal facilities. However, like other radioactive waste it still needs to be contained to protect people and the environment
- Higher Activity Radioactive Waste (HAW) Higher activity radioactive waste is a broad term, which includes all HLW and ILW, as well as certain wastes categorized as LLW but which are not currently suitable for disposal in existing LLW facilities. The evaluation has been done based on the activity concentration of radionuclides i.e. its evaluated based on radionuclide peak energy peak energy
- Comparing the maximum seismic magnitude of Ethiopia seismic design with average effective peak ground acceleration of the disposal site selection for radioactive waste as per EBC (2000), Mitchell (1970), Karol (1985) and Tjandrajana (1989) limiting the disposal site is the major work for designing Radioactive waste disposal where D_{100} is the sieve size for which all the grout particles are finer. For the present research groundmass of quarry blasting was estimated based on Littlejohn (1985) relation; $\frac{10.3m}{s} = s/\sqrt{w}$ where, S= effective distance travelled by seismic wave considering isotropic site or homogeneous site effect of induced seismicity of uncontrolled quarry blasting. For this research, three suspension cement types are considered treatment (conditioning); normal Portland cement (Ethiopian Muger Ordinary Portland Cement (OPC), Muger Portland Pozzolan Cement (PPC), and ultrafine cement (MC-500) concrete.

In the case of approach (ii), existing records of ongoing foundation grouts, and insitu permeation test was attempted using sodium silicate grout and later was analyzed and interpreted. Existing records are thoroughly examined for the type of grout used, occurrence of ‘grout filtration’, applied grouting pressure sufficiency and any related ‘hydro-fracturing. Analysis and interpretation of insitu permeation test results emphasized on radius of influence grout penetration, improvement in engineering performance of the material, and the magnitude of injection pressure.

4.2.2. Data types and sources

Required and relevant data characterizing the radioactive waste, medium and grout component considered in analysis were obtained from laboratory and insitu tests, and reviewing literatures or other secondary sources. Grain size, porosity and Permeability values characterizing the grouting medium are among the basic input data used for the analysis in the present research. Besides, the grain size distribution of the major components of three suspension grout types (normal Portland cement, ultra-fine cement, and bentonite/clay) was adopted. Moreover, the review of records of previously executed radioactive waste inventory data in the area has contributed valuable information used for the fortification of the analysis and interpretation for the present research.

4.2.3. Laboratory and insitu tests

Radionuclide identification half and mean life of radioactive element, dose and activity concentration and radiation leakage, were determined from insitu and laboratory tests conducted during the present research.



Figure 4.1.1 representative sample of cement raw material

Representative samples, of the industrial raw material such as limestone, gypsum, clay, coal, tantalite ore distribution were analyzed. The Laboratory testing were carried

out in Ethiopian radiation protection authority and nuclear technology agency in Addis Ababa. Area dose rate assessment was done at the Raw material stockpiles, at different parts of the fabrication steps/processes and product storages. The safety assessment included measurements of area dose rate for both gamma (γ) in $\mu\text{Sv/h}$, alpha (α) and beta (β) particles in **Becquerel per centimeter square** (Bq/cm^2) and collection of soil and Rock samples and information about the day to day work environment management in general.



Figure 4.1.3 In situ density test of concrete pre-cast for conditioning (treatment).

An insitu permeation-grouting test, using sodium silicate grout, was attempted on a block sample prepared with dimensions of 60cm depth x 60cm width x 50 cm Length. Environs of the block were backfilled with granular material with the view to monitor any possible leakage. A 50 cm deep and 60 mm wide test section was bored using a hand operated auger at the center of the above mentioned test block. The test initially was planned and modelled to be done with the application of safe grouting pressure. However, during the actual test the chemical grout was observed to permeate by gravity only and no expected resistance was offered by the test section, thus no external pump pressure was applied.

4.2.4. Spent Fuel Storage

Spent Fuel is the fuel that no longer has the ability to undergo nuclear activity. The storage method adopted for spent fuel and other radioactive materials in the nuclear site ensures minimal exposure to on site operators as well as the public. Initially, this spent fuel is stored in pools in the reactor site. The storage of spent fuel requires a lot of facilities like 1: shielding to handle the fuel, 2: cooling to maintain fuel temperature, 3: criticality safety control, 4: controlling water chemistry, 5: decontamination of these, maintaining fuel temperature and cooling it is the most important of them all.

There are two commonly accepted types of fuel storage; Pool storage method and the Dry Storage method. The two methods are briefly discussed below; Pool Storage: Storing spent fuel assemblies under at least 20 feet of water. It provides adequate shielding from the radiation. The assemblies are moved into the water pools from the reactor along the bottom of water canals, so that the spent fuel is always shielded to protect workers. About one-fourth to one-third of the total fuel load from the pools is spent and removed from the reactor every 12 to 18 months and replaced with fresh fuel and sent to radioactive storage or permanent deep well injection.

- **Dry Storage:** For increasing spent fuel storage capacity, utilities began looking at options such as dry cask storage as the need for alternative storage began to grow when pools at many nuclear reactors began to fill up. Spent fuel that has already been cooled in the pool for at least one year, is stored in a container called a cask. The cask contains an inert gas that surrounds the fuel. The casks are typically cylinders made up of steel that are either welded or bolted closed. The steel cylinder provides a leak-tight confinement of the spent fuel. To provide radiation shielding to workers and the public, each cylinder is surrounded by additional steel, concrete, or other material. Some of the cask designs can also be used for safe transportation

There are specific features that characterize the safety of each type of spent fuel storage. The following factors define the main safety requirements:

- Minimizing the danger of hermetic sealing damage (leading to release of radioactivity by the fuel)
- Minimizing the release of radioactivity and its migration to the environment in case of fuel-element cladding damage

Minimizing the mechanical damage of spent nuclear fuel elements as a result of external impacts (earthquakes, fires, explosions, projectiles, fall of flying objects).

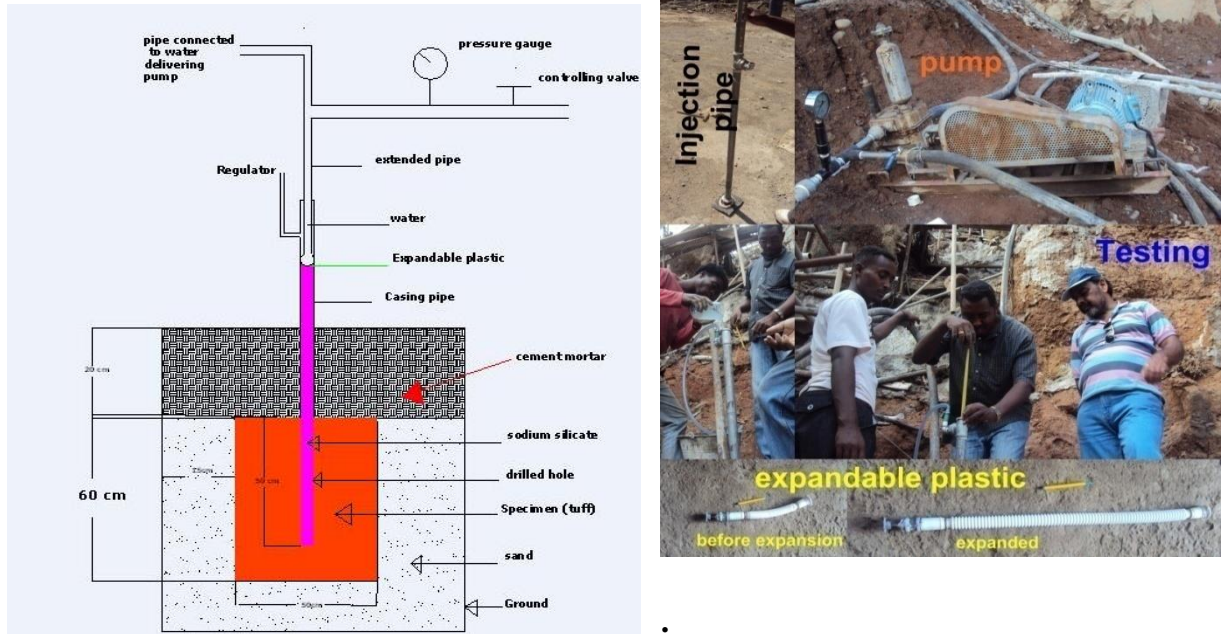


Figure 4.1.4 In situ permeation (low-pressure) test of geological radioactive disposal

Next observations, each for 5 minutes run, were recorded by falling head technique after reasonable time delay of 10, 20 and 30 minutes. Injection was continuous for 2 hrs. and 20 minutes. Besides, 3 days after the injection, sample was recovered and observed for the radius of influence of penetration, improvement in strength and for leakage conditions of radiation if any.

4.2.5 Grain size distribution of suspension grout components

The research has considered three types of particulate/suspension grouts (normal Portland cement, Micro fine cement and betonies grouts) while analyzing radioactive treatment and conditioning.

The present technique followed for the permeation groutability analysis inquires the grain size distribution of each particulate grout considered. Accordingly, representative gradation data for each major grout component were procured from available sources. The normal Portland cements (OPC and PPC), grain size data was obtained from local cement factory, Ethiopian Mughher Cement Factory Enterprise,

and ten independent batch representative grain size analysis records were plotted (Fig. 5.1.5) and their average curve have been considered for the purpose of analysis. The average grain size distribution for micro fine cement and bentonite were adapted from IS-Tokyo'96. The 85% finer (D85) and the maximum grain size diameter (D100) of each type of grout component aforementioned were utilized while analyzing permeation groutability of the tuff and volcanic breccia on the basis of the groutability ratio (N), and comparing void size with maximum grain of grout particle in suspension, respectively.

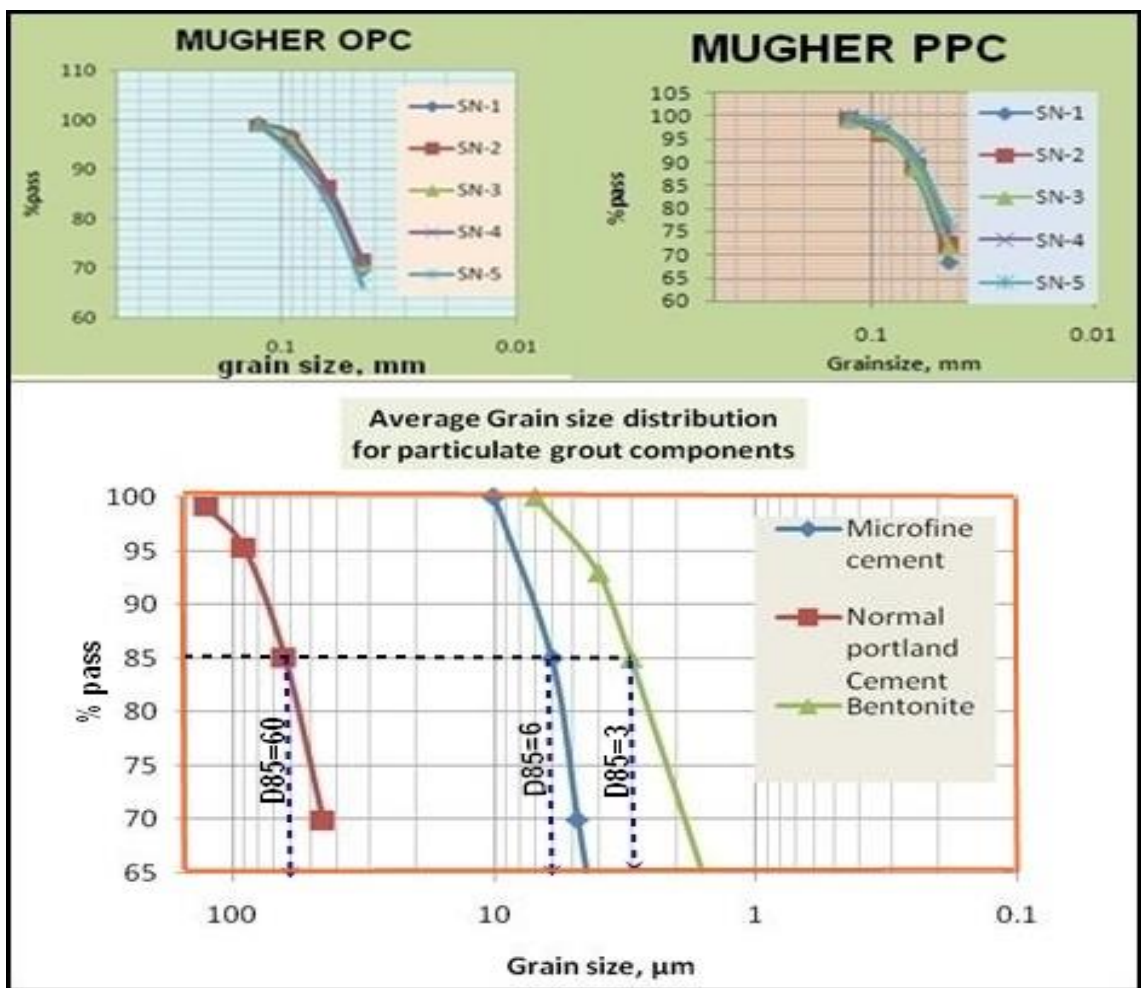


Figure 4.1.1 Grain size distribution of suspension grout components

Represents the materials that were used in conditioning of Cs-137. It is observed that the good sand can be used in Cs-137 cementation was sand. This type of sand is considered as most effective in radiation attenuation as other types. The lower dose was $5\mu\text{Sv/h}$ while the maximum was $9(\mu\text{Sv/h})$. This indicates that C-15 is the best one. In addition, it is shown that the C-10 Concrete is best concrete in radiation attenuation

as dose was 3($\mu\text{Sv/h}$ the lowest one in concrete material. Mixture of sand and concrete were good than other in Cs-137 conditioning. The results showed that the obtained values were within the acceptable level for most of the samples. It's within the permissible limits of the radiation level adopted by the IAEA of 10pSv/h.20.

Table 4.3; Concrete Mix Design to disposal of radioactive waste

material	Dose $\mu\text{sv/hr.}$
C-5 concrete mix design	3-5 $\mu\text{sv/hr.}$, average 4 $\mu\text{sv/hr.}$
C-10 concrete mix design	5-9 $\mu\text{sv/hr.}$, average 7 $\mu\text{sv/hr.}$
C-15 concrete mix design	9-13 $\mu\text{sv/hr.}$, average 11 $\mu\text{sv/hr.}$
C-20 concrete mix design	13-17 $\mu\text{sv/hr.}$ average 15 $\mu\text{sv/hr.}$
C-30 concrete mix design	17-25 $\mu\text{sv/hr.}$ average 21 $\mu\text{sv/hr.}$
C-40 concrete mix design	25-35 $\mu\text{sv/hr.}$ average 30 $\mu\text{sv/hr.}$
C-50 concrete mix design	≥ 35 $\mu\text{sv/hr.}$

According to the IAEA principle it's enough to use C-5 -C-15 for such type of waste, thus the maximum radiation release is around 5 $\mu\text{sv/hr.}$ For critical cases such as the Akaki steel factory max reading before conditioning 140 $\mu\text{sv/hr.}$ and after conditioning 4 $\mu\text{sv/hr.}$ Cs -137 orphan source. So conditioning is the best option to treat radioactive waste. Table 4.7 orphan source NDG (disposed waste near Akaki river) radiation leakage and radioactive release has been detected using portable gamma spec. Shallow ground disposal without effective engineered barriers is nowadays allowed only for LLW. A modern shallow ground disposal facility consists of monolithic vaults made of reinforced concrete; isolated from the geological medium by means of a clay layer or similar. The waste packages are transferred into the vaults with a crane Spaces between packages are backfilled with cement grout. Finally, the top cover of concrete and other protective layers above the vault are made. However, when we see our country Ethiopia, shallow ground disposal has been done without considering these phenomenological criteria.

Table 4.1.7 orphan source of radioactive material

Radiation type	Source Cs -137	Before geological deposal (before shielding)	After deposal (after shielding)
γ ($\mu\text{sv/hr.}$)	Source Cs -137	140 γ ($\mu\text{sv/hr.}$)	3 γ ($\mu\text{sv/hr.}$)
γ ($\mu\text{sv/hr.}$)	Source Cs -137	@ surface	@4m depth

The reading indicates that the radiation released to the environment was beyond the ALARA principle, which is beyond the threshold limits of the environmental and occupational exposure; hence, it has been disposed near Akaki River without geotechnical investigation of the site effect. Akaki river has shallow ground water aquifer system, which is susceptible to radioactive radiation leakage, despite the fact that, the akaki area is located in the main Ethiopian rift system, which needs special attention of earth's dynamism such as ductile and brittle shear zone. In such condition, geological disposal is not advisable but in Ethiopia, we lack expertise in regard to radioactive waste Disposal technology. The principal approaches to the predisposal management of radioactive waste are commonly termed 'delay and decay', 'concentrate and contain' and 'dilute and disperse'. 'Delay and decay' involves holding the waste in storage. Until the desired reduction in activity has occurred through radioactive decay of the radionuclides contained in the waste. 'Concentrate and contain' means reduction of volume and confinement of the radionuclide content by means of a conditioning process to prevent or substantially reduce dispersion in the environment. 'Dilute and disperse' means discharging effluent to the environment in such a way that environmental conditions and processes ensure that the concentrations of the radionuclides are reduced to such levels in the Environment that the radiological impacts of the released material are acceptable. The approaches 'delay and decay' and 'concentrate and contain' often involve the holding of waste in a storage facility or the emplacement of waste in a disposal facility. Radioactive waste therefore has to be processed, as necessary, in such a way that it can be safely placed and held in a storage facility or a disposal facility. The approach 'dilute and disperse' is a legitimate practice in the management of radioactive waste, but only when carried out within authorized limitations established by the regulatory body. Various factors, including

the nature and the amount of radioactive waste, occupational and public exposures, environmental effects, and human health, safety, and social and economic factors, are to be considered when deciding between options in the predisposal management of radioactive waste. However, the preferred option, as far as is reasonably practicable, is to concentrate and contain the waste and to isolate it from the biosphere. In the predisposal management of radioactive waste, decisions often have to be made at a time when no disposal facility is available and the waste acceptance criteria for disposal are unknown. A similar situation would arise if radioactive waste were to be stored over long periods of time for reasons of safety or for other reasons. In both cases, consideration has to be given to whether, for the purposes of safety, the radioactive waste will be stored in a raw, a treated or a conditioned form. The anticipated needs for any future steps in radioactive waste management have to be taken into account as far as possible in making decisions on the processing of the waste. The radioactive waste of Ethiopia has been categorized (see appendix III)

CHAPTER FIVE

5.1.1 EVALUATION, TECHNIQUES, DATA ANALYSIS AND DATA INTERPRETATION.

5.1.2 OVERVIEW OF SAMPLES ANALYSIS

The experimental frame work for this research work in the analysis of samples commencing from detector calibration to analytical calculation is summarized in Figure.5.1.1.

