NATURAL RADIOACTIVITY AND RADON CONCENTRATION IN TAPIOCA AND SWEET POTATO IN KEDAH AND PULAU PINANG: TRANSFER FACTOR AND RADIOLOGICAL RISKS

ALMUTAIRI ABDULMAJEED MUIDH S

UNIVERSITI SAINS MALAYSIA

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ALMUTAIRI ABDULMAJEED MUIDH S

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LIST OF SYMBOLS

A	Sample surface area (used for CR-39)
AED_{in}	Indoor annual effective dose
AED_{ingest}	Annual effective dose for ingestion
AED_{out}	Outdoor annual effective dose
A_i	Initial activity of source
A_o	Area of field of view
A_s	Specific activity of radionuclide
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_{ex}	External hazard index
H_{in}	Internal 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)
ε	Efficiency of HPGe detector at gamma-ray line
η	Absolute full energy detection efficiency
κ	Porosity of soil
λ	Decay constant of radionuclide
λ_{Rn}	Decay constant of radon
ω	Back diffusion constant for soil

LIST OF ABBREVIATIONS

CRM	Continuous Radon Monitor
DOE	Department of Environment
EC	European Commission
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

KERADIOAKTIFAN TABII DAN KEPEKATAN RADON DALAM UBI KAYU DAN UBI KELEDEK DI KEDAH DAN PULAU PINANG: FAKTOR PEMINDAHAN DAN RISIKO RADIOLOGI

ABSTRAK

Tanih merupakan sumber takungan utama radionuklid semula jadi yang boleh dipindahkan kepada manusia melalui rantai makanan. Dos yang di akibatkan oleh pengambilan radionuklid melalui makanan bergantung kepada penyerapan radionuklid melalui aktiviti pertanian. Dalam kajian ini, kepekatan aktiviti semula jadi radionuklid ^{226}Ra, ^{232}Th, ^{40}K bagi tanah, ubi kayu, ubi keledek, dan baja dari Pulau Pinang dan Kedah telah dikaji. Pengukuran kepekatan radioaktiviti dilakukan dengan menggunakan Pengesan Germanium Berketulenan Tinggi (HPGe). Faktor pemindahan radionuklid dari tanih ke sayuran rizom juga ditentukan. Kepekatan aktiviti ^{226}Ra, ^{232}Th dan ^{40}K bagi sampel tanih dari Kedah masing-masing adalah 92.46 ± 46.88, 65.59 ± 11.43 dan 583.11 ± 121.89 Bqkg^{-1}. Kepekatan aktiviti bagi sampel tanih dari Pulau Pinang masing-masing adalah 80.87 ± 9.22, 57.60 ± 4.41 dan 512.09 ± 113.24 Bqkg^{-1}. Purata kepekatan radionuklid ^{226}Ra, ^{232}Th dan ^{40}K adalah tinggi bagi sampel tanih dari Kedah berbanding sampel tanih pertanian Pulau Pinang. Nilai yang ditentukur telah dibandingkan dengan dapatan penyelidik lain. Kepekatan aktiviti ^{226}Ra, ^{232}Th dan ^{40}K bagi ubi kayu dan ubi keledek dari Kedah masing-masing adalah 2.61 ± 0.44, 6.62 ± 1.78 dan 162.82 ± 46.33 Bqkg^{-1}. Kepekatan aktiviti bagi sampel ubi kayu dan ubi keledek dari Pulau Pinang masing masing adalah 1.57 ± 0.09, 5.29 ± 1.23 dan 134.71 ± 29.06 Bqkg^{-1}. Faktor pemindahan radionuklid semula jadi ^{226}Ra, ^{232}Th dan ^{40}K dari tanah ke sayuran rizom ditentukur dan masing-masing berada dalam julat 0.017-0.055, 0.063-0.165 dan Tanih

merupakan sumber takungan utama radionuklid semula jadi yang boleh dipindahkan kepada manusia melalui rantai makanan. Dos yang di akibatkan oleh pengambilan radionuklid melalui makanan bergantung kepada penyerapan radionuklid melalui aktiviti pertanian. Dalam kajian ini, kepekatan aktiviti semula jadi radionuklid ^{226}Ra, ^{232}Th, ^{40}K bagi tanah, ubi kayu, ubi keledek, dan baja dari Pulau Pinang dan Kedah telah dikaji. Pengukuran kepekatan radioaktiviti dilakukan dengan menggunakan Pengesan Germanium Berketulenan Tinggi (HPGe). Faktor pemindahan radionuklid dari tanih ke sayuran rizom juga ditentukan. Kepekatan aktiviti ^{226}Ra, ^{232}Th dan ^{40}K bagi sampel tanih dari Kedah masing-masing adalah 92.46 ± 46.88, 65.59 ± 11.43 dan 583.11 ± 121.89 Bqkg^{-1}. Kepekatan aktiviti bagi sampel tanih dari Pulau Pinang masing-masing adalah 80.87 ± 9.22, 57.60 ± 4.41 dan 512.09 ± 113.24 Bqkg^{-1}. Purata kepekatan radionuklid ^{226}Ra, ^{232}Th dan ^{40}K adalah tinggi bagi sampel tanih dari Kedah berbanding sampel tanih pertanian Pulau Pinang. Nilai yang ditentukur telah dibandingkan dengan dapatan penyelidik lain. Kepekatan aktiviti ^{226}Ra, ^{232}Th dan ^{40}K bagi ubi kayu dan ubi keledek dari Kedah masing-masing adalah 2.61 ± 0.44, 6.62 ± 1.78 dan 162.82 ± 46.33 Bqkg^{-1}. Kepekatan aktiviti bagi sampel ubi kayu dan ubi keledek dari Pulau Pinang masing masing adalah 1.57 ± 0.09, 5.29 ± 1.23 dan 134.71 ± 29.06 Bqkg^{-1}. Faktor pemindahan radionuklid semula jadi ^{226}Ra, ^{232}Th dan ^{40}K dari tanah ke sayuran rizom ditentukur dan masing-masing berada dalam julat 0.017-0.055, 0.063-0.165 dan sampel dari Kedah dan Pulau Pinang. Hasil kajian juga memberi pengetahuan baru berkenaan faktor pemindahan kepekatan radionuklid bagi sampel tanih, ubi keledek, ubi kayu, dan baja dari Kedah dan Pulau Pinang. Berdasarkan kajian penyelidikan ini, adalah disarankan agar tanih yang hendak digunakan dalam kegiatan pertanian haruslah terdiri dari tanih dengan indeks bahaya luaran (Hex) kurang dari 1.

