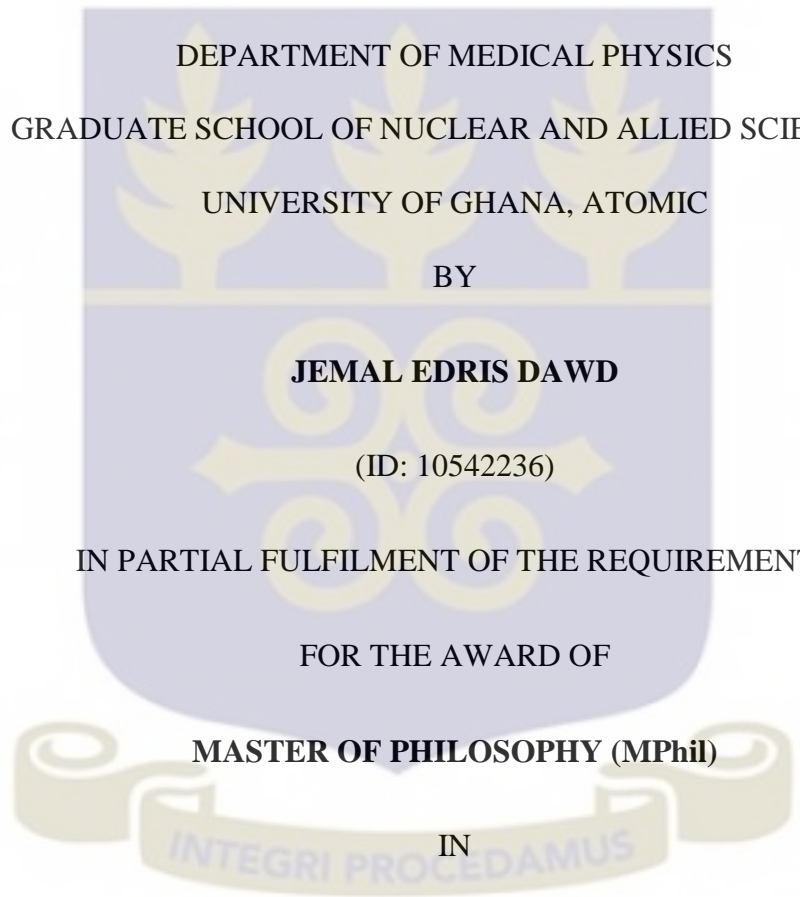


EVALUATION OF THE LEVEL OF NORMS AND ASSOCIATED
RADIOLOGICAL HAZARDS & RISKS FROM MINING ACTIVITIES OF
KENTICHA TANTALUM MINES IN ETHIOPIA

THIS THESIS IS SUBMITTED TO THE
DEPARTMENT OF MEDICAL PHYSICS
GRADUATE SCHOOL OF NUCLEAR AND ALLIED SCIENCES,
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BY
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IN PARTIAL FULFILMENT OF THE REQUIREMENTS
FOR THE AWARD OF
MASTER OF PHILOSOPHY (MPhil)
IN
NUCLEAR SCIENCE AND TECHNOLOGY

The background of the text area features a large, semi-transparent watermark of the University of Ghana crest. The crest is a shield-shaped emblem with a blue background and yellow/gold decorative elements, including a central circular motif and a banner at the bottom with the Latin motto "INTEGRI PROCEDAMUS".

July 2016

DECLARATION

This thesis is the result of research work undertaken by Jemal Edris Dawd towards the award of the degree of Master of Philosophy in Nuclear Science and Technology in the Department of Medical Physics under the School of Nuclear and Allied Sciences, University of Ghana, under close supervision of Prof. Emmanuel Ofori Darko and Rev. (Dr.) Samuel Akoto Bamford.

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SUPERVISOR'S DECLARATION

We hereby declare that the preparation and presentation of this thesis were supervised in accordance with guidelines on supervision of thesis laid down by the University of Ghana.

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(CO-SUPERVISOR)

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DATE

DEDICATION



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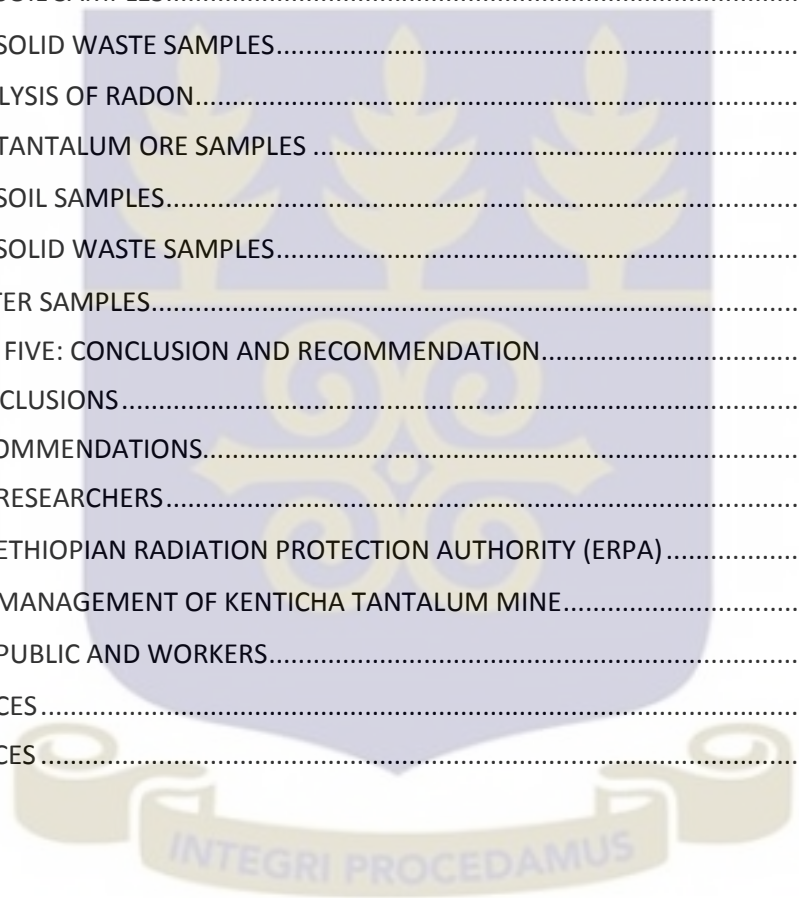
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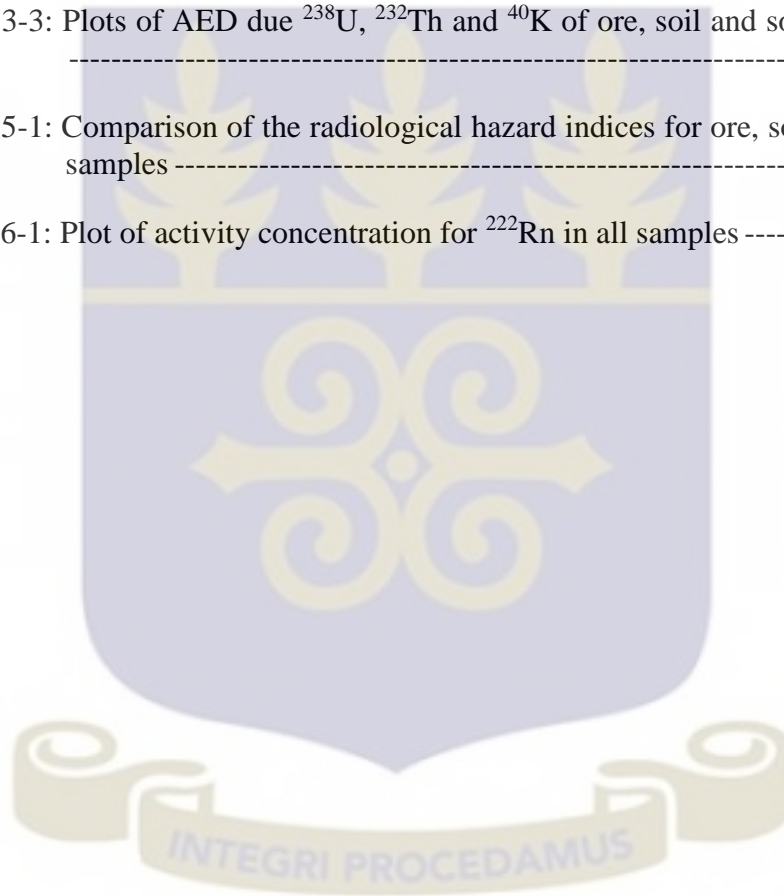
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LIST OF ABBREVIATIONS

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
1M HNO ₃	1 Molar nitric acid
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} Rn	Randon-222, radon-220 and radon-219
^{226,224,223} 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
$\text{Bq/m}^2\text{s}$	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
$D_{,ext}$	average external gamma dose rate in $\mu\text{Gy/h}$
e_A	Error in activity concentration (Bq/kg)
EC	Electron capture/radon emanation coefficient
e.g.	For example

EMDSC	Ethiopian Mineral Development Share Company
ERPA	Ethiopian Radiation Protection Authority
Err	Error
etc.	et cetera
f	Radon emanation factor
Fe ₂ O ₃	Iron (III) oxide
Fe ₃ O ₄	Iron (II, III) oxide, magnetite
FWHM	Full Width at Half Maximum
GAEC	Ghana Atomic Energy Commission
GPS	Global Positioning System
H _{ext}	External hazard index
HF	Hydrofluoric acid
H _{int}	Internal hazard index
HNRC	hereditary nominal risk coefficient and
HPGe	High Purity Germanium Detector
I ₁	Activity of radon at t ₁
I ₂	Activity of radon at secular equilibrium
IAEA	International Atomic Energy Agency
ICRP	International Commission on Radiological Protection
I	representative gamma index
kBq/kg	kilo Becquerel per kilogram
kBq/m ³	kilo Becquerel per cubic meter
K-electron	K-shell electron

K-salt	potassium fluorotantalate, potassium fluorotantalate, potassium heptafluorotantalate, or potassium tantalum fluoride
k_T	Partition coefficient of radon between the water and air phases
L	Diffusion length
LET	(Low) linear Energy Transfer
LNT model	Linear Non-threshold model
m	Fraction of the porosity that is water filled/weight of the sample container/
m/s	Meter per second
m^2/s	meter square per second
MCA	Multi channel analyzer
MDA	Minimum detectable activity
MeV	Mega electron volt
mg/L	Milligram per liter
mm	Millimeter
MME	Ministry of Mine and Energy of Ethiopia
MnO_2	Manganese (IV) oxide
N	^{222}Rn activity at radioactive equilibrium i.e. at t_2 , and
NAS	National Academy of Sciences
N_B	background counts in the region of interest for a particular radionuclide
Nb_2O_5	Niobium pentoxide
N_D	net counts of the radionuclide in the samples
nGy/h	Nano gray per hour
NORM	Naturally Occurring Radioactive Material

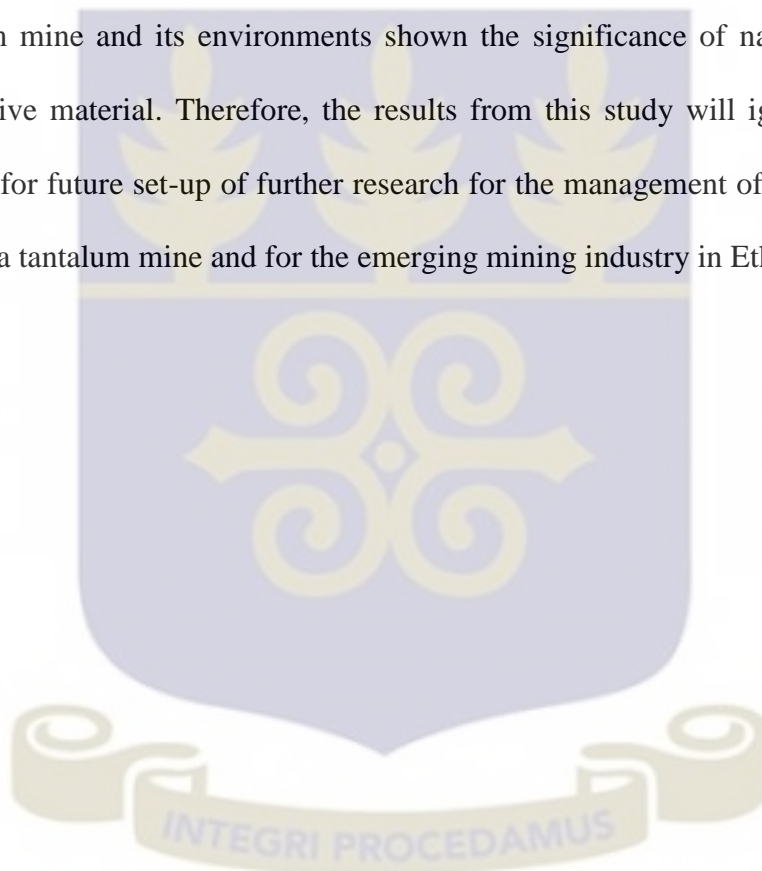
N-S	North to South
°C	degree Celsius
Ore	Tantalum ore
P_{γ}	gamma line emission probability
ppm	Parts per million
QDL	Qualitative Detection Limit
Ra	Radium
Rn	Radon
RPI	Radiation Protection Institute of Ghana
S	second
SEAMIC	Southern and eastern African mineral center
SNAS	(Graduate) School of Nuclear and Allied Sciences
STD	standard deviation
Sv	Sievert
Sv/Gy	Sievert per gray
Sv/y	Sieverts per year
Ta	Tantalum
t_1	Counting time after sealing
t_2	Counting time after 30 days (10 hours),
Ta ₂ O ₅	Tantalum pent oxide
t_d	delay time between sampling and counting
TE-NORM	Technologically enhanced naturally occurring radioactive
T_{exp}	annual exposure time

Th	Thorium
TIC	Tantalum-Niobium international Study Center
TiO ₂	Titanium (IV) oxide
U	Uranium
T _{std}	counting time and
UK	United Kingdom
UN	United Nations
UNSCEAR	UN Scientific Committee on the Effects of Atomic Radiation
US	United State
USEPA	US Environmental Protection Agency
USGS	United State Geological Survey
V	Volt
VDU	Visual Display Unit
WAV	Worldwide Average Value
WNA	World Nuclear Association
W _{samp}	mass of the sample (kilogram) or volume (liter)
	Alpha
	Decay constant for the specific radionuclide
s	Soil grain density
T _C	counting time and
ε _f	photo peak efficiency,

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 ± 1.431 Bq/kg, 24.945 ± 0.492 Bq/kg, 603.170 ± 55.013 Bq/kg, 69.478 ± 31.0 Bq/kg and 112.554 ± 50.249 kBq/m³, respectively. In soil the activity concentrations were 69.354 ± 1.081 Bq/kg, 15.479 ± 0.231 Bq/kg, 718.880 ± 65.531 Bq/kg, 68.923 ± 1.7 Bq/kg and 111.655 ± 2.681 kBq/m³, respectively and in solid waste samples 110.496 ± 1.907 Bq/kg, 15.009 ± 0.274 Bq/kg, 607.269 ± 55.375 Bq/kg, 98.300 ± 38.6 Bq/kg and 159.246 ± 62.607 kBq/m³ 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 ± 0.003 mSv, 0.020 ± 0.001 mSv and 0.023 ± 0.004 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 ± 25.7 mBq/m².s for ores, from 56.2 ± 1.4 to 58.1 ± 1.4 mBq/m².s for soil and from 53.3 ± 32.1 to 120.2 ± 32.1 mBq/m².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.



CHAPTER ONE

INTRODUCTION

This chapter provides a brief introduction to the NORMs, a brief background to the study, the associated problem statement, the objectives, the relevance and the justification of this study.

1.1 BACKGROUND OF THE STUDY

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.

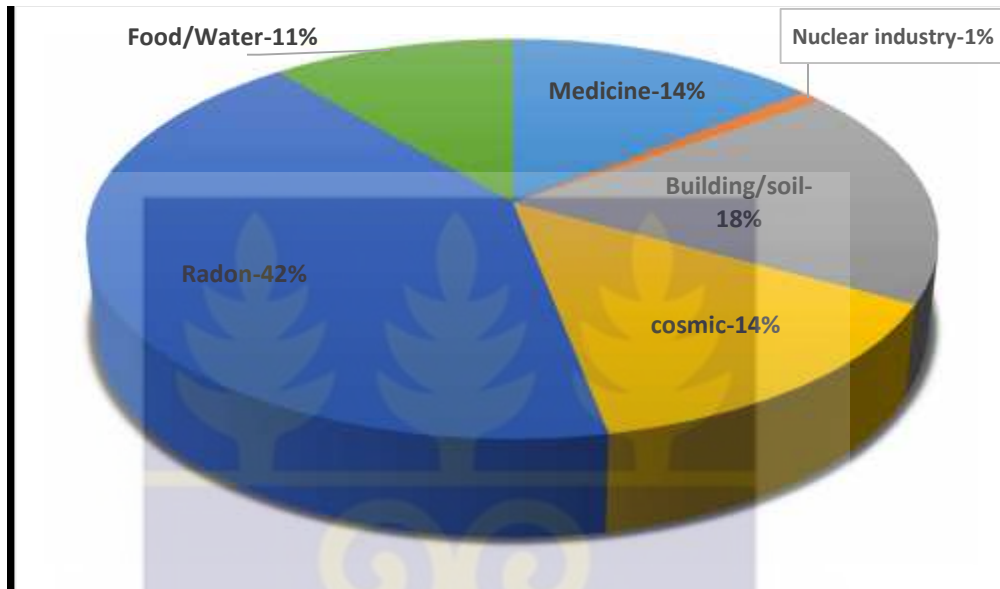


Figure 1.1-1: The world average annual dose from various sources (Todsadol, 2012).

