NATURAL RADIOACTIVITY IN DOMESTIC WATER SOURCES IN NORTHERN PENINSULAR MALAYSIA

by

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Thesis submitted in fulfillment of the requirements
for the degree of
Doctor of Philosophy

March 2012

KERADIOAKTIFAN ASLI DALAM SUMBER AIR DOMESTIK DI UTARA SEMENANJUNG MALAYSIA

Oleh

BASHIR GIDE MUHAMMAD

Tesis yang diserahkan untuk
Memenuhi keperluan bagi
Ijazah Doktor Falsafah

Mac 2012

DEDICATION

This work is dedicated to my late parents Alhaji Gide Muhammad and Malama Zainab Gide Muhammad for their amazing foresight, industry and endurance and more importantly for their unyielding faith in education. To my dear wife Hauwa'u Suleiman for her patience and understanding. Finally, to my children Muhammad, Mahmud, Fadimatu and Khadijah.

ACKNOWLEDGEMENT

I would like to express my sincere gratitude to my supervisors, Prof. Mohamad Suhaimi Jaafar, Deputy Dean (Academic and Student Affairs), School of Physics, Universiti Sains Malaysia and Dr. Azhar Abdul Rahman, Chairman of Medical Physics Programme for the opportunity to undertake this project and for their support and guidance that led to the success of this project. My sincere gratitude also goes to Dr. Eid Mahmoud for his useful suggestions in some of the laboratory work.

I would also like to express my appreciation for the valuable assistances given to me by Encik Azmi Abdullah of Biophysics Laboratory, and Encik Yahya Ibrahim of Medical Physics Laboratory for taking the huge task of driving me around for the purpose of this project. A general thanks to the entire staff of Centre for Energy Research and Training, Ahmadu Bello University, Zaria, Nigeria and the staff of School of Physics, Universiti Sains Malaysia for their valuable help in my experimental work and their useful suggestions. My sincere gratitude also goes to the staff of PBA Penang, Bahagian Bekalan Air Perlis, Jabatan Kerja Raya Negeri Perlis, Jabatan Bekalan Air Negeri Kedah and Lembaga Air Perak for approving my access to the water sources used in this project.

Thanks are also due the management of Umaru Musa Yar'adua University, Katsina, Nigeria and School of Postgraduate studies, Universiti Sains Malaysia for financial support through postgraduate research grant (1001/PFIZIK/844023).

Finally, I would also like to thank my family and all my friends whose direct or indirect contribution to this work made it a success. May Allah (SWT) reward all abundantly.

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LIST SYMBOLS

α	alpha particle
β	beta particle
$\sum$	summation
ϕ	neutron flux (cm^{-2} s^{-1})
σ	activation cross section of target isotope (cm^2)
θ	fractional isotopic abundance of the isotope of interest
γ	gamma-ray branching ratio
λ	decay constant of the product nuclide (s^{-1})
ε	detector efficiency at the photopeak
W_{std}	mass of the element in the standard
W_{sample}	mass in the sample
µSv	micro Sievert
µGy	microgray
µg	microgram
µm	micrometre
A	activity
a	annum
A_c	radionuclide activity concentration (Bql^{-1})
A_i	annual intake of water (ly^{-1})
A_{Ra}	radium activity
A_{Th}	thorium activity
A_K	potassium activity
Bq	Becquerel

C_e	Concentration in environment
C_f	dose conversion factor for the radionuclide (Sv Bq^{-1})
C_o	Concentration in organism
C_{std}	count rates for the standard
C_{sample}	count rates for the sample
cm	centimetre
D	total committed effective dose (Sv y^{-1})
E_d	effective dose
E_γ	gamma energy
eV	electron volt
H_{ex}	external hazard index
H_{in}	internal hazard index
H	hour
Iγ	gamma radiation representative level index
K	kelvin
kBq	kilobecquerel
keV	kilovolt
L	litre
M	atomic weight of the element of interest (g mol^{-1})
m	metre
mBq	millibecquerel
mg	milligram
ml	millilitre
mm	millimetre
mSv	milli Sievert

N	avogadro's number (6.023 x 1023 mol^{-1})
nGy	nanogray
pCi	picocurie
Ra_{eq}	radium equivalent activity concentration indices
s	second
t_c	counting time (s)
t_d	decay time (s)
t_i	irradiation time (s)
V	volt
W	mass of element of Interest (g)
y	year

ACRONYM DEFINITIONS

ALARA	-	As low as reasonably achievable
BAT	-	Best available technologies
BCF	-	Bio-concentration factor
DCF	-	Dose conversion factor
DNA	-	Deoxyribonucleic acid
FAO	-	Food and agriculture organization
FWHM	-	Full width at half maximum
GPS	-	Global positioning system
HPGe	-	High-Purity Germanium
IAEA	-	*International Atomic Energy Agency*
ICRP	-	*International Commission on Radiological Protection*
ISO	-	International Organization for Standardization
LET	-	Linear energy transfer
LNT	-	Linear non threshold
LSA	-	Liquid scintillation assembly
MCA	-	Multi-channel analyzer
MCGL	-	Maximum contaminant goal limit
MCL	-	Maximum contaminant LEVEL
MDA	-	Minimum detectable activity
NAA	-	Neutron activation analysis
NaI(Tl)	-	Thallium doped sodium iodide
NDWQS	-	National drinking water quality standards
NIRR-1	-	Nigerian research reactor-1
NORM	-	Naturally occuring radioactive materials

NRC	-	National Research Council
NRPB	-	National Radiological Protection Board
ppm	-	Parts per million
RDL	-	Reference dose level
RH	-	Relative humidity
TDS	-	Total dissolved solids
TENORM	-	Techincally enhanced NORM
UK	-	United Kingdom
USA	-	United State of America
USEPA	-	United States Environmental Protection Agency
WHO	-	World Health Organisation

KERADIOAKTIFAN ASLI DALAM SUMBER AIR DOMESTIK DI UTARA SEMENANJUNG MALAYSIA

ABSTRAK

Bioakumulasi bahan radioaktif terlarut melalui air minuman dianggap sebagai faktor penting terhadap masalah kesihatan jangka panjang. Sehubungan dengan perkara ini, Pertubuhan Kesihatan Sedunia (WHO) telah meletakkan satu garis panduan kualiti radiologi bagi air minuman, digunapakai untuk menskrin sumber air yang digunakan secukupnya di kawasan utara Semenanjung Malaysia untuk keradioaktifan asli. Lokasi pensampelan adalah 12, 26, 16 dan 19, masing-masing dari negeri-negeri Pulau Pinang, Perlis, Kedah dan Perak. Pertamanya, kepekatan kasar aktiviti alfa dan beta telah diukur dalam sumber-sumber bekalan domestik air mentah dan seterusnya air terawat dengan menggunakan prosedur Pertubuhan Piawaian Antarabangsa (ISO). Suatu sistem pembilang berkadaran aliran gas 8 saluran (EURYSIS-IN20) telah digunakan dalam analisis ini. Kepekatan kasar aktiviti alfa dan beta dalam air mentah didapati berjulat masing-masing dari 90 ± 6.30 Bq m^{-3} hingga 490 ± 21.20 Bq m^{-3} dan 370 ± 3.00 Bqm^{-3} hingga 1890 ± 26.00 Bq m^{-3} masing-masing. Nilai kasar alfa didapati berada di bawah MCL 500 Bq m^{-3} sebagaimana yang disyorkan oleh ICRP dan WHO. Manakala, nilai kasar beta dalam beberapa sampel adalah melebihi nilai MCL 1000 Bq m^{-3} (nilai yang sama yang digunapakai oleh Piawaian Kualiti Air Minuman Kebangsaan, Malaysia). Nilai kepekatan aktiviti beta yang tinggi di beberapa kawasan, mendorong kepada perlunya penskrinan selanjutnya. Dalam hal ini, analisis spektroskopi gamma telah digunakan untuk menganalisis radionuklid asli (U-238, Th-232, K-40 and Ra-226) dalam sampel air dan tanah daripada sumber air domestik. Seterusnya, kebersandaran umur dos berkesan komited yang disebabkan oleh pemakanan langsung nuklid ini dianggarkan dengan