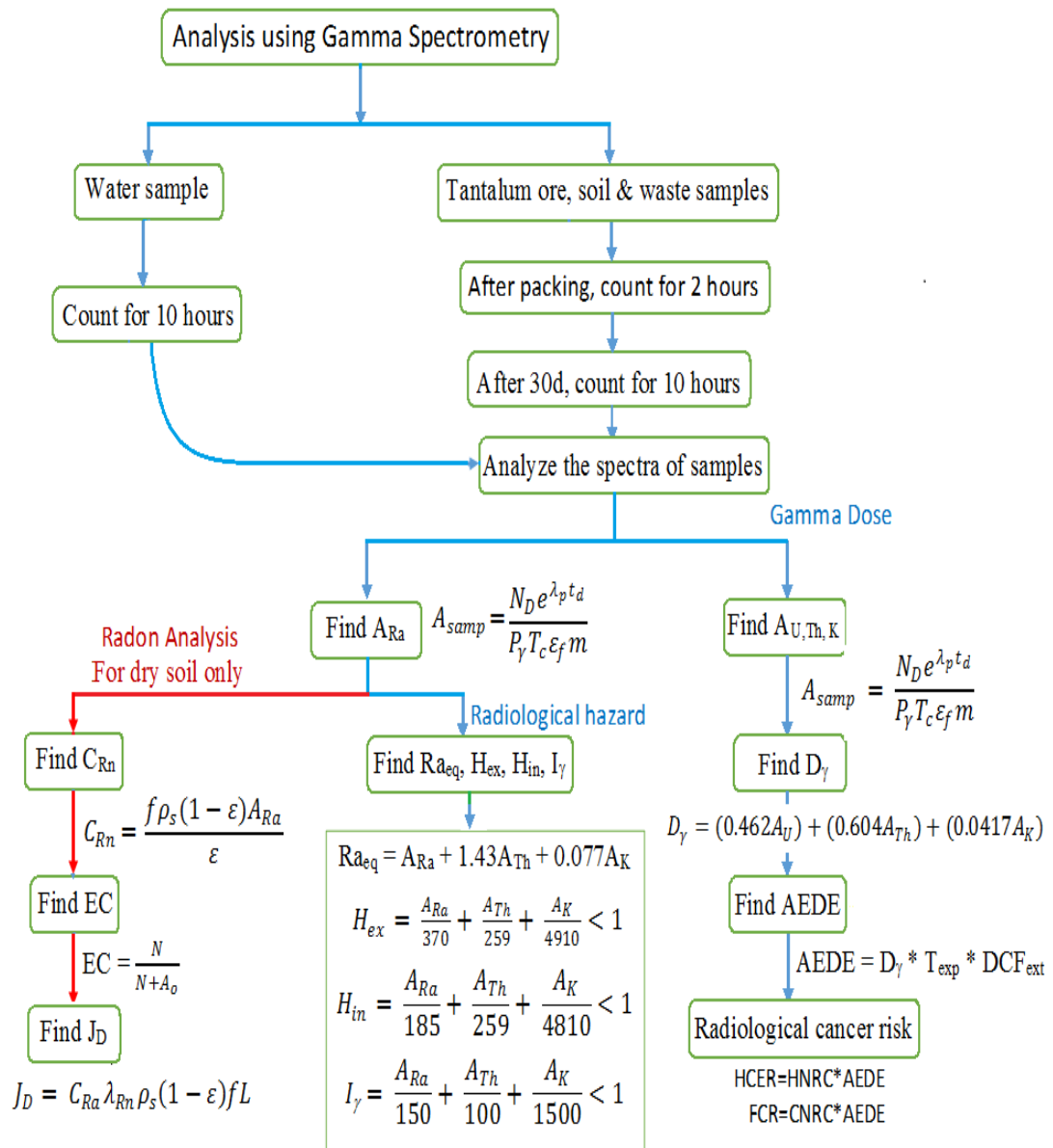


Figure 5.1.1 Flowchart for the experimental sample analysis5

5.1.3. ACTIVITY CONCENTRATION AND RATIOS

In secular equilibrium, the activity concentrations of ^{214}Pb and ^{214}Bi represent uranium-238 or radium-226 (S.Todsadol, 2012) and that of ^{212}Pb , ^{208}Tl and ^{228}Ac represent activity concentration of ^{232}Th . Therefore, the activity concentration of ^{238}U radionuclide was estimated from average activities of ^{214}Pb at gamma peaks of [351.92keV (35.6%) and 295.21keV (18.42%)] and ^{214}Bi [609.31keV (45.5%), 1120.3 keV (14.9%), 1238.1keV (5.8%) and 1764.49keV (15.3%)], while ^{232}Th radionuclide was estimated from peaks of ^{212}Pb [238.63keV (43.6%)], ^{208}Tl [583.19keV (85%) and 860.56 keV (12.5%)] and ^{228}Ac [338.3 keV (11.27%), 911.1keV (25.8%) and 968.97 keV (15.8%)]. The ^{40}K photo peak energy [1460.83keV (10.7%)] was used for computing its activity concentration (UNSCEAR, 2000b). The unit of measurements was/kg activity concentration of tantalum ore, solid waste and soil; and Bq/L for water samples. The worldwide Population-weighted average activity concentrations of ^{238}U , ^{232}Th and ^{40}K are 33, 45 and 420 Bq/kg, respectively (Saher et al, 2013; UNSCEAR, 2008a). The $^{238}\text{U}/^{232}\text{Th}$ and $^{238}\text{U}/^{40}\text{K}$ concentration ratios will also be compared with the average values reported for the soils of other countries (Saher et al, 2013). Therefore, from direct counting of samples using HPGe detector, the specific activity concentrations of ^{238}U , ^{232}Th and ^{40}K were determined using equation (3.6-1) (Darko et al., 2013; Darko et al, 2010, 2013).

$$A_{\text{samp}} = \frac{N_D \cdot e^{\lambda_p t_d}}{P_\gamma \cdot T_c \cdot \epsilon_f \cdot W_{\text{samp}}} \quad (5.1.1)$$

Where,

- N_D is the net counts of the radionuclide in the samples, ($e^{\lambda_p t_d}$) is the decay correction factor for delay between time of sampling and counting,
- λ_p is the decay constant of the parent radionuclide,
- t_d is the delay time between sampling and counting,
- P_γ is the gamma ray emission probability (gamma ray yield),
- T_c is the sample counting time,
- ϵ_f is the absolute counting efficiency of the detector system and
- W_{samp} is the mass of the sample (kilogram) or volume (liter).

5.1.4 UNCERTAINTY CALCULATION

A measurement result is complete only when is accompanied by a quantitative statement of its uncertainty. The estimation of result's uncertainty is the most important issue in analytical techniques especially in the determination of radioactivity in environmental samples in which their activity concentration is mainly low (30Bq/kg in soil sample). Therefore, identifying the sources of uncertainty in nuclear analytical techniques is an important step for reporting high quality data. The most important sources of uncertainty and the magnitude of their contribution in the case of gamma spectrometry are sample weight, detector efficiency, counting statistics, half-life, emission probability, coincidences correction, attenuation correction, sample geometry, background peak area and net count peak area(Bakr and Ebaid, 2011). For this work, the combined uncertainty was numerically obtained by applying the usual method for each of corresponding counting rate in all samples activity concentration and it was computed by equation (3.6-2):

$$e_A = A_{s\text{amp}} * \sqrt{\left(\frac{\Delta N}{N_D}\right)^2 + \left(\frac{\Delta \varepsilon}{\varepsilon_f}\right)^2 + \left(\frac{\Delta p_\gamma}{P_\gamma}\right)^2 + \left(\frac{\Delta W}{W_{s\text{amp}}}\right)^2} \quad (5.1.2)$$

Where,

- e_A is error in activity concentration (Bq/kg),
- ΔN is the error in the net count,
- $\Delta \varepsilon$ is the errors in absolute-efficiency of the detector?
- ΔP_γ is the error in gamma emission probability and
- ΔW is the error in the net weight of the sample.

5.1.5 EXTERNAL GAMMA DOSE RATE IN AIR DUE TO NORM

The direct relationship between radioactivity concentrations of natural radionuclides and their exposure is called absorbed dose rate in air at 1m above the ground. Therefore, The mean activity concentrations of ^{40}K , ^{238}U and ^{232}Th in the tantalum ore , soil, solid waste (and water samples)calculated in equation (3.6-1) was used to calculate the external gamma dose rate in air; which was calculated by equation (3.6-3) (Darko et al, 2013; Nadia, 2014; Todsadol, 2012).

$$D_{\gamma,\text{ext}} = DCF_U A_U + DCF_{Th} A_{Th} + DCF_K A_K \quad (5.1.3)$$

Where,

- A_u , A_{Th} and A_k are the activity concentrations of ^{238}U , ^{232}Th and ^{40}K respectively and
- $DCF_U = 0.462$, $DCF_{Th} = 0.604$ and $DCF_K = 0.0417$ in nGy/h per Bq/kg are dose conversion factors for ^{238}U , ^{232}Th and ^{40}K respectively.

The external radiation ^{238}U and ^{232}Th decay series and ^{40}K make approximately produce equal contribution to the externally incident gamma radiation dose to individuals in typical situations both outdoors and indoors. The population-weighted values give an average absorbed dose rate in air outdoors from terrestrial gamma radiation of 60 nGy/h. The average values range from 18 to 93 nGy/h. A typical range of variability for measured absorbed dose rates in air is from 10 to 200 nGy/h (UNSCEAR, 2000b).

5.1.6 ANNUAL EFFECTIVE DOSE (AED) FOR EXTERNAL γ -RADIATION

Effective dose is a measure of the whole body dose, which is the sum of the doses from both external and internal exposures (Lab, 2011). Annual effective dose is the effective dose in a year multiplied by radiation weighting factor. For any sample, terrestrial gamma radiation (absorbed dose rate in air) has been measured 1m above the ground surface. These data cannot directly provide the radiological risk to which an individual is exposed (Aliyu et al, 2015; Todsadol, 2012). The absorbed dose of external terrestrial gamma radiation calculated in section (3.6-3) can be converted into AED. To estimate annual effective doses: (a) the conversion coefficient from absorbed dose in air to effective dose, DCF_{ext} , and (b) the outdoor occupancy factor, which is 0.2 is applied (UNSCEAR, 2000b). The average numerical values of these parameters vary with the age of the population and the climate at the location considered. For this research, AED was computed by equation (3.6-4) (Faanu, 2011; Darko et al, 2010; IAEA, 2005; Todsadol, 2012):

$$AED_{\gamma,ext} = D_{\gamma,ext} \cdot T_{exp,ext} \cdot DCF_{ext} \quad (5.1.4)$$

Where;

- $D_{\gamma,ext}$ is the average external gamma dose rate in $\mu Gy/h$,
- T_{exp} is the annual exposure time (h);

- DCF_{ext} is the conversion factor from dose rate to effective dose of 0.7 Sv/Gy for environmental exposure to gamma rays (Faanu, 2011; IAEA, 2005; Nadia, 2014; UNSCEAR, 2000b).

For this research, if the workers assumed to spend 8 hours per day and their maximum working days per year is 245, then the total exposure duration is estimated to be 1960 h/y (8h/dx245d/y=1960 h/y); and applying outdoor occupancy factor of 0.2 (i.e. $T_{exp} = 1960 \times 0.2 = 392$ h/y). The worldwide average outdoor annual effective dose from total external terrestrial radiation except radon and cosmic is 0.07 mSv and from total external terrestrial radiation except radon is 0.48mSv (i.e. outdoor, 0.07 mSv, plus indoor, 0.41mSv), with the results for individual countries being generally within the 0.3 - 0.6mSv range. For children and infants, the values are about 10% and 30% higher, in direct proportion to an increase in the value of the conversion coefficient from absorbed Dose in air to effective dose (Al Murgan, 2015; UNSCEAR, 2000b). Specifically, the worldwide average annual effective dose from ^{238}U and ^{232}Th and ^{40}K are 1.34 mSv/y, 0.34 mSv/y and 0.33 mSv/y, respectively(UNSCEAR, 1993, 2000b). Therefore, the external effective dose rate in unit of $\mu\text{Sv/y}$ in a simplified to the following formulas:

$$AED_{\gamma,ext} = D_{\gamma,ext}(n\text{Gy/h}) \times 392\text{h/y} \times 0.7 \text{ Sv/Gy} \quad (5.1.5)$$

$$AED_{\gamma,ext} = D_{\gamma,ext} \times 2.744 \times \mu\text{Sv/y} \quad (5.1.6)$$

5.1.7 RADIOLOGICAL CANCER RISK ASSESSMENT DUE TO NORM

The long term effects of receiving radiation exposure impacts probability of cancer or death. These impacts are quantified in terms of stochastic and non-stochastic effects and their associated health risks. Therefore, the radiological fatality cancer risks and severe hereditary effects due to exposure to NORMs were assessed from the Kenticha tantalum mine. This was done by using the ICRP recommended risk assessment technique and the use of appropriate nominal probability coefficients for stochastic effects (ICRP, 2007). The ICRP recommended nominal risk coefficients for stochastic effects are given in Table 3.6-1 (Hanlon, 2012; Bevelacqua, 2009). Fatality cancer risk (FCR) and hereditary cancer effects risks (HCER) were calculated by equation (3.6-7 and 3.6-8) respectively.

$$\text{FCR} = \text{CNRC} \times \text{AED} \quad (5.1.7)$$

$$\text{HCER} = \text{HNRC} \times \text{AED} \quad (5.1.8)$$

Where,

- CNRC is cancer nominal risk coefficient,
- HNRC is hereditary nominal risk coefficient and
- AED is annual effective dose of the radiation received.

Table 5.1.1: Detriment adjusted nominal risk coefficients for cancer and heritable effects (ICRP, 2007)

Exposed population	Cancer fatality		Hereditary effects		Total	
	ICRP publication	ICRP publication	ICRP publication	ICRP publication	ICRP publication	ICRP publication
ICRP publication	103	60	103	60	103	60
Whole	5.5	6	0.2	1.3	5.7	7.3
Adult	4.1	4.8	0.1	0.8	4.2	5.6
Note: Detriment adjusted nominal risk coefficients ($1 \times 10^{-2} \text{ Sv}^{-1}$) for stochastic effects after exposure to radiation at low dose rate						

The ICRP risk assessment technique was employed in the estimation of the radiological fatality cancer risks for the individual and whole population as well as severe hereditary effects (ICRP, 2007). The practical arrangement of radiological risk recommended by ICRP is established on the assumption that at doses below 100 mSv, a specified increment in dose will yield a directly proportionate in the probability of incurring cancer or hereditary effects attributed to ionizing radiation. The model is commonly known as Linear-Non-Threshold dose response for which any dose greater than zero has an optimistic probability of producing an effect (Faanu, 2011; Cember, 2009; ICRP, 2007).

4.1.8 RADIOLOGICAL HAZARD ASSESSMENT DUE TO NORM

Table 5.1.2 Gamma Spectrometer analysis on the ore Sample in the jig box of tantalum mining

Nuclide	Type	Activity concentration (Bq/gm)	Dose (μ Sv/hr)	Remarks
Ra ²²⁶ +dau	Norm	252	2.42	Not acceptable
U ²³⁸ +dau	Norm	180	1.52	Not acceptable
Th ²³²	Norm	125	0.45	Not acceptable

Table 5.1.3. Gamma Spectrometer analysis on the dryer and temporary storage

Nuclide	Type	Activity concentration (Bq/gm)	Dose (μ Sv/hr)	Remarks
Ra ²²⁶ +dau	Norm	1200	2.42	Not acceptable
U ²³⁸ +dau	Norm	800	1.52	Not acceptable
Th ²³²	Norm	500	0.45	Not acceptable

Table 5.1.4 Gamma spectrometer analysis on the iron ore waste

Nuclide	Type	Activity concentration (Bq/gm)	Dose (μ Sv/hr)	Remarks
Ra ²²⁶ +dau	Norm	3.74	0.625	Not acceptable
U ²³⁸ +dau	Norm	1.50	0.250	acceptable
Th ²³²	Norm	0.75	0.800	Not acceptable

A measurement on the amount of ambient radiation has been measured to estimate the annual radiation exposure of workers. Besides, Environmental samples were collected to be analyzed in the ERPA laboratory to know which radionuclide is found in the collected sample. So the result of measurement on the amount of ambient radiation shows that the activities conducted on the factory is not safe from radiological point of view especially those work at the jig box (hand picking area), The final point, the heat box (Temporary storage), the main storage, the driver who transport the ore and so on. The maximum radiation readings for alpha, beta and gamma taken at the site were higher than that of the upper limit set for occupational exposure. The laboratory analysis findings indicate that radium (Ra²²⁶+dau), (U²³⁸+dau) and thorium (Th²³²) radio nuclides are found on the tantalum ore, solid iron waste, tailing dam samples with high activity concentration and radiation doses higher than that of the background radiation measured at that site and this is an indication that the mining activities in the Factory have an impact on the environment. The other samples collected from different sites around the mining areas even though they contain radium (Ra²²⁶+dau), (U²³⁸+dau) and thorium Th²³² radio nuclides, their activity concentration is less and radiation dose lower than that of the background radiation measured at the sites. Measurement at the stockpile; The stockpile is part of the mining where the raw material is collected from the ground surface using the loading machineries

Table 5.1.3. insitu non- destructive test radiation leakage readings of NORM waste at variable distance factor (Derban cement factory).