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.

1.2 STATEMENT OF THE PROBLEM

This research proposed to evaluate NORM at Kenticha tantalum mine, 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 radium ^{226}Ra and its daughter, ^{238}U and daughter and ^{232}Th 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 coltan, 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 TENORM 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.

1.3 RESEARCH OBJECTIVES

PRIMARY 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 its immediate environs.
 - 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.

1.4 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 had 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.

1.5 SCOPE OF THE STUDY

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, waste, waste tailing and water samples from Kenticha tantalum mine in Ethiopia by deciding on independent sample location to have representative sample.

1.6 STRUCTURE OF THIS RESEARCH

This research paper consists of five 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 results 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 five concludes the study and gives an overall summary of the research, recommendations and lessons learned. Any limitations of the experimental design of this research were elaborated in this chapter.



CHAPTER TWO

LITERATURE 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 REVIEW OF NORM

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 40 Bq/kg. Granites and black-shales are occasionally rich in these elements, with typical concentrations of up to 200 Bq/kg while Monazite can contain uranium and thorium at up to 200 kBq/kg (Sobanski, 2004). The world nuclear association report said, the Australian mineral concentrate contains thorium and uranium with activity concentration of 0.3 - 3.0 Bq/kg and 0 - 0.8 Bq/kg, respectively. Also it said that the processed tantalum 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 ^{232}Th . In one niobium facility in Brazil, activity levels in waste ranged up to 200,000 Bq/kg of ^{228}Ra (in barium

sulphate) and 117,000 Bq/kg of ^{232}Th (in the slag) (UNSCEAR, 2008a). Whereas, phosphate ores typically contain about 1,500 Bq/kg of uranium and radium; although some phosphate rocks contain upto 20,000 Bq/kg of U_3O_8 . 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 2.1-1: 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 Ramsar in 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 2.1-2: 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 doses 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 ^{40}K . 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 78 kBq/m^3 (UNSCEAR, 2000b).

2.2 INFORMATION ON TANTALUM

Tantalum is a rare, hard, and 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 almost completely 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 coefficient 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, doesn't 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.

2.3 USES OF TANTALUM

Tantalum was discovered in 1802 by Swedish scientist, A.G. Ekeberg (Nete, 2013) but commercial use of tantalum began in Germany in 1903 with the production of tantalum wire to replace carbon in incandescent light filaments (Cunningham, 2000) before the advent of cheap tungsten wire (Hayes and Burge, 2003). The 1940s saw the introduction of tantalum to its key role in the production of capacitors and demand for the metal increased dramatically concurrent with the development of radar and military radio communications (Ndoli, 2013; Hayes and Burge, 2003). Since then, the demand for tantalum has increased steadily over the past two decades due to their importance in the production of modern industrial materials and high tech consumer products (Ndoli, 2013; BGS, 2011). Tantalum is used in components for items such as cell phones, hearing aids, and hard drives. Tantalum's low mechanical strength and high biocompatibility allow it to coat stronger substrates, like stainless steel, for medical applications. It is used for blood vessel support stents, plates, bone replacements, and suture clips and wire. In the chemical industry, tantalum's corrosion resistance makes it useful as a lining for pipe, tanks, and vessels. Tantalum oxide can increase the refractive index of lens glass, while the hardness of tantalum carbide makes it an ideal component in the manufacture of cutting tools (Ndoli, 2013; EMDSC, 2013; Cunningham, 2000; Luis, 2012). Currently, Ethiopia has been exporting tantalum concentrate to western countries without any value added and not used as an input for industrial manufacturing in Ethiopia.

2.4 SOURCES AND RESERVES OF TANTALUM

Even though, the International Strategic Minerals Inventory Summary Report said the world estimates of niobium and tantalum resources are fragmentary and incomplete, there is about 150,000 tons of tantalite with 0.7 ppm (BGS, 2011) contained in ores

from resources around the world (Joel, 2005; Richard et al, 1993). Today, tantalum reserves amount is almost 53,000 tons at existing mines (Joel, 2005). In addition, Fauna and Flora International report indicates that the world tantalum supply is obtained from mine production (58%), synthetic concentrates (9%), recycling (24%) and stockpiles (9%) (Hayes and Burge, 2003). There are no precise reported reserve evaluations that have been established for African deposits although some countries have been producing for more than 40 years (Joel, 2005). Tantalum mineralization has also been reported in 17 countries on the African continent including Ethiopia. In Ethiopia, a deposit in weathered crust was delineated in 1988 with proven reserves of 25,850 tons (17.2% of the world reserve) of columbo– tantalite ore at 0.02 to 0.03% Ta₂O₅ (SEAMIC, 2006). Whereas the reserve of Ta₂O₅ in the weathered zone was calculated to be 2,400 tons (14.12% of country reserve) at 0.015% Ta₂O₅. Currently, mining is operated by a state-owned company, Ethiopia Mineral Development Share Company (EMDSC) since 1990, at Kenticha, about 600 km south of Addis Ababa and 50km southeast of Shakiso in the Adola Greenstone Belt. The Kenticha tantalum production is around 190 tons of Ta₂O₅ per year (MME, 2009). Figure 2.4-1 shows tantalum and niobium mineral deposits in the globe. Brazil and Australia are the major world producers of these metals followed by Canada, Mozambique and Ethiopia (Hayes and Burge, 2003; Nete, 2013).



Figure 2.4-1: The world coltan distribution (■ symbol) (Nete, 2013)

2.5 RADIOACTIVE NATURE OF TANTALUM

Tantalum ores at Kenticha derived from pegmatite and granite rocks, comprise a wide variety of more than a hundred minerals. Environmental concerns of mining activities are mainly associated with solid/liquid/gaseous wastes containing radioactive substances which may elevate the background radiation from naturally occurring materials and thus may affect human radiation dose. Due to this potential risk, there have been recent changes to the regulations recommended by the International Atomic Energy Agency (IAEA) resulting in a lowering of the radioactivity limit in tantalum materials transported across the globe as normal goods due to varying amounts of ^{238}U and ^{232}Th decay chains as well as ^{40}K . These elements remain in the concentrate after the initial ore treatment. The industry association of Tantalum-Niobium international Study Center (TIC) has conducted a survey and found that the majority of concentrates produced have radioactivity levels of up to 40,000Bq/kg (Joel, 2005). The IAEA new regulation has moved the ceiling of radioactivity units in tantalum materials from

70,000 down to 10,000 Bq/kg and possibly less (Joel, 2005). This implies that many offered concentrates are classified as dangerous goods. This is a serious issue that the tantalum industry should work on a more manageable limit of radioactivity for the whole industry (Joel, 2005).

2.6 MINERAL PROCESSING OF TANTALUM ORE

Of the more than 70 identified tantalum containing minerals, tantalite $[(\text{Fe},\text{Mn})(\text{Ta},\text{Nb})_2\text{O}_6]$, microlite $[(\text{Na},\text{Ca})\text{Ta}_2\text{O}_6(\text{O},\text{OH},\text{F})]$, and wodginite $[(\text{Ta},\text{Nb},\text{Sn},\text{Mn},\text{Fe})\text{O}_2]$ are identified as greatest economic importance in the world for tantalum (USGS, 2014b). However, the major source is the columbite-tantalite mineral, also called coltan, $(\text{Fe},\text{Mn})(\text{Nb},\text{Ta})_2\text{O}_6$, (Figure 2.6-1). Coltan contains (5 – 30 Ta_2O_5 , wt.% in columbite and 42 – 84 Ta_2O_5 , wt.% in tantalite) and (55 – 78 Nb_2O_5 , wt.% in columbite and 2 – 40 Nb_2O_5 , wt.% in tantalite)(Richard et al, 1993). Owing to the low grades of tantalum ore, it is beneficiated (a physical process that doesn't chemically change the material) to tantalum concentrate that is 20 – 40% Ta_2O_5 equivalent.

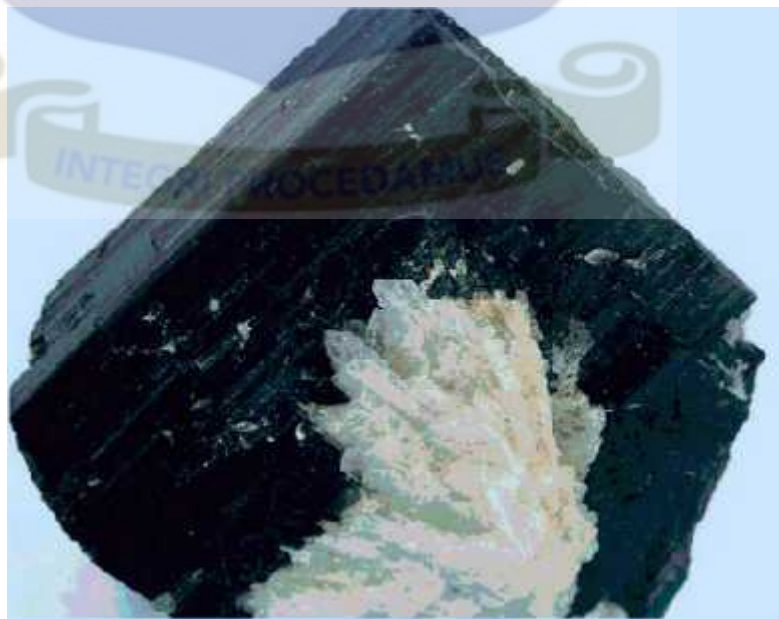


Figure 2.6-1: Dark colored tantalite mineral with pale colored albite (BGS, 2011)

BENEFICIATION ROUTES FOR TANTALUM

The beneficiation process adopted in concentrating a particular ore is a function of the nature of the ore in terms of physicommechanical and chemical characteristics of the ore. Basically, there are three stages that are involved and these are pre-concentration, primary concentration and concentration clean up (Amuda et al., 2007). The choice of any or all of these depends on the characteristics of the ore (particularly the content of the principal concentrate) relative to associated minerals and impurities. The beneficiation process may be carried out by any or combination of the mineral beneficiation techniques. These techniques include wet gravity, magnetic, electrostatic and flotation and the associated contending technologies are pyrometallurgy, chlorination and hydrometallurgy. The correlation existing among the three variables of grades of tantalite, beneficiation and chemical extraction routes are provided in Table (2.6-1).

Table 2.6-1: Correlation of tantalum beneficiation routes (Adetunji et al., 2005)

Tantalite Grades %	Raw-material input	Beneficiation Routes	Chemical Extraction Route
2-10	Low-medium grade in tin slags	Pre concentration (sizing, gravity separation)	<i>Pyrometallurgy</i>
40-100	Alloys, scraps		<i>Chlorination</i>
20-40	Natural ores	Flotation, leaching, magnetic, electrostatic separation. Primary concentration classifications, stage treatment	<i>Hydrometallurgy</i>
20-40	Synthetic concentrates		
> 15	High grade tin slags		

Fortunately, the tantalite ore reserves investigated at Kenticha tantalum mine is 40 - 70% tantalite ore concentrate. This average tantalite implies that the Kenticha tantalite deposits are candidates for hydrometallurgical beneficiation. The conventional method for the beneficiation of tantalite mineral is the gravity separation technique (Amuda et al., 2007) due to the density of tantalum minerals which allows concentration with other heavy metals. However, in the present effort, the focus is the concentration of multi-constituents of tantalum bearing minerals with a view to

generating economic value for the secondary ore concentrates in the tantalum bearing minerals.

Kenticha beneficiation effort proposed a model that combines the traditional gravity method of beneficiation of tantalite mineral with techniques that are capable of equally beneficiating TiO_2 , MnO_2 and Fe_2O_3 as adjunct recovered concentrates. A scheme of the concentration model is presented in Figure 2.6-2, which incorporates mainly gravity, magnetic and electrostatic separation techniques with leaching as adjunct beneficiation technique to generate the various secondary ore concentrates.

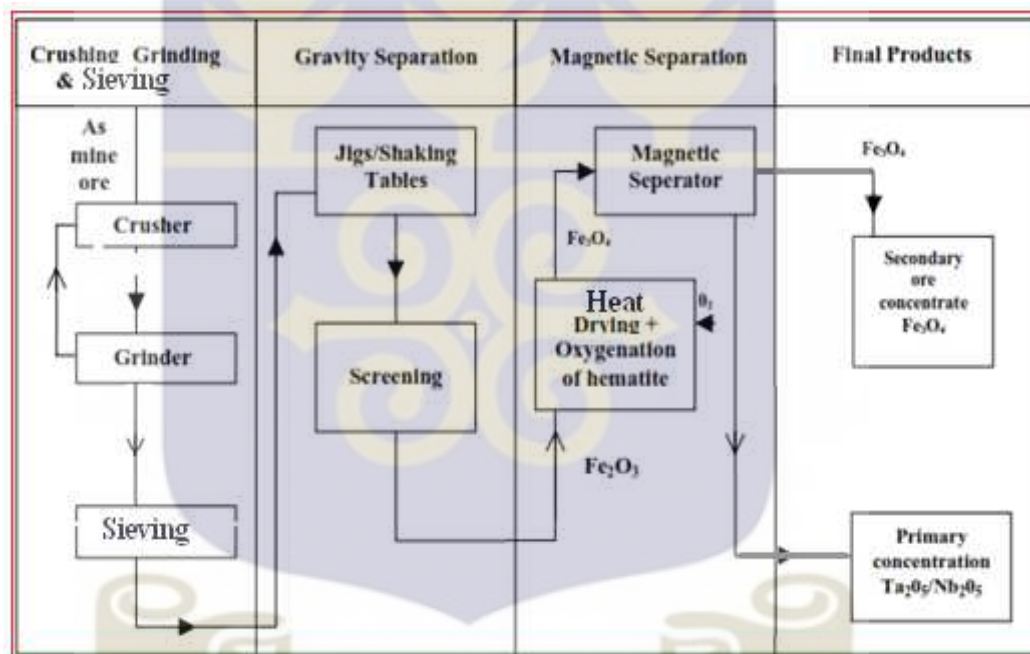


Figure 2.6-2: Generalized Concentration flow charts of $\text{Ta}_2\text{O}_5/\text{Nb}_2\text{O}_5$

In the concentration process, the ore is simply dug out by a bulldozer, scooped up onto trucks and driven to a plant where the ore is mixed with water (MME, 2012). Then, the ore is crushed, ground and sieved in appropriate system through a series of steps of screening, jigging and final treatment on shaking tables (MME, 2012). The screened ore is subjected to gravity separation technique where Ta_2O_5 and other associated minerals are concentrated, which is approximately 70 percent, Ta_2O_5 , (see Figure 2.6-

3). The Ta_2O_6 is separated from associated heavy metals such as wolframite, ilmenite which are electrically conducting through electrostatic separation.



Figure 2.6-3: Tantalum concentrate manufactured at Kenticha (MME, 2010)

The other heavy metals such as hematite, manganese oxide and rutile are roasted, dried and subsequently subjected to magnetic separation where the hematite transformed to ferromagnetic magnetite is concentrated from paramagnetic manganese and titanium ore minerals. The titanium/manganese ore are subsequently beneficiated through hydrometallurgy using leaching.

The remaining 30 percent is stored in a vast tailings pond for later use (MME, 2012). Generally, the concentrate ore, which may be 0.02% -0.04% Ta_2O_5 , consisting of a number of other radioactive isotopes, such as uranium and thorium families and potassium – 40, has been synthesized together (BGS, 2011).

Once the tantalum ore is recovered and concentrated, it is processed (treated chemically at high temperature) in several different ways to extract the tantalum in a form usable by industry. Tantalum concentrate is dissolved in acid at high temperature after which the solution is reacted with chemicals that yield K-salt (also called potassium fluortantalate, potassium fluorotantalate, potassium heptafluorotantalate, or potassium tantalum fluoride) or Ta_2O_5 (USGS, 2014a).

