NATURAL RADIOACTIVITY AND RADON CONCENTRATION IN SOIL AND WATER FROM NON-CULTIVATED AND CULTIVATED AREA OF SEBERANG PERAI, MALAYSIA

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by

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LIST OF ABBREVIATIONS

CRM	Continuous Radon Monitor
DOE	Department of Environment
EC	European Commission
FAAS	Flame Atomic Absorption Spectrometer
HPGe	High Purity Germanium
IAEA	International Atomic Energy Agency
ICRP	International Commission on Radiological Protection
NCRP	National Council on Radiation Protection and Measurements
NORM	Naturally Occurring Radioactive Materials
NRPB	National Radiological Protection Board
NTDs	Nuclear Track Detectors
OECD	Organization of Economic Cooperation and Development
RTC	Radon Tight Chamber
UNSCEAR	United Nations Scientific Committee on the Effects of Atomic Radiation
WHO	World Health Organization

LIST OF SYMBOLS

λ	Decay constant of radionuclide
λ_{Rn}	Decay constant of radon
ε	Efficiency of HPGe detector at gamma ray line
η	Absolute full energy detection efficiency
ω	Back diffusion constant for soil
κ	Porosity of soil
A	Sample surface area (used for CR-39)
A_o	Area of field of view
A_i	Initial activity of source
A_s	Specific activity of radionuclide
AED_{in}	Indoor annual effective dose
AED_{ingest}	Annual effective dose for ingestion
AED_{out}	Outdoor annual effective dose
C_{eq}	Equilibrium radon concentration
C_K	Activity concentration of ^{40}K
C_{Ra}	Activity concentration of ^{226}Ra
C_{Rn}	Concentration of radon
C_{Th}	Activity concentration of ^{232}Th
D_{in}	Indoor absorbed dose
D_{out}	Outdoor absorbed dose
E	Energy of gamma line
E_R	Exhalation rate of radon
H_{in}	Internal hazard index
H_{ex}	External hazard index

I_α	Alpha index
I_γ	Gamma index
m	Mass of soil sample
$P_\gamma(E)$	Gamma emission probability at energy E
Ra_{eq}	Radium equivalent
S	Surface of soil sample (used for CRM)
V	Void space's volume in container (used for CR-39)
V_{eff}	Air effective volume of RTC
z_o	Thickness of soil (used for CR-39)

KERADIOAKTIFAN TABII DAN KEPEKATAN RADON DALAM TANIH DAN AIR DARIPADA KAWASAN PENANAMAN DAN BUKAN PENANAMAN DI SEBERANG PERAI, MALAYSIA

ABSTRAK

Kesejahteraan manusia dikompromi dan terjejas apabila lebih terdedah kepada radionuklid tabii (^{226}Ra, ^{232}Th, ^{40}K) dalam tanih dan air dan gas radon (^{222}Rn) dalam tanih. Tesis ini berusaha untuk memperoleh data asas kepekatan radionuklid yang terdapat secara tabii (^{226}Ra, ^{232}Th, ^{40}K), dos radiologi dan indeks bahaya radionuklid ini. Empat puluh sampel tanih penanaman dan tiga puluh sampel tanih bukan penanaman, dan tiga puluh dua sampel air yang digunakan untuk pengairan dan air paip diperoleh dari Seberang Perai, Malaysia. Sampel tersebut dinilai menggunakan pengesan Germanium ketulenan tinggi (HPGe); pengesan (nombor model GEM-M7040P4, Canberra, Inc.) memperoleh kecekapan relatif 40% dan resolusi tenaga 1.9 keV pada 1.3322 MeV daripada ^{60}Co. Selain itu, kepekatan gas radon (^{222}Rn) dan kadar ekshalasi radon dinilai bagi sampel tanih penanaman dan bukan penanaman dengan menggunakan Monitor Pengesan Radon (CRM) dan Pengesan Trek Nuklear (CR-39 NTDs). Pertamanya, purata aktiviti kepekatan ^{226}Ra, ^{232}Th dan ^{40}K dalam tanih penanaman dinilai sebanyak 85.01 ± 42.14, 59.09 ± 22.75 dan 384.86 ± 216.28 Bq kg^{-1}. Manakala, dalam tanih bukan penanaman didapati sebanyak 54.21 ± 34.15, 55.19 ± 47.22 dan 276.87 ± 203.43 Bq kg^{-1}, masing-masing. Oleh itu, purata kepekatan radionuklid (^{226}Ra, ^{232}Th, ^{40}K) didapati lebih tinggi dalam tanih penanaman berbanding tanih bukan penanaman. Walau bagaimanapun, beberapa tanih yang bukan penanaman telah menunjukkan bacaan yang tinggi di Kampung Mengkuang, Kubang

Semang di Seberang Perai Tengah sepadan dengan julat bacaan yang dilaporkan untuk negara-negara lain di seluruh dunia. Selain itu, indeks bahaya luaran (H_{ex}) dianggarkan melebehi satu untuk tanih bukan penanaman di Kampung Mengkuang, Kubang Semang; sebaliknya, bacaan adalah kurang daripada satu. Keduanya, kepekatan radon berubah dari 18 Bq m^{-3} hingga 1381.48 Bq m^{-3}, setanding dengan nilai di seluruh dunia. Tambahan pula, kadar ekshalasi radon dari kedua-dua tanih penanaman dan bukan penanaman dianalisis dengan menggunakan CRM dan CR-39 NTD dan didapati berada di bawah had keselamatan 57.6 Bq m^{-2} j^{-1}. Secara perbandingan, purata aktiviti kepekatan ^{226}Ra, ^{232}Th dan ^{40}K di perairan (sungai, saliran, tasik, air paip) dinilai sebanyak 1.12 ± 0.46, 3.14 ± 1.13 dan 136.56 ± 19.07 Bq l^{-1} serta kepekatan radionuklid (^{226}Ra, ^{232}Th, ^{40}K) dalam semua jenis air didapati lebih rendah berbanding nilai yang sama di seluruh dunia. Walau bagaimanapun, nilai dos berkesan tahunan untuk pengingesan ($AED_{ingest.}$) dalam air paip didapati lebih tinggi berbanding dos berkesan tahunan yang dicadangkan bagi konsumsi air minuman sebanyak 0.1 mSv y^{-1} seperti yang disyorkan oleh IAEA dan WHO. Akhirnya, penemuan ini memberikan tinjauan komprehensif tentang kesan radionuklid yang terdapat secara tabii di dalam tanih dan air dan kesan kepekatan radon di udara terhadap kesihatan penduduk di kawasan Seberang Perai. Dapatan ini membantu mengelakkan risiko kesihatan daripada sinaran dengan memilih perumahan yang sesuai, tanah pertanian dan bahan binaan yang sesuai. Oleh itu, adalah disyorkan bahawa beberapa tanih yang digunakan dalam aktiviti pertanian dan bahan binaan harus dipilih apabila indeks bahaya luaran (H_{ex}) adalah kurang daripada satu. Juga, air adalah selamat selepas pemprosesan dan penurasan, dan sesuai untuk kegunaan rumah dan keperluan industri.