Limestone		
Radiation type	Reading at 1m above the ground	Reading at 0.5 m above the ground
α and β (Bq/cm ²)	1.55	3.77
γ (μ sv/hr)	0.053	0.065
Gypsum		
Radiation type	Reading at 1m above the ground	Reading at 0.5 m above the ground
α and β (Bq/cm ²)	9.5	11.2
γ (μ sv/hr)	0.014	0.052
Basalt		
Radiation type	Reading at 1m above the ground	Reading at 0.5 m above the ground
α and β (Bq/cm ²)	2.10	6.95
γ (μ sv/hr)	0.070	0.073
Coal		
Radiation type	Reading at 1m above the ground	Reading at 0.5 m above the ground
α and β (Bq/cm ²)	2.03	5.77
γ (μ sv/hr)	0.0837	0.0866
Pumice		
Radiation type	Reading at 1m above the ground	Reading at 0.5 m above the ground
α and β (Bq/cm ²)	12.10	14.20
γ (μ sv/hr)	0.034	0.037
Sand stone		
Radiation type	Reading at 1m above the ground	Reading at 0.5 m above the ground
α and β (Bq/cm ²)	10.60	11.35
γ (μ sv/hr.)	0.021	0.037

Measurements on the amount of ambient radiation level have been conducted to estimate the annual radiation exposure of workers. Besides, environmental samples; soils and rocks from the surrounding were collected to be analyzed at ERPA laboratory. So far the results of direct measurements on the amount of ambient radiation shown on the activities at the factory are conducted safely from radiological point of view. The maximum reading taken at the site for gamma and particle radiation was 0.0866 μ sv/hr. and 14.20 Bq/cm² respectively. This is much lower than the upper limit set for occupational exposure code of conduct. There are three basic methods for controlling external radiation hazards. These are:

The nucleus reacts with a subatomic particle or another nucleus to form a product nucleus that is more massive than the starting material. Occur spontaneously only under special conditions. Each of the three general classes of radioactive nuclei is characterized by a different decay process or set of processes.

- Neutron-rich nuclei

Have too many neutrons and have a neutron-to-proton ratio that is too high to give a stable nucleus. These nuclei decay by a process that converts a neutron to a proton, thereby decreasing the Neutron-to-proton ratio.

- Neutron-poor nuclei

Have too few neutrons and have a neutron-to-proton ratio that is too low to give a stable nucleus.

These nuclei decay by processes that convert a proton to a neutron, thereby increasing the neutron-to-proton ratio.

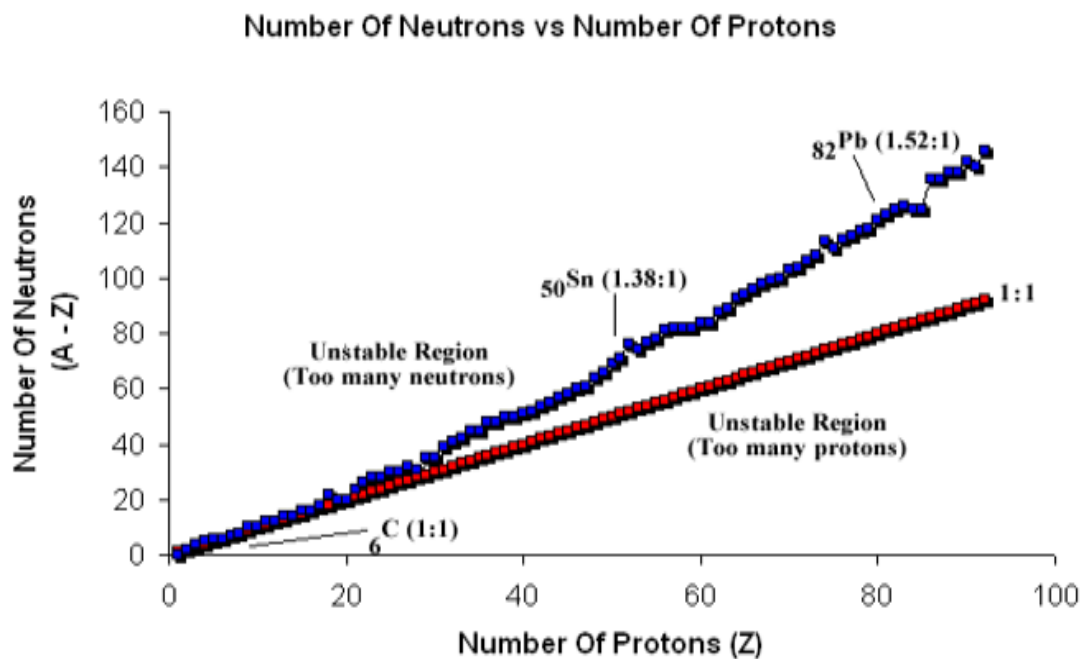


Fig 5.1.2 nuclear stability

As a nuclear reaction occurs, it has the ability to produce a chain reaction. A **chain reaction** is a reaction where the products are able to produce more products in a self-sustaining reaction series. In order to achieve a chain reaction there must be:

- A sufficient mass.

- A large concentration of fissionable nuclei

The **critical mass** is when the mass and concentration are high enough to sustain a chain reaction. A **sub-critical mass** is one that is too small to achieve a chain reaction. We can predict the stability of a nucleus by using some simple rules. All isotopes heavier than atomic number 83 have an unstable nucleus. Isotopes with 2, 8, 20, 28, 50, 82, or 126 protons or neutrons in their nucleus occur in the most stable isotopes. Nuclei are the most stable with pairs of protons and neutrons. So those with all protons and all neutrons paired up are the most stable. Isotopes with an atomic number less than 83 are most stable when the ratio of protons to neutrons is 1:1.

5.1.9. Radiation hazards and permissible limits to radiation dose

There are two principal hazards, which arise from radioactivity, regardless of the type of radiation. These are irradiation and contamination. Irradiation is mainly a problem with materials, which emit gamma radiation, or from X-ray machines. Ionizing energy is radiated out from the source and passes through a person's body. As it does so, the body tissues absorb some of the energy and the resulting ionization may have the effect of producing somatic or genetic changes on the exposed individual. Radiation is part of the environment. Humans have been living with the natural background radiation. Therefore, it is not possible to lower the dose incurred, by someone, to zero. Radiation protection is the setting of permissible dose to the public and to special groups of the society (like workers, pregnant women and children). The setting of maximum limits is a matter of establishing a balance between the benefits to be gained from ionizing radiations versus the risks incurred. This implies the equilibrium point may be different for different people, localities etc. Because of these subjective judgements, the ICRP presents its limits as recommendations only.

Table 5.1.4 Dose limits as recommended by the ICRP

Occupational		General Public
Whole body	100 mSv in 5 yrs, but not more than 50 mSv in any year.	1 mSv/yr averaged over any consecutive 5 years.
Single Organs		
Lens of the eye	150 mSv/yr	15 mSv/yr
Skin (100 cm ²)	500 mSv/yr	50 mSv/yr
Other Organs or tissues	500 mSv/yr	50 mSv/yr

In ICRP 60, two sets of limits are defined: one for individuals exposed occupationally and one for the public (Table 5.1.4)

The maximum annual dose to all radiation workers (male and female) is 20 mSv. There is an additional limit on females of reproductive capacity of 13 mSv in any three-month period. The reason for this additional restriction is to protect a recently conceived fetus within a woman who may be unaware of her pregnancy. In this sense, it is not so much a restriction designed for the protection of females but is a restriction designed to protect a fetus.

a) CALCULATION OF RADIUM EQUIVALENT ACTIVITY

Radium equivalent activity (Ra_{eq}) index represents a weighted sum of activities of ^{226}Ra , ^{232}Th and ^{40}K in NORM (Nadia, 2014). Due to uneven distribution of these natural radionuclides in the Earth crust, the actual activity level of ^{226}Ra , ^{232}Th and ^{40}K in the samples can be evaluated by means of this index. The worldwide average concentrations of the radionuclides of ^{226}Ra , ^{232}Th and ^{40}K are 370, 259 and 4810 Bq/kg, respectively (UNSCEAR, 2000a). Based on the estimation, 1Bq/kg of ^{226}Ra , 0.7Bq/kg of ^{232}Th , and 13Bq/kg of ^{40}K produces the same gamma-ray dose rate (Avwiri et al, 2012) in the environment. The radium equivalent concept is a widely used hazard index to assess the radiation hazards due to exposure of gamma radiation from different mixtures of uranium, thorium and potassium in the samples collected from different locations. This index was calculated by Equation (3.6-9)

$$Ra_{eq} = A_{Ra} + 0.077A_K + 1.43A_{Th} \quad (5.1.9)$$

Where,

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

The permissible maximum value of the radium equivalent activity is 370Bq/kg sources, which correspond to an effective dose of 1mSv/y for the public. The above criterion only considers the external hazard due to gamma rays in building materials.

b) EXTERNAL HAZARD INDEX (H_{ex})

The worldwide public exposure to natural sources of radiation (NORM) is estimated to annual average effective dose of 2.4mSv/y per person and generally expected to be in the range 1 -10mSv, (UNSCEAR, 2000b). If the dwelling is constructed, using these investigated materials and the estimate radiation dose expected to be delivered externally above this limit, hazard will be recorded. The external hazard index based

on a criterion are introduced using a model proposed by (Krieger, 1981), which was given by equation (3.6-10)(UNSCEAR, 2000c).

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (5.2.1)$$

Where,

- A_{Ra} , A_{Th} and A_K are the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K , respectively.

For construction materials to be considered safe for construction of dwellings, the external hazard index should be less than unity. This implies, the value of external hazard index must not exceed unity, in order for the external gamma radiation hazard to be insignificant. External hazard index of $H_{ex} = 1$ corresponds to a $Ra_{eq} = 370$ Bq/kg (Dragovic et al., 2006; Nada et al., 2009).

c) INTERNAL HAZARD INDEX (H_{int})

This is because, radon and its radioactive progenies are hazardous to the respiratory organs in dwellings and gamma rays emanated from construction materials. The internal exposure in (mGy/y) is quantified by the internal hazard index, H_{int} , which was calculated by the equation (3.6-11)(Saher et al, 2013):

$$H_{int} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (5.2.2)$$

Where,

- A_{Ra} , A_{Th} and A_K are the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K respectively.

For construction materials of dwellings to be considered safe, H_{int} should be less than unity, and the maximum value of H_{int} to be equal to unity (Iqbal et al, 2000).

d) REPRESENTATIVE GAMMA INDEX (I_γ)

The representative level (radioactivity, gamma) index is used to estimate the level of radiation hazard associated with the natural radionuclides of ^{226}Ra , ^{232}Th and ^{40}K in specific investigated samples. Specific levels of NORM are related to the types of rock from which the ores and soils originate (Harb et al, 2008; Al-Murgen, 2015; NEA-OECD, 1979).the gamma rays emanating from building materials can pose a health hazard. The restriction on building materials for gamma radiation is based on a dose range of 0.3 to 1 mSv/y (Ademola, 2009; Avwiri et al, 2012). In order to examine

whether a building material meets these limit of dose criteria. The activity index or gamma index I_γ of the samples under investigation was found from the Equation (3.6-12):

$$I_\gamma = \frac{A_{Ra}}{150} + \frac{A_{Th}}{100} + \frac{A_K}{1500} \leq 1 \quad (5.2.3)$$

Where,

- A_{Ra} , A_{Th} and A_K are activity concentrations for nuclides ^{226}Ra , ^{232}Th and ^{40}K , respectively (Taher and Madkour, 2011).

This gamma index is also used to correlate the annual dose rate due to the excess external gamma radiation caused by superficial materials. It is a screening tool for identifying materials that might be health concern when used for construction. Values of $I_\gamma = 1$ corresponds to an annual effective dose of less than or equal to 1mSv, while $I_\gamma = 0.5$ corresponds to annual effective dose less or equal to 0.3mSv (Avwiri et al, 2012). In order for the radiation hazard to be negligible, the value of I_γ must be less than unity (Harb et al, 2008).

CHAPTER SIX

RESULTS AND DISCUSSION

In this chapter, the results of the energy calibration, efficiency calibration and determination of the MDA of the HPGe system are presented. The discussions also cover the analysis of activity concentration, dose rate, AED, radiological hazard indices and radiological cancer risk assessments of experiment to ^{238}U , ^{232}Th and ^{40}K . The results were compared with similar facilities and other relevant work done elsewhere in the world.

6.1 HPGe DETECTOR CHARACTERIZATION

6.1.1 ENERGY CALIBRATION

Linear relationship existed between photon energy of radionuclide and produced channel number (centroid of each full energy event) and the result of the energy calibration is shown in Figure 4.1-1. The equation displayed on the energy calibration curve (equation 4.1-1) was used to determine the peak energies, E , of gamma spectra from the samples within the energy range of 0 – 2000 KeV:

$$E = 0.244 * \text{Cn} - 0.0998, \text{ where } R^2 = 1 \quad (6.1-1)$$

Where, Cn is channel number that correspond centroid of each full energy event and peak energy in energy calibration based (y-axis).

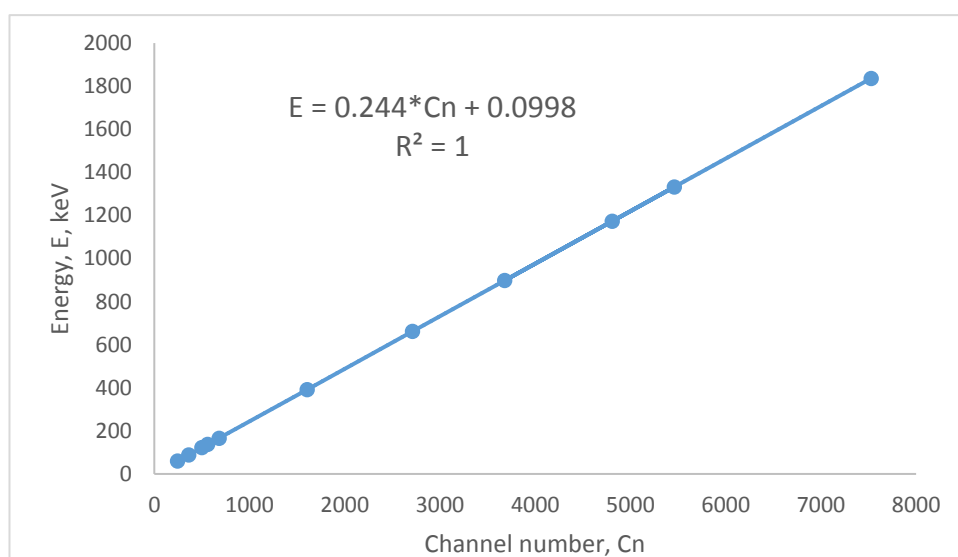


Figure 6.1.1: Energy calibration curve using mixed radionuclides standard

Hence, energy calibration makes it possible to identify the elemental composition of unknown samples (qualitative analysis).

6.1.2 EFFICIENCY CALIBRATION

In Figure 4.1-2, the efficiency values plotted against energy, resulted in an exponential curve. The displayed equation on the absolute efficiency calibration curve (equation 4.1-2) was used to calculate the detection efficiency of radionuclide at any energy, E, within the energy range (0 – 2000 KeV):

$$\varepsilon_f = 1.3803 E^{-0.694}, \quad \text{where } R^2 = 0.9992 \quad (6.1-2)$$

Where, • ε_f is detection efficiency of radionuclide interest (y-axis),

- E is the peak energy of radionuclide interest (x- axis) (y-axis).

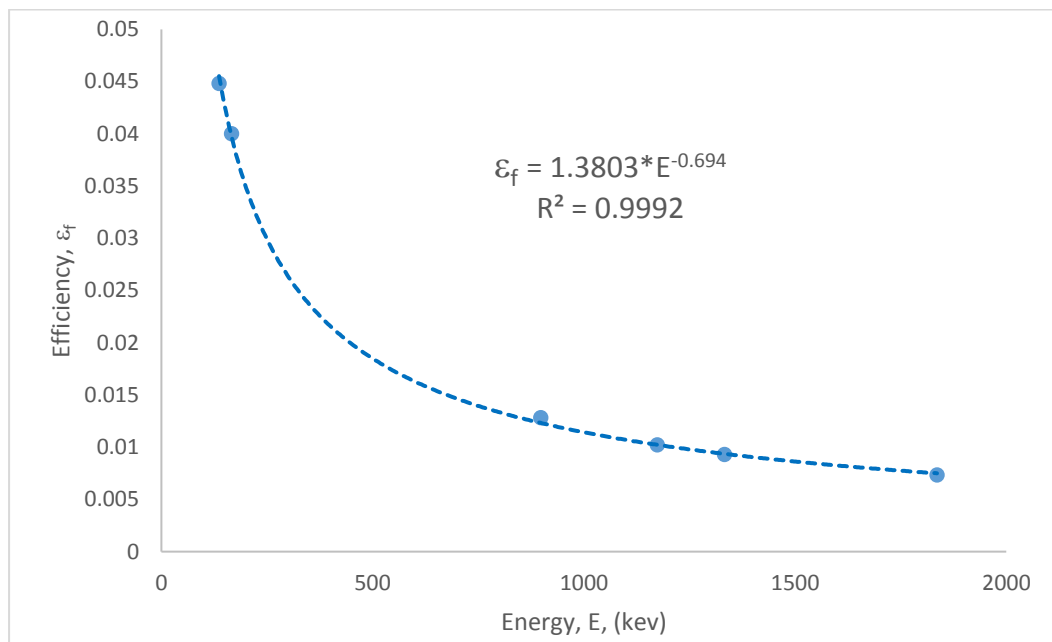


Figure 6.1-1: Efficiency calibration curve using mixed radionuclides standard

The plot shows that the efficiency of photon detection by the HPGe detector decreases with increasing photon energy. This trend conforms to the expected behavior of HPGe detectors.