menggunakan Faktor Penukaran Dos (DCF) yang disediakan oleh UNSCEAR. Hasil kajian menunjukkan min dos berkesan komited adalah masing-masing 0.112 mSv, 0.104 mSv dan 0.100 mSv untuk bayi, kanak-kanak dan orang dewasa. Nilai dos tersebut di beberapa kawasan kajian adalah didapati lebih tinggi berbanding had dos rujukan (RDL) ICRP dan WHO iaitu 0.1 mSv daripada air minuman, yang merupakan 10% RDL daripada sumber asli di atas latar belakang asli. Monitor Rad 7 dan aksesori Rad H_2O radon dalam air juga digunakan untuk mengukur kepekatan aktiviti di dalam air dan menilai kesan kelembapan relatif dan kebolehubahan suhu dalam pemantauan berterusan radon. Kepekatan radon yang relatifnya tinggi didapati dalam beberapa sumber air mentah berbanding kawasan lain di dunia, bagaimanapun nilainya masih jauh di bawah paras yang disyorkan oleh WHO, iaitu aras rawatan 100 Bq L^{-1}. Daripada data ini, kebergantungan umur yang berkaitan dengan dos berkesan komited yang disebabkan oleh pemakanan ^{222}Rn, akibat penggunaan secara langsung air minuman juga dihitung. Kepekatan aktiviti masing-masing yang diukur bagi radionuklid asli di dalam tanah menyebabkan aktiviti yang setara dengan radium (Ra_{eq}) dan nilai indeks bahaya luaran (H_{ex}) adalah masing-masing berjulat dari 81 – 185 Bq kg^{-1} dan 0.22 – 0.49. Nilai ini adalah lebih rendah berbanding had yang dibenarkan ICRP, iaitu 370 Bq kg^{-1} dan 1 masing-masing. Kadar dos terserap akibat daripada aktiviti nuklid asli juga didapati berada dalam julat 36.90 – 82.59 nSv h^{-1} dengan min di bawah nilai purata dunia (59 nSv h^{-1}). Analisis Pengaktifan Neutron (NAA) dan Spektrometri Jisim Plasma Gandingan Induktif (ICP-MS) telah digunakan untuk menganalisis tahap kepekatan unsur surih dalam sampel sedimen dan bio-akumulasinya, masing-masing dalam beberapa spesis akuatik. Bioakumulasi tertinggi diperhatikan dalam *Sciaera* (Gelama) terkumpul lebih kurang 0.66% daripada tahap kepekatan arsenik terkesan di dalam sedimen, diikuti oleh *Rastrelliger kanagurta*

(kembung) dengan bioakumulasi lebih kurang 0.64% dan kemudian *Lutianus argentimaculatus* (Ikan Merah) lebih kurang 0.48% . Unsur-unsur lain seperti U, Cs, Co dan Fe menunjukkan nilai bioakumulasi yang jauh lebih rendah, menandakan keselamatan spesies dan manusia sebagai pengguna tearkhir.

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ABSTRACT

Bioaccumulation of dissolved radioactive materials through drinking water is considered an important pathway for long-term health concerns. Thus, the World Health Organization (WHO) guidelines for drinking water radiological quality were adopted to screen the adequately utilized surface water resources in northern peninsular Malaysia for natural radioactivity. The sampling locations were 12, 26, 16 and 19 from the states of Penang, Perlis, Kedah and Perak, respectively. Firstly, gross alpha and gross beta activity concentrations were measured in the raw and subsequently the treated domestic water supply sources using International Standard Organisation (ISO) proceedures. An eight channel gas flow proportional counting system (EURYSIS-IN20) was employed in this analysis. The gross alpha and beta activity concentrations in the raw water were found to range from 90 ± 6.30 Bq m^{-3} to 490 ± 21.20 Bq m^{-3} and 370 ± 3.00 Bq m^{-3} to 1890 ± 26.00 Bq m^{-3}, respectively. The gross alpha values fall below the ICRP and WHO recommended MCL of 500 Bq m^{-3} while the gross beta values in some samples exceeds the MCL value of 1000 Bq m^{-3}(similarly adopted by the Malaysian National Drinking Water Quality Standards). The high beta activity concentration values in some areas prompted the need for further screening. In this regard, gamma spectroscopic analysis was employed to analyze the natural radionuclides (U-238, Th-232, K-40 and Ra-226) in the water and soil samples from the domestic water sources. Subsequently, the age dependent committed effective doses due to direct

ingestion of these nuclides were estimated using the Dose Conversion Factors (DCF) provided by UNSCEAR. The results indicated the mean committed effective doses to be 0.112 mSv, 0.104 mSv and 0.100 mSv for infants, children and adults, respectively. The values in few areas were found to be higher than the ICRP and WHO reference dose limits (RDL) of 0.1 mSv from drinking water, which is 10% of the RDL from natural sources above the natural background. The study also utilizes Rad 7 radon monitor and its Rad H_2O radon-in-water accessories to measure ^{222}Rn activity concentration in the water and to assess the effects of relative humidity and temperature variability in continuous radon monitoring. Relatively high radon concentrations were obtained, however, the values still falls below the WHO recommended treatment level of 100 Bq L^{-1}. From this data, the age-dependent associated committed effective doses due to the ingestion of ^{222}Rn as a consequence of direct consumption of drinking water were also calculated. The respective measured activity concentrations of the natural radionuclides in soil led to the radium equivalent activity (Ra_{eq}) and the external hazard index (H_{ex}) ranging from 81 – 185 Bq kg^{-1} and 0.22 – 0.49 respectively. These values are lower than the ICRP corresponding permissible limits of 370 Bq kg^{-1} and 1, respectively. The absorbed dose rate resulting from these natural nuclides was also found to be in the range of 36.90 – 82.59 nSv h^{-1} with a mean below the world average value (59 nSv h^{-1}). Neutron Activation Analysis (NAA) and Inductively Coupled Plasma Mass Spectrometry (ICP-MS) were employed to analyze the level of trace elements concentrations in sediment samples and their bioaccumulation in some aquatic species respectively. The highest bioaccumulation was observed in *Sciaera* (Gelama) accumulating about 0.66%

of the deteceted arsenic concentration levels in the sediments, followed by *Rastrelliger kanagurta* (Kembung) with a bioaccumulation of about 0.64% and then *Lutianus argentimaculatus* (Ikan Merah) having about 0.48%. Other elements such as U, Cs, Co and Fe shows far lower bioaccumulation values indicating the safety of the species and the humans being the ultimate consumers.

CHAPTER ONE

INTRODUCTION

1.1 BACKGROUND

NORM, an acronym for naturally occurring radioactive materials, refers to the long-lived radioactive elements such as uranium, thorium and potassium and their decay products, such as radium and radon that existed since the formation of the earth. These elements have always been present in the earth's crust and subsequently, in all environmental media. Some minerals attributable to the origin of these materials include coal that is known to contain traces of uranium and thorium, as well as potassium-40, lead-210, and radium-226. Mineral sands are also known to contain significant amount of monazite, a rare earth material that contains thorium. Tantalum ores, also contain many minerals, some of which contain uranium and thorium. Phosphate used in the production of agro-chemicals such as fertilizer, herbicides and pesticides also constitute a major source of NORM in the environment. Building materials, inevitably found widely distributed and closer to humans may also contain elevated levels of natural radionuclides.