NATURAL RADIOACTIVITY AND RADON CONCENTRATION IN SOIL AND WATER FROM NON-CULTIVATED AND CULTIVATED AREA OF SEBERANG PERAI, MALAYSIA

ABSTRACT

Human well-being is compromised and jeopardized when over exposed to natural radionuclides (^{226}Ra, ^{232}Th, ^{40}K) in soil and water and radon gas (^{222}Rn) in soil. This thesis endeavors to acquire fundamental data of naturally occurring radionuclides concentrations (^{226}Ra, ^{232}Th, ^{40}K), radiological doses and hazard indexes of these radionuclides. Forty samples of cultivated soil and thirty samples of non-cultivated soil, and thirty-two samples of water utilized for irrigation and tap water were acquired from Seberang Perai, Malaysia. The samples were evaluated using High Purity Germanium detector (HPGe); the detector (model no. GEM-M7040P4, Canberra, Inc.) obtained 40% relative efficiency and 1.9 keV energy resolution at 1.3322 MeV of ^{60}Co. Additionally, radon gas (^{222}Rn) concentrations and radon exhalation rates were evaluated for both non-cultivated and cultivated soils samples by employing a Continuous Radon Monitor (CRM) and Nuclear Track Detectors (CR-39 NTDs). Firstly, the average concentrations activity of ^{226}Ra, ^{232}Th and ^{40}K in cultivated soils were evaluated to be 85.01 ± 42.14, 59.09 ± 22.75 and 384.86 ± 216.28 Bq kg^{-1}, while in non-cultivated soil were found to be 54.21 ± 34.15, 55.19 ± 47.22 and 276.87 ± 203.43 Bq kg^{-1}, respectively. Thus, the average concentrations of radionuclides (^{226}Ra, ^{232}Th, ^{40}K) were exhibited higher in cultivated soils than non-cultivated soils. However, some non-cultivated soils have been manifested high in Kampung Mengkuang, Kubang Semang in Seberang Perai Tengah corresponding with

the range of those reported for other countries across the world. Moreover, the external hazard index (H_{ex}) is estimated above unity from non-cultivated soils in Kampung Mengkuang, Kubang Semang; otherwise, it is registered below unity. Secondly, radon concentrations varied from 18 Bq m^{-3} to 1381.48 Bq m^{-3}, which were comparable to the values worldwide. Furthermore, radon exhalation rates from both cultivated and non-cultivated soils were analyzed by using CRM and CR-39 NTDs and found to be below the safety limit of 57.6 Bq m^{-2} h^{-1}. Comparatively, the average concentrations activity of ^{226}Ra, ^{232}Th and ^{40}K in waters (river, stream, lake, tap) were evaluated to be 1.12 ± 0.46, 3.14 ± 1.13 and 136.56 ± 19.07 Bq l^{-1} as well as the concentrations of radionuclides (^{226}Ra, ^{232}Th, ^{40}K) in all types of water were found lower compared to the corresponding values worldwide. However, the values of annual effective doses for ingestion ($AED_{ingest.}$) in tap water were found higher than the recommendation annual effective dose for ingestion of drinking water of 0.1 mSv y^{-1} as recommended by the IAEA and WHO. Finally, the findings gave a comprehensive survey of the effect of naturally occurring radionuclides in soil and water and the impact of radon concentrations in the air on people's health in Seberang Perai region. This finding helps to avoid the health risks from radiations by selecting suitable housing, arable land and suitable building materials. Thus, it is recommended that some soils used in agriculture activities and building materials should be opted when the external hazard index (H_{ex}) is less than unity. Also, water is safe after processing and filtration, and appropriate for household use and industrial purposes.

CHAPTER 1: INTRODUCTION

1.1 Background

Knowledge of radioactivity contents in various types of soils and water forms an integral part of Health Physics. According to a report by Healthy Environments and Consumer Safety, radionuclides are found in the environment as naturally occurring radionuclides and as byproducts of artificial radionuclides (Health Canada, 1995). Both soils and water act as the primary sources of the Naturally Occurring Radioactive Materials (NORM). These radioactive materials can be categorized into three groups, which comprise of primordial or terrestrial, cosmogenic and anthropogenic nature (UNSCEAR, 1988). There is a high potential of these materials to contribute appreciably to the dose received by humans. This dose could occur through internal exposure as a result of their ingestion or inhalation, or external exposures (Eisenbud & Gesell, 1997). Thus, it is important to incorporate appropriate methods in minimizing the hazardous effects of these high quantities of these radionuclides (Herranz, Abelairas, & Legarda, 1999; Sorg, 1990).

Natural radionuclides particularly those found in decay chains of ^{238}U and ^{232}Th are highly radiotoxic. Notable among these is ^{226}Ra and ^{228}Ra. Comparatively, several human activities have introduced artificial radionuclides. These activities include nuclear power plants, nuclear weapons testing and manufacture and use of radioactive isotopes in medicine and industry (Al-Qasmi et al., 2016). In addition to these activities such as mining, milling and processing of uranium ores and mineral sand, manufacture and using of fertilizer, burning of fossils fuels and metal refining

have increased the amounts of NORM in the environment to levels that pose a threat to human health (Pujol & Sanchez-Cabeza, 2000).

Fertilizer industry relies on materials such as phosphates. There are high quantities of natural radionuclides, such as uranium and thorium, originate from phosphate rocks. Thus, most of commercial fertilizers include large concentrations of natural radionuclides. These fertilizers are used in the soils to raise the level of fertility in plants, leading to an increase in the abundance of plants and their productivity (Ghosh et al., 2008). Therefore, plants take up a significant amount of the radioactive substance that ends up being consumed by man. Regarding external and internal exposure to phosphate rocks and fertilizers, human is exposed externally to gamma rays from phosphate rocks and fertilizers. Comparatively, internal exposure involves the ingestion of food contaminated with radioactive materials and inhalation of radon gas and fertilizer dust, can affect human by alpha particles and gamma rays. For instant, farmers are exposed to the dust of phosphate fertilizers in agricultural land by direct inhalation (Ahmed & El-Arabi, 2005; Scholte & Timmermans, 1996).

People inhaling radioactive gasses are at high risk in respect to their health. These gasses headed by radon gas which originates in soils and rocks beneath the houses, building materials, underground and surface water and natural gas. The radon (^{222}Rn) produced in the uranium (^{238}U) series can decay into short-lived daughters (^{218}Po and ^{214}Po) by half-life ($T_{1/2}$ = 3.82 days). Radon emits alpha particles during its decay and is considered as a notable source of lung cancer for non-smokers in the world. Most of the radon gas is out of the human body before it decays during inhalation and exhalation processes and that under a very short period.

