

**EVALUATION OF NATURAL RADIOACTIVITY AND HAZARD
INDICES IN THE SOIL COLLECTED FROM THE RESIDENTIAL
COLLEGE AREAS OF UNIVERSITY MALAYA, MALAYSIA**

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**FACULTY OF SCIENCE
UNIVERSITY OF MALAYA
KUALA LUMPUR**

2017

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HAZARD INDICES IN THE SOIL COLLECTED FROM
THE RESIDENTIAL COLLEGE AREAS OF UNIVERSITY
MALAYA, MALAYSIA**

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**DISSERTATION SUBMITTED IN PARTIAL
FULFILMENT OF THE REQUIREMENTS FOR
THE DEGREE OF MASTER OF TECHNOLOGY
(MATERIALS SCIENCE)**

**DEPARTMENT OF PHYSICS
FACULTY OF SCIENCE
UNIVERSITY OF MALAYA
KUALA LUMPUR**

2017

UNIVERSITY OF MALAYA
ORIGINAL LITERARY WORK DECLARATION

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Field of Study: **Experimental Physics (Radiation)**

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ABSTRACT

Realizing the heterogeneous distribution of naturally occurring radioactive materials (NORM) in all earth born media, it is important to assess their concentrations in residential hostels environment to control the occupants' health from radiation exposure. In connection with this, soil samples collected from all residential colleges of University of Malaya were assessed to obtain the concentrations of NORM by using HPGe gamma-ray spectrometry. The activity concentrations of ^{226}Ra ranged from 42.16 to 61.27 Bq/kg, ^{232}Th ranged from 52.89 to 81.80 Bq/kg and ^{40}K ranged from 73.07 to 306.43 Bq/kg. Radium-226, Thorium-232 and Potassium-40 have average activity concentration of 52.31 ± 2.40 Bq/kg, 68.42 ± 1.92 Bq/kg and 224.58 ± 2.28 Bq/kg respectively. The results show that the average concentrations of the primordial radionuclides were lower than Malaysia average values. But ^{232}Th and ^{226}Ra are higher than the world average while ^{40}K is lower compared with values giving in UNSCEAR 2000. The dose rate and radiological indices were obtained and compared with worldwide recommended values

ABSTRAK

Atas kesedaran mengenai taburan heterogen bahan radioaktif terwujud semulajadi (NORM) pada semua media bawakan bumi, adalah penting penilaian ketumpatan bahan tersebut di persekitaran asrama penginapan Universiti Malaya dikaji bagi menjamin kesihatan penghuni. Berkaitan ini, sampel tanah dipungut dari semua kawasan asrama penginapan di Universiti Malaya untuk penilaian NORM menggunakan spectrometer sinar gamma HPGe. Ketumpatan aktiviti dari 42.16 ke 61.27 Bq/kg bagi 226-Ra, 52.89 ke 81.80 Bq/kg bagi 232-Th dan 73.07 ke 306.43 Bq/kg bagi 40-K didapati. Ketumpatan aktiviti 52.31 ± 2.40 Bq/kg, 68.42 ± 1.92 Bq/kg and 224.58 ± 2.28 Bq/kg diukur bagi 226-Ra, 232-Th dan 40-K masing-masing. Hasil pengukuran mendapati nilai ketumpatan purata radionuclid kuno adalah lebih rendah dari nilai purata. Negara Malaysia. Walaubagaimana pun nilai bagi 232-Th dan 226-Ra adalah lebih tinggi dari purata dunia, manakala nilai bagi 40-K adalah lebih rendah, berpandukan UNSCEAR 2000. Kadar dos dan indeks radiologi didapati dari hasil pengukuran dan dibandingkan dengan nilai yang anjurkan sedunia.

ACKNOWLEDGEMENTS

This dissertation is dedicated to my beloved wife, Sarah Nneji and wonderful children (Obiageri Nneji, Chinonye Nneji and Ihechukwu Nneji). They really sacrificed and encouraged me in the study and writing of this dissertation. My wife's understanding and encouragement kept me working when I wanted to give up.

My special thanks and appreciation goes to Prof. Dr. Mayeen Uddin Khandaker, my ideal supervisor. His sage advice, insightful criticisms, and patient encouragement aided me so much. I would also like to thank Prof Yusoff Bin Mohd Amin whose support of this project was greatly needed and deeply appreciated.

I would like to thank my mom and my brothers and sister for supporting me spiritually throughout this study and my life in general

I must acknowledge as well the many friends, colleagues, fellow students and teachers, especially Dr. Matthew Tikpangi Kolo, who assisted, advised, and supported my research and writing efforts.

TABLE OF CONTENTS

Abstract	iii
Abstrak	iv
Acknowledgements	v
Table of Contents	vi
List of Figures	x
CHAPTER 1: INTRODUCTION.....	1
1.1 Soil and Radionuclides	1
1.2 Radioactivity.....	1
1.3 NORM and TENORM.....	2
1.3.1 Cosmogenic radionuclides and radiation.....	3
1.3.2 Primordial (Terrestrial) Radionuclides.....	5
1.3.2.1 Radioactivity decay series.....	6
1.3.2.2 Potassium-40 (⁴⁰ K).....	13
1.4 Man-made (Artificial) Sources of Radiation	13
1.5 Radionuclides transfer in the environment.....	14
1.6 Biological effect of ionizing radiation.....	16
1.7 Classification of radiation effect on biological system.....	17
1.7.1 Stochastic effects.....	17
1.7.2 Deterministic effects.....	18
1.8 Justification/research significant.....	19
1.9 Aims and objectives.....	20
CHAPTER 2: LITERATURE REVIEW.....	21
2.1 Atoms and radiation.....	21

2.2	Radioactive half life.....	22
2.3	Activity	23
2.4	Radioactive equilibrium.....	24
2.5	Types of radiation	25
2.5.1	Alpha (α) particles.....	26
2.5.2	Beta (β) particle.....	26
2.5.3	Gamma (γ) radiation.....	27
2.5.4	Differences between Alpha, beta and gamma	27
2.5.5	Characteristics of alpha beta and gamma rays	28
2.6	Spontaneous fission	28
2.7	Intensity	29
2.8	Interaction of gamma rays with matter.....	29
2.8.1	Photoelectric effect.....	30
2.8.2	Compton effect.....	31
2.8.3	Pair production.....	33
2.9	Review of existing knowledge	34
CHAPTER 3: MATERIALS AND METHODS		37
3.1	Radiation detection techniques	37
3.2	Gamma ray spectroscopy.....	37
3.2.1	High purity germanium detector (HPGe)	38
3.2.2	Preamplifier	39
3.2.3	Main amplifier	39
3.2.4	Multi-channel analyser (MCA)	40
3.3	Detector resolution.....	40
3.4	Calibration of the detector	41

3.4.1	Energy calibration	41
3.4.2	Efficiency calibration	42
3.4.3	Determination of minimum detectable activity	43
3.4.4	Background counting	44
3.5	Area of Study	44
3.6	Sample Sampling	45
3.7	Sample preparation	48
CHAPTER 4: MEASUREMENTS AND RESULTS.....		50
4.1	Measurement of ^{226}Ra , ^{232}Th and ^{40}K radioactivities	50
4.2	Activity concentration	51
4.3	Hazard indices	52
4.3.1	Radium equivalent activity.....	52
4.3.2	Absorbed dose rate (D_R).....	53
4.3.3	Annual effective dose equivalent (AEDE).....	54
4.3.4	Annual gonadal dose equivalent (AGDE).....	55
4.3.5	Activity utilization index (AUI)	56
4.3.6	External and Internal hazard indices (H_{ex} and H_{in})	57
4.3.7	Representative gamma index ($I_{\gamma r}$).....	58
4.3.8	Excess lifetime cancer risk (ELCR)	59
CHAPTER 5: DISCUSSION		72
5.1	Radioactivity levels of ^{226}Ra , ^{232}Th and ^{40}K in the residential college areas of University of Malaya	72
5.2	Radium equivalent Index	74
5.3	Absorbed dose rate	74
5.4	Annual effective dose equivalent.....	74

5.5	Annual gonadal dose equivalent.....	75
5.6	Activity utilization index	75
5.7	External hazard indices (H_{ex}).....	75
5.8	Internal hazard indices (H_{in})	76
5.9	Representative gamma index.....	76
5.10	Excess lifetime cancer risk	76
 CHAPTER 6: CONCLUSION.....		77
6.1	Limitation and suggestion for further studies.....	78
	References	79

University of Malaya

LIST OF FIGURES

Figure 1.1 Thorium 232 decay series.....	7
Figure 1.2 The uranium-238 decay series.....	9
Figure 1.3 Actinium decay series.....	11
Figure 1.4 Neptunium decay series.....	12
Figure 1.5 Routes of human exposure to sources of natural and man made.....	15
Figure 3.1 Diagram of Gamma Spectrometer Experimental setup.....	38
Figure 3.2 Detection efficiency curve of the detector.....	43
Figure 3.3 Location of University Malaya (N3°7'15''E101°39'23'') Kuala Lumpur, Malaysia.....	47
Figure 3.4 Collection of samples at the sample sites.....	48
Figure 3.5 Photos of samples preparation (oven drying, weighing and storage).....	49
Figure 4.1 Photo of P-type Coaxial ORTEC, GEM-25 high purity germanium Gamma ray detector with the MCA and gamma vision PC.....	51

LIST OF TABLES

Table 1.1 Average radiation dose from natural sources (UNSCEAR 2000)	5
Table 1.2 Average radioactive decay series (Tait, 1980).....	6
Table 1.3 Thorium decay series with half-life.....	7
Table 1.4 Natural abundance of uranium isotopes.....	8
Table 1.5 Uranium -238 decay series with half-life.....	9
Table 1.6 Uranium-235 (Actinium) decay series with half-life.....	10
Table 1.1 Neptunium decay series with half-life.....	12
Table 2.1 Characteristics of alpha beta and gamma rays	28
Table 3.1 Energy calibration	42
Table 3.2 Radionuclide, Energy and Detection efficiency	43
Table 3.3 Sampling sites coordinates.....	45
Table 4.1 Mean of activity concentration for ^{226}Ra , ^{232}Th and ^{40}K in the soil samples from residential college areas of University of Malaya	52
Table 4.2 Mean of radium equivalent activity for ^{226}Ra , ^{232}Th and ^{40}K in the Soil samples from residential college areas of University of Malaya.....	53
Table 4.3 Mean of absorbed dose rate values in the soil samples from Residential college areas of University of Malaya.	54
Table 4.4 Mean of annual effective dose equivalent (AEDE) values in the soil samples from residential college areas of University of Malaya.	55
Table 4.5 Mean of annual gonadal dose equivalent (AGDE) values in the soil Samples from residential college areas of University of Malaya.	56
Table 4.6 Mean of activity utilization index (AUI) in the soil samples from residential college areas of University of Malaya.....	57

Table 4.7 Mean of external and internal hazard indices values in the soil samples from residential college areas of University of Malaya	58
Table 4.8 Mean of representative gamma index indices values in the soil samples from residential college areas of University of Malaya	59
Table 4.9 Mean of excess lifetime cancer risk (ELCR) values in the soil samples from residential college areas of University of Malaya.	60
Table 4.10 summary of activity concentration and radiological hazard parameters in the soil samples from residential college areas of University of Malaya	61

University of Malaya

CHAPTER 1: INTRODUCTION

1.1 Soil and Radionuclides

Soil is typically a mixture of many substances like water, air, organic remains, clay, particles of rock and inorganic materials. Organic remains are found as soil components in a decay of living matters while the inorganic constituents exist in form of minerals substances. Soil formation is based on addition, loss and transfer of materials either by natural means or human activities. Oxygen, silicon, aluminum, calcium and iron are the major uncontaminated components of soil (Manzoor et al., 2013) Soil serves as medium of transport for radionuclides through food chain which depends on their chemical properties and the uptake process by the plants and animals. The radioactive elements in the soil provide large percentage of ionizing radiation from natural sources. And natural radiation is the largest contributor of external dose (80%) to the world population which depends on the local geological characteristics and conditions of soils (UNSCEAR, 1993). The radioactivity in the soil is mainly from the naturally occurring radionuclides ^{238}U (99.2745% of uranium), ^{235}U (0.72% of uranium), ^{232}Th (100% of thorium) and their decay products, and the primordial radionuclide ^{40}K (0.0117 % of potassium) found in the earth crust (Rabesiranana et al., 2008). In the world average soil, potassium, uranium and thorium has mass fraction of 14%, $\sim 2\mu\text{g g}^{-1}$ and $9\mu\text{g g}^{-1}$ (Bowen, 1993). According to Rohit et al., (2009) natural radioactive materials can reach unsafe radiological level under certain levels.

1.2 Radioactivity

X-ray discovered by Roentgen brought about a great enthusiasm in the scientific circles. According to Bernard (1969), Becquerel reports that certain elements particularly uranium, emit radiations that are similar to x-ray in 1896 initiated scientific understanding of radioactivity. Later, Marie curie, investigating a mineral of uranium

discovered that other elements such as thorium (Th), polonium (Po) and radium (Ra) are also radioactive (Gasparini, 1984). Of all these elements, the primordial radionuclides of Uranium (U), Thorium (Th) and isotope of potassium (K) are of significant importance to the radiological status of the environment. This is because gamma radiation emitted from these primordial radionuclides and their decay progeny is one the main external source of radiation exposure to man. Radionuclides, radioactivity and radiation have been an indispensable component of the globe since its creation. And all living things existing in the world are exposed to radiation. The nucleus found at the centre of an atom consists of protons and neutrons. These protons and neutrons are held together by strong nuclear force but some nuclei still appear to be unstable. The unstable atomic nuclei undergo radioactive decay (spontaneous nuclear transformation) to form more stable nuclei thereby emitting energy in the form of particles and electromagnetic wave. The decomposition process is called radioactivity while energy and particles which are released during the process is called radiation. The sources of radioactivity are classified into two types natural and artificial.

1.3 NORM and TENORM

Radionuclides are classified based on their origin. The radioisotopes that contribute to natural radiation are called Naturally Occurring Radioactive Materials (NORMs) while technologically enhanced radionuclides are Technologically Enhanced Naturally Occurring Radioactive Materials (TENORMs).

Naturally Occurring Radioactive Materials (NORM)

A nuclide which is radioactive in its natural physical state is referred as naturally occurring radioactive material. NORMs are part of nature and it is found everywhere in the globe. NORMs exist in man's environment at different quantities/levels. And humans are continually exposed to it through food, soil, solar and water. Therefore,

everything in nature has some amount of natural radioactivity. Natural radiation are classified into two categories namely; Extra-terrestrial radiation (cosmogenic radionuclides, cosmic radiation etc) and terrestrial (primordial) radiation.

Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM)

Technologically Enhanced Naturally Occurring Radioactive Materials (TENORMs) is formed as a result of technology and human activities based on quest for economic growth and development. Such activities includes oil and gas exploration, uranium and phosphate mining and milling, coal fired power plants, tobacco, air travel, metal ore processing, manufacture of building materials and others, that could enhance and modify the concentration of NORMs, their environmental distribution and radiation exposure dose to human beings. Generally, some of the non-nuclear industrial processes causes a considerable contribution to the radio-ecological pollution such as phosphate ore mining and phosphate fertilizers manufacture and agricultural application (UNSCEAR,1988) and they also contribute technologically. TENORM contributors includes waste water treatment sludge, phosphate fertilizer and potash, phosphate industry waste, oil and gas production scale and sludge, paper and pulp industry, scrap metal release and recycling, coal ash, uranium overburden and mines spoils, geothermal energy production waste, metal mining and production waste.