6.1.3 MINIMUM DETECTABLE ACTIVITY

The minimum detectable activity, MDA, of the counting system for the radiation dose was calculated using Equation (3.5-3) and the results are shown in Table 6.1-1. These values were evaluated from the blank sample for its minimum activity.

Table 6.1-1 Calculated MDA of ^{238}U , ^{226}Ra , ^{232}Th and ^{40}K .

Nuclides	Minimum Detectable Activity, (Bq/kg)
Uranium-238	0.110
Radium-226	0.110
Thorium-232	0.025
Potassium-40	0.340

In this case, there is real activity present with the first approximation that all signals are small compared with background. The MDA depends on the counting efficiency of the detector and the emission probability per disintegration of the selected gamma line. The MDA for the counting system ranged from 0.110 Bq/kg to 0.340 Bq/kg. These value means the minimum activity needed from the source to ensure a false-negative rate is not larger than 5% (Knoll, 1999).

6.1.4 ANALYSIS OF ACTIVITY CONCENTRATION

6.1.5 TANTALUM ORE SAMPLE

The radionuclide activity concentrations for the composite samples prepared from five (5) tantalum ore samples collected from different locations across Kenticha tantalum mine were determined using equation (3.6-1) and the results are shown in Table 6.2-1. Each of corresponding counting rates in all samples activity concentration has its own error and it was computed using equation (3.6-2). Table 6.2-1 shows the measured ranges and average specific activity concentration values (\pm error) in Bq/kg for ^{238}U , ^{232}Th and ^{40}K in tantalum ore samples; and compared with the worldwide average values (WAV) reported by UNSCEAR 2000 and IAEA 2003.0914352658.

Table 6.1.4-1: Activity of ^{238}U , ^{232}Th decay series and ^{40}K in tantalum ore

Samples code	Activity concentration, Bq/kg		
	²³⁸ U±Error	²³² Th±Error	⁴⁰ K±Error
TaO1	54.990±1.125	25.650±0.490	629.870±57.440
TaO2	69.791±1.456	26.213±5.000	627.005±57.179
ROC	111.177±1.712	22.970±0.487	552.634±50.419
Mean ± Err (This work)	78.653±1.431	24.945±0.492	603.170±55.013
Range (This work)	54.990-111.177	22.970-26.213	552.634-629.870
Worldwide average^a	33	45	420
IAEA 2003	40	70	>1000
Worldwide Range	16 – 110	11 - 64	140 – 850

a: UNSCEAR 2000

From Table 6.2-1, the average specific activity concentrations of ²³²Th are almost two times lower than the world average value. However, ²³⁸U was found to be two times higher than the worldwide average reported by UNSCEAR 2000; and it was also two times higher than the values of crustal average of granite rocks reported by IAEA 2003(see Tables 2.10-1). Moreover, the specific activity concentrations of ⁴⁰K were also found to be higher than the worldwide average activity concentrations; but lower than IAEA 2003 report for ⁴⁰K concentration for granite rocks. The tantalum ore of Kenticha tantalum mine is obtained from granite-pegmatite rocks found in the Adola greenstone belt ,which are highly radioactive and contain trace amounts of ²³⁸U decay series(UNSCEAR, 2000b). Therefore, this high activity concentration of ²³⁸U and ⁴⁰K in the tantalum ore can be attributed to the presence of the granite-pegmatite rocks of the study area which generally known for the high value metal content including uranium.

6.1.6 SOIL SAMPLE

Composite soil samples prepared from six (6) different soil samples collected from different locations across Kenticha tantalum mine were analyzed to calculate the range and mean values (±error) of activity concentration in Bq/kg for ²³⁸U, ²³²Th and ⁴⁰K in soil samples as shown in Table 4.2-2. The values were compared with the worldwide average values reported by UNSCEAR 2000 and IAEA 2003.

Table 6.1.4-2: Activity of ^{238}U , ^{232}Th decay series and ^{40}K in soil samples

Samples code	Activity concentration, Bq/kg		
	$^{238}\text{U}\pm\text{Error}$	$^{232}\text{Th}\pm\text{Error}$	$^{40}\text{K}\pm\text{Error}$
SS1	70.866±1.104	16.583±0.257	723.177±65.923
SS2	67.842±1.059	14.374±0.205	714.583±65.139
Mean ± err (This work)	69.354±1.081	15.479±0.231	718.880±65.531
Range (This work)	67.842-70.866	14.374-16.583	714.583-723.177
Worldwide average^a	33	45	420
IAEA 2003	66	37	400
Worldwide Range	16 - 110	11 - 64	140 – 850

From Table 6.2-2, it can be clearly understood that the calculated average activity concentration for ^{238}U and ^{40}K were found to be almost two times higher than the worldwide mean activity concentrations reported by UNSCEAR 2000. However, the computed activity of ^{238}U and ^{40}K were almost equivalent and two times higher than the IAEA 2003 report for soil. The activity concentration of ^{232}Th was three times lower than the world average reported by UNSCEAR 2000 and two times lower than IAEA 2003, see Table (2.10-1). According to UNSCEAR 2000 report, all soils contain terrestrial radionuclides present at trace levels, therefore, the soil bearing granite-pegmatite rocks of Kenticha may contain trace amounts of highly radioactive ^{238}U decay series and ^{40}K , in which the specific activity levels are related to the types of rock from which the soils originated (UNSCEAR, 2000b). Hence, the high activity concentration of ^{238}U and ^{40}K in soil samples will be due to the granite-pegmatite rocks origin of the Kenticha soil.

6.1.7 SOLID WASTE SAMPLES

Table 4.2-3 displays the range and average values ($\pm\text{err}$) of activity concentration in Bq/kg for ^{238}U , ^{232}Th and ^{40}K for the composite solid waste samples prepared from eight (8) solid waste samples collected from different locations across Kenticha tantalum mine. The values obtained were compared with the worldwide average values reported by UNSCEAR 2000.

Table 6.1.4-3: Activity of ^{238}U , ^{232}Th decay series and ^{40}K in solid waste samples

Samples code	Activity concentration, Bq/kg		
	$^{238}\text{U}\pm\text{Error}$	$^{232}\text{Th}\pm\text{Error}$	$^{40}\text{K}\pm\text{Error}$
WJT	64.235±0.922	19.989±0.286	723.033±65.910
FW	60.821±0.872	17.906±0.258	717.305±65.387
WR	137.315±2.333	11.756±0.239	534.232±48.744
WT1	148.331±2.518	13.085±0.270	559.991±51.095
WT2	141.779±2.889	12.311±0.317	501.784±45.737
Mean±err (This work)	110.496±1.907	15.009±0.274	607.269±55.375
Range (This work)	60.821 - 148.331	11.756 – 19.989	501.784– 723.033
Worldwide average	33	45	420
Worldwide Range	16 – 110	11 – 64	140 – 850

Comparing the calculated average activity concentration values of ^{238}U and ^{40}K to UNSCEAR report, the values were found to be three times higher than and almost two times higher than the worldwide average activity concentration of ^{238}U and ^{40}K , respectively. As indicated in section (2.1), in one niobium industry of Brazil, the activity concentration level of ^{232}Th in the slag was 117,000 Bq/kg. In contrast, the solid waste from Kenticha has very low levels of ^{232}Th activity concentration than the world average. This shows the granite-pegmatite rocks of Kenticha have a very low level of thorium content.

Figure 6.2-1 shows the graphical comparison for the natural radionuclide activity concentration for ^{238}U , ^{232}Th and ^{40}K in ore, soil and solid waste samples collected from the study area.

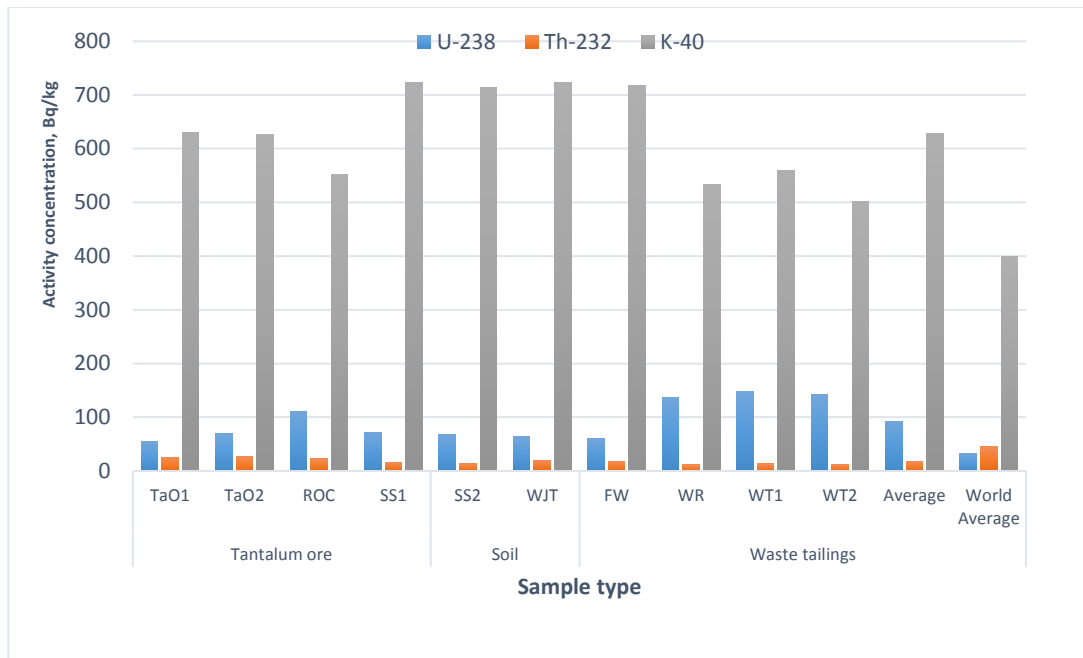


Figure 6.1.4-1: Plot of activity concentration for ^{232}Th , ^{238}U and ^{40}K in all samples

Generally, the specific activity concentrations of ^{232}Th and ^{40}K for solid waste samples were lower than the concentrations in soil and tantalum ore samples. However, the concentrations of ^{40}K in soil samples were the highest of all the other samples. Similarly, the concentration of ^{232}Th in the tantalum ore samples was higher than soil and solid waste samples. Additionally, the concentrations of ^{238}U in solid waste samples were the highest of all the others. This implies the processing of the tantalum ore increase the concentration of ^{238}U and slightly decreases the ^{232}Th and ^{40}K concentrations. Therefore, large quantities of tantalum mine solid waste may subsequently lead to increased levels ^{238}U content Kenticha area. Figure 6.2-2 also shows the graphical comparison for the natural radionuclide activity concentration of ^{238}U , ^{232}Th and ^{40}K of the study area with selected countries. The sources of data are Darko et al. 2010 for Ghana (Mine 1 and Mine 2 values) and UNSCEAR 2000 for other countries.

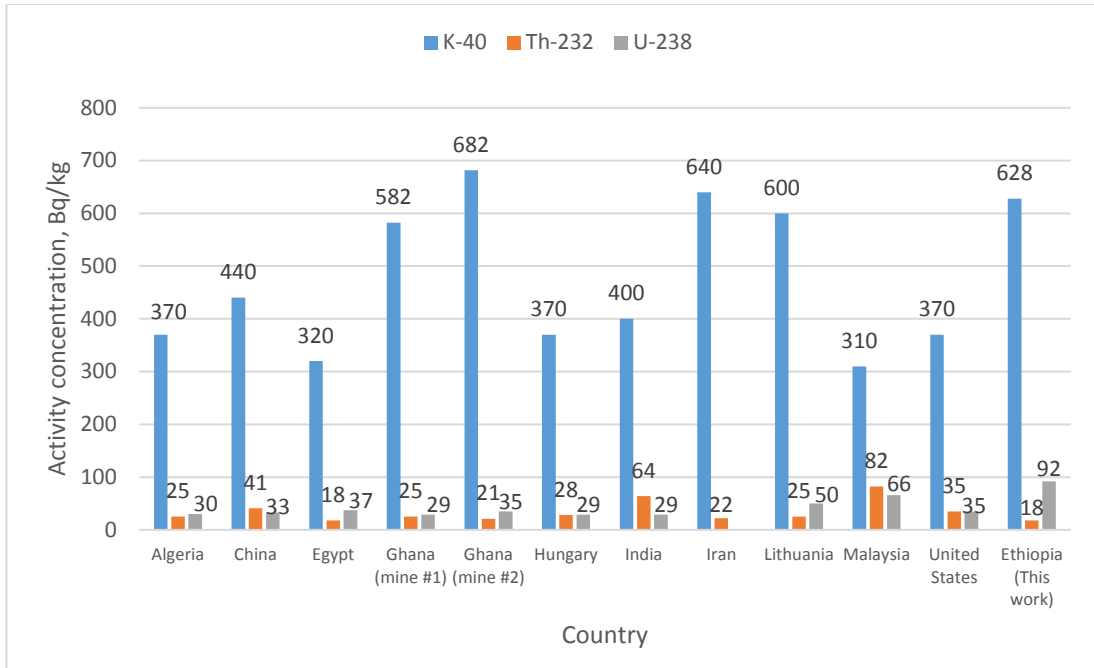


Figure 6.1.4-2: Comparison of concentrations of ^{238}U , ^{232}Th and ^{40}K in soils with published data (Darko et al, 2010; UNSCEAR, 2000c)

Figure 5.2-2 shows that the specific activity concentrations of ^{232}Th and ^{40}K for Egypt was the lowest compared to the rest. Similarly, the concentrations of ^{238}U and ^{232}Th for Malaysia were the highest as compared to all countries. Additionally, the concentration of ^{40}K in soils for Ghana (mine #2) was the highest of all the countries. Therefore, when the records of Kenticha tantalum mine was compared with these countries, the concentration of ^{238}U was higher than the highest records of Malaysia and ^{40}K was lower than the highest records of Ghana (mine #2). However, the concentration of ^{232}Th was equivalent to the lowest records of Egypt. This will be due to variation in the geology, chemistry, P^{H} , nature and redox potential of mine in those countries.

a) ACTIVITY CONCENTRATION RATIOS

Table 4.3-4 shows concentration ratios of ^{238}U to ^{232}Th and ^{238}U to ^{40}K for the composite tantalum ores, soil and solid waste samples collected from different locations in Kenticha tantalum mine were compared with the records of different countries in the world. These values were used to show abundance of ^{238}U in comparison with ^{232}Th and ^{40}k . Based on this study, ^{238}U is more abundant than ^{232}Th in all samples collected from the study area. The $^{238}\text{U}/^{40}\text{K}$ ratio for samples of the study

area was found in proportion compared with the ratio for other countries as shown in Figure 6.3-3.

Table 6.1.4-4: Concentration ratios of ^{238}U to ^{232}Th and ^{238}U to ^{40}K in tantalum ores

Nuclides	Samples code											Mean±ST D
	Tantalum ore			Soil		Solid waste samples						
	Ta O1	Ta O2	R O C	SS 1	SS2	W JT	F W	W R	WT 1	WT 2		
$^{238}\text{U}/^{232}\text{Th}$	2.1 4	2.66	4.8 4	4.2 7	4.7 2	3.2 1	3.4 0	9.4 0	11.3 4	11.5 2	5.75±3.60	
$^{238}\text{U}/^{40}\text{K}$	0.0 9	0.11	0.2 0	0.1 0	0.1 0	0.0 9	0.0 9	0.2 1	0.27	0.28	0.15±0.08	

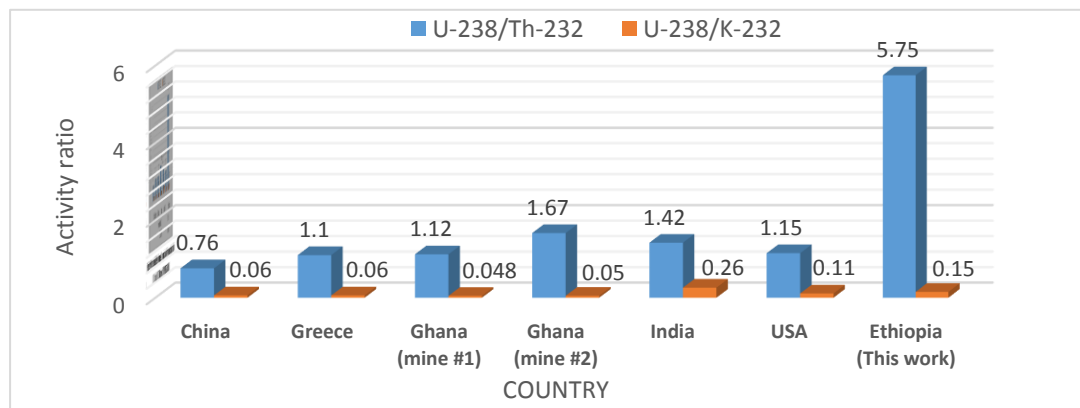


Figure 6.1.4-3: comparison of activity concentration ratios for $^{238}\text{U}/^{232}\text{Th}$ and $^{238}\text{U}/^{40}\text{K}$ (Darko et al, 2010; UNSCEAR, 2000c)

As shown in Figure 4.2-3, the result obtained for $^{238}\text{U}/^{232}\text{Th}$ and $^{238}\text{U}/^{40}\text{K}$ ratio of this work were compared with the average values reported for the soils of Greece, USA, India, China (Saher et al, 2013), Ghana mine #1 and Ghana mine (Darko et al, 2010). The $^{238}\text{U}/^{232}\text{Th}$ ratio for this work (5.75 ± 3.60) was much higher than all the aforementioned countries. This could be due to several factors like variation in the geology, chemistry, pH , and nature of mine in these countries.