The problem lies in the products of radon and their depositions in the lung, discharging energy in the form of alpha particles. Alpha particles could cause double-stranded DNA breaks or generate free radicals that can also destruct the DNA and thus cause lung cancer (Brüske-Hohlfeld, 2009; Steck, 2005).

The interaction of the human body with radiation from external and internal sources leads to biological and health effects. The external and internal exposures cause two kinds of health effects resulting from changes in atoms and molecules of body tissues. One of the effects occurs in which the severity of the tissue damage is proportional to the dose and the other, which a threshold dose exists below which they do not occur. These later show up as clinical symptoms. The nature and severity of these symptoms and the rate at which they appear depends on the amount of radiation absorbed and the rate at which it is received. Injuries resulting from radiation can be divided into two classes, somatic and genetic effects. In somatic effects, damages appear in the irradiated person while genetic effects arise only in the offspring of the irradiated person. This occurs as a result of radiation damaging germ cells in the reproductive organs (the gonads)(Dalci, Dorter, & Guclu, 2004).

Recently, reports highlight on high-level exposure arising from natural radionuclides particularly ^{238}U, ^{232}Th, and ^{40}K. This level is based on an observation of their annual contributions to the accumulated radiological dose (Chambers, 2015). Investigations on terrestrial natural radiation in soils, water, and radon gas impacts, have received significant reasons interest globally (Ahmad, Jaafar & Khan, 2014; Al-Ghamdi, 2014; Almayahi, Tajuddin & Jaafar, 2012b; Bleise, Danesi & Burkart, 2003; Dusane, Mishra, Sahu & Pandit, 2014; Saleh, Ramli, Alajerami & Aliyu, 2013). However, limited reports have been supported in Seberang Perai in Penang to

track the source and nature of minerals causing enhanced levels of natural radiation in both soils and water. Alsaffar et al. (2015) found natural radioactivity in soils used for rice plant in Seberang Perai with the maximum values of 208.51 Bq kg^{-1} for ^{226}Ra, 194.13 Bq kg^{-1} for ^{232}Th and 943.11 Bq kg^{-1} for ^{40}K and minimum values of 49.4 Bq kg^{-1} for ^{226}Ra, 68.22 Bq kg^{-1} for ^{232}Th and 138.31 Bq kg^{-1} for ^{40}K (Alsaffar, Jaafar, Kabir & Ahmad, 2015).

1.2 Problem Statement

The external exposure from gamma rays from soil and internal exposure from alpha particles from food, water and inhaling radon gas in air are two factors to cause cancer like skin and lung cancer. Lung cancer is the common killer in Malaysia because of radon gas (Almayahi et al., 2012a). Study about radioactivity of soil and radon concentrations has been conducted in Pulau Pinang, however the study is restricted to the part of Penang in the territory. The results of the study were high in concentrations of the radionuclides and radon concentration in Penang Island. Also, the geological nature of Seberang Perai contain the rocks which might have radioactivity similar to other studies in Malaysia (Sanusi et al., 2014). These reasons drew the attention of the analyst to concentrate on other part of Penang state, which is Seberang Perai. The research novelty is based on the study of concentrations of radionuclides in non-cultivated soil and cultivated soil and their association with the concentration of radon in Seberang Perai to bridge the gap of radiological data in this region and protect people's health.

Natural radionuclides concentrations were observed to be higher in both cultivated and non-cultivated soils collected from northern parts of Malaysian

Peninsula especially in Penang and Cameron Highlands (Almayahi, Tajuddin & Jaafar, 2012a; Murtadha Sh Aswood, Jaafar & Bauk, 2013). These areas had some common factors such as practicing agricultural activities using different types of fertilizers to improve the qualities of the crops. Extensive use of fertilizer leads to water pollution, which ends up affecting people's health in the case that they consume the polluted water. Additionally, the accumulation of radionuclides in both soils and water act as a potential source of environmental pollution. Therefore, it is essential to measure concentrations of these radionuclides with the aim of protecting individuals' health (Murtadha Sh Aswood et al., 2013).

It is an essential to think of the contributions of the earth crust and geological areas regarding radionuclides. Water sources are contaminated directly through the earth's crust containing radionuclides like ^{238}U and ^{232}Th, and their daughters like ^{226}Ra and ^{228}Ra, respectively. The natural non-series radionuclide ^{40}K is also found. Artificial pollution of these water sources occurs through radioactive wastes. Therefore, natural radionuclides are highly toxic and contribute to the doses received by humans through both internal and external exposure (Eisenbud & Gesell, 1997). Thus, this research, through investigation addresses the following problems:

a. What is the level of NORM in both non-cultivated and cultivated soils, and water?

b. By using active (CRM) and passive (CR-39) techniques, what is the level of radon concentrations in both non-cultivated and cultivated soils?

1.3 Objectives of Research

This study involves the following objectives:

1. To measure concentrations of natural radionuclides ^{226}Ra, ^{232}Th and ^{40}K in soils (non-cultivated soil and cultivated soil) and water (rivers, streams, lakes, taps) in Seberang Perai, Penang.

2. To measure concentrations and exhalation rates of radon gas ^{222}Rn in soils (non-cultivated soil and cultivated soil).

3. To compare the data obtained to the international world averages and other countries.

1.4 Scope of Research

The research will focus on measuring concentrations and distributions of natural radionuclides ^{226}Ra, ^{232}Th, and ^{40}K in soils and water from different locations in Seberang Perai, Penang. Emphasis is to assess the level of background radiation arising from these radionuclides and fill the gap of studies in Seberang Perai and provide the data for protecting people's health. Also, the comparison between non-cultivated and cultivated soils was employed to study the impact of fertilizer to increase the radioactivity in non-cultivated soil. A hyper pure Ge-detector will be used to measure concentrations and distributions of the natural radionuclides. The study is important as it provides ^{222}Rn concentration in different soils samples and the exhalation rate using a Continuous Radon Monitor (CRM) and CR-39 detectors.

1.5 Outline of Thesis

This thesis comprises of five chapters covering different sections. Chapter 1 focuses on the general introduction of natural radionuclides, problem statement and objectives. At the end of Chapter 1 is the scope of research and an outline of the thesis. Chapter 2 is a summary of sources of background radiations, radon emanation and exhalation and exposure pathways as well as literature review in natural radionuclides in soils and water, and radon in soils. A description of the study area, locations and preparation of samples, measurement of natural radionuclides using HPGe in soils and water and measurement of radon concentration in soils using CRM and CR-39 detectors are comprehensively discussed in Chapter 3. Chapter 4 provides a summary of the results and discussion. Lastly, Chapter 5 covers the conclusion and future work relating to the research.

CHAPTER 2 : THEORETICAL BACKGROUND AND LITERATURE REVIEW

2.1 Sources of Natural Radionuclides Radiations

Natural radionuclides radiations can be classified into three categories according to their behavior on the environment. This includes radiations resulting from the natural radionuclides on the earth's surface (the terrestrial radiation), radioactivity in water and airborne radioactivity.