1.3.1 Cosmogenic radionuclides and radiation

Cosmogenic radionuclides are natural radionuclides that are currently originated by nuclear reaction when high energy cosmic radiation passes through the earth's atmosphere. The highly energetic cosmic ray particles collide with stable elements in the atmosphere and in the ground. According to Alatisse et al 2008, the entire geosphere, the atmosphere and all parts of the earth that directly exchange materials with atmosphere contains cosmogenic radionuclides with the major production being from

the interaction of cosmic rays with atmospheric gases. These radionuclides are produced through bombardment of the upper atmosphere by high energy heavy particles. The cosmogenic radionuclides comprise ^{14}C , ^{22}Na , ^7Be and tritium. Only ^{14}C and tritium contribute to any significant natural radiation exposure to the global population as internal exposure through inhalation. The exposure from these sources is relatively low and uniform over the surface of the planet (Benneth, 1997). ^{14}C is present in carbon dioxide in the air, in the terrestrial biosphere, and in bicarbonates in the ocean. This radionuclide is produced in the atmosphere by the interaction of cosmic ray particles/energy with ^{14}N . The neutron spectrum covers a wide energy range in the lower atmosphere, from thermal to 100MeV (UNSCEAR, 1993).

Cosmic radiation

Cosmic radiation refers to both the primary energetic particles of extra-terrestrial origin and to secondary particles generated by the interaction of primary particles with the atmosphere. Cosmic rays radiation consists of 85% protons, 14% alpha particles and about 1% nuclei of atomic number between 4 and 26 (James, 1996). These particles are highly penetrating and have high energies. The annual external dose rates from cosmic rays depend slightly on latitude and strongly on altitude. Biehl et al., (1949) studied the effects of geomagnetic latitudes on the total cosmic rays and found that the ratio of latitude effects at low geomagnetic latitudes to those at higher latitudes is roughly 65:100. Annual effective dose from cosmic ray radiation around the world is estimated between the ranges of 0.26 to 2.00 mSv/year. The table 1.1 below shows natural sources of average radiation dose worldwide.

Table 1.1 Average radiation dose from natural sources (UNSCEAR 2000)

Source	Worldwide average annual effective dose (mSv)	Typical range (mSv)
External exposure		
Cosmic rays	0.4	0.3 - 1.0
Terrestrial gamma rays	0.5	0.3 – 0.6
Internal exposure		
Inhalation(mainly radon)	1.2	0.2 – 10 ^b
Ingestion (food, drinking water)	0.3	0.2 – 0.8
Total	2.4	1 – 10

1.3.2 Primordial (Terrestrial) Radionuclides

Primordial radionuclides are the radionuclides found in the earth's crust. They originated with other (stable) nuclei in the course of cosmic nucleogenesis by thermonuclear reactions in the core of a star, which then exploded as a supernova and enriched the nucleus cloud from which the sun and the solar system were formed about 4.5×10^9 years ago. Radiation from primordial sources constitutes about 85% of the natural background radiation exposure received by individuals in the environment (IAEA, 1996, Obed et al., 2005). About 70 out of 340 naturally occurring nuclides on the earth are radioactive therefore humans are continually exposed to radiation due to natural radioactivity in the terrestrial environment (Olomo, 2006). The specific activity levels of terrestrial sources of radiation are related to the composition of each lithological area and to the content of the rock from which the soils originated (Akhtar, 2004 and Tahir, 2005). These radionuclides when ingested or inhaled enter the human body and are distributed among body organs according to the metabolism of the element involved. The organs normally exhibit varying sensitivities to the radiation and thus, varying doses and risks result from their consumption or inhalation.

1.3.2.1 Radioactivity decay series

These are radionuclides that are headed by parent radionuclides that decay in sequence to other radionuclides with different half live and decay modes, and finally end to stable isotopes (NCRP,1992). There are four naturally occurring radioactive decay series (Table 1.2). These include Thorium (^{232}Th), Uranium (^{238}U), Neptunium (^{237}Np) and Actinium (^{235}U). These radionuclides do not decay to stable isotope in one step, but give rise to decay series. And not all nuclides of the series emit gamma radiations (Firestone, 1998). The members of natural radioactive series are genetically related by alpha decay (Ghoshal, 2005).

Table 1.2 Natural radioactive decay series (Tait,1980)

Name of series	Type	Stable end product	Parent radionuclide	Half-live (years)
Thorium	4n	^{208}Pb	^{232}Th	1.3×10^{10}
Neptunium	4n+1	^{209}Bi	^{237}Np	2.20×10^6
Uranium	4n+2	^{206}Pb	^{238}U	4.47×10^9
Actinium	4n+3	^{207}Pb	^{235}U	7.1×10^8

(a) *Thorium (Th) (4n) series*

Thorium-232 occurs naturally with atomic number 90 and has half life of 1.4×10^{10} years significantly longer than the age of the earth. It has a long decay series that contains important radionuclides such as radium-228 and radon-220. Radium-228, a beta emitter is a direct descendant of thorium-232, with half life of 5.75 years enters the body mainly through food. The isotope of Radon-220 that appears in the decay series of thorium has a half life time of 55.6 seconds which is too short for significant escape. Thorium-232 undergoes ten steps, six alpha and four beta decay steps with possible 346 gamma ray emissions before becoming a stable isotope, ^{208}Pb as shown in figure 1.1 and table 1.3 in page 7. Thorium is essentially insoluble. Therefore, concentration of this

radionuclide in biological material is almost negligible. This radionuclide also is not mobile in the environment. The highest concentrations of thorium in the body have found predominantly in the pulmonary lymph nodes and lungs. The presence of high concentrations in this area of the body indicates that infiltration occurs mainly as a result of inhalation of soil and dust particles (NCRP, 1992).

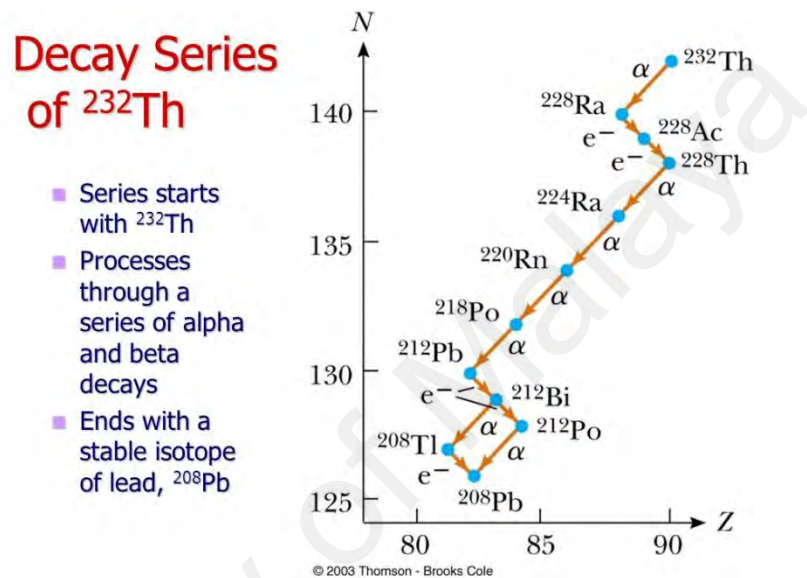


Figure 1.1 Thorium 232 decay series. (Nancy Walton (Modified 2016).

Table 1.3 Thorium decay series with half-life

Nuclide	Mode of decay	Half-life
^{232}Th	α	1.4×10^{10}
^{228}Ra	β	5.8 years
^{228}Ac	β	6.1 hours
^{228}Th	α	1.9 years
^{224}Ra	α	3.7 days
^{220}Rn	α	55.6 seconds
^{216}Po	α	0.15 seconds
^{212}Pb	β	10.6 hours
^{212}Bi	36% α and 64% β	60.5 minutes
^{212}Po	α	3.0×10^{-7} seconds
^{208}Tl	β	3.1 minutes
^{208}Pb	stable	stable

(b) *Uranium (U) (4n+2) series*

Uranium has atomic number of 92 and three naturally occurring isotopes namely; ^{234}U , ^{235}U and ^{238}U as show in table 1.4 in page 8 which already undergo radioactive decay by emission of alpha particle accompanied by weak gamma radiation. The uranium isotopes are all radioactive and their decays produce a number of secondary radioactive elements that continue to decay until they reach stable nuclei. Of the three isotopes of uranium, ^{235}U and ^{238}U are the most important radionuclides of primary origin in the earth's crust. Uranium-238 has a half life of 4.47×10^9 . ^{238}U (Uranium-radium) series takes about 14-16 steps to reach ^{208}Pb , with possible 458 gamma rays as shown in figure 1.2 and table 1.5 in page 9. The decay products of ^{238}U include important radionuclides such as radium-226, and radon-222. The release of radiation during the decay process raises health concerns. Humans could be exposed to uranium through food and water ingestion or inhaling of contaminated air.

Table 1.4 Natural abundance of uranium isotopes

Isotopes	^{238}U	^{235}U	^{234}U
Natural Abundance (%)	99.27	0.72	0.0055
Half-life (years)	4.47×10^9	7×10^8	2.46×10^5

(c) *Actinium (4n+3) series*

Actinium series is a daughter of Uranium-235. The decay of ^{235}U (Uranium to Actinium) series goes through 11- 14 radionuclides to ^{207}Pb as shown in figure 1.3 and table 1.6 below.

Table 1.6 Uranium-235 (Actinium) decay series with half-life.

Isotope	Symbol	Proton number	Nucleon number	Mode of decay	Half life
Uranium	U	92	235	α	704 yrs
Thorium	Th	90	231	β	25.5 hrs
Protactinium	Pa	91	231	α	32,760 yrs
Actinium	Ac	89	227	α, β	21.7 yrs
Francium	Fr	87	223	α, β	21.8 mins
Thorium	Th	90	227	α	18.7 days
Radium	Ra	88	223	α	11.4 days
Radon	Rn	86	219	α	4 secs
Astatine	At	85	219	α, β	56 secs
Bismuth	Bi	83	215	β	7.7 mins
Polonium	Po	84	215	β	1.8 mins
Astatine	At	85	215	α	0.1 min
Lead	Pb	82	211	β	36.1 mins
Bismuth	Bi	83	211	α, β	2.1 mins
Polonium	Po	84	211	α	0.5 secs
Thallium	Tl	81	207	β	4.7 mins
Lead	Pb	82	207	Stable	Stable

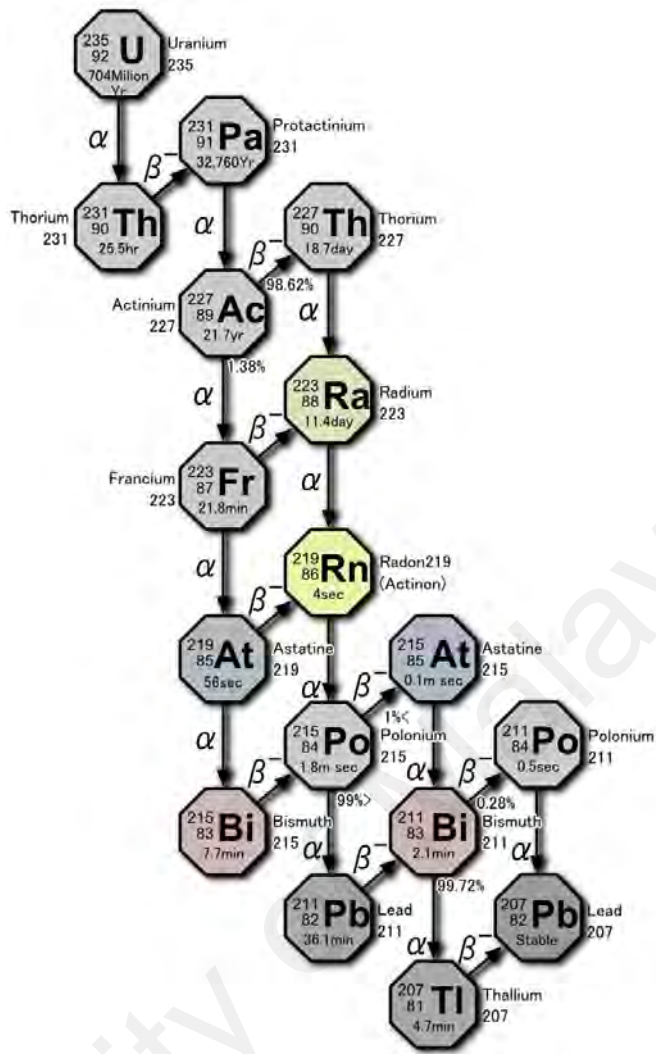


Figure 1.3 Actinium decay series. (Chloe Reynolds (n.d)).

(d) *Neptunium* (^{237}Np) ($4n+1$) series

Neptunium has atomic number of 93. It is a radioactive series that was brought to light after the synthesis of trans uranium element. It takes 12 steps from ^{237}Np to bismuth 209 which is a stable nuclei as shown in figure 1.4 and table 1.7 in page 12. The series contains some important radionuclides like uranium, thorium, actinium, radium, radon, etc.

Table 1.7 Neptunium decay series with half-life

Isotope	Symbol	Proton number	Nucleon number	Mode of decay	Half life
Neptunium	Np	237	93	α	2.14×10^6 years
Protactinium	Pa	233	92	β	27 days
Uranium	U	233	91	α	159200 years
Thorium	Th	225	90	α	7304 years
Radium	Ra	225	88	β	15 days
Actinium	Ac	225	89	α	10 days
Francium	Fr	221	87	α	5 minutes
Astatine	At	217	85	α	32 seconds
Bismuth	Bi	213	83	α, β	46 minutes
Polonium	Po	213	84	α	4.2×10^{-6} minutes
Thallium	Tl	81	209	β	2.2 minutes
Lead	Pb	82	209	β	3.25 minutes
Bismuth	Bi	83	209	stable	1.9×10^9 years

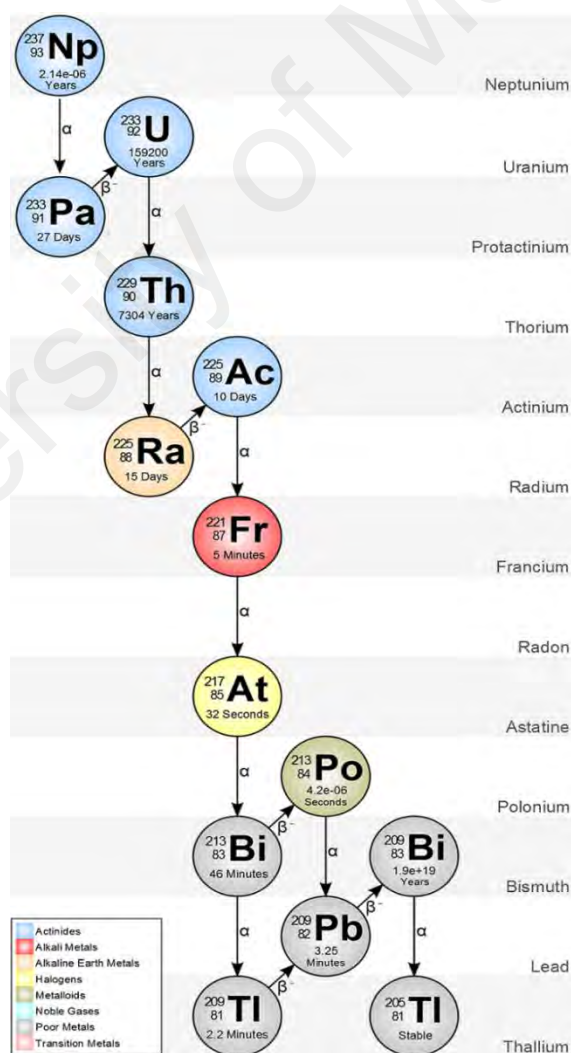


Figure 1.4 Neptunium decay series. (Chloe Reynolds (n.d))

1.3.2.2 Potassium-40 (^{40}K)

Potassium-40 has been found to be the most significant primordial radionuclide of terrestrial origin. It is so important in terms of dose associated with naturally occurring radionuclides. Potassium is found in large amount throughout nature. It is the seventh most abundant element in the crust of the earth and the sixth most abundant element in solutions in the oceans (Argonne National Laboratory, EVS 2005). It has a half-life of 1.3×10^9 years. It is a both beta and gamma emitter. The main decay modes of ^{40}K are β -decay to stable ^{40}Ca and electron capture to an excited state of ^{40}Ar , emitting 89% of 1.314 MeV of β - particles most of the time (Kathren, 1998). ^{40}Ar decays to its ground state by the emission of a gamma ray of 1.461 MeV which happens in 10.67% of all decays. This photon value makes it easy to identify and quantify potassium-40 by gamma ray spectroscopy. It is an excellent calibration point because of the presence of potassium in essentially all environmental sample (Alatise et al, 2008).