6.1.8 DOSE ASSESSMENT DUE TO NORM ABSORBED DOSE RATE FROM TANTALUM ORE, SOIL AND SOLID WASTE SAMPLES

The calculated range and average of gamma absorbed dose rate in air outdoors (D_γ) and annual effective dose (AED) from ^{238}U , ^{232}Th and ^{40}K in the composite ore, soil and solid waste samples from Kenticha tantalum mine compared with the permissible limits recommended by UNSCEAR- 2000 are shown in Table 4.3-1. The world average dose rate in air outdoor from ^{238}U , ^{232}Th and ^{40}K are 18 nGy/h, 15 nGy/h and 27 nGy/h, respectively; and the worldwide average AED from ^{238}U and ^{232}Th and ^{40}K are 1.34 mSv/y, 0.34 mSv/y and 0.33 mSv/y, respectively (UNSCEAR, 1993, 2000b). Table 6.1.8-1: D_γ and AED due to ^{238}U , ^{232}Th and ^{40}K compared with the WAV

Nuclides	External γ -dose, nGy/h			AED, mSv/y		
	Ore	Soil	Waste	Ore	Soil	Waste
U-238	36.61	32.04	51.05	0.010	0.009	0.014
Th-232	14.58	9.32	9.04	0.004	0.003	0.003
K-40	25.22	29.98	25.32	0.007	0.008	0.007
Sum (This work)	76.41	71.34	85.41	0.021	0.020	0.023
Mean\pmSTD(This work)	77.72 \pm 7.13			0.021 \pm 0.002		
Worldwide average	60 ^a			0.48 (0.07+0.41) ^{2b}		
Worldwide Range	18 – 93 ^b			0.3 - 0.6 ^b		

In Table 6.3-1, the calculated gamma absorbed dose rates in ore, soil and solid waste samples were in range of 71.337 and 85.408 nGy/h with an average value of 77.717 nGy/h. This value indicates that the absorbed dose rate from terrestrial radiation in air outdoors in the investigated area were slightly higher than the worldwide average value recommended by UNSCEAR-2000 of 60 nGy/h; this difference could be attributed to vast differences exhibited in Kenticha granite –pegmatite rocks in texture, mineralogy, geology and geochemical state (Zerihun, n.d.) of the sampling sites.

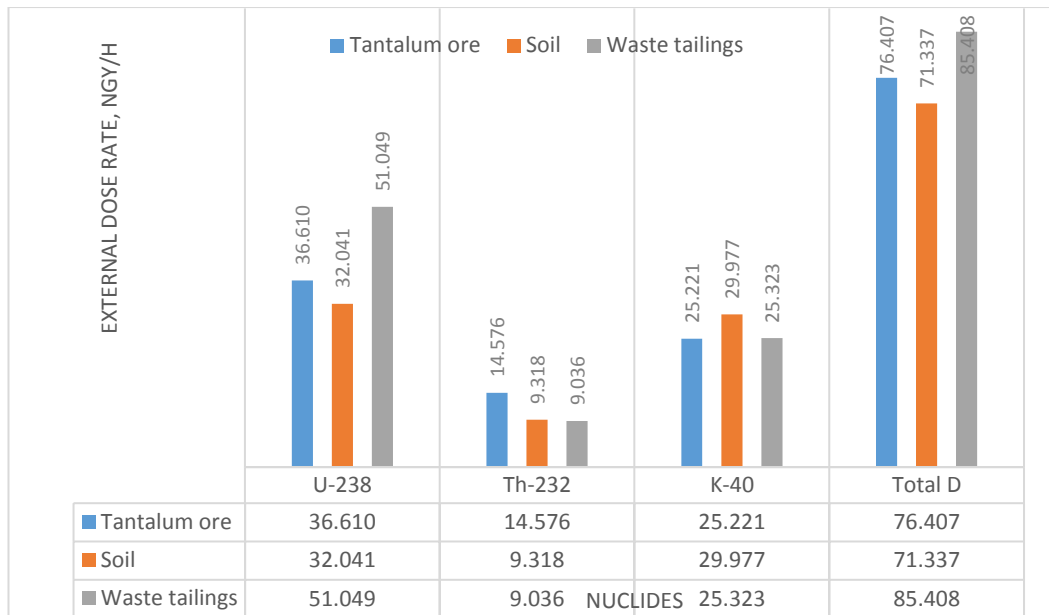


Figure 6.1.8-1: Comparison of γ - dose due ^{238}U , ^{232}Th and ^{40}K in ore, soil and solid waste samples.

Figure 6.3-1 shows the graphical comparison of the external absorbed dose rate due to ^{238}U , ^{232}Th and ^{40}K in tantalum ore, soil and solid waste samples collected from Kenticha tantalum mine. As the figure shows, the external gamma dose due to uranium-238 is the highest of the rest followed by dose due to potassium-40. The gamma dose rate originating from thorium-232 is the lowest. Even though, the total gamma dose rate due to ^{238}U , ^{232}Th and ^{40}K was above the worldwide average reported by UNSCEAR 2000, the average gamma dose rate was within the worldwide range of 18 – 93 nGy/h (UNSCEAR, 1993). However, this value reveals the significance of high NORM activity concentration of the area.

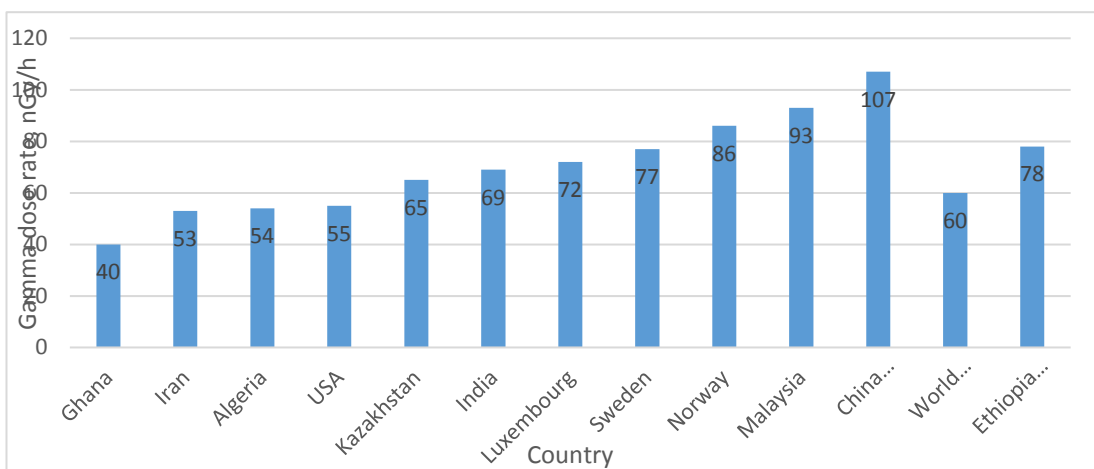


Figure 6.1.8-2: Comparison of absorbed dose rate from NORMs in soil by country(UNSCEAR, 2000d)

Figure 6.3-2 compares the gamma absorbed dose rate of the study area with some countries in the world. The comparison revealed that the external gamma dose rate of Ghana, Iran, Algeria and USA are slightly lower than the world average. However, the gamma dose rate for Kazakhstan, India, Luxembourg, Sweden, Norway, Malaysia and China, increases from 65 nGy/h to 107 nGy/h which is higher than the world average of 60 nGy/h so is the 78 nGy/h recorded for the study area (Ethiopia). As also indicated in (section 2.1), Gerais and Guarapari in Brazil, Yangiang in China, the states of Kerala and Madras in India, the Nile delta in Egypt, Niue Island in the Pacific, parts of Italy and central massive in France have the highest natural background radiation exposure levels. According to UNSCEAR 2000 report, there are various causes of these elevated exposure levels for high natural background; one is due to differences in features of the geology. For instance, monazite sand deposits, volcanic soils, granitic rocks, schistic rocks and some associated with uranium minerals in soil are the main causes of high natural background radiation. It should be noted that exposures in high background areas can vary in time as deposits or beach sands are replenished by springs and tides (UNSCEAR, 2000b).

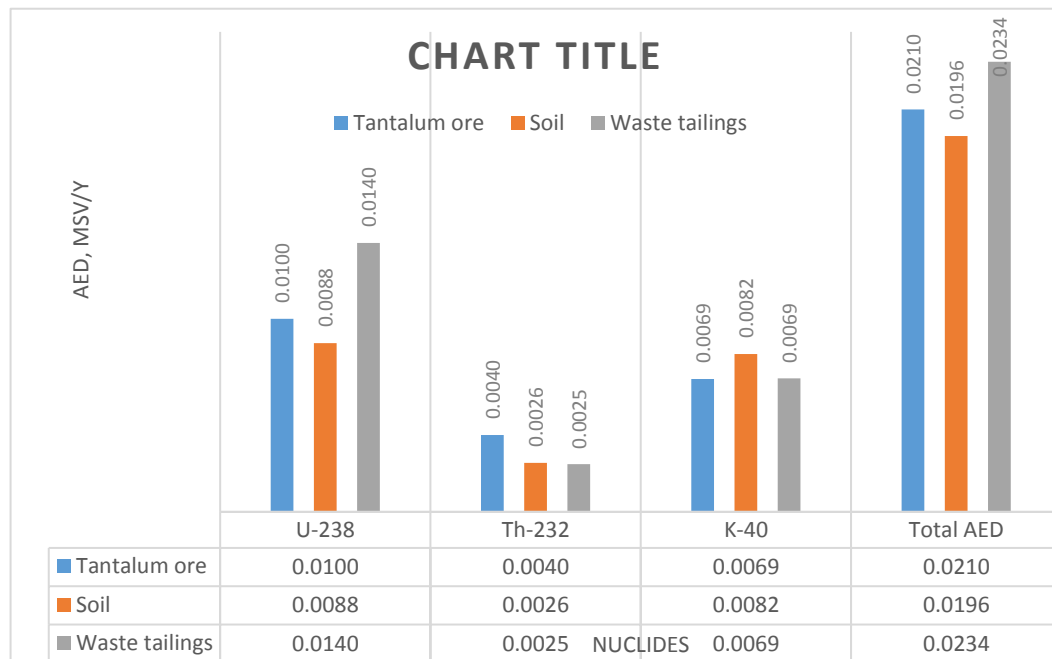
6.1.9 ANNUAL EFFECTIVE DOSE FROM TANTALUM ORE, SOIL AND SOLID WASTE SAMPLES

Table 6.3-1 provides the corresponding annual effective dose (AED) attributable to terrestrial gamma radiations due to ^{238}U , ^{232}Th and ^{40}K activity concentrations. The average outdoor Aero the study area is three times lower than the worldwide average value of 0.07 mSv/year (Al Murgan, 2015; UNSCEAR, 2000b). Therefore, the AED from ^{238}U , ^{232}Th and ^{40}K activity concentrations falls within the public AED limit of 1

Figure 6.1.8-3: Plots of AED due ^{238}U , ^{232}Th and ^{40}K of ore, soil and solid waste samples

mSv (IAEA, 2010). Figure 5.3-3 show the graphical comparison of the AED incurred from ^{238}U , ^{232}Th and ^{40}K in tantalum ore, soil and solid waste samples collected from the study area. Generally, the graph shows that the AED received from the solid waste was higher than the AED received from tantalum ore and soil. However, the total AED received from all types of materials due to ^{238}U , ^{232}Th and ^{40}K were below the worldwide reported average.

6.1.10 RADIOLOGICAL CANCER RISK ASSESSMENT



The evaluation of risk associated with the various exposure pathways considered in this research are reported. Table 4.4-1 shows the average annual effective dose for the composite samples of tantalum ore, soil and solid wastes well as the estimated risk components. The risk of exposure to low doses and dose rates of radiation was estimated using the ICRP recommended risk coefficient (ICRP, 2007, 2012) and an assumed 70 years lifetime of continuous exposure of the population to low level radiation.

Table 6.1.10-1: Estimated stochastic risk assessments for external irradiation of ^{238}U , ^{232}Th and ^{40}K in ore, soil and solid waste samples

Samples code	Annual effective dose, (Sv)	Fatality cancer risk to population per year	Life time fatality cancer risk to Adult	Severe hereditary effect per year, population	Estimated lifetime hereditary effect, adult
TaO1	1.84E-05	1.06E-06	8.20E-07	1.38E-07	8.29E-08
TaO2	2.04E-05	1.17E-06	9.06E-07	1.53E-07	9.16E-08
ROC	2.42E-05	1.39E-06	1.08E-06	1.82E-07	1.09E-07
SS1	2.00E-05	1.15E-06	8.90E-07	1.50E-07	9.00E-08
SS2	1.92E-05	1.10E-06	8.53E-07	1.44E-07	8.62E-08
WJT	1.97E-05	1.13E-06	8.78E-07	1.48E-07	8.88E-08
FW	1.89E-05	1.09E-06	8.40E-07	1.42E-07	8.50E-08
WR	2.21E-05	1.27E-06	9.82E-07	1.66E-07	9.93E-08
WT1	2.74E-05	1.57E-06	1.22E-06	2.05E-07	1.23E-07
WT2	2.58E-05	1.48E-06	1.15E-06	1.93E-07	1.16E-07
Mean	2.16E-05	1.24E-06	9.61E-07	1.62E-07	9.72E-08
STD	3.14E-06	1.81E-07	1.40E-07	2.35E-08	1.41E-08
Min	1.84E-05	1.06E-06	8.20E-07	1.38E-07	8.29E-08
Max	2.74E-05	1.57E-06	1.22E-06	2.05E-07	1.23E-07

The fatality cancer risks for all samples were in the range of 1.06E-06 - 1.57E-06 with an average value of 1.24E-06. This implies that approximately 1 person out of a 1,000,000 people is likely to suffer from cancer per year as a result of external irradiation from tantalum ore, soil and solid waste and this is considered to be insignificant. The lifetime fatality cancer risk for all samples were in a range of 8.20E-07 - 1.22E-06 with an average value of 9.61E-07 which also meaning that approximately 1 person out of 1,000,000 people is likely to suffer from cancer. The corresponding severe hereditary effects per year and estimated lifetime hereditary effects were in a range of 1.38E-07 - 2.05E-07 and 8.29E-08 - 1.23E-07 with an average values of 1.62E-07 and 9.72E-08, respectively. Similarly, this suggests that approximately 2 persons out of 10,000,000 people are likely to suffer from severe hereditary diseases per year and approximately 1 person out of 10,000,000 people are likely to suffer from lifetime hereditary related diseases due to low background radiation exposure, this is considered to be insignificant compared with the EPA acceptable range of risks of 1.0E-6 – 1.0E-4 values for the population of the study area. The EPA acceptable risk value of 1.0E-6, i.e. 1 case out of 1 million people dying from cancer, is considered as inconsequential (Faanu, 2011).

6.1.11 RADIOLOGICAL HAZARD ASSESSMENT

This sub-section outlines the computed radium equivalent activity (R_{eq}), external hazard index (H_{ext}) and internal hazard index (H_{int}) using Equations 3.8, 3.9, 3.10 and 3.11, respectively, in tantalum ore, soil and solid waste tailing samples from Kenticha tantalum mine in Ethiopia. It is important to assess the gamma radiation hazards on humans from the point of view of the use of some of the above samples as building material. The values of R_{eq} , H_{ext} and H_{int} for all samples in the study were compared with the recommended limits by UNSCEAR-2000.

6.1.12 TANTALUM ORE SAMPLES

The composite samples prepared from the five different tantalum ore samples collected from the study area were identified with the sample codes TaO1, TaO2 and ROC. Table 6.5-1 shows a summary of the results obtained for tantalum ore samples from the Kenticha tantalum mine area.

Table 6.1.11-1: Hazard indices due to ^{238}U , ^{232}Th and ^{40}K in ore samples

Sample code	$R_{eq}, \text{Bq/kg}$	H_{ex}	H_{in}	I_{γ}
TaO1	138.1	0.373	0.516	1.029
TaO2	136.0	0.367	0.503	1.015
ROC	180.7	0.488	0.773	1.300
Mean±STD(This work)	151.6±25.20	0.409±0.0 7	0.597±0.1 5	1.115±0.1 6
Range±STD (This work)	136 - 180.7±25.20	0.367 - 0.488±0.0 7	0.503 - 0.773±0.1 5	1.015 - 1.30±0.16
World average	<370	<1	<1	<1

The use of a material whose radium equivalent activity concentration exceeds 370Bq/kg is discouraged to avoid radiation hazards(Avwiri et al, 2012). However, the average $R_{activity}$ concentration of the tantalum ore sample was below the permissible level of 370 Bq/kg recommended(UNSCEAR, 2000a). Therefore, the use of these materials for building is not expected to cause any significant hazard. Similarly, the computed external and internal hazard indices for the tantalum ore were less than unity. However, the representative gamma index is almost equal to the upper limit of

unity. Generally, the external radiation field and internal radon exposure due to NORM in the terrestrial tantalum ore containing thorium and uranium head series and potassium - 40 which all human beings are exposed to is safe for use.

