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NATURAL RADIOACTIVITY LEVEL IN SOIL, CROPS, RIVER SEDIMENTS, AND SELECTED AQUATIC SPECIES IN SOUTH AFRICA'S OIL-PRODUCING AREAS

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By

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ABSTRACT

This research was conducted to estimate natural radionuclide contents in various environmental samples collected from farmlands and rivers of South Africa's oil and non-oil-producing areas. The radiological analysis was also performed to assess the effect of radiation on the population that consumed these crops and fish species. A high-resolution Hyper Pure Germanium (HPGe) detector was used to conduct the gamma-ray measurements for all the well-prepared samples collected.

Bree, Klein-Brak, and Bakens rivers were selected from the oil-producing areas for the fish and river sediment samples. The uMngeni river was selected for the non-oil producing area serving as the control. Philippi, Uitenhage, and Hartenbos farms were selected for the oil-producing areas for the farm soil and crop samples. In comparison, the Ukulinga farm was selected for the non-oil producing area.

The mean activity concentrations for the selected fish samples from the examined areas for ^{238}U, ^{232}Th and ^{40}K ranged from (8.60 ± 2.97), (4.26 ± 1.18), (105.66 ± 47.77) $Bq.kg^{-1}$; (8.06 ± 3.64), (4.84 ± 2.00), (126.88 ± 47.30) $Bq.kg^{-1}$; (8.30 ± 3.64), (3.48 ± 1.44), (90.42 ± 29.35) $Bq.kg^{-1}$; (6.48 ± 2.05), (5.26 ± 1.79), (78.38 ± 20.55) $Bq.kg^{-1}$ for the Bree; Klein-Brak; Bakens, and uMngeni rivers. The annual effective ingestion dose ranged from 0.050 $mSv.y^{-1}$ to 0.100 $mSv.y^{-1}$; 0.033 $mSv.y^{-1}$ to 0.118 $mSv.y^{-1}$; 0.034 $mSv.y^{-1}$ to 0.090 $mSv.y^{-1}$; 0.046 $mSv.y^{-1}$ to 0.082 $mSv.y^{-1}$ for the Bree, Klein-Brak, Bakens, and uMngeni rivers.

Also, the estimated values for the annual equivalent dose of gonads, bone marrow and bone surface cells due to ingestion of fish samples ranged from 58.77 to 127.27 $mSv.y^{-1}$; 42.14 to 125.94 $mSv.y^{-1}$; 39.34 to 84.97 $mSv.y^{-1}$; 54.54 to 71.97 $mSv.y^{-1}$ for Bree; Klein-Brak; Bakens, and uMngeni rivers.

The results of the study indicated that the mean activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in the sediment samples from the oil-rich areas are 11.13, 7.57, 22.5 $Bq.kg^{-1}$; 5.51, 4.62, 125.02 $Bq.kg^{-1}$; 7.60, 5.32, 24.12 $Bq.kg^{-1}$ and 4.13, 3.28, 13.04 $Bq.kg^{-1}$for the Bree, Klein-Brak, Bakens, and UMngeni rivers. The average excess lifetime cancer risks are 0.394 x 10^{-3}, 0.393 x 10^{-3}, 0.277 x 10^{-3}, and 0.163 x 10^{-3} for sediment samples at Bree, Klein-Brak, Bakens, and uMngeni rivers.

The mean activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the farm soils were 30.71, 31.97, 345.97 $Bq.kg^{-1}$; 18.67, 31.55, 191.93 $Bq.kg^{-1}$; 38.03, 41.18, 381.89 $Bq.kg^{-1}$; 8.47, 8.65, 94.22 $Bq.kg^{-1}$ for Philippi, Uitenhage, Hartenbos, and Ukulinga farms.

The mean activity concentration of ^{226}Ra, ^{232}Th and ^{40}K for crop samples are 4.54 ± 1.47, 4.87 ± 1.69, 140.18 ± 35.38 $Bq.kg^{-1}$; 9.17 ± 4.79, 3.85 ± 1.87, 136.75 ± 22.04 $Bq.kg^{-1}$; 7.97 ± 2.91, 4.62 ± 2.40, 105.97 ± 48.65 $Bq.kg^{-1}$; 4.23 ± 1.63, 2.72 ± 1.19, 48.36 ± 15.55 $Bq.kg^{-1}$ for Philippi, Uitenhage, Hartenbos, and Ukulinga farms.

The activity concentration and soil-to-crop transfer factors for ^{40}K were found to be much higher, possibly because this element is critical in crop growth. The results showed that the crop samples' transfer factor is in the order cowpea>potato>maize.

The activity concentrations and radiological dose estimates reported for the studied areas were within the values of 33, 45, and 450 $Bq.kg^{-1}$ for ^{226}Ra (^{238}U), ^{232}Th, and ^{40}K recommended by the United Nations Scientific Committee on the Effects of Atomic Radiation. However, all obtained values indicated a significant difference between the natural radionuclide concentrations in the samples from the in oil-rich areas compared to that of

the non-oil-rich area. Hence, the result of the activity concentrations in the selected environmental samples at the time of this study does not pose any radiological risks. The results of this research may be used as a benchmark and reference data for future investigations.

PREFACE

This thesis describes the work undertaken by Abiola Olawale Ilori in the School of Chemistry and Physics, the discipline of Physics, University of KwaZulu-Natal, Pietermaritzburg Campus, South Africa under the supervision of Prof. Naven Chetty.

This thesis's contents are my original work unless otherwise indicated where due reference has been supplied in the text to use others' work. No part of this thesis has been submitted in part, or whole, to any other University for degree purposes.

Abiola Olawale Ilori (Student): Signature [redacted] Date: 19-11-2020

Dr. Oluwabamise Adeleye (Co-supervisor): Signature [redacted] Date: 29-01-2021

Prof. Naven Chetty (Supervisor): Signature [redacted] Date: 30-01-2021

DECLARATION: PLAGIARISM

I, Abiola Olawale Ilori, declare that:

[1]. The research results reported in this thesis, except where otherwise indicated, are my original research results.

[2]. The thesis has not been submitted in full/in part for any degree or examination to any other university (s).

[3]. This thesis does not contain data, pictures, or graphs from other persons unless acknowledged/cited as originating from others.

[4]. This thesis does not contain other persons writing unless expressly acknowledged as being sourced from other researchers. Where other written sources have been quoted, then:

(a) Their words have been re-written, but the general information attributed to them has been referenced.

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[5]. Where I have reproduced a publication of which I am an author, I have indicated which part of the publication was contributed by me and have acknowledged other authors' contributions.

Signature (Student):  …… **Date**: …19-11-2020…

DECLARATION: 4-PUBLICATIONS

I, Abiola Olawale Ilori, with this, declare the details of contribution to publications that form part of the research presented as chapters in this thesis.

i Abiola Olawale Ilori, Naven Chetty & Bamise Adeleye (2020): Radiological dose assessments of fish samples due to the presence of NORMs in oil-rich areas of South Africa. *Environmental Forensics*, https://doi.org/10.1080/15275922.2020.1806150

ii Abiola Olawale Ilori, Naven Chetty & Bamise Adeleye (2020): Activity Concentration of Natural Radionuclides in Sediments of Bree, Klein-Brak, Bakens, and uMngeni Rivers and it's Associated Radiation Hazard Indices. *Transactions of the Royal Society of South Africa*, https://doi.org/10.1080/0035919X.2020.1815894

iii Abiola Olawale Ilori & Naven Chetty: Soil-to-crop Transfer of Natural Radionuclides in Farm Soil of South Africa. *Environmental Monitoring and Assessment*, https://10.1007/s10661-020-08756-7

iv Naven Chetty & Abiola Olawale Ilori: Estimation of Natural Radionuclides and its Radiological Hazard Assessment in South Africa's Farm Soils (*under peer review*).

Signature (Student): ..[redacted]..... **Date**: ...19-11-2020...

CONFERENCE CONTRIBUTIONS

[1]. Abiola Olawale Ilori, Naven Chetty, Bamise Adeleye. Radionuclides in Fish Samples from the Oil-producing Areas of South Africa and their Dose Assessments. CUKUROVA 5th International Scientific Researches Conference, Adana, Turkey (Oral Presentation through Zoom), October 9-11, 2020.

[2]. Abiola Olawale Ilori, Naven Chetty. Transfer of Naturally Occurring Radionuclides from Soil to Indigenous Food Crops of South Africa's Oil Producing Areas. Flash Presentation at the Postgraduate Research & Innovation Symposium (Online), College of Agriculture, Engineering and Science, University of KwaZulu-Natal, South Africa, December 10-11, 2020.

Signature (Student):  …… **Date**: …19-11-2020…

DEDICATION

This work is dedicated to my wife, Mofoluwasho, and my children: David, Davina, and Dasha.

For their love, support, and encouragement.

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LIST OF ABBREVIATION AND SYMBOLS

ALARA	As Low As Reasonably Achievable
$Bq.kg^{-1}$	Becquerel per kilogram
Ci	Curie
DNA	Deoxyribonucleic acid
FAOSTAT	Food and Agriculture Organization Corporate Statistical Database
FBS	Food Balance Sheets
Gy	Gray
HPGe	High Purity Germanium Radiation Detector
IAEA	The International Atomic Energy Agency
ICRP	International Commission for Radiological Protection
^{40}K	Potassium
LSA	Low Specific Activity
MCA	Multichannel Analyzer
NORM	Naturally Occurring Radioactive Materials
NRC	National Research Council
PMT	Photomultiplier Tube
^{226}Ra	Radium
SAES	South African Energy Sector

Sv	Sievert
TENORM	Technologically Enhanced Naturally Occurring Radioactive Materials
^{232}Th	Thorium
UNSCEAR	United Nations Scientific Committee on the Effects of Atomic Radiation
USEPA	The United States Environmental Protection Agency
^{238}U	Uranium
WHO	World Health Organization
τ	The mean or average life of a radioactive atom
λ	Half-life
α	Alpha particles
β	Beta particles
γ	Gamma rays

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CHAPTER 1

INTRODUCTION

1.1 Background of the study

The occurrence of naturally occurring radioactive material (NORM) in the atmosphere with activity concentrations higher than the reference standards recommended by the United Nations Scientific Committee on the Effects of Atomic Radiation has been in recognition to be harmful to humans and the environment [1-6]. One of the United Nations' targets for sustainable food security is to ensure access to affordable, adequate nutrition, and healthy food for everyone [7-8]. The petroleum industry has been one of the largest importers and consumers of radioactive materials that caused radioactive waste to be released into the environment [9].

There have been reports of these impacts on achieving the United Nation's environmental sustainability goals [10]. Significant studies have been conducted worldwide to determine the activity levels of naturally occurring radioactive materials at different locations in soil, marine organisms, and sediments [2, 11-16]. Such research has shown that a significant quantity of natural radionuclide and their decay elements constitute the materials that make up the atmosphere [1, 17].

The leading external cause of human body irradiation is the terrestrial background radiations released from the naturally occurring radioactive materials [1]. The environmental assessments of naturally occurring radioactivity activity levels provide us with vital information on the concentrations of radioactive elements in the local environment [18]. Assessing exposure to the population living in an environment suspected of being too or moderately radioactive is of great importance. It plays a vital role in exploring radioactivity-related health risks. Hence, its future responses to environmental radioactivity from human activities, especially industrialization, are guaranteed [19].

Radiation from external exposures and radionuclides, absorbed via internal exposures, is the significant way humans are exposed to radiation. These exposure sources come either from cosmic rays emitted from outer space, the sun's surface, or terrestrial radionuclide in Earth's crust [1, 20]. Conclusions could then be made that radioactivity is part of everyday life because it is inhaled and consumed every day. Food crops widely grown and consumed in South Africa include maize, wheat, sugar cane, sunflowers, etc. Many fish and shellfish such as kingklip, snoek, red roman, hake, cod, sole, tuna caught in South African waters are also widely consumed. Hence, fish are not the only foodstuffs consumed by the man from the aquatic environment; many invertebrates are also consumed. Many aquatic species are harvested for direct consumption or other products used in the manufacture of various foods. Not

all are consumed directly but are used as a fish meal fed to domesticated animals [21, 22].

The direct and indirect radiation exposure pathways due to human consumption of foods containing radioactive elements lead to both external and internal radiation exposure. The description of the paths leading to external and internal exposure of man is shown in Figure 1.1. It will affect the communities' health and economies in these regions and the public outside those population that eat the aquatic species. Information on yearly quantities of foodstuff consumed is obtained from the document on food consumption per capita in South Africa given on FAOSTAT food balance sheets (FBS) [22].

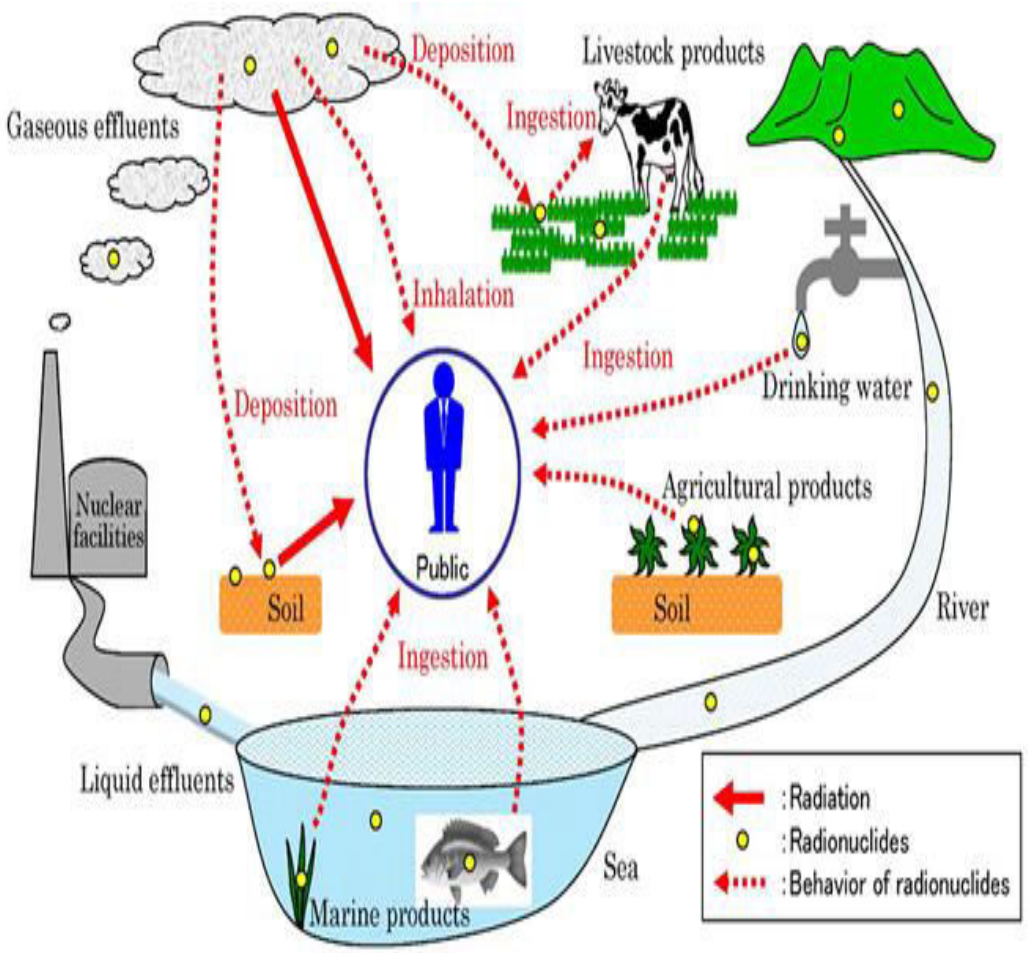

Fig. 1.1: Exposure pathways due to radionuclides released from nuclear plants [23].

Natural radioactivity in the soil results in human exposure, both internal and external [3, 24], and people are not aware of the sources of radiation within their environments. Humans have also introduced natural background radiation to specific artificial sources in the last century [25]. Naturally occurring radiation sources have about four to five times the quantity compared to the human-made sources [26]. Organisms have continuously been exposed to cosmic rays through radionuclide produced by cosmic ray collisions and ionizing radiation spread across all living and non-living components of the environment. Modern-day life has adapted to all environmental variables and restrictions, including natural radiation in the background.

While high radiation levels are harmful to organisms, some environmental radiations, such as background radiation is essential to life. This has contributed to the fundamental processes of chemical and biological evolution [1, 17, 20]. The main concern is that the Earth's heat content is primarily supplied and retained by the heat of primordial radionuclides decay [1, 17].

The primary sources of ionizing radiation are cosmic radiation and terrestrial radiation. Cosmic rays also provide 13 percent natural external exposure to Earth's gamma radiation, giving 15 percent natural external radiation exposure [27]. A small proportion of the exposure may seem insignificant; however, every measure must control the radiation released into the environment. At higher altitudes, the atmosphere that shields cosmic rays decreases, leading to increased radiation dose exposures to the environment [28].

The infinite number of stars in the universe is the possible origin of cosmic rays, as shown by the increased strength of cosmic rays observed on Earth from solar flares [29]. Cosmic ray interactions with matter continuously create many radionuclides in the atmosphere [29]. A few of these radionuclides are released as fragments and others through the activation of stable neutron atoms. The radionuclides produced from cosmic rays are ^{10}Be, ^{36}Cl, ^{14}C, ^{3}H, ^{22}Na, ^{39}Cl, etc. [30]. The predominant radiation source found in natural

materials such as soil, rocks, construction materials, and drinking water is terrestrial radiation, also known as primordial radiation. Many contributors to these terrestrial radionuclide sources include natural radium, uranium, and thorium [17, 20].

Notwithstanding the ubiquity of terrestrial radiation, their concentrations differ considerably with location [26]. The distribution of primordial radionuclides and their progeny in natural environments is influenced by many factors, including the chemical properties of nuclides, physical environmental conditions, ecological, and the corresponding environmental attributes [31]. The consumption of crops and animals contributes a significant pathway to radionuclide concentration ingested by high radioactive environments' dwellers. The radiation dose depends on the food's radiation concentration and the quantity consumed [20, 32].

1.2 Research motivation

The extensive use of ionizing radiations throughout the world, particularly in the medical area and in the industry, such as petroleum industries, agriculture, and research, causes significant concern [25]. While South Africa has minimal proven oil and gas reserves, its oil and gas exploration started as far back as 1913. However, oil production and research drove years ago in the Orange Basin off the west coast and the Bredasdorp Basin on the south coast of Cape Province of South Africa [33]. It is due to the well-developed infrastructure of South Africa and the crude oil reserves in the region. The extent and rate at which this industry is implementing modern technology in South Africa are very significant.

The oil industry is one of the largest importers of radioactive materials in the economic sectors [33]. There are numerous radioactive contaminants in the environment from radioactive materials in the industry that include nuclear

well-logging, nuclear density gages, radiography, oil well maintenance radiotracers, and pipeline leakages [14, 34]. Crude oil extraction and other human activities are known to boost natural radionuclides in an environment [34, 35].

Given numerous precautions based on accident, mishandling of equipment, inappropriate discharge, loss, and theft, the radioactive materials from natural and artificial sources may pollute the oil-producing region's air and the aquatic environment. The environmental contamination caused by these radioactive materials can lead to both internal and external sources of radiation to aquatic organisms through various routes, which can inevitably impact the economy and health of the residents of the oil-producing regions [16, 18, 31].

One of the United Nations' goals is to provide sustainable food security to ensure that all people have access to adequate, nutritionally sufficient, and nutritious food [7, 8]. Therefore, due to the oil production industry's operations by the heavy use of radioactive substances, the concentrations of radioactive elements in the atmosphere would have been increased [36]. These radioactive substances make their way into humans, mostly through food and water, in the natural environment [37]. It is a known fact that the condition of the soil on which food crops are grown mostly determines the quality of the food produced [38, 39].

1.3 Aim and objectives of the thesis

The petroleum industry's operational activities across the Orange Basin off the west coast and the Bredasdorp Basin on the south coast of South Africa's Cape Province include radioactive materials of different proportions and chemical compositions half-lives. Despite the numerous safety precautions to guarantee protection in applying radionuclides in the oil sector or any industry, there is always a finite possibility of accidents, abuses, and leakages during which the

environment may be polluted [34]. Therefore, the aquatic animals are exposed to radiation sources, and their lives are threatened due to the ionizing radiation's detrimental impact. The exposures to radioactivity may affect the health and economies of the regions and the public outside those consuming the aquatic species.

This study aims to take samples of soil, crops, river sediments, and selected fish samples in the rivers at South Africa's oil-producing and non-oil producing area and analyze them for their radioactivity concentrations.

The objectives of the analyses are to:

(i) Measure the activity concentration of NORMs in river sediments and selected fish samples from the studied areas.
(ii) Evaluate the radiation hazard indices from the sediment and fish samples relatives to human health due to radionuclides in the aquatic environment.
(iii) Measure the activity concentration of NORMs from the farm soil and locally grown crops within the study area.
(iv) Assess the impact of radionuclide in the farm soil and crops and the implications for the human population through the radiological dose assessments.
(v) Compare the radionuclides' level within the studied areas with the average world average values and those of other countries.

1.4 Organization of the thesis

The organization of this thesis has been structured into seven chapters. Chapter 1 gives the background to the research work, the research motivation, and the study's aim and objectives. Chapter 2 provides a comprehensive theoretical knowledge as a literature review on radiation, its sources, and the principle of its detection, measurements, and protection. It discussed the bio-effects of radiation exposure and an overview of the application of radionuclides

in oil exploration. Chapter 3 is a published journal article concentrating on radiological dose evaluations of fish samples due to the presence of NORMs in South Africa's oil-rich areas. It was carried out to estimate the natural radionuclide content of fish samples collected from rivers in oil-rich and non-oil-rich areas of South Africa. The effect of radiation on the population eating the fish was determined through radiological analysis. Chapter 4 is a published journal article that focuses on the activity concentration of natural radionuclides in sediments of Bree, Klein-Brak, Bakens, and uMngeni rivers and its associated radiation hazard indices. Radiation hazard indices were estimated and excess lifetime cancer risk. Chapter 5 is a journal article under a peer review that focuses on the soil-to-crop transfer of natural radionuclides in farm soil of South Africa. In the study, the activity concentration of natural radionuclides in farm soil and the most common indigenous food crops in oil-producing (Philippi, Uitenhage, Hertenbos farms) non-oil producing (Ukulinga farm) areas of South Africa was measured.

Consequently, the transfer of these radionuclides from soil-to-crops was estimated. Chapter 6 is a journal article under a peer review that focuses on the estimation of natural radionuclides and its radiological hazard assessment in South Africa's farm soils. The radiological hazard assessments of radium equivalent, the absorbed dose rate, the annual effective dose rate, and the representative level index for all the study samples were calculated. Finally, Chapter 7 summarizes the study's significant findings, draws a precise conclusion, and provides recommendations for future work.

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CHAPTER 2

LITERATURE REVIEW

2.1 Radiation

Transmission of energy in waves via space or tangible medium is called radiation [1, 2]. The emission of this energy as electromagnetic waves causes ionization, particularly high-energy ones [3]. Radiation comprises electromagnetic radiation (radio waves, microwaves, infrared, visible light, ultraviolet), X-rays, gamma radiation, alpha, beta, neutron radiations, acoustic radiation (ultrasound, echo, seismic waves), and gravitational radiation [4, 5]. Therefore, radiations are generated from the influence of waves passing in all directions from an origin [3].

Wilhelm Conrad Roentgen originally discovered X-rays radiation in 1895, after which the X-ray was incorporated into the detection and treatment of diseases [6, 7]. After discovering radioactivity, some accidents from prolonged exposure to radiation were identified immediately in pioneer radiation employees,

unaware of the possibility of such impact [8]. The first of these injuries were mainly skin reactions on the hands and several other forms of injuries documented within a decade, including cancer due to radiation [6-8].

Radiation is classified as either ionizing or non-ionizing based on the radiated particles' energy and their effects on materials [9, 10]. The primary difference is that ionizing radiation has sufficient energy to ionize atoms while not ionizing radiation. Although radiation cannot be felt, it is rapidly detected and measured to measure exposure [11].

2.1.1 Ionizing radiation

Ionizing radiation originates from atoms' nuclei and is the basic building block of matter [9, 11]. Individual atoms disintegrate into entirely new particles making them unstable, as such are radioactive. The unstable atoms release energy in the form of gamma rays as radiation. As this nucleus decays with alpha or beta particles' emissions, it becomes a new entity that can emit gamma rays simultaneously [12]. In different stages, the atoms progress to a stable state where they are no longer radioactive. Ionizing radiation is carcinogenic due to the interaction between environmental factors, such as smoke or radiation, and our makeup as determined by our genes [13].

Two protons and two neutrons are composed of alpha particles, doubly charged (coming from the two protons being charged). In line with the relatively slow speed and high mass of alpha particles, this effect means that they interact with matter more readily than beta particles or gamma rays and lose their energy rapidly. There is little penetrating ability for an alpha particle that can be prevented by the first skin surface or paper sheet. An Alpha particle can, however, cause more significant harm to the body than other forms of radiation [4, 5, 13].

Beta particles are the fast-moving electrons of several kinds of radioactive atoms ejected from the nuclei [14]. Such particles are charged individually and ejected at much higher velocities than alpha particles. However, a few millimeters thick layers of aluminum could indeed stop them [13, 14].

Gamma rays represent energy transmitted without material movement, heat, and light activity within a wave [4]. X-ray radiation originates from the transitions between electrons whereas Gamma-rays are generated by nuclear transitions. In addition, gamma-rays have in general shorter wavelengths and therefore, higher energies as compared to X-rays. Thus, gamma rays, unlike light, have a massive penetration and can pass through human flesh. Thick, dense concrete, lead, or water shielding is necessary to protect humans against gamma rays [9, 15].

2.1.2 Non-ionizing radiation

Electromagnetic radiation is known as non-ionizing radiation, which is the energy propagated through a material medium in the form of electromagnetic waves, such as radio waves, visible light, and gamma rays [16, 17]. It is a collection of energy waves consisting of electric or magnetic oscillatory fields that travel at the speed of light. Non-ionizing radiation is a form of electromagnetic radiation, a natural component of the atmosphere by which man is almost adapted. However, a change in radiation's intensity and character can adversely affect living organisms [18, 19]. Rather than an atom which produces charged ions when it passes through a matter, the electromagnetic radiation seems to have enough excitation energy. Moving an electron to a higher energy state requires electrical and magnetic fields, radio-waves, microwaves, infrared, ultra-violet, and visible radiation [13].

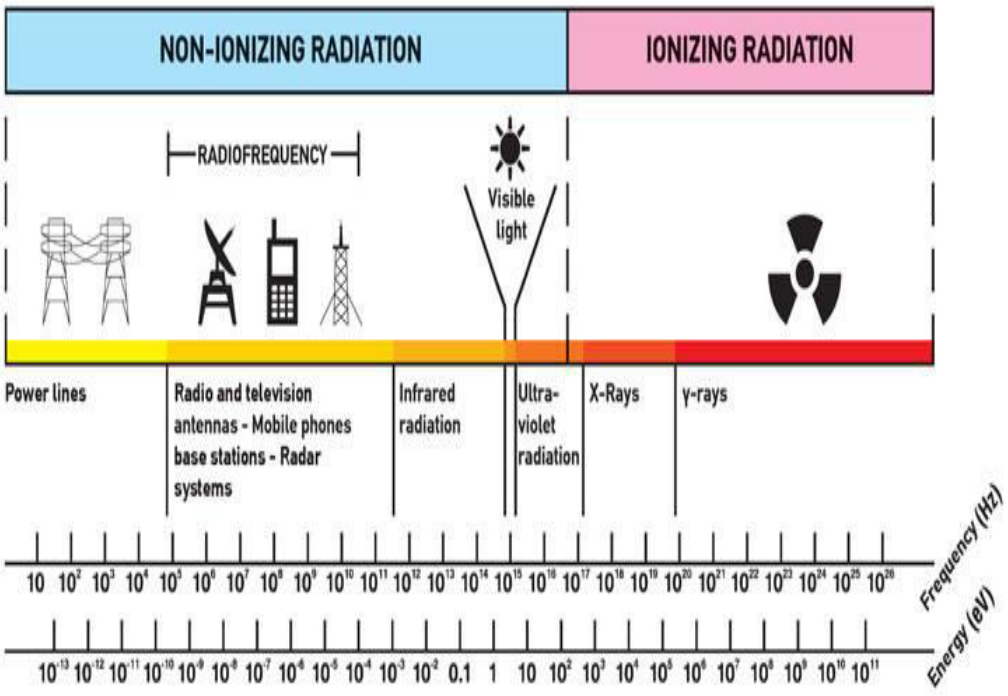

Fig. 2.1: Categories of ionizing and non-ionizing radiation [20].

2.2 Sources of radiation

After discovering radioactivity in 1896, an extensive study was conducted on the science of radiation by Henrie Becquerel [21, 22]. Several radioactive sources have been here since the planet was created, although many artificial sources were added to those natural sources in the last century. Radionuclides release radioactive waste that has become a part of our daily lives [23]. The radiation properties have been widely applied to different aspects of human life, including medicine, industry, electricity generation, and agriculture [24]. Humans may be exposed to radiation emitted from various radioactive sources, depending on their activities and environment [25]. The ionizing radiation that results from radionuclides on the earth's surface and cosmic rays in the earth's

atmosphere is the most prominent radiation source to which all people are exposed [9, 13, 20].

Natural sources have contributed four to five times as much as artificial sources have contributed [9, 26]. Natural radiation in the atmosphere is also the most important cause of human radiation exposure [26]. Human exposure to natural radiation accounts for around 85% of the annual radiation exposure dose [12, 13]. Exposure to natural radiation remains of little concern to the public except for those working with mineral ores and radioactive materials [27, 28]. However, the possible danger to human health is posed by any radiation dose. Simultaneously, the level of personal exposure to naturally occurring radioactive elements is typically statistically insignificant at the individual level [27]. Recently, the International Atomic Energy Agency (IAEA) has primarily focused on protecting the public from the risk of radiation resulting from natural radiation [26-28].

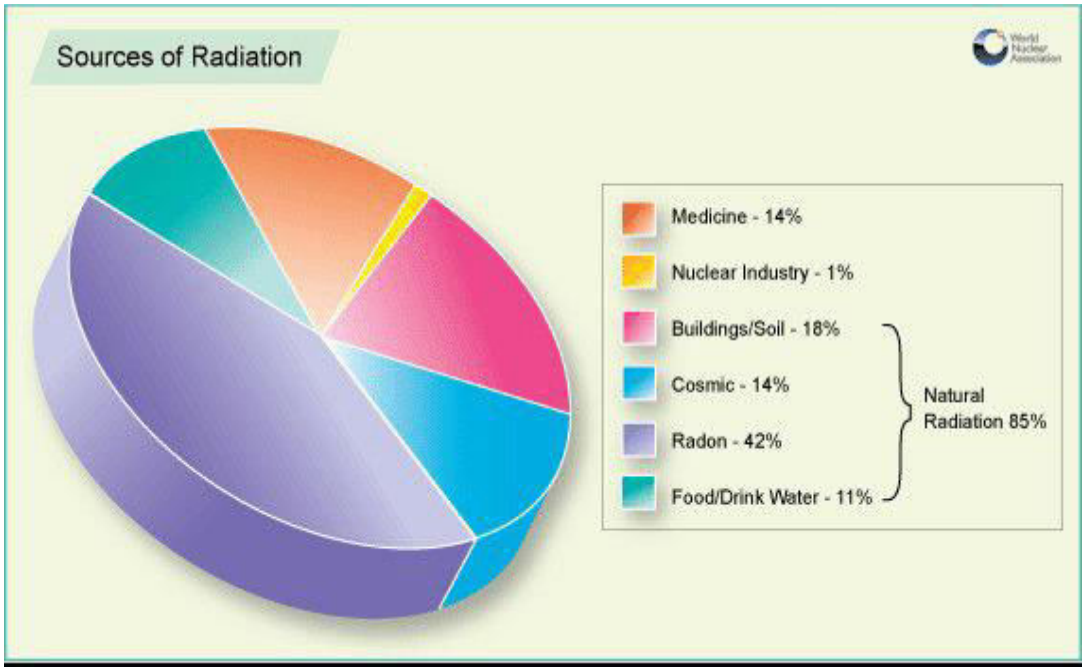

Fig. 2.2: Mean annual worldwide exposure of radiation from various sources [27].

2.2.1 Natural sources

It is essential to consider the degree of radioactive material that an individual receives from the natural environment since radiation can be found in soils, air, and water that human consumes [28]. Natural sources formed their ways, even in the building materials and items we commonly use. Cosmic, terrestrial, and internal radiation sources are the three primary sources of naturally occurring radiation exposure to humans.

Both the primary energetic protons and extraterrestrial alpha particles that hit the earth's atmosphere come from cosmic radiation. It also comes from the cosmic rays by bombarding stable nuclides in the atmosphere. The amount of ozone that blocks cosmic rays reduces and thereby increases the dose. The near-infinite numbers of stars in the universe are the potential cause of cosmic radiation [29-30].

Terrestrial sources are natural sources of radiation in the soil, minerals, construction materials, and drinkable water referred to as primordial [16]. Even though terrestrial radiations (primordial radionuclides) are ubiquities, their concentration varies substantially with location [16, 28]. The primordial distribution of radionuclides is influenced by many factors, including chemical composition, physical elements, ecological attributes, and biota physiology. The primary source of internal terrestrial exposure is from Potassium-40, which declines to Calcium-40 and Argon, both positively and negatively beta-decay [31]. These reach the body by ingesting foodstuffs or drinking water in which radionuclides are distributed in varying quantities of traces. Our bodies as living beings emit radiation; that is, our bodies contain natural radionuclides.

Of these natural radionuclides, the principal factors to the annual dose rates are ^{40}K (Potassium), ^{208}Tl (Thallium) and ^{228}AC (Actinium) in the ^{232}Th (Thorium) series and two short-lived decay products of ^{222}Rn (Radon), namely ^{214}Pb (Lead) and ^{214}Bi (Bismuth) in the ^{238}U (Uranium) series [32]. The natural radiation level is dependent on the soils in the region and the types of rock

from which the rock originates. Both isotopes from uranium and thorium have half-life equivalent to the earth's age and are therefore abundant. ^{238}U isotope is the most prevalent in naturally occurring uranium, with a ratio of 99.28 percent to others [33].

Worldwide average activity concentrations of radionuclides resulting from terrestrial radionuclides are 30, 400, and 35 $Bgkg^{-1}$ for ^{232}Th, ^{40}K, and ^{238}U, respectively [13]. About 95 percent of the world's population lives in places with absorbed air exposure levels ranging from 30 to 70 $Gy.h^{-1}$ [12, 13, 34]. However, there were reports of several areas having dose rates that are significantly higher than the estimated background value [35].

A body's internal exposure to radiation is via ingestion of food from drinking water and radionuclide inhalation. Of these radionuclides, which are sources of internal exposure, the most important is ^{40}K and the decaying daughter of ^{232}Th and ^{238}U. Potassium (^{40}K) is known to constitute the primary naturally occurring source of internal radiation; it is found everywhere in both living and non-living matters [36].

2.2.2 Artificial sources

Artificial sources emanating from artificial radionuclides augment natural radiation in the environment [37]. Human has artificially introduced several hundred radionuclides into the atmosphere over the past decades, while effects from artificial radioactive materials differ widely. For artificial sources, this variation is usually more significant than for the natural sources. The artificial radioactive materials were predominantly the result of medical x-ray equipment.

Many artificial radiation materials come from nuclear weapons, nuclear plants, television, and much other radiation-producing equipment [9, 13, 24, 38]. The use of nuclear sources by man has been in existence for a long. For medical diagnosis and therapy, artificial and natural radioactive elements have been used efficiently. Patients and medical staff are subjected to ionizing radiation

and subsequent exposures using an x-ray for treatment and therapy. Most of these are generally not available to the public. Still, they may find their way into the environment through infringements of transport regulations, routine releases, accidents, thefts, losses, misapplications, or disposal. Like the naturally occurring sources of radiation that are commonly spread around the world, artificial sources are typically found when it affects a fraction of the population.

Research and technology in some fields have brought about the use of radionuclides to affect research and technology. These radionuclides decay and later bring about the release of radiation and increase the environmental effect of radiation. The application of these fertilizers that are themselves radioactive to the farm causes food contamination [39]. The contamination level is higher if the fertilizer is applied to the farm soil in liquid form than in grain form. This contaminated food can release ionizing radiation inside the body of a man that takes it [25, 28].

Also, there are many naturally deposited minerals in the soil that contain radioactive elements in one form or the other. Among these radioactive minerals are granite and cement, to mention a few. The radioactive elements are found in small quantities in these radioactive materials. The abundant radioactive elements found in these minerals are Uranium and Thorium, mostly in Uranite Oxide and Thorianite Oxide, respectively [40]. The mining of these radioactive materials for human use will bring about the exposure of the radioactive elements (radionuclides), hence, decay later to give radiation into the environment.

Some industries also use radiation in one form or another. The example is made of construction companies where X-rays are used to check dislocation in construction works like bridges, buildings, and engine production in a specific industry. There are also atomic energy industries that release one form of radiation or another into the environment. These radiations emitted from the

nuclear energy industries result from the excitation of most material used for nuclear energy production.

In summary, if there is a nuclear weapon experiment or attack, it results in the discharge of radioactive materials into the debris attached to atmospheric dust and water particles [41]. The heavier of those particles landed on earth near the denotation location to cause negative impacts. The lighter ones stay in the upper atmosphere before falling into the lower atmosphere.

2.3 Radiation decay

Atoms are known to be radioactive in substance decay randomly with the probability of its nucleus decaying at a time interval independent of that time [20, 42]. The intensity of radioactive decay that occurs in a material is called the activity (A). Hence, activity is the number of nuclei decaying in such material [34, 36, 43].

The rate of radioactive decay associated with the activity can be expressed by the fundamental law of radioactive decay [44, 45]:

$$A = -\frac{dN}{dt} = \lambda N \qquad (2.1)$$

A is the activity of a pure radioactive source equal to the number, dN, at a given time, dt, and proportional to the amount, N, of radioactive nuclei present at a time, t. λ is the decay constant, which is equal to the probability of decay of an atom per unit of time. The negative sign means that the number of radioactive nuclei decreases [44-46]. The radiation activity unit was originally known as curie (Ci), based on 1 gram of Radium (^{226}Ra) corresponding to $3.7\text{x}10^{10}$ disintegrations per second. The Becquerel (Bq) has become the standard radiation activity unit defined as decay per second [47]. Therefore, $1Ci = 3.7\text{x}10^{10}Bq$ [48].

It is possible to resolve this same exponential law of radioactive decay based on equation 2.1, given as [42, 48]:

$$N(t) = N_0 e^{-\lambda t} \qquad (2.2)$$

where N_0 represents the number of nuclei originally present at the time $t = 0$.

A specific period known as the half-life characterizes the rate of radioactive decay ($t_{\frac{1}{2}}$) [45], which is the time it takes to decay one-half of the original nuclei [47]. The rate of decaying is associated with the half-life, as shown in equation 2.3 [49]:

$$t_{\frac{1}{2}} = \frac{ln2}{\lambda} = \frac{0.693}{\lambda} = \tau . ln2 \qquad (2.3)$$

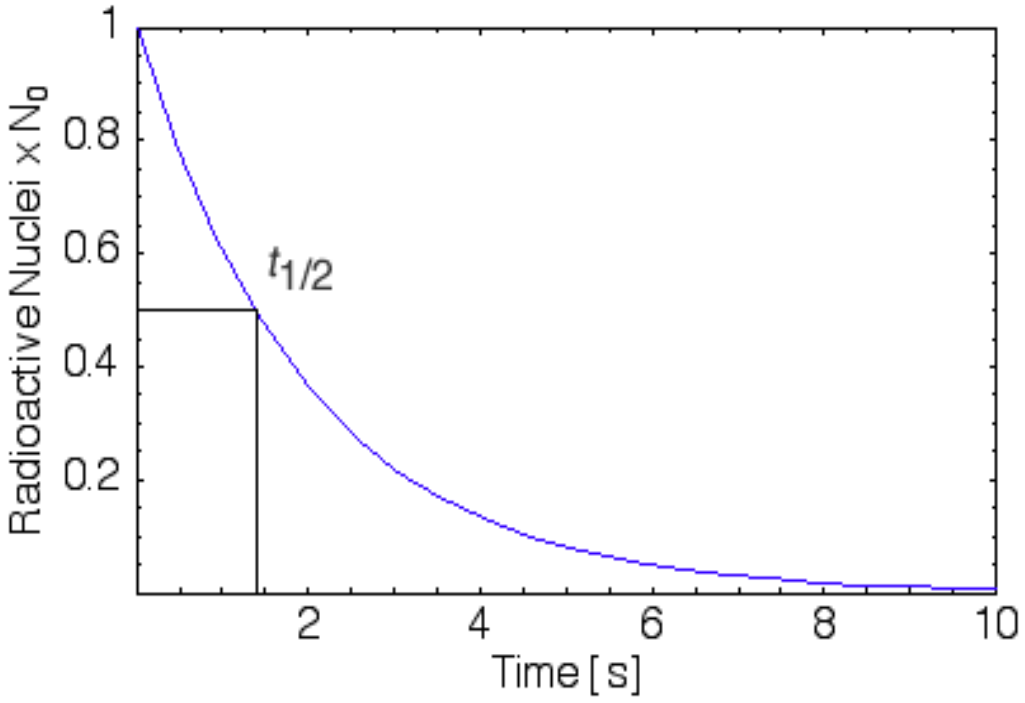

Fig. 2.3: Typical radioactive decay curve [27].

The mean or average life, τ, of a radioactive atom could be determined as some atoms survive much longer than others. Therefore, the atoms' numerical average age as the number of atoms decreases from N_0 to 0 is estimated. Consideration of the radioactive decay curve in Figure 2.3 might do this. Let N be the number of atoms that have survived for a time t, and $N - dN$ those still existing at the time $t + dt$; then dN represents the number of atoms that

disintegrate at time dt. Therefore, the combined ages of the atoms in this group at the time of disintegration tdN. The average life of an atom will be the sum of the combined ages of all the age groups of atoms from N_0 to 0 divided by the total number of atoms; in mathematical terms, this is [49]:

$$\tau = \frac{\int_{N_0}^{0} tdN}{\int_{N_0}^{0} dN} \qquad (2.4)$$

For integrating, t is used as a function of N. Substituting it from equation (2.2) and noting that the denominator in equation (2.4) equals$-N_0$, we have [49]:

$$\tau = \frac{1}{\lambda}\frac{\int_{N_0}^{0}[-ln(N/N_0)]dN}{(-N_0)} = \frac{1}{\lambda}\int_{N_0}^{0}\left(ln\frac{N}{N_0}\right)d\left(\frac{N}{N_0}\right) \qquad (2.5)$$

Integrating this by parts, then

$$\tau = \frac{1}{\lambda}\left[\frac{N}{N_0}\left(ln\frac{N}{N_0}\right) - \frac{N}{N_0}\right]_{N_0}^{0} \qquad (2.6)$$

This is indeterminate at the upper limit, but by using l'Hospital's rule, we have

$$\tau = \frac{1}{\lambda} \qquad (2.7)$$

This $\tau = \frac{1}{\lambda}$ is the mean life or life expectancy of an individual radioactive atom. Since λ represents the probability that an atom will decay in unit time, it follows at once that τ is the possible time of disintegration per atom. The mean lifetime can, therefore, be represented as a half-life [49]:

$$\tau = \frac{t_{\frac{1}{2}}}{0.693} \qquad (2.8)$$

The half-life and mean a lifetime of radioactive materials can differ widely from fractions of seconds to billions of years, depending on the particular radionuclide that decreases [45]. The intensity of the radioactivity is achieved by differentiating equation (2.2). It is achieved provided that the time interval *dt* during which the decay occurs is much lower than the half-life ($t_{\frac{1}{2}}$) [45]:

$$A = \left|\frac{dN}{dt}\right| = \lambda N_0 e^{-\lambda t} \quad (2.9)$$

$$A(t) = \lambda N(t) = A_0 e^{-\lambda t} \quad (2.10)$$

where $A_0 = \lambda N_0$ and is the initial activity at $t = 0$.

2.4 The radioactive series

When the unstable nuclei's excess energy has converted into a more stable state, nuclear radiation is released. The three significant forms of radiation emitted by radioactive substances are alpha (α), beta (β), and gamma (γ) decays [24, 46]. It was widely assumed that whenever a new half-life was identified, a new radioelement could be involved, and attempts were made to isolate it chemically and, if possible, classify it by an atomic weight determination [50]. Observation of evolutionary relatedness among activities after chemical separation also implied sequences of transmutations. The implementation of the theories on displacement allowed the deduction of new bodies' atomic weights from the established.

In this regard, the radioactive elements mentioned in Table 2.1 have been classified, focusing on the natural α-emitted radioelements; these are the thorium and radium series.

Table 2.1: Natural α-emitters (heavy metals) [51]

Elements	**Nuclear symbol**	**α-disintegration energy (MeV)**	$t_{\frac{1}{2}}$ **(=**$\frac{0.693}{\lambda}$**)**
Uranium I	$^{238}_{92}U$	4.27	$4.51\text{x}10^{9}\ y$
Uranium II	$^{234}_{92}U$	4.85	$2.48\text{x}10^{5}\ y$
Ionium	$^{230}_{90}Th$	4.76	$8.0\text{x}10^{4}\ y$
Radium	$^{226}_{88}Ra$	4.86	$1622\ y$
Radon	$^{222}_{86}Em$	5.59	$3.823\ d$
Radium A	$^{214}_{84}Po$	6.11	$3.05\ m$

Radium C (0.04%)	$^{214}_{83}Bi$	5.61	19.7 m
Radium C'	$^{214}_{84}Po$	7.83	1.64x10^{-4} s
Radium F (polonium)	$^{210}_{84}Po$	5.40	138.401 d
Thorium	$^{232}_{90}Th$	4.08	1.39x10^{10} y
Radiothorium	$^{228}_{90}Th$	5.52	1.91 y
Thorium X	$^{224}_{88}Ra$	5.78	3.64 d
Thoron	$^{220}_{86}Em$	6.40	5.5 s
Thorium A	$^{216}_{84}Po$	6.90	0.158 s
Thorium C	$^{212}_{83}Bi$	6.21	60.5 m
Thorium C'	$^{212}_{84}Po$	8.95	3.04x10^{-7} s
Actino-uranium	$^{235}_{92}U$	4.64	7.1x10^{8} y
Protoactinium	$^{231}_{91}Pa$	5.14	3.43x10^{4} y
Radioactinium	$^{227}_{90}Th$	6.14	18.2 d
Actinium X	$^{223}_{88}Ra$	5.97	11.7 d
Actinon	$^{219}_{86}Em$	6.94	3.92 s
Actinium A	$^{215}_{84}Po$	7.50	1.83x10^{-3} s
Actinium C	$^{211}_{83}Bi$	6.75	2.16 m
Actinium C'	$^{211}_{84}Po$	7.59	0.52 s
Potassium	$^{40}_{19}K$	1.32	1.3x10^{9} y

As all changes throughout the mass numbers were due to the release of α-particles, each series have masses that differ by multiples of four units. The naturally occurring series originates from nuclei with long lives relative to

earthly ones [52]. Neptunium has a reasonably short life, but it is its series' longest-lived member [53]. Each sequence ends when the cycle of decay contributes to creating a stable nucleus that becomes the product; the parents and end products of specific radioactive series occur naturally.

Table 2.2: Parents and end products of radioactive series [54]

Name of series	Mass number	Parent	Half-life	Product
Thorium	$4n$	^{232}Th	$1.4\text{x}10^{10}\ y$	^{208}Pb
Neptunium	$4n+1$	^{237}Np	$2.2\text{x}10^{6}\ y$	^{298}Bi
Uranium-radium	$4n+2$	^{238}U	$4.5\text{x}10^{9}\ y$	^{206}Pb
Uranium-actinium	$4n+3$	^{235}U	$7.2\text{x}10^{8}\ y$	^{207}Pb

Thus, the naturally occurring radioactive isotopes end with lead; this accounts for lead and helium in the radioactive ores. Most radioactive elements disintegrate in a definite manner with the ejection of an α- or β-particle. As shown in figure 2.4-2.7, the four radioactive series's main decay chain was shown. The $4n$ (thorium), $4n+2$ (uranium-radium) and $4n+3$ (uranium-actinium) series are found in nature but the $4n+1$ (neptunium) series must be prepared artificially.

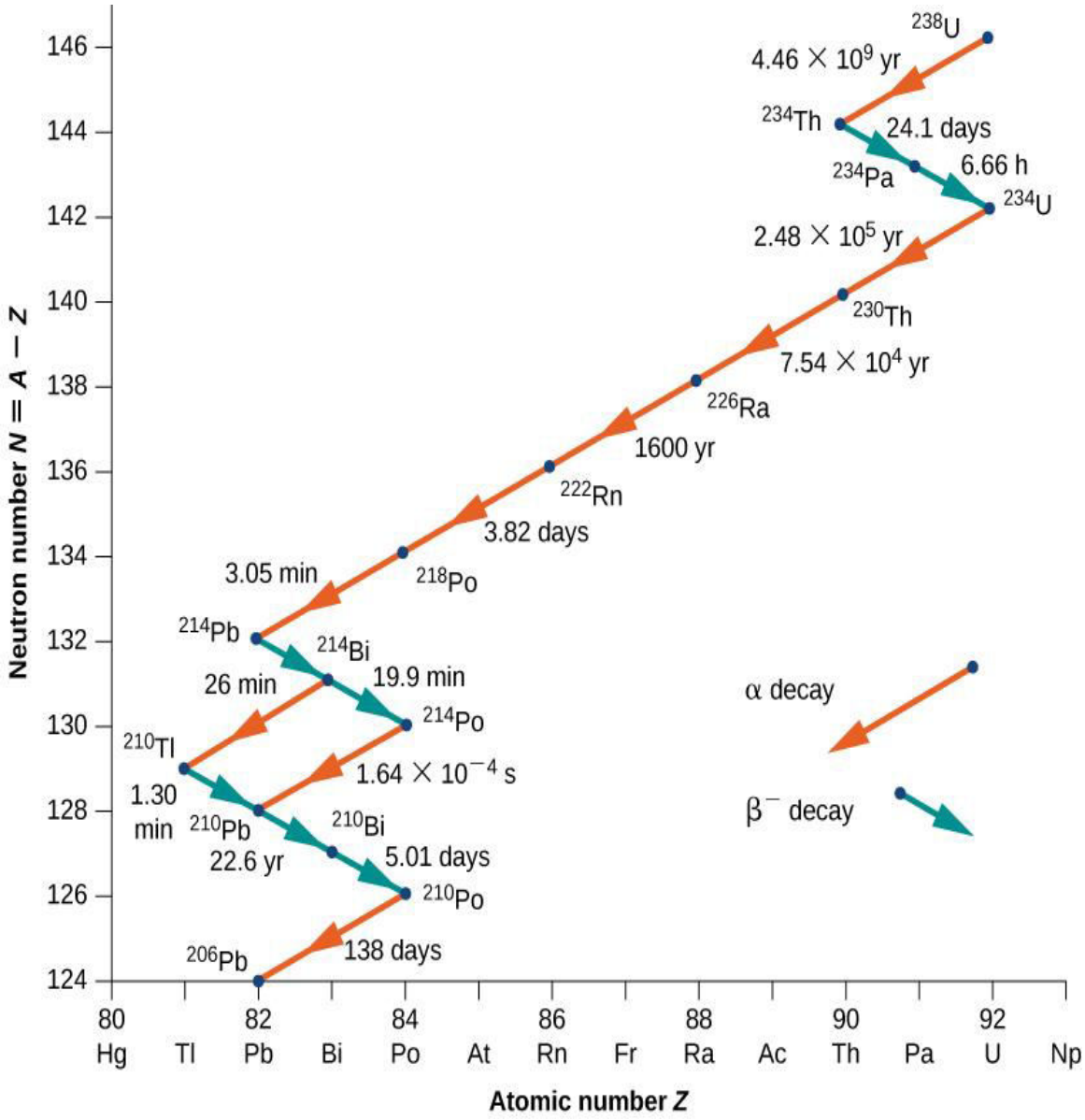

Fig. 2.4: Series of decaying (^{238}Uranium) [55]

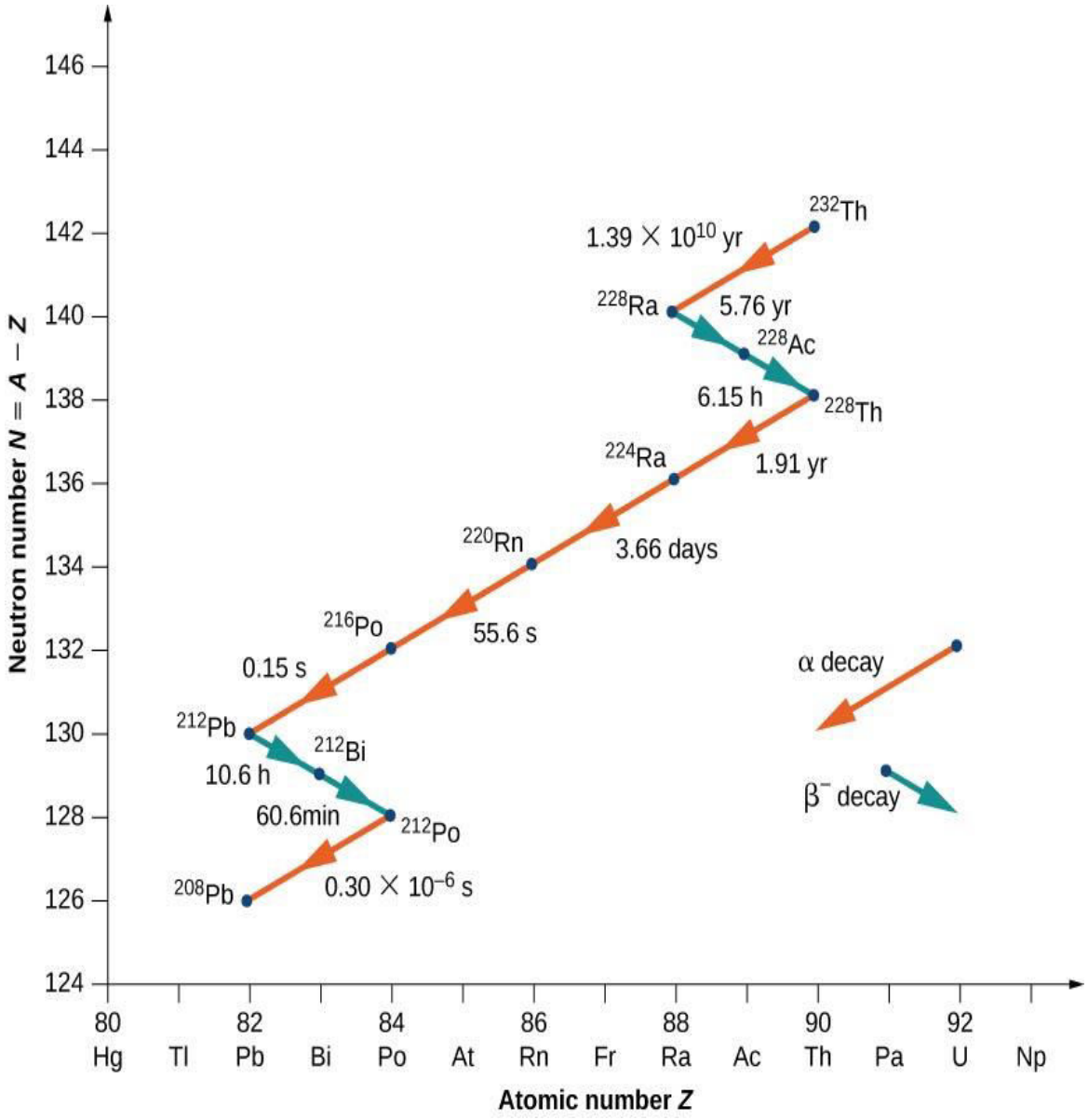

Fig. 2.5: Series of decaying (^{232}Thorium) [55]

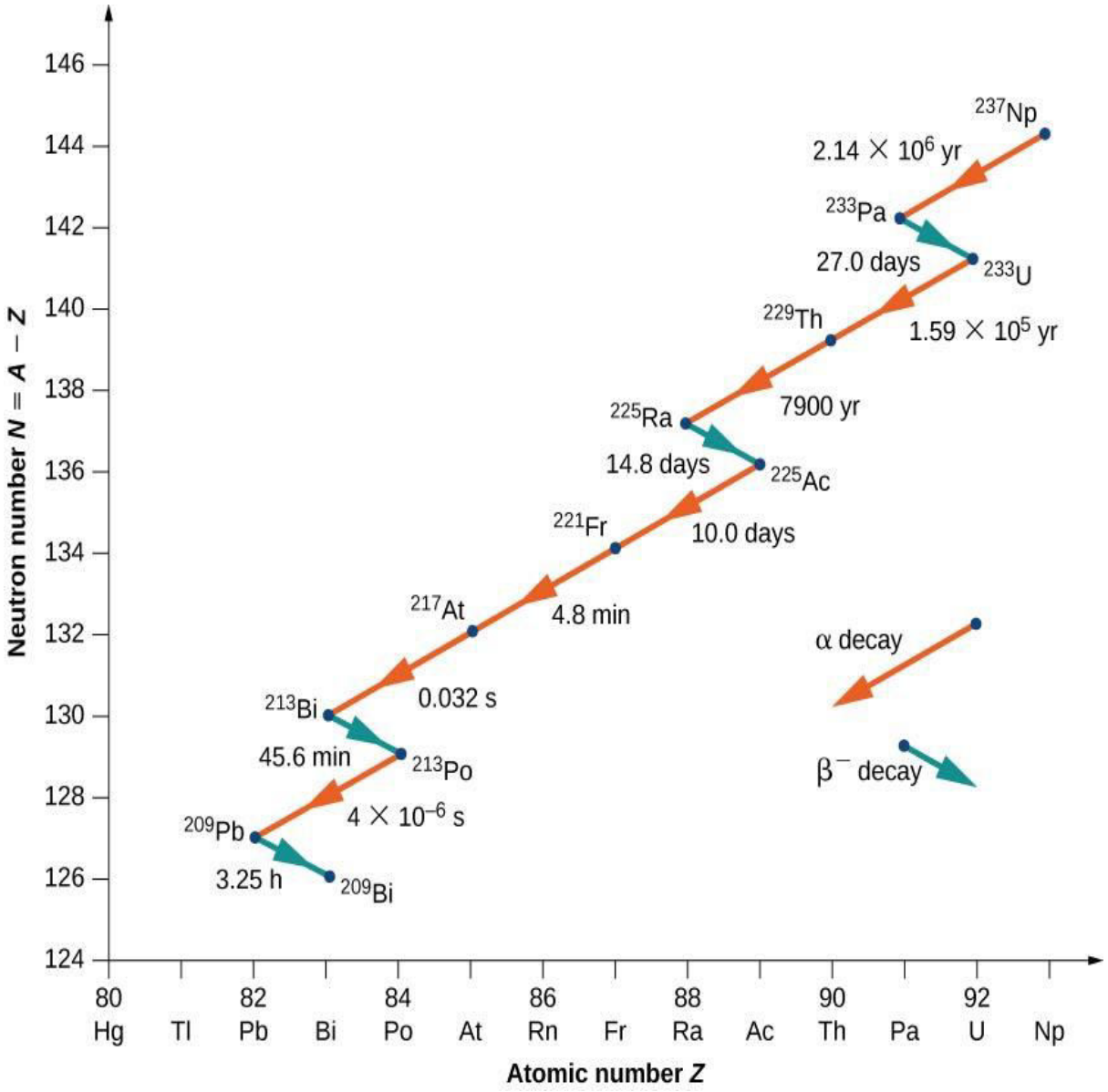

Fig. 2.6: Series of decaying (^{93}Neptunium) [55]

2.5 NORMs in oil and gas production

Atoms with an unstable nucleus are defined by unstable energy that allows them to be surmounted inside the nucleus to produce new radioactive particles [56]. Such atoms are known as radionuclides. The radionuclide that may occur naturally or artificially undergoes radioactive decay and thus emits gamma or subatomic particles. Radionuclides play a significant role in the processes that provide humans with food, water, and good health. Still, they can also constitute actual or potential hazards.

Radionuclides fall into three major categories, which are radionuclides of primordial, secondary, and cosmogenic origins. Primordial radionuclides derive mostly from the stars' interiors, such as uranium, which are around as they have long half-lives. Secondary radionuclides arising from the decay of primordial radionuclides have shorter half-lives. Due to cosmic rays, cosmogenic radionuclides are formed continuously in the atmosphere. They are contained in nature as their quantities are still being replenished even though they have short lives [47, 53].

The International Atomic Energy Agency (IAEA) describes NORMs as radioactive materials containing no substantial amounts of radionuclides other than those naturally occurring [57]. In the past, different names have been applied to these radioactive materials, such as Low Specific Activity (LSA) and Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM) [5, 57]. Many human activities indirectly increase NORMs, for instance, the use of building materials containing high activity concentrations in buildings and workplaces or the use of radioactive materials in industries such as various oil and gas production activities [58]. The IAEA concluded that regulatory control on activity concentration below 1 Bqg^{-1} is not necessary. If the activity concentrations are at or above the 1 Bqg^{-1}, the activity may still be excluded from regulatory checks if the occupational exposure is less than 1 $mSvy^{-1}$ [60]. Considering also that the worldwide effective dose rate of public

exposure from the soil with weighted mean concentrations of 30, 35, and 400 $Bqkg^{-1}$ for ^{238}U, ^{232}Th, and ^{40}K respectively is 0.460 $mSvy^{-1}$ [13]. NORM occurs in minerals and its sands, coal, bauxite, and oil explorations [61]. Oil exploration processes require several radioactive sources, which can alter the environment's radioactivity concentration if it escaped into it [62].

The first reports of NORMs linked to oil and gas production appeared in 1904, while several later releases referenced the incidences of ^{226}Ra in oil and gas field reservoir water [63]. The material analysis shows that the solids in oil production plants' down-hole and surface structures, such as tubular inside walls, wellheads, valves, pumps, and separators, seem to contain nuclides of the ^{238}U and ^{232}Th series [64].

Oil and gas-induced NORMs can cause external exposure during production due to accumulations of gamma-emitting radionuclides; and internal exposure to workers and other persons, particularly during maintenance, waste transportation, and contaminated equipment [59, 64, 65]**,** thereby leading to different kinds of biological effect.

2.6 Application of radionuclides in oil exploration

The petroleum industry is a growing sector of the South African economy, which comprised some activities, including extracting and importing crude oil, coal, and gas [66]. The exploration of this oil requires the extensive use of radioactive sources [67]. Radioactive sources used in several industries, including the petroleum industry, have raised concerns about its safety to the environment and humans. Oilfield radioactive wastes are incredibly mobile, transported worldwide, even to remote locations through rivers and streams [68].

Radiation from radioactive sources is used in many ways. Still, there were complaints about radioactive sources that have been stolen, shackled, or abused by users in the sense of industry and government safeguards and

regulation [68]. These include non-destructive weld testing and inspection of critical metal components for cracks and other defects as gauges for monitoring and often regulating the density of mud and cement grout; monitoring and controlling the thickness of materials petrochemical plants. Down-hole logging systems use numerous radioactive sources to record various characteristics of a well.

Devices incorporating sealed sources are installed to detect smoke and measure or eliminate static electricity and emergency signs. Unsealed radioactive substances are also used in several ways. These include the uses as foils grout into the wells' lining to act as a depth marker and liquids and powder to trace the labeled grout movement. Other applications of unsealed sources are to mark the point where a casing has been entered as liquids for water flooding tracing of wells and investigation reservoir parameters, leakages in pipelines, and product reservoir tanks. These applications involve radioactive materials of various strengths, half-lives, and compositions [69].

2.6.1 Application of sealed radioactive sources

Gamma radiography and X-radiography are used by the petroleum industry to ensure that all design and manufacturing is completed to the required standard [66-68]. Radiography is necessary during the fabrication of machinery used to servicing rigs and platforms since the welds and joints must withstand the extremely high physical stresses associated with petroleum production [67]. The technique employs reliable gamma sources, notable ^{192}Ir ($T_{1/2}$~$74\ d$), with the order of TBq.

Nuclear gauges are extensively mounted on oil and gas-related plants and equipment [70]. They are often designed to track and regulate fluid levels in vessels and detect the interface between different densities' interfaces, such as the interfaces in separators for liquid, oil, and natural gas. Each gauge usually comprises a radioactive source and detector.

Radioisotope density gauges are often used to show the existence and precisely calculate the cement grout density. This density gauge is based on calculating the intensity of a gamma radiation beam after passing through grout as it passes between a radioactive source and a gamma radiation detector. Iridium-192 is another radionuclide widely used for gauges in the oil production industry [71].

Highly technical instruments are installed in the well for well-logging, determining the rocks' physical parameters and geological features around the borehole and the rocks' elemental constituents. All these devices have one or more radiation detectors, radioactive sources, or ionizing radiation producing systems [60, 63]. Nuclear logging techniques involve measuring and labeling the gamma rays released by naturally occurring radioactive material in rocks. The log records the content of the rocks in uranium, thorium, and potassium.

A long skid pushes the instrument hydraulically against the wall of the well in another technique. Two detectors of radiation, positioned at different distances from the tool's source, test the neutron backscattered from the rock formation. The two readings' relationship provides the rock with a porosity index, determining if the rock will likely contain hydrocarbons or water. This technique requires ^{241}Am/Be source with activity to emit 4-5 MeV neutrons [60].

The third technique comprises a gamma-gamma or density tool that contains two detectors and a source of ^{137}Cs. The sum of back-scattered gamma rays from the formation provides the log density that a log is a reliable measure of the gas presence [72].

2.6.2 Application of unsealed radioactive sources

Oil and gas processing in the industry includes the use of unsealed radioactive solids (powder and granular forms), liquids, and gasses to examine or track other products' movement, including in closed and often inaccessible pipes and

vessels [63, 70]. Most of these radiotracers are readily detectable and measured by emissions. Gamma emitters such as ^{46}Sc, ^{140}La, ^{56}Mn, ^{24}Na, ^{124}Sb, ^{192}Ir, ^{99m}Tc, ^{131}I, ^{110m}Ag, ^{41}Ar, and ^{131}Xe are used due to the ease with which they can be identified and measured [63]. Such gamma emitters often have relatively short half-lives. They are injected into the wells to track the fluid movement in the formation of rocks or behind the well's enclosure and tubing [73]. Detectors mounted outside of the device rapidly track the radiotracers. They require non-invasive manufacturing processes involving minimal disruption.

2.7 Bio-effect of radiation exposure

Interactions between human body systems and the exposed radiation from naturally occurring sources regularly include transmitting radiation energy to body tissues. Radiation exposure can affect living tissue by altering the structure of the cells and destroying DNA. As radiation travels through living cells, the cell structure triggers ionization and atom molecules' excitation, thereby breaking the bonds between the atoms and the particles. The breaking of the bonds results in the formation of ions or radicals, which are highly reactive. They damage the cell, resulting in either somatic or genetic, depending on whether the damage is done on autosomal or germ cells, respectively. The exposed person suffers from somatic effects, while effects are passed to the progeny due to its damage [16].

The amount of damage depends on the intensity of radiation absorbed, its type, and the overall amount. Harm to a cell by radiation depends on the cell's radio-sensitivity and the amount of radiation absorbed. Cells that divide quickly are more prone to radiation than cells that divide slowly. However, bone marrow cells, lymphatic system, and reproductive systems are among the most radiosensitive ones in the body. Most genetic and carcinogenic effects of radiation exposure are stochastic, with a latent period varying between 2 years and 25 years after exposure. The impact of low exposure may not have been

observable as the damage is at the cellular level. These lead to indirect bio-molecular damages in a 10^{-7} second to many hours' timeframe [74]. Additionally, visible symptoms will occur for several days to weeks, leading to the eventual death of cells or even the whole organism. Specific biological effects that are hard to notice could lead to cancer and possible transmission to coming generations of some genetic disorders [4, 27].

The effects of ionizing radiation can occur to biological systems due to internal and external radiation exposure. In general, internal exposure is through the ingestion of food or water or particle or aerosol inhalation. Inhalation of radon gas (^{222}Rn) from ^{238}U series may lead to internal exposure. Indeed, due to long half-lives, most primordial radionuclides such as ^{238}U, ^{232}Th, and ^{40}K can still be found in trace quantities and eventually find their way into food and drinks [5, 9].

The determined internal dose depends on both the radiant and the absorbing material's biological and physical conditions. The physical considerations include the radioactive source's energy and half-life, while the biological consists of the body's radioisotope distribution. It also depends on its bio-kinetic behavior in the various organs and tissues, such as absorption, high turnover, and retention times.

Bio-effects of radiation have been under study for several decades [5, 16, 27, 57]. Because of its use in several aspects of our lives, evaluating its adverse effects is essential, intending to set safety limits for exposure and good work practice. In terms of radiation dose calculations, an estimate of the amount of ionization that occurred and the energy consumed by a cell associated with biological effects can be considered. Radioactive contamination of the aquatic environment has a very high probability of causing direct exposure to man and other organisms in the ecosystem, thereby making a man stand the risk of various biological effects.

2.7.1 Deterministic effect of radiation exposure

The deterministic effect is due to a body that causes unnecessary cell damage, hampering an irradiated tissue or organ's proper functioning. The degree of harm caused by a deterministic event depends on the exposed person's distribution. The deterministic impact threshold with sufficient magnitude in even more susceptible individuals will occur at a low dose rate when various individuals with different susceptibilities are exposed to radiation [75]. However, if the dose rate increases, more individuals are likely to experience the same effects before the other degree of deterministic impact at a high dose is exhibited by the entire population.

Not all organs are equally susceptible to radiation, so in an over-exposure scenario, the reaction pattern or disease syndrome depends on the severity of the dose [35]. According to a joint study published by the IAEA (1989) on occupational safety, causing temporary sterility in normal males, it is approximately 0.15 Gy for short-term exposure. For more prolonged exposure of about 0.4 Gy. A dose rate of 3.5 to 6.0 Gy is above the threshold, which may lead to permanent sterility upon acute exposure.

The threshold for a woman to experience permanent infertility is between 2.5 to 6.0 Gy [63]. The threshold dose rate of clinical effect is 0.5 Gy for whole bone marrow acute exposure, with 0.4 Gy assigned to the threshold dose rate of extended exposure. In the existing radiation safety system, this problem of dose rate limitation aims to avoid deterministic effects [4, 63].

2.7.2 The stochastic effects of doses of radiation

The stochastic effects of radiation exposure arise by chance and primarily consist of cancer, and genetic factors with stochastic results occur years after exposure. Therefore, as the dose increases for an organism, so does the risk of cancer or a genetic effect. There is, however, no guarantee that even a high dose of radiation will at any time lead to cancer or genetic damage [76]. There

is also no threshold dose, where there is relative confidence that exposure to radiation does not have adverse effects.

Nevertheless, in individuals who have not been exposed to radiation above background levels, stochastic effects can occur. Therefore, it can never be known that specific radiation exposure was due to the incidence of cancer or genetic damage. Radiation exposure can be predicted to have a stochastic effect even though it is not entirely definable. Fetal exposure to radiation during pregnancy has been reported to raise the risk of leukemia in infants and during particular early pregnancy times, leading to mental retardation and congenital malformations if the radiation is high enough [77].

2.8 Principles of radiation detection and measurements

Most radiation detection and measurement instruments are based on the ionization and excitation they produce in the atoms and molecules of the materials they traverse. Coulomb's force may direct this for charged particles and indirectly for neutral particles. These uncharged particles produce secondary charged particles in their passage through matter. The type of radiation strongly determines the detection and measurement of ionizing radiations due to the different modes of interaction.

Most environmental samples contain some α, β and γ-emitting radionuclides up to measurable levels. Therefore, it is possible to detect any of the particles α, β, and γ-ray of the daughter nuclides to assay the parent radioisotope. Many factors determine the choice of the detection method. Among the factors is the nature of the source, the purpose of measurement, and, most importantly, the facilities available.

The detection and measurement of radionuclides in environmental samples have often been undertaken by many authors using either a Sodium Iodide

(NaI) or a Germanium detector [78-80]. The spectroscopic technique of gamma-ray scintillation was employed in this work using a High Purity Germanium (HPGe) Radiation Detectors. The choice is based primarily on the low cost of this method and the availability of resources. The detector has been reported as suitable for detecting and measuring effectiveness, the radionuclide contents of environmental samples [81].

2.8.1 Scintillation spectroscopy

A scintillation detector is a transducer that converts the ionizing radiation's kinetic energy into a flash of visible light [82, 83]. Molecules exhibit the same luminescence effect despite that their scintillation processes have essential variations. Some organic and inorganic crystals used in scintillation spectroscopy are Anthracene, Quaterphenyl, Stilbene, Terphenyl, Diphenyl, Acetylene, Naphthalene, Chloroanthracene, ZnS(Ag), CdS(Ag), NaI(TI), NaCl(Ag), LiI(Ti, Sn or Eu), CsI(TI) and $CaSO_4$.

NaI(TI) is the commonest scintillation detector due to its unique suitability in gamma-ray measurement and its high efficiency. It is highly hygroscopic, so it is usually enclosed in an airtight covering. The covering is often a low-density metal like Al and made thin enough not to cause considerable attenuation of the γ-radiation being detected. The casing has an optical window through which it is coupled to a photomultiplier (PM) tube.

In the scintillation detection system, an incident gamma photon dissipates its energy (E) entirely in the scintillator producing photons (Ew_0q_0). Where w_0 is the average energy that creates a single photon, and q_0 is the luminescence quantum's efficiency. For a scintillation detector, the value of w_0 is about 3eV. A fraction G of the incident photons, called light collector efficiency, impinging on the photocathode, is converted to electrons. This conversion's effectiveness is mC_{pe}, where m is a factor between 0 and 1, according to the spectral match between the scintillation spectrum and the photo-cathode response. C_{pe} is the

photo quantum efficiency of the window-cathode system. The number N_p of photoelectrons at the photocathode of the photomultiplier tube (PMT) is given by [83]:

$$N_p = \frac{E}{w_0} q_0 m C_{pe} G \qquad (2.11)$$

These electrons are collected with efficiency g_c by the first dynode. The total number N_t of particles arriving at the dynode is therefore given by [83]:

$$N_t = \frac{E}{w_0} q_0 m C_{pe} G g_c \qquad (2.12)$$

Several factors are affecting the superior efficiencies. G is determined by self-absorption, reflection losses, light trapping, optical flaws, and the photocathode's optical geometry. Self-absorption is small in $NaI(TI)$ crystal, and hence G can be made nearly unity by coating the detector with a reflector (MgO). The efficacy factor $mC_{pe}g_c$ depends on the cathode's wavelength and incidence level of the photons. Photocathode with an excellent spectral match has a large. The photo quantum efficiency C_{pe} depends on the cathode material and thickness, while g_c relies on the dynode's structure and its potential.

The N_t electrons given by equation 2.12 are multiplied at k successive dynodes with an overall gain M gave as [49]:

$$M = \pi_i^k m_i \qquad (2.13)$$

where m_i is the multiplication at the i^{th} dynode. m_i is proportional to the voltage within the dynodes. The total number Q_0 of electrons at the anode is given as [45]:

$$Q_0 = M \frac{E}{w_0} q_0 m C_{pe} G g_c \qquad (2.14)$$

Q_0 is a linear function of the energy E of the initial incident photon.

Irrespective of the number of electrons given by equation 2.14, several electrons are due to thermionic emission. This number n_T is a function that varies exponentially with temperature and expresses as [84]:

$$n_T = AT^2 e^{\left(\frac{-Qe}{kT}\right)} \qquad (2.15)$$

T is the absolute temperature, e is the electronic charge, k is Boltzmann's constant, A *and* Q are characteristics of the cathode material.

The number of electrons with thermal energy more significant than the working function of common photo-cathode materials at room temperature is about the order 10^3per cm^2 per second. These thermionic electrons are multiplied in the photomultiplier tube (PMT) and constitute the dark current that forms part of the energy spectrum background. The dark current's contribution has been identified to pose a significant problem, especially when low energy radiation or weak sources are being counted [85].

2.8.2 Pulse shaping and amplitude selection

The number of electrons reaching the anode, expressed in equation 2.14, suffers an exponential decay according to the expression [47]:

$$N = Q_0 e^{\left(-t/T_d\right)} \qquad (2.16)$$

T_d is the scintillator's modified decay time; this decay time constant is changed by decay time spread effects within the PMT. The output of the PMT is, therefore, a pulse, shown in Figure 2.7.

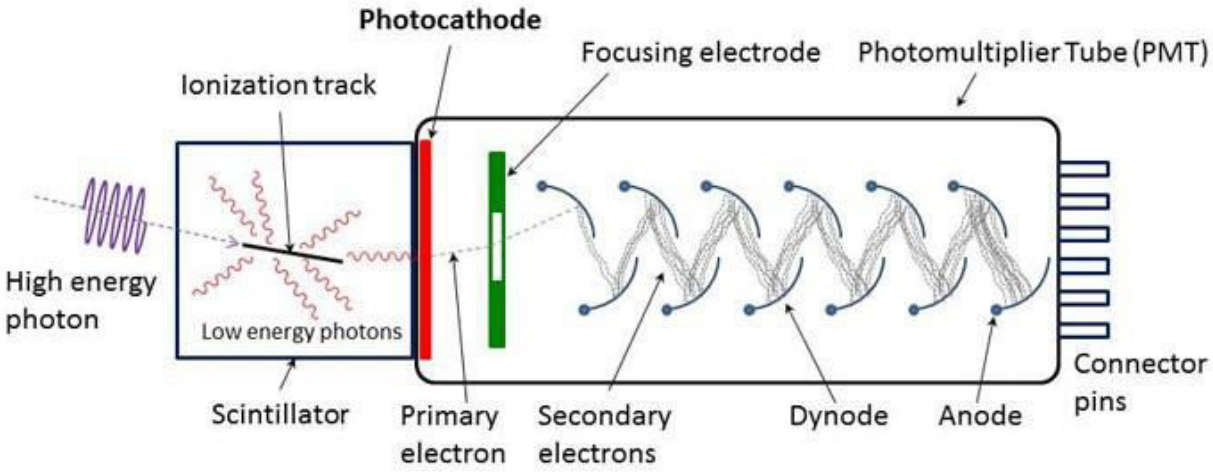

Fig. 2.7: A scintillator crystal coupled with a PMT with ten dynodes [86].

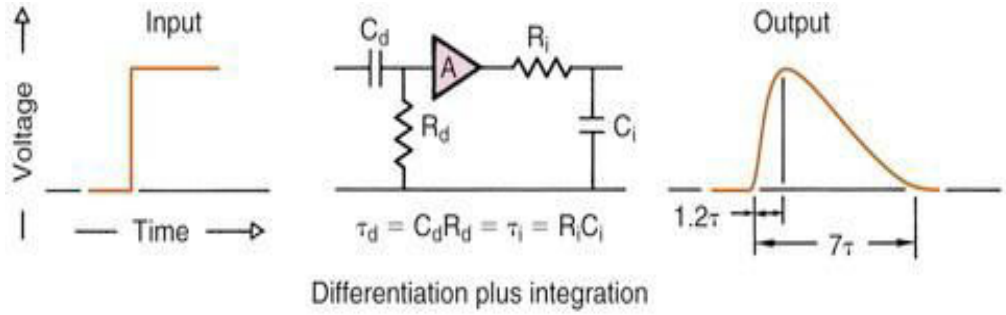

Fig. 2.8: A schematic representation of a pulse shaping circuit [87].

These pulses may be subjected to a pile-up; that is, a new pulse may arrive while the system is still responding to a previous pulse, as shown in figure 2.8. It is thus essential to collect the information about a pulse as quickly as possible. This is usually achieved by a pulse shaping RC circuit, as shown in figure 2.8, which is placed after the amplifier; RC is the time constant of the circuit. After shaping, the number $N(t)$ of electrons in the shaped output is given by [48]:

$$N(t) = Q_0 \frac{RC}{(RC - T_d)} (e^{-t/RC} - e^{-t/T_d}) \quad (2.17)$$

In gamma spectrometer applications, the energy spectrum is of importance. The voltage pulse is of greater value than the current pulse. Hence the voltage $\left(\frac{Q}{C}\right)$ is given by [47]:

$$V(t) = \frac{Q_0}{C_s} . q . \frac{RC}{(RC - T_d)} (e^{-t/RC} - e^{-t/T_d}) \quad (2.18)$$

where C_s is the capacitance of the capacitor, and q is the electronic charge.

Equation 2.17 implies that the amplitude of the pulse depends on C_s, T_d, and RC; and that the pulse is of random occurrence both in the amplitude and time while also revealing that the peak $V(t)_p$ of the voltage pulse is proportional to the dissipated energy due to the initial radiation. The distribution $N(E)$ is the number of pulses with energy E. The spectrum is acquired by converting the analog information in $V(t)_p$ into digital data, which is more convenient to process.

The signals with pulse amplitude less than some preset minimum energy E are filtered off using a discriminator such as a Schmitt-Trigger circuit or an analog comparator circuit. The real signals are analyzed according to their heights. The elemental pulse height analyzer is called a single channel analyzer, while modern pulse analysis is conducted using multichannel analyzers.

2.8.3 Gamma-ray spectroscopy

Interactions of photons with the detector material determine the shape of the spectrum obtained. Twelve different processes by which γ-rays can be absorbed or scattered have been reported [88]. However, in the region of 0.01 - 10MeV, which is encountered in a familiar environment, most interactions are explained in terms of just three processes: the photoelectric effect, Compton scattering, and pair production [89].

2.8.3.1 Photoelectric effect

In high atomic number materials, the mode of interaction is pronounced as the photoelectric effect is predominant at low energy below 0.5 MeV [90, 91]. In the photoelectric effect, a γ-ray photon passes all its energy to an electron and vanishes. The photoelectron is emitted with energy, $E_e = hv_0 - B_e$, which is the electron's binding energy. The effect only happens with a bound electron where the electron and atom can conserve the momentum. The excited atom loses its energy through Auger electrons emission and X-ray characteristics from filling the inner shells where the photoelectric effect often occurs [92].

The highest absorption of light in scintillation spectroscopy is due to the photoelectric effect since the photon's total radiation is transferred to excite the crystal [93]. The peak resulting from this is termed photopeak, and its position in the spectrum is related to the incident γ-ray energy by equations 2.13 and 2.17. Other peaks related to photoelectric absorption are those due to the Auger electrons and characteristics X-rays, which occur in the spectrum's low energy region. They are both characteristics of the absorbing materials.

2.8.3.2 The Compton scattering

The photon undergoes elastic dispersion with a free or weakly bound electron in the Compton scattering [94]. Suppose the small effects due to the binding of an atomic electron to the nucleus are neglected. In that case, the scattering of such incident photon energy hv_0 by an electron can be treated as a simple two-dimension elastic scattering. The gamma-ray is losing part of its energy to the electron for the recoil. The energy of the recoil electron can be derived from energy and momentum conservation principles [94]:

$$E_r = hv_0 \left[\frac{hv_0(1-cos\theta)}{M_eC^2+hv_0(1-cos\theta)}\right] \qquad (2.19)$$

where θ is the scattering angle, and M_e is the mass of the electron. From equation 2.19, E_r can have a value in the continuous range of zero to a

maximum of E_c, called the Compton edge, which corresponds to a back-scattering angle ($\theta = 180^0$). At this point [94]:

$$E_c = \left[\frac{2hv_0}{M_eC^2+2hv_0}\right] \qquad (2.20)$$

The fractional energy loss in the Compton scattering is quite significant for energetic γ-rays. It takes precedence in the energy spectrum of 0.6 – 2.5 MeV and leads to a continuous broad distribution of pulses termed Compton plateau, from zero up to E_c. Compton plateaus of higher energies always form part of the background of the photo-peaks of lower energies. This usually poses a problem to an accurate evaluation of the net area due to photoelectric absorption in gamma spectroscopy. Calculating the detector's background count while no sample has been put in it and deducting it from the overall count to calculate the net count in each counting period makes the real count [85].

2.8.3.3 Pair production

Pair production becomes increasingly important above photon energy, 1.02 MeV, or $2mc^2$[95]. During pair production, the photon is fully absorbed and replaced by a pair of positrons and electrons whose total energy is equal to the initial photon energy. That is

$$hv_0 = (T_e + mc^2) + \left(T_p + mc^2\right) \qquad (2.21)$$

where $T_e$and T_p is the electron and positron kinetic energies, respectively. For this method, a nucleus' presence is important to preserve momentum, although the energy the nucleus takes is minimal. The pair production intersection is approximately proportional to $ln\,E$.

Both particles ($e^{\pm}$) lose their kinetic energies, T_e and T_p, by excitation of the absorbing crystal. Positron then annihilates an electron to produce two electrons, each having an energy of $0.511\,MeV$, to conserve momentum in

approximately 180^0. The final light output following the initial pair production is usually due to both gammas photons resulting from the annihilation interaction losing their kinetic energy to the crystal. Since the time interval between pair production and annihilation is as short as $10^{-6}s$, a light pulse size given by the incident photon's total energy hv_0, a photopeak, is a possibility. This is added to the photopeak due to photoelectric absorption.

Apart from the particularities of the gamma spectrum due to these possibilities of interaction in the detecting crystal, a 0.511 MeV peak may be observed if an appreciable pair production occurs in the detector's vicinity (e.g., shield wall). A photopeak at 0.511 MeV is therefore not usually suitable for the activity measurement of a source. Other features of gamma spectra are sum peaks. They result when more than one single photon is interacting with the detector within a sufficiently short period. The combined output pulses are one single pulse by the detector system. This may be the case for gamma emitted in a cascade, or when the activity being measured is very high, or the detector's dimension is substantial. These features are not likely to be observed in the low-level measurement of environmental radioactivity conducted in the present work.

2.9 The concepts of radiation protection and recommended limits.

The risk involved with radiation constitutes significant public attention, and regulatory radiation safety programs are being established to enhance radiation [77]. Component of this requires that users of radioactive material maintain exposure levels to the dose limits defined and ensure that users must be approved and monitored by the body in compliance with the regulatory guidelines on the use of radioactive materials. The use of radioactive sources must justify the procedure involved by enforcing individual exposure limits

according to the International Commission for Radiological Protection (ICRP) guidelines on the use of ionizing radiation.

Unjustified radiation exposure could be prevented by following the fundamental principles of time, distance, and shielding (TDS) in radiation safety to protect people who deal directly with radioactive materials. The policy ensures that the exposure time, distance, and adequate shielding from the source translates into the recommended acceptable dose limits [2, 4, 9]. ICRP has recommended that workers' occupational exposure be controlled so that the recommended limits are not exceeded [9].

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CHAPTER 3

RADIOLOGICAL DOSE ASSESSMENTS OF FISH SAMPLES DUE TO THE PRESENCE OF NORMS AT OIL-RICH AREAS OF SOUTH AFRICA

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Radiological dose assessments of fish samples due to the presence of NORMs at oil-rich areas of South Africa.

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Abstract

This study was conducted to estimate natural radionuclide contents in twenty-four fish samples collected from rivers of South Africa's oil-rich and non-oil-rich areas. Also, the radiological analysis was performed to assess the effect of radiation on the population that consumed these fish. A high-resolution Hyper Pure Germanium (HPGe) detector was used to conduct the gamma-ray measurements. The mean activity concentrations for the selected fish samples from the examined areas for ^{238}U, ^{232}Th and ^{40}K ranged from (8.60 ± 2.97), (4.26 ± 1.18), (105.66 ± 47.77) $Bq.kg^{-1}$ for the Bree; (8.06 ± 3.64), (4.84 ± 2.00), (126.88 ± 47.30) $Bq.kg^{-1}$ for Klein-Brak; (8.30 ± 3.64), (3.48 ± 1.44), (90.42 ± 29.35) $Bq.kg^{-1}$ for Bakens and (6.48 ± 2.05), (5.26 ± 1.79), (78.38 ± 20.55) $Bq.kg^{-1}$ for uMngeni rivers. The annual effective ingestion dose ranged from 0.050 $mSv.y^{-1}$ (*Argyrosomus japonicas*) to 0.100 $mSv.y^{-1}$ (*Lichia amia*) for the Bree; 0.033 $mSv.y^{-1}$ (*Pomadasys commersonnii*) to 0.118 $mSv.y^{-1}$ (*Pomatomus saltatrix*) for Klein-Brak; 0.034 $mSv.y^{-1}$ (*Enteromius pallidus*) to 0.090 $mSv.y^{-1}$(*Anguilla marmorata*) for Bakens and 0.046 $mSv.y^{-1}$ (*Anguilla marmorata*) to 0.082 $mSv.y^{-1}$ (*Hypseleotris cyprinoides*) for uMngeni river respectively. Also, the estimated values for the annual equivalent dose of gonads, bone marrow and bone surface cells due to ingestion of fish samples ranged from 58.77 to 127.27 $mSv.y^{-1}$ for Bree; 42.14 to 125.94 $mSv.y^{-1}$ for Klein-Brak; 39.34 to 84.97 $mSv.y^{-1}$ for Bakens and 54.54 to 71.97 $mSv.y^{-1}$ for uMngeni rivers (control) respectively. The activity concentrations and radiological dose estimates reported for the studied areas were within the values recommended by the United Nations Scientific Committee on the Effects of Atomic Radiation. Hence, the result of the activity concentrations in the selected fish samples at the time of this study does not pose a radiological risk. The results could also be used as reference data for radioactivity pollution in the study area.

Keywords: dose assessments, fish samples, NORMs, oil, radioactivity.

3.1 Introduction

A higher degree of naturally occurring radioactive materials (NORMs) in the atmosphere has been reported in the past half-century (Samreh *et al.,* 2014; Ali *et al.*, 2019). Such NORMs' activity concentrations are higher than the radiological reference levels recommended for living organisms by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR, 2000). It was further documented that the different materials making the atmosphere contain a variable amount of NORMs and their decay components (N.R.C., 1999). Therefore, gamma radiations from these NORMs are regarded as the predominant external source of irradiation to the human body (UNSCEAR, 2000; Valkovic, 2019).

Artificial sources of radiation raise natural sources in the environment. These artificial sources originate from human-made radionuclides such as those used in medical procedures, the production of nuclear power, and even the effects of a nuclear explosion (Ojovan and Lee, 2014; Wada *et al.*, 2016; WHO, 2016). Radionuclide from these artificial sources may find its way into the environment through periodic releases, injuries, breaches, losses, and inappropriate disposal or violation (Vlado, 2019). Naturally occurring radionuclides emit ionizing radiation that has become part of our daily lives (Ajayi and Dike, 2016). Due to their solubility in water, run-off from the soil, exchange, and extraction of pollutants with the atmosphere, some degree of artificial radionuclide and NORMs is contained in the aquatic environment (Lilley, 2001; IAEA, 2003).

Potassium (^{40}K) is the most abundant radionuclide found in the aquatic environment, followed by Uranium (^{238}U) due to their higher degree of solubility in the water medium. However, Thorium (^{232}Th) is practically insoluble in water as its series are not present in large amounts in sediment samples (Ademola and Ehiedu, 2010).

Aquatic animals, including fish species, ingest radionuclide at concentrations higher than terrestrial levels (Arai, 2014). Fish are not the only aquatic animals' man consumes from the aquatic environment; a wide range of invertebrates are also consumed. Many of these aquatic animals are harvested for direct consumption or as products for certain food products (Jennings *et al.,* 2016). These direct and indirect pathways of the consumption of aquatic animals by man may lead to both external and internal sources of radiation exposure when those aquatic animals contain radioactive elements (Toshihiro *et al.,* 2015).

South Africa has minimal proven oil and gas reserves (SAES, 2018) and the use of radioactive sources in the oil industry involves its usage in well-logging, density gauges, radiography, radiotracers and leak detection for pipelines (ICRP, 1990; IAEA, 2003; Oni et al., 2011). The massive presence of crude oil has been accounted for within South Africa's southern coast and southwest of Mosel Bay (Shannon and Chapman, 1983). Moreover, crude oil reserves at Bredasdorp and deep marine basins have been primarily explored, leading to massive oil produces in South Africa since 1987 (Van, 1989).

The oil industry is one of the leading importers and users of radioactive materials in the sectors of the economy (SAES, 2018). The exploration and exploitation of crude oil and other human activities are known to technologically increase the radionuclides that exist naturally in an environment (IAEA, 2005; Oni *et al.,* 2011; Emumejaye, 2015). Geological formations containing oil and gas deposits contain natural radionuclides, and geologists have recognized their presence and used it as a deposit-finding tool

(IAEA, 2003). Consequently, a higher degree of radionuclides would be released into the aquatic environment by the oil producers' heavy use of radioactive sources (IAEA, 2007; Iwetan *et al.,* 2019). Thus, there may be an increase in the natural radionuclide concentrations in the sediments and aquatic animals of the study areas with additional concern that it may impact both humans and the ecosystem (Babatunde *et al.,* 2015).

The environmental assessment of NORMs provides vital information on the concentration of radioactive elements in the local environment (Alaamer, 2008). Hence, determining the radiological risk to the population living and feeding on the aquatic animals of South Africa's oil-rich environments is of great importance. It plays a critical role in understanding radioactivity-related health risks and paves the way for observation of future changes in environmental radiation activities.

The goal of this study was to determine the level of radioactivity due to the presence of NORMs (^{238}U, ^{232}Th, and ^{40}K) in selected fish samples from South Africa's oil-rich areas. Additionally, radiological analysis, such as the annual effective ingestion dose ($H\ .A.$) and the annual gonadal dose equivalent (AGDE), due to the ingestion of fish samples from the studied areas, were also calculated.

3.2 Materials and Methods

3.2.1 Collection of Samples

Twenty-four fish samples that are widely present in the selected four rivers of the study areas were collected for this study. Fig 3.1 shows the sampling map of the study areas. The fish samples were collected from each selected river with the help of local fish hunters and anglers using hooks and nets, which were eighteen (18) samples from the oil-rich areas (Table 3.1) and six (6)

samples from the non-oil-rich area (Table 3.2). Two samples of each species were collected and were preserved in 40% formaldehyde in labeled containers (Thorp and Rogers, 2016). The edible part of the fish samples was used for the study. The common names, species, and family of the samples are in Tables 3.1 and 3.2.

Table 3.1: The sampled fish species from rivers of oil-rich areas.

Samples Codes	Common Name	Species	Family	River Name	G.P.S. Locations
F1, F2	Spotted grunter	*Pomadasys commersonnii*	Haemulidae		
F3, F4	Kabeljou	*Argyrosomus japonicus*	Sciaenidae	Bree	$34^0 23' 52.4'' S$
F5, F6	Leervis	*Lichia amia*	Carangidae		$20^0 49' 20.1'' E$
F7, F8	Leervis	*Lichia amia*	Carangidae		
F9, F10	Elf	*Pomatomus saltatrix*	Pomatomus	Klein-Brak	$34^0 05' 22.4'' S$
F11, F12	Grunter	*Pomadasys commersonnii*	Haemulidae		$22^0 08' 25.9'' E$
F13, F14	Longfin eel	*Anguilla marmorata*	Anguillidae		
F15, F16	Banded tilapia	*Tilapia sparrmanni*	Cichlidae	Bakens	$33^0 57' 22.0'' S$
F17, F18	Goldie barb	*Enteromius pallidus*	Cyprinidae		$25^0 32' 40.5'' E$

Table 3.2: The sampled fish species from the river of non-oil-rich area (control).

Sampling Code	Common Name	Species	Family	River Name	G.P.S. Locations
F19, F20	Scally yellowfish	*Hypseleotris cyprinoides*	Eleotridae		
F21, F22	Longfin eel	*Anguilla marmorata*	Anguillidae	uMngeni	$29^0 48' 32.8'' S$
F23, F24	Banded tilapia	*Tilapia sparrmanni*	Cichlidae		$31^0 01' 09.5'' E$

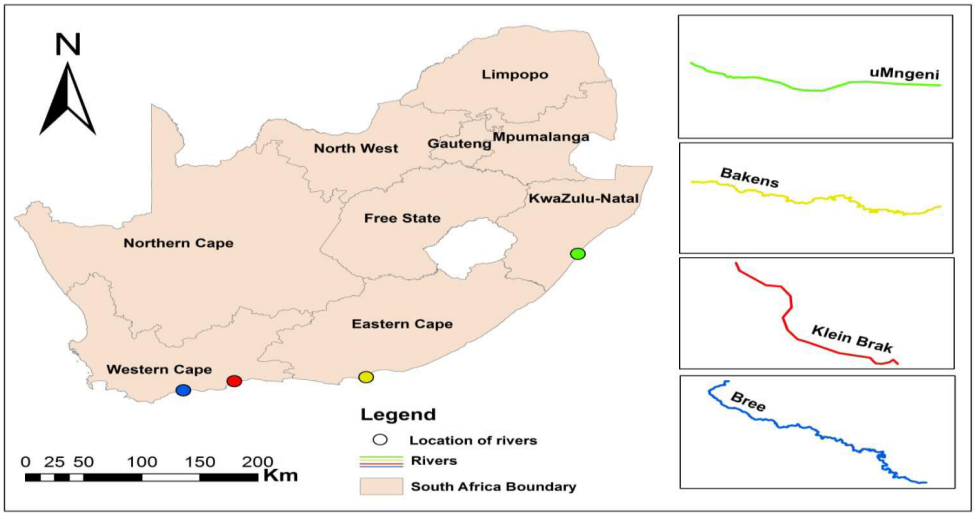

Fig 3.1: South Africa map showing the rivers that were being sampled.

3.2.2 Samples preparation

The fish samples were well-prepared and oven-dried at a temperature of 80^0C (Ng and Dougherty, 1983; Ademola *et al.*, 2010). The dried samples of each sampling were pulverized, weighed, and packed into air-tight Polyvials clear 100 ml plastic pill bottles. The pill bottles were sealed and kept for a minimum of 28 days in other to allow natural radionuclide and their short-lived progeny to obtain secular radioactive equilibrium (Ademola *et al.,* 2010). All samples were prepared at the Radiation Laboratory of the University of KwaZulu-Natal, Pietermaritzburg Campus, South Africa.

3.2.3 Instrumentation and calibration

In this study, the HPGe detector was used to count and detect the radionuclide content in the samples. The detector model is GC4520, a P-type co-axial detector with pre-amplifier and amplifier models 2002CSL and ORTEC 572). This analysis was carried out at the Environmental Radiation Laboratory (E.R.L.) of iThemba LABS Cape Town, South Africa. The detector was cooled to liquid nitrogen temperature, producing spectroscopic data and pulses

proportional to the photon energy captured. The HPGe detector used is 62.5 mm in diameter, 59.5 mm in length with 45 percent relative efficiency, and 2.2 KeV resolutions on the 1332 KeV ^{60}Co line. Connected to the detector was a fully equipped multichannel analyzer (M.C.A.), which includes a pre-amplifying stage, amplifier stage, and a display terminal. Data was collected and analyzed using computer installed PalmtopMCA software.

The detector undergoes a full energy peak and efficiency calibration using generic ^{232}Th, ^{238}U, and ^{40}K reference sources having 4938.8 Bq, 3252 Bq, and 13910.8 Bq. The calibration was performed to determine the presence of radionuclide in the samples. Therefore, a volume source with the same geometry as the sample (100 ml plastic pill bottle) was used for the calibration. The standard reference source was measured for 3600 seconds, and the obtained spectrum was used to produce the efficiency curve (Fig. 3.2). Power fitting was performed to get the best R^2-value.

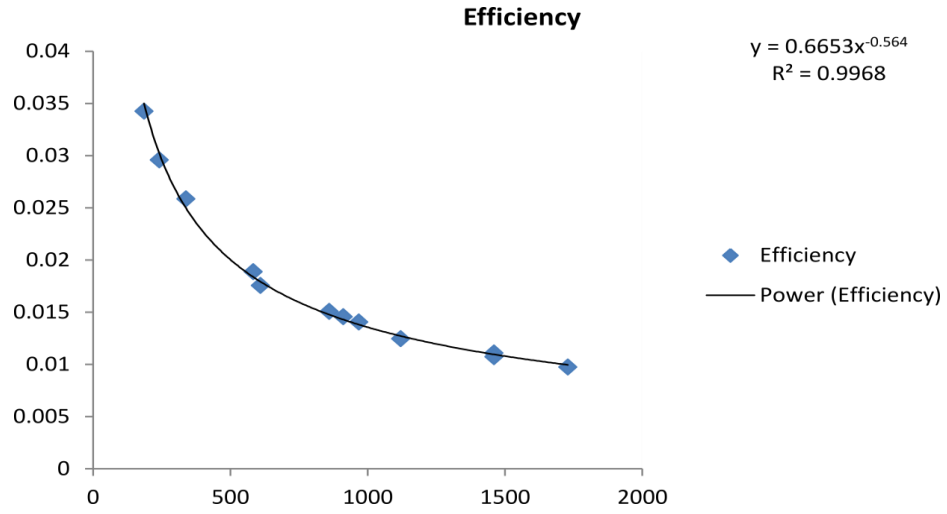

Fig. 3.2: Measurement efficiency curve showing the detection efficiency as a function of the gamma-ray energy used by the HPGe detector.

3.2.4 Calculation of activity concentration

The activity concentration of ^{238}U, ^{232}Th, and ^{40}K in $Bq.kg^{-1}$ (dry weight) was calculated based on measured efficiency, net count rate, mass, and sample count time of the detector. This is presented in the expression (Jibiri and Fasae, 2012; Billa *et al.*, 2016):

$$A_c = \frac{C_\gamma}{P_\gamma . m_s . E_f . t_c} \quad (3.1)$$

Where A_c is the activity concentration for each sample, C_γ is the net peak energy, P_γ is the probability of gamma-ray decay, m_s is the mass of the sample in kg, E_f is the efficiency of the detector and t_c is the total counting time in seconds.

For the gamma analysis, each sample was placed directly on the detector for a 36000-seconds exposure duration. The gamma-ray significance transition defined from the data spectra and the uncertainty associated with each particular nuclide was to assess the activity concentration of the radionuclides. A better approximation of activity concentration was obtained by utilizing the weighted average of each nuclide of interest's specific activity.

3.3 Radiological Dose Assessments

3.3.1 The annual effective ingestion dose assessment (H .A.)

The annual effective ingestion dose was calculated to estimate the resulting internal dose of radiation to the study area's inhabitants because of the fish consumption. This is useful in assessing the health risk posed by ionizing radiation to the population using fish products. “The total dose following ingestion may be calculated by summing the doses for each ^{238}U and ^{232}Th radionuclide.” ^{40}K was excluded in the annual effective dose assessment of ingestion, as it is homeostatically regulated in the human cells as an isotope of

an essential element (Jibiri and Abiodun, 2012). "The addition of any quantity of ^{40}K in living organisms results in a decrease in the biological half-life of ^{40}K." As such, no additional dose is obtained from adding ^{40}K to the body (Billa *et al.*, 2016).

The following equation, adapted from an ICRP analysis, gives the effective dietary intake dose (ICRP, 2007; Fasae and Isinkaye, 2018):

$$H_A(mSv.y^{-1}) = \sum\sum C_f^i\ (Bq.kg^{-1}) \text{ x } U^i(\text{kg.y}^{-1}) \text{ x } {}_gT_y\ (\text{Sv.Bq}^{-1}) \qquad (3.2)$$

where $H_A(mSv.y^{-1})$ is the annual effective ingestion dose, C_f^i $(Bq.kg^{-1})$ is the activity concentrations of radionuclide of interest in the fish sample, U^i (kg.y^{-1}) is the annual consumption rate per capita and ${}_gT_y$ (Sv.Bq^{-1}) is the dose conversion coefficient for the ingestion of radionuclide. The dose conversion coefficients for the radionuclides determined in this work are given as 2.8×10^{-7} and 2.3×10^{-7} Sv.Bq^{-1} for ^{238}U and ^{232}Th, respectively (ICRP, 2007; Khandaker *et al.*, 2015). Fish consumption rate per capita in South Africa was assessed on FAOSTAT Food Balance Sheets (F.B.S.) as 7.5 $kg.y^{-1}$ (Ronquest-Ross *et al.,* 2015).

3.3.2 Annual Gonadal Equivalent Dose (AGED)

For the estimation of the annual equivalent dose of gonads, bone marrow, and the bone surface cells, the organ of interests as specified by (UNSCEAR, 2000; Shetty and Narayana, 2007; Khan *et al.,* 2011), were used in equation (3) considering the activity concentrations of ^{238}U, ^{232}Th and ^{40}K in the samples (Ghose *et al.,* 2000; Ademola and Ehiedu, 2010):

$$AGDE\ (\mu Sv.yr^{-1}) = 3.09C_U + 4.18C_{Th} + 0.314C_K \qquad (3.3)$$

C_U, C_{Th}, and $C\ .K.$ are the radioactivity concentration of ^{238}U, ^{232}Th, and ^{40}K.

3.4 Result and Discussions

The activity concentration of ^{238}U, ^{232}Th and ^{40}K for species of fish collected from the Bree river varied from (6.06 ± 6.54) to (12.57 ± 10.16), (3.18 ± 3.56) to (6.36 ± 7.80) and (73.74 ± 37.02) to (201.67 ± 36.48) $Bq.kg^{-1}$ with a mean activity concentration of (8.60 ± 2.97), (4.26 ± 1.18) and (105.66 ± 47.77) $Bq.kg^{-1}$, respectively.

For fish samples collected from Klein-Brak river, the activity concentration varied from (4.11 ± 3.71) to (14.88 ± 11.75), (1.95 ± 4.90) to (6.85 ± 4.21) and (67.79 ± 30.52) to (169.12 ± 26.53) $Bq.kg^{-1}$ with a mean activity concentration of (8.06 ± 3.64), (4.84 ± 2.00) and (126.88 ± 47.30) $Bq.kg^{-1}$ for ^{238}U, ^{232}Th and ^{40}K, respectively.

For samples collected from Bakens river, the activity concentration varied from (3.68 ± 6.12) to (11.84 ± 6.29), (1.71 ± 5.78) to (5.66 ± 5.43) and (50.34 ± 28.26) to (115.89 ± 27.24) $Bq.kg^{-1}$ with a mean activity concentration of (8.30 ± 3.64), (3.48 ± 1.44) and (90.42 ± 29.35) $Bq.kg^{-1}$ for ^{238}U, ^{232}Th and ^{40}K, respectively.

For samples collected from uMngeni river (control site), the activity concentration varied from (4.19 ± 9.69) to (8.91 ± 4.79), (2.71 ± 4.97) to (7.33 ± 3.77) and (50.45 ± 25.09) to (105.35 ± 26.35) $Bq.kg^{-1}$ with a mean activity concentration of (6.48 ± 2.05), (5.26 ± 1.79) and (78.38 ± 20.55) $Bq.kg^{-1}$ for ^{238}U, ^{232}Th and ^{40}K, respectively (Table 3.3).

The measured activity concentration for rivers at the oil-rich areas (Bree, Klein-Brak, and Bakens rivers) was 12.6%, 21.4%, and 15.6% higher than that of the control site, i.e., non-oil-area (uMngeni river) for ^{238}U, ^{232}Th, and ^{40}K, respectively.

Table 3.3: Activity concentration of natural radionuclides in fish samples of the areas studied.

Samples Codes	^{238}U $(Bq.kg^{-1})$	^{232}Th $(Bq.kg^{-1})$	^{40}K $(Bq.kg^{-1})$
F1	6.19 ± 4.22	4.03 ± 4.98	91.19 ± 34.80
F2	7.91 ± 20.24	4.85 ± 4.68	98.85 ± 32.98
F3	6.06 ± 6.54	3.18 ± 3.56	85.20 ± 25.13
F4	6.76 ± 7.09	3.52 ± 3.48	83.31 ± 24.57
F5	12.57 ± 10.16	3.60 ± 5.29	73.74 ± 37.02
F6	12.09 ± 9.50	6.36 ± 7.80	201.67 ± 36.48
Range	**6.06 - 12.57**	**3.18 - 6.36**	**73.74 - 201.67**
Mean	**8.60 ± 2.97**	**4.26 ± 1.18**	**105.66 ± 47.77**
F7	8.41 ± 4.65	2.78 ± 2.70	130.86 ± 17.19
F8	7.44 ± 5.41	6.29 ± 3.82	169.12 ± 26.53
F9	6.92 ± 9.02	5.44 ± 5.22	68.77 ± 29.45
F10	14.88 ± 11.75	6.85 ± 4.21	163.48 ± 33.25
F11	6.59 ± 6.21	5.75 ± 4.60	161.24 ± 33.27
F12	4.11 ± 3.71	1.95 ± 4.90	67.79 ± 30.52
Range	**4.11 - 14.88**	**1.95 - 6.85**	**67.79 - 169.12**
Mean	**8.06 ± 3.64**	**4.84 ± 2.00**	**126.88 ± 47.30**
F13	7.25 ± 6.23	5.66 ± 5.43	50.34 ± 28.26
F14	11.84 ± 6.29	4.53 ± 6.02	93.80 ± 29.11
F15	11.46 ± 10.37	2.37 ± 6.59	110.10 ± 26.80
F16	10.99 ± 7.95	3.06 ± 5.73	114.65 ± 30.64
F17	3.68 ± 6.12	3.54 ± 4.43	115.89 ± 27.24
F18	4.55 ± 5.16	1.71 ± 5.78	57.76 ± 27.67
Range	**3.68 - 11.84**	**1.71 - 5.66**	**50.34 - 115.89**
Mean	**8.30 ± 3.64**	**3.48 ± 1.44**	**90.42 ± 29.35**
F19	8.91 ± 4.79	4.69 ± 6.56	60.34 ± 26.26

F20	8.25 ± 7.75	7.33 ± 3.77	50.45 ± 25.09
F21	5.69 ± 4.81	2.71 ± 4.97	81.56 ± 26.54
F22	7.59 ± 4.25	4.09 ± 4.56	105.35 ± 26.35
F23	4.19 ± 9.69	7.08 ± 7.19	77.79 ± 32.52
F24	4.27 ± 4.33	5.68 ± 4.96	94.76 ± 30.65
Range	**4.19 - 8.91**	**2.71 - 7.33**	**50.45 - 105.35**
Mean	**6.48 ± 2.05**	**5.26 ± 1.79**	**78.38 ± 20.55**

Table 3.4: Annual effective ingestion dose and annual gonadal dose equivalent in samples of fish from the studied areas.

Samples Codes	$\mathbf{H_U}$ $(\boldsymbol{mSv.y^{-1}})$	$\mathbf{H_{Th}}$ $(\boldsymbol{mSv.y^{-1}})$	$\mathbf{H_A}$ $(\boldsymbol{mSv.y^{-1}})$	**AGDE** $(\boldsymbol{\mu Sv.yr^{-1}})$
F1	0.036	0.019	0.055	64.61
F2	0.046	0.023	0.068	75.75
F3	0.035	0.015	0.050	58.77
F4	0.039	0.017	0.056	61.76
F5	0.072	0.017	0.089	77.04
F6	0.070	0.030	0.100	127.27
Mean	**0.049**	**0.020**	**0.070**	**77.53**
F7	0.048	0.013	0.062	78.70
F8	0.043	0.030	0.073	102.39
F9	0.040	0.026	0.066	65.72
F10	0.086	0.032	0.118	125.94
F11	0.038	0.027	0.065	95.03
F12	0.024	0.009	0.033	42.14
Mean	**0.046**	**0.023**	**0.069**	**84.98**
F13	0.042	0.027	0.068	61.87
F14	0.068	0.021	0.090	84.97

F15	0.066	0.011	0.077	79.89
F16	0.063	0.014	0.078	82.75
F17	0.021	0.017	0.038	62.56
F18	0.026	0.008	0.034	39.34
Mean	**0.048**	**0.016**	**0.064**	**68.56**
F19	0.051	0.022	0.073	66.08
F20	0.047	0.035	0.082	71.97
F21	0.033	0.013	0.046	54.52
F22	0.044	0.019	0.063	73.63
F23	0.024	0.033	0.058	66.97
F24	0.025	0.027	0.051	66.69
Mean	**0.037**	**0.025**	**0.062**	**66.64**

Table 3.4 shows the annual effective ingestion and annual gonadal doses equivalent in samples of fish from the studied areas. The annual effective ingestion dose (H .A.) due to the content of the natural radionuclides (^{238}U, ^{232}Th, and ^{40}K) in the fish samples from the rivers in the oil-rich areas varied.

i For Bree river, it varied from 0.050 $mSv.y^{-1}$ (*Argyrosomus japonicas*) to 0.100 $mSv.y^{-1}$ (*Lichia amia*).

ii For Klein-Brak river, it varied between 0.033 $mSv.y^{-1}$ (*Pomadasys commersonnii*) and 0.118 $mSv.y^{-1}$ (*Pomatomus saltatrix*).

iii For Bakens river, it varied between 0.034 $mSv.y^{-1}$ (*Enteromius pallidus*) and 0.090 $mSv.y^{-1}$ (*Anguilla marmorata*).

The activity concentrations of fish samples from uMngeni river (control) varied between 0.046 $mSv.y^{-1}$ (*Anguilla marmorata*) and 0.082 $mSv.y^{-1}$ (*Hypseleotris cyprinoides*). The estimated annual effective ingestion dose for all fish samples is found to be less than 1 $mSv.y^{-1}$, as recommended by UNSCEAR (2000). Therefore, from a radiological point of view, the radiation exposure received from the ingestion of the fish samples from the areas examined did not present

any major health risk to the population. Figure 3.3 shows the activity concentrations of ^{238}U, ^{232}Th, and ^{40}K in the fish samples from the areas studied.

The calculated values of the annual equivalent dose of gonads, bone marrow and the bone surface cells due to the ingestion of fish samples from the oil-rich areas ranged from 58.77 to 127.27 $mSv.y^{-1}$ with an average of 77.53 $mSv.y^{-1}$ for the Bree, 42.14 to 125.94 $mSv.y^{-1}$ with an average of 84.98 $mSv.y^{-1}$ for Klein-Brak and 39.34 to 84.97 $mSv.y^{-1}$ with an average of 68.56 $mSv.y^{-1}$ for Bakens rivers, respectively.

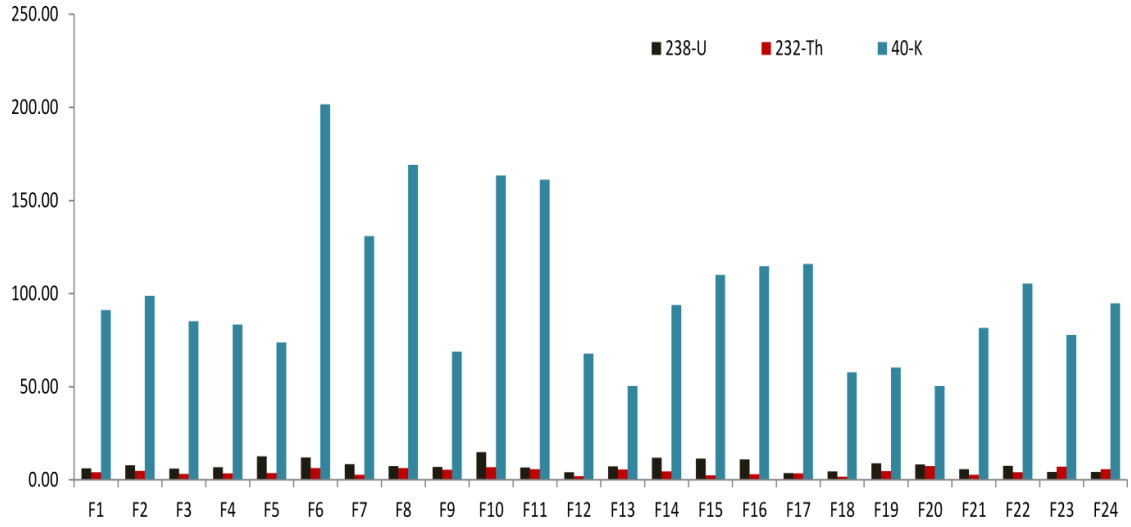

Figure 3.3: Concentrations of activity of ^{238}U, ^{232}Th, and ^{40}K in fish samples from the areas studied.

The value calculated for the control area (uMngeni river) ranged between 54.54 to 71.97 $mSv.y^{-1}$, with an average of 66.64 $mSv.y^{-1}$. The contribution of ^{238}U and ^{232}Th to the annual effective ingestion dose due to the consumption of fish species in the studied areas is shown in Figure 3.4. While the values calculated for rivers at the oil-rich areas were higher than the control river, all values were lower than the UNSCEAR (2000) recommended standard value of 300 $\mu Sv.y^{-1}$.

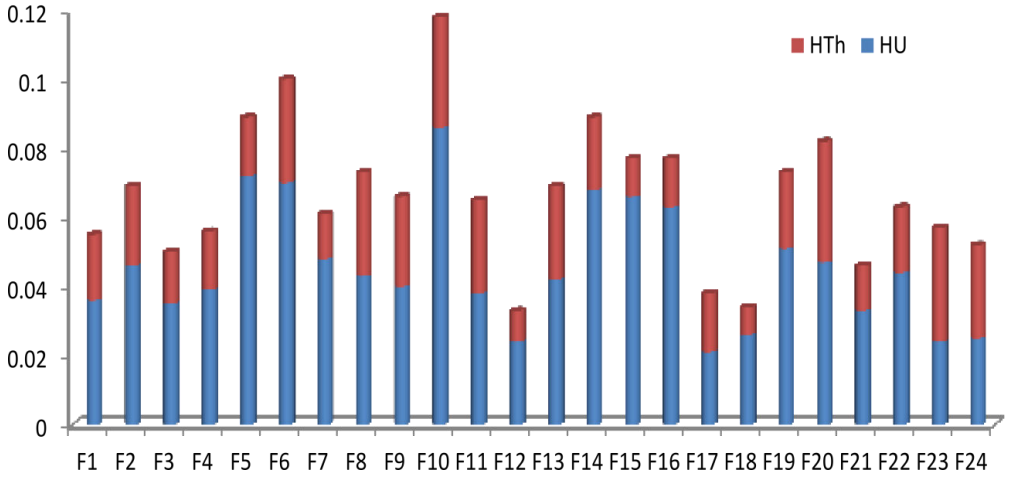

Figure 3.4: Contribution of Uranium and Thorium to the annual effective ingestion dose due to the consumption of fish species in the studied areas.

Table 3.5: Comparison of activity concentrations in fish samples of the studied areas with other studies in different countries.

		Activity Concentration ($Bq.kg^{-1}$,)			
Country	**Location**	**^{238}U**	**^{232}Th**	**^{40}K**	**References**
South Africa	Breede river	6.06-12.57	3.18-6.36	73.74-201.67	Present study
	Klein-Brakriver	4.11-14.88	1.95-6.85	67.79-169.12	
	Baakens river	3.68-11.84	1.71-5.66	50.34-115.89	
	Umgeni river	4.19-8.91	2.71-7.33	50.45-105.35	
Bangladesh	Bengal	1.05	0.77	61.5	Ghose *et al.*, 2000
Brazil	Ceara river	0.190-0.650	0.017-0.033	-	Pereira *et al.*, 2010
Nigeria	Igbokoda river	17.7-86.9	13.8-86.8	385-952	Ademola & Ehiedu, 2010
United States	Amchitka Island	0.21 – 0.112	-	-	Burger *et al.*, 2007
Portugal	North Atlantic Ocean	2.0 – 30.2	-	32 - 152	Carvalho *et al.*, 2011
World Average		33	45	420	UNSCEAR, 2008

The activity concentrations of natural radionuclides of interest in fish samples from the present study compared to values reported for different parts of the world and average world values were shown in Table 3.5.

3.5 Conclusion

The activity concentration of natural radionuclide (^{238}U, ^{232}Th, and ^{40}K) was investigated using the Hyper Pure Germanium (HPGe) detector in fish samples of rivers from oil-rich and non-oil-rich (control) areas of South Africa. The annual effective ingestion dose and the annual gonadal equivalent dose due to radionuclide intake by the ingestion of fish from the studied area were also evaluated. The measured activity concentrations for rivers at the oil-rich areas (Bree, Klein-Brak, and Bakens rivers) were 12.6%, 21.4%, and 15.6% higher than that of the control site, the non-oil-area (uMngeni river) for ^{238}U, ^{232}Th, and ^{40}K, respectively. The results obtained for the equivalent doses were lower than the UNSCEAR recommended doses of 1 $mSv.y^{-1}$ for the effective annual intake dose and 300 $\mu Sv.yr^{-1}$ for the annual gonadal equivalent dose. The results of activity concentration in fish samples from the study areas do not pose a radiological risk at the time of the study. However, industrial activity in the study area may increase the activity concentrations and the radiological hazard due to the ingestion of fish and other aquatic animals. The study's findings may be used as reference data in the study area for potential monitoring of possible future radioactivity pollution.

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CHAPTER 4

ACTIVITY CONCENTRATION OF NATURAL RADIONUCLIDES IN SEDIMENTS OF BREE, KLEIN-BRAK, BAKENS, AND UMNGENI RIVERS AND ITS ASSOCIATED RADIATION HAZARD INDICES

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Activity Concentration of Natural Radionuclides in Sediments of Bree, Klein-Brak, Bakens, and uMngeni Rivers and its Associated Radiation Hazard Indices.

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Abstract

The Hyper Pure Germanium (HPGe) detector was used to measure the activity concentrations in sediment samples of rivers in South Africa, and its associated radiological hazard indices were evaluated. The results of the study indicated that the mean activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in the sediment samples from the oil-rich areas are 11.13, 7.57, 22.5 $Bq.kg^{-1}$; 5.51, 4.62, 125.02 $Bq.kg^{-1}$ and 7.60, 5.32, 24.12 $Bq.kg^{-1}$ for the Bree, Klein-Brak and Bakens Rivers, respectively. In contrast, the control site (UMngeni River) was 4.13, 3.28, and 13.04 $Bq.kg^{-1}$ for ^{226}Ra, ^{232}Th, and ^{40}K. The average excess lifetime cancer risks are 0.394 x 10^{-3}, 0.393 x 10^{-3}, 0.277 x 10^{-3}, and 0.163 x 10^{-3} for sediment samples at Bree, Klein-Brak, Bakens, and uMngeni rivers. All obtained values indicated a significant difference between the natural radionuclide concentrations in the samples from the rivers in oil-rich areas compared to that of the non-oil-rich area. The values reported for the activity concentrations and radiological hazard indices were below the average world values; hence, the risk of radiation health hazard was negligible in all study areas.

Keywords: concentrations, hazard indices, natural radionuclides, radiation, sediments.

4.1 Introduction

Human exposure to radiation from naturally occurring radioactive materials has been investigated by several authors to determine the impact of the environment on exposure (Karahan and Bayulken, 2000; Khan *et al.,* 2003; Oni *et al.,* 2011; Ajayi and Dike, 2016). The degree of exposure from these natural radionuclides varies with location and is often enhanced by artificial sources through industrial activities (Cenci and Martin, 2004; Paschoa and Steinhäusler, 2010; Krishnamoorthy *et al.,* 2014; Ajanaku *et al.,* 2018). Natural radiation impacting human safety is most often those from the ^{238}U and ^{232}Th series radionuclides and their decay products and ^{40}K (Eisenbud and Gesell, 1997).

The existence of other natural resources, such as oil and gas, will further increase natural radionuclides in such environments due to the excessive use of radiation sources in the oil and gas exploration activities (Whicker and Schultz, 1982; Paschoa *et al.,* 2010). South Africa has minimal proven oil and gas reserves; however, oil and gas exploration in the country started in 1913. The oil industry is one of the leading importers and users of radioactive materials in many sectors of the economy (SAES, 2018). The use of radioactive sources in the industry involves operations in nuclear well-logging, nuclear density gauges, radiography, and radiotracers in the maintenance of oil wells, studies of reservoirs, and pipelines for leak detection (Oni *et al.,* 2011; IAEA, 2003; ICRP, 1991). The massive presence of crude oil has been accounted for within South Africa's southern coast and southwest of Mosel Bay (Shannon and Chapman, 1983). Moreover, crude oil reserves at Bredasdorp and deep marine basins have been primarily explored, leading to massive oil produces in South Africa since 1987 (van Wyk, 1989).

There may be a high degree of radionuclide contamination in the study areas sediment samples with additional concern that it may impact both humans and the environment. Thus, determining the radiological risk to the population living around oil-rich environments is of great importance. It plays a critical role in understanding radioactivity-related health risks and paves the way for observation of future changes in environmental radiation activities.

This study aimed to determine the level of natural radionuclides (^{226}Ra, ^{232}Th, and ^{40}K) in the river sediments of oil-rich areas (Bree, Klein-Brak, Bakens) and that of non-oil-rich area (uMngeni), serving as a control site. The following associated hazard indices were calculated: the radium equivalent, absorbed dose rates at outdoor and indoor, annual outdoor and indoor effective doses, and excess lifetime cancer risk of sediment samples from the studied areas.

4.2 Materials and methods

4.2.1 Samples collection

For this study, sixteen samples of sediments were collected using grab samplers (Ravisankar *et al.,* 2015) from four rivers within the studied areas. The sediment samples collected consists of particulate organic and inorganic matter. The samples were collected at an average distance of 10 m from the riverside and 1-2 m depth. Four sediment samples of each river were collected to obtain an average of the radioactivity concentration for uniform distribution across the area of interest. For the Bree, Klein-Brak, Bakens, and uMngeni rivers' sediments, the sampling codes are S1-S4, S5-S8, S9-S12, and S13-S16, respectively. Sediment samples were collected for the oil-rich and non-oil-rich areas. The water in each sediment sample was drained, packed, labeled, and transported for further laboratory processing. Figure 4.1 illustrates the locations of the rivers selected for the study in South Africa.

4.2.2 Preparation of samples

The sediment samples were well prepared and oven-dried until pulverized at a temperature of 105^{0}C (Iwetan *et al.,* 2015). Dried samples of each sampling were weighed and packed into airtight Polyvials clear 100 ml plastic pill bottles (IAEA, 1989). The pill-bottles were sealed and stored for at least 28 days in others to allow natural radionuclides and their short-lived progeny to achieve secular radioactive equilibrium (ICRP, 1990; Xinwei and Xiaolan, 2006). All samples were prepared at the Laboratory in the Physics discipline at the University of KwaZulu-Natal, Pietermaritzburg, KwaZulu-Natal, South Africa.

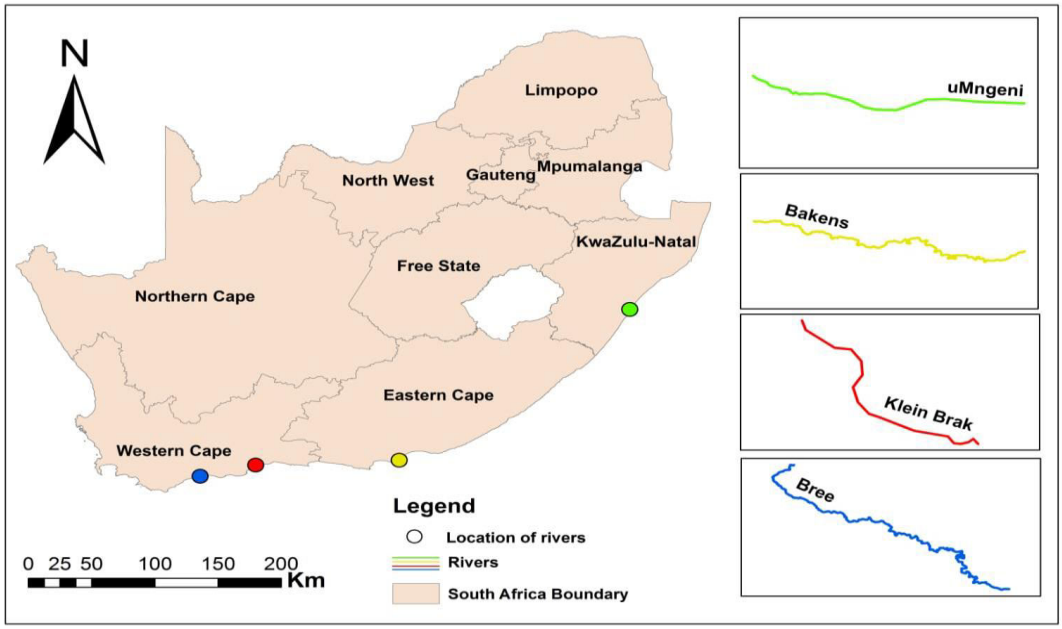

Fig 4.1: South Africa map showing the rivers that were being sampled.

4.2.3 Radioactivity measurements

In this study, HPGe detector was used to count and detect the radionuclide content in the samples. Measurements of the samples' radioactivity level were performed at the Environmental Radiation Laboratory (ERL) of iThemba LABS Cape Town, South Africa. The detector was cooled to liquid nitrogen's temperature, producing spectroscopic data and pulses proportional to the photon energy captured (Wallbrink *et al.,* 2002; Simon *et al.,* 2012; Guembou

et al., 2017). The HPGe detector used on the 1332 KeV ^{60}Co line is 62.5 mm in diameter, 59.5 mm in length, and 45 percent relative efficiency and 2.2 KeV resolutions. The detector was connected to a fully equipped multichannel analyzer (MCA), which includes a pre-amplifying stage, amplifier stage, and a display terminal. Data was collected and analyzed using computer installed PalmtopMCA software.

The detector undergoes a full energy peak and efficiency calibration using generic ^{232}Th, ^{226}Ra, and ^{40}K reference sources with 4938.8 Bq, 3252 Bq, and 13910.8 Bq. The calibration was done to assess the presence of radionuclide in the samples. Thus, a volume source with the same configuration as the sample, which was 100 ml plastic bottles, was used for calibration. The standard reference source was calculated for 3600 seconds, and the spectrum obtained was used to generate the efficiency curve.

The minimum detection limit (MDL) of the gamma-ray detector system is its operational capability without the sample's influence (Khandaker *et al.,* 2012; Ibitola *et al.,* 2018). It is the smallest amount of radionuclide of interest that can be accurately measured, which was estimated according to the equation below (Knoll, 2010; Kirkpatrick *et al.,* 2015):

$$MDL(Bq.kg^{-1}) = \frac{k_{\propto}\sqrt{N_B}}{n(E).P_{\gamma}.T_c.m} \quad (4.1)$$

Where $k_{\propto}$ is the factor that converts cps (counting per seconds) to Bq given as 1.96 at 95% confidence level, N_B is the net background count rate at the region of interest of radionuclide of interests, $n(E)$ is the counting photo-peak efficiency ($cps.Bq^{-1}$), P_{γ} is the probability of gamma-ray emission, T_c is the counting time (s), and m is the dry weight of the sample (kg). The MDL for the radionuclides of interest were 0.75, 0.84, and 2.21 $Bq.kg^{-1}$ for ^{226}Ra, ^{232}Th, and ^{40}K, respectively.

4.2.4 Activity concentration

The activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K in $Bq.kg^{-1}$ was determined based on the detector's measured efficiency, net count rate, mass and sample count time. It is presented in the expression (Jibiri and Fasae, 2012; Mekongtso *et al.,* 2016):

$$A_c = \frac{C_\gamma}{P_\gamma . m_s . E_f . t_c} \quad (4.2)$$

Where A_c is the activity concentration for each sample, C_γ is the net peak energy, P_γ is the probability of gamma-ray decay, m_s is the mass of the sample in kg, E_f is the detector's efficiency, and t_c is the total counting time in seconds. For the gamma analysis, each sample was put directly on the detector over 36000-seconds exposure duration. In addition to the uncertainty associated with each nuclide, the gamma-ray transition of interest detected from the data spectra was used to assess the activity's exact concentration. A better estimate of activity concentration has been obtained using the weighted average of each nuclide of interest's specific activity and results shown in Table 4.1.

4.3 Radiation hazard indices

4.3.1 Radium equivalent (R_{eq})

The radium activity index provides a valuable mechanism for monitoring safety standards related to both the external and the internal radon doses and its progeny. The index of activity equivalent to radium was calculated as given the equation (El-Taher *et al.,* 2019):

$$R_{eq}(Bq.kg^{-1}) = C_{Ra} + 1.43C_{Th} + 0.077C_K \quad (4.3)$$

Where C_{Ra},C_{Th}, and C_K are the radioactivity concentration in $Bq.kg^{-1}$ of ^{226}Ra, ^{232}Th, and ^{40}K.

4.3.2 Absorbed dose rates - $D_{(out)}$ and $D_{(in)}$

The absorbed dose rates establish a relationship between the standardized distribution of natural radionuclides (^{226}Ra, ^{232}Th, and ^{40}K) by a sample and its human exposure at 1 m above ground level (IAEA, 2010; El-Taher *et al.,* 2019). The absorbed dose rates (D) in $nGyh^{-1}$ are determined based on the guidelines provided by the European Commission by adding outdoor and indoor dose conversion factors for ^{226}Ra, ^{232}Th, and ^{40}K in $Bq.kg^{-1}$ (dry weight), respectively (UNSCEAR, 2000):

$$D_{(out)}(nGyh^{-1}) = 0.0417C_K + 0.599C_{Th} + 0.436C_{Ra} \quad (4.4)$$

$$D_{(in)}(nGyh^{-1}) = 0.081C_K + 1.1C_{Th} + 0.92C_{Ra} \quad (4.5)$$

Where C_K, C_{Th}, and C_{Ra} stand for the specific activities of corresponding radionuclides.

4.3.3 Annual outdoor & indoor effective doses ($E_{(out)}$ and $E_{(in)}$)

In the study, the annual outdoor and indoor effective doses related to gamma radiation have been evaluated to complement the environmental risk associated with absorbed dose (UNSCEAR, 2000; Jibiri *et al.,* 2007). The conversion coefficient from the absorbed dose, the indoor, and the outdoor occupancy variables were parameters considered to calculate the annual effective doses (Xinwei *et al.,* 2006; Celik *et al.,* 2010; Huang *et al.,* 2015).

The conversion coefficient of 0.7 $Sv.Gy^{-1}$ was used to calculate the effective dose ingested in the air obtained by adults. 0.2 (20%) was used as the time of stay in the outdoor and 0.8 (80%) as the time of stay in the indoor in a year (Gilbert *et al.,* 2018). The associated annual indoor and outdoor effective doses were estimated with the following equation (Eisenbud *et al.,* 1997; IAEA, 2003; Oni *et al.,* 2011):

$E_{(out)}\ (\mu Sv.yr^{-1}) = D_{(out)}\ (nGy.h^{-1})$ x 8760 $(h.y^{-1})$ x 0.2 x 0.7 $(Sv.Gy^{-1})$ x 10^{-3} (4.6)

$E_{(in)}\ (\mu Sv.yr^{-1}) = D_{(in)}\ (nGy.h^{-1})$ x 8760 $(h.y^{-1})$ x 0.8 x 0.7 $(Sv.Gy^{-1})$ x 10^{-3} (4.7)

4.3.4 Excess lifetime cancer risk (ELCR)

Over a lifetime, the risk of cancer is characterized as an excessive chance of lifetime cancer due to radiation exposure factors (Dragović *et al.,* 2006; Aziz *et al.,* 2014; Adeleye and Chetty, 2017). ELCR was calculated using the equation provided in the International Commission on Radiological Protection (ICRP) publication 60 (ICRP, 1990; Iwetan *et al.,* 2015).

$ELCR_{(out)} = (E_{(out)}$ x DL x $RF)$ x 10^{-3} (4.8)

$ELCR_{(in)} = (E_{(in)}$ x DL x $RF)$ x 10^{-3} (4.9)

In the above, DL is the average lifespan (estimated at 70 years), while RF is the risk factor ICRP gave in 1991 as 0.05.

4.4 Results and discussion

Table 4.1: Activity of natural radionuclides in sediment samples from the study areas.

Samples Codes	Name of River	GPS Coordinate	^{226}Ra $(Bq.kg^{-1})$	^{232}Th $(Bq.kg^{-1})$	^{40}K $(Bq.kg^{-1})$
S1	Bree	$34^0 23' 52.4'' S$ $20^0 49' 20.1'' E$	11.06 ± 1.02	8.43 ± 2.28	15.65 ± 10.52
S2			8.94 ± 1.04	5.95 ± 1.90	28.55 ± 10.70
S3			11.10 ± 3.42	7.98 ± 6.54	24.53 ± 28.31
S4			13.42 ± 4.27	7.92 ± 9.27	21.36 ± 12.49
Range			**8.94 – 13.42**	**5.95 – 8.43**	**15.65 – 28.55**
Mean			**11.13 ± 1.83**	**7.57 ± 1.10**	**22.52 ± 5.45**
Total			**44.52**	**30.28**	**90.09**
S5	Klein-Brak	$34^0 05' 22.4'' S$ $22^0 08' 25.9'' E$	6.10 ± 1.75	4.62 ± 1.14	132.09 ± 10.32
S6			3.16 ± 4.27	3.87 ± 1.45	87.39 ± 9.12
S7			6.53 ± 5.44	5.99 ± 6.22	154.73 ± 33.26
S8			6.27 ± 7.01	4.01 ± 3.06	125.88 ± 25.58

Range			**3.16 – 6.53**	**3.87 – 5.99**	**87.39 – 154.73**
Mean			**5.51 ± 1.58**	**4.62 ± 0.97**	**125.02 ± 27.99**
Total			**22.06**	**18.49**	**500.09**
S9	Bakens	$33^0 57' 22.0'' S$ $25^0 32' 40.5'' E$	7.84 ± 3.45	4.69 ± 1.19	12.45 ± 7.49
S10			6.67 ± 2.50	6.43 ± 1.45	39.21 ± 7.67
S11			6.46 ± 3.75	4.95 ± 2.51	22.99 ± 32.65
S12			9.45 ± 7.73	5.22 ± 3.92	21.82 ± 34.27
Range			**6.46-9.45**	**4.69-6.43**	**12.45-39.21**
Mean			**7.60 ± 1.37**	**5.32 ± 4.77**	**24.12 ± 11.11**
Total			**30.42**	**21.29**	**96.47**
S13	uMngeni (control)	$29^0 48' 32.8'' S$ $31^0 01' 09.5'' E$	4.52 ± 1.67	3.19 ± 4.96	13.60 ± 6.72
S14			3.12 ± 4.61	2.85 ± 6.99	13.35 ± 6.95
S15			4.58 ± 3.39	3.02 ± 4.57	13.48 ± 7.42
S16			4.29 ± 11.42	4.06 ± 3.81	11.73 ± 10.75
Range			**3.12-4.58**	**2.85-4.06**	**11.73-13.60**
Mean			**4.13 ± 0.68**	**3.28 ± 2.54**	**13.04 ± 1.88**
Total			**16.51**	**13.12**	**52.16**

Natural radionuclide specific activities were measured in selected sediment samples from various rivers in South Africa's oil-rich areas (Bree, Klein-Brak, and Bakens) and non-oil-rich area (uMngeni). Results, sampling positions, GPS coordinates, and sampling codes are displayed in Table 4.1.

The activity concentration values for sediment samples at Bree river ranged from 8.94 ± 1.04 to 13.42 ± 4.27 $Bq.kg^{-1}$ with a mean value of 11.13 ± 1.83 $Bq.kg^{-1}$, 5.95 ± 1.90 to 8.43 ± 2.28 $Bq.kg^{-1}$ with a mean value of 7.57 ± 1.10 $Bq.kg^{-1}$ and 15.65 ± 10.52 to 28.55 ± 10.70 $Bq.kg^{-1}$ with a mean value of 22.52 ± 5.45 $Bq.kg^{-1}$for ^{226}Ra, ^{232}Th and ^{40}K respectively.

The activity concentration values for sediment samples at Klein-Brak river ranged from 3.16 ± 4.27 to 6.53 ± 5.44 $Bq.kg^{-1}$ with a mean value of 5.51 ± 1.58 $Bq.kg^{-1}$, 3.87 ± 1.45 to 5.99 ± 6.22 $Bq.kg^{-1}$ with a mean value of 4.62 ± 0.97 $Bq.kg^{-1}$ and 87.39 ± 9.12 to 154.73 ± 33.26 $Bq.kg^{-1}$ with a mean value of 125.02 ± 27.99 $Bq.kg^{-1}$for ^{226}Ra, ^{232}Th and ^{40}K respectively.

The activity concentration values for sediment samples at Bakens river ranged from 6.46 ± 3.75 to 9.45 ± 7.73 $Bq.kg^{-1}$ with a mean value of 7.60 ± 1.37 $Bq.kg^{-1}$, 4.69 ± 1.19 to 6.43 ± 1.45 $Bq.kg^{-1}$ with a mean value of 5.32 ± 0.77 $Bq.kg^{-1}$ and 12.45 ± 7.49 to 39.21 ± 7.67 $Bq.kg^{-1}$ with a mean value of 24.12 ± 11.11 $Bq.kg^{-1}$for ^{226}Ra, ^{232}Th and ^{40}K respectively.

The activity concentration values for sediment samples at uMngeni river, the non-oil-rich area (control), ranged from 3.12 ± 4.61 to 4.58 ± 3.39 $Bq.kg^{-1}$ with a mean value of 4.13 ± 0.68 $Bq.kg^{-1}$, 2.85 ± 6.99 to 4.06 ± 3.81 $Bq.kg^{-1}$ with a mean value of 3.28 ± 0.54 $Bq.kg^{-1}$ and 11.73 ± 10.75 to 13.60 ± 6.72 $Bq.kg^{-1}$ with a mean value of 13.04 ± 5.88 $Bq.kg^{-1}$for ^{226}Ra, ^{232}Th and ^{40}K respectively. Figure 4.2 shows the mean activity concentrations of natural radionuclides and the radium equivalent in sediment samples from the selected sampled river sediments of South Africa.

The estimated average radium equivalent values in rivers of oil-rich areas (Bree, Klein-Brak, and Bakens) in the present study are 23.69, 21.75, and 17.07 $Bq.kg^{-1}$, respectively. In contrast, the non-oil-rich area (Umgeni river) was 9.82 $Bq.kg^{-1}$ (Table 4.2). The reported values are lower than 370 $Bq.kg^{-1}$, the value for the world average radium equivalent activity given by UNSCEAR (UNSCEAR, 2000).

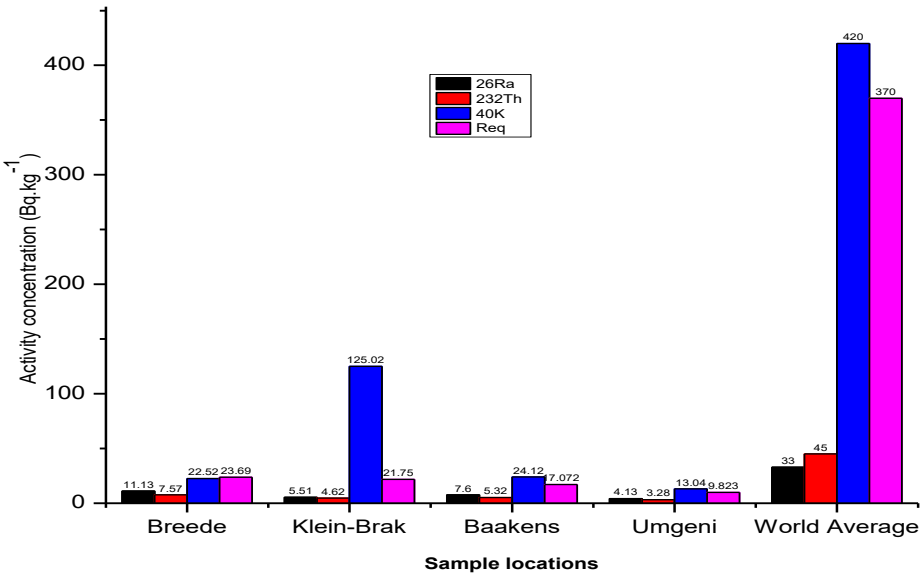

Figure 4.2: Mean activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K, and R_{eq} in sediment samples from locations studied.

The average outdoor absorbed dose recorded for sediment samples of the rivers Bree, Klein-Brak, Bakens, and uMngeni due to natural radionuclides was 10.326, 10.386, 7.509, and 4.308 $nGyh^{-1}$ (Table 4.2). The average indoor absorbed dose recorded for sediment samples of the rivers Bree, Klein-Brak, Bakens, and uMngeni due to the presence of natural radionuclides was 20.391, 20.284, 14.804 and 8.426 $nGyh^{-1}$ (Table 4.2). All values recorded are below the world average of 59 $nGyh^{-1}$ and 84 $nGyh^{-1}$ (UNSCEAR, 2000) for an outdoor and indoor absorbed dose.

The average outdoor effective dose for Bree, Klein-Brak, Bakens, and uMngeni rivers is 12.664, 12.737, 9.209, and 5.284 $\mu Sv.yr^{-1}$, as shown in Table 4.3. The average world value reported in UNSCEAR Report (2000) for outdoor effective dose is 70 $\mu Sv.yr^{-1}$, which is higher than values reported in the present study. The average indoor effective dose for Bree, Klein-Brak, Bakens, and uMngeni rivers is 100.031, 99.504, 77.621, and 41.513 $\mu Sv.yr^{-1}$, as shown in Table 4.3. The average world value reported in UNSCEAR report (UNSCEAR, 2000) for an

effective indoor dose is 410 $\mu Sv.yr^{-1}$, which is higher than the present study's values. The values are more than four times lower than the average world value for all the sediment samples used for this study.

The average excess lifetime cancer risk due to outdoor exposures are 0.044 x 10^{-3} , 0.045 x 10^{-3}, 0.032 x 10^{-3} and 0.018 x 10^{-3} while that of indoor exposures are 0.350 x 10^{-3}, 0.348 x 10^{-3}, 0.254 x 10^{-3} and 0.145 x 10^{-3} with a total average excess lifetime cancer risk for each sampling location as 0.394 x 10^{-3}, 0.393 x 10^{-3}, 0.277 x 10^{-3} and 0.163 x 10^{-3} for Bree, Klein-Brak, Bakens and uMngeni rivers respectively (Table 4.3).

Figure 4.3 shows the contribution of natural radionuclide of interest in the sampled river sediments of South Africa. The activity concentrations of natural radionuclides of interest in sediment samples from the present study compared to values reported for different parts of the world and average world values were shown in Table 4.4.

Table 4.2: Activity of natural radionuclides (^{226}Ra, ^{232}Th and ^{40}K), radium equivalent dose (R_{eq}), outdoor $D_{(out)}$, and indoor $D_{(in)}$ absorbed dose rates of sediment samples from the study areas.

Samples Codes	**^{226}Ra ($Bq.kg^{-1}$)**	**^{232}Th ($Bq.kg^{-1}$)**	**^{40}K ($Bq.kg^{-1}$)**	**$D_{(out)}$ ($nGyh^{-1}$)**	**$D_{(in)}$ ($nGyh^{-1}$)**	**R_{eq} ($Bq.kg^{-1}$)**
S1	11.06 ± 1.02	8.43 ± 2.28	15.65 ± 10.52	10.524	20.716	24.320
S2	8.94 ± 1.04	5.95 ± 1.90	28.55 ± 10.70	8.652	17.082	19.647
S3	11.10 ± 3.42	7.98 ± 6.54	24.53 ± 28.31	10.643	20.978	24.402
S4	13.42 ± 4.27	7.92 ± 9.27	21.36 ± 12.49	11.486	22.789	26.390
Mean	**11.13 ± 1.83**	**7.57 ± 1.10**	**22.52 ± 5.45**	**10.326**	**20.391**	**23.690**
Total	**44.52**	**30.28**	**90.09**	**41.306**	**81.565**	**94.759**
S5	6.10 ± 1.75	4.62 ± 1.14	132.09 ± 10.32	10.935	21.393	22.878
S6	3.16 ± 4.27	3.87 ± 1.45	87.39 ± 9.12	7.340	14.243	15.423
S7	6.53 ± 5.44	5.99 ± 6.22	154.73 ± 33.26	12.884	25.123	27.002
S8	6.27 ± 7.01	4.01 ± 3.06	125.88 ± 25.58	10.385	20.376	21.697

Mean	**5.51 ± 1.58**	**4.62 ± 0.97**	**125.02 ± 27.99**	**10.386**	**20.284**	**21.750**
Total	**22.06**	**18.49**	**500.09**	**41.544**	**81.135**	**87.000**
S9	7.84 ± 3.45	4.69 ± 1.19	12.45 ± 7.49	6.747	13.380	15.505
S10	6.67 ± 2.50	6.43 ± 1.45	39.21 ± 7.67	8.395	16.385	18.884
S11	6.46 ± 3.75	4.95 ± 2.51	22.99 ± 32.65	6.738	13.246	15.303
S12	9.45 ± 7.73	5.22 ± 3.92	21.82 ± 34.27	8.157	16.203	18.595
Mean	**7.60 ± 1.37**	**5.32 ± 4.77**	**24.12 ± 11.11**	**7.509**	**14.804**	**17.072**
Total	**30.42**	**21.29**	**96.47**	**30.036**	**59.215**	**68.287**
S13	4.52 ± 1.67	3.19 ± 4.96	13.60 ± 6.72	4.449	8.769	10.129
S14	3.12 ± 4.61	2.85 ± 6.99	13.35 ± 6.95	3.624	7.087	8.223
S15	4.58 ± 3.39	3.02 ± 4.57	13.48 ± 7.42	4.370	8.631	9.940
S16	4.29 ± 11.42	4.06 ± 3.81	11.73 ± 10.75	4.792	9.363	10.999
Mean	**4.13 ± 0.68**	**3.28 ± 2.54**	**13.04 ± 1.88**	**4.308**	**8.462**	**9.823**
Total	**16.51**	**13.12**	**52.16**	**17.234**	**33.849**	**39.292**

Table 4.3: Activity of natural radionuclides, annual outdoor and indoor effective doses $E_{(out)}$ and $E_{(in)}$, excess lifetime cancer risk (ELCR) of sediment samples from the study areas.

Samples Codes	**^{226}Ra** $(Bq.kg^{-1})$	**^{232}Th** $(Bq.kg^{-1})$	**^{40}K** $(Bq.kg^{-1})$	$E_{(out)}$ $(\mu Sv.yr^{-1})$	$E_{(in)}$ $(\mu Sv.yr^{-1})$	$ELCR_{(out)}$	$ELCR_{(in)}$
S1	11.06 ± 1.02	8.43 ± 2.28	15.65 ± 10.52	12.907	101.624	0.045	0.356
S2	8.94 ± 1.04	5.95 ± 1.90	28.55 ± 10.70	10.611	83.799	0.037	0.293
S3	11.10 ± 3.42	7.98 ± 6.54	24.53 ± 28.31	13.053	102.910	0.046	0.360
S4	13.42 ± 4.27	7.92 ± 9.27	21.36 ± 12.49	14.086	111.792	0.049	0.391
Mean	**11.13 ± 1.83**	**7.57 ± 1.10**	**22.52 ± 5.45**	**12.664**	**100.031**	**0.044**	**0.350**
Total	**44.52**	**30.28**	**90.09**	**50.657**	**400.124**	**0.177**	**1.400**
S5	6.10 ± 1.75	4.62 ± 1.14	132.09 ± 10.32	13.411	104.947	0.047	0.367
S6	3.16 ± 4.27	3.87 ± 1.45	87.39 ± 9.12	9.002	69.869	0.032	0.245
S7	6.53 ± 5.44	5.99 ± 6.22	154.73 ± 33.26	15.801	123.246	0.055	0.431
S8	6.27 ± 7.01	4.01 ± 3.06	125.88 ± 25.58	12.736	99.955	0.045	0.350
Mean	**5.51 ± 1.58**	**4.62 ± 0.97**	**125.02 ± 27.99**	**12.737**	**99.504**	**0.045**	**0.348**
Total	**22.06**	**18.49**	**500.09**	**50.950**	**398.017**	**0.178**	**1.393**
S9	7.84 ± 3.45	4.69 ± 1.19	12.45 ± 7.49	8.274	65.638	0.029	0.230
S10	6.67 ± 2.50	6.43 ± 1.45	39.21 ± 7.67	10.295	80.380	0.036	0.281

S11	6.46 ± 3.75	4.95 ± 2.51	22.99 ± 32.65	8.263	64.978	0.029	0.227
S12	9.45 ± 7.73	5.22 ± 3.92	21.82 ± 34.27	10.004	79.487	0.035	0.278
Mean	**7.60 ± 1.37**	**5.32 ± 4.77**	**24.12 ± 11.11**	**9.209**	**72.621**	**0.032**	**0.254**
Total	**30.42**	**21.29**	**96.47**	**36.837**	**290.484**	**0.129**	**1.017**
S13	4.52 ± 1.67	3.19 ± 4.96	13.60 ± 6.72	5.456	43.017	0.019	0.151
S14	3.12 ± 4.61	2.85 ± 6.99	13.35 ± 6.95	4.445	34.765	0.016	0.122
S15	4.58 ± 3.39	3.02 ± 4.57	13.48 ± 7.42	5.359	42.339	0.019	0.148
S16	4.29 ± 11.42	4.06 ± 3.81	11.73 ± 10.75	5.876	45.931	0.021	0.161
Mean	**4.13 ± 0.68**	**3.28 ± 2.54**	**13.04 ± 1.88**	**5.284**	**41.513**	**0.018**	**0.145**
Total	**16.51**	**13.12**	**52.16**	**21.136**	**166.052**	**0.074**	**0.581**

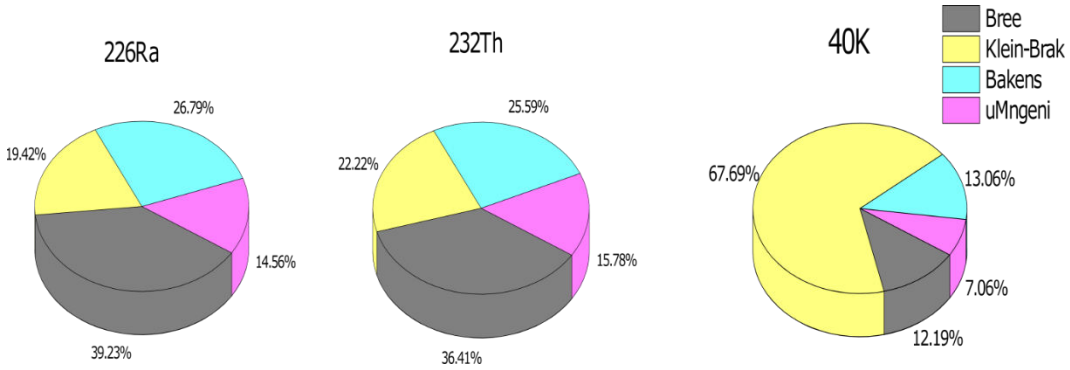

Figure 4.3: Contribution of each natural radionuclide in the sampled location in the present study.

Table 4.4: Activity of natural radionuclides and radiation hazard indices in sediment samples from the studied areas compared with those from other parts of the world.

Country	**Location**	**^{226}Ra**	**^{232}Th**	**^{40}K**	$D_{(out)}$	$D_{(in)}$	R_{eq}	$ELCR$	**References**
South Africa	Bree river	11.13	7.57	22.52	10.326	20.391	23.690	0.394	Present Study
	Klein-Brak river	5.51	4.62	125.02	10.386	20.284	21.750	0.393	
	Bakens river	7.60	5.32	24.12	7.509	14.804	17.072	0.286	
	uMngeni river	4.13	3.28	13.04	4.308	8.462	9.823	0.163	
	Average	7.09	5.20	46.18	8.132	15.985	18.084	0.309	
Nigeria	Delta	8.66	11.66	302.15			35.92-86.23		Iwetan *et al.*, 2015
Egypt	Nile river	16.30	12.94	200.21				0.100	El-Taher *et al.*, 2019
Pakistan	Northern Pakistan	50.66	70.15	531.70	87.47	165.39	190.89	3.21	Aziz *et al.*, 2014
India	Ponnaiyar river	7.31	46.85	384.11	47.07	73.37		0.202	Ramasamy *et al.*, 2009
Ghana	Tono	7.31	6.91	379.94		24.59-	32.25-		Agalga *et al.*, 2013

						45.04	55.14		
Turkey	Kirklardi	37	40	667	118			0.500	Taskin *et al.*, 2009
World Average		33	45	420	59	84	370	1.45	UNSCEAR, 2008

4.5 Conclusion

Natural radioactivity concentrations were determined from sixteen sediment samples collected from oil-rich and non-oil-rich areas of South Africa. Twelve sediment samples are from oil-rich areas (Bree, Klein-Brak, and Bakens rivers), whereas the remaining four are from the non-oil-rich area (uMngeni river) which serves as the control site. The activity concentration values observed in the oil-rich areas sediment samples for ^{226}Ra, ^{232}Th, and ^{40}K are higher for all samples than those of the non-oil-rich area. All concentration values for activity in the river sediments of the studied area were lower than the world average concentrations of 33, 45, and 420 $Bq.kg^{-1}$, provided by the United Nations Scientific Committee on the Effects of Atomic Radiation in 2008 for ^{226}Ra, ^{232}Th, and ^{40}K, respectively. Radiological risk indices calculated were observed to be lower than the maximum acceptable values; thus, there is a low risk of people developing cancer within the studied areas. Also, values reported for this study were within the recommendations of the United Nations Scientific Committee on the Effects of Atomic Radiation. More industrial activities, particularly those involving different radioactive sources usage, may increase the radiation threat in the future that will require attention. This study's findings can be used as a benchmark for possible studies and can serve as reference data for tracking potential radioactivity contamination in the areas studied.

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CHAPTER 5

SOIL-TO-CROP TRANSFER OF NATURAL RADIONUCLIDES IN FARM SOIL OF SOUTH AFRICA

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Soil-to-crop Transfer of Natural Radionuclides in Farm Soil of South Africa

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Abstract

The activity concentration of natural radionuclides in farm soil and most common indigenous food crops (maize, potato, cowpea) in oil-producing (Philippi, Uitenhage, and Hertenbos farms) and non-oil producing (Ukulinga farm) areas of South Africa was measured using a Hyper Pure Germanium detector. Consequently, the transfer of these radionuclides from soil-to-crops was estimated. The mean activity concentration of ^{226}Ra, ^{232}Th and ^{40}K for farm soil samples are 30.71 ± 11.77, 31.97 ± 8.90, 345.97 ± 98.62 $Bq.kg^{-1}$ for Philippi; 18.67 ± 6.70, 31.55 ± 11.48, 191.93 ± 33.39 $Bq.kg^{-1}$ for Uitenhage; 38.03 ± 17.44, 41.18 ± 31.54, 381.89 ± 163.40 $Bq.kg^{-1}$ for Hartenbos, and 8.47 ± 2.87, 8.65 ± 3.52, 94.22 ± 25.97 ± 25.97 $Bq.kg^{-1}$ for Ukulinga. The mean activity concentration of ^{226}Ra, ^{232}Th and ^{40}K for crop samples are 4.54 ± 1.47, 4.87 ± 1.69, 140.18 ± 35.38 $Bq.kg^{-1}$ for Philippi; 9.17 ± 4.79, 3.85 ± 1.87, 136.75 ± 22.04 $Bq.kg^{-1}$ for Uitenhage; 7.97 ± 2.91, 4.62 ± 2.40, 105.97 ± 48.65 $Bq.kg^{-1}$ for Hartenbos, and 4.23 ± 1.63, 2.72 ± 1.19, 48.36 ± 15.55 $Bq.kg^{-1}$ for Ukulinga. The activity concentration and soil-to-crop transfer factors for ^{40}K were found to be much higher, possibly because this element is critical in crop growth. The results showed that the crop samples' transfer factor is in the order cowpea>potato>maize. This study showed that activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in crops and the corresponding transfer factors depend on activity concentrations of the same radionuclides in soil.

Keywords: activity concentration, crops, farm soil, HPGe detector, NORMs, transfer factor.

5.1 Introduction

The environment contains different degrees of natural radioactive materials (NORMs), which vary by geographical location and are enhanced by human activities through industrialization (Karahan and Bayulken, 2000; IAEA, 2007). Natural radionuclides include the primordial radionuclides ^{235}U, ^{238}U, ^{232}Th and their decay chains, ^{14}C, and ^{3}H cosmogenic radionuclides, and ^{40}K (Morcos *et al.*, 1992; UNSCEAR, 2000; Larivière and Guérin, 2010; Ajanaku *et al.,* 2018; Ilori and Alausa, 2019). Naturally occurring radionuclides are available in various samples of the environment, including air, water, plant, and soil (Delko, 1996; Poschi *et al.,* 2007). Oil exploration, extensive uses of fertilizers on farmlands, and mining activities have also been established as primary sources of radiation to the environment and a source of radiological risk to humans (IAEA, 1994; NRC, 1999; Carvalho, 2017).

South Africa's oil and gas sector has been the leading importers and consumers of radioactive materials from 1913 principally for its oil exploration (SAES, 2018). The oil and gas reserves at Bredasdorp and deep marine basins have been primarily explored, leading to massive oil produce in South Africa since 1987 (van Wyk, 1989). These industrial activities may lead to an increase in the background radiation of an environment such as the rivers, soils, and a transfer to humans (ICRP, 1991; IAEA, 2007; Sunday, *et al.* 2019; Ali *et al.,* 2019).

Natural radionuclides present in the soil contributes to its uptake and translocation into edible parts of plants, driven by several factors, such as soil characteristics, plant types, atmosphere, environmental contamination, and

agricultural practices (Thabayneh and Jazzar, 2013; Sunday *et al.,* 2019). Radionuclide transfers from soil-to-crops are known as the main route by which radionuclides are transferred to humans through crops' ingestion. This transfer is defined as a transfer factor and is known to be the most significant human contribution to the dose of radiation (IAEA, 1994; Hany *et al.,* 2019).

Assessment of radionuclides in food crops grown in areas suspected of high radiation is critical in evaluating radionuclides' transfer from soil-to-crops and their risk levels to public health (Khan *et al.,* 1992; Khan *et al.,* 2010; Gilbert *et al.,* 2018). Governments are required to concentrate not only on appropriate food supplies for their people but also on food sources that are chemically and radiological safe (UNSCEAR, 2000). This is part of the UN's primary objective of sustainable food security, which is to help the Member States ensure that their citizens have access to sufficient, nutritionally suitable and, considerably safe foods for human consumption (Jibiri *et al.,* 2007; Pérez-Escamilla, 2017; Yadav *et al.,* 2018; El-Bilali *et al.,* 2019).

South Africa has one of the most diverse and comprehensive crop farming systems growing mainly in vegetables, fruit, nuts, and grain (Abalu and Hassan, 1998; Dredge, 2015). South African climate varies from subtropical to the Mediterranean, allowing for a multitude of opportunities for agriculture (Singh and Singh, 2017). Thus, the most common indigenous food crops (maize, potato, and cowpea) grown and consumed in South Africa were collected for this present study.

Therefore, this study aims to estimate the activity concentrations of natural radionuclides (^{226}Ra, ^{232}Th, and ^{40}K) in farm soils and crops grown in oil-producing (Philippi, Uitenhage, and Hartenbios farms) and non-oil producing (Ukulinga farm) areas of South Africa. Also, the transfer of these radionuclides from soil-to-crops is evaluated.

5.2 Materials and methods

5.2.1 Collection of samples

This study's samples were selected from farms at oil-producing areas and a farm from the non-oil-producing area in South Africa. Soil samples were collected at random within clear boundaries areas of the farmland. The soil samples were taken at a depth of 5-10 cm using a well-cleaned field trowel, where the crop roots are located (Jibiri *et al.,* 2007; Senthilkumar *et al.,* 2010; Usikalu *et al.,* 2014). The farm soil samples for each sampling point were each packaged in labeled polythene packets. The samples of crops (cowpea, maize, and potato) were picked randomly from each selected farm. The samples collected for this study are ready to be harvested and consumed (Jibiri *et al.,* 2007; Tchokossa *et al.,* 2013; Adedokun *et al.,* 2019). The crops were washed thoroughly, packed into labeled polythene packets. The sealed polythene packets containing the soil and crop samples were transferred to the physics discipline laboratory at the University of KwaZulu-Natal, Pietermaritzburg, South Africa. The coordinates were measured and recorded at each sampling location using the Geological Position System (GPS) device. The sample codes, sampling locations, and GPS coordinates are shown in Tables 5.1 and 5.2. Figure 5.1 illustrates the areas of the farmlands selected for the study in South Africa.

5.2.2 Preparation of samples

The soil samples collected from the farms were air-dried for five days at a laboratory temperature of approximately 27^0C and relative humidity of about 70% (IAEA, 1989). Extraneous materials such as plant roots, stones, and decaying plant materials were removed from each of the samples and then dried in an electronic oven at a temperature of 105 ^{0}C until moisture was extracted from all soil samples, and a constant weight was obtained (Tufail *et*

al., 2006; Abu-Khadra *et al.,* 2008; Noli *et al.,* 2017). The crop samples were further cleaned while the edible parts were cut into pieces that were air-dried in the laboratory for over seven days (Gilbert *et al.,* 2018; Adedokun *et al.,* 2019). The crop samples were then dried in an oven at 70^0C until a constant dried weight was obtained for each sample (IAEA, 1989; Jwanbot *et al.,* 2013). The dried samples were blended into fine powders using an electric blender and sieved through a 2 mm pore size mesh to homogeneity (Darko *et al.,* 2015). The sieved parts were weighed into previously weighed Polyvials clear 100 ml plastic pill bottles (IAEA, 1989) to obtain each soil and crop samples' actual weight. The pill-bottles were sealed and stored for at least 28 days to allow natural radionuclides and their short-lived progeny to achieve secular radioactive equilibrium (Hague and Ferdous, 2017; IAEA, 2007; Gilbert *et al.,* 2018). The samples were counted for a 3600 second using the Hyper Pure Germanium (HPGe) detector to estimate the radionuclide activity concentration in the dry samples (Doyi *et al.,* 2017).

Table 5.1: The sampled crops from farmlands within oil-producing regions

Sample Codes	Common Names	Species	Family	Locations	GPS coordinates
C1, C2	Cowpea (Legume)	*Vigna unguiculata*	Euphorbiaceae	Philippi Farms	$34^0 01' 10.9'' S$
C3, C4	Potato (Stems)	*Solanum tuberosum*	Solanacaea		$18^0 33' 46.5'' E$
C5, C6	Maize (Grain)	*Zea mays*	Poaceae		
C7, C8	Cowpea (Legume)	*Vigna unguiculata*	Euphorbiaceae	Uitenhage Farms	$33^0 54' 55.5'' S$
C9, C10	Potato (Stems)	*Solanum tuberosum*	Solanacaea		$25^0 18' 44.6'' E$
C11, C12	Maize (Grain)	*Zea mays*	Poaceae		
C13, C14	Cowpea (Legume)	*Vigna unguiculata*	Euphorbiaceae	Hartenbos Farms	$34^0 06' 13.2'' S$
C15, C16	Potato (Stems)	*Solanum tuberosum*	Solanacaea		$22^0 03' 43.9'' E$
C17, C18	Maize (Grain)	*Zea mays*	Poaceae		

Table 5.2: The sampled crops from farmlands at non-oil-producing region (control)

Sample Codes	Common Names	Species	Family	Locations	GPS coordinates
C19, C20	Cowpea (Legume)	*Vigna unguiculata*	Euphorbiaceae	Ukulinga Farms	$29^0 39' 45.3'' S$
C21, C22	Potato (Stems)	*Solanum tuberosum*	Solanacaea		$30^0 24' 17.7'' E$
C23, C4	Maize (Grain)	*Zea mays*	Poaceae		

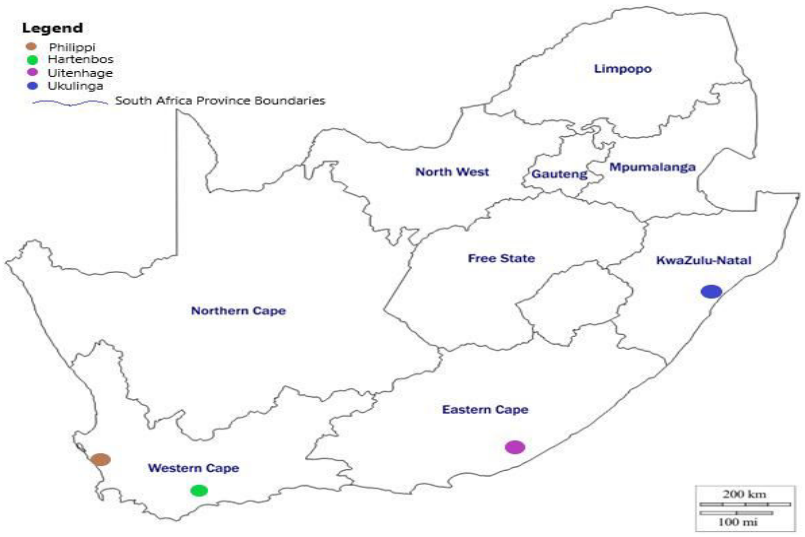


Fig 5.1: South Africa map showing the sampling locations for this study.

5.2.3 Instrumentation

The Hyper Pure Germanium (HPGe) detector was used in this analysis for counting and detecting the radionuclide content in the samples. The detector was cooled to liquid nitrogen temperature, yielding spectroscopic data and pulses proportional to the photon energy captured (Wallbrink *et al.,* 2002; Simon *et al.,* 2012; Guembou *et al.*, 2017). The detector used is 62.5 mm in diameter, 59.5 mm in length with 45 percent relative efficiency, and 2.2 KeV resolutions on the 1332 KeV ^{60}Co line. A fully fitted multichannel analyzer (MCA) was connected to the detector, including a pre-amplification stage, amplifier stage, and display terminal. For the gamma-ray detection experiment,

each sample was placed directly on the detector for 36000 seconds of exposure (Adedokun *et al.,* 2019). In addition to the uncertainty associated with each particular nuclide, the gamma-ray value transition defined from the data spectra was used to determine the specific activity concentration for each radionuclide of interest (Turhan and Gürbüz, 2008; Joel *et al.,* 2016). An estimate of the specific activity concentration was obtained using the weighted average of each nuclide of interest. Data were gathered and analyzed using PalmtopMCA software, which was installed on the computer. The measurements were performed at the Environmental Radiation Laboratory (ERL) of iThemba LABS Cape Town, South Africa.

5.2.4 Energy and efficiency calibration

For the calibration, a volume source with the same geometry as the sample was used to determine the activity concentration of radionuclide present in the samples. The energy calibration was performed by comparing the specific gamma-ray energies in the standard reference material spectrum with the spectrometer channel number. The detector undergoes a full energy peak and efficiency calibration using generic ^{226}Ra, ^{232}Th, and ^{40}K reference sources with an activity concentration of 3252 Bq, 4938.8 Bq, and 13910.8 Bq, respectively. This expression gives the equation relating to the energy and channel number (Joel *et al.,* 2016):

$$E_\gamma = C_1 + C_2 C_N \qquad (5.1)$$

where E_γ is the energy in KeV, C_N is the channel number for a given radionuclide, while C_1 and C_2 are calibration constants for a given geometry.

The efficiency calibration was performed by acquiring a calibration standard spectrum until the total absorption peak count rate can be determined with a statistical uncertainty of less than 1 percent at a 95 percent confidence point.

For the calculation of photo peaks, the net count rate was established to evaluate the output for all the energies used at the measurement time. The output was linked by the count rate correlation and the standard source (Adukpo *et al.*, 2010; Darko *et al.*, 2015):

$$\left(E_\gamma\right)_\varepsilon = \frac{N_e}{(A_c * P_b * t_c)} \qquad (5.2)$$

where N_e is the full energy peak net count corresponding to the energy probability of gamma photons E_γ and gamma emission P_b, A_c is the standard source activity, and the counting time is t_c.

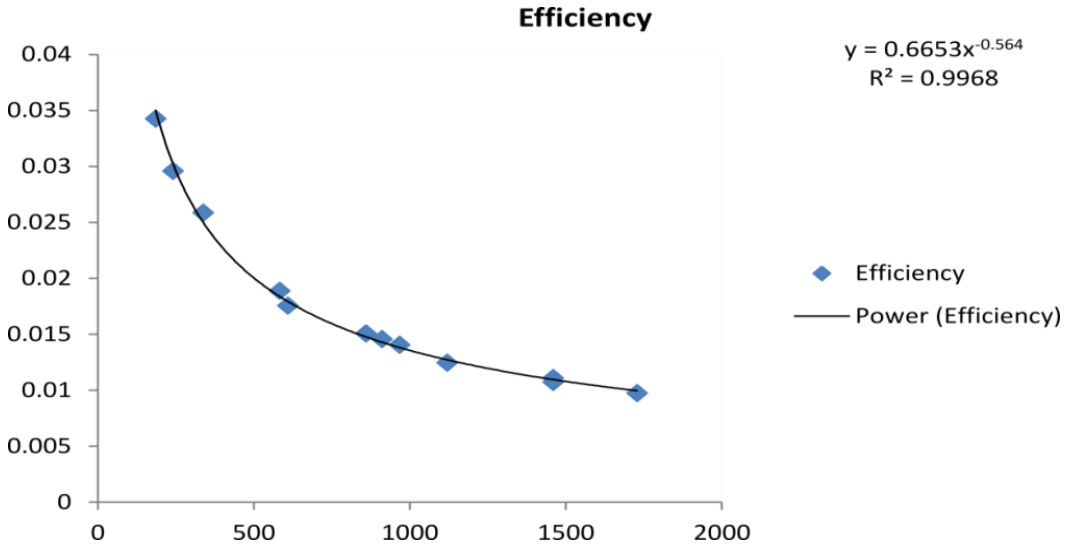


Fig. 5.2: Efficiency calibration curve showing the detection efficiency as a function of the gamma-ray energy used for the HPGe detector.

Therefore, the energy efficiency was plotted as a function of the peak energy and extrapolated for the measurement geometry used to calculate the efficiencies at other peak energies (Chowdhury *et al.,* 1999; Adukpo *et al.,* 2010). The standard reference source was measured for 3600 seconds (Jibiri and Fasae, 2012; Darko *et al.,* 2015; Mekongtso *et al.,* 2016), and the spectrum obtained was used to generate the efficiency curve, and power fitting was performed to get the best R^2-value (Fig. 5.2).

5.2.5 Calculation of activity concentration

The activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K in $Bq.kg^{-1}$ (dry weight) was calculated based on measured efficiency, net count rate, mass, and sample count time of the detector. It is presented in the expression (Jibiri and Fasae, 2012):

$$A_c = \frac{C_\gamma}{P_\gamma . m_s . E_f . t_c} \quad (5.3)$$

where A_c is the activity concentration for each sample, C_γ is the net peak energy, P_γ is the probability of gamma-ray decay, m_s is the mass of the sample in kg, E_f is the efficiency of the detector and t_c is the total counting time in seconds.

For the gamma analysis, each sample was placed directly on the detector for a 36000-seconds exposure duration. The gamma-ray significance transition defined from the data spectra and the uncertainty associated with each particular nuclide was to assess the radionuclides' activity concentration (Solak *et al.,* 2014). A better approximation of activity concentration was obtained by utilizing each nuclide's weighted average of interest's specific activity.

5.2.6 Transfer factor (TF)

The transfer of natural radionuclides from farm soils to crops is determined from the measured concentration of activity in farm soils and corresponding crops. Hence, the soil samples' radionuclides interact with the soil composition and are passed to the soil solutions and soil particles. The proportion of these radionuclides that are passed to the soil solution may be incorporated into crops through plants' roots (Abdulaziz and El-Taher, 2013; Gilbert *et al.,* 2018). The transfer factor values were calculated using equation 4 from the measured radionuclide in the crops with the farm soils:

$$TF = \frac{A_c}{A_s} \quad (5.4)$$

where A_c is the activity of radionuclides in crops, and A_s is the activity of radionuclides in farm soils, in $Bq.kg^{-1}$ dry weight, respectively. The radionuclide transfer factor from soil-to-crop can be used as an index for evaluating trace elements' retention or the transfer of elements from soil to crop (Sabine and Gerald, 2002; Yadav *et al.*, 2018).

5.3 Results and discussion

Table 5.3 displays the results of the naturally occurring radionuclide activity in farm soils in different areas of South Africa. Figure 5.3 shows the distribution of ^{226}Ra, ^{232}Th, and ^{40}K in farm soil samples from the areas studied.

The activity concentration values for farm soil samples at Philippi farm ranged from 14.26 ± 1.19 to 48.89 ± 8.17 $Bq.kg^{-1}$ with a mean value of 30.71 ± 11.77 $Bq.kg^{-1}$ for ^{226}Ra, 22.30 ± 1.41 to 45.11 ± 3.22 $Bq.kg^{-1}$ with a mean value of 31.97 ± 8.90 $Bq.kg^{-1}$ for ^{232}Th, and 237.68 ± 10.89 to 486.51 ± 40.05 $Bq.kg^{-1}$ with a mean value of 345.97 ± 98.62 $Bq.kg^{-1}$for ^{40}K.

The activity concentration values for farm soil samples at Uitenhage farm ranged from 10.52 ± 1.12 to 25.82 ± 3.02 $Bq.kg^{-1}$ with a mean value of 18.67 ± 6.70 $Bq.kg^{-1}$ for ^{226}Ra, 13.06 ± 1.93 to 44.33 ± 5.21 $Bq.kg^{-1}$ with a mean value of 31.55 ± 11.48 $Bq.kg^{-1}$ for ^{232}Th, and 140.19 ± 10.92 to 229.79 ± 12.08 $Bq.kg^{-1}$ with a mean value of 191.93 ± 33.39 $Bq.kg^{-1}$for ^{40}K.

The activity concentration values for farm soil samples at Hartenbos farm ranged from 16.47 ± 1.28 to 64.86 ± 3.01 $Bq.kg^{-1}$ with a mean value of 38.03 ± 17.44 $Bq.kg^{-1}$ for ^{226}Ra, 16.83 ± 1.52 to 88.60 ± 1.17 $Bq.kg^{-1}$ with a mean value of 41.18 ± 31.54 $Bq.kg^{-1}$ for ^{232}Th, and 135.20 ± 17.49 to 604.80 ± 13.42 $Bq.kg^{-1}$ with a mean value of 381.89 ± 163.40 $Bq.kg^{-1}$for ^{40}K.

The activity concentration values for farm soil samples at Ukulinga farm ranged from 5.59 ± 2.21 to 12.96 ± 2.91 $Bq.kg^{-1}$ with a mean value of 8.47 ± 2.87 $Bq.kg^{-1}$ for ^{226}Ra, 4.52 ± 2.05 to 14.11 ± 2.73 $Bq.kg^{-1}$ with a mean value of 8.65 ± 3.52 $Bq.kg^{-1}$ for ^{232}Th, and 62.70 ± 22.58 to 126.51 ± 21.21 $Bq.kg^{-1}$ with a mean value of 94.22 ± 25.97 $Bq.kg^{-1}$for ^{40}K.

The activity concentrations of radionuclide in soil have significant variations that can be due to soil types, soil composition, and extensive fertilizer applications in the farmlands, geological features, and presence of natural resources such as oil (Ghazwa *et al.,* 2016; Adjirackor *et al.,* 2017). Potassium is abundant in all soil samples compared to uranium and thorium, which may be due to its presence in the soil as solutions. ^{40}K also occurs as exchangeable K^+ ion adsorbed or released from soil particle surfaces and organic matter (Ashley *et al.*, 2006). All values reported for the farm soils at the non-oil producing area (Ukulinga farm) were below the world average values. In contrast, some values reported for the farm soils at the oil-producing areas (Philippi, Uitenhage, and Hartenbos farms) are above the world average values of 33, 45, and 450 $Bq.kg^{-1}$ for ^{226}Ra, ^{232}Th, and ^{40}K, respectively.

Table 5.3: Activity concentration of natural radionuclides in samples of farm soils from the studied areas.

Sample Codes	**^{226}Ra ($Bq.kg^{-1}$)**	**^{232}Th ($Bq.kg^{-1}$)**	**^{40}K ($Bq.kg^{-1}$)**
S1	27.62 ± 3.42	29.07 ± 1.02	237.68 ± 10.89
S2	14.26 ± 1.19	31.29 ± 1.47	288.06 ± 9.48
S3	29.33 ± 4.41	45.11 ± 3.22	486.51 ± 40.05
S4	38.25 ± 2.84	39.78 ± 1.18	287.17 ± 10.24
S5	48.89 ± 8.17	22.30 ± 1.41	330.00 ± 15.68
S6	25.94 ± 6.22	24.25 ± 2.01	446.38 ± 37.81
Range	**14.26 – 48.89**	**22.30 – 45.11**	**237.68 – 486.51**
Mean	**30.71 ± 11.77**	**31.97 ± 8.90**	**345.97 ± 98.62**
S7	23.52 ± 2.91	44.33 ± 5.21	166.85 ± 12.46
S8	24.61 ± 1.13	13.06 ± 1.93	214.42 ± 11.14
S9	14.14 ± 1.52	30.09 ± 3.38	140.19 ± 10.92

S10	25.82 ± 3.02	27.00 ± 2.49	210.05 ± 10.47
S11	10.52 ± 1.12	43.00 ± 2.53	190.31 ± 15.22
S12	13.41 ± 1.67	31.83 ± 7.02	229.79 ± 12.08
Range	**10.52 – 25.82**	**13.06 – 44.33**	**140.19 – 229.79**
Mean	**18.67 ± 6.70**	**31.55 ± 11.48**	**191.93 ± 33.39**
S13	16.47 ± 1.28	17.46 ± 2.39	455.05 ± 31.32
S14	22.58 ± 1.44	16.83 ± 1.52	135.20 ± 17.49
S15	64.86 ± 3.01	88.60 ± 1.17	604.80 ± 13.42
S16	42.85 ± 1.81	73.75 ± 1.21	382.54 ± 29.24
S17	35.26 ± 1.27	25.41 ± 1.09	447.27 ± 42.51
S18	46.16 ± 2.31	25.00 ± 1.36	266.47 ± 15.07
Range	**16.47 – 64.86**	**16.83 – 88.60**	**135.20 – 604.80**
Mean	**38.03 ± 17.44**	**41.18 ± 31.54**	**381.89 ± 163.40**
S19	7.90 ± 1.66	7.94 ± 3.21	62.70 ± 22.58
S20	6.16 ± 1.40	14.11 ± 2.73	93.25 ± 22.18
S21	5.59 ± 2.21	4.52 ± 2.05	105.95 ± 20.41
S22	7.34 ± 1.26	8.68 ± 5.29	64.68 ± 15.27
S23	12.96 ± 2.91	11.00 ± 3.22	112.24 ± 11.38
S24	10.86 ± 1.51	5.63 ± 2.41	126.51 ± 21.21
Range	**5.59 – 12.96**	**4.52 – 14.11**	**62.70 – 126.51**
Mean	**8.47 ± 2.87**	**8.65 ± 3.52**	**94.22 ± 25.97**

Table 5.4 shows the measurement of activity concentrations of natural radionuclides in crop samples of different areas of South Africa. Figure 5.4 shows the distribution of ^{226}Ra, ^{232}Th, and ^{40}K in crop samples from the areas studied.

The activity concentration values for crop samples at Philippi farm ranged from 3.08 ± 2.60 to 7.18 ± 4.08 $Bq.kg^{-1}$ with a mean value of 4.54 ± 1.47 $Bq.kg^{-1}$ for ^{226}Ra, 3.58 ± 1.19 to 8.25 ± 2.17 $Bq.kg^{-1}$ with a mean value of 4.87 ± 1.69 $Bq.kg^{-1}$ for ^{232}Th, and 53.12 ± 30.93 to 209.20 ± 23.26 $Bq.kg^{-1}$ with a mean value of 140.18 ± 35.38 $Bq.kg^{-1}$for ^{40}K.

The activity concentration values for crop samples at Uitenhage farm ranged from 4.99 ± 2.38 to 18.33 ± 2.09 $Bq.kg^{-1}$ with a mean value of 9.17 ± 4.79 $Bq.kg^{-1}$ for ^{226}Ra, 2.25 ± 1.68 to 7.32 ± 2.36 $Bq.kg^{-1}$ with a mean value of

3.85 ± 1.87 $Bq.kg^{-1}$ for ^{232}Th, and 105.94 ± 28.27 to 169.39 ± 18.95 $Bq.kg^{-1}$ with a mean value of 136.75 ± 22.04 $Bq.kg^{-1}$ for ^{40}K.

The activity concentration values for crop samples at Hartenbos farm ranged from 5.27 ± 2.28 to 11.54 ± 2.58 $Bq.kg^{-1}$ with a mean value of 7.97 ± 2.91 $Bq.kg^{-1}$ for ^{226}Ra, 2.75 ± 1.91 to 8.86 ± 4.24 $Bq.kg^{-1}$ with a mean value of 4.62 ± 2.40 $Bq.kg^{-1}$ for ^{232}Th, and 40.10 ± 16.38 to 182.02 ± 20.21 $Bq.kg^{-1}$ with a mean value of 105.97 ± 48.65 $Bq.kg^{-1}$for ^{40}K.

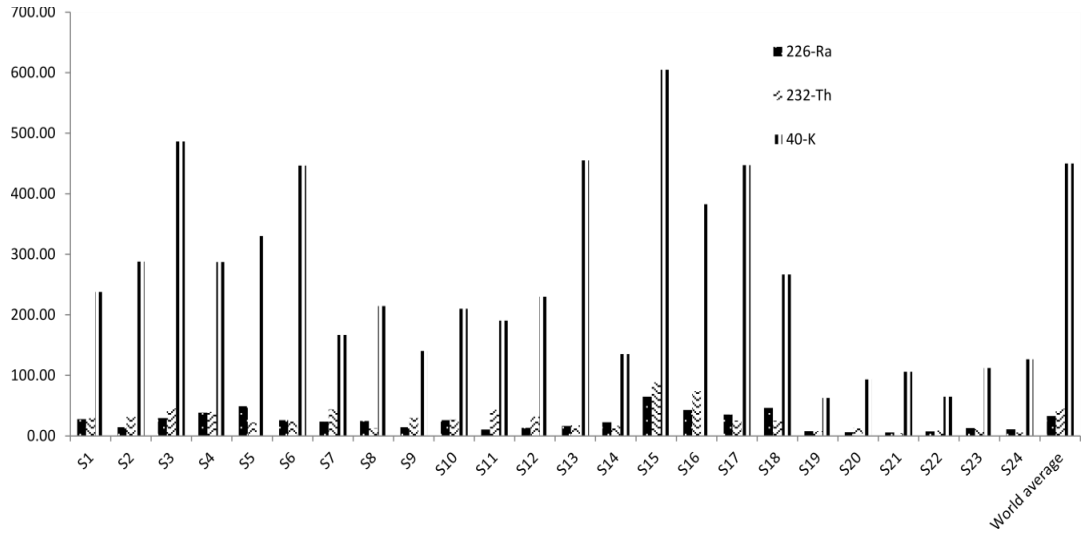
The activity concentration values for crop samples at Ukulinga farm ranged from 3.06 ± 1.77 to 4.94 ± 2.06 $Bq.kg^{-1}$ with a mean value of 4.23 ± 1.63 $Bq.kg^{-1}$ for ^{226}Ra, 1.27 ± 1.04 to 3.90 ± 1.56 $Bq.kg^{-1}$ with a mean value of 2.72 ± 1.19 $Bq.kg^{-1}$ for ^{232}Th, and 28.06 ± 8.87 to 68.51 ± 11.84 $Bq.kg^{-1}$ with a mean value of 48.36 ± 15.55 $Bq.kg^{-1}$for ^{40}K.

The results showed that the crops predominantly absorb the natural radionuclides of ^{226}Ra, ^{232}Th, and ^{40}K. Potassium (^{40}K) appears highest in all crop samples because it is an essential resource for plant growth and crops take up significant quantities of potassium during their life cycle (Jibiri *et al.*, 2007; White and Brown, 2010; Parikh and James, 2012). ^{40}K was highest in potato (*Solanum tuberosum*) of C3 with a value of 209.20 ± 23.26 $Bq.kg^{-1}$ at Philippi farm, ^{232}Th was highest in potato (*Solanum tuberosum*) of C15 with a value of 8.86 ± 4.24 $Bq.kg^{-1}$ at Hartenbos farm, and ^{226}Ra was highest in potato (*Solanum tuberosum*) of C10 with a value of 18.33 ± 2.09 $Bq.kg^{-1}$ at Uitenhage farm, respectively. Hence in the present study, natural radionuclides are the highest in potato samples. ^{40}K has the highest concentration of activity, while ^{226}Ra has a higher concentration of activity than ^{232}Th.

The geological location, soil formation properties, chemical characteristics, soil pH in which the crops are grown, and other natural resources such as oil and gas may also influence the variations in radionuclide concentration in crop samples from different farmlands of the study areas.

Table 5.4: Activity concentration of natural radionuclides in samples of crops from the studied areas.

Sample Codes	^{226}Ra ($Bq.kg^{-1}$)	^{232}Th ($Bq.kg^{-1}$)	^{40}K ($Bq.kg^{-1}$)
C1	7.18 ± 4.08	4.36 ± 2.66	183.01 ± 17.85
C2	4.42 ± 4.11	4.40 ± 1.25	195.88 ± 34.58
C3	3.08 ± 2.60	4.06 ± 1.93	209.20 ± 23.26
C4	3.26 ± 2.74	3.58 ± 1.19	120.61 ± 22.67
C5	4.58 ± 2.98	8.25 ± 2.17	53.12 ± 30.93
C6	4.70 ± 1.43	4.54 ± 1.68	79.26 ± 29.92
Range	**3.08 – 7.18**	**3.58 – 8.25**	**53.12 – 209.20**
Mean	**4.54 ± 1.47**	**4.87 ± 1.69**	**140.18 ± 35.38**
C7	9.33 ± 3.57	2.25 ± 1.68	133.48 ± 20.58
C8	7.17 ± 2.22	4.57 ± 2.74	169.39 ± 18.95
C9	6.08 ± 2.11	2.98 ± 1.31	131.78 ± 14.79
C10	18.33 ± 2.09	3.30 ± 2.26	153.34 ± 15.53
C11	4.99 ± 2.38	2.70 ± 1.41	126.58 ± 28.74
C12	9.12 ± 5.97	7.32 ± 2.36	105.94 ± 28.27
Range	**4.99 – 18.33**	**2.25 – 7.32**	**105.94 – 169.39**
Mean	**9.17 ± 4.79**	**3.85 ± 1.87**	**136.75 ± 22.04**
C13	5.27 ± 2.28	2.86 ± 1.45	182.02 ± 20.21
C14	5.42 ± 3.20	3.03 ± 1.22	75.71 ± 17.93
C15	9.08 ± 4.88	8.86 ± 4.24	120.96 ± 31.81
C16	5.57 ± 3.98	5.90 ± 2.58	91.81 ± 31.73
C17	11.54 ± 2.58	4.32 ± 2.50	40.10 ± 16.38
C18	10.93 ± 2.62	2.75 ± 1.91	125.24 ± 36.86
Range	**5.27 – 11.54**	**2.75 – 8.86**	**40.10 – 182.02**
Mean	**7.97 ± 2.91**	**4.62 ± 2.40**	**105.97 ± 48.65**
C19	4.40 ± 1.26	2.58 ± 1.94	54.20 ± 32.85
C20	3.06 ± 1.77	1.27 ± 1.04	58.03 ± 18.95
C21	4.40 ± 1.66	3.31 ± 1.09	32.17 ± 21.63
C22	4.15 ± 2.63	1.35 ± 1.01	49.20 ± 27.14
C23	4.94 ± 2.06	3.88 ± 1.70	28.06 ± 8.87
C24	4.43 ± 2.01	3.90 ± 1.56	68.51 ± 11.84
Range	**3.06 – 4.94**	**1.27 – 3.90**	**28.06 – 68.51**
Mean	**4.23 ± 1.63**	**2.72 ± 1.19**	**48.36 ± 15.55**



. Fig. 5.3: Distribution of ^{226}Ra, ^{232}Th, and ^{40}K in farm soil samples from the areas studied.

Table 5.5 shows naturally occurring radionuclide transfer factor values from soil-to-crop samples in the areas under this study. Potassium has the highest transfer factor value, then uranium, followed by thorium (^{40}K > ^{226}Ra > ^{232}Th). In the oil-producing areas, the highest transfer factor values of 0.71, 0.37, and 0.94 were recorded for ^{226}Ra, ^{232}Th, and ^{40}K. In contrast, in the non-oil-producing area (control), the highest transfer factor values of 0.79, 0.73, and 0.86 were recorded for ^{226}Ra, ^{232}Th, and ^{40}K. Uranium has been reported to exhibit a higher transfer factor value than thorium due to soil pH and textures (Saeed *et al.,* 2012; Elsaman *et al.,* 2020). Potassium has the highest transfer factor values in all the areas studied, due to its significance in crop growth towards adapting to environmental stresses (Hasanuzzaman *et al.,* 2018). The amount of fertilizers administered to farmlands can also account for higher potassium values in all soil and crop samples. The activity concentrations of natural radionuclides in the soil of the study area and its soil-to-crop transfer values do not pose any radiological threats to human health within the areas studied. Table 5.6 shows the values for the activity concentration of natural radionuclides in farm soil samples from the studied areas compared with those from other parts of the world.

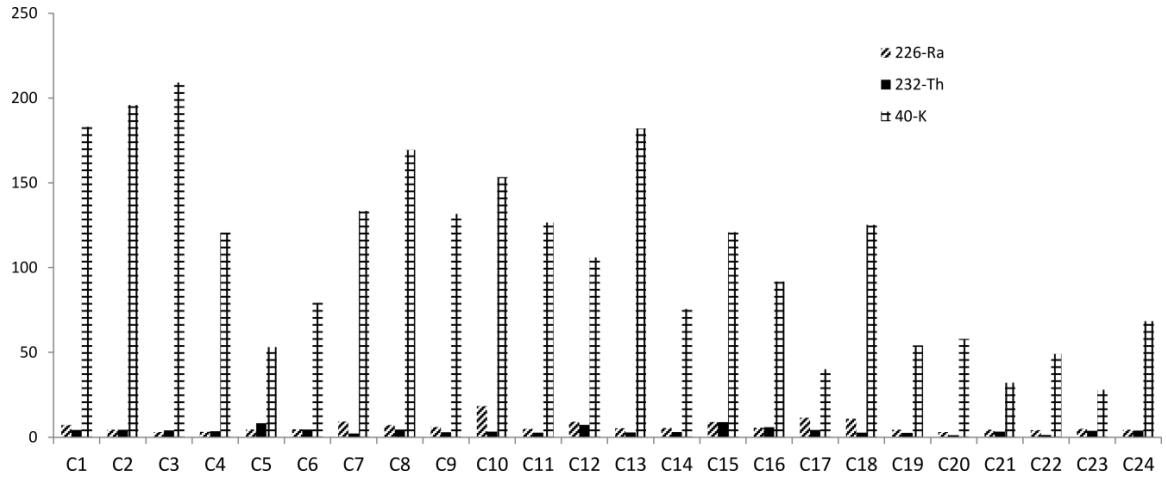


Fig. 5.4: Distribution of ^{226}Ra, ^{232}Th, and ^{40}K in crop samples from the areas studied.

Table 5.5: Soil-to-crops transfer factors in the samples from the studied areas.

Sample Codes	^{226}Ra	^{232}Th	^{40}K
S1 – C1	0.26	0.15	0.77
S2 – C2	0.31	0.14	0.68
S3 – C3	0.11	0.09	0.43
S4 – C4	0.09	0.09	0.42
S5 – C5	0.09	0.37	0.16
S6 – C6	0.18	0.19	0.18
Range	**0.09 – 0.31**	**0.09 – 0.37**	**0.16 – 0.77**
Mean	**0.17**	**0.17**	**0.44**
S7 – C7	0.40	0.05	0.80
S8 – C8	0.29	0.35	0.79
S9 – C9	0.43	0.10	0.94
S10 – C10	0.71	0.12	0.73
S11 – C11	0.47	0.06	0.67
S12 – C12	0.68	0.23	0.46
Range	**0.29 – 0.71**	**0.05 – 0.35**	**0.46 – 0.94**
Mean	**0.50**	**0.15**	**0.73**
S13 – C13	0.32	0.16	0.40
S14 – C14	0.24	0.18	0.56
S15 – C15	0.14	0.10	0.20
S16 – C16	0.13	0.08	0.24
S17 – C17	0.31	0.17	0.09

S18 – C18	0.25	0.11	0.47
Range	**0.13 – 0.32**	**0.08 – 0.18**	**0.09 – 0.56**
Mean	**0.23**	**0.13**	**0.33**
S19 – C19	0.56	0.32	0.86
S20 – C20	0.50	0.09	0.62
S21 – C21	0.79	0.73	0.30
S22 – C22	0.57	0.16	0.76
S23 – C23	0.38	0.35	0.25
S24 – C24	0.41	0.69	0.54
Range	**0.38 – 0.79**	**0.09 – 0.73**	**0.25 – 0.86**
Mean	**0.53**	**0.39**	**0.56**

Table 5.6: Activity of natural radionuclides in farm soil samples from the studied areas compared with those from other parts of the world.

Country (location)	**^{226}Ra**	**^{232}Th**	**^{40}K**	**References**
Malaysia	45.11 – 111.4	51.83 – 127.35	99.2 – 172.85	Ghazwa *et al.,* 2016
Turkey	7.4 – 79.8	9.5 – 170.8	35.7 – 913.8	Ayse *et al.,* 2017
India	18.22 – 90.30	34.80 – 124.68	80.42 – 181.41	Singh *et al.,* 2005
Pakistan	30.3 – 38.7	50.6 – 64.0	560 – 635.6	Akhtar *et al.,* 2005
Algeria	23.72 – 65.47	26.45 – 27.10	220.80 – 260.70	Bramki *et al.,* 2018
Philippi	14.26 – 48.89	22.30 – 45.11	237.68 – 486.51	Present Study (South Africa)
Philippi	10.52 – 25.82	13.06 – 44.33	140.19 – 229.79	
Philippi	16.47 – 64.86	16.83 – 88.60	135.20 – 604.80	
Philippi	5.59 – 12.96	4.52 – 14.11	62.70 – 126.51	
World Average	33	45	450	UNSCEAR, 2008

5.4 Conclusion

In this study, Hyper Pure Germanium (HPGe) detector was used to measure the activity concentration of natural radionuclides in farm soils and crops grown in oil-producing (Philippi, Uitenhage, and Hertenbos farms) and non-oil-producing (Ukulinga farm) areas of South Africa, and consequently the transfer of these radionuclides from soil-to-crops was estimated. The values reported showed a higher activity concentration in farm soils collected at the oil-producing areas compared to the non-oil-producing area. All values reported for the farm soils

at the non-oil producing area are below the recommended world average values. In contrast, some values reported for the farm soils at the oil-producing areas are above the recommended world average values of 33, 45, and 450 $Bq.kg^{-1}$ (UNSCEAR, 2008) for ^{226}Ra, ^{232}Th, and ^{40}K, respectively. Potassium appears highest in all soil and crop samples for the estimated transfer factors. It is an essential resource for plant growth, and crops take up large quantities of this potassium during their life cycle. ^{40}K has the highest value in C3, ^{232}Th has the highest value in C15, and ^{226}Ra has the highest value in C10 at Philippi, Hartenbos, and Uitenhage farm soils, all within the oil-producing areas. The results showed that the crop samples' transfer factor is the order cowpea>potato>maize. The varying transfer factor in crops depends on some factors such as soil characteristics, climatic conditions, type of plants, part of the plant concerned, the physical-chemical form of the radionuclides, and the interfering element (IAEA, 1989; Hany *et al.*, 2019). This study showed that activity concentration ^{226}Ra, ^{232}Th, and ^{40}K in crops and the corresponding transfer factors depend on activity concentrations of the same radionuclides in soil. This study's results can be used as baseline and reference evidence for future investigations in the areas studied.

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CHAPTER 6

ESTIMATION OF NATURAL RADIONUCLIDES AND ITS RADIOLOGICAL HAZARD ASSESSMENT IN SOUTH AFRICA'S FARM SOILS

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Naven Chetty & Abiola Olawale Ilori: Estimation of Natural Radionuclides and its Radiological Hazard Assessment in South Africa's Farm Soils (*under review*).

Estimation of Natural Radionuclides and its Radiological Hazard Assessment in South Africa's Farm Soils

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Abstract

This study aimed to measure the activity concentrations and assess the radiological hazard due to the presence of natural radionuclides in farm-soils collected from South Africa's oil-producing (Philippi, Uitenhage, and Hartenbos) and non-oil producing (Ukulinga) areas. A high-resolution Hyper Pure Germanium (HPGe) detector was used to conduct the gamma-ray measurements. The mean activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the farm soils were 30.71, 31.97, 345.97 $Bq.kg^{-1}$ for Philippi; 18.67, 31.55, 191.93 $Bq.kg^{-1}$ for Uitenhage; 38.03, 41.18, 381.89 $Bq.kg^{-1}$ for Hartenbos and 8.47, 8.65, 94.22 $Bq.kg^{-1}$ for Ukulinga, respectively. The values reported for the farm soils at the non-oil producing area were below the world average values. In contrast, some values reported for the farm soils at the oil-producing areas were above the world average values of 33, 45, and 450 $Bq.kg^{-1}$ for ^{226}Ra, ^{232}Th, and ^{40}K recommended by the United Nations Scientific Committee on the Effects of Atomic Radiation. The radiologic hazard assessments (radium equivalent, absorbed dose rate, annual effective dose, and representative level index) for all the farm soils in the studied areas were within the recommended values. Therefore, at the time of this study, the reported values pose no radiological hazards to humans. In conclusion, it is recommended that the level of activity concentrations of specific radionuclides in the farmlands' soils be periodically monitored. The results in this study may be used as a benchmark and reference data for future investigations.

Keywords: activity concentration, environment, farm soils, HPGe detector, radiation hazards.

6.1 Introduction

Humans have been reported to be routinely exposed to varying degrees of radiation from natural and artificial sources [1, 2]. However, the vast majority of human radiation exposures originate from natural sources, especially from the environment [3, 4]. The natural radiation associated with irradiation's primary external source with the human body comprises the ^{238}U and ^{232}Th series and their decay products and ^{40}K [5, 6].

Exploring natural resources as well as mining activities, and the widespread use of phosphate fertilizers on farmlands will further increase the degree of natural radionuclides present in an environment [7-10]. South Africa has minimally proven oil and gas reserves; however, its oil exploration activities date back decades [11]. The oil and gas industry of the country has been reported to be a leading importer and consumer of radioactive sources that are used in well-logging, density gauges, radiography, radiotracers and leak detection for pipelines [11, 12-15].

The agricultural sector in South Africa is one of the most diverse globally, consisting of extensive, comprehensive, corporate, and private crop-farming systems [16]. Agricultural operations involve broad soils exposure to diverse radiation sources. Thus, the soil is a vital predictor of any potential radiation hazard to humans in the foreseeable future [17]. The measurement of radiation in soils also allows for the identification of areas that are hazardous to human health [18, 19].

In the soil samples of the study areas, there may be a high degree of radionuclide exposure with an additional concern that this could affect both humans and the environment. Measuring the degree of radionuclides in farm

soils may also help monitor human-ingested radionuclides in crops [20, 21]. Therefore, the radiological hazard assessment to the population living in oil-producing areas is of considerable significance because it plays a crucial role in identifying the risks associated with radioactivity present in the environment. It also paves the way for the detection of future changes in environmental radiation activity.

Thus, this study aims to measure natural radionuclides (^{226}Ra, ^{232}Th, and ^{40}K) in the farm soil samples collected from the oil-producing and non-oil producing areas of South Africa. The corresponding radiological hazard assessments were estimated for the radium equivalent, the absorbed dose rate, the annual effective dose, and the farm soil's representative level index in the studied areas.

6.2 Materials and methods

6.2.1 Collection of samples

Soil samples from selected farmlands at oil-producing areas (Philippi, Uitenhage, and Hartenbios farms) and farmland from the non-oil-producing area (Ukulinga farm), South Africa were collected for this study. The sample codes, sampling locations, and GPS coordinates are shown in Table 6.1. The soil samples were collected at random with a well-cleaned hand trowel at a depth of 5-10 cm within clear boundaries of the farmlands [22, 23]. The coordinates were measured and recorded at each sampling location using the Geological Position System (GPS) device. The samples were packaged separately in labeled polythene packets and sealed. The sealed polythene packets containing the farmlands soil samples were transferred to the Laboratory in the Physics discipline at the University of KwaZulu-Natal, Pietermaritzburg, South Africa. Figure 6.1 illustrates the locations of the farmlands selected for the study in South Africa.

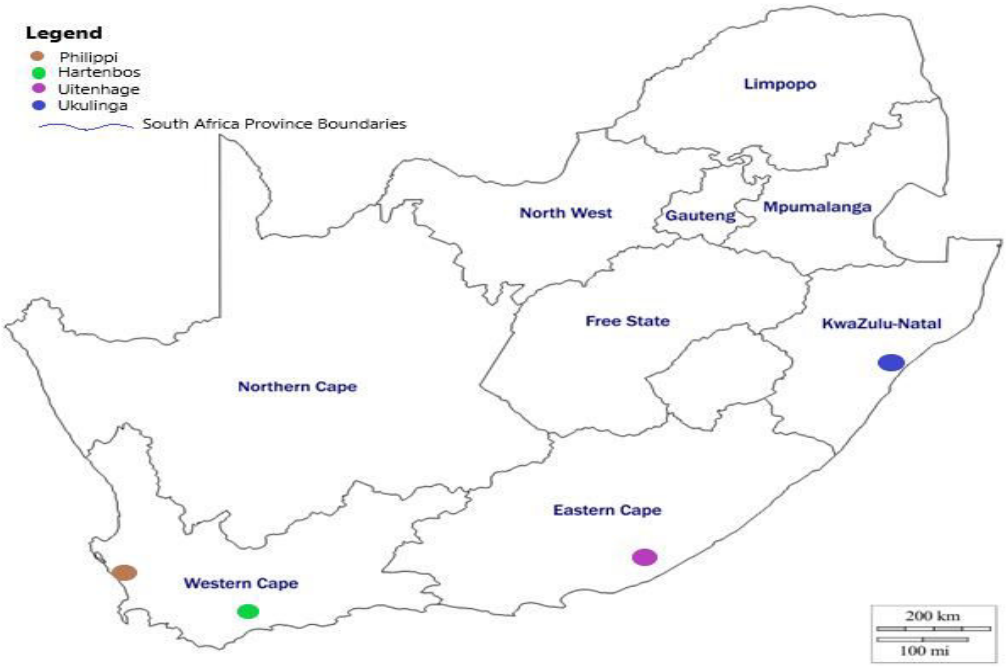

Fig. 6.1: South Africa map showing the location of the farmlands that were being sampled.

6.2.2 Preparation of the samples

The soil samples collected from the farmlands were air-dried for five days at a laboratory temperature of approximately 27 ^{0}C and relative humidity of about 70% [24]. Extraneous materials such as plant roots, stones, and decaying plant materials were removed from each of the samples and then dried in an electronic oven at a temperature of 105 ^{0}C until moisture was extracted from all the soil samples, and a constant weight was obtained [7, 25, 26]. The dried soil samples were then crushed into a fine powder and sieved using a 2 mm pore size mesh to homogeneity. The well-prepared samples of each sampling were weighed and packed into air-tight Polyvials clear 100 ml plastic pill bottles [24]. The pill-bottles were sealed and stored for at least 28 days to allow

natural radionuclides and their short-lived progeny to achieve secular radioactive equilibrium [27-30].

6.2.3 Instrumentation

The Hyper Pure Germanium (HPGe) detector was used in this analysis for counting and detecting the radionuclide content in the samples. The detector was cooled to liquid nitrogen temperature, yielding spectroscopic data and pulses proportional to the photon energy captured [31, 32]. The detector used is a 62.5 mm in diameter, 59.5 mm in length with 45 percent relative efficiency, and 2.2 KeV resolutions on the 1332 KeV ^{60}Co line. A fully fitted multichannel analyzer (MCA) was connected to the detector, including a pre-amplification stage, amplifier stage, and display terminal.

For the gamma-ray detection experiment, each sample was placed directly on the detector for 36000 seconds of exposure [33]. In addition to the uncertainty associated with each particular nuclide, the gamma-ray value transition defined from the data spectra was used to determine the specific activity concentration for each radionuclide of interest [34, 35]. An estimate of the specific activity concentration was obtained using the weighted average of each nuclide of interest. Data were gathered and analyzed using PalmtopMCA software, which was installed on the computer. The measurements were performed at the Environmental Radiation Laboratory (ERL) of iThemba LABS Cape Town, South Africa.

6.2.4 Energy and efficiency calibration

For the calibration, a volume source with the same geometry as the sample was used to determine the activity concentration of radionuclide present in the samples. The energy calibration was performed by comparing the specific

gamma-ray energies in the standard reference material spectrum with the spectrometer channel number. The detector undergoes a full energy peak and efficiency calibration using generic ^{226}Ra, ^{232}Th, and ^{40}K reference sources with an activity concentration of 3252 Bq, 4938.8 Bq, and 13910.8 Bq, respectively. This expression gives the equation relating to the energy and channel number [35]:

$$E_\gamma = C_1 + C_2 C_N \tag{6.1}$$

where E_γ is the energy in KeV, C_N is the channel number for a given radionuclide, while C_1 and C_2 are calibration constants for a given geometry.

The efficiency calibration was performed by acquiring a calibration standard spectrum until the total absorption peak count rate can be determined with a statistical uncertainty of less than 1 percent at a 95 percent confidence point. For the calculation of photo peaks, the net count rate was established to evaluate the output for all the energies used at the measurement time. The output was linked by the count rate correlation and the standard source [36, 37]:

$$\left(E_\gamma\right)_\varepsilon = \frac{N_e}{(A_c * P_b * t_c)} \tag{6.2}$$

N_e is the full energy peak net count corresponding to the energy probability of gamma photons E_γ and gamma emission P_b, A_c is the standard source activity, and the counting time is t_c.

Therefore, the energy efficiency was plotted as a function of the peak energy and extrapolated for the measurement geometry used to calculate the efficiencies at other peak energies. The standard reference source was measured for 3600 seconds [37], the spectrum obtained was used to generate the efficiency curve, and power fitting was performed to get the best R^2-value.

6.3 Results and discussion

6.3.1 Activity concentration

The activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K in $Bq.kg^{-1}$ (dry weight) was determined based on the detector's measured efficiency, net count rate, mass, and sample count time. It is presented in the expression [38, 39]:

$$A_c = \frac{C_\gamma}{P_\gamma . m_s . E_f . t_c} \quad (6.3)$$

where A_c is the activity concentration of each sample; C_γ is the net peak energy; P_γ is the probability of gamma-ray decay; m_s is the mass of the sample in kg; E_f is the detector's efficiency; t_c is the total counting time in seconds.

The results of assessing the activity concentrations of natural radionuclides in the farm soils of the studied areas are shown in Table 6.1. The result of the distribution of ^{226}Ra, ^{232}Th, and ^{40}K in farm soil samples from the areas studied is shown in Figure 6.2.

Table 6.1: The activity concentration of natural radionuclides in samples of farm soils from the studied areas.

Sample Codes	Farm Names	GPS coordinates	**^{226}Ra ($Bq.kg^{-1}$)**	**^{232}Th ($Bq.kg^{-1}$)**	**^{40}K ($Bq.kg^{-1}$)**
S1	Philippi	$34^0 01' 10.9'' S$ $18^0 33' 46.5'' E$	27.62 ± 3.42	29.07 ± 1.02	237.68 ± 10.89
S2			14.26 ± 1.19	31.29 ± 1.47	288.06 ± 9.48
S3			29.33 ± 4.41	45.11 ± 3.22	486.51 ± 40.05
S4			38.25 ± 2.84	39.78 ± 1.18	287.17 ± 10.24
S5			48.89 ± 8.17	22.30 ± 1.41	330.00 ± 15.68
S6			25.94 ± 6.22	24.25 ± 2.01	446.38 ± 37.81
Range			**14.26 – 48.89**	**22.30 – 45.11**	**237.68 – 486.51**
Mean			**30.71 ± 11.77**	**31.97 ± 8.90**	**345.97 ± 98.62**
S7	Uitenhage	$33^0 54' 55.5'' S$ $25^0 18' 44.6'' E$	23.52 ± 2.91	44.33 ± 5.21	166.85 ± 12.46
S8			24.61 ± 1.13	13.06 ± 1.93	214.42 ± 11.14
S9			14.14 ± 1.52	30.09 ± 3.38	140.19 ± 10.92
S10			25.82 ± 3.02	27.00 ± 2.49	210.05 ± 10.47

S11			10.52 ± 1.12	43.00 ± 2.53	190.31 ± 15.22
S12			13.41 ± 1.67	31.83 ± 7.02	229.79 ± 12.08
Range			**10.52 – 25.82**	**13.06 – 44.33**	**140.19 – 229.79**
Mean			**18.67 ± 6.70**	**31.55 ± 11.48**	**191.93 ± 33.39**
S13			16.47 ± 1.28	17.46 ± 2.39	455.05 ± 31.32
S14		$34^0 06' 13.2'' S$	22.58 ± 1.44	16.83 ± 1.52	135.20 ± 17.49
S15	Hartenbos	$22^0 03' 43.9'' E$	64.86 ± 3.01	88.60 ± 1.17	604.80 ± 13.42
S16			42.85 ± 1.81	73.75 ± 1.21	382.54 ± 29.24
S17			35.26 ± 1.27	25.41 ± 1.09	447.27 ± 42.51
S18			46.16 ± 2.31	25.00 ± 1.36	266.47 ± 15.07
Range			**16.47 – 64.86**	**16.83 – 88.60**	**135.20 – 604.80**
Mean			**38.03 ± 17.44**	**41.18 ± 31.54**	**381.89 ± 163.40**
S19			7.90 ± 1.66	7.94 ± 3.21	62.70 ± 22.58
S20		$29^0 39' 45.3'' S$	6.16 ± 1.40	14.11 ± 2.73	93.25 ± 22.18
S21	Ukulinga	$30^0 24' 17.7'' E$	5.59 ± 2.21	4.52 ± 2.05	105.95 ± 20.41
S22			7.34 ± 1.26	8.68 ± 5.29	64.68 ± 15.27
S23			12.96 ± 2.91	11.00 ± 3.22	112.24 ± 11.38
S24			10.86 ± 1.51	5.63 ± 2.41	126.51 ± 21.21
Range			**5.59 – 12.96**	**4.52 – 14.11**	**62.70 – 126.51**
Mean			**8.47 ± 2.87**	**8.65 ± 3.52**	**94.22 ± 25.97**

The activity concentration values for farm soil samples at Philippi farm ranged from 14.26 ± 1.19 to 48.89 ± 8.17 $Bq.kg^{-1}$ with a mean value of 30.71 ± 11.77 $Bq.kg^{-1}$, 22.30 ± 1.41 to 45.11 ± 3.22 $Bq.kg^{-1}$ with a mean value of 31.97 ± 8.90 $Bq.kg^{-1}$ and 237.68 ± 10.89 to 486.51 ± 40.05 $Bq.kg^{-1}$ with a mean value of 345.97 ± 98.62 $Bq.kg^{-1}$for ^{226}Ra, ^{232}Th and ^{40}K.

The activity concentration values for farm soil samples at Uitenhage farm ranged from 10.52 ± 1.12 to 25.82 ± 3.02 $Bq.kg^{-1}$ with a mean value of 18.67 ± 6.70 $Bq.kg^{-1}$, 13.06 ± 1.93 to 44.33 ± 5.21 $Bq.kg^{-1}$ with a mean value of 31.55 ± 11.48 $Bq.kg^{-1}$ and 140.19 ± 10.92 to 229.79 ± 12.08 $Bq.kg^{-1}$ with a mean value of 191.93 ± 33.39 $Bq.kg^{-1}$for ^{226}Ra, ^{232}Th and ^{40}K.

The activity concentration values for farm soil samples at Hartenbos farm ranged from 16.47 ± 1.28 to 64.86 ± 3.01 $Bq.kg^{-1}$ with a mean value of 38.03 ± 17.44 $Bq.kg^{-1}$, 16.83 ± 1.52 to 88.60 ± 1.17 $Bq.kg^{-1}$ with a mean value of 41.18

± 31.54 $Bq.kg^{-1}$ and 135.20 ± 17.49 to 604.80 ± 13.42 $Bq.kg^{-1}$ with a mean value of 381.89 ± 163.40 $Bq.kg^{-1}$for ^{226}Ra, ^{232}Th and ^{40}K.

The activity concentration values for farm soil samples at Ukulinga farm ranged from 5.59 ± 2.21 to 12.96 ± 2.91 $Bq.kg^{-1}$ with a mean value of 8.47 ± 2.87 $Bq.kg^{-1}$, 4.52 ± 2.05 to 14.11 ± 2.73 $Bq.kg^{-1}$ with a mean value of 8.65 ± 3.52 $Bq.kg^{-1}$ and 62.70 ± 22.58 to 126.51 ± 21.21 $Bq.kg^{-1}$ with a mean value of 94.22 ± 25.97 $Bq.kg^{-1}$for ^{226}Ra, ^{232}Th and ^{40}K.

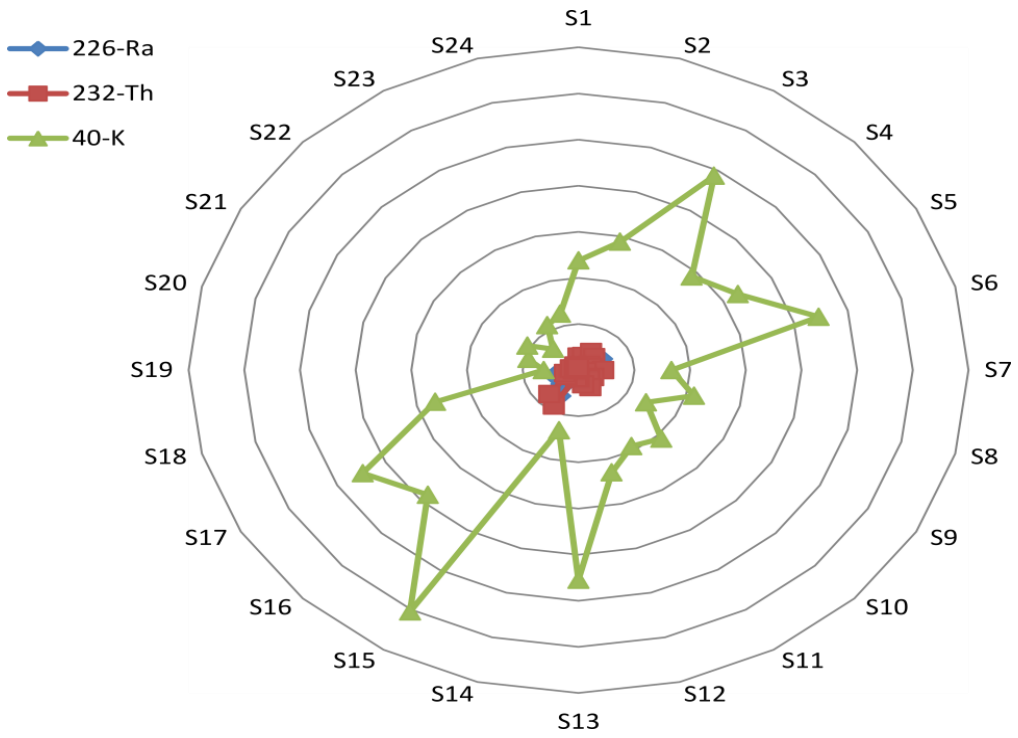

Fig. 6.2: Distribution of ^{226}Ra, ^{232}Th, and ^{40}K in farm soil samples from the areas studied.

All values reported for the farm soils at the non-oil producing area (Ukulinga farm) were below the world average values. In contrast, some values reported for the farm soils at the oil-producing areas (Philippi, Uitenhage, and Hartenbos farms) are above the world average values of 33, 45, and 450 $Bq.kg^{-1}$ for ^{226}Ra, ^{232}Th, and ^{40}K [1].

Figure 6.2 confirms that naturally occurring potassium (^{40}K) is the primary radiation source for humans and is considered an essential element in farm soils [6, 24]. The high concentration of potassium in soil samples was due to the tiny potassium ions trapped inside the clay particle composition of the soil, attributed to the crystalline forces that hold these potassium ions [40, 41]. The potassium of large amounts has also been reported in various agricultural fertilizers [42, 43]. The high presence of potassium in farm soils of Philippi and Hartenbos (Western Cape) may be attributed to the massive application of fertilizers within the farmlands to grow different crops.

6.4 Radiological hazard assessments

6.4.1 Radium equivalent (R_{eq})

The index of radium activity provides a valuable framework for controlling safety standards, as it applies to both the external and internal radon doses and its progeny. The index of activity equivalent to radium was calculated as given the equation [1]:

$$R_{eq}(Bq.kg^{-1}) = C_{Ra} + 1.43C_{Th} + 0.077C_K \quad (6.4)$$

C_{Ra}, C_{Th}, and C_K are the radioactivity concentration in $Bq.kg^{-1}$ of ^{226}Ra, ^{232}Th, and ^{40}K.

The estimated radium equivalent R_{eq} ($Bq.kg^{-1}$) values in farm soils at the oil-producing areas (Philippi, Uitenhage, and Hartenbos) ranged from 81.183 to 131.304 with a mean value of 103.064 $Bq.kg^{-1}$, 59.793 to 99.768 with a mean value of 78.567 $Bq.kg^{-1}$ and 57.065 to 238.125 with a mean value of 126.316 $Bq.kg^{-1}$, respectively. In contrast, farm soils at the non-oil-producing area (Ukulinga) ranged from 20.200 to 37.331, with a mean value of 28.087 $Bq.kg^{-1}$ (Table 6.2). The reported values were lower than 370 $Bq.kg^{-1}$ [1], the recommended value for the world average radium equivalent activity.

6.4.2 Absorbed dose rate (D)

The absorbed dose rate D ($nGyh^{-1}$) at 1 m above ground level was calculated to ensure a uniform distribution of radionuclides ^{226}Ra, ^{232}Th, and ^{40}K in $Bq.kg^{-1}$ [1, 44, 45]. This parameter can assess the radiological risk of radionuclides present in agricultural soils [46]. Therefore, the absorbed dose rate was calculated using the equation [22, 47, 48].

$$D\ (nGyh^{-1}) = 0.427C_{Ra} + 0.623C_{Th} + 0.043C_K \quad (6.5)$$

where D is the absorbed dose rate ($nGyh^{-1}$). C_{Ra}, C_{Th}, C_K is the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in $Bq.kg^{-1}$, respectively.

As shown in Table 6.2, the absorbed dose rate determined for ^{226}Ra, ^{232}Th and ^{40}K radioactivity concentrations in farm soils at the oil-producing areas ranged from 37.968 to 61.550 with a mean value of 47.906 $nGyh^{-1}$ for Philippi, 27.864 to 44.839 with a mean value of 35.882 $nGyh^{-1}$ for Uitenhage and 25.944 to 108.898 with a mean value of 58.312 Gyh^{-1} for Hertenbos, respectively. In contrast, the absorbed dose rate in farm soils at the non-oil-producing area ranged from 9.754 to 17.213, with a mean value of 13.054 $nGyh^{-1}$ for Ukulinga. The values reported for farm soil samples were below the recommended world average value of 60 $nGyh^{-1}$ [1]. However, the absorbed dose rates reported for farm soil samples S3 (61.550 $nGyh^{-1}$), S15 (108.898 $nGyh^{-1}$), and S16 (80.691 $nGyh^{-1}$) from the oil-producing areas are above the recommended world average value.

6.4.3 Annual effective dose rate (E)

The annual effective dose rate estimates the stochastic impact of radiation dose exposure to humans [49]. The conversion coefficient from the absorbed dose rate was considered in estimating the annual effective dose rate. The conversion coefficient of 0.7 $Sv.Gy^{-1}$ was used to calculate the annual effective dose rate in $\mu Sv.yr^{-1}$, while 0.2 (20%) was used as the time of stay in the

outdoor in a year [50, 51]. The corresponding annual effective dose rate was calculated with the equation [1, 29, 52]:

$$E\ (\mu Sv.yr^{-1}) = D(nGy.h^{-1})\ \text{x}\ 8760\ (h.y^{-1})\ \text{x}\ 0.2\ \text{x}\ 0.7\ (Sv.Gy^{-1})\ \text{x}\ 10^{-3} \qquad (6.6)$$

The annual effective dose rate calculated for the farm soils at the oil-producing areas ranged from 46.564 to 75.484 with a mean value of 58.752 $\mu Sv.yr^{-1}$ for Philippi, 34.172 to 54.990 with a mean value of 44.005 $\mu Sv.yr^{-1}$ for Uitenhage and 31.817 to 133.553 with a mean value of 71.514 $\mu Sv.yr^{-1}$ for Hertenbos, respectively. In contrast, the annual effective dose rate for the farm soils at the non-oil-producing area ranged from 11.962 to 21.109, with a mean value of 16.009 $\mu Sv.yr^{-1}$ for Ukulinga. The average annual effective dose rate values reported for farm soils at Philippi, Uitenhage, and Ukulinga were below the recommended average world value of 70 $\mu Sv.yr^{-1}$ [1]. However, the average annual effective dose rate reported value for Hertenbos is above the world average.

6.4.4 Representative level index ($I_{\gamma r}$)

The radiation hazards attributable to the ^{226}Ra, ^{232}Th, and ^{40}K radionuclides can also be measured using the index called the representative level index. The representative level index (Iyr) is used to estimate the level of gamma radiation hazard associated with the natural radionuclides in specific soil of an environment [53-56]. The ensuing equation was used to determine the representative level index for the farm soil samples collected from the study areas [15, 55, 57]:

$$I_{\gamma r} = \left(\frac{1}{150}\right) C_{Ra} + \left(\frac{1}{100}\right) C_{Th} + \left(\frac{1}{1500}\right) C_K \qquad (6.7)$$

where C_{Ra}, C_{Th}, and C_K are the radioactivity concentration in $Bq.kg^{-1}$ of ^{226}Ra, ^{232}Th, and ^{40}K, respectively.

The representative level index reported for farm soils at oil-producing areas ranged from 0.600 to 0.971 with a mean value of 0.755 for Philippi, 0.438 to

0.711, with a mean value of 0.568 for Uitenhage and 0.409 to 1.72 with a mean value of 0.920 for Hertenbos, respectively. In contrast, the representative level index for farm soils at the non-oil-producing area ranged from 0.153 to 0.271, with a mean value of 0.206 for Ukulinga. Values of index reported in farm soil samples except for samples S15 (1.722) and S16 (1.278) are within the recommended dose of 1, which is the recommended index value for the population given by the European Commission Radiation Protection [58]. Figure 6.3 shows the mean activity concentration of ^{226}Ra, ^{232}Th, ^{40}K, and Ra_{eq} in farm soil samples from the areas studied and recommended world average values.

Table 6.2: Radium Equivalent Dose (R_{eq}), Absorbed Dose Rate (D), Annual Effective Dose Rate (E), and Representative Level Index ($I_{\gamma r}$) of farm soil samples from the study areas.

Sample Codes	R_{eq} $(Bq.kg^{-1})$	D $(nGyh^{-1})$	E $(\mu Sv.yr^{-1})$	$I_{\gamma r}$
1	87.482	40.120	49.204	0.633
2	81.183	37.968	46.564	0.600
3	131.304	61.550	75.484	0.971
4	117.244	53.462	65.566	0.844
5	106.184	48.957	60.041	0.769
6	94.987	45.378	55.651	0.713
Range	**81.183-131.304**	**37.968-61.550**	**46.564-75.484**	**0.600-0.971**
Mean	**103.064**	**47.906**	**58.752**	**0.755**
7	99.768	44.839	54.990	0.711
8	59.793	27.864	34.172	0.438
9	67.964	30.812	37.788	0.489
10	80.601	36.877	45.226	0.582
11	86.660	39.463	48.397	0.627
12	76.617	35.435	43.458	0.561
Range	**59.793-99.768**	**27.864-44.839**	**34.172-54.990**	**0.438-0.711**
Mean	**78.567**	**35.882**	**44.005**	**0.568**
13	76.478	37.478	45.963	0.588
14	57.065	25.944	31.817	0.409
15	238.125	108.898	133.553	1.722
16	177.764	80.691	98.959	1.278
17	106.037	50.119	61.466	0.787

18	102.428	46.743	57.326	0.735
Range	**57.065-238.125**	**25.944-108.898**	**31.817-133.553**	**0.409-1.722**
Mean	**126.316**	**58.312**	**71.514**	**0.920**
19	24.080	11.015	13.509	0.174
20	33.519	15.431	18.925	0.244
21	20.200	9.754	11.962	0.153
22	24.742	11.327	13.892	0.179
23	37.331	17.213	21.109	0.271
24	28.649	13.583	16.658	0.213
Range	**20.200-37.331**	**9.754-17.213**	**11.962-21.109**	**0.153-0.271**
Mean	**28.087**	**13.054**	**16.009**	**0.206**

Table 6.3: Activity of natural radionuclides and radiation hazard indices in farm soil samples from the studied areas compared with those from other parts of the world.

Country	**Location**	**^{226}Ra**	**^{232}Th**	**^{40}K**	R_{eq}	D	E	I_{yr}	**References**
Malaysia	Kedah	102.08	133.96	136.98	458.785	141.62	169	1.07	[22]
Yemen	Abyan Delta	33.15	77.25	1220.59	-	-	-	-	[59]
Turkey	Rize	7.4-79.8	9.5-170.8	35.7-913.8	125.0	10.7-156.4	13.1-191.8	-	[60]
Qatar	-	17	10	201	47	22	27	-	[61]
India	Kanyakumari	44.07	215.0	1585	437	200	-	3.3	[62]
Nigeria	Ibadan	12.46	16.73	207.19	56.59	26.38	32	0.42	[63]
Saudi Arabia	Jeddah	44.87	54.59	2652.30	337.58	-	169.73	-	[64]
Pakistan	Faisalabad	30.3-38.7	50.6-64.0	560 - 635.6	-	68-83	-	-	[65]
Egypt	Nile Delta	35.53	23.59	266.41	-	-	-	-	[66]
South Africa	Philippi	30.71	31.97	345.97	103.064	47.906	58.752	0.755	Present Study
	Uitenhage	18.67	31.55	191.93	78.567	35.882	44.005	0.568	
	Hartenbos	38.03	41.18	381.89	126.316	58.312	71.514	0.920	
	Ukulinga	8.47	8.65	94.22	28.087	13.054	16.009	0.206	
	Average	23.97	28.34	253.50	84.01	38.79	47.57	0.6124	
World Average		33[a]	45[a]	450[a]	370[b]	60[b]	70[b]	1.00[c]	[a][67] [b][1] [c][58]

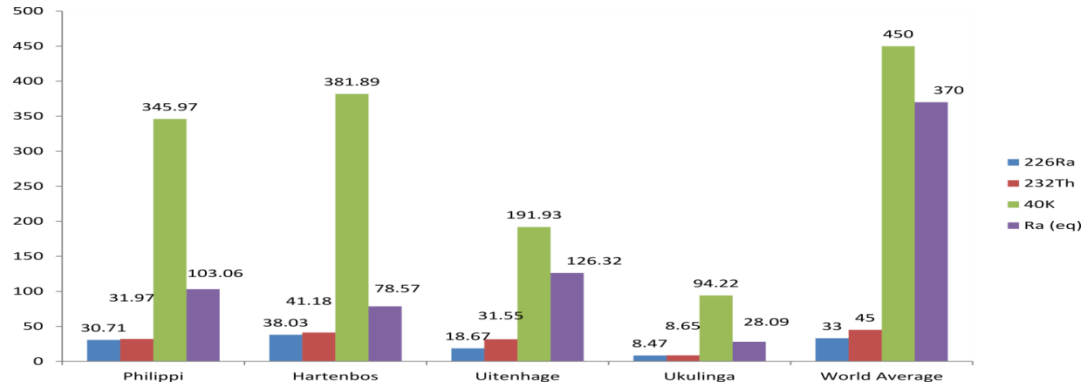

Fig. 6.3: Mean activity concentration of ^{226}Ra, ^{232}Th, ^{40}K, and Ra(eq) in farm soil samples from the areas studied and recommended world average values.

6.5 Conclusion

The activity concentration of natural radionuclides in farm soils from the oil and non-oil producing (control site) areas of South Africa was measured using a Hyper Pure Germanium (HPGe) detector. The radiological hazard assessments were also calculated due to the existence of these radionuclides. The mean radionuclide concentrations recorded for the farm soils in the oil and non-oil producing areas were below the world average values of 33, 45, and 450 $Bq.kg^{-1}$ for ^{238}U, ^{232}Th, and ^{40}K, respectively. However, in the oil-producing areas, the radiation levels recorded for farm soil samples S3, S15, and S16 were above these recommended world average values. The radiological hazard assessments of radium equivalent, the absorbed dose rate, the annual effective dose rate, and the representative level index for all the study samples were within the recommended safe radiological values. The activity concentrations of radionuclides in the farm soil samples reported for these study areas do not pose any radiological hazard risks. However, excessive use of radioactive sources, the widespread use of fertilizers on farmlands, and industrial activities in the study area may increase the radiological burden, leading to increases in radiation exposure. This analysis's findings can be used in the areas studied as a benchmark and reference data for future investigations.

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CHAPTER 7

Summary and future work

7.1 Summary

This study was conducted to measure the activity concentrations of the naturally occurring radioactive materials in various environmental samples such as the farm-soil, crops, river sediments, and selected fish samples collected from South Africa's oil-producing/non-oil producing area and estimates its radioactivity hazard indices relative to human health.

The presence of natural radioactivity in the environment may result in internal and external radiation exposure to humans. People are not aware of the sources of radiation within their territories. Although high radiation levels are harmful to organisms, some environmental radiations are essential to life, such as background radiation, which has contributed significantly to the fundamental processes of chemical and biological evolution. The presence of natural resources such as oil and gas have also added to the natural background radiation. The naturally occurring sources contribute approximately four to five times the human-made sources.

South Africa has minimal proven oil and gas reserves; however, oil production and research have driven for specific years back in the Orange Basin off the west coast and the Bredasdorp Basin on the south coast of Cape Province of South Africa. These oil industries are among the largest importers and users of radioactive materials in its economic sectors that often increase the oil-producing areas' background radiation. Hence, it increases the radiation risk hazard to humans and other living organisms within such environments. The natural radionuclides enhanced their way into the human body, mainly via food and water in the natural surroundings.

Bree, Klein-Brak, and Bakens rivers were selected from the oil-producing areas for the fish and river sediment samples. Also, the UMngeni river was selected for the non-oil producing area serving as the control. Philippi, Uitenhage, and Hartenbos farms were selected for the oil-producing areas for the farm soil and crop samples. In comparison, the Ukulinga farm was selected for the non-oil producing area.

The HPGe detector was used to count and detect the radionuclide content in the environmental samples collected for this study. The detector model is GC4520, a P-type co-axial detector with pre-amplifier and amplifier models 2002CSL and ORTEC 572).

The measured activity concentrations for river sediments at the oil-rich areas (Bree, Klein-Brak, and Bakens rivers) were 12.6%, 21.4%, and 15.6% higher than that of the control site, the non-oil-area (uMngeni river) for ^{238}U, ^{232}Th, and ^{40}K, respectively. The results obtained for the radiation risk hazard doses due to the presence of NORMs in all the fish samples were lower than the UNSCEAR recommended doses of 1 $mSv.y^{-1}$ for the effective annual intake dose and 300 $\mu Sv.yr^{-1}$ for the annual gonadal equivalent dose.

A higher activity concentration in farm soils collected at the oil-producing areas was recorded compared to the non-oil-producing area. Potassium is an essential resource for plant growth, and crops take up large quantities of this

potassium during their life cycle. Therefore, it appears highest in all soil and crop samples for the estimated transfer factors. The study showed that activity concentration ^{226}Ra, ^{232}Th, and ^{40}K in crops and the corresponding transfer factors depend on activity concentrations of the same radionuclides in soil.

Conclusively, most activity concentrations and radiological dose estimates recorded were within the values of 33, 45, and 450 $Bq.kg^{-1}$ for the studied areas for ^{226}Ra (^{238}U), ^{232}Th, and ^{40}K recommended by the United Nations Scientific Committee on the Effects of Atomic Radiation. However, all the values obtained showed a substantial difference between the natural radionuclide concentrations of the oil-rich samples relative to those of the non-oil-rich areas. Therefore there are no radiological threats due to the activity concentrations in the selected environmental samples at the time of this analysis. As a benchmark and reference data for future inquiries, the findings of this study may be used.

7.2 Future work

As discussed in this study, our research focused on the assessment of naturally occurring radioactive materials (NORMs) such as uranium (^{238}U), thorium (^{232}Th), and potassium (^{40}K) in environmental samples of South Africa's oil-producing areas and its radiological dose assessment concerning the health of human and aquatic species.

Future research involving a wide range of samples from aquatic species and food crops widely consumed by humans living in the areas sampled could contribute to a more accurate reflection of radiological impact assessments due to the existence of NORMs. Due to the pervasive use of radioactive sources in oil exploration by industries, Technically Enhanced NORMS and artificial sources may also be investigated in the future. Hence, various pollutants from radioactive materials in the environment may be studied, and their corresponding impacts on humans and the environment.

Finally, these findings should be viewed in the sense of this research, given the limitations of the sampling size from the areas studied, the High Purity Germanium Detector (HPGe) system's calibration, and its parameters. Future research elsewhere would be of importance to corroborate these results.