The presence of NORM in the environment is known to be responsible for a large radiation exposure to humans as reported by many previous studies (UNSCEAR, 2000b, Abo *et al*., 2008, Amrani and Cherouati, 1999, Briner, 2010, Collman *et al*., 1991). The human health hazards posed by NORM usually comes as a result of internal irradiation of sub-cellular organelles from ingested as well as inhaled natural radioactive materials and their decay products found in the environment through air, food and mainly drinking water. In a study on the incubation period for the induction of cancer by

internally deposited radionuclides, it was gathered that irradiation from internally deposited radionuclides induces malignant tumors by accumulation in specific organs, that are irradiated over a life long period. The study also suggested that indirect radiation effects such as effects of highly reactive species resulting from radiolysis of water is another factor responsible for human radiation exposure (Yamamoto *et al*., 2010). In another study to assess fatal cancer risk after external and internal exposure from natural radionuclides in soil, Zupunski and co workers evaluated the cancer risk for NORM exposure in the case of both inhalation and ingestion. The estimated cancer risks from both inhalation and ingestion exposure were found to have effects on all tissues, especially to deposition sites in the body (Zupunski *et al.*, 2010). The release of significant quantities of long-lived radionuclides to the environment during the Chernobyl nuclear power plant accident in 1986 was also assessed by a previous study. The study evaluated how radionuclides contributed to radiation doses due to ingestion of contaminated foods and external exposure from the ground deposition. The study further showed that ingestion of the long-lived radionuclides, primarily radiocesium, typically contributed a small percentage of the total thyroid dose (Minenko *et al*., 2006). A review on the toxicity of depleted uranium as an emerging environmental pollutant also enumerated the chemical toxicity associated with the original element and stressed that large doses seek kidney as the target organ for the acute chemical toxicity. Adult animals that were exposed to depleted uranium during development were found to display persistent alterations in behavior, even after cessation of depleted uranium exposure (Briner, 2010). Collman and collegues also explored the association between groundwater radon levels and

childhood cancer mortality. The study suggested the existence of a strong association between waterborne radon and leukaemia (Collman *et al.*, 1991).

Generally, all living organisms on earth are exposed to radiation from different natural radioactive sources such as cosmic radiation; external radiation from natural radionuclides present in soils, rocks and subsequently building materials; and internal radiation due to ingested and inhaled radionuclides present in food, air and water. The presence of various radioactive materials in the body, by accumulation of trace amounts as a consequence of ingestion and inhalation, continuously irradiate the sub cellular organelles with alpha and beta particles as well as gamma rays of varying energies. Evidences from both human and animal studies also indicate that radiation exposure at low to moderate doses may increase the long-term incidence of cancer. Animal studies in particular suggest that the rate of genetic malformations may be increased by radiation exposure.

1.2 STATEMENT OF THE PROBLEM

Some parts of the northern Malaysian peninsular are characterised by large deposits of Amang tin tailings. The tailings which mainly consists of a mixture of tin ore, sand and minerals were initially left as waste products of tin mining activities in the area. Amang, by-product of tin minerals reprocessing, has been found to contain valuable minerals such as zircon, monazite, xenotime, columbite and struvirite that are known to contain uranium and thorium of invarying concentrations. Similarly, many other surface water resources utilised for domestic purposes in the area are either dams, whose contents are characterised by high residence time, hence high dissolved solids,

or rivers, which are also heavily characterised by wash-off of contaminants in their flow process. These instances, coupled with facts from a number of studies, prompted the need for screening of the domestic water sources in the area for radiological safety. The carbon radiometric survey conducted during 1995 and 1996 in parts of the states of Pahang and Kelantan, using the GR650 spectrometer provided by IAEA indicated some parts of the area to have uranium potential (OECD Nuclear Energy Agency, 2001). Similarly, seawater, sediments and some marine species along the coastal areas of Malaysia were found to contain uranium and thorium of considerable specific activities (Akyil and Mohd, 2007, Termizi, 2004). A study conducted on the influence of tin tailings on NORM in the states of Selangor and Perak, Malaysia also revealed some radiological risk associated with amang processing and the accumulated effluent in the recycling ponds in the area. Sediment samples from the recycling ponds of two amang plants in the states of Selangor and Perak, Malaysia, were found to have traces of U-238, Ra-226, Th-232 as well as K-40 higher than many other parts of the world. Correspondingly, the mean radium equivalent activity concentration indices, Ra(eq), and gamma radiation representative level index, I(gamma), were found to be higher than the world's average (Bahari et al., 2007). Lee and co-workers in a survey of the natural background gamma radiation and radioactivity concentrations in Kinta District, Perak, Malaysia also reported an external gamma dose rates ranging from 39 to 1039 nGy h^{-1}, a mean external gamma dose rate of 222 ± 191 $nGyh^{-1}$ and respective activity concentrations of U-238, Th-232 and K-40 of 12-426 Bq kg^{-1}, 19-1377 Bq kg^{-1} and <19-2204 Bq kg^{-1}(Lee *et al.*, 2009*)*. The observed primordial nuclides in the area are known to be the parents of the

natural decay series, resulting in a chain of radionuclides. The potential health hazards from ingestion of radionuclides through drinking water have been well established, with many countries adopting the guideline activity concentration recommendation of the World Health Organization (WHO, 1988). The above findings constitute the reasons that prompted the need to investigate the abundantly utilised domestic water resources in the area for radiological safety associated with the NORM activity concentration level.

This baseline research evaluates the direct impact of the natural radioactivity level in domestic water supply sources in the northen part of the peninsular Malaysia. The Drinking Water Quality Surveillance Unit, under the Ministry of Health, Malaysia is the body vested with the responsibility management of drinking water quality control throughout the country. The ministry established the National Drinking Water Quality Standards (NDWQS). The standards stipulate limits for physical, chemical, microbiological and radiological parameters and mandated compliance with these standards for all private water suppliers. Consequently all the manufacturers of the treated domestic water samples used in this study claimed to have been approved by the Ministry. The emphasis of the drinking water quality by the manufacturers is on the physico-chemical parameters rather than the limits of NORM. Evaluation of the radioactivity level of the NORM in the surface water in the northern part of Peninsular Malaysia is therefore an important venture not only to the health safety of the members of public in the area but rather, it also provides a basis for the understanding numerous radiological processes in the region. The results obtained from the study is also aimed at providing a baseline data on the NORM level in the much utilised

surface water resources in the area which constitute more than 90% of the total water supply by the residents in the area. The baseline data so provided can also serve as a basis for more ideal regulations in the Malaysian National Drining water Quality Standards (NDWQS). Similarly, this study may suggests a possibility that the radioactivity level of the domestic water in the area to be above the acceptable Maximum Contaminants Level.

1.3 OBJECTIVES

The main objective of the study is to assess the level of NORM in the domestic water supplies of the study area through evaluation of the contribution from gross alpha and gross beta activity concentrations, identifying the radionuclides present and measuring their activity concentrations as well as studying their bioaccumulation in aquatic species. An estimate can therefore be deduced of the radiation risk indices such as the committed effective doses, external hazad index, external gamma dose rate as well as bioconcentration factors to the population resulting from consumption of the water. These estimates can then be ultimately compared with guidance levels set aside by radiological protection agencies such as the International Commisssion for Radiological Protection (ICRP), World Health Organisation (WHO) as well as the national guidance level on radiological protection to the human population. The ideal approach would be through analysis of samples from various locations throughout the study area aimed at achieving the underlisted objectives.