NATURAL RADIOACTIVITY AND RADON CONCENTRATION IN TAPIOCCA AND SWEET POTATO IN KEDAH AND PULAU PINANG: TRANSFER FACTOR AND RADIOLOGICAL RISKS

ABSTRACT

Soil is the principal reservoir of natural radionuclides that can be transmitted via the food chain to humans. In this work, the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in soil, tapioca and sweet potato, and fertilizer from Penang and Kedah were studied. Measurement of the activity concentrations was made using high purity germanium detector (HPGe). The transfer factors of the radionuclides were determined from soil to root vegetables. The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in soil samples from Kedah are 92.46 ± 46.88, 65.59 ± 11.43 and 583.11 ±121.89 Bq kg^{-1}, respectively. The corresponding activity concentration of the samples from Penang are 80.87 ± 9.22, 57.60 ± 4.41 and 512.09 ±113.24 Bqkg^{-1}. The average radionuclide concentrations were significantly higher in Kedah soil than in Penang. Measured values were compared with studies worldwide. The activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in root tapioca and sweet potato from Kedah are 2.61 ± 0.44, 6.62 ± 1.78 and 162.82 ±46.33 Bqkg^{-1}, respectively. The corresponding concentration from Penang are 1.57 ± 0.09, 5.29 ± 1.23 and 134.71 ±29.06 Bq kg^{-1}. The transfer factors of ^{226}Ra, ^{232}Th and ^{40}k from the soil to root vegetables are in the range of 0.017–0.055, 0.063–0.165 and 0.137–0.500, respectively. In the tapioca and sweet potato samples, the largest dose contribution was found to be from ^{232}Th (~48%), followed by ^{40}K (33%) and the lowest was due to ^{226}Ra (19%). The annual effective dose of tapioca and sweet potato are 3793 and 4504 nSv, respectively. These are well below the average total ingestion dose

reported by UNSCEAR 2000 at 290 μSv. Concentration and exhalation levels for radon gas ^{222}Rn were measured for the soil samples. Continuous radon monitor (CRM) and nuclear track detectors (CR-39 NTD) were used in the measurements. Radon concentrations were found to be 31 - 821.53 Bqm^{-3}. The soil exhalation rates were below the safety limit of 57.6 $Bqm^{-2}h^{-1}$. The results show the impact of natural radionuclides in soil, root vegetables, and fertilizer have on air radon concentrations. This study also contributes new knowledge on the transfer factor of natural radionuclides from soil to tapioca and sweet potato. Based on the results, it is recommended that soils used in farming activities should be chosen when the external hazard index (H_{ex}) is less than 1.

CHAPTER 1

INTRODUCTION

1.1 Background

Radioactive materials exist in the atmosphere naturally and are part of our everyday life (IAEA, 1999; UNSCEAR, 2000; UNSCEAR, 2012). They are present in the atmosphere and generally in all living and non-living matter, including the human body (Hunter-Smith, 2012). Naturally occurring radionuclides materials (NORM) are known to be the major causes of public exposure. These come from two main sources: the incident of cosmic rays in the atmosphere (cosmogenic origin) and the crust of the earth (terrestrial origin) (Al-Sulaiti, 2011; Canbazoğlu and Doğru, 2013; UNSCEAR, 2000). The terrestrial radionuclides are either from naturally occurring series, such as ^{238}U and ^{232}Th, or from the radioactive isotope ^{40}K, and these are usually long-lived radionuclides (half-lives in the order of 100 million years). Cosmogenic radiation is formed by spalling the atoms in the atmosphere. Some of the radionuclides formed due to cosmic rays have long half-lives, but the majority have shorter half-lives than those that originated from the earth crust. These nuclides with cosmic origin are ultimately deposited in the soil through dry or wet deposition processes. Fertilizers are also known to contain a considerable amount of natural radioactivity (IAEA, 2006). Therefore, agricultural land is a depository for various radionuclides which can be transferred to humans through the consumption of plant cultivated in these soils, thus posing a health risk to humans.

The transfer of radionuclides into plants mainly takes place through two pathways. The first is through the root-uptake of these radionuclides directly from the soil. Uptake of radionuclides by the roots occurs either through passive or active

transfer mechanism since some elements are selectively taken up by the roots, e.g., potassium (and hence ^{40}K), and the use of high potassium content fertilizer will add to this uptake by the plants through the roots. It is typical to characterise their transfer to the plants through transfer factors (TF) that are both radionuclide and plant-dependent. Afterwards, consumption of these plants can lead to an effective internal dose that poses health risks (Chambers, 2015). The other pathway is the direct deposit of radionuclides on the leaves through either wet or dry deposition. This is of particular importance for leafy vegetables. Either one or both pathways are of varying importance to radionuclide accumulation in plants depending on the type of the plant and the geological nature of the land. Ultimately, humans consume these plants, leading to the accumulation of various radionuclides in different organs depending on the radionuclide. In addition to the risk of consuming food containing NORM, the inhalation of radionuclides poses another external health risk to humans. The main radionuclide that is responsible for an internal dose from inhalation is radon.

Different monitoring and regulatory agencies have realised the potential danger of exposure to or ingestion of NORM and thus have put forward certain recommendations and regulations regarding public and occupational exposure to NORM as well as other radionuclides. The International Commission on Radiological Protection (ICRP) is the principal international organisation concerned with radiation protection. It is an advisory organization, and its recommendations have significant influence on the development of radiation protection standards in many nations. The latest publication of the ICRP regarding NORM was published in 2019 (ICRP, 2019). In addition to the ICRP, there are other important international organisations such as the International Atomic Energy Agency (IAEA) and the

Commission of the European Communities (CEC). One of the IAEA's objectives is to establish radiation protection standards, and it has published a series on radiation safety standards for the protection of the people and the environment, the latest published in 2018 (IAEA 2018a, IAEA 2018b, IAEA 2018c). An effective dose of 1 mSv in a year is the current reference level for public exposure in specific existing exposure situations, e.g., exposure to radionuclides in commodities such as food or drinking water. The CEC is aimed at member states of the European Community, and the current radiation protection standards developed by the CEC are based mainly on the ICRP recommendations. Another important monitoring agency is the UNSCEAR which is not a regulatory agency but collects and reviews many results from research worldwide on the effects of ionising radiation on humans and the environment. In addition to these international agencies, some regional organisations of importance need to be mentioned, such as the Environmental Protection Agency (EPA) and the National Council on Radiation Protection and Measurements (NCRP) in the United States.