2.1.1 Terrestrial Radiation

Terrestrial radiation occurs naturally through the presence of NORM within the earth's crust. Terrestrial radiation includes primordial radionuclides of two types; the series primordial and non-series radionuclides (Haber, 2015). The series primordial radionuclides contain mainly ^{238}U series, ^{232}Th series and Actinium (^{235}U) series. The ^{238}U series starts with ^{238}U and transferring to ^{226}Ra and then ends with stable element ^{206}Pb. The process is facilitated by the decaying activity of alpha, and beta particles alongside gamma radiations as illustrated in Figure 2.1. The relative abundance of ^{238}U is found to be 99.3 % in terrestrial sources. Similarly, ^{232}Th series starts with ^{232}Th, proceeding to ^{228}Ra and ends with a stable element ^{208}Pb through alpha and beta decays alongside gamma radiations. A clear presentation of the events that covers this process is presented in Figure 2.2. Relative abundance of ^{232}Th is almost 100% in terrestrial sources compared to that of ^{228}Th, which is 1.35×10^{-8} %. The ^{235}U series starts with ^{235}U and ends with stable element ^{207}Pb through alpha and beta decays alongside gamma radiations as in Figure 2.3. The relative abundance of ^{235}U is 0.7 % in terrestrial sources (IAEA, 1990). The concentration of uranium and

thorium varies considerably depending on the type of rock formation. These elements are present in water and soils of the earth's strata in small quantities. A high level of uranium in phosphate rocks corresponds to its high concentrations in commercial phosphate fertilizer. On a similar account, shales containing organic matter are found to be highly radioactive (Boyle, 2013; Hamilton, 1989).

The most abundant groups of ^{40}K and ^{87}Rb comprise the non-series primordial radionuclides. Other members included in this group are ^{50}V, ^{142}Ce, ^{209}Bi, ^{190}Pt, and ^{115}In. These members contain no dosimetric significance. Additionally, several of these elements decay directly into a stable nuclide. The radionuclide ^{40}K occurs only to the extent of 0.0118% isotopic abundance in natural potassium. Its character in being ubiquitous in living systems influences its contribution to as much as one-third of the external terrestrial and internal dose from natural background. Comparatively, the isotopic abundance of ^{87}Rb is found to be higher than that for ^{40}K, although its contribution to dose is limited by its relative scarcity within the earth's crust (Alpen, 1997).

Other terrestrial radionuclides are found to exist in low levels; therefore, limiting their contribution to dose in humans is minimal. These radionuclides include the examples of; ^{235}U series, ^{138}La, ^{I47}Sm, and ^{176}Lu.

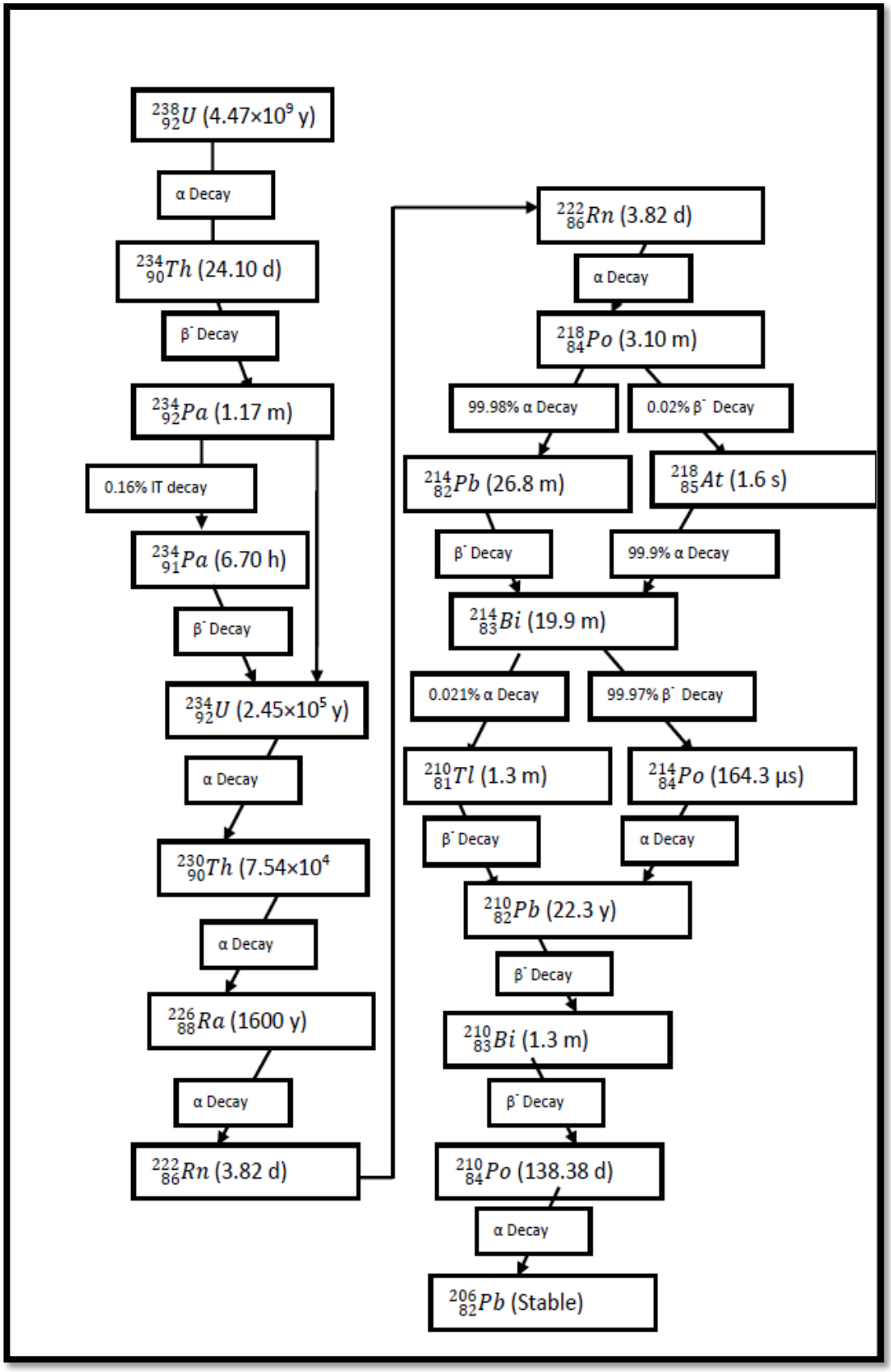

Figure 2.1: Uranium-238 decay series.