6.1.13 SOIL SAMPLES

The composite samples prepared from the six different soil samples collected from the Kenticha mine were identified with the sample codes SS1 and SS2. Table 4.5-2 shows a summary of the results obtained for analysis of these samples.

Table 6.1.11-2: Hazard indices due to ^{238}U , ^{232}Th and ^{40}K in soil samples

Sample code	R_{eq} , Bq/kg	H_{ex}	H_{in}	I_{\square}
SS1	149.5	0.404	0.593	1.115
SS2	143.3	0.387	0.570	1.072
Mean±STD (This work)	146.4±4.36	0.395.01	0.582±0.0 2	1.094±0.0 3
Range±STD (This work)	143.3 - 149.5±4.36	0.387 - 0.404±0.01	0.570 - 0.593±0.0 2	1.072 - 1.125±0.0 3
World average	<370	<1	<1	<1

From Table 6.5-2, the estimated radium equivalent activity of soil samples was below the permissible level of 370 Bq/kg recommended by (UNSCEAR, 2000a). However, the use of a material whose radium equivalent activity concentration exceeds 370Bq/kg is discouraged to avoid radiation hazards(Avwiri et al, 2012). Therefore, soil is safe to use it as building materials. Similarly, the computed external and internal hazard indices for the tantalum ore was less than unity, but the representative gamma index were almost equal to the upper limit of unity. In general, the gamma radiation field that assesses the level of exposure due to radionuclides from the superficial surface of the building materials and internal radon exposure arising from radon inhalation and originating from radionuclides are more than the external hazard index that assesses the exposure arising from gamma emitting radionuclides in building materials. However, the use of soil as building material can be considered safe.

6.1.14 SOLID WASTE SAMPLES

Table 6.5-3 shows a summary of the results obtained for solid waste samples from the Kenticha tantalum mine area. The composite samples prepared from eight different

samples collected from the different location of the study area were identified with the sample codes as indicated in the table below.

Table 6.1.11-3: Hazard indices due to ^{238}U , ^{232}Th and ^{40}K in solid waste samples

Sample code	R_{eq},Bq/kg	H_{ex}	H_{in}	I_γ
WJT	148.5	0.401	0.575	1.11
FW	141.6	0.383	0.547	1.06
WR	190.2	0.514	0.871	1.36
WT1	206.8	0.559	0.951	1.47
WT2	145.5	0.393	0.634	1.05
Mean±STD (This work)	166.5±29.9	0.450±0.08	0.716±0.18	1.21±0.19
Range±STD(This work)	141.7 - 210.2	0.383 - 0.559	0.547 - 0.951	1.05 - 1.47
Worldwide average	<370	<1	<1	<1

The estimated radium equivalent activity of solid waste samples was below the permissible level of 370 Bq/kg recommended by (UNSCEAR, 2000a). However, the use of a material whose radium equivalent activity concentration exceeds 370Bq/kg is discouraged to avoid radiation hazards(Avwiri et al, 2012). Therefore, solid waste of Kenticha tantalum mine will be safe for use as building materials. Correspondingly, the computed external and internal hazard indices for the solid waste were less than unity, but the representative gamma index is slightly higher than the upper limit of unity. Generally, the external radiation field and internal radon exposure due to NORM in the terrestrial soil containing thorium and uranium head series and potassium 40 which all human beings are exposed to is safe for use.

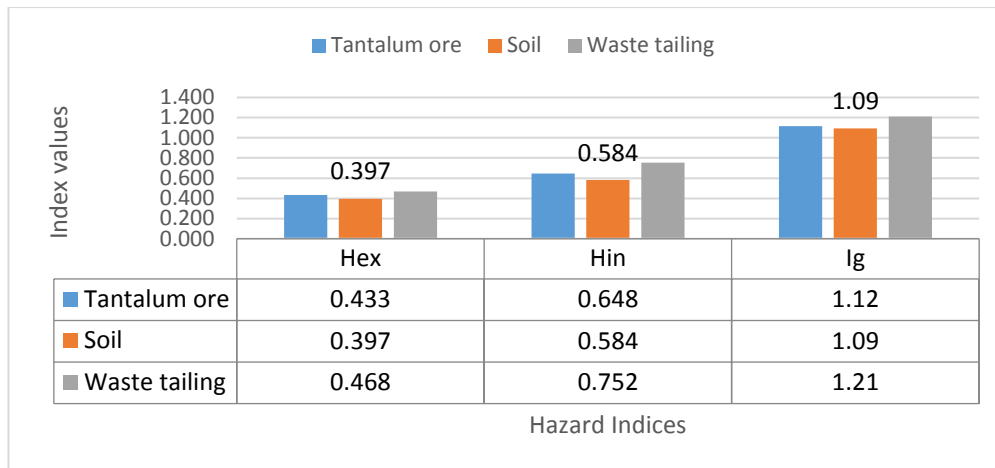


Figure 6.1.11-1: Comparison of the radiological hazard indices for ore, soil and solid waste samples γ

Figure 6.5-1 shows the graphical comparison of the radiological hazard indices due to ^{238}U , ^{232}Th and ^{40}K in tantalum ore, soil and solid waste samples. As the figure shows, the average representative gamma index that accesses the level of gamma exposure due to nuclides from the superficial surface of the building materials was almost equal to unity. The internal radon exposure arising from radon inhalation originating from radionuclides and the external hazard index that accesses the exposure arising from gamma emitting radionuclides in building materials were all less than unity. All building materials must have all hazard indices less than unity for it to be considered safe for use. Therefore, the use of ore, soil and solid waste as building material can be considered safe.

Table 6.1.11-4: Comparison of the average activity concentrations ^{238}U , ^{232}Th , ^{40}K and radium equivalent Activities (Ra_{eq}) of soil, rocks, solid waste and tailings of the study area with published data (Faanu, 2011).

Country	No. of samples	Activity concentration, Bq/kg			Ra_{eq} , Bq/kg
		^{226}Ra	^{232}Th	^{40}K	
Canada	7	51.5	48.1	114.7	129.4
Austria	18	26.7	14.2	210	63.1
Algeria	12	41	27	422	112
Brazil	1	61.7	58.5	564	188.8
China	46	56.5	36.5	173.2	122
Egypt	85	78	33	337	151
Japan	38	12.5	23.9	206.2	62.5
India	1	37	24.1	432.2	104.7
Tunisia	2	21.5	10.1	175.5	49.7
Denmark	145	40	28	248.3	99.1
Ethiopia	10	78.9	18.5	643.1	154.8

According to Table 6.5-4, the radium equivalent activity concentration in all the aforementioned countries is lower than the recommended value of 370 Bq/kg (UNSCEAR, 2000b). Relatively, the highest was recorded in Brazil, while the lowest was recorded in Tunisia. Additionally, the records of this work were three times higher than the lowest records of Tunisia and lower than the highest record of Brazil. Hence, the materials of the study area may be being safe for use as building material.

6.2 ANALYSIS OF RADON

6.2.1 TANTALUM ORE SAMPLES

Composite samples were prepared from the five different tantalum ore samples collected from different location of Kenticha tantalum mine of Ethiopia. Then, the activity concentration, exhalation rate (J_D) and emanation coefficient of ^{222}Rn in the composite ore sample matrix was calculated from the activity concentration of ^{226}Ra . The results of the experiments are summarized in Table 5.6-1.

Table 6.2-1: ^{226}Ra concentration and corresponding C_{Rn} , EF and J_D in ore samples.

Samples code	$A_{\text{Ra-226}}$, Bq/kg	C_{Rn} , kBq/m ³	EC, %	J_D , mBq/m ² .s
TaO1	52.9	85.7	85.4	43.9
TaO2	50.2	81.4	81.8	41.7
ROC	105.3	170.5	84.5	87.3
Mean±STD (This work)	69.5±31.0	112.6±50.2	83.9±1.9	57.6±25.7
Range±STD (This work)	50.2 - 105.3±31.0	81.4 - 170.5±50.2	81.8 - 85.4±2.9	41.7 - 87.3±25.7
World average	35	78	22	33
Worldwide range	17 – 60		5 - 70	

Average activity concentration of radium-226 from the tantalum ore samples were two times higher than the worldwide average value of 35 Bq/kg (UNSCEAR, 2000b). While, the mean activity concentration of ^{222}Rn was much higher than the world average value of 78 kBq/m³ (UNSCEAR, 2000b).

The average radon emanation coefficient for crushed and homogenized ore sample under dry condition was significantly higher than the UNSCEAR report for the typical emanation coefficients for rocks and soils are in a range of 5 – 70% with a representative value of 22% (Faanu, 2011; UNSCEAR, 2000b). Correspondingly, the

average ^{222}Rn exhalation rate of tantalum ore was almost two times higher than the UNSCEAR 2000 reported value of $33\text{mBq/m}^2\text{s}$ (UNSCEAR, 2000b).

In general, the concentration, emanation coefficient and exhalation rate of radon in tantalum ore samples are dependent on several factors, the most important of which are the ore's radium content and distribution, grain size, porosity, permeability to gas movement, and moisture content. These characteristics are, in turn, determined by the ore's parent-material composition, climate, and the ore's age or maturity (Schumann, 1995).

6.2.2 SOIL SAMPLES

Two composite samples from the six different soil samples collected from the different location of Kenticha tantalum mine ($\text{SS}_1 - \text{SS}_6$) were used for analysis to calculate the range and mean ($\pm\text{STD}$) values for activity concentration of ^{226}Ra and concentration (C_{Rn}), emanation coefficient (EC) and exhalation rate (J_{D}) of ^{222}Rn . Table 6.6-2 shows a summary of the results obtained for the composite soil samples from the study area.

Table 6.2-2: ^{226}Ra concentration and corresponding C_{Rn} , EC and J_{D} in soil samples.

Samples code	$A_{\text{Ra-226}}$, Bq/kg	C_{Rn} , kBq/m ³	EC, %	J_{D} , mBq/m ² .s
SS1	70.1	113.6	84.2	58.1
SS2	67.8	109.8	81.5	56.2
Mean \pm STD (This work)	68.9 \pm 1.7	111.7 \pm 2.7	82.9 \pm 1.9	57.2 \pm 1.4
Range \pm STD (This work)	67.8 - 70.1 \pm 1.7	109.8 – 113.6 \pm 2.7	81.5 - 84.2 \pm 1.9	56.2 - 58.1 \pm 1.4
World average	35	78	22	33
Worldwide range	17 – 60		5 - 70	

According to Table 4.6-2, the calculated mean activity concentration of ^{226}Ra in the soil samples was two times higher than the worldwide average value of 35 Bq/kg (UNSCEAR, 2000b). Similarly, the average activity concentration of ^{222}Rn in the soil was higher than 78 kBq/m³(UNSCEAR, 2000b). Correspondingly, the average radon emanation coefficient of soil samples was significantly higher than UNSCEAR report, which is typically 22% for rocks and soils (Faanu, 2011; UNSCEAR, 2000b). Whereas the average ^{222}Rn exhalation rate of soil was two times higher than the value of $33\text{mBq/m}^2\text{s}$ (UNSCEAR, 2000b). Normally, the concentration, emanation coefficient and exhalation rate of radon in soil are dependent on the soil's radium content and

distribution, grain size, porosity, permeability to gas movement, and moisture content (Schumann, 1995). These characteristics are, in turn, determined by the soil's parent-material composition, climate, and the soil's age or maturity.

6.2.3 SOLID WASTE SAMPLES

Five composite solid waste samples were prepared from the eight different solid waste samples (i.e. WT1-WT4, FeOW, FW, WR, WJT) collected from the different location of Kenticha tantalum mine were used to compute the mean and range values (Mean±STD) for concentration (C_{Rn}), emanation coefficient (EC), exhalation rate (JD) of ^{222}Rn as well as the activity concentration radium-226. Table 4.6-3 shows a summary of the results obtained for solid waste samples from the study area.

Table 6.2-3: ^{226}Ra concentration and corresponding C_{Rn} , EF and solid waste samples.

Samples code	A_{Ra-226} , Bq/kg	C_{Rn} , kBq/m ³	EC, %	JD, mBq/m ² .s
WJT	64.3	104.1	52.7	53.3
FW	60.8	98.5	54.0	50.4
WR	132.2	214.2	72.6	109.7
WT1	145.0	234.8	83.4	120.2
WT2	89.2	144.5	83.2	74.0
Mean±STD (This work)	98.3±38.6	159.2±62.6	69.2±15.1	81.5±32.1
Range±STD (This work)	60.8 - 145.0±38.6	98.5 – 234.8±62.6	52.7- 83.4±15.1	50.4 - 120.2±32.1
World average	35	78	22	33
Worldwide range	17 – 60		5 - 70	

From Table 5.6-3, the radium – 226 mean activity concentration was two time higher than the worldwide average value of 35 Bq/kg (UNSCEAR, 2000b). The mean activity concentration of ^{222}Rn was higher than 78 kBq/m³ (UNSCEAR, 2000b). Correspondingly, the average radon emanation coefficient of solid waste sample was almost three times higher than UNSCEAR report for rocks and soils are with a representative value of 22% (Faanu, 2011; UNSCEAR, 2000b). While the average radon - 222 exhalation rate of solid waste sample was two times higher than the value of 33 mBq/m².s (UNSCEAR, 2000b). Geologic and climatic factors, including radium content, grain size, siting of radon parents within soil grains or on grain coatings, and soil moisture conditions, determine the soil's emanating power and radon transport characteristics. Similarly, the calculated values for the solid waste samples collected

from Kenticha tantalum mine would be affected by these factors. Figure 5.7-1 illustrates the comparison for the radon concentration in the analyzed samples.

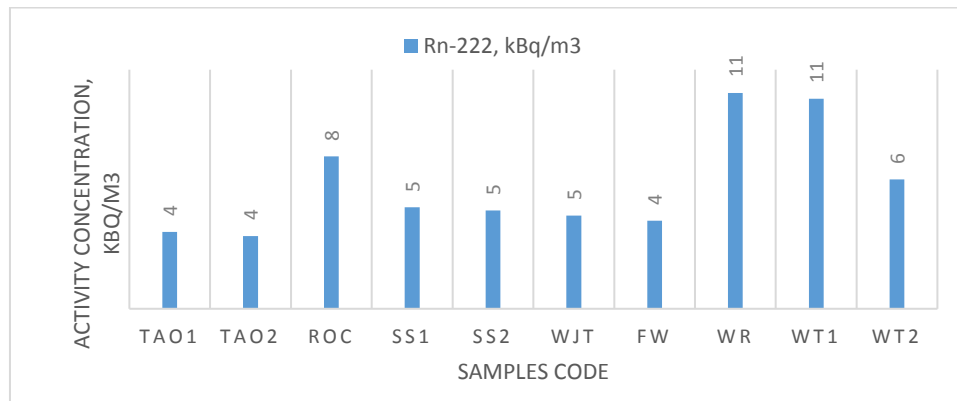


Figure 6.2-1: Plot of activity concentration for ²²²Rn in all samples

Generally, the concentrations of ²²²Rn for WR and WT1 were higher than all the other samples followed by ROC. As indicated in Figure 4.2-1, the concentration of ²³⁸U in solid waste samples was higher than all the others. This implies, the quantities of ²²⁶Ra produced from ²³⁸U is high that subsequently lead to increased levels ²²²Rn in that samples.

6.2.4 Comparison with other works

When the radon emanation coefficients of ore, soil and solid waste samples were compared with the records of UNSCEAR 2000 report as shown in Table 4.6-4, the values of this study were higher than the UNSCEAR 2000 values for granite, uranium mill tailings, petroleum solid waste and different types soil samples of aforementioned countries. *Table 6.2-4: Radon EC in soil, rock and building materials (Nabil et al, 2009)*

Country	Materials they used	Emanation coefficient (%)
Japan, Misasa	Granite	29
Austria, Badgastein	Granite	2.1
U.S.A., San Jose	Soil	25
New Mexico	Soil	38 ± 8.0
Finland	Soil	17-23
Italy	Soil	6-60
Austria, Jabiluka	Uranium mill tailing	7.2
U.S.A.	Uranium mill tailing	15 ± 1.9
Saudi Arabia	Petroleum waste	15 – 57
Ethiopia (this work)	Solid waste	69.2±15.1
Ethiopia (this work)	Soil	82.8±1.9
Ethiopia (this work)	Tantalum ore	83.9±1.9

The radon emanation coefficient of this study was two times higher than the New Mexico record for soil and five times higher than the U.S.A uranium tailing and Saudi Arabia petroleum reports. However, the Japan Misasa report is three times lower than the Kenticha radon emanation coefficient of Kenticha. All these variations depend on the geologic, climatic factors and sample matrix differences of the samples considered by each country. Table 6.2-5: Comparison of ^{226}Ra activity concentration and ^{222}Rn emanation coefficient of the study area with different samples from various industrial activities (Darko et al, 2011).