The level of NORM in soil and food have been investigated thoroughly in recent years, and the values reported for their levels have widely varied. For example, the reported values for the activity concentrations of the members of the decay series (^{238}U, ^{226}R and ^{232}Th) in root vegetables and fruits (which are typically grouped together) worldwide were (in mBq/kg) in the ranges of 0.4–900, 5–400 and 0.08–7.1, respectively (UNSCEAR, 2000). The corresponding values of the activity concentrations for the radionuclides ^{238}U, ^{226}R and ^{232}Th reported in Asia were in the ranges of (in mBq/kg) 0.4–77, 11–63 and 2.3–4.7, respectively. The recommended reference values for the concentrations of these radionuclides are 1, 5 and 5 mBq/kg. In leafy vegetables, the recommended values are much higher, and the values for the

activity concentrations for the radionuclides (in the same order) are 20, 50 and 15 mBq/kg. With regard to the activity concentration in the soil, and the worldwide averages are 400 Bq/kg for ^{40}K, 35 Bq/kg for ^{238}U and 30 Bq/kg for ^{232}Th. It is necessary to point out here that the activity concentration in soil generally varies over a wide range even in the same region depending on the type of soil and the geological nature of the crust. Radon is considered the main contributor to exposure to natural radioactivity. The primary pathway for radon exposure is through inhalations. The worldwide average of external inhalation exposure is 1.26 mSv (UNSCEAR, 2000), of which 1.15 mSv is due to radon.

It should be noted that indoor exposure due to radon makes most of the inhalation exposure. The World Health Organization (WHO) recommends a national reference level of 100 Bq/m^3 for radon in dwellings and that the level should not exceed the maximum value of 300 Bq/m^3, which gives an annual effective dose of 10 mSv (ICRP, 2011). Radon levels worldwide vary considerably, and a comparison of various measurements will be made later in the thesis. However, it should be noted that radon levels exceeding the recommended values have been reported in many regions around the world, including Malaysia. It should also be noted that radon levels show seasonal variation. The annual effective dose can rise from different pathways: external exposure, inhalation or food ingestion.

The dose resulting from inhalation is dependent upon the content of dust particles in the air which contain the radionuclides of ^{238}U and ^{232}Th series as well as ^{40}K (Misdaq et al., 2001). Globally, the average annual committed effective dose from inhalation of both the Uranium and Thorium series to be (in μSv) is 5.0, 6.0 and 5.8 for infants, children and adults, respectively. The corresponding values resulting from ingestion are 260, 200 and 110 μSv, which clearly shows that the contribution

of the uranium and thorium series is predominately due to ingestion. In addition to the dose from the decay series, ^{40}K contributes 110 µSv to the total. The total effective dose due to ingestion is 240 µSv, which is considered safe for human health.

In Malaysia, NORM has been studied by different researchers. Table 1.1 below summarises the locations, samples investigated, and the methods used in the investigation.

Table 1.1 Summary of NORM studies in Malaysia

Location	Sample	Method of investigation	Reference
Penang	Soil	Gamma-ray spectroscopy	(Almayahi, 2012)
Pontian (Johor)	Soil	Gamma-ray spectroscopy	(Saleh, 2013c)
Cameron Highlands and Penang	Soil and vegetable	NAA	(Aswood, 2013)
North and West Peninsular, Malaysia	Soil and rice	Nal (TI)	(Asaduzzaman, 2015)
Sungai Petani (Kedah)	Soil	Gamma-ray spectroscopy	(Ahmad et al., 2015)
Sekinchan, Selangor	Soil and vegetable	Gamma-ray spectroscopy	(Solehah, 2016)
Malaysia, Selangor	Soil and vegetable	Gamma-ray spectroscopy	(Priharti, 2016)
Northern Peninsular Malaysia	Soil	CRM	(Almayahi et al., 2013)
Kedah, Malaysia	Soil	CR-39	(Ahmad et al., 2014).

In Malaysia, tapioca (Manihot esculenta also known as cassava or manioc) and sweet potato (Ipomoea batatas also known as Dioscorea species) are the fourth and fifth most consumed crops after rice, wheat and corn. Tapioca is used as a nutritional supplement and is essential for biofuel, animal feed and chemicals . In the year 2011, an estimated area of around 2230 ha with a potential yield of about 26,600

tones was allocated for sweet potato plantation in Malaysia. In the same year, a total area of 2596 ha was devoted to tapioca production with a potential yield of 33,200 metric tons (Department of Agriculture Peninsular Malaysia, 2011). In the last few years, several researchers have investigated various aspects of radioactivity in soil and plants in Malaysia. Many past studies have shown significant NORM of crops and plantation samples taken from the northern peninsular Malaysia, therefore this study is significant to further investigate NORM activity in other major crops planted in the northern peninsular Malaysia.

In this thesis, a research study was conducted to determine radioactivity concentrations of NORM in the root vegetables of tapioca and sweet potato, agricultural soil and the fertilizers used, in Penang and Kedah. The transfer factors of radioactivity from soil to plant was calculated, and the effective internal dose resulting from the ingestion (based on food consumption data) of NORM was assessed. In addition, an assessment of the health risk to farmers due to inhalation of the radon gas was performed. This study focuses on radiation risk due to NORM resulting from the ingestion (from available data) of sweet potato and tapioca in Kedah and Penang in which the contribution from fertilizers was taken into account, the transfer of radionuclides from soil to plant was calculated and the radon concentrations in farms were evaluated.

1.2 Problem Statements

Exposure to NORM is one of the main sources of radiation exposure for human beings. It is also hazardous to the environment and contributes to higher background dose. Moreover, NORM is a hazard to humans and results in effective internal dose due to ingestion of food containing NORM. Therefore, it is significant

to assess the level of NORM in food. An intensive study of these radionuclides in both soil and food will provide a better understanding of the protection of human beings as well as the environment.

Regulation bodies focused mainly on NORM levels produced by specific industries (e.g., mining). However, there has been a growing interest in NORM from non-industrial activity lately. There are few data concerning the level of these radionuclides in the root vegetables from Kedah and Penang. Therefore, this research is important to provide a better understanding of non-industrial NORM in this region.