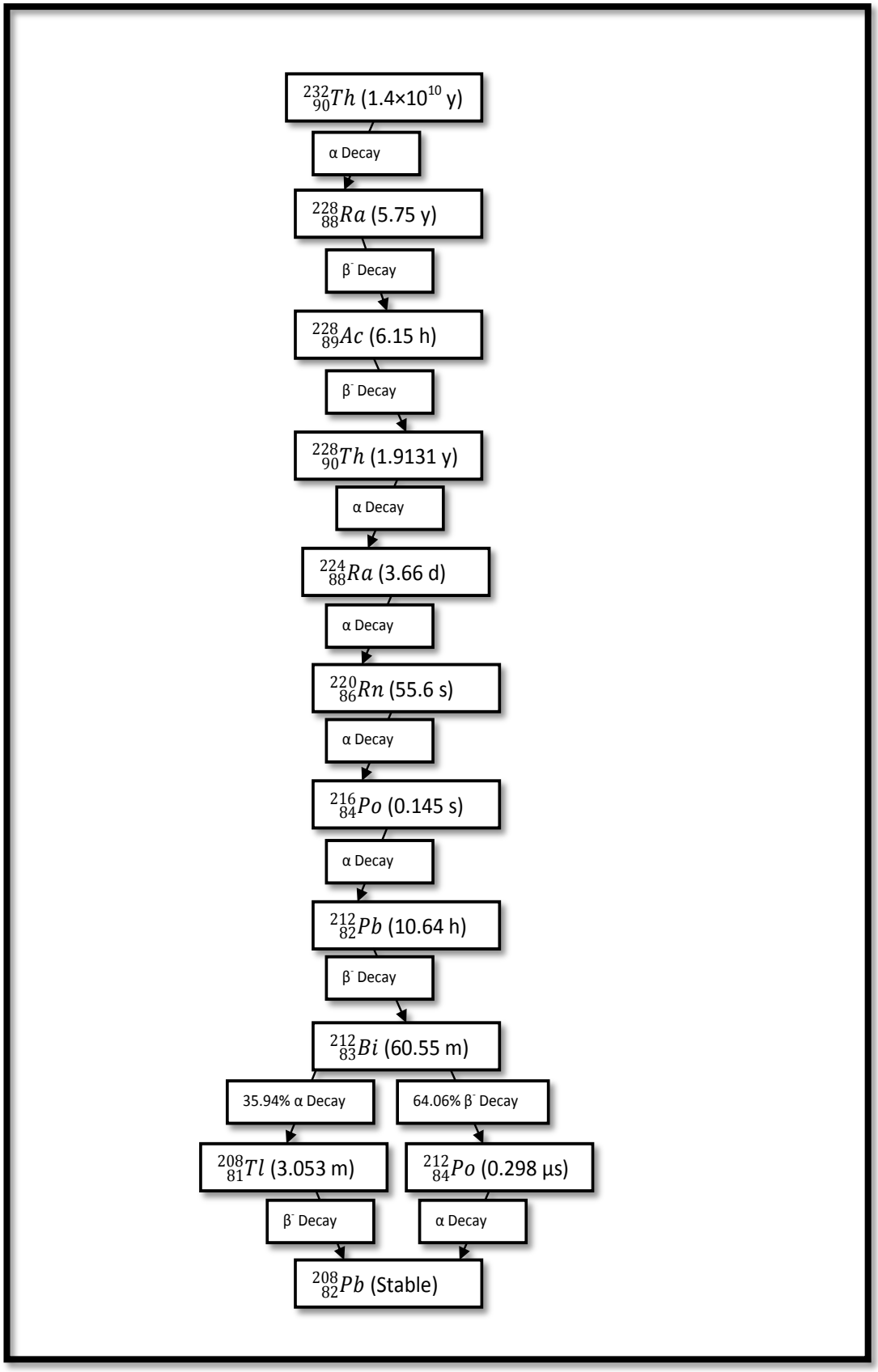

Figure 2.2: Thorium-232 decay series.

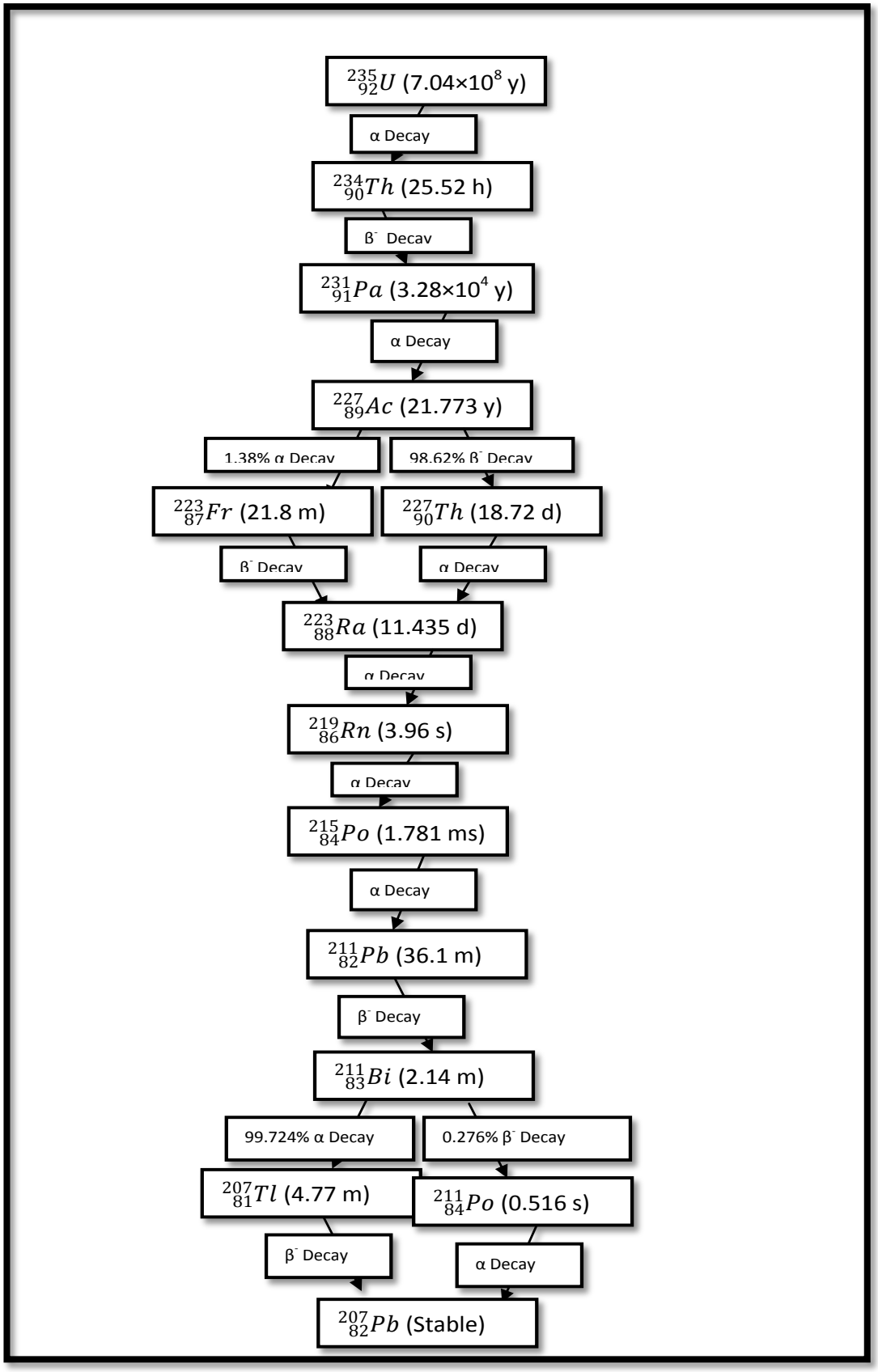

Figure 2.3: Uranium-235 decay series.