1.4 Man-made (Artificial) Sources of Radiation

Artificial radioactivity is a radioactivity that is generated through human activities which varies with time and location. The technological advancement and research in environment produces more radiation sources other than natural sources. The sources of man-made radiation includes; nuclear power plants (for energy production and nuclear weapons for warfare) and reprocessing facilities, radiation generating devices such as x-rays machine for medical diagnosis and therapy, nuclear accelerators for the study of nuclear transmutations and sources used for industrial and agricultural applications (UNSCEAR, 1988). The environment gets contaminated through transport, routine release, accidents, loss and disposal or misuse of radioactive materials. A typical example is the major nuclear accident at Chernobyl-4 nuclear power installation in 1986 when huge amounts of various radionuclides escaped into the atmosphere (Olomo, 2006). Radiation used in medicine for both diagnostic and therapeutic purposes

especially the management of cancer in humans make a significant contribution to man's exposure (Paschal, 2006).

Man-made sources of radiation can only affect a small size of the population at any time under controlled management. Some common consumer products enhanced man's exposure, like the luminous watches and clock which contains ^3H , ^{147}Pm or ^{226}Ra as activating agent (UNEP, 1991; NCRP, 1977). Television sets produce x-rays, but modern television sets have designed to produce negligible amounts when used correctly and serviced appropriately (Larmash, 1983). Also, smoke detectors contain alpha emitting sources such as Americium-241. Some porcelain dentures and eyes glasses which contain uranium and thorium (NCRP, 1977), also enhanced exposure. Starters for fluorescent tube lights and electrical appliances contain sealed radionuclides although they do not cause any hazard unless they are broken (NCRP, 1977), X-rays machines used for screening travellers (Mettler and Sinclair, 1990), cigarette smoke and tobacco which contain Pb-210 and Po-210 (Larmash, 1983; NCRP, 1977, Paschal, 2006) and combustible fuels as well as building materials which could be mixed with uranium, thorium and potassium containing waste etc (NCRP, 1977). All these radiation sources could contaminate the human body through irradiation, inhalation and ingestion leading to varying doses of radiation to man (Pascal, 2006; NCRP, 1977).

1.5 Radionuclides transfer in the environment

The level of radiation dose to man can be increased due to the radionuclides in the environment. Man is either exposed to radiation by external or internal means. External exposure is a direct exposure of man from the environment while internal exposure is an exposure taken up by man through various mean such as inhalation of contaminated dust, ingestion of dirt and dust, inhalation of radon diffusing from material and skin contamination (see figure 1.5). Radioactive materials can be released into air or directly

into water or soil. When they are released in air, they can travel some distance, depending on some factors like wind speed and direction and altitude of the release. The products of airborne releases can be transported to humans by several ways. Man can directly inhale them or the materials will finally deposit themselves on the ground, where they will find their ways into plant and animal life, as a result of that into the food chain. Deposition of airborne contaminants into water can reach humans either by direct ingestion or through the food chain. Likewise, depositions on the soil and water find their way into the food chain through plant and animal life. Rain water runoff can carry soil into oceans, rivers, lakes and streams, thereby transporting any soil (sediment) contamination to the water. Additionally, radioactive materials can leach into porous soils and into ground water (Doendara, 2007). Apart from all these ways, radionuclides in the aquatic environment could cause external exposure via the use of riverbed sand (sediments) as building materials. It is a well-known knowledge that sediments from rivers, lakes and beaches are used as materials for construction of buildings (Xinwei and Xiaolan, 2006).

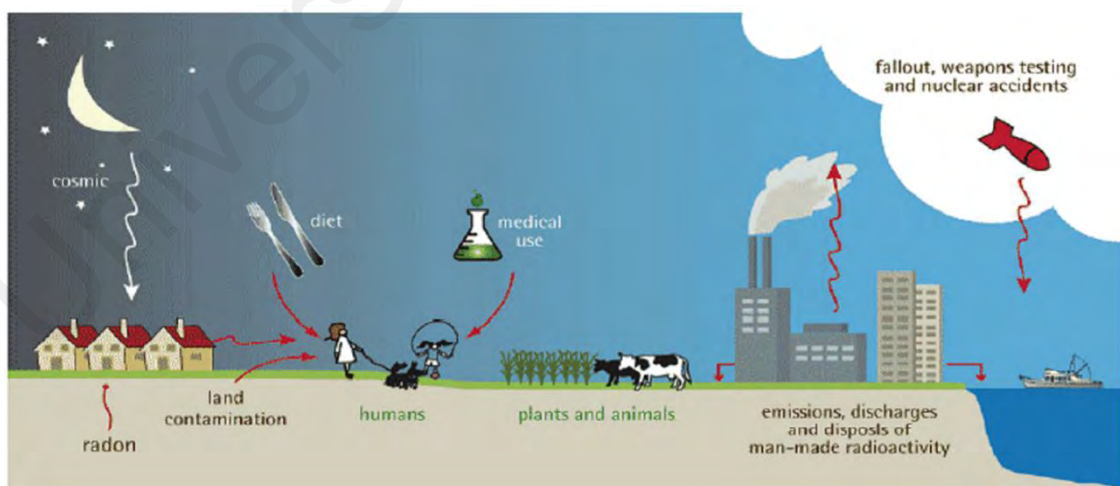


Figure 1.5 Routes of human exposure to sources of natural and man made (www.google.com/search?q=Routes+of+human+exposure+to+sources+of+natural+and+man+made).

1.6 Biological effect of ionizing radiation

Ionizing radiation is a form of energy which transfers enough energy above the threshold energy to cause ionization. The subsequent health effects when radiation interacts with any biological entity are due to physical and chemical changes that came as a result of ionization. The basic building blocks of human body are the cells that form tissues and organs. Living tissues are very sensitive to change and the ionization can alter to damage them. A unit cell in human body consists of nucleus, which is surrounded by about 70% colourless fluid called cytoplasm that contains variety of compounds such as salts, fats, carbohydrates, amino acids and proteins. A cell is injured when exposed to radiation which eventually interacts with other non-irradiated cells and thereby causing damage to the entire biological entity. The injury incurred as a result of exposure to ionizing radiation may lead to molecular changes and formation of chemical species or radicals (H^+ and OH^-) which have deleterious effects on the chromosomes materials of the cells. The H^+ and OH^- attack Deoxyribonucleic acids (DNA) causing the breakage of the molecules and the rupturing of the molecular bonds. The breakage of the DNA molecules ends up to the sudden random change in genetic code, and as a consequence causes genetic mutation. Such mutated cells may be repaired in a process called DNA degeneracy. Mutated cells that are not properly repaired may die through apoptosis or survive as viable but transferred from a parent to an offspring (Muller 1927). The effects of radiation exposure of human (multi-cellular organism) cells are complicated. The water content in the cell experiences ionization and excitation within 10^{-16} seconds when radiation transfers energy to a biological medium. The resulting ions interact with other water molecules and cause a number of new products like H^+ and OH^- and strong oxidizing agent H_2O_2 (hydrogen peroxide). The nature and extent of damage caused by ionizing radiation depends on the amount of exposure, the frequency of exposure, types of radiation, radio-sensitivity of the cell and

the penetrating power of radiation to which an individual is exposed. Low doses of radiation over a long time can cause various types of cancer, such as thyroid, breast, lung cancer and leukemia. Shahid in 2012 reported the biological effect of ionizing radiation as shown in figure 1.6

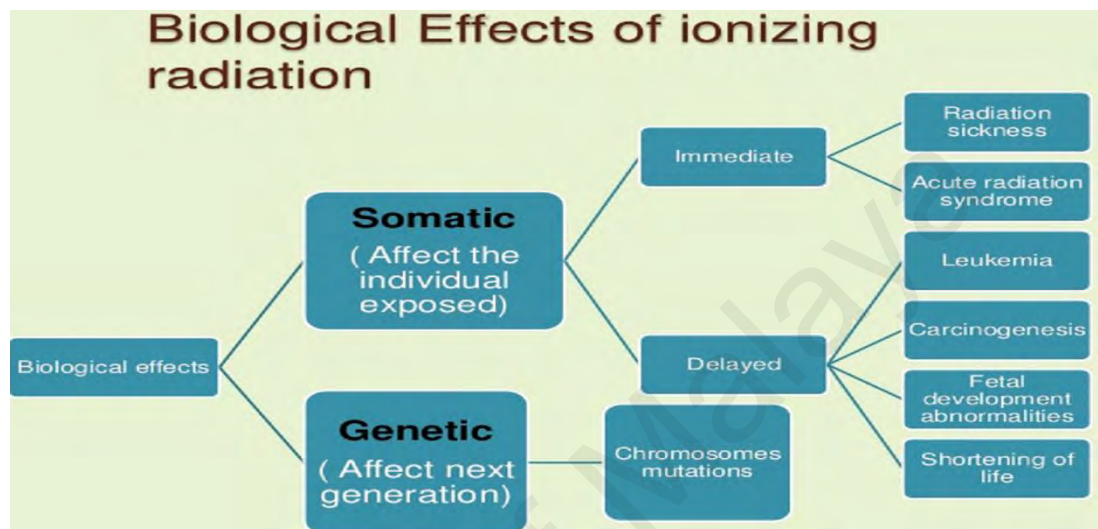


Figure 1.6 Biological effects of ionizing radiation

1.7 Classification of radiation effect on biological system

Radiation effect on biological systems can be classified using different criteria. It is classified recently into two categories (stochastic and deterministic effects) based on presence and absence of a threshold radiation dose to produce the effect. When radiation effects occur with a threshold level of dose it is said to be deterministic effects, while the radiation effects occur without a threshold level is known as stochastic.

1.7.1 Stochastic effects

Stochastic effects include malignant and hereditary diseases for which the probability of an effect occurring rather than its severity is considered to be directly proportional to the effective dose level. In stochastic effects, any radiation dose no matter how small is capable of initiating an effect. Genetic mutations are examples that

can result from stochastic effects. A gene mutation occurs when Deoxyribonucleic Acid (DNA) is altered. In 1927, Muller discovered the mutagenic properties of ionizing radiation and reported that radiation can cause alteration to the genetic information contain in a germ cell. Genetic mutation caused by radiation exposure can be transferred from a parent to an offspring. If the mutant gamete is successfully fertilized and the zygote (fertilized ovum) developed into a life offspring, then the mutation is carried into the progeny. Radiogenic cancer is a stochastic effect of ionizing radiation and the risk of incurring cancer from radiation exposure depends on factors like; the dose administered over time, the age, sex and genetic background of the exposed person. In recent times, cancer has assumed greater importance in the health agenda throughout the world and it has been observed that exposure to ionizing radiation increases the risk of incurring cancers (Farai et al., 2000, Brenner et al., 2003).

1.7.2 Deterministic effects

Deterministic effects of radiation is predictable and its severity is an inevitable consequences of exceeding a given threshold radiation dose. In other words, the severity of the deterministic effect is a function of radiation dose. Examples of deterministic effects are non-malignant skin damage (erythema), and hematological effects (changes in the composition of the blood). Somatic effect is deterministic and may be observed when an individual is irradiated. The damage due to somatic effect is only limited to the exposed individual, and in essence the individual suffers and die with the damage. Somatic effects may take a longer time to develop and become evident after ionizing radiation had been administered either acutely or over an extended period. This is referred to as delayed somatic effect. Another delayed somatic effect is cataract which affects the opacity of the lens of the eye. A radiogenic cataract is a deterministic effect because there is practical threshold of ionizing radiation dose below which cataract is

not produced or manifested; and its severity, when it occurs, is related to the magnitude of the radiation dose and the time over which it is administered (Nobuyuki et al 2014).

1.8 Justification/research significant.

All the habitat of the globe has presence of radioactive materials (radionuclides). Therefore, the earth is a source of natural radiation. Natural radiation is mostly due to the activity level of primordial radioactive materials such as Uranium-238, Thorium-232 and their daughters, in addition potassium-40 that is naturally present in the earth crust. Man has always been exposed to natural radiation emerging from within and outside the earth surface. It is a common knowledge that the irradiation due to ionizing radiations from natural sources happens because of the primordial radioactive elements in the soil and rocks, cosmic rays gaining access into the earth's atmosphere from outer space and the internal exposure from radioactive elements through food, water and air. Natural radioactivity is widespread in the earth's environment and it exists in various geological formations in soil, plants, rocks, water and air (Surinder et al., (2005). Human activities may cause increment of natural radioactivity level in an environment. In certain situations, natural radioactive materials can attain dangerous radiological levels. It is therefore necessary to measure the natural radioactivity in the residential hostels environment to control the occupants' health from radiation exposure. And have a baseline for future changes in the environmental radioactivity due to human activity.

1.9 Aims and objectives

The aim of this research is to evaluate natural radioactivity and hazard indices in the soil collected from the residential college areas of University of Malaya, Malaysia. The objectives of the study are as follows:

1. To evaluate the activity concentrations of natural radionuclides ^{232}U , ^{228}Ra and ^{40}K in the surface soil samples collected from different areas of UM using High-purity Germanium (HPGe) detectors system.
2. To calculate radiation levels and associated gamma-absorbed dose rates by members of the public
3. To determine the areas in terms of radiological implication associated with studied radionuclides.
4. To evaluate the radiological threats on members of the public utilizing the environment.
5. To develop research dexterity on NORMs investigation and measurement.