Acid Leaching: chemical processing routes

The chemical separation and quantification of the different elements require the sample to be in solution (Nete, 2013). Leaching is the removal of material by dissolving them away from the tantalite ore by using HF acid (Olushola and Folahan, 2011). Concentrated HF and high temperature fusion digestion techniques are commonly used for this mineral dissolution. The presence of fluoride ions from the HF dissolution is extremely important in the subsequent separation of tantalum.

Despite its success in leaching the main elements in the tantalite mineral while also providing the necessary fluoride ions for the separation of tantalum, HF digestion procedure often results in incomplete dissolution of the mineral samples. This method involves the conversion of the target impurities (Fe_2O_3) into soluble salts that can be separated from the solid residue by filtration, decantation or centrifugation.

2.7 NORM ASSOCIATED WITH TANTALUM MINING

The primary NORM radionuclides are uranium, thorium, potassium, radium, and radon (Jason, 1998). Usually, these naturally occurring radionuclides are present at only trace levels (approximately 10 ppm by mass). However, their presence throughout the environment is responsible for approximately half of the radiation dose (approximately 1mSv/year) to people (Long et al, 2012).

Radioactive material containing no significant amounts (the term 'significant amounts' would be a regulatory decision) of radionuclides other than naturally occurring radionuclides is called NORM (IAEA, 2007). Material in which the activity concentrations of the naturally occurring radionuclides have been changed by a process is included in naturally occurring radioactive material. On the other hand, the term Technologically Enhanced Naturally Occurring Radioactive Materials

(TENORM) is used to describe situations where human activities have increased the potential for exposure to these radionuclides in comparison to the naturally occurring situation. These activities which can enhance NORM levels directly are the mining, milling and processing of uranium ores and mineral sands, fertilizer manufacture and use, phosphate manufacture, burning of fossil fuels, metal ore processing (including tinstone (tin), tantalite, columbite, fergusonite, koppite, asenopyrites, etc) and general underground mining and open-pit mining activities like tantalum mining (O'Brien and Cooper, 1998). This implies that, in the process of tantalum ore mining at Kenticha tantalum mine can cause the concentration of NORM activity to increase and used to exposure of inhabitants of the area. In addition, the use of buildings material containing NORMs to construct dwellings and workplaces can also lead to exposure to NORM. Furthermore, the use of disposed waste materials (e.g. mine tailings, phosphor-gypsum, and fly ash) and is associated NORM concentrations could pose significant radiological problems if use for construction purposes (O'Brien and Cooper, 1998). These activities may bring people into closer contact with ores containing higher concentrations of radionuclides and the processing of ores may concentrate one or more of NORMs into a particular product or waste stream. NORM can also be present in consumer products, including common building products (like brick and cement blocks), granite counter tops, glazed tiles, phosphate fertilizers and tobacco products (CNSC, 2014).

The human activities mentioned above could produce NORM that are associated with at several pathways by which the radioactive material can reach humans. The pathway largely depends on the processes involved and can be broadly categorized into; on-site, off-site, airborne, waterborne, food products, etc. For on-site pathways, the exposures tend to be direct from external γ -radiation or internal exposure resulting

from inhalation of radioactive dust or radon progeny. Due to the presence of NORM in most soils and rocks, underground mining activities can lead to enhanced levels of radioactive dust, and radon isotopes and other radioactive isotopes. These NORMs can reach humans via several pathways, including the food chain, inhalation or ingestion of airborne radioactive dust and the inhalation of radon isotopes and their progeny which reach the atmosphere as a result of the exhalation of radon isotopes from the ground surface or from the surface of building materials (O'Brien and Cooper, 1998).

2.8 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 radioactivity (those come from ground).

2.8.1 COSMIC RADIATION AND COSMOGENIC RADIONUCLIDES

The radiation penetrating earth's atmosphere and originating from space (galaxies and sun) outside the earth is called cosmic radiation (Choppin et al, 2002; Gerti, 2012) and continually bombarded by high-energy particles. These cosmic rays interact with the nuclei of atmospheric constituents, producing a cascade of interactions and secondary reaction products that contribute to cosmic ray exposures which decrease in intensity with depth in the atmosphere, from aircraft altitudes to ground level. The cosmic ray interactions also produce a number of radioactive nuclei known as cosmogenic radionuclides.

Most of the cosmogenic radionuclides are relatively scarce and have insignificant contribution to the dose from naturally occurring radioactivity. Table (2.8-1) shows some of the cosmogenic radionuclides produced by the interaction of cosmic radiation with the atmosphere. Best known of these are ^3H and ^{14}C contributing significant exposures to the worldwide population (UNSCEAR, 2008a).

Table 2.8-1: Long-lived cosmogenic radionuclides in atmosphere (Choppin et al, 2002)

Nuclides	Half-life(years)	Decay mode	Particle energy (MeV)	Production rate (atoms/m ² s)
^3H	12.32	β^-	0.0186	2500
^{10}Be	1.52×10^6	β^-	0.555	300
^{14}C	5730	β^-	0.1565	17000-25000
^{22}Na	2.605	β^+	0.545	0.5
^{26}Al	7.1×10^5	β^+	1.16	1.2
^{32}Si	160	β^-	0.213	1.6
^{35}S	0.239	β^-	0.167	14
^{36}Cl	3.01×10^5	β^-	0.709	60
^{36}Ar	268	β^-	0.565	56
^{53}Mn	3.7×10^6	EC	0.596	
^{81}Kr	2.2×10^5	EC	0.28	

2.8.2 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 ^{238}U , ^{232}Th and ^{40}K .

URANIUM SERIES (4n +2 SERIES)

The uranium atom consists of three different isotopes: ^{238}U (99.3%), ^{235}U (0.72%) and ^{234}U (0.005%). The ^{238}U and ^{234}U 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).

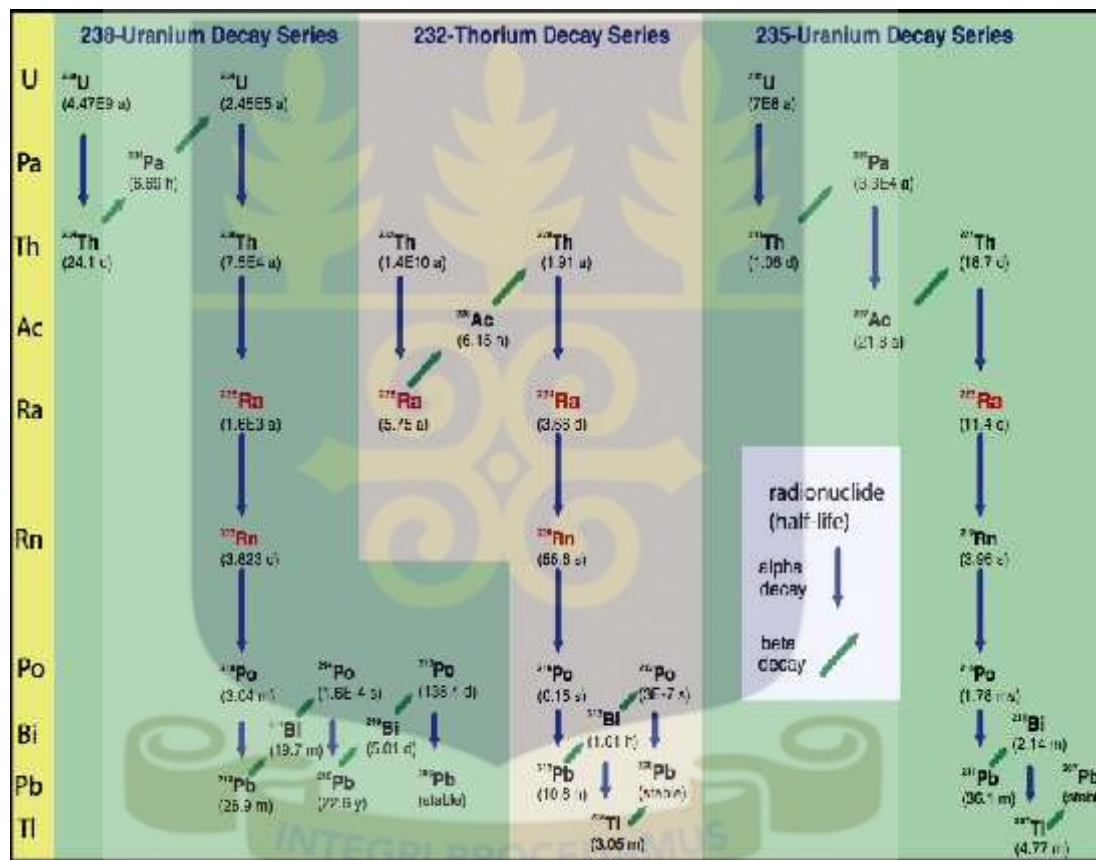


Figure 2.8-1: Decay series of the natural ^{238}U , ^{235}U and ^{232}Th ¹

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

¹ (<http://www.daviddarling.info/encyclopedia/A/Emain.html>)

$^{235}\text{U}/^{238}\text{U}$ ratio will be constant and all nuclides in each of the series will be in equilibrium (Read and Hayder, 2011).

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 end 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 .

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).

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).

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 (Cember, 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 a 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}K with half-life of 1.3 billion years decay 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 decay into the excited state of ^{40}Ar . Secondly, the excited nuclide ^{40}Ar , decayed 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).

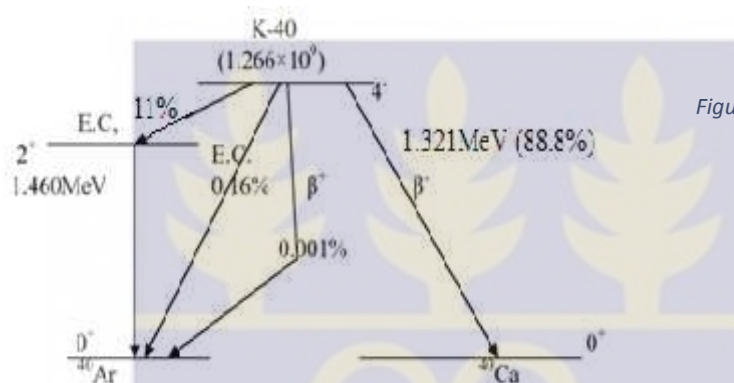


Figure 2.8-2: Decay scheme of potassium - 40

2.8.3 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 as a result 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 that “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).

2.8.4 RADON

The three naturally occurring radon isotopes are $^{222, 220, 219}\text{Rn}$ (with half-lives 3.824 d, 55.6 s and 4 s) are produced by the alpha decay of radium isotopes ($^{226, 224, 223}\text{Ra}$) which are decay products of ^{238}U , ^{232}Th and ^{235}U , respectively (Ishimori et al, 2013). It occurs at low levels in virtually all rock, soil, water, plants, and animals. Radon-222 is slightly soluble in water ($510 \text{ cm}^3/\text{L}$ at 0°C) to form compounds under laboratory conditions. It has a density of 9.73 g/L at 0°C . The α -emitter ^{220}Rn and ^{222}Rn production in

terrestrial materials is depends on the activity concentrations of ^{228}Ra and ^{226}Ra present, respectively. It has four short-lived decay products: ^{218}Po (3.05 min), ^{214}Pb (26.8 min), ^{214}Bi (19.9 min), and ^{214}Po (164 μs). Both polonium isotopes are α -emitters. Radon is the most significant element of human irradiation by natural sources. The most significant mode of exposure is the inhalation of its short-lived decay products of ^{210}Pb and ^{210}Po . The relatively long half-life of one of its decay products, ^{212}Pb (10.6 h), allows this isotope time to deposit on surfaces or migrate away from its source before producing the important alpha-emitter ^{212}Bi (60.6 min) (UNSCEAR, 2000b).

2.8.4.1 RADON CONCENTRATION

The ^{222}Rn concentrations in open air are quite variable with time-average concentrations in the range of 2-30 Bq/m³ and in the soil and root zone, the concentrations higher by 1000 times than in the open air (Faanu, 2011). Depending on the composition of the soil and the bedrock, the average concentration ^{222}Rn varies widely(UNSCEAR, 2000b). Factors that may influence the levels of ^{222}Rn concentration in soil, water and air are: grain or particle size and shape of soil, moisture content, temperature, geology of soil formation, permeability of the soil, seasonal variation and wind pressure. For soil with an average ^{226}Ra concentration of 40 Bq/kg, the average ^{222}Rn concentration in the soil water would be about 60 Bq/m³. Much higher values of 8000 Bq/m³ and 50, 000 Bq/m³ have been measured in deep ground waters in areas such as in the United States of America (Maine) and in Finland, respectively (UNSCEAR, 1996). UNSCEAR reported 78 kBq/m³ for soil (UNSCEAR, 2000b). The action level of radon recommended by the ICRP for which intervention is necessary is 1000 Bq/m³ for working area (ICRP, 2007). This value is based on an assumed occupancy of 2000 hours per year and this is equivalent to an

effective dose of 6 mSv per year. This value is also the midpoint of a range of 500-1500 Bq/m³ (ICRP, 1993).

2.8.4.2 RADON EMANATION COEFFICIENT

Radium is a family of NORM produced by the decay of uranium and thorium, mainly from uranium, which in turn produces daughter radon. Inhalation exposure dominates all other pathways of radiological exposure for human being, which is caused by radon and its decay products emanating from soil and rock (UNSCEAR, 2000a). The rate of escape or emanation of radon into the surrounding air from solids is called radon emanation rate or radon exhalation rate of solid (Pandit, 2010). The radon emanation coefficient is defined as fraction of the total number of radon atoms generated from a radium-bearing grain that are released into the pore space of rock or soil (Schumann, 1993; UNSCEAR, 1988, 2000b). According to UNSCEAR 2000 report, the typical emanation coefficient for rock and soil typically range from 0.05 to 0.7 with a representative value of 0.22 (UNSCEAR, 2000b). The radon emanation is enhanced by radium distribution, solid grain size, water content and temperature (Nabil et al, 2009; UNSCEAR, 2000b). Radon concentrations in soil depend on the distribution and concentrations of the parent radium radionuclides in the bedrock and overburden and on the permeability of the soil. Generally the radon emanation coefficient is inversely proportional to grain size (UNSCEAR, 2000b).

2.8.4.3 RADON EXPOSURE

The presence of naturally occurring radionuclides in soil, rock, water, and air along with cosmic radiation results in continuous and largely unavoidable radiation exposures of all humans. Exposures larger than these due to undisturbed natural background can result from human activities that move naturally occurring radionuclides from normally inaccessible locations to locations where humans are

present or concentrate naturally occurring radionuclides. For instance, radon and its short-lived alpha emitting progeny in the atmosphere are the most important contributors to human exposure from natural sources (UNSCEAR, 2000b). According to world nuclear association report, NORMs give rise to a very much larger radiological effect to the public (by about four orders of magnitude) compared with that caused by the nuclear industry (WNA, 2009) via several pathways, including the food chain, inhalation or ingestion and the inhalation of radon isotopes and their progeny (UNSCEAR, 2000c). This exposure is mostly produced typically by alpha particles emitted by the inhaled short-lived decay products of ^{222}Rn and ^{220}Rn radionuclides, and their subsequent deposition along the walls of the various airways of the bronchial tree provide the main pathway for radiation exposure of the lungs (UNSCEAR, 2000b). Based on UNSCEAR 2000 report, there is general agreement among scientists that it is the alpha particle irradiation of the secretory and basal cells of the upper airways that is responsible for the lung cancer risk seen in miners, although there remains some uncertainty as to exactly which cells are most important for the subsequent induction of lung cancer (UNSCEAR, 2000b). While the health risks associated with high radon exposures in underground mines have been known for a long time, relatively little attention was paid to environmental radon exposures until the 1970s (UNSCEAR, 2000b).

2.8.4.4 HAZARDS ASSOCIATED WITH RADON

The hazards and risk due to exposure to radon gas from NORM contaminated material is estimated from the radon emanation fraction; which was defined in section (2.7.3.2). The radon gas diffuses out of the earth crust into the air and becomes widely dispersed throughout the local atmosphere. Thus, radon and its progeny are the principal source of naturally occurring atmospheric radioactivity. The radioactive radon daughters,

which are solids under ordinary conditions, attach themselves to atmospheric dusts. Atmospheric concentrations of radioactivity from this source vary widely around the earth and are dependent on the local concentrations of uranium and thorium in the earth. Although the average atmospheric radon concentration is on the order of 2×10^{-6} Bq/mL (5×10^{-11} μ Ci/mL), concentrations 10 times greater are not uncommon (Cember, 2009).