2.1.2 Radioactivity in Water

Abundance and properties of radionuclides and their behavior in the environment appear as influential factors on the ecosystem. Physical, chemical and biological mechanisms of radionuclides in the environment affect their abundance and behavior. The influence of these factors is more pronounced in aquatic environments (Buesseler et al., 2012). Most of the radionuclides released in an aquatic system are readily adsorbed on the outer surface suspended particulates. The fast rate of absorption is a result of their low water solubility that also facilitates their extraction from the water column through sedimentation process. A good example of these radionuclides is isotopes of ^{131}Ce, ^{54}Mn, ^{55}Fe, ^{57}Co and the actinides including thorium and uranium. Other elements in this category are observed to remain in solution form in water. These elements include ^{85}Sr, ^{51}Cr and ^{125}Sb. Thus, depending on the chemical properties of contaminants, these radionuclides may accumulate in water sources to levels of great concern especially in threatening human health (Health Canada, 1995). Radioactivity in natural waters is usually low, although contaminated sediments can serve as sources of radionuclides contamination. This pollution could occur even after a long period after an effective removal of dissolved radionuclides (Coetzee, Winde, & Wade, 2006).

Parent and daughter radionuclides have varying patterns of behavior, thus it will be the difference in their events in water sources. Taking an example of ground water with high levels of radium, this water tends to have low concentration levels of uranium as known that ^{238}U is the parent of ^{226}Ra. Higher levels of ^{226}Ra in water can be expected in areas containing uranium mining and milling operations. Similarly, high concentrations of the natural radionuclides are in the case of direct water contact

with rocks or soils. Generally, surface water will always register lower levels of radionuclides than underground water (Bem et al., 2014).

In studying the relationship between radon gas and water, the gas is found to emerge from rocks containing uranium and radium found in the water. Through activities such as tapping this water into houses, man introduces the gas to the environment. The gas is released through the use of the tapped water. It is estimated about 50% of the gas is released during showering and a total of 100% during the performance of cooking and washing activities. Short-lived daughters such as ^{218}Po, ^{214}Pb, and ^{214}Bi are also generated following these activities. Inhalation of the gas contributes to the exposure of the population to respiratory problems, which is far much a contributing factor than the act of drinking the contaminated water (Health Canada, 1995).

2.1.3 Airborne Radioactivity

The background radiation can originate from radioactivity carried by the ambient air, as trace amounts of radioactive gases or dust particles. The noble gas ^{222}Rn can become airborne before decaying. Research shows that soil and rocks beneath the houses contribute to the presence of the gas, which is about four to five times more concentrated than outdoor levels. Outdoor levels are less concentrated due to the frequent air dilution following the free flow of air. Building materials, outside air, use of water and natural gas contribute to the high concentration of indoor Radon. Exceptions to generalization are frequent since circumstances are found to vary with different places and time.

The ^{222}Rn decay process involves a series of short-lived daughters, two of which are ^{218}Po and ^{214}Po and are found to be alpha emitters. Different radioactive

isotopes are also generated from other series of naturally occurring radionuclides. However, these isotopes are of less radiological importance. Thorium series generates ^{220}Rn, also referred to as thoron. The parent nuclide, ^{232}Th, more abundant compared to ^{238}U, but with a longer half-life. As a result of this longer half-life, the average rate of production of ^{220}Rn in the ground is close to being similar to ^{222}Rn. However, the shorter half-life of ^{220}Rn, ($T_{1/2}$ = 56 second), as compared with ($T_{1/2}$ = 3.82 days) for ^{222}Rn, gives it a much greater chance of its decaying before being airborne. Actinium series produces ^{219}Rn, also called action after several transformations from the relatively rare origin nuclide ^{235}U. Its half-life last about 4 seconds, thus its contribution to airborne radon is insignificant. Comparatively, radioactive dust consists of either natural radionuclides or atmospheric fallout. These can easily be eliminated through filtration of the air supply system (Godish, 1989; Knoll, 2010).

2.1.3(a) Radon Emanation Phenomenon

Radon emanation refers to the process of releasing of radon atoms, emerging through alpha decay of radium-grained into pore spaces of grain. Emanation coefficient or fraction is defined as the ratio of escaped radon atoms to originating radon atoms numbers. Emanation occurs as a result of two factors; alpha recoil and diffusion. The emanation of radon as a result of recoil and diffusion in grains is influenced by factors such as the temperature of the grains, surrounding pore spaces, radiation damage, density and composition of the materials and radium distribution in grains. It is noted that the highest percentage in alpha recoil as a result of very low diffusion coefficients of radon in the solid grains (10^{-31}–10^{-69} m^2 s^{-1}) is directly influenced by the outlined factors (W. W. Nazaroff, 1992; W. W. Nazaroff & Nero,

1988). Other factors such as moisture content, atmospheric pressure, grain size, and pore size influence the emanation of radon from surrounding pore space.

Figure 2.4 illustrates the mechanism of radon emanation. Radon atoms can be released into pore spaces without the occurrence of any obstacle as shown in case A. Liquids such as water can also be existed in the way of radon atoms, which depend on their residual energies in striking a nearby grain as illustrated in case B. In addition to this, transition of radon atoms from pocket generated through their recoil path into pore spaces can also occur as in case E. The same process could occur through the escape of the gas into inner pore followed by diffusion to outer pores as presented in case F. All these previous cases can lead to presence of radon emanation phenomenon. Despite the many ways of generating radon atoms, some atoms cannot find to be released and become embedded within the grain as in case C or be transformed to outer pores as in case G. Others can be absorbed in the inner surfaces of grains as illustrated in case D. Radon emanation phenomenon disappears in pervious three cases (Sakoda & Ishimori, 2014).