In this work, the study of concentrations of NORM in vegetables that form an essential part of the Malaysian population's diet was performed as well as in agricultural soil and fertilizer. Thus, the transfer factor was calculated, and the concentrations of the detected radionuclides were compared to worldwide accepted values. Therefore, research will give an overview of exposure through food in Malaysia.

This research addresses the following questions:

i. What is the level of NORM in soil, sweet potato, tapioca and fertilizer?

ii. What is the level of radon concentrations in sweet potato and tapioca agricultural soil in Penang and Kedah?

iii. How are the radionuclides transferred from soil to sweet potato and tapioca?

iv. How do the radiological hazard indices and external and internal doses are compared to acceptable values set by the radiation protection agencies and with the values reported worldwide?

The measurement of levels of NORM in the soil will be used to assess the external radiation exposure to the farmers working in the agricultural land, while the measurement of NORM in root vegetables will be used to assess the internal radiation dose due to ingestion of these vegetables. Based on the measured values and the calculated doses, the health risk of population is identified.

The areas under investigation in the present work are Kedah and Penang, of which have not been investigated before based on the parameters aimed in the present work. In this study, radon concentration was measured by two different techniques along with the measurement of gamma spectroscopy.

1.3 Objectives of the Study

This research aims to assess the radiological risk of natural radionuclides in plantation farms in two states in Malaysia, Penang and Kedah. In addition radon concentration will be measured because radon is the second leading cause of lung cancer worldwide. The studied samples include root vegetables, agricultural soil and fertilizers. The aim can be achieved by executing the following objectives:

1. to measure the radionuclide activity concentrations in tapioca, sweet potato, soil and fertilizer samples.
2. to calculate the transfer factor for natural radionuclides from agricultural soil to tapioca and sweet potato.

3. to measure the concentrations and exhalation rates of radon gas ^{222}Rn from agricultural soil .

4. to study radiological hazard indices, radium equivalent activity, annual outdoor effective dose equivalent, exposure rate and internal and external hazard indices of tapioca, sweet potato, soil and fertilizer.

1.4 Scope of Research

The research is focused on evaluating the level of radiological hazards related to the natural radioactivity in root vegetables, soil and fertilizer in tapioca and sweet potato farms in Penang and Kedah, Malaysia. Radioactivity is present in different parts of nature, requiring an extensive study of the radiation levels and assessing the dose acquired by population. Naturally occurring radiation and environmental radioactivity were studied in different worldwide countries (UNSCEAR, 2000). This research is dedicated to evaluating the rate of the public dose and the performance of epidemiological studies. This work is significant as it establishes baseline data from the levels of natural radiation in Malaysia. The study uses a CRM and a CR-39 detector to analyse ^{222}Rn concentrations in different soil samples and its exhalation rate. The study also uses Hyper Purity Germanium (HPGe) gamma-ray spectroscopy to measure the activity concentrations of the natural radionuclides^{226}Ra, ^{232}Th and ^{40}K in soil and root vegetable (tapioca and sweet potato) samples. The current study is different than other similar by adopting a comprehensive approach of assessing the radiological risk and calculating the transfer factors for the same regions the difference is in the comprehensive approach of risk assessment carried out for the area Kedah and Penang and the calculation of transfer factors for the same regions.

1.5 Thesis Outline

This thesis consists of five chapters. Chapter 1 provides a general introduction to natural radionuclides, the description of problems and the goals. At the end of Chapter 1, there is the research framework and a thesis outline. Chapter 2 offers a list of sources of background radiation, radon emanation and exhalation and exposure mechanisms as well as a literature review of natural radionuclides in root vegetables, soils and fertilizers and soil radon.

A summary of the field of analysis, locations and sample preparation, measurement of natural radionuclides using HPGe in root vegetables, soil and fertilizer, measurement of radon concentration in soil using CRM and CR-39 detectors is discussed in detail in Chapter 3. Chapter 4 includes a description and explanation of the findings. Finally, Chapter 5 covers the conclusion and future work related to the research.

CHAPTER 2

THEORY AND LITERATURE REVIEW

In this chapter, extensive review on NORM, radon, radionuclides pathway to human body and relevant past works concerning this research are deliberated.

2.1 Environmental Radiation

The world is naturally radioactive and its formation is due to radioactive materials that occur naturally (NORM) present in the environment; Existence on earth has, therefore, settled under the pervasive with the existence of ambient radiation to which all living things belong constantly exposed (Ahmad et al., 2015). Radioactivity of natural origin is commonly dispersed in the atmosphere and can be present in varying amounts in environmental variety materials, such as rock, soil, plant and food, air, water and construction materials (Abojassim et al., 2014; Kant et al., 2015). Various biochemical pathways and behaviour of some individuals move these distinct radionuclides geological medium to the biosphere. They are in cumulated bio in the chain of food and function as the key path of human exposure. People are constantly subjected to radioactivity in the atmosphere, which can be divided into two major sources: natural and anthropogenic (human-made) sources. Standard radioactivity sources derive mainly relative half-lives of long-lived terrestrial radionuclides in relative terms to the earth and widely dispersed in the crust and extra-terrestrial roots of the earth originate from the bombing of cosmic beams. (NCRP, 1975; Lilley, 2001; UNSCEAR, 2000). In addition to natural sources, human operations are the use of radiation and radioactive materials involved (e.g., applications for industrial and medical purposes) radionuclides can be released

into the atmosphere and radioactivity in nature can be increased. (Eisenbud and Gesell, 1997; UNSCEAR, 2010).

2.1.1 Natural radionuclides

It is possible to separate natural radionuclides into two groups: cosmogenic and terrestrial, depending on their origin. Since the long half-lives of such terrestrial radionuclides are long (one hundred million or more years), a significant some of these radionuclides still exist, today. Such radionuclides are normal in rock, soil, food and water, and the ocean (UNSCEAR, 2010). Once again, there are two groups of terrestrial radionuclides, namely primordial (non-series) and decay chain (series) radionuclides (Eisenbud and Gesell, 1997; UNSCEAR, 2010).