According to ICRP publication 65, radiation is a causative agent of cancer in many organs and tissues of the body (ICRP, 1993). Specially National and international scientific organizations have concluded that radon causes lung cancer in humans (EPA, 2000). According to the NAS, breathing indoor radon in homes is estimated to cause about 15,000 to 22,000 lung cancer deaths each year in the United States (EPA, 2000). As it was mentioned above, health hazard come from the exposure of radon decay products (progeny) not from radon itself (Cember, 2009) and there is a delay of many years between the initiation of a cancer by radiation and its growth to a size which can be observed clinically (ARPANSA, 2015). The primary adverse health effect associated with chronic exposure to radon is lung cancer (Cember, 2009). The risk of developing lung cancer depends on how much radon we breathe in. The more radon there is in the air, the bigger the risk. Similarly, the longer we spend breathing in that radon, the bigger the risk (ARPANSA, 2015). According to EPA recommended guideline, the average indoor radon level is estimated to be about 1.3 pCi/L, and about 0.4 pCi/L of radon is normally found in the outside air (EPA, 2012). The overall risk of radon exposure is related not only to its average level in the home, but also to the occupants and their lifestyles (ARPANSA, 2015).

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 results 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).

2.10 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 in different 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 2.10-1: 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

2.11 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.

2.11.1 HPGe DETECTOR FOR GAMMA RAY SPECTROMETRY

Solid state detectors with a p-n junction are made from semiconductor materials, which have electrical conducting properties midway between a “good conductor” and an “insulator.” The most commonly used semiconductor materials which are primarily used as detecting media for radiation detectors are silicon and germanium (Cember, 2009; Todsadol, 2012). Detectors made from ultrapure germanium material are generally known as intrinsic or hyper-pure germanium (HPGe) detectors (Knoll, 1999; Todsadol, 2012). Currently, HPGe detector is commonly applied to detect gamma radiation. Detection and measurement of gamma radiation generally requires a greater sensitive volume, that is, a thicker depletion layer (Cember, 2009; Todsadol, 2012).

Figure 2.11-1 is a block diagram of simple γ -ray spectrometry setup.

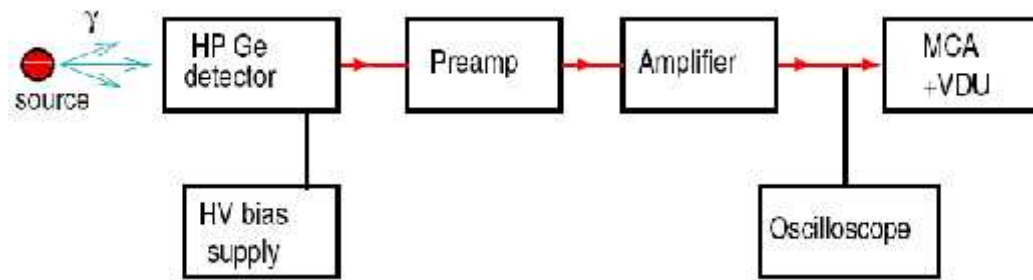


Figure 2.11-1: Scheme of HPGe detector system. (Rhul, 2007)

2.11.2 COUNTING SYSTEM FOR GAMMA SPECTROMETRY

Neither ^{238}U nor ^{232}Th emit gamma rays, and gamma ray emissions of their radioactive daughter (^{214}Pb and ^{214}Bi for ^{238}U and ^{212}Pb , ^{208}Tl and ^{228}Ac for ^{232}Th , respectively) products are used to estimate their concentrations. The ^{40}K photo peak energy [1460.83keV (10.7%)] will be used for computing its activity concentration (IAEA, 2003b; UNSCEAR, 2000b). Each gamma ray photon has a discrete energy, and this energy is characteristic of the source isotope. Hence, nuclear spectroscopy is based on the analysis of radioactive isotopes by measuring the energy distribution of the source. When incident gamma radiations interact with a detector active volume undergoing any of the three principal interaction processes (i.e. photoelectric, Compton and pair production) generates a number of electronic secondary charges (electron-hole pairs) which are proportional to the amount of gamma-ray energy deposited in the detector. The electrical charge output from the detector will be collected and counted through an electronic counting system as illustrated in Figure 2.11-1 and then displayed in the form of the gamma-ray spectrum.

A detector bias supply is connected to a detector in order to withdraw the electron-hole pairs created within the depletion region of a detector and then all charge carriers will be collected by the pre-amplifier. The preamplifier converts the collected charge pulse to a voltage pulse. The shape and size of the preamp pulse are modified to be suitably processed by the amplifier. Due to the amplifier being linear, the pulse height

is proportional to the gamma-ray energy which was absorbed by the active volume of the detector. The pulses from the amplifier are sorted by their pulse height and then converted to a digital number referred as a channel (bins) by the analog-to-digital converter (ADC) in the multichannel analyzer (MCA). The number of pulses according to each pulse height range is counted and stored in each channel number that contributes to a gamma-ray spectrum. The total number of channels in MCAs varies by factors of 2 over the range of 128 to 16384 each with a storage capacity of 10^5 to 10^6 Radiation counts per channel (Cember, 2009).

In nuclear spectroscopy, the resolution of the detector is important if spectral lines closed together are to be separated and observed. Energy resolution may be viewed as the extent to which a detector is able to distinguish between two closely lying energies (radioisotopes). The HPGe detectors achieve the best energy resolution but due to their small active volume, their sensitivity is low and takes several minutes to record a spectrum. Its energy resolution is expressed as full width at half maximum (FWHM); in which the narrower the peak, the lower the FWHM, the better the ability of the detector to resolve and separate close peaks that is said to be better resolution. Energy resolution is given by equation (2.11-1):

$$\text{Resolution} = \frac{\text{Full width at half maximum (FWHM)}}{\text{Peak centroid}} * E_{\text{analysis}} \quad (2.11-1)$$

Since germanium has a low band gap, the HPGe detector is cooled with liquid nitrogen to temperature of 77°K to reduce the thermal generation of charge carriers and leakage current that creating noise (Knoll, 1999).

CHAPTER THREE

MATERIALS AND METHODS

This chapter discussed about geographical location, geology and hydrology of the study area. Moreover, it provided information about the materials and methods used for sampling, sample preparation, measurements of activity using HPGe detector, dose rate, AED as well as radiological hazard and cancer risk assessments.

3.1 DESCRIPTION OF THE STUDY AREA

The study was conducted at Kenticha tantalum mine. It is found in Odo Shakiso with the geographical location of 6°20'N 39°10'E latitude and longitude, in Southern Ethiopia as shown in Figure 3.1-1. The mine is located about 600 km south of Addis Ababa and 50 km Southeast of Shakiso, administrative capital of Odo Shakiso.

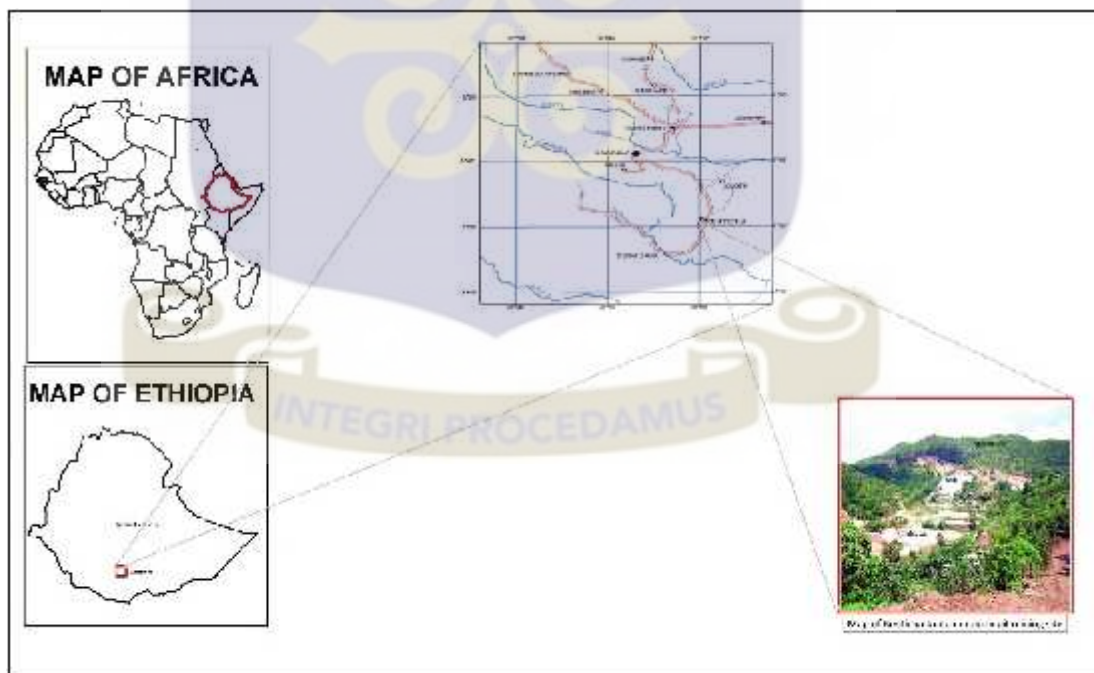


Figure 3.1-1: Location map of Kenticha tantalum mine in Odo Shakiso (GSE, 2010)

Odo Shakiso covers an area of about 168,682 km², where mixed farming and other economic activities are the source of major livelihood of the people. The expansion of

social services, secondary economic activities and modern means of transportation and communication are in their early stages of development (EMDSC, 2013).

The 2007 national census reported a total population of 206,372 for this area with an estimated population density of 32.2 people per square kilometer, which is greater than the Zone average of 21.1. The four largest ethnic groups reported in Odo Shakiso were the Oromo, the Amhara, the Gedeo, and the Soddo Gurage (CSAE, 2007).

3.1.1 KENTICHA TANTALUM MINE

The Kenticha tantalum mine in Ethiopia is considered to be one of the largest world class tantalum producing assets in Eastern Africa. The mineral is currently experiencing global boom. This mine is operated by a state-owned company, Ethiopia Mineral Development Share Company (EMDSC) since 1990 in the Adola Greenstone Belt. A deposit in weathered crust was delineated in 1988 with proven reserves of 25,850 tons of columbo– 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 pentaoxide and 2300 tons of niobium pentaoxide, and primary ore with proven reserve of 2393 ton Ta_2O_5 and 2362.5 ton Nb_2O_5 . 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 in Africa, Kenticha tantalum mine in southern Ethiopia and the distribution of sampling points; Appendix I also described sampling co-ordinates for ore, soil, solid waste, waste tailings and water samples.

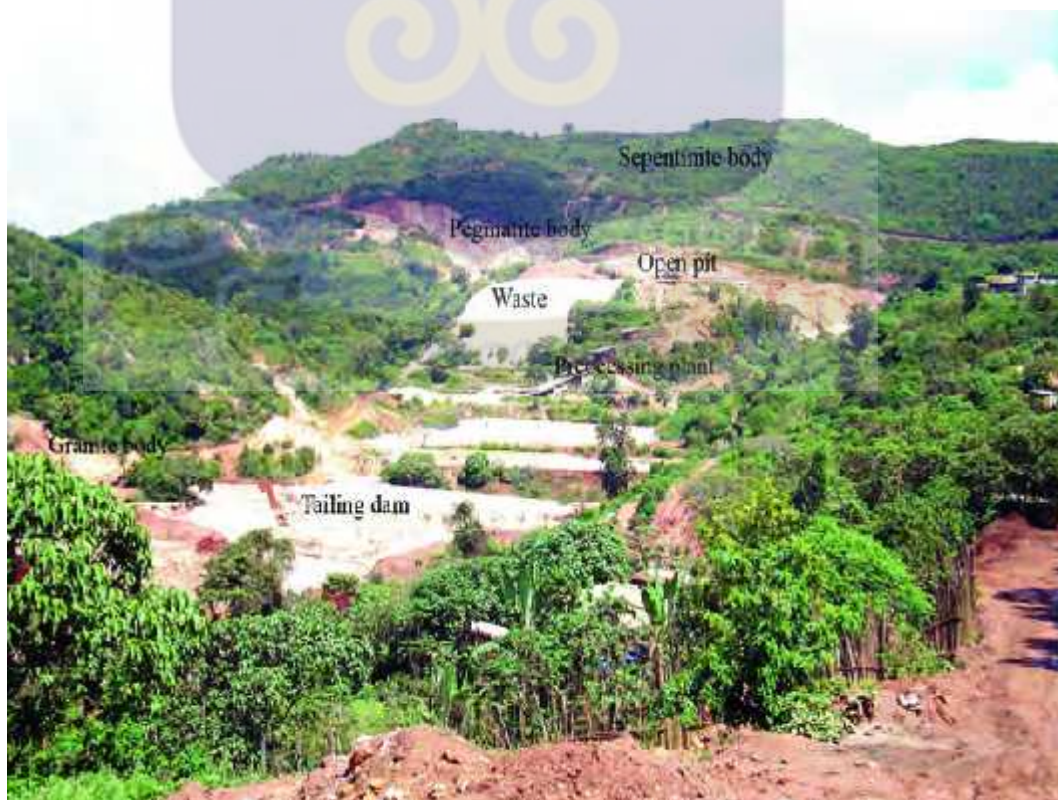


Figure 3.1-2: Map of Kenticha tantalum open pit mining site (MME, 2010)

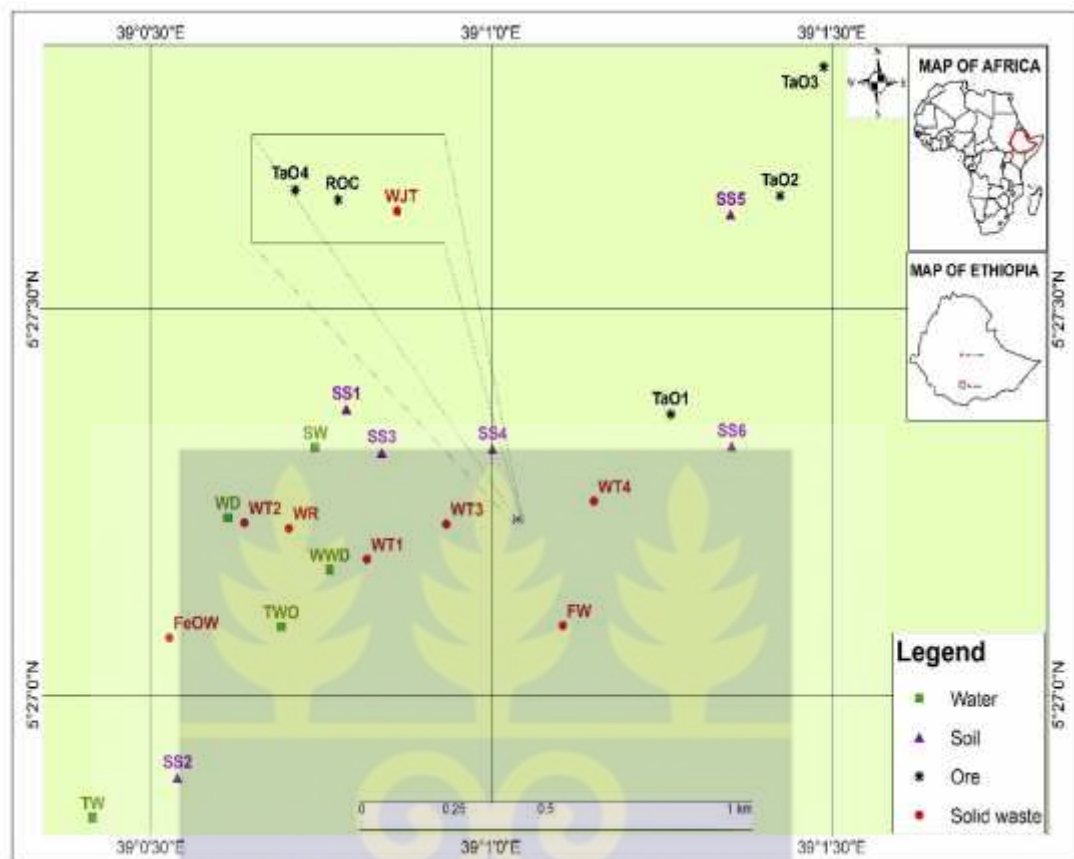


Figure 3.1-3: Outline of Kenticha tantalum mine showing sampling points

3.1.2 GEOLOGY OF KENTICHA TANTALUM DEPOSIT

According to Kozyrev et al. 1982, the Kenticha greenstone belt is part of the rock of Adola area, which is an integral part of the Mozambique belt extending from South Africa across Tanzania and Kenya into Ethiopia and then into North Eastern Africa (Kozyrev et al; 1982). Moreover, the Adola area's rocks are summarized to be Gneissic Terrain and Green-stone Belt. The Gneissic terrain consists of relatively high grade rocks, which occupy three N-S trending anticlinal zones separated by two intervening narrow N-S trending synclinal Greenstone Belts. With reference to the Greenstone Belts, Western, Central and Eastern Gneissic terrains are distinguished. Similarly, Woldai classified the two greenstone belts into western (Megado Zone Greenstone belt) and Eastern (Kenticha Zone greenstone belt) (Woldai, 1989). The

greenstone belt forms the eastern most portion of the Adola area and it lies across the boundary of the gneisses belts of Kenticha formation and Aflata formation, (Fig. 3.1-4) (Zerihun, n.d.).