1. To evaluate the level of gross alpha and gross beta activity concentrations in the domestic water sources in the area.

2. To evaluate the activity of U-238, Th-232, Ra-226 and K-40 in the domestic water sources in the area and other associated radiological risk indices in the soil.
3. To estimate the age dependent committed effective doses resulting from direct consumption of the water.
4. To determine the concentration of the radioactive elements and heavy metals in sediments and the bioconcentration factors of some hazardous water contaminants in some aquatic species in the area.

1.4 SCOPE AND LIMITATIONS OF THE STUDY

The study was focussed on evaluation of the level, distribution pattern as well as health risk associated with the NORM existing in the domestic water sources in the northern part of peninsular Malaysia. Thus, the study was limited only to the results of the laboratory measurements of the radionuclides activity concentrations in samples obtained from the the northern states of Perlis, Kedah, Penang and Perak.

1.5 ORGANIZATION AND FLOW OF THE THESIS

This thesis is structured in seven chapters, as summarized below.

Chapter One is the general introduction, primarily intended to provide a general overview of the research problems statement, objectives as well as scope of the research and to present the general flow of the whole research project and organization of the thesis. Chapter Two provides the background

and overview of the subject area, in particlar, occurrence and health effects associated with ingestion of radionuclides through domestic water supply. As a general literature review, the chapter further highlights some very important topics and findings related to the research area. Chapter Three presents the general measurements and analysis of the gross alpha and gross beta activity concentration levels in the domestic water sources in the study area; being the first recommended step in the radiological aspect of drinking water quality. Chapter Four presents the experimental proceedure and outcomes of the gamma spectroscopic analysis conducted to identify and quantify the natural radionuclides present in the domestic water in the study area. The Chapter also present the estimates of the committed effective dose due to these radionuclides resulting from direct consumption of the water in the study area. Chapter Five presents the use of a convenient and reliable method for measurement of waterborne radon activity concentrations and the estimation of committed effective dose from the waterborne radon in the domestic water in the study area. The Chapter further highlights the effects of relative humudity and temperature variability in environmental radon measurements. Chapter Six presents the experimental proceedure and results of the neutron activation analysis (NAA) in determination of trace elements in sediments as well as gamma spectroscopic analysis of soil from the domestic water sources in the area. Also presented in this Chapter is the ICP-MS multi elemental analysis of bioaccumulated trace elements in some aquatic species to determine their bioconcentration levels. Finally, Chapter Seven presents general concluding remarks, offers recommendations and suggests future research associated with this study.

CHAPTER TWO

LITERATURE REVIEW

2.1 INTRODUCTION

Naturally occurring radionuclides of terrestrial origin exist in different compositions in all environmental media throughout the world. Radionuclides from uranium and thorium natural decay series (^{238}U, ^{235}U and ^{232}Th), whose half-lives are comparable to the age of the earth, exist in significant quantities virtually everywhere within the earth's crust (IAEA, 1979). ^{40}K, the only radioisotope of the several natural isotopes of potassium has a relative isotopic abundance of only 0.0118%. Its decay scheme is characterised by a single gamma energy of 1.46 MeV also referred to as monoenergetic decay. The activity concentration of ^{40}K in soil is an order of magnitude higher than that of ^{238}U and ^{232}Th (UNSCEAR, 2000b). Uranium on the other hand, has principally two radioisotopes, ^{238}U and ^{235}U, of which the first is most abundant (99.73%) and is therefore the radiosotope of interest when considering natural uranium exposure. The natural uranium decay series results in a number of radioisotopes leading through 14 decays to the final decay product of stable ^{206}Pb end product as shown in Figure 2.1 with its corresponding decay data depicted in Table 2.1. The principal gamma emission from ^{238}U is associated with its respective eighth and ninth daughters of ^{214}Pb and ^{214}Bi and not directly from ^{238}U. Figure 2.2 and subsequent Table 2.2 gives the decay scheme and decay data of ^{235}U respectively. Thorium, with a radioisotope ^{232}Th also has a complex decay scheme which leads to a stable end product of ^{208}Pb as depicted in Figure 2.3, with subsequent decay data depicted by Table 2.3. An equally important progeny from the above

discussed uranium decay series present in the environment is ^{226}Ra which is the potential emitter of natural radioisotope ^{222}Rn, the radon gas. Radon is present virtually everywhere on earth. ^{222}Rn is an innert gas, emanated through the decay of ^{226}Rn, their short half-lives gives ^{222}Rn progenies the ability to attain rapid radioactive equilibrium with ^{222}Rn. Radon is extremely volatile and is readily released from water. Radon gas can also dissolve and accumulate in water until aerated. ^{222}Rn is a daughter of special interest from this decay chain. This is for the fact that it is responsible for a large percentage of natural radiation exposure; it comes mainly due to decay of radium contained in rocks and soil as part of the uranium radionuclide chain. The radiological consequence due to radium in water could be viewed in two folds; inhalation of emanated radon and its decay products following their release from water into household air and direct ingestion through drinking. Their health effects have been adequately reported (Gosink *et al*., 1990, Banzi *et al.*, 2000, Al-Kazwini and Hasan, 2003, Auvinen et al., 2005, Ajayi and Achuka, 2009,). Radium when absorbed into the body, behaves like calcium, it thus collect in bones thereby leading to bone cancer.

To protect human population from the potential hazards associated with radiation exposure, various radition protection agencies proferred recommendations and various governments made legislations on the protection of the public against radiation exposure above maximum contaminants levels (MCL). The World Health Organisation (WHO) and International Commission on Radiological Protection (ICRP) proposed recommendations on the limitations of domestic exposure above the natural background (WHO, 1988, ICRP, 1991). These recommendations were adopted by many countries

through their radiological protection agencies. The United States Environmental Protection Agency (USEPA) the National Radiological Protection Board (NRPB) in the UK are a few examples. In line with these recommendations, similar attempts were made by the Drinking Water Quality Surveillance Unit, under the Ministry of Health, Malaysia, a body charged with the responsibility of management of drinking water quality control throughout the country. USEPA, in its radiation protection efforts, established a maximum contaminant level (MCL) for radium in public water supplies of 5 pCi/L. The MCL for radium has been set well below levels for which health effects have been observed and is therefore assumed to be protective of public health.