CHAPTER 2: LITERATURE REVIEW

2.1 Atoms and radiation

All matter is made up of atoms and their effective diameters are about $3 \times 10^{-10} \text{m}$ (Abraham 1986). Nearly all the mass of the atom is concentrated in the nucleus which is centrally placed within the atom. The nucleus of an atom contains protons which carry a positive electric charge and neutrons which carry no charge at all. They are held together by a strong nuclear force. Each atom contains equal number of protons and electrons and is therefore electrically neutral. The number of electrons in the atom and hence the number of protons in the nucleus called the atomic number, Z gives an element its unique properties. Since the protons and neutrons have almost the same mass and are much heavier than electrons, most of all atom's mass is concentrated in the nucleus, thus the total number of protons added to neutrons is referred to as the mass number, A of the particular atom. Thus, the mass number minus the proton number gives the neutron number, N ; that is $N = A - Z$. Although many nuclides are stable, most are not, the stability of nuclei is determined mainly by the neutron to proton ratio ($\frac{N}{Z}$) of a nuclide. For light nuclei, neutron to proton ratio, $\frac{N}{Z} = 1$ while for heavy nuclei, neutron to proton ratio, $\frac{N}{Z} = 1.5$. This is the ideal situation for stability of a nucleus. Radioactivity is a natural and spontaneous process that occurs when unstable nuclei of an element emit or radiate excess energy in the form of particles or waves. The particles are called ionizing radiation because they have the ability to ionize or change the physical and chemical structure of an atom they pass through. In some cases, one or more isotopes of an element are radioactive, are referred as radionuclides. Radionuclides are unstable elements which undergo radioactive decay, by emitting radiation in the form of alpha or beta particles and gamma rays. They undergo

spontaneous nuclear transformation which results in the formation of new elements. These spontaneous transformations of a nucleus is called radioactivity and the excess energy emitted is a form of ionizing radiation. The capacity of radioactive emissions to cause ionization of molecules is the basis for health hazards and provides the means by which radiation can be detected and evaluated. A radionuclide can be identified by the characteristics of the radiation it emits. These characteristics include the rate of decay or half-life of the radionuclide and the type of energy of radiation emitted.

2.2 Radioactive half life

The rate at which particles are disintegrated is expressed by the half-life of the radionuclide. The half-life, $T_{1/2}$ is the period of time it takes for an unstable atom to decay to half its initial value. The half-life is related to the decay constant by λ , $T_{1/2} = \frac{0.693}{\lambda}$. As a radionuclide decays, it becomes an isotope of an element, which can be deduced as follows:

If a number of nuclei, N is given at a time, t is proportional to the number of radioactive atoms, dN in time, dt , then:

$$\frac{dN}{dt} \propto N \quad (2.1)$$

Introducing the disintegration (decay) constant λ which has a characteristics value for every radionuclide and minus sign because of the fact that number of radioactive nuclei decreases with time, equation (2.1) can be rewritten as:

$$\frac{dN}{dt} = - \lambda N \quad (2.2)$$

To find N as a function of time t , we arrange equation 2.2 as:

$$\frac{dN}{N} = - \lambda dt \quad (2.3)$$

Integrating both sides of the above equation,

$$\int_{N_0}^N \frac{dN}{N} = - \int_0^t \lambda dt \quad (2.4)$$

Which gives;

$$\ln\left(\frac{N}{N_0}\right) = - \lambda(t-t_0) \quad (2.5)$$

where N_0 is the number of radionuclides in a sample at some arbitrary initial time t_0 .

Since $t_0 = 0$, the equation above become

$$\ln\left(\frac{N}{N_0}\right) = - \lambda t \quad (2.6)$$

The above equation can be written as exponential form as:

$$N = N_0 e^{-\lambda t} \quad (2.7)$$

N is the quantity of radionuclides that remain after and has not yet decay after a time t .

Equation (2.6) is the radioactive decay law.

$$\text{Since } A_t = \lambda N_t \quad (2.8)$$

$$A = A_0 e^{-\lambda t} \quad (2.9)$$

where the activity A , of a radionuclide is the total number of decay per second; A_0 is the activity at a time $t = 0$.

2.3 Activity

One of the most important quantities associated with a sample of radioactive material is its activity. Activity is the rate at which the nuclei within the sample undergo disintegration and can be expressed in terms of the numbers of disintegration per

seconds (dps). The Becquerel (Bq) is the S.I unit and is equivalent to 1dps. The probability that a nucleus will decay in a certain time interval does not depend on the age of the nucleus, the state of the chemical combination, temperature, pressure or the presence of other atoms or nuclei but it is a property of the individual isolated nucleus. The rate of radioactive decay from a sample of any radioactive substance must be proportional only to the number of nuclei present, if radioactivity is a property of isolated nuclei.

2.4 Radioactive equilibrium

Radioactive equilibrium or disequilibrium is an important consideration in all gamma ray spectrometric measurement. Gamma ray spectroscopy can be used to determine the concentrations of uranium, thorium and potassium in the rock or soil samples because gamma rays of specific energies are associated with each radioelement. By looking at the peak in the energy spectrum of gamma rays being emitted by the source, the radioelement content of the source can be inferred. The method involves the counting of gamma ray photons with specified energies. The gamma ray count rate can then be related to the amount of parent by assuming there is a direct relation between the amount of daughter and parent. The assumption is valid when the radioactive decay series is in the state of secular equilibrium.

Secular equilibrium is established in a radioactive series when the number of atoms of each daughter being produced in a series is equal to the number of atoms the daughter's parent lost by radioactive decay. The rate of loss by decay is proportional to the amount of radioelement present.

$$\frac{dN_1}{dt} = \lambda_1 N_1 \quad (2.10)$$

where N_1 is the amount of element 1 (parent) and λ_1 is the decay constant for element 1 (parent)

The rate of formation of daughter element is also given as

$$\frac{dN_1}{dt} = \lambda_2 N_2 \quad (2.11)$$

where N_2 = amount of element 2 (daughter) and λ_2 = decay constant for element 2 (daughter)

In a radioactive series N_1 is decaying into N_2 at the above rate while at the same time N_2 will decay with decay constant λ_2 into N_3 and so on. For a parent with a relatively long half-life, after a long period of time, the amount of any given daughter becomes constant. The rate of production from its parent becomes equal with its rate of decay. The series is then said to be in the state of secular equilibrium and it requires that;

$$\lambda_1 N_1 = \lambda_2 N_2 = \lambda_3 N_3 \dots \dots \dots \lambda_n N_n \quad (2.12)$$

When this condition is obtained, it is possible to determine the amount of the parent of the decay series by measuring the radioactivity from any of the daughter elements. i.e activity of the parent is equal to the activity of the daughter. It is important to know the length of time required for a secular equilibrium to be established in a case of soil and rock samples, which are crushed and sealed for laboratory analysis using gamma ray spectrometry.

2.5 Types of radiation

The instability of the atomic nucleus in certain atoms makes their nuclei to experience as spontaneous disintegration by emission of energy and particles. The emission of various types of radiation is an atomic property rather than a molecular or

bulk phenomenon, because the rate of emission does not depend on the state of chemical composition of the radioactive element concerned. Also, it was found that radioactive substances emits three (3) kinds of radiations; alpha particles (helium nuclei), beta particles (electrons and positrons) and gamma radiations. These rays could be distinguished from one another basically in two ways; the difference in the case with which the rays could pass through matter and by the direction in which their path was bent by the application of magnetic field (Parasnis, 1986).

2.5.1 Alpha (α) particles.

These α are helium (He) nuclei of two protons and two neutrons and a charge of +2, which can be stopped in its path by a thin sheet of paper. Alpha particles exist as a tightly bound entity within the nuclei and it is heavier than helium, which may be ejected during disintegration and expressed as



where P, α , X and Y are the number of protons, alpha particle, initial element and final element respectively.

The above represents transition of element X to Y by alpha emission. The velocity of α particle is rather small and the mechanism for the energy loss by α particles passing through matter is by elastic-inelastic collision with atomic electron. The most significant feature of α particle is its discrete energy and such energy is used to identify the specific nuclide that emits it.

2.5.2 Beta (β) particle

These are electrons with a charge of -1. They are ejected when a neutron splits into a positron (β^+) and an electron (β^-) as represented by the equation below;



where P, N X, Y and e^- are number of proton, number of neutron, initial element, final element and ejected electron respectively.

The proton stays in the nucleus and the electron is ejected in certain radioactive disintegration. The result of transmutation is that there will be a gain in charge +1 with no change in mass. The penetrating power of β particle is about 100 times that of α particle but can be stopped by few millimeters of aluminum foil.

2.5.3 Gamma (γ) radiation

These are not discrete particles but pure electromagnetic radiation of the same kind like x-rays, light and radio waves but of shorter wavelength. The gamma-ray photons are of high energy. They have much more penetrating power compared with beta-particles. Sizeable thick lead (Pb) is required to stop gamma rays. Their emission does not affect the charge and mass of the nucleus but only a decrease in its energy content. In most cases, the emission of gamma rays is a secondary consequence of either an alpha decay or beta decay.

2.5.4 Differences between Alpha, beta and gamma

Alpha and beta particles lose their energy in passing through matter collision, ionization etc. they are brought to a virtual stop within certain distance, which is called their range. The range of alpha particle is only a few centimeters or about 30 μ m in denser particles, while a thin sheet of lead or a few centimeters of sand will stop beta particles. The intensity of gamma rays traversing matter decreases exponentially with distance, so that we cannot speak of a definite range in the case.

2.5.5 Characteristics of alpha beta and gamma rays

Table 2.1 Characteristics of alpha beta and gamma rays

Characteristic	Alpha	Beta	Gamma
Emission of	2 P+2N	1 electron – High kinetic energy	High frequency photon (e-m rad.)
Changes from	Uranium - Plutonium	Radium - Polonium	unchange
Charge (C)	2	-1	0
Mass (kg)	4	5.41×10^{-4}	0
Speed (km/s)	15000	3×10^9	300,000
% of speed of light	5%	Approx. 100%	100
Kinetic energy (MeV)	5	5.0×10^{-3} MeV – 1 MeV	0.1 MeV - < 10MeV
Penetration power	Low- has large mass and can be easily stopped by a thin sheet of paper	Moderate- average mass and charge can be stop by metal with little mm thickness.	Very high- No mass, no charge and can be stopped only by a large thick cement, steel or lead.
Ionization power	Very-high (large charge)	Average (Low charge)	Low (no charge)

2.6 Spontaneous fission

Some heavy radio-nuclei can decompose through spontaneous fission. It is like neutron-induced fission which occurs in reactors, but the difference is that at the starting point of this fission process, neutron is not important. Heavy nucleus undergo through this kind of radioactive decay by splitting into two lighter nuclei releasing many neutrons. For example;



The determination of spontaneous fission products is not consistent like alpha and beta decay, but the distribution is in statistical form.

2.7 Intensity

Radiation intensity is the amount of energy that goes through a known area which is at right angle to the direction of radiation travel at a given time unit. The strength of radioactive sources is not easy to be measured using its activity. The detector makes it easier and convenient to measure the intensity of gamma ray or X-ray sources. The intensity of a source is measured by sampling the number of photons emitted from the source at a given time, which is directly related to the number of disintegrations at the same given time (activity).

2.8 Interaction of gamma rays with matter

The earth contains many radionuclides that decay and emit charged particles such as alpha, beta radiations and neutral particle like gamma radiations. To calculate the radioactivity in the soil, we need to focus on detecting the emissions of charged and neutral particles from the decaying radionuclides. The emitted particles are easily observed when they interact with the matter through the excitation and ionization effects they cause in the matter. The methods in which these effects are used for radiation detection depend on the mode of interaction, type and energy of radiation. Therefore, a good knowledge of the response of a specific type of detector must be based on the familiarity of fundamental mechanism by which radiation interacts and lose their energies in matter since the main objective in radiation spectrometry is to measure the energy distribution of the incident radiation. Gamma ray photon is uncharged and creates no direct ionization or excitation of the material they pass through. Therefore, the detection of gamma ray hang on causing the gamma-ray photon to undergo an interaction that transfer all or part of the photon energy ($h\nu$) to electron in the absorbing material.

Absorption of gamma rays in matter occurs by mechanisms that are completely different from the absorption of charged particles. There are many modes of interaction but only three mechanisms (Photoelectric effect, Compton scattering and pair production) are important in radiation measurement.

2.8.1 Photoelectric effect

The experience of ejecting electrons from an atom after the absorption of light photon is called photoelectric effect. Here, the incident gamma rays interact (collide) with atomic electron in the absorber matter and transfer all of its energy to the electron. The atom gets ionized if the absorbed energy is enough to release the electron from its atomic shell. The kinetic energy of the emerging electron equals to the total energy of the photon minus its binding energy.

$$K.E_e = E_\gamma - E_B \quad (2.16)$$

where $K.E_e$, E_γ , and E_B are kinetic energy of the photoelectron, energy of incident photon and binding energy of the electron respectively.

Photoelectric emission occurs when energy of incident photon (E_γ) is greater or equal to the binding energy of the electron (E_B) in the material. The tightly bound shells like K- shell always have vacancy due to the photoelectrons production. The immediate filling of the vacancy by electrons from the higher shells leads to the production of X-ray characteristics. As stated by Tait (1980) and James (1995), Photoelectric effect has a cross section written as;

$$\sigma_{p.e} = \delta E_\gamma^{7/2} \rho Z^5 \quad (2.17)$$

where, σ , ρ and Z are the constant, density of the absorbing material and the atomic number of the absorbing material respectively.

At lower frequencies, light is incapable of ejecting electrons, thus the critical frequency for light to eject an electron is called the threshold frequency. It is evident from equation (2.16) above that photoelectric effect is more for materials at low photon energies (<0.1 MeV) and photons are absorbed more strongly in high atomic number (Z) materials. i.e, the absorption due to the photoelectric effect decreases sharply with increasing gamma ray energy but increases rapidly with increasing absorbing material's atomic number. Example, lead (Pb) used in gamma ray spectroscopy shields the detector from gamma rays.

2.8.2 Compton effect

The interaction takes place between the incident gamma ray photon with energy (0.1-10 MeV) and an electron in the absorbing materials. Here, the photon undergoes elastic scattering with a free or loosely bound electron in the outmost atomic shell. After transferring some portions of its energy to the electron, the energy of the photon reduces from $h\nu$ to $h\nu'$ with also a change in frequency and wavelength (λ to λ'). Some part of the original photon remains as lower energy which may undergo a further photoelectric or Compton interaction but in a different direction from the parent photon. The energy of the electron scattered at different angle is written as $h\nu-h\nu'$. The energy $h\nu-h\nu'$ that is deposited in the material in the interaction can be calculated by applying the law of conservation of energy and momentum. The change in wavelength of the photon according to Briks (1964) and Tait (1980) is

$$\Delta \lambda = \lambda' - \lambda = \frac{h}{m_0c} [1 - \cos\theta] \quad (2.18)$$

where $\Delta \lambda$, λ' , λ , m_0 , h and θ are the change in wavelength, wavelength of the scattered photon, wavelength of the incident photo, the initial mass of the electron, the plank's constant and the angel of scatter of photon respectively.