The Kenticha granites formation consists of a rare metal element class pegmatite barren to Nb, Ta, Be, Li enriched types (Zerihun, n.d.). The rare earth metal occurrences are hosted in a long and linear belt for over 100 km with pegmatite field area coverage of roughly 2,500 km². While, the pegmatite formation constitutes biotite-muscovite gneisses, quartz amphibole gneiss, quartzo-feldspatic biotite schist, two mica schist and marble (Kozyrev et al, 1982). The rare earth metal associated granites corresponds to muscovite and alaskite. In general three types of ore such as primary ore (tantalite bearing granite-pegmatite with complex Ta-Nb-Li-Be mineralization), lateritic ore and eluvial-deluvial and alluvial placer ore have been recognized at Kenticha. The weathered ore developed over the primary ore of pegmatite represents the huge rare metal resources of the Kenticha deposit.

Several other works (Zerihun and Tadesse, 1996; Zerihun et al, 1995; Zerihun, n.d.) had been carried out on the mineralogical and compositional characterization of major tantalum bearing mineral deposits in the country. These works revealed the extensive abundance of elemental tantalum in association with niobium as the respective oxides.

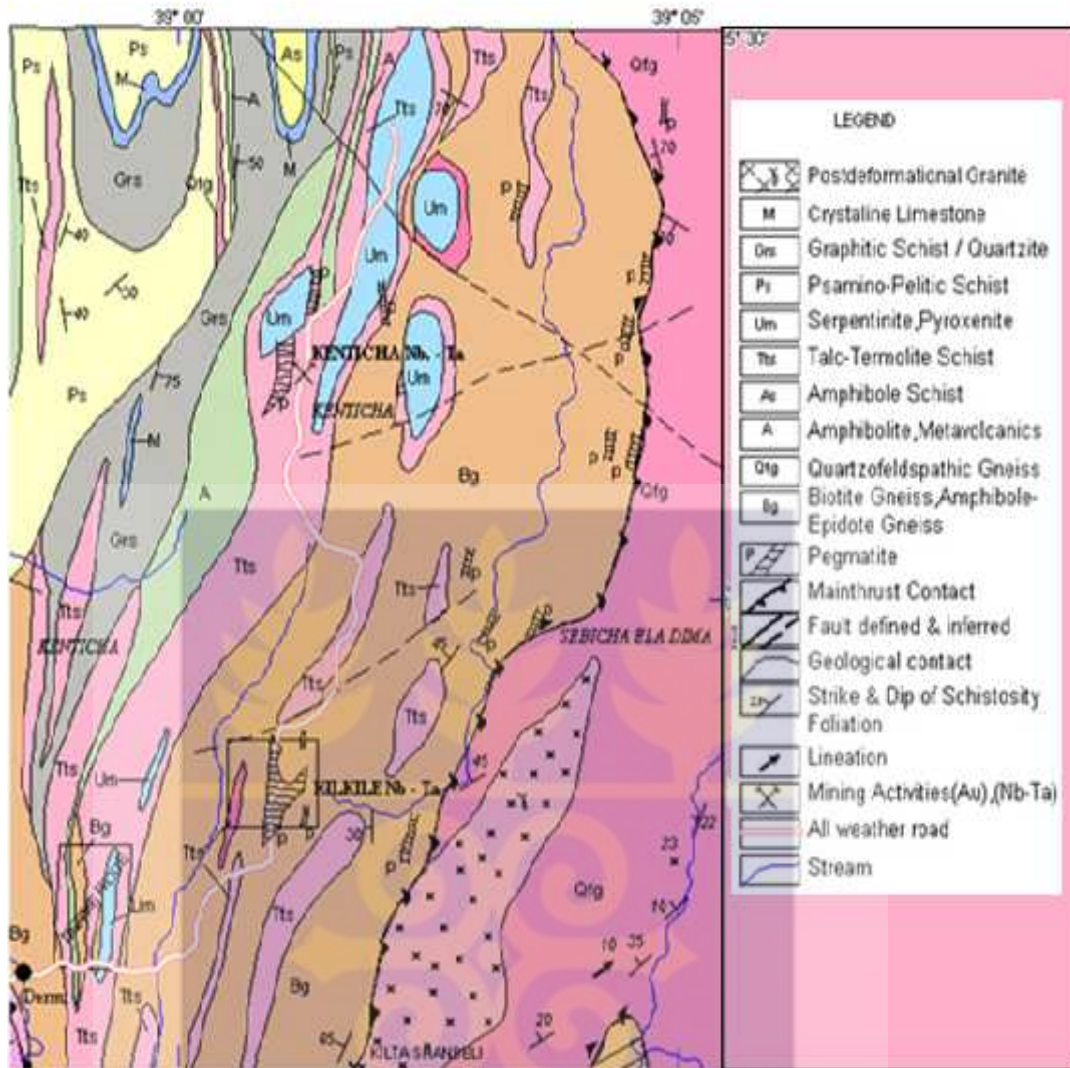


Figure 3.1-4: Geologic map of the Kenticha Area²

3.1.3 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 rain water. Drainage is well-developed and mainly gets water from rain. The Mormora River is the largest perennial river in the region about 8 km west of the Kenticha tantalum deposit (source).

² Extracted from Geological Map of Adola area, 1:100, 000, 1992

3.2 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.

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.



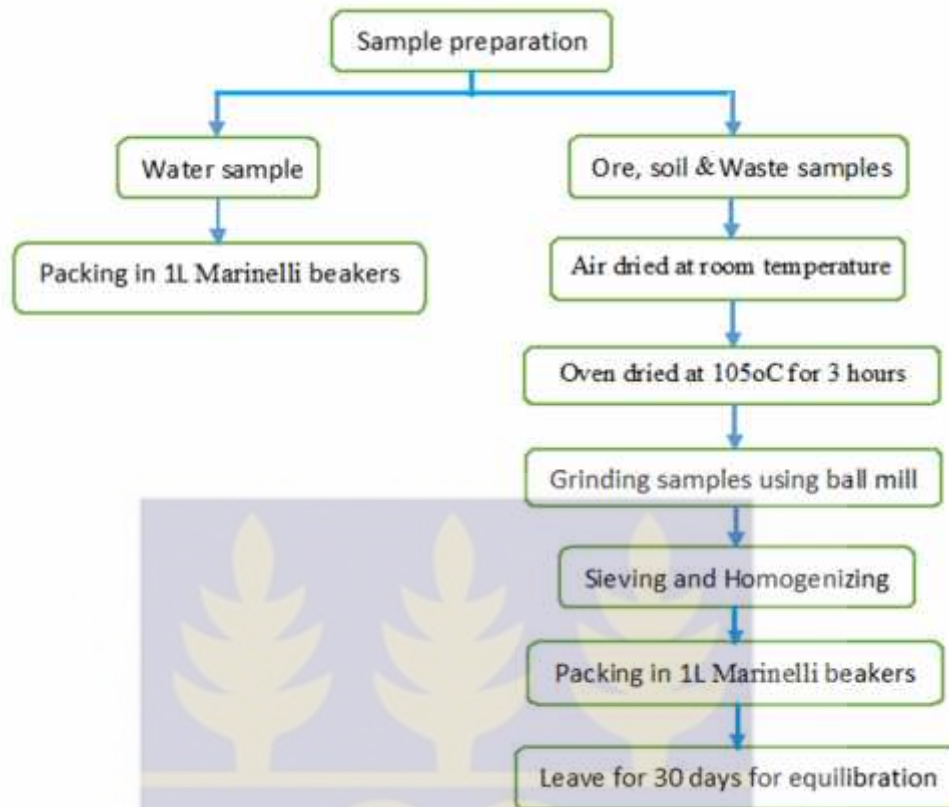


Figure 3.2-1: Flow chart for sample preparation and packing

3.3 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 3.3-1: Kenticha open pit tantalum mine

The water samples were also collected into clean labelled 1.5L polyethylene bottles from the Mormora Rivers, tap water and spring water. Each sample were immediately spiked (acidified) with 1M HNO_3 to prevent sorption on surface of the bottle. Then, the bottle were 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 were washed with detergent to remove grease, then with water following further washing with 10% HNO_3 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 3.3-2: Water dam at Kenticha for use of the mine

3.4 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) Marinelli beakers were recorded using weighing balance (METTLER TOLEDO, Model: XP2001S, SNR: B044082756, mass range: 0.1g to 2100 g, 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 3.4-1: Sample filled in a Marinelli beaker and sealed with plastic tape

Moreover, the 1L water samples were filled into weighed Marinelli 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 3.4-2: Water sample filled in a Marinelli beaker and sealed with plastic tape

3.5 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.



Figure 3.5-1: Gamma spectrometry set-up at GAEC

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 .

3.5.1 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 very 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).

3.5.2 EFFICIENCY CALIBRATION

An accurate efficiency calibration of the system is necessary for accurate quantification of radionuclides present in the sample. The working principle is measuring an output pulse for each radiation event that interacts within the active volume in a detector. The mixed radionuclides standard contained in Marinelli beakers with the same geometry as that of samples used from the NORM measurement that was used for the energy calibration was also used for the efficiency calibration. By counting the mixed standard radionuclides on the detector for 36,000 seconds with

energies between 60 keV – 2 MeV (Faanu, 2011), the net counts in the full-energy peaks was determined and their corresponding energies used in the determination of the efficiencies. For each source, the efficiencies at various energies was calculated by using equation (3.5-1) (Faanu, 2011):

$$\epsilon_f = \frac{\text{Net count}}{A_{std}P_\gamma T_{std}} \quad (3.5-1)$$

Where,

- ϵ_f is the full-energy peak efficiency (number of counts detected per number of γ -ray emitted by source),
- P_γ is gamma line emission probability,
- T_{std} is the counting time and A_{std} is the present activity of the standard, which is given by equation (3.5-2):

$$A_{std} = A_{std}^0 e^{-\lambda t} \quad (3.5-2)$$

Where,

- A_{std}^0 is the the initial activity of the standard at the time of manufacture (i.e. Becquerel's),
- λ is the decay constant and
- t is the delay time.

Generally, the efficiency calibration of the HPGe detector shows that efficiency decreases as the energy increases (Todsadol, 2012) as shown in Figure 4.1-2, which is an indication that the system is operating properly and it is geometric dependent (Faanu, 2011). The radionuclides and their activities in the standard are shown in Appendix III.

3.5.3 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 [351.92 keV (35.6%)] and ^{214}Bi [609.31 keV (45.5%) and 1764.49 keV (15.3%)], while ^{232}Th was estimated from peaks of ^{212}Pb [238.63 keV (43.6%)] and ^{208}Tl [583.19 keV (85%)]. Moreover, the ^{40}K photo peak energy [1460.83 keV (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):

$$MDA = \frac{1.645 \overline{N_B}}{\epsilon_f P_\gamma T_C m} \quad (3.5-3)$$

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).

3.6 ANALYSIS OF SAMPLES USING GAMMA SPECTROMETRY

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 3.6-1.

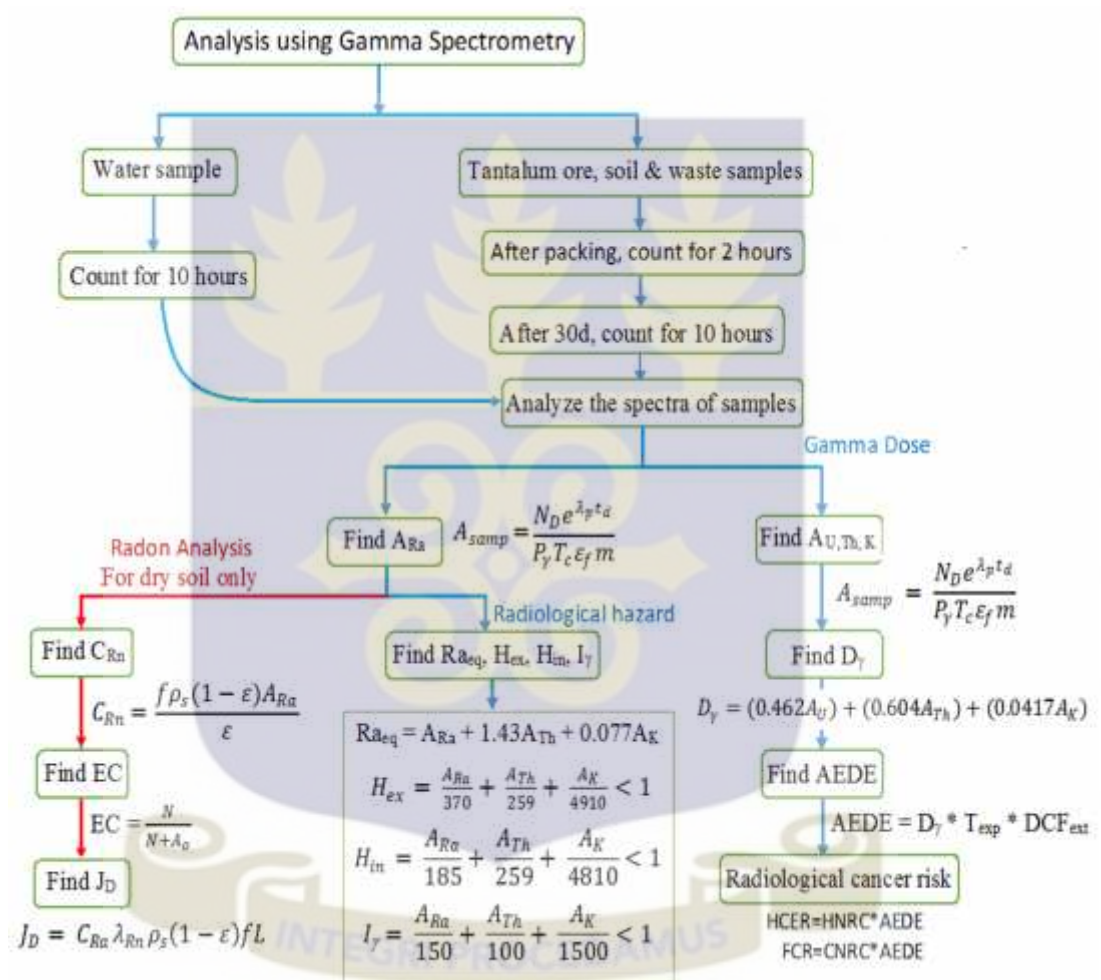


Figure 3.6-1: Flowchart for the experimental sample analysis

3.6.1 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 [351.92

keV (35.6%) and 295.21 keV (18.42%)] and ^{214}Bi [609.31keV (45.5%), 1120.3 keV (14.9%), 1238.1 keV (5.8%) and 1764.49 keV (15.3%)], while ^{232}Th radionuclide was estimated from peaks of ^{212}Pb [238.63 keV (43.6%)], ^{208}Tl [583.19 keV (85%) and 860.56 keV (12.5%)] and ^{228}Ac [338.3 keV (11.27%), 911.1 keV (25.8%) and 968.97 keV (15.8%)]. The ^{40}K photo peak energy [1460.83 keV (10.7%)] was used for computing its activity concentration (UNSCEAR, 2000b). The unit of measurements was Bq/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 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).

$$A_{\text{samp}} = \frac{N_D \cdot e^{\lambda_p t_d}}{P_\gamma \cdot T_c \cdot \epsilon_f \cdot W_{\text{samp}}} \quad (3.6-1)$$

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).

3.6.2 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 (30 Bq/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):

$$e_A = A_{s\text{amp}} * \sqrt{\frac{\Delta N}{N_D}^2 + \frac{\Delta \epsilon}{\epsilon_f}^2 + \frac{\Delta p_\gamma}{P_\gamma}^2 + \frac{\Delta W}{W_{s\text{amp}}}^2} \quad (3.6-2)$$

Where,

- e_A is error in activity concentration (Bq/kg),
- ΔN is the error in the net count,
- $\Delta \epsilon$ 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.

3.6.3 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).

$$D_{\gamma,\text{ext}} = \text{DCF}_{\text{U}}A_{\text{U}} + \text{DCF}_{\text{Th}}A_{\text{Th}} + \text{DCF}_{\text{K}}A_{\text{K}} \quad (3.6-3)$$

Where,

- A_{u} , A_{Th} and A_{k} are the activity concentrations of ^{238}U , ^{232}Th and ^{40}K respectively and
- $\text{DCF}_{\text{U}} = 0.462$, $\text{DCF}_{\text{Th}} = 0.604$ and $\text{DCF}_{\text{K}} = 0.0417$ 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 93nGy/h. A typical range of variability for measured absorbed dose rates in air is from 10 to 200 nGy/h (UNSCEAR, 2000b).