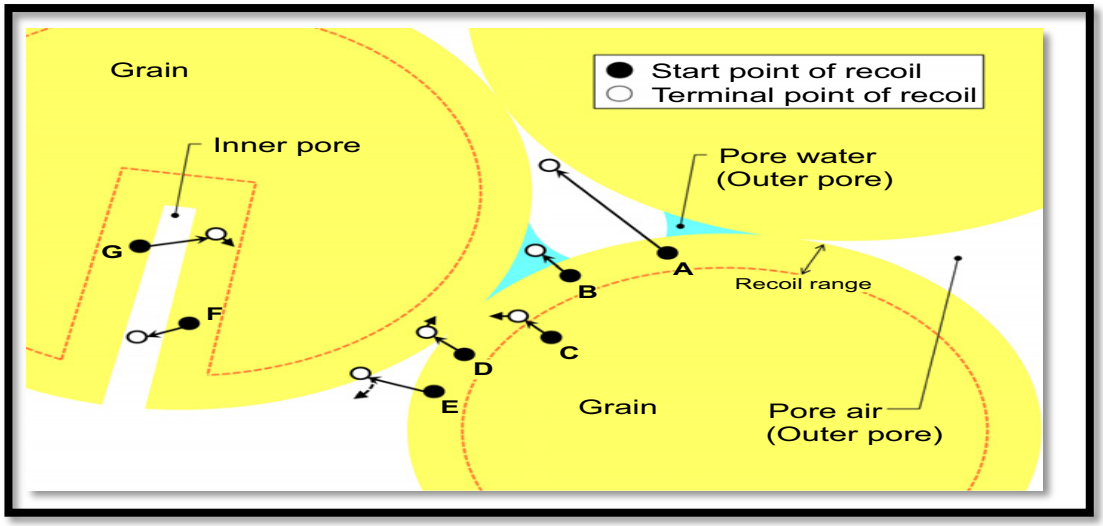

Figure 2.4: Scheme of radon emanation phenomenon.

2.1.3(b) Radon Exhalation

Radon exhalation indicates to the flow of radon from environmental sources to indoor enclosures. Environmental sources include building materials, rocks, and soils. Based on Sun, Guo & Zhuo (2004), 60.4% of indoor radon arises from the ground and soils surrounding the buildings. The depth from which radon atoms are discharged from the soil into the air relies on the nature of soil, its moisture content, and structural geology. For ^{222}Rn, the depth is regularly about (1-2) m in unsaturated soils, deeper for sands and shorter for saturated and compacted soils (Ahmad, Jaafar, & Alsaffar, 2015). Therefore, the rate of exhalation is defined as the number of atoms emerging from soil surface boundary per unit surface area in unit time. This expected exhalation rate of radon is mostly managed by atmospheric pressure, humidity, forces of wind and temperature (Sun, Guo, & Zhuo, 2004).

2.2 Exposure Pathways

The source of radiation categorizes as exposure to radiation falls into two distinct groups; external and internal exposure. External exposure results from sources outside the human body, whereas internal exposure is a result of radioisotopes deposited into the system of the exposed individual. The possibility of measuring the doses of external exposure, either directly or indirectly using available detection instruments makes it easier to deal with external exposures than internal. The challenge in determining internal doses is attributed to the assumptions made while performing calculations of the amount of radioisotope involved and its distribution within the body. Thus, the dose equivalent from internal exposure should usually be the best choice to evaluate internal doses (Knoll, 2010).

2.2.1 External Exposure

External exposure to radionuclides by humans is majorly a result of uranium-thorium series. The local concentration of these radionuclides and their decay products varies widely depending on the geologic characteristics of the region. Similarly, ^{40}K contributes to the human exposures, especially to the dose originating from internal exposures. A less significant contribution of ^{40}K occurs through the environment and thus contributing to external exposure. The doses resulting from external sources of the body are entirely a contribution of gamma rays emitted during the decaying process of radionuclides. Based on the low power of penetration of both beta and alpha particles, their emissions from the decaying natural radionuclides will not significantly contribute to the dose received externally. Consequently, only minor contributions of beta rays are received by the skin (Kirby, Downing & Gohary, 2010).

The average annual effective dose in worldwide from external terrestrial radiation both outdoors and indoors amount to a total of 0.48 mSv y^{-1} (UNSCEAR, 2000). A particular circumstance occurs in daughters of the decay of ^{222}Rn. Radon gas is diffusing into the atmosphere, decays in a non-equilibrium fashion, leading to an external exposure originating principally from lead and bismuth radionuclides. These elements are produced from the decaying process of radon decay. For a typical time-averaged outdoor radon concentration of 7.5 Bq m^{-3} (200 pCi m^{-3}), of the two Radon daughters, it is estimated that the absorbed dose rate in the air following these radon daughters would be 23.2 mrad y^{-1} (232 μGy y^{-1}) (NCRP, 1988).

The history of geological composition and the history of the area determine the variation of terrestrial radioactivity doses to human beings. Comparatively, indoor exposure from gamma rays resulting from crustal radionuclides is regulated depending on the materials used in construction. The position of individuals within the building is also a considerable factor regarding the doses received. In the last few decades, peculiar sources of human exposure have become noticeable. Some of these sources include diagnostic radiology, therapeutic radiology, use of isotopes in medicine, using fertilizers, radioactive waste, the fall-out from nuclear weapon tests, and occupational exposures from nuclear reactors and accelerators (UNSCEAR, 2000).

2.2.2 Internal Exposure

Radioactive materials gain access to the body through three main routes namely; inhalation, ingestion and through the skin to the bloodstream and lymphatic system. Transmission of radionuclides from the environment to man can occur through gaseous, food and water intake. Once inside the body, the radionuclides are absorbed, metabolized, and distributed to tissues according to their chemical properties for elements and compounds. Thus, physical and biological entities determine the ultimate biological effects of internal exposure. Physical entities include the physical properties of radionuclide (half-life), type and energy of radiation emitted; the linear energy transfer (LET), spatial distribution of radiation energy absorption and microdosimetric consideration. Comparatively, biological factors comprises of chemical properties of radionuclide, transportation of radionuclide through body, translocation from one tissue to another, the localization in target tissue or organ, transit time in body organs, excretion pathways outside the body, biological half-life and effective half-life, radiation response of tissues of

disposition and other determinants such as age, sex, pregnancy and disease that naturally increases both chance and magnitude of infection. A significant application of this is the fact that physical and biological factors can be used to calculate the absorbed doses to organs and tissues, thus assist in the construct mathematical models in the assessment of internal dose (El-Naggar, 1998).

Doses by inhalation result from the presence of dust particles in air containing radionuclides belonging to the ^{238}U and ^{232}Th decay chains. After inhaling the radon, the dominant component of exposure is the short-lived decay products of radon. Ingestion doses are mainly as an outcome of ^{40}K and to the ^{238}U and ^{232}Th series radionuclides present in foods and drinking water. The dose rate from ^{40}K can be determined directly and accurately from external measurements of its concentration in the body. To perform an analysis of the content of uranium- and thorium- series radionuclides in the body, it requires complicated chemical analyses of tissues exposed to these elements. The performance of this analysis faces the challenge of the few data available. As an alternative to dose estimation, there is the analysis of the radionuclide contents in foods and water, alongside carrying out a bioassay data guided by the knowledge of metabolic behavior (UNSCEAR, 2000).