The energy of the scattered gamma ray, E'_γ in terms of the scattering angle Θ is given by;

$$E'_\gamma = \frac{E_\gamma}{1 + \frac{E_\gamma}{m_0 c^2} [1 - \cos\theta]} \quad (2.19)$$

Kinetic energy of the electron as $K.E_e$

$$K.E_e = E_\gamma - E'_\gamma \quad (2.20)$$

Thus,

$$K.E_e = \frac{\frac{E_\gamma^2}{m_0 c^2} [1 - \cos\theta]}{1 + \frac{E_\gamma}{m_0 c^2} [1 - \cos\theta]} \quad (2.21)$$

where m_0 is the initial mass and $m_0 c^2$ is the initial energy of the electron. Taking from equation 2.6, the minimum value of $K.E_e$ is zero when $\theta = 0$. And the maximum value called Compton edge, E_c corresponds to a head-on collision in which the photon is scattered backwards, is when $\theta = 180^\circ$

The maximum energy value is obtained as;

$$E_c = E_\gamma \left[\frac{2E_\gamma}{m_0 c^2 + 2E_\gamma} \right] \quad (2.22)$$

Note, the probability of Compton scattering per atom of the absorber depends on the number of electrons available as scattering agent, and therefore increases linearly with atomic number (Z) and decreases with increasing energy. Thus, Compton scattering makes only a small contribution to absorption of high energy photons.

2.8.3 Pair production

In pair production, a photon of sufficient energy materializes into an electron and a positron. The energy equation of the process is written as;

$$h\nu = E_e + E_p + 2m_0c^2 \quad (2.23)$$

Energy greater than 1.022MeV may be absorbed entirely and be replaced by an electron-positron pair. Pair production is predominately confined to high energy gamma rays. The process occurs in the field of a nucleus of the absorbing material. Because the positron will be subsequently annihilated after slowing down in the absorbing medium, two annihilation photons are normally produced as a secondary product of the interaction.

Unlike the other two photon interactions, pair production has a cross section σ_{pp} which increases, although slowly with photon E_γ and the interaction tends to be dominant at high energies.

The cross section is given by:

$$\sigma_{pp} = cZ^2\rho \ln E_\gamma \quad (2.24)$$

$c = \text{constant.}$

The net effect of the above three interactions is an exponential attenuation in the intensity of a beam of gamma rays passing through a thickness t of an absorbing material. This is described by the equation

$$I = I_0e^{-\sigma t} \quad (2.25)$$

where I_0 is the initial intensity at $t = 0$ and σ is the linear absorption co-efficient due to all the effects.

2.9 Review of existing knowledge

The existence of natural radioactivity in the soil and rock has long been recognized. It has been found that uranium-238, thorium 232 and potassium are present in the earth crust in parts per million (ppm) levels. There has been rapid increase in human population. Human activities can increase the natural radioactivity of the surface soil through waste dumping that contain radioactive material and opening of the earth crust thereby exposing substances naturally buried in the soil. According to (UNSCEAR, 2000), mining of all kinds affect the environment adversely, this made mining one of the major sources that enhance the natural radioactivity level of the environment. Gamma radiation emitted from primordial radionuclides and their progeny is one of the main external sources of radiation exposure to the human. Terrestrial radioactivity, and associated external exposure due to gamma radiation depend primarily on the geological formation and soil type of the location; and these factors greatly influence the dose distribution from natural radiation.

Since natural radiation is the largest contributor of external dose to the world population, measurement of gamma radiation dose from natural sources is of particular importance. The concentration of ^{232}Th , ^{226}Ra and ^{40}K vary widely depending on the location. In addition, soil acts as a source of transfers of radionuclides through the food chain depending on their chemical properties and the uptake process by the roots to plants and animals; hence it is the basic indicator of radiological status of the environment. These radionuclides take part in several biogeochemical processes that determine their mobility and availability for biological uptake. The major potential hazard from natural radiation is from external exposure either by direct exposure to the soil or as they enter in many building materials. [UNSCEAR, 1993; Beir VII, 2006; UNSCEAR, 2000; Klein, C.K and Hurlbut, C.S., 1985; Jabbar, A., et al., 2010; Mandic, L.J., et al., 2010]

Nowadays, studies on health effects due to ionizing radiations have produced substantial evidences that exposures to high level of radiation can cause illness or even death. Despite a well known effect of cancer, scientists have long known that ionizing radiations with high doses may cause mental retardations in children's mother exposed to radiations during pregnancy period. Radium and its ultimate parent uranium in the ground lead to the production of radon a radioactive gas. The inhalation of radon progenies at a high level may cause an increase of lung cancer (UNSCEAR, 1993).

In an earlier work done by Ravisankar et al 2012, on activity concentration of natural radionuclides that are in different locations of Yelagiri hills shows that thorium-232 is 1.19 times higher than world median value. And the spatial distribution and lifetime cancer risk due to gamma radioactivity in the same Yelagiri hills in India by Chandrasekaran A., et al 2014 reported Th/U concentration to be 5.074 and U/Th ratio 0.43 which are higher than the global ratio of 3.5 and 0.26 respectively. In their further work on natural gamma radiation in beach sediments of north east coast of Tamilandu, India 2014, they reported that average concentration of natural radionuclides, all calculated radiological parameters and hazard indices except external hazard index are higher than the recommended level. Therefore the sediments of this beach pose significant threat to the people utilizing it.

As cited in an overview on measurement of natural radioactivity in Malaysia by Nisar et al 2015, one of the major health problems in Malaysia is cancer (Stomach, breast, lungs, liver, leukaemia and thyroid). Medically, it been certified that the fourth leading cause of death is cancer. Taking from the progress and development that has been achieved in Malaysia, cancer has become a very serious health concern. The result of a study done in 1994 in Penang, Malaysia showed that the age standardized incidence rate was 119.3 per 100,000 for all types of cancers. From 2004-2008, about 9692 cases

were identified in Penang, Malaysia. Sequel to the above, a study was carried out by Almayahi et al in 2012 and reported in his conclusion that high concentration of natural radioactivity and $^{226}\text{Ra}/^{238}\text{U}$ disequilibrium are the main cause of cancer in Penang (Almayahi et al 2012a). Therefore, it is important to know the natural radioactivity of every environment and monitor the radiation hazards that may appear to the inhabitants due to the use of soil; which happen to be the aim of the present study.

University of Malaya

CHAPTER 3: MATERIALS AND METHODS

3.1 Radiation detection techniques

The general principle involved in radiation detection is the interaction of radiation with matter which produces ionization and electronic excitation. The radiation either gives off some or all its energy to the medium of the detector by either ionizing it directly or causing emission of charged particles which later produces ionization of the medium.

Ionizing radiation can be measured through the physical and chemical effects of its interaction with matter. Field and laboratory methods are based mainly on the ionizing properties of radiation and the use of instruments that convert the radiation to electrical signals. Ionization chambers, proportional counters, Geiger-Muller tubes, scintillation counters, semiconductor detectors (for instance HPGe), thermoluminescence detector and various mechanical and chemical track detectors are used to monitor and quantify alpha, beta, gamma and neutron radiation in the environment.

3.2 Gamma ray spectroscopy

Gamma ray spectrometer is a powerful technique which identifies and quantifies specific energy photons (gamma rays), in environment and geological samples thereby quantitating specific radionuclides. It utilizes the direct proportionality between the energy of an incoming gamma ray and the pulse amplitude at the output of the detector. The interaction between the gamma rays from the sample and the detector atoms are amplified and converted into a voltage pulse proportional to the photon energy. Then, the pulse amplitudes are analysed, and the output of the spectrometer is an energy spectrum of detected radiation. Since individual radionuclides emit specific gamma

energies, the spectrum can be used to identify and quantify specific radionuclide that releases the radiation.

Gamma ray spectrometer consists of a detector, preamplifier, pulse-height analyser system, data readout capability and shield sample enclosure. The pulse height analyser consists of a linear amplifier, an analog to digital converter (ADC), memory storage and logic control mechanism which allows the storage of data in various modes and displays or recall. The amplitude gain stabilization in gamma ray spectrometers helps to the effect of energy spectrum drift

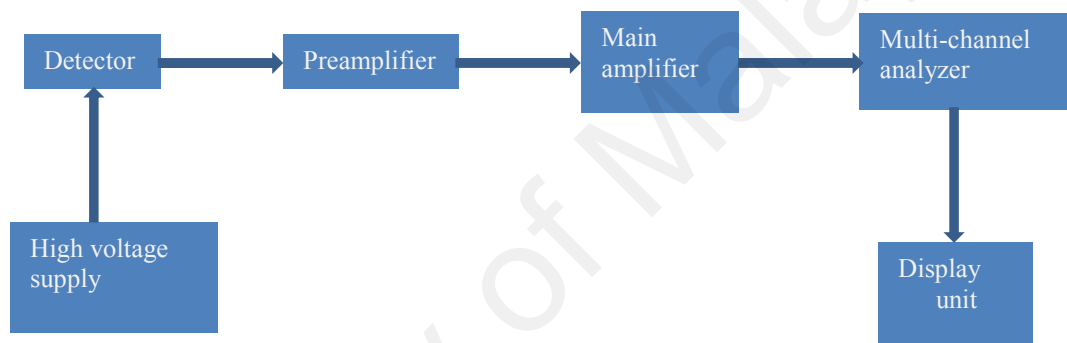


Figure 3.1 Diagram of Gamma Spectrometer Experimental setup

3.2.1 High purity germanium detector (HPGe)

HPGe is a semiconductor detector with high purity germanium. It approximately has the properties of theoretical true intrinsic semiconductor material and $< 10^{10}$ atoms/cm³ impurity. High purity germanium is capable of yielding up to 10mm depletion layer with 1000 volts of reserve bias. The germanium detector diode has the ability to endure high reverse bias voltage at crystal temperature.

The detector used in this experiment is a P-type Coaxial ORTEC, GEM-25 high purity germanium gamma ray detector which has 57.5mm crystal diameter and 51.5mm thickness, shielded with a cylindrical shaped lead which helps to minimize background radiation intrusion with the measured spectrum. The operating voltage of this detector is set at +2800V. The HPGe detector is cooled with Ortec X-Cooler II. The detector element absorbs the energy from incident gamma ray and produces a current pulse whose integral is proportional to the absorbed energy. According to Mayeen et al 2012, these types of detector present a more uniform, efficient cross-section of the active detector volume to the samples that are counted at a short distance from the detector.

3.2.2 Preamplifier

The preamplifier is the first element in a signal- processing chain. It converts the ionization charge developed in the detector during each absorbed nuclear event to a step function output pulse whose amplitude is proportional to the total charge accumulated in that particular event. The preamplifier is located very close to detector to achieve good performance. This will reduce the attenuation of the output signal in a way that maximizes the electronic signal to the noise ratio, hence amplifying the signal before additional noise or signal distortion can occur.

3.2.3 Main amplifier

The primary function of the amplifier is shaping of the pulse from the preamplifier. The amplifier enlarges the amplitude of the pulse coming from the preamplifier into adequate amplitude that can be easily and accurately measured. It shapes and filters the pulse to improve the signal-to noise ratio. It also prevents overlapping. The amplifier must be non-overloading, which means that it must amplify equally well at high count rates as it does at low count rates.

3.2.4 Multi-channel analyser (MCA)

The multi channel analyser does a great job in this experiment. It performs the function of pulse height analysis. The operation of multi channel analyser is basically on the principle of converting analog to digital. The analogy to digital converter (ADC) in MCA measures and sorts out the incoming pulses according to their amplitudes (Paschal, 2006). The output is then stored in the computer-type memory, which has many addressable locations and the number of channels into which recorded spectrum can be divided. The channels storage is done in counts per seconds. The output information is either displayed as data to be analysed or final result form in real time.

3.3 Detector resolution

The energy resolution of a detector is a measure of its ability to differentiate between two gamma rays that vary with very little energies difference. The width of a peak is determined by the resolution of the detector. The measure of the detector resolution is given by the sharpness of the photo peak. The energy resolution (R) is defined as full width of the photopeak at half maximum amplitude (FWHM) divided by its energy (E) corresponding to the highest point in the photopeak of a reference radioisotope expressed as a percentage.

i.e

$$R = \frac{E_{fwhm}}{E} \times 100\% \quad (3.1)$$

where E_{fwhm} is the full width at half maximum amplitude.

The resolution of the detector used in this study was 28.2% and 1.67keV FWHM energy resolution at 1.33 MeV (^{60}Co). This an excellent resolution to distinguish the gamma ray energies considered during this experiment

3.4 Calibration of the detector

The calibration of the detector is a very important aspect of gamma ray spectroscopy for radioactivity measurements. This is to ensure that the gamma ray spectra are accurately interpreted in terms of energy and specific activity. An essential requirement for the measurement of gamma emitters is the exact identity of photopeaks present in a spectrum produced by the detector system. The procedure for identifying the radionuclides within a spectrum relies upon methods which match the energies of the principal gamma rays emitted by known radionuclides.

3.4.1 Energy calibration

The energy calibration was done so to establish a relationship between peak position in the spectrum and the corresponding gamma ray energy. The height of each pulse output from a photomultiplier tube and hence the channel corresponding to it is directly proportional to the initial gamma energy producing the pulse. The calibration process involved different gamma emitter sources of known energies, these includes Cobalt-60 (1173.22 keV,1332.492keV) Americium-241(59.541 keV), Cadmium-109 (88.040 keV), Cobalt-57(122.061 keV,136.474 keV), Mercury-203 (279.195 keV), Strontium 85 (514.007keV), Cesium-137 (661.657keV), Yttrium-88 (898.042keV,1836.063keV),and Tin-113 (391.698keV). The gamma emitter sources were exposed to the HPGe detector and gamma spectra were obtained under suitable counting time. The gamma source and their corresponding energies are shown in table 3.1

Table 3.1 Energy calibration

Source	Energy (keV)
⁶⁰ Co	1173.22 and 1332.492
²⁴¹ Am	59.54
¹⁰⁹ Cd	88.04
⁵⁷ Co	122.061 and 136.474
²⁰³ Hg	279.195
¹¹³ Sn	391.698
⁸⁵ Sr	514.007
¹³⁷ Cs	661.657
⁸⁸ Y	898.042 and 1836.063

3.4.2 Efficiency calibration

In gamma ray spectrometry, the peak in the spectrum is related to the amount of radioactivity it represents. This requires the absolute full-energy peak efficiency. This gives a relationship between the peak area and the number of gamma rays emitted by the source. The efficiency was determined by using Ra-226 source only with energy range 186.211 to 1764.491 keV. It was ensured that the calibration standard reference sample represented the soil samples to be counted. The counting was done for 1 hour to determine the detector efficiency. The detection efficiency was determined for each of the gamma energies under investigation in this study and their values are tabulated in table 3.2. The dependence of the detection efficiency on the gamma ray energy is shown in figure 3.2 in page 43

Table 3.2 Radionuclide, Energy and Detection efficiency

Radionuclide	Energy (keV)	Detection efficiency
Pb-214	295.22	0.0188271
Pb-214	351.93	0.0170268
Bi-214	609.32	0.0124387
Bi-214	1120.29	0.0087799
Ac228	338.32	0.0174153
Ac228	911.20	0.0098811
Ac228	968.97	0.0095398
Tl-208	583.18	0.0127546
K-40	1460.82	0.0075432