3.6.4 ANNUAL EFFECTIVE DOSE (AED) FOR EXTERNAL X-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):

$$AED_{\gamma,ext} = D_{\gamma,ext} \cdot T_{exp,ext} \cdot DCF_{ext} \quad (3.6-4)$$

Where;

- $D_{,ext}$ is the average external gamma dose rate in $\mu\text{Gy/h}$,
- 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 ($8\text{h/d} \times 245\text{d/y} = 1960\text{ h/y}$); and applying outdoor occupancy factor of 0.2 (i.e. $T_{exp} = 1960 \times 0.2 = 392\text{ h/y}$). 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 ²³⁸U and ²³²Th and ⁴⁰K 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 μSv/y in a simplified to the following formulas:

$$AED_{y,ext} = D_{y,ext}(nGy/y) \times 392 \mu Sv/y \times 0.7 Sv/Gy \quad (3.6-5)$$

$$AED_{y,ext} = D_{y,ext} \times 2.744 \mu Sv/y \quad (3.6-6)$$

3.6.5 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.

$$FCR = CNRC \times AED \quad (3.6-7)$$

$$HCER = HNRC \times AED \quad (3.6-8)$$

Where,

- CNRC is cancer nominal risk coefficient,

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

Table 3.6-1: Detriment adjusted nominal risk coefficients for cancer and heritable effects (ICRP, 2007)

Exposed population	Cancer fatality		Hereditary effects		Total	
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).

3.6.6 RADIOLOGICAL HAZARD ASSESSMENT DUE TO NORM

A radiological dose assessment calculates the amount of radiation energy that might be absorbed by a potentially exposed individual as a result of a specific exposure. ^{226}Ra , ^{232}Th and ^{40}K activity concentrations (98.5% of the radiological hazard of U series is due to Ra and its decay products) measured in each of the studied samples indicate the quantity of radioactivity present but do not provide a measure of radiation hazard. The hazard associated with radioactivity in samples is assessed through hazard indices. A hazard index is a parameter that is represented by a single value that takes into account the measured activity concentrations of ^{226}Ra , ^{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, dose rate, annual effective dose and total absorbed dose rate (Saher et al, 2013). In this section, the radiological hazard of radium-equivalent activity index (R_{eq}), external (H_{ext}) and internal (H_{int}) indices and radioactivity level index (I_γ) associated with radioactivity in all samples were evaluated and compared with recommended values. The natural radionuclides considered in this radiological assessment are ^{226}Ra , ^{232}Th and ^{40}K .

a) CALCULATION OF RADIUM EQUIVALENT ACTIVITY

Radium equivalent activity (R_{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, 1 Bq/kg of ^{226}Ra , 0.7 Bq/kg of ^{232}Th , and 13 Bq/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)

$$R_{eq} = A_{Ra} + 0.077A_K + 1.43A_{Th} \quad (3.6-9)$$

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 370 Bq/kg sources which correspond to an effective dose of 1mSv/y for the general 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).

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (3.6-10)$$

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 based on the fact that, 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):

$$H_{int} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (3.6-11)$$

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):

$$I_\gamma = \frac{A_{Ra}}{150} + \frac{A_{Th}}{100} + \frac{A_K}{1500} \leq 1 \quad (3.6-12)$$

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 must be less than unity (Harb et al, 2008).

3.7 ANALYSIS OF RADON IN THE SAMPLES

3.7.1 CALCULATION OF RADON CONCENTRATION

Tantalum ore, soil and solid waste samples was used to study radon in the environment. The samples were first dried by air followed by oven and then cooled. Then, the samples were completely sealed in one liter (1L) Marinelli beakers. Each sealed container containing the samples was counted by gamma-ray spectrometry for 7200 seconds after two hours of samples preparation. Finally, the samples were kept for one month to achieve the secular equilibrium between radium (half-life of 1600years) and radon (half-life of 3.86 days), and then they were counted for 36,000 seconds. Finally, the activity concentration of ^{226}Ra was determined from ^{214}Pb [351.92 keV (35.6%) and 295.21 keV (18.42%)] and ^{214}Bi [609.31 keV (45.5%)] average peak areas. This helps to estimate the concentration of radon in each samples using the activity concentration of ^{226}Ra by equation (3.7-1) (UNSCEAR, 2000b):

$$C_{Rn} = \frac{A_{Ra} f \rho_s (1-\epsilon)}{\epsilon m k_T - 1 + 1} \quad (3.7-1)$$

Where,

- A_{Ra} is the activity concentration of ^{226}Ra in (Bq/kg),
- f is the radon emanation factor, (0.2),
- ρ_s is the density of the tantalum ore grains, (2700 kg/m³),
- ϵ is the total porosity, (0.25),

- m is the fraction of the porosity that is water filled, (0.95),
 - k_T is the partition coefficient of radon between the water and air phases, (0.23)
- and m is zero if the sample is dry.

For this study, m is zero because the samples were dried before the activity concentrations were measured. Therefore, equation (3.7-2) becomes:

$$C_{Rn} = \frac{A_{Ra} f \rho_s (1-\epsilon)}{\epsilon} \quad (3.7-2)$$

In view of the latest scientific data on radon in soil, the average activity concentration of ^{222}Rn should be less than 78 kBq/m^3 (UNSCEAR, 2000b).

3.7.2 DETERMINATION OF RADON EXHALATION RATE

It is clear that after radon has been produced from the decay of radium in solid grains, a fraction of them escaped to the pore space. With time the radon in this pore will migrate to the atmosphere in a process called exhalation rate which is defined as the number of radon isotope atoms left per unit surface area of material per unit time ($\text{Bq/m}^2.\text{s}$), or mass exhalation rate which is defined as the number of radon isotopes atoms left per unit mass of the material per unit time (Bq/kg.s).

The key mechanism for the entry of radon into the atmosphere is molecular diffusion (IAEA, 2005; Nabil et al, 2009; UNSCEAR, 2000b). For a porous mass of homogeneous material semi-infinite in extent, the flux density of radon per unit area of the bulk porous medium (radon exhalation rate of pores plus solids), rather than per unit area of the pore air, migrated through the surface of dry soil, J_D ($\text{Bq/m}^2\text{s}$), is given by equation (3.7-3):

$$J_D = C_{Ra} \lambda_{Rn} \rho_s (1 - \epsilon) f L \quad (3.7-3)$$

Where,

- C_{Ra} is the activity concentration of ^{226}Ra in earth material (Bq/kg),
- λ_{Rn} is the decay constant of radon ($2.0974 \times 10^{-6} S^{-1}$),
- f is the emanation fraction for earth material (0.2),
- ρ_s is the soil grain density (2700 kg/m^3) and
- ϵ is the porosity of dry earth material (0.25).

This formula is valid only for dry soil. The range of diffusion (diffusion length, L , in meter) is equal to $\sqrt{D_e/\lambda_{Rn}}$, where $D_e = 2 \times 10^{-6} \text{ m}^2/\text{s}$.

3.7.3 DETERMINATION OF RADON EMANATION COEFFICIENT

The samples were prepared as illustrated in section 3.3, and γ rays of ^{214}Pb and ^{214}Bi of each sample were measured at 2 hours after sealed (I_1) to allow the short lived ^{214}Pb and ^{214}Bi isotopes to grow into secular equilibrium with the radon still remaining in the sample. Then, they were measured again at 30 days (I_2) after the radioactive secular equilibrium between radon and radium was achieved. Then, based on White and Rood methods (Faanu, 2011; Nabil et al, 2009), the radon emanation coefficient was calculated by dividing radon activity concentration (Bq) in that space by the radium activity concentration (Bq) in those materials.

$$I_1 = A_0 + N(1 - e^{-\lambda t_1}) \quad (3.7-4)$$

$$I_2 = A_0 + N(1 - e^{-\lambda t_2}) \quad (3.7-5)$$

Where;

- I_1 is the activity of radon at t_1 (Bq/m^3),
- I_2 is the activity of radon at secular equilibrium,
- A_0 is the bound ^{222}R activity of at $t=0$,
- t_1 is the counting time after sealing (2 hours),
- t_2 is the counting time after 30 days (10 hours),

- N is the ^{222}Rn activity at radioactive equilibrium i.e. at t_2 , and
- λ is the ^{222}Rn decay constant.

Based on the concentration of ^{222}Rn during the time interval between sealing (t_1) and after 30 days (t_2), the emanation factor of ^{222}Rn can be derived from equation (3.7-4) and (3.7-5) (Faanu, 2011; M.Nabil et al, 2009) as follows:

$$N = \frac{I_1 - I_2}{1 - e^{-\lambda t_1} - 1 - e^{-\lambda t_2}} \quad (3.7-6)$$

$$A_0 = \frac{I_2 1 - e^{-\lambda t_1} - I_1 1 - e^{-\lambda t_2}}{1 - e^{-\lambda t_1} - 1 - e^{-\lambda t_2}} \quad (3.7-7)$$

Radon emanation coefficient, EC , is defined as mathematically as follows:

$$EC = \frac{\text{net radon released}}{\text{total radon released}} \quad (3.7-8)$$

$$EC = \frac{N}{N + A_0} \quad (3.7-9)$$



CHAPTER FOUR

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.

4.1 HPGe DETECTOR CHARACTERIZATION

4.1.2 ENERGY CALIBRATION

A 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:

$$E = 0.244 * C_n - 0.0998, \quad \text{where } R^2 = 1 \quad (4.1-1)$$

Where,

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

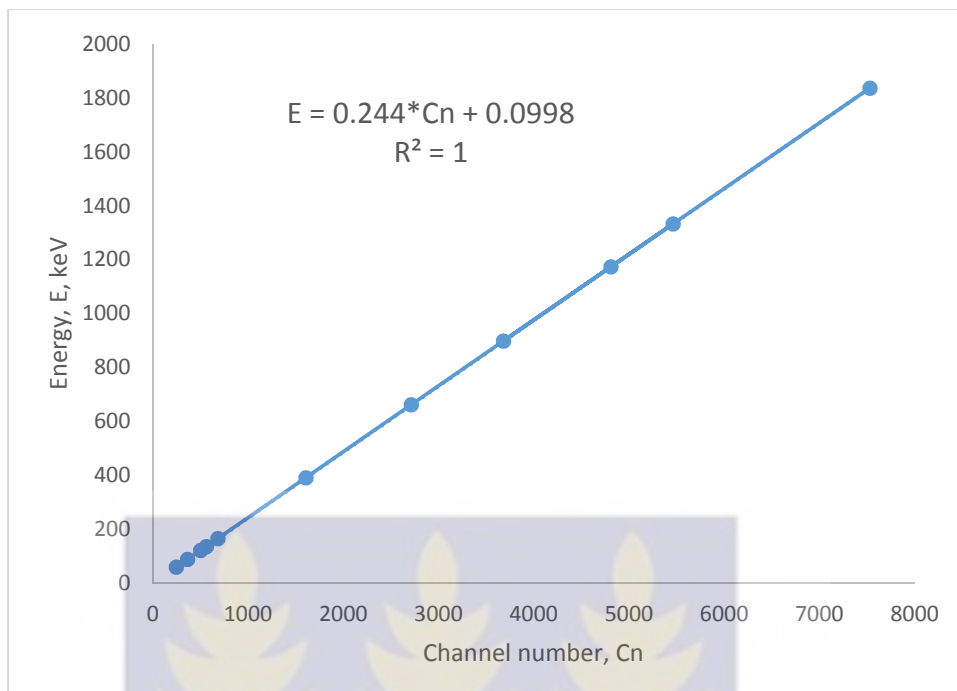


Figure 4.1-1: Energy calibration curve using mixed radionuclides standard

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

4.1.2 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):

$$\varepsilon_f = 1.3803 E^{-0.694}, \quad \text{where } R^2 = 0.9992 \quad (4.1-2)$$

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

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

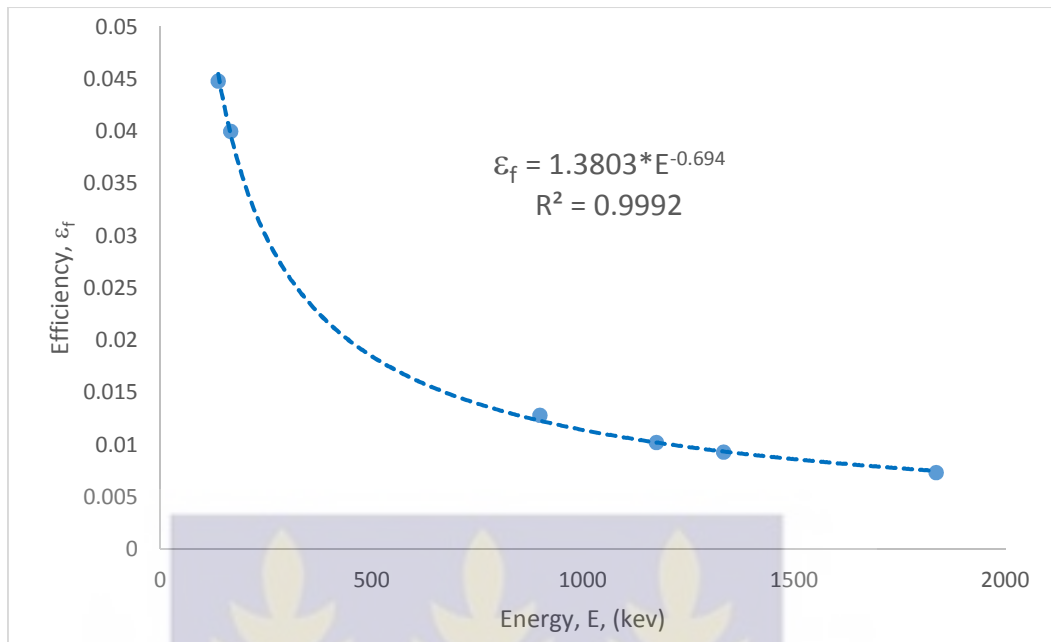


Figure 4.1-2: Efficiency calibration curve using mixed radionuclides standard

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.

4.1.3 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 4.1 -1. These values were evaluated from the blank sample for its minimum activity.

Table 4.1-1 Calculated MDA of ²³⁸U, ²²⁶Ra, ²³²Th and ⁴⁰K.

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).

4.2 ANALYSIS OF ACTIVITY CONCENTRATION

4.2.1 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 4.2-1. Each of corresponding counting rates in all samples activity concentration has its own error and it was computed using equation (3.6-2). Table 4.2-1 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 4.2-1: Activity of ^{238}U , ^{232}Th decay series and ^{40}K in tantalum ore

Samples code	Activity concentration, Bq/kg		
	$^{238}\text{U}\pm\text{Error}$	$^{232}\text{Th}\pm\text{Error}$	$^{40}\text{K}\pm\text{Error}$
TaO1	54.990 \pm 1.125	25.650 \pm 0.490	629.870 \pm 57.440
TaO2	69.791 \pm 1.456	26.213 \pm 5.000	627.005 \pm 57.179
ROC	111.177 \pm 1.712	22.970 \pm 0.487	552.634 \pm 50.419
Mean \pm Err (This work)	78.653 \pm 1.431	24.945 \pm 0.492	603.170 \pm 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 4.2-1, the average specific activity concentrations of ^{232}Th are almost two times lower than the world average value. However ^{238}U 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 ^{40}K were also found to be higher than the worldwide average activity concentrations; but lower than

IAEA 2003 report for ^{40}K 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 ^{238}U decay series (UNSCEAR, 2000b). Therefore, this high activity concentration of ^{238}U and ^{40}K 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.

4.2.2 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 (\pm error) of activity concentration in Bq/kg for ^{238}U , ^{232}Th and ^{40}K 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 4.2-2: 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 \pm 1.104	16.583 \pm 0.257	723.177 \pm 65.923
SS2	67.842 \pm 1.059	14.374 \pm 0.205	714.583 \pm 65.139
Mean \pm err (This work)	69.354 \pm 1.081	15.479 \pm 0.231	718.880 \pm 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

a: UNSCEAR 2000

From Table 4.2-2, 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 UNSCAER 2000 report, all soils contains terrestrial radionuclides present at trace levels, therefore, the soil bearing granite-pegmatite rocks of Kenticha may contains 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.

4.2.3 SOLID WASTE SAMPLES

Table 4.2-3 displays the range and average values (\pm 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 4.2-3: Activity of ^{238}U , ^{232}Th decay series and ^{40}K in solid waste samples

Samples code	Activity concentration, Bq/kg		
	$^{238}\text{U}\pm$ Error	$^{232}\text{Th}\pm$ Error	$^{40}\text{K}\pm$ Error
WJT	64.235 \pm 0.922	19.989 \pm 0.286	723.033 \pm 65.910
FW	60.821 \pm 0.872	17.906 \pm 0.258	717.305 \pm 65.387
WR	137.315 \pm 2.333	11.756 \pm 0.239	534.232 \pm 48.744
WT1	148.331 \pm 2.518	13.085 \pm 0.270	559.991 \pm 51.095
WT2	141.779 \pm 2.889	12.311 \pm 0.317	501.784 \pm 45.737
Mean\pmerr (This work)	110.496 \pm 1.907	15.009 \pm 0.274	607.269 \pm 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 show the granite-pegmatite rocks of Kenticha has very low level of thorium content.