2.1.1(a) Primordial radionuclides

Several single primary radionuclides were observed (non-series) naturally and decay directly into stable nuclides. Most of them are long half-life radioactive isotopes of (10^{10}-10^{15} years), equivalent to the earth's Age, Low and substantially low abundance of isotopes, resulting in negligibly fewer activities and generally not considered to be significant in relation to human exposure to radiation (Lilley, 2001). Radioactive isotopes ^{40}K and ^{87}Rb are however, important normal radioactivity sources. Some of them decay due to beta and alpha emissions, while others follow electron capture (NCRP, 1975; Eisenbud and Gesell, 1997). ^{40}K is commonly dispersed in the surface of the earth and is the dominant in natural radionuclides food and tissues of human beings. It is also an important source of internal radiation exposure due to its high-energy beta emissions, but the human body is

homeostatically controlled by keeping the ^{40}K the dosage constant inside the man's body (Eisenbud and Gesell, 1997; UNSCEAR, 2000).

2.1.1(b) Decay chains (series) radionuclides

Natural decay radionuclides series (^{238}U, ^{232}Th and ^{235}U) and their subsequent radioactive degradation progeny and radionuclide non-series ^{40}K are the most frequently found radionuclides in nearly all products for the environment, including soil, rock and food, the dosage of exposure arising mainly in drinking water from these radionuclides and food alpha and beta particles that irradiate the different organs internally (Awudu et al., 2011; Canbazoğlu and Doğru, 2013; IAEA, 2003; UNSCEAR, 2000). The three-decay series of radionuclides (^{238}U, ^{232}Th and ^{235}U) with a half-life relative to the earth's Age it could still be present, in the environment that forms the main part of our radiation from nature (NCRP, 1975; Krane, 1988; Lilley, 2001).

2.1.2 Man-made radionuclides

Artificial sources are deposited into the environment by human activities which are typically time-dependent, along with nuclear activities and other influences. Such activities include but are not limited to, nuclear weapons research, nuclear power plants, mining (especially uranium) and oil exploration. Artificial sources can also be used for a number of applications: medical, commercial, research or agricultural (Eisenbud and Gesell, 1997; UNSCEAR, 1988).

2.2 NORM

2.2.1 Uranium

The atomic group has a dense grey-silver metallic element known as Uranium, with an atomic number of 92 and an atomic mass of 238.03(U). Natural background radiation is of low intensity in the environment and is caused by the degradation of Uranium materials. It has been noted that the abundance Uranium in the crust of the earth is not as scarce as expected, with an average concentration of 2.7 ppm, which is currently more than mercury, silver or cadmium. Minerals such as Pitchblende, Uraninite, Carnotite and Autunite contain Uranium as well. It can also be derived commercially from lignite, Sands of phosphate rock and monazite.

The three radioactive isotopes of natural Uranium are ^{238}U, ^{235}U and ^{234}U, with respective abundances of 99.27%, 0.72 % and 0.0055%, respectively. The corresponding half-lives are : 4.47×10^9, 7.04×10^8 and 2.46×10^5 years. Due to its lengthy half-life, Uranium is extremely radiotoxic. Natural Uranium has a specifc activity of 25 Bq mg^{-1} (Aswood, 2013). ^{238}U decays to ^{234}Th and eventually to stable lead (^{206}Pb) by a series of decays. This decay chain involves 14 nuclear decay steps resulting in the emission of eight α-particles and six β-particles. The ^{238}U and ^{234}U are part of a family called the uranium series . The isotope ^{235}U belongs to another series called the actiniums series (Cember and Johnson, 2009). Decay schemes for primordial ^{238}U and ^{235}U are shown in Figure 2.1 and Figure 2.2, respectively (Poschl and Nollet, 2006). Natural isotopes that are products of decay of the ^{238}U and ^{232}Th radioactive series that ecological system can also be affected by ^{226}Ra with a 1600 years' half-life and ^{228}Ra with a half-life of 5.7 years. The early discovery of radium is closely linked to primary radioactivity research; in addition, both radium isotopes

with the atomic number 88 are radioactive. Due to high radiotoxicity, radium is hazardous to the environment, although it is present in very small quantities. ^{226}Ra and ^{238}U are generally balanced in undisturbed natural soil and may not be stable in disturbed soils. In undisturbed environments, the formation of natural radionuclides is in the form of secular equilibrium (the parent and daughter activities are equal). The secular radiological balance of each series may be unstable due to the fact that physicochemical patterns in crust of the earth, such as emanation and leaching (Cember and Johnson, 2009).

2.2.2 Thorium

Naturally and/or artificially, there are 25 radioisotopes of thorium, Th.The characteristics of thorium isotopes can differ from seconds to 10^{10} years as well as their half-lives. The atomic number of these isotopes is 90 and their atomic mass ranges from 212 to 236 and in all redox situations, the constant oxidation state is (+4) in natural water (Santschi et al., 2006). There are six thorium isotopes, naturally, with β-emitting ^{234}Th (24.10 days) and a-emitting ^{230}Th (7.54 x 10^4 years) Uranium (^{238}U) series; α-emitting ^{232}Th, (1.41 x 10^{10} years) and ^{228}Th (1.913 years) thorium (^{232}Th) series; and β-emitting ^{231}Th (25.52 h) and α-emitting ^{227}Th (18.72 days) in ^{235}U series.

^{232}Th has a relatively higher natural abundance or concentration, longer half-lives, of activity, it is more significant (Jia et al., 2008). Because of its degradation, a variety of other α-, β-, and/or γ-emitting progeny One of the extremely radiotoxic elements is known as ^{232}Th (Talip et al., 2009). ^{232}Th is the first member of the thorium series in a long series (4n) (Cember and Johnson, 2009). Decay series of Thorium (^{232}Th) have been shown in Figure 2.3.

In crust of the world, thorium in general, distribution is distributed at a small and average lithospheric concentration of 8 - 12 mg g^{-1}, at an average concentration of 6 mg g^{-1} in soil. Therefore, in quantity, it is approximately twice as much as Uranium (Jia et al., 2008). Its concentration in sedimentary rocks is just a few parts per million (ppm), although it can be ten times higher in acid igneous rocks (Aswood, 2013).