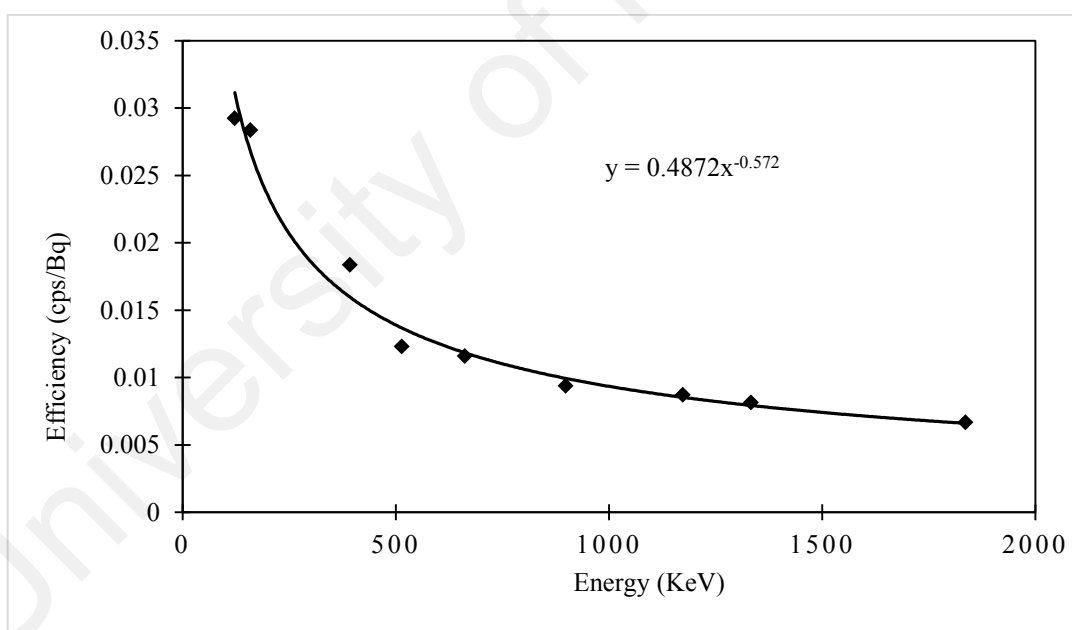


Figure 3.2 Detection efficiency curve of the detector

3.4.3 Determination of minimum detectable activity

The minimum detectable activity (MDA) of a measuring system describes its operating capacity without the influence of any sample. The detectable limit is expressed in Bq/kg, which is required to estimate the minimum detectable activity

concentration in a sample at 95% confidence level, using equation 3.1 (Khandaker et al, 2012)

$$MDA(Bq/kg) = \frac{K_{\alpha} \sqrt{N_B}}{\eta E P_{\gamma} T_c M} \quad (3.2)$$

where K_{α} , N_B , M , η (E) and P_{γ} are the statistical coverage factor equivalent to 1.645, the background count (cps), the sample mass (kg), the photo-peak efficiency, the probability of gamma emission and the counting time(s) respectively.

With the measurement system used in this study, the minimum detectable activity (MDA) for the targeted radionuclides was calculated to be 0.60Bq/kg, 0.70Bq/kg and 2.40Bq/kg for Radium-226, Thorium-232, and potassium-40 respectively. Any activity concentration values below these numbers are taken in this study as below detection limit (BDL) of the detector.

3.4.4 Background counting

Background radiation are the radiations that arise due to the presence of natural radionuclides in the environment, cosmic rays entering the atmosphere, radionuclides present in the building materials used in building the laboratory, electronic noise and other sources.

In this work, the environmental background was reduced with the help of the cylindrical shaped lead that shielded the detector. The background radiation was automatically deducted from the acquired spectrum by the machine.

3.5 Area of Study

University of Malaya, Malaysia is a multidisciplinary university with 17 faculties and research centres which covers the whole spectrum of learning from Arts, Science, Humanities, Engineering and Medicine. The university dates back 1957 and the first

choice of Malaysia's top students with international students population from more than 80 different countries. University of Malaya is seized 750 acre (309 hectare) with 12 residential colleges and strategically located (N3°7'15''E101°39'23'') within the heart of Kuala Lumpur_which makes it attractive and advantageous for her international academic staff, students and visitors. The population of University of Malaya as at February 2016 was 17580 students and 7245 staff.

3.6 Sample Sampling

Thirty six (36) soil samples were randomly collected from the twelve (12) residential college areas (samples from three sampling sites of each college) of University of Malaya, Malaysia, at a depth of 0-5 cm using hand auger as shown in figure 3.4 in page 48. The sampling sites (hostel, car park and recreational areas) were chosen due to dense human activities in the areas. The soil sample from each sampling site were thoroughly mixed together to provide a representative sample for the site. Thereafter, about 1kg of the composite sample were packed in a polythene bag and labeled with a sample identity using a coding that reflects both the area name, college and sample matrix. Then the collected soil samples were taken to the laboratory for preparation and spectroscopic analysis of the targeted radionuclides. The distance between each sampling site in a college was about 200-300 metres. The map showing the locations where the soil samples were collected across University of Malaya (UM) is shown in figure 3.3. The location of each sampling point was taken by means of a Global Positioning System as recorded in table 3.3

Table 3.3 Sampling sites coordinates.

Colleges in University of Malaya	Sample code	Location coordinates
1	C1C	N03° 07' 01'' E 101° 39' 30''
“	C1H	N03° 07' 01'' E 101° 39' 33''

“	C1R	N03° 07' 03'' E 101° 39' 35''
2	C2C	N03° 07' 05'' E 101° 39' 27''
“	C2H	N03° 07' 04'' E 101° 39' 28''
“	C2R	N03° 07' 04'' E 101° 39' 26''
3	C3C	N03° 07' 27'' E 101° 39' 2''
“	C3H	N03° 07' 25'' E 101° 39' 1''
“	C3R	N03° 07' 24'' E 101° 39' 3''
4	C4C	N03° 07' 31'' E 101° 38' 58''
“	C4H	N03° 07' 29'' E 101° 39' 0''
“	C4R	N03° 07' 29'' E 101° 39' 0''
5	C5C	N03° 07' 37'' E 101° 39' 29''
“	C5H	N03° 07' 40'' E 101° 39' 30''
“	C5R	N03° 07' 38'' E 101° 39' 33''
6	C6C	N03° 06' 55'' E 101° 39' 18''
“	C6H	N03° 06' 54'' E 101° 39' 16''
“	C6R	N03° 06' 53'' E 101° 39' 20''
7	C7C	N03° 07' 30'' E 101° 39' 0''
“	C7H	N03° 07' 34'' E 101° 39' 2''
“	C7R	N03° 07' 33'' E 101° 39' 3''
8	C8C	N03° 07' 48'' E 101° 38' 59''
“	C8H	N03° 07' 49'' E 101° 38' 56''
“	C8R	N03° 07' 47'' E 101° 38' 55''
9	C9C	N03° 07' 14'' E 101° 38' 41''
“	C9H	N03° 07' 17'' E 101° 38' 45''
“	C9R	N03° 07' 17'' E 101° 38' 42''
10	C10C	N03° 07' 50'' E 101° 39' 2''
“	C10H	N03° 07' 54'' E 101° 39' 2''
“	C10R	N03° 07' 51'' E 101° 39' 0''
11	C11C	N03° 07' 44'' E 101° 39' 39''
“	C11H	N03° 07' 48'' E 101° 39' 38''
“	C11R	N03° 07' 42'' E 101° 39' 40''
12	C12C	N03° 07' 33'' E 101° 39' 38''
“	C12H	N03° 07' 14'' E 101° 38' 41''
“	C12R	N03° 07' 17'' E 101° 38' 45''

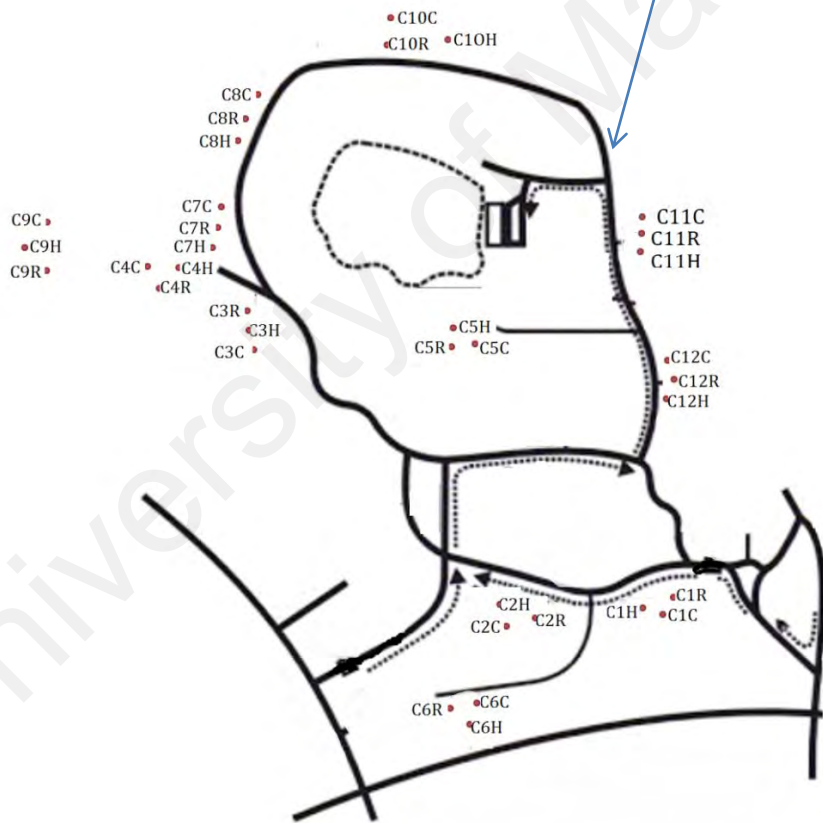
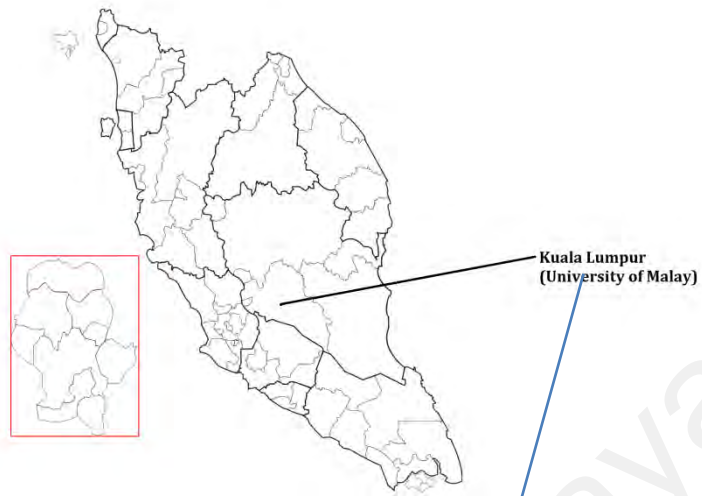


Figure 3.3 Location of University Malay (N3°7'15''E101°39'23'') Kuala Lumpur, Malaysia



Figure 3.4 Collection of samples at the sample sites

3.7 Sample preparation

In order to remove excess moisture, the soil samples were air dried in the laboratory at room temperature for 24 hours. They were further dried in the oven at 45-600 degree Celsius. The dried soil samples were pulverized to fine powder and sieved with a 1mm mesh sieve to achieve homogeneity. Then 500g of the homogenized soil sample were transferred into uncontaminated empty polyethylene plastic marinelli beakers; sealed, labeled and kept for a period of four (4) weeks to achieve secular radioactive equilibrium between short lived members of radium and thorium series as shown in figure 3.5 in page 49.



Drying of samples in the Oven



Weighing of samples



Sealed and labelled samples



Stored samples

Figure 3.5 Photos of samples preparation (oven drying, weighing and storage)

University of M

CHAPTER 4: MEASUREMENTS AND RESULTS

4.1 Measurement of ^{226}Ra , ^{232}Th and ^{40}K radioactivities

A gamma spectrometry system consisting of a P-type Coaxial ORTEC, GEM-25 high purity germanium gamma ray detector connected to an ADCM data acquisition system, coupled with PCAII multichannel analyser and gamma vision computer system was used for analysis in this study. The detector has a good resolution of about 28.2% at energy of 1.33 MeV. This is sufficient enough to distinguish the gamma energies of interest in this study.

Each of the samples were placed in the detector and measured for a period of 86400 seconds (24hrs). Then, the activity concentration of daughter radionuclides ^{214}Pb (determined from its 295.22 keV and 351.93 keV gamma ray peak) and ^{214}Bi (determined from its 609.32keV and 1120.29 keV gamma ray peak) were used to calculate the ^{226}Ra in the soil sample. While ^{228}Ac (determined from its 338.32keV, 911.20keV and 968.97keV gamma ray peak) and ^{208}Tl (determined from its 583.18keV gamma ray peak) were taken as indicators of ^{232}Th , ^{40}K was determined by measuring the 1460.82keV gamma rays emitted during the decay of ^{40}K . The photo of P-type Coaxial ORTEC, GEM-25 high purity germanium gamma ray detector is shown in figure 4.1.

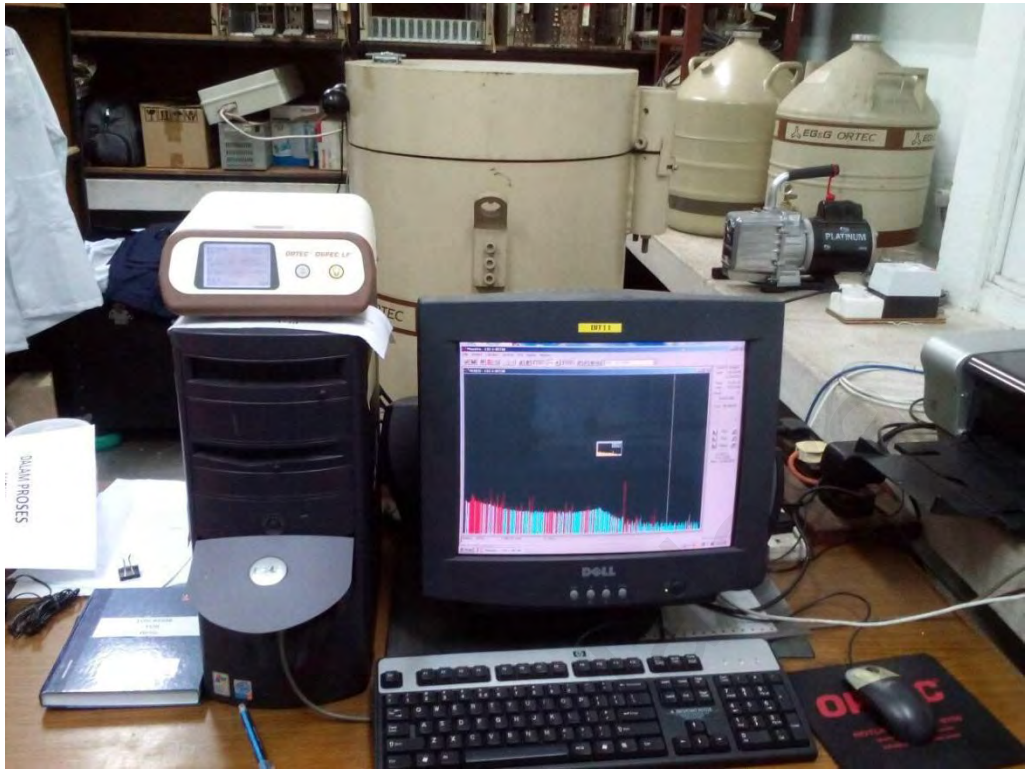


Figure 4.1 Photo of P-type Coaxial ORTEC, GEM-25 high purity germanium gamma ray detector with the MCA and gamma vision PC

4.2 Activity concentration

The radioactive source is described by its activity, which is the number of nuclear disintegration per unit of time. The activity concentration of each radionuclide is calculated using the below as expressed by Khandaker et al. 2012;

$$Ac \text{ (Bq/kg)} = \frac{CPS \times 1000}{\epsilon_{\gamma} \times I_{\gamma} \times W} \quad (4.1)$$

where Ac (Bq/kg), CPS, ϵ_{γ} , I_{γ} and W are specific activity, net counts per second for each sample investigated, the detector photo-peak efficiency at respective gamma-ray peak, the corresponding gamma-ray intensity and the mass of sample in gram respectively.