Figure 4.2-1 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.

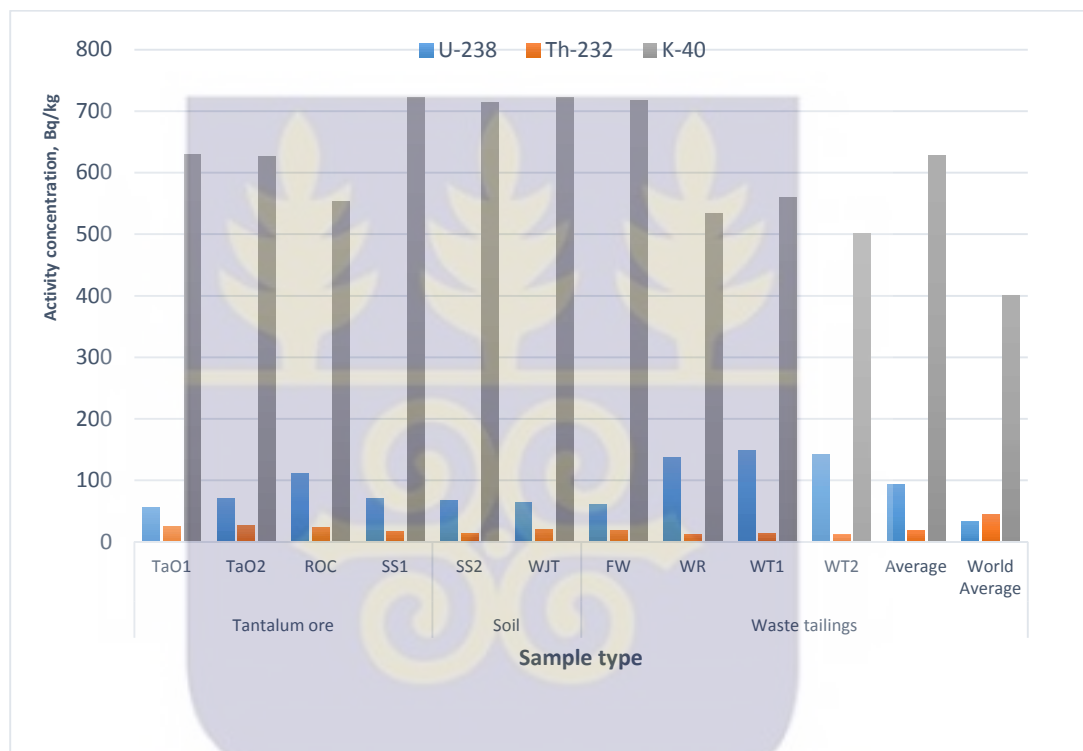


Figure 4.2-1: Plot of activity concentration for ^{232}Th , ^{238}U and ^{40}K in all samples

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 4.2-2 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.

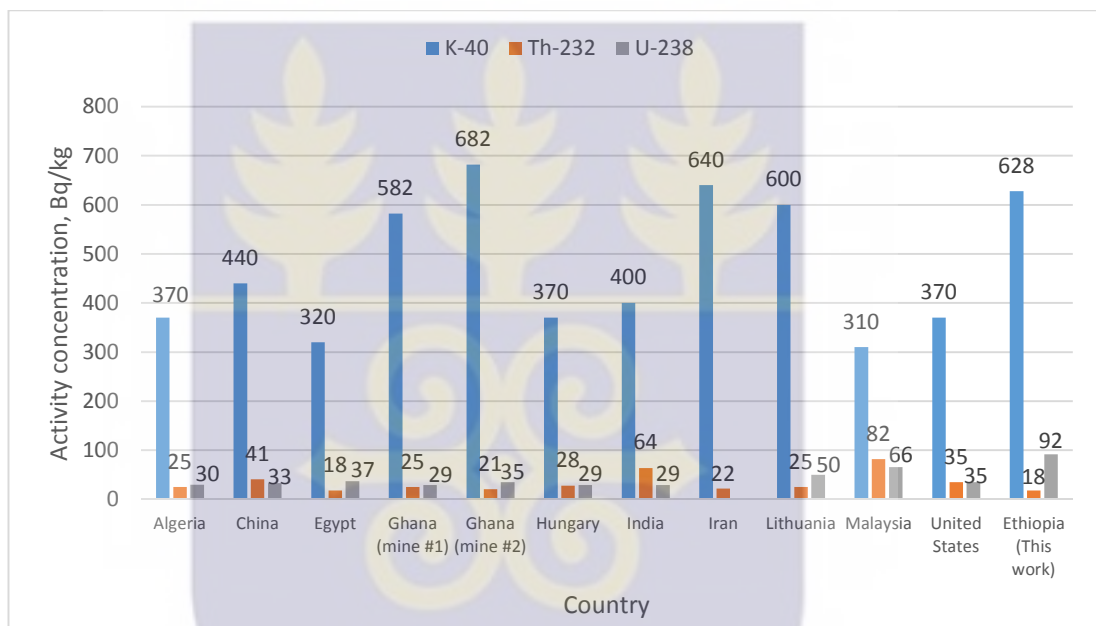


Figure 4.2-2: Comparison of concentrations of ^{238}U , ^{232}Th and ^{40}K in soils with published data (Darko et al, 2010; UNSCEAR, 2000c)

Figure 4.2-2 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 $^{238}U/^{40}K$ ratio for samples of the study area was found in proportion compared with the ratio for other countries as shown in Figure 4.3-3.

Table 4.2-4: Concentration ratios of ^{238}U to ^{232}Th and ^{238}U to ^{40}K in tantalum ores

Nuclides	Samples code										Mean±STD
	Tantalum ore			Soil		Solid waste samples					
	TaO1	TaO2	ROC	SS1	SS2	WJT	FW	WR	WT1	WT2	
$^{238}U/^{232}Th$	2.14	2.66	4.84	4.27	4.72	3.21	3.40	9.40	11.34	11.52	5.75±3.60
$^{238}U/^{40}K$	0.09	0.11	0.20	0.10	0.10	0.09	0.09	0.21	0.27	0.28	0.15±0.08

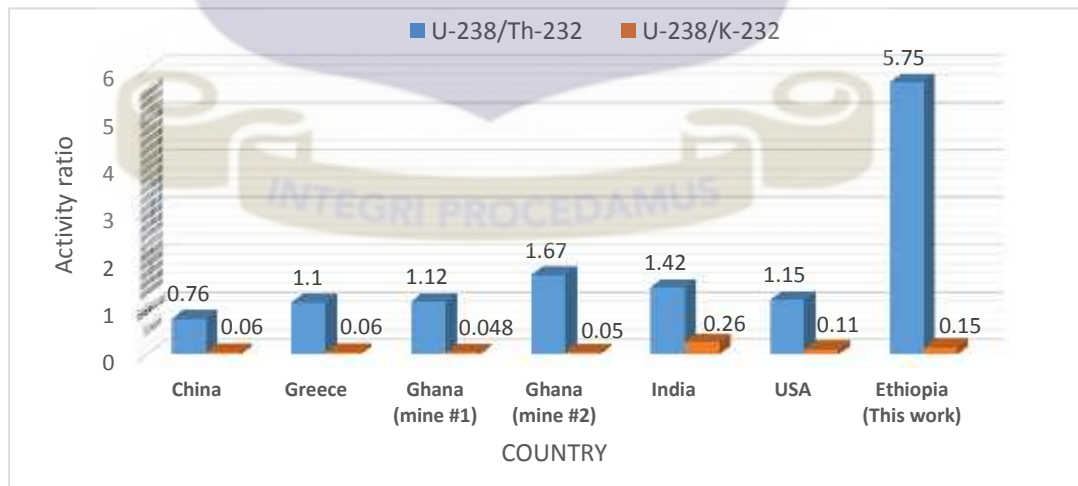


Figure 4.2-3: comparison of activity concentration ratios for $^{238}U/^{232}Th$ and $^{238}U/^{40}K$ (Darko et al, 2010; UNSCEAR, 2000c)

As shown in Figure 4.2-3, the result obtained for $^{238}U/^{232}Th$ and $^{238}U/^{40}K$ 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, P^H , and nature of mine in these countries.

4.3 DOSE ASSESSMENT DUE TO NORM

4.3.1 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 4.3-1: D_γ and AED due to ^{238}U , ^{232}Th and ^{40}K compared with the WAV

Nuclides	External α -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) ^{3b}		
Worldwide Range	18 – 93 ^b			0.3 - 0.6 ^b		

In Table 4.3-1, 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

³ Parenthesis shows the sum of outdoor and indoor annual effective dose, respectively, a: UNSCEAR 2000, b: UNSCEAR 1993

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.

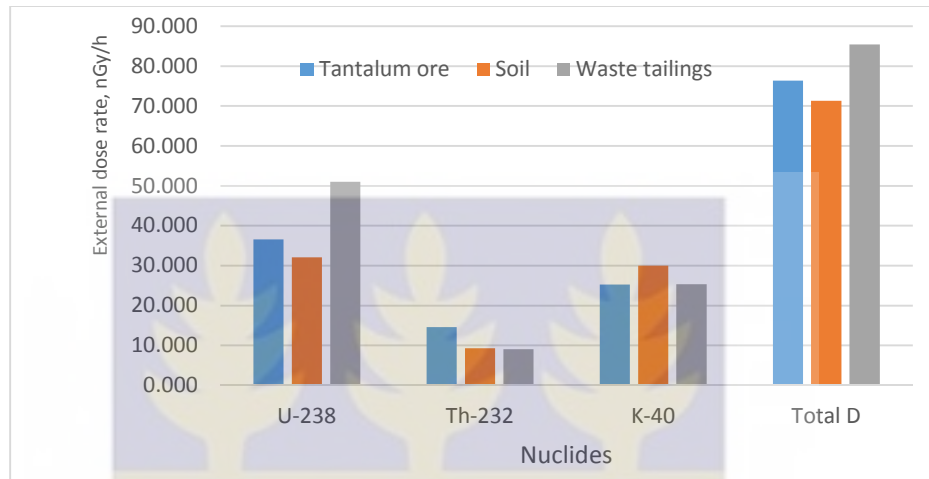


Figure 4.3-1: Comparison of γ - dose due ^{238}U , ^{232}Th and ^{40}K in ore, soil and solid waste samples.

Figure 4.3-1 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 reveal the significance of high NORM activity concentration of the area.

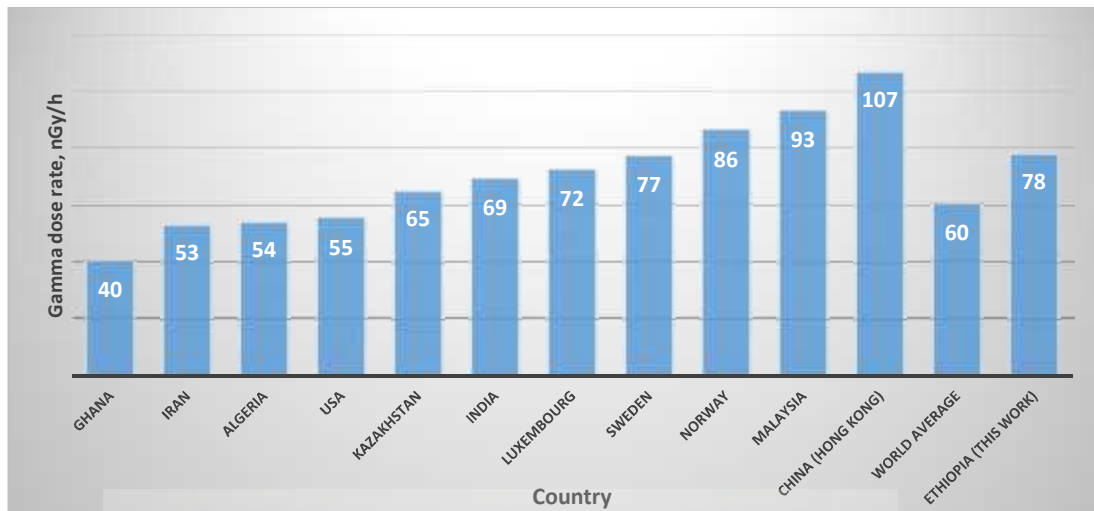


Figure 4.3-2: Comparison of absorbed dose rate from NORMs in soil by country (UNSCEAR, 2000d)

Figure 4.3-2 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).

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

Table 4.3-1 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 AED of the study area is three times lower than the worldwide average value of 0.07 mSv/year (Al Murgen, 2015; UNSCEAR, 2000b). Therefore, the AED from ^{238}U , ^{232}Th and ^{40}K activity concentrations falls within the public AED limit of 1 mSv (IAEA, 2010).

Figure 4.3-3 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.

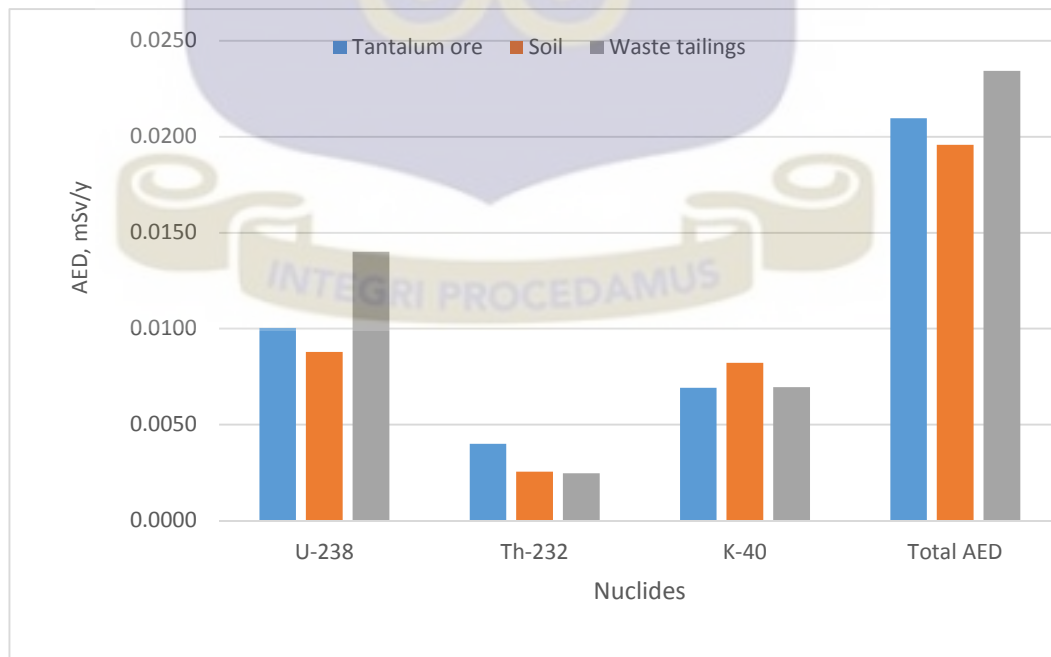


Figure 4.3-3: Plots of AED due ^{238}U , ^{232}Th and ^{40}K of ore, soil and solid waste samples

4.4 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 waste as 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 4.4-1: Estimated cancer 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 $1.06\text{E}-06$ - $1.57\text{E}-06$ with an average value of $1.24\text{E}-06$. 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 $8.20\text{E}-07$ - $1.22\text{E}-06$ with an average value of $9.61\text{E}-07$ 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 $1.38\text{E-}07$ - $2.05\text{E-}07$ and $8.29\text{E-}08$ - $1.23\text{E-}07$ with an average values of $1.62\text{E-}07$ and $9.72\text{E-}08$, 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 $1.0\text{E-}6$ – $1.0\text{E-}4$ values for the population of the study area. The EPA acceptable risk value of $1.0\text{E-}6$, i.e. 1 case out of 1 million people dying from cancer, is considered as inconsequential (Faanu, 2011).

4.5 RADIOLOGICAL HAZARD ASSESSMENT

This sub-section outline 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.

4.5.1 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 4.5-1 shows a summary of the results obtained for tantalum ore samples from the Kenticha tantalum mine area.

Table 4.5-1: Hazard indices due to ^{238}U , ^{232}Th and ^{40}K in ore samples

Sample code	Ra_{eq} , Bq/kg	H_{ex}	H_{in}	I_{x}
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\pmSTD (This work)	151.6 \pm 25.20	0.409 \pm 0.07	0.597 \pm 0.15	1.115 \pm 0.16
Range\pmSTD (This work)	136 - 180.7 \pm 25.20	0.367 - 0.488 \pm 0.07	0.503 - 0.773 \pm 0.15	1.015 - 1.30 \pm 0.16
World average	<370	<1	<1	<1

The use of a material whose radium equivalent activity concentration exceeds 370 Bq/kg is discouraged to avoid radiation hazards (Avwiri et al, 2012). However, the average Ra_{eq} 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.