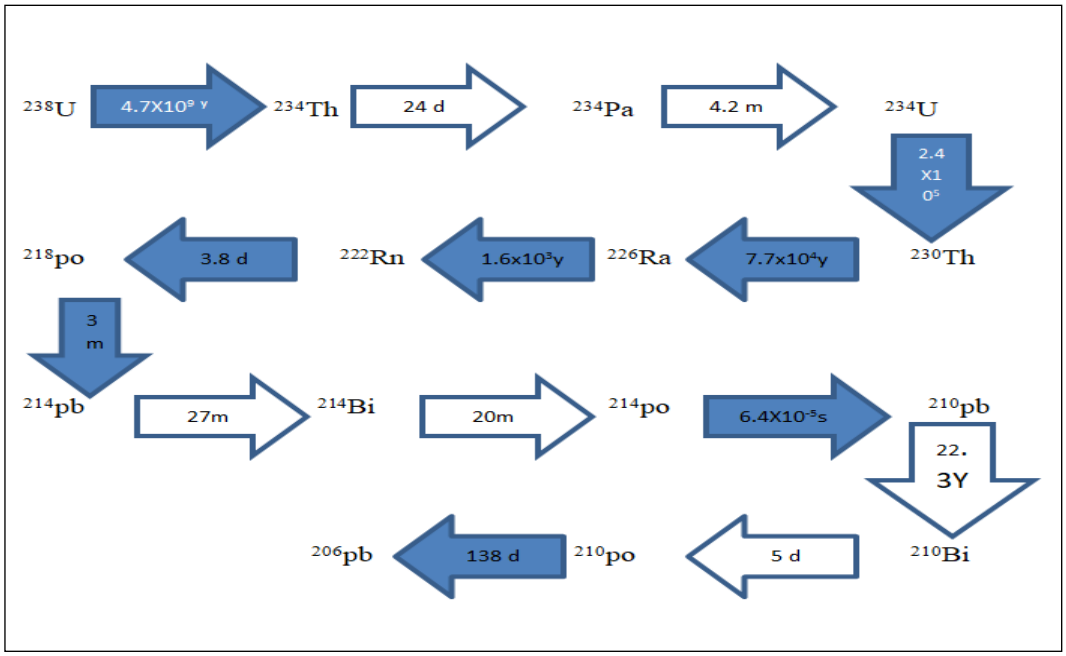

Figure 2.1 Uranium (^{238}U)decay series (4n+2)

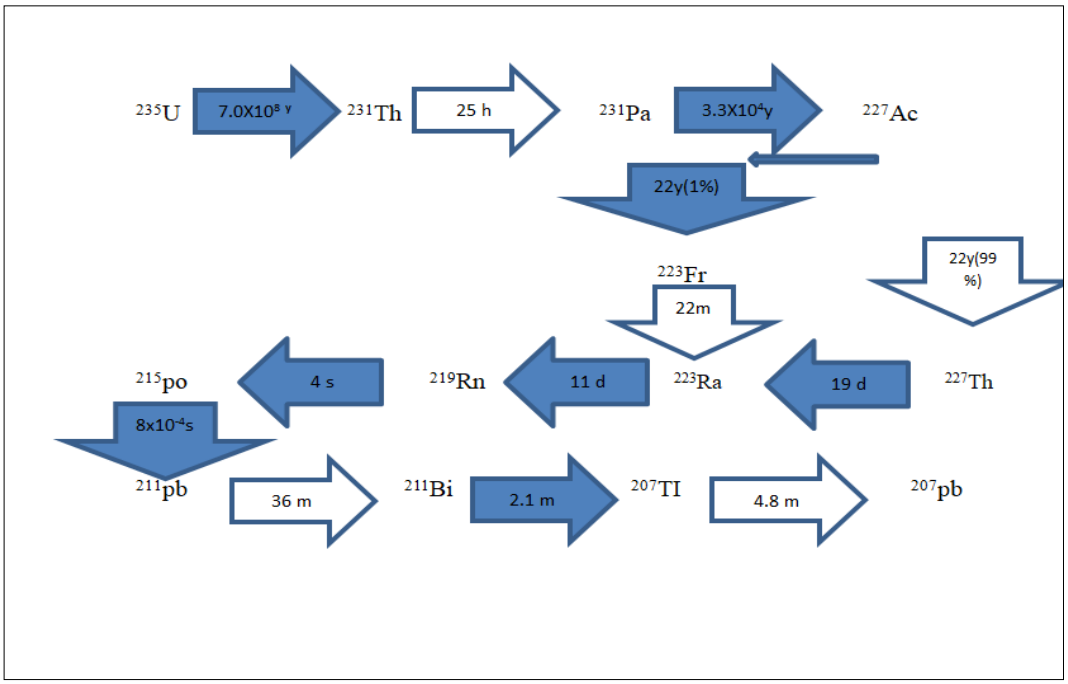

Figure 2.2 Uranium (^{235}U) decay series (4n+3)

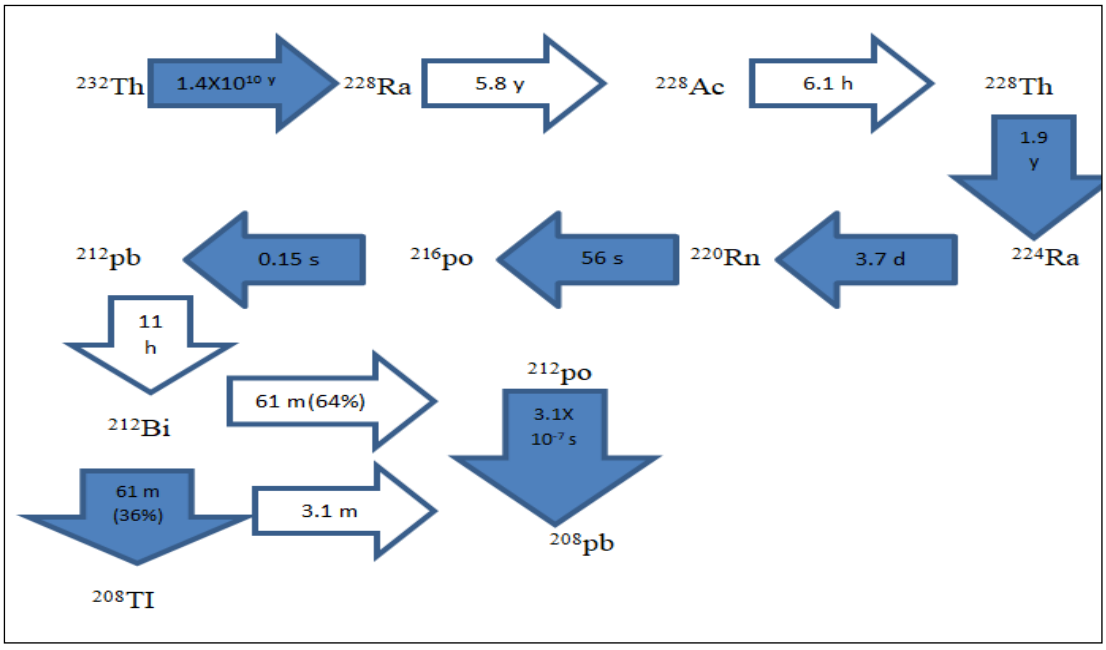

Figure 2.3 Thorium (^{232}Th) decay series (4n)