The mean activity concentration for ^{226}Ra , ^{232}Th and ^{40}K in the soil samples from residential college areas of University of Malaya are presented in table 4.1.

Table 4.1 Mean of activity concentration for ^{226}Ra , ^{232}Th and ^{40}K in the soil samples from residential college areas of University of Malaya

College	^{226}Ra (Bq/kg)	^{232}Th (Bq/kg)	^{40}K (Bq/kg)
C ₁	54.60±2.50	57.87±2.55	306.43±4.77
C ₂	61.27±2.35	78.44±1.60	180.19±2.14
C ₃	42.16±2.86	55.24±2.38	131.50±2.48
C ₄	54.33±2.69	80.27±1.95	237.43±1.92
C ₅	53.41±1.88	64.85±1.54	152.99±1.40
C ₆	50.38±1.89	69.58±1.48	212.55±1.32
C ₇	43.41±2.15	65.45±1.70	303.82±1.21
C ₈	57.7±2.24	78.16±1.48	270.09±1.42
C ₉	53.5±2.81	81.8±1.93	294.55±1.59
C ₁₀	54.09±2.67	52.89±2.58	262.41±1.76
C ₁₁	42.73±2.65	57.13±2.03	270.18±1.24
C ₁₂	60.15±2.05	79.4±1.75	73.07±4.91
Mean	52.31±2.40	68.42±1.92	224.58±2.28

Note: Total mean is the value of the average of the concentrations of particular radionuclides in all the colleges.

4.3 Hazard indices

In order to represent the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K by a single quantity, radiation hazards associated with them must be taken into consideration, which includes radium equivalent activity (Ra_{eq}), Absorbed dose rate (D_R), Annual effective dose equivalent (AEDE), Annual gonadal dose equivalent (AGDE), Activity utilization index (AUI), External and Internal hazard indices (H_{ex} and H_{in}), Representative gamma index ($I_{\gamma r}$), Excess lifetime cancer risk (ELCR)

4.3.1 Radium equivalent activity

According to Agbalagba and Onoja 2011; Dabayneh et al. 2008, radium equivalent activity (Ra_{eq}) it has been assumed that 259 Bq/kg of ^{232}Th or 370 Bq/kg of ^{226}Ra or

4810 Bq/kg of ^{40}K produces the same gamma-ray dose. Radium equivalent index is measured in becquerel per kilogram (Bq/kg). It is mathematically defined by;

$$\text{Ra}_{\text{eq}} = C_{\text{Ra}} + 1.43C_{\text{Th}} + 0.77C_{\text{K}} \quad (\text{UNSCEAR, 2000}) \quad (4.2)$$

where Ra_{eq} is the radium equivalent activity and C_{Ra} , C_{Th} and C_{K} are the measured specific activity concentration of ^{226}Ra , ^{232}Th , and ^{40}K respectively.

The radium equivalent activity for ^{226}Ra , ^{232}Th and ^{40}K in the soil samples from residential college areas of University of Malaya is shown in table 4.2.

Table 4.2 Mean of radium equivalent activity for ^{226}Ra , ^{232}Th and ^{40}K in the soil samples from residential college areas of University of Malaya.

College	Ra_{eq} (Bq/kg)
C ₁	160.95
C ₂	187.31
C ₃	131.28
C ₄	187.4
C ₅	157.93
C ₆	166.25
C ₇	160.4
C ₈	190.27
C ₉	193.15
C ₁₀	149.93
C ₁₁	145.23
C ₁₂	179.32
Total Mean	167.45

4.3.2 Absorbed dose rate (D_R)

The activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K calculated only shows the levels of the radionuclides present in the soil sample but do not represent the effect of such levels on bio-system. Therefore, to assess the radiation risk to a bio-system, absorbed dose rate should be calculated. The absorbed dose rates (D_R) of gamma radiations in air

at 1 m above the ground due to the concentrations of ^{226}Ra , ^{232}Th and ^{40}K was calculated using the equation (UNSCEAR, 2008);

$$D_R \text{ (nGy/h)} = 0.462 C_{\text{Ra}} + 0.604 C_{\text{Th}} + 0.0417 C_{\text{K}} \quad (4.3)$$

D_R is the absorbed dose and C_{Ra} , C_{Th} and C_{K} are the measured specific activity concentration of ^{226}Ra , ^{232}Th , and ^{40}K respectively.

The absorbed dose rate is measured in nanogray per hour and the calculated values are presented in table 4.3.

Table 4.3 Mean of absorbed dose rate values in the soil samples from residential college areas of University of Malaya.

College	D_R (nGy/h)
C ₁	72.96
C ₂	83.2
C ₃	58.33
C ₄	83.48
C ₅	70.22
C ₆	74.17
C ₇	72.26
C ₈	85.13
C ₉	86.41
C ₁₀	67.88
C ₁₁	65.51
C ₁₂	78.79
Total Mean	74.86

4.3.3 Annual effective dose equivalent (AEDE)

Reference to UNSCEAR 2000, determination of biological hazard due to individual exposure to radiation, gray is converted to Sievert with 0.7 SvGy^{-1} used as conversation factor for the biological effectiveness of the dose causing damage in human tissue and the occupancy factor of 0.2 that specifies the proportion of the total time spent outdoors and indoors. It is estimated that an individual spends an average of 80 % of his time indoor. The annual effective dose equivalent (AEDE) in outdoor air is measured in

millisievert per year (mSvy^{-1}) and the formula for its calculation as stated by UNSCEAR 2000 was;

$$\text{AEDE (mSvy}^{-1}\text{)} = \text{DR (nGyh}^{-1}\text{)} \times 8760\text{hr}^{-1} \times 0.7 \times \text{Sv/Gy} \times 0.2 \times 10^{-6} \text{ mSvy}^{-1}/\text{Sv} \quad (4.4)$$

The calculated values of Annual effective dose equivalent (AEDE) is show in table 4.4

Table 4.4 Mean of annual effective dose equivalent (AEDE) values in the soil samples from residential college areas of University of Malaya.

College	AEDE (mSvy^{-1})
C ₁	0.09
C ₂	0.1
C ₃	0.07
C ₄	0.1
C ₅	0.08
C ₆	0.09
C ₇	0.09
C ₈	0.1
C ₉	0.1
C ₁₀	0.08
C ₁₁	0.08
C ₁₂	0.1
Total Mean	0.09

4.3.4 Annual gonadal dose equivalent (AGDE)

In Ravisankar et al., 2014, annual gonadal dose equivalent (AGDE) is seen as the degree of genetic importance of yearly dose equivalent received by the population reproductive organs (gonads). Moreover, active bone marrow and bone surface cells were seen as organs of interest UNSCEAR (1988).

Using the formula stated by (Chandrasekaran et al., 2014; Ravisankar et al., 2014), annual gonadal dose equivalent (AGDE), due to the specific activities of ^{232}Th , ^{226}Ra and ^{40}K was estimated as

$$\text{AGDE } (\mu\text{Svy}^{-1}) = 3:09C_{\text{Ra}} + 4:18C_{\text{Th}} + 0:314C_{\text{K}} \quad (4.5)$$

Where AGDE is the annual gonadal dose equivalent and C_{Ra} , C_{Th} and C_K are the measured specific activity concentration of ^{226}Ra , ^{232}Th , and ^{40}K respectively. The annual gonadal dose equivalent measured in microsievert per year ($\mu Sv y^{-1}$) and its mean values obtained in this study are reported in table 4.5 below.

Table 4.5 Mean of annual gonadal dose equivalent (AGDE) values in the soil samples from residential college areas of University of Malaya.

College	AGDE ($\mu Sv y^{-1}$)
C ₁	506.83
C ₂	573.78
C ₃	402.47
C ₄	577.96
C ₅	484.15
C ₆	513.26
C ₇	503.12
C ₈	589.81
C ₉	599.73
C ₁₀	470.62
C ₁₁	455.68
C ₁₂	540.7
Total Mean	518.18

4.3.5 Activity utilization index (AUI)

The activity utilization index (AUI) is the dose rates in air from different combination of ^{226}Ra , ^{232}Th and ^{40}K (natural radionuclides) in soil samples. The formula prescribed by (Ravisankar et al., 2014) can be used to calculated AUI;

$$AUI = \left[\frac{C_{Ra}}{50 Bq/kg} \right] f_U + \left[\frac{C_{Th}}{50 Bq/kg} \right] f_{Th} + \left[\frac{C_K}{500 Bq/kg} \right] f_K \quad (4.6)$$

where C_{Ra} , C_{Th} and C_K are the measured specific activity concentration of ^{226}Ra , ^{232}Th , and ^{40}K respectively and f_{Th} (0.604), f_K (0.041) and f_U (0.462) are the individual fractional contributions to the total gamma radiation dose rate from the actual radioactivity level these radionuclides (Chandrasekaran et al., 2014). NEA-

OECD(1979) recorded that Typical activities per unit mass of primordial radionuclides in soils are 50 Bq/kg for ^{232}Th , 50 Bq/kg for ^{226}Ra and 500 Bq/kg for ^{40}K .

The calculated mean value of activity utilization index (AUI) in the soil samples from residential college areas of University of Malaya is tabulated in table 4.6.

Table 4.6 Mean of activity utilization index (AUI) in the soil samples from residential college areas of University of Malaya.

College	AUI
C ₁	1.23
C ₂	1.53
C ₃	1.07
C ₄	1.49
C ₅	1.29
C ₆	1.32
C ₇	1.22
C ₈	1.5
C ₉	1.51
C ₁₀	1.16
C ₁₁	1.11
C ₁₂	1.52
Total Mean	1.33

4.3.6 External and Internal hazard indices (H_{ex} and H_{in})

In radiological health evaluation surveys, external hazard index is measured to estimate the radiological hazard caused by external exposure to gamma radiation from the soil samples. As stated by UNSCEAR (2000), external hazard index (H_{ex}) can be calculated with the equation;

$$H_{\text{ex}} = \frac{C_{\text{Ra}}}{370} + \frac{C_{\text{Th}}}{259} + \frac{C_{\text{K}}}{4810} \quad (4.7)$$

Furthermore, radiations from radon and its short-lived daughter radionuclides present radiological hazard to the respiratory organ. UNSCEAR (2000), equation computed internal radiation exposure (H_{in}) as;

$$H_{in} = \frac{C_{Ra}}{185} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \quad (4.8)$$

where C_{Ra} , C_{Th} and C_K are the measured specific activity concentration of ^{226}Ra , ^{232}Th , and ^{40}K respectively. The radiation hazard is considered insignificant when the calculated values of H_{ex} and H_{in} are less than unity. Table 4.7 shows the H_{ex} and H_{in} values in the studied soil samples.

Table 4.7 Mean of external and internal hazard indices values in the soil samples from residential college areas of University of Malaya

College	$H_{ex} \leq 1$	$H_{in} \leq 1$
C ₁	0.43	0.58
C ₂	0.51	0.67
C ₃	0.35	0.47
C ₄	0.51	0.65
C ₅	0.43	0.57
C ₆	0.45	0.59
C ₇	0.43	0.55
C ₈	0.51	0.67
C ₉	0.52	0.67
C ₁₀	0.4	0.55
C ₁₁	0.39	0.51
C ₁₂	0.48	0.65
Mean	0.45	0.59

4.3.7 Representative gamma index ($I_{\gamma r}$)

It is important to take a look at the concentrations of ^{226}Ra , ^{232}Th and ^{40}K in relation to gamma radiation hazards. The representative gamma index ($I_{\gamma r}$) estimates the radiation hazard of gamma rays due to the individual radioactivity levels of the targeted primordial radionuclides in the soil samples. ($I_{\gamma r}$) can be computed using the equation given by (NEA-OECD1979; Ravisankar et al., 2014)

$$I_{\gamma r} = \frac{C_{Ra}}{150} + \frac{C_{Th}}{100} + \frac{C_k}{1500} \quad (4.9)$$

where C_{Ra} , C_{Th} and C_K are the measured specific activity concentration of ^{226}Ra , ^{232}Th , and ^{40}K respectively. The representative gamma index of this study is reported in table 4.8

Table 4.8 Mean of representative gamma index indices values in the soil samples from residential college areas of University of Malaya

College	lyr \leq 1
C ₁	1.15
C ₂	1.31
C ₃	0.92
C ₄	1.32
C ₅	1.11
C ₆	1.17
C ₇	1.15
C ₈	1.35
C ₉	1.37
C ₁₀	1.06
C ₁₁	1.04
C ₁₂	1.24
Total Mean	1.18

4.3.8 Excess lifetime cancer risk (ELCR)

The significance of exposure from natural radioactivity in soil and the possible health risk especially cancer, has gained global attention. Some regulatory bodies make use of quantitative risk assessment process to determine excess cancer risk over a lifetime (ELCR). Two of these bodies are United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) and Committee on the Biological Effects of Ionizing Radiation (BEIR) Both organizations agreed that their risk estimates should be reduced for a low dose exposures protracted over several months or years to account for a reduced effectiveness of the cell damage mechanism (Brian et al., 1995).

Therefore, there is need to determine the excess cancer risk over a lifetime (ELCR). The probability of developing cancer over a lifetime at a given exposure level can be calculated mathematically. It is quantified as a value representing the number of extra

cancers expected in a given population exposure to carcinogen (effective cancer causing radionuclides, substances or radiations) at a stated dose. As stated in Ravisankar et al., (2014) excess lifetime cancer risk (ELCR) can be estimated using the following formula

$$\text{ELCR} = \text{AEDE} + \text{DL} + \text{RF} \quad (4.10)$$

where AEDE, DL, and RF are annual effective dose equivalent, average lifetime duration (70 years) and risk factor (0.05 Sv^{-1}). i.e fatal cancer risk per Sievert respectively.

The excess lifetime cancer risk (ELCR) of this study is reported in table 4.9

Table 4.9 Mean of excess lifetime cancer risk (ELCR) values in the soil samples from residential college areas of University of Malaya.

College	ELCR ($\times 10^{-3}$)
C ₁	0.31
C ₂	0.35
C ₃	0.25
C ₄	0.35
C ₅	0.3
C ₆	0.31
C ₇	0.31
C ₈	0.36
C ₉	0.37
C ₁₀	0.29
C ₁₁	0.28
C ₁₂	0.33
Total Mean	0.32

Table 4.10 below is the summary of activity concentration and radiological hazard parameters in the soil samples from residential college areas of University of Malaya.