4.5.2 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 4.5-2: Hazard indices due to ^{238}U , ^{232}Th and ^{40}K in soil samples

Sample code	Ra_{eq} , Bq/kg	H_{ex}	H_{in}	I_{x}
SS1	149.5	0.404	0.593	1.115
SS2	143.3	0.387	0.570	1.072
Mean\pmSTD (This work)	146.4 \pm 4.36	0.395.01	0.582 \pm 0.02	1.094 \pm 0.03
Range\pmSTD (This work)	143.3 - 149.5 \pm 4.36	0.387 - 0.404 \pm 0.01	0.570 - 0.593 \pm 0.02	1.072 - 1.125 \pm 0.03
World average	<370	<1	<1	<1

From Table 4.5-2, 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 370 Bq/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.

4.5.3 SOLID WASTE SAMPLES

Table 4.5-3 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 4.5-3: 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 370 Bq/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.

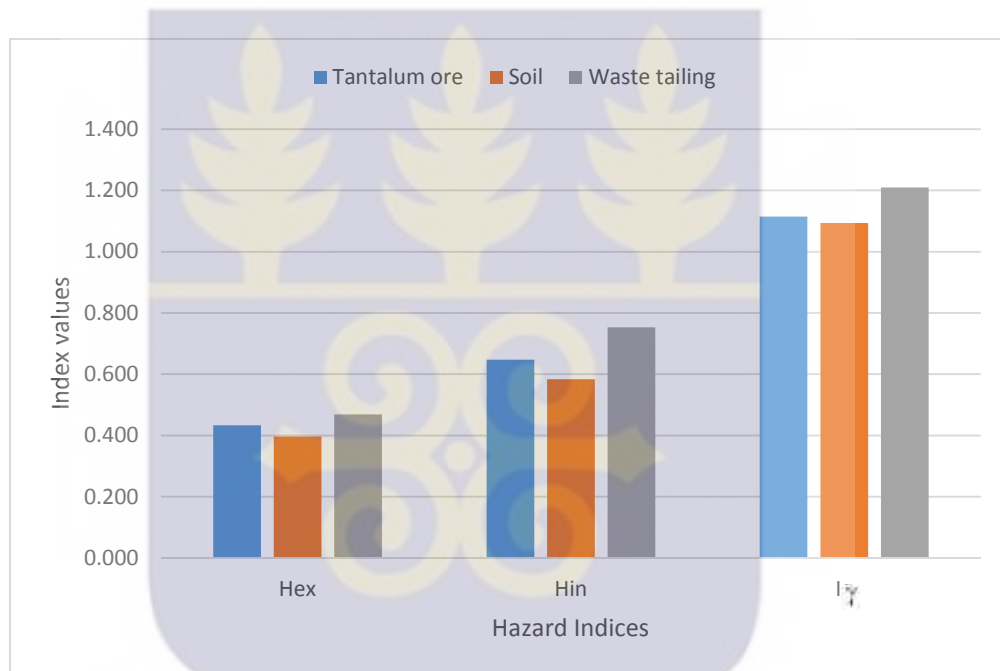


Figure 4.5-1: Comparison of the radiological hazard indices for ore, soil and solid waste samples

Figure 4.5-1 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 4.5-4: 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	
Australia	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
Ghana	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
Turkey	145	40	28	248.3	99.1
Ethiopia (This work)	10	78.9	18.5	643.1	154.8

According to Table 4.5-4, 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 safe for use as building material.

4.6 ANALYSIS OF RADON

4.6.1 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 4.6-1.

Table 4.6-1: ^{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 mBq/ m².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).

4.6.2 SOIL SAMPLES

Two composite samples from the six different soil samples collected from the different location of Kenticha tantalum mine (SS₁ – SS₆) were used for analysis to

calculate the range and mean (\pm STD) values for activity concentration of ^{226}Ra and concentration (C_{Rn}), emanation coefficient (EC) and exhalation rate (J_{D}) of ^{222}Rn . Table 4.6-2 shows a summary of the results obtained for the composite soil samples from the study area.

Table 4.6-2: ^{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/m³	EC, %	J_{D}, mBq/m².s
SS1	70.1	113.6	84.2	58.1
SS2	67.8	109.8	81.5	56.2
Mean\pmSTD (This work)	68.9 \pm 1.7	111.7 \pm 2.7	82.9 \pm 1.9	57.2 \pm 1.4
Range\pmSTD (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 time 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 mBq/ m²s (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.

4.6.3 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 (J_D) 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 4.6-3: ^{226}Ra concentration and corresponding C_{Rn} , EF and J_D in solid waste samples.

Samples code	A_{Ra-226}, Bq/kg	C_{Rn}, kBq/m³	EC, %	J_D, mBq/m².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 4.6-3, 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 33mBq/ 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 4.7-1 illustrates the comparison for the radon concentration in the analyzed samples.

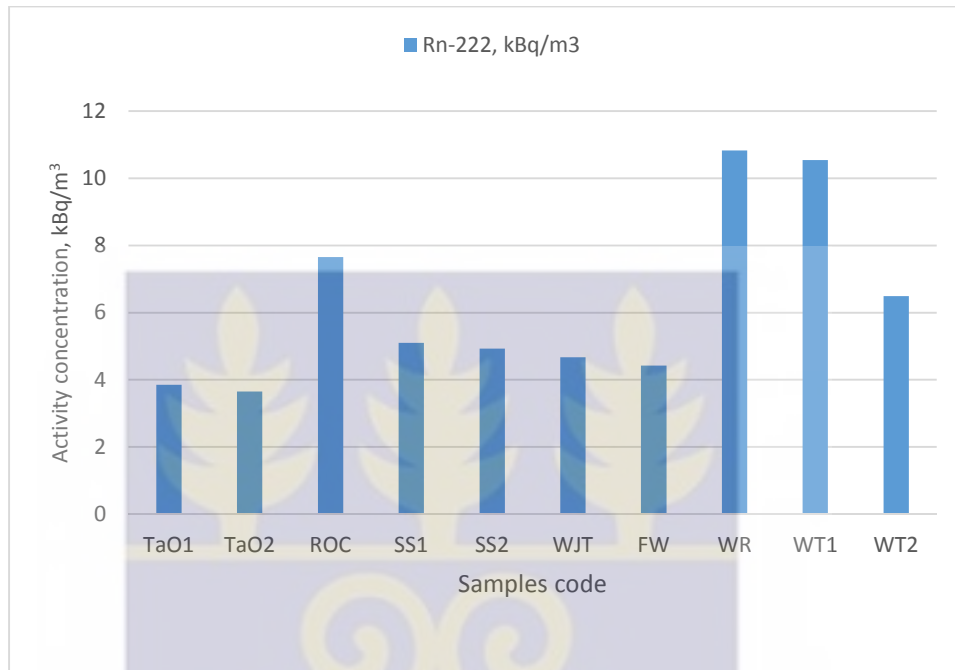


Figure 4.6-1: Plot of activity concentration for ^{222}Rn in all samples

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

a) 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 4.6-4: 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 4.6-5: 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 4.6-6, 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 phosphogypsum samples of some countries in the world (UNSCEAR, 2000b).

Table 4.6-6: 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 4.6-6, 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.

4.7 WATER SAMPLES

Water samples collected from Kenticha tantalum mine waste tailing dam, tap water from office, spring water, pure water dam, and Mormora 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 FIVE

CONCLUSION AND RECOMMENDATION

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

5.1 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/y, 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 slightly 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.

5.2 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.

5.2.1 RESEARCHERS

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.

5.2.2 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 plays 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.

5.2.3 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 mine as 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.

5.2.4 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.



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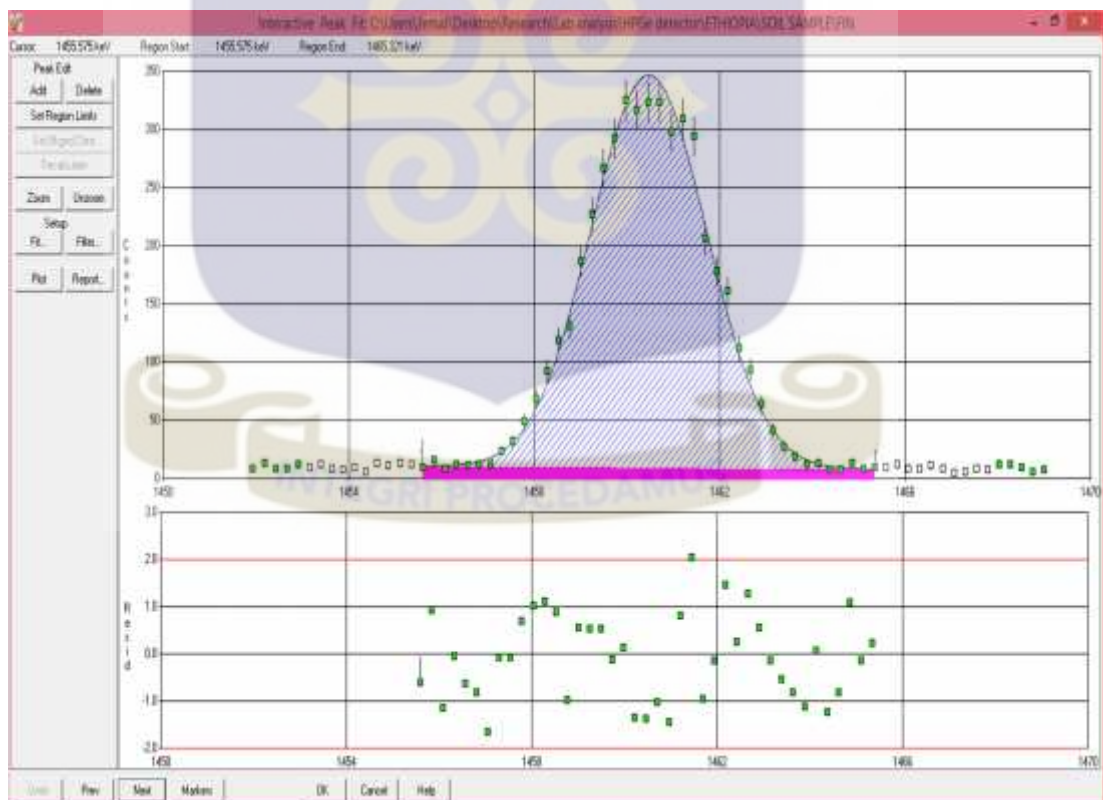
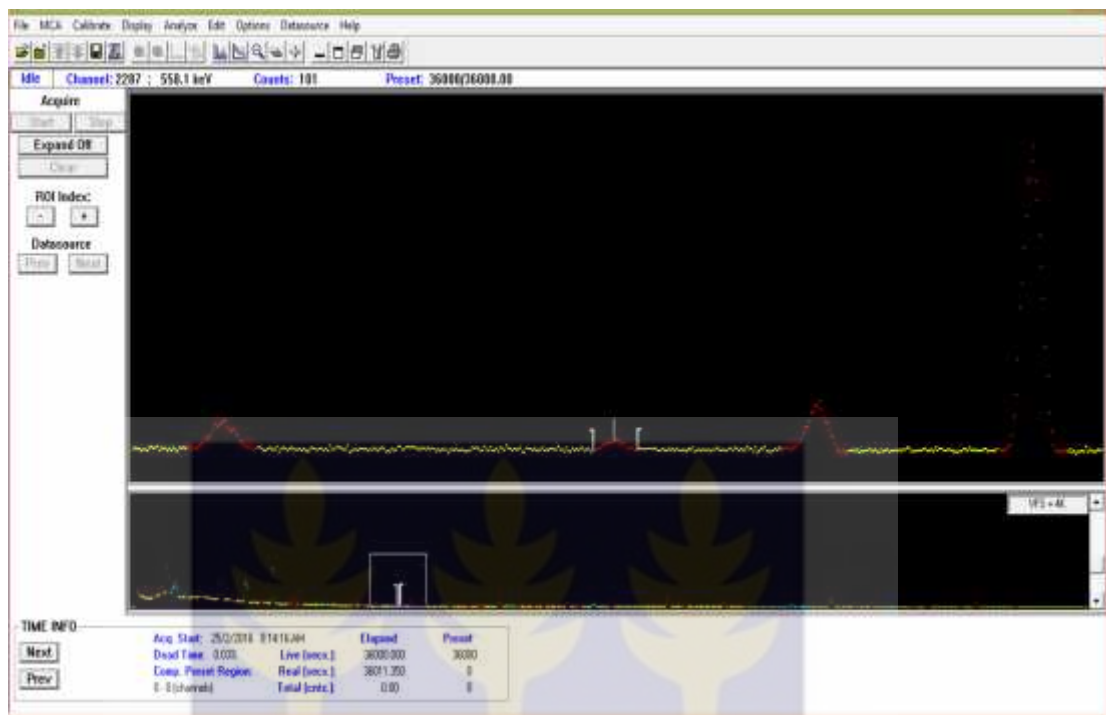


APPENDICES

**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
SS6	Soil sample taken from around the open pit	5°27'19.34"N	39° 1'21.21"E

APPENDIX II: Screenshot of Genie 2000 user interface for Soil sample, SS1



APPENDIX III: Mixed radionuclide standard specifications used for HPGe detector calibration



CZECH METROLOGY INSTITUTE
INSPECTORATE FOR IONIZING RADIATION



Radiová 1, 102 00 Praha 10

CERTIFICATE

Cert. No: 9031 - OL - 146/14 Type: MBSS 2 Prod. No: 050214-1425039

Radionuclide	Half life days	Activity kBq	Combined standard uncertainty, %
Am-241	157800	4,694	1,1
Cd-109	462,6	14,54	1,4
Ce-139	137,5	1,355	1,1
Co-57	271,26	1,156	1,1
Co-60	1925,4	2,697	1,1
Cs-137	11019	2,689	1,3
Sn-113	115,1	4,000	2,2
Sr-85	64,78	4,570	1,5
Y-88	106,6	5,323	1,2

Mass: 980,0 g Density: 0,98 g/cm³ Volume: 1000 cm³
Radionuclide impurities: gamma < 0,1 % Homogeneity better than: 1 %
Reference date: 20.3.2014

Description:
Radioactive material is homogeneously dispersed in silicone resin. Composition of the matrix: C - 0,324
H - 0,0816 O - 0,216 Si - 0,379 (mass ratio).

Measuring method:
Preparation issues from standard ER solutions whose activities were determined by suitable absolute method. Final control is based on gamma spectrometry on HPGe detector.

Note:
As the criterion of homogeneity standard deviation of the activity value of 1 cm³ element was chosen (n=10). The volume is calculated from the mass and the density.

Date of the certificate issue: 25.2.2014 Validity: 3 years

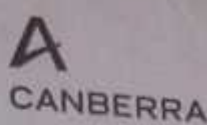
Customer:
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Wienersiedlung 6
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Austria


 Ing. Jiří Suráň, MBA
 director

Control: RNDr. Richard Blud'ovský, CSc., RNDr. Pavel Dryák, CSc.

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Appendix IV: DETECTOR SPECIFICATIONS AND PERFORMANCE DATA



DETECTOR SPECIFICATION AND PERFORMANCE DATA

Specifications

Detector Model GX4020 Serial number 614120
 Cryostat Model 7500SL
 Preamplifier Model 2002CSL

The purchase specifications and therefore the warranted performance of this detector are as follows:

Nominal volume cc Relative efficiency 40%
 Resolution 2.0 keV (FWHM) at 1.33 MeV
 keV (FWHM) at 1.33 MeV
 1.10 keV (FWHM) at 122 keV
 keV (FWHM) at
 Peak/Compton 56:1 Cryostat well diameter mm Well depth mm
 Cryostat description or Drawing Number if special 7500SL

Physical Characteristics

Geometry Coaxial one open end, closed end facing window
 Diameter 60.5 mm Active volume cc
 Length 61.5 mm Crystal well depth mm
 Distance from window (outside) 6 mm Crystal well diameter mm

Electrical Characteristics

Depletion voltage (+4000) Vdc
 Recommended bias voltage Vdc (+4500) Vdc
 Leakage current at recommended bias 0.01 nA
 Preamplifier test point voltage at recommended voltage 0.8 Vdc

Resolution and Efficiency

With amp time constant of 4 μ s

Isotope	⁶⁰ Co	⁶⁰ Co		
Energy (keV)	122	1332		
FWHM (keV)	878	1.92		
FWTM (keV)		1.51		
Peak/Compton		63.4:1		
Rel. Efficiency		44.2%		

- Tests are performed following IEEE standard test ANSI/IEEE std325-1996
 - Standard Canberra electronics used - See Germanium detector manual Section 7

Tested by: [Signature] Date: January 29, 2014
 Approved by: [Signature] Date: January 29, 2014

12/03/2007

GD/AME001/F