2.2.3 Potassium

Potassium is a soft silver-white alkali metal that can easily be oxidized to its known (+1) oxidation state in the air (K). Potassium is a simple cation present in minerals as a positive counterion (K+) in a number of preparations and is a natural element. It is particularly susceptible to water and is a vital part of the human body as well because it controls the electrical system with the help of Na^+ to balance water in the body. This is the total of 28 identified potassium isotopes, of which only three are natural, are known to ^{39}K (93.3%); ^{40}K (0.012%) (terrestrially important) and ^{41}K (6.7%). With regard to natural radiation, the element is of great significance due to the half-life of ^{40}K as 1.28 x 10^9 years (Aswood, 2013). Figure 2.4 demonstrates the decay of ^{40}K into stable ^{49}Ar (10.75%) by electron capture and by positron emission, and the decay to stable ^{40}Ca (89.25%) by beta emission. For every 100 disintegrations in this phase of decay, approximately 11 of these emit gamma photons with a maximum energy of 1.46 MeV and 89 emit beta particles with a maximum energy of 1.31MeV (Vearrier et al., 2009).

The activity of natural potassium is 31 Bqg^{-1}. ^{40}K is in healthy animals and humans, the main cause of radioactivity, and more than just that, ^{14}C. The mass of 70 kg of the human body is about 4,400 nuclei of ^{40}K each second of decay (Knoll, 2010). However, the body's potassium content is under homeostatic regulation and is barely influenced by environmental changes, resulting in a relatively constant dose of ^{40}K in the body (Vearrier et al., 2009).

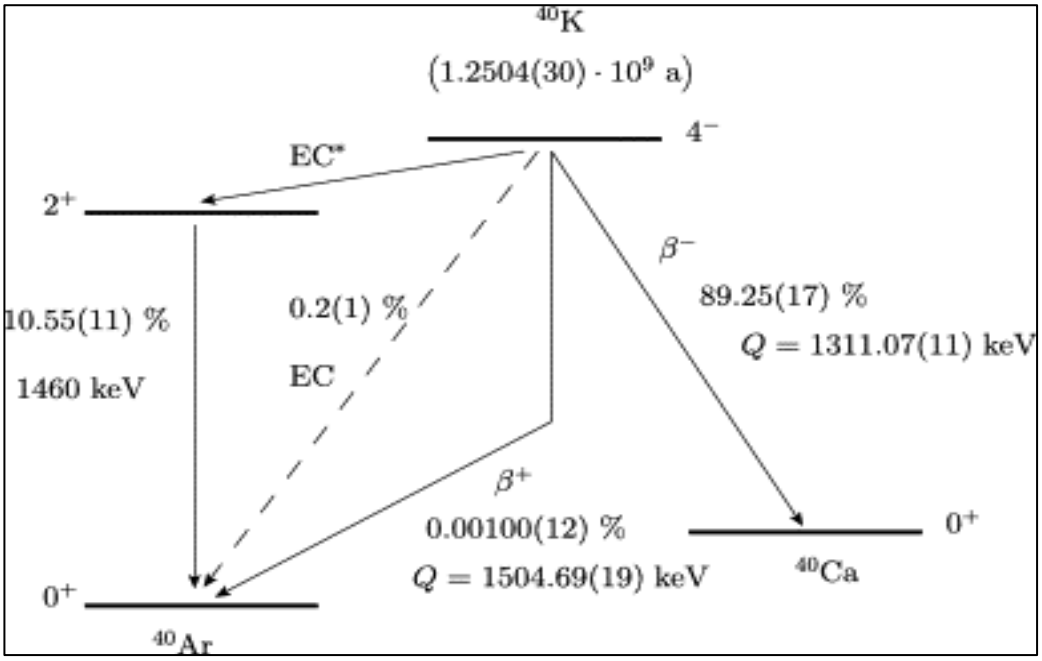

Figure 2.4 **^{40}K** decay series (IAEA,1989)

2.2.4 Radium

Radium and its daughter products are responsible for a significant fraction of the doses obtained by human beings from naturally occurring internal emitters. ^{226}Ra emits an alpha particle with a half-life of 1622 years. The radium decay yield is ^{222}Rn, which is responsible for exposure to radiation.

2.2.5 Radon

^{222}Rn is produced by the alpha decay of ^{226}Ra. It originates from the natural decay of uranium found in almost all types of soil, although the concentrations vary widely (NCRP, 1984). It normally moves up from the ground to the air. Radon decays with a half-life of 3.8 days, producing shorter daughters, namely ^{218}Po, ^{214}Pb, ^{214}Bi and ^{214}Po. Some atoms are leaving the soil and entering the surrounding air or water. Radon is therefore available indoors and outdoors. The emission of alpha particles occurs within the lungs as the radon progeny is inhaled. Radon decay

products are the most important and dominant contributors to the inhalation dose. Radon and its decay products are the main natural sources of human irradiation in the air, as shown in Table 2.1. Radon concentrations are higher indoors, but there is also a significant concentration outdoors. In addition to affecting the public, radon has been noted to be responsible for many reported cancerous linked cases among uranium miners.

Table 2.1 Typical volume activities of ^{222}Rn and ^{220}Rn (outdoors and indoors) and typical annual effective dose (outdoors and indoors) [adopted from UNSCEAR, 2001].

Nuclide	**Activity Outdoor (Bqm^{-3})**	**Activity Indoor (Bqm^{-3})**	**Effective dose Outdoor ($\mu Sv y^{-1}$)**	**Effective dose Indoor ($\mu Sv y^{-1}$)**
^{222}Rn	10	10–100	100	1000
^{220}Rn	10	2–20	≈ 10	≈ 90

2.3 Recommendations and Regulations of NORM

NORM in the environment may be inadvertently acquired by the body through inhalation, ingestion or absorption (IAEA, 1989). Consequently, they may be deposited in various parts of the body. There are already several international and regional organisations formed to track and alert the public to the effects of radiation exposure. These organisations investigate the most efficient methods by working for the governments and the international commissions formed to reduce the associated effects due to radiation.