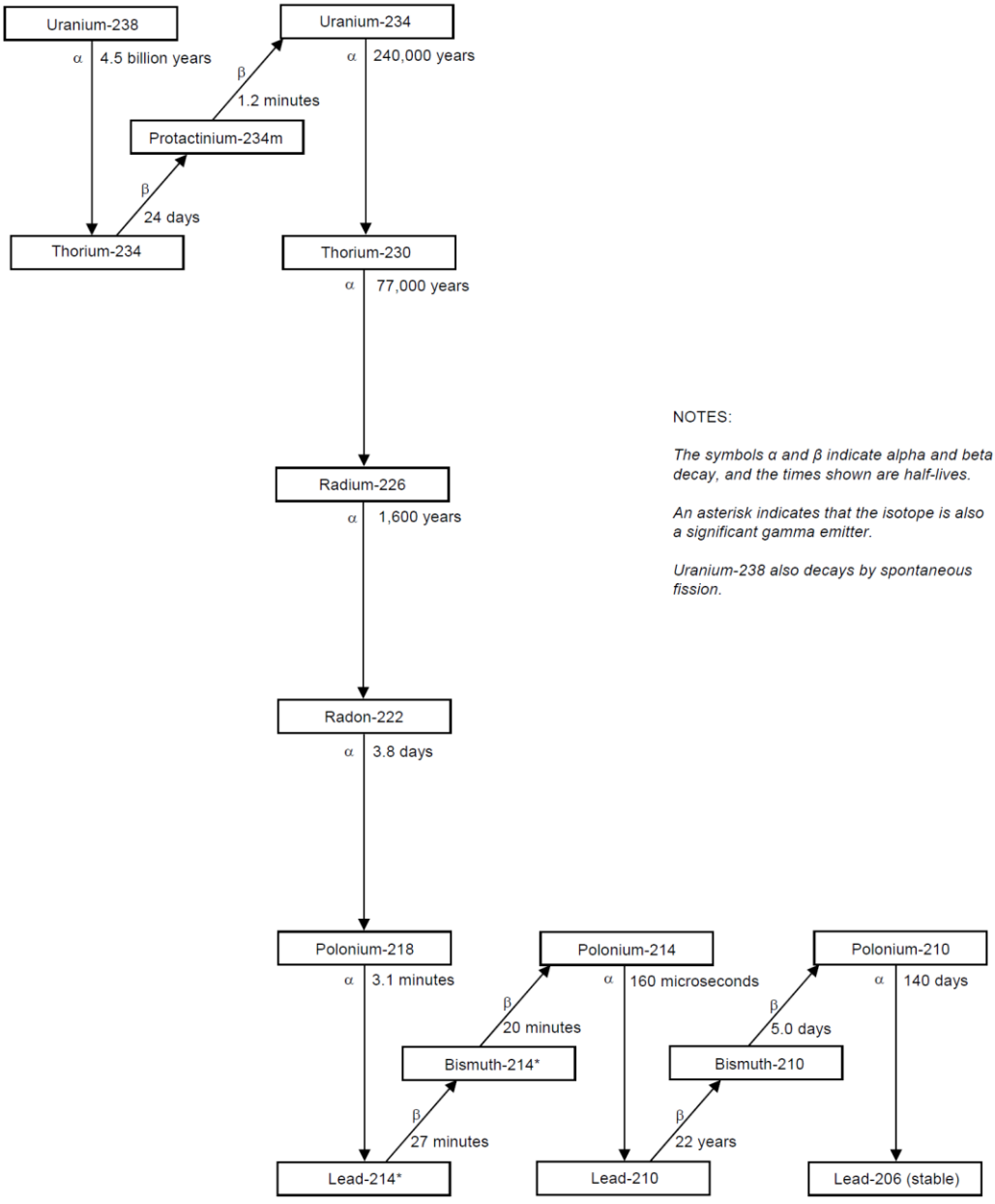
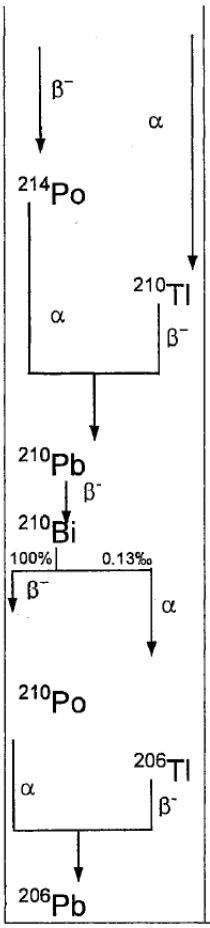


Figure 2.1 U-238 decay scheme

Table 2.1 U-238 decay data

Nuclide	Half-life	Major radiation energies (MeV) and intensities		
		Alpha (α)	Beta (β)	Gamma (γ)
^{238}U	4.51×10^9 a	4.15 (25%)		0.0480 (0.08%)
α		4.20 (75%)		
^{234}Th	24.1 d		0.103 (21%)	0.0633 (4%)
β^-			0.193 (79%)	0.0924 (3%)
				0.0928 (3%)
^{234m}Pa	1.17 min		2.29 (98%)	0.0136 (0.44%)
^{234}Pa	6.75 h		0.53 (66%)	0.0947 (15%)
			1.13 (13%)	0.0984 (25%)
β^-				0.1110 (8%)
				0.1313 (20%)
				0.5693 (10%)
				0.8832 (12%)
				0.9267 (11%)
				0.9460 (20%)
^{234}U	2.47×10^5 a	4.72 (28%)		0.0531 (0.12%)
α		4.77 (72%)		
^{230}Th	8.0×10^4 a	4.62 (24%)		0.0677 (0.38%)
α		4.68 (76%)		0.1436 (0.05%)
^{226}Ra	1602 a	4.60 (6%)		0.0838 (0.31%)
α		4.78 (95%)		0.1859 (3%)
^{222}Rn	3.823 d	5.49		0.5100 (0.08%)
α		(100%)		
^{218}Po	3.05 min	6.00	0.33 (0.019%)	
99.98% 0.02%		(100%)		
β^-				
^{214}Pb	26.8 min		0.65 (50%)	0.0748 (6%)
			0.71 (40%)	0.0771 (11%)
β^-			0.98 (6%)	0.2419 (7%)
				0.2952 (19%)
				0.3519 (37%)
^{218}At	2 s	6.65 (6%)		
		6.70 (94%)		
β^-				
^{214}Bi	19.7 min	5.45	1.00 (23%)	0.6093 (46%)
99.98% 0.02%		(0.012%)	1.51 (40%)	0.7684 (5%)

		5.51 (0.008%) 7.69 (100%) 3.72 (0.002‰) 4.65 (0.07‰) 4.69 (0.05‰) 5.305 (100%)	3.26 (19%) 1.3 (25%) 1.9 (56%) 2.3 (19%) 0.016 (85%) 0.061 (15%) 1.16 (100%) 1.571 (100%)	1.1202 (15%) 1.2381 (6%) 1.3778 (4%) 1.7645 (16%) .0799 (0.014%) 0.2960 (80%) 0.7950 (100%) 1.0800 (19%) 1.2100 (17%) 1.3100 (21%) 0.803 (0.0011%)
	164 ms 1.3 min 21 a 5.01 d 138.4 d 4.19 min Stable			

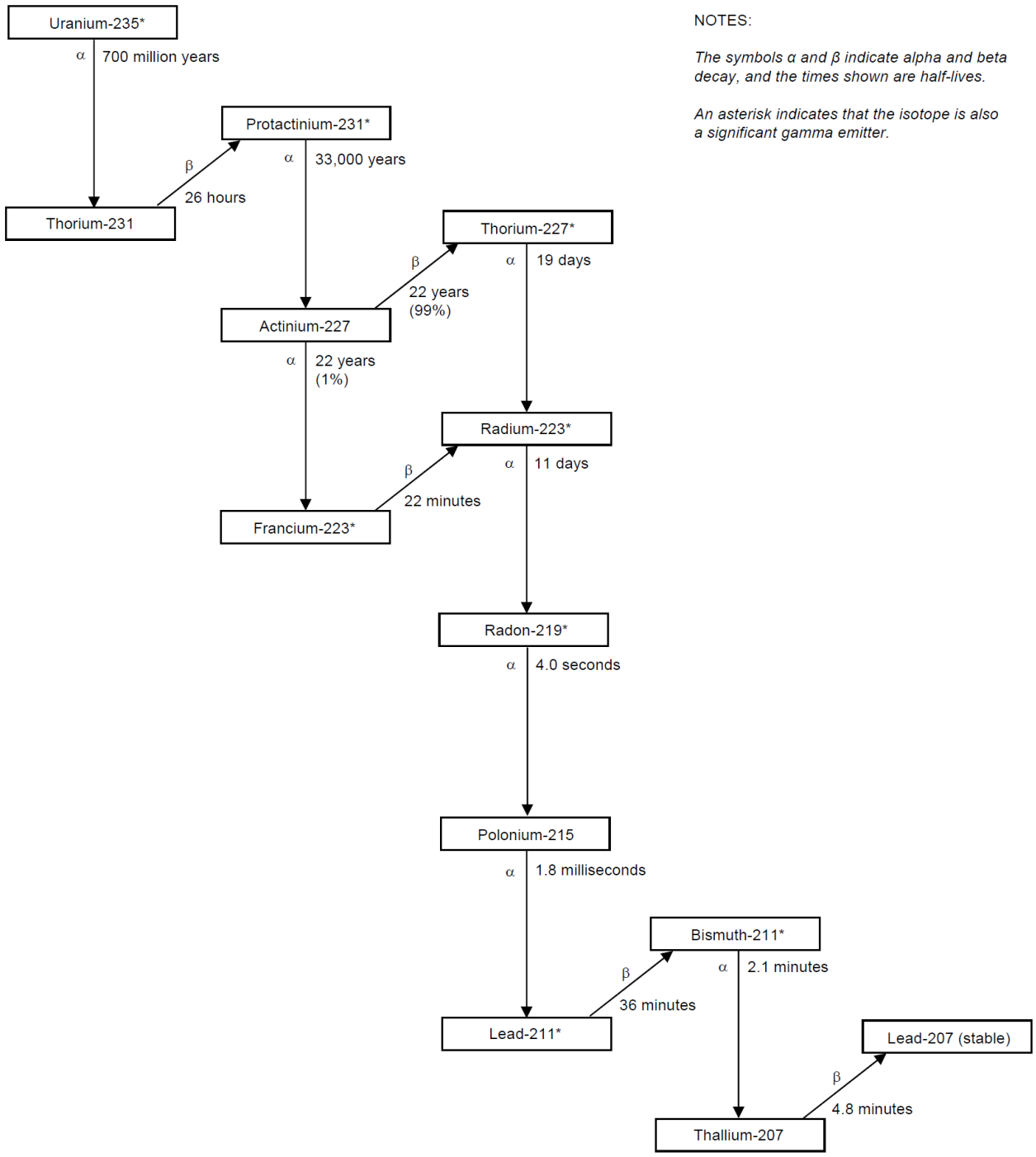


Figure 2.2 U-235 decay scheme

Table 2.2 U-235 decay data

Nuclide	Half-life	Major radiation energies (MeV) and intensities		
		Alpha (α)	Beta (β)	Gamma (γ)
^{235}U	7.1×10^8 a	4.37 (18%) 4.40 (57%) 4.58 (8%)		0.1438 (10%) 0.1633 (5%) 0.1857 (54%) 0.2053 (5%)
α				
^{231}Th	25.5 h		0.140 (45%) 0.220 (15%) 0.305 (40%)	0.0842 (8%) 0.0899 (1 %)
β^-				
^{231}Pa	3.25×10^4 a	4.95 (22%) 5.01 (24%) 5.02 (23%)		0.3000 (2%) 0.3027 (2%)
α				
^{227}Ac	21.6 a	4.86 (0.18%) 4.95 (1.2%)	0.043 (99%)	0.1604 (0.02%)
98.6% 1.4%				
β^-				
^{227}Th	18.2 d	5.76 (21%) 5.98 (24%) 6.04 (23%)		0.0885 (3%) 0.2360 (11%) 0.2563 (7%) 0.2999 (2%) 0.3298 (4%)
α				
α				
^{223}Fr	22 min	5.44 (0.005%)	1.15 (100%)	0.0800 (10%) 0.0815 (8%) 0.0886 (7%) 0.2346 (4%)
β^-				
^{223}Ra	11.43 d	5.61 (26%) 5.71 (54%) 5.75 (9%)		0.0811 (15%) 0.0838 (25%) 0.0947 (9%) 0.1543 (6%) 0.2696 (14%) 0.3241 (4%)
α				
^{219}Rn	4.0 s	6.42 (8%) 6.55 (11%) 6.82 (81%)		0.2712 (10%) 0.4018 (7%)
α				
^{215}Po	1.78ms	7.38 (100%)	0.74 (0.23‰)	0.4388 (0.04%)
100% 0.00023%				
β^-	36.1 min		0.29 (1.4%) 0.56 (9.4%) 1.39 (87.5%)	0.4048 (3%) 0.8318 (3%)
^{211}Pb α				
α ^{215}At β^-	0.1 ms	8.01 (100%)		0.4040 (0.05%)
^{211}Bi	2.15 min	6.28 (16%) 6.62 (84%)	0.60 (0.28%)	0.3511 (12%) 0.4048 (4%) 0.8318 (3%)
0.28% 99.7%				
β^-				
^{211}Po α	0.52 s	7.45 (99%)		0.5700 (0.5%) 0.9000 (0.5%)
α ^{207}Tl β^-	4.8 min			0.8976 (0.27%)
^{207}Pb	Stable			

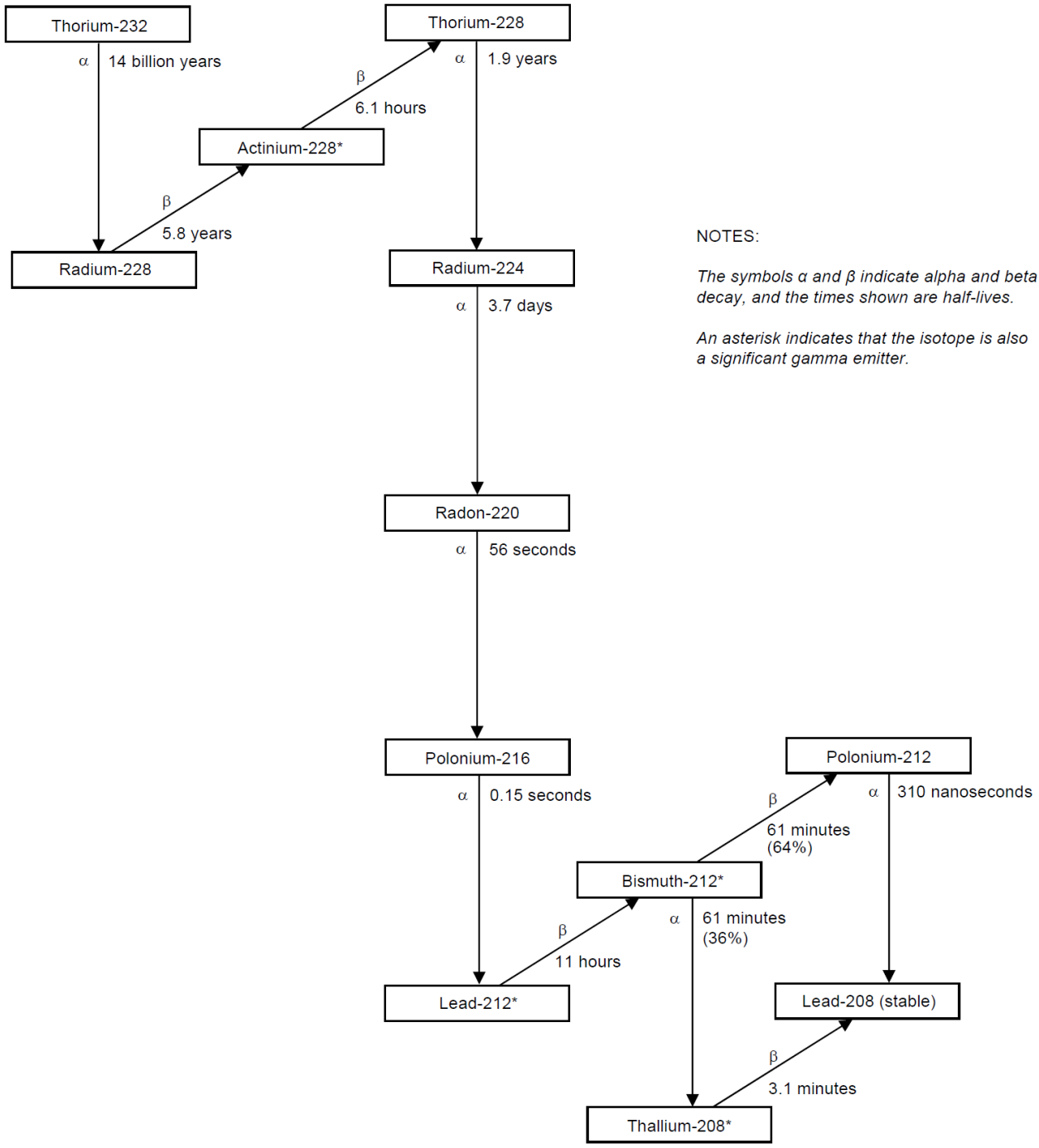


Figure 2.3 Th-232 decay scheme

Table 2.3 Th-232 decay data

Nuclide	Half-life	Major radiation energies (MeV) and intensities		
		Alpha (α)	Beta (β)	Gamma (γ)
^{232}Th	1.41×10^{10} a	3.95 (24%) 4.01 (76%)		0.0590 (0.19%)
α ↓				
^{228}Ra			0.055 (100%)	
β⁻ ↓	6.7 a			
^{228}Ac			1.18 (35%) 1.75 (12%) 2.09 (12%)	0.2094 (6%) 0.3384 (12%) 0.4630 (5%) 0.7948 (5%) 0.9111 (29%) 0.9689 (17%)
β⁻ ↓	6.13 h			
^{228}Th		5.34 (28%) 5.43 (71%)		0.0844 (1.19%) 0.2159 (0.27%)
α ↓	1.910 a			
^{224}Ra		5.45 (6%) 5.68 (94%)		0.0838 (0.21%) 0.2410 (4%)
α ↓	3.64 d			
^{220}Rn	55 s	6.29 (100%)		0.5497 (0.10%)
α ↓				
^{216}Po	0.15 s	6.78 (100%)		
α ↓				
^{212}Pb			0.346 (81%) 0.586 (14%)	0.0748 (10%) 0.0771 (17%) 0.0872 (6%) 0.2386 (43%) 0.3000 (3%)
β⁻ ↓	10.64 h			
^{212}Bi		6.05 (25%) 6.09 (10%)	1.55 (5%) 2.26 (55%)	0.0399 (1%) 0.7271 (12%) 0.7854 (2%) 1.6206 (3%)
64.0% | 36.0%				
β⁻ ↓	60.6 min			
^{212}Po	304 ns	8.78 (100%)		0.5700 (2%) 2.6100 (3%)
α ↓				
^{208}Tl			1.28 (25%) 1.52 (21%) 1.80 (50%)	0.0728 (2%) 0.0749 (3%) 0.2774 (6%) 0.5107 (22%) 0.5831 (86%) 0.8605 (12%)
β⁻ ↓	3.10 min			
^{208}Pb	Stable			

Radioactive decay occurs when an unstable (radioactive) isotope transforms to a more stable isotope, generally by emitting a subatomic particle such as an alpha, beta particles or photons. Inevitably, the presence of these elements from the natural radioactivity series in the earth's crust makes the natural background radiation present in our environment at greatly varying levels, heavily depending on geology. High granite areas or mineralized sands always tend to have more terrestrial radiation than other areas. Terrestrial radiation is traceable to formation of the earth some six billion years ago, with the earth containing many radioactive isotopes. Since then, all the shorter lived radionuclides have decayed with the exception of those radionuclides with very long half lives remaining along with the radionuclides formed from the decay of the long lived radionuclides. These naturally-occurring radionuclides which include isotopes from uranium and thorium and their decay products, such as radon greatly enhance external gamma ray exposure and internal exposure from inhalation and ingestion of the radionuclides. The amount of uranium and radium in soil varies greatly with geographic location and soil type. Some areas with high natural radiation background in terms of gamma absorbed dose rates as reported by the United Nations Scientific Committee on Effects of Atomic Radiation are summarized in Table 2.4 (UNSCEAR 2000).

Table 2.4 Areas of high natural radiation background (UNSCEAR 2000)

Country	Area	Characteristics of area	Approx. Population	Absorbed dose rate in air ($nGyh^{-1}$)
Brazil	Mineas Gerais and Goias Pocos de Caldas Araxa	Monazite sands Volcanic intrusives	73 000 350	90-170 (streets) 90-90 000 110-1 300 340 average 2 800 average
China	Yangjiang Quangdong	Monazite particles	80 000	370 average
Egypt	Nile delta	Monazite sands		20-400
France	Central region Southwest	Granitic, Schistous, sandstone area Uranium minerals	7 000 000	20-400 10-10 000
India	Kerala Madras Ganges delta	Monazite sands, Coastal areas 200 km long, 0.5km wide	100 000	200-4 000 1 800 average 260-440
Iran	Ramsar Mahallat	Spring waters	2 000	70-17 000 800-4 000
Italy	Lazio Campania Orvieto town South Toscana	Volcanic soil	5 100 000 5 600 000 21 000 100 000	180 average 200 average 560 average 150-200
Switzerland	Tessin, Alps, Jura	Gneiss, Verucano, Ra-226 karst soil	300 000	100-200

The origin of radionuclides in domestic water sources can be traceable to the trace deposits of naturally occuring or artificial radioactive material that inevitably exist within the environment. Their presence in form of NORM, that is sometimes technically enhanced, resulting to what is generally termed as TENORM (Technically enhanced NORM), occur mainly in domestic water sources due to contamination as a result of leaching of minerals in the earth crust while the artificial enter the aqueous media mainly through waste disposal practices, spills, and land application of chemicals. These contaminants vary in concentrations in water sources are heavily depending on hydrogeological conditions as well as human activities. The years, the behaviour of radionuclides in water, soils and sediments has been the subject of considerable scientific interest and numerous investigations were carried out as a result. These investigations provide some basic understanding of radionuclide distribution and dynamics in lakes and rivers, as well as in their respective catchments, in different hydrogeologic systems and geographic regions. A number of researches conducted confirms that existance and mobility of radionuclides through the surface and the ground water systems is dependent on the physical and chemical properties of the contaminant, and on the rock and sediment characteristics (Garcia-Sanchez, 2008, Jeffree *et al*., 2007, Semizhon *et al*., 2010). Radionuclides that commonly occur in the aqueous phase were found to be very mobile within the aquatic environment thereby distributing their concentrations. In some cases, radionuclides strongly interact with the particulate matter suspended in water and the bottom sediments and consequently transported via flowing water (Yaron et al., 2010).

Most of the drinking water sources for the inhabitants of northern peninsular Malaysia comes from the abundant surface water resources in the area. Radionuclides of natural origins usually find their way into surface water as a result of leaching of the earth crust. The earth crust contains small amounts of uranium, thorium and radium as well as radioactive isotopes of uranium. Similarly, a number of radionuclides find their way into the drinking water sources due to human activities of agriculture, medicine and industry (Larry, 1996). Uranium is one of the most abundant radionuclides in the surface of the earth, and water constitutes the principal route by which uranium is incorporated into humans, due to its commonly high solubility. In general, a wide range of radionuclides are known to occur in water, these include cesium-137, chromium-51, cobalt-60, iodine-131, iron-59, lead-210, phosphorous-32, plutonium-238, radium-226 radon-222, ruthenium-106, scandium-46, strontium-90, thorium-232, uranium-238, zinc-65 and zirconium-96 (Larry, 1996).

The potential health hazards associated with inhalation and ingestion of radionuclides through indoor air and drinking water respectively have been well established, with many countries adopting the guidelines of activity concentration (MCL) recommendations of the World Health Organization (WHO, 1988, Ajayi and Achuka, 2009, Fatima *et al*., 2007). Studies of environmental activity concentration levels often utilize the much debated linear no threshold (LNT) hypothesis on radiation risk. This hypothesis provides the fundamental basis used in predicting the risk associated with radiation exposure, LNT model also gives the basis for radiation protection practices (ICRP, 1991). Dose limitation associated with human exposure is a

direct reflection of the assumption that risk is proportional to total dose, without a threshold as depicted by the LNT model. Though the concept of LNT is considered to be 'hypothetical' due to lack of evidence to support the assumption that irrepairable biological damage occurs even at very low doses, the LNT hypothesis has been adopted by every national and international body that offers radiation protection recommendations or interprets radiological data. Bodies such as the ICRP, the NCRP, IAEA, and the UNSCEAR all adopted the LNT model. No serious radiological health effects are expected from consumption of drinking-water if the concentrations of radionuclides are below the guidance levels. The LNT dose-response relationship reveals that there is no safe radiation dose; accumulation of low doses due to radionuclides (such as those found in contaminated water) in the body overtime may lead to several types of radiological health effects such as reduced blood cell counts, vulnerability to kidney damage leading to renal dysfunction as well as chromosomal aberration which may later lead to carinogenic effects (Briner, 2010).

One important instance associated with radiation in humans is the ionization of the abundant water molecules in the body by the high Linear Energy Transfer (LET) nuclides leading to the production of highly reactive species such as H_2O_2 as depicted by the radiolysis of water shown in Figure 2.4. Figure 2.4 expresses how exposure due to ionizing radiation induces high-energy radiolysis of water molecules into hydrogen and hydroxyl radicals, which are known to be highly reactive species. These reactive species recombine to produce a series of highly reactive combinations such as

superoxide (HO_2) and peroxide (H_2O_2), which produce oxidative damage within the cell.

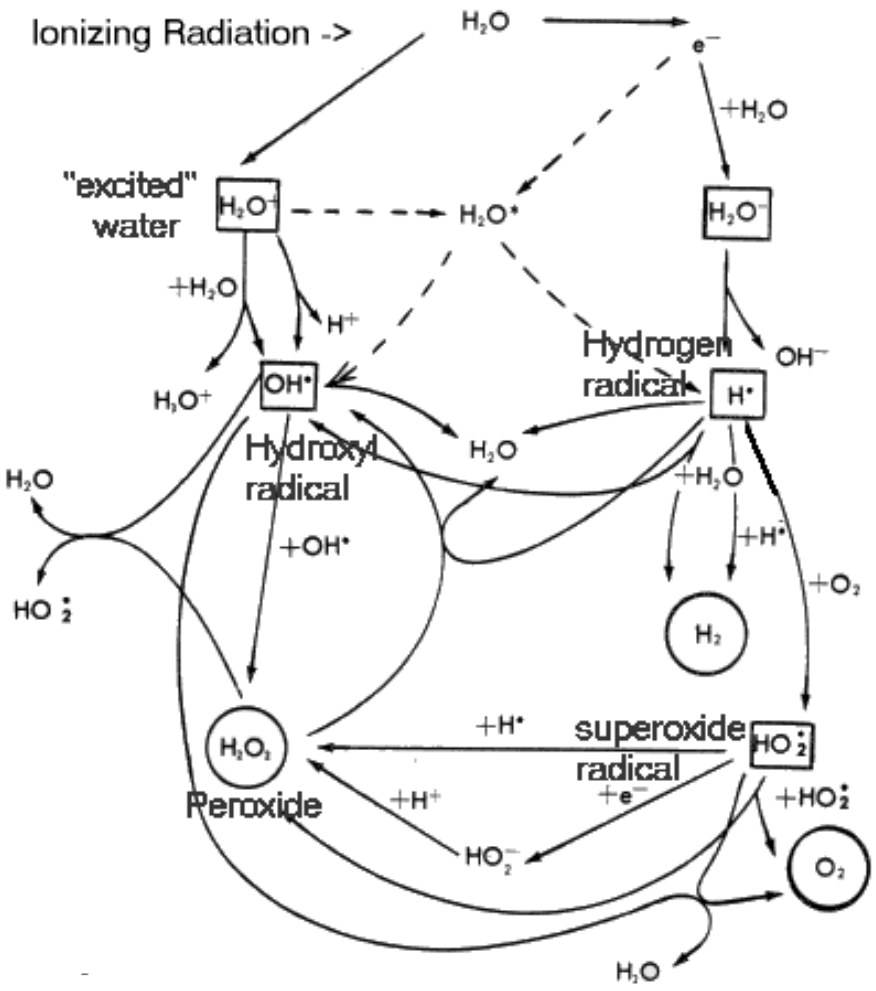


Figure 2.4 Radiolysis of water

The radionuclides, which are known to decay by alpha, beta and other emissions pose hazards when ingested into human body through drinking water. Alpha particles, for instance, are among the most hazardous forms of radiation within human tissue. They have high LET, meaning that they loose their energy within a very short distance in dense media thereby causing significant damage to the surrounding biomolecules when ingested into the body. A typical 5.8 MeV alpha emission from ^{222}Rn which is by hundreds of

thousands times more energetic than the weak hydrogen bond between the purine and pyrimidine bases of the DNA, it is similarly more energetic than all the molecular binding energies of the phosphoric acid backbone of the DNA and could therefore cause devastating result, on molecular scale when deposited into the nucleus. Studies indicated that exposure to radiation at low and moderate doses, which is typical for public exposure from water and other environmental media, can cause stochastic detrimental health effects in form of malignancies and hereditary effects. Both human and animal studies indicates that radiation exposure at low to moderate doses may increase the long-term incidence of cancer. Animal studies in particular suggest that the rate of genetic malformations may be increased by radiation exposure.

The world annual natural radioactivity level, including external exposure, consumption of food and water containing natural radionuclides, and inhalation of radon with its daughter products, amounts on average to 2.4 mSv, with a range from 1 to 10 mSv (UNSCEAR, 2000b). The UNSCEAR report also indicate that the average worldwide exposure to natural sources in foods and drinking water is 0.29 mSv y^{-1} (about 0.17 mSv y^{-1} from K-40 and about 0.12 mSvy^{-1} from U-238 and Th-232) (UNSCEAR, 2000b). The international recommendation of annual dose limit for controlled exposures of the general public, above the annual natural radiation background is 1 mSv (Wrixon, 2008, WHO, 1988). For prolonged exposure situations in case of all environmental radioactive sources, including natural and human-made sources, the ICRP recently recommended a generic intervention level of annual dose of 10 mSv as the level below which intervention is unjustified, taking into account radiological, economic and social factors (Wrixon, 2008).