On an average, the annual effective dose in worldwide from uranium and thorium series through inhalation exposure amounts to 6 $\mu Sv\ y^{-1}$, compared to values acquired through ingestion totaling to 0.12 $mSv\ y^{-1}$. The average annual effective dose from ^{40}K in ingestion exposure is 0.17 $mSv\ y^{-1}$. The average annual contribution from all internal sources of natural radionuclides amounts to 1.55 $mSv\ y^{-1}$. This number means that 1.15 $mSv\ y^{-1}$ of the total inhalation exposure results from radon gas and its decay products, while an estimated value of 0.1 $mSv\ y^{-1}$ results from

inhaled thoron alongside its decay products. The remainder of the total estimates is attributed to ingestion of other radionuclides found present in food and drinking water (UNSCEAR, 2000).

2.3 Research on Natural Radionuclides in Soil

Radionuclides arising in soil are distributed widely in earth's crust. The concentration of these materials in soils and depends on geological formulations and geographical conditions. This reason is behind different levels of radionuclides in the soils from various regions in Malaysia and other countries as shown in Table 2.1.

Table 2.1: Natural radioactivity in non-cultivated (NC) and cultivated (C) soil in Malaysia and other countries.

Sample location	Sample type	Methods	Results	Reference
First: Malaysia				
Kinta District, Perak	NC soil	HPGe detector	Activity concentration ranges were 12–426 Bq kg^{-1} for ^{238}U, 19–1377 Bq kg^{-1} for ^{232}Th and <19–2204 Bq kg^{-1} for ^{40}K. The world's mean values of ^{226}Ra, ^{232}Th and ^{40}K in soil are 32, 45 and 420 Bq kg^{-1}, respectively .The gamma dose rates varied from 39 to 1039 nGy h^{-1} in the District. The average of gamma dose rate was found to be 222 ± 191 nGy h^{-1}, which is double the value obtained in Malaysia and four times the value of the world average, of 59 nGy	(Lee et al., 2009; UNSCEAR, 2000).
Penang	NC soil	HPGe detector	The average activity concentrations of ^{238}U, ^{226}Ra, ^{232}Th, and ^{40}K in soil samples were obtained to be 184 ± 11, 396 ± 22, 165 ± 14, and 835 ±	(Almayahi et al., 2012a)

			28 Bq kg^{-1}, respectively. The average external gamma dose rate is 315 ± 44 nGy h^{-1}, which is more than quintuple of worldwide average	
Northern Malaysian Peninsula	NC soil	HPGe detector	The average values of activity were found to be 57 ± 2, 68 ± 4 and 427 ± 17 Bq kg^{-1}, respectively. The mean value of outdoor absorbed dose rate gained from soil was 88 nGy h^{-1}. Both mean external and internal hazard values were 0.50 and 0.65, respectively, which they were less than unity. The average of radium equivalent activity was 186 Bq kg^{-1}, which is less than the recommended values according to the guidelines of OECD (1979), whish 370 Bq kg^{-1}.	(Almayahi et al., 2012b).
Segamat District, Johor	NC soil	HPGe detector	The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K were found to be (12 ± 1 – 968 ± 27), (11 ± 1 - 1210 ± 41) and (12 ± 2 – 2450 ± 86) Bq kg^{-1}, respectively. The external gamma dose rates ranged from 25 to 1281 nGy h^{-1}. The outdoor average gamma dose rate measured 276 nGy h^{-1}. The lowest average dose rate in Sungai Segamat Mukim was found to be 115 nGy h^{-1}, while the highest average gamma dose rate was 375 nGy h^{-1} in Buloh Kasap Mukim. This value is six times the average worldwide dose rate of 59 nGy h^{-1}	(Saleh et al., 2013)
Sepang District, Selangor	C soil	HPGe detector	The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in soil sampled for the assessment	(Asaduzzaman et al., 2014)

			was valued at 92 – 142, 7.2 – 102 and 385 -1023 Bq kg^{-1}, respectively	
Muar District, Johor	NC soil	HPGe detector	The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K were found to be (6 ± 1 – 244 ± 9), (11 ± 1 – 583 ± 18) and (13 ± 6 – 830 ± 13) Bq kg^{-1} respectively. The average external gamma dose rate was 151 nGy h^{-1}.	(Saleh et al., 2014)
Sungai Petani, Kedah	NC soil & C soil	HPGe detector	On average, the radioactivity for ^{226}Ra, ^{232}Th, and ^{40}K in virgin soils were 51.06±5.83, 78.44±6.42, and 125.66±7.26 Bq kg^{-1}, respectively, while those in agricultural soils were found to be 80.63±5.78, 116.87±7.87, and 200.66±18.2 Bqkg^{-1}, respectively. The corresponding concentrations in agricultural soils were higher than those in virgin soils. The average values of outdoor and indoor absorbed dose rates in agricultural soils were 116.04 and 218.46 nGy h^{-1}, respectively.	(Ahmad et al., 2015)
Seberang Perai, Penang	C soil	HPGe detector	Concentrations of ^{226}Ra, ^{232}Th and ^{40}K in agricultural soils used for rice plant in Seberang Perai in Penang ranged 49.4 - 208.51, 68.22 - 194.13 and 138.31 - 943.11 Bq kg^{-1}, respectively	(Alsaffar et al., 2015)
Kampung Sakan, Kedah - Kampung Permatang	C soil	NaI(Tl) detector	The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in agricultural soil in Sungai Besar were found to be 7.5 – 11.3, 17.3 – 25.2 and	(Asaduzzaman et al., 2015)

Tok Labu, Pulau Pinang & Sungai Besar, Selangor			106.0 – 124.8 Bq kg^{-1} respectively. Also, The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in agricultural soil in Kampung Permatang Tok Labu were found to be 6.5 – 11.6, 12.8 – 19.6 and 78.5 – 100.5 Bq kg^{-1}, respectively. Moreover, the concentrations of ^{226}Ra, ^{232}Th and ^{40}K in agricultural soil in Kampung Sakan were found to be 5.1 – 9.2, 9.6 – 13.5 and 70.5 – 81.7 Bq kg^{-1}, respectively.	
Langkawi Island, Kedah-Tumpat Kelantan, Pasir Panjang Negeri Sembilan, Klang Selangor & Cameron Highlands, Pahang	C soil	HPGe detector	^{226}Ra, ^{232}Th and ^{40}K concentrations ranged 1.33 – 30.90, 0.48 – 26.80 and 7.99–136.5 Bq kg^{-1}, respectively.	(Khandaker et al., 2016)
Seberang Perai, Penang	NC soil & C soil	HPGe detector	The average concentrations activity of ^{226}Ra, ^{232}Th and ^{40}K in cultivated soils were evaluated to be 85.01 ± 42.14, 59.09 ± 22.75 and 384.86 ± 216.28 Bq kg^{-1}, while in non-cultivated soil were found to be 54.21 ± 34.15, 55.19 ± 47.22 and 276.87 ± 203.43 Bq kg^{-1}, respectively.	This Study