Table 4.10 summary of activity concentration and radiological hazard parameters in the soil samples from residential college areas of University of Malaya

College	Activity concentrations (Bq kg ⁻¹)				Dose			Hazard indices (≤ 1)				ELCR (x10 ⁻³)
	²²⁶ Ra	²³² Th	⁴⁰ K	Ra _{eq}	D _R (nGyh ⁻¹)	AEDE (mSvy ⁻¹)	AGDE (μ Sv y ⁻¹)	AUI	H _{ex}	H _{in}	I _{rr}	
C ₁	54.6	57.87	306.43	160.95	72.96	0.09	506.83	1.23	0.43	0.58	1.15	0.31
C ₂	61.27	78.44	180.19	187.31	83.2	0.1	573.78	1.53	0.51	0.67	1.31	0.35
C ₃	42.16	55.24	131.5	131.28	58.33	0.07	402.47	1.07	0.35	0.47	0.92	0.25
C ₄	54.33	80.27	237.43	187.4	83.48	0.1	577.96	1.49	0.51	0.65	1.32	0.35
C ₅	53.41	64.85	152.99	157.93	70.22	0.08	484.15	1.29	0.43	0.57	1.11	0.3
C ₆	50.38	69.58	212.55	166.25	74.17	0.09	513.26	1.32	0.45	0.59	1.17	0.31
C ₇	43.41	65.45	303.82	160.4	72.26	0.09	503.12	1.22	0.43	0.55	1.15	0.31
C ₈	57.7	78.16	270.09	190.27	85.13	0.1	589.81	1.5	0.51	0.67	1.35	0.36
C ₉	53.5	81.8	294.55	193.15	86.41	0.1	599.73	1.51	0.52	0.67	1.37	0.37
C ₁₀	54.09	52.89	262.41	149.93	67.88	0.08	470.62	1.16	0.4	0.55	1.06	0.29
C ₁₁	42.73	57.13	270.18	145.23	65.51	0.08	455.68	1.11	0.39	0.51	1.04	0.28
C ₁₂	60.15	79.4	73.07	179.32	78.79	0.1	540.7	1.52	0.48	0.65	1.24	0.33
Mean	52.31	68.42	224.58	167.45	74.86	0.09	518.18	1.33	0.45	0.59	1.18	0.32

Table 4.11 shows a summary of ^{226}Ra , ^{232}Th and ^{40}K activity concentration, absorbed dose rates, annual gonadal dose equivalent and excess lifetime cancer risk in different areas of Malaysia compared with the world values.

Table 4.11 Summary of activity concentration ^{226}Ra , ^{232}Th and ^{40}K , absorbed dose rates, annual gonadal dose equivalent and excess lifetime cancer risk in different areas of Malaysia and world values.

Study area	Activity concentrations (Bq kg^{-1})				Radiological dose and risk			Reference
	^{226}Ra	^{232}Th	^{40}K	Ra_{eq}	D_R (nGyh^{-1})	AGDE ($\mu\text{Sv y}^{-1}$)	ELCR ($\times 10^{-3}$)	
Pontain District	37	53	293	158.36	69	237	0.344	Muneer et al 2013
UluTriram Johor	44				200	245	1	Abdulrahman et al 2007
Melaka State					183	225	0.915	Sahrone et al 2005
Kinta District Perak	112	246	277		222	272	1.11	Lee et al 2009
Sungai Kampung	196	628	475		458	562	2.29	Apriantoro et al 2009
Palong					500	614	2.5	Khalik et al 2005
Johor State				174.64	163	200	0.815	Abdel et al 2001
Selama District	178	353	296		273	335	1.36	Ramli et al 2009b
UM college Areas	52.31	68.42	224.58	167.45	74.86	518.18	0.32	Present study
World	33	45	420		59	72	0.29	UNSCEAR 2000

CHAPTER 5: DISCUSSION

The HPGe radiological analysis to measure/investigate the radioactivity concentration and radiation hazard indices in the soil collected from the residential college areas of University of Malaya, Malaysia, was well-designed, properly carried out and results obtained. The results of the study are discussed under the following subheadings;

5.1 Radioactivity levels of ^{226}Ra , ^{232}Th and ^{40}K in the residential college areas of University of Malaya

Comparing the activity concentration values of the radionuclides in the twelve (12) residential college areas of University of Malaya, (Table 4.1), ^{226}Ra was highest at college two (C_2) with mean concentration value of 61.27 ± 2.35 Bq/kg, followed by colleges; C_{12} , C_8 , C_1 , C_4 , C_{10} , C_9 , C_5 , C_7 , C_{11} and lastly C_3 with concentrations values of 60.15 ± 2.05 Bq/kg, 57.7 ± 2.24 Bq/kg, 54.60 ± 2.50 Bq/kg, 54.33 ± 2.69 Bq/kg, 54.09 ± 2.67 Bq/kg, 53.5 ± 2.81 Bq/kg, 53.41 ± 1.88 Bq/kg, 43.41 ± 2.15 Bq/kg, 42.73 ± 2.65 Bq/kg and 42.16 ± 2.86 Bq/kg respectively. The high level of ^{226}Ra recorded in C_2 , C_{12} and C_8 may be due to the clay nature of the soils and fertilizer applied for horticultural purposes. The total average concentration of ^{226}Ra in all the twelve (12) residential colleges is 52.31 ± 2.40 Bq/kg. Average concentrations of ^{226}Ra in all the colleges are higher than the world average of 33 Bq/kg but lower than the Malaysian average value of 66 Bq/kg (UNSCEAR, 2000; Muneer et al., 2013).

The mean activity concentration of ^{232}Th in the twelve residential colleges varied between 81.8 ± 1.93 Bq/kg to 52.89 ± 2.58 Bq/kg. The highest value of ^{232}Th was recorded in college nine (C_9) and least value in college ten (C_{10}). The total average activity concentration of ^{232}Th was 68.42 ± 1.92 Bq/kg. The study showed that the mean activity concentration values of ^{232}Th in all the areas studied are higher than the world

average but lower than Malaysia average of 82 Bq/kg. The range of average activity concentration of ^{40}K was 306.43 ± 4.77 Bq/kg to 73.07 ± 4.91 Bq/kg with total mean activity concentration of all the colleges at 224.58 ± 2.28 Bq/kg. Colleges one (C_1) and twelve (C_{12}) had the highest and lowest values of ^{40}K activities respectively. The average radioactivity level of ^{40}K in this study was seen lower than the world average of 420 Bq/kg and Malaysia average of 310 Bq/kg. The high values in the average concentration of ^{226}Ra , ^{232}Th and ^{40}K recorded in some colleges may be due fertilizer applied for horticultural purposes. The loamy and clay nature of the soils may also affect the concentration of the radionuclides.

Comparing the result obtained from this study with results from other surveys in Malaysia, as shown in table 4.11, it was observed that the values of ^{232}Th , ^{226}Ra and ^{40}K in this study were lower than values in most of the previous studies. UNSCEAR (2000), reported crustal concentrations of areas with normal background radiation levels around the world as 16-110Bq/kg for ^{238}U , 11-64 Bq/kg for ^{232}Th and 140-820Bq/kg for ^{40}K . Therefore, the activity concentrations of the three radionuclides obtained in this study were consistent with world range and the soil can be classified under area of normal background radiation.

The radioactivity levels of natural radionuclides in the area of this study were in agreement with the global trend on the distribution of natural radionuclides in soil. The variations observed in different colleges could be attributed to geological characteristics and conditions of the soil of the study area. Besides these natural factors, human activities such as agricultural practices (horticulture) in which fertilizers are applied to improve the soil nutrients for plant growth had been known to be a contributor to the variations of activity concentrations of natural radionuclides.

5.2 Radium equivalent Index

Examining table 4.2, the highest mean value of activity was recorded at college nine (C₉) 193.15Bq/kg while the least mean value was observed in college three (C₃) with 131.28 Bq/kg. The overall mean value of radium equivalent activity in all the twelve (12) residential college areas of University Malaya was found to be 167.45 Bq/kg.

Report from (Sam & Abbas, 2001; Shiva Prasad et al., 2008) stated that the use of materials whose radium equivalent concentration exceeds 370 Bq/kg is discouraged in order to avoid radiation hazards. The radium equivalent of 370 Bq/kg corresponds to the dose limit of 1mSv for the general public. As it is evident in table 4.3, none of the colleges had mean radium equivalent activity value that exceeds 370Bq/kg. Therefore, soil from residential college areas of University of Malaya is safe from radiation hazards since the radium equivalent is less than 370Bq/kg.

5.3 Absorbed dose rate

The average value of absorbed dose rates of gamma radiations in air at 1 m above the ground due to the concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K were found to be 72.96 nGy/h, 83.20 nGy/h, 58.33 nGy/h, 83.48 nGy/h, 70.22 nGy/h, 74.17 nGy/h, 72.26 nGy/h, 85.13 nGy/h, 86.41 nGy/h, 67.88 nGy/h, 65.51 nGy/h, 78.79 nGy/h for colleges one to twelve respectively. The range of the mean gamma absorbed dose rate of all the college areas was 86.41 nGy/h to 58.33 nGy/h with highest and lowest values recorded at C₉ and C₃ respectively. The overall mean gamma absorbed dose rate from University Malaya residential college area soil was 74.86 Bq/kg. This value is higher than the global standard average value of 59 nGy/h but lower than maximum permissible value. The value is lower than the Malaysian average of 92 nGy/h.

5.4 Annual effective dose equivalent

In table 4.4, the highest values of outdoor annual effective dose equivalent values were recorded in C₂, C₄, C₈, C₉, & C₁₂ at 0.10 mSv⁻¹ and the least value reported in C₃

at 0.07mSvy^{-1} soil samples from residential college areas of University of Malaya. The calculated mean values of all the colleges ranged 0.07 to 0.1 mSvy^{-1} and the overall mean was 0.09 mSvy^{-1} which is lower than the global average value of 0.45 mSvy^{-1} and also less than the dose criterion limit of 1.0 mSvy^{-1} . Since the result obtained from this survey is less than the criterion limit, the soil from the area of study does not pose any radiological hazard to the public utilizing the environment.

5.5 Annual gonadal dose equivalent

The mean values of Annual gonadal dose equivalent are presented in table 4.5. College nine (C_9) and College three (C_3) showed highest and lowest values in this study with $599.73\text{ }\mu\text{Svy}^{-1}$ and $402.47\text{ }\mu\text{Svy}^{-1}$ respectively. The overall mean value of annual gonadal dose equivalent was $518.18\text{ }\mu\text{Svy}^{-1}$. This value was far higher than the USCEAR value of $72\text{ }\mu\text{Svy}^{-1}$ reported as global average but within the range of Malaysia values.

5.6 Activity utilization index

Table 4.5 presented the mean values of the activity utilization index of residential college area of University of Malaya. The values varied from 1.07 to 1.53 with the highest value observed in college two (C_2) and least in college three (C_3). The overall average mean of all the colleges was 1.33. This value is above 1 which is the value of AUI that corresponds to an annual effective dose safety limit. Although the mean value of AUI in this study was high but it is still below the maximum permissible limit.

5.7 External hazard indices (H_{ex})

From table 4.7, the overall average of the external hazard index calculated from the twelve residential college areas of University of Malaya was 0.45, while individual college values varies from 0.35 to 0.52. The highest value was observed in College nine (C_9) and least value in college three (C_3). Since the world safe limit is put at 1,

therefore, the soil of University of Malaya college areas with 0.45 average values is safe from radiation hazards.

5.8 Internal hazard indices (H_{in})

Table 4.7 reported the highest values of internal hazard indices at colleges; two (C_2), eight (C_8) and nine (C_9) with values of 0.67. The least values was college three (C_3) with 0.47 and the overall mean value of all the colleges was found to be 0.59. The global recommended safe limit value for internal hazard indices was reported at 1 and all the values from the colleges are lower.

5.9 Representative gamma index

Considering all the values in table 4.8, the highest mean value of the representative gamma index was found at college nine (C_9) with 1.37 value and the least value was at college three (C_3) with 0.92. The total average value of representative gamma index for University of Malaya residential college areas soil was calculated to be 1.18. The value was above the recommended unit value to keep the effective dose rate below or at 1.0mSvy^{-1} .

5.10 Excess lifetime cancer risk

Making reference to table 4.9, the excess lifetime cancer risk values of University of Malaya residential college areas soil ranged from 0.25×10^{-3} to 0.37×10^{-3} . The highest value was observed at college nine (C_9) and least value at college three (C_3). The total value of excess lifetime cancer risk for University of Malaya residential college areas soil was calculated as 0.32×10^{-3} . The excess lifetime cancer risk for UM college areas was a higher when compared with the world average value of 0.29×10^{-3} . The chances of developing cancer over a lifetime by members of the public utilizing the environment are 10.3%.

CHAPTER 6: CONCLUSION

The activity concentrations of natural radionuclides radium-226, thorium-232 and potassium-40 in the twelve (12) residential colleges of University of Malaya, Malaysia have been measured using a gamma-ray spectrometer with high purity germanium (HPGe) detector. The activity concentrations of the three radionuclides have been used to calculate the radium equivalent activity, absorbed dose rate, annual effective dose equivalent, annual gonadal dose equivalent, activity utilization index, external hazard index, internal hazard index, representative gamma index and excess lifetime cancer risks to estimate the radiological implications on the occupants.

The following conclusions were made from the survey:

1. The mean activity concentration of ^{226}Ra , ^{232}Th and ^{40}K were found to be within the normal background level with UNSCEAR crustal concentration range.
2. The average value for each of the assessed radiological hazard parameter in the areas was found to be below world safety limit set by UNSCEAR; therefore, the areas have no harmful radiation effect associated with studied radionuclides.
3. The calculated $\frac{Th}{U}$ overall mean concentration ratio was 1.308 which is lower compared with the world's ratio of 3.5. And the $\frac{U}{Th}$ concentration ratio is 0.765 which is higher than the global average of 0.26.
4. The values of absorbed dose rate, annual gonadal dose equivalent, activity utilization index, representative gamma index, and excess lifetime cancer risk were higher than the world average values but were below the permissible limit.
5. Radium equivalent activity, annual effective dose equivalent, external and internal hazard indices values were lower than the world's values which shows

that the soil of residential college areas of University of Malaya, Malaysia pose no potential health threat connected to natural radioactivity.

6. The occupants of the hostels are safe from radiological problems.
7. The results of this study provide a general background level for the area studied and can be used as baseline data and guide for future evaluation and assessment of radiological hazards to human health.
8. The analysis provides good experience and skills for naturally occurring radioactivity measurement and monitoring.

6.1 Limitation and suggestion for further studies

The major constraint encounter in this study was the frequent breaking down of the detector. This limitation affected the expected time of completion of this research. With the experiences gained from this survey, the following suggestions are recommended for further study:

1. A formal notification should be given to hostels wardens and students to inform them about the survey, so to grantee the safety and integrity of the researcher.
2. More detailed research on natural radioactivity can be carried out in University of Malaya hostels since an individual spend 80% of his time indoor.
3. Regular assessment and monitoring of the studied area due to those radionuclides higher than world average.

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