It should be remembered that sections of the United Nations have worked to define radioactivity levels. In 1955, the United Nations (UN) Scientific Committee on Atomic Radiation (UNSCEAR) was formed to estimate and monitor the rates and

effects of exposure to ionising radiation on humans. Over the years, these studies are issued by UNSCEAR to analyse radiation exposure from various sources, including nuclear power plants, nuclear weapons tests in the 1960s, natural and medical radiation exposure and occupational radiation exposure. The Food and Agriculture Organization (FAO), which is part of the UN, plays an important role in putting forth guidelines for food safety standards. The International Atomic Energy Agency (IAEA) also plays a key part in protecting consumers from potential food-related radiation hazards (Poschl and Nollet, 2006). UNSCEAR calculated the reference dose level (RDL) of the committed effective dose to be 0.3 mSv per year in its guidance on the ingestion of food and water. The RDL of 0.3 mSv corresponds to about 30% of the dose limit (which is 1 mSv) for the population, which is suggested by both the international basic safety standards (IAEA, 1996) and the International Commission on Radiological Protection (ICRP, 1991). In Malaysia, regulations of NORM is done by Atomic Energy Licensing Board (AELB). The dose limit for the public is 1 μSv/y and the critical concentrations of radionuclides in the uranium and thorium series is 1 Bq/g and for K-40 is 10 Bq/kg.

The dosages of radiation are divided into two groups. First, is the exposure from natural extra-terrestrial sources such as cosmic radiation and radiation from terrestrial radioactivity. The second group is dosage received by the human body from internal sources composed of the naturally occurring radionuclides. UNSCEAR (UNSCEAR, 2000) reported that, as shown in Table 2.2, the global average annual human exposure from natural sources is around 2.4 mSv y^{-1}. Human exposure from natural sources, as shown in Table 2.2, is 2.4 mSv y^{-1}.

Table 2.2 Average radiation dose from natural sources (UNSCEAR, 2000)

Sources of Natural Radioactivity	Annual Effective Dose (mSv)	Typical Range (mSv)
External		
Cosmic rays	0.4	0.3–1.0
Terrestrial gamma ray	0.5	0.3–0.6
Internal		
Inhalation (principally radon)	1.2	0.2–10.0
Ingestion	0.3	0.2–0.8
Total	2.4	1–10

2.3.1 ICRP

ICRP recommends an annual ingestion dose of 1 mSv for the public. It also recommends a soil radon concentration of 200–600 Bqm^{-3}. The main recommendations are the dose conversion factors for different age groups for the calculation of ingestion dose, and the main values are 280, 230 and 6.2 for ^{226}Ra, ^{232}Th and ^{40}K, respectively.

2.3.2 UNSCEAR

UNSCEAR monitors the radioactivity concentrations reported in the literature and provides worldwide averages for various radiological quantities. The average concentrations for NORM in soil worldwide are 35, 30 and 400 $Bqkg^{-1}$ for ^{226}Ra, ^{232}Th and ^{40}K, respectively. The soil to plant transfer factors reported by UNSCEAR are 0.03 for ^{226}Ra and 0.0005 for ^{232}Th, respectively. The dose rate for the public recommended by UNSCEAR from various sources is given in Table 2.1. The Ra_{eq} recommended by UNSCEAR is 370, and the recommended value for external and internal hazard indices of soil is less than 1.

2.3.3 Others

Other organisations, like WHO, have made recommendations based on the those of other agencies, like the ICRP, and it recommends an annual ingestion dose in the range of 250–400 μSv. The IAEA provides values for transfer factor worldwide in the ranges of 0.002–0.0049 for ^{226}Ra, 0.00033–0.0018 for ^{232}Th and 0.55–1 for ^{40}K.

2.4 NORM Exposure

2.4.1 Background radiation

The radiation source is divided into two different groups: internal and external exposure. Outside exposure is the sources outside the human body result, while internal exposure is the result of radioisotopes accumulated in the environment of the exposed person. The possibility of making measurements external exposure doses using the open detection tools either directly or indirectly, external exposures are better to deal with than internal exposures. In measuring the amount of radioisotope involved and its distribution within the body, the difficulty of assessing the internal doses is due to the assumptions made. As a consequence, the dosage equivalent of internal exposure should generally be the best choice for internal dose assessment (Knoll, 2010).

2.4.2 Internal exposure

Three primary paths, namely inhalation, absorption, and the bloodstream and lymphatic system via the skin, provide radioactive contaminants with access to the body. By ingesting gas, food and water, the transmission of radionuclides from the atmosphere to the human body can occur. Once within the body, depending on the

chemical properties of the elements and compounds, radionuclides are absorbed, metabolized and spread to tissues. Thus, the final physiological effects of internal exposure are determined by physical and biological entities. The physical properties include physical entities of radionuclides (half-life), the form and energy of radiation emitted and the linear energy (Dovlete and Povinec). Comparatively, biological variables include the chemical features of radionuclides, the transport of radionuclides through the body. Translocation between one tissue and another. The Target organ or tissue position, transit time in internal organs, out-of-body respiration pathways, biological half-life and effective half-life, radiation reaction of disposable tissues and other determinants such as age, gender, pregnancy, and disease, which naturally increase the likelihood of absorption. Inhalation doses are the result of airborne dust particles containing radionuclides belonging to the ^{238}U and ^{232}Th decay chains. Inhalation of radon or short-lived decay products from radon is one of the main contributions of doses to respiratory tract.

Ingestion doses are primarily the result of radionuclides ^{40}K, ^{238}U and ^{232}Th in food and drinking water. The ^{40}K dose rate can be directly calculated and precisely in the body, from external measures of its concentration. Complicated chemical analysis of the tissues exposed to these elements is needed to analyse the radionuclide content of the uranium and thorium series in the body.

Investigation of radionuclides uptakes from soil to plants becomes necessary due to their large contribution to human internal radiation dose and in the understanding of the factors that influence the uptake process. Therefore, it is necessary to determine and estimate the activity of various radionuclides present in soil and their transfer factors TFs in different food samples.