Industrial Activity	A_{Ra} , in KBq/kg	^{222}Rn EC (%)
Oil and gas production		
• Oklahoma	76.1	8.7
• Michigan	15.4	13.8
Phosphate industry		
• Gypsum	1.2	20.0
• Slag	1.26	1.0
Power plant generation		
• Cool ash	0.14	2.0
Metallurgical processing		
• Uranium mining	0.92	30.0
• Gold mining	0.013	55.4
• Tantalum mine (This work)	0.078	78.6

Correspondingly, the radon exhalation rates of ore, soil and solid waste samples results were compared with UNSCEAR 2000 records of different samples in the world as shown in Table 5.6-6, which reveals that the exhalation rate obtained in this analysis was higher than the radon exhalation rate records of UNSCEAR 2000 for different kinds of soil, granite, sediment rocks and phosphor gypsum samples of some countries in the world (UNSCEAR, 2000b). Table 6.6.6 Exhalation Rate from different rock and soil samples (Nabil et al, 2009)

Country	Materials they used	Exhalation rate(mBq/m ² .s)
Belgium	Phosphogypsum	43×10^{-2}
Bangladesh	Sediment rocks	$(1.3 - 8.0) \times 10^1$
China, Hong Kong	Granite	$(1.1 - 37) \times 10^{-1}$
Egypt	Granite	$(1.0 - 19) \times 10^{-2}$
The moon surface	Soil	4.5×10^{-1}
Mean world value	Soil	1.6×10^1
Osaka, Japan	Soil	2.0×10^{-3}
Island of Hawaii	Soil	$(25 \pm 6.3) \times 10^{-2}$
Mean world value	Soil	$(1.5 - 2.3) \times 10^1$
New Mexico	Soil	$(32 \pm 4.1) \times 10^0$
Germany	Soil	$(0.20 - 10) \times 10^1$
Mean world value	Soil	$(2.0 - 70) \times 10^0$
Ethiopia (this work)	Solid waste	50.4– 120.2±32.1
Ethiopia (this work)	Soil	56.2 – 58.1±1.4
Ethiopia (this work)	Tantalum ore	41.7– 87.3±21.9

According to Table 6.6-6, the radon-222 exhalation rate for the different rocks and soil samples of the aforementioned countries in the world was significant compared with tantalum ore, soil and solid waste samples collected from the study area. This might be due to the variation in geologic, climatic factors and sample matrix differences of the samples. The maximum annual dose to all radiation workers (male and female) is 20 mSv. There is an additional limit on females of reproductive capacity of 13 mSv in any three-month period. The reason for this additional restriction is to protect a recently conceived fetus within a woman who may be unaware of her pregnancy. In this sense, it is not so much a restriction designed for the protection of females but is a restriction designed to protect a fetus. The estimated annual effective dose at the spots is given in Table 4.6.7 In order to estimate the annual effective dose, over a year, a worker is assumed to be exposed to gamma radiation for 400 h, unshielded, at a distance of 1 m from a large volume of material and to inhale dust from the material for 2000 h. As can be seen from Table 6.6.7, the annual effective dose to workers is above the average permissible 20mSv/year limit

5.3.5 WATER SAMPLES

Water samples collected from Kenticha tantalum mine waste tailing dam, tap water from office, spring water, pure water dam, and Marmora River were identified with the sample codes WD, SW, TWO, TW and WWD. The samples were prepared and counted by gamma spectrometry for 36,000 seconds. However, the resulting gamma spectrum was not appreciably significant for the gamma analysis. It may require other methods of analysis such as gross-alpha and gross-beta counting following alpha spectrometry.

CHAPTER SEVEN

CONCLUSION AND RECOMMENDATION

This chapter provides the main conclusions from the study of NORMs in ore, soil, solid waste and water samples from different mining area of, Ethiopia. It also focuses on recommendations addressed to the various stakeholders that were made based on results from this study.

7.1 CONCLUSIONS

This work considered the evaluation of the level of NORMs and associated radiological hazards and risks from mining activities of Kenticha tantalum mines in Ethiopia, in terms of the exposure to both workers and the public living in and around the vicinity of the mine. This study comprised analysis of samples such as tantalum ore, soil, solid waste, waste tailing and water samples within and around the mine. The NORMs identified in the samples were ^{238}U , ^{232}Th and ^{40}K . The corresponding ranges of activity concentration of ^{238}U , ^{232}Th and ^{40}K were estimated to be 55 – 148 Bq/kg, 12 – 26 Bq/kg and 500 – 730 Bq/kg with average values of 92 Bq/kg, 18 Bq/kg and 628 Bq/kg, respectively. In comparison with the worldwide average value reported by UNSCEAR, the values of ^{238}U and ^{40}K were almost two times higher than the recommended values of 33 Bq/kg and 420 Bq/kg, respectively (UNSCEAR,2000). However, the concentration of ^{232}Th is almost three times lower than the worldwide average of 45 Bq/kg (UNSCEAR, 2000b). According to UNSCEAR 2000 report, the average activity concentration values of the study area are also relatively three times higher than the records of Algeria, China, Egypt, Ghana, Hungary and India; two times higher than Lithuania, Malaysia and United States (see section 4.2). This implies that the NORMs activity concentration at Kenticha tantalum mine is relatively higher with respect to the concentrations of the nuclides; this could be due to geology of granite – pegmatite rocks of the study area, as well as the P^{H} , matrix effect and redox potential of the area. Therefore, these high activity concentrations of the nuclides will influence background radiation to the environment. As a result, special attention should be given to the solid waste, waste tailings and liquid waste, which might be disposed to the environment.

The average absorbed doses rates of the study area from ^{238}U , ^{232}Th and ^{40}K was estimated to be 78 nGy/h, which was relatively higher than 60 nGy/h recommended (UNSCEAR, 2000). This implies that, Kenticha area has higher exposure of background radiation emanating from the significant levels of natural radionuclides. Correspondingly, the annual effective dose (excluding AED from water, food, dust, radon intake; which were not addressed by this research work) of the study area was estimated to be 0.021 mSv/which was much lower than world reported average of 0.07 mSv/y (UNSCEAR, 1993). This value is also lower than the 1 mSv/y and 20 mSv/y dose limit recommended by the ICRP for members of the public and occupational workers, respectively (ICRP, 2007). Concerning the risks to members of the public, the average fatality cancer risks per year, lifetime fatality cancer risk, severe hereditary effects per year and lifetime hereditary effects for this exposure pathway were assessed according to ICRP method. Comparing the results obtained with the EPA acceptable risk factor, all the risk factors revealed no significant hazard to the population of the study area. The average radium equivalent activity values for the ore, soil and solid waste samples were 160 Bq/kg, which was below the internationally accepted value of 370 Bq/kg (UNSCEAR, 2000a). Additionally, the average external and internal hazard indices were below the internationally accepted value of unity (Dragovic et al., 2006; Nada et al., 2009). However, the average representative gamma index was equal to the acceptable value of unity. The recommendation proposed for all building materials must have external hazard, internal hazard and gamma indices would be less than or equal to unity. Therefore, soil or rocks from the study area can be used for construction purposes without posing any significant radiological hazards to humans.

The average radium activity concentration for the ore, soil and solid waste samples was 79 Bq/kg which was two times higher than the world average value of 35 Bq/kg (UNSCEAR, 2000b), while, the average activity concentration of ^{222}Rn in the samples were 126 kBq/m³, this value was relatively higher than the recommended value of 78 kBq/m³ (UNSCEAR, 2000b). The average radon emanation coefficient of the study area was also 79%, this value is significantly higher than the typical emanation coefficients for rocks and soils of 22% in a range of 5% to 70% (Faanu, 2011; Nazaroff et al., 1988; UNSCEAR, 2000). Moreover, the average ^{222}Rn exhalation rate was 64 mBq/m²s; and this is two times higher than the value of 33 mBq/m²s (UNSCEAR, 2000b). These results compared well with results published in UNSCEAR 2000

reports, all are above worldwide values and records of some countries as mentioned in chapter 4 (subsection 4.6). This implies that the area under study has significant levels of ^{222}Rn gas in its environment.

7.2 RECOMMENDATIONS.

Based on the reviews of available data describing NORM occurrence in soil, rocks and other different types of samples from published paper and comparing with the conclusions drawn from this study, preliminary conclusions can be drawn regarding the need for additional research, the need for regulatory authority commitment, the need for Kenticha tantalum mine management body commitment and the need for worker and public responsibilities in order to mitigate impacts of TENORMs on the environment from the mine activities. Hence, the following recommendations are made to the relevant stakeholders.

The facility doesn't incorporate seismic design, I recommend the authority to have a seismic design of PGA 0.75g, thus the facility is situated in seismically active area which needs serious attention of monitoring and follow up, beside the fact that the facility must be at least 30km far from the residential area but its built in the residential hot spot., so the authority must give awareness for the society about the hazardous effects of radioactive waste.

Thus, in order to minimize the radiation doses of the workers and impact on the environment, the authority must; work on the following IAEA code of practice and code of conduct. Proper radiation monitoring exercise to be conducted on the site every six months in order to check the possible rise in radiation level due to accumulation of mineral from various quarry sites. Accurate record keeping is an important part of the control of all radioactive materials. It is important that the presence and subsequent treatment and/or disposal of radioactive waste is recorded. This will ensure the waste can be controlled and reduces the possibility of causing uncontrolled exposure in the environment. The licensee/owner/responsible person should maintain an up-to-date record of all radioactive material in the store. This can be an inventory of current material and a record of their location at all times. There is no disposal policy or strategy so far and no radioactive waste has been formally disposed in Ethiopia. There is no disposal policy or strategy in the State so I recommend the Ethiopian government to have disposal policy and strategy regarding to radioactive source/waste. It appears that there have been no known facilities or practices, which could have radioactive residue or generate radioactive waste in Ethiopia. However, it appears that no formal assessments have ever been made.

Future works

In future studies, determination of gross alpha and gross beta activity concentration in drinking water sources should be done in the study area using a gross alpha and beta counter. For a variety of food products and vegetation of the study area (e.g. teff, enset, maize, sorghum, cabbage, etc), determination of activity concentrations of ^{238}U , ^{232}Th and ^{40}K by gamma spectrometry would be required. This research has also not taken into account NORMs exposure from inhalation of dusts. The annual effective dose from radon was not addressed by this study. Hence, researchers should consider these to enable calculation of overall risk of the study area and avoid over/under estimations of radiation from NORMs. Moreover, researchers could also make their future study in the area more comprehensive by including natural radioactivity baseline (reference) data for ^{238}U , ^{232}Th and ^{40}K using math lab software or other methods. Researchers should also consider other methods of analysis with high precision of sample matrix to confirm the higher values of absorbed doses, activity concentration of ^{238}U , ^{232}Th and ^{40}K which was included in this research. Researchers should also consider determination of rare earth element for the study area, which was not included in this research. Generally, the study area needs further research. The study should gradually be implemented at all other mining areas in the country too. so Assessment of radon concentration and external

Gamma radiation level at different NORM mining area and radiation map of Ethiopia in general is the potential future work.

7.2.1 ETHIOPIAN RADIATION PROTECTION AUTHORITY (ERPA)

The radioactivity associated with NORMs, specifically with ^{238}U and ^{40}K as shown in this study has activity concentrations exceeding the world average. Therefore, it is recommended that the ERPA play the supervisory role to ensure occupational, public and environmental protection. The ERPA should organize further research on NORMs in the study area and other mining industries in the country, such as in potash, salt, gold, etc. mines. The ERPA should also organize basic radiation protection training for the relevant Kenticha tantalum mine management bodies and workers. Results from this study and other similar research could aid in the development of NORM regulations for Ethiopia.

7.2.2 MANAGEMENT OF KENTICHA TANTALUM MINE

Although the estimated values of the radium equivalent activity, hazard indices and annual effective doses all fell within internationally accepted recommended limits, activity concentration and absorbed dose rate as well as the radium and radon concentration, emanation coefficient and exhalation rate of the study area were higher than the worldwide average values. Therefore, the mine plant management needs to ensure the radiological protection of workers and the public by managing solid and liquid wastes of the mines well. Refresher training related to NORMs should be arranged for the personnel regularly to develop their ability of minimizing radiation exposures from mining activities. Proper radiation monitoring exercise to be conducted on the site in order to check the possible rise in radiation level due to accumulation of mineral from various quarry sites; and workers could be provided personal monitoring service (Thermoluminescent dosimeter - TLD) and personal protective equipment like safety glasses, dust masks, safety boots, and gloves to all workers.

7.2.3 PUBLIC AND WORKERS

The annual effective doses from the samples of this study were all within the annual effective dose limit of 20 mSv and 1 mSv for occupationally exposed workers and public, respectively; and all hazard indices as well as the radium equivalent activity were within internationally accepted limits. Based on radiological point of view, it is concluded that all workers within Kenticha tantalum mine are not exposed to any significant radiological hazard. However, there is a need for constant and systematic monitoring of the environment in the study area.

REFERENCES

International Atomic Energy Agency, Manual on Decontamination of Surfaces, Safety Series No 48, Vienna (1979).

International Atomic Energy Agency, Handling, Conditioning and Storage of Spent Sealed Radioactive Sources, Iaea-Tecdoc 1145, Iaea, Vienna (2000).

International Standard – Radiation Protection – Sealed Radioactive Sources – Leakage Test Methods - Iso 9978:1992.

International Atomic Energy Agency, Manual on Decontamination of Surfaces, Safety Series No 48, Vienna (1979).

International Atomic Energy Agency, Handling, Conditioning and Storage of Spent Sealed Radioactive Sources, Iaea-Tecdoc-1145, Iaea, Vienna (2000).

International Standard – Radiation Protection – Sealed Radioactive Sources – Leakage Test Methods - Iso 9978:1992.

International Atomic Energy Agency (2000), Regulations for The Safe Transport of Radioactive

Material, 1996 Edition (Revised), Safety Standards Series No. Ts-R-1 (St-1, Rev.), Iaea, Vienna (2000).

International Atomic Energy Agency (2002), Predisposal Management of Low and Intermediate Level Radioactive Waste, Safety Standards Series No. Ws-G-2.5, Iaea, Vienna.

International Atomic Energy Agency (2011), Radiation Protection and Norm Residue Management in the Production of Rare Earths from Thorium Containing Minerals

International Commission On Radiological Protection (1991), Recommendations of The International Commission on radiological Protection, Publication 60, Pergamon Press, Oxford And New York.

International Commission On Radiological Protection (2000), Protection of The Public In Situations of Prolonged Radiation Exposure, Publication 82, Elsevier Science, Oxford.

International Atomic Energy Agency (1996a), International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources. Safety Report Series 115

International Atomic Energy Agency (2006), Assessing The Need for Radiation Protection Measures in Work Involving Minerals and Raw Materials. Safety Report Series 49.

International Atomic Energy Agency (2002), Management of Radioactive Waste from the Mining and Milling of Ores. Safety Guide Ws-G-1.2, Iaea, Vienna.

International Atomic Energy Agency (1996b), Regulations for the Safe Transport of Radioactive Material. Safety Standards Series St-1.

International Atomic Energy Agency (2003), Extent of Environmental Contamination by Naturally Occurring Radioactive Material (Norm) and Technological Options for Mitigation Technical Report.

International Atomic Energy Agency (2004), Application of the Concepts of Exclusion, Exemption and Clearance, Iaea Safety Standards Series.

APPENDICES

APPENDIX I: Sampling co-ordinates for ore, soil, and solid waste and water samples from study area

Sample code	Description of sampling location	Location coordinates	
		Latitude	Longitude
WD	pure water from water dam, dam 2	5°27'13.79"N	39° 0'36.82"E
SW	Spring water from the spring	5°27'19.24"N	39° 0'44.52"E
TWO	Tape water from office area	5°27'5.27"N	39° 0'41.53"E
TW	Tape water taken from village around mine	5°26'50.46"N	39° 0'24.91"E
WWD	Waste water taken from waste dam, Dam1	5°27'9.73"N	39° 0'45.81"E
WT1	Waste sample from dam 1	5°27'10.53"N	39° 0'49.07"E
WT2	Waste samples from dam 2	5°27'13.37"N	39° 0'38.29"E
WT3	Waste samples from dam 1	5°27'13.27"N	39° 0'56.08"E
WR	Waste for recycle found between the dam 1 and dam 2	5°27'12.95"N	39° 0'42.21"E
WT4	Waste samples around processing plant	5°27'15.05"N	39° 1'9.07"E
WJT	Waste sample taken from beneath Jigging table	5°27'13.63"N	39° 1'2.42"E
FeOW	Iron oxide waste from waste iron ore store	5°27'4.45"N	39° 0'31.68"E
FW	Full waste disposed to the environment	5°27'5.38"N	39° 1'6.34"E
ROC	Rich ore concentrated taken processing plant root	5°27'13.84"N	39° 1'2.03"E
TaO1	Tantalum ore from open pit 1	5°27'21.84"N	39° 1'15.85"E
TaO2	Tantalum ore from open pit 2	5°27'38.84"N	39° 1'25.47"E
TaO3	Tantalum ore from open pit 3	5°27'48.87"N	39° 1'29.30"E
TaO4	Tantalum concentrate	5°27'13.99"N	39° 1'1.69"E
SS1	Soil sample taken near to spring water	5°27'22.24"N	39° 0'47.26"E
SS2	Soil sample taken from around the village	5°26'53.56"N	39° 0'32.38"E
SS3	Soil sample taken from near to waste tailing dam	5°27'18.86"N	39° 0'50.38"E
SS4	Soil sample taken from around processing plant	5°27'19.13"N	39° 1'0.14"E
SS5	Soil sample taken from around waste disposed	5°27'37.37"N	39° 1'21.10"E

Appendix II. Disused radioactive waste in the storage facility (Transferred to ERPA Interim Waste Storage)

No	I Practice	II Radionuclied	III Quantity in TBq	IV D-value TBq	V Ratio of A/D	VI category	
						A/D based	Assigne d
1	Brachythera py low doserate	Cs-137	1.74E-03	1.00E-01	1.74E-02	4	4
		Cs-137	2.80E	1.00E-01	2.80E-02	4	4
		Cs-137	3.95E-03	1.00E-01	3.95E-02	4	4
		Cs-137	2.81E-03	1.00E-01	2.81E-02	4	4
		Cs-137	1.73E-03	1.00E-01	1.73E-02	4	4
2	Calibration	Cs-137	7.40E-03	1.00E-01	7.40E+00	3	
	Check Source	Sr-90	3.33E-02	4.00E+00	8.33E-03	5	
		Sr-90	1.11E-05	4.00E+00	2.78E-06	5	
3	Level Guage	Am-241	1.67E-03	6.00E-02	2.78E-02	4	
		Am-241/Be	1.67E-03	6.00E-02	2.78E-02	4	
		Am-241	1.67E-03	6.00E-02	2.78E-02	4	
		Am-241	1.67E-03	6.00E-02	2.78E-02	4	
		Am-241	1.67E-03	6.00E-02	2.78E-02	4	
4	Mositure density gauges	Cs-137	3.70E-04	1.00E-01	3.70E-03	5	4
		Am-241/Be	1.48E-03	6.00E-02	2.47E-02	4	4
		Cs-137	3.70E-04	1.00E-01	3.70E-03	5	4
		Am-241/Be	1.85E-03	6.00E-02	3.08E-02	4	4
		Cs-137	2.96E-04	1.00E-01	2.96E-03	5	4
		Am-241/Be	1.48E-03	6.00E-02	2.47E-02	4	4
		Cs-137	7.40E-05	1.00E-01	7.40E-04	5	
		Am-241/Be	1.48E-03	6.00E-02	2.47E-02	4	4
		Cs-137	2.96E-04	1.00E-01	2.96E-04	5	4
		Am-241/Be	1.48E-03	6.00E-02	2.47E-02	4	4
		Cs-137	2.96E-04	1.00E-01	2.96E-03	5	4
		Am-241/Be	1.48E-03	6.00E-02	2.47E-02	4	4
		Cs-137	2.96E-04	1.00E-01	2.96E-03	5	4
		Am-241/Be	1.48E-03	6.00E-02	2.47E-02	4	4
		Cs-137	2.96E-04	1.00E-01	2.96E-03	5	4
		Am-241/Be	1.48E-03	6.00E-02	2.47E-02	4	4
		Cs-137	3.70E-04	1.00E-01	3.70E-03	5	4
		Am-241/Be	1.85E-03	6.00E-02	3.08E-02	4	4
		Cs-137	3.70E-04	1.00E-01	3.70E-03	5	4
		Am-241/Be	2.96E+00	6.00E-02	4.93E+01	4	4
		Cs-137	2.96E-04	1.00E-01	2.96E-03	5	4
		Am-241/Be	1.48E-03	6.00E-02	2.47E-02	4	4
		Cs-137	2.96E-04	1.00E-01	2.96E-03	5	4
		Am-241/Be	1.48E-03	6.00E-02	2.47E-02	4	4
		Cs-137	2.96E-04	1.00E-01	2.96E-03	5	4
		Am-241/Be	1.48E-03	6.00E-02	2.47E-02	4	4
		Cs-137	2.96E-04	1.00E-01	2.96E-03	5	4
Am-241/Be	1.48E-03	6.00E-02	2.47E-02	4	4		
Cs-137	2.96E-04	1.00E-01	3.70E-03	5	4		
Am-241/Be	1.48E-03	6.00E-02	2.47E-02	4	4		
Cs-137	3.70E-04	1.00E-01	2.96E-03	5	4		

No	I Practice	II Radionuclied	III Quantity in TBq	IV D-value TBq	V Ratio of A/D	VI category	
4	Mositure density gauges	Am-241/Be	1.48E-03	6.00E-02	2.47E-02	4	4
		Cs-137	2.96E-04	1.00E-01	2.96E-03	5	4
		Am-241/Be	1.48E-03	6.00E-02	2.47E-02	4	4
		Cs-137	2.96E-04	1.00E-01	2.96E-03	5	4
		Am-241/Be	1.48E-03	6.00E-02	2.47E-02	4	4
		Cs-137	2.96E-04	1.00E-01	2.96E-03	5	4
		Am-241/Be	1.48E-03	6.00E-02	2.47E-02	4	4
		Cs-137	2.96E-04	1.00E-01	2.96E-03	5	4
		Am-241/Be	1.48E-03	6.00E-02	2.47E-02	4	4
		Cs-137	2.96E-04	1.00E-01	2.96E-03	5	4
		Am-241/Be	1.48E-03	6.00E-02	2.47E-02	4	4
		Cs-137	2.96E-04	1.00E-01	2.96E-03	5	4
		Am-241/Be	1.48E-04	6.00E-02	2.47E-02	4	4
		Cs-137	2.96E-04	1.00E-01	2.96E-03	5	4
		Am-241/Be	1.48E-04	6.00E-02	2.47E-02	4	4
		Cs-137	2.96E-04	1.00E-01	2.96E-03	5	4
		Am-241/Be	1.48E.03	6.00E-02	2.47E-02	4	4
		Cs-137	3.70E-04	1.00E-01	3.70E-03	5	4
		Am-241/Be	1.58E-02	6.00E-02	2.63E-01	4	4
		Cs-137	3.70E-04	1.00E-01	3.70E-03	5	4
		Am-241/Be	1.48E.03	6.00E-02	2.47E-02	4	4
		Cs-137	3.70E-04	1.00E-01	3.70E-03	5	4
		Am-241/Be	1.48E.03	6.00E-02	2.47E-02	4	4
		Cs-137	3.70E-04	1.00E-01	3.70E-03	5	4
		Am-241/Be	1.48E.03	6.00E-02	2.47E-02	4	4
		Cs-137	3.70E-04	1.00E-01	3.70E-03	5	4
		Am-241/Be	1.48E.03	6.00E-02	2.47E-02	4	4
		Cs-137	3.70E-04	1.00E-01	3.70E-03	5	4
		Am-241/Be	1.48E.03	6.00E-02	2.47E-02	4	4
		Cs-137	3.70E-04	1.00E-01	3.70E-03	5	4
		Am-241/Be	1.85E-03	6.00E-02	3.08E-02	4	4
		Cs-137	3.70E-04	1.00E-01	3.70E-03	5	4
		Am-241/Be	1.85E-03	6.00E-02	3.08E-02	4	4
		Cs-137	2.96E-04	1.00E-01	2.96E-03	5	4
		Am-241/Be	1.48E.03	6.00E-02	2.47E-02	4	4
		Cs-137	3.70E-04	1.00E-01	3.70E-03	5	4
		Am-241/Be	1.85E-03	6.00E-02	3.08E-02	4	4
		Cs-137	3.70E-04	1.00E-01	3.70E-03	5	4
		Am-241/Be	1.85E-03	6.00E-02	3.08E-02	4	4

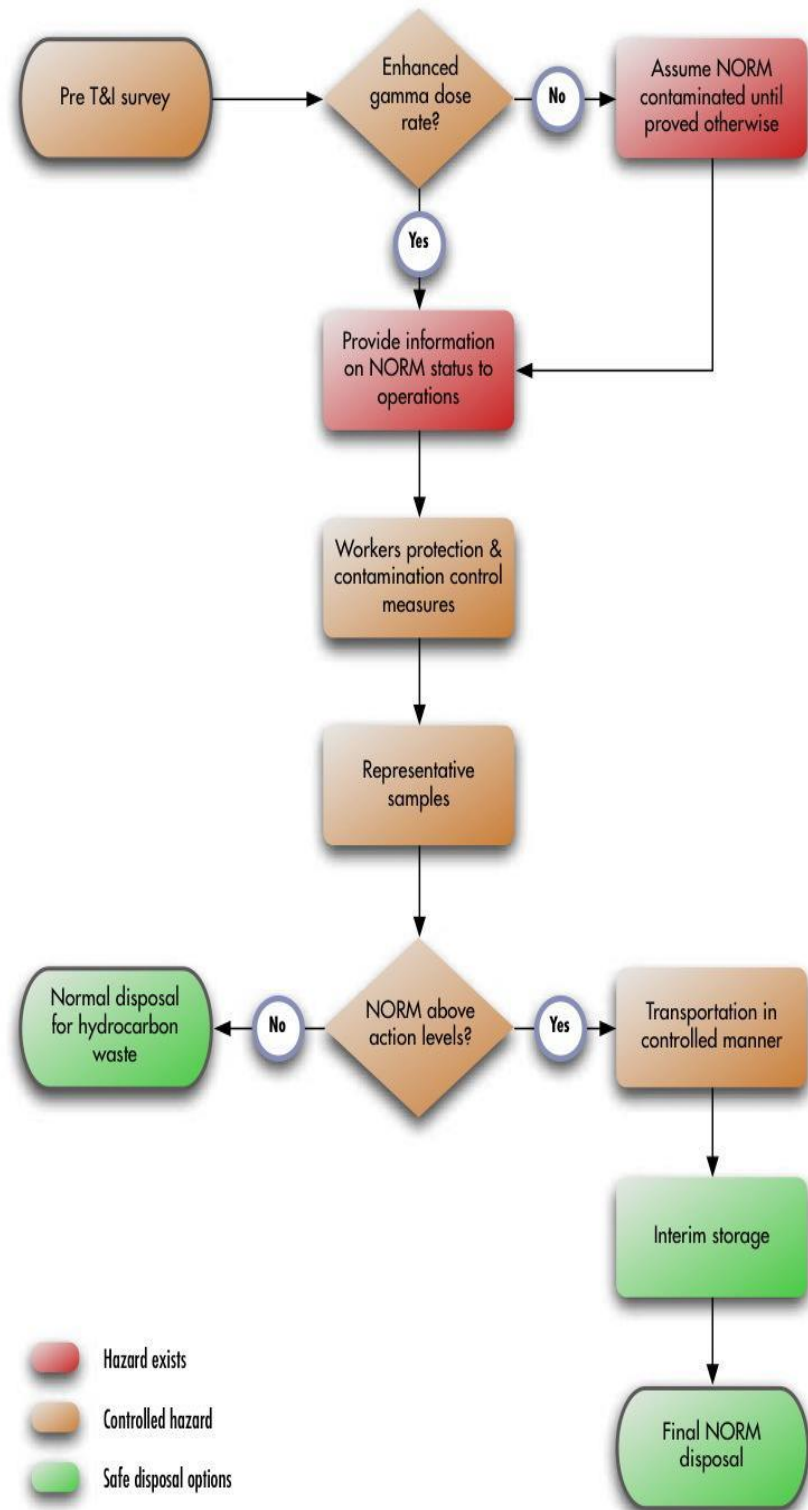
No	I Practice	II Radionuclied	III Quantity in TBq	IV D-value TBq	V Ratio of A/D	VI category			
4	Mositure density gauges	Am-241/Be	1.85E-03	6.00E-02	3.08E-02	4	4		
		Cs-137	3.70E-04	1.00E-01	3.70E-03	5	4		
		Am-241/Be	1.85E-03	6.00E-02	3.08E-02	4	4		
		Cs-137	3.70E-04	1.00E-01	3.70E-03	5	4		
		Am-241/Be	1.85E-03	6.00E-02	2.47E-02	4	4		
		Cs-137	3.70E-04	1.00E-01	3.70E-03	5	4		
		Am-241/Be	1.85E-03	6.00E-02	3.08E-02	4	4		
		Cs-137	3.70E-04	1.00E-01	3.70E-03	5	4		
		Am-241/Be	1.85E-03	6.00E-02	3.08E-02	4	4		
		Cs-137	2.96E-04	1.00E-01	2.96E-03	5	4		
		Am-241/Be	1.48E.03	6.00E-02	2.47E-02	4	4		
		Cs-137	2.96E-04	1.00E-01	2.96E-03	5	4		
		Am-241/Be	1.48E.03	6.00E-02	2.47E-02	4	4		
		Cs-137	2.96E-04	1.00E-01	2.96E-03	5	4		
		Am-241/Be	1.48E.03	6.00E-02	2.47E-02	4	4		
		Cs-137	2.96E-04	1.00E-01	2.96E-03	5	4		
		Am-241/Be	1.48E.03	6.00E-02	2.47E-02	4	4		
		Cs-137	3.70E-04	1.00E-01	3.70E-03	5	4		
		Am-241/Be	1.85E-03	6.00E-02	3.08E-02	4	4		
		Cs-137	2.96E-04	1.00E-01	2.96E-03	5	4		
		Am-241/Be	1.48E.03	6.00E-02	2.47E-02	4	4		
		Cs-137	2.96E-04	1.00E-01	2.96E-03	5	4		
		Am-241/Be	1.48E.03	6.00E-02	2.47E-02	4	4		
		Cs-137	2.96E-04	1.00E-01	2.96E-03	5	4		
		Am-241/Be	1.48E.03	6.00E-02	2.47E-02	4	4		
		Cs-137	2.96E-04	1.00E-01	2.96E-03	5	4		
		Am-241/Be	1.48E.03	6.00E-02	2.47E-02	4	4		
		Cs-137	2.96E-04	1.00E-01	2.96E-03	5	4		
		Am-241/Be	1.48E.03	6.00E-02	2.47E-02	4	4		
		Cs-137	2.96E-04	1.00E-01	2.96E-03	5	4		
		Am-241/Be	1.48E.03	6.00E-02	2.47E-02	4	4		
		Cs-137	2.96E-04	1.00E-01	2.96E-03	5	4		
		Am-241/Be	1.48E.03	6.00E-02	2.47E-02	4	4		
		Cs-137	1.11E-04	4.00E-02	2.78E-03	5	4		
				Am-241/Be	7.40E-.03	6.00E-02	1.23E-03	4	

No	I Practice	II Radionuclied	III Quantity in TBq	IV D-value TBq	V Ratio of A/D	VI category	
						A/D based	Assigned
5	Research	Co-60	3.70E-04	3.00E-02	1.23E+01	2	
		Co-60	9.25E+00	3.00E-02	3.08E+02	2	
6	Sterilization	Co-60	9.04E+02	3.00E-02	3.01E+04	1	1
		Co-60	2.00E+02	3.00E-02	6.67E+03	1	1
7	Teletherapy	Co-60	3.70E+02	3.00E-02	1.23E+04	1	1
		Cs-137	7.40E-02	1.00E-01	1.23E+04	3	
8	Well logging	Am-241/Be	8.51E-04				
		Am-241/Be	2.04E-02				
		Am-241/Be	7.40E-01				
9	Density gauges	Cs-137	1.11E-02	1.00E-01	1.11E-01	4	3
		Cs-137	1.11E-02	1.00E-01	1.11E-01	4	3
		Cs-137	1.11E-02	1.00E-01	1.11E-01	4	3

Appendix III, concrete mix design for conditioning of radioactive source.

Strength	Cement	Sand	W:C	Slump	28 days (N/mm ²)								
					MPa	kg/m ³	%	mm	1	2	3	Average	4
C-30	410	39	0.5	165	17.1	19.8	21.1	19.3					
C-30	400	39	0.5	130	20.3	20.6	20.0	20.3	31.0	30.5	30.3	30.6	
C-15	280	34	0.80	105	10.7	12.7	12.9	12.1	20.6	19.9	18.1	19.5	
C-15	280	30	###	65	11.4	11.7	12.1	11.7	20.9	18.4	18.1	19.1	
C-30	400	33	0.53	120	25.5	25.8	22.3	24.5	35.4	31.5	29.6	32.1	
C-30	400	33	0.48	130	26.0	26.3	24.8	25.7	37.8	38.9	37.9	38.2	
C-15	280	33	0.60	130	20.3	14.6	19.1	18.0	27.4	28.6	26.2	27.4	
C-15	280	33	0.6	105	20.1	21.5	19.8	20.4	26.3	28.7	26.6	27.2	
C-15	280	37	0.6	140	9.8	11.7	11.8	11.1	16.4	17.4	17.9	17.2	
C-30	400	37	0.48	110	30.2	30.5	30.5	30.4	37.1	42.2	39.9	39.7	

Appendix IV: Process cycle of NORM Management



Appendix V: Radiation Dose level at the Project site

	Description	U-238			Th-232					
	Concentration in the feedstock (ppm) 0.81% U3O5	8100			700					
	Activity (Bq/gm of concentrate)	84.84			2.13					
Hot Spot	Over a year, a worker is assumed to be exposed to gamma radiation for 400 h, unshielded, at a distance of 1 m from a large volume of material and to inhale dust from the material for 2000 h.	U-238 Activity (Bq/g)	Effective dose (mSv/a per Bq/g)			Th-232 Activity (Bq/g)	Effective dose (mSv/a per Bq/g)			ANNUAL EFFECTIVE DOSE (TOTAL) mSv/a
			Gamma Radiation	Dust Inhalation	Total Effective Dose (Rounded)		Gamma Radiation	Dust Inhalation	Total Effective Dose (Rounded)	
1	Transport: Mining to Storage @	84.84	0.328		27.83	2.13	0.176		0.38	28.20
2	Unloading to store	84.84	0.328	0.51	8.89	2.13	0.176	0.108	0.08	8.96
3	Store @ Diba	84.84	0.328	0.51	4.44	2.13	0.176	0.108	0.04	4.48
4	Store to crusher	84.84	0.328	1.02	114.36	2.13	0.176	0.216	0.84	115.20
5	Crushing	84.84	0.328	0.51	71.09	2.13	0.176	0.108	0.61	71.70
6	Crusher to Digester	84.84	0.328	0.51	71.09	2.13	0.176	0.108	0.61	71.70
7	Digester to Filter	84.84	0.328	0.51	71.09	2.13	0.176	0.108	0.61	71.70
8	Filtering Process	282.79	0.328		92.75	7.12	0.176		1.25	94.01
9	Filter to Solid waste pile	282.79	0.328		23.19	7.12	0.176		0.31	23.50
10	Filter to Liquid Waste (Transport)	0.00	0.328	0.51	0.00	0.00	0.176	0.108	0.00	0.00
11	Solid waste interim store	282.79	0.328	0.51	14.81	7.12	0.176	0.108	0.13	14.94
12	Solid waste transport	282.79	0.328		92.75	7.12	0.176		1.25	94.01
13	Solid waste disposal	282.79	0.328	0.51	236.98	7.12	0.176	0.108	2